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35

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Assessing recycling strategies for  
critical raw materials in waste  
electrical and electronic equipment

*Maximilian Ueberschaar*

# Assessing recycling strategies for critical raw materials in waste electrical and electronic equipment

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vorgelegt von  
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von der Fakultät III – Prozesswissenschaften  
der Technischen Universität Berlin

zur Erlangung des akademischen Grades  
Doktor der Ingenieurwissenschaften  
– Dr.-Ing. –

genehmigte Dissertation

## **Promotionsausschuss**

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Tag der wissenschaftlichen Aussprache: 09. Juni 2017

Berlin 2017

## **Bibliografische Information der Deutschen Nationalbibliothek**

Die Deutsche Nationalbibliothek verzeichnet diese Publikation in der Deutschen Nationalbibliografie; detaillierte bibliografische Daten sind im Internet über <http://dnb.dnb.de> abrufbar.

## **Bibliographic information published by the Deutsche Nationalbibliothek**

The Deutsche Nationalbibliothek lists this publication in the Deutsche Nationalbibliografie; detailed bibliographic data are available in the Internet at <http://dnb.dnb.de>.

Schriftenreihe des	Institut für Technischen Umweltschutz Band 35, 2017
Herausgeber	Institut für Technischen Umweltschutz der Geschäftsführende Direktor, Sekr. Z1 Straße des 17. Juni 135 10623 Berlin <a href="http://www.itu.tu-berlin.de">www.itu.tu-berlin.de</a>
Redaktion	Heike Otto

Zugl.: Berlin, Technische Universität, Diss., 2017

## **D83**

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1. Auflage, 2017

**ISSN 1864-5984**

**ISBN 978-3-86948-582-9**

## Abstract

The circular economy is a strategy for reducing risks that result from the criticality of commodities in our economic system. Therefore, anticipatory planning of recycling infrastructure must consider the historical and future use of metals as well as detailed product characterizations. For bulk materials like plastics, industrial base metals (IBMs) such as steel, aluminum, copper, and precious metals (PM), e.g. gold, silver, palladium, well-established methodologies and a profound knowledge base exist that enable a detailed process modeling. Similar information is missing for a selected set of critical raw materials (S-CRMs), which are applied to a large extent in modern electrical and electronic equipment (EEE). Although, being highly important for the producing industry and being related to potential future supply risks, a functional recycling from waste electrical and electronic equipment (WEEE) is currently not practiced.

This doctoral thesis focuses on the provision of WEEE product-centric information with a special emphasis on S-CRMs via the consolidation and further development of available methods to support the derivation of recycling strategies. Research is needed explicitly for gallium in integrated circuits (ICs), indium applied as indium tin oxide (ITO) in liquid crystal displays (LCD), rare earth elements (REEs) used in NdFeB magnets applied in hard disk drives (HDDs), and tantalum in tantalum capacitors applied on printed circuit boards (PCBs).

Following a consecutive methodology, in a first step, S-CRMs will be identified and localized in WEEE flows via material flow analysis (MFA), and single WEEE products via recycling-oriented product characterization. In a second step, theoretically available mass flows of these S-CRMs and by-applied materials are quantified for EEE put-on-market (worldwide/Germany) and WEEE collected (Germany) in 2013. Recyclability of S-CRMs in selected WEEE applications is assessed via customized recycling processes. Data are then collated in a recycling barrier analysis investigating the framework for a successful recovery of S-CRMs.

WEEE flow results indicate a lower relevance of S-CRM recovery from WEEE, as significantly higher yields can be gained by reducing losses in the primary production of S-CRMs, in subsequent refining steps, and in the manufacture of semi-finished products. However, supply risks typically accompanying the primary production routes of S-CRMs indeed suggest considering S-CRMs recycling from WEEE. Current recycling strategies of WEEE are solely optimized for the recovery of bulk and valuable materials. Results on the general pre-processing of WEEE show that S-CRM recovery is not the core focus, which results in their dilution with other materials in mechanical treatment. A later S-CRM recovery from generated output fractions is hardly possible. Together with the loss of S-CRMs, other valuable materials, such as gold or copper, are lost within those output fractions as well, due to limited liberation and separation techniques in mechanical treatment.

Results from WEEE product analyses confirm the earlier indicated lower relevance of S-CRM recovery, as the mass fractions in the WEEE devices is rather low. However, selected liberated components contain high S-CRM mass fractions of up to 30-50% and are worth further end-of-life treatment. The typical design of WEEE devices often impedes a feasible recycling of the S-CRMs. Also, the technical identification of smaller components (e.g. tantalum capacitors and in particular ICs), which contain S-CRMs, is complex due to lacking well identifiable characteristics.

Chemical analyses prove heavily fluctuating mass fractions of S-CRMs in WEEE components. Furthermore, they have a limited selectivity, as with the removal of materials carrying S-CRMs, other elements are unintentionally removed. This applies to valuable materials such as gold, copper in ICs and silver in tantalum capacitors. Also, contaminating elements are affected such as nickel in NdFeB magnets or even toxic heavy metals such as arsenic, chromium, lead, and antimony in LCD panels.

Resulting from WEEE flow and WEEE product analyses, theoretically available mass flows of gallium from ICs, of indium from LCD panels, of REEs from NdFeB magnets in HDDs and of tantalum from tantalum capacitors are estimated. Results show that WEEE cannot solely cover the S-CRM demand to produce new similar goods currently occurring on the market. Furthermore, the subsequently performed recyclability assessments reveal that S-CRMs cannot be recycled with current practice. However, within this thesis individual approaches are presented, which enable the concentration of S-CRMs and other by-applied materials to provide them for appropriate end-processing steps. For instance, a thermal treatment of ICs, manual removal of tantalum capacitors and a chemical fractionation of LCD panels show promising recycling processes for liberation and separation of components and materials carrying S-CRMs.

In conclusion, the generated fundamental product-centric information for WEEE products allows for the development of recycling strategies and supports the holistic recovery of all materials applied in WEEE such as S-CRMs, PMs, IBMs, and other bulk materials. Thus, an extension with additional non-product-related information enables the further development of general political recycling strategies implemented e.g. in the European WEEE directive, national laws or in in-house business plans of recycling facilities. Also, technical and economic recycling strategies are supported, which are used for the technical set-up in recycling plants and the selection of materials to be recovered in context of processing costs and expected revenues.

## Acknowledgements

In the first place, I want to thank two very important women. The first one is Vera Susanne Rotter, who gave me the opportunity to work for her and doing my doctoral thesis with a lot of creative freedom and trust in my work. Sometimes, discussions were necessary to align our opinions, which almost always ended in much better results. Thank you, Susanne, for the intense and inspiring time at your chair. The second very important person did not less. Thank you, Sabi, for being there all the time and for not going crazy or getting mad at me for speaking so much about the work and the thesis and so on. You were always listening and cheered me up when necessary. This is just an acknowledgment in my doctoral thesis and cannot express, what I feel. You know, that I am not good with words, but I know you understand.

I have to thank Nathalie and Paul for their support and giving me host and years of fun in our shared office. The “Kleeblatt” was ripped apart, but this gave space for your indoor jungle. May the oxygen be with you. Call me, once the first monkey moved in.

Birgit and Claudia, thank you so much for your patience with me and all the samples I brought to you. This work wouldn't have been possible without you. I hope the next scientific assistants won't have such sophisticated materials and won't insist on measuring all possible elements.

I want to thank the whole chair of Circular Economy and Recycling Technology for being such a perfect place to work. This bunch of various types of people makes this chair an open-minded and welcoming place for everyone. This, of course, includes all the students, who were working for and with me and were feeding me with data.

Nathalie and Alex, thank you for the work you put in the review and the comments, which helped to improve this thesis a lot.

Finally, I want to thank Prof. Sabine Flamme and Prof. Helmut Antrekowitsch for being such enthusiastic, helpful and uncomplicated supervisors.

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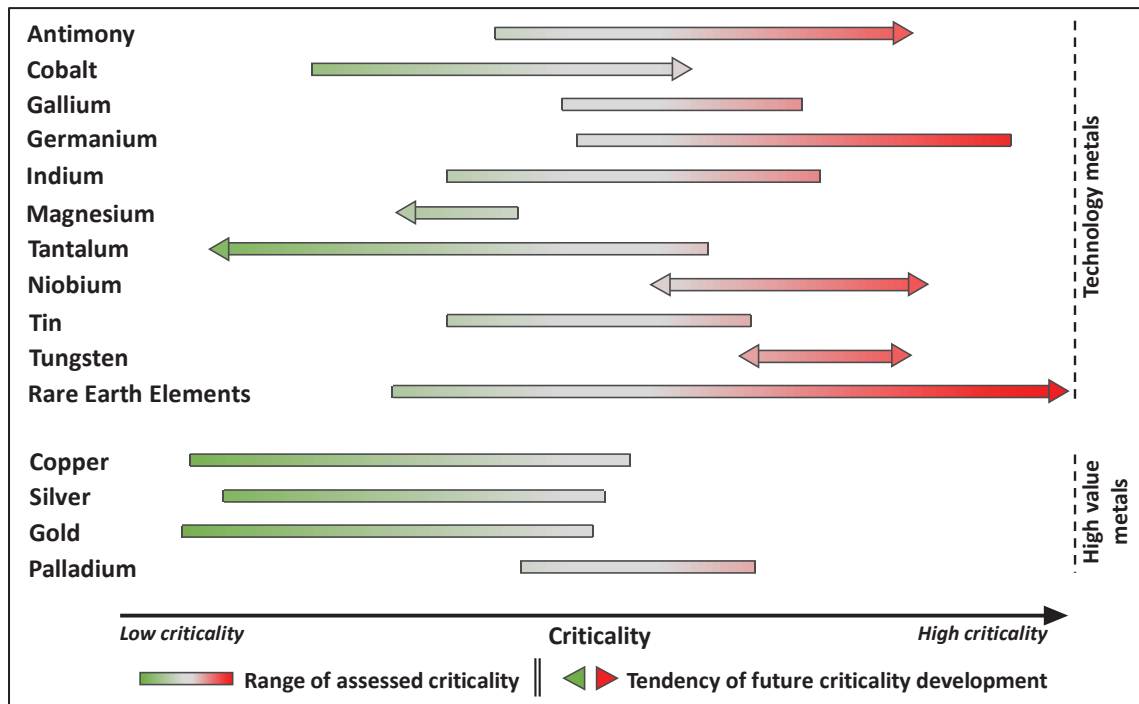
## Abbreviations

Abbreviation	Full name
AAS	Atomic Absorption Spectroscopy
AS	Automated Sorting
ATR-FTIR	Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy
C	Concentrate
CCFL	Cold Cathode Fluorescence Lamp
CIGS	Copper Indium Gallium (di)Selenide (solar cell)
CRM	Critical Raw Materials
CRT	Cathode Ray Tube
CW	Chip Production Waste
EEE	Electrical and Electronic Equipment
EOL	End-Of-Life
GaAs	Gallium arsenide
GaP	Gallium phosphide
GaN	Gallium nitride
GaSb	Gallium antimonide
HDD	Hard Disk Drive
IBM	Industrial Base Metals
IC	Integrated Circuit
ICP-AES, ICP-OES	Inductively Coupled Plasma – Atomic Emission Spectroscopy
ICP-MS	Inductively Coupled Plasma – Mass Spectroscopy
IR	Infrared (optics)
IT	Information and Telecommunication
ITO	Indium Tin Oxide
LCD	Liquid Crystal Display
LED	Light Emitting Diode
LOD	Limit of detection
LOI	Loss Of Ignition
MFA	Material Flow Analysis
MS	Manual Sorting
NdFeB	Neodymium-Iron-Boron
NWEE	Not Collected WEEE
PC	Personal Computer
PCB	Printed Circuit Boards
PG	Primary Gallium
PGM	Platinum Group Metals
PM	Precious Metals
POM	Put-On-Market
PW	Primary Waste
R	Recyclate
REE	Rare Earth Elements
RefW	Refining Waste
Res	Primary Resource Potential
RG	Refined Gallium
RW	Recycling Waste
SC	Separate Collection
S-CRM	Selected Critical Raw Materials
SFA	Substance Flow Analysis
SG	Secondary Gallium
S-OES, Spark-OES	Spark Optical Emission Spectroscopy
TV	Television
VIC	Visually Identifiable Capacitor
W	Wafer
WEEE	Waste Electrical and Electronic Equipment
WLAN	Wireless Local Area Network
WW	Wafer Production Waste
XRF	X-Ray Fluorescence

## 1. Introduction and objectives

Current and future innovations in the information, telecommunication, and entertainment sectors are reliant on technology metals<sup>1</sup> such as gallium, indium, tantalum and rare earth elements (REE). Increasing sales of electrical and electronic equipment (EEE) and the increasing use of such technology metals in them deteriorate the future supply reliability. Although representing an appropriate source for secondary raw materials, technology metals are usually not recovered from waste electrical and electronic equipment (WEEE), due to economic and technical recycling<sup>2</sup> barriers. In fact, functional recycling<sup>3</sup> rates were found to be under 1% (UNEP - International Resource Panel, 2011). A number of institutions assessed the supply situation and the substitutability of various elements, which are most important for the producing industry of countries lacking of their own natural resources (European Commission, 2010; European Commission, 2014a; Hatayama and Tahara, 2015; Graedel and Reck, 2016; Erdmann and Behrendt, 2011).

Results show a significant criticality for technology metals such as antimony, gallium, germanium, cobalt, indium, niobium, tantalum, REEs and even high-value materials<sup>4</sup> like platinum group metals (PGM) leading to the term “critical raw materials<sup>5</sup>” (CRMs). Figure 1 shows a summary of the results of relevant studies.



Own illustration based on: (European Commission, 2010; European Commission, 2014a; U.S. Department of Energy, 2011; Hatayama and Tahara, 2015; Erdmann and Behrendt, 2011)

Figure 1: Qualitative range of assessed criticality for technology and high-value metals from various sources including a tendency of future criticality development

<sup>1</sup> *Technology metal*: generally-rare metals that are essential for the production of 'high tech' devices and engineered systems (Prinsloo, 2016). See glossary for further explanation.

<sup>2</sup> *Recycling*: “Recirculation of production and consumption waste (also: waste heat) into the economic cycle” (Springer Gabler Wirtschaftslexikon, 2017)

<sup>3</sup> *Functional recycling* is recycling in which the physical and chemical properties that made the material desirable in the first place are retained for subsequent use (Reuter et al., 2011). See glossary for further explanation.

<sup>4</sup> “*Material* serves as an umbrella term for both substances and goods” (Brunner and Rechberger, 2004). See glossary for further explanations.

<sup>5</sup> *Critical raw materials*: raw materials are labeled “critical”, when the risks of supply shortage and their impacts on the economy are higher compared with most of the other raw materials. (European Commission, 2010)

Some of the institutions repeat their assessments frequently so that a time trend of criticality can be derived. Due to different perspectives of the researchers, such as local raw material availability or present producing industry, these trends are partially contrary. However, a possible trend to a higher criticality is noticeable.

Definitions and terms for material groups differ in these criticality studies and generally in literature. Therefore, in this thesis following terms will be used: “industrial base metals<sup>1</sup> (IBMs)” for aluminum, iron, copper, etc.; “precious metals (PMs)” for example gold, silver and palladium; and a selected “set of critical metals (S-CRMs<sup>2</sup>)” for example gallium, indium, rare earth elements (REEs), tantalum, cobalt, etc.). (Ueberschaar et al., 2017b)

In the context of criticality of materials, the circular economy can be seen as an imperative to reduce related risks affecting the criticality of commodities in our economic system. However, any effort in a priori planning of new infrastructure for recycling has to be based on a good understanding of historical and future use of applied materials in EEE and detailed WEEE products information. For bulk materials like plastics, steel, aluminum, copper, and PMs, such as gold, silver, palladium, well-established methods and a deep knowledge base exist that enables detailed process<sup>3</sup> modeling (Rotter et al., 2013). Similar information for S-CRMs still has to be gained, and recycling strategies<sup>4</sup> have to be developed. Recycling strategies address three interdependent dimensions: 1) *political recycling strategies* that focus on a strategic resource handling via e.g. further development of the European WEEE directive and national laws or e.g. business plans of single recycling facilities; 2) *technical recycling strategies*, which describe the individual technical set-up for the recovery<sup>5</sup> of selected target materials based on available techniques; c) *economic recycling strategies* defined by the framework conditions based on revenues for processed and generated materials in context to all costs.

Recycling strategies for S-CRMs in WEEE should consider the recovery of any by-applied bulk and valuable materials. Losses can lower the purity of output materials and the economic and environmental yield (UNEP, 2013). However, due to highly varying mass fractions and total loads, and the sophisticated application of liberation<sup>6</sup> and separation<sup>7</sup> processes on complex components<sup>8</sup> and WEEE products, the recycling of all applied materials is either technically not possible or not reasonable from an economic or ecological perspective. Figure 2 shows example recycling potentials from a theoretical, technical, ecological and economic perspective.

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<sup>1</sup> *Industrial base metal*: a common metal in the producing industry that is not considered precious, such as aluminum, copper, iron, tin, or zinc. (Oxford Dictionaries, 2017)

<sup>2</sup> *S-CRM*: a reduced set of those metals rated as critical by the European Commission (European Commission, 2014b; European Commission, 2010). This set excludes materials, which are in focus of current recycling strategies (i.e. precious metals).

<sup>3</sup> “A *process* is defined as the transformation, transport, or storage of materials.” (Brunner and Rechberger, 2004)

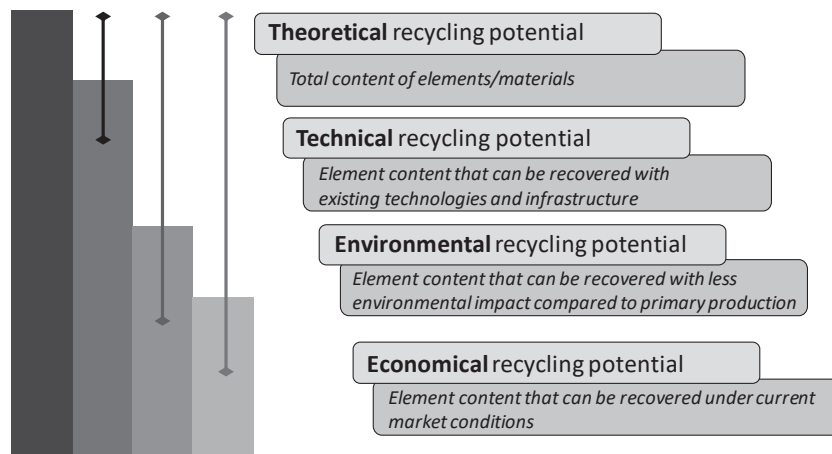
<sup>4</sup> The *recycling strategy* describes the goals of recycling efforts (political, economic) and the related selection of recycling processes or the recycling chain (technical).

<sup>5</sup> “(Resource) *Recovery* is the selective extraction of disposed materials for a specific next use, such as recycling, composting or energy generation in order to extract the maximum benefits from products, delay the consumption of virgin resources, and reduce the amount of waste generated.” (Grand Traverse County, 2017)

<sup>6</sup> *Liberation* refers to all manual, mechanical, chemical or physical measures to strip down unwanted materials and obtain the single target appliance, material or substance.

<sup>7</sup> *Separation* refers to any categorization process (see sorting in glossary), but with a focus on target materials. The separation follows typically after the liberation.

<sup>8</sup> *Component*: an essential part of an appliance providing major functions



adapted from (Rotter et al., 2016)

Figure 2: Perspectives for the use of recycling potentials

Thus, the physical and chemical properties of WEEE products limit the possible recycling depth. Product-centric information represents the key element for the development of recycling strategies. However, adequate S-CRM data is missing (Chancerel et al., 2013) and general guidelines or common methodologies for their generation rarely exist for WEEE products. Therefore, the objective of this doctoral thesis is the provision of WEEE product-centric information with a special emphasis on S-CRMs via the consolidation and further development of available methods to support the derivation of recycling strategies.

## 2. Background

### 2.1. S-CRMs in WEEE

S-CRMs are versatily applied in EEE. However, information about recycling potentials and strategies are not covered sufficiently in current research. Fundamental data gaps exist for the chemical composition with a focus on metal contents in relevant WEEE products and components, and for batch analyses of critical metals in complex equipment mixes in WEEE processing (Chancerel et al., 2013). Table 1 shows an overview of applied S-CRMs in WEEE and the related purpose of the application. Actual data inaccuracies highlight the need for research.

*Table 1: S-CRM use in WEEE and accuracy of available data*

Metal	Application	Accuracy of the data already available
<b>Antimony</b>	Synergist for flame retardants	Indicative values based on measurements available for casings and PCBs
	Package of electronic components	Indicative values based on estimations and standards
	ATO transparent conductive layer	No data
	Glass of CRT	Indicative values based on measurements
<b>Cobalt</b>	Cathode of lithium-ion batteries	Indicative values based on estimations, measurements, and producer information
	Metal hydride alloy of NiMH batteries	Indicative values based on estimations and measurements
	Cathode of nickel–cadmium batteries	Indicative values based on estimations
	Magnetic material	Indicative values based on industry standards specifications
	Electrical contact	Indicative values based on estimations and measurements of PCBs
	Active part of electronic components	Indicative values based on estimations and measurements of PCBs
	Glass of CRT	Indicative values based on measurements
<b>Gallium</b>	Semiconductor material in integrated circuits	No data on chips, indicative values based on measurements of PCBs
	Semiconductor material in chips for white LEDs	Indicative values based on measurements and estimations
	Semiconductor material in laser diodes	No data
<b>Germanium</b>	Semiconductor material in integrated circuits	No data
	Phosphor	No data
<b>Indium</b>	ITO transparent conductive layer	Detailed data based on measurement for different equipment types
	Semiconductor material in chips for white LEDs	Indicative values based on measurements and estimations
	Solder material	No data
<b>Rare earth elements</b>	Phosphor in fluorescent lamps	Indicative values predominantly based on estimations
	Phosphor in LEDs	Indicative values predominantly based on estimations
	Magnetic material	Values based on measurements and standards
	Anode and cathode of NiMH batteries	Indicative values based on rough estimations
	Material in electronic components	No data
<b>Tantalum</b>	Capacitor anode	Indicative values based on measurements and outdated standards
	Material in electronic components	No data

Based on: (Chancerel et al., 2013)

Note: ATO: antimony–tin–oxide; CRT: cathode ray tube; PCBs: printed circuit boards; NiMH: nickel–metal hydride; LED: light-emitting diode; ITO: indium–tin–oxide.

According to this overview, knowledge gaps exist inter alia for gallium in integrated circuits<sup>1</sup> (ICs) on printed circuit boards (PCBs), indium in liquid crystal displays (LCDs), REEs in neodymium iron boron (NdFeB) magnets from hard disk drives (HDD) and tantalum in tantalum capacitors on PCB.

#### 2.1.1. Integrated circuits

Gallium arsenide (GaAs), Gallium phosphide (GaP), Gallium nitride (GaN), and Gallium antimonide (GaSb) represent by far the most applied gallium semiconductor compounds (Roskill, 2012). Almost 99 % of refined gallium is applied in the form of GaAs and GaN (Jaskula, 2014).

The main end-use-sector of gallium is in EEE sector being approx. 90 % of the total production. In the EEE sector, almost 68 % of the applied gallium is used for integrated circuits (ICs), which are required for high-frequency wireless communication (e.g. mobile phones – 4G, 3G, 2G, etc. or WLAN). The remaining material is mostly used for optoelectronic components such as light emitting diodes (LEDs) or laser diodes for background lighting in computers, TV, mobile phones or photodetectors and solar panels (Jaskula, 2014). An overview of the Ga consumption worldwide in 2012 is shown in Table 2.

Table 2: Share and amount of the worldwide Ga-consumption per end-use-sector

End-use-sector	Share in 2014	Consumption in 2012
	[%]	[t]
Integrated circuit and field-effect transistors	50	177
LED for background and general lighting applications	38	135
Photovoltaics	4	14
Other	8	28

Based on: (Roskill, 2012)

© (Ueberschaar et al., 2017c)

#### 2.1.2. LCD panels

Up until 2025, the yearly demand for indium will grow by 70% relative to the year 2015 (Licht et al., 2015). In 2011, 1,220 Mg indium was primarily mined worldwide, from which 660 Mg indium was refined (Licht et al., 2015). In recent years, the output of refined indium increased to 800 Mg in 2013 and 820 Mg in 2014 (U.S. Geological Survey, 2015). Over 55% of the worldwide production of indium is used in the ITO (indium tin oxide) target industry, which is a major component in the manufacture of liquid crystal displays (LCD) (Licht et al., 2015; Li et al., 2011; Nakashima and Kumahara, 2002; U.S. Geological Survey, 2015) (cf. Figure 3). For further information about the use of indium see the supporting information S1 in (Ueberschaar et al., 2017d).

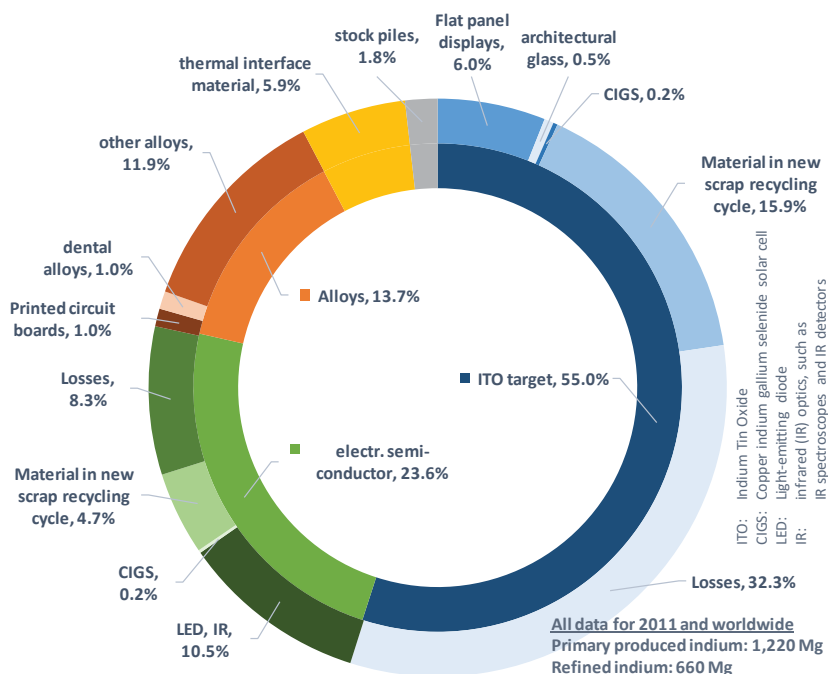
End-of-life<sup>2</sup> (EOL) LCDs account for up to 90% of the indium-bearing components in WEEE. This concentration<sup>3</sup> in a particular set of WEEE devices could provide a basis for successful recycling (Swain et al., 2016; Chou and Huang, 2009; Wang, 2011). Although a number of studies have been published, a full recycling of LCD panels has not been developed (Zhan and Xu, 2014; Wang et al., 2015; Sun et al.,

<sup>1</sup> "An integrated circuit (IC) is a small semiconductor-based electronic device consisting of fabricated transistors, resistors and capacitors. Integrated circuits are the building blocks of most electronic devices and equipment." (Techopedia, 2017)

<sup>2</sup> End-of-life (EOL) of a product is a term with different meanings dependent from the user's profession. From a vendor's perspective, EOL indicates the end of the planned lifetime of a product purchased by a customer or the planned end of support of a product, which results in the stop of marketing, selling, or rework sustaining the product. From a customer's perspective, EOL of a product stands for the end of wanted use of an owned product, due to outdated technics or loss of its functionality.

<sup>3</sup> Concentration: "In chemistry, concentration is the abundance of a constituent divided by the total volume of a mixture. The term concentration can be applied to any kind of chemical mixture, but most frequently it refers to solutes and solvents in solutions" (IUPAC, 2014)

2017; Ciacci et al., 2015; Zeng et al., 2015; Rasenack and Goldmann, 2014; Rocchetti et al., 2014; Silveira et al., 2015; Virolainen et al., 2011; Fontana et al., 2015; Lee et al., 2013; Kato et al., 2013; He et al., 2014; Savvilotidou et al., 2014; fem Forschungsinstitut Edelmetalle und Metallchemie and Institut für Energie- und Umwelttechnik e.V., 2011; Götze and Rotter, 2012; Zhang et al., 2015; Zhang and Xu, 2016) (cf. supporting information S1, S2, and S3 in (Ueberschaar et al., 2017d)). Hence, recovery of indium from WEEE is not yet being carried out (Chancerel et al., 2013; European Commission, 2010).



Own illustration based on: (Licht et al., 2015)

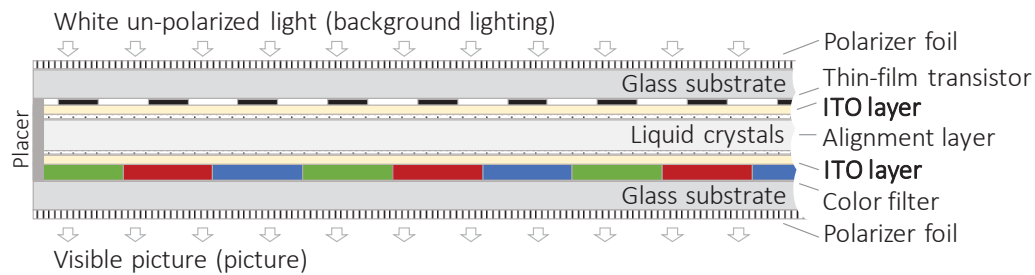
Figure 3: Overview of application of refined indium worldwide in 2011

Based on the screen size and the primary function of screen devices, the shares of applied LCD panels vary. Own disassembly operations revealed mass fractions<sup>1</sup> of LCD panels in notebooks of 9.5% (n=8), 8.5% for mobile phones (n=38), 19% for tablets (n=16) and 14.7 % for smartphones (n=26). Other studies show different values. EMPA & SWICO (2011) and Westerdahl et al. (2011) state a 19% LCD panel share in notebooks. Furthermore, Westerdahl et al. (2011) determined 19% for TV sets. In contrast, (EMPA & SWICO (2011)) show only 6% for TVs and 9% for PC monitors. The varying shares are most probably related to different levels of disassembly. The LCD panels are usually assembled in frames, including the background lighting and electronic components like PCB / electronics. For tablets and smartphones (devices with touch functionality), the LCD panels are partially bonded with surface mounted glass, which cannot be separated completely. Hence, the category “LCD display” or “LCD panel” can be interpreted in different ways.

Although the shares of LCD panels vary, in the screen devices investigated the structure and design of the panels are standardized. Each panel consists of several layers in a sandwich structure. Two glass panes made of sodium- or boron-silicate glass form the basis. The inner sides are populated with color pigments, thin film transistors, and the conductive indium tin oxide (ITO) layer. (Jalalpoor et al., 2013)

<sup>1</sup> Mass fraction: “The mass fraction is the ratio of one substance’s mass to the mass of the total mixture” (IUPAC, 2014)

Figure 4 shows the sandwich design of an LCD panel in profile.



Own illustration (Ueberschaar et al., 2017d)

Figure 4: Design of LCD panels with ITO layers and polarization foils in profile

© (Ueberschaar et al., 2017d)

### 2.1.3. Hard disk drives

REEs in HDDs are applied in small amounts in PCBs and NdFeB (neodymium iron boron) magnets located in the spindle and the linear motor (voice-coil actuator) (cf. Figure 5).



Own illustration (Ueberschaar and Rotter, 2015)

Figure 5: Location PCB (l.) and NdFeB magnets (r.) in linear (below) motor and spindle motor (midst)

Depending on required physical properties, NdFeB magnets contain various REEs and other alloying elements. The base alloy for NdFeB magnets is  $\text{Nd}_2\text{Fe}_{14}\text{B}$  (Zepf, 2013). Specific additions of elements are used to adjust the material properties (Zepf, 2013; Goldman, 1999). Cobalt substitutes REEs and Fe materials up to over 5% to increase the Curie temperature, which describes the point at which the magnetic field is offset and permanently destroyed (Rotter et al., 2013; Hatch, 2011). Dysprosium increases the temperature characteristics of the magnet as the compound has a better stability against demagnetization. This does not mean a significant increase in the Curie temperature. Moreover, adding dysprosium decreases the residual induction of the magnet, which leads to lower magnetic field properties (Hatch, 2011; Goldman, 1999).

Praseodymium was used to lower production costs as it can substitute neodymium up to 20-25%. This changed in recent years as quoted trade prices for praseodymium are even higher by now. (MetaErden GmbH, 2014a; MetaErden GmbH, 2014b)

NdFeB magnets in voice-coil actuators are sintered products for which powdered material in a mold is heated to temperatures below the melting point. At the same time, an external magnetic field is applied. The end product is a solid piece with desired dimensions (Kim et al., 2013). In this way manufactured magnets have the best results regarding the magnetic properties.

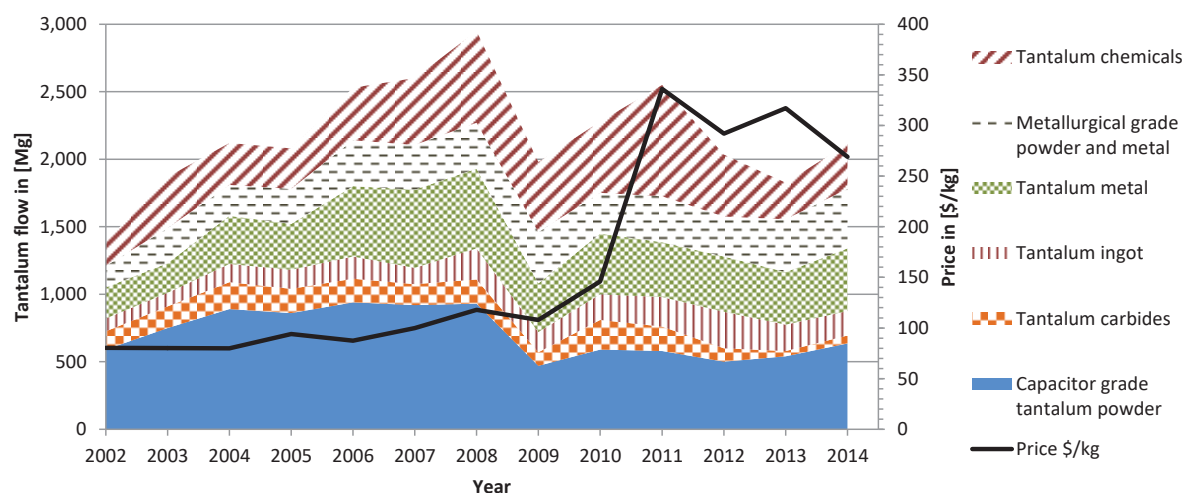
Other manufacturing methods are polymer-bonding and compression-molding, which are used for the production of spindle motor magnets. Compression-molding is similar to sintering processes, but with this method, the powder is heated to over the melting point, and the material is consolidated with high pressure. Polymer-bonded magnets are made by mixing a special binder to the NdFeB powder. This blend can be pressed to a precise desired shape. With this approach, no or only a little post-processing is necessary. Magnets produced with this technique are highly suitable for the spindle motor. The typical magnetic properties of these materials are lower compared to sintered magnets (Hatch, 2011).

The magnet material, mainly the REE materials are highly oxidizing in contact with air. Because of this, the permanent magnets are coated with a protective layer. This can be thin polymeric coatings, but mainly sacrificial anodes made of nickel or tin/zinc are used. Sacrificial anodes as coatings are not applied for bonded magnets in the spindle motor.

☛ (Ueberschaar and Rotter, 2015)

#### 2.1.4. Tantalum capacitors

Tantalum is used for various applications. Figure 6 shows the global shipments for tantalum as raw material.



Own illustration (Ueberschaar et al., 2017a), based on: (Schwela, 2010; Buckingham et al., 2014; Tantalum-Niobium International Study Center, 2013a; Tantalum-Niobium International Study Center, 2013b; Tantalum-Niobium International Study Center, 2011; Tantalum-Niobium International Study Center, 2016; U.S. Geological Survey, 2016a; infoMine, 2016), Price conversion based on: (U.S. Geological Survey, 2016b)

The indicated price applies for the tantalum content in the raw materials only. Current price from 2015: 236,91 \$/kg (U.S. Geological Survey, 2016a; U.S. Geological Survey, 2016b; infoMine, 2016)

Figure 6: Tantalum shipments and price development between 2002 and 2012

Tantalum capacitors represent the major application for tantalum in the EEE sector. They consist of a fireproof and mostly inert epoxy resin cover containing  $\text{SiO}_2$  and a tantalum-rich anode. This anode is based on sintered tantalum with tantalum oxide covering the surface and is connected with a tantalum wire to the cathode material through silver connectors (see Figure 7). The porous structure of the sintered tantalum anode is impregnated with a  $\text{MnO}_2$  electrolyte and surrounded by a graphite layer. In addition to ca. 48% - 49% tantalum, the capacitors contain carbon, silicon, manganese, and silver in large quantities. (Rotter et al., 2013; Spitzczok von Brisinski et al., 2014)

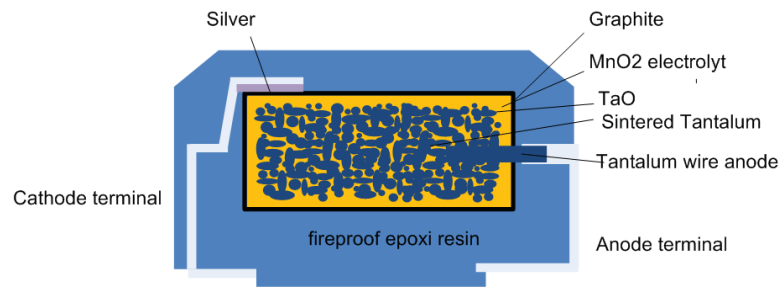


Figure 7: Design of a typical tantalum capacitor (Chancerel et al., 2013)

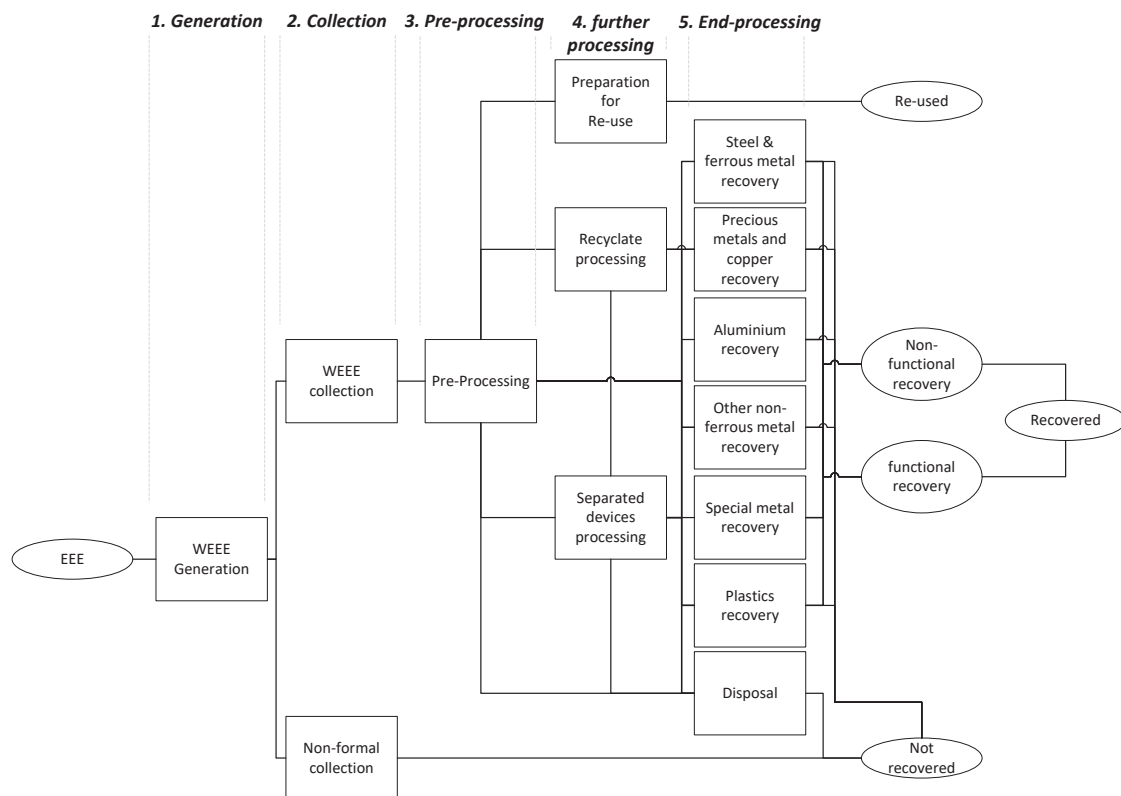
© (Ueberschaar et al., 2017a)

## 2.2. Recycling of S-CRMs

### 2.2.1. Status-quo of WEEE processing

The recycling chain<sup>1</sup> for WEEE consists of various subsequent steps: 1. generation of WEEE, 2. collection, 3. pre-processing, 4. further processing of treated material, and 5. end-processing. Usually, for each step specialized facilities exist. The recycling efficiency<sup>2</sup> (see 2.3.2 “Recycling efficiency”) of the entire recycling chain depends on the efficiency of each step and on how well the interfaces between these interdependent steps are managed (Schluep et al., 2009). With the end of the recycling chain, WEEE products are re-used, processed materials either functionally or non-functionally recovered, or not recovered. Figure 8 shows a simplified overview of the WEEE recycling chain.

Pre-processing represents one of the central steps in the recycling chain of WEEE as well as of other complex appliances. Via liberation and separation, materials are channeled into designated recycling processes<sup>3</sup>. Generated outputs have to be either processed further or can be used directly in end-processing steps.



Own illustration

Figure 8: Simplified recycling chain from WEEE generation to recovered material

The recycling of WEEE faces various political, technical and economic limitations. To fulfill the regionally mandatory recycling quotas<sup>4</sup>, current technical recycling strategies target IBM like copper, iron, and aluminum, and also plastics, as bulk materials. Those recycling quotas influence the recycling, due to tradeoffs between the recovery rates and recovery grades. Commercial recycling systems cannot create completely pure recyclate streams (van Schaik and Reuter, 2004a; van Schaik and Reuter, 2004b)

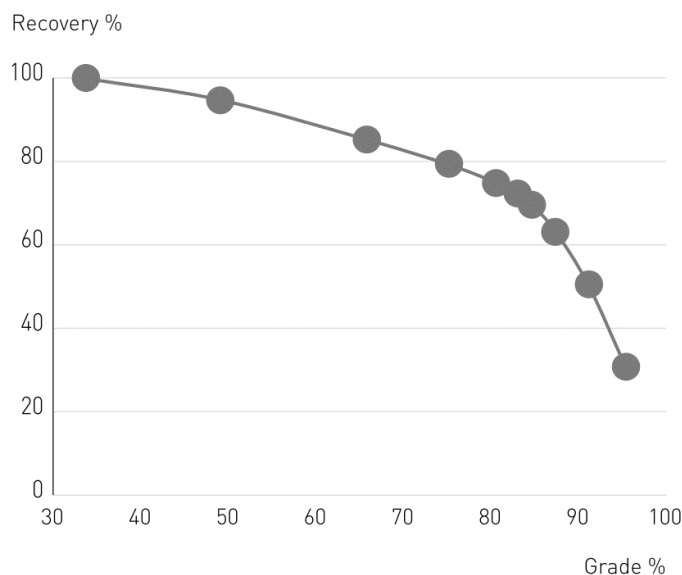
<sup>1</sup> *Recycling chain*: the combination of recycling processes enabling the recycling of material streams or appliances.

<sup>2</sup> *Recycling efficiency*: the efficiency of all related processes in the recycling chain for the recycling of a target substance.

<sup>3</sup> *Recycling process*: single process, which is used for the processing (transformation) of a waste stream. For the recycling of waste, a combination of recycling processes, the so-called recycling chain, is typically necessary.

<sup>4</sup> *Recycling quota*: the share of recycled substances based on the recycling efficiency.

and never achieve a 100% material recovery. Figure 9 shows how much material can be recovered, depending on the required grade (purity) of the output. With higher recovery rates, the grade of the target materials decreases in the output. (UNEP, 2013)



(UNEP, 2013)

Figure 9: Maximum grade-recovery curve

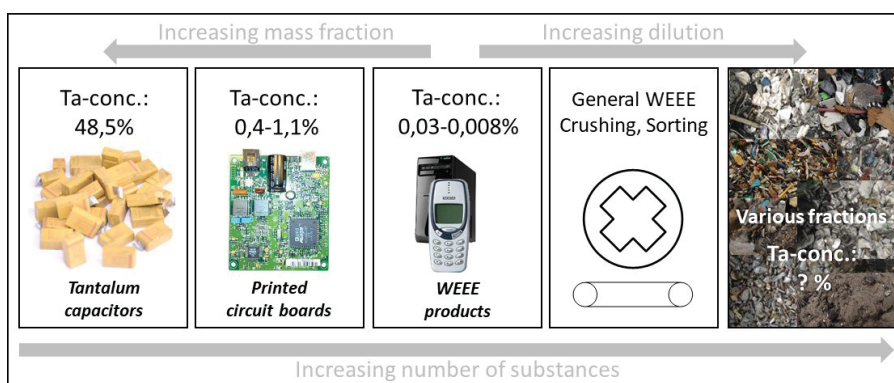
Further recycling limitations base on the WEEE product properties. The design of WEEE materials and appliances does often not comply with physical limits in manual, physical and chemical processing (Schaik and Reuter, 2013; Reuter and van Schaik, 2015) and results in lower recovery rates and the need for more complex recycling processes.

However, although being trace materials, materials with a high economic value, such as precious metals (gold, silver, palladium), are of high interest from an economic perspective. A recovery of such materials takes place in end-processing steps, which are usually limited to a particular set of materials. For example, integrated smelters are capable of recovering copper, precious metals, and some additional elements. However, other materials and elements carried in the same material stream are diluted to the slag and irretrievably lost. Undesignated materials may remain in the target product and are non-functional recycled as they are not usable for their original purpose anymore. Because of this, the transfer of non-target metals into recyclates for end-processing leads to a loss of these materials for a functional recycling, higher energy demands and lower recovery rates due to impurities and potential end-products of lower quality (UNEP, 2013). This applies in particular to most of the metals defined as S-CRM and whose recovery is discussed on the political agenda.

© (Ueberschaar et al., 2017b)

### 2.2.2. General recycling quotas for S-CRMs

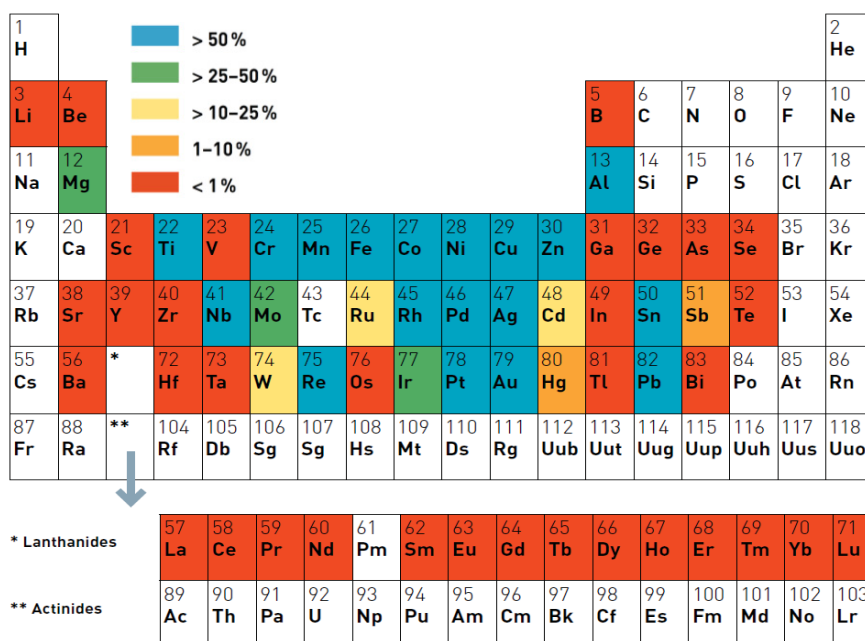
In the first instance, WEEE processing usually consists of comminution and sorting<sup>1</sup> of categorized material streams. Categories comply with bulk and generally valuable materials. S-CRMs are not targeted in recycling and are by-processed only. Through this, S-CRMs mechanically processed are usually diluted with other materials to low mass fractions, which are partially under detectable limits (Ueberschaar et al., 2017b). Furthermore, the number of by-processed elements increase. Figure 10 shows the possibilities of concentrating or diluting depending on the way of treatment of WEEE devices on the example of tantalum capacitors applied on PCBs in WEEE appliances. A separation of tantalum capacitors is not possible with current automated systems. Only a manual separation provides a pure output fraction.



Own illustration

Figure 10: Concentration and dilution related to S-CRM carrying appliances in the case of tantalum capacitors

However, mechanical processing represents the standard in pre-processing of WEEE. A subsequent recovery of S-CRMs is related to high costs, high energy demands and low recovery rates (UNEP, 2013). UNEP - International Resource Panel (2011) states current functional recovery rates of under 1% for most of the materials classified as S-CRM (see Figure 11).



(UNEP - International Resource Panel, 2011)

Figure 11: EOL functional recycling rates of various investigated elements

<sup>1</sup> Sorting is any process of arranging items systematically. See glossary for further explanation.

### 2.2.3. Status of gallium, indium, tantalum and REE recovery

#### Gallium in ICs

The functional end-of-life recycling rate of gallium is under 1 % (UNEP - International Resource Panel, 2011). The primary reason is the application of this element only in minor mass fractions in relevant equipment types. First recycling approaches for single components like LEDs are presented by Swain et al., 2015 or Rotter et al., 2016b. However, in general recycling processes, gallium is usually further diluted and subsequently lost. Due to this, gallium from WEEE is classified as not recyclable (Ciacci et al., 2015).

