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Assessment of element-specific recycling efficiency in WEEE pre-processing

Article, Postprint

This version is available at <https://doi.org/10.14279/depositonce-6365>.



Suggested Citation

Ueberschaar, Maximilian; Geiping, Julia; Zamzow, Malte; Flamme, Sabine; Rotter, Vera Susanne: Assessment of element-specific recycling efficiency in WEEE pre-processing. - In: Resources, Conservation, and Recycling. - ISSN: 0921-3449. - 124 (2017). - pp. 25-41. - DOI: 10.1016/j.resconrec.2017.04.006. (Postprint version is cited, page numbers may differ.)

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Abstract

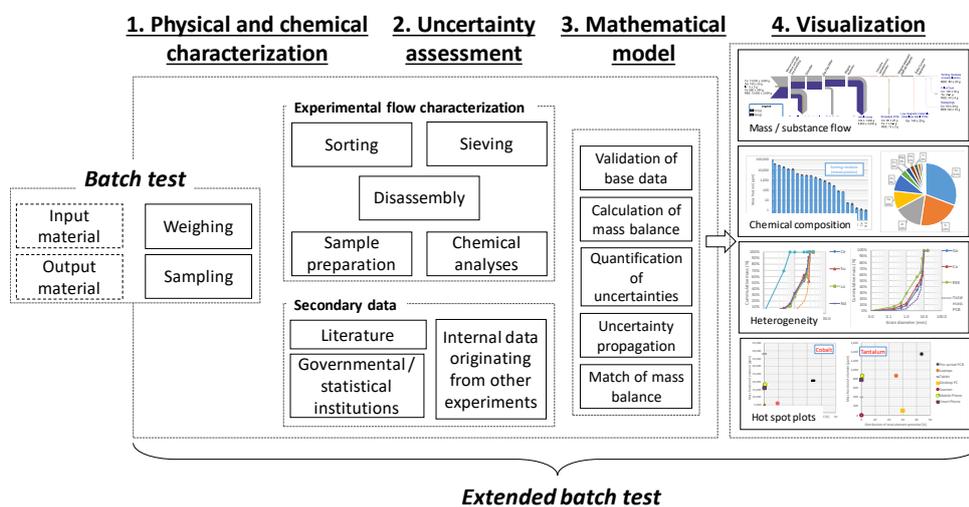
Pre-processing is a crucial step to ensure the efficiency of subsequent processes and the quality of recyclates. The efficiency of pre-processing can be affected by high losses to undesigned output fractions. Standard batch tests usually provide mass balances and are a good proxy for bulk materials balances (iron/steel, aluminum, plastics).

This article aims at harmonizing methodologies and recommends a strategy for further study in pre-processing on a plant scale. We have developed an “extended batch test” method, which should help to

- describe the fates of materials and elements,
- assess the quality of output fractions,
- identify access points for critical metals and other valuable elements to enable their recovery.

A methodical approach was compiled with common material flow analysis methods and an extended set of methods, which improve the reliability via the assessment of uncertainties. This applies to systematic effects and random effects. This extended batch test was performed with a 40 Mg Waste Electrical & Electronic Equipment (WEEE) batch to trace the flows of industrial base metals, precious metals and critical metals in a WEEE pre-processing plant.

Results show that one-third of the input was separated and sorted manually, while the remaining material was subsequently crushed and automatically sorted. Copper and precious metals are distributed to various output fractions but are most concentrated in the sorting residues. Critical metals like cobalt and rare earth elements are mainly concentrated in the manually sorted materials but also appear in the ferrous metals scrap and the shredder light fraction.



Graphical abstract

1. Introduction

Pre-processing is one of the central steps in the recycling chain of Waste Electrical and Electronic Equipment (WEEE) as well as of other complex products. Through liberation and separation, secondary raw materials are channeled into designated recovery processes. Generated outputs have to be either processed further or can be used directly in final recovery operations. To fulfill the regionally mandatory recycling quotas, current recycling strategies target industrial base metals like copper, iron, and aluminum, and also plastics, as bulk materials (Chancerel, 2010; Reuter and van Schaik, 2012; Rotter et al., 2016). Furthermore, trace materials with a high economic value, such as precious metals (gold, silver, palladium), are of high interest from an economic and ecologic perspective (Li et al., 2015). A recovery of such materials takes place in subsequent end-refining processes, which are usually limited to a particular set of materials (Khaliq et al., 2014). 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. Because of this, the transfer of non-target metals into recyclates for end-processing leads to a loss of these materials, 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 critical raw materials (CRM) by the European Commission (2010, 2014), the U.S. Department of Energy (2011) and the Japan Institute of Metals and Materials (Hatayama and Tahara, 2015) and whose recovery is discussed on the political agenda. The set and classification of assessed materials differ in these criticality studies. Therefore, we will distinguish them as follows: “industrial base metals (IBM)” (aluminum, iron, copper), “precious metals (PM)” (for example gold, silver, and palladium) and a selected “set of critical metals (S-CRM)” (for example, rare earth elements (REE), indium, cobalt, etc.). S-CRMs are mainly applied in complex WEEE products (Chancerel et al., 2015, 2013). Due to highly sophisticated processes for their recovery, no recycling strategies are implemented yet (Kumar and Holuszko, 2016; Li et al., 2017; Zeng and Li, 2016).

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 checking the efficiency of processes. Recycling efficiency is usually tested with batch tests. Due to local 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). 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 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 (WEEForum, 2013).

Experimental material flow analysis has been introduced as a systematic approach to track goods/materials and substances to understand their origin and fate in investigated processes. It is based on the same procedures used for conventional batch tests, 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)

In order to compare commonly used methodologies and the quality of the results of MFA on a substance level, various studies using experimental batch tests have been compared in Table 1. The trials took place in different facilities processing household waste, WEEE and ELV (end-of-life-vehicles).

Table 1: Comparison of experimental MFA on a substance level in the form of a batch test applied on various waste streams

Study	Waste input	Objective	Size of batch	Target elements	Sampling method	Sample preparation	Digestion	Analytical methods	Input sampling	Output sampling	Statistical analysis
(Rotter et al., 2004)	Household waste	Assessment of selective Heavy metal Separation in RDF	4-8 Mg	Cl, Hg, Cd, Pb, Zn, (Sb)	3 samples per fraction	Grinding <0,5 mm	Acid based Microwave digestion	AAS	Sorting analysis of waste fraction	Yes, all fractions	Calculation of interval of confidence (95%)
(Morf and Taverna, 2004)	sWEEE	Assessment of metallic and non-metallic materials	230 Mg	Al, Sb, Pb, Cd, Cr, Fe, Cu, Ni, Hg, Zn	16-20 single samples per fraction	Manual sorting, grinding, partially re-melting	Acid based Microwave digestion	ICP-MS, ICP-OES, XRF	Sorting analysis	Yes, only relevant fractions	Calculation of interval of confidence (95%)
(Chancerel, 2010; Chancerel et al., 2011, 2009)	sWEEE, ICT & consumer electronics	Identification of PM losses in pre-processing	27 Mg	Ag, Au, Pd, Cu, Fe, Al	Umicore standard including fire assaying and re-melting with recuperation of target elements. X-ray fluorescence analysis and acid based digestion prior to determination with inductively coupled plasma mass spectrometry (ICP-MS) and atomic emission spectroscopy (ICP-AES)				No (data on average input composition assessed)	Full sampling, dismantling tests, literature data, visual estimation	Data reconciliation of material balance based on Gaussian distribution
(Schoeps et al., 2010)	Mono batch: PC	Identification of PM losses in pre-processing	2.9 Mg	Ag, Au, Pd					Sorted mono batch	No info.	Not applied
(Oguchi et al., 2013)	WEEE: PCB & CRT	Metal fates in actual waste treatment processes	No info.	Full analysis	No info.	Grinding <0,25 mm	Acid based Microwave digestion	ICP-OES, ICP-MS	Sorting analysis	Yes, all fractions	Not applied
(Morf et al., 2013)	Municipal solid waste	Precious metals and rare earth metals flow	No batch trial	Full analysis	Method following (Bauer, 1995; Gy, 1992)	Partially divided; grinding <0,1 mm	Acid based Microwave digestion	ICP-MS	Only homogeneous materials; full chemical analysis		Data reconciliation based on Gaussian distribution
(Arena and Di Gregorio, 2014)	Municipal solid waste	Mass flow rates and composition for decision making	100 Mg	C, Cd, Pb	Literature review				No	No info.	Data reconciliation based on Gaussian distribution
(Widmer et al., 2015)	EE in ELV	Distribution of scarce metals. Plausibility of results	95,3 Mg; 100 ELV	31 scarce metals	No info.	Grinding <0,5 mm	Acid based Microwave digestion	XRF, ICP-OES, ICP-MS	Generic input	6 from 7 sampled	ISO GUM class c, 12-24%
(Habib et al., 2015)	Hard disk drives	Track of REE based permanent magnets	244 kg; 1050 HDD	REE	No info.	No info.	-	WD-XRF	Yes	Visual sorting	Not applied

Note: household waste = residual household waste is mixed waste collected after source separations, WEEE = small WEEE, ICT = information and communications technology, consumer electronics = entertainment electronics

Some conclusions from previous MFA studies can be summarized as follows:

- Assessment of uncertainties and an appropriate uncertainty propagation received only minor attention.
- In order to generate data with a high level of detail, a combination of various methodologies has to be used. Through this, uncertainties like systematic effects can be reduced.
- Furthermore, it is important to validate results. One possibility is the sampling of the input material in order to match the results of input and output.
- Dissipation of lower value critical metals in pre-processing has not yet been investigated.

Therefore, the objective of this article is to demonstrate a suitable methodology for WEEE pre-processing that supports improvements in recycling efficiencies for bulk and trace materials along the recycling chain with particular attention to CRM, which presently has low recycling rates. For this, we took the standard WEEELABEX batch test (WEEEforum, 2013) as a basis and developed an “extended batch test”, which should help to

- describe the fate of materials and elements during pre-processing,
- assess the quality of output fractions from pre-processing relative to the subsequent process steps and
- identify options for further concentration of CRMs and other valuable elements to enable their recovery

Furthermore, this article should help to harmonize existing methodologies and recommend a useful strategy for further study in pre-processing on a plant scale.

2. Material and Methods

2.1. Research concept

For this study, a batch test was conducted in a WEEE pre-processing plant. The processes used covered 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 1 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.

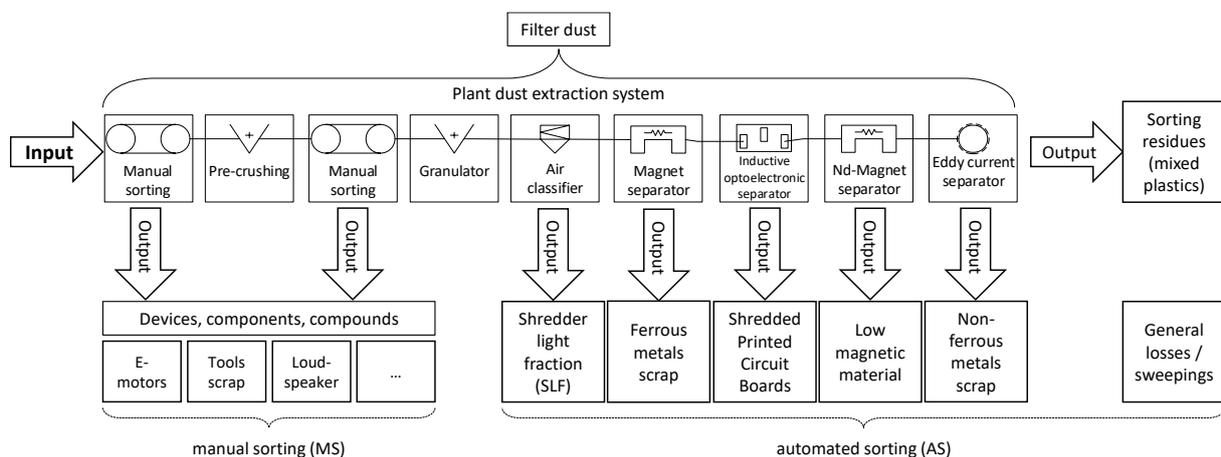


Figure 1: Process set-up of recycling plant investigated

Target metals of this investigation include ‘industrial base metals’ (IBM) like aluminum, iron and copper, ‘precious metals’ (PM) gold, silver, palladium and platinum and a selected ‘set of critical metals’ (S-CRM) like cobalt, gallium, indium, tantalum and the WEEE relevant REE (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 the information in Table 1, a general methodology for carrying out a batch test was developed. Figure 2 depicts the complete approach.

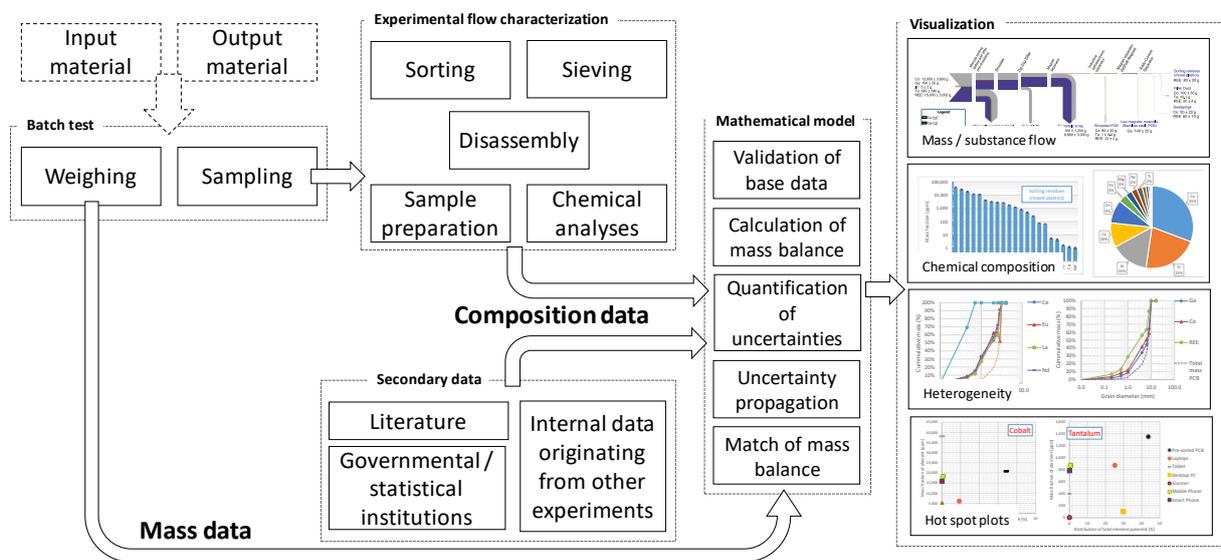


Figure 2: Experimental design of the “extended batch test” with subsequent data collection and evaluation

Based on a chemical characterization, 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.

Uncertainties are usually not part of MFA studies (Laner et al., 2014). However, in order 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.

2.2. Sampling methodologies

In this study, sampling followed the method of LAGA PN 98, which represents the only national regulation for the sampling of waste materials in Germany. It 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.

2.2.1. Calculation of required primary sample amounts

Relevant parameters were established prior to the batch test, 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 equal volume and the total size of the sample taken per output were calculated. The amount of single samples taken was divided by the duration of the batch test (cf. supporting information S3).

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 1). Increasing the reliability of the sampling procedure, the calculation of the amount of increments was carried out conservatively. Furthermore, the volume of the increments was increased.

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 stream, LAGA 98PN suggests a single sampling of these fractions. Therefore, material from the manual separation was sampled in high quantities, to carry out product-centric recyclability assessments with disassemblies and partial chemical analyses (Rotter et al., 2013). Table 2 shows a sampling procedure overview and the sampled masses.

Table 2: 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/m ³]	[m ³]	[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 Printed Circuit Boards	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 Printed Circuit Boards	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

2.2.2. Subsampling from primary samples

In order 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). Table 3 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 3: Sample splitting for chemical 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.

2.3. Characterization of input and output 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.

2.3.1. Physical characterization

(1) 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 shows the sorting protocol used.

(2) 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 printed circuit boards (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 shows the disassembly protocol draft. The composition data assessed for small household tools, loudspeaker drivers, and e-motors is depicted in S7.

(3) 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 minutes 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 minutes 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.

(4) 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). 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.

2.3.2. Chemical characterization

(1) 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.

(2) 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 previous batch tests. The supporting information S7 shows the data aggregated for further calculations.

(3) Characterization of sieving steps (AS)

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

(4) 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. Also, 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 to compare the results with laboratory 1. For sample preparation, samples AS4 - 7 were milled in a cross beater mill (Retsch SK1) and an ultracentrifugal 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 HNO₃-H₂O and aqua regia in a microwave-assisted digestion with 1.600 W, 20 bar, 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 performed with a QQQQ-ICP-MS in O₂-mode.

2.4. Uncertainty assessment

2.4.1. 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 3). 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.

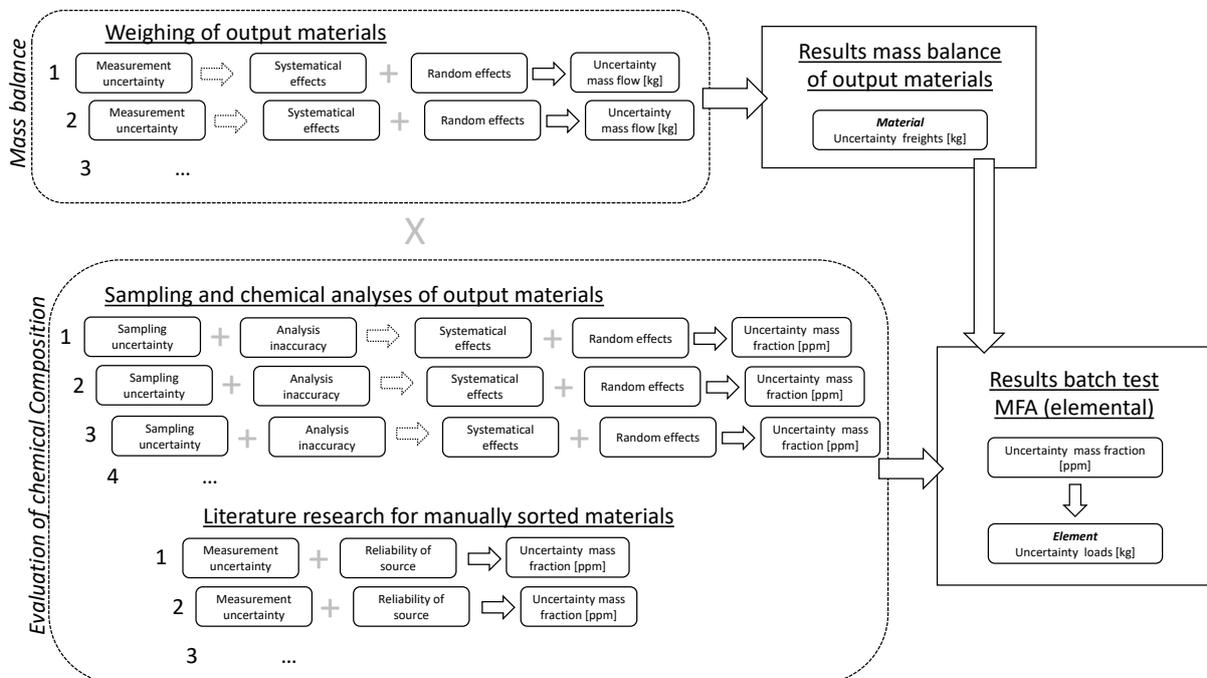


Figure 3: Mathematical model for the calculation and uncertainty propagation of an MFA on an elemental basis

2.4.2. 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.

2.4.3. 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. Formula 1 shows the basis for further calculations.

Formula 1: Calculation of the sampling uncertainty following (Francois-Bongarcon and Gy, 2002; Gy, 1998)

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

With:

f = shape factor, g = size range parameter, a_L = decimal proportion of target component A in lot L, δ_A = density of target component A, δ_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).

2.4.4. 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 4 shows the uncertainties according to the data source reliability and the level of specificity used in this study.

Table 4: Uncertainties according to the data source reliability (Laner et al., 2015)

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

2.5. Calculation of the mass balance

2.5.1. General calculation

The general approach for determining the mass balance on the elemental level is described in Formula 2. Formula 3 shows the calculation of element specific transfer coefficients following Chancerel et al., 2009 and Rotter et al., 2004.

Formula 2: Calculation of mass balance on element level

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

Formula 3: 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

2.5.2. 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, 2012, 2008; 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. Formula 4 and Formula 5). Formula 6 describes the calculation of the overall input quantity.

Formula 4: Calculation of uncertainty for manually sorted materials

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

Formula 5: Calculation of uncertainty for automatically sorted materials

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

Formula 6: Calculation of uncertainty for overall input quantity

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

With:

σ_{MS} = uncertainty for manually sorted materials, σ_{AS} = uncertainty for automatically sorted materials, σ_{IQ} = uncertainty for the overall input quantity, σ_{LS} = uncertainty of data in literature sources (cf. supporting information S7), σ_{DSR} = uncertainty according to data source reliability (cf. Table 4), σ_{FSE} = sampling uncertainty (cf. Table 5), σ_{MU} = measurement uncertainty (cf. supporting information S8)

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.

3. Results

3.1. Uncertainty assessment

3.1.1. Assessment of systematic effects

Comparison of material to element distribution

With the information from the sorting analyses, the results of the chemical analyses could be validated. To this end, mass fractions of target metals in the sorted materials were assumed. The detailed list is attached in the supporting information S13.

For IBMs like Fe and Al, the results were almost in the same range. For substances applied only in low concentrations in the original components or devices, some of the findings from the sorted materials differed substantially from the results of the chemical analyses. This effect was also noticeable for cobalt and REE. Here, a significant share was possibly contributed by magnet material (mostly NdFeB magnets). These components were pulverized during the shredding processes. Occasionally, single clots of magnetic material stuck together with ferrous metals were found in the ferrous metals scrap. A closer examination with an XRF handheld (Thermo Fisher/Analyticon XL3 air) revealed high contents of REE and Co and identified this material partially as NdFeB magnets. As this material was highly contaminated, it was not sorted as a separate material fraction in the sorting analysis carried out.

Figure 4 shows the examples aluminum and gold. The results for the other metals investigated are depicted in the supporting information S13.

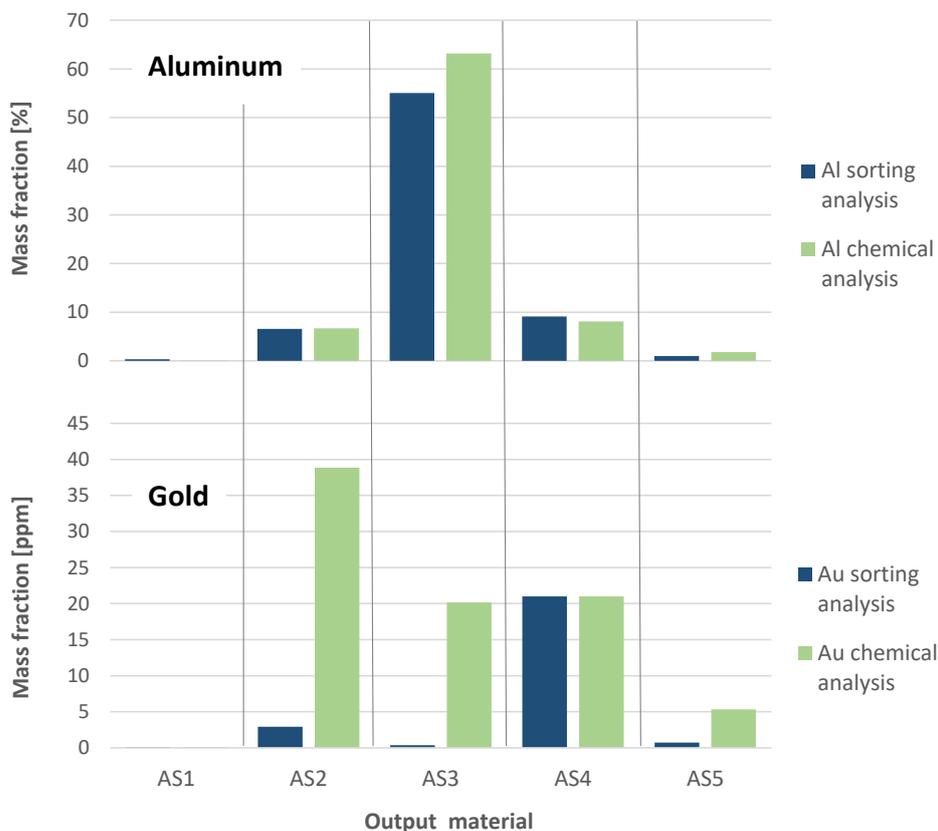


Figure 4: Comparison of Al content in output materials based on chemical analyses and sorting analyses

Due to higher findings using the chemical analyses, trace metals were suspected in the fines of the output fractions, as only material over 5 mm was sorted. This non-sorted material constituted a significant proportion of the total material with a range between 2% and 19% (cf. Figure 8).

Gold analysis

The determination of the element gold was related to high systematic effects. This applied in particular to the sample AS1. Here, laboratory 1 determined mass fractions of about 67 ppm Au. In line with the high masses of this output fraction, the gold load was expected to be extremely high. Further analyses confirmed these high concentrations. A further sample from another test at the same facility had comparable results. As the sorting test showed no corresponding material composition in the form of PCBs, etc., washing tests with subsequent chemical analyses were carried out, to investigate the fines for gold content. Results showed gold mass fractions of from 1×10^{-3} to 3×10^{-2} ppm. This did not confirm the high gold content in the ferrous metals scrap. Subsequently, laboratory 3 (cf. Table 3) developed a specific methodology for reliably determining the gold content of sample AS1. The results revealed an actual gold mass fraction of 0.9 ppm. This results was verified by the comparison of input-output loads.

