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Resource recovery from residual household waste: An application of exergy flow analysis and exergetic life cycle assessment

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ABSTRACT

Exergy is based on the Second Law of thermodynamics and can be used to express physical and chemical potential and provides a unified measure for resource accounting. In this study, exergy analysis was applied to four residual household waste management scenarios with focus on the achieved resource recovery efficiencies. The calculated exergy efficiencies were used to compare the scenarios and to evaluate the applicability of exergy-based measures for expressing resource quality and for optimizing resource recovery. Exergy efficiencies were determined based on two approaches: (i) exergy flow analysis of the waste treatment system under investigation and (ii) exergetic life cycle assessment (LCA) using the Cumulative Exergy Extraction from the Natural Environment (CEENE) as a method for resource accounting. Scenario efficiencies of around 17-27% were found based on the exergy flow analysis (higher efficiencies were associated with high levels of material recycling), while the scenario efficiencies based on the exergetic LCA lay in a narrow range around 14%. Metal recovery was beneficial in both types of analyses, but had more influence on the overall efficiency in the exergetic LCA approach, as avoided burdens associated with primary metal production were much more important than the exergy content of the recovered metals. On the other hand, plastic recovery was highly beneficial in the exergy flow analysis, but rather insignificant in exergetic LCA. The two approaches thereby offered different quantitative results as well as conclusions regarding material recovery. With respect to resource quality, the main challenge for the exergy flow analysis is the use of exergy content and exergy losses as a proxy for resource quality and resource losses, as exergy content is not per se correlated with the functionality of a material. In addition, the definition of appropriate waste system boundaries is critical for the exergy efficiencies derived from the flow analysis, as it is constrained by limited information available about the composition of flows in the system as well as about secondary production processes and their interaction with primary or traditional production chains. In the exergetic LCA, resource quality could be reflected by the savings achieved by product substitution and the consideration of the waste's upstream burden allowed for an evaluation of the waste's resource potential. For a comprehensive assessment of resource efficiency in waste LCA, the sensitivity of accounting for product substitution should be carefully analyzed and cumulative exergy consumption measures should be complimented by other impact categories.

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1. Introduction

Security of supply of material and energy resources is extremely important for industrialized countries. In Europe and other highly developed economies more sustainable management of resource flows and recovery of resources embedded in waste materials are high on the political agenda (e.g. EC, 2014), with a focus on resource efficiency and improved utilization of secondary

management plays an increasingly important role as the provider of secondary raw materials for industry. In order to design and optimize waste management systems from a resource perspective, not only the waste amounts collected for recycling but rather the quality of the recovered resources is an important basis for prioritizing between individual material and energy recovery options. Thus, both the resource potential of a waste flow and relevant quality related aspects (e.g. purity, chemical speciation, mixing) need to be reflected in the assessment of resource recovery from

resources in society (cf. EC, 2011a,b). In this context, waste

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waste. Often, these aspects are neglected in assessments of waste systems.

Several thermodynamically based approaches have been suggested to address resource depletion and/or resource quality (cf. Ayres et al., 1998; Liao et al., 2012). Exergy-based measures have been put forward on various levels of analysis, ranging from classical process engineering thermodynamics to macro-scale systems such as the global ecosystem (cf. Szargut, 2005). Whereas energy analysis does not consider the usefulness of energetic resources and cannot account for material resources, exergy can serve as a unified measure for all kinds of resources (cf. Dewulf et al., 2008). Resource consumption indicators based on cumulative exergy scores, such as the Cumulative Exergy Demand (CExD, Bösch et al., 2007) or the Cumulative Extraction of Exergy from the Natural Environment (CEENE, Dewulf et al., 2007) have been developed for Life Cycle Impact Assessment (LCIA). The loss of exergy in a system has been used to account for resource consumption and resource efficiency (cf. Gößling-Reisemann, 2008), with applications for entire economic sectors (e.g. Ayres et al., 2011) as well as for specific recycling processes (e.g. Castro et al., 2007). Previously, the exergy efficiency of waste treatment has been addressed with respect to relatively homogenous waste types, such as used cooking oil (e.g. Peiró et al., 2008), and technology networks for the recycling of specific end-of-life (EOL) products, such as EOL vehicles (e.g. Ignatenko et al., 2007). However, with respect to resource recovery from complex waste types, such as household waste composed of all kinds of materials with a range of potential management options available, the applicability of exergy analysis for assessment of resource efficiency and resource quality has not been evaluated systematically.