Currently, a recovery is carried out only for material streams with high gallium mass fractions. These concentrates arise only in the processing of semi-finished products in the form of processing residues, mainly coming from GaAs wafer production. About 45 % of the gallium input in the production originates from this new scrap. (U.S. Geological Survey, 2015)

☞ (Ueberschaar et al., 2017c)

#### Indium in LCD panels

Practical recycling of indium is conducted in the production of semi-finished and intermediate products only (see supporting information S2 in (Ueberschaar et al., 2017d)). Recycling of indium from EOL LCD panels is not yet carried out on an industrial scale (Chancerel et al., 2013; Chancerel et al., 2015; European Commission, 2010). At least, much research has been recently carried out on the topic of indium recovery from LCD panels. Various approaches have been tested with the main focus on indium recovery. Appendix 1 gives an overview of the most recently discussed studies.

The first step for most indium recovery approaches is the transfer of the ITO into a liquid state. The main parameters are the preparation of the LCD panels, a particular concentration of the leachate used, temperature, reaction time and repetition of the chosen process. The primary leachates used are sulfuric acid, nitric acid, and hydrochloric acid while the chosen temperature is between 80-90° C. With this approach, up to 99% of the indium can be extracted. (Rocchetti et al., 2015; Silveira et al., 2015; Rasenack and Goldmann, 2014; Virolainen et al., 2011; Zeng et al., 2015)

Most of the approaches tested use a high temperature for a better and more efficient reaction. To reduce the energy input, the leaching process can also be executed at room temperature. Here, a specific sample preparation such as the crushing of the LCD panels plays an important role. Further concentration steps via hydrometallurgical procedures such as solvent extraction purify the indium output and separate undesigned substances<sup>1</sup>.

Recovery of applied polarizer foils and the glass substrate is only occasionally investigated. Most of the studies state that a separation of foils took place but was executed manually. The reason behind this was the fear that the plastics might disturb further processes. However, a focused recovery of polarizer foils and glass substrate has not been broadly investigated in detail nor have any successful approaches been implemented in current recycling strategies. FEM (2011), Götze and Rotter (2012), Zhang et al. (2015) and Zhang and Xu (2016) summarized recently investigated strategies for a recycling of LCD panels (Götze and Rotter, 2012; Zhang and Xu, 2016; fem Forschungsinstitut Edelmetalle und Metallchemie and Institut für Energie- und Umwelttechnik e.V., 2011; Zhang et al., 2015).

☞ (Ueberschaar et al., 2017d)

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<sup>1</sup> "A *substance* is any (chemical) element or compound composed of uniform units. All substances are characterized by a unique and identical constitution and are thus homogeneous." (Brunner and Rechberger, 2004)

### REEs in HDD

Regarding the recovery of REEs in HDDs respectively NdFeB magnets, the major problem is the liberation of the NdFeB magnets. All approaches showing successful recycling in literature are based on the processing of already liberated magnet material. In practice, a major part of collected HDD is shredded due to data protection reasons and as it is the most economical way of treatment to separate the bulk materials with post-shredder technologies. This procedure dilutes minor metals<sup>1</sup> into the main output streams. Magnetic materials will be transferred to the ferromagnetic fraction or dust fraction (Westphal, 2015; Habib et al., 2015). This leads to a complete loss of REE materials. Manual disassembly is one option to face this problem. However, still low prices for REE scrap lead to non-economical processes. Currently, only social-working companies are able to perform this commissioned work for lower prices.

One additional issue beyond liberation is the availability of recovery and refining options. Currently, most approaches are still in development stage. Largely research studies deal with the recovery REEs in NdFeB magnets. Appendix 1 shows a structured overview of given options for the functional respectively elemental recovery of REEs based on the input of already liberated, single magnets.

Methods for re-use or functional recycling of magnet materials are most energy efficient and less technology-intensive approaches. Furthermore, no loss of material is expected. As a direct re-use of magnets limits the fields of application and the general application of magnets changes regarding electric mobility and wind power stations, a functional recycling with re-sintering processes is most expedient. Adding 1% of Neodymium to each re-sintering step, newly manufactured magnets maintain their physical properties and reach the same magnetic field strengths (Zakotnik et al., 2008a).

With a few exceptions like pyrometallurgical processes or gas phase extraction, the presented methods recover only REE concentrates. To obtain high purity mixed REE materials or single REEs for direct application in the production of new electrical and electronic appliances or components, the concentrates have to be subsequently processed. Adapted from the production of primary resources these processes are mainly hydrometallurgical methods. Therefore, a direct utilization of hydrometallurgical approaches for the recovery of REEs in NdFeB magnets of HDD is reasonable. Developed by Elwert et al. and tested under laboratory conditions, a high-purity recovery of other applied materials like iron and cobalt is possible (Elwert and Goldmann, 2013).

All mentioned approaches are based on the input of liberated NdFeB magnets. Feasible approaches for the treatment of whole HDDs filling the gap to apply developed recycling processes have rarely been developed. The most expedient way is the manual dismantling. Hitachi presents another possibility to separate complete magnets. This new invention works with a spinning drum-type unit which shakes and swirl the HDDs so that the inner connections of the devices come loose. The end product of this method are the structural components of the HDDs (Nemoto et al., 2011). One problem is here, that the magnets are still connected to the carrying steel plates (cf. Figure 47).

Regarding this, technical approaches for an economical preparation of the HDDs must be developed to apply the presented recovery methodologies.

© (Ueberschaar and Rotter, 2015)

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<sup>1</sup> *Minor metals* is a widely used term in the metal industry that generally refers to not common, primary metals not traded on formal exchanges, such as the London Metal Exchange (LME). (minor metals trade association, 2017)

### Tantalum in tantalum capacitors

Recycling options have been established only for new tantalum capacitor scrap and concentrated slags (H.C. Starck, 2011), with recycling rates claimed to be approximately 20-30% (Schwela, 2010). Recycling of tantalum from WEEE is not practiced yet, but first approaches were investigated and tested. The overall recycling procedure for tantalum capacitors consists of three main steps after the collection and liberation of the PCBs: Firstly, the identification, liberation, and separation of the tantalum capacitors. Secondly, mechanical processing to remove contaminants like polymers, etc. Third and final step: metallurgical processing in order to generate a tantalum concentrate. This material may be subsequently processed so that it can be used as a secondary raw material in new products (Cymorek, 2015; Stenzel, 2016).

#### *Identification, liberation, and separation of tantalum capacitors*

The liberation and separation of tantalum capacitors are usually done by manually removing the visibly identifiable components. This requires significant labor input and is currently carried out only by cross-funded sheltered workshops for test purposes (Rotter et al., 2016) since high labor costs make this approach non-cost-effective.

Various approaches for mechanical processing have been developed with a view to automating the process. Some of these approaches involve crushing the PCBs and the separation of the capacitors through sieving and classification by mass and magnetism (OKI, 2013; Kwon et al., 2016). Other approaches involve liberation via underwater explosion, and these show promising results (Fujita et al., 2014). The tantalum capacitors are then sorted with an eddy-current and a gravity separator.

Hayashi and Oki, 2014 have investigated the separation of the different compounds after their detachment from the PCBs and have invented a pneumatic technique that enables high separation efficiencies for tantalum capacitors.

Even robotic approaches are being tested at some research institutions, for some of the more common standardized PCBs (Kreibe and Förster, 2016).

#### *Recycling and tantalum recovery from WEEE capacitor scrap*

Most established recycling processes for tantalum capacitors apply acid leaching to separate tantalum from all other parts like electrolyte or coating material. Specifically, the  $\text{MnO}_2$  content is transferred into water-soluble  $\text{MnCl}_2$  using an HCl treatment (Wang et al., 2009; Smokovich and Hafner, 2009). The silver content is thereby recovered from the waste slag.

Mineta and Okabe, 2005 have developed a recycling process by adapting the processes of primary production. Here, complete oxidation of the capacitor scrap is followed by a magnetic separation of iron and nickel containing materials. After the addition of water, the process involves two sieving steps and a pulverizing process. Leaching agent  $\text{HNO}_3$  is then used to remove all compounds except tantalum and tantalum oxides. A second approach was developed recently by Spitzok von Brisinski et al., 2014. In this process, an ionic liquid composed of di-alkyl imidazolium halides and  $\text{AlCl}_3$  is used to dissolve all components of the capacitor except the tantalum anode. Unfortunately, this approach is only applicable to through-hole capacitors, because surface-mounted device capacitors show no reaction.

Fujita et al., 2014 suggest a direct and easy treatment of the tantalum capacitors with heat and a subsequent size screening. The different heat stabilities of the applied materials lead to a sequential separation. The result is a tantalum powder with a Ta grade of 16%. The authors claim a total recovery rate of 70%.

An entirely different and older approach is given by Stumpf and Seyfert, 1979. Here, capacitors are dissolved in molten aluminum, and the remaining tantalum is separated. Other materials like silver can also be recovered.

All the studies presented describe initial approaches for the identification, liberation, separation, and concentration of tantalum from capacitors applied to PCBs. However, full recovery is not possible yet. In particular, identification and separation are challenging. Therefore, this study focuses on the outputs of manual separation, which represents the safest methodology. Particularly worthwhile equipment types for successful recycling will also be evaluated.

© (Ueberschaar et al., 2017a)

## 2.3. Methodological background

### 2.3.1. Material flow analysis

Experimental material flow analysis<sup>1</sup> (MFA) has been introduced as a systematic approach to track goods<sup>2</sup>/materials and substances in order to understand their origin and fate in investigated processes. It is based on the same procedures used for conventional batch tests<sup>3</sup>, which means a mass balance of goods. Also, the MFA can focus on individual substances (chemical elements, alloys, compounds, etc.) which is often called substance flow analysis (SFA) (Brunner and Rechberger, 2004). Information about individual substances in processed goods is mainly based on individually executed investigations and chemical measurements. (Brunner, 2012)

### 2.3.2. Recycling efficiency

Recycling efficiency describes yields of processes and is used to assess state-of-the-art processes (actual practice). The overall WEEE recycling efficiency can be measured at three levels:

1. collection rate, which represents the ratio between generated WEEE and WEEE collected for recycling.
2. recycling process efficiency rate, which is the quotient of a recycled material and that material collected with WEEE for recycling.
3. (element-specific) recycling rate, which generally refers to functional recycling and is defined by the ratio of recycled material (or element) and the total amount of this material (or element) in generated WEEE. (UNEP - International Resource Panel, 2011)

Although Reuter and van Schaik highlighted that recovery rates could only be assessed by considering the overall process chain, an assessment of the performance at the pre-processing level is a useful tool to identify design-related reasons for resource losses due to insufficient liberation (Reuter and van Schaik, 2015; van Schaik and Reuter, 2010) or to optimize plant operations (Chancerel et al., 2009; Chancerel and Rotter, 2009).

In process engineering, mass and energy balances represent essential tools for assessing process efficiency. Recycling efficiency is usually tested with batch tests. Due to regional legislation, recycling schemes or internal quality management, plant operators are increasingly required to perform such batch tests (NVMP Association, 2014), with WEEELABEX standards being one example for technical implementation. These are defined as the “manual or mechanical processing of a definite and well-defined amount of WEEE or fractions thereof to determine the yields and compositions of the resulting output fractions and de-pollution performance” (WEEEforum, 2013). Basically, such batch tests represent a mass balance of processed material. The informative value of this approach is limited, as no full information about material or substance flows<sup>4</sup> can be provided. A higher level is achieved through partially conducted sorting analyses of particular generated output fractions. Furthermore, the quality of relevant waste fractions regarding de-pollution can be verified by carrying out chemical analyses (WEEEforum, 2013).

© (Ueberschaar et al., 2017b)

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<sup>1</sup> “*Material flow analysis (MFA)* is a method to describe, investigate, and evaluate the metabolism of anthropogenic and geogenic systems. MFA defines terms and procedures to establish material balances of systems. [...] MFA includes balances of both goods and substances.” (Brunner and Rechberger, 2004)

<sup>2</sup> “*Goods* are defined as economic entities of matter with a positive or negative economic value. Goods are made up of one or several substances” (Brunner and Rechberger, 2004)

<sup>3</sup> “*A batch test* is manual or mechanical processing of a definite and well-defined amount of appliances or fractions thereof to determine the yields and compositions of the resulting output fractions” (NVMP Association, 2014)

<sup>4</sup> “*A flow* is defined as a “mass flow rate.” This is the ratio of mass per time that flows through a conductor.” (Brunner and Rechberger, 2004)

### 2.3.3. Recycling-oriented product characterization

Recycling-oriented product characterization is defined as a systematic approach to support the design and the operation of recycling processes. It provides and requires a deep understanding of the needs for recycling processes, specifically focusing on the prevention of avoidable material mixing and dilution while also providing relevant information for the design of new EEE (Chancerel and Rotter, 2009). The characterization of WEEE is structured using following levels:

1. General data on the equipment: equipment type, producer, model name or number and year of production
2. Physical and mechanical properties: weight and size of the whole equipment, material fractions, assemblies, components, connection types
3. Chemical analysis of the assemblies and components: e.g. polymer characterization and element analysis

The focus on critical metals affects for instance level 3, in which the content of critical metals will be measured. However, recyclers report that not only the metal concentration is relevant for the process design and operation, but also:

- ➔ Mechanical properties of the metal-containing assembly (for example tantalum-containing capacitor): What is the particle size? Which other materials are connected, and how are they connected? Are the other materials abrasive?
- ➔ Can assemblies or components that contain critical metals be identified visually?

☞ Conference proceeding (Rotter et al., 2013)

### 2.3.4. Recyclability assessment

Maris and Froelich (2013) analyzed existing methods for recyclability assessment. Still, a consistent definition of recyclability is missing. Ardente and Mathieux (2012) describe recyclability as the ability of waste products to be recycled, based on actual practices. The scope consists of similar approaches used for the ‘recycling-oriented product characterization’. While the ‘recycling-oriented product characterization’ focuses on the chemical analysis of single components and materials and relates to recycling processes only in theory, the recyclability assessment calculates a recyclability rate testing practically the recycling efficiency of state-of-the-art processes. The recyclability rate is defined by the ratio of the recyclable product mass to total product mass. (Ardente and Mathieux, 2012; International Electrotechnical Commission, 2012)

### 2.3.5. Uncertainties and statistical, mathematical modeling

A measurement has imperfections, which lead to a discrepancy between the measured value and the real (unknown) value. Traditionally, this phenomenon consists of two components, namely, a random effect<sup>1</sup> and a systematic effect<sup>2</sup> (Joint Committee for Guides in Metrology, 2008). To ensure statistically correct results, these effects have to be determined for each measurement. In this thesis, one

<sup>1</sup> “Random error presumably arises from unpredictable or stochastic temporal and spatial variations of influence quantities. The effects of such variations, called the “random effects”, give rise to variations in repeated observations of the measurand. Although it is not possible to compensate for the random error of a measurement result, it can usually be reduced by increasing the number of observations; its expectation or expected value is zero.” (Joint Committee for Guides in Metrology, 2008)

<sup>2</sup> “The systematic error cannot be eliminated but it can often be reduced. If a systematic error arises from a recognized effect of an influence quantity on a measurement result, called the “systematic effect”, the effect can be quantified and, if it is significant in size relative to the required accuracy of the measurement, a correction or correction factor can be applied to compensate for the effect. It is assumed that, after correction, the expectation or expected value of the error arising from a systematic effect is zero.” (Joint Committee for Guides in Metrology, 2008)

measurement includes typically the sampling<sup>1</sup>, meaning the sampling procedure and the mass sampled, as well as the chemical analyses, which comprise the mass of the sample taken and the analytical methodologies applied.

The uncertainties are calculated for own analyses and have to be determined for data from literature research. Due to this, an uncertainty propagation with an appropriate mathematical model has to be applied, which provides one value for overall uncertainty.

In general, systematic effects have to be minimized through careful, practical work. They can also be eliminated afterward through the use of correction factors (Joint Committee for Guides in Metrology, 2008). Therefore, uncertainties provided for measurement results should be based mainly on random effects.

In this doctoral thesis, distributions were modeled as normal and mathematically independent. The necessary uncertainty propagation bases on the Gaussian concept partially simplified with Goodman's expression. In contrast to the principle of maximum uncertainties, relative uncertainties are calculated for all random effects determined.

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<sup>1</sup> “*Sampling* is a necessary physical handling step in many fields of industry and the science behind it has its basis in chemistry, physics, and mathematics. [...] The main idea of taking a correct sample includes the withdrawal of a sufficiently enough amount of material for it to be representative for the total amount of material. From a practical point of view, it is essential to know the minimum amount of material that can be regarded as being representative. Simultaneously, it is of importance to know how accurate an obtained sample is, i.e., preferably, the deviation between the true properties of the total batch and the amount that has been withdrawn as a sample needs to be known.” (Dihalu, 2012)

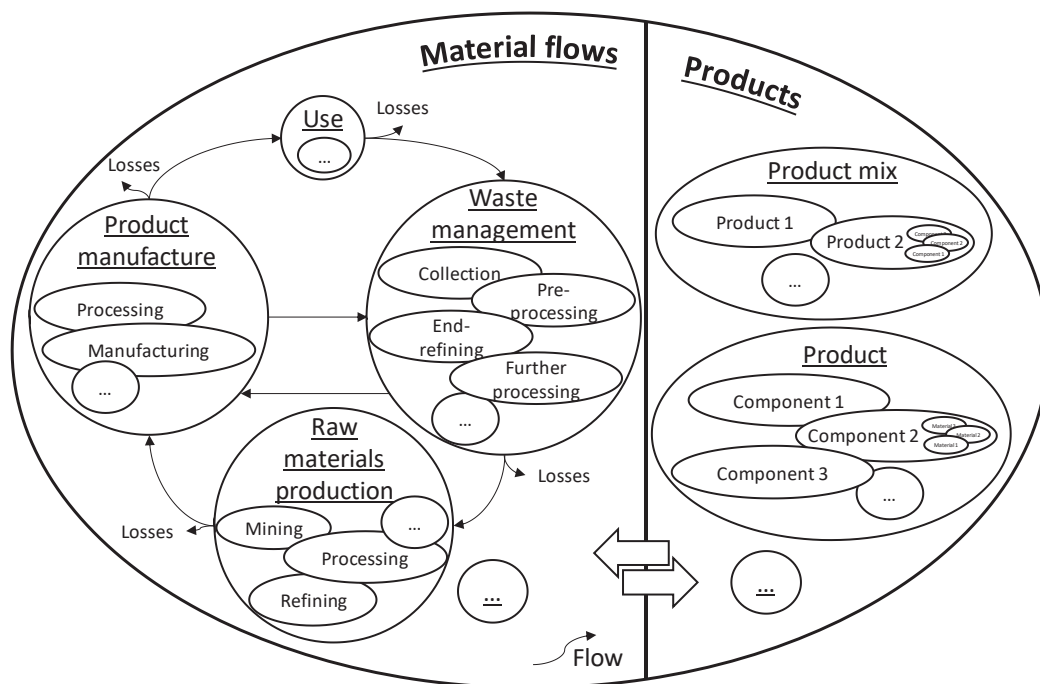
### 3. Material and methods

#### 3.1. Research concept

WEEE product-centric information must provide a fundamental data base, which is applicable for the derivation of recycling strategies by extending them with external, non-product related, data such as material purchase and retail prices, investments, or processing costs. WEEE product-centric information used for the political, technical and economic dimension of recycling strategies are interdependent.

The basis for all further investigations is the generation of holistic product and material flow information. Focusing on S-CRMs, hotspots<sup>1</sup> in WEEE products and flows can be identified. In this thesis, S-CRM hotspots are defined by their suitability being an access point for S-CRM recycling basing on a minimum mass fraction of target metals, mass fractions of by-applied materials, the design complexity of the WEEE product as well as other chemical and physical properties.

This hotspot analysis is applicable on WEEE material flows and WEEE products (cf. Figure 12). Investigating material flows, usually MFAs (cf. 2.3.1 “Material flow analysis”) are used focusing on the whole life-cycle<sup>2</sup> of any material in single systems<sup>3</sup> such as waste management or even subsystems such as pre-processing of WEEE. Studies on WEEE products are based on a recycling-oriented product characterization (cf. 2.3.3 “Recycling-oriented product characterization”) to determine physical and chemical properties. This can be done for a product mix, single products, and disassembled components or materials.



Own illustration

Figure 12: Systems and subsystems in WEEE products and material flows. Left: Material flows depicted as life-cycle. Right: Products and product mixes

<sup>1</sup> *Hotspot*: a part of a WEEE material flow or product, which holds a relatively high amount or share of a target substance.

<sup>2</sup> *Life-cycle* refers to the stages of a product's life from raw material extraction through materials processing, manufacture, distribution, use, repair and maintenance, and disposal or recycling.

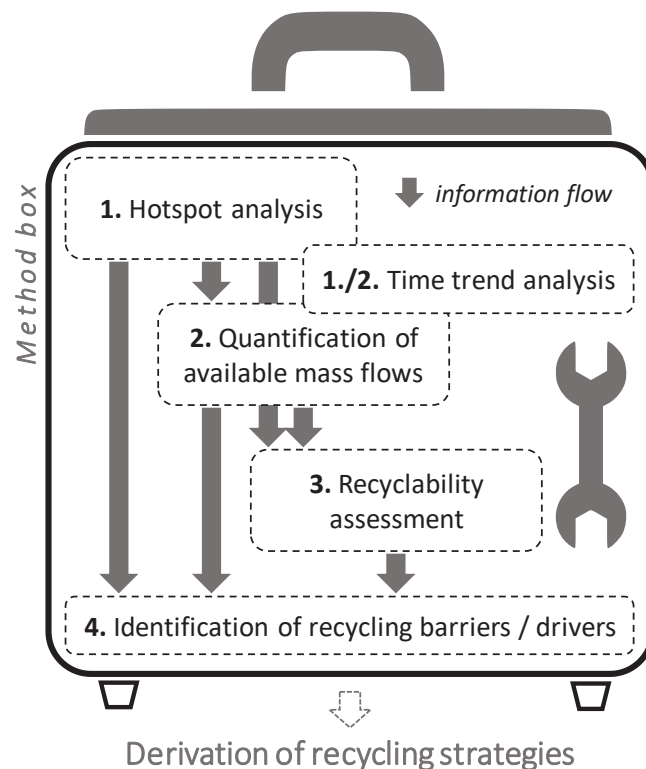
<sup>3</sup> “The *system* is the actual object of an MFA investigation. A system is defined by a group of elements, the interaction between these elements, and the boundaries between these and other elements in space and time. It is a group of physical components connected or related in such a manner as to form and/or act as an entire unit.” (Brunner and Rechberger, 2004)

Using WEEE material flow and WEEE product information, masses of S-CRMs available for recycling purposes or put-on-market (POM) embedded in new EEE products can be calculated. Depending on the quality of the generated data, recorded manufacturing dates of WEEE products allow for the derivation of possible time trends in the application of target materials in WEEE products and subsequently in the calculated masses POM and available for recycling.

In the context of the WEEE product design, a subsequent recyclability<sup>1</sup> assessment (cf. 2.3.4 “Recyclability assessment”) checks for the recyclability of targeted S-CRMs and other materials.

Finally, a recycling barrier analysis consolidates all gathered information and sets them into context with general recycling boundary conditions. Barriers and drivers, which hinder or support the recycling of targeted S-CRMS and other applied materials in WEEE products can be identified and evaluated.

This set of measures results in a consecutive order of methodologies enabling the derivation of recycling strategies. Figure 13 shows the overall methodological approach.



Own illustration

Figure 13: Consecutive methodologies for the generation of product-centric information enabling the derivation of recycling strategies

The consecutive set of methodologies for the generation of product-centric information base on reviewed, consolidated and further developed methodologies. In this thesis, this overall approach is applied in various practical case studies.

<sup>1</sup> *Recyclability*: the ability of waste products to be recycled, based on product attributes and used recycling techniques. (International Electrotechnical Commission, 2012; Ardente and Mathieux, 2012)

### 3.2. System boundaries

#### 3.2.1. WEEE products and target materials

Chancerel et al. (2013) investigated the need for research in the field of S-CRM recovery from WEEE. 26 applications were listed using the selected S-CRMs, from which eight applications were identified as relevant for further research. These were antimony used as flame retardant in plastics, gallium, and germanium in ICs on PCBs, REEs in phosphors of cold cathode fluorescent lamps (CCFLs), and LEDs in NdFeB permanent magnets, cobalt in batteries, tantalum in tantalum capacitors and indium in ITO in LCDs. Moreover, Chancerel et al. (2013) state a lack of research for batch analyses of critical metals in complex equipment mixes in WEEE processing. This doctoral thesis, therefore, focuses on:

#### 1. Material flows

- a. Metal life-cycle MFA on the example of gallium
- b. MFA of S-CRMs, PMs, and IBMs in pre-processing of WEEE (waste management)

#### 2. Products

- a. Gallium in ICs on PCBs in various WEEE devices
- b. Indium applied as ITO in various WEEE devices using LCD panels
- c. REEs used for NdFeB magnets applied in HDDs in various WEEE devices
- d. Tantalum in tantalum capacitors on PCBs in different WEEE devices

#### 3.2.2. Time and regional boundaries for quantification of available mass flows

Based on available POM and collection data, system boundaries for the quantification of available mass flows were set to Germany (POM and WEEE), worldwide (POM) in the year 2013. The quantification is carried out for indium, REEs, and tantalum. The potential analysis for gallium is part of the gallium life-cycle MFA. Here, most data were available for the year 2011. The regional boundary is Germany.

### 3.3. S-CRM hotspot analysis via material flow analysis

#### 3.3.1. MFA gallium life-cycle

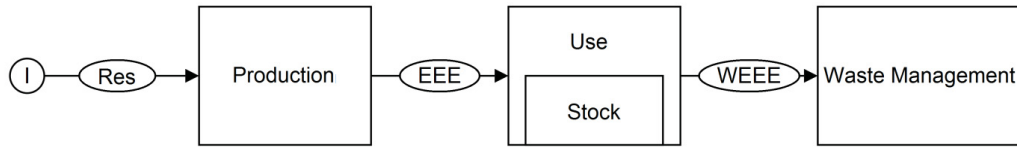
This MFA study focuses on gallium in the EEE and WEEE sector. Recent studies by Chancerel et al., 2015, 2013 or Oguchi et al., 2011 highlighted the relevant equipment types from information and telecommunications equipment (IT) and consumer equipment, which were investigated in detail (cf. Table 3).

Table 3: Relevant WEEE categories for GaAs potential

Category	Equipment types investigated	UNU key
Equipment investigated carrying gallium in PCBs	Desktop PC	0302
	Notebook	0303
	Printer	0304
	Mobile phone	0306
	Video Cassette Recording (VCR)	0404
	DVD-Player	0404
	Radio/Recorder	0403
	Digital camera	0406
	Video camera	0406
	Video games	0702
Equipment investigated carrying gallium in LED for background lighting	Notebook	0303
	TFT-TV	0408
	Flat screen monitor	0309

Based on: (Chancerel et al., 2013; Oguchi et al., 2011)

For carrying out an MFA, the life-cycle of gallium was split into three sub-systems, namely “production”, “use” and “waste management” (cf. Figure 14). The use phase is not considered in this MFA. The systems “production” and “waste management” were modeled individually, since the major gallium share from production is exported as semi-finished products and most gallium-bearing products are imported.

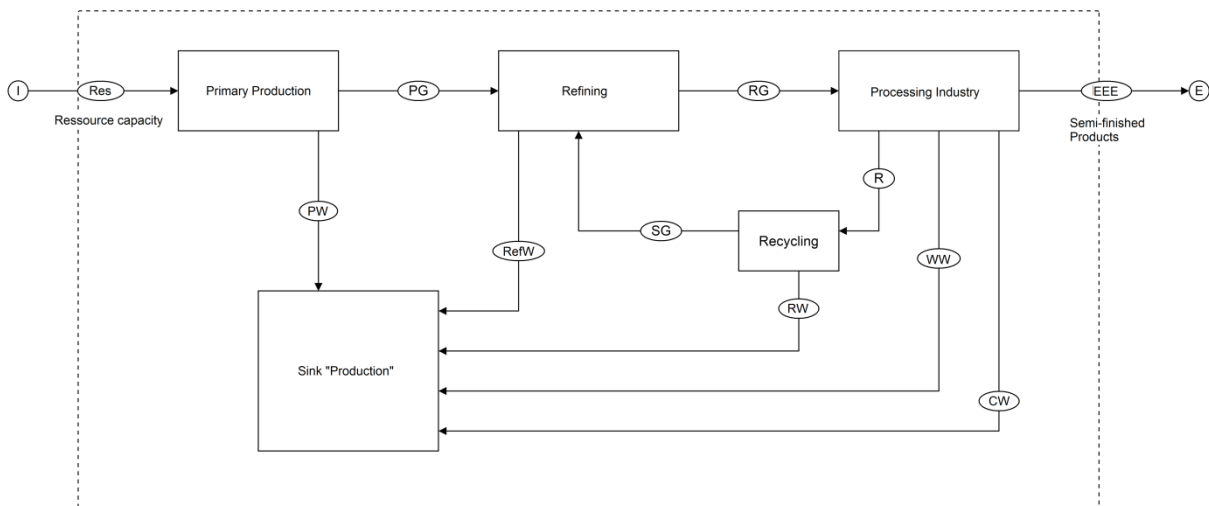


Own illustration (Ueberschaar et al., 2017c)

Figure 14: Lifecycle of gallium (Res = primary resource)

As regional system and time boundary Germany with the reference year 2012 was chosen. The focus is set on IT and entertainment equipment containing gallium-bearing PCBs or LEDs (cf. Table 2). Not all data, necessary for calculating needed transfer coefficients are available for this year. Thus, approximated and fitted data from other years were used.

As there is no current recycling of gallium from WEEE, the system ends with the waste management and is not reconnected to the production side. Only recycling efforts in the production processes feed back directly into the production system. Figure 15 shows the detailed qualitative material flow model of the system „production“, which is used for further modeling. It contains five process steps that are linked to each other, wherein the sink represents the sum of all losses. The model starts with the primary resource potential (Res) of gallium in bauxite, which is related to efficiencies of status-quo processes. Here, losses (primary waste: PW) are transferred to the red mud (cf. supporting information S2.2.1 in Ueberschaar et al. (2016c)). The extracted gallium (primary gallium: PG) is then refined to obtain desired purities (cf. supporting information S2.2.2 in Ueberschaar et al. (2016c)). Losses through refining waste (RefW) are summed up in the “sink production”.



Own illustration (Ueberschaar et al., 2017c)

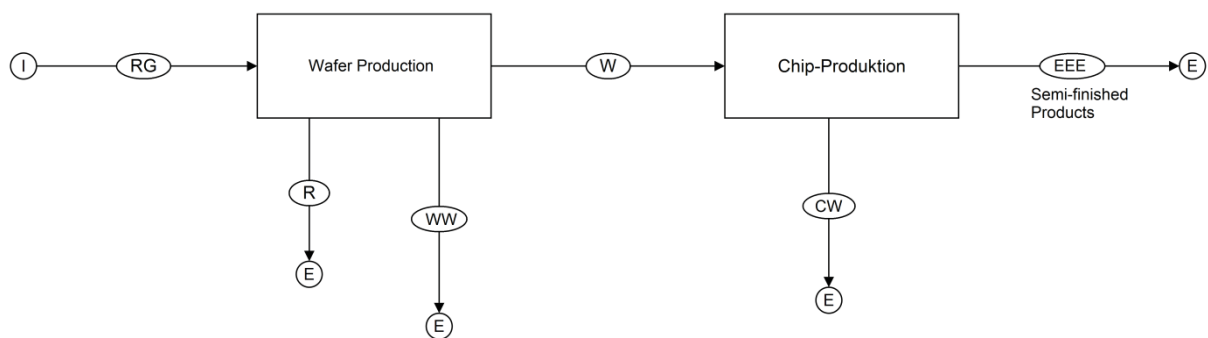
Figure 15: Qualitative material flow model of the gallium “production” system (PG: primary Ga, RG: refined Ga, R: Recycle, SG: secondary Ga, PW: primary production waste, RefW: refining waste, RW: recycling Waste, WW: production waste (Wafer), CW: production waste (chip))

Germany has only low refining capacities (Roskill, 2012). The process itself is not bounded to a specific location (Wuppertal Institut für Klima - Umwelt - Energie GmbH, 2011). This leads to considerable import and export masses. The import constitutes the major source of refined gallium for Germany. The export is dominated by unrefined material (cf. supporting information S2.1 in Ueberschaar et al.

(2016c)) (Liedtke, 2015). These mass flows are not part of the model, as only Germany's self-supply was investigated.

The refined gallium (RG) is then transferred to the producing industry and applied in wafers (W), which are passed on to the chip producing industry for further processing steps. In addition to losses in both steps (WW and CW), the wafer production generates a recyclate (R) as a by-product, which can be recycled subsequently to secondary gallium. This material is fed to the refining processes of gallium. A final semi-finished product represents the output from the subsystem "production". Supporting information S2.2 in Ueberschaar et al. (2016c) provides more detailed information for this subsystem.

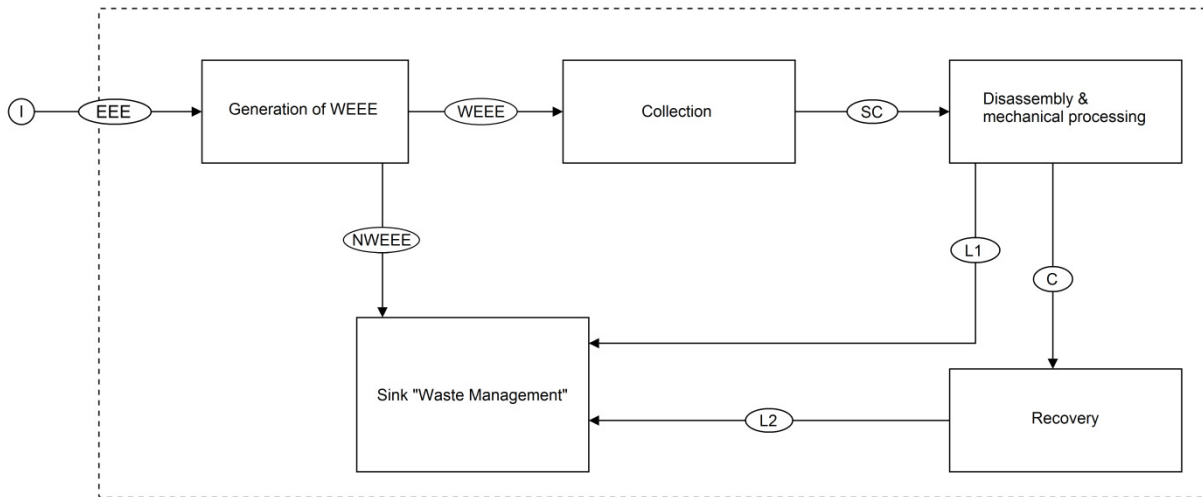
The system "production" includes another sub-system named "processing industry" (see Figure 16). Here, one individual system was developed based on the wafer and chip production to harmonize various existing processes with fluctuating losses. So far, only in the production of wafers, a recycling of new scrap takes place without considering CIGS solar panels.



Own illustration (Ueberschaar et al., 2017c)

Figure 16: Detailed scheme of the sub-system "processing industry" (RG: refined Ga, W: wafer, R: recyclate, WW: wafer waste, CW: chip waste)

Figure 17 depicts the system "waste management", which refers to all relevant IT and entertainment equipment types (cf. Table 3). This sub-system starts right after the use phase of EEE. In order to apply an appropriate recycling, WEEE is collected by public waste management authorities, distributors and manufacturers which share the product responsibility (Bundesministerium für Umwelt Naturschutz Bau und Reaktorsicherheit, 2015). Here, first losses occur due to low collection rates of relevant devices. The collected material is then pre-processed using manual and mechanical treatment. PCBs represents the major carrier of gallium and are therefore characterized as a concentrate (C). The separation process of these components is related to losses, basing on low efficiencies in the sorting processes or losses through dust during a mechanical treatment. Pyrometallurgical processes are used for the recovery of metals in PCBs. Here, an extraction of gallium is possible but not executed yet. Therefore, no recovery flows of gallium are included in this system. Like in the system "production", the sink is the sum of all losses.



Own illustration (Ueberschaar et al., 2017c)

Figure 17: Qualitative material flow model of the system "waste management" (NWEEE: not collected WEEE, SC: separate collection, C: concentrate, L1 and L2: losses)

The system "waste management" differentiates between the WEEE streams, which are officially collected and the transfer to the informal sector. However, due to inconsistent and fragmentary background data, the waste flows in the informal sector are not part of this study. Supporting information S2.3 in Ueberschaar et al. (2016c) provides more information for the subsystem "waste management".

#### Data collection and processing

The quantitative determination of required data followed two different methodical approaches. Process input and output flows were determined by literature research. Data gaps were filled with generally assessed transfer coefficients. These two approaches were used complementarily in this study. Data on transfer coefficients were collected by literature research, expert consultations and reasonable assumptions (cf. supporting information S2.2 in Ueberschaar et al. (2016c)).

Furthermore, uncertainties in the data sets have been considered, and an error<sup>1</sup> propagation has been applied. As the data sources for this MFA are heterogeneous, they should not be used equivalently. Following Laner et al., 2015, sources have been rated regarding their reliability and appropriate uncertainties have been set up (c.f. Appendix 4).

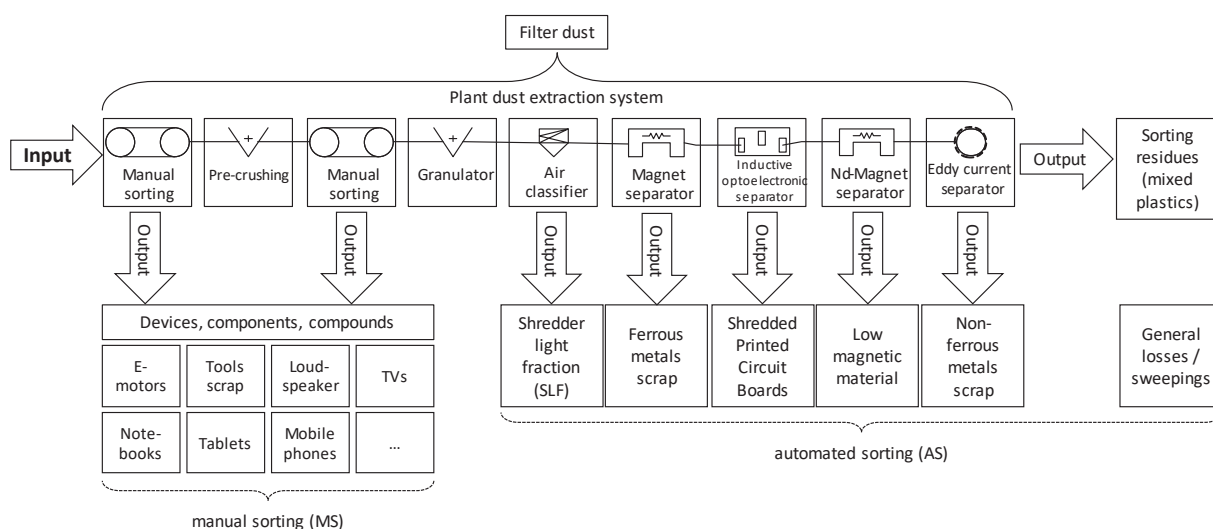
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<sup>1</sup> Error: "In general, a measurement has imperfections that give rise to an error. The error is the result of a measurement minus a true value of the measurand" (Joint Committee for Guides in Metrology, 2008). See glossary for further explanation.

### 3.3.2. Extended batch test in WEEE pre-processing

For this study, a batch test was conducted in a WEEE pre-processing plant. Processes cover liberation, and manual and automated sorting, which can be regarded as typical in the pre-processing sector. The input material was 40 Mg WEEE comprising a mix of information and telecommunication technology devices, consumer entertainment devices, small household appliances, electrical and electronic tools, toys, and electric motors, etc. Only a few non-designated devices were present in the batch, such as air conditioning and cooling appliances and lamps, which were separated in the manual sorting step.

Figure 18 shows the process scheme of the study plant with the input and output streams generated by the various separation processes. The output materials are grouped as output fractions from “manual sorting” (MS) and “automated sorting” (AS). A coding for all output fractions in the extended batch test is depicted in the supporting information S2 in Ueberschaar et al. (2016a).



Own illustration (Ueberschaar et al., 2017b)

Figure 18: Process set-up of recycling plant investigated

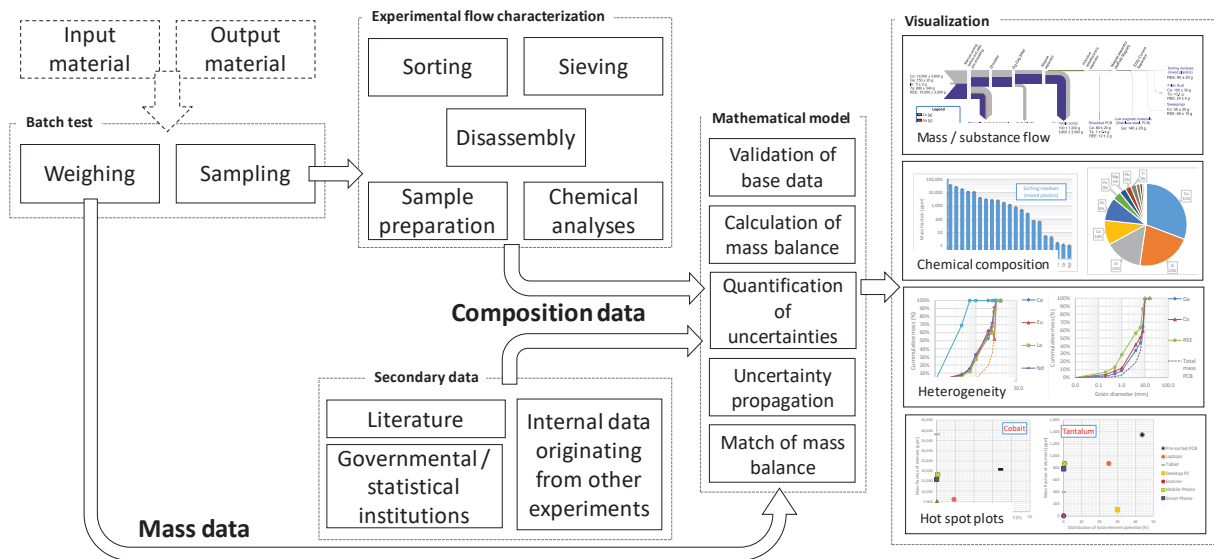
Target metals of this investigation include IBMs like aluminum, iron, and copper, PMs (gold, silver, palladium, and platinum) and S-CRMs like cobalt, gallium, indium, tantalum and the WEEE relevant REEs (neodymium, praseodymium, dysprosium, lanthanum, cerium, europium, lutetium, etc.).

Various experimental MFA have been conducted in recent years with different objectives and waste streams. Based on a literature review on various studies using experimental batch tests, a general methodology for carrying out a batch test was developed (cf. Ueberschaar et al., 2016a). Figure 19 depicts the complete approach.

Based on chemical characterization<sup>1</sup>, the composition of all input and output materials can be assessed. For output fractions consisting of whole devices or components, disassembly prior to chemical analyses might be necessary. Additional sorting and sieving analyses represent adequate tools to increase information density and enable a validation of data.

Chemical analyses of all output fractions are not always possible. In such cases, sufficient data must be available from own studies or external research. This secondary data has to be calculated with the experimental data within an appropriate mathematical model. Through this, the overall flows and stocks can be assessed.

<sup>1</sup> Chemical characterization consists of all procedures for the determination of the chemical composition of any substance. This includes the sampling, sample preparation and chemical analysis.



Own illustration (Ueberschaar et al., 2017b)

Figure 19: Experimental design of the "extended batch test" with subsequent data collection and evaluation

Uncertainties are usually not part of MFA studies (Laner et al., 2014). However, to provide comprehensible quantitative data, the apportioning of uncertainties is necessary. Therefore, the mathematical model used must contain an adequate assessment and propagation of uncertainties.

### Sampling

In this study, sampling followed the methodology of LAGA PN 98, which provides a general guideline for procedures for the investigation of physical, chemical and biological properties of waste (Länderarbeitsgemeinschaft Abfall, 2001). According to the grain size distribution, heterogeneity of material and the substances to be investigated, appropriate sampling procedures needed to be chosen according to the kind of transport or storage (head, belt, falling stream, etc.) and material characteristics. On this basis, minimum sample quantities for each output fraction were calculated following LAGA PN 98. Table 4 shows the sample codes for the related output fractions and the masses sampled in the batch test. Appendix 2 describes the sampling strategy in detail.

### Subsampling from primary samples

To provide sufficient representative material for the characterization of the output fractions, the samples taken were split for chemical analyses, sieving analyses, and sorting analyses, with some partial samples retained. The division of the samples was carried out using a ripple divider for handling large sample volumes (cf. supporting information S4 in Ueberschaar et al. (2016a)).

Table 5 shows the sample masses for the characterization tests subsequently carried out.

The chemical analyses were conducted in up to three laboratories, depending on the elements to be measured and the complexity of the sample.

Table 4: Overview of sampled fractions over an 11 h sampling period

Sampled Output			Output batch test	Overall sampling quantity before reduction
Sample code	Sample name		[kg]	[kg]
Manual sorting	MS1	Tools scrap	789	33
	MS2	E-Motors	1,645	549
	MS3	Loudspeaker	397	237
	MS4	Copper rich fraction	3,271	47
	MS5	Pre-sorted PCBs	221	283
Automated sorting	AS1	Ferrous metals scrap	9,789	228
	AS2	Low magnetic material	709	77
	AS3	Non-ferrous metals scrap	1,105	108
	AS4	Shredded PCBs	625	87
	AS5	Sorting residues (mixed plastics)	12,498	208
	AS6	SLF (Fluff)	1,691	67
	AS7	Filter dust	96	8
	AS8	Sweepings	~100	33

Table 5: Sample splitting for chemical, sieving and sorting analyses

Sample code	Mass for sieving and sorting analysis [kg]	Chemical analyses			
		Total mass for chemical analysis [kg]	Laboratory 1 [kg]	Laboratory 2 [kg]	Laboratory 3 [kg]
AS1	66	84	74	10	10
AS2	37	41	41	-	-
AS3	54	54	54	-	-
AS4	22	50	43	7	-
AS5	51	71	58	13	-
AS6	14	33	30	3	-
AS7	-	8	6	2	-
AS8	-	33	33	-	-
MS5	-	67	67	-	-

The unlisted output fractions were not split and were used directly for the chemical analyses.

### Characterization of output and input fractions from the extended batch test

Characterization of output materials provides the database for the MFA. The information was gathered through physical and chemical characterization of the input materials and output fractions from manual sorting (MS) and automated sorting (AS). A full analysis is not always possible or feasible. Therefore, different methodologies were used.

#### *Physical characterization*

##### → Batch input characterization

A total of 40 Mg input material was manually sorted prior to the batch test to identify the equipment types present (for example cooling device, CRT TV, tablet, smartphone, etc.) and to determine average weights of the WEEE products in the batch. The material was later remixed to provide a realistic input. The supporting information S1 in Ueberschaar et al. (2016a) shows the sorting protocol used.

##### → Disassembly and material quantification (MS)

Dismantling trials were used as a tool to assess the material composition of whole devices and components from manual sorting. Samples were disassembled until manual separation with mechanical tools was no longer possible. Target end materials were components like PCBs and batteries, but mostly homogeneous materials like ferrous metals, non-ferrous metals, plastics, and composite materials, which represent an inseparable compound of homogenous materials. The assessment of the output fractions was based on visual identification in combination with a magnet for verifying ferrous metals.

The supporting information S6 in Ueberschaar et al. (2016a) shows the disassembly protocol draft. The composition data assessed for small household tools, loudspeaker drivers, and e-motors is depicted in S7 in Ueberschaar et al. (2016a).

##### → Sieve analysis (AS)

The output fractions AS1 - 6 were sieved in a Haver "Test Sieve Shaker EML 450 DIGITAL PLUS" using sieves with nominal mesh widths of 0.125; 0.25; 0.5; 1.0; 2.0; 5.0; 8.0; 10; 16; 20 mm. Sieving time was about 5 min with self-readjusting amplitude up to maximal 2 mm.

Due to high shares of fines between 0.25 and 5.0 mm, an additional sieve analysis with nominal mesh sizes 0.2; 0.5; 1.0; 4.0; 6.3; 8.0; 10; 16 mm was carried out for the materials AS4-7 with a KH "Tfk. Rö/W-B" in combination with a subsequent determination of the chemical composition. Sieve time was 10 minutes for 4.0-16 mm and 12 min for <4.0 mm with sample sizes AS4 (7.3 kg), AS5 (12.5 kg), AS6 (2.3 kg) and AS7 (1.6 kg), which did not originate from the extended batch test, but from the same processing plant with comparable input material.

##### → Sorting analysis (AS)

Output fractions from the automated sorting processes >5 mm were sorted by visual identification. This excluded the materials AS6 - 8. A sorting protocol was developed, which was used for the analysis (cf. supporting information S5 in Ueberschaar et al. (2016a)). Main categories for sorting were: ferromagnetic material; aluminum; other non-ferrous metals; colored plastics, black plastics; foils, rubber, etc.; PCBs; batteries; cables; glass, ceramics; organic material; and the rest.

### *Chemical characterization*

#### → Hotspot characterization of disassembled materials (MS)

In addition to the visual material quantification in the dismantling trials, hotspots like metallic pieces or other materials like magnets were determined via an XRF handheld (Thermo Fisher/Analyticon XL3 air) to quantify alloying elements and verify first results.