Comparison of input-output loads

Using the example of gold, the input and output loads of the extended batch test were compared. Approximately 210 g gold related to the input of PCBs and ~0.2 g to the gold connectors. From this, a total of 210 g gold was estimated for the input. The calculated gold content in the output is about 240 g. This verified the analytical approach and the data basis of this study.

3.1.2. Assessment of sampling uncertainties

Using Formula 1 as presented above and the sample masses, a sampling uncertainty was calculated for each target element and output fraction of the automated sorting process. Table 5 shows the results. As various sample sizes were used for the analyses, each laboratory refers to different uncertainties. The sampling uncertainties were assessed only for those target elements which were over the detection limit. Therefore, only the S-CRM Co and the REE are depicted.

Table 5: Sampling uncertainties for target elements according to sample size and output fraction

Target material	Responsible laboratory	Sampling uncertainties calculated as %							
		AS1	AS2	AS3	AS4	AS5	AS6	AS7	AS8
Au, Ag, Pd, Ga	Lab 1	2.0	0.4	1.1	0.1	0.6	0.04	0.03	0.04
	Lab 2, 3	5.4	-	-	0.3	1.3	0.2	0.06	-
Cu	Lab 1	4.1	1.5	1.1	0.4	0.9	0.04	0.05	0.04
	Lab 2	11.3	-	-	0.9	1.9	0.2	0.08	-
Fe	Lab 1	0.1	0.2	2.4	1.0	3.8	0.04	0.03	0.04
	Lab 2	0.2	-	-	2.5	8.0	0.2	0.06	-
Al	Lab 1	2.3	0.9	0.2	1.0	1.3	0.04	0.03	0.04
	Lab 2	6.4	-	-	2.5	2.7	0.2	0.06	-
Co, REE	Lab 1	9.8	2.0	4.7	8.9	6.0	0.04	0.03	0.04
	Lab 2	26.6	-	-	22,0	12.7	0.2	0.6	-

According to Francois-Bongarcon and Gy (2002), uncertainties of under 7% are acceptable for a reliable sampling. As depicted, some laboratories were linked to higher uncertainties as they received lower sample quantities for determining particular elements. Results from laboratory 1 were considered to

be the most accurate. Only for cobalt and REE in AS1 and AS4 were the uncertainties slightly higher than 7%.

3.2. Fate of materials and elements in extended batch test

3.2.1. Calculated element input

Figure 5 shows the element input calculated in the extended batch test with a total mass of 42,860 kg. The highest share of metals was represented by the IBMs, in particular iron, followed by copper and aluminum. PMs and S-CRMs constituted only a small quantity in the extended batch test. About 4 kg of PMs, mostly silver, were processed. REE and cobalt dominated the S-CRMs.

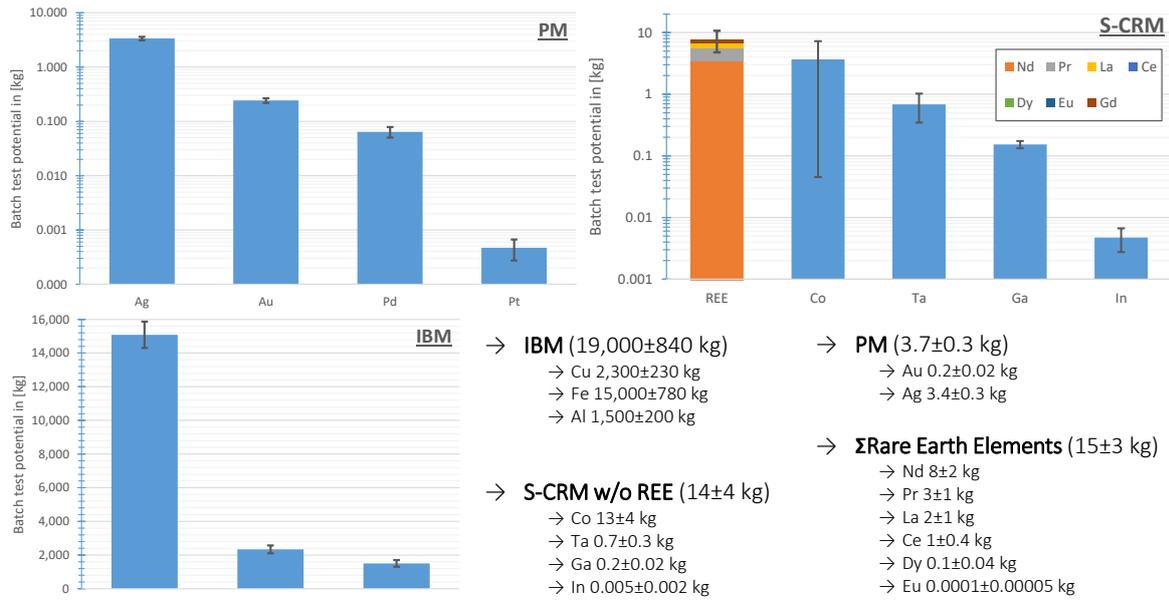


Figure 5: Calculated material input quantities (errors bars based on all assessed uncertainties)

3.2.1. 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 transfer coefficients calculated are a highly important tool for the evaluation of the data generated. Supporting information S12 shows the data in detail. Figure 6 shows the aggregated data on all mass flows. The elemental material flow analysis depicts the elemental distribution of 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.

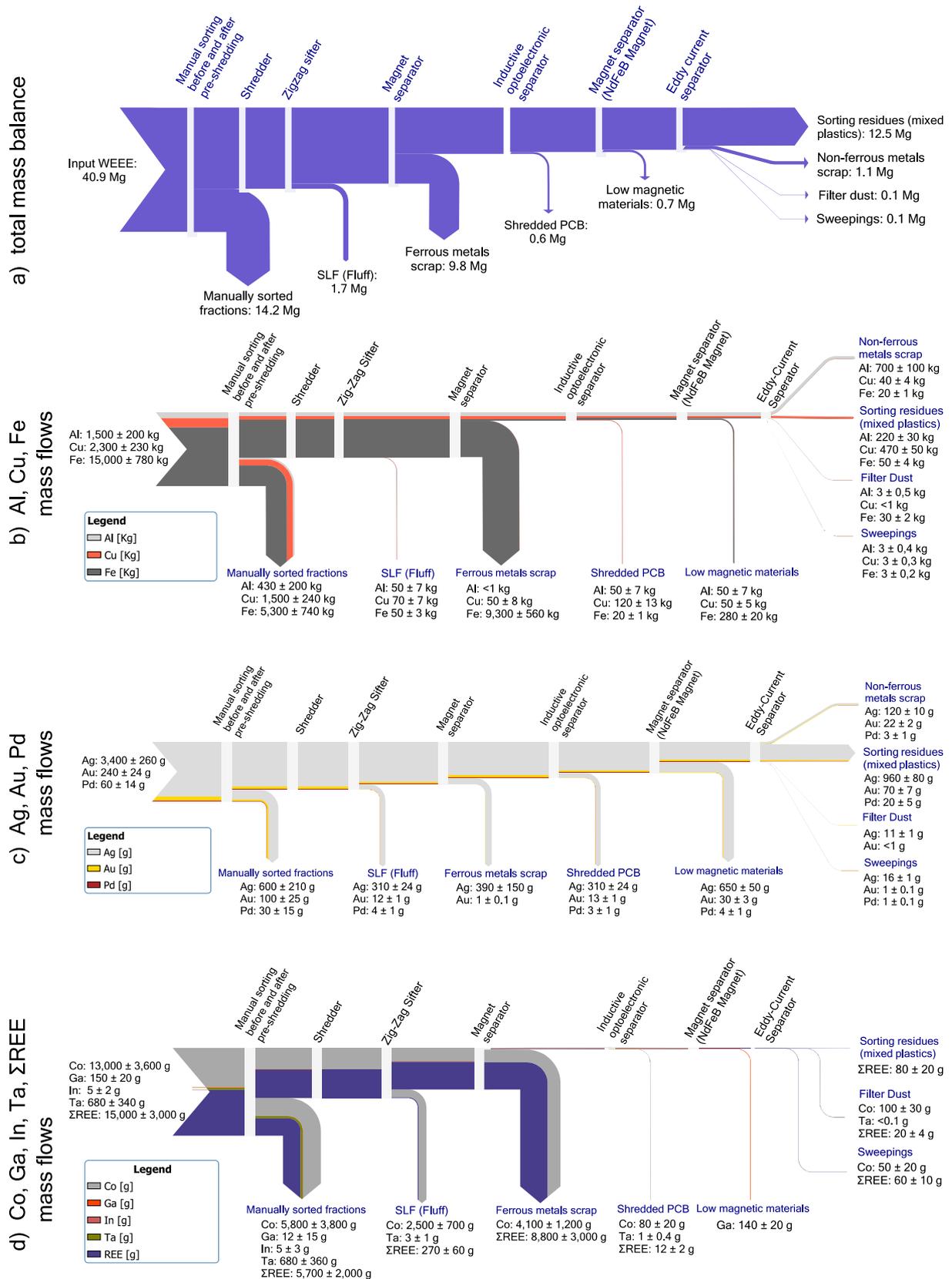


Figure 6: Mass balance and mass flows in extended batch test a) total mass, b) aluminum, copper and iron c) silver, gold, palladium and platinum, d) cobalt, gallium, indium, tantalum and ΣREE

More than one-third of the material was separated manually before 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, 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 before mechanical processing, but 20% still ended in the sorting residues AS5. Due to the very efficient 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.

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

Figure 7 shows the general splitting of the target metals into manually sorted materials before 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, while the mass fractions for the automated sorted output fractions are depicted in the supporting information S8.

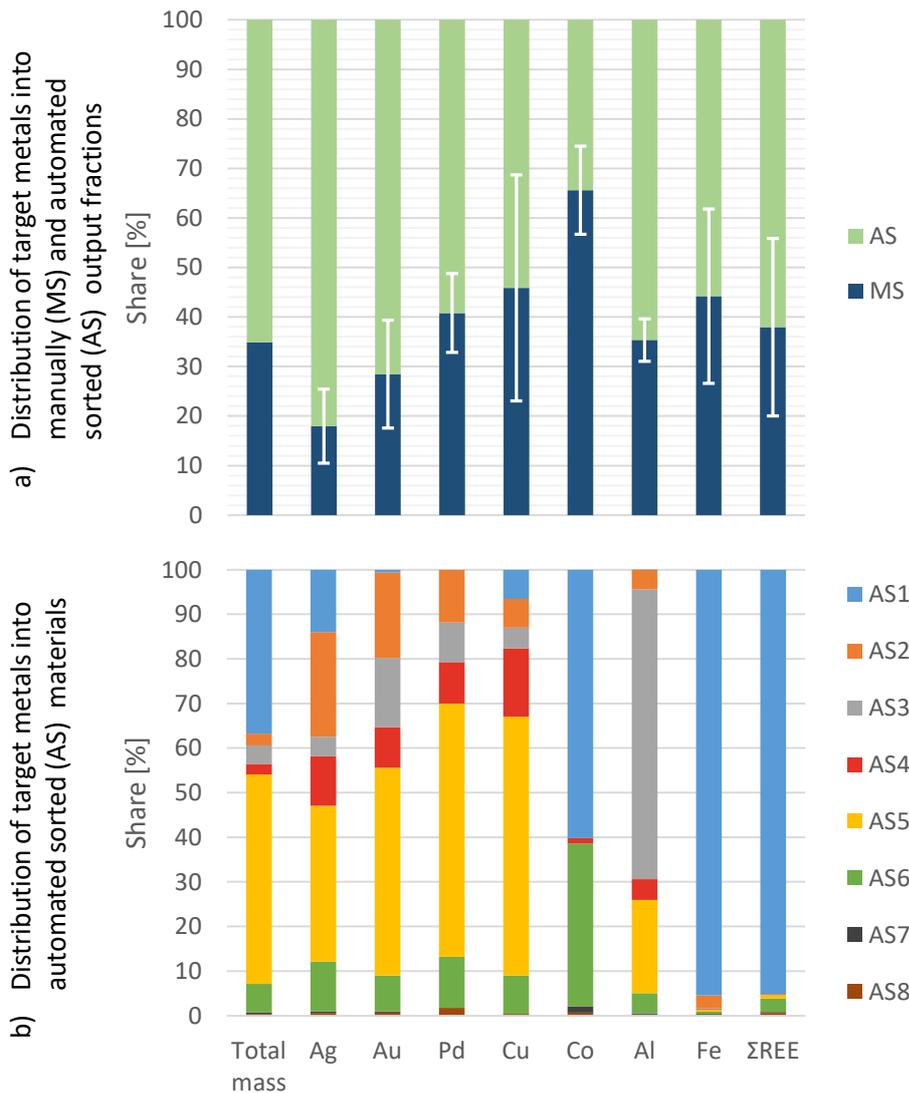


Figure 7: Splitting of IBM, PM, and S-CRM through manual and automated sorting and distribution in output fractions of automated sorting processes

Except for copper, all elements investigated were mostly concentrated in the automated sorting materials. The main reason for this was the high mass share 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 ΣREE.

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 significant in quantity and contained a high load of target elements even with low determined mass fractions.

3.3. Quality assessment of output fractions from pre-processing

3.3.1. 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 share of 3.5% in AS2 (see Figure 8).

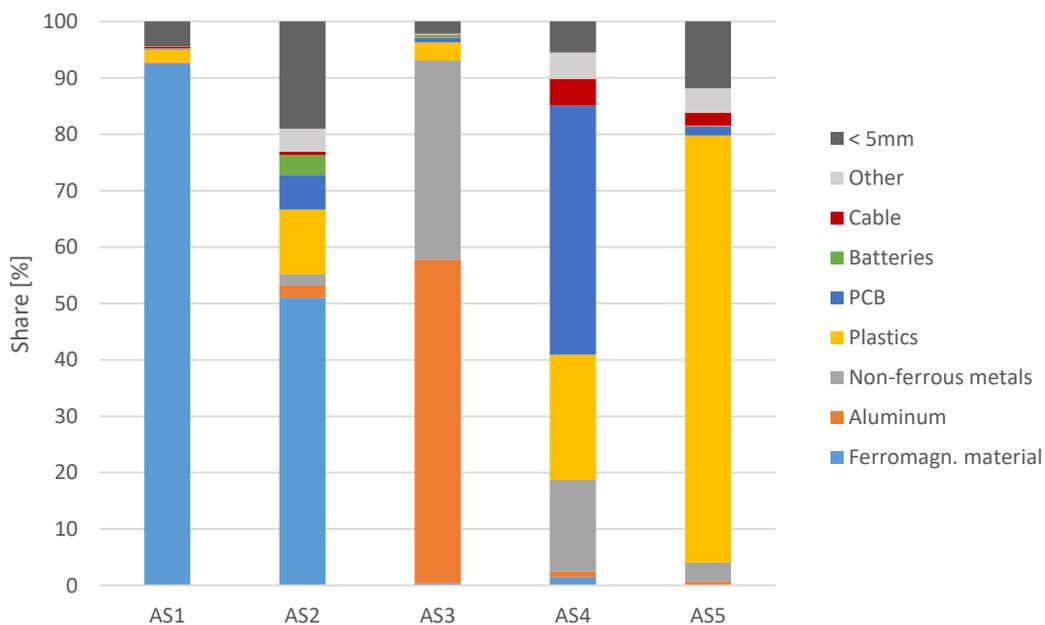


Figure 8: Composition of recyclates from mechanical processing based on sorting analysis

3.3.2. 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 CRMs were located more in the smaller grain sizes. In general, 30-50% of the S-CRMs and up to 90% of the PMs enrich in grain sizes below 5 mm. Figure 9 shows the distribution of Ga, Co, and Σ REE in the output fraction AS4.

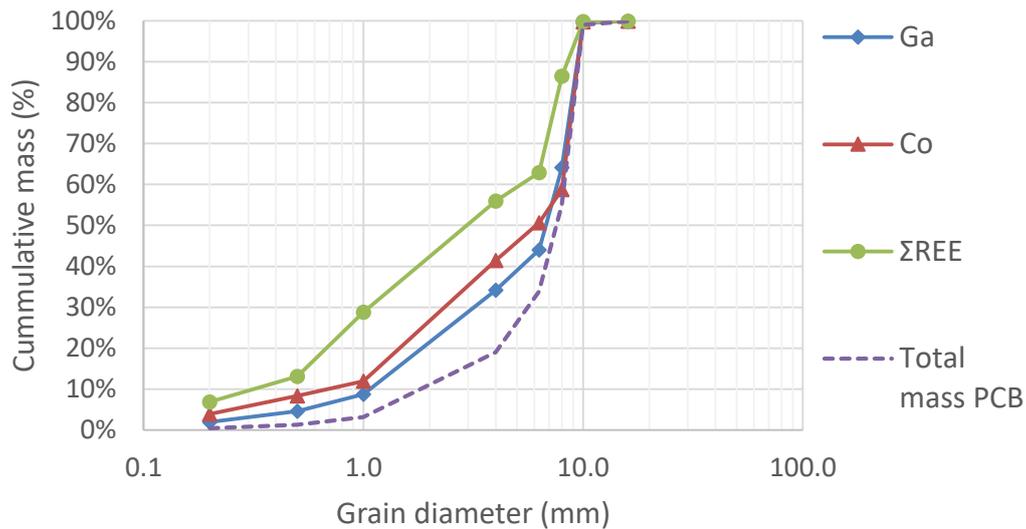


Figure 9: Example of sieve analysis of shredded PCBs (AS4) and cumulative masses of Ga, Co, and ΣREE in screening steps

The supporting information S11 depicts the results for all other studied output fractions regarding IBMs, PMs, and S-CRMs. Supporting information S10 shows the results of the sieve analysis.

3.4. 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).

Figure 10 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 before the mechanical processing. Also, a single battery fraction (MS7) was 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.

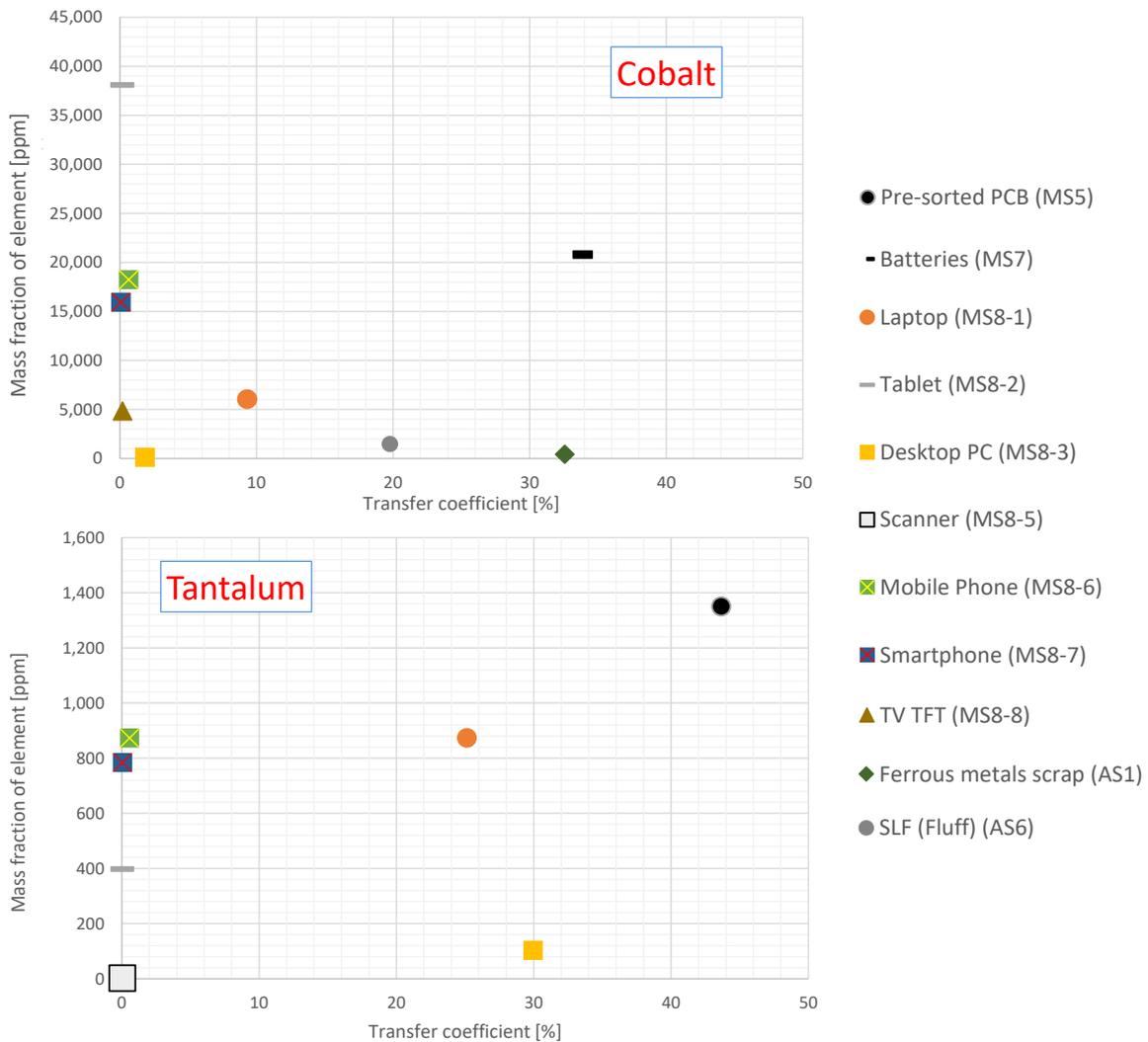


Figure 10: Hotspot plot for cobalt and tantalum in the output fractions with the highest mass fractions

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 significant share of the tantalum loads, with between 25 and 30%. However, the total weight of the laptops and PCs reduced the relative mass fractions. This would appear to be another useful source, but a subsequent separation of the PCB from the device is necessary. This processes 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 11 shows the hotspot diagram of all REE 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 REE. However, the mass fraction was very low. In contrast, about 30% of the Σ 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 REE, whereby the set of applied REE differs for NdFeB magnets, batteries or even lighting products (Rotter et al., 2016; Sommer et al., 2015; Ueberschaar and Rotter, 2015). 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 Σ REE.

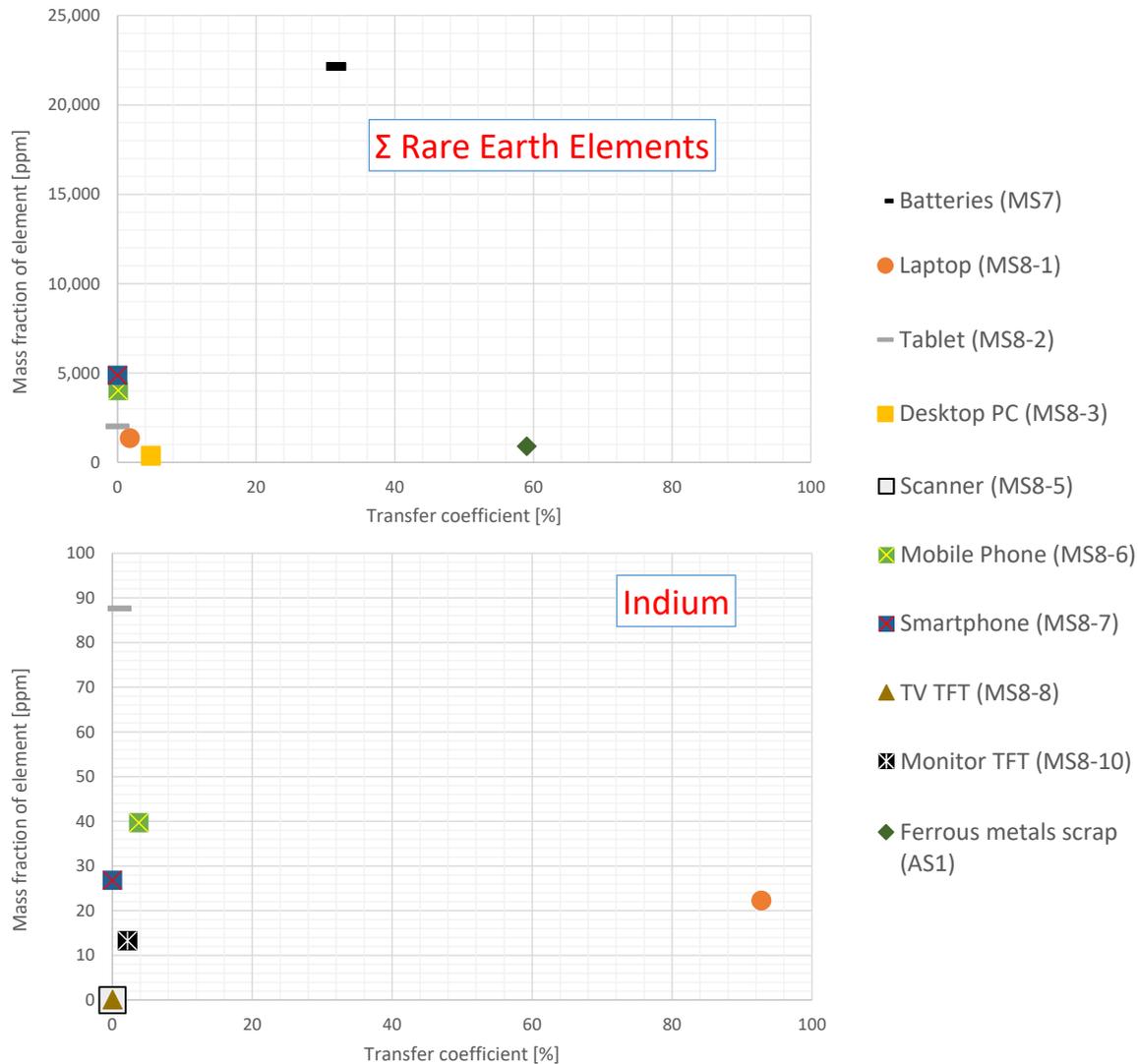


Figure 11: Hotspot plot for Σ REE and indium in the output fractions with the highest mass fractions

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, the highest indium loads were contributed by laptops. 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. Iron and aluminum were dispersed among all output fractions investigated. A presentation in the same form would not be feasible.