It is the goal of this study to provide an improved basis for the exergy-based evaluation of resource recovery from waste. Therefore, two approaches for exergy analysis using different resource accounting methods and system definitions are applied and discussed with respect to key methodological choices. The approach of mapping all the exergy flows and losses within the system under investigation is termed "exergy flow analysis", and the approach of exergy accounting following life cycle assessment (LCA) principles by applying indicators, such as CEENE, to the materials and resources included in the system is termed "exergetic life cycle assessment".

In this study, four scenarios involving increasing levels of source-segregation and recycling of selected resources (metals, plastics, and organic waste materials) from residual household waste are defined and evaluated using the two exergy analysis approaches. For exergy flow analysis, a step-wise procedure for quantification of exergy efficiencies is applied: (i) first a material and energy flow analysis is performed for each scenario, (ii) then the exergy contents of each flow in the scenarios are determined, and (iii) the resulting exergy values for all flows (input, internal, and output) are used for calculation of the recovery efficiency for each scenario by dividing the exergy contents of the useful outputs by the exergy contents of the scenario inputs. In addition to the exergy flow analysis an exergetic LCA applying the CEENE method is carried out, thereby extending the waste system to include all processes from natural resource extraction to final disposal or utilization of all outputs. Here, the resource recovery efficiency is derived by dividing the CEENE of the useful outputs by the CEENE of the inputs into the waste system. Finally, two key methodological aspects specifically related to the assessment of material quality in the two approaches are evaluated: (i) the number of compounds considered in the exergy flow analysis (i.e. how detailed the composition of each material flow is addressed), and (ii) the choice of product substitution in the exergetic LCA (i.e. which types of virgin raw materials are to be off-set by the recovered waste materials).

2. Materials and methods

2.1. Scenario analysis

Four scenarios are defined for the management of residual waste from private households. The scenarios range from direct incineration of all the residual waste in a waste-to-energy (WtE) plant in the simplest case (Scenario A) to source-segregation and recovery of metals, hard plastics, and organic waste in the most elaborate treatment system (Scenario D). The scenarios are designed in an additive manner: starting from Scenario A, each further scenario addresses the effect of source-segregation and recovery of an additional waste fraction using a specific technology (see Table 1). The scenarios are defined in a generic way using literature data from various sources, however, from a Danish perspective. This is reflected, for instance, by the waste composition data used for the scenarios (cf. Table 2) as well as by waste-to-energy being the default solution for treating residual household waste (overall around 50% of municipal solid waste was incinerated in 2012 in Denmark, Eurostat, 2014). It should be noted that the scenarios are not defined to identify the best waste treatment system, but as a basis to investigate the potentials and challenges of using exergy analysis to optimize resource recovery from waste. Hence, other scenarios or different technology combinations would be possible and the choice of scenarios in this study is not intended to be all-embracing with respect to resource recovery options from waste. In addition, a specific plant may be quite different in reality compared to the scenario models, because the scenarios are defined based on average data from literature.

Table 1Layout of the scenarios to evaluate resource recovery from residual household waste.

| | Separately collected waste | Recovery technologies |
|------------|--|--|
| Scenario A | Residual waste | Waste-to-energy (WtE) + bottom ash treatment (BAT) |
| Scenario B | Residual waste | Waste-to-energy (WtE) + bottom ash treatment (BAT) |
| | Metals (segregation efficiency: 60% for Al cans, 40% for Al containers and foils, 50% for Fe cans, 10% for other metals) | Metal sorting facility (MSF) |
| Scenario C | Residual waste | Waste-to-energy (WtE) + bottom ash treatment (BAT) |
| | Metals (segregation efficiency: 60% for Al cans, 40% for Al containers and foils, 50% for Fe cans, 10% for other metals) | Metal sorting facility (MSF) |
| | Plastics (segregation efficiency: 50% for hard plastics, 10% for soft and non-recyclable plastics (misssorted)) | Plastics sorting facility (PSF) |
| Scenario D | Residual waste | Waste-to-energy (WtE) + bottom ash treatment (BAT) |
| | Metals (segregation efficiency: 60% for Al cans, 40% for Al containers and foils, 50% for Fe cans, 10% for other metals) | Metal sorting facility (MSF) |
| | Plastics (segregation efficiency: 50% for hard plastics, 10% for soft and non-recyclable plastics (misssorted)) | Plastics sorting facility (PSF) |
| | Organic waste (segregation efficiency: 50% for food and vegetable waste) | Anaerobic digestion facility (ADF) |