#### → Literature data on chemical composition (MS)

Not all output fractions from the manual sorting were assessed through disassembly trials with subsequent chemical analyses. Generally, for well-investigated output fractions, in particular whole devices and components, a literature research was carried out. This applied to IT devices like mobile phones, notebooks, etc., and also for single components like batteries or particular output materials, which were assessed in former batch tests. The supporting information S7 in Ueberschaar et al. (2016a) shows the data aggregated for further calculations.

#### → Characterization of sieving steps (AS)

For the characterization of the output fraction of each individual sieving step in the sieve analysis, samples were dissolved microwave-assisted in aqua regia with 1.600 W, 20 bars, 15:00 min in a CEM MARS 5. For the elemental determination, an ICP-AES (Thermo Scientific iCAP 6000 Series) was used.

#### → Full analysis of output fractions (AS)

Full analyses were carried out on the output fractions from the automated sorting processes (AS1 – 8). To address matrix influences on the measured contents of specific elements, chemical analyses were carried out by three different laboratories using different sample assaying and analytical methods for Ag, Al, As, Au, Ba, Bi, Ca, Cd, Ce, Co, Cu, Eu, Dy, Fe, Ga, Ge, La, Mg, Mn, Mo, Nd, Ni, Pb, Pd, Pr, Pt, Sb, Si, Sm, Sn, Ta, Tb, Te, Ti, V, W, Zn, Zr. In addition, loss of ignition (LOI) was determined by laboratory 1 during the assaying step.

Laboratory 1 applied a fire assay and remelting with a recuperation of target elements. An X-ray fluorescence analysis (XRF) and a wet-chemical digestion with a subsequent determination through inductively coupled plasma mass spectrometry (ICP-MS) and atomic emission spectroscopy (ICP-AES) were carried out.

Laboratory 2 analyzed smaller sample sizes in order to compare the results with laboratory 1. For sample preparation, samples AS4 - 7 were milled in a cross-beater mill (Retsch SK1) and an ultra centrifugal mill (Retsch ZM1) with the addition of dry ice in the case of a high plastics content to <0,2 mm powder. Sample AS1 was completely melted at 1700°C without any additives in a standard atmosphere, then homogenized and subsequently cast in a homogeneous solid state. The product was analyzed with an XRF handheld (Thermo Fisher/Analyticon XL3 air). Single pieces were drilled out for a wet-chemical analysis at laboratories 2 and 3. Subsequently, all samples were digested with HNO<sub>3</sub>-H<sub>2</sub>O and aqua regia in a microwave-assisted digestion with 1.600 W, 20 bars, 15:00 min in a CEM MARS 5. For the elemental determination, an ICP-AES (Thermo Scientific iCAP 6000 Series) was used.

Laboratory 3 received the prepared single pieces of the cast AS1 sample from laboratory 2. Here, an optimized determination for gold was carried out. For this purpose, the sample was digested in microwave-assisted aqua regia at 215°C. The measurement was carried out with a QQQQ-ICP-MS in O<sub>2</sub>-mode.

## Data processing and uncertainty assessment

### *Calculation of the mass balance*

The general approach for determining the mass balance on the elemental level is described in Equation 1. Equation 2 shows the calculation of element specific transfer coefficients following Chancerel et al., 2009 and Rotter et al., 2004.

*Equation 1: Calculation of mass balance on element level*

$$m_{i,input} = m_{input} * x_{i,input} = \sum_{j=1}^k m_{i,fraction_j} * x_{i,fraction_j}$$

*Equation 2: Calculation of element specific transfer coefficients*

$$TC_{i,fraction_j} = \frac{m_{i,fraction_j} * x_{i,fraction_j}}{m_{input} * x_{i,input}}$$

*With:*

*x = mass fraction (mg/kg), m = mass (kg); indices: j = output fraction from batch test, i = substance / element, k = number of output fractions*

The uncertainty assessment is presented in Appendix 3.

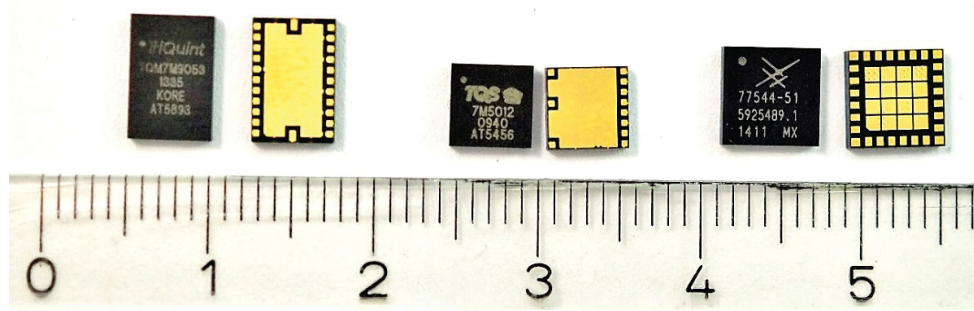
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### 3.4. S-CRM hotspot analysis via product analysis

Following chapter 2.3.3, the recycling-oriented product characterization was carried out on separated ICs, LCD panels, HDDs and tantalum capacitors.

#### 3.4.1. Integrated circuits

Three different state-of-the-art IC used for mobile radio standards like 2G, 3G, 4G or WLAN were investigated in detail. These components are applied in modern smartphones, mobile phones, tablets, etc. Figure 20 shows example pictures of the IC types investigated. Table 6 lists the properties of the components and the investigated quantities.



Own illustration (Ueberschaar et al., 2017c)

Figure 20: Overview of investigated chip types (from left to right: Type 1, Type 2, Type 3)

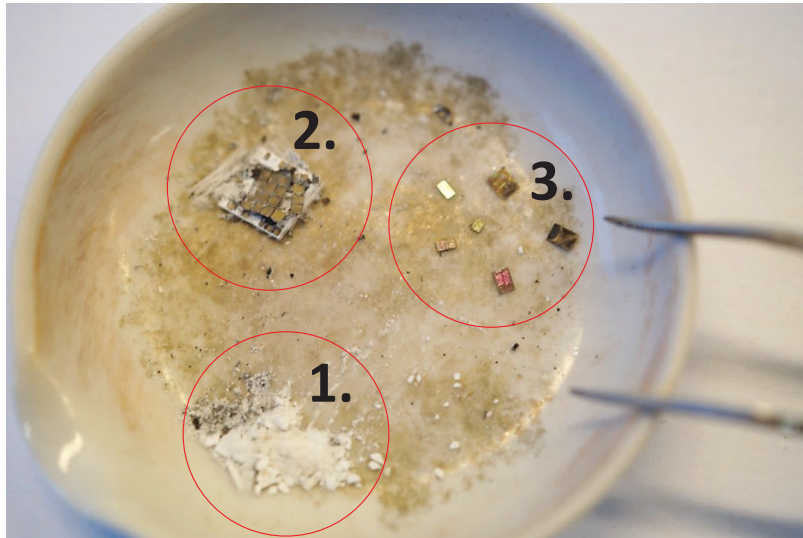
All chips consist of labeled synthetic material at the front and a metal plate at the back. The area is less than 1 cm<sup>2</sup> for all chips. Supporting information S3.2 in Ueberschaar et al. (2016c) depicts the single weights of the samples.

Table 6: Amount and details of investigated radio chips

Chip-Type	Manufacturer	Model no.	Specifications	Average weight	Number of samples
Type 1	TriQuint	TQM7M9053	Quad-band GSM-EDGE and Tri-Band W/CDMA/HSPA+/LTE	86±2 mg	4
Type 2	TriQuint	TQM7M5012	Quad-Band GSM850/GSM900/DCS/PCS Power Amplifier Module	61±0.3 mg	3
Type 3	Skyworks	SKY77544	Quad-Band GSM / GPRS / EDGE – Triple-Band WCDMA	81±0.1 mg	2
Total			-		9

Pre-tests showed that no manual, mechanical nor chemical liberation of single components was possible in order to liberate containing materials and components. Therefore, a thermal treatment was applied. The samples were heated at 250° C for 110 min and again at 550° C for 360 min in a muffle furnace. With this procedure, the plastics casings were burned and the inboard components were liberated. The thermal exposure revealed single fractions, which were clearly separable (cf. Figure 21). For a subsequent chemical analysis, the liberated components have been sorted according to visually identifiable characteristics like form or color. Four clusters were identified:

1. Isolation with copper connectors
2. Metal plates (back of the chip)
3. Electronic modules (chip)
4. Others



Own illustration (Ueberschaar et al., 2017c)

Figure 21: Liberated and sorted components of a high-frequency radio chip after thermal treatment

All fractions were weighted and chemically analyzed with an X-Ray fluorescence analysis (XRF) (Thermo Fisher/ Analyticon XL3 Air) to determine the chemical composition. The samples were measured several times from various angles without any further processing to avoid losses related to milling and decanting.

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### 3.4.2. LCD panels

#### Chemical analysis of indium and tin in the panel sandwich

An optimized test method to quantify the indium mass fraction in the LCD panel was earlier developed (Jalalpoor et al., 2013). This approach avoided any pretreatment of the LCDs to minimize indium losses prior to analysis. Consequently, the panels were analyzed without a crushing step or the removal of the polarizer films. As the large panels could not be analyzed as a whole, single pieces were cut out with masses of around 0.5 to 1 g and a cross-section of approximately 1 cm<sup>2</sup>. A microwave assisted leaching of the indium from the glass surface was carried out (CEM Ultra-High Throughput Microwave Digestion System MarsXpress) with 10 ml nitric acid (HNO<sub>3</sub>, 65%, Rotipuran Supra; Fa. Roth) at 180 °C. Further parameters were a maximal pressure of 20 bars, 20 min preheating time, 20 min retention time at 180 °C and 20 min cooling time. Three parallel measurements per sample were carried out. In order to avoid systematic effects from analysis procedures and to check the quality of the results, the chemical analysis was carried out in parallel with an ICP–OES (Inductively Coupled Plasma – Optical Emission Spectrometry) (Thermo Fisher iCAP 6300 Duo) and a flame AAS (atomic absorption spectroscopy) (Perkin Elmer F-AAS 1100 B). The validation of the chemical analyses is provided in the supporting information S11.

This methodology was applied to six different LCD device groups originating from dismantling studies: mobile phones (UNU key 0306-01; n=11), smartphones (UNU key 0306-02, n=26), tablets (UNU key 0303-02, n=26), notebooks (UNU key 0303-01, n=20), PC monitors (UNU key 0309-01, n=11) and LCD TVs (UNU key 0408-01, n=9).

#### Chemical analysis of toxic heavy metals and strontium

The samples were prepared as described for the determination of indium in the LCD panels. After cutting out a 1 cm<sup>2</sup> large piece from the center of the panel, the panels disintegrated into 2 halves. Due to the application of the elements to be measured in the glass panels, a wet chemical analysis using an ICP would have only been possible after a total digestion of the panels with hydrofluoric acid. For safety reasons and to facilitate the measurements, analyses were conducted via X-ray fluorescence (XRF). The two halves were each analyzed from both sides with a handheld device from Thermo Fisher / Analyticon XL3 air.

Devices from the six different LCD equipment groups mentioned above were analyzed: mobile phones (0306-01, n=26), smartphones (0306-02, n=27), tablets (0303-02, n=26), notebooks (0303-01, n=18), PC monitors (0309-01, n=10) and LCD TVs (0408-01, n=5).

#### Polymer analysis of polarizer foils

The type of polymers applied as polarizer foils in LCD panels plays an important role in the economic efficiency of future recycling strategies. Cellulose acetate represents high-value polymers, which would support recycling efforts if applied throughout the range of relevant LCD devices. Therefore, small samples of polarizer foils were manually separated from ten LCD panels obtained from smartphones and tablets and investigated regarding their polymer type to check for the regular use of cellulose acetate. Polarizer foils were subjected to an Attenuated Total Reflectance Fourier Transform Infrared spectroscopy (ATR-FTIR, Spectrum One, Perkin Elmer) without further pretreatment. Resulting spectra were matched with spectra from the "ATR of polymer" database from Perkin Elmer for identification of polymer type.

☞ (Ueberschaar et al., 2017d)

#### 3.4.3. Hard disk drives

Following the recycling-oriented product characterization, this study focuses on Personal Computers (PCs) (UNU Subkey 3021) and notebooks (UNU Subkey 3031). An in-depth analysis of in total 49 3.5" HDD from desktop PCs and 6 2.5" HDD from notebooks was conducted to determine the product structure and material composition followed by a semi-quantitative analysis of the casing material, a chemical analysis of the PCBs and the magnet components.

109 desktop PCs were disassembled in two German pre-treatment recycling plants to obtain the share of 3.5" HDDs. Also, 6 notebooks have been dismantled for comparison reasons. Subsequently, the HDDs were investigated in detail.

#### Manual disassembly of hard disk drives

Manual disassembly is a crucial step for characterizing end-of-life devices to get information about the structure and furthermore about opportunities for subsequent recycling strategies and to separate components of interest for chemical analyses. For investigating the REE potential of HDDs, NdFeB magnets have to be liberated. The weight of the dismantled components is used in pursuing chemical analyses for an estimation of the overall share of focused materials.

To obtain further information about the disassembled components in the HDDs and to characterize the material, an elemental semi-quantitative determination was carried out with an XRF device (Thermo Fisher / Analyticon XL3 air).

### Chemical composition of NdFeB magnets

Each material applied in EEE is embedded in a specific matrix related to its functional use. These matrices influence analytical results and the quality of obtained results. The most common approach for facing these matrix effects is to develop calibrations using standard samples with the same matrix as the sample to be measured and known concentrations of the elements which have to be determined (Fifield and Haines(editors), 2000). This is explicitly important for solid samples for which the matrix effects are contrastable higher (Nomura and Oliveira, 2007). In order to assess these matrix effects, different methods for the sample preparation and varied solvents for the leaching processes were tested to evaluate the best methodology for the determination of REE in NdFeB magnets. This approach leads to the most accurate results working with an ICP-OES or ICP-MS.

Furthermore, first evaluations for the use of XRF analysis were investigated. Utilizing other measuring principles for the determination of the chemical composition, this technique was intended to obtain a second set of comparable results of REE in NdFeB magnets (cf. supporting information 1 in Ueberschaar and Rotter, (2015)).

Following the optimized methods for determination of REE in NdFeB magnets described in supporting information 1 in Ueberschaar and Rotter, (2015), the samples were prepared. Former tests with X-ray and wet-chemical analyses showed roughly the elemental content of the magnets. In further tests with sampling material, the dissolving behavior for voice-coil actuator magnets was better with aqua regia, and best results were achieved with HNO<sub>3</sub>-H<sub>2</sub>O for spindle motor magnets. This can be traced back to different production methods for the magnets and the specific dissolubility of REEs.

The developed approaches for determination of REE in NdFeB magnets in HDDs were applied separately on 35 voice-coil actuator magnets and 32 spindle motor magnets. The chemical analyses were carried out for a broad range of elements to obtain highest detection rate as possible. Following elements were measured: Metals: Al, Sn; Semi-metals: B; Transition Metals: Co, Cu, Fe, Ni, Zn; Lanthanides: Dy, Gd, Nd, Pr, Sm, and Tb.

The samples of the voice-coil actuator have been crushed and subsequently milled in an agate vibrating tube mill. The acidulation was conducted with 300mg sample material. 6,66ml ultrapure water and 3,33ml HNO<sub>3</sub> (69% Rotipuran Supra, Roth) were added. The vessels were covered and let rest overnight. Subsequently, the vessels were locked to perform a microwave-assisted dissolution (1.600W, 20bar, 200°C, 15:00min).

The samples of the spindle motor have been simply crushed without further milling processes. The acidulation was carried out with 300mg sample material and 10ml aqua regia consisting of HNO<sub>3</sub> (69% Rotipuran Supra, Roth) and HCL (37%, Merck) in ratio 1:3. The vessels were let rest for approximately 1h until no gas formation was noticeable anymore. Subsequently, the vessels were locked, and a microwave-assisted dissolution (1.600Wm 20bar, 200°C, 15:00min) was performed.

For the measurement with ICP-OES a multi-element standard in HNO<sub>3</sub>:H<sub>2</sub>O (1:2) with stated elements was prepared.

To investigate the change in compositions of applied elements in both magnets, the results of the chemical analyses were put into context to time periods. The chemical composition was split up according to the manufacturing dates of the HDDs. This approach was worked out separated for magnets out of the voice-coil motor and the spindle motor in stacked histograms. Through these methods, not only time trends but also the different blending of magnets can be derived.

### Chemical composition of PCBs

Not only the magnets contain REEs. Suspecting small quantities of these elements, also all dismantled PCBs have been investigated as a mixed sample. In total a sample mass of 1,450g was prepared to determine PMs: Ag, Au, Pd; IBMs: Al, Cu, Fe, Ni, Ti, Zn; S-CRMs: Ce, Nd, Pr, Eu, Tb, Dy, La, Sm, Gd, and Ta. The chemical analyses were carried out by a third-party laboratory as optimized approaches are already applied for PCBs. The concentration of the PMs Au and Pd was measured with ICP-OES and additionally for Au, Ag, and Pd with a Spark Optical Emission Spectroscopy (Spark-OES). All base and other metals like REE were determined with a laboratory XRF.

© (Ueberschaar and Rotter, 2015)

#### 3.4.4. Tantalum capacitors

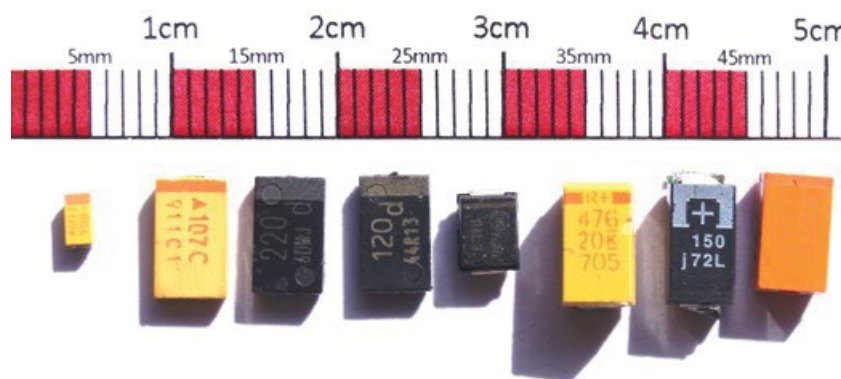
In this study, a total of over 600 WEEE components belonging to mobile phones, smartphones, tablets, notebooks, desktop PCs, HDDs, flat screen monitors, and servers were investigated with disassembly trials regarding their capacitor content. The devices were chosen according to an expected high tantalum capacitor content and a high share of new EEE put-on-market (Chancerel et al., 2013; Chancerel et al., 2015). A complete overview of a total of ten component types is supplied in Table 7.

Table 7: Overview of investigated devices and subunits with production years 1993-2014

	Mobile Phone	Smart-phone	Tablet	Notebook		Desktop PC			Flat Screen Monitor	Server
				Main PCB	Sec. PCB	HDD	Main PCB	Graphic Card		
<b>UNU key</b>	0306-01	0306-02	303-02	0303	0303	0301	0302	0302	0309	0307
<b>Number of devices disassembled in total</b>	59	26	16	63	128	99	90	56	70	14
<b>Production years</b>	1998-2011	2007-2014	2010-2015	1993-2010		1998-2008	1996-2010	1995-2012	no data available	

Note: Mobile phones comprise phones without touch screens; smartphones with touch screens. The PCB obtained from notebooks were classified into main PCB, containing the central processing unit (CPU), and secondary PCB, comprising all other contained PCB (on average a ratio of 1 : 1.1 (primary : secondary) PCB was found). HDDs comprise storage devices used in household applications covering both internal and external devices, but excluding solid state drives

The tantalum capacitors were visually identified by shape, color, and coding. A crucial characteristic for identification is the dash on the surface of the capacitor, indicating the plus and minus connectors. Example photos of the most common surface-mounted yellow and black tantalum capacitors are shown in Figure 22.



Own illustration (Ueberschaar et al., 2017c)

Figure 22: Example pictures of tantalum capacitors (typical length of 2 mm to 7 mm)

As capacitors were removed after a visual evaluation of each PCB, the term “visually identifiable tantalum capacitor” (**VIC**) for the separated target fraction will be used.

For each device, a recycling-oriented product characterization was carried out. The product masses, the number of PCBs, PCB masses, the number of VICs per PCB, and the VIC masses were determined by both weighing and counting. Furthermore, the year of production was assessed for each single device. This was done either by accessing the production date on the unit or PCB with imprints or via the product names or model number of the PCB.

The devices investigated were classified into UNU-Keys, as developed by the United Nations Universities, Institute for the Advanced Study of Sustainability (Baldé et al., 2015).

### Chemical analyses

In order to assess both element specific separation and the selectivity of VIC removal trials, the elemental compositions of the VICs and the PCBs after capacitor removal were analyzed chemically. This allowed the precision of capacitor separation to be assessed. The remaining tantalum content on the PCB and the transferred precious or other metals connected to a capacitor separation were quantified. The chemical composition of VIC and PCB was assessed using the methodologies described in the following paragraphs.

After VIC removal, the PCBs of all devices per equipment type investigated were merged into one mixed sample to obtain a maximum sample size for a wet-chemical analysis. This was also carried out for the VIC sample, which consists of all VIC taken from all equipment types. The chemical analysis took place in two different laboratories. As the sample sizes for smartphones and tablets were too small for laboratory 1, their PCBs were analyzed in laboratory 2. Supporting information S6 in Ueberschaar et al. (2016b) shows all sample sizes and the laboratory responsible for the relevant measurements.

Each PCB sample and the mixed VIC sample processed by laboratory 1 underwent a shredding process with dust capture, followed by a fire assay procedure (collection of Pb), followed by analysis with ICP-AES, ICP-MS, and XRF. In addition to determining tantalum content, 35 more elements were analyzed, such as the PMs Au, Ag, Pt, and Pd.

The two PCB samples processed by laboratory 2 were first prepared using a shredding process that generated two fractions of different grain sizes. The coarse material consisted of hard metals, which were not shreddable, and was separated prior to the milling step. It was then transferred to a liquid phase by an open aqua regia digestion. The other, finer, material was milled and processed in both a fluxing digestion after an incineration of volatile organic compounds at 550° C and a microwave-assisted digestion with aqua regia. The measurements were carried out with an ICP-OES and ICP-MS. In total, the mass fractions of 54 elements were determined.

Also, the loss of ignition for all samples was determined.

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### 3.5. Quantification of available mass flows

The potential S-CRM mass flows were calculated for put-on-market (POM) product flows, and WEEE flows through EOL devices collected for recycling purposes. Furthermore, for some other main components of the devices investigated the available mass flows were calculated as well.

The analysis of the available mass flows was calculated for both Germany and the global market in 2013 according to equation 1 and 2. Literature, own data, and data from statistical institutions have been reviewed (Cemix, 2013; Statista, 2015a; Statista, 2015b; Stiftung elektro-altgeräte register, 2015; Jalalpoor et al., 2013; Chancerel, 2010; Schischke et al., 2013).

*Equation 3: Available mass flow POM*

$$m_{j,i \text{ POM}} = \sum_{i=1}^n N_i * \overline{m}_i * \overline{s}_{i,x} * \overline{s}_{x,j}$$

*Equation 4: Available WEEE mass flow*

$$m_{j,i \text{ WEEE}} = \sum_{i=1}^n \overline{m}_i * \overline{s}_{i,x} * \overline{s}_{x,j}$$

With:

$m_{j,i \text{ POM}}$ : mass of material j (S-CRM or other elements and materials) in products i POM (kg);  $m_{j,i \text{ WEEE}}$ : mass of material j in WEEE products i (kg);  $N_i$ : number of product i POM (-);  $\overline{m}_i$ : average mass of product i (kg);  $\overline{s}_{i,x}$ : average mass fraction of S-CRM bearing component x in product i (%);  $\overline{s}_{x,j}$ : average mass fraction of material j in component x (%).

The calculation was carried out for LCD panels, HDDs, and tantalum capacitors. Mass potentials for ICs were considered separately in chapter 3.3.1 “MFA gallium life-cycle”.

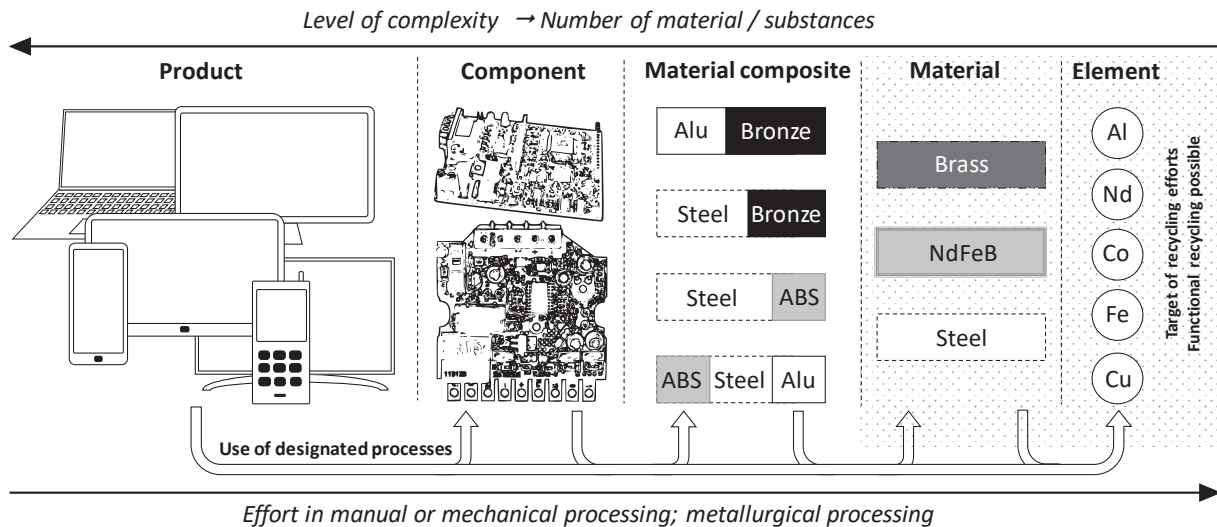
### 3.6. Time trend analysis

In order to identify product trends regarding the use of elements investigated, the dates of manufacture of all devices investigated were evaluated and compared with the individual mass fraction of the elements determined.

Time trend analyses were carried out for LCD panels, HDDs, and tantalum capacitors. For ICs, no time trend analysis was possible due to identification problems of ICs on PCBs.

### 3.7. Recyclability assessment

Recyclability assessment is applicable on single products, assembled components, material composites, single material, and on mixtures of those, or processed materials. Figure 23 shows these single levels as a recycling chain of one WEEE product. The levels ‘material’ and ‘element’ represent the basis for a functional recycling and are generally the target of any recycling efforts.



Own illustration

Figure 23: Levels of processing single WEEE products on which a recyclability assessment is applicable

The S-CRM hotspot analyses revealed S-CRM hotspots on component and material levels. The conducted recycling-oriented product characterization provides fundamental information about the product design and its physical and chemical properties. This information allows for the development of recycling processes to reduce the complexity of WEEE products and to transfer WEEE products or components to a material or element level. In this thesis, the recyclability is assessed via theoretical evaluations and practical separation tests to liberate and concentrate target materials in the form of single recycling processes (cf. Table 8).

Table 8: Materials and components assessed in recyclability assessment

Starting material / component		Target S-CRMs	Target IBMs and PMs and other materials	Methodology
Processed materials	Output materials from WEEE pre-processing	Co, Ga, In, REE, Ta	Al, Cu, Fe, Au, Ag, Pd	Theoretical assessment of recyclability in state-of-the-art end-processing steps
	Integrated circuits	Ga	Au, Ag, Cu	Thermal treatment and sorting of output fractions
WEEE components	LCD Panels	In	Glass substrate, polarizer foils	Chemical treatment of LCD panels
	Tantalum capacitors	Ta	Ag	Manual separation of tantalum capacitors from PCBs

### 3.7.1. Output materials from WEEE pre-processing

Based on the recycling processes of WEEE pre-processing, metals are dispersed to various output fractions. Here, a big number of different elements with varying mass fractions can be present. According to the recycling strategy target materials, output fractions are guided to different actors in the recycling chain to recover valuable materials as well as to separate contaminants. Depending on the chosen route, single materials are recovered or are not recovered due to technical or economic barriers.

Based on the results of the conducted extended batch test (cf. 3.3.2 “Extended batch test in WEEE pre-processing”) and literature research on mechanical and pyro- or hydrometallurgical processes, an estimation of the recyclability of target metals in state-of-the-art processes is carried out. For this purpose, the used recycling processes for the output materials from WEEE pre-processing will be traced back.

Appendix 5 compares the most common state-of-the-art end-processing steps, which are used for the further concentration and purification of IBMs, PMs, and S-CRMs in WEEE pre-processing output fractions. Target and conflict materials, which hinder the recycling of S-CRMs in end-processing steps, are investigated.

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### 3.7.2. Integrated circuits

Manual, mechanical, thermal, and chemical treatment of ICs was tested to separate single components and to concentrate gallium into a separate output. After pre-tests, the approach of thermal treatment was further investigated. The samples were heated at 250° C for 110 min and again at 550° C for 360 min in a muffle furnace. With this procedure, the plastics casings were burned and the inboard components were liberated. The thermal exposure revealed single modules, which were clearly separable. For a subsequent chemical analysis, the liberated components have been sorted according to visually identifiable characteristics like form or color. Four clusters were identified:

1. Isolation with copper connectors
2. Metal plates (back of the chip)
3. Electronic modules (chip)
4. Others

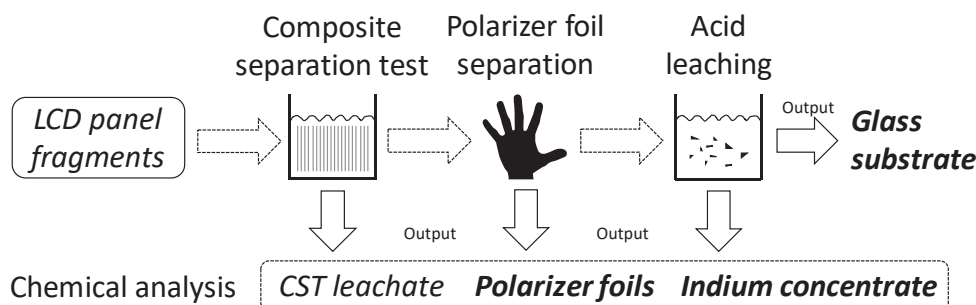
All fractions were weighted and chemically analyzed with an X-Ray fluorescence analysis (XRF) (Thermo Fisher/ Analyticon XL3 Air) to determine the chemical composition. The samples were measured several times from various angles without any further processing to avoid losses related to milling and decanting. Through this, the flows of Cu, Ga, As, Au and Ag into the four output fractions through thermal treatment and visual sorting was examined. Assuming a processing of the chips 1 – 3 in the same ratio, an MFA assessed.

The approach of thermal treatment and visual sorting was also used for the S-CRM hotspot analysis of ICs. Chapter 3.4.1 “Integrated circuits” provides information in more detail.

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## 3.7.3. LCD panels

The tested recycling process approach is carried out in two separate steps (cf. Figure 24 ). The first step is the composite separation test (CST) in which the polarizer foils are removed from the glass substrate. In the second step, the ITO is separated from the glass substrate. The recycling approach was exemplarily carried out on various LCD panel samples from two different equipment types: smartphones and tablets. For tablets, the sample mass was 8 g and for smartphones 10 g. All samples were cut into pieces of between 2-4 cm<sup>2</sup>.



Own illustration (Ueberschaar et al., 2017d)

Figure 24: Methodological sequence of new recycling step for LCD panels on laboratory scale

#### Composite separation / polarizer foils separation

For the separation of polarizer foils, the samples were treated with a water / base solution with 10 vol.-% NaOH over 72 hours at room temperature. This treatment caused a separation of polarizer foils from the glass substrate by interfacial unbinding without digesting the polymer. At the end of the reaction time, the polarizer foils could be separated easily. Residues represented the glass substrate with the applied ITO layer.

To stop any reaction of the base with the plastics and to prevent any indium transfers, the polarizer foils were subsequently washed with an aliquot volume of distilled water. The leachate from the NaOH solution and the aliquot washing water were subsequently combined, weighed and prepared for chemical analysis. The same procedure was carried out for the polarizer foils, which were dried beforehand.

#### Acid leaching / ITO separation

For the separation of the ITO from the glass residues, the same methodologies were used as described for the chemical analysis of the whole LCD panels. This methodology consists of a microwave assisted (CEM Ultra-High Throughput Microwave Digestion System MarsXpress) leaching of the indium from the glass surface with 10 ml nitric acid (HNO<sub>3</sub>, 65%, Rotipuran Supra; Fa. Roth) at 180 °C, maximal pressure 20 bar, 20 min preheating time, 20 min retention time at 180 °C and 20 min cooling time. Three parallel measurements per sample were carried out.

#### Chemical analysis of output fractions

In total, four output fractions were generated in the composite separation test of the new recycling approach: 1. foils fraction, 2. foil leachate residues, 3. ITO + liquid crystals (organics) and 4. a cleaned glass fraction. To evaluate the efficiency of the process and to investigate the detailed flows of In, Sn, Sr and the toxic heavy metals As, Cr, Sb, Pb, chemical analyses of the output fractions were subsequently carried out. The remaining glass fraction is ITO free and is not investigated in detail any further.

For the foils fraction, a methodology for an optimized determination of the target elements with an ICP-OES had to be developed. To transfer the elements of interest to the liquid phase, three different

acid digestions were tested with the foil fraction samples from both smartphones and tablets. The following standard leaching agents were chosen: hydrochloric acid (HCl), nitric acid (HNO<sub>3</sub>) and aqua regia (volume ratio conc. HNO<sub>3</sub> to conc. HCl 1:3).

The foil leachate already represents a liquid, which can be chemically analyzed directly.

☞ (Ueberschaar et al., 2017d)

#### 3.7.4. Tantalum in Ta capacitors

Tantalum capacitors have optical indicators, which were used for a visual identification and manual separation (cf. 3.4.4 “Tantalum capacitors”). Over 600 PCBs originating from mobile phones, smartphones, tablets, notebooks, desktop PCs, HDDs, flat screen monitors, and servers were processed with this approach. The efficiency and selectivity of this process were assessed via the calculation of transfer coefficients and the selectivity of this manual recycling approach (Ueberschaar et al., 2017a).

Following this analysis, the total tantalum content of the PCBs and the tantalum content, which is potentially recoverable via a removal of the tantalum capacitors, is calculated using the average amount of tantalum capacitors for each device and the average tantalum content.

☞ (Ueberschaar et al., 2017a)

### 3.8. Identification of recycling barriers and drivers

Potential recycling barriers and drivers for the recycling of targeted S-CRMs in WEEE products were assessed via the consolidation of all generated product-centric information and further stakeholder interviews and workshops.

Boundary conditions regarding product related properties, available and used technology for liberation, separation, and end-processing of the material as well as economic parameters were considered. Table 9 shows an overview of all investigated issues.

Table 9: Basis elements for a barrier analysis to be investigated

Product related analysis	Product design			Material composition		
	Change in product design	Available product information or labeling	Dissipation through product design	Recycling conflict with other valuable or relevant materials	Complexity of material composition	Recycling conflicts with contaminants
Recycling chain related analysis	Collection of end-of-life devices					
	Collection rates of relevant devices			Trends in collection rates		
	Pre-processing			End-processing		
	Mechanical liberation of target materials	Availability of identification and sorting technology	Complexity of metallurgical liberation	Separability after liberation		End-refining channels available
Economy related analysis	Economy					
	Financial drivers for the recovery of target material			Energy consumption and waste water/residues production		

Results were categorized in a modified SWOT (strengths, weaknesses, opportunities, and threats) matrix (cf. Table 10). While a normal SWOT analysis differentiates between internal and external impacts, a consistent classification of recycling barriers and drivers to the SWOT groups is not possible. Barriers to S-CRM recycling may give rise to opportunities and have a positive environmental or economic impact and drivers may lead to unwanted side effects.

Table 10: Modified SWOT analysis matrix

	Strengths	Weaknesses
Opportunities	Drivers for the recovery of applied S-CRMs in components investigated.	Weaknesses identified for the recovery of S-CRMs but may be canceled out via technical progress.
Threats	Drivers or reasons for a lower relevance of S-CRMs recycling. These aspects may impede the recovery of the applied S-CRMs in components investigated.	Aspects impede the recycling of the component investigated carrying S-CRMs or the recovery of applied S-CRMs.

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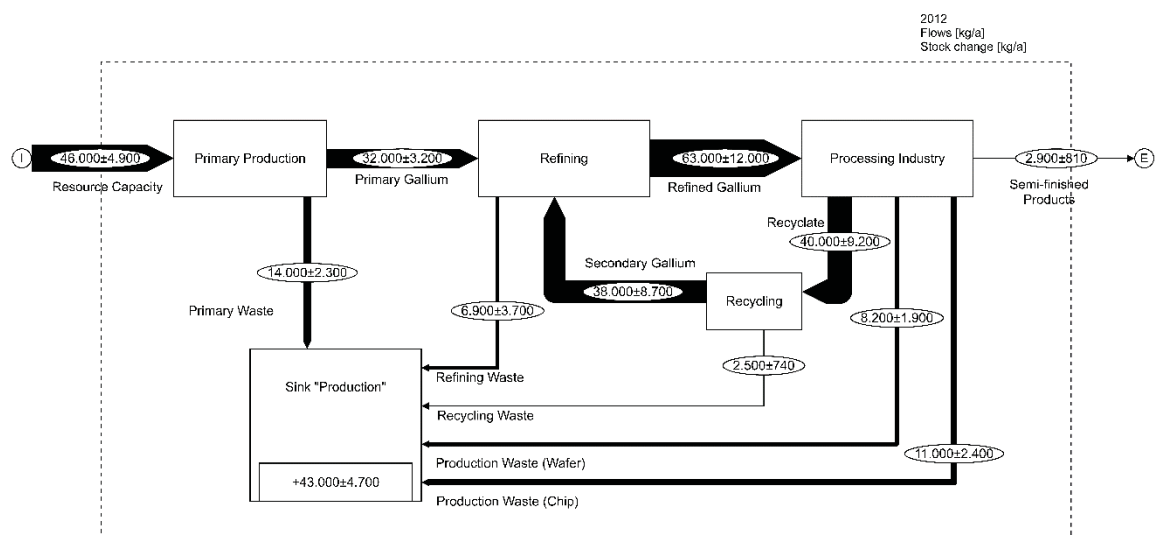
## 4. Results

### 4.1. S-CRM hotspot analysis via material flow analysis

#### 4.1.1. MFA gallium life-cycle

##### Material flows in system "production"

This model consists of five processes, while one process represents the losses occurring during the primary production of gallium and the further processing. In 2012,  $32,000 \pm 3,200$  kg of gallium was extracted as a by-product of the aluminum production (cf. Figure 25). Only 70 % of the gallium potential is extracted from the lye in the Bayer process. Through this,  $14,000 \pm 2,300$  kg gallium remain in the red mud and is subsequently landfilled. This represents the highest loss in the system 'production' (cf. supporting information S2.2.1 in Ueberschaar et al. (2016c)).



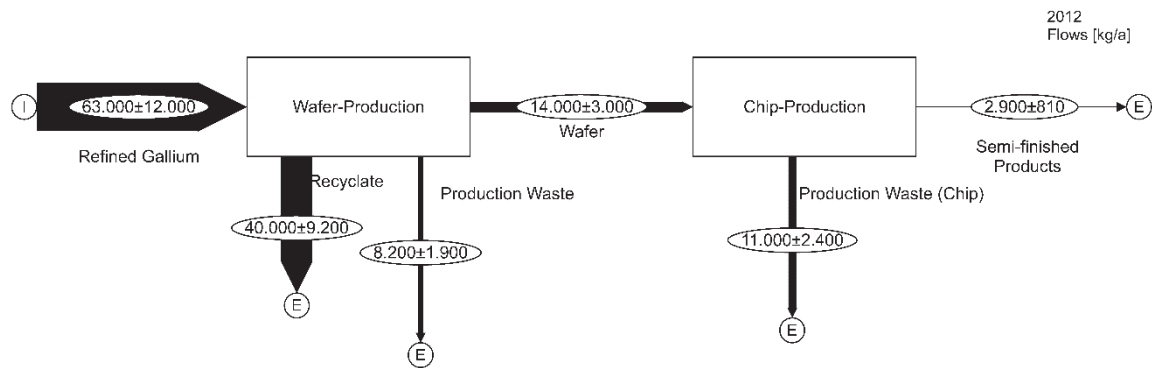
Own illustration (Ueberschaar et al., 2017c)

Figure 25: Quantitative material flow model of the system "production" for gallium

To concentrate and to obtain required grades, the gallium is subsequently refined (cf. supporting information S2.2.2 in Ueberschaar et al. (2016c)). Here, the refining losses are much smaller with  $6,900 \pm 3,700$  kg. From the refining step, about  $63,000 \pm 12,000$  kg of gallium is transferred to the processing industry. This increased amount of gallium is subjected to recycled new scrap, which arises mainly in the wafer production. This material can be recycled and is led back to the refining process. This saves  $38,000 \pm 8,700$  kg of the containing gallium. Here, only 3 %, i.e.  $2,500 \pm 740$  kg is lost during the recycling. This is the lowest quantity of lost gallium in the system "production".

Figure 26 shows the subsystem "processing industry" (cf. supporting information S2.2.3 in Ueberschaar et al. (2016c)). Two of the four output flows depict losses related to the wafer production (cf. supporting information S2.2.4 in Ueberschaar et al. (2016c)) and to the chip production (cf. supporting information S2.2.5 in Ueberschaar et al. (2016c)). In the chip production process, the substrate thickness must be reduced or partially removed. Here, a highly concentrated gallium material is lost. This flow accounts to  $11,000 \pm 2,400$  kg of a total of  $14,000 \pm 3,000$  kg transferred into this process. Herewith, the losses in the chip production are comparable to the losses in the primary production of gallium.

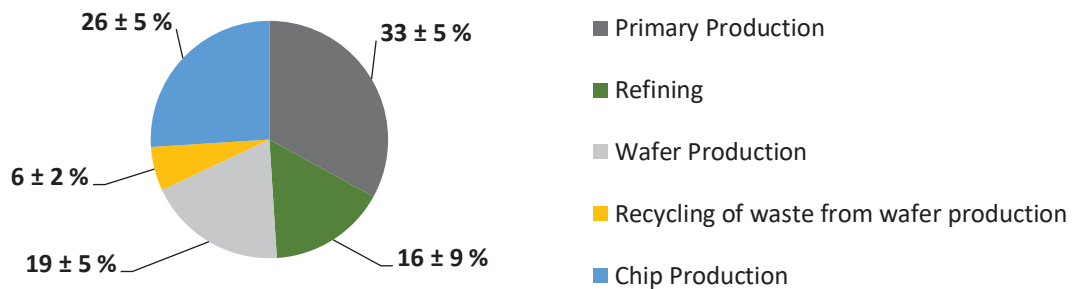
Although a high percentage of the production waste is recycled after the wafer production, about  $8,200 \pm 1,900$  kg gallium is not recovered. In total, only 8 % of the refined gallium is applied in EEE products in the form of chips.



Own illustration (Ueberschaar et al., 2017c)

Figure 26: Quantitative material flow model of the sub-system "processing industry"

In 2012, the system "production" had a total loss of  $43,000 \pm 4,700$  kg, which is 93 % of the total gallium input. These flows are supposed to be transferred to e.g. landfills as red mud representing a stock. As stock transfers prior to 2012 are unknown, this study shows only the change in stock. For a direct comparison, Figure 27 shows the relative values.



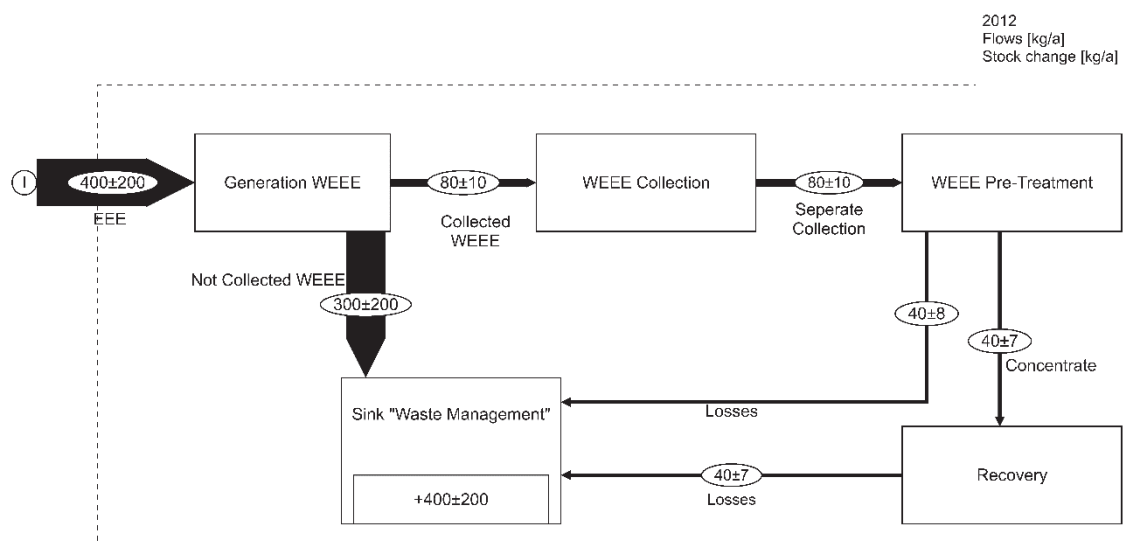
Own illustration (Ueberschaar et al., 2017c)

Figure 27: Relative shares of all gallium losses in the system "production"

The highest losses with  $33 \pm 5$  % occur during the primary production of gallium, followed by the chip production with  $26 \pm 5$  % and wafer production with  $19 \pm 5$  %. Only low losses are assigned to the refining process ( $16 \pm 9$  %) and the recycling of the new scrap ( $6 \pm 2$  %).

#### Material flows in system "waste management"

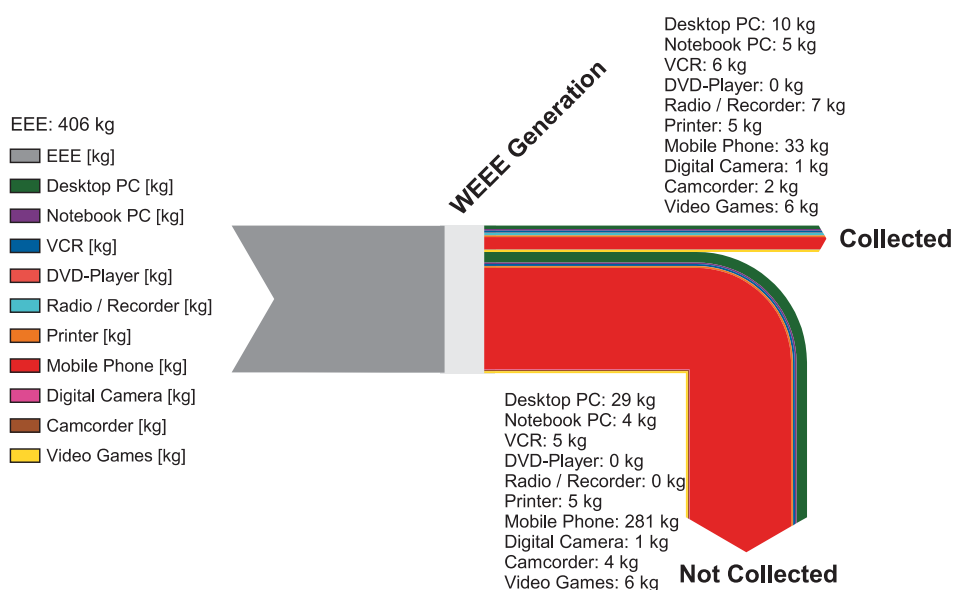
Major losses in the system "waste management" base on an insufficient collection of arising WEEE (cf. Figure 28). Not collected WEEE is assigned to losses over the residual waste collection, at home stored end-of-life devices and a transfer of WEEE into the informal sector (cf. supporting information S2.3.1 in Ueberschaar et al. (2016c)).



Own illustration (Ueberschaar et al., 2017c)

Figure 28: Quantitative material flow model of the system "waste management"

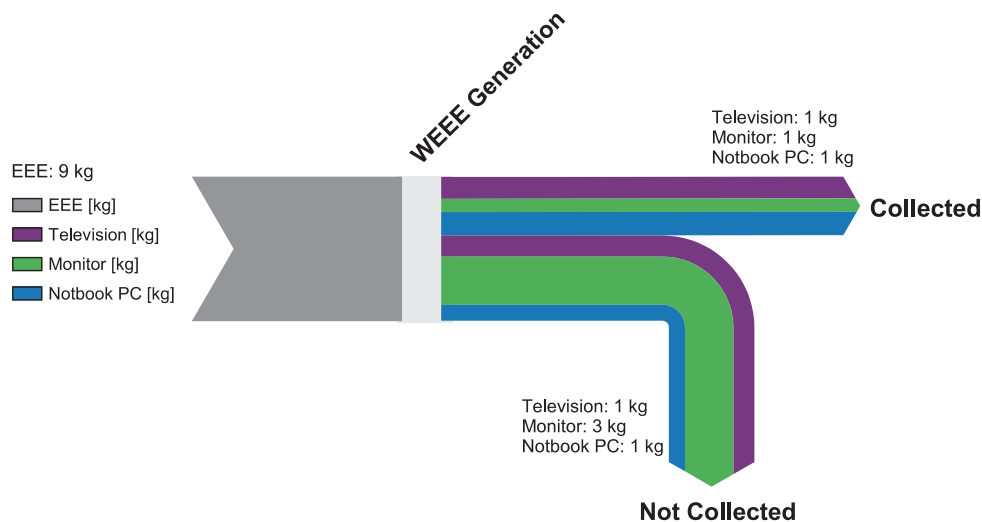
All flows in this system were determined individually for devices carrying PCBs and LEDs. Figure 29 depicts the gallium flows related to PCBs in equipment types investigated. In Germany in 2012, over 400 kg gallium arose with the generation of WEEE. About 340 kg of this potential was not collected. Mobile phones represent the major share with over 280 kg. Only almost 80 kg gallium was collected through the formal collection of end-of-life devices.