Recycling of the investigated elements from the output fractions generated in the pre-processing of WEEE is not always possible or economically feasible. Therefore, a recyclability assessment for each element and output fraction was carried out. Supporting information S15 presents the results.

4. Discussion

Due to the application of a variety of methodologies summarized to one data set to investigate the flows of particular elements, some approaches have to be discussed.

4.1. Sampling

Taking single samples and cumulating them into one total quantity in order to sample continuous material streams can lead to unexpected failures. The characteristics of the single samples investigated represent only a small share of all the material processed. During sampling breaks, significant variations in the composition and subsequently in the mass fractions measured can take place. This “nugget effect” is a chaotic component and can be considered as the variance of an entirely random component (François-Bongarçon, 1994; Pitard, 1994). Supporting information S16 shows an example of a variation of grain sizes and the mass fraction for one element over time. Depending on the sampling schedule, single extreme variations of the mass fraction can pass undiscovered. This leads to an over- or underestimate of the real value.

An approach with much more statistical power could be premised on a sampling methodology based on many single samples taken randomly over time that are subsequently chemically analyzed. By using a large amount 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 the risk of a nugget effect.

4.1.1. Sampling uncertainty for SLF (fluff)

In this study, Gy’s formula was used to determine sampling uncertainties. This methodology focused initially on compact materials. As the shredder light fractions have a very fluffy appearance with some very long fibers, there were doubts about whether the calculations were suited to this kind of output. Similar application problems are known to have arisen for other formulas in some areas (Abzalov, 2011; Bloch von Blottnitz, 1999; Bunge and Bunge, 1999; Dihal, 2012). This suggests that a new development or an adaption of a practicable formula is needed to apply calculated sampling uncertainties to such materials as SLF (fluff).

4.1.2. Sampling uncertainty for chemical analyses

The sample masses directly influence the quality of the results. Based on the physical and chemical properties of the material to be analyzed, uncertainty can be calculated. This has been done for all output fractions sampled in this study.

However, the sampling uncertainty for the chemical analyses is not considered. For a wet-chemical digestion and a subsequent chemical analysis, usually a mass between 0.1-1 g is used, as the prepared sample is assumed to be homogenous. An according sampling uncertainty is considered to be low and so was not part of any calculation in this study.

4.2. Chemical characterization

The main focus for the extended batch test described is the chemical characterization of all flows. However, varying results based on random or systematic effects can lead to much higher or lower loads of the element being investigated. This particularly applies to flows with large shares of the total input. Examples in this study are the ferrous metals scrap AS1 and the sorting residues output AS5.

Also, the detection limits of measurement devices raise similar problems, as element mass fractions below those limits are not considered mass flows. Consequently, total flows can be systematically underestimated for the overall MFA if this flow is set to zero, or systematically overestimated if the limit of detection (LOD) is considered as the average value. In order to improve the sensitivity of the

analysis, a further fractioning of waste fractions through sorting, sieving, and disassembly might be useful prior to a chemical characterization to achieve fractions above the detection limit and to identify hotspots.

4.3. Mathematical model

MFA studies aggregate data from multiple data origins with accompanying uncertainties. Different methodologies and sources can provide such uncertainties in various data formats. However, usually, the applied mathematical model is limited regarding the handling of extended uncertainty information like variability or inherent randomness and unpredictability, which cannot be expressed in a simple value for the mean and standard deviation (Laner et al., 2014). Consequently, different data sets have to be transformed, which always results in a loss of information (Schwab et al., 2016). 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. The previously discussed approach of using developed 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 (Heukelem et al., 2004; Laner et al., 2014; Müller et al., 2014). 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. The related losses of information are the reason why societal MFA are sometimes criticized for an overly simplistic treatment of material flows and processes (Reuter et al., 2006, 2005).

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.

4.4. Method comparison with other MFA studies

To avoid major systematic and random effects, sampling procedures, sample preparation, and chemical analyses play key roles in the execution of experimental MFA studies. A method comparison with other MFA studies (cf. Table 1) reveals a partially insufficient application. In some studies, a sampling plan which includes both the sampling procedures and corresponding sample masses is not documented in detail. This means that sampling uncertainties cannot be deduced and an assessment of sufficient sample masses per output fraction is not possible.

The main function of the sample preparation for chemical characterization is the homogenization associated with comminution and/or remelting. Prior tasks can include sorting, sieving, disassembly, etc. Such processes greatly influence the volatility of particular substances to be measured. Due to a usually uniform application of one chosen sample preparation procedure, process related losses remain unnoticed, which leads to an underestimation of results.

The same applies to the subsequent chemical characterization. Some studies used only one methodological approach for the chemical analyses, meaning the use of just one measurement device and also the same laboratory sample preparation, e.g. acid digestion for wet chemical analyses. As a result, the determination of systematic effects related to the method used is not possible. In most cases, the systematic effect was assessed by an input-output comparison. This can prove the overall

correctness. However, if the input and output are characterized using the same methodologies, the same systematic effects can occur on both sides, which leads to an incorrect mass balance. Therefore, appropriate methods, like those shown in this study, need to be adopted for validating single measurements.

In general, the determination of systematic and random effects plays a minor role in most of the studies reviewed. The application of an uncertainty assessment at all levels of an MFA, like the determination of sampling uncertainties or systematic measurement uncertainties with an appropriate uncertainty propagation, is not usual but is hugely important.

5. Conclusion

Extending the scope of recycling efficiency assessment for pre-processing from bulk materials like IBMs to trace materials like PMs and S-CRMs requires expanded test methodologies to produce meaningful results. Standard methodologies have not yet been established. This applies to sampling, sample preparation, chemical analysis and statistical evaluation.

Batch tests applied for WEEE processing as in the context of WEEELABEX certification can be extended by combining various analytical tools to input and output fractions. This increases the statistical significance of the results and also provides additional information to assess recyclability and technical processes adaptations better.

Some methodological core findings can be summarized as follows:

- Gy's sampling theory is principally applicable for determining sampling strategies and sample sizes but requires a thorough knowledge of the particle characteristics of the batch to be sampled.
- Additional characterization of batch input material by sorting and dismantling helps to identify the systematic effects of the output sampling and the subsequent chemical analysis. Literature data on material composition can support the assessment of the input composition.
- Sorting analysis with particle characterization of pre-processing output fractions (recyclates) supports the evaluation of the material matrix relevant to the recyclability assessment and also identifies deficiencies in the selectiveness of separation steps during pre-processing.
- The combination of sieve analysis with chemical analysis helps to identify hotspots of trace metal distribution. On the one hand, this approach supports lower detection limits for the overall batch. On the contrary, it provides the fractioned chemical analysis with valuable information for further processing to increase the concentration and the purity of output fractions.
- Chemical analysis of PMs and S-CRMs is very sensitive to matrix effects due to the presence of accompanying elements that can lead to an over- or underestimate of elements. Element and matrix specific adaptation of analytical methods is required.

This study further revealed a good separation of the bulk materials for the pre-processing plant investigated. However, valuable materials like copper and PMs are not sufficiently selectively separated. Instead, they are distributed to various output fractions, mainly to the manual sorting output fractions, or to two fractions from automatic sorting (shredded PCBs and sorting residues). Neither necessarily results in an irreversible loss as these materials can be recovered in subsequent processes. The S-CRMs are mostly separated during the manual sorting, with recovery depending on the subsequent processing steps. Exceptions are cobalt and REE. Large proportions of these are concentrated in the ferrous metals scrap and to a lesser extent in the SLF, from which recovery is not feasible.

The systematic methodological assessment approach provided to improve element-specific recycling efficiency is to a large extent based on *a priori* knowledge of materials and material distribution. 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.

Finally, we want to highlight that element extended batch testing can support specific recycling efficiency assessment, but the results represent only a one-off snapshot of a specific plant, for a specific pre-processing configuration and a specific material input. Results should not be generalized and extrapolated.

6. Funding

This work was funded by the German Federal Ministry of Education and Research (BMBF) (UPgrade - Project reference number 033R087)

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Supporting information

Assessment of element-specific recycling efficiency in WEEE pre-processing

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1. S1 Sorting protocol of input material

Table 1: Sorting protocol for input material used for extended batch test

Sample category	Number [-]	Total weight [kg]	Weight [%]	Weight per item [kg]	Notes
<i>Microwave oven</i>	147	2.222	5.47	15.12	incl. combi
<i>Extraction hood</i>	37	449	1.11	12.14	incl. Built-in
<i>Cooking plate</i>	37	272	0.67	7.35	
<i>Barbecue/grill (oven small)</i>	198	926	2.28	4.68	incl. Small grill ovens
<i>Fan/heater (separate)</i>	262	820	2.02	3.13	e.g. radiator heater, table ventilator
<i>Centrifuge</i>	18	212	0.52	11.78	often old centrifuges, no dryers
<i>Fryer</i>	240	872	2.15	3.63	
<i>Coffee/espresso machine</i>	436	1,150	2.83	2.64	incl. combi with water boil function
<i>Vacuum/floor cleaner</i>	700	3,610	8.89	5.16	vacuum/carpet/floor cleaner, carpet sweeper
<i>Personal care appliance</i>	491	439	1.08	0.89	e.g. hair dryer, shaver, weighing devices
<i>Household appliance (small)</i>	1,149	1,924	4.74	1.67	e.g. water cooker, clock, iron, kitchen device
<i>Fabric processing appliance</i>	44	426	1.05	9.68	all textile machines
<i>Audio/video device (not portable)</i>	2,700	9,892	24.36	3.66	e.g. video-/DVD player, hi-fi, loudspeaker
<i>Audio/video device (portable)</i>	685	644	1.59	0.94	e.g. Walkman, mp3 player, camera, headset
<i>Musical instrument</i>	9	46	0.11	5.11	e.g. electrical piano, organ, guitar
<i>Electric (hand) tool</i>	877	3,006	7.40	3.43	e.g. drill, small high pressure cleaner, grass trimmer
<i>Electric toy</i>	100	158	0.39	1.58	very diversified , e.g. electrical trains
<i>Game computer</i>	135	206	0.51	1.53	e.g. Gameboy, PlayStation, incl. Accessories
<i>Desktop Computer</i>	357	3,624	8.92	10.15	
<i>Laptop</i>	153	357	0.88	2.33	
<i>Tablet</i>	2	1	0.00	0.50	
<i>IT-/Office appliance (small)</i>	2,291	1,669	4.11	0.73	e.g. mouse, modem, telephone, calculator, answering machine
<i>Printer/-scanner</i>	716	4,563	11.24	6.37	not the industrial big ones
<i>Mobile phone / smartphone</i>	95	9	0.02	0.09	
<i>Sport-/relaxing device</i>	3	121	0.30	40.33	e.g. home trainer
<i>Household luminaire</i>	841	849	2.09	1.01	e.g. floor/ceiling/ desk lamp
<i>Prof or built-in: Ventilation</i>	0	0	0.00		e.g. window/ wall / roof ventilator
<i>Prof or built-in: Heating/hot water appliance</i>	25	178	0.44	7.12	e.g. boiler, heat radiator panel, terrace heater
<i>Prof: IT-/Office appliance</i>	3	86	0.21	28.67	e.g. copier, mainframe, server, fax

Sample category	Number [-]	Total weight [kg]	Weight [%]	Weight per item [kg]	Notes
<i>Prof: Electrical tool (large)</i>	49	507	1.25	10.35	e.g. compressor, industrial high pressure cleaner
<i>Prof: Medical/health appliance</i>	0	0	0.00		e.g. hospital, dentist equipment
<i>Prof or built-in: Measure/control appliance</i>	29	194	0.48	6.69	e.g. security camera
<i>Prof or built-in: Detector/sensor/switch</i>	0	0	0.00		e.g. smoke/ fire detector, movement sensor
<i>Prof: Luminaire (inside)</i>	87	356	0.88	4.09	e.g. TL luminaire, emergency lighting
<i>Prof: Luminaire (outside)</i>	28	34	0.08	1.21	e.g. street light, traffic light
<i>Washing machine/dishwasher/dryer</i>	3	116	0.29	38.67	excl. Loose parts of large household appliance
<i>Cooling/freezing device (& doors)</i>	9	290	0.71	32.22	e.g. fridge, deep freezer, air conditioner
<i>TV/monitor</i>	64	378	0.93	5.91	incl. Broken ones
Total	13,020	40,606	100		

2. S2 Output fraction coding

Table 2: Codes for all individual output fractions in the extended batch test partially grouped for better readability

Treatment step	Output fraction description	Output fraction code	Sub-category descriptions	Sub-category code
Manual sorting (MS)	Tools scrap	MS1	-	
	E-Motors	MS2	-	
	Loudspeaker	MS3	-	
	Copper rich fraction	MS4	Mixed cables and Cu/Brass	MS4-1
			Cu-anchors, transformers	MS4-2
	Pre-sorted PCB	MS5	-	
	Ferrous metals rich materials	MS6	Rotary shear scrap	MS6-1
			Sheet-metal scrap	MS6-2
	Batteries	MS7	-	
	IT devices	MS8	Laptop	MS8-1
			Tablet	MS8-2
			Desktop PC	MS8-3
			Printer	MS8-4
			Scanner	MS8-5
			Mobile Phone	MS8-6
			Smartphone	MS8-7
			TV TFT	MS8-8
TV CRT			MS8-9	
Monitor TFT			MS8-10	
Monitor CRT			MS8-11	
Mowing machines	MS9	-		
Cooling and air conditioning appliances	MS10	-		
Metal-poor material	MS11	Wooden boxes and wood	MS11-1	
		Commercial and residual waste	MS11-2	
Hazardous material	MS12	Capacitors	-	
Automated sorting (AS)	Ferrous metals scrap	AS1	-	
	Low magnetic material	AS2	-	
	Non-ferrous metals scrap	AS3	-	
	Shredded PCB	AS4	-	
	Sorting residues (mixed plastics)	AS5	-	
	SLF (Fluff)	AS6	-	
	Filter dust	AS7	-	
	Sweepings	AS8	-	

Note: The sub-categories of the output fraction IT-devices do not represent individual output fractions, but are listed as those due to their potentials of precious metals like gold, silver, palladium and critical metals in general like indium, tantalum or rare earth elements.

3. S3 Calculation of required primary sample amounts

Table 3: Overview of fractions to be sampled with according properties, sample volumes, masses and reliability factors

Sampled Output	Predicted output	Bulk density	Predicted volume of total output	Single sample volume	Number single samples	Calc. sampling quantity	
	[kg]	[kg/m ³]	[m ³]	[l]		[l]	[kg]
Sorting residues (mixed plastics)	10,000	440	22.7	2	8	16	7
Ferrous metals scrap	6,000	1,500	4.0	2	8	16	24
Non-ferrous metals scrap	1,100	1500	0.7	2	8	16	24
SLF (Fluff)	1,000	210	4.8	5	8	40	8
Low magnetic material	600	1,000	0.6	2	8	16	16
Shredded PCB	460	500	0.9	3	8	24	12
Filter dust	170	210	0.8	1	8	8	2

4. S4 Design of sample divider for large sample masses

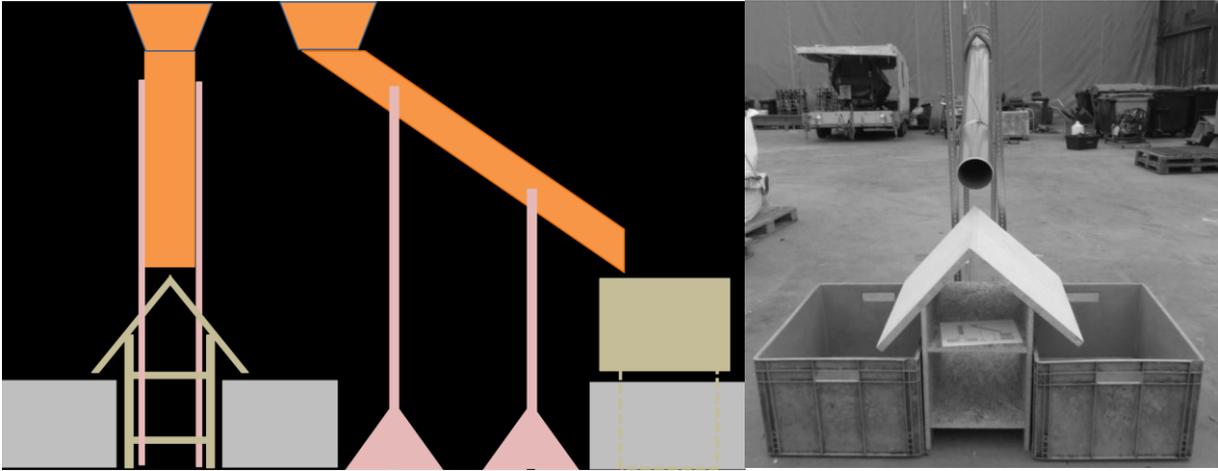


Figure 1: Design of divider for large sample masses

5. S5 Sorting analysis protocol

Table 4: Sorting analysis protocol

Sorting analysis - Composition of materials			
Sorting fraction	Weight [g]	Photo number	Comment
Ferromagnetic particles			
Aluminum			
Other non-ferrous metals			
Plastics colored			
Plastics black			
Printed circuit boards			
Batterys / Electronics			
Organic / textile			
Cable			
Foam / rubber / foils			
Glass / ceramics			
Compounds			
Rest			
Sum not sorted material (<5 mm)			
Sum			

6. S6 Disassembly protocols

This disassembly form was used for e-motors, electric tools scrap (drilling machines with cable and battery, angle grinder, electric chain saw, electric hedge trimmer, sewing machine, grass trimmer with battery) and loudspeaker (driver).

Table 5: Disassembly protocol used for disassemblies of components and devices from manually sorted output

Sample number				
Disassembly object/device				
Total mass [g]				
Ferrous metals in g	Stator from motor			
	Rotor from motor			
	Switch			
	Fe/Al			
	Fe/plastics			
	Fe/Cu/plastics			
		Plastics/Fe		
		misc		
	Total Fe metals			
	Al	NF metals / plastics		
		Plastics / NF metals		
		Plastics / Al		
		Fe/Al		
		misc		
	Total Al			
	Cu	Cu/plastics		
		Cu from stator		
		Cu from rotor		
		Switch		
		Plastics / Cu		
		Fe/Cu		
		Fe/Cu/Plastics		
		Misc		
	Total Cu			
	Total non-ferrous metals			

Sample number			
Disassembly object/device			
Misc in g	Battery		
	Capacitor		
	Printed Circuit Board		
	Misc		
	Total Misc		
Plastics in g	Switch		
	Plastics / Fe		
	Plastics / Al		
	Plastics / Cu		
	Plastics / NF metals		
	Al / plastics		
	NF / plastics		
	Cu / plastics		
	Fe / plastics		
	Fe/Cu/plastics		
	Misc		
	Total plastics		
Magnet in g	Weight		
	Total magnet		
Σ			

7. S7 Data on composition of output materials from manual sorting

This data was generated by literature review, but also dismantling studies of poorly described devices, components, and materials with subsequent material and element characterization via wet-chemical approaches (ICP-OES) and/or XRF.

7.1. Composition IT devices

Table 6: Composition of IT devices

%	Lap-tops	Tablet	Desk-top PC	Printer	Scanner	Mobile Phone	Smart Phone	TV TFT	TV CRT	Monitor TFT	Monitor CRT
Ag	1.11E-02	2.01E-02	1.33E-02	5.48E-04	1.14E-03	1.79E-01	3.96E-02	2.87E-03	3.25E-03	4.46E-03	1.43E-03
StDev	7.38E-03	1.81E-03	7.27E-03	6.07E-05	0.00E+00	1.08E-01	0.00E+00	1.74E-03	4.53E-03	3.36E-03	
Al	2.80E+00	1.18E+01	9.84E+00	1.79E+00	1.79E+00	4.16E+00	2.10E+01	5.39E+00	1.37E+00	4.64E+00	2.05E+00
StDev		1.29E+01	6.61E+00	1.49E+00	1.49E+00	3.79E+00	2.69E+01	1.26E+00	9.73E-01		
As	3.25E-04	1.70E-03	1.00E-03			1.49E-03	3.05E-03	1.01E-04	0.00E+00	3.02E-04	
StDev	2.99E-04	1.22E-04				1.01E-03	0.00E+00	8.17E-05		2.41E-04	
Au	7.08E-03	8.25E-03	1.70E-03	3.04E-04	3.65E-04	3.26E-02	1.62E-02	1.04E-03	3.34E-04	3.63E-03	
StDev	2.15E-03	7.45E-04	5.29E-04	8.49E-05	1.74E-04	1.37E-02	0.00E+00	8.55E-04	4.29E-04	4.26E-04	
Ba	7.14E-02	7.97E-02	2.45E-02	2.37E-02		4.30E-01	1.57E-01	3.05E-02	1.03E-02	2.33E-02	
StDev	1.70E-02	7.20E-03	7.78E-03	1.73E-03		3.30E-01	0.00E+00	3.86E-03	1.45E-02	1.26E-02	
Be	0.00E+00	6.28E-05	2.00E-02			3.65E-03	1.24E-04	0.00E+00	1.20E-03	0.00E+00	
StDev		5.67E-06				2.28E-03	0.00E+00	0.00E+00	1.70E-03	0.00E+00	
Bi	1.50E-03	3.69E-03	2.29E-03	7.17E-05			7.26E-03				
StDev		3.33E-04	3.22E-03								
Ca		9.99E-02					1.97E-01				
StDev		9.03E-03									
Cd	0.00E+00	1.24E-05	9.00E-03			7.85E-05	1.97E-01	0.00E+00	3.35E+00	0.00E+00	
StDev	0.00E+00	1.12E-06				1.00E-04		0.00E+00	4.74E+00	0.00E+00	
Ce	5.45E-06	3.39E-04				8.10E-02	6.68E-04	1.26E-05			
StDev		3.06E-05						1.56E-05			
Co	6.03E-01	3.81E+00	1.17E-02	3.04E-04	4.08E-04	1.82E+00	1.59E+00	4.83E-01	3.64E-04	8.57E-05	1.23E-03
StDev	1.48E+00	4.39E-01	1.17E-02	9.53E-05		2.76E+00		1.18E+00	3.97E-04	2.27E-04	
Cu	3.81E+00	3.90E+00	5.93E+00	6.10E+00		1.19E+01	4.72E+00	1.38E+00	3.16E+00	1.96E+00	4.93E+00
StDev	1.48E+00	4.02E-01	2.08E+00	5.00E+00		3.30E+00		8.64E-01	1.05E+00	1.87E+00	

%	Lap-tops	Tablet	Desk-top PC	Printer	Scanner	Mobile Phone	Smart Phone	TV TFT	TV CRT	Monitor TFT	Monitor CRT
Dy	3.34E-03		4.00E-03			1.59E-02					
StDev											
Eu	2.23E-06					2.27E-03		2.16E-05		1.43E-05	
StDev								2.98E-05			
Fe	2.00E+01	8.86E+00	1.97E+01	3.88E+01	3.88E+01	7.87E+00	3.22E+00	3.92E+01	1.00E+01	3.71E+01	8.12E+00
StDev		5.71E-01	1.55E+01	8.77E+00	8.77E+00	4.31E+00		5.44E+00	7.04E+00		
Ga	1.10E-04	5.86E-04	5.50E-04	1.22E-05	2.52E-06	5.30E-03	1.16E-03	8.60E-06	0.00E+00	1.65E-05	
StDev	4.23E-05	5.30E-05	6.36E-04	5.37E-06				1.49E-05			
Gd	3.76E-05					1.41E-01					
StDev						1.44E-01					
In	2.23E-03	8.77E-03			1.26E-07	3.97E-03	2.68E-03	1.49E-05		1.33E-03	
StDev		1.04E-03			7.25E-07			0.00E+00			
La	6.12E-07	5.80E-03					1.14E-02				
StDev		5.24E-04									
Li		7.14E-01					9.43E-04				
StDev		8.23E-02									
Mg		2.60E+00					4.83E-02				
StDev		2.62E+00									
Mn			3.00E-02			1.72E-01	0.00E+00				
StDev						1.26E-01					
Mo	1.18E-03	6.05E-04					1.19E-03				
StDev	2.09E-03	5.46E-05									
Nd	1.17E-01	1.62E-01	3.00E-02			1.41E-01	3.95E-01				
StDev		9.30E-03				1.44E-01					
Ni	1.03E-01	4.34E-01	5.25E-01			1.34E+00	5.78E-01	1.33E-02	2.70E-02	2.04E-02	
StDev	2.25E-02	4.12E-02	4.60E-01			1.16E+00		9.86E-03	2.36E-02	2.35E-02	
Pt	2.23E-04	1.13E-03					2.23E-03				
StDev		1.02E-04									
Pd	2.51E-03	1.73E-03	6.67E-04	1.62E-04	1.41E-04	1.10E-02	3.41E-03	8.79E-05	2.16E-04	7.37E-04	3.28E-05
StDev	4.06E-04	1.56E-04	5.51E-04	1.21E-05	4.49E-04	3.49E-03		1.11E-04	5.04E-05	9.91E-05	