Table 2Composition of the household waste input distinguishing 17 waste fractions (based on Petersen et al., 2012).

| Name of waste fraction | g/100 g of waste | Water content (% wet mass) | Ash content (% TS) | LHV (MJ/kg TS) |
|--|---------------------|-------------------------------|--------------------------|----------------------|
| Vegetable & food waste | 47 | 72 | 6.0 | 20 |
| Aluminum foil & containers | 0.5 | 19 | 76 | 6.8 |
| Aluminum cans | 0.4 | 8.3 | 100 | 0.0 |
| Diapers & hygiene products | 5.6 | 46 | 8.0 | 22 |
| Glass | 2.4 | 7.1 | 100 | 0.0 |
| Paper and cardboard | 19 | 20 | 17 | 15 |
| Paper-plastic- composites | 1.7 | 17 | 3.2 | 22 |
| Plastics (other) | 2.2 | 11 | 4.6 | 36 |
| Plastics (hard) | 4.5 | 8.5 | 5.5 | 37 |
| Cat litter | 1.4 | 16 | 93 | 0.0 |
| Stones and other non- combustibles | 2.8 | 15 | 83 | 1.1 |
| Fe metals | 0.9 | 13 | 100 | 0.0 |
| Other metals | 0.9 | 8.3 | 100 | 0.0 |
| Textiles, shoes, rubber | 2.3 | 6.2 | 5.4 | 21 |
| Garden waste & wood | 4.9 | 46 | 23 | 14 |
| Other combustibles | 3.4 | 35 | 33 | 17 |
| Hazardous waste (batteries) | 0.1 | 8.9 | 86 | 0.6 |
| Total | 100 | 46 ^a | 24 ^a | 17 ^a |

Abbreviations: LHV = lower heating value; TS = total solids.

The waste input considered is identical for all the scenarios and represents a typical composition of Danish household waste from single-family residential structures, after the source-segregation of paper and container glass (Petersen et al., 2012). In Scenario A all the waste is directed to a waste-to-energy plant, where heat and electricity are produced from waste incineration (cf. Table 1). The bottom ash is treated to separate ferrous and non-ferrous metals from the incineration residues. The latter are subsequently deposited at an inorganic waste landfill together with fly ashes from the WtE plant. The dewatered filter cake from air pollution control is landfilled at an underground repository. The metal scraps from bottom ash treatment are transported to a metal smelter and used in secondary production. In Scenario B metals are sourcesegregated with the specific efficiencies shown in Table 1 (based on Bernstad et al., 2012; Møller et al., 2013; Table SI-2 of the Supporting Info). No miss-sorting is assumed for metals. Thus, no other materials are contained in the metal bins. The residual waste is directed to a waste-to-energy plant for energy recovery following the same route as in Scenario A. The collected metal scraps are transported to a metal sorting facility (MSF), which splits the input into Fe, Al, Cu scraps, and solid residues. The metal scraps are directed to secondary production plants and the solid residues are directed to an inorganic waste landfill. In Scenario C, hard plastics (bottles, containers, and various plastic products) are source-segregated in addition to metals (efficiencies based on Møller et al., 2013). The collected plastics are directed to a plastics sorting facility (PSF), where polyethylene terephthalate (PET) bottles and high-density polyethylene (HDPE) plastics are sorted out for material recycling and residues are used as a residue-derived fuel. Finally, in Scenario D half of the organic household waste is also collected in a separate bin (cf. Table 1, segregation efficiency based on Møller et al., 2013). The organic waste is directed to an anaerobic digestion facility (ADF), where biogas is produced from biodegradable matter and converted to heat and electricity in a combined heat and power (CHP) plant. The solid digestion residue is subsequently ripened and spread on land. In addition to the material and energy flows of treatment processes, fuel use for waste collection and transport is included in the scenario modeling.