Own illustration (Ueberschaar et al., 2017c)

Figure 29: Collected and not collected gallium amounts with PCBs in WEEE

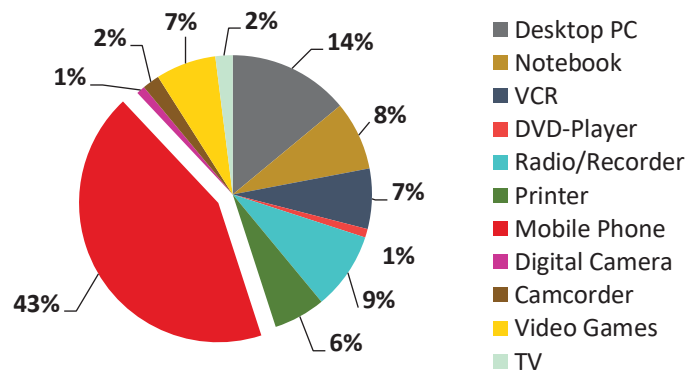
Figure 30 depicts the gallium flows related to WEEE LEDs. Here, a total of almost 10 kg arose with WEEE in one year. The losses range from 1.3 kg to 2.5 kg gallium.



Own illustration (Ueberschaar et al., 2017c)

Figure 30: Collected and not collected gallium amounts with LEDs used for background lighting in WEEE

The total amount of collected PCB and LED related gallium sums up to almost 80 kg. Figure 31 shows the distribution according to the equipment types investigated. Mobile phones clearly account the major share with 43 % of the total gallium collected with WEEE, followed by desktop PCs (14 %). All other equipment types have a share less than 10 % each.

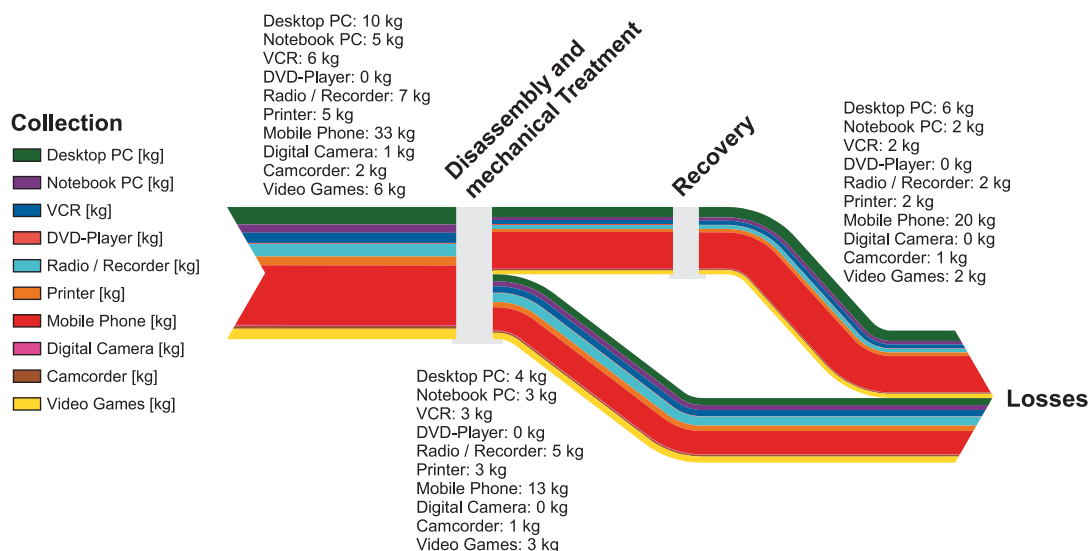


Own illustration (Ueberschaar et al., 2017c)

Figure 31: Share of gallium related equipment types in collected WEEE

Figure 32 and Figure 33 show the gallium flows for relevant equipment types carrying PCBs and LEDs within the treatment of WEEE (cf. supporting information S2.3.2, S2.3.3, and S2.3.4 in Ueberschaar et al. (2016c)). Here, Figure 32 shows the flows of collected gallium from PCBs, Figure 33 for gallium from LEDs for background lighting. After the collection step, all material is directed to a manual pre-treatment, followed by mechanical processing. Here, losses relate to the incorrect sorting of PCBs and general dust discharges. After pre-treatment, recyclates are further processed with metallurgical approaches to recover single metals. Used pyrometallurgical processes are not optimized for gallium recovery. Thus, gallium is subsequently lost. So far, only one industrial scale process for the recovery of gallium exist. Operated by Umicore, only CIGS solar modules are being recycled.

Figure 32 shows, that highest gallium losses related to PCBs originate from mobile phones with 13 kg. All other equipment investigated types account for 0.3 kg to a maximum of 5 kg lost gallium. Since no gallium in WEEE is recycled the sum of the losses equals to the input into the system “waste management”.

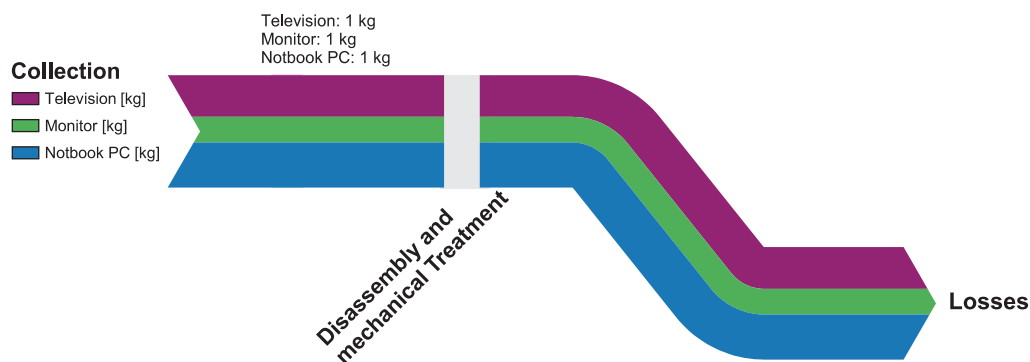


Own illustration (Ueberschaar et al., 2017c)

Figure 32: Flows of collected gallium from PCB in the treatment of WEEE

In total,  $40 \pm 8$  kg gallium is lost during the manual disassembly and the mechanical treatment. This is almost the same amount, which is lost during the metallurgical processes.

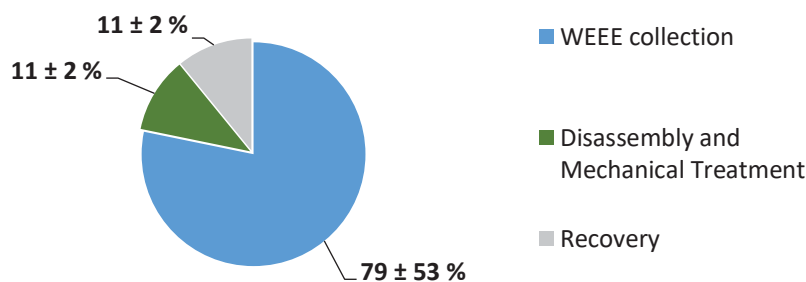
While PCBs are separated to be processed individually in subsequent processes, this does not apply for LEDs, which leads to complete loss of gallium after the disassembly and mechanical treatment (cf. Figure 33). Thus, losses related to LEDs equal to the input amount of 3.7 kg gallium.



Own illustration (Ueberschaar et al., 2017c)

Figure 33: Flows of collected gallium from LED in the treatment of WEEE

The cumulated gallium losses in the system “waste management” related to LEDs and PCBs sum up to  $400 \pm 200$  kg. Figure 34 shows the individual shares. Here, WEEE collection relates to almost 80 % of the total losses. WEEE treatment holds lower shares, although no gallium is recovered here.



Own illustration (Ueberschaar et al., 2017c)

Figure 34: Relative proportions of the losses related to the system “waste management”

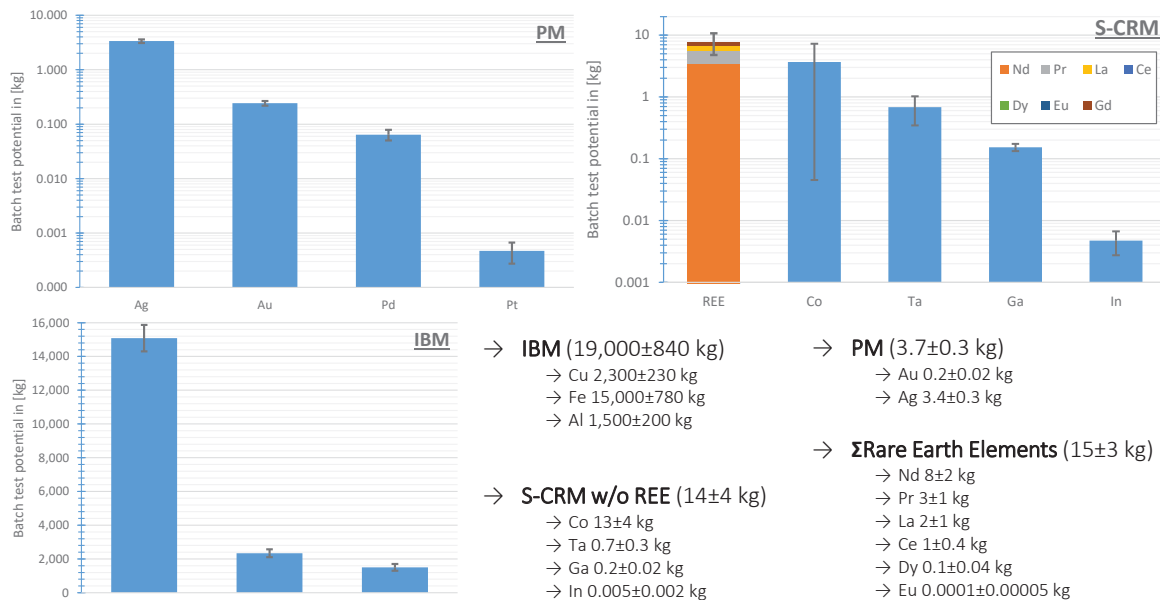
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#### 4.1.2. Extended batch test in WEEE pre-processing

##### Fate of materials and elements in extended batch test

##### *Calculated element input*

Figure 35 shows the element input calculated in the extended batch test with a total mass of 42,860 kg. The IBMs represented the highest share of metals, in particular iron, followed by copper and aluminum. PMs and S-CRM constituted only a small quantity in the extended batch test. About 4 kg of PMs, mostly silver, were processed. The S-CRM were dominated by REE and cobalt.

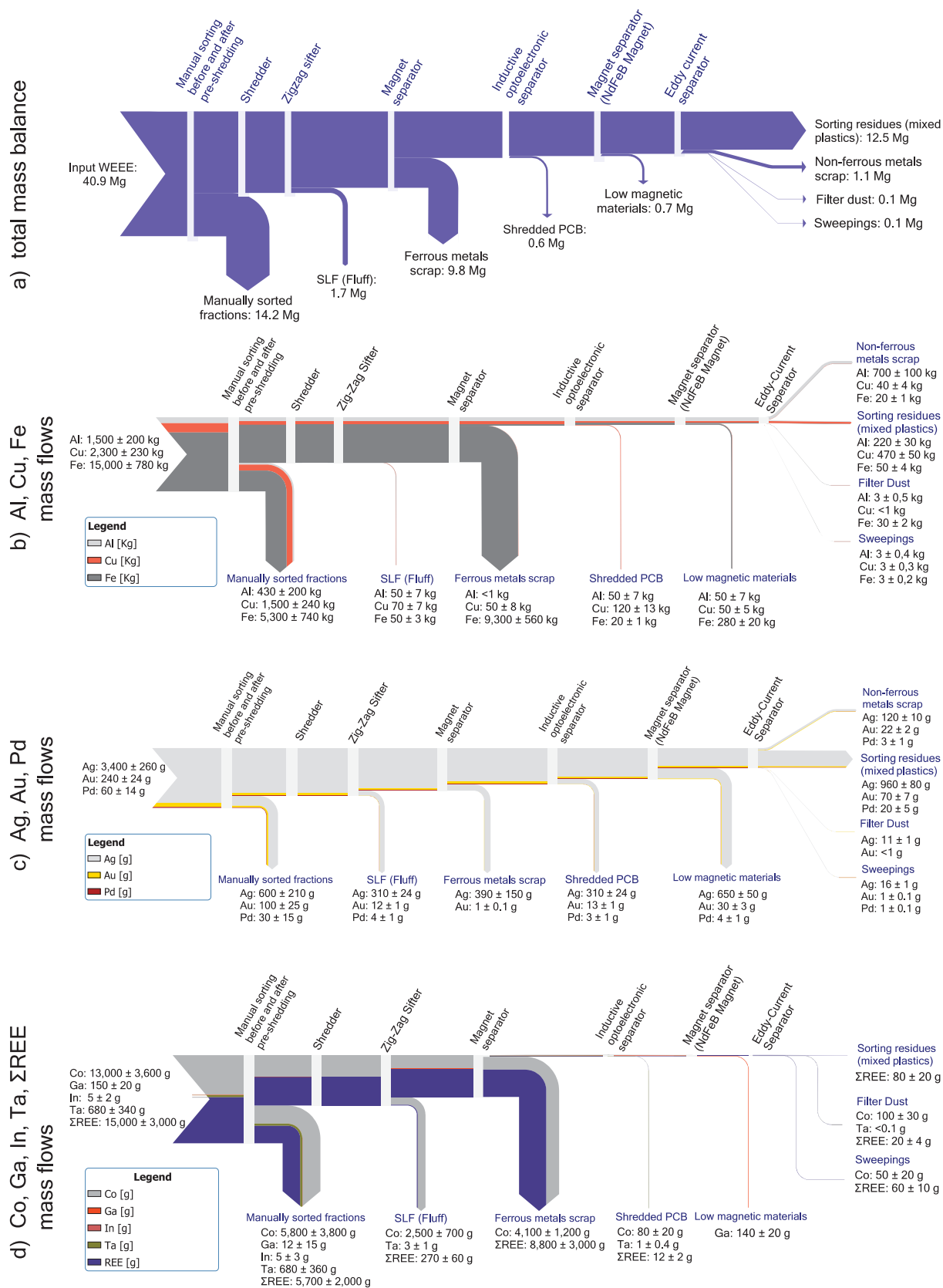


Own illustration (Ueberschaar et al., 2017b)

Figure 35: Calculated material input quantities for IBMs, PMs, and S-CRMs in extended batch test (errors bars based on all assessed uncertainties)

##### *Distribution of metals in output fractions*

With the validated data, not only the general mass flows in the extended batch test, but also the flows of the elements studied, along with corresponding uncertainties, can be presented. The calculated transfer coefficients are a highly important tool for the evaluation of the generated data. Supporting information S11 in Ueberschaar et al. (2016a) shows the data in detail. The aggregated data on all mass flows is shown in Figure 36. The elemental material flow analysis depicts the elemental distribution from input material to output fractions. The schematic presentation is structured by IBM, PM and S-CRM. The REE are depicted as a cumulated mass ΣREE.



Own illustration (Ueberschaar et al., 2017b)

Figure 36: Mass balance and mass flows in extended batch test a) total mass, b) aluminum, copper, and iron c) silver, gold, palladium and platinum, d) cobalt, gallium, indium, tantalum and ΣREE

More than one-third of the material was separated manually prior to mechanical processing. These manual sorting fractions consisted of whole devices like IT devices MS8 (smartphones, tablets, notebooks, etc.) or tools MS1 and single components like drivers from loudspeakers MS3, single batteries MS7 or power supplies MS4-2. The other two main quantities were ferrous metals scrap AS1 and the sorting residues AS5, which consisted mostly of plastics.

About 47% of aluminum ended up in the non-ferrous scrap AS3, up to 15% in the sorting residues AS5 and 17% in the manually sorted IT devices MS8. Most of the copper was separated manually prior to mechanical processing, but 20% still ended in the sorting residues AS5. Due to the very effective magnetic separator, 45% of iron was removed in manual separation, and 65% from the ferrous metals scrap AS1.

PMs like silver were distributed to all output fractions. Most of the significant loads of gold and Pd were separated during the pre-sorting but were distributed to almost all output fractions in the automated sorting. Highest loads were found in manually sorted materials, but also in low magnetic materials AS2 and in particular in the sorting residues AS5.

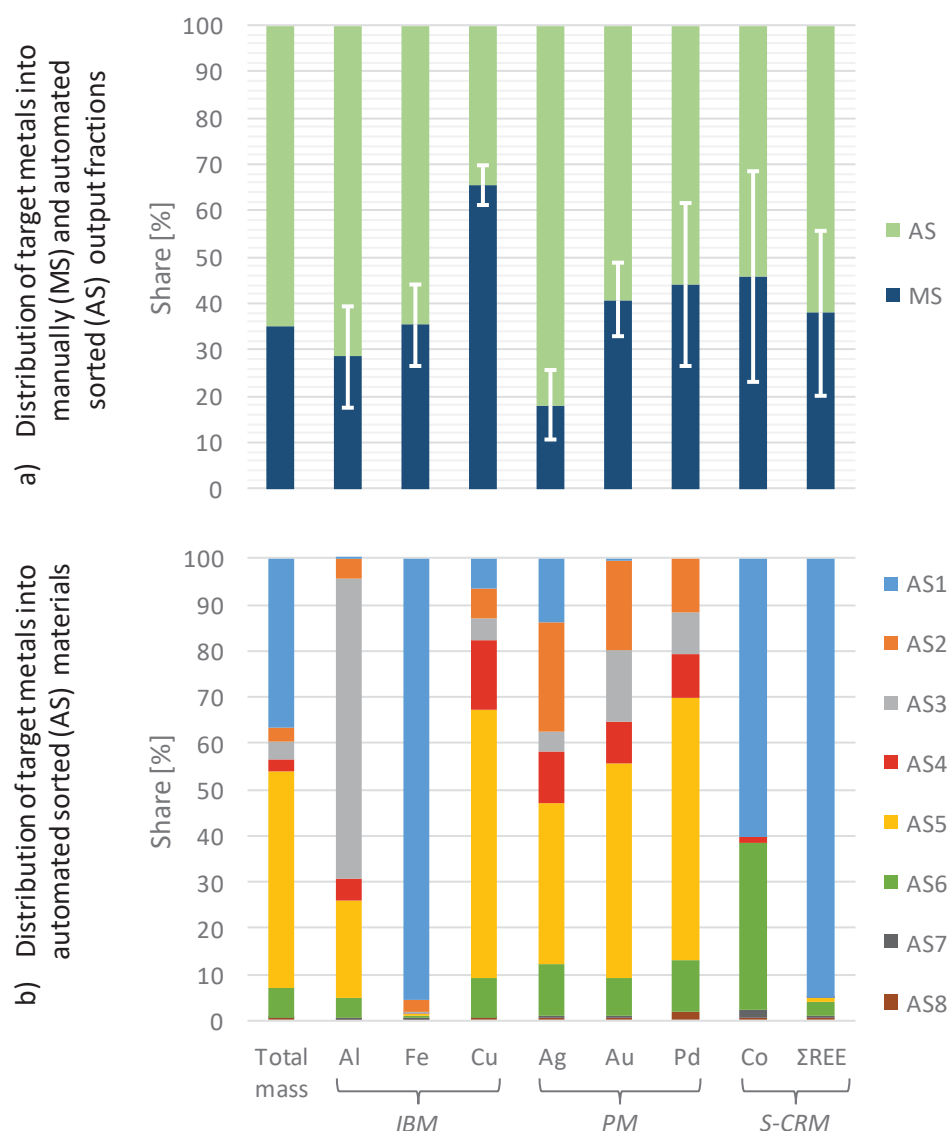
$\Sigma$ REE and Cobalt represented the highest share of the S-CRMs. Most of it was concentrated in the pre-sorting step ( $\Sigma$ REE 38%, Co 46%) due to battery removal (MS7). SLF (fluff) AS6 also contained a large share of Co, at 20%. Over 30% of Co and about 60% of  $\Sigma$ REE was enriched in the ferrous metals scrap AS1.

Figure 37 shows the general splitting of the target metals into manually sorted materials prior to the mechanical processing and materials from the automated sorting, including the corresponding uncertainties. As described earlier, no uncertainties were assessed for the total mass. These distribution figures relate to the mass fractions in the output investigated and to the transported masses. The mass fractions for the manually sorted materials are shown in the supporting information S7 in Ueberschaar et al. (2016a), while the mass fractions for the automated sorted output fractions are depicted in the supporting information S8 in Ueberschaar et al. (2016a).

Except for copper, all elements investigated were mostly concentrated in the automated sorting materials. The main reason for this was the high mass fraction of about two-thirds that went to the mechanical processing but was also due to the high mass fractions of elements in one of the output fractions.

Depending on the aims of the sorting process, the agglomeration of compatible elements in output fractions is to be desired, but the presence of some other elements can have a contaminating effect on the material. For example, iron was concentrated in the ferrous metals scrap but was related to high loads of Co and  $\Sigma$ REE.

Also noticeable was the high transfer of PMs as well as copper and aluminum to the sorting residues AS5, which consisted mostly of mixed plastics. As in the case of ferrous metal scrap AS1, these sorting residues were large in quantity and contained a high load of target elements even with low determined mass fractions.



Own illustration (Ueberschaar et al., 2017b)

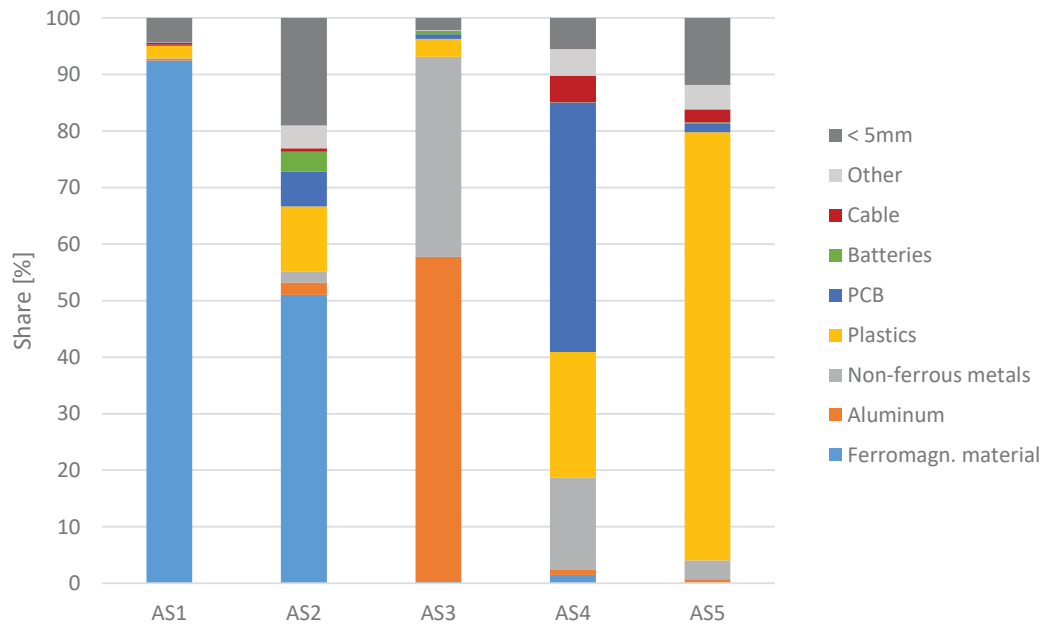
Figure 37: Splitting of IBMs, PMs, and S-CRMs through manual and automated sorting and distribution in output fractions of automated sorting processes

### Quality assessment of output fractions from pre-processing

#### Sorting analysis of mechanically processed materials

The two plant output fractions ferrous metals scrap (AS1) and non-ferrous scrap (AS3) showed a purity above 92-93% of designated materials, while other output fractions had much higher contaminations from undesigned materials. In particular, in shredded PCBs (AS4) contaminations existed in all grain sizes.

Carriers of target metals like PCBs for PM, tantalum or gallium or batteries for cobalt and REE were found in various output fractions. PCB pieces were dispersed amongst almost all plant output fractions, despite the fact that AS4 and AS2 (low magnetic materials) were the intended routes to ensure the recovery of PMs. Also, 1.5% of the sorting residues AS5 and about 0.8% of AS3 also consisted of these materials. Batteries accounted for a mass fraction of 3.5% in AS2 (see Figure 38).



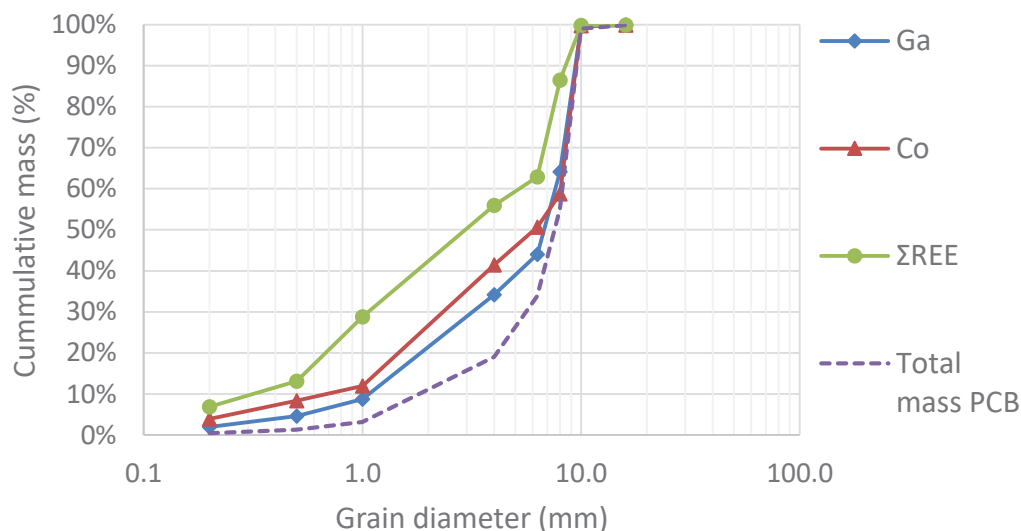
Own illustration (Ueberschaar et al., 2017b)

Figure 38: Composition of recyclates from mechanical processing based on sorting analysis

#### Sieve analysis with determination of chemical composition

Results show that the mass fractions of S-CRMs and PMs were partially higher in smaller grain sizes. However, the overall share of fines in output fractions from the automated sorting was usually low, with an average of 8.5% for the samples AS1-5. Exceptions were filter dust and SLF (fluff). IBMs were distributed more evenly in the single sieve ranges. For example, copper was usually evenly concentrated in larger screen sizes. In contrast, the S-CRMs were located more in the smaller grain sizes. In general, 30-50% of the S-CRM and up to 90% of the PMs enrich in grain sizes below 5 mm. Figure 39

shows the distribution of Ga, Co and  $\Sigma$ REE in the output fraction AS4.



Own illustration (Ueberschaar et al., 2017b)

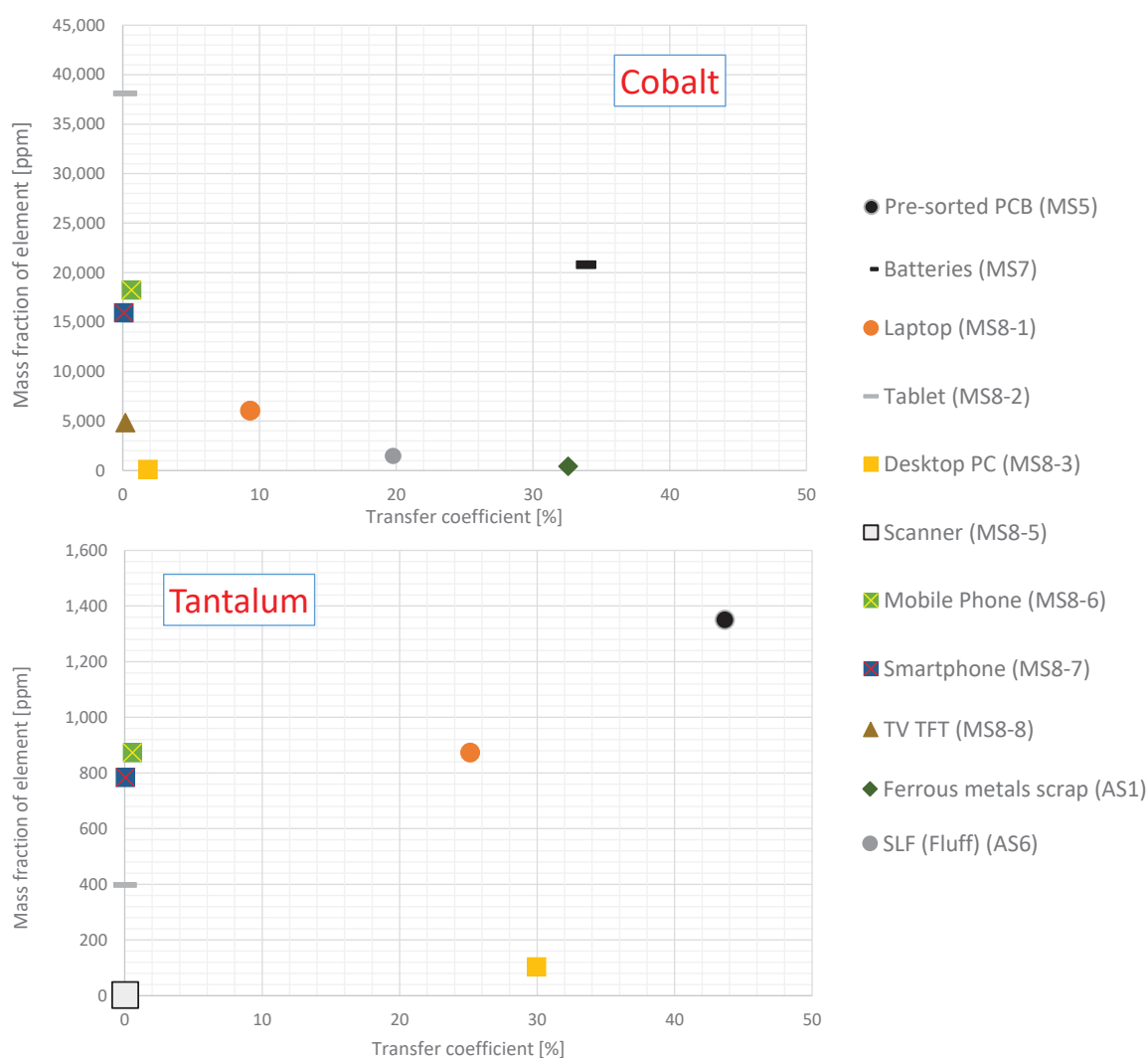
Figure 39: Example of sieve analysis of shredded PCBs (AS4) and cumulative masses of Ga, Co, and  $\Sigma$ REE in screening steps

Regarding IBMs, PMs, and S-CRMs, the results for all other output fractions studied are depicted in the supporting information S11 in Ueberschaar et al. (2016a). Supporting information S10 in Ueberschaar et al. (2016a) shows the results of the sieve analysis.

### Identification of hotspots for the recovery of target metals

As shown, target elements were scattered to various output fractions. To assess the potential to separate and recover them, plant output fractions were given an element specific assessment based on their “grade” and the “transfer coefficient” plotted in a hotspot diagram (cf. supporting information S7, S8, and S12 in Ueberschaar et al. (2016a)).

Figure 40 shows both indicators for cobalt and tantalum. Cobalt was mostly found in magnet materials and batteries, which are applied in various mobile devices. These were manually sorted prior to the mechanical processing. Also, a single battery fraction (MS7) was also separated, representing the major load of cobalt, with almost 35% of total cobalt input and a high mass fraction of over 20,000 ppm. Ferrous metals scrap AS1 was found to hold over 30% of the cobalt load, but with only a minor mass fraction.



Own illustration (Ueberschaar et al., 2017b)

Figure 40: Hotspot plot for cobalt and tantalum in the output fractions with the highest mass fractions

Most of the tantalum probably derived from alloys and tantalum capacitors used in PCBs. Relevant output materials for tantalum were mainly located in the pre-sorted materials. The highest share of over 40% of the total tantalum loads, with a grade of nearly 1,400 ppm, was accounted for by pre-sorted PCBs (MS5). This might provide a useful basis for the recovery of tantalum in the future.

In addition to this output, laptops and desktop PCs also held a large share of the tantalum loads, with between 25 and 30%. However, the total weight of the laptops and PCs reduced the relevant mass fractions. This would appear to be another useful source, but a subsequent separation of the PCB from

the device is necessary. This process did not take place in the plant investigated and is not a part of this study. Other devices like mobile phones, smartphones, and tablets have high mass fractions but were related to low transferred masses due to low collection rates.

Figure 41 shows the hotspot diagram of all REEs investigated as a sum and indium. Almost 60% of the total REE were concentrated in the ferrous metals scrap (AS1). This suggests that this output is the most promising regarding the recovery of REEs. However, the mass fraction was very low. In contrast, about 30% of the  $\Sigma$ REE was concentrated in the batteries MS7 in the manual sorting step. Here, the mass fractions were much higher. Based on this, batteries would appear to be a significantly better source for the recovery of REEs, whereby the set of applied REEs differs for NdFeB magnets, batteries or even lighting products (Ueberschaar and Rotter, 2015; Sommer et al., 2015; Rotter et al., 2016). All other battery containing materials like mobile phones, smartphones and tablets had only low concentrations and held only a small percentage of the total amount of  $\Sigma$ REE.

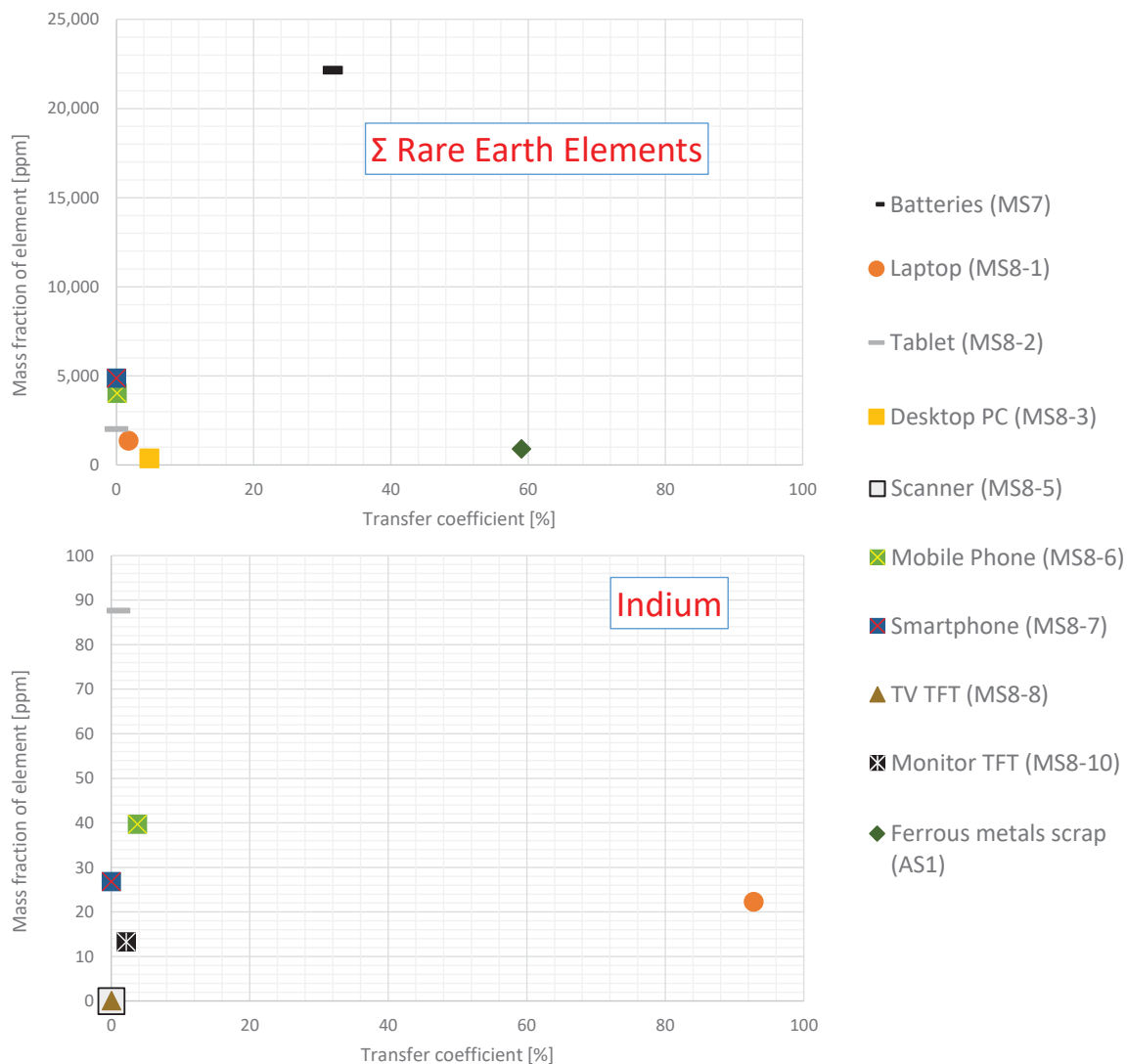


Figure 41: Hotspot plot for  $\Sigma$ REE and indium in the output fractions with the highest mass fractions

Indium is applied mostly in display devices and was only detected in the manually sorted materials. The various indium mass fractions in the flat screen / TFT displays and the screen dimensions directly influence the indium loads. The highest mass fractions of indium were found in tablets (MS8-2), mobile phones (MS8-6) and smartphones (MS8-7). Due to the low collection rates of these devices and, more

specifically, to the low amounts in this batch, laptops contributed the highest indium loads. The impact of monitors and TVs was limited.

The distribution figures for PMs (gold, silver), copper and the S-CRM gallium are shown in the supporting information S14 in Ueberschaar et al. (2016a). Iron and aluminum were dispersed among all output fractions investigated. A presentation in the same form would not be feasible.

© (Ueberschaar et al., 2017b)

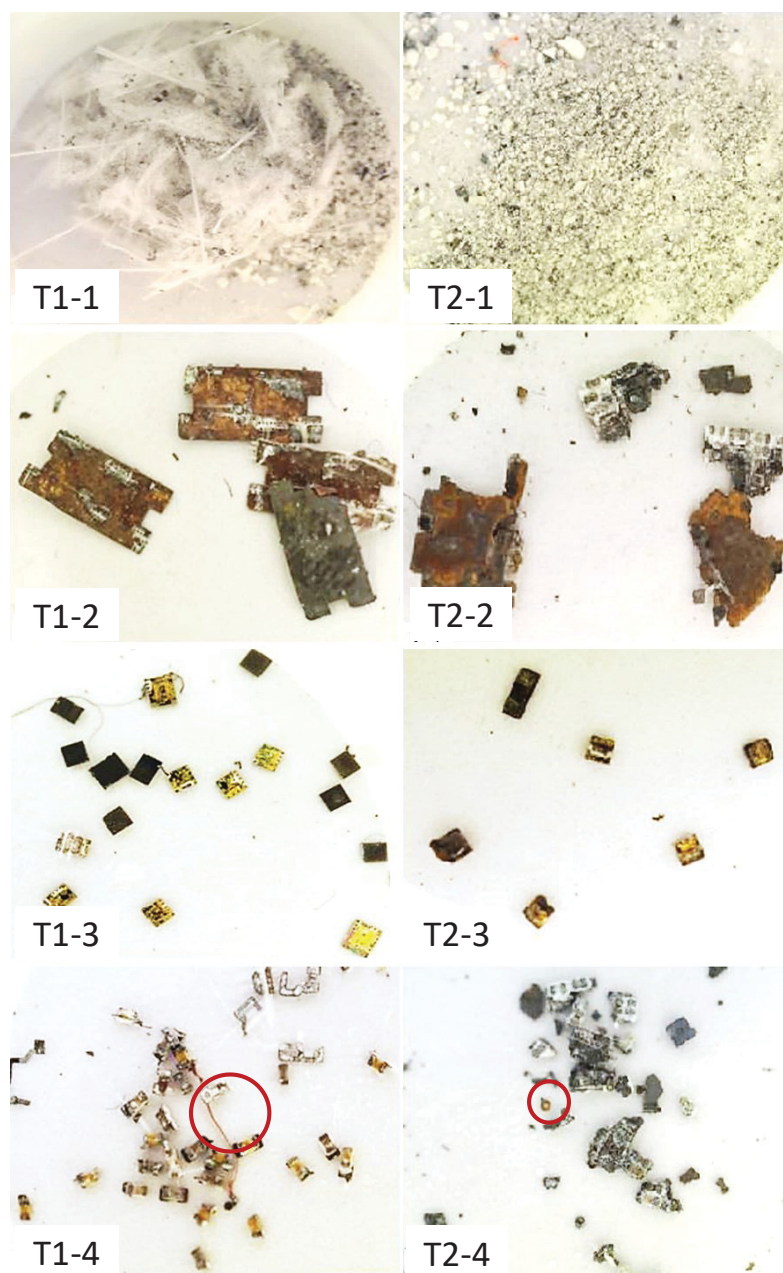
## 4.2. S-CRM hotspot analysis via product analysis

### 4.2.1. Integrated circuits

The inner structure of the ICs investigated is very complex, varying for each chip type. Appendix 6 describes the physical properties of the chips investigated.

Table 11 shows the obtained sorting fractions after the thermal treatment and the assigned sample numbers, which are used for a further description.

Figure 42 depicts exemplary the sorting fractions of chip type 1 and 2. Type 3 is optically similar to the first type. Red circles mark single components with unlikely appearance, which were categorized to the most probable fraction.



Own illustration (Ueberschaar et al., 2017c)

Figure 42: Comparison of the liberated fractions from chip type 1 and type 2

Table 11: Designation of the isolated fractions

	Isolation with copper connections	Metal plate (back of the Chip)	Electronic Module	Other
<b>Type 1</b>	T1-1	T1-2	T1-3	T1-4
<b>Type 2</b>	T2-1	T2-2	T2-3	T2-4
<b>Type 3</b>	T3-1	T3-2	T3-3 [T3-3.1, T3-3.2, T3-3.3, T3-3.4, T3-3.5]	-

#### Determination of chemical composition with XRF analysis

Results of the chemical analyses showed a mixture of various elements, including high shares of PMs in some fractions. Here, six most relevant elements according to their mass fraction and economic significance were chosen to be investigated in more detail. Those are copper, gallium, germanium, arsenic, silver and gold. Figure 43 shows the results for each sample in detail (cf. supporting information S3.4 in Ueberschaar et al. (2016c)).

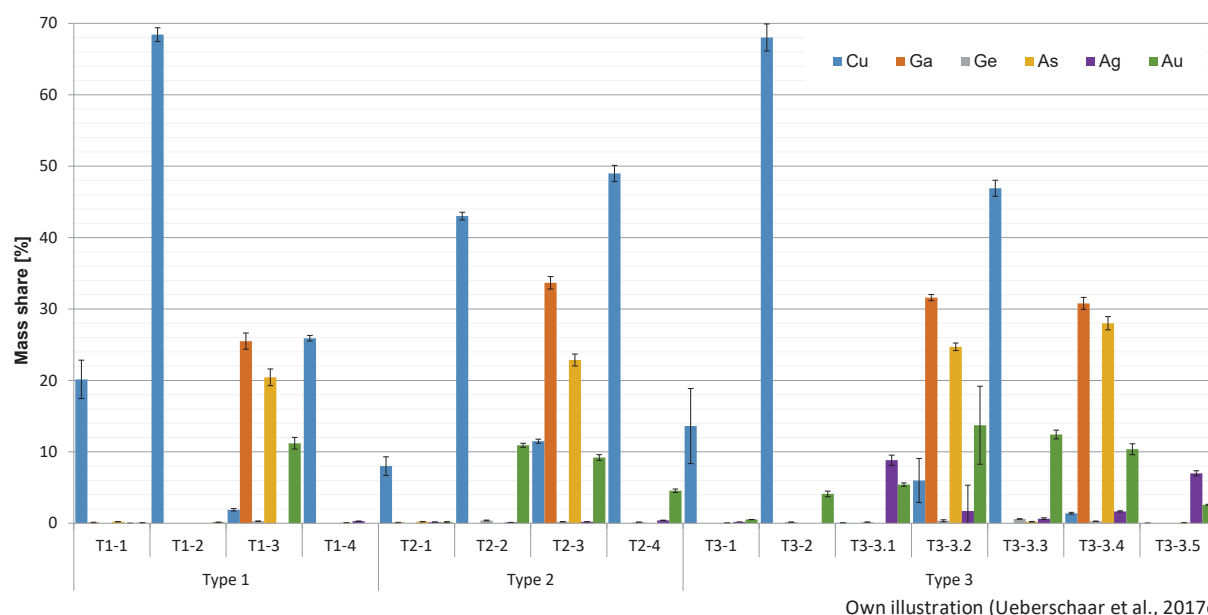


Figure 43: Mass fractions of Cu, Ga, Ge, As, Ag and Au in the separated components of the three investigated radio chips

In each IC chip, at least one fraction holds a gallium mass fraction between 25-34 %, but also an arsenic share 20-28 %. In all chips, the “electronic component” fraction holds gallium and arsenic in a similar ratio. In six fractions, gallium was detected, four had higher mass fractions, from which two fractions were found in chip type 3.

Another critical element, which is also defined as S-CRM is germanium. Ge was found in 9 of 15 fractions in very low mass fractions ranging from 0.15 to 0.6 %.

Highest copper loads were found in the metal plate on the back of each chip (T1-2, T2-2, T3-2), while a smaller amount was applied in the isolation with copper connectors (T1-1, T2-1, T3-1).

Gallium and arsenic can be assigned to particular sorting fractions. Furthermore, the results prove the possibility of concentrating gallium and arsenic with the tested approach. In all investigated chips, the GaAs rich fractions were assigned to “electronic modules”, which can be recognized by its light golden glimmer. Besides gallium, this fraction contains economically valuable metals like gold or copper. Here, further studies must evaluate a combined recovery of these applied metals.

Based on the weights of the fractions and the determined mass fractions of applied elements, potential loads of target metals have been calculated (cf. Table 12). These results are only approximate values, as the measurement accuracy of the used XRF technique is limited.

Table 12: Loads of Cu, Ga, Ge, As, Ag and Au and total mass of chip in mg per unit

	Total chip	Cu	Ga	Ge	As	Ag	Au
	Load in mg						
<b>Type 1</b>	85	15.3	1.3	<0.1	1.2	<0.1	0.6
<b>Type 2</b>	61	12.9	1.8	<0.1	1.3	0.1	2.3
<b>Type 3</b>	82	12.9	0.9	<0.1	0.8	0.3	1.2

No significant differences were determined for copper loads in the investigated chips. Copper constitutes the most applied metal in all investigated chips, followed by the sum of gallium and arsenic. In type 3, the gallium and arsenic load is much lower, whereas the gold load is higher compared to type 1. In two of three chips, the mass of gold content is higher than the load of gallium.

© (Ueberschaar et al., 2017c)

#### 4.2.2. LCD panels

102 LCD devices from six different product groups were investigated for their mass fraction of polarizer foils and the glass substrate. Due to the design, glass substrate and polarizer foils account for the major mass fraction in LCD panels. Only minor shares are related to liquid crystal and ITO layers. On average,  $20 \pm 5\%$  of LCD panels consist of polarizer foils, while  $80 \pm 5\%$  is related to the glass substrate (cf. Table 13).

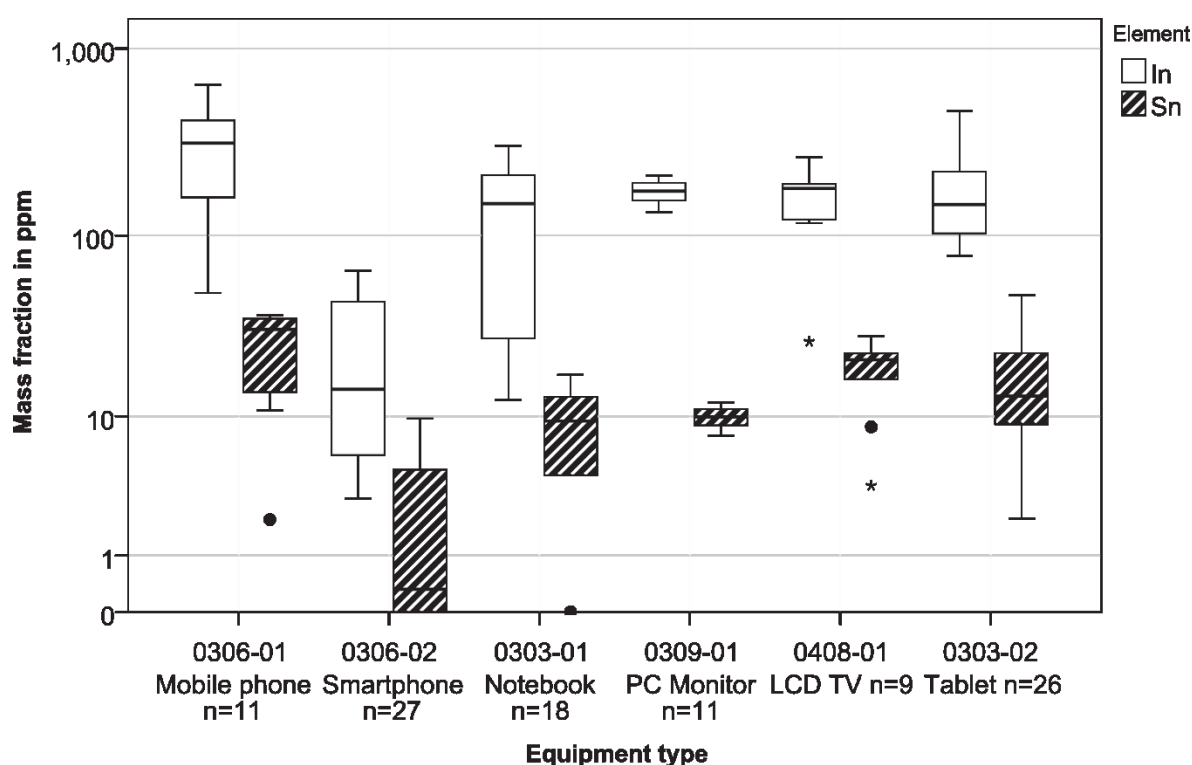
Table 13: Share of the main components polarizer foils and glass substrate in LCD panels from various screen devices

Equipment type	UNU key	Number of samples [-]	Share polarizer foil [%]	Share glass substrate incl. ITO and organics [%]
Mobile phone	0306-01	11	$21.3 \pm 3.5$	$78.6 \pm 3.2$
Smartphone	0306-02	27	$18.3 \pm 10$	$81.7 \pm 10$
Notebook	0303-01	18	$13.9 \pm 3$	$86 \pm 3$
PC Monitor	0309-01	11	$15.6 \pm 1.6$	$83.3 \pm 1.6$
LCD TV	0408-01	9	$20.6 \pm 3.5$	$79 \pm 3.1$
Tablet	0303-02	26	$23.6 \pm 7.6$	$76.4 \pm 7.6$

#### Indium in LCD panels

In Figure 44 is shown that the indium mass fraction in LCD panels of various LCD devices varies between 3-660 ppm. By far the most indium per LCD panel mass is applied in mobile phones. Indium mass fractions in tablets, PC monitors, LCD TVs, and notebooks are with 10 to 500 ppm in a similar range. Significant lower mass fractions of indium we found in smartphones. A more detailed picture of the distribution and information about the indium mass per panel weight and screen we provide in the supporting information S4, S5, S6 in Ueberschaar et al. (2016d).