%	Lap-tops	Tablet	Desk-top PC	Printer	Scanner	Mobile Phone	Smart Phone	TV TFT	TV CRT	Monitor TFT	Monitor CRT
Pb	1.48E-01	7.49E-02	3.50E+00	7.88E-02		2.84E-01	1.47E-01	7.89E-02	1.66E+00	5.86E-02	6.08E-02
StDev	4.19E-02	6.76E-03	3.10E+00	1.73E-02		1.81E-01		6.03E-02	2.12E+00	3.17E-02	
Pr	1.50E-02	3.24E-02	2.00E-03				7.89E-02	0.00E+00		2.22E-07	
StDev		1.86E-03						0.00E+00			
Sb	2.18E-02	8.18E-03	2.55E-01			5.67E-02	1.61E-02	5.51E-03	3.70E-02	5.29E-03	2.06E-02
StDev	1.06E-02	7.39E-04	3.47E-01			3.83E-02		2.72E-03	1.49E-02	4.28E-03	
Si		3.91E-01					7.70E-01				
StDev		3.53E-02									
Sm						1.65E-02					
StDev											
Sn	2.20E-01	2.49E-01	5.90E-01	1.22E-01	1.67E-01	7.74E-01	3.14E-01	1.97E-01	1.03E-01	1.04E-02	5.53E-03
StDev		3.39E-03	5.80E-01	1.91E-02		3.30E-01		1.88E-01	5.10E-02		
Sr	5.20E-03	5.63E-03	3.50E-03	1.38E-03		3.92E-02	1.11E-02	3.50E-03	4.90E-03		
StDev		5.09E-04		1.91E-04		3.23E-02					
Ta	8.73E-02	3.98E-02	1.03E-02		2.00E-04	8.73E-02	7.84E-02	0.00E+00	0.00E+00		
StDev	1.03E-02	3.59E-03	1.38E-02		1.15E-03	1.59E-02					
Tb	2.23E-07					2.04E-03				3.97E-06	
StDev											
Te											
StDev											
Ti	0.00E+00	1.20E-01	2.00E-02				2.36E-01				
StDev		1.08E-02									
V	0.00E+00	1.34E-04					2.64E-04				
StDev		1.21E-05									
W											
StDev											
Y	9.02E-05	1.01E-03			1.26E-07	2.04E-03	1.98E-03			2.03E-04	
StDev		9.08E-05			7.25E-07						
Zn	4.21E-02	5.44E-02	1.14E+00	3.27E-02		6.03E-01	8.91E-02	5.35E-02	8.46E-02	2.90E-03	1.77E-01
StDev	9.39E-02	2.88E-03	1.09E+00	3.99E-03		3.34E-01		9.70E-02		7.66E-03	
Zr		1.63E-02					3.22E-02				
StDev		1.48E-03									

%	Lap-tops	Tablet	Desk-top PC	Printer	Scanner	Mobile Phone	Smart Phone	TV TFT	TV CRT	Monitor TFT	Monitor CRT
Plastics	3.36E+01	9.92E+00	1.53E+00	4.58E+01	4.58E+01			2.91E+01			
StDev	2.58E+01	6.60E+00		6.87E+00	6.90E+00			1.45E+01			

Table based on: (Anindya et al., 2014; Arshadi and Mousavi, 2015; Buchert et al., 2012a, 2012b; Christian et al., 2014; Cui and Forssberg, 2007; Cui and Zhang, 2008; Dervišević et al., 2013; Fitzpatrick et al., 2015; Huisman, 2004; Juchneski et al., 2013; Kim et al., 2013; Laboratory and Hazardous Material Laboratory, 2004; Lim and Schoenung, 2010; Maragkos et al., 2013; Matsuto et al., 2004; Milovantseva and Saphores, 2013; Oguchi et al., 2011; Savvilotidou et al., 2014; Schischke et al., 2013; Song et al., 2013; Townsend et al., 2004; Tuncuk et al., 2012; Ueberschaar et al., 2017; Unep, 2009; United Nations University, 2007; Valero Navazo et al., 2014; Wu et al., 2008; Xiu et al., 2015) and own studies

7.2. Composition of household tools

Table 7: Composition of household tools

%	Battery-powered screwdriver	Battery trimmer	Hedge trimmer	Drilling machine	Chain saw	Angle grinder	Sewing machine	Power supply	misc. Electronics
Fe	6.20E-01	1.24E-01	3.29E+00	3.21E+00	2.01E-01	1.41E+00	1.08E+00	1.58E+00	4.04E-03
StDev	1.32E-01	2.43E-02	3.27E-01	1.22E+00	2.00E-02	1.33E-01	6.06E-01	5.09E-01	1.61E-03
Cu	5.63E-02	1.94E-02	3.77E-01	8.75E-01	2.31E-02	4.59E-01	3.41E-01	5.20E-01	5.18E-02
StDev	4.33E-02	4.20E-03	4.29E-03	3.34E-01	2.62E-04	5.77E-02	1.94E-01	1.93E-01	4.21E-03
Al	3.89E-02	7.54E-03		8.91E-02		3.07E-01	7.58E-01	4.76E-02	3.30E-03
StDev	4.57E-02	4.26E-03		3.34E-01		2.94E-01	7.67E-01	3.92E-02	1.43E-03
Sn	8.67E-05							2.93E-03	3.20E-03
StDev	1.37E-04							5.98E-03	1.19E-03
Au	2.13E-06							7.18E-05	7.85E-05
StDev	1.01E-05							4.40E-04	8.76E-05
Ag	3.43E-06							1.16E-04	1.26E-04
StDev	6.85E-05							2.99E-04	5.96E-05
Pd	2.77E-07								1.02E-05
StDev	6.18E-07								5.37E-06
Pt	5.55E-08								2.05E-06
StDev	1.09E-07								9.47E-07
Pb	5.34E-05							1.80E-03	1.97E-03
StDev	8.64E-05							3.77E-03	7.51E-04
Zn	4.02E-05	1.32E-03						1.12E-02	1.48E-03
StDev	1.07E-04	2.61E-04						5.33E-03	9.30E-04
Ni	3.82E-05	1.91E-02						1.29E-03	1.41E-03
StDev	8.46E-05	1.28E-03						3.69E-03	7.35E-04
Co	6.73E-07	6.50E-03						2.27E-05	2.48E-05
StDev	1.92E-06	5.52E-04						8.39E-05	1.67E-05
Ce	1.13E-06	4.22E-03						3.83E-06	4.19E-06
StDev	3.21E-07	5.52E-04						1.40E-05	2.79E-06
La	6.81E-08	7.47E-03						2.30E-06	2.51E-06
StDev	1.93E-07	1.25E-03						8.41E-06	1.67E-06
Nd		6.15E-03							
StDev		1.63E-03							

%	Battery-powered screwdriver	Battery trimmer	Hedge trimmer	Drilling machine	Chain saw	Angle grinder	Sewing machine	Power supply	misc. Electronics
Pr		1.76E-03							
StDev		4.94E-04							
Sr	4.34E-03	9.15E-06							
StDev	3.75E-05	1.31E-07							
Ba	1.57E-04	5.43E-03							
StDev	3.83E-06	3.93E-05							
Plastics	3.69E-01	1.32E-01	4.13E-01	1.94E+00	2.52E-02	2.52E-01	7.14E-02	6.87E-01	6.05E-02
StDev	1.37E-01	3.76E-02	3.21E-01	9.99E-01	1.96E-02	1.83E-01	8.70E-02	2.72E-01	1.24E-02
Misc									
StDev									

Based on: own studies and (Chancerel, 2010; Chancerel et al., 2010, 2009)

Note: critical metals were suggested in batteries, magnets, and printed circuit boards. Only in battery driven devices (here trimmer) NdFeB magnets were found! The mixed tools fraction is depicted in 7.4.

7.3. Composition of batteries

Table 8: Composition and share of battery types

%	NiCd	NiMH	Li-Ion	Pb	Primary cells	misc.
Share in average battery mix	14	10	9	32	30	3
Al		8.50E-01	9.31E+00			
StDev		3.54E-01	6.57E+00			
Co	1.07E+00	3.76E+00	1.73E+01			
StDev	4.04E-01	1.12E+00	4.22E+00			
Fe	3.89E+01	1.83E+01	1.67E+01	2.00E+01	2.05E+01	2.00E+01
StDev	5.10E+00	1.41E+01	1.63E+01	5.00E+00	5.00E+00	
Ni	2.19E+01	3.95E+01	3.75E+00			
StDev	2.04E+00	7.73E+00	3.28E+00			
Zn		1.45E+00				
StDev		9.00E-01				
Cu			9.69E+00		2.00E+01	1.00E-01
StDev			1.84E+00			
Mn		1.54E+00	1.00E-01			
StDev		6.07E-01				
Cd	1.49E+01	2.80E+00				
StDev	1.30E+00					
Ce		4.75E+00				
StDev		1.91E+00				
La		8.45E+00				
StDev		4.31E+00				
Nd		6.95E+00				
StDev		5.59E+00				
Pr		2.00E+00				
StDev		1.70E+00				
Lithium			2.94E+00			
StDev			1.33E+00			
Plastics	5.95E+00	7.00E+00	4.13E+00	6.00E+00		
StDev	2.76E+00	2.83E+00	4.07E+00	3.00E+00		
Pb				6.00E+01		
StDev				1.50E+01		

Based on: (Sommer et al., 2015) and other own studies

7.4. Composition of mixed and other important fractions

Table 9: Composition of mixed and other important fractions

%	Cooling and air conditioning appliances	Tools scrap	Loudspeaker	E-Motors	Mower	Batteries
Ag	5.00E-04	2.45E-04				
StDev		3.13E-04				
Al	1.31E+01	1.57E+00	4.09E-01	3.82E+00	1.34E+00	9.23E-01
StDev	5.59E+00	9.44E-01	2.98E-01	3.78E+00	1.32E+00	6.27E-01
As						
StDev						
Au	3.90E-05	1.52E-04				
StDev		4.49E-04				
Ba	8.20E-04	5.59E-03	3.83E+00			
StDev		3.95E-05	6.79E-02			
Be						
StDev						
Bi	0.00E+00					
StDev						
Ca						
StDev						
Cd						2.37E+00
StDev						1.83E-01
Ce		4.23E-03				4.75E-01
StDev		5.52E-04				1.91E-01
Co	8.80E-05	6.55E-03				2.08E+00
StDev		5.59E-04				5.49E-01
Cu	1.25E+01	4.66E+00	9.65E-01	2.22E+01	8.48E+00	6.88E+00
StDev	7.78E+00	5.27E-01	3.19E+00	3.48E+00	2.02E+00	1.65E-01
Dy						
StDev						
Eu						
StDev						
Fe	5.10E+01	7.74E+01	7.93E+01	7.28E+01	6.05E+01	2.19E+01
StDev	5.66E+00	6.80E+00	2.44E+01	5.03E+00	1.18E+01	6.69E+00
Ga	0.00E+00					
StDev						
Gd						
StDev						
In						
StDev						
La		7.48E-03				8.45E-01
StDev		1.25E-03				4.31E-01
Li						2.65E-01
StDev						1.20E-01

%	Cooling and air conditioning appliances	Tools scrap	Loudspeaker	E-Motors	Mower	Batteries
Mg						
StDev						
Mn						1.63E-01
StDev						6.07E-02
Mo						
StDev						
Nd		6.15E-03				6.95E-01
StDev		1.63E-03				5.59E-01
Ni		2.18E-02	6.21E-03			7.34E+00
StDev		3.98E-03	5.82E-03			1.35E+00
Pt		2.10E-06				
StDev		9.53E-07				
Pd	0.00E+00	1.05E-05				
StDev		5.40E-06				
Pb	1.70E-02	3.82E-03				1.92E+01
StDev		3.85E-03				4.80E+00
Pr		1.76E-03				2.00E-01
StDev		4.94E-04				1.70E-01
Sb						
StDev						
Si						
StDev						
Sm						
StDev						
Sn	5.00E-02	6.21E-03				
StDev		6.10E-03				
Sr	7.00E-01	4.35E-03	1.30E+00			
StDev		3.75E-05	1.88E-02			
Ta	0.00E+00					
StDev						
Tb						
StDev						
Te						
StDev						
Ti						
StDev						
V						
StDev						
W						
StDev						
Y						
StDev						

%	Cooling and air conditioning appliances	Tools scrap	Loudspeaker	E-Motors	Mower	Batteries
Zn	1.20E-02	1.41E-02	9.69E-02			1.45E-01
StDev		5.42E-03	2.97E-03			9.00E-02
Zr						
StDev						
Plastics	1.90E+01	5.35E+00	2.97E+00	5.79E-01	1.65E+01	3.82E+00
StDev	1.77E+01	1.11E+00	5.07E+00	5.02E-01	1.10E+01	1.99E+00
Misc		3.77E+00	9.26E-02	5.34E-01	1.87E-01	
StDev		1.42E-01	4.24E-01	9.25E-01	3.24E-01	

Based on (Hester and Harrison, 2008; Oguchi et al., 2011; Sommer et al., 2015); tools, loudspeaker and e-motors have been dismantled and relevant materials determined by an XRF analysis.

8. S8 Chemical analysis of output fractions from automated processing

This data represents the results of chemical analyses carried out on the generated output fraction from the automated separation within the batch test.

8.1. Manually pre-sorted Printed Circuit Boards (MS5)

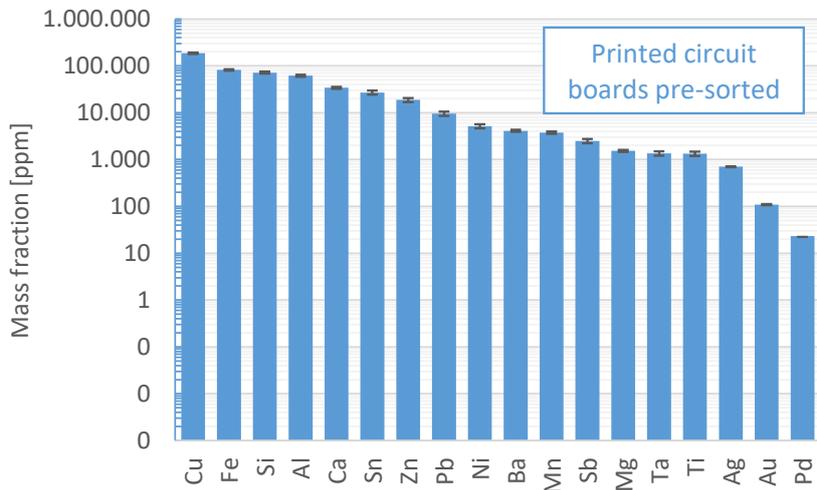


Figure 2: Composition of manually pre-sorted printed circuit boards (MS5)

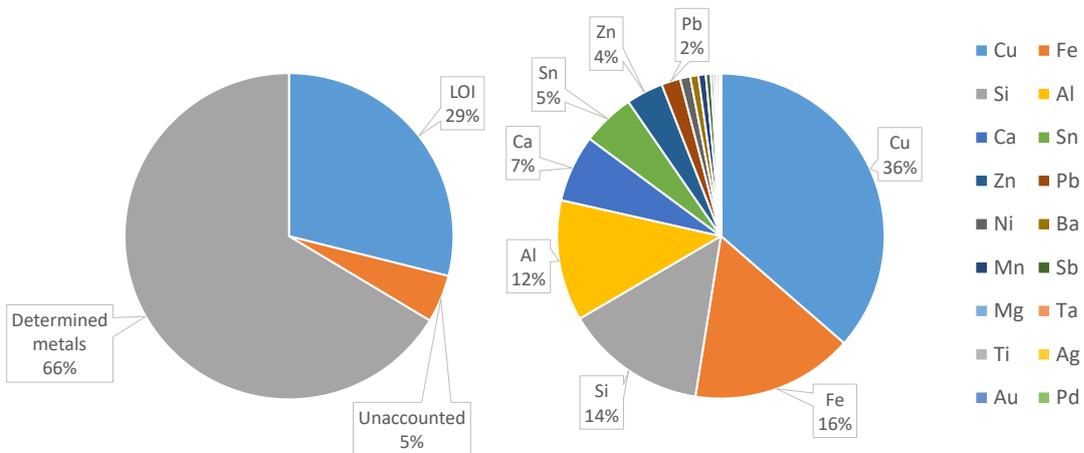


Figure 3: Overview of accounted materials and loss of ignition for manually pre-sorted printed circuit boards (MS5)

8.2. Ferrous metals scrap (AS1)

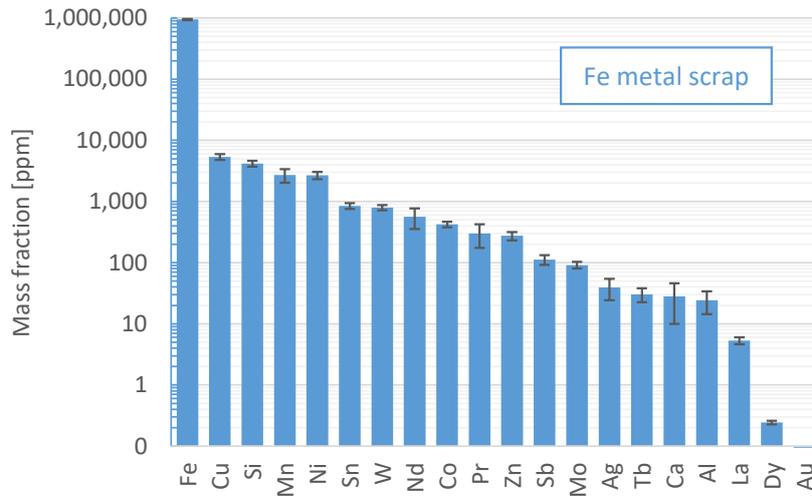


Figure 4: Composition of ferrous metals scrap (AS1)

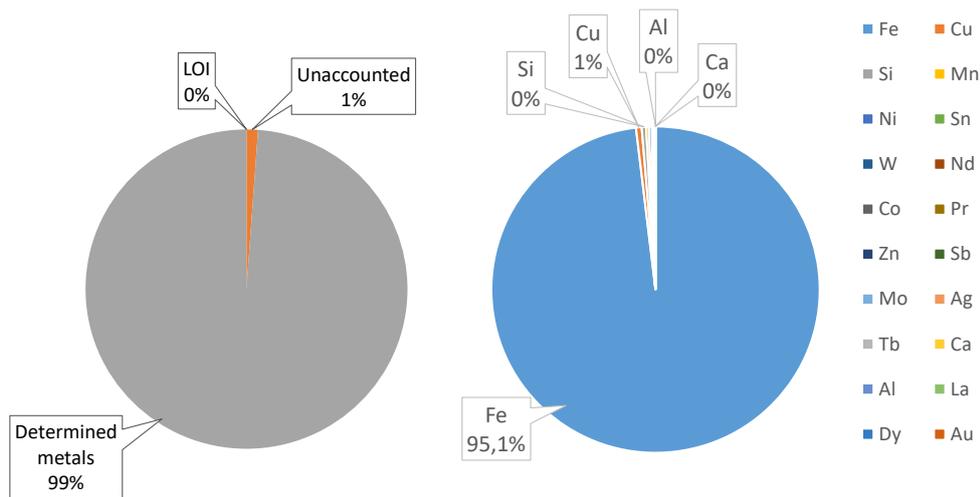


Figure 5: Overview of accounted materials and loss of ignition for ferrous metals scrap (AS1)

8.3. Low magnetic material (AS2)

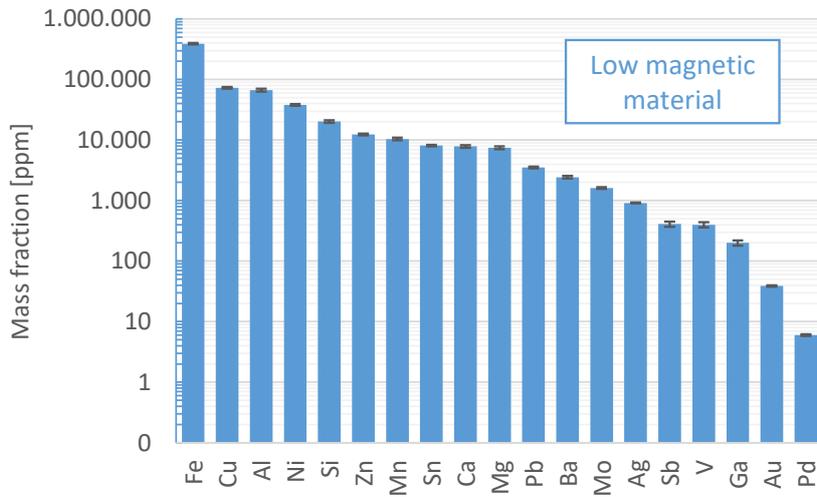


Figure 6: Composition of low magnetic material (AS2)

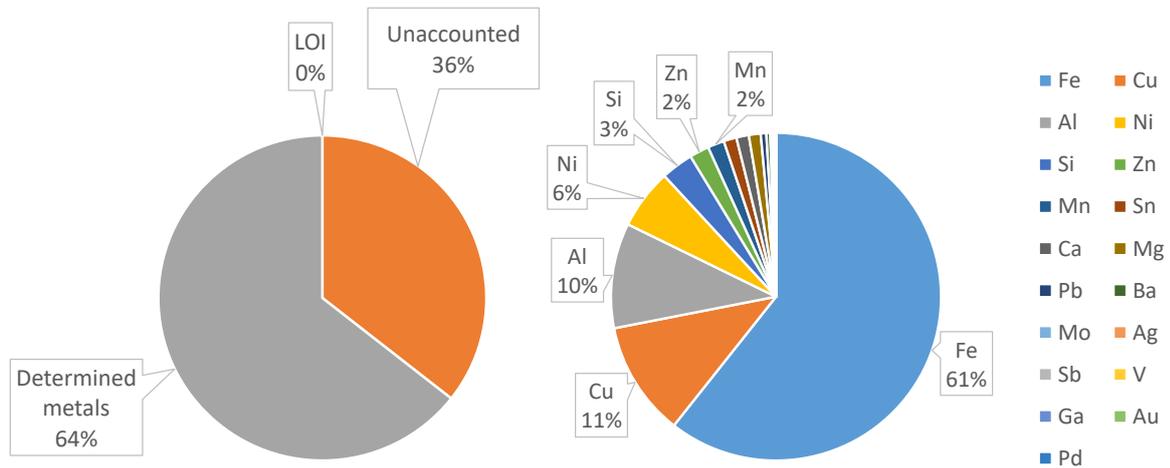


Figure 7: Overview of accounted materials and loss of ignition for low magnetic material (AS2)

8.4. Non-ferrous metals scrap (AS3)

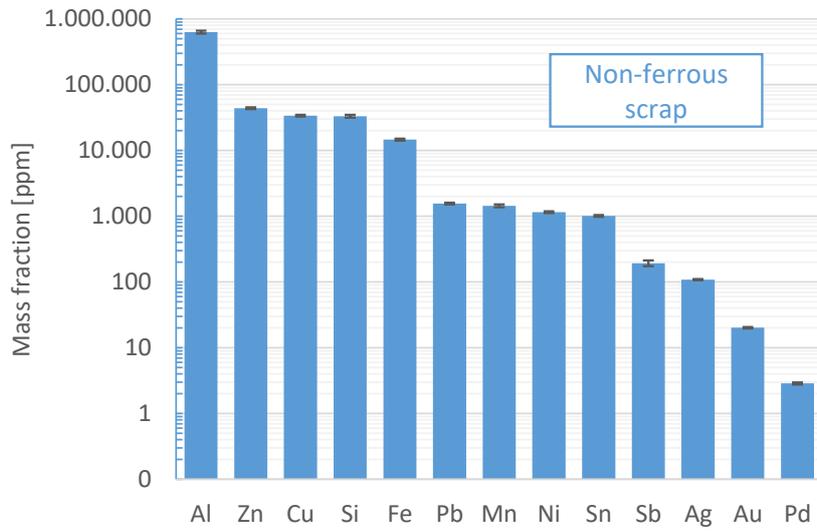


Figure 8: Composition of non-ferrous metals scrap (AS3)

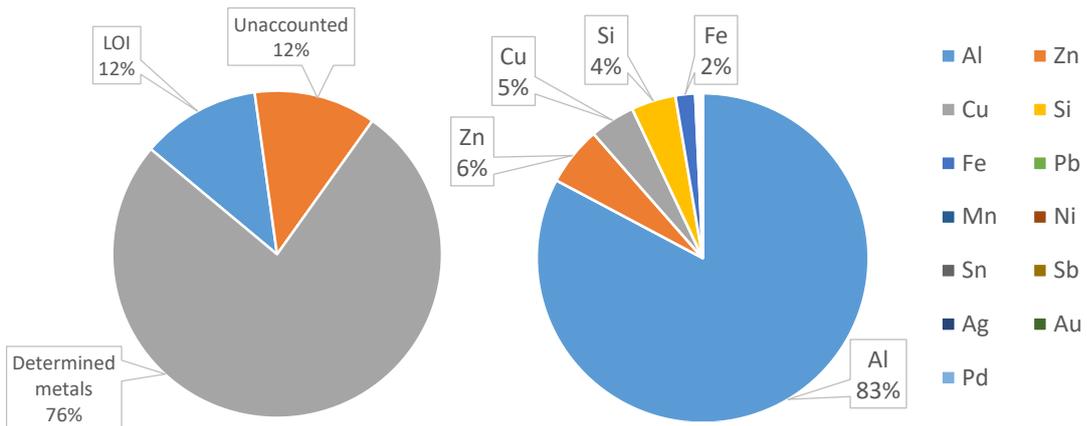


Figure 9: Overview of accounted materials and loss of ignition for non-ferrous metals scrap (AS3)