2.2. Material and energy flow modeling

2.2.1. Modeling approach

Material flow analysis is performed for various waste fractions based on the mass balance principle (cf. Brunner and Rechberger, 2004) using the STAN software (Cencic and Rechberger, 2008). Mass and energy balances are established for each process. As an example, the qualitative model for Scenario D is shown in Fig. 1. because Scenario D comprises the maximum of separately collected wastes and treatment pathways (cf. Table 1). Material and energy flows are determined with respect to the treatment of 1000 kg of household waste, thus, representing the functional unit of the analysis. Uncertainty is considered in the model with respect to the energy consumption of waste collection, treatment, and transport, on the one hand, and with respect to the material and energy recovery efficiencies of the individual waste treatment plants, on the other hand. Uncertain quantities are defined using triangular or normal probability distributions reflecting probable ranges of variation for model parameters. The probable ranges are in some cases based on specific literature sources and in other cases they represent estimates by the authors qualified by the typical range of values found in the literature. The uncertainty of the resulting material and energy flows are computed using Monte Carlo simulations with 10.000 iterations using the MS Excel®based @Risk software (Palisade Corporation 2013). In contrast, as the specific scenario assumptions are not the focus of this evaluation, aspects such as the composition of the waste input (cf. Table 1), the source-segregation efficiencies, and the material transfer coefficients related to non-resource flows (e.g. effluent from ADF) are not addressed in the uncertainty analysis.

2.2.2. Waste input

The waste input is defined based on the composition of Danish household waste, after the source-segregation of paper and container glass. The data originates from a study by Petersen et al. (2012) about household waste of single-family residential structures in Denmark (Aarhus). The dataset is available from the EASE-TECH database (cf. Clavreul et al., 2014), where the originally sorted 123 characteristic material fractions have been merged to 48 material fractions (cf. Table SI-1 of the Supporting Information). For these material fractions, the contents of 46 chemical elements are given in the database. Hence, the waste flows are modeled down to the level of elements present in each material fraction. However, for presenting the analysis and its results the 48 material fractions are further aggregated to 17 fractions (see Table 2). The average water content of the input waste is rather high (46% on a wet mass basis), which is due to the relatively large proportion of food waste (47% of the wet waste is food waste). Therefore, the lower heating value (LHV) of the wet waste is only 8.2 MJ/kg compared to 17.3 MJ/kg of the dry waste (Table 2).

2.2.3. Waste treatment processes

The waste-to-energy (WtE) plant model within the scenarios is a grate furnace with a typical wet system for flue gas treatment with fly ash separation and a two-stage acidic pollutant removal in aqueous solution and an on-site wastewater treatment of wet-scrubber effluents (cf. Koehler et al., 2011). The bottom ash is quenched in water after its discharge from the grate. Material

^a The average water content, ash content, and LHV are calculated as weighted averages according to the waste composition.