Tin and indium are applied with comparable relative shares. The ratio of tin to indium fluctuates slightly for mobile phones  $7 \pm 2\%$ , tablets  $9 \pm 3\%$ , PC monitors  $6 \pm 1\%$ , LCD TVs  $11 \pm 2\%$ , notebooks  $8 \pm 2\%$  and smartphones  $9 \pm 7\%$ . Literature refers to a usual tin dopant concentration in indium between 8-10% to achieve sufficient conductivities. The results of the chemical analysis of the LCD panels from various LCD devices show varying indium contents.



Own illustration (Ueberschaar et al., 2017d)

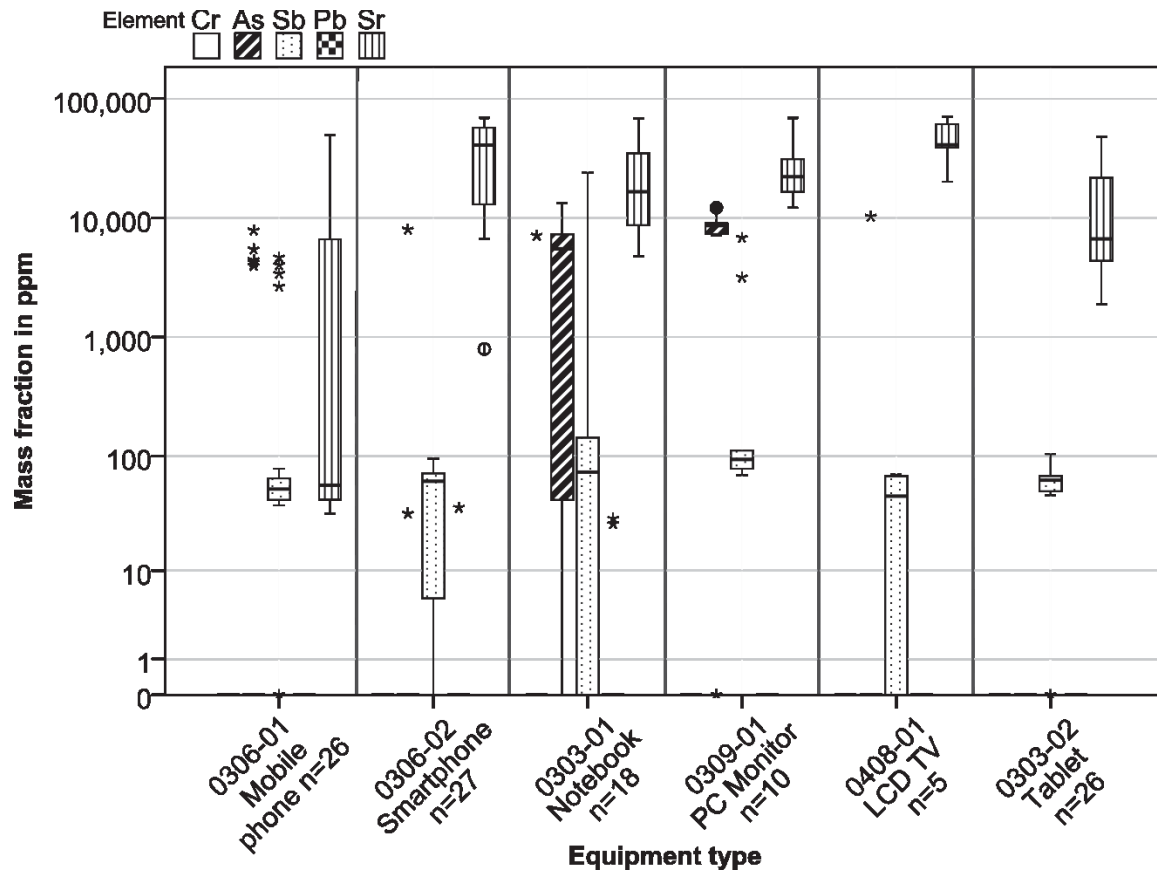
Figure 44: Indium and tin mass fractions in LCD panels from various LCD devices in ppm depicted as boxplot with median, interquartile range (IQR) (25/75 %) and whiskers 1.5 IQR (circle: outlier >1.5 IQR, asterisk: extreme value >7 IQR)

Indium mass fractions vary widely within such equipment groups investigated as smartphones, mobile phones, and notebooks, but these variations do not correlate with the screen size. An overview of the relationship between screen size and indium mass fraction is shown in the supporting information S5 & S6 in Ueberschaar et al. (2016d). While both the highest and some of the lowest values for indium concentrations are related to mobile phones, results for smartphones and PC monitors show only small ranges. One explanation might be the different LCD technologies applied. For example, the mobile phones investigated had both monochrome and colored pictures.

#### Toxic heavy metals + Sr in LCD panel glass

An XRF screening of the LCD panel glass for toxic heavy metals + Sr revealed the presence of As, Sb, Pb, and Sr, which might hinder the recycling of the glass fraction. Not all elements were detected in all equipment types nor all the panels investigated in any relevant equipment group. While the presence of strontium was verified for each LCD panel investigated in all equipment types, chromium could not be detected at all. Also, lead was used in only a few panels from smartphones and notebooks. Arsenic was not used in tablets and only 20% of the mobile phones and LCD TVs investigated. Higher shares were determined for notebooks and in particular for PC monitors. Antimony was determined in most LCD panels; lead only in a few panels from smartphones and notebooks. A full overview is given in the supporting information S8 in Ueberschaar et al. (2016d).

Figure 45 shows mass fractions of chemically analyzed toxic heavy metals + Sr in the panel glass of equipment types investigated. The supporting information S9 in Ueberschaar et al. (2016d) shows a detailed boxplot and histogram assessment for each element for all equipment types investigated.



Own illustration (Ueberschaar et al., 2017d)

Figure 45: Mass fractions of toxic heavy metals + Sr in LCD glass of various LCD devices in ppm depicted as boxplot with median, interquartile range (IQR) (25/75 %) and whiskers 1.5 IQR (circle: outlier >1.5 IQR, asterisk: extreme value >7 IQR)

The highest mass fractions of the toxic heavy metals and Sr in all LCD devices investigated was determined for strontium. Arsenic in considerable amounts was found only in notebooks and to a lesser extent in PC monitors. In other equipment types, higher mass fractions were determined in only a few of the panels analyzed. Antimony was determined in all equipment types with roughly the same mass fractions, though some showed greater fluctuations in measured values. No mass fractions could be identified for chromium, but in all devices measured there was a high level of error for this element. This is discussed in the supporting information S11 in Ueberschaar et al. (2016d). Furthermore, chemical analyses carried out on the output materials generated by the composite separation test on tablets and smartphones revealed traces of Cr. A direct comparison of results with other studies was hardly possible. Savvilotidou et al. compared literature and own data for heavy metals. Cr and As showed fluctuating results from 1-600 ppm in laptops, PC monitors, and mobile phones. (Savvilotidou et al., 2014)

#### Polymer types of polarizer foils

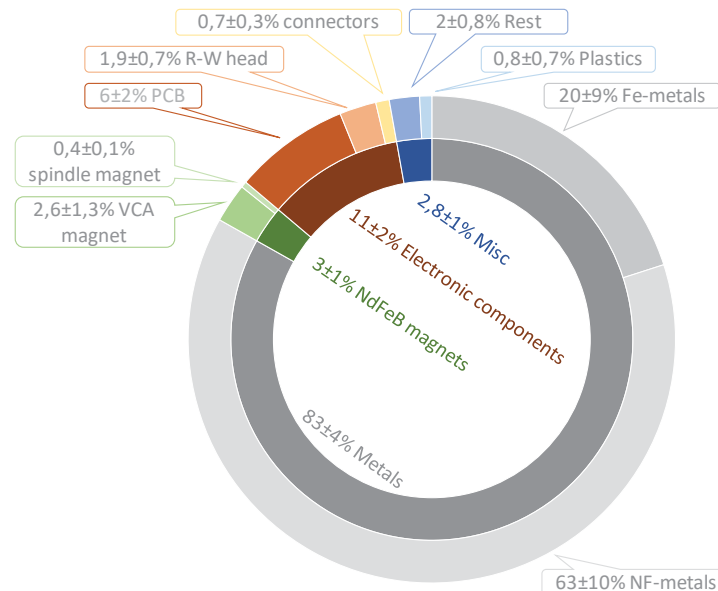
The measured spectra from the ATR-FT-IR measurements of polarizer foils compared well. Cellulose acetate and cellulose triacetate were the highest rated matches (95%) in the ATR of polymers library (Perkin Elmer), which was used to identify polymer type. The match of a cellulose triacetate database spectrum and an exemplary measured spectrum is depicted in Figure 82. Based on this small-scale evaluation, waste polarizer foils may represent an excellent input for plastic recycling, if separated thoroughly from the glass substrate.

© (Ueberschaar et al., 2017d)

#### 4.2.3. Hard disk drives

##### Structure of desktop PCs

The dismantled desktop PCs consist of 64% metals, 31% electronic components and 5% plastics (Figure 46). HDDs have with in average 542±68g a share of 3,3% of an average PC with a weight of 12,2kg.



Own illustration (Ueberschaar, 2015)

Figure 46: Results for test disassemblies of 109 collected desktop Computers in two German pre-treatment recycling plants

The results of dismantled notebooks with an average weight of 3,2kg show a share of 3,6% HDDs, which is comparable to desktop PCs. Absolute weight of HDDs in notebooks is 134±26g.

##### Structure and composition of hard disk drives

For the disassembly of the main components (casing, PCBs, hard disks, etc.) only form closure connections, mostly screws were found. Structures with REE materials, NdFeB magnets, are mounted with material closures and frictional locking connections. The access to magnets in voice-coil actuators is hindered by the construction as usually two magnets are applied, which are locked to each other by their magnet fields. Furthermore, the magnets are bonded on a carrying steel plate. In order to enable the removal of magnets, the components were heated with a gas burner (<2000°C) to incinerate the organic bonding and to destroy the magnetic field.

Based on different coefficients of thermal expansion of the different assembled materials, the same procedure was used to open the form and frictional locking connections of the spindle motor. Figure 47 shows the procedure for the liberation of NdFeB magnets.

Additional to the magnets in the voice-coil actuator and spindle motor, a very small, cubic magnet is applied in HDDs. As the test disassemblies showed an average weight of under 0,2g per magnet, this component was not considered any further.

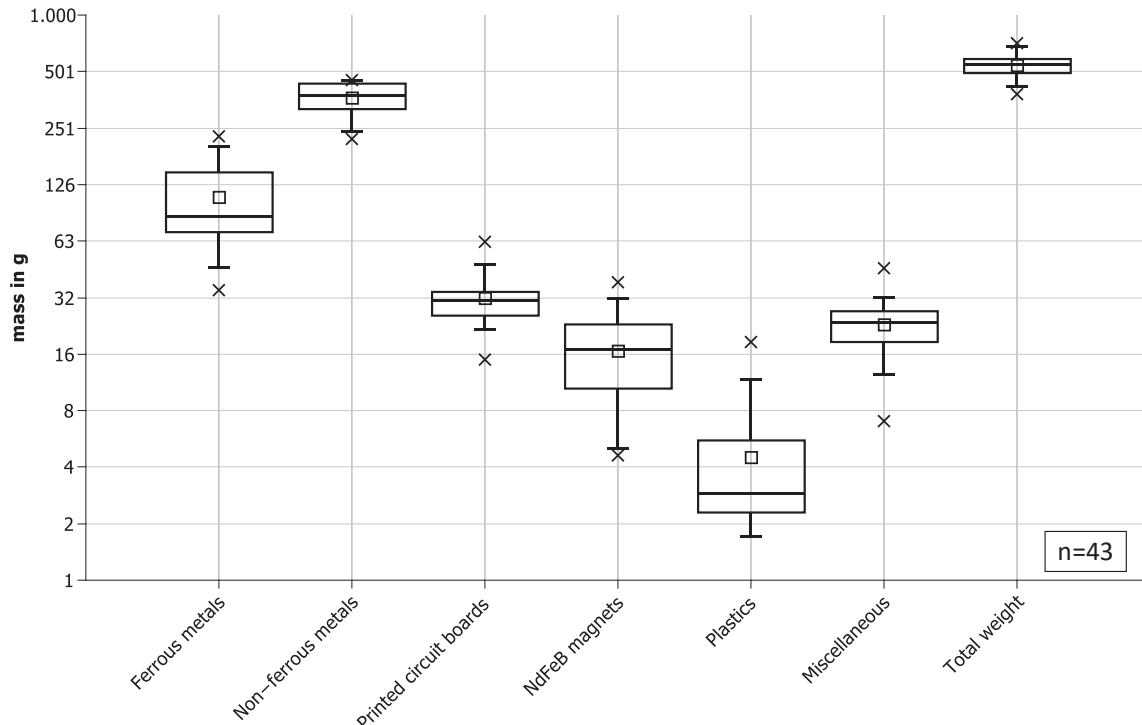


Own illustration (Ueberschaar and Rotter, 2015)

Figure 47: Liberation of NdFeB magnets in voice-coil actuator (top) and spindle motor (bottom)

#### Material composition of hard disk drives

Over 40 dismantled HDDs (average mass  $544 \pm 68\text{g}$ ) from desktop PCs showed following composition: ferrous metals  $20 \pm 9\%$ ; non-ferrous metals  $67 \pm 13\%$ ; PCBs  $6 \pm 2\%$ , NdFeB magnets  $3 \pm 1,5\%$  (voice-coil actuator  $2,6 \pm 1,5\%$ ; spindle motor  $0,4 \pm 0,06\%$ ); plastics  $0,8 \pm 0,7\%$ ; miscellaneous  $5,5 \pm 1,7\%$ . The miscellaneous fraction consists inter alia of different multi-material components like copper coils with plastics, aluminum compounds or the read-writing head. The high variety of the weight of the voice-coil actuator magnet is caused by differing numbers of applied magnets in this component. In most cases, two magnets are applied. Test disassemblies showed that ca. 18% of investigated HDDs have only one magnet in the linear motor. In those cases, the value for weight is cut in half. Figure 48 shows the statistical evaluation of the results of the manual disassembly.



Own illustration (Ueberschaar and Rotter, 2015)

Figure 48: Heterogeneity of disassembled HDDs

The non-ferrous metals represent the biggest share in HDDs. Except for a few wires and screws, this fraction consists largely of aluminum. Primarily the cast aluminum casing ( $210 \pm 30\text{g}$ ) influence the mass significantly. Measurements with an XRF device for the determination of the alloys showed a mixture

of approximately 96-98% Al and the missing 2-4% of Cu and Zn as alloying elements to ensure a high mechanical strength and a high-temperature range. Small non-magnetic steel inlets applied in the casing have a high chromium and nickel content (approximately Fe 71-74%, Cr 17-18% and Ni 7-9%).

The storage hard disks contain a pure aluminum core. The coating consists of several very thin layers to provide the required magnetic storage capabilities and mechanical surface properties. Measurements with the XRF proved the contents of Cr, Co, Ni, Fe, and Zn of the soft magnetic under layers and the magnetic storage layers. Further, the spacer between the single hard disks ( $5,9 \pm 3,7$ g in total) consist of a high purity aluminum of the alloy type Al-1000.

Another component made of aluminum is the basis and head of the spindle motor without bearing ( $26 \pm 13$ g), consisting of 95-99% Al and small amounts of Fe, Cu, and Mn.

Two third of the investigated coverings applied to close the body with the aluminum casing consist of brass (~65% Cu, ~35% Zn) with a Ni, eventually Ni-Cu coating ( $120 \pm 60$ g). The remaining one-third ( $120 \pm 25$ g) is made of steel with high Fe (~70%), Cr (~20%) and Ni (~8%) share but also small amounts of Mn.

The voice-coil magnets are applied on steel plates ( $46 \pm 25$ g). Mostly these components consist of pure steel (Fe-C) with a nickel coating for corrosion protection.

In both fractions, the ferrous and non-ferrous metals, screws produced with different steel alloy materials, mostly Fe, can be found.

A big share in the miscellaneous fraction usually holds the read-writing head ( $10 \pm 4$ g) for performing the data reading and writing operations on the hard disks. This component consists of a relatively big copper coil for executing the movement of the head relative to the voice-coil motor and hard disks. The basic structure consists of aluminum with Cu and Ni as alloy and coating materials. The top of the head is based on Fe-Cr-Ni. This unit is connected to a small applied printed circuit board with thin Au and Cu wires.

The comparative disassembly of HDDs from notebooks (~134g average weight) showed slightly higher shares for the magnets with  $5,8 \pm 0,7\%$  (voice-coil:  $4,7 \pm 1\%$ ; spindle:  $1,2 \pm 0,4\%$ ) and the PCB with  $13,1 \pm 0,2\%$ . This is based on the lightweight construction of HDDs in notebooks. One example for this is the very thin housing material. Owed to this design, fewer screws were used. Components are more often connected with pressure bonds. This leads to problems in a manual disassembly and can also influence the mechanical treatment.

#### Chemical composition of NdFeB magnets

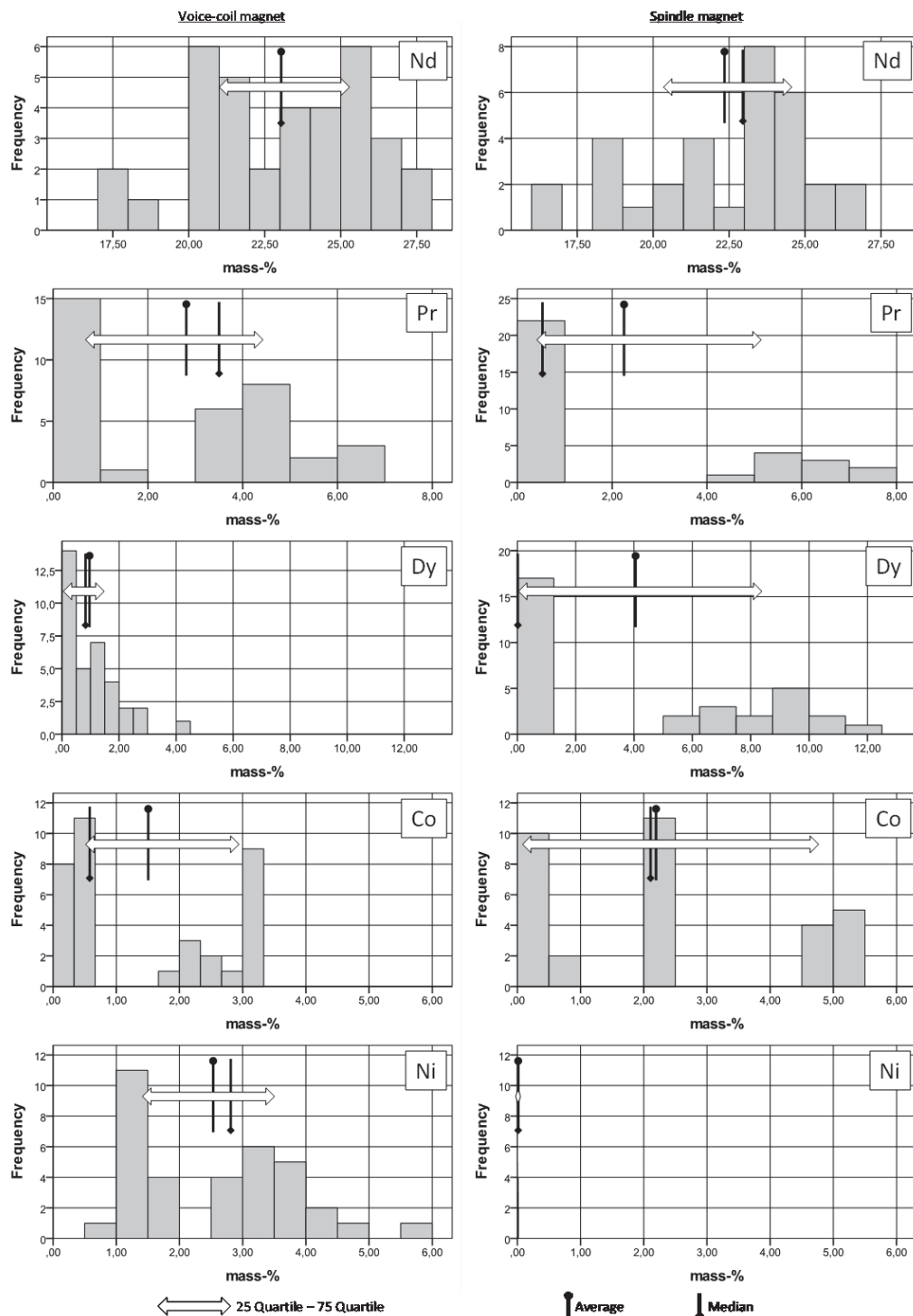
The determined main shares of elements are Fe, Nd, Pr, Dy, Co and nickel, which is only applied in the voice-coil actuator magnet. Except for Iron (cf. supporting information 2 in Ueberschaar and Rotter, (2015)), Figure 49 shows a statistical evaluation of these elements applied in the voice-coil actuator and the spindle motor.

Notable is the diversification of the results for Co and Dy. This applies especially for the spindle motor magnets. These elements represent the possible admixtures for improving the physical properties of NdFeB magnets. The results show a varying Nd content correlating to percentages of added alloy elements. Therefore, the overall share of REEs is nearly constant. As there are higher variations in the alloy composition in the spindle motor, the overall share of REE is stronger influenced. This applies especially to the use of high percentages of dysprosium and cobalt. Dysprosium with a share up to 12% increase the overall REE content which can go up to measured values of about 38%.

Moreover, the addition of Pr and Co varies. While Pr is applied irregularly to substitute Nd, the addition of Co correlates to the application of Nd. Therefore, higher percentages of Co are connected to lower overall REE contents. This applies explicitly for the voice-coil actuators (cf. Appendix 8)

Other REEs like Tb, Gd, and Sm were found only in traces below 0,1% for all samples. Due to other manufacturing methods, a special coating for preventing spindle motor magnets from oxidizing is not necessary. Therefore, no nickel is applied.

Just as a little Cu, Sn, and Zn are used in both kind of magnets.



Own illustration (Ueberschaar and Rotter, 2015)

Figure 49: Comparison of applied critical elements in NdFeB magnets

### Chemical composition of HDD PCBs

Based on a batch of almost 50 PCBs the results show the element's mass fractions for a mix of HDDs with manufacturing dates between 1994-2008.

Due to detection limits of the applied methodologies and as elements are partially not applied not all elements were determined. Table 14 shows the main shares of applied metals in PCBs from HDDs in PCs.

*Table 14 Chemical composition of applied metals in PCBs from HDDs*

Precious metals			Base metals							Critical metals			
ppm			mass-%							mass-%			
Ag	Au	Pd	Al	Cu	Fe	Ni	Sn	Ti	Zn	Ce	Nd	Sb	Ta
3440	1020	210	2.2	31.6	7.1	2.5	2.4	0.6	0.4	0.5	0.2	0.1	0.8

In addition to low mass fractions of Ce and Nd as REEs, relatively high contents of PMs such as Ag and Au were measured.

☞ (Ueberschaar and Rotter, 2015)

#### 4.2.4. Tantalum capacitors

### PCB and VIC content in different device categories

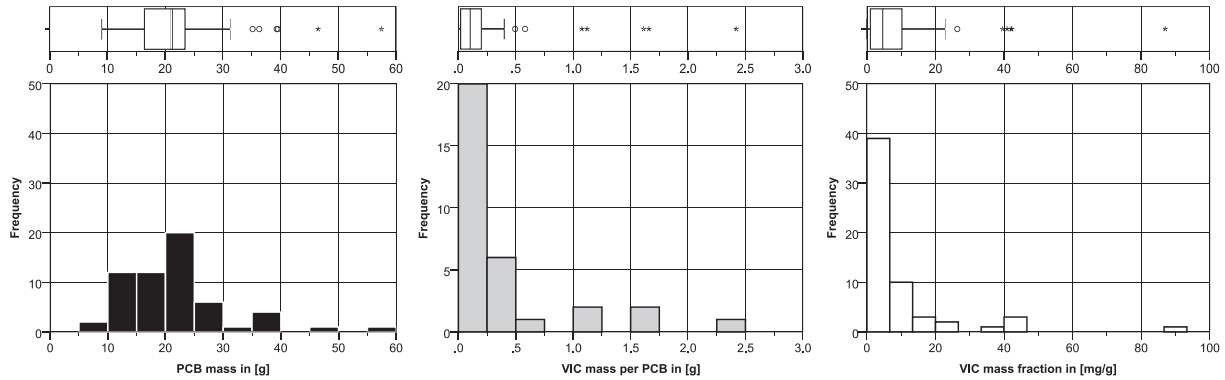
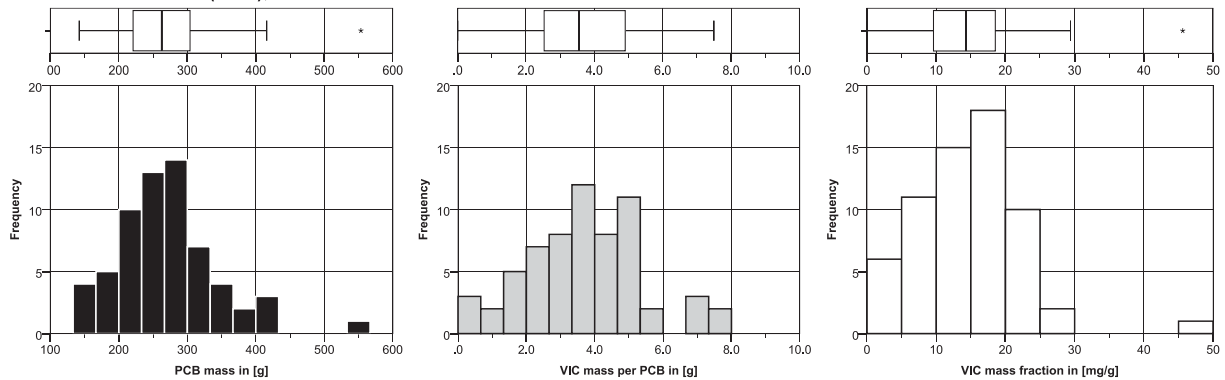
The mass fractions and total masses of tantalum in different equipment types were analyzed and the elemental mass fractions of a combined VIC sample and combined PCB samples per equipment type investigated after VIC removal were determined.

Figure 50 shows PCB masses per device, VIC masses per device and mass fractions of VICs applied on PCBs, taking the example of mobile phones and laptops. For each data set, a histogram and a boxplot have been produced. The boxplots consist of a median, interquartile range (IQR) between 25% and 50% and whiskers within 1.5 times of IQR. Depending on the data, whiskers can also end with the last value between the quartile and 1.5 IQR. Furthermore, outliers are marked with dots. The results for smartphones, tablets, desktop PCs, servers, graphic cards, HDDs, and flat screen monitors are summarized in the supporting information S8 in Ueberschaar et al. (2016b).

The amounts of tantalum and the weight of the assembled PCBs vary significantly. The values of the VIC mass per device and PCB are generally lower for mobile phones than for notebooks. The distribution of VIC mass per device and VIC mass per PCB is strongly shifted to small values for mobile phones, while values are more evenly distributed for notebooks.

This leads to a more sophisticated data processing and statistical evaluation. For non-normal (Gaussian) distributions, expectation values, uncertainties, etc. have to be determined through distribution fittings and the application of appropriate calculation models for each equipment type (Korf, 2016).

In general, the highest number of VICs is found on the main PCBs of notebooks and on the PCBs from server devices, which showed the highest median with about 30 VICs per PCB. In the samples investigated, VICs were found in a high fraction of PCBs, whereas a larger fraction of PCBs from other equipment held no VICs. The average number of VICs is also low for these devices. Smartphones and tablets contain the lowest amounts of VICs (median = 1). A total overview of the number of VICs per device type is shown in the supporting information S9 in Ueberschaar et al. (2016b).

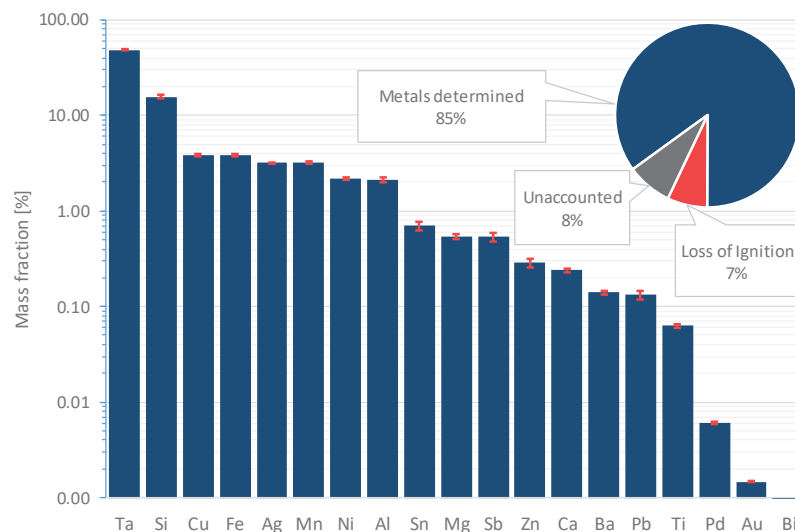
**Mobile phones (n=59); UNU=306-01****Notebooks mainboard (n=90); UNU=303**

Own illustration (Ueberschaar et al., 2017a)

Figure 50: PCB mass per device (left), VIC per device (middle) and VIC mass fraction per PCB (right) for mobile phones and notebooks. With interquartile range (25/75 %); whiskers 1.5 IQR; asterisk = extremum; circle = outlier.

**Assessment of VIC composition**

All VICs separated from the equipment types investigated were analyzed as a mixed sample to obtain an overall mass fraction for tantalum, precious metals, and other substances. Figure 51 shows the results of the chemical analyses of the VIC sample. The depicted values represent the average mass fraction per element and the related uncertainty given by the measurement uncertainty. Full analysis results are attached in the supporting information S10 in Ueberschaar et al. (2016b).



Own illustration (Ueberschaar et al., 2017a)

Figure 51: Elemental composition of VIC (LOI: Loss on ignition. Unaccounted: Share of elements not determined in the analyses; error bars show measurement uncertainty)

The inset in Figure 51 shows the amount of metals determined in the analyses in relation to the total mass of the sample. In addition to the loss of ignition (LOI), the total fraction of identified contents was nearly 92%. The LOI is most probably caused by the shell of the capacitors, which is composed of a fire-proof resin, and the graphite material in the core of the capacitor.

The average tantalum content is  $48.5 \pm 0.5\%$ , a figure in line with the 40% - 50% cited by the Tantalum-Niobium International Study Center (T.I.C.), whose data is based on a rough estimation using the number of tantalum capacitors sold and the amount of tantalum-grade powder (Schwela, 2010). Other studies also show results in the same range (Spitzcok von Brisinski et al., 2014; Kwon et al., 2016). This verifies the quality of VIC separation and can be used for further calculations.

In addition to tantalum, major contents are silicon, aluminum, copper, iron, manganese, nickel, and a surprisingly high content of silver with a mass fraction of  $3.2 \pm 0.04\%$ . Hence, considering their scarcity and market value, target elements for possible recycling are manganese, silver, and tantalum. Silicon, copper, iron, nickel, and aluminum contents are comparably low and not suitable for a single metal or mixed metal recycling process.

☞ (Ueberschaar et al., 2017a)

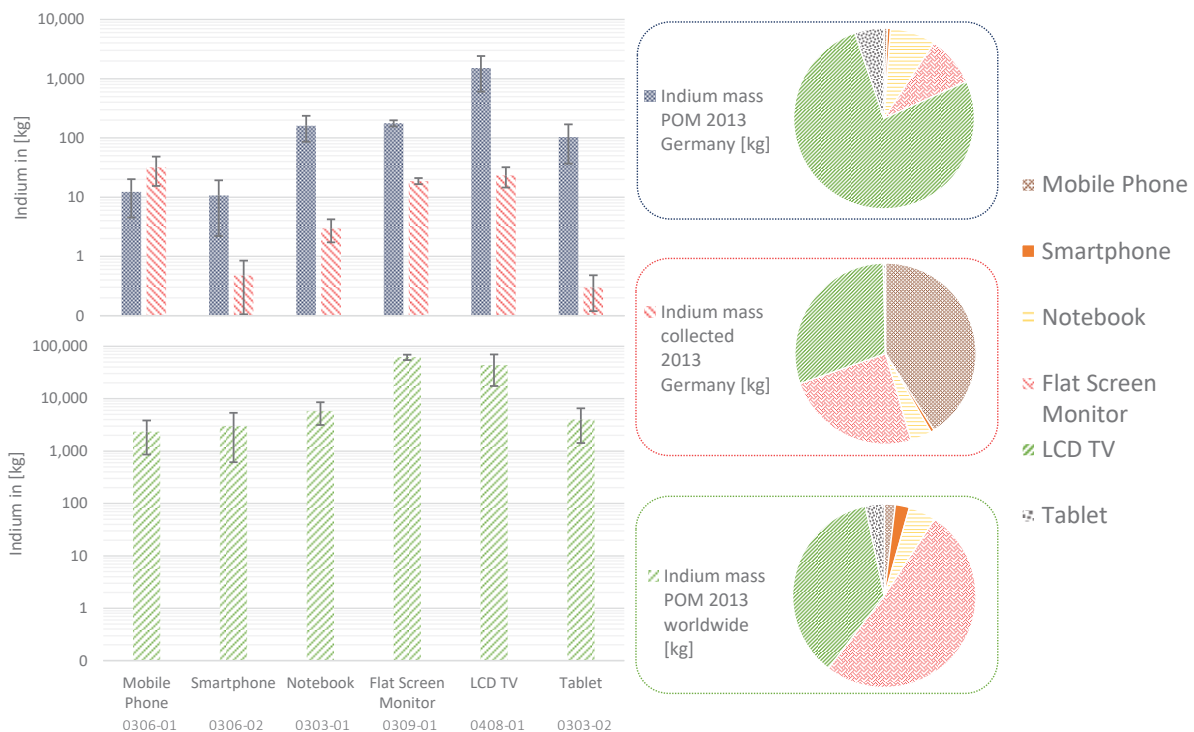
### 4.3. Available mass flows

#### 4.3.1. LCD panels

Mass potentials for indium, polarizer foils and glass substrate in LCD panels were assessed. The results revealed a total of  $2 \pm 1$  Mg indium POM in Germany in 2013. These values relate to over  $12,000 \pm 7,400$  Mg LCD panels. The quantities are correspondingly higher for POM worldwide with  $120 \pm 30$  Mg indium ( $710,000 \pm 220,000$  Mg LCD panels).

In contrast, the collection rate for LCD panels is much lower with  $390 \pm 70$  Mg. Here, only  $0.08 \pm 0.02$  Mg indium was collected in Germany in 2013. The complete calculation is shown in the supporting information S13 in Ueberschaar et al. (2016d).

Figure 52 shows the differentiated collection and POM data for indium from LCD panels in each equipment type investigated. Additional mass data for polarizer foils and glass substrate is shown in Figure 53 and Figure 54.



Own illustration (Ueberschaar et al., 2017d)

Figure 52: Total indium potential from LCD panels in various put-on-market devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany in 2013

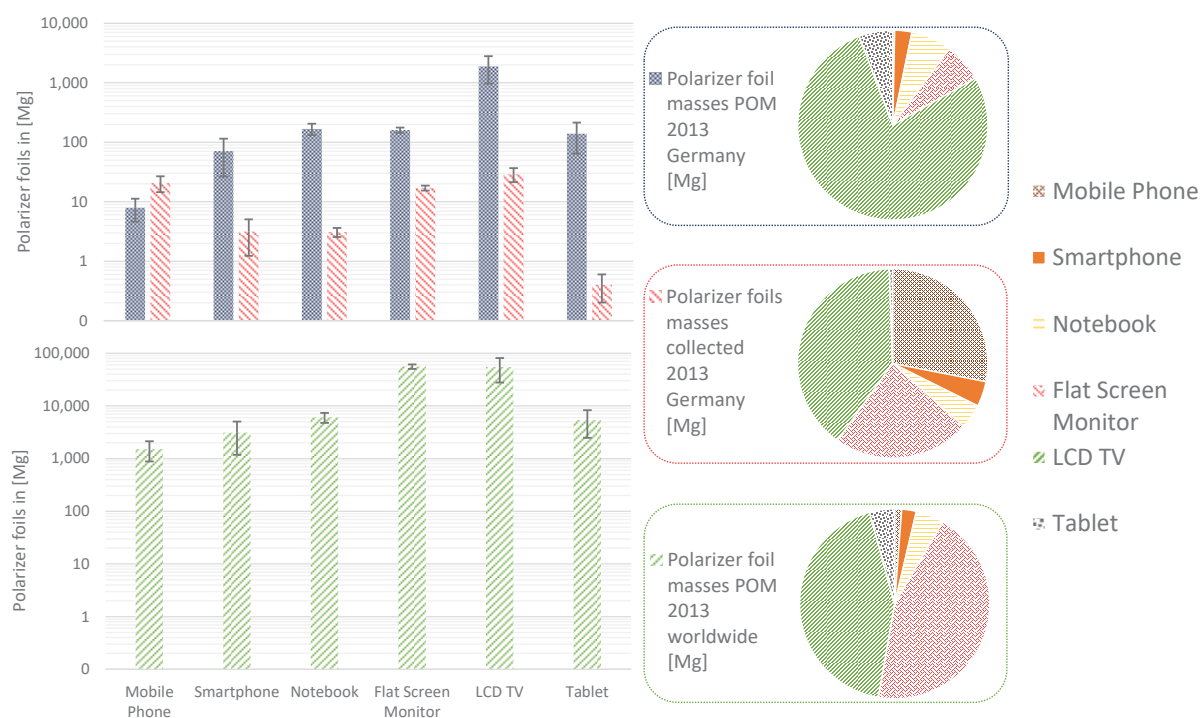
The differences in indium contribution potential of the various equipment types are remarkable. While in Germany the POM indium potential is dominated by LCD TVs, followed by far lower quantities of LCD monitors, notebooks, and tablets, the worldwide potential is mainly driven by LCD panels from flat screen monitors and LCD TVs. The share of tablets, notebooks, smartphones and mobile phones is much lower but more evenly distributed.

The indium potential relating to WEEE LCD devices collected for recycling purposes derives mainly from mobile phones, LCD TVs, and flat screen monitors. Far lower quantities are provided by the collection of notebooks. Smartphones and tablets play a minor role as no large-scale collection of these devices currently takes place. Consequently, recovery strategies should focus on those equipment types with the highest potential.



Own illustration (Ueberschaar et al., 2017d)

Figure 53: Total glass potential from LCD panels in various put-on-market devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany in 2013



Own illustration (Ueberschaar et al., 2017d)

Figure 54: Total polarizer foils potential from LCD panels in various put-on-market devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany in 2013

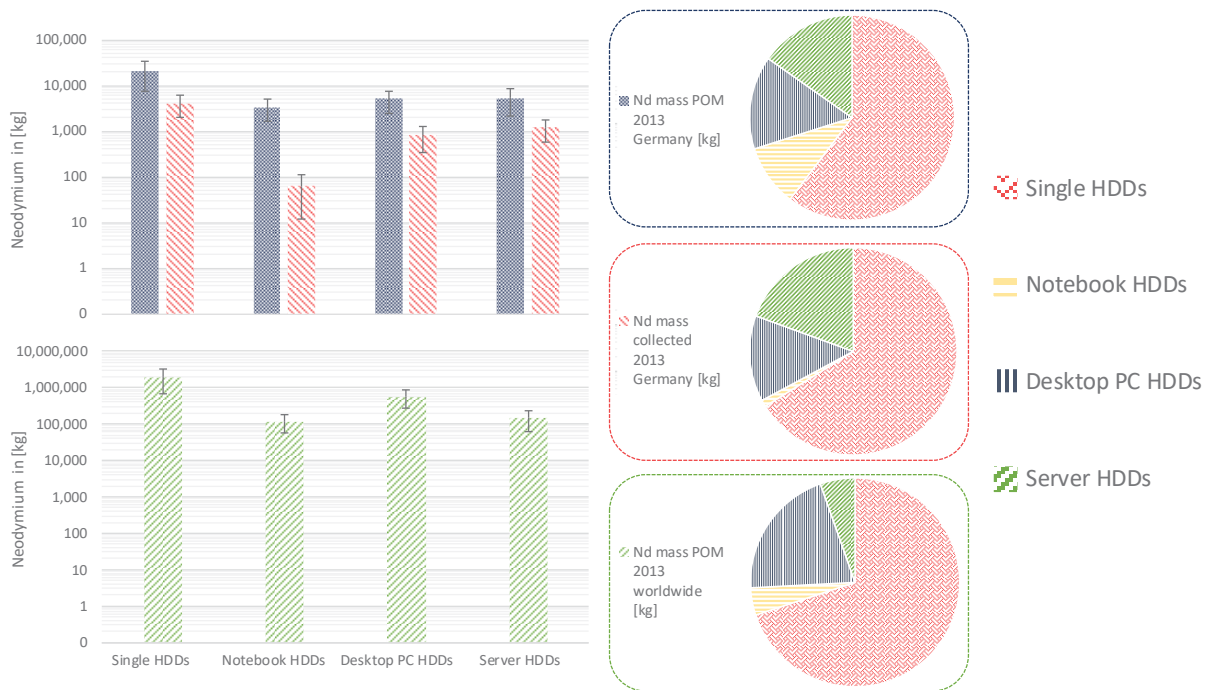
© (Ueberschaar et al., 2017d)

## 4.3.2. HDDs

The S-CRM hotspot analysis for HDDs revealed two important REE materials applied in NdFeB magnets originating from the spindle motor and the voice coil actuator. Therefore, the S-CRM mass potential analysis was carried out for neodymium and dysprosium in single HDD, and HDD assembled in notebooks, PCs, and servers.

Figure 55 and Figure 56 depict the results. In total  $34 \pm 14$  Mg neodymium and  $2 \pm 1$  Mg dysprosium were POM in Germany in 2013. Masses related to collected WEEE are much lower with only  $6 \pm 2$  Mg Nd and  $0.4 \pm 0.2$  Mg Dy. Worldwide,  $2,800 \pm 1,300$  Mg Nd and  $170 \pm 110$  Mg Dy were POM in 2013.

Compared to indium from screen devices and tantalum from PCB carrying devices, the share of contributing devices to neodymium and dysprosium masses POM in Germany and worldwide as well as collected WEEE is very similar. Single HDDs carry most Nd and Dy in NdFeB magnets. HDDs applied in notebooks, PCs, and servers together sum up to a maximum of 40% Nd and Dy contribution.



Own illustration

Figure 55: Total neodymium potential from HDD in various put-on-market devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany in 2013



Own illustration

Figure 56: Total dysprosium potential from HDD in various put-on-market devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany in 2013

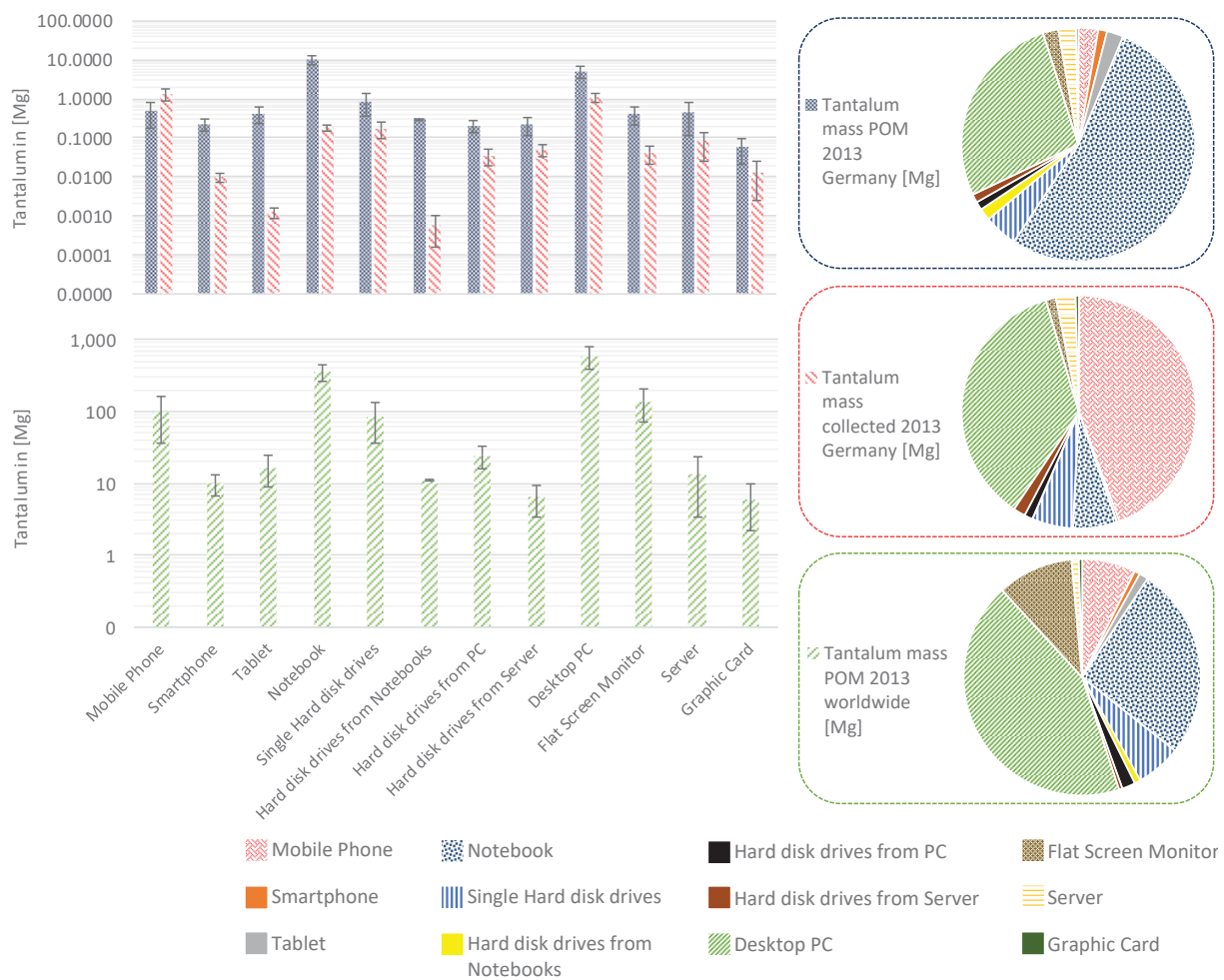
#### 4.3.3. Tantalum capacitors

Results reveal a total of  $6 \pm 1$  Mg of collected tantalum capacitors and subsequently  $3 \pm 1$  Mg of tantalum.

In contrast, POM data differs. This is important for developing a circular economy as secondary tantalum will be able to meet the production needs of new electrical and electronic goods. In Germany in 2013, the POM figures were  $39 \pm 7$  Mg tantalum capacitors and  $19 \pm 3$  Mg tantalum.

Worldwide POM figures for 2013 were  $2,800 \pm 500$  Mg of tantalum capacitors and subsequently  $1,300 \pm 250$  Mg of tantalum.

Figure 57 shows the differentiated collection and POM data for tantalum masses obtained through VIC separation in each equipment type investigated (results for PCB residues and PCBs including VICs are provided in the supporting information S14 and S15 in Ueberschaar et al. (2016b). The calculation for the data extrapolation is also depicted in S12.)



Own illustration (Ueberschaar et al., 2017a)

Figure 57: Total tantalum potential through VIC separation in POM devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany

The highest tantalum collection potential among tantalum bearing devices is found in mobile phones. Although tantalum content in this device type is average for the equipment types investigated, high collection rates lead to high availability of tantalum masses. Mobile phones constitute an exception as POM tantalum masses are generally lower than those of WEEE. This is because of a shift in technology. The market for mobile phones is shrinking while the market for smartphones is increasing and

significant quantities of smartphones are not yet being collected. As a result, the gap between tantalum masses for POM and collection is high. This also applies to tablets. Therefore, smartphones and tablets, as well as HDDs from notebooks, have the lowest potential for tantalum collection.