8.5. Shredded printed circuit boards (AS4)

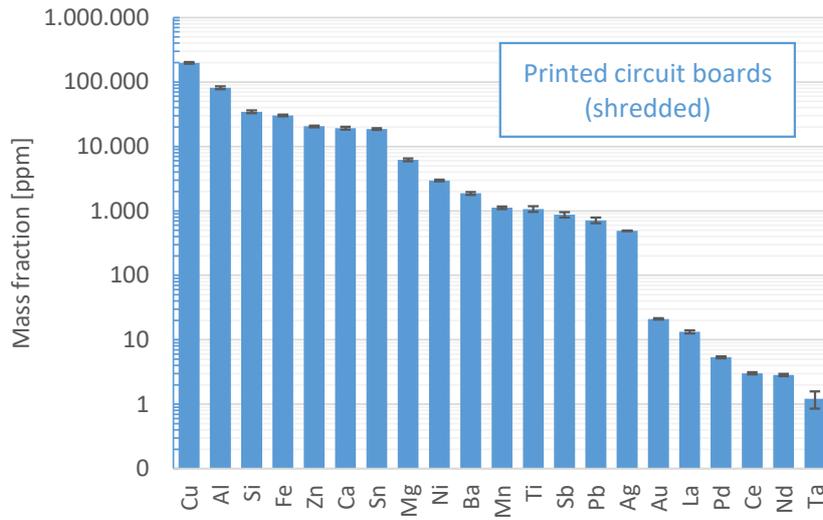


Figure 10: Composition of shredded printed circuit boards (AS4)

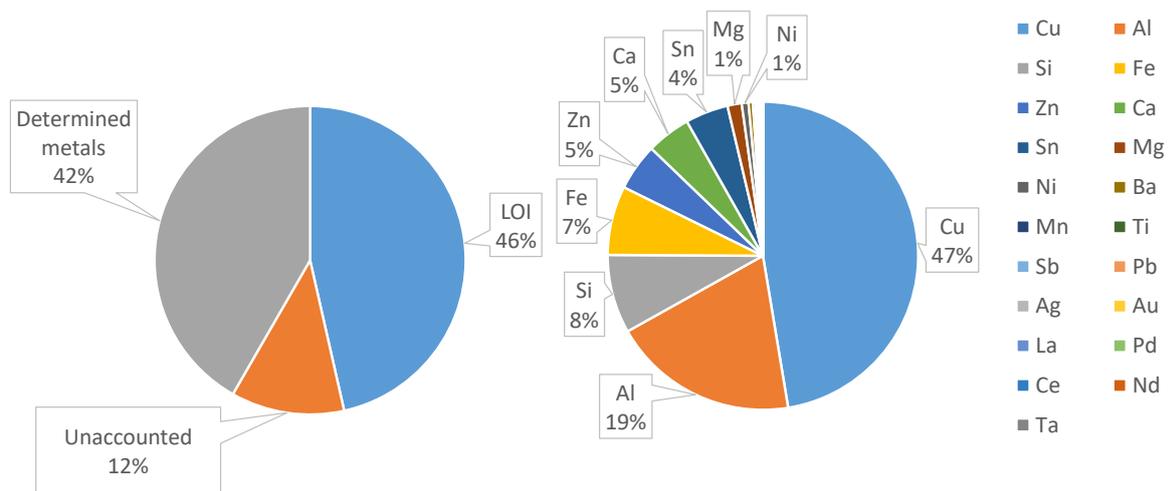


Figure 11: Overview of accounted materials and loss of ignition for shredded printed circuit boards (AS4)

8.6. Sorting residues (mixed plastics) (AS5)

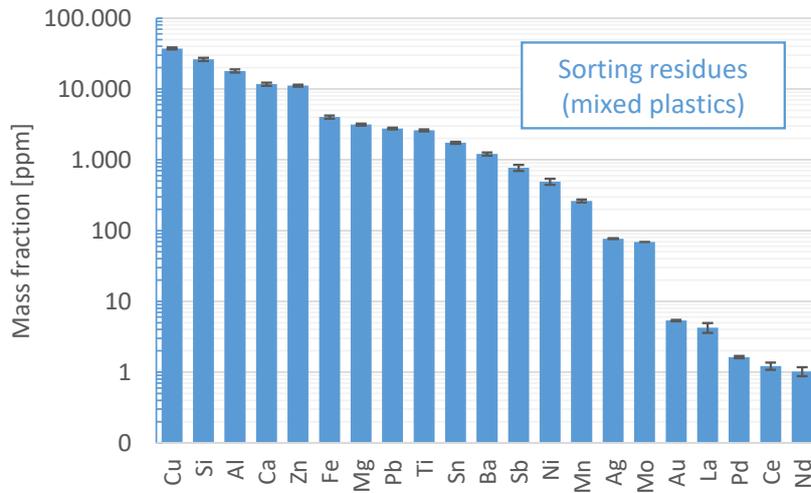


Figure 12: Composition of sorting residues (mixed plastics) (AS5)

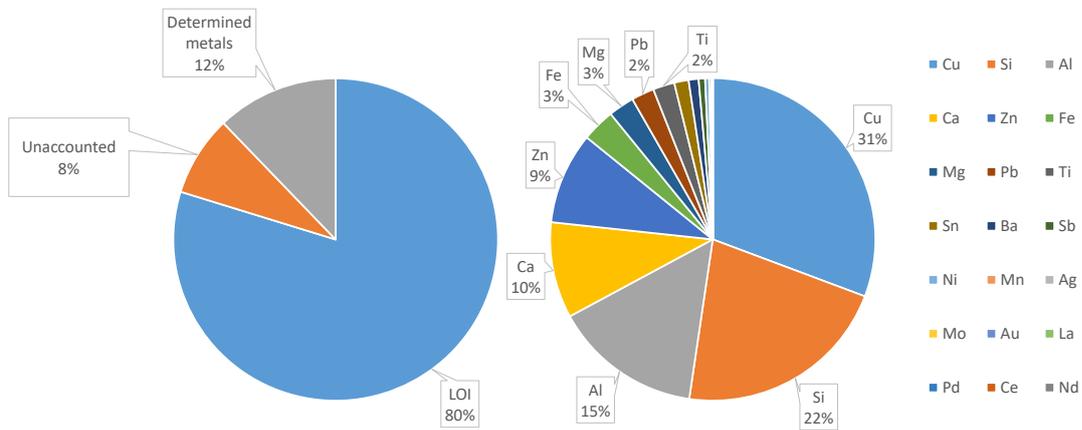


Figure 13: Overview of accounted materials and loss of ignition for sorting residues (mixed plastics) (AS5)

8.7. Shredder light fraction (fluff) (AS6)

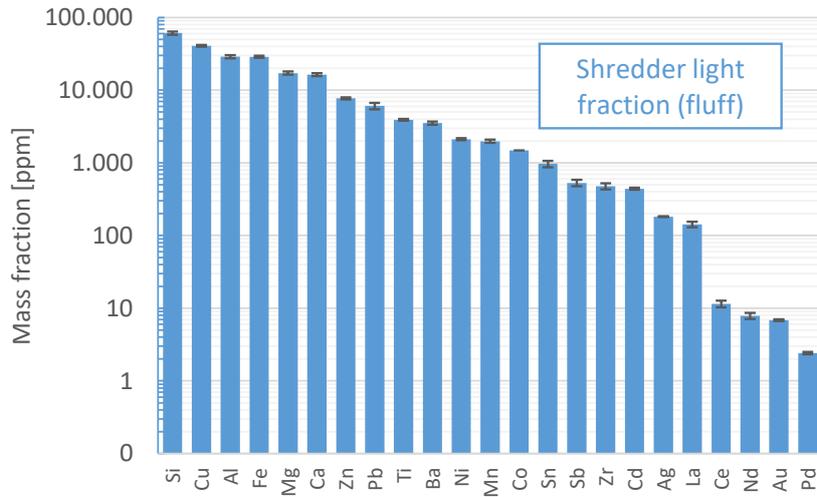


Figure 14: Composition of shredder light fraction (fluff) (AS6)

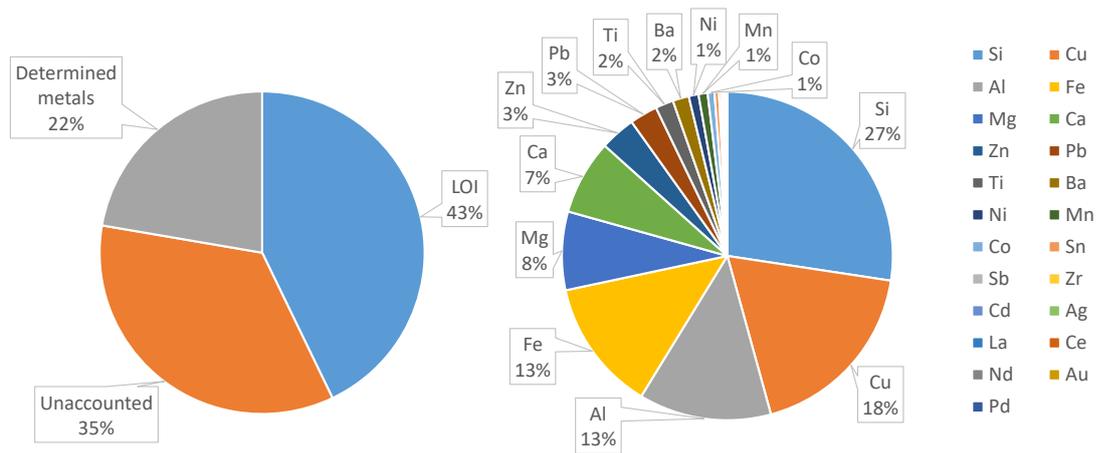


Figure 15: Overview of accounted materials and loss of ignition for shredder light fraction (fluff) (AS6)

8.8. Filter dust (AS7)

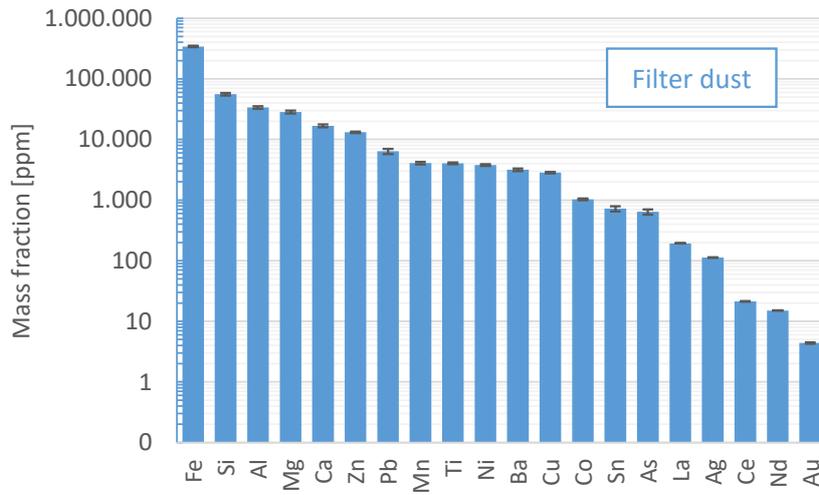


Figure 16: Composition of filter dust (AS7)

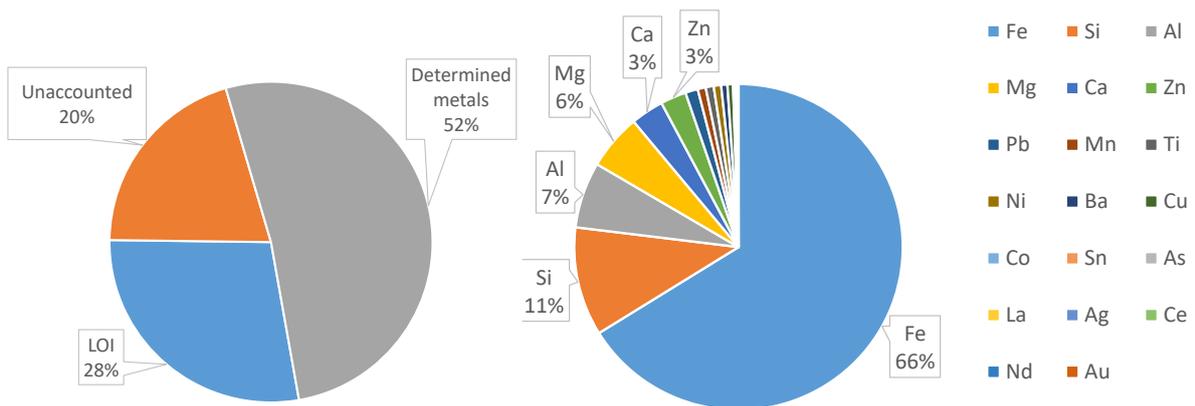


Figure 17: Overview of accounted materials and loss of ignition for filter dust (AS7)

8.9. Sweepings (AS8)

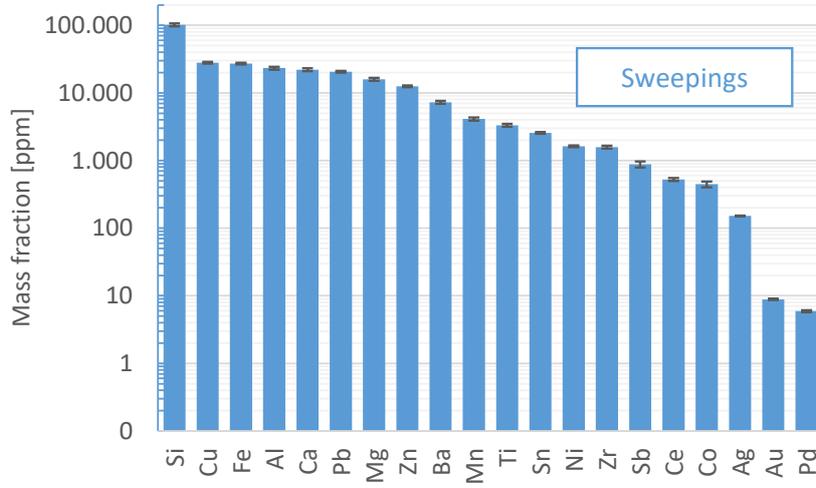


Figure 18: Composition of sweepings (AS8)

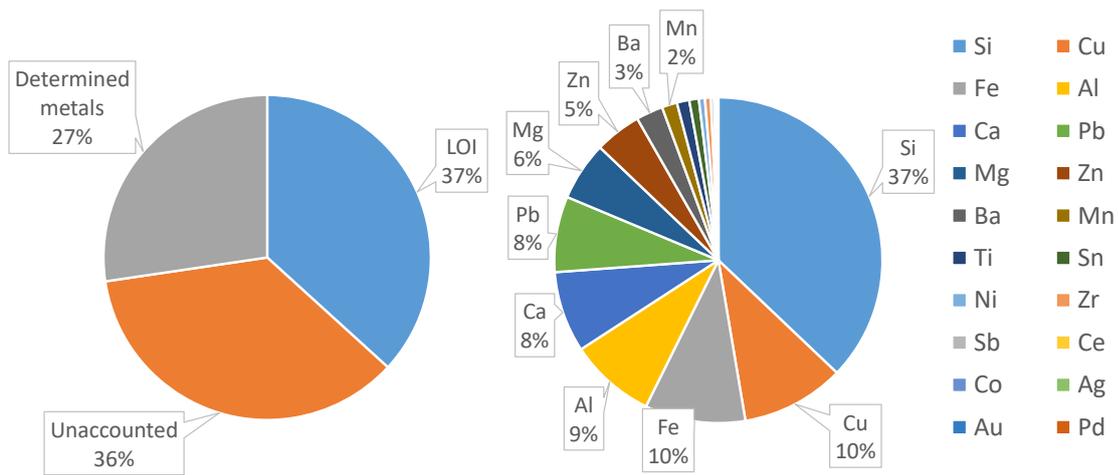


Figure 19: Overview of accounted materials and loss of ignition for sweepings (AS8)

9. S9 Example sampling uncertainty calculation for AS1 in laboratory 1

Table 10: Sampling uncertainty calculation for Au, Ag, Pd, and Cu in PCBs in AS1

Component to be sampled			
Au, Ag, Pd, Cu based on PCB			
Variables	Abbr.	Unit	Value
Density critical component	δ_A	g/cm^3	2.00
Density Rest of lot	δ_G	g/cm^3	6.78
Share component in lot	a_L	-	0.00
Liberation diameter	d_L	cm	0.25
Nominal diameter	d	cm	1.20
Shape factor	t	-	0.20
Size range parameter	g	-	0.35
Constitution parameter	c	g/cm^3	1171.11
Liberation parameter	β	-	0.21
Variance of fundamental Error	S^2_{FE}	-	0.00
Fundamental Error	s	-	0.02
		%	2.00

Table 11: Sampling uncertainty calculation for Cu in cables in AS1

Component to be sampled			
Cu based on cables			
Variables	Abbr.	Unit	Value
Density critical component	δ_A	g/cm^3	3.90
Density Rest of lot	δ_G	g/cm^3	6.78
Share component in lot	a_L	-	0.00
Liberation diameter	d_L	cm	0.05
Nominal diameter	d	cm	1.20
Shape factor	t	-	0.25
Size range parameter	g	-	0.35
Constitution parameter	c	g/cm^3	1519.86
Liberation parameter	β	-	0.04
Variance of fundamental Error	S^2_{FE}	-	0.00
Fundamental Error	s	-	0.01
		%	1.14

Table 12: Sampling uncertainty calculation for Cu in non-ferrous metals in AS1

Material to be sampled			
Cu based on Non-ferrous metals			
Variables	Abbr.	Unit	Value
<i>Density critical component</i>	δ_A	g/cm^3	8.92
<i>Density Rest of lot</i>	δ_G	g/cm^3	6.77
<i>Share component in lot</i>	a_L	-	0.00
<i>Liberation diameter</i>	d_L	<i>cm</i>	0.10
<i>Nominal diameter</i>	d	<i>cm</i>	1.20
<i>Shape factor</i>	t	-	0.15
<i>Size range parameter</i>	g	-	0.35
<i>Constitution parameter</i>	c	g/cm^3	11628.72
<i>Liberation parameter</i>	β	-	0.08
Variance of fundamental Error	S^2_{FE}	-	0.00
Fundamental Error	s	-	0.03
		%	3.45

Table 13: Sampling uncertainty calculation for Fe in AS1

Material to be sampled			
Fe			
Variables	Abbr.	Unit	Value
<i>Density critical component</i>	δ_A	g/cm^3	8
<i>Density Rest of lot</i>	δ_G	g/cm^3	2.36
<i>Share component in lot</i>	a_L	-	0.92
<i>Liberation diameter</i>	d_L	<i>cm</i>	0.30
<i>Nominal diameter</i>	d	<i>cm</i>	1.20
<i>Shape factor</i>	t	-	0.10
<i>Size range parameter</i>	g	-	0.35
<i>Constitution parameter</i>	c	g/cm^3	2.23
<i>Liberation parameter</i>	β	-	0.25
Variance of fundamental Error	S^2_{FE}	-	0.00
Fundamental Error	s	-	0.00
		%	0.07

Table 14: Sampling uncertainty calculation for Al in AS1

Material to be sampled			
Al			
Variables	Abbr.	Unit	Value
Density critical component	δ_A	g/cm^3	2.7
Density Rest of lot	δ_G	g/cm^3	6.78
Share component in lot	a_L	-	0.002
Liberation diameter	d_L	cm	0.40
Nominal diameter	d	cm	1.20
Shape factor	t	-	0.15
Size range parameter	g	-	0.35
Constitution parameter	c	g/cm^3	1344.62
Liberation parameter	β	-	0.33
Variance of fundamental Error	S^2_{FE}	-	0.00
Fundamental Error	s	-	0.02
		%	2.34

Table 15: Sampling uncertainty calculation for Co and REE in batteries in AS1

Component to be sampled			
Co, REE based on batteries			
Variables	Abbr.	Unit	Value
Density critical component	δ_A	g/cm^3	5
Density Rest of lot	δ_G	g/cm^3	6.78
Share component in lot	a_L	-	0.000851
Liberation diameter	d_L	cm	1.20
Nominal diameter	d	cm	1.20
Shape factor	t	-	0.20
Size range parameter	g	-	0.35
Constitution parameter	c	g/cm^3	5865.51
Liberation parameter	β	-	1.00
Variance of fundamental Error	S^2_{FE}	-	0.01
Fundamental Error	s	-	0.10
		%	9.79

Table 16: Sampling uncertainty calculation for all target elements in fine material in AS1

Material mix to be sampled			
General based fine material			
Variables	Abbr.	Unit	Value
<i>Density critical component</i>	δ_A	g/cm^3	6
<i>Density Rest of lot</i>	δ_G	g/cm^3	6.81
<i>Share component in lot</i>	a_L	-	0.037444
<i>Liberation diameter</i>	d_L	cm	0.05
<i>Nominal diameter</i>	d	cm	0.10
<i>Shape factor</i>	t	-	0.50
<i>Size range parameter</i>	g	-	0.35
<i>Constitution parameter</i>	c	g/cm^3	148.72
<i>Liberation parameter</i>	β	-	0.50
Variance of fundamental Error	S^2_{FE}	-	0.00
Fundamental Error	s	-	0.00
		%	0.04

Table 17: Cumulated sampling uncertainty for IBMs, PMs, and S-CRMs in AS1

Element	Cumulated uncertainty	Based on:
<i>Au, Ag, Pd</i>	2.00%	Table 10, Table 16
<i>Cu</i>	4.1%	Table 10, Table 11, Table 12, Table 16
<i>Fe</i>	0.08%	Table 13, Table 16
<i>Al</i>	2.3%	Table 14, Table 16
<i>Co, REE</i>	9.8%	Table 15, Table 16

10. Sieve and sorting analysis of output fractions from automated processing

10.1. S10 Sieve analysis

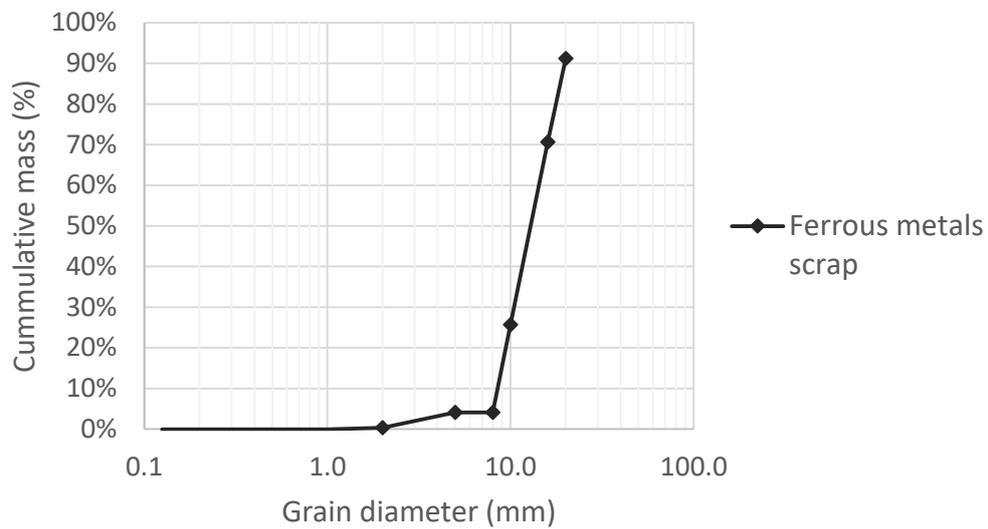


Figure 20: Results of sieve analysis for ferrous metals scrap (AS1)

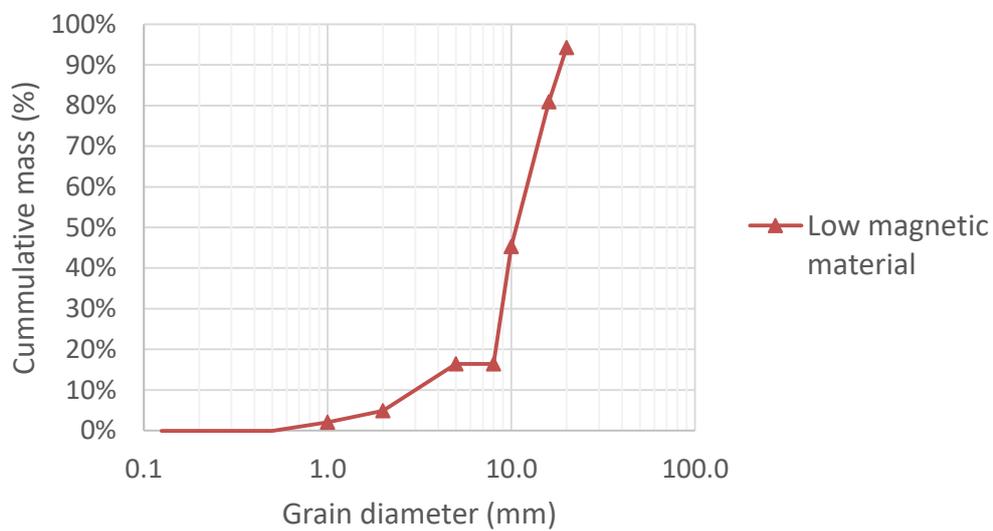


Figure 21: Results of sieve analysis for low magnetic material (AS2)