Tantalum masses linked to POM devices in Germany and worldwide have almost the same distribution. Highest potentials are connected to notebooks and desktop PCs. The lowest potentials are calculated for servers and associated HDDs as well as smartphones and tablets.

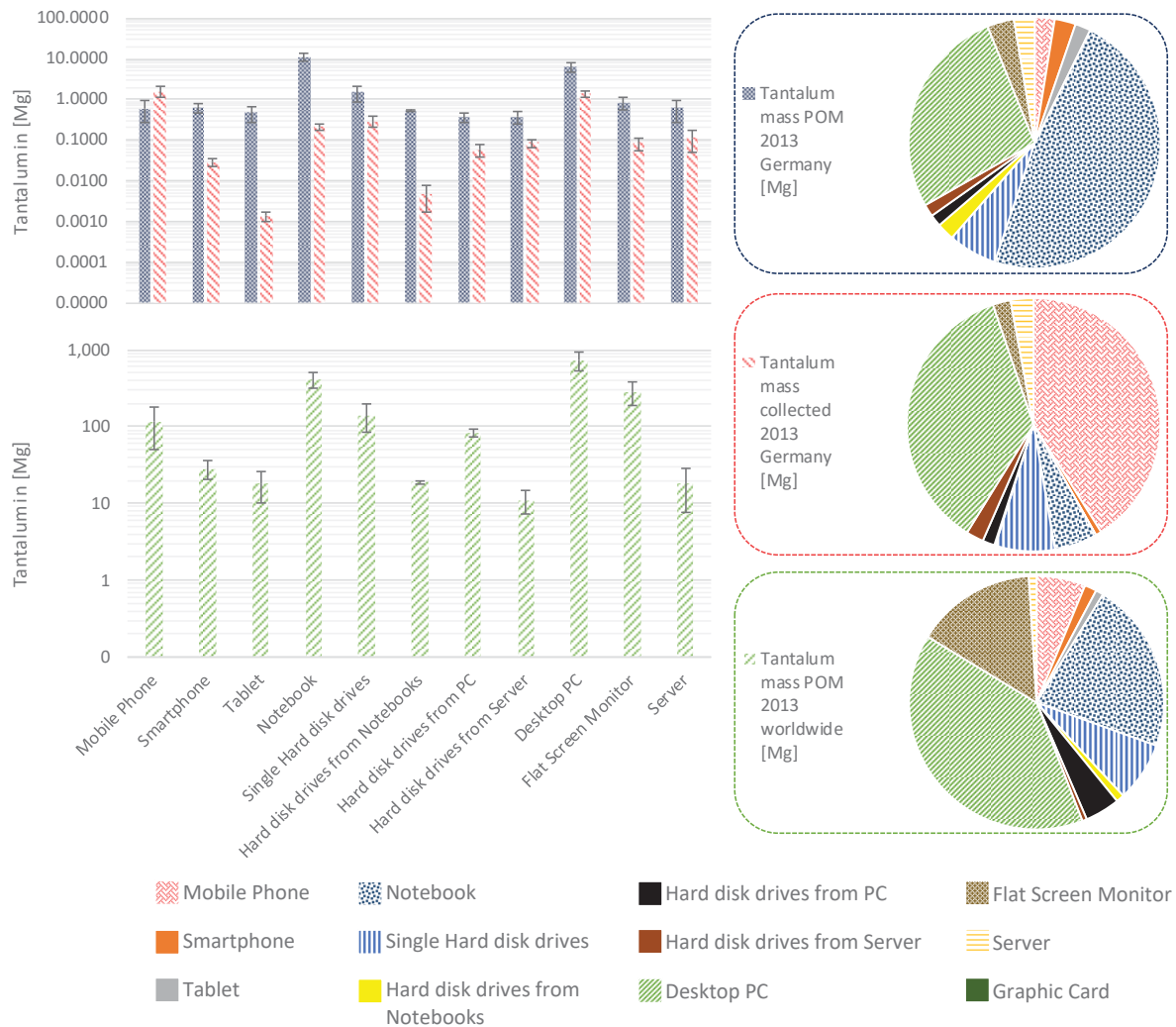
This data applies only for the year 2013. Depending on market shifts, the potentials can change significantly.

The POM and returned tantalum flows are closely related to the composition of sold or collected equipment types with accordingly different loads of tantalum. In order to compare this distribution, Figure 57 also shows the relevant shares for tantalum POM and collected in 2013 in Germany and POM worldwide. This data is based only on tantalum flows through VICs. The distribution for PCB residues and PCBs including VIC are shown in Figure 58 and Figure 59.



Own illustration (Ueberschaar et al., 2017a)

Figure 58: Total tantalum potential in PCB after VIC separation in POM devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany

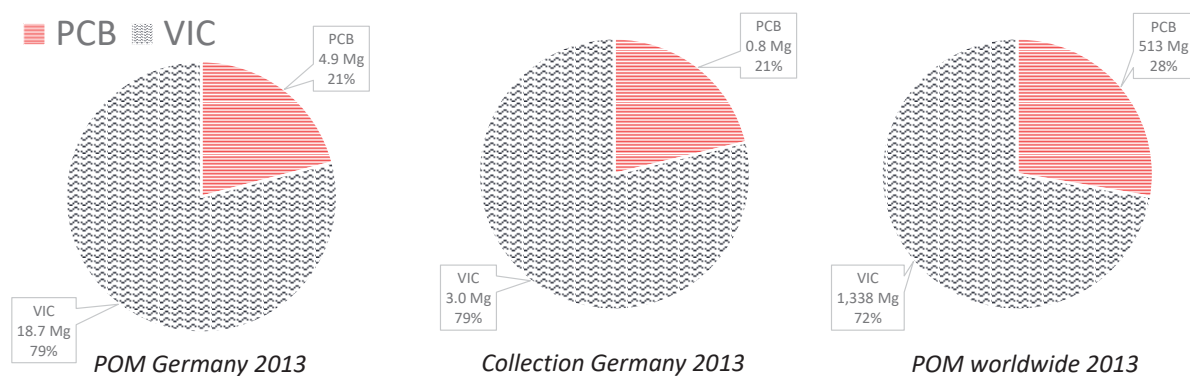


Own illustration (Ueberschaar et al., 2017a)

Figure 59: Total tantalum potential in PCB and VIC in POM devices (Germany / worldwide) versus WEEE devices collected for recycling purposes in Germany

Due to high sales of notebooks in Germany in 2013 (~5 million devices), the share of tantalum POM is dominated by this type of equipment. Although sales of smartphones are much higher (~22 million devices), the contribution of these tantalum flows is only about 1.2 % due to the low number of applied VICs. In contrast, the tantalum flows collected for recycling purposes are dominated by mobile phones. Background data is given in the supporting information S12 in Ueberschaar et al. (2016b).

The tantalum load from PCB residues is partially higher than in the separated VICs. This applies, for example, to smartphones and desktop PCs. For HDDs and flat screen monitors the ratio of tantalum in PCB residue and VICs is almost the same. Therefore, tantalum flows related to PCBs and VICs in POM and WEEE must be distinguished from each other. A comparison of all equipment types investigated, except for graphic cards, is shown in Figure 60.



Own illustration (Ueberschaar et al., 2017a)

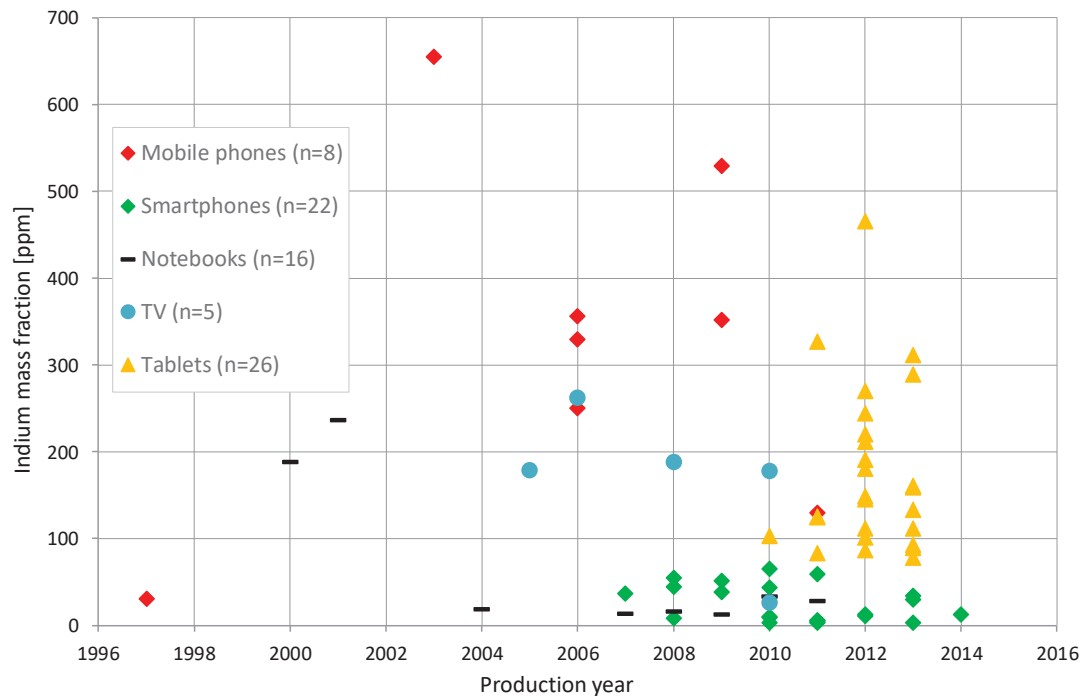
Figure 60: Share of PCB residues and VIC on the total tantalum flows POM and collection in Germany and POM worldwide in 2013

© (Ueberschaar et al., 2017a)

#### 4.4. Time trends

##### 4.4.1. LCD panels

It was not possible to date all of the devices; only a reduced data set could be analyzed. The only time trends that could be interpreted were those for TV sets and notebooks, where a negative trend was noticeable. For tablets and smartphones, only short-term data sets were available, due to the recent market entry of such devices. Generally, a significant drop in the indium mass fractions over time did not reveal itself and any potential reduction of applied indium and tin content could not be verified (cf. Figure 61).

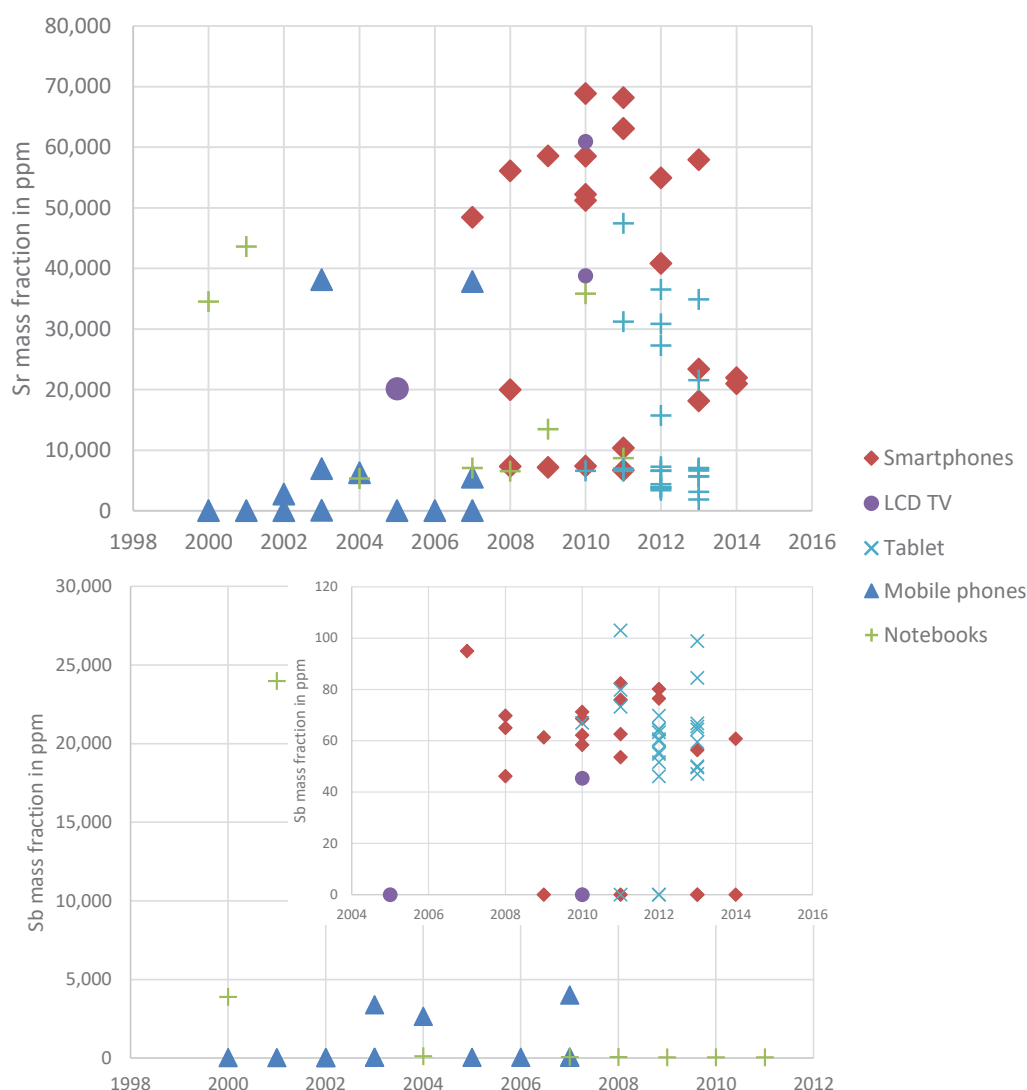


Own illustration (Ueberschaar et al., 2017d)

Figure 61: Indium mass fraction vs. production year of panels from various screen devices investigated

Several reasons could influence the use of toxic heavy metals + Sr the manufacture of LCD panels over time. Among the main ones are efforts to reduce the use of harmful substances like RoHS. Therefore, a possible time trend of toxic heavy metals + Sr used was also investigated.

As in the application of ITO in LCD panels, no time trend can be derived. The peaks of toxic heavy metals + Sr used occur randomly. Figure 62 shows the results for Sb and Sr. All other graphical depictions are shown in the supporting information S10 in Ueberschaar et al. (2016d).



Own illustration (Ueberschaar et al., 2017d)

Figure 62: Mass fractions of Sr and Sb in ppm versus manufacturing date of various investigated screen devices

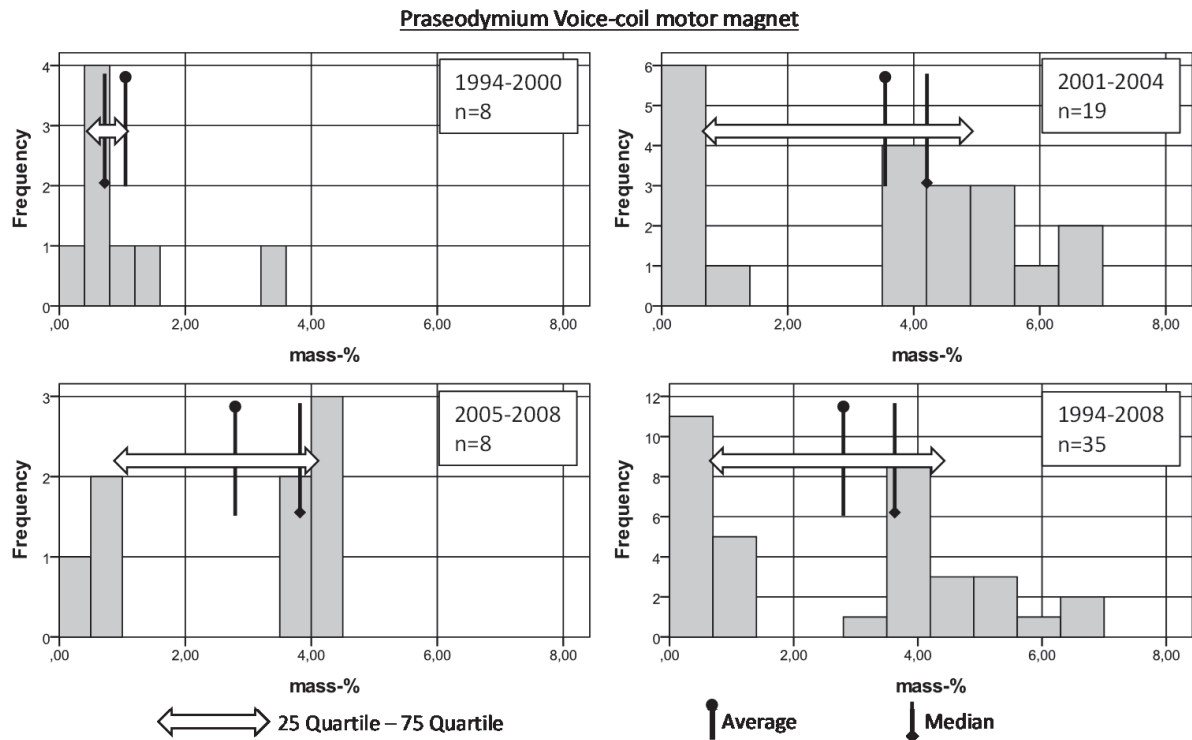
© (Ueberschaar et al., 2017d)

#### 4.4.2. Hard disk drives

The development of blends for the manufacturing of NdFeB magnets changed over time due to economic reasons or higher demands on magnet materials. Chemical compositions related to time periods were assessed with developed stacked histograms separated for the voice-coil and spindle motor magnets.

One example for the detailed results is shown with element Neodymium for the voice-coil magnets in Figure 63. The assessment for all element separated for the voice-coil and the spindle magnet is attached in the supporting information 3 in Ueberschaar and Rotter, (2015).

The chronological trend for voice-coil magnets shows that the admixture of Praseodymium increases until 2005. Like stated before, Praseodymium was used for economical reasons. As the prices raised significantly from 2005-2006 (MetaErden GmbH, 2014a) on, the substitution for Neodymium with this material decreased. The same applies for substances with technical benefits like Cobalt. Not only the admixture in each magnet drops in the years 2000 to 2008 from nearly 3% to 1,4-1,6%, but also the number of voice-coil magnets in which this substance is generally applied decreases.



Own illustration (Ueberschaar and Rotter, 2015)

Figure 63: Time trend for the application of Praseodymium in voice-coil motor magnets

Dysprosium decreases a little bit in the years 2000-2004 before the values nearly recover.

Neodymium holds the highest share of the REEs in the magnets. As it is the most important substance to obtain the typical strong magnetic field, it behaves very constant in the distribution in all measured magnets and generally over time. In average, it decreases a little bit from 25,5% (1994-2000) to 22,5% (2000-2008). This is traced to a general reduction of the REE overall content in NdFeB magnets: 28,6% (1994-2000) to 26,4% (2000-2008).

As Nickel represents the coating of the voice-coil magnets, the application is very constant over time.

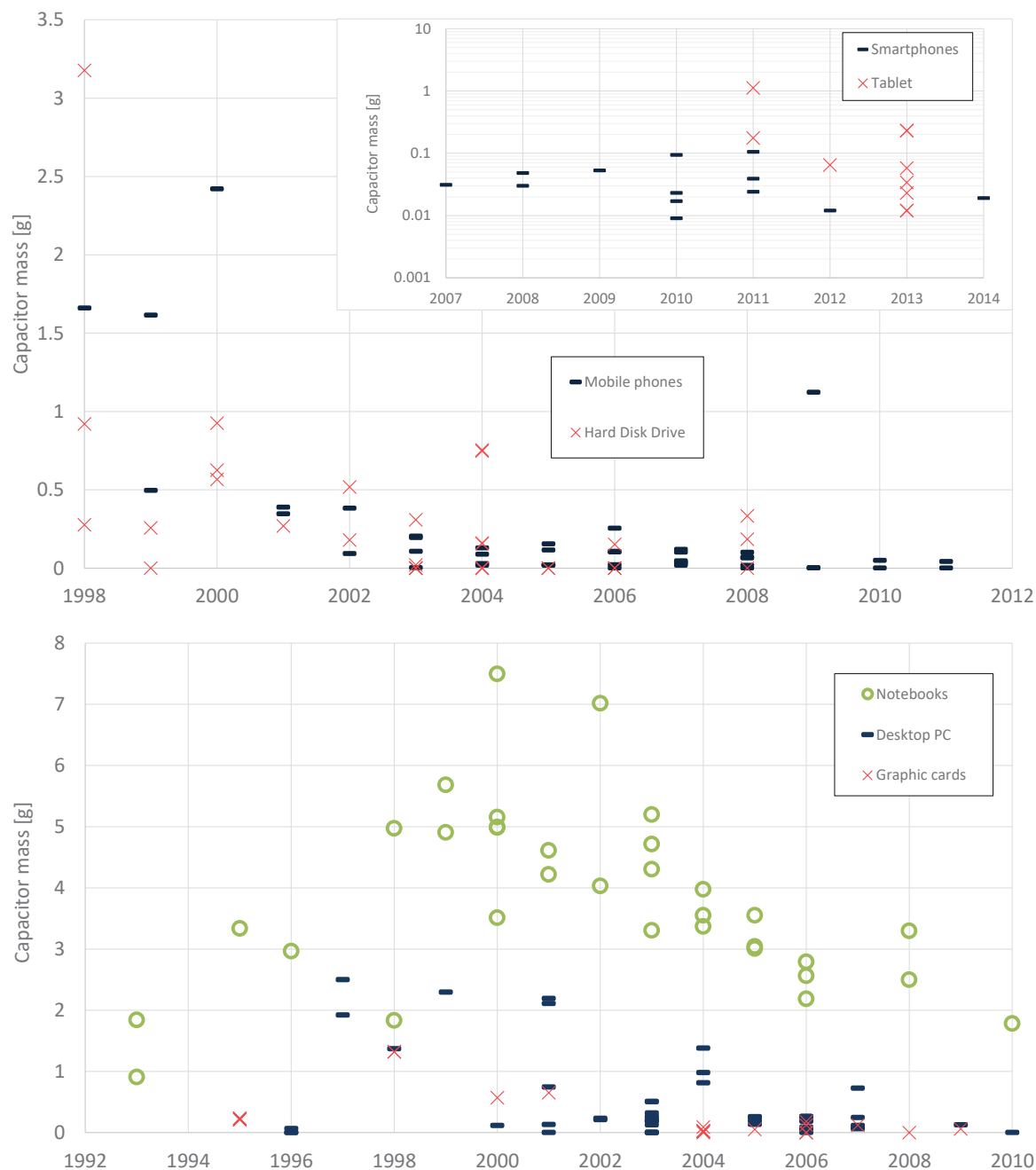
Like for the voice-coil magnets, Dysprosium has a drop in the years 2000-2005 and is again more often applied in spindle motor magnets from the year 2005 and later. Also, the chronological trend for Praseodymium is comparable. In the years 1994-2000 admixtures were under 1% and raised from 2000 on to percentages of 6-7%. The progress of application of Neodymium in spindle motors is similar to the linear motors. Only in the last years 2000-2008, the application of Neodymium decreased to under 20% while Praseodymium and Dysprosium were used for its substitution. Likely results were shown for Cobalt. Nearly all investigated magnets from 1994-2004 have high contents of this substance with about 3%. After 2005, values fall far below 1%.

Different kinds of blending are accounted for the manufacturing of both types of magnets. Depending on market prices and desired physical properties, Neodymium is substituted to a specific level with Dysprosium, Praseodymium, or Cobalt. Mainly, Dysprosium and Cobalt are applied together with low concentrations of Praseodymium. In the years from 2000 on, higher concentrations of Praseodymium were determined. In those samples, the Dysprosium content was enhanced while Cobalt content is very low.

© (Ueberschaar and Rotter, 2015)

#### 4.4.3. Tantalum capacitors

In order to devise a trend over time of applied tantalum in the equipment types investigated, the mass fractions of VICs were compared to the data gathered on recycling-oriented product characterization. Figure 64 shows a comparison by year of manufacture of the total VIC masses separated from the PCBs per single device investigated.



Own illustration (Ueberschaar et al., 2017a)

Figure 64: Mass of capacitors per PCB of mobile phones, smartphones, tablets, HDDs, graphic cards and mainboards from desktop PCs, and notebooks compared by date of manufacture

In general, the total mass of VICs applied decreases over time. This applies to notebooks as well, although an increase in total VIC mass was seen between 1993 and 1999. This may be attributed to the development of computing capacity and an increasing miniaturization of the devices. However, less data is available for these years which may result in an incomplete representation of the total mass of VICs applied.

A stronger trend is shown for mobile phones, HDDs, desktop PCs, and graphic cards. Here, a generally decreasing trend in capacitor mass per PCB and unit is visible.

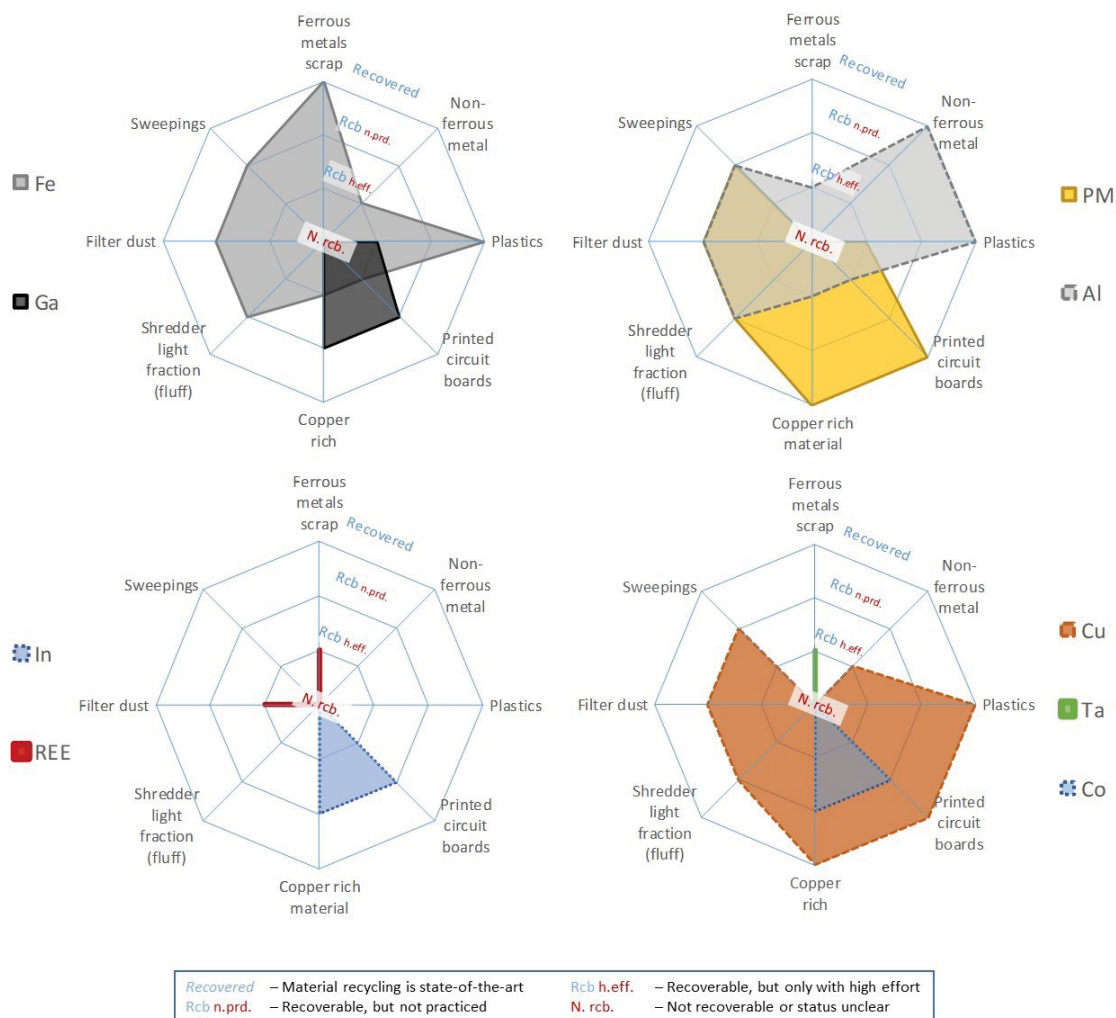
Smartphones and tablets represent a very young generation of mobile devices. Since these equipment types are still in the market saturation phase (particularly tablets), figures are available only for the years between 2007 and 2014. In the case of tablets, data is available for only three years. Development in product technology may be less over this short time span. Therefore, variations in VIC content are also low.

☞ (Ueberschaar et al., 2017a)

## 4.5. Recyclability assessment

### 4.5.1. Output materials from WEEE pre-processing

Figure 65 summarizes the results of the theoretically performed conceptual recyclability assessment for IBMs, PMs, and S-CRMs, in the output fractions from the mechanical pre-treatment of WEEE (cf. 4.1.2 “Extended batch test in WEEE pre-processing”).



Own illustration (Ueberschaar et al., 2017b)

Figure 65: Recyclability of IBMs, PMs, and S-CRMs in the output fractions of the automated sorting process in the extended batch test

It is distinguished between a practiced recovery, non-recycling, and possible recycling strategies, which are simply not used or connected to disproportionate high efforts. These results do not considerate recycling efficiencies.

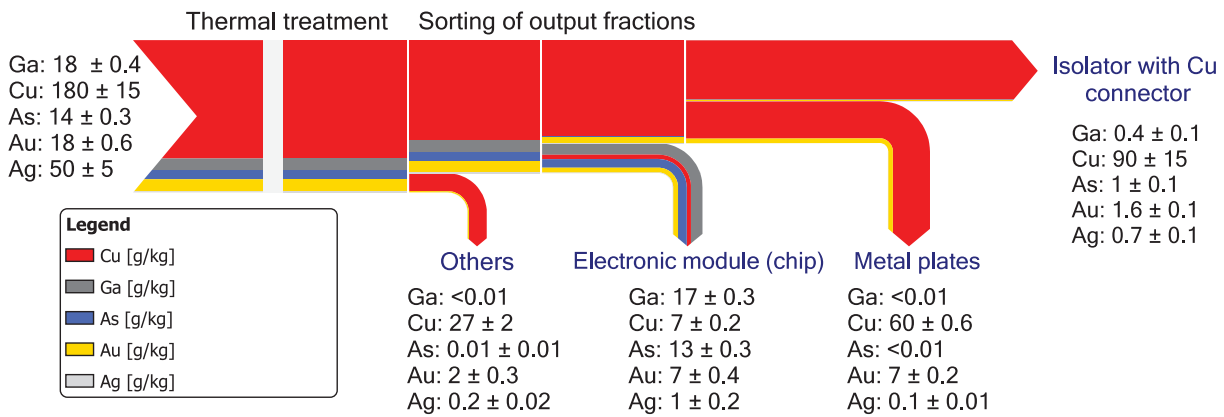
Results show output fractions for the optimized recovery of IBMs and PMs. Highly valuable materials like gold or copper may be even separated in subsequent recycling processes. This does not apply to S-CRMs. Most output fractions are related to a total loss of these minor metals. Ta or REE cannot be recovered if dispersed to any of the present output fractions. A pre-separation of according components or WEEE devices is mandatory. A recovery is at least possible for Ga or In from the PCB or Cu-rich fraction. However, due to low mass fractions and other present minor metals, the recovery is highly energy and cost intensive.

© (Ueberschaar et al., 2017b)

#### 4.5.2. Integrated circuits

Through thermal treatment, components and materials in ICs were liberated. Output fractions were then visually sorted into 4 categories: isolator, metal plates, electronic modules and others. An MFA on element level was developed for Ga, Cu, As, Au and Ag to assess the selectivity of this process.

Figure 66 shows the mass flow diagram. The data is depicted in g and is normalized to 1,000 g input material (ICs).



Own illustration (Ueberschaar et al., 2017c)

Figure 66: Mass flows of Ga, Cu, As, Au and Ag in output fractions after thermal treatment of ICs. All data normalized to 1,000 g input material

For Ga and As, a very high selectivity is noticeable. Being an essential part of the chip, 98% of Ga and 94% of As concentrate in the electronic module. As represents the dopant in this type of semiconductor. Thus, it represents an inherent part of the semiconductor material and is therefore linked to Ga. High shares of Au are present in the electronic module and the metal plates output with about 40% in each case.

Table 15 shows the transfer coefficients for all elements investigated. Results prove a worse selectivity for Cu and Ag.

Table 15: Transfer coefficients of elements investigated in recyclability assessment of ICs via thermal treatment

	Cu		Ga		As		Ag		Au	
%	Avg.	StDev	Avg.	StDev	Avg.	StDev	Avg.	StDev	Avg.	StDev
Isolator with Cu connector	50	8	2	1	6	1	36	2	9	1
Metal plates (back of chip)	31	1	0	-	0	0	3	1	39	1
Electronic modules (chip)	4	1	98	2	94	2	49	12	41	2
Others	15	1	0	-	0.1	1	12	1	11	2

Recycling of gallium from WEEE is technically feasible. However, due to the design and use of components carrying gallium, various process steps are required. In particular the identification and separation of according components are complex. However, the use of gallium is limited to only a few applications, which might facilitate such processes. Material flows of similar components could be combined to supply larger mass flows to recycling facilities.

The relatively small shares of gallium-bearing components in PCBs or LEDs lead to a dilution with other materials in conventionally applied recycling processes. Ending in the pyrometallurgical process for

copper and precious metals refining, gallium is transferred oxidized to the slag. Based on this, gallium rich components must be separated prior to any mechanical processing with other material.

Results in this thesis prove the technical possibility of liberation and separation of gallium concentrates. Selected subsequent processes would enable the recovery of gallium and other valuable materials like gold and copper. This combines economical and ecological requirements for developing recycling strategies for critical raw materials.

☞ (Ueberschaar et al., 2017c)

#### 4.5.3. LCD panels

The newly developed recycling strategy consists of a composite separation and dissolution of the ITO conducted on samples taken from smartphones and tablets. For both steps, a mass flow analysis was carried out, depicted on a material level in % and on an element level as mg/kg input material. Figure 67 shows the results.

The mass flow analysis for smartphones and tablets shows that all polarizer foils were separated successfully in an individual output. The same applies to the glass substrate. After the base leaching step, the glass residues were leached to wash off all surface applied substances. This allows a clean LCD glass fraction to be provided. Due to reactions during the base leaching, the separated liquid phase represents an indium concentrate with organics like liquid crystals, adhesives, or potentially converted polarizer foils. Compared to the smartphone samples, the mass flow of tablets shows a lower overall quantity of glass and comparable quantities of ITO and polarizer foils. Only the organic material, which could consist of adhesives or degenerated polarizer foils, has a much higher share in the tablet test with  $190 \pm 80$  g/kg input material compared to  $10 \pm 80$  g/kg in smartphones.

The material flow analysis on an elemental level shows that a selective separation of indium and copper is possible. About 80% of the total indium was transferred to the ITO output in the composite separation test for smartphones and tablets. Only minor masses were found in the foils fraction and the leachate. Interestingly, tin was not selectively separated and was distributed to all output fractions. Table 16 shows the transfer coefficients for these three elements.

Table 16: Transfer coefficients of In, Sn, Cu, As, Cr, Pb, Sb, and Sr with individual mobility expressed as related uncertainties within the output fractions in two separately conducted composite separation tests for smartphones and tablets

Type	Output	Target metals						Toxic heavy metals + Sr									
		In		Sn		Cu		As		Cr		Pb		Sb		Sr	
		[%]	±	[%]	±	[%]	±	[%]	±	[%]	±	[%]	±	[%]	±	[%]	±
Smartphone	Foils fraction	14		57		20		4		10		37		61		11	
	Foil leachate residues	4	12	13	22	12	12	46	7	0.7	13	63	15	13	23	52	7
	ITO + Organics	82		30		67		50		90		ND		26		37	
Tablet	Foils fraction	17		44		14		32		45		62		99		31	
	Foil leachate residues	3	12	16	16	3	12	68	11	31	15	38	20	1	30	17	13
	ITO + Organics	80		40		83		ND		25		ND		ND		52	

**Note:** The uncertainties indicated represent a possible shift of values to other output fractions in the related experimental procedure. ND = not determined

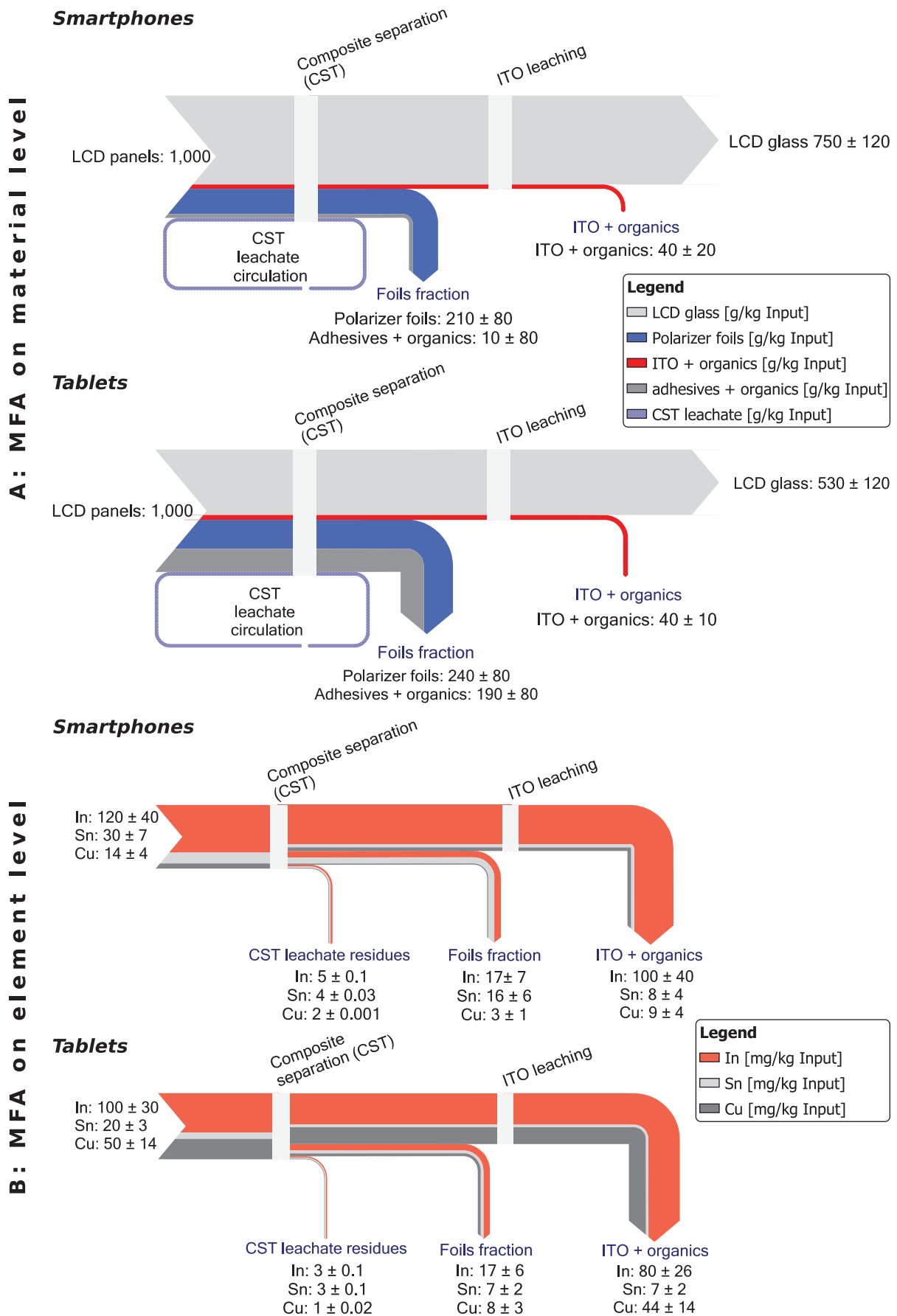


Figure 67: Mass flow on a material and an element level of a composite separation test carried out with samples from smartphones and tablets: **A** material level (top) and **B** element level (below). All data normalized to 1,000 g input material

#### Cross contaminations through toxic heavy metals + Sr

Not only the flows of strategically important elements like indium and tin or valuable materials like copper are of interest. Flows of possibly present toxic heavy metals + Sr must be investigated as well. Although these materials are supposed to be applied only to the glass, traces can be washed out and transferred to the generated output fractions. In total  $1,400 \pm 300 \mu\text{g As/kg}$  input material,  $20,000 \pm 8,000 \mu\text{g Cr/kg}$  input,  $450 \pm 70 \mu\text{g Pb/kg}$  input,  $9,400 \pm 2,500 \mu\text{g Sb/kg}$  input and  $38,000 \pm 6,000 \mu\text{g Sr/kg}$  input were dissolved and distributed in the separation test for smartphones. For tablets,  $400 \pm 40 \mu\text{g As/kg}$  input,  $500 \pm 100 \mu\text{g Cr/kg}$  input,  $350 \pm 70 \mu\text{g Pb/kg}$  input,  $4,200 \pm 1,400 \mu\text{g Sb/kg}$  input and  $55,000 \pm 10,000 \mu\text{g Sr/kg}$  input was mobile in the separation test. Sb was mostly transferred to the foils fraction as it was apparently applied to the polarizer foils. Pb was not found in the indium-rich output fraction. Further general statements cannot be deduced. The distribution of most of the toxic heavy metals + Sr into the various output fractions seems to be random. Table 16 shows the transfer coefficients of all substances investigated and chemically analyzed in both composite separation tests with smartphones and tablets.

#### Transformation of polarizer foils

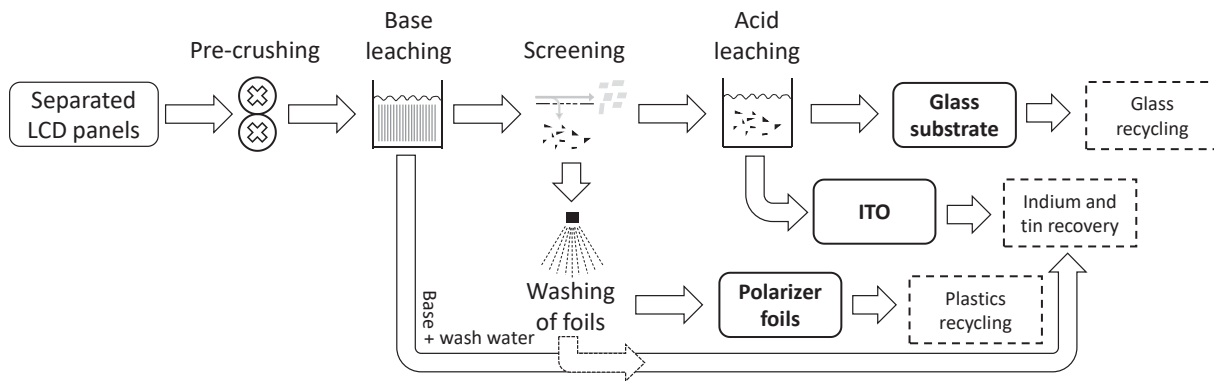
The base leaching process might affect the quality of the polarizer foils. A comparison of the flexibility of untreated polarizer foils with those from the separation tests revealed that treated foils were much more brittle. Therefore, 50 base-treated polarizer foils were investigated again by ATR-FT-IR. Only 7 out of 50 treated foils showed a spectrum of cellulose triacetate, while the majority turned out to be a material like cellophane which results from hydrolysis of the cellulose triacetate.

As polymer recycling was not a focus of this study, only minor work was performed to improve the quality of treated polarizer foils. One approach for optimization involves a shorter base treatment or the application of lower base concentrations. However, the authors do not expect that base treatment can be applied without the danger of hydrolyzing the target polymer, which would dramatically reduce the material value for recycling.

Therefore, another approach was tested, on a small laboratory scale, using a proprietary CreaSolv® formulation (Schlummer et al., 2006). Treatment of LCD devices with this solvent dissolved the polarizer films and enabled a separation of foil free glass substrates. After evaporation of the solvent from the polymer solution, the resulting recycled polarizer films turned out to be non-hydrolyzed cellulose acetate. Such a solvent based treatment step could be integrated into a future recycling concept for LCDs as it enables closed-loop-usage of the solvent and the production of purified recycled polymers if the polymer solution is subjected to appropriate filters before drying. However, the suitability and economic feasibility of such an approach need to be investigated in more detail.

#### Recommended processing of LCD panels

A first recommendation for the processing of LCD panels using the described approaches is shown in Figure 68. The pre-crushing step is supposed to break only the glass substrate, leaving the polarizer foils intact. The foils can then be sieved off after the base leaching step.



Own illustration (Ueberschaar et al., 2017d)

Figure 68: Recommended processing of LCD panels from screen devices for the recovery of indium, tin, glass substrate and plastics

Next, an explicit washing step of the separated polarizer foils is introduced to rinse off any potentially remaining traces of ITO which could lead to a lower transfer of indium in the output fraction. Furthermore, the base neutralized with the wash water can be forwarded to a subsequent indium recovery. The above discussed solvent approach for separation of glass and polarizer foils could replace the base leaching and the washing of the foils without affecting the other unit operations.

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#### 4.5.4. Tantalum capacitors

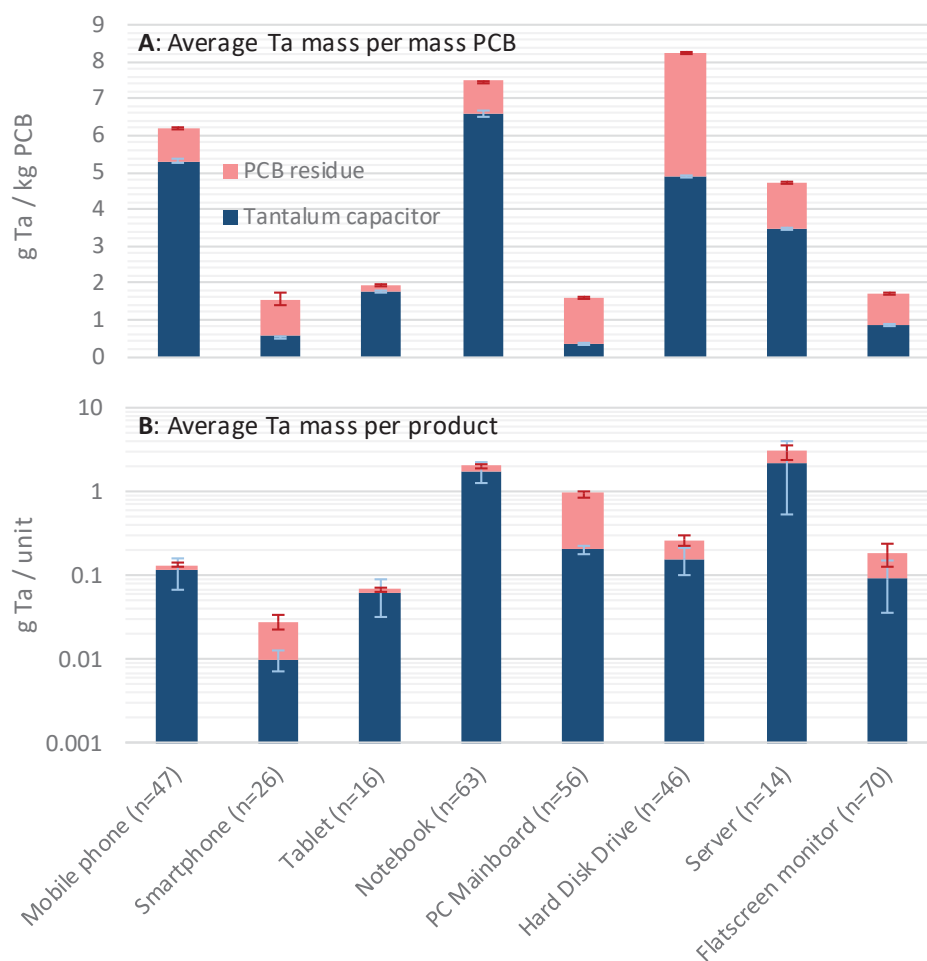
The removal of VICs does not transfer the total content of tantalum available on PCBs to a separate stream. Apparently, depending on their properties, not all applied tantalum capacitors can be identified. Furthermore, use of Ta in other components on a PCB is possible. Therefore, chemical analyses for the separated VICs and the remaining PCBs have been carried out (cf. supporting information S10 and S11 in Ueberschaar et al. (2016b)).

Figure 69 shows the resulting tantalum mass fraction per PCB and product type differentiated by the origin of the tantalum. The variations in results are much higher for g per product and are depicted on a logarithmic scale.

Across almost all devices investigated most tantalum was found in the VICs. Depending on the equipment type, different uncertainties for the separation of tantalum are given.

PCBs from notebooks display the highest overall tantalum content determined in PCBs (PCB including VICs) of almost 8 g/kg PCB. Furthermore, high levels can also be found in HDDs, servers, and mobile phones. HDDs contain the highest overall Ta content. Here, the PCB residues still have a tantalum load of 3 g/kg PCB<sub>residue</sub>. Therefore, the total content is over 8 g/kg PCB including VICs.

In smartphones and desktop PC mainboards, levels of Ta in PCB residues are higher than in the separated VICs. In contrast, very low overall Ta content was found in smartphones, with 1.5 g/kg PCB, and in PC mainboards and flat screen monitors, with 1.6 g/kg PCB. These figures are based on a considerably fluctuating tantalum content per unit, as the mass fraction of the PCB to the related unit varies, as does the number and mass of applied VICs.