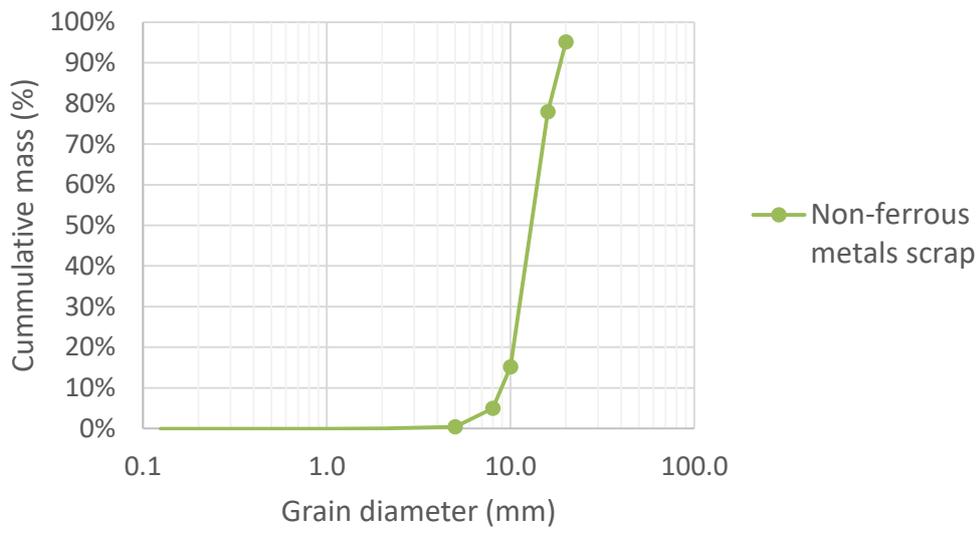


Figure 22: Results of sieve analysis for non-ferrous metals scrap (AS3)

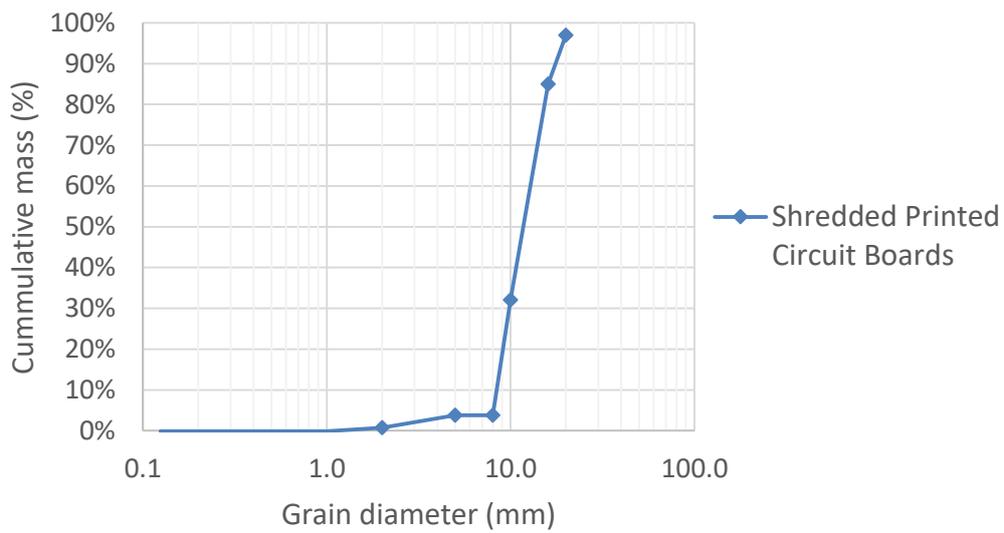


Figure 23: Results of sieve analysis for shredded printed circuit boards (AS4)

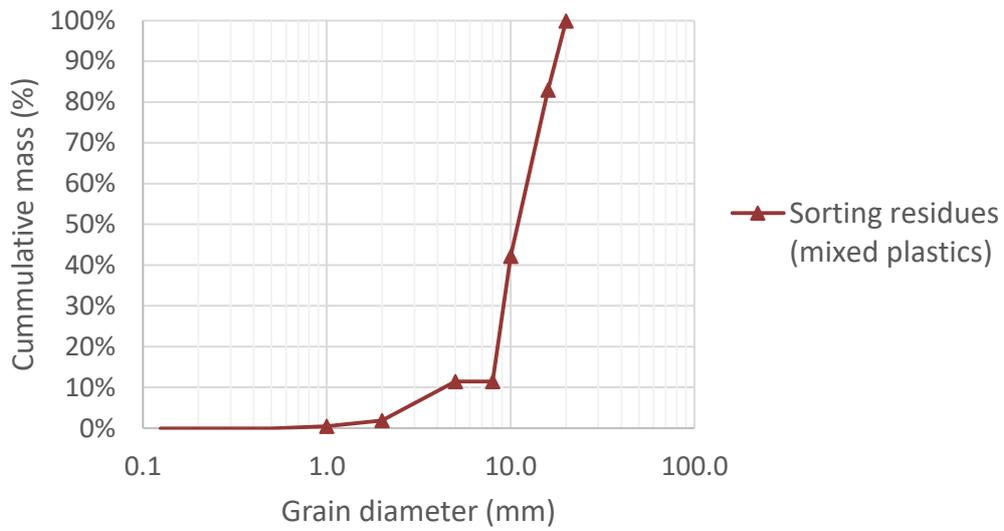


Figure 24: Results of sieve analysis for sorting residues (mixed plastics) (AS5)

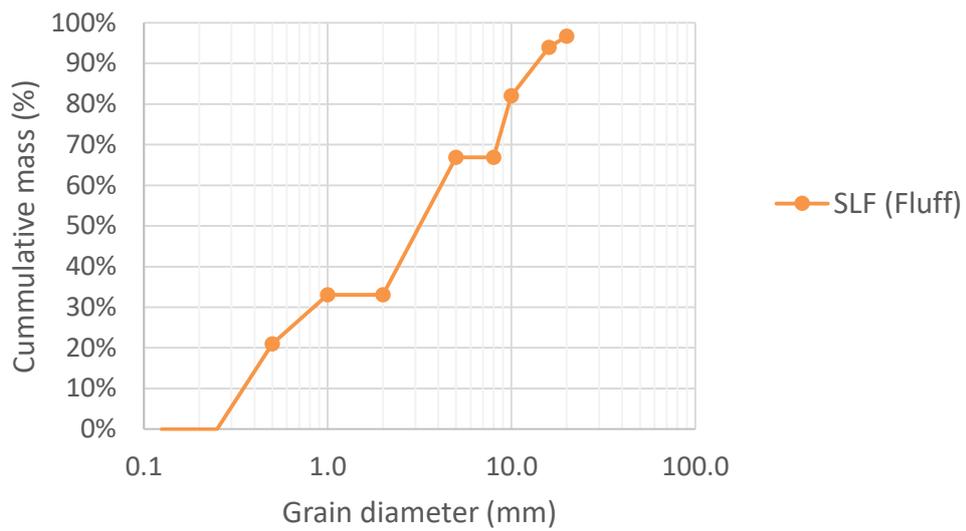


Figure 25: Results of sieve analysis for SLF (Fluff) (AS6)

10.2. S11 Sieve analysis and cumulative masses of metals

10.2.1. Shredded printed circuit boards (AS4)

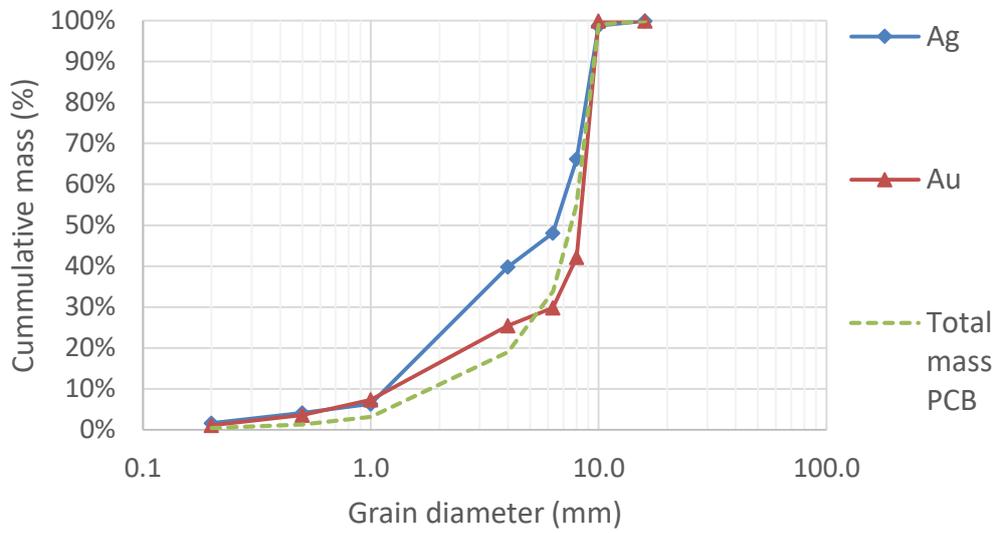


Figure 26: Sieve analysis of shredded PCB (AS4) and cumulative masses of Ag and Au in screening steps

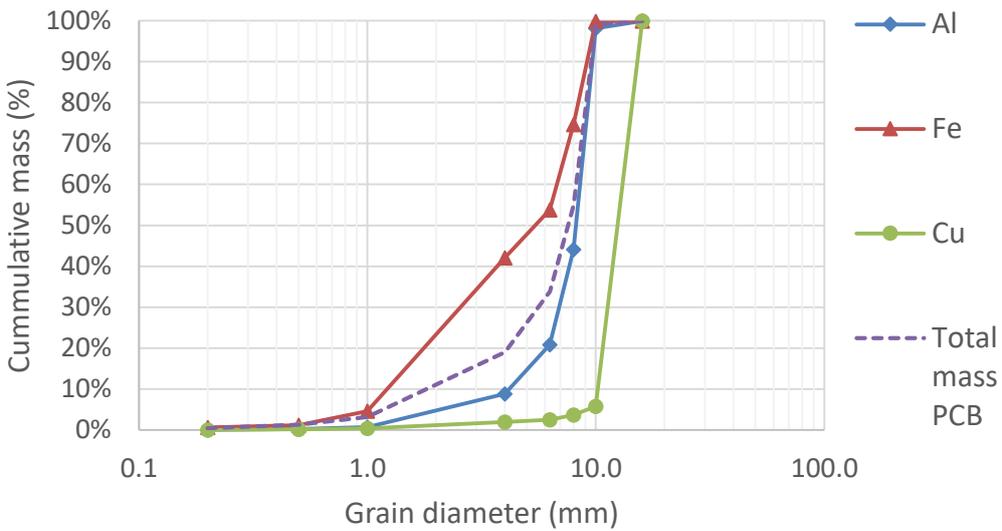


Figure 27: Sieve analysis of shredded PCB (AS4) and cumulative masses of Al, Fe, and Cu in screening steps

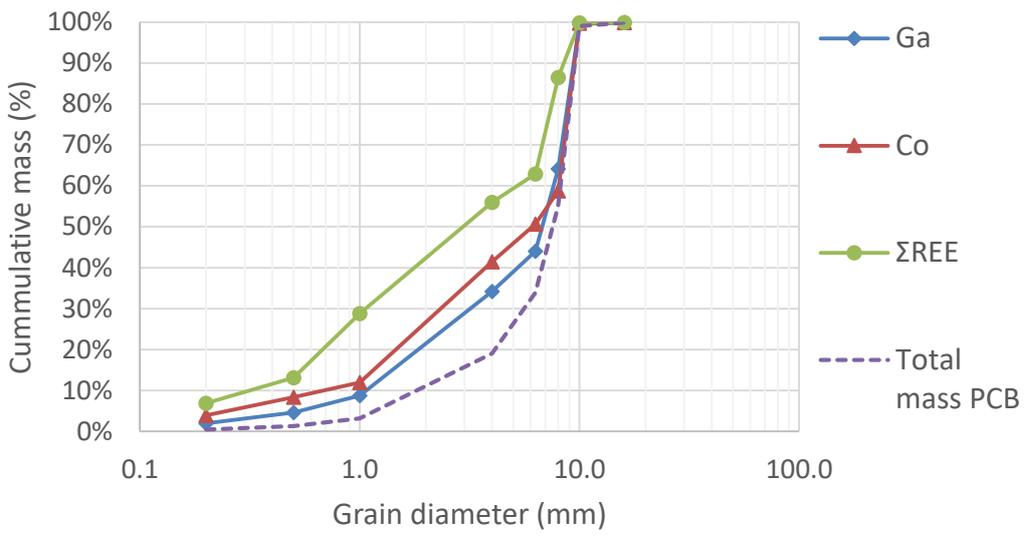
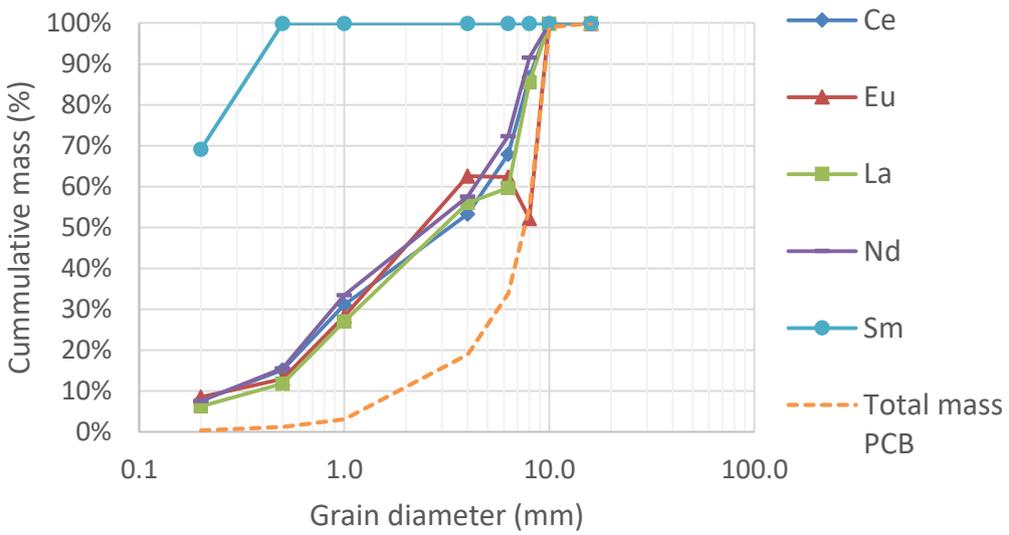


Figure 28: Sieve analysis of shredded PCB (AS4) and cumulative masses of Ga, Co and ΣREE in screening steps



Sieve analysis of shredded PCB (AS4) and cumulative masses of the rare earth elements Ce, Eu, La, Nd and Sm in screening steps

10.2.2. Sorting residues (mixed plastics) (AS5)

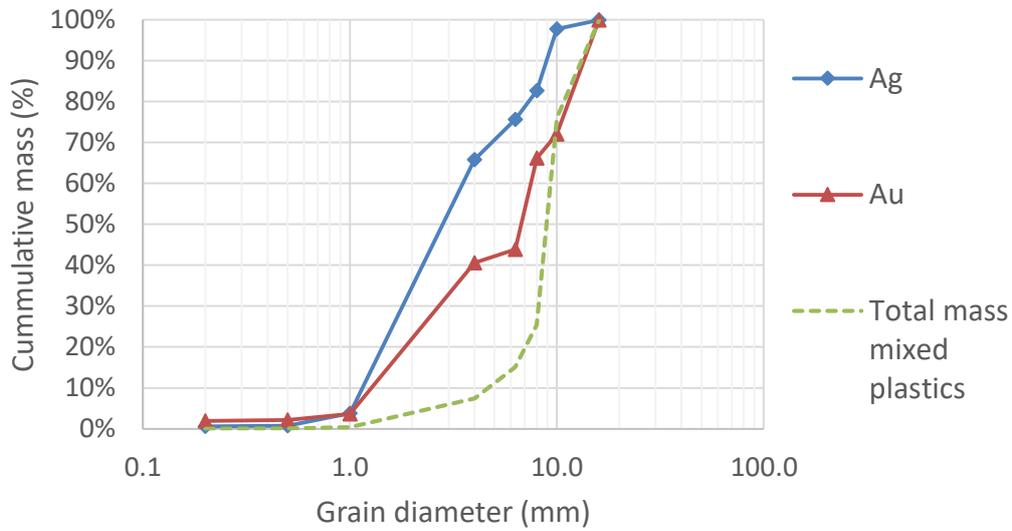


Figure 29: Sieve analysis of sorting residues (mixed plastics) (AS5) and cumulative masses of Ag and Au in screening steps

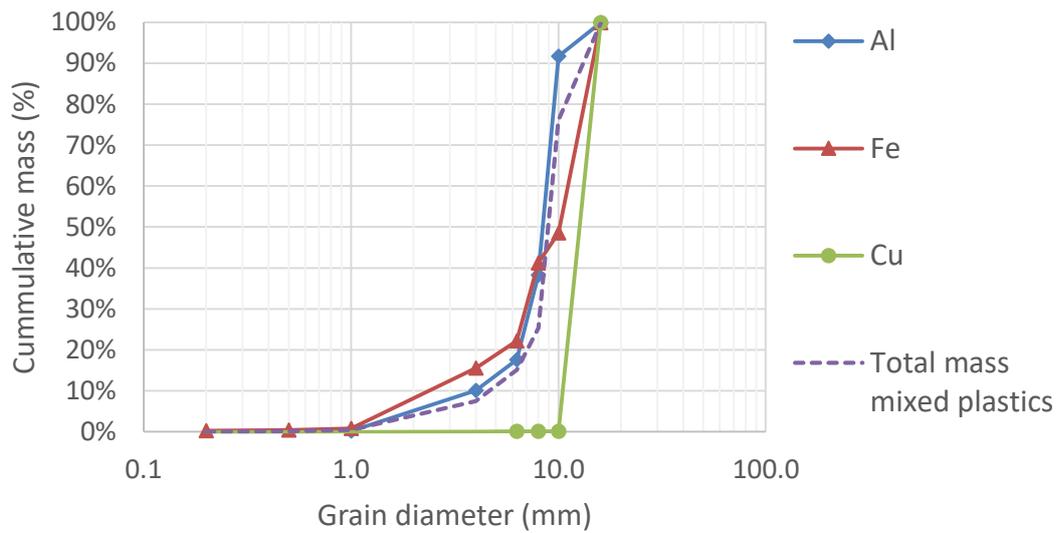


Figure 30: Sieve analysis of sorting residues (mixed plastics) (AS5) and cumulative masses of Al, Fe, and Cu in screening steps

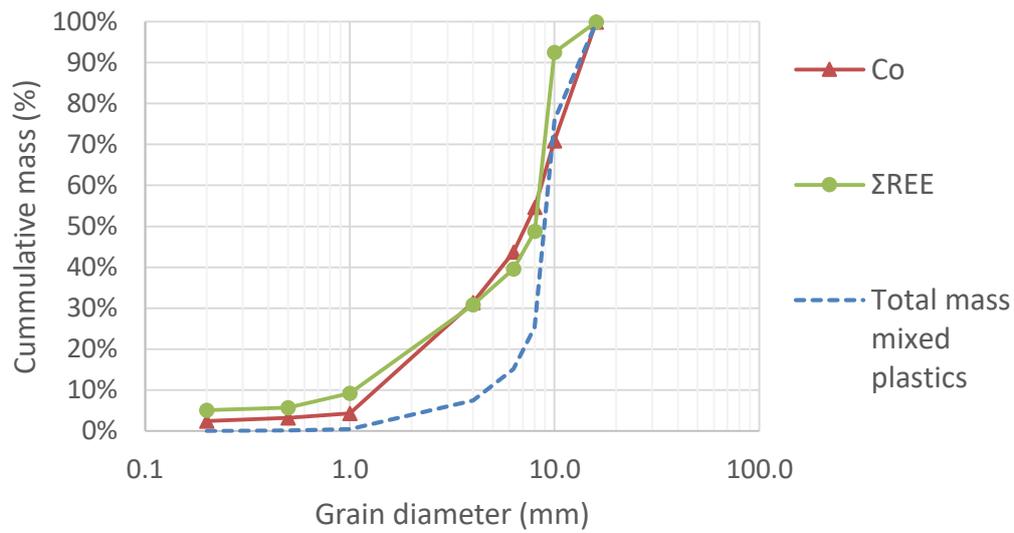


Figure 31: Sieve analysis of sorting residues (mixed plastics) (AS5) and cumulative masses of Co and Σ REE in screening steps

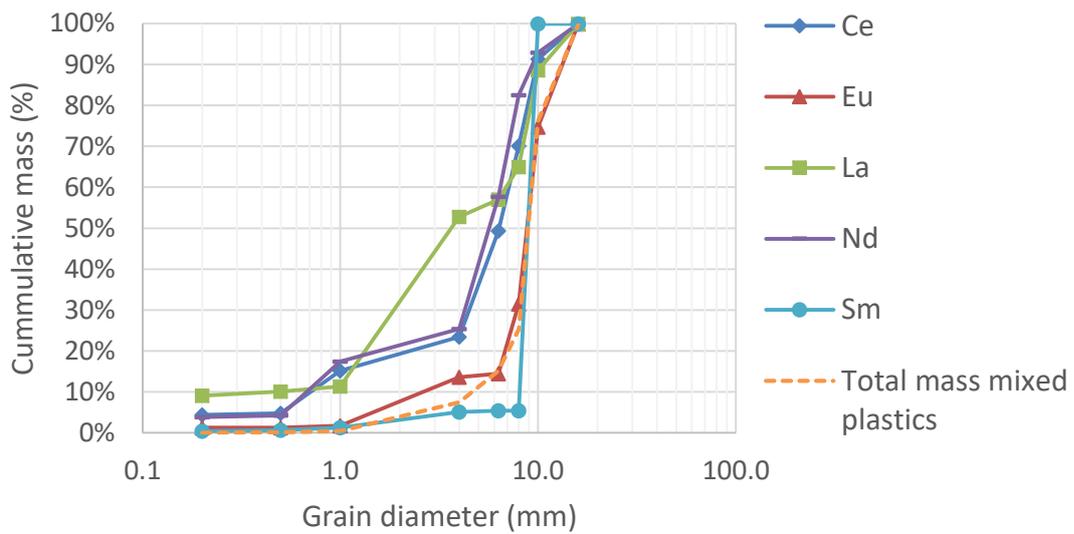


Figure 32: Sieve analysis of sorting residues (mixed plastics) (AS5) and cumulative masses of the rare earth elements Ce, Eu, La, Nd and Sm in screening steps

10.2.3. Shredder light fraction (fluff) (AS6)

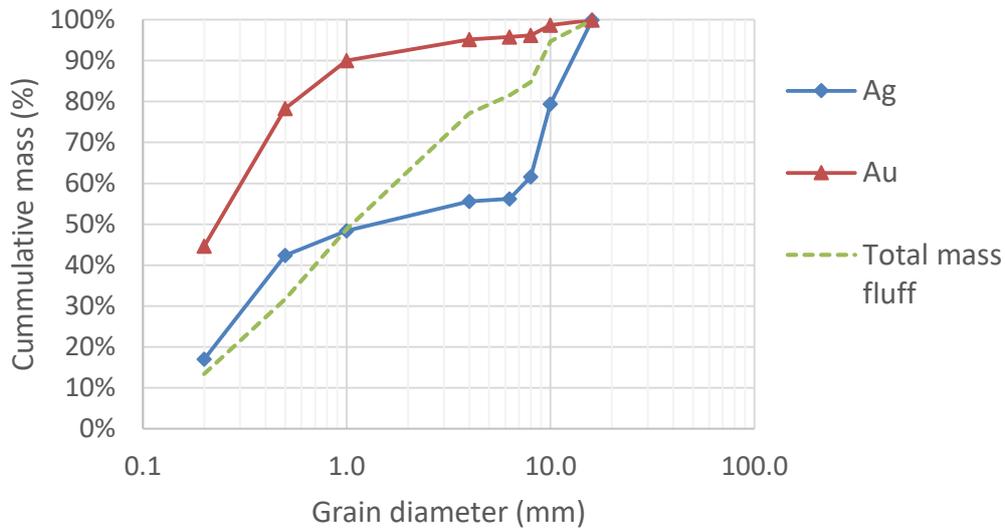


Figure 33: Sieve analysis of shredder light fraction (fluff) (AS6) and cumulative masses of Ag and Au in screening steps