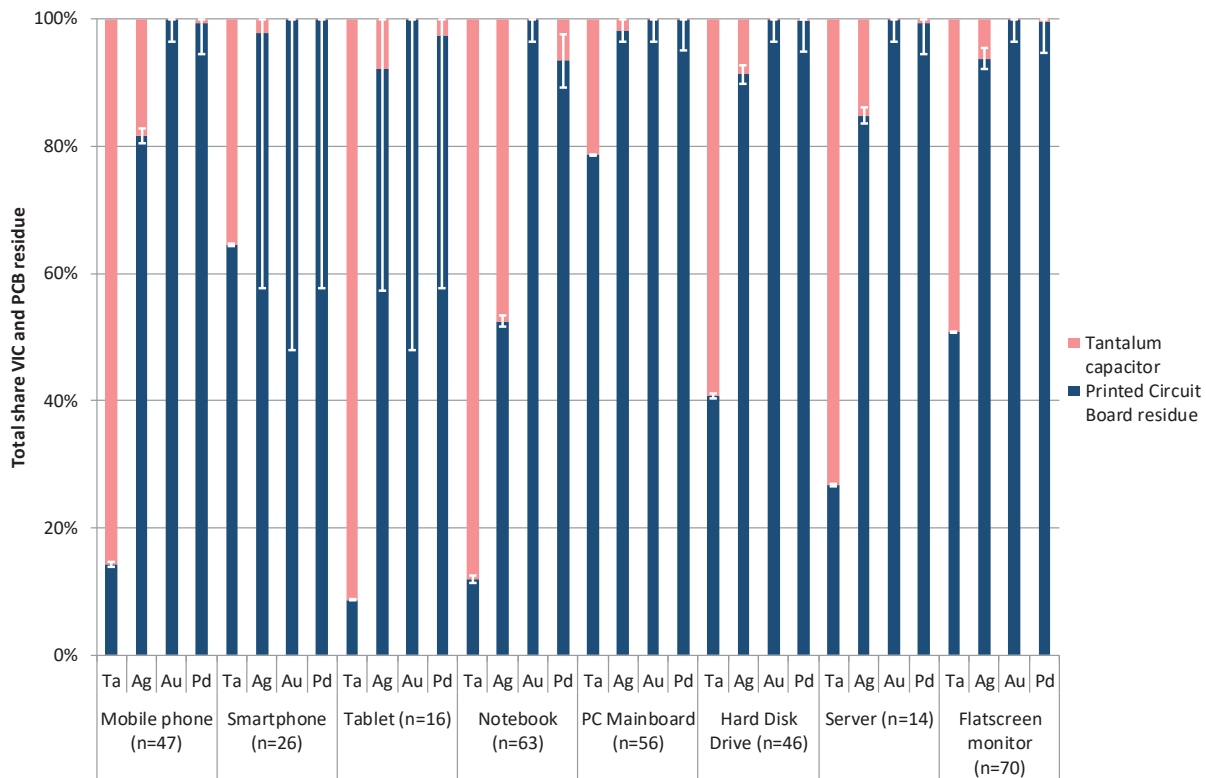
Own illustration (Ueberschaar et al., 2017a)

Figure 69: Average tantalum content per mass PCB (A) and product (B). Error bars show standard deviation

The results show a good correlation with results from other studies. Fujita et al., 2014 determined about 1 g/kg PCB for desktop PCs. PCBs from various sources have only minor tantalum concentrations, with about 0.2 g/kg PCB (Xuefeng Wen et al., 2005). In comparison, general tantalum grades of coltan range between 0.1 and 0.5 g/kg and are much lower than for the PCBs investigated in this study but correlate well with a mixed PCB input (U.S. Geological Survey, 2013).

The tantalum content in g can be calculated relative to the assembled PCB (g Ta per PCB), and also to the whole device (g Ta per device). With this information, the relevance of processing particular equipment types to separate the applied VICs can be compared. Highest contents were found in servers with 4-5 g/unit, followed by notebooks with ca. 1 g/unit. The tantalum content per unit of the very small PCBs in mobile phones, smartphones, tablets, HDDs, and flat screen monitors was low. Particularly in smartphones and tablets, many fewer VICs are applied.

The overall mass fraction of identified capacitors and hence the total quality of the removal process was investigated. For this, the load of one metal in the VIC and the load of the same metal in the PCB were added together to find a total load. The single loads from the two fractions were calculated as a share of this total value. The results are depicted in Figure 70. Here, the contrast in selectivity of tantalum and PM (gold, silver, and palladium) for the equipment types investigated is shown.



Own illustration (Ueberschaar et al., 2017a)

Figure 70: Comparison of the selectivity and retention of tantalum, silver, gold, and palladium (error bars show standard deviation)

The chemical determination of smartphones and tablets was carried out by a different laboratory from the one used for the analyses of the other equipment types. Therefore, other uncertainties related to the methodologies used influenced the results.

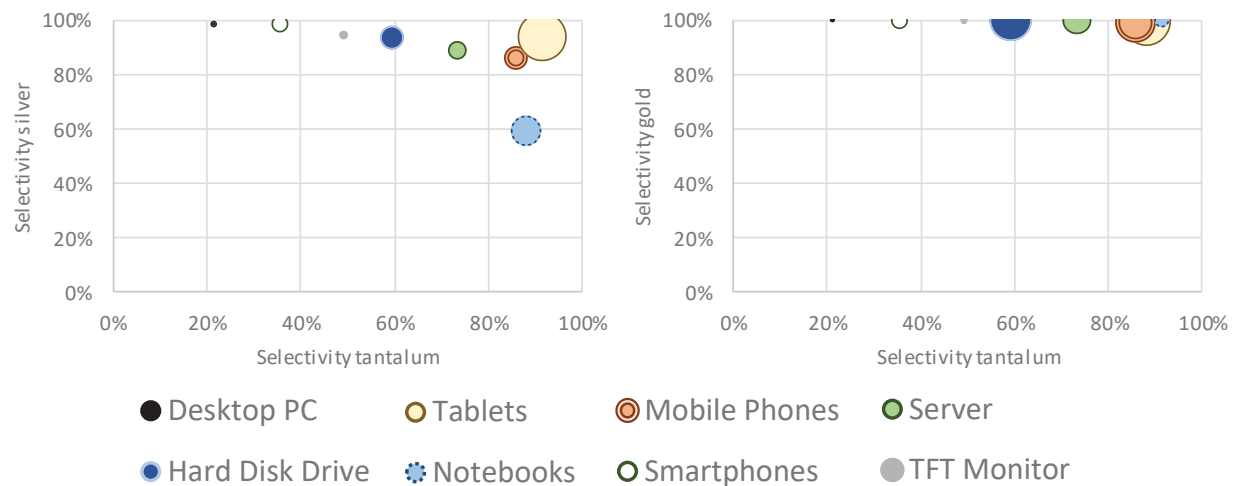
The tantalum content in the VIC fraction for tablets, mobile phones, notebooks, and servers is very high, which means a good visual identification of applied capacitors. In contrast, the tantalum load in the separated VICs compared to the load in the PCB residues is very low with under 50% for flat screen monitors, under 40% for smartphones and just under 20% for desktop PCs. This data reveals a very low visual identification and separation quota of tantalum-bearing components.

In most of the devices investigated a high percentage of the silver applied to the tantalum capacitors is removed. The example notebook shows that up to ca. 50% of silver can be removed via VIC separation. The lowest mass fraction of silver removed is encountered in desktop PCs and smartphones, for which the mass fraction of separated tantalum is also very low.

As expected, the separated mass fractions of other precious metals like gold and palladium are low and show that these elements are mainly applied in the residual PCB.

#### Selectivity tantalum and silver

The surprisingly high content of silver in tantalum capacitors was investigated in more detail, in order to check the impact of the removal of VICs on the recycling efficiency of silver, applying established processes on the entire PCB. The results of a comparison of the selectivity of tantalum and silver through VIC removal are shown in Figure 70 and covered in more detail in Figure 71.



Own illustration (Ueberschaar et al., 2017a)

Figure 71: Selectivity for silver and gold vs. tantalum related to a removal of tantalum capacitors

It is obvious that selective removal of tantalum is not possible for all the equipment types investigated. Silver is always applied in tantalum capacitors and is therefore removed with the VICs. The mass fraction of the silver removed compared to the total content of silver is dependent on the PCB of the equipment type investigated and on the number and weight of applied tantalum capacitors.

The trend of silver separation alongside VIC removal is positive. In other words, the higher the amount of VIC separation, the higher the amount of silver removal. Therefore, regular removal of VICs for purposes of tantalum recycling would lead unavoidably to a loss of silver. Interviews with tantalum producers, who also recycle new scrap, reveal that recycling of both tantalum and silver is not yet possible or economic (Marwede et al., 2016a; H.C. Starck, 2011).

A good example of this is notebooks. Here, a high selectivity of tantalum meets a relatively low selectivity of silver with only 50%. This means that a large percentage of the total silver available is transferred to the VIC fraction.

This has to be taken into account in any development of recycling processes as capacitor removal may interfere with the operating efficiency of established PCB recycling processes. The establishment of a recycling infrastructure for tantalum from capacitors requires the development of side-by-side recovery processes for tantalum and silver.

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#### 4.6. Recycling barriers and drivers

Results obtained from the recycling barrier analysis were converted into a modified SWOT diagram to depict opportunities and risks for the recycling of S-CRMs comprehensively.

##### 4.6.1. Integrated circuits

The relatively small shares of gallium-bearing components on PCBs lead to a dilution with other materials in recycling processes used conventionally. Ending in the pyrometallurgical process for copper and precious metals refining, gallium is transferred oxidized to the slag. Thus, gallium rich components must be separated prior to any mechanical processing with other material. In particular mobile phones and newly tablets are important gallium sources, bearing more than 40 % of the total gallium loads in the IT and entertainment equipment.

Major recycling barriers are lacking identification and separation technologies (cf. Table 17). The distinction between ICs and other components applied on PCBs is complex. Due to the chip design and connection type, a separation after removal is also complicated. Low chip masses relate to low loads of gallium with approximately only 1-2 mg Ga per chip.

Recycling drivers are the possibly increasing application of ICs, the growing market and increasing collection rates for mobile devices.

Table 17: Recycling barriers and drivers for gallium in ICs applied on PCBs

	Strengths		Weaknesses					
Opportunities	Increasing collection rates in future	High potential in mobile phones	First approaches for treatment of chips tested	Other radio technologies are available, increasing substitution of Ga by e.g. silicon-based technologies		Currently, relevant devices are collected in insufficient quantities		Currently, no recycling channels available
Threats	Recycling conflicts with gold and copper	IC and LED are main applications of Ga	Complex design of chips	No identification technology for chips available	No explicit labeling for doping elements	Gallium applied as trace metal, arsenic as contaminant for metallurgy	Material value low	Energy and chemicals consumption for thermal and chemical processing increasing for low-grade input

For separated ICs, this thesis proved the possibility of liberation and separation of gallium-containing components in ICs as a first approach. Relevant components with high gallium shares were separated. This material must be treated subsequently for a selective isolation of gallium from the copper, gold matrix. Arsenic as a contaminant has to be investigated in detail as well.

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#### 4.6.2. LCD panels

Main constraints for the recycling of LCD panels and a related recovery of indium are high costs and the generation of potentially high amounts of waste water in chemical and hydrometallurgical processes. Here, contaminants such as toxic heavy metals can wash out and influence subsequent processes. In general, the material revenues are low at the moment. Therefore, a newly developed and designed recycling process has to be very efficient not to exceed cost limits.

Strengths are a good basis of developed processes for the separation of the sandwich compound and a subsequent recovery of target metals. LCD panels are main components in relevant devices. Due to the depollution process for cold cathode background lighting in the pre-treatment, LCD panels are casually separated and are available for recycling processes. However, the transition to LED background lighting may hamper this driver. Also, a substitution of ITO would influence the indium recovery, as substance flows will be diluted.

WEEE collection rates of relevant screen devices are low at the moment. An increasing available mass flow of screen devices and LCD panels is expected in future.

Table 18 depicts the obtained results from the recycling barrier analysis for indium from LCD panels

Table 18: Recycling barriers and drivers for indium in LCD panels

	Strengths					Weaknesses		
Opportunities	LCD panels have to be separated due to possibly applied CCFL background lighting	No labeling of LCD panels necessary	No recycling conflicts with other valuable or strategically important materials	First approaches for the separation of foils and glass tested	Various approaches for indium separation and concentration tested	Materials applied in a sandwich compound	Collection rates of relevant screen devices low, but will increase in future	Currently, no recycling channels available
Threats	Transition to LED background lighting would hamper the casual separation of LCD panels		Possible substitution with ATO or other materials			Contaminants applied in LCD panel glass	Material value low	High input of chemicals for processing necessary

Note: CCFL = cold cathode fluorescence lamps

#### 4.6.3. Hard disk drives

The mass and share of NdFeB magnets in HDDs is relatively high. With almost 30% REE share, this magnet type constitutes a good basis for recycling. However, the liberation of NdFeB magnets from HDDs is complex. This applies in particular for the smaller magnets from the spindle motor (high Dy content). Liberation and separation technologies are not available on the market but are under research. The same applies for the recycling of the magnets. Currently, recycling channels do not exist. Hydrometallurgical approaches for single REE recovery consume a high amount of chemicals and energy.

Except for the nickel coating, the magnet is directly usable for a functional recycling, if the material is re-sintered to a new magnet. However, the composition of magnets changes according to demands for specific physical properties and market prices of raw material substitutes. This includes the use of dysprosium, which was applied in only a few magnets investigated. Moreover, this element was mostly found in magnets applied to the spindle motor.

Table 19 shows the results of the recycling barrier analysis for Nd, Table 20 for Dy.

Table 19: Recycling barriers and drivers for neodymium in NdFeB magnets in HDDs

	Strengths			Weaknesses		
Opportunities	Liberation technologies and recovery options in research	Only minor recycling conflicts with Al, Co and Ni	High mass fraction of Nd in magnets	Relevant WEEE mass flows low, but stable in future	Currently, no recycling channels available	No identification technology for magnets available
Threats	No valuable elements by-applied (Dy, if applied)			Miniaturization of magnets	Increasing variety of chemical composition	Sophisticated design, limited access

Table 20: Recycling barriers and drivers for dysprosium in NdFeB magnets in HDDs

	Strengths		Weaknesses			
Opportunities	Liberation technologies and recovery options in research	Only minor recycling conflicts with Al and Co	Less used in future due to high prices	Relevant WEEE mass flows low, but stable in future	Currently, no recycling channels available	No identification technology for magnets available
Threats	No valuable elements by-applied	High value of Dy	Sophisticated access to magnet and low mass	Dysprosium not always applied	Sophisticated design, limited access	Mass of Dy very low

#### 4.6.4. Tantalum capacitors

VICs analyzed consist almost half of tantalum. Plastics, silicon, copper, iron and silver represent other major materials. This simple design facilitates recycling processes. Usually, VICs on PCBs are directed to copper smelters with subsequent precious metals refining. Recovery of tantalum in this process is not feasible. However, VICs contribute to this process due to copper and silver contents. A prior separation of VICs is crucial for the recovery of tantalum but would lower the value of the PCB and slightly lower the overall efficiency of the refining process. Economically feasible technologies for the identification and separation of VICs are not yet available. Recovery processes for tantalum in VICs do exist but are related to high energy and chemicals input.

Investigated WEEE products showed an uneven distribution of VICs. Therefore, a focus on particular products leads to higher yields in recycling. However, the use of VICs seems to be decreasing. Miniaturization of tantalum capacitors will further reduce total tantalum loads and value. Moreover, due to its criticality, initial approaches have been made to substitute tantalum with niobium. This might reduce the need for a circular economy for tantalum in a long-term perspective.

Table 21 summarizes the results obtained from the recycling barriers analysis.

Table 21: Recycling barriers and drivers for tantalum in VICs

	Strength				Weaknesses			
Opportunities	Simple design of Ta caps	Available recovery strategies for tantalum exist	Less used materials in caps	No contaminants applied	Currently, relevant devices are collected in insufficient quantities	Reduction of applied tantalum through miniaturization and less numbers of used capacitors	Currently, no recycling channels available	
Threats	Recycling conflict with silver	Substitution of Ta with Nb	Capacitors have only a few, but mainly the same characteristics		No separation technology available through complex identification	High labor costs for manual separation	Energy and chemicals consumption for thermal and chemical processing increasing for low-grade input	Unsteady distribution in relevant equipment types Material value low

Major recycling barriers are lacking identification and separation technologies. First approaches are tested in research but are not applicable on an industrial scale for PCBs originating from consumer products. Due to the design, current processes cannot recover all elements applied such as tantalum, copper, and silver. New technologies have to be developed and are most possibly related to high consumption of chemicals and energy.

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## 5. Discussion

Discussions related to particular methodologies and results are part of the single articles of this cumulative thesis and are available in Ueberschaar et al. (2016a), (2016b), (2016c), (2016d); Ueberschaar and Rotter, 2015). The general adaptability and applicability of the results will be discussed in the following.

### 5.1. Comparison of recycling strategies

This thesis gives a fundamental background for developing holistic recycling strategies aiming at a circular economy for all materials applied in WEEE devices with a special emphasis on S-CRMs. Results show that the recovery of S-CRMs is often related to trade-offs, resulting in an inferior recovery efficiency of by-applied materials such as PMs and IBMs. However, to reduce investments and to increase the acceptance in the recycling sector, optimized recycling strategies must be modularly designed enabling the integration as a single recycling step in the recycling chain.

First approaches for optimized recycling processes have been tested in this thesis. A direct comparison of different recycling strategies is needed to assess the most suitable process for the recycling of WEEE. Summarizing all information from hotspot analyses, analyses of available mass flows and assessment for recyclability and recycling barriers, example processes are qualitatively compared. The fields “recyclability”, “technology”, “material economics” and the “market situation” are evaluated for state-of-the-art processes and processes optimized for S-CRM recovery.

Table 22 shows the evaluation matrix of this qualitative assessment for the comparison of recycling strategies.

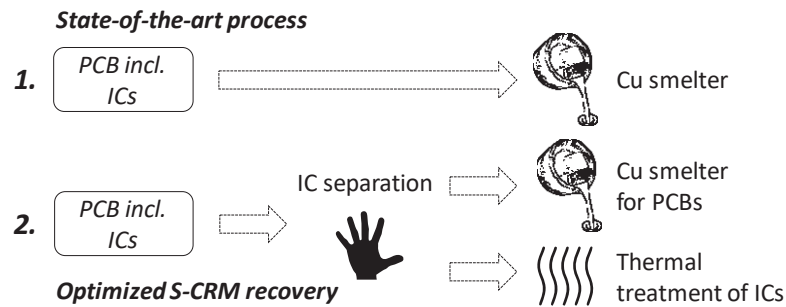
Table 22: Qualitative assessment of research results for the comparison of recycling strategies

Field	Focus	Assessment	
		State-of-the-art process	Process optimized for S-CRM recovery
Recyclability	IBM	<i>Estimation of the recyclability of IBMs, PMs, S-CRMs, and OMM (other major materials)</i>	
	PM		
	S-CRM		
	OMM		
Technology	Low residues	<i>Estimation of generated residue amounts</i>	
	Low chemicals	<i>Estimation of chemicals consumption</i>	
	Low energy	<i>Estimation of energy consumption</i>	
Material economics	Low investment	<i>Estimation of necessary investment for optimized process</i>	
	Revenue pricing	<i>Estimation of revenue for all generated output fractions</i>	
	Low purchase pricing	<i>Estimation of purchase price for input material</i>	
Market situation	Output	<i>Estimation of selling possibilities for all output materials</i>	
	Input	<i>Estimation of availability of sufficient input material</i>	

The listed points were qualitatively rated with ++ (very good), + (good), o (fair), - (bad), -- (very bad) for recycling strategies applied individually on ICs, LCD panels, HDDs, and tantalum capacitors (cf. Appendix 9). Results were then transferred to a graphical depiction.

### 5.1.1. Integrated circuits

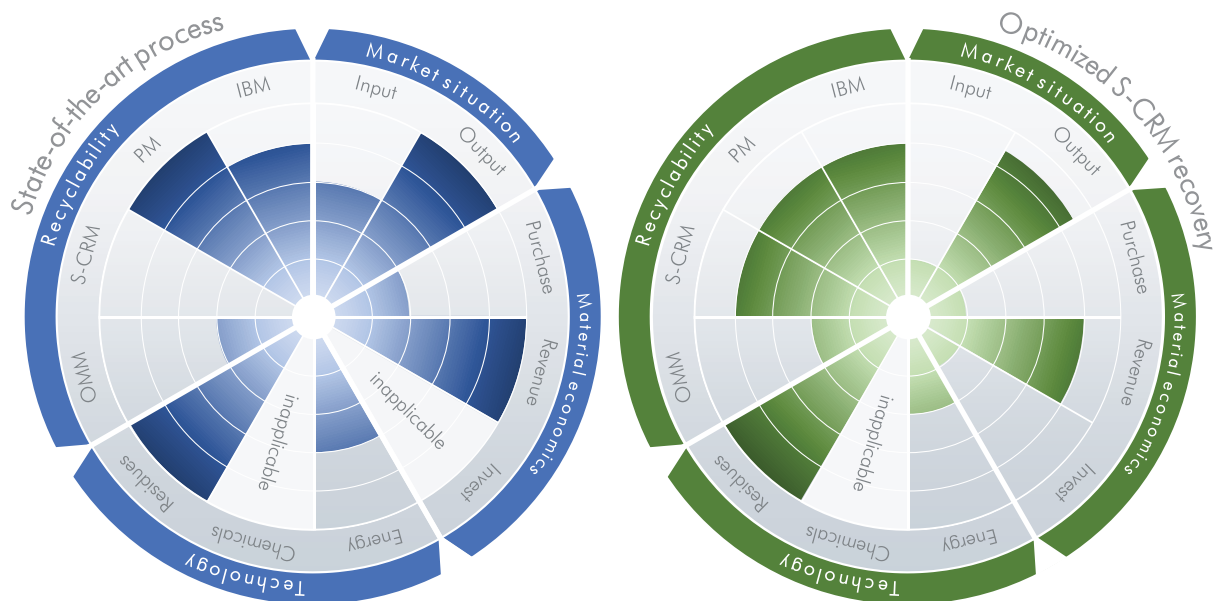
ICs are applied on PCBs, which are usually processed in a copper smelter. As there is no removal of the ICs prior to this process, the copper smelter can be seen as the state-of-the-art process for IC treatment. An optimized IC recycling approach is developed in this thesis described in chapter “4.5.2”. The compared technical recycling strategies include the PCB treatment in a copper smelter and an optimized process including additionally a thermal treatment of separated ICs (cf. Figure 72).



Own illustration

Figure 72: Recycling strategies compared for IC processing (integrated copper smelter vs. ICs separation + integrated copper smelter)

Appendix 9 shows the detailed comparison of the recycling strategies, while the results are quantitatively presented in Figure 73.



Own illustration

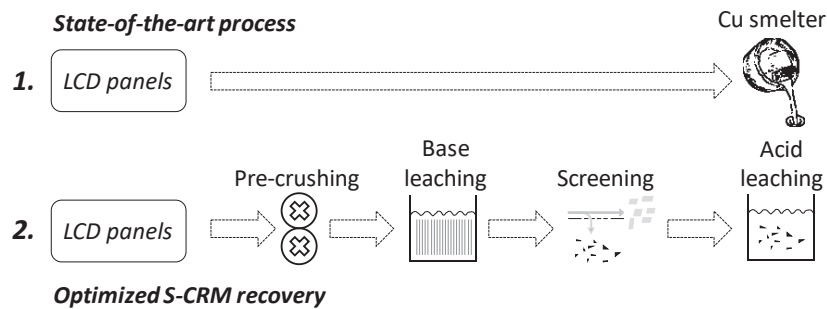
Figure 73: Recycling strategy comparison for ICs applied on PCBs (integrated copper smelter vs. ICs separation + integrated copper smelter) - Note: This figure depicts only a qualitative assessment; a high amplitude represents an optimized situation

Due to missing optical characteristics, the identification of ICs on PCBs is complex. This requires a high investment in new technology and lead to only moderate achievable efficiencies for gallium recovery in the optimized process. While the recyclability of IBMs is comparable, small amounts of gold are transferred to the IC output lowering the PM recyclability of the overall process. Due to the high mass fraction of plastics in PCBs, which is fueling the smelting process, only low amounts of residues are generated in both cases.

While the state-of-the-art process focusses on IBM and PM recovery from any PCB input, an optimized process for gallium recovery must focus on PCBs with high IC mass fraction. This jeopardizes the acquisition of input material and increases the purchase prices.

### 5.1.2. LCD panels

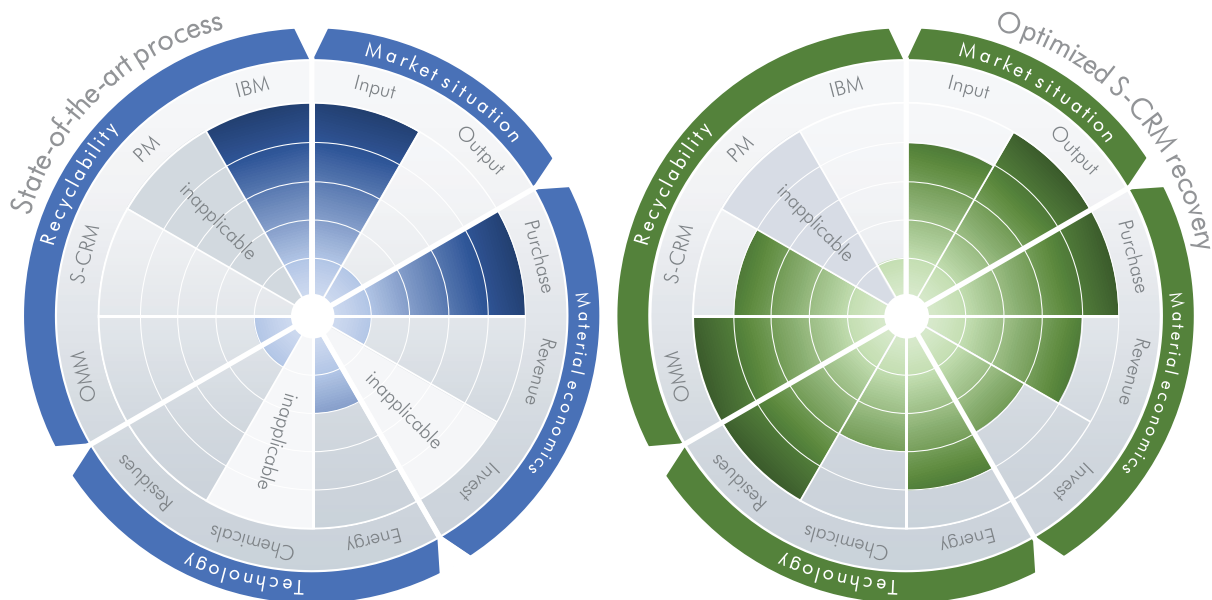
Currently, there is no standard recycling process for LCD panels. Due to the low content of valuable material, recyclers use various routes to discard disassembled LCD panels. Known routes are cement factories (plastics for energy input), copper smelter (plastics for energy input, copper content recoverable, and glass used as slag builder), and general landfilling. Here, the copper smelter is compared to the LCD panel process presented in chapter “4.5.3” (cf. Figure 74).



Own illustration

Figure 74: Recycling strategies compared for LCD panels processing (integrated copper smelter vs. base treatment + acid leaching step)

Appendix 9 shows the detailed comparison of the recycling strategies, while the results are quantitatively presented in Figure 75.



Own illustration

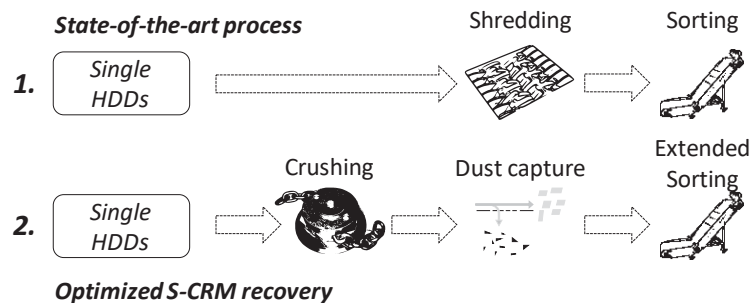
Figure 75: Recycling strategy comparison for LCD panels (integrated copper smelter vs. base treatment + acid leaching) - Note: This figure depicts only a qualitative assessment; a high amplitude represents an optimized situation

The optimized treatment of LCD panels for the recovery of indium, polarizer foils, and panel glass represents the best example of an upscalable process. While the processing of LCD panels in a copper smelter focusses on the copper recovery and using the plastics only as fuel, the optimized process can provide almost all materials applied in LCD panels as separate output fractions. Aiming at low-temperature reactions in the base leaching (polarizer foils separation) and in the acid leaching (ITO

recovery), energy can be saved. Currently, there is no appropriate route for LCD panels in the recycling chain, which leads to a cheap supply of input material. The plastics (cellulose acetate) and indium are perfectly saleable outputs. Only the presence of toxic heavy metals in the glass fractions might hinder a full recycling of LCD panels and impedes the sale of this output fraction.

### 5.1.3. HDD

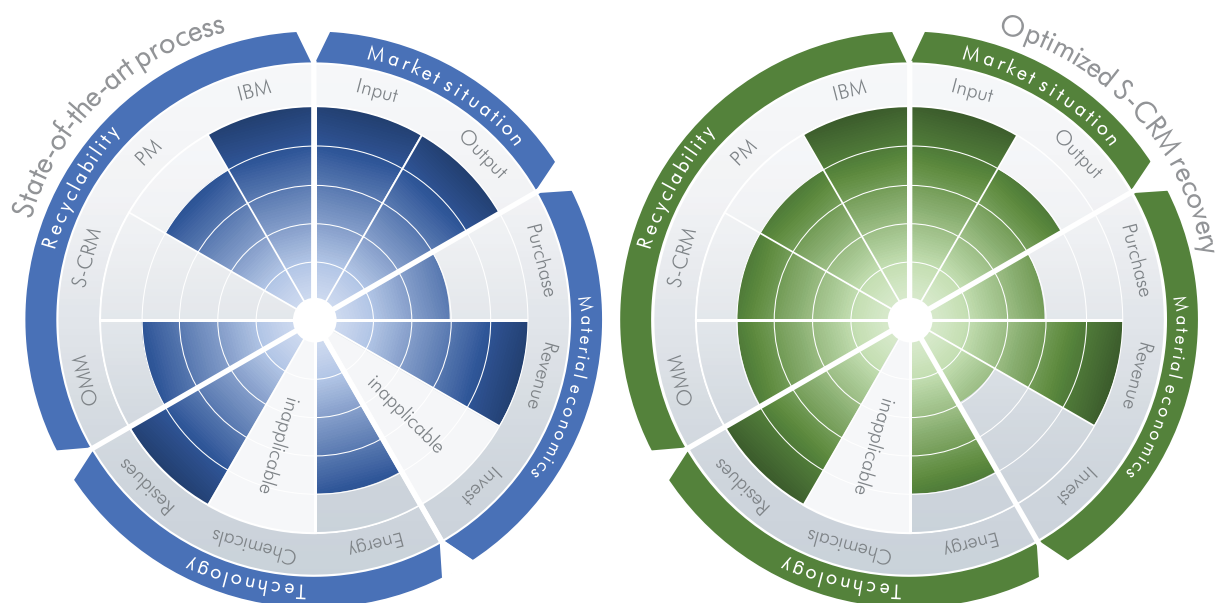
HDDs consist of a high share of IBMs, which are excellently separable with conventional mechanical processes. Due to applied PCBs in HDDs, additional high mass fractions of PMs increase the intrinsic value of these devices. Standard processes aim at a total destruction of the whole devices to gain data security. For a higher value added, PCBs can be separated beforehand and passed on to specialized processes. Manually working companies disassemble HDDs to recover PCBs, NdFeB magnets and to obtain a higher purity of the applied IBMs. As this is not a cost-effective procedure, the considered state-of-the-art process consists of shredding the HDDs including the PCBs and of subsequent sorting processes. This approach is compared to a specialized mechanical treatment of HDDs with a focus on NdFeB magnets separation to recover REEs (cf. Figure 76). Habib, Parajuly and Wenzel, 2015; Westphal, 2015; Muchow, 2016 traced back REEs in the mechanical treatment of HDDs.



Own illustration

Figure 76: Recycling strategies compared for HDD processing (shredding + sorting vs. optimized crushing + optimized sorting)

Appendix 9 shows the detailed comparison of the recycling strategies, while the results are quantitatively presented in Figure 77.



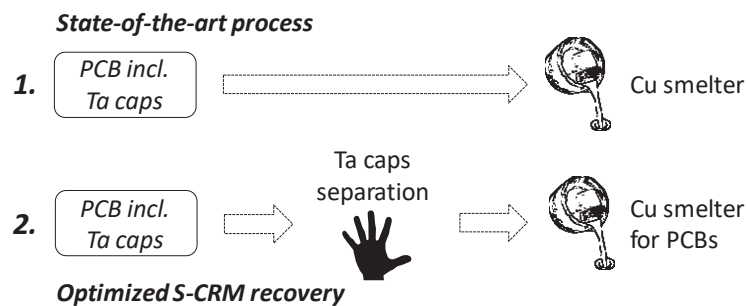
Own illustration

Figure 77: Recycling strategy comparison for HDDs (shredding + sorting vs. optimized crushing + optimized sorting) - Note: This figure depicts only a qualitative assessment; a high amplitude represents an optimized situation

Both recycling strategies consist solely of mechanical treatment and sorting processes and are therefore highly comparable. The optimized recycling process aims at a specialized crushing process to liberate the applied NdFeB magnets. High impacts pulverize the magnets, which leads to an enrichment of the magnets in the fine grains. Dust capture and sizing processes provide a REE concentrate. The high share of NdFeB magnets in HDDs supports the feasibility of this approach.

#### 5.1.4. Tantalum capacitors

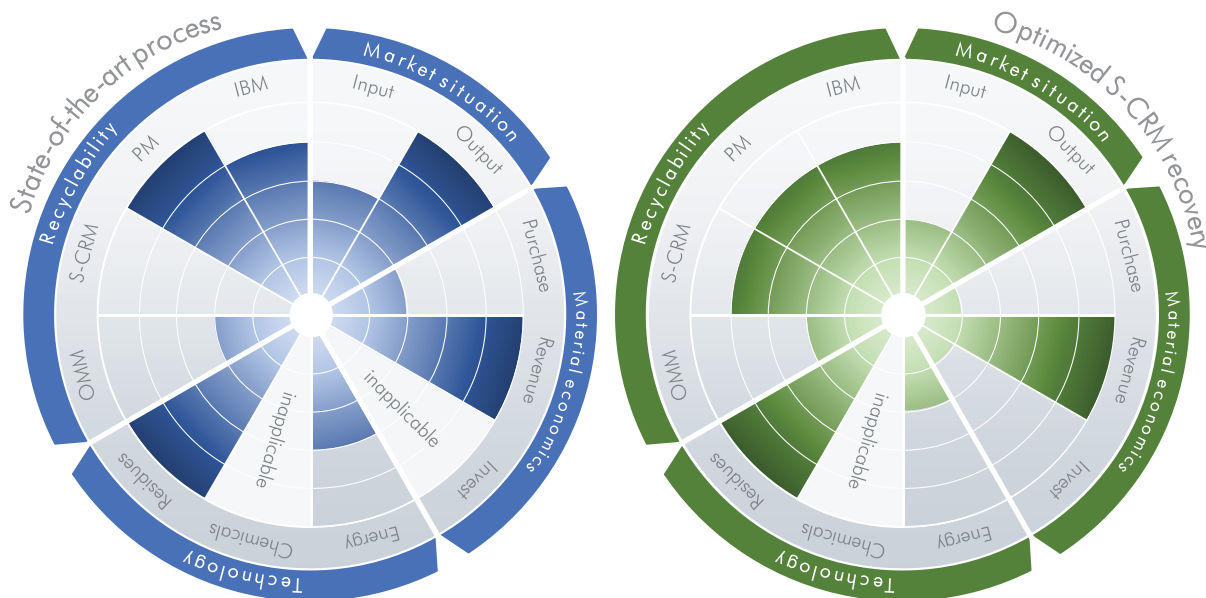
Tantalum capacitors are applied on PCBs, which are usually processed in a copper smelter. First approaches for a separation of tantalum capacitors prior to this process are tested (Kreibe and Förster, 2016; Stenzel, 2016; Rotter et al., 2016). A manual separation was evaluated in chapter “4.5.4”. The compared recycling strategies include the PCB treatment in a copper smelter, while the optimized process additionally includes the separation of the tantalum capacitors from the PCBs cf. Figure 78).



Own illustration

Figure 78: Recycling strategies compared for tantalum capacitor processing (integrated copper smelter vs. separation of tantalum capacitors + integrated copper smelter)

Appendix 9 shows the detailed comparison of the recycling strategies, while the results are quantitatively presented in Figure 79.



Own illustration

Figure 79: Recycling strategy comparison for tantalum capacitors (integrated copper smelter vs. separation of tantalum capacitors + integrated copper smelter) - Note: This figure depicts only a qualitative assessment; a high amplitude represents an optimized situation

Due to the high tantalum grade, tantalum capacitors are highly suitable for tantalum recycling. The characteristic design of tantalum capacitors enables an optical separation. The recyclability study in

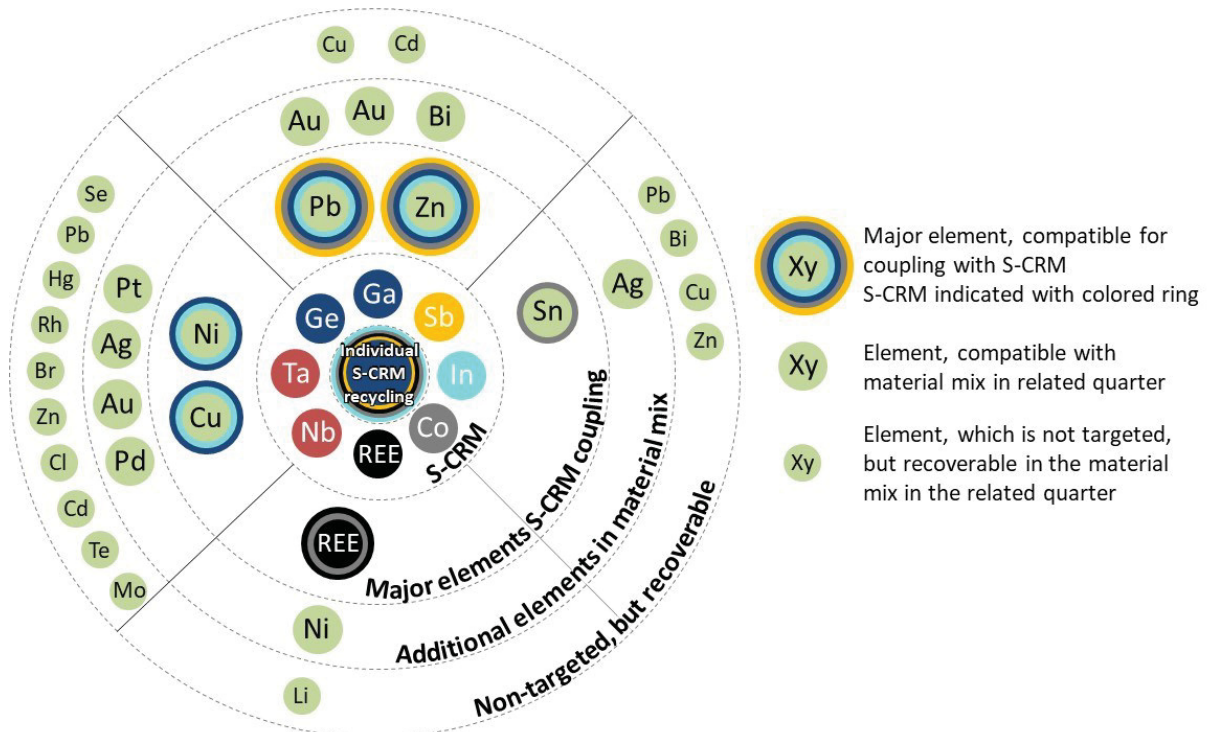
this thesis revealed a limited separation of tantalum from PCB via tantalum capacitors removal. Therefore, higher investments in new technologies are necessary. Due to the transfer of silver to the tantalum capacitor output, the recyclability of PMs is limited.

While the state-of-the-art process focusses on IBM and PM recovery from any PCB input, an optimized process for tantalum recovery must focus on PCBs with high tantalum capacitor mass fractions. This jeopardizes the acquisition of input material and increases the purchase prices.

## 5.2. Design-for-recycling<sup>1</sup> recommendations

The recycling-oriented product characterization and the conceptual recyclability assessment revealed the complexity of investigated WEEE products and the limits of current, but also new recycling approaches, for S-CRMs. Main technical recycling barriers are low mass fractions of S-CRMs and the presence of other accompanying elements. The first reduction of complexity is usually achieved via manual disassembly or mechanical liberation of S-CRM carrying components such as LCD panels / ITO, tantalum capacitors, NdFeB magnets, etc. Design-for-recycling can support the accessibility and the liberation of such components. First recommendations are depicted in Marwede et al., 2016b by using inter alia results of the case studies presented in this thesis.

The main focus of design-for-recycling strategies should be set on a general product design, which uses selective element mixes, which are compatible with end-processing steps. The conceptual recyclability assessment proved the incompatibility of various elements applied in WEEE components investigated. Via such “element design-for-recycling” recommendations, manufacturers would be able to design components building element clusters, which are separable in hydro- or pyrometallurgical processes. Basing on current recovery routes of metals, an approach for such element clustering is depicted in Figure 80.



Own illustration based on (Reuter et al., 2011; Nakajima et al., 2009; Nakajima et al., 2011; Shuva et al., 2016)

Figure 80: Compatible elements for S-CRM clusters in current recovery processes for an “element design-for-recycling”

<sup>1</sup> “The designed-for-recycling method incorporates recycling and recyclability criteria into the design phase of products, with the aim of obtaining recycled and/or recyclable products” (CCR Information and query response service, 2003)

The radar chart shows the S-CRMs and compatible elements for clustering in materials, material composites or components, indicated by colored rings in corresponding colors. Thus, Ga, Ge are compatible with Ni, Cu, Pb, and Zn. Further additional elements are allowed in the material cluster and are sometimes even welcome like PMs. Further minor shares of other elements may be present but are not targeted in the recovery process. Other examples are: Co with REE or Pb and Zn; Sb with Pb and Zn or In in Ni, Cu or Pb, Zn. Ta and Nb are exceptions as there are no compatible standard recovery routes. Thus, Ta and Nb cannot be clustered and have to be processed individually.

A clustering as suggested here does not lead necessarily to a recovery of S-CRMs. However, it enables the technical possibility and facilitates recovery processes. Bundling waste streams with similar S-CRM contents could increase related mass fractions in the material mixes processed. Existing metallurgical routes were usable for the recovery of S-CRMS.

### 5.3. Further use of data

In this thesis, potentials and recycling strategies were assessed for WEEE products carrying S-CRMs and other materials to be considered for further the development of recycling strategies. Studies were carried out using the practical examples of gallium, copper & gold in ICs, indium, glass & polarizer foils in LCD panels, neodymium & dysprosium in NdFeB magnets, and tantalum & silver in tantalum capacitors. All exemplary conducted studies were carried out individually for each product based on their physical and chemical properties. Thus, used methodologies can be adapted for the characterization of any other WEEE product to investigate S-CRM potentials and develop recycling strategies.

Third party use of results provided with this thesis is wanted. However, the individual suitability of data has to be checked first, before using them for other purposes.

#### 5.3.1. Material flow data

MFA data provided in this thesis represents very specific data and should therefore not be used for any extrapolations in its native form. This applies explicitly to the presented transfer coefficients. To ensure a high significance for further calculations, data have to be aggregated and compared with other MFA results. A standardized classification of the data sets is mandatory. Databases like the “UMKDP - Urban mine knowledge data platform” of the ProSUM project (Horizon 2020 - N° 641999) represents a centralized database for all available data and information on arising, stock and treatment of WEEE, but also EOL vehicles (ELVs), batteries and mining waste.

#### 5.3.2. Product data

A high number of WEEE products were sampled and investigated to obtain product-centric information on gallium, indium, tantalum and REE contents. However, the chemical composition changes for each investigated WEEE product group and also within WEEE products in the same product group. This applies for S-CRMs, but also for other potentially valuable elements, which are in focus of current recycling strategies.

The samples investigated represents a very specific set of devices and components from a limited number of manufacturers and manufacturing dates. Extrapolations on this basis are feasible. For a higher significance, sample quantities have to be extended and further compared with other product data.

## 5.4. General methodological discussion

### 5.4.1. Sampling

Results in this thesis show a great influence of sampling procedures on obtained results. In chapter 3.3.2 “Extended batch test in WEEE pre-processing”, methodologies were described how to handle the sampling of heterogeneous materials in batch tests and how sampling uncertainties can be calculated theoretically and assessed practically. In contrast, uncertainties obtained via recycling-oriented product characterization base on the sample’s heterogeneity only. A sampling uncertainty in this context is not addressed. However, there are no guidelines for taking representative samples with unknown quantity and heterogeneity. The only approach to generate reliable results is to increase the number of samples respectively of WEEE products investigated.

### 5.4.2. Chemical characterization

Most information provided in this thesis bases on chemical characterization. Chemical analysis of S-CRMs and other minor metals in WEEE represents a very young subject in this research field. No standardized approaches or guidelines are available. Thus, methodologies have to be developed and validated individually for each new material to be measured. Varying results based on random or systematic effects can lead to much higher or lower loads of the element being investigated.

Also, the detection limits of measurement devices raise similar problems, as element mass fractions below those limits are not considered mass shares or mass flows. Consequently, total flows can be systematically underestimated if this flow is set to zero, or systematically overestimated if the limit of detection (LOD) is considered as the average value. To improve the sensitivity of analyses, further fractioning of materials might be useful prior to a chemical characterization to achieve fractions above the detection limit and to identify hotspots.

☛ (Ueberschaar et al., 2017b)

### 5.4.3. Mathematical model

Data usually provided in literature is expressed as a range, or arithmetic mean with median and standard deviation. This depiction of data is easy to use but lacks background information about the sample characteristics. Many approaches exist for providing more detailed information. By generating large amounts of single values, the variability of each material investigated can be determined (Laner et al., 2014). Related distribution patterns can then be drawn which not only supply information about the expected average values, including uncertainties but also about the variability of the mass flow being studied, minimizing systematic effects. However, to achieve compatibility with other datasets, data has to be aggregated, which always results in a loss of information (Schwab et al., 2017). This is particularly true in the case of variability data retrieved for example by using sampling methodologies or aggregated literature data from various sources. Conventionally, further calculations assume a statistically uniform distribution of values like a normal distribution, which simplifies the uncertainty propagation but might lead to incorrect results. Mass fractions of REEs in NdFeB magnets and amount of VICs applied on PCBs are good examples in this thesis for not uniformly distributed data sets.

Using distribution patterns helps to evaluate the data statistically. If needed, a subsequent mathematical processing like distribution fitting and a calculation of the expected value as well as a determination of the variability of material flow properties can be carried out (Korf, 2016). However, a conventional uncertainty propagation cannot be calculated on this kind of data basis. One possible solution is the adapted use of the Monte Carlo simulation, which estimates the probability distribution of the calculated model (Müller et al., 2014; Laner et al., 2014; Heukelem et al., 2004). This would provide greater statistical safety for the overall results.

Case specific mathematical models need to be developed for each application. However, appropriate mathematical operations able to handle these data sets are not yet available in a structured way. The models currently in use result in a high-level aggregation of data.

Further research is needed on the topic of mathematical models, particularly regarding uncertainty propagation using extended uncertainty information. A consistent guideline on the use of appropriate tools for case-specific models should be provided.

© (Ueberschaar et al., 2017b)

## 6. Conclusions and outlook

### 6.1. General conclusions and outlook

Due to their criticality, S-CRMs need to be recycled. Successful identification, liberation, and separation allow for an individual recovery of S-CRMs or other functional materials, increase the purity of end-products, and help to keep processing costs down. This thesis provides a consecutive methodology applicable on material flows and WEEE products for assessing product-centric information and general recycling barriers and drivers. This fundamental knowledge base allows for the development of recycling strategies and supports the holistic recovery of all materials applied in WEEE such as S-CRMs, PMs, IBMs, and other bulk materials. Thus, an extension with additional non-product-related information enables the further development of general political recycling strategies implementable e.g. in the European WEEE directive, national laws or in in-house business plans of recycling facilities. Also, technical and economic recycling strategies are supported, which are used for the technical set-up in recycling plants and the selection of materials to be recovered.

However, practical results from product analyses prove a limited access to relevant components in WEEE and only low S-CRMs mass fractions. Furthermore, a standardization of products is not practiced. Due to manufacturers' demands and developments in manufacturing and product design, the application of S-CRMs and also contaminants changes from product to product and over time. In this thesis, a time trend towards less applied material and a general miniaturization is evidenced for some products. The continuous technology shift in new consumer products amplifies this effect. In short time intervals, new products replace conventional products, which have not yet started to return to the end-of-life material pool.

Recyclability assessments confirm the technical possibility of S-CRM recovery from WEEE. Tested approaches provide S-CRM mass fractions up to 30-50% in generated outputs. However, results show that the recovery of S-CRMs is often related to trade-offs, resulting in an inferior recovery efficiency of by-applied material such as PMs and IBMs, higher environmental impacts, or increased costs. Consequently, a direct comparison of newly developed and state-of-the-art recycling strategies is mandatory to assess the most suitable process for the recycling of WEEE. To reduce investments and to increase the acceptance in the recycling sector, optimized recycling strategies must be modularly designed enabling the integration as a single recycling process in the recycling chain.