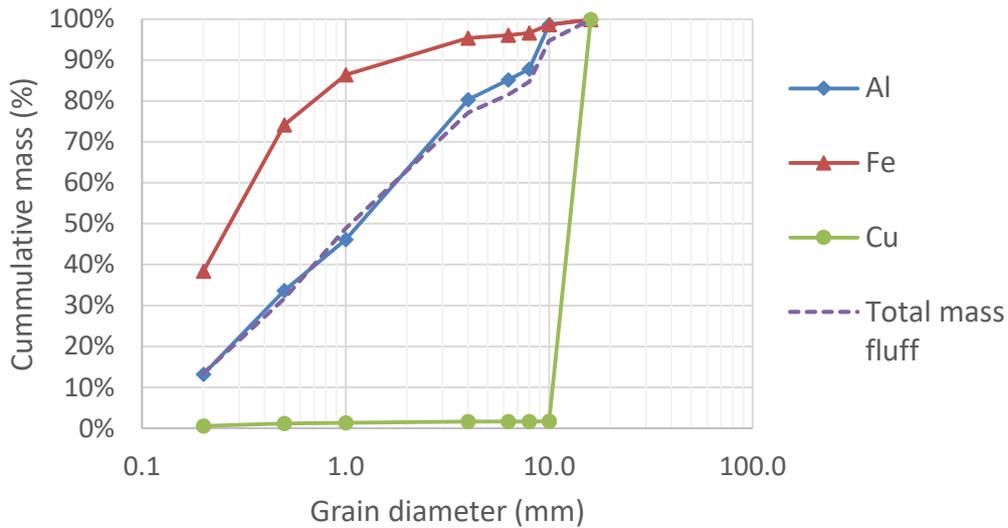


Figure 34: Sieve analysis of shredder light fraction (fluff) (AS6) and cumulative masses of Al, Fe, and Cu in screening steps

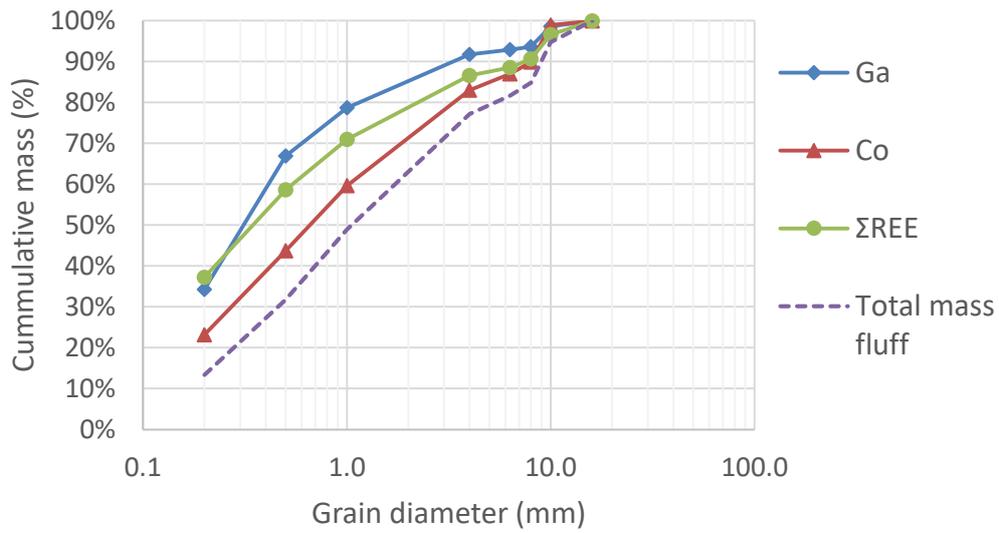


Figure 35: Sieve analysis of shredder light fraction (fluff) (AS6) and cumulative masses of Ga, Co and ΣREE in screening steps

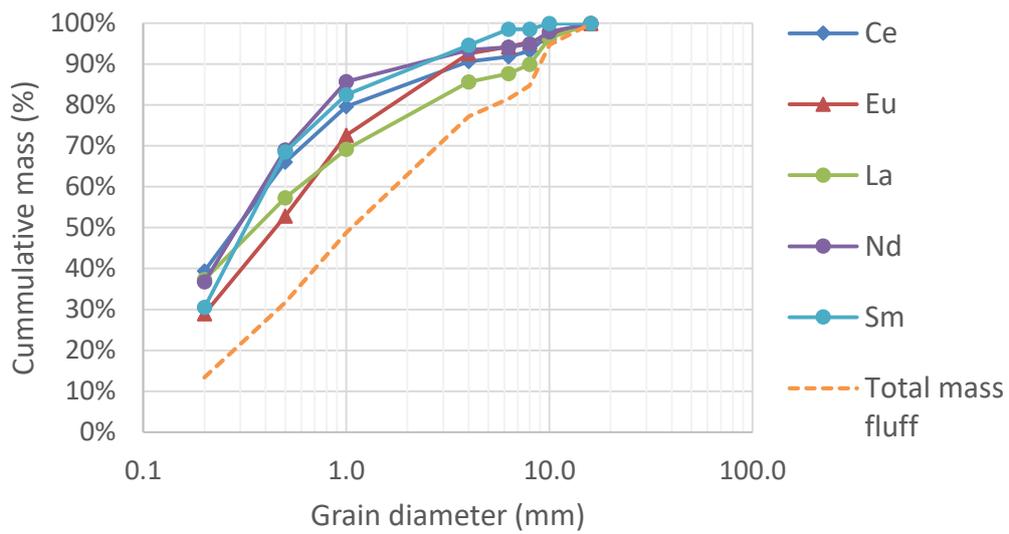


Figure 36: Sieve analysis of shredder light fraction (fluff) (AS6) and cumulative masses of the rare earth elements Ce, Eu, La, Nd and Sm in screening steps

10.2.4. Filter dust (AS7)

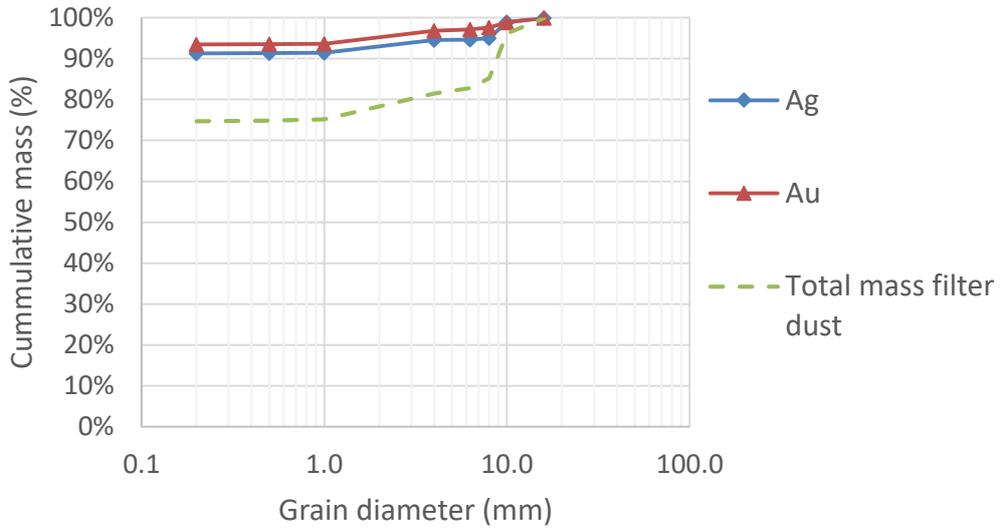


Figure 37: Sieve analysis of filter dust (AS7) and cumulative masses of Ag and Au in screening steps

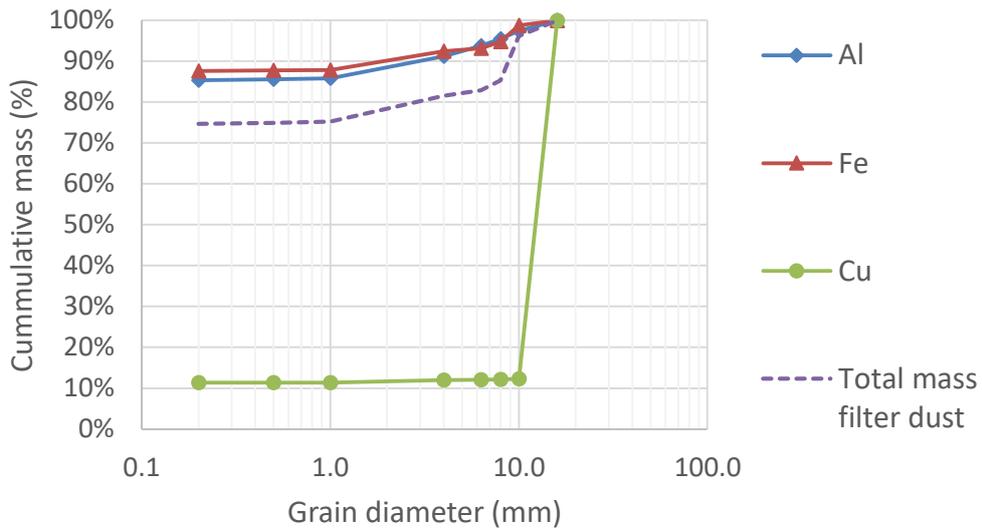


Figure 38: Sieve analysis of filter dust (AS7) and cumulative masses of Al, Fe, and Cu in screening steps

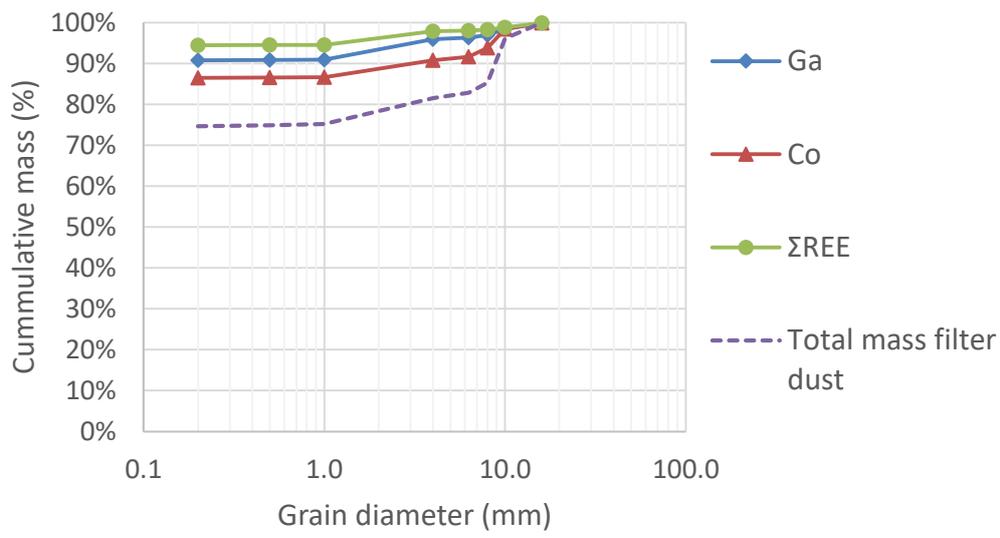


Figure 39: Sieve analysis of filter dust (AS7) and cumulative masses of Ga, Co and ΣREE in screening steps

11. S12 Transfer coefficients

Table 18: Transfer coefficients for investigated elements in manually sorted output fractions

Output fractions manual sorting		Mass [kg]	Industrial base metals			Precious metals			Lower value critical metals				
			Fe	Al	Cu	Ag	Au	Pd	Co	Ga	In	Ta	ΣREE
Manual sorting	IT-Devices	4,921	10±4 %	17±12 %	12±7 %	9±6 %	24±9 %	36±24 %	12±27 %	8±10 %	100±60 %	56±56%	7±2 %
	Cu-rich material	4,916	15±2 %	6±5 %	45±8 %	0.7±0.8 %	4±4 %	0.0%	0.04±0.06 %	0.0%	0.0%	0.0%	0.01±0.01 %
	Metal-poor material	2,078	2±1 %	2±1 %	3±2 %	0.8±0.5 %	3±2 %	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
	Tools scrap	1,043	5±0.5 %	1±0.6 %	3±0.4 %	0.1±0.1 %	0.5±2 %	0.1±0.1 %	0.4±0.1 %	0.0%	0.0%	0.0%	1±0.2 %
	Ferrous rich metals	618	3±1 %	0.1±0.1 %	0.2±0.6 %	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
	Printed Circuit Boards	221	0.1±0.01 %	1±0.1 %	2±0.2 %	5±0.4 %	10±1 %	8±2 %	0.0%	0.0%	0.0%	44±22 %	0.0%
	Batteries	203	0.3±0.1 %	0.1±0.1 %	0.6±0.1 %	0.0%	0.0%	0.0%	33±14 %	0.0%	0.0%	0.0%	30±13 %
	Cooling and air conditioning appliances	111	0.4±0.1 %	1±0.5 %	0.6±0.4 %	0.02±0.001 %	0.02±0.002 %	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
	Hazardous material	40	0.1±0.03 %	0.5±0.3 %	0.0%	2±1 %	0.0%	0.0%	0.001±0.0002 %	0.0%	0.0%	0.0%	0.0%
Total manual sorting	14,151	35±5 %	28±13 %	66±2 %	18±6 %	41±10 %	44±24 %	46±30%	8±10 %	100±60 %	99.5±61 %	38±13 %	

Background information on chemical composition based on "S7 Data on composition of output materials from manual sorting", "S8 Chemical analysis of output fractions from automated processing", and (Anindya et al., 2014; Arshadi and Mousavi, 2015; Buchert et al., 2012a, 2012b; Chancerel, 2010; Chancerel et al., 2010, 2009; Chancerel and Rotter, 2009; Christian et al., 2014; Cui and Forssberg, 2007; Cui and Zhang, 2008; Dervišević et al., 2013; Fitzpatrick et al., 2015; Hester and Harrison, 2008; Huisman, 2004; Juchneski et al., 2013; Kim et al., 2013; Laboratory and Hazardous Material Laboratory, 2004; Lim and Schoenung, 2010; Maragos et al., 2013; Matsuto et al., 2004; Milovantseva and Saphores, 2013; Oguchi et al., 2011; Savvilitidou et al., 2014; Schischke et al., 2013; Sommer et al., 2015; Song et al., 2013; Townsend et al., 2004; Tuncuk et al., 2012; Ueberschaar et al., 2017; Unep, 2009; United Nations University, 2007; Valero Navazo et al., 2014; Wu et al., 2008; Xiu et al., 2015), and own studies.

Table 19: Transfer coefficients for investigated elements in mechanically processed output fractions

Output fraction		Mass [kg]	Industrial base metals			Precious metals			Lower value critical metals				
			Fe	Al	Cu	Ag	Au	Pd	Co	Ga	In	Ta	ΣREE
Mechanical processing	Ferrous metals scrap	9,789	62±4 %	0.02±0.01 %	2±0.3 %	12±5 %	0.4±0.04 %	0.0%	33±10 %	0.0%	0.0%	0.0%	59±20 %
	Low magnetic material	709	2±0.1 %	3±0.5 %	2±0.2 %	19±2 %	11±1 %	7±2 %	0.0%	92±16 %	0.0%	0.0%	0.0%
	Non-ferrous metals scrap	1,105	0.1±0.01 %	47±7 %	2±0.2 %	4±0.3 %	9±1 %	5±1 %	0.0%	0.0%	0.0%	0.0%	0.0%
	Sorting residues (mixed plastics)	625	0.3±0.02 %	15±2 %	20±2 %	29±2 %	28±3 %	32±7 %	0.0%	0.0%	0.0%	0.0%	0.5±0.1 %
	Shredded printed circuit boards	1,691	0.1±0.01 %	3±0.5 %	5±1 %	9±1 %	5±1 %	5±1 %	0.6±0.2 %	0.0%	0.0%	0.1±0.1 %	0.1±0.02 %
	SLF (Fluff)	96	0.3±0.02 %	3±0.5 %	3±0.3 %	9±1 %	5±0.5%	6±1 %	20±6 %	0.0%	0.0%	0.4±0.2 %	2±0.4 %
	Filter dust	12,498	0.2±0.01 %	0.2±0.03 %	0.01±0.001 %	0.3±0.03 %	0.2±0.02 %	0.0%	0.8±0.2 %	0.0%	0.0%	0.0%	0.1±0.03%
	Losses / Sweepings	108	0.02±0.001 %	0.2±0.02 %	0.1±0.01 %	0.5±0.04 %	0.4±0.04 %	1±0.2 %	0.4±0.1 %	0.0%	0.0%	0.0%	0.4±0.1 %
Total mechanical processing	26,621	65±4 %	72±7 %	34±2 %	82±5 %	59±3 %	56±8 %	54±11 %	92±16 %	0.0%	0.5±0.2 %	62±20 %	

Background information on chemical composition based on "S8 Chemical analysis of output fractions from automated processing"

12. S13 Comparison of sorting analysis vs. chemical analysis

12.1. Estimation of contents of manually sorted materials

Table 20: Estimated contents for manually sorted materials

Values in %	Fe	Al	Cu	Au	Ag	Pd	Ta	Co	ΣREE	Ga	Based on
Ferromagnetic material	9.5E +01							1.0E -03			Estimation
Aluminium		9.5E +01								1.0E -02	Estimation
Other non-ferrous metals			6.0E +01								Estimation
Plastics colored											Estimation
Plastics black											Estimation
Printed circuit boards	6.9E +00	1.8E +01	4.5E +01	4.8E -03	1.1E -01	1.2E -03	1.2E -04				Chemical analysis results of this study
Batteries	2.2E +01	9.2E -01	6.9E +00					2.1E +00	2.2E +00		(Sommer et al., 2015)
Organic											Estimation
Cable			3.5E +01								Estimation
Foam, Gum, Foils											Estimation
Glass, ceramics											Estimation
Rest											Estimation

12.2. Comparison of sorting analysis vs. chemical analysis per metal

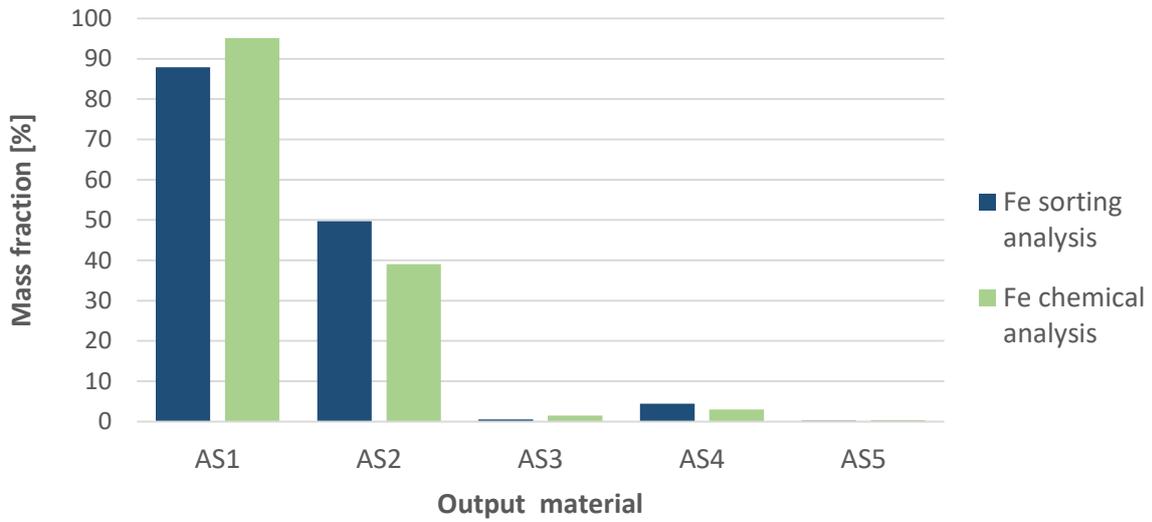


Figure 40: Comparison of sorting analysis vs. chemical analysis for Fe

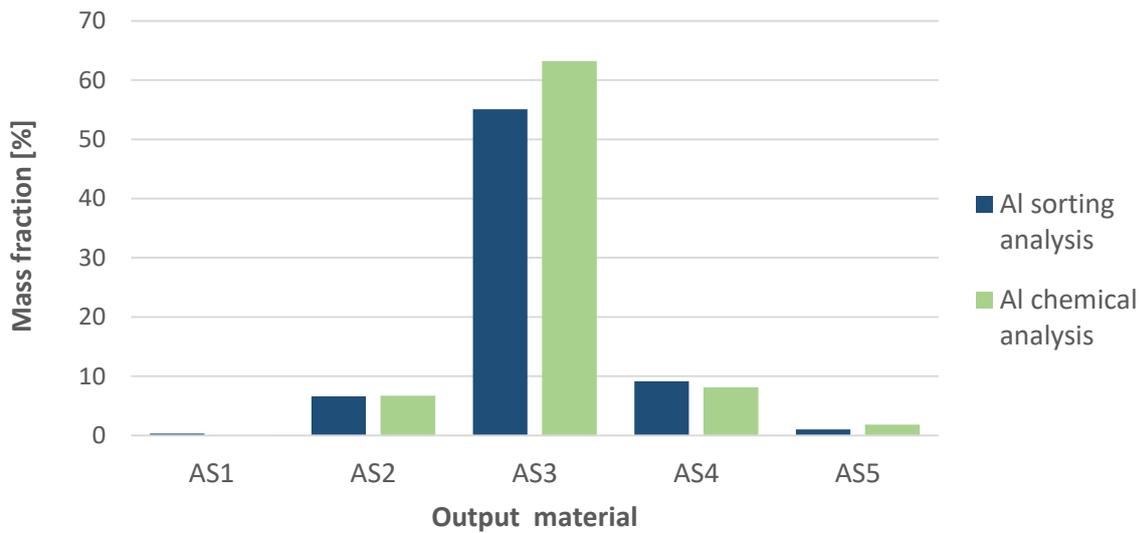


Figure 41: Comparison of sorting analysis vs. chemical analysis for Al

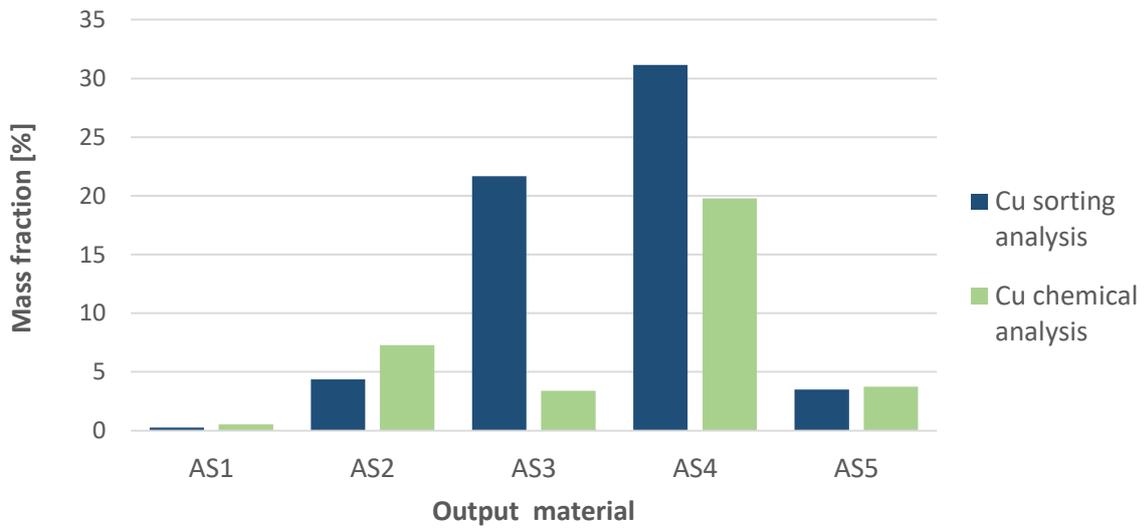


Figure 42: Comparison of sorting analysis vs. chemical analysis for Cu

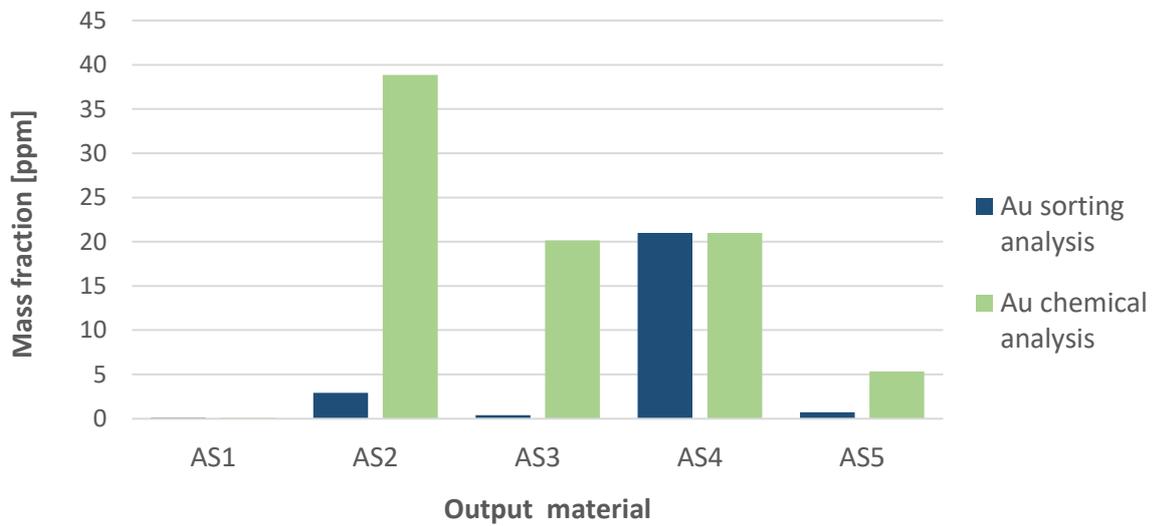


Figure 43: Comparison of sorting analysis vs. chemical analysis for Au

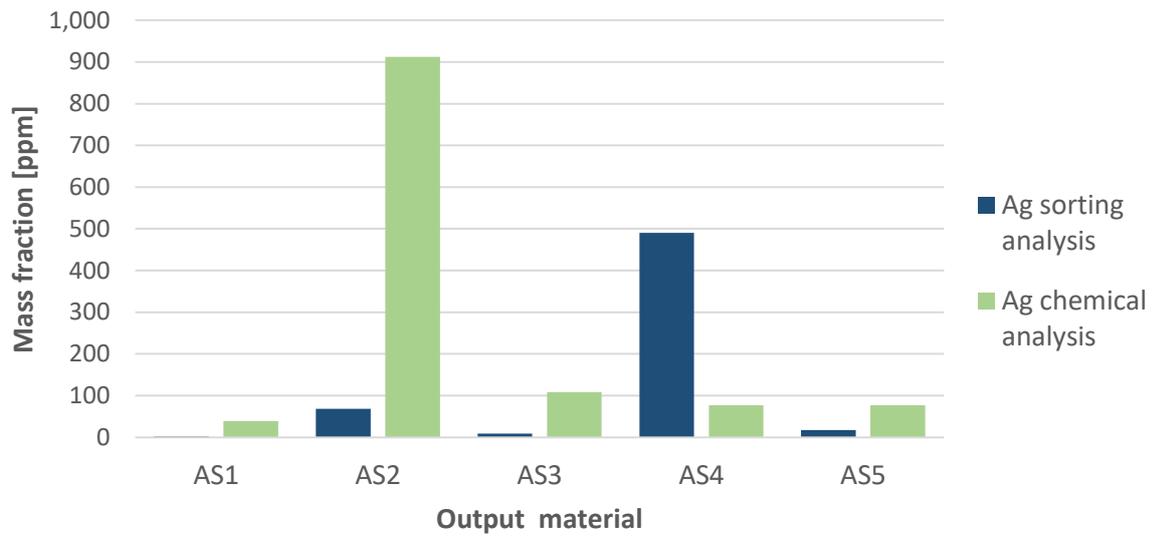


Figure 44: Comparison of sorting analysis vs. chemical analysis for Ag

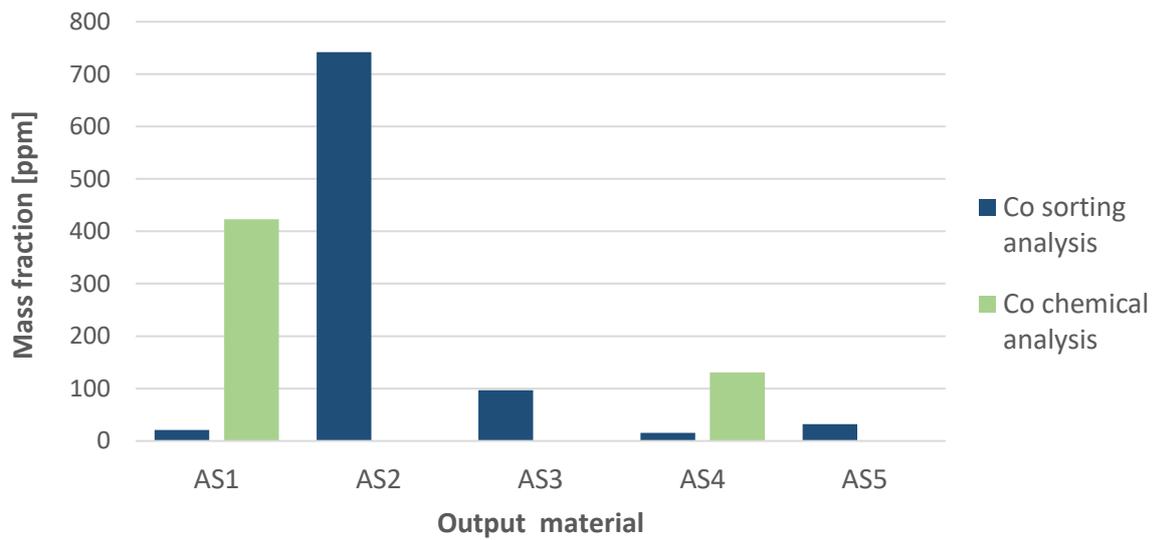


Figure 45: Comparison of sorting analysis vs. chemical analysis for Co

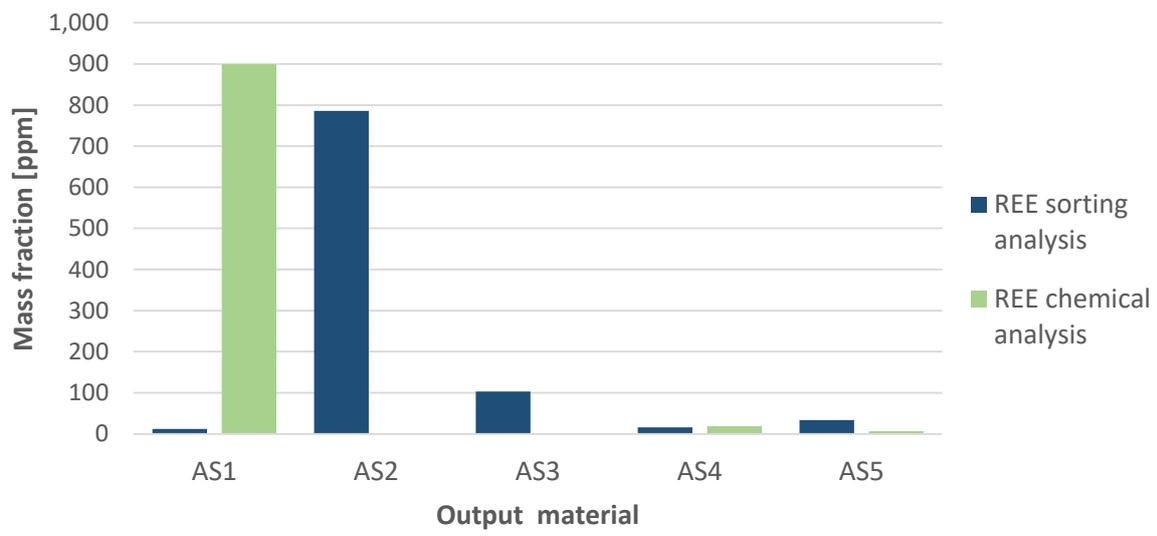


Figure 46: Comparison of sorting analysis vs. chemical analysis for Σ REE

13. S14 Identification of key output fractions for the recovery of target metals

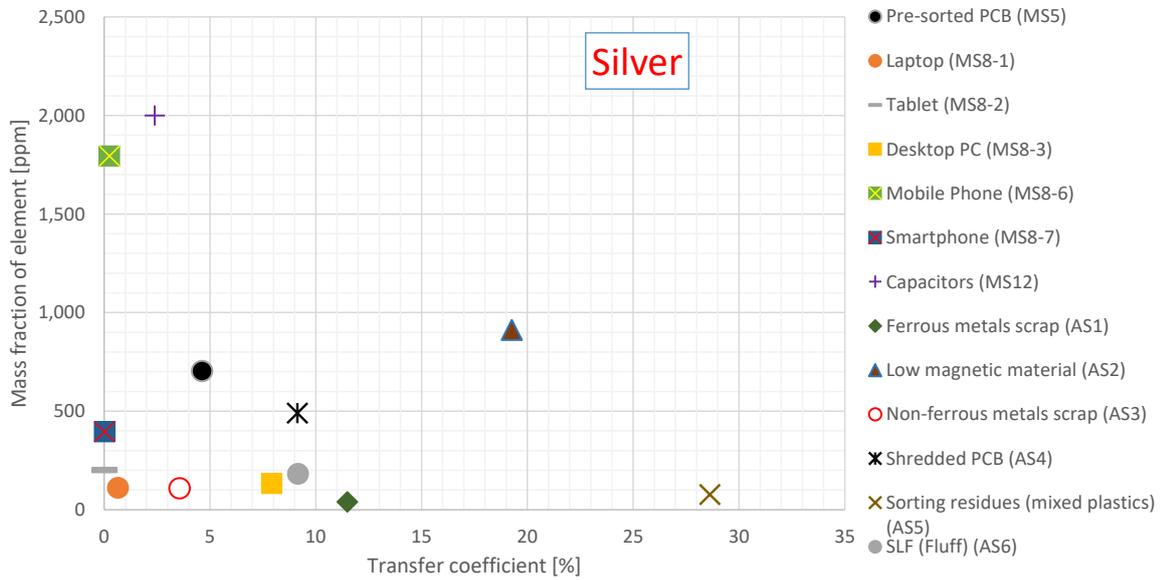


Figure 47: Hotspot plot for silver in the output fractions with the highest mass fractions

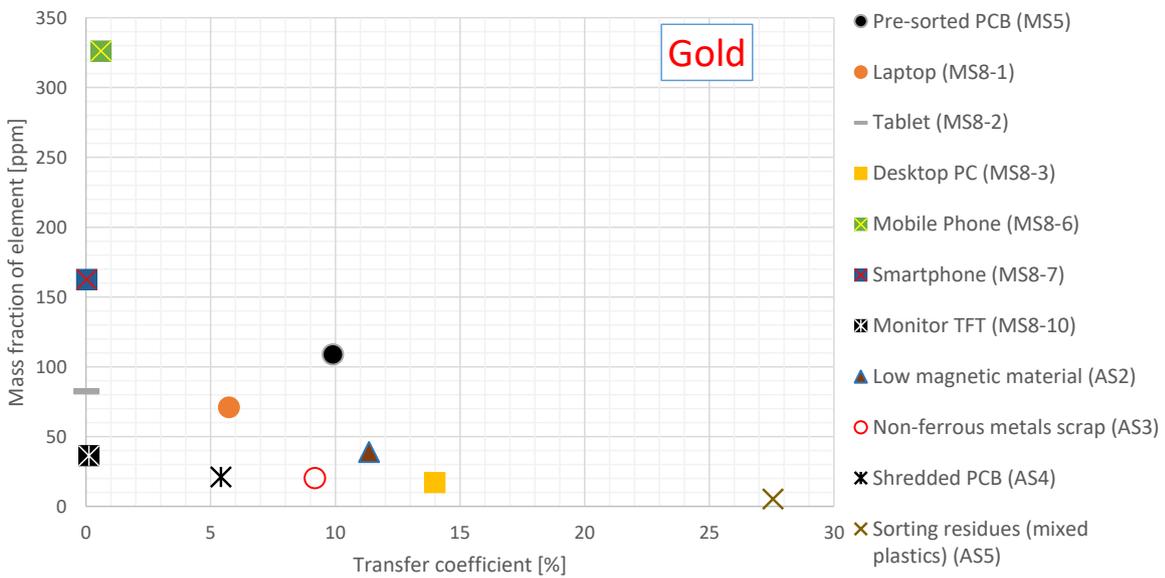


Figure 48: Hotspot plot for gold in the output fractions with the highest mass fractions

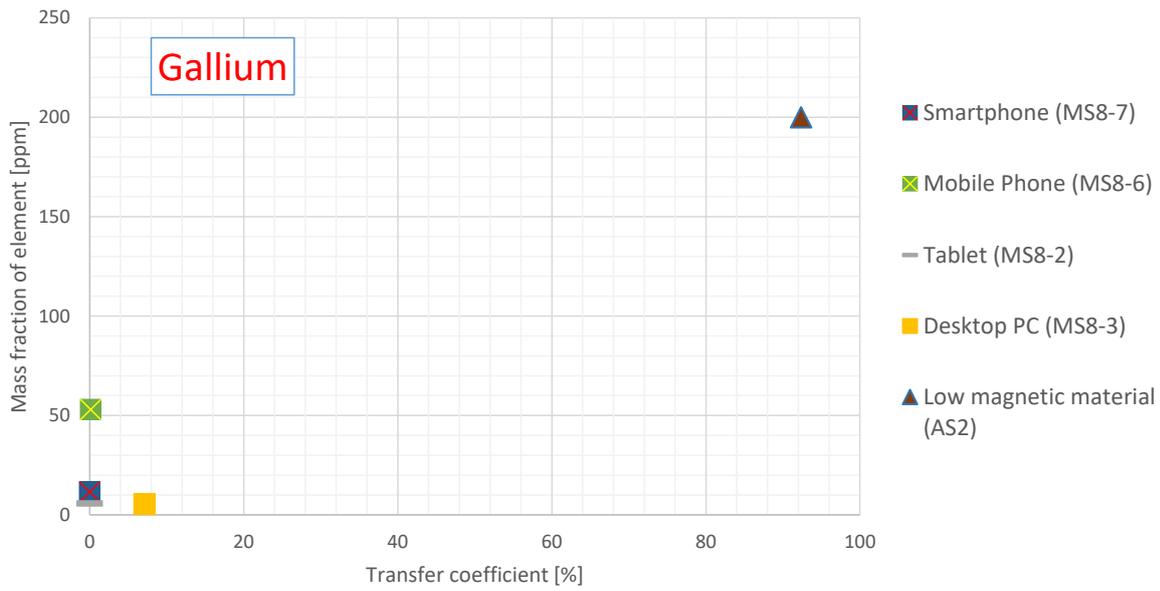


Figure 49: Hotspot plot for gallium in the output fractions with the highest mass fractions

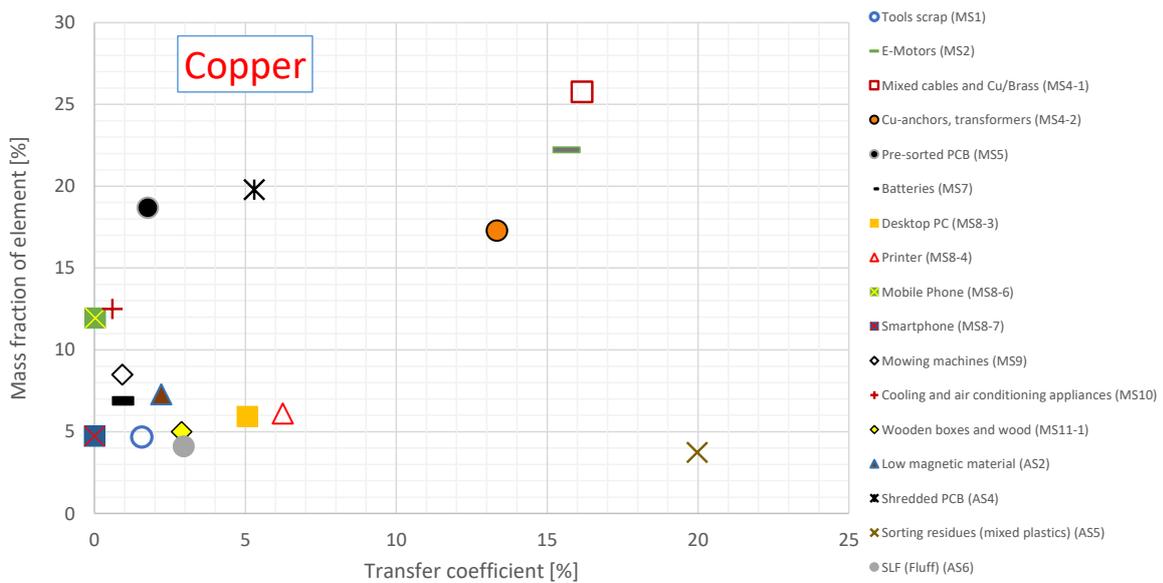


Figure 50: Hotspot plot for copper in the output fractions with the highest mass fractions

14. S15 Recyclability assessment

Based on the techniques of processing WEEE, metals are dispersed to various output fractions. Here, a big number of different elements with varying mass fractions can be present. According to the target-materials, output fractions are guided to the various actors in the recycling chain to recover valuable materials as well as to release 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 this batch test and external research about mechanical and pyro- or hydrometallurgical processes, an estimation of the recyclability of target metals in this study will be carried out. For this purpose, the particular routes will be traced back to question subsequent processes.

Intending to investigating the recyclability of the target materials, the dispersion into other materials has to be examined. Based on the most valuable substance or substance mix, the material is usually forwarded to an appropriate recycler. Those subsequent recovery processes may enable the enrichment of critical metals or other trace metals. However, this can be still connected to a high effort. Pyrometallurgy usually constitutes the next step after the pre-treatment of WEEE for metals. Here, the recovery of critical metals is mostly not even possible.

The further paths of the output materials has been investigated in order to reveal opportunities for the recycling of critical metals. Figure 51 shows a simplified recycling chain and the routes of the material from WEEE to a secondary raw material.

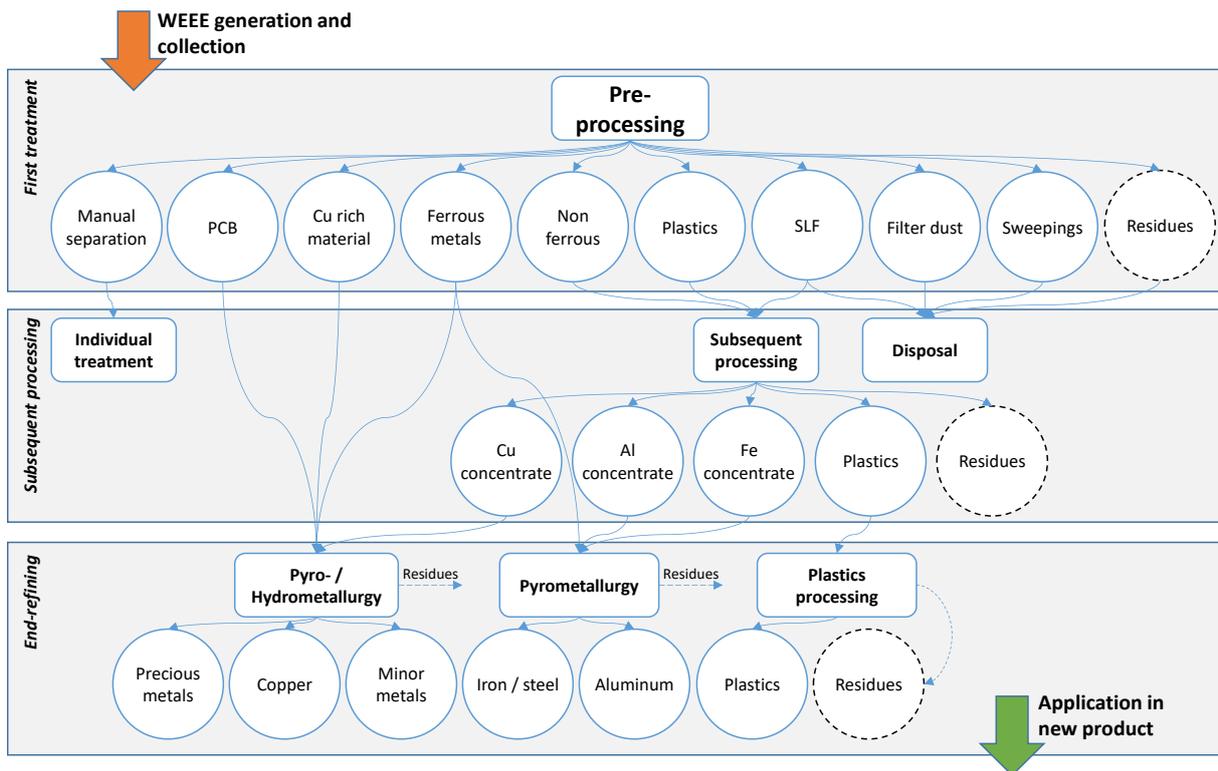


Figure 51: Simplified recycling chain based on the investigated recycling process

Table 21 shows the most common state-of-the-art end refining processes, which are used for the further concentration and purification of target metals. The used methodologies were assessed from a technical and economic perspective, whether subsequent processes allow a recovery of the investigated target elements cobalt, gallium, indium, tantalum and rare earth elements.

Table 21: 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
<i>Converter of steel making</i>	Fe plus alloying materials	Co, Ga, In, REE, Ta		x
<i>Copper smelter as de-oxidant with subsequent precious metals refining</i>	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
<i>Aluminum refining / re-melting</i>	Aluminum and alloying materials	Co, Ga, In, REE, Ta		x
<i>Copper smelter with subsequent precious metals refining</i>	Cu, Au, Ag, Pd, Pt plus alloying materials	Co, Ga, In, REE, Ta		x
<i>Integrated smelter (Cu, Pb smelter, and subsequent refining)</i>	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	
<i>Copper smelter with subsequent precious metals refining</i>	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
		Ta		x
		In		x
		REE		x
		Ga		x
<i>Integrated smelter (Cu, Pb smelter, and subsequent refining)</i>	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	
<i>Copper smelter with subsequent precious metals refining</i>	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
<i>Converter of steel making</i>	Fe plus alloying materials	Co, Ga, In, REE, Ta		x
<i>Plastics recovery</i>	Various types of plastics	Co, Ga, In, REE, Ta		x
<i>Copper smelter with subsequent precious metals refining</i>	Cu, Au, Ag, Pd, Pt	Co, Ga, In, REE, Ta		x
<i>Converter of steel making</i>	Fe plus alloying materials	Co, Ga, In, REE, Ta		x
<i>Aluminum refining / re-melting</i>	Aluminum and alloying materials	Co, Ga, In, REE, Ta		x
<i>Discarded</i>	none	Co, Ga, In, REE, Ta		x

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

The indicated conflict materials represent mostly valuable elements, which usually define the used metallurgical route and can be connected 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.

Figure 52 summarizes up the recyclability of the investigated base, precious and critical metals entered the output fractions from the mechanical treatment of WEEE.

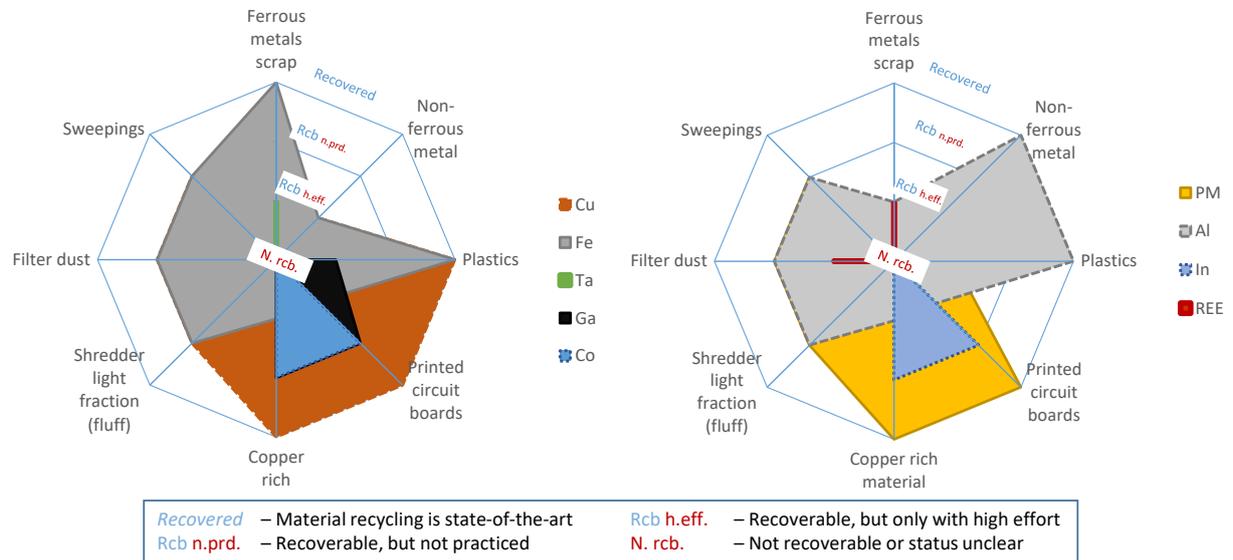


Figure 52: Recyclability of base, precious and critical metals in the output fractions of the automated sorting process

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

15. S16 Example of variation of grain sizes and material composition during batch test

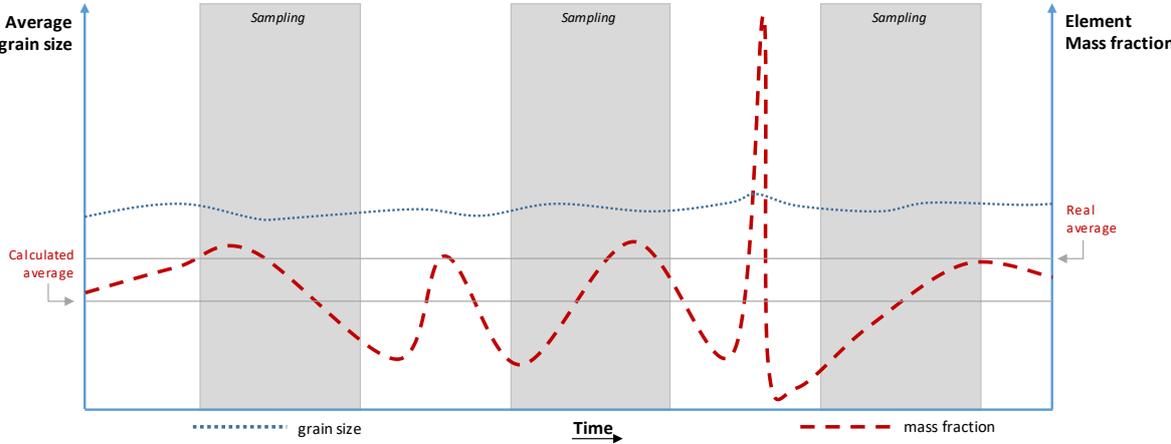


Figure 53: Example of variation of grain sizes and material composition during batch trial

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