Only low masses are available for recycling purposes dependent on the focused S-CRM and the carrying WEEE product. Data from 2013 show, that WEEE cannot currently cover the S-CRM demand to produce new similar goods. From a long-term perspective, market saturation of related products will reduce this gap. Furthermore, results from the material flow analyses prove a generally lower relevance of S-CRM recovery from WEEE. Higher yields can be gained by reducing losses in the primary production of S-CRMs, in refining steps, and in the manufacture of semi-finished products.

Design-for-recycling represents a key factor for improving the recyclability of S-CRMs. First approaches for structuring recommendations were made in literature but focus on accessibility and liberation of components carrying S-CRMs in WEEE products only. This thesis proves the presence of various by-applied materials and elements, which cannot be recovered in end-process steps. Thus, potential guidelines must include overviews, which elements can accompany others to be separable in subsequent metallurgical processes. As a consequence, circular economy could include the recovery of all elements applied and could replace the current practice of focusing on most valuable materials only.

## 6.2. Methodological conclusions and outlook

Methodologies applied in this thesis are not standardized and were either individually developed or adapted to provide detailed and reliable information about WEEE material flows and products. Some methodological core findings can be summarized as follows:

- WEEE material flow and product analyses require the development of new approaches (e.g. extended batch test in WEEE pre-processing) and the adaption of procedures already in use (e.g. recycling-oriented product characterization).
- The assessment of uncertainties and an appropriate uncertainty propagation receives only minor attention in comparable studies. Traditionally, discrepancies between the measured value and the real (unknown) value base on two components:
  - o Systematic effects base on recognized effects, which shift and falsify the results. This effect cannot be eliminated, but minimized. For example, by using correction factors.
  - o Random effects arise from unpredictable or stochastic temporal and spatial variations. Unlike the systematic effects, these effects cannot be compensated, but they can be reduced by increasing the number of observations.
- Analytical application on samples originating from WEEE is challenging due to heterogeneous material and due to highly fluctuating mass fractions of elements to be measured and of other by-applied elements. Moreover, analytical methodologies for WEEE are not yet well investigated.
  - o Sampling is one of the key factors for reliable data, basing to a large extent on a priori knowledge of materials and material distribution. Sampling uncertainties depending on sample size and way of sampling have to be determined practically or theoretically.
  - o Mechanical or thermal sample preparation can change the original composition of the sample.
  - o Additional chemical characterization with complementary techniques helps to identify and to understand systematic effects, which lead to an over- or under-determination of present mass fractions and loads. This is important in particular for PMs and S-CRMs.
  - o The chemical characterization is very sensitive to matrix effects due to the presence of accompanying elements. Element and matrix specific adaptation of analytical methods is required.
- Statistical evaluation is highly important to generate reliable data
  - o Data sets obtained from investigated WEEE products and material flows are often not uniformly distributed. Conventional (Gaussian) error propagation is not always applicable but commonly used.
  - o Typically calculated uncertainties lead to a loss of information as data sets are aggregated to just one value or range. An alternative enabling the use of extended uncertainty information including the variability of measured samples is required.
  - o New mathematical approaches are needed for providing and presenting aggregated data and for the propagation of errors accordingly.

Further research should focus on enlarging the knowledge base to improve the accuracy and precision of future studies. Additionally, the focus should be on standardized matrix adaptation of chemical analytical methods and mathematical models for a reliable statistical evaluation.

## Glossary

<b>Batch test</b>	A batch test is manual or mechanical processing of a definite and well-defined amount of appliances or fractions thereof to determine the yields and compositions of the resulting output fractions. (NVMP Association, 2014)
<b>Chemical characterization</b>	Chemical characterization consists of all procedures for the determination of the chemical composition of any substance. This includes the sampling, sample preparation and chemical analysis.
<b>Component</b>	An essential part of an appliance providing major functions.
<b>Concentration</b>	In chemistry, concentration is the abundance of a constituent divided by the total volume of a mixture. The term concentration can be applied to any kind of chemical mixture, but most frequently it refers to solutes and solvents in solutions. (IUPAC, 2014)
<b>CRM</b> - critical raw materials	Raw materials are labeled “critical”, when the risks of supply shortage and their impacts on the economy are higher compared with most of the other raw materials. Definition following (European Commission, 2010).
<b>Design-for-recycling</b>	The designed-for-recycling method incorporates recycling and recyclability criteria into the design phase of products, with the aim of obtaining recycled and/or recyclable products (CCR Information and query response service, 2003).
<b>EEE</b> – electrical and electronic equipment	Electrical and Electronic Equipment is defined as equipment which is dependent on electric currents or electromagnetic fields in order to work properly and equipment for the generation, transfer and measurement of such currents and fields. As a general rule of thumb, if it has a battery or needs a power supply to work properly, it is EEE and there are structures in place to reuse/recycle this equipment when it reaches end of life. (EPA, 2017)
<b>End-of-life (product)</b>	End-of-life (EOL) of a product is a term with different meanings dependent from the user’s profession. From a vendor’s perspective, EOL indicates the end of the planned lifetime of a product purchased by a customer or the planned end of support of a product, which results in the stop of marketing, selling, or rework sustaining the product. From a customer’s perspective, EOL of a product stands for the end of wanted use of an owned product, due to outdated technics or loss of its functionality.
<b>Error</b>	In general, a measurement has imperfections that give rise to an error. The error is the result of a measurement minus a true value of the measurand. Traditionally, an error is viewed as having two components, namely, a random component and a systematic component. The error is an idealized concept and errors cannot be known exactly. Adapted from (Joint Committee for Guides in Metrology, 2008)

<b>Extended batch test</b>	An extended batch aims at the same goals and procedures of a batch test, but includes additional methodologies to provide reliable data.
<b>Functional recycling</b>	Functional recycling is that portion of EOL recycling in which the metal in a discarded product is separated and sorted to obtain recyclates that are returned to raw material production processes that generate a metal or metal alloy. Often it is not the specific alloy that is remelted to make the same alloy but any alloys within a certain class of alloys that are remelted to make one or more specific alloys. For example, a mixture of austenitic stainless-steel alloys might be remelted and the resulting composition adjusted by addition of reagents or virgin metal to make a specific stainless-steel grade. (Graedel et al., 2011)
<b>Flow</b>	A flow is defined as a “mass flow rate.” This is the ratio of mass per time that flows through a conductor, e.g., a water pipe. The physical unit of a flow might be given in units of kg/sec or t/year. (Brunner and Rechberger, 2004)
<b>Goods</b>	Goods are defined as economic entities of matter with a positive or negative economic value. Goods are made up of one or several substances. (Brunner and Rechberger, 2004)
<b>Hotspot</b>	A part of a WEEE material flow or product, which holds a relatively high amount or share of a target substance.
<b>IBM</b> – industrial base metals	A common metal in the producing industry that is not considered precious, such as aluminum, copper, iron, tin, or zinc. Adapted from (Oxford Dictionaries, 2017)
<b>IC</b> – integrated circuit	An integrated circuit (IC) is a small semiconductor-based electronic device consisting of fabricated transistors, resistors and capacitors. Integrated circuits are the building blocks of most electronic devices and equipment. An integrated circuit is also known as a chip or microchip. (Techopedia, 2017)
<b>Liberation</b>	Liberation refers to all manual, mechanical, chemical or physical measures to strip down unwanted materials and obtain the single target appliance, material or substance.
<b>Life-cycle</b>	Life-cycle refers to the stages of a product's life from raw material extraction through materials processing, manufacture, distribution, use, repair and maintenance, and disposal or recycling.
<b>Mass fraction</b>	The mass fraction is the ratio of one substance's mass to the mass of the total mixture. (IUPAC, 2014)
<b>Material</b>	MFA material serves as an umbrella term for both substances and goods. (Brunner and Rechberger, 2004)
<b>Minor metals</b>	Minor metals is a widely used term in the metal industry that generally refers to not common, primary metals not traded on formal exchanges, such as the London Metal Exchange (LME). Adapted from (minor metals trade association, 2017)

<b>MFA</b> – material flow analysis	Material flow analysis (MFA) is a method to describe, investigate, and evaluate the metabolism of anthropogenic and geogenic systems. MFA defines terms and procedures to establish material balances of systems. [...] MFA includes balances of both goods and substances. (Brunner and Rechberger, 2004)
<b>Non-functional recycling</b>	Nonfunctional recycling is that portion of EOL recycling in which the metal is collected as old metal scrap and incorporated in an associated large-magnitude material stream as a “tramp” or impurity elements. This prevents dissipation into the environment but represents the loss of the metal’s function, as it is generally impossible to recover it from the large-magnitude stream. Although nonfunctional recycling is termed a type of recycling, it leads to an open metal life cycle, as discussed above. Examples are small amounts of copper in iron recyclates that are incorporated into recycled carbon steel. (Graedel et al., 2011)
<b>Process(ing)</b>	A process is defined as the transformation, transport, or storage of materials. Materials are transformed throughout the entire industrial economy on different levels. Transformation takes place in primary production processes such as in the mining and metal industry, where metals are extracted from mineral ores. Consumption processes such as private households transform goods into wastes and emissions. (Brunner and Rechberger, 2004)
<b>Pre-processing</b>	Pre-processing describes the first treatment of WEEE in the recycling chain via manual or mechanical processes. Through liberation and separation, secondary raw materials are channeled into designated recycling processes. Generated outputs have to be either processed further or can be used directly in final recovery processes.
<b>Random effect</b>	In general, a measurement has imperfections that give rise to an error in the measurement result. Traditionally, an error is viewed as having two components, namely, a random component and a systematic component. Random error presumably arises from unpredictable or stochastic temporal and spatial variations of influence quantities. The effects of such variations, called the “random effects”, give rise to variations in repeated observations of the measurand. Although it is not possible to compensate for the random error of a measurement result, it can usually be reduced by increasing the number of observations; its expectation or expected value is zero. (Joint Committee for Guides in Metrology, 2008)
<b>Recovery</b>	(Resource) Recovery is the selective extraction of disposed materials for a specific next use, such as recycling, composting or energy generation in order to extract the maximum benefits from products, delay the consumption of virgin resources, and reduce the amount of waste generated. (Grand Traverse County, 2017)

<b>Recyclability</b>	The ability of waste products to be recycled, based on product attributes and used recycling techniques. Adapted from (International Electrotechnical Commission, 2012; Ardente and Mathieux, 2012)
<b>Recycling</b>	Recirculation of production and consumption waste (also: waste heat) into the economic cycle (Springer Gabler Wirtschaftslexikon, 2017)
<b>Recycling chain</b>	The combination of recycling processes enabling the recycling of material streams or appliances.
<b>Recycling efficiency</b>	The efficiency of all related processes in the recycling chain for the recycling of a target substance.
<b>Recycling process</b>	Single process, which is used for the processing (transformation) of a waste stream. For the recycling of waste, a combination of recycling processes, the so-called recycling chain, is typically necessary.
<b>Recycling strategy</b>	The recycling strategy describes the goals of recycling efforts (political, economic) and the related selection of recycling processes or the recycling chain (technical).
<b>Recycling quota</b>	The share of recycled substances based on the recycling efficiency.
<b>Recycling-oriented product characterization</b>	Recycling-oriented characterization is defined as a systematic approach to support the design and the operation of recycling processes. It provides and requires a deep understanding of the needs for recycling processes, specifically focusing on the prevention of avoidable material mixing and dilution while also providing relevant information for the design of new EEE (Chancerel and Rotter, 2009)
<b>Sampling</b>	Sampling is a necessary physical handling step in many fields of industry and the science behind it has its basis in chemistry, physics, and mathematics. [...] The main idea of taking a correct sample includes the withdrawal of a sufficiently enough amount of material for it to be representative for the total amount of material. From a practical point of view, it is essential to know the minimum amount of material that can be regarded as being representative. Simultaneously, it is of importance to know how accurate an obtained sample is, i.e., preferably, the deviation between the true properties of the total batch and the amount that has been withdrawn as a sample needs to be known. (Dihalu, 2012)
<b>S-CRM – selected critical raw materials</b>	A reduced set of those metals rated as critical by the European Commission (European Commission, 2014b; European Commission, 2010). This set excludes materials, which are in focus of current recycling strategies (i.e. precious metals).
<b>Separation</b>	Separation refers to any categorization process (see sorting), but with a focus on target materials. The separation follows typically after the liberation.

<b>Sorting</b>	<p>Sorting is any process of arranging items systematically, and has two common, yet distinct, meanings:</p> <ol style="list-style-type: none"> <li>1. ordering: arranging items in a sequence ordered by some criterion;</li> <li>2. categorizing: grouping items with similar properties.</li> </ol>
<b>Substance</b>	<p>A substance is any (chemical) element or compound composed of uniform units. All substances are characterized by a unique and identical constitution and are thus homogeneous. (Brunner and Rechberger, 2004)</p>
<b>System</b>	<p>The system is the actual object of an MFA investigation. A system is defined by a group of elements, the interaction between these elements, and the boundaries between these and other elements in space and time. It is a group of physical components connected or related in such a manner as to form and/or act as an entire unit. (Brunner and Rechberger, 2004)</p>
<b>Systematic effect</b>	<p>In general, a measurement has imperfections that give rise to an error in the measurement result. Traditionally, an error is viewed as having two components, namely, a random component and a systematic component.</p> <p>The systematic error cannot be eliminated but it can often be reduced. If a systematic error arises from a recognized effect of an influence quantity on a measurement result, called the “systematic effect”, the effect can be quantified and, if it is significant in size relative to the required accuracy of the measurement, a correction or correction factor can be applied to compensate for the effect. It is assumed that, after correction, the expectation or expected value of the error arising from a systematic effect is zero. Adapted from (Joint Committee for Guides in Metrology, 2008)</p>
<b>Technology metal</b>	<p>Generally-rare metals that are essential for the production of 'high tech' devices and engineered systems, such as:</p> <ul style="list-style-type: none"> <li>- The mass production of miniaturized electronics and associated devices;</li> <li>- Advanced weapons systems and platforms for national defense;</li> <li>- The generation of electricity using 'alternative' sources such as solar panels and wind turbines;</li> <li>- The storage of electricity using cells and batteries. (Prinsloo, 2016)</li> </ul>
<b>WEEE</b> – waste electrical and electronic equipment	<p>Waste of electrical and electronic equipment (WEEE) such as computers, TV-sets, fridges and cell phones is one the fastest growing waste streams in the EU. WEEE is a complex mixture of materials and components that because of their hazardous content, and if not properly managed, can cause major environmental and health problems. (European Commission, 2016)</p>

<b>XRF – X-Ray fluorescence</b>	<p>In this method, the sample is irradiated by an intense and focused x-ray beam. The energy of the x-rays is sufficient to expel electrons from the inner shells (close to the atomic nucleus) in an atom. Electrons from outer shells fall subsequently into the holes left by the expelled electrons. These electrons then emit x-rays and the energy of the emitted x-rays is determined by the energy states of the electron shells of the specific atom. As every atom has its own particular structure of its electron shells the energy of the emitted x-rays is characteristic for each element and can be used for its identification. (ColourLex, 2017)</p>
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## Appendix 1: Comparison of recycling approaches

Table 23: Comparison of recycling approaches for LCD panels

Focus of process	Process output	Methodology	Indium yield	Reference
- Indium	- Indium concentrate - Glass substrate - Residues	- Leaching with sub-critical water - T 360° C, RT 1h	>10%	(Yoshida et al., 2014)
	- Indium concentrate - Residues	- HCL leaching - Chloride-Induced Vaporization - T 700°C, RT 90min	>84%	(Hall et al., 2009)
	- Indium concentrate - Residues	- LCD glass crushed to micron size particles - HCL leaching - T room temperature, RT ~30min	<90 %	(Lee et al., 2013)
	- Condensed indium product - Residues	- Vacuum carbon-reduction - T 950°C, RT 30min	<90%	(He et al., 2014)
	- Indium concentrate - Residues	- H <sub>2</sub> SO <sub>4</sub> leaching - Varying T and RT - Ion exchange for indium concentration	<95%	(Rasenack and Goldmann, 2014)
	- Indium concentrate - Residues	- H <sub>2</sub> SO <sub>4</sub> leaching - T 80° - 90° C, varying RT	<96,4%	(Rocchetti et al., 2015; Virolainen et al., 2011; Zeng et al., 2015)
	- Indium concentrate - Residues	- Leaching with o HCl o HNO <sub>3</sub> o H <sub>2</sub> SO <sub>4</sub> - Varying T and RT - Solvent extraction for indium concentration	>99%	(Virolainen et al., 2011; Yang et al., 2013; Kato et al., 2013; Yang et al., 2014)
	- Solid indium product - Solid zinc product - Residues	- LCD glass crushed to micron size particles - H <sub>2</sub> SO <sub>4</sub> leaching - T 90°C, 120min - Zinc plate cementation for indium recovery; T 65°C - Sulfide precipitation for zinc recovery; T 60°C, RT 10min	<99%	(Li et al., 2011)
- Indium - Tin	- Solid indium product - Residues	- H <sub>2</sub> SO <sub>4</sub> leaching - Cementation with zinc powder; T 55-60°C, varying RT	99.8%	(Rocchetti et al., 2014)
	- Indium concentrate - Glass substrate - Residues	- HCL leaching - T 75° C, RT 2h	76.16 x 10 <sup>3</sup> g/L	(Swain et al., 2016)
- Indium - Arsenic - Antimony - Polarizer foils	- Polarizer foils - Indium, arsenic, antimony concentrate - Residues	- Thermal shock for foil recovery - Leaching with: o HNO <sub>3</sub> o HCl - Varying T and RT	~60%	(Savvilitidou et al., 2015)
- Indium - Polarizer foils	- Indium concentrate - Glass substrate - Polarizer foils - Residues	- Thermal and chemical treatment for polarizer foil removal - HCL leaching - T 25°C, RT 6h - Solvent extraction for indium concentration	<90%	(Fontana et al., 2015)
	- Polarizer foils - Indium product - Residues	- Solvent bath for foil recovery - H <sub>2</sub> SO <sub>4</sub> leaching - T 90°C, RT 1h - Indium precipitation with NH <sub>4</sub> OH	>96%	(Silveira et al., 2015)

Note: H<sub>2</sub>SO<sub>4</sub> = sulfuric acid, HCl = hydrochloric acid, HNO<sub>3</sub> = nitric acid, NH<sub>4</sub>OH = ammonia water, T = temperature, RT = reaction time

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Table 24: Comparison of recycling approaches for NdFeB magnets

Method	Input material	Output REE material	Disadvantages
<b>Direct re-use of magnets</b>	Intact and liberated magnets	Single magnets for re-use, same chemical and physical properties	<ul style="list-style-type: none"> <li>- Practicable for only large quantities</li> <li>- Only applicable for same products</li> </ul>
<b>Re-sintering after hydrogen decrepitation</b>	<ul style="list-style-type: none"> <li>- Liberated magnets with same chemical composition</li> <li>- Only magnet material without coatings</li> </ul>	Magnet powder in same chemical and physical properties, respectively new magnets in eligible forms	<ul style="list-style-type: none"> <li>- No mixed scrap feed</li> <li>- No oxidized magnets</li> <li>- Coatings consisting of Zn, Sn, Cu, or Mg have to be removed</li> </ul>
<b>Hydrometallurgical methods</b>	All types of NdFeB magnets with different alloys	Mixed REE concentrate, single REE concentrates and high purity REEs	<ul style="list-style-type: none"> <li>- Complex processes</li> <li>- High quantities of chemicals and water are necessary</li> <li>- Depending on process: high energy input</li> </ul>
<b>Pyrometallurgical methods</b>	All types of NdFeB magnets with different alloys	REE alloys, REE concentrates	<ul style="list-style-type: none"> <li>- High purity of input material required to prevent solid waste</li> <li>- High energy input</li> <li>- Depending on process: no oxidized magnets applicable</li> </ul>
<b>Gas phase extraction</b>	All types of NdFeB magnets with different alloys	REE concentrate for selected elements	<ul style="list-style-type: none"> <li>- Recovery of single elements</li> <li>- Subsequent processing of residues</li> <li>- Depending on process: High quantities of chlorine gas required</li> </ul>

Based on (Binnemans et al., 2013; Nemoto et al., 2011; Zakotnik et al., 2008a; Zakotnik et al., 2006; Zakotnik et al., 2008b; Ishioka et al., 2014; Elwert and Goldmann, 2013; Rabatho et al., 2012; Takeda et al., 2006; Takeda et al., 2004)

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## Appendix 2: Sampling and subsampling for the extended batch test in WEEE pre-treatment plant

### Calculation of required primary sample amounts

Relevant parameters were established prior to the batch test, in order to obtain information about the homogeneity of the materials to be sampled, predicted volume and mass per output, bulk density, and grain size distribution. From this, a minimum number of single samples of comparable volume and the total size of the sample taken per output were calculated. The number of single samples taken was divided by the duration of the batch test.

The pre-assessment revealed a high inhomogeneity for particular output materials. According to Chancerel, 2010, sorting residues and ferrous metals scrap are carriers of high loads of target metals (cf. Figure 18). Therefore, a safety factor for each sampled output was set up, which was multiplied by the sample volumes (cf. supporting information S3 in Ueberschaar et al. (2016a)).

The sampling of single devices and components of input material and material deriving from manual separation is unusual in sampling procedure. As these flows represent no continuous streams, LAGA 98PN suggests a single sampling of these fractions. Therefore, material from the manual separation was sampled in relatively high quantities, in order to carry out a recycling-oriented product characterization with disassemblies and partial chemical analyses.

Table 25 shows the sampling masses used. The single sample volumes and the number of single samples were adjusted to the given safety factors. Therefore, the overall sampling mass per output was higher than calculated in the pre-assessment (cf. supporting information S3 in Ueberschaar et al. (2016a)).

Table 25: Detailed overview of sampled fractions over an 11 h sampling period

Sampled Output			Output batch test	Bulk density	Volume of total output	Single sample volume	Number single increments	Average interval of sampling	Overall sampling quantity before reduction	
Sample code	Sample name		[kg]	[kg/m³]	[m³]	[L]	[-]	[min]	[L]	[kg]
Manual sorting	MS1	Tools scrap	789	not applied						33
	MS2	E-Motors	1,645							549
	MS3	Loudspeaker	397							237
	MS4	Copper rich fraction	3,271							47
	MS5	Pre-sorted PCBs	221							283
Automated sorting	AS1	Ferrous metals scrap	9,789	1,500	6.5	7	22	30	154	228
	AS2	Low magnetic material	709	1,000	0.7	3.5	22	30	77	77
	AS3	Non-ferrous metals scrap	1,105	1,500	0.7	4	22	30	88	108
	AS4	Shredded PCBs	625	500	1.3	8	22	30	176	87
	AS5	Sorting residues (mixed plastics)	12,498	440	28.4	22	22	30	484	208
	AS6	SLF (Fluff)	1,691	210	8.1	6	22	30	132	67
	AS7	Filter dust	96	210	0.5	5	6	100	30	8
	AS8	Sweepings	~100	not applied						33

Note: SLF = shredder light fraction

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### Appendix 3: Uncertainty assessment for the extended batch test in WEEE pre-treatment plant

#### General concept

A measurement has imperfections, which lead to a discrepancy between the measured value and the real (unknown) value. Traditionally, this phenomenon consists of two components, namely, a random effect and a systematic effect (Joint Committee for Guides in Metrology, 2008). To ensure statistically correct results, these effects have to be determined for each measurement. Random effects represent unpredictable discrepancies to the real value and cannot be influenced. Systematic effects usually represent an offset to the real value. They can be afterward eliminated by using (determined) correction factors. Systematic effects have to be revealed through carefully applied practical work or additional methods, and eliminated or at least minimized. Not identified systematic effects are considered and treated as random effect. Therefore, final results of uncertainties should base on random effects only (Joint Committee for Guides in Metrology, 2008).

Uncertainties have to be assessed for own measurements and data from literature research. Due to this, an uncertainty propagation with an appropriate mathematical model has to be applied, which provides one value for overall uncertainty (see Figure 81). In this study, measurement uncertainties for weighing the output materials were not considered as systematic and random effects are usually very low for such operations.

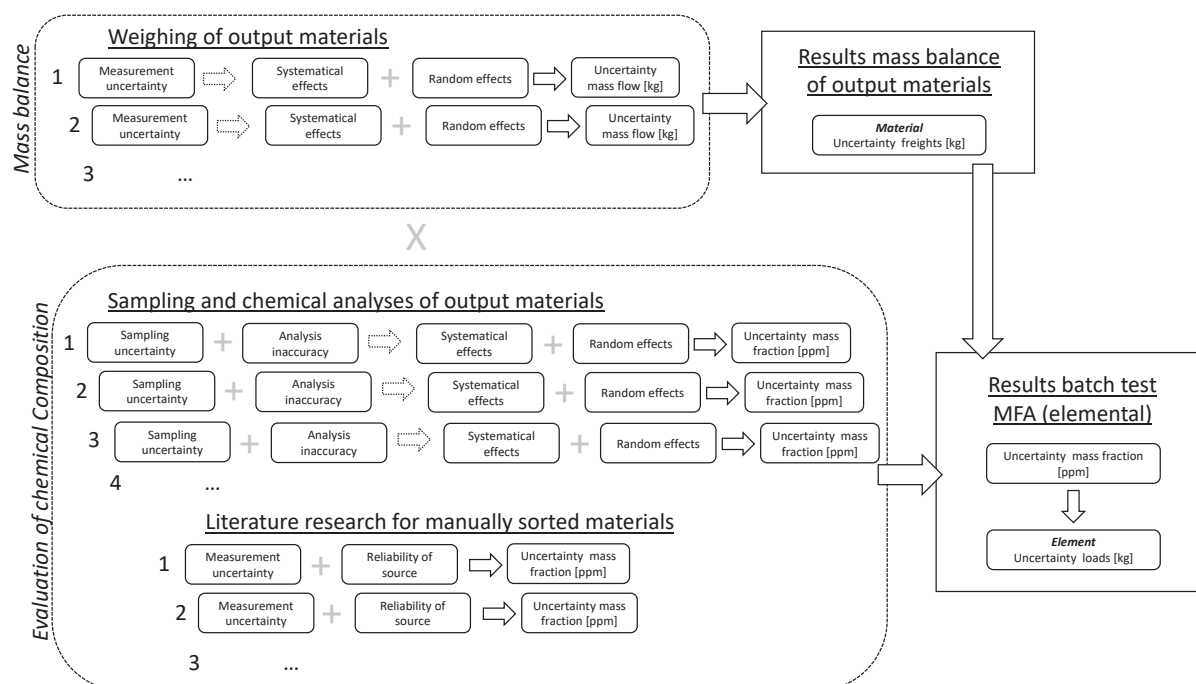


Figure 81: Mathematical model for the calculation and uncertainty propagation of an MFA on an elemental basis

#### Assessment of systematic effects

In this extended batch test study, systematic effects were identified via (1) redundancy of chemical analyses, (2) a comparison of material and element distribution, (3) case-specific methods, and (4) a comparison of the input-output loads.

##### 1. Redundancy of chemical analyses

In this extended batch test, up to three different laboratories were assigned to perform the chemical analyses of the output fractions. A comparison of the results revealed systematic effects.

## 2. *Comparison of material to element distribution*

Data from the sorting analyses was used to calculate the expected mass fractions and loads for the elements investigated, along with the literature data for each output fraction in the batch test. The results were then compared directly to the results of the chemical analyses. As only material >5 mm was sorted, additional data from the sieving analyses was used to verify the results. Inconsistent results for single elements had to be checked subsequently with appropriate methodologies like further chemical characterization using other techniques.

## 3. *Case-specific methods*

The chemical analyses of the sample AS1 for the element gold revealed high variances, which were due to systematic effects. Results from the “comparison of material to element distribution” did not clearly verify high gold mass fractions. To avoid misinterpretation, subsequent washing tests were conducted, to investigate the fines in the sample, which were not sorted. For this, three representatively divided samples from the ferrous metals scrap were treated with a 1% and a 2% aqua regia solution over a given period in an overhead shaker at room temperature. After 15, 30 and 60 minutes, samples of the liquid were taken and analyzed for gold with an ICP-AES (Thermo Scientific iCAP 6000 Series).

## 4. *Comparison of input-output loads*

A comparison of input-output loads verifies the MFA model and shows potential systematic effects. In this study, this comparison was carried out using the example of gold, as most analytical problems were related to this element. In order to compare the cumulated loads from the output fractions, an estimate of the gold content in the input material was carried out. PCBs and gold connectors were identified as carriers and corresponding mass shares in the input were approximated based on the results from the input sorting. Oguchi et al., 2011 and Ueberschaar et al., 2017 indicate PCB contents in WEEE devices and related gold mass fractions. For the estimation, we differentiate in two PCB qualities. PCBs in ordinary household devices were calculated conservatively with 100 ppm gold content. Generously equipped PCBs in information and telecommunication equipment such as mobile phones, desktop PCs, notebooks, etc. were conservatively calculated with 500 ppm gold.

Literature research gave an indication of gold loads for connectors. By estimated surface areas covered and a layer thickness of 0,02-0,08  $\mu\text{m Au}$  (Vincenz et al., 2010), the gold loads were assessed. The results of this input estimate were compared with the cumulated gold loads, based on the chemical characterization of the output fractions.

### Assessment of random effects

In this study, random effects were determined via (1) calculation of sampling uncertainties, and (2) determination of measurement uncertainties.

#### 1. *Sampling uncertainty*

Uncertainties were assessed for all samples taken from the automated sorting processes following Gy, 1992a, 1992b, 1998 and Geelhoed and Glass, 2004. This factor depended on the physical and chemical properties and was calculated for each target element. Equation 5 shows the basis for further calculations.

Equation 5: Calculation of the sampling uncertainty following (Gy, 1998; Francois-Bongarcon and Gy, 2002)

$$\sigma_{FSE} = \sqrt{\frac{f g \left( \frac{1 - a_L}{a_L} \right) [(1 - a_L) \delta_A + a_L \delta_G] d_l d^2}{M_S}}$$

With:

$f$  = shape factor,  $g$  = size range parameter,  $a_L$  = decimal proportion of target component A in lot L,  $\delta_A$  = density of target component A,  $\delta_G$  = mean density of remaining, non-target component G,  $d_l$  = liberation diameter,  $d$  = average grain diameter,  $M_S$  = mass of sample taken [g]

For the assessment of all other necessary parameters, the material was investigated prior to the extended batch test. With this information, a sampling uncertainty was calculated for each output fraction from automated sorting and only for those elements which were listed as target elements for this study and were over the detection limit in the chemical analyses. The supporting information S9 depicts an example calculation for the output fraction AS1 and the sample mass for laboratory 1.

## 2. Measurement uncertainties

“A random error is associated with the fact that when a measurement is repeated, it will generally provide a measured quantity value that is different from the previous value” (Joint Committee for Guides in Metrology, 2009). Via multiple measurements or a calculated measurement device dependent uncertainties, each laboratory provided a measurement uncertainty for each element analyzed (cf. supporting information S8).

### Uncertainties for literature-derived data

External sources always carry particular uncertainties since the data gathering method is frequently not well described and uncertainties not quantified. The use of various methodologies on different sample types by other research teams produces random rather than systematic effects. Following Laner et al., 2015, Table 26 shows the uncertainties according to the data source reliability and the level of specificity used in this study.

Table 26: Uncertainties according to the data source reliability (Laner et al., 2015)

Source / reliability	Specificity / representativeness	Coefficient of variance [%]
<b>National statistical office or independent institutions Research</b>	National data Data based on numerous measurements of the quantity of interest	1.5
<b>Official statistics from interest groups/ associations Research studies</b>	National data Data based on several measurements of the quantity of interest	4.5
<b>Individual organizations Research studies</b>	Company-specific (fractional) data Few measurements or measurements not fully representative for the quantity of interest (but transferable)	13.75
<b>Expert estimates Research studies</b>	Data based on aggregation of expert estimates Measurements of limited representativeness (unknown transferability)	41.5
<b>Rough estimates or educated guesses</b>	Specific, but based on scarce information	>50

## Calculation of the mass balance

### 1. General calculation

The general approach for determining the mass balance on the elemental level is described in Equation 6. Equation 7 shows the calculation of element specific transfer coefficients following Chancerel et al., 2009 and Rotter et al., 2004.

*Equation 6: Calculation of mass balance on element level*

$$m_{i,input} = m_{input} * x_{i,input} = \sum_{j=1}^k m_{i,fraction_j} * x_{i,fraction_j}$$

*Equation 7: Calculation of element specific transfer coefficients*

$$TC_{i,fraction_j} = \frac{m_{i,fraction_j} * x_{i,fraction_j}}{m_{input} * x_{i,input}}$$

*With:*

$x$  = mass fraction (mg/kg),  $m$  = mass (kg); indices:  $j$  = output fraction from batch test,  $i$  = substance / element,  $k$  = number of output fractions

### 2. Uncertainty calculation

Due to the methods used for sampling and analysis, only one value is provided by the chemical analyses of the output fractions. The approach used cannot give any insight in the statistical distribution of the sampled output fraction. Following usual practice, we assume a normal distribution (Chancerel et al., 2009; Joint Committee for Guides in Metrology, 2008; Joint Committee for Guides in Metrology, 2012; Laner et al., 2014). Therefore, the necessary uncertainty propagation is based on the Gaussian concept. In contrast to the principle of maximum uncertainties, relative uncertainties are calculated for all random effects determined.

The uncertainty calculation is performed separately for manually sorted and automatically sorted materials (cf. Equation 8 and Equation 9). Equation 10 describes the calculation of the overall input quantity.

*Equation 8: Calculation of uncertainty for manually sorted materials*

$$\sigma_{MS} = \sqrt{\sigma_{LS}^2 + \sigma_{DSR}^2}$$

*Equation 9: Calculation of uncertainty for automatically sorted materials*

$$\sigma_{AS} = \sqrt{\sigma_{FSE}^2 + \sigma_{MU}^2}$$

*With:*

$\sigma_{MS}$  = uncertainty for manually sorted materials,  $\sigma_{AS}$  = uncertainty for automatically sorted materials,  $\sigma_{LS}$  = uncertainty of data in literature sources (cf. supporting information S7),  $\sigma_{DSR}$  = uncertainty according to data source reliability (cf. Table 26),  $\sigma_{FSE}$  = sampling uncertainty (cf. Equation 5),  $\sigma_{MU}$  = measurement uncertainty (cf. supporting information S8)

Equation 10: Calculation of uncertainty for overall input quantity

$$\sigma_{IQ} = \sqrt{\sigma_{MS}^2 + \sigma_{AS}^2}$$

With:

$\sigma_{MS}$  = uncertainty for manually sorted materials,  $\sigma_{AS}$  = uncertainty for automatically sorted materials,  
 $\sigma_{IQ}$  = uncertainty for the overall input quantity

This kind of error propagation usually adds a covariance coefficient. The variables are completely independent, which results in a nullification of the correlation and covariance coefficients. Thus, the covariance coefficient was not included.

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## Appendix 4: Uncertainty assessment for heterogeneous data sources for gallium MFA

Table 27: Data sources for the MFA and the according uncertainties used for error propagations

Source	Uncertainty	
Concrete indication of quantities from reliable sources	10 %	Very low
Official statistical reports (e.g. federal environmental agency, Roskill reports, etc.) or quantifications for transfer coefficients based on expert interviews or scientific articles	20 %	Low
Conversion of data	50 %	Medium
General assumptions	60 %	High

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## Appendix 5: Recyclability of S-CRMs in WEEE pre-processing output fractions

Table 28 shows the most common state-of-the-art end-refining processes, which are used for the further concentration and purification of target metals. The methodologies used were assessed from a technical and a rough economic perspective, whether subsequent processes allow a recovery of the investigated S-CRMs.

The indicated conflict materials represent mostly valuable elements, which usually define the used metallurgical route. These routes may lead to a loss of critical metals. These losses can constitute an entry into alloying materials, but are mostly a transfer to the gas or slag phase. The slag phase bears the potential for a subsequent recovery. The oxidization and the co-existence of various other substances would lead to an energy intensive and therefore costly process.

Here, only an integrated smelter with Copper smelting, following lead smelting and subsequent refining steps could recover Co, In and Ga. Due to low concentrations in the input material, this is not done yet.

Table 28: State-of-the-art of end refining with conflicting materials

State-of-the-art end-refining	Target metal of recycling – mostly in conflict with critical metal	Non-target metal of recycling	Possible recovery	Recovery <u>not</u> possible or feasible
Converter of steel making	Fe plus alloying materials	Co, Ga, In, REE, Ta		x
Copper smelter as de-oxidant with subsequent precious metals refining	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
Aluminum refining / re-melting	Aluminum and alloying materials	Co, Ga, In, REE, Ta		x
Copper smelter with subsequent precious metals refining	Cu, Au, Ag, Pd, Pt plus alloying materials	Co, Ga, In, REE, Ta		x
Integrated smelter (Cu, Pb smelter and subsequent refining)	Cu, Au, Ag, Pd, Pt, Rh, Ru, Ir, In, Se, Te, Pb, Sn, Sb, Bi, Ni, As	Co	x	
		Ta		x
		In	x	
		REE		x
		Ga	x	
Copper smelter with subsequent precious metals refining	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
		Ta		x
		In		x
		REE		x
		Ga		x
Integrated smelter (Cu, Pb smelter and subsequent refining)	Cu, Au, Ag, Pd, Pt, Rh, Ru, Ir, In, Se, Te, Pb, Sn, Sb, Bi, Ni, As	Co	x	
		Ta		x
		In	x	
		REE		x
		Ga	x	
Copper smelter with subsequent precious metals refining	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
Converter of steel making	Fe plus alloying materials	Co, Ga, In, REE, Ta		x
Plastics recovery	Various types of plastics	Co, Ga, In, REE, Ta		x
Copper smelter with subsequent precious metals refining	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
Converter of steel making	Fe plus alloying materials	Co, Ga, In, REE, Ta		x
Aluminum refining / re-melting	Aluminum and alloying materials	Co, Ga, In, REE, Ta		x
Discarded	none	Co, Ga, In, REE, Ta		x

Based on: (Reuter et al., 2011; Nakajima et al., 2009; Nakajima et al., 2011; Shuva et al., 2016)

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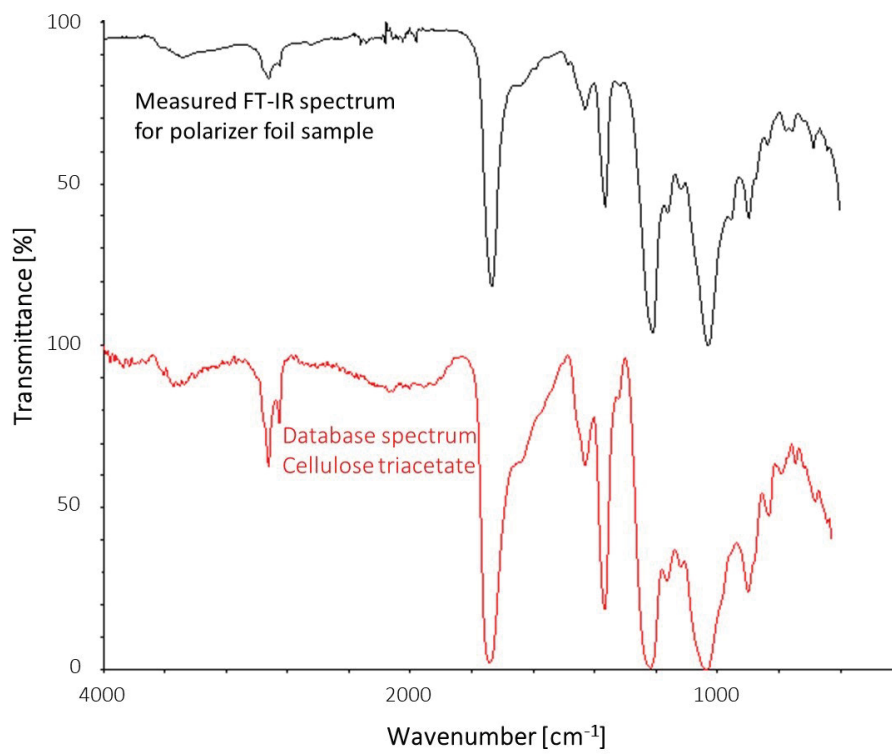
## Appendix 6: Physical and mechanical properties of investigated ICs

Table 29: Physical and mechanical properties of investigated chip-types

Properties		Type 1	Type 2	Type 3
Dimension	Weight [mg]	85	61	82
	Length [mm]	7.5	5	6
	Width [mm]	5	5	6
	Thickness [mm]	0.9	1	0.9
Stability	Untreated	Very stable	Very stable	Very stable
	After thermal treatment	Easily removable fractions	Moderately removable fractions	Easily removable fractions
Loss of weight	Due to thermal treatment [%]	14.4	8.7	11.6
	Due to separation and decanting [%]	6.1	5.5	3.3
No. of fractions after sorting process	Isolation with copper connections	1	1	1
	Metal plate (back of the Chip)	1	1	1
	Electronic module	1	1	5
	Other	1	1	-

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## Appendix 7: Example of an FT-IR spectrum recorded for a polarizer foil



Own illustration (Ueberschaar et al., 2017d)

Figure 82: Example of an FT-IR spectrum recorded for a polarizer foil (upper graph) plotted versus a database spectrum Cellulose triacetate (lower graph)

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## Appendix 8: Chemical composition of NdFeB magnets investigated

Table 30 shows the results for all elements determined and the various admixtures in voice-coil actuator and spindle motor magnets and for the total magnet content in HDDs.

Table 30 Overview of most important elements in NdFeB magnets

		NdFeB magnet in linear motor		NdFeB magnet in spindle motor		Combined	
		Average	StDev	Average	StDev	Average	StDev
Weight in g		14.3	8.2	2.3	0.3	16.6	8.3
Mass fraction in %	Al	0.37	0.11	0.24	0.24	0.35	0.13
	Fe	63.03	2.02	63.11	5.18	63.02	2.46
	Cu	0.13	0.14	0.03	0.03	0.11	0.12
	Sn	0.01	0	0.01	0	0.01	0
	Pr	2.77	2.13	2.18	2.68	2.69	2.2
	Nd	23.04	2.77	22.35	2.81	22.93	2.77
	Tb	0.14	0.16	0.02	0	0.12	0.14
	Dy	0.97	0.98	4.01	4.48	1.39	1.46
	B	0.58	0.03	0.66	0.19	0.59	0.05
	Gd	0.1	0.02	0.06	0.05	0.1	0.02
	Ni	2.54	1.24	0	0	2.19	1.07
	Sm	0.22	0.04	0.28	0.13	0.23	0.05
	Co	1.5	1.26	2.22	1.98	1.6	1.36
	Zn	0.02	0	0.02	0.02	0.02	0.01
	Sum REE	27.23	1.48	28.91	4.75	27.46	1.93

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## Appendix 9: Comparison of recycling strategies

### Integrated Circuits

Table 31: Comparison of two recycling strategies for ICs

Field	Focus	PCBs in Copper smelter		Separation of ICs from PCBs and processing of both	
		Assessment	Qualitative rating	Assessment	Qualitative rating
Recyclability	IBM	Typical process, high efficiency for Cu recovery, but not for Fe and Al.	+	Typical process, high efficiency for Cu recovery, but not for Fe and Al.	+
	PM	Typical process, high efficiency for PM recovery.	++	Traces of gold are separated with the ICs, but recoverable in subsequent recycling steps.	+
	S-CRM	No recovery of S-CRMs.	--	Recovery of gallium. But ICs are not identifiable with standard procedures.	+
	OMM	Plastics can be separated	-	Plastics can be separated	-
Technology	Low residues	Low amounts of residues.	++	Low amounts of residues, but maybe higher in subsequent gallium recovery processes.	++
	Low chemicals	No chemicals; only in subsequent recovery processes.	Not applicable	IC separation mechanical or manual only. Gallium recovery through thermal treatment.	Not applicable
	Low energy	Plastics used as fuel; still high energy input of smelter.	o	Additional processes, which consume energy + subsequent thermal treatment of ICs.	-
Material economics	Low investment	State-of-the-art process; existing technology can be used.	Not applicable	Either high labor costs or sophisticated technique for IC identification and separation.	--
	Revenue pricing	High revenues for IBM, PM contents.	++	High revenues for PM (- gold losses) and copper, lower revenues for gallium concentrate.	+
	Low purchase pricing	Highly competitive market for PCBs.	-	Potentially higher prices for purchase, as specific PCBs have to be bought (tablets, smartphones, mobile phones, notebooks).	--
Market situation	Output	Good market for PM and Cu.	++	Gallium concentrate contains gold. Therefore, specific recovery processes needed, which are not yet available.	+
	Input	Highly competitive market for PCBs.	o	Only a few PCB types worth of gallium recovery. High masses needed as only low mass fraction of gallium in ICs on PCBs.	--

LCD panels

Table 32: Comparison of two recycling strategies for LCD panels

Field	Focus	Copper smelter		CST + Indium separation	
		Assessment	Qualitative rating	Assessment	Qualitative rating
Recyclability	IBM	Typical process, high efficiency for Cu recovery.	++	Copper not in focus, but may be recovered from the indium concentrate.	--
	PM	No PM in LCD panels.	Not applicable	No PM in LCD panels.	Not applicable
	S-CRM	No recovery of S-CRMs	--	Indium recovery, but losses to undesigned output fractions	+
	OMM	Plastics burned, glass used as slag builder.	--	Glass and plastics recoverable, tin lost.	++
Technology	Low residues	Slag has to be landfilled (mostly glass).	--	Low amounts of residues, if glass is recycled.	++
	Low chemicals	No chemicals; only in subsequent recovery processes.	Not applicable	Base leaching for CST; Acid leaching for indium separation. May be used in circular process.	o
	Low energy	Plastics used as fuel; still high energy input of smelter.	-	Base and acid leaching can be conducted at room temperature.	+
Material economics	Low investment	State-of-the-art process; existing technology can be used.	Not applicable	Base leaching can be conducted in very simple ways. Acid leaching process more sophisticated.	o
	Revenue pricing	Revenue for copper minus landfilling for glass (slag).	--	Revenue from indium and cellulose acetate + glass, if recycled.	+
	Low purchase pricing	No trade channel for LCD panels. Low prices.	++	No trade channel for LCD panels. Low prices.	++
Market situation	Output	Slag has to be landfilled.	--	If glass recyclable, only minor masses not saleable.	++
	Input	Independent, depends on market.	++	Only lower mass flows available in WEEE sector, but currently no competitors.	+

HDD

Table 33: Comparison of two recycling strategies for LCD panels

Field	Focus	Shredding for data destruction		Specialized mechanical + metallurgical treatment	
		Assessment	Qualitative rating	Assessment	Qualitative rating
Recyclability	IBM	IBMs in standard mechanical processes with standard sorting processes well recoverable	++	IBMs in standard mechanical processes with standard sorting processes well recoverable	++
	PM	PMs in PCBs from HDDs can be recovered, if separated. If shredded, losses via dust exposure and undesigned output fractions.	+	PMs in PCBs from HDDs can be recovered, if separated. If shredded, losses via dust exposure and undesigned output fractions.	+
	S-CRM	No recovery of S-CRMs	--	REE recovery from NdFeB magnets possible, but losses through distribution in other output streams	+
	OMM	Plastics can be separated	+	Plastics can be separated	+
Technology	Low residues	HDDs consist mostly of metals, which are effectively recycled in mechanical processes	++	HDDs consist mostly of metals, which are effectively recycled in mechanical processes	++
	Low chemicals	Mechanical process only	Not applicable	Mechanical process only	Not applicable
	Low energy	Mechanical process only	+	Mechanical treatment + classification processes + metallurgical REE recovery	+
Material economics	Low investment	State-of-the-art process; existing technology can be used.	Not applicable	Different crushing unit needed and further classification steps + eventually pyrometallurgical step	-
	Revenue pricing	High revenues for IBM, PM contents. But maybe some PM losses in mechanical treatment.	++	High revenues for IBM, PM contents. But maybe some PM losses in mechanical treatment.	++
	Low purchase pricing	Relatively high prices for purchase of HDD	0	Relatively high prices for purchase of HDD	0
Market situation	Output	Due to high metal contents, all materials perfectly saleable	++	Market for REE output not existent; reuse in primary metal production to be evaluated.	+
	Input	Independent, depends on market.	++	Predictable and stable WEEE mass flows. Subsequent REE recovery process dependent from input.	++

## Tantalum capacitors

Table 34: Comparison of two recycling strategies for tantalum capacitors

Field	Focus	PCBs in Copper smelter		Separation of tantalum capacitors from PCBs and processing of both.	
		Assessment	Qualitative rating	Assessment	Qualitative rating
Recyclability	IBM	Typical process, high efficiency for Cu recovery, but not for Fe and Al	+	Typical process, high efficiency for Cu recovery, but not for Fe and Al	+
	PM	Typical process, high efficiency for PM recovery	++	High shares of silver separated with tantalum caps and subsequently lost	+
	S-CRM	No recovery of S-CRMs	--	Recovery of tantalum. But not all tantalum bearing components identifiable.	+
	OMM	Plastics can be separated	-	Plastics can be separated	-
Technology	Low residues	Low amounts of residues	++	Low amounts of residues, but maybe higher in subsequent tantalum recovery processes	++
	Low chemicals	No chemicals; only in subsequent recovery processes.	Not applicable	Tantalum capacitor separation mechanical or manual only	Not applicable
	Low energy	Plastics used as fuel; still high energy input of smelter	o	Additional processes for separation of tantalum capacitors necessary	-
Material economics	Low investment	State-of-the-art process; existing technology can be used.	Not applicable	Either high labor costs or sophisticated technique for tantalum capacitors identification and separation	--
	Revenue pricing	High revenues for IBM, PM contents	++	High revenues for PM (- silver losses) and copper, low revenues for tantalum	++
	Low purchase pricing	Highly competitive market for PCBs	-	Potentially higher prices for purchase, as specific PCBs have to be bought	--
Market situation	Output	Good market for PM and Cu	++	No market for tantalum capacitors	+
	Input	Highly competitive market for PCBs	o	Only a few PCB types worth of tantalum recycling	-

ISSN 1864-5984  
ISBN 978-3-86948-582-9