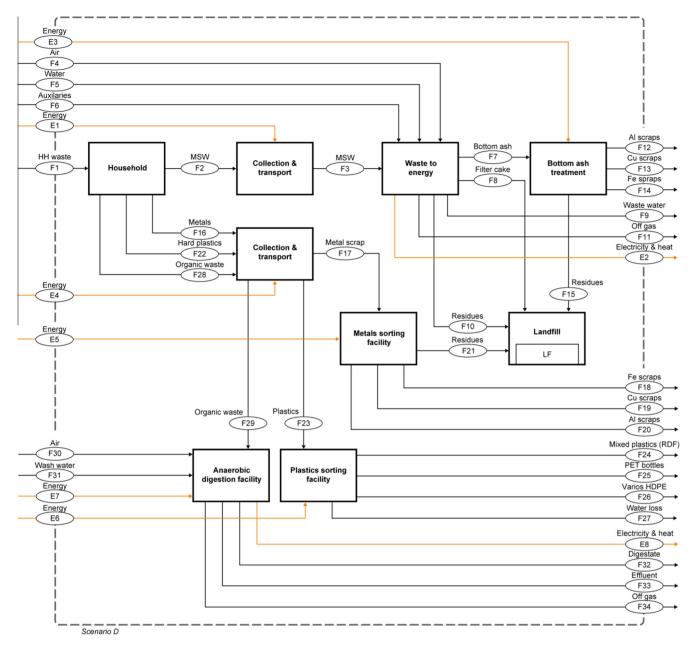


Fig. 1. Qualitative illustration of the material and energy flows of Scenario D. Flows (F = material flows, E = energy flows) are shown as arrows, processes are shown as boxes. All the energy (=fuel) spent on waste collection and material transport is considered in the processes "Collection & transport". Waste system boundary is illustrated via the gray dashed line with rounded corners.

transfer coefficients and net efficiencies for heat and power production are based on average European data. The net efficiencies of electricity and heat production related the LHV of the residual waste are defined via triangular distributions with a mode of 15% and a range from 5% to 25% for electricity and with a mode of 37% and a range from 3% to 72% for heat, respectively (based on Reimann, 2013). The respective shares of electricity and heat are inversely correlated, so the higher the amount of electricity produced, the lower the amount of heat produced. Thus, the overall energy efficiency ranges between 28% (max. electricity and min. heat) and 77% (min. electricity and max. heat) with an average of 52% (the probability distribution of the overall energy efficiency is illustrated in Fig. SI-1 of the Supporting Information). It should be noted, that at some Danish WtE plants overall efficiencies might be higher due to very efficient heat utilization (cf. Rambøll, 2006). The material and substance flows of the WtE plant are modeled

based on the probabilistic transfer coefficients (TC) reported by Koehler et al. (2011) for European municipal solid waste incineration plants. The transfer coefficients used to model the material flows of the WtE plant and the major input and output flows are shown in Tables SI-3 and SI-4 of the Supporting Information (SI).

The bottom ash treatment (BAT) plant is located at the WtE plant and is based on the Danish state of the art as described by Allegrini et al. (2014). The modes for the metal recovery efficiencies are 85% for Fe, 62% for Al, and 43% for Cu (cf. Allegrini et al., 2014, 2015). The recovery ratios relate to the metal loads in the input and in the output and are defined as triangular distributions ranging from 0.7 to 0.95 for Fe, 0.5 to 0.7 for Al, and 0.2 to 0.6 for Cu, respectively (modes see above). The output flows of Fe, Al, and Cu equal the amount of pure metal in the scrap. Hence, the actual scrap flows will be higher due to accompanying elements in alloys, impurities and contaminants in the scrap (cf. Allegrini et al., 2015).

In order to calculate the (approximate) amount of scrap produced from the bottom ash treatment, a correction factor of 0.75 is assumed. Hence, 25% of the material in the scrap metal flow is not constituted by the respective metal but by other substances such us elements bound to the metals in chemical compounds (e.g. oxygen in iron oxide) or dust particles adhering to the metal pieces (e.g. SiO₂). The grain size fractions of Al scraps are used to calculate the oxidation ratio of Al scrap from bottom ash and are also adopted from Allegrini et al. (2014). Thus, 24% of Al is present in the fraction <5 mm, 23% of Al in the fraction 5-10 mm, and 53% of Al in the fraction above 10 mm. With respect to the Al scrap, the oxidation rates are different for the three scrap size classes, with 34% for <5 mm, 28% for 5-10 mm, and 19% for >10 mm (based on Allegrini et al., 2014). For Fe scrap an average oxidation rate of 25% is assumed in agreement with data reported by López-Delgado et al. (2003). Finally, no oxidation is considered for Cu scrap. For Al. the oxidized fractions are considered as losses in the recycling process, where only the metallic contents are directed to the secondary product. For the Fe scrap, 90% of the iron in the scrap is directed to the secondary product (cf. Allegrini et al., 2015). For the Cu scrap, all the elemental Cu ends up in the product. The energy consumption (electricity and fuel) data for the bottom ash treatment processes are retrieved from Allegrini et al. (2015), who provide specific data for a Danish state of the art treatment plant (0.24 kW h/Mg of bottom ash, 2.1 L diesel/Mg of Fe scrap, 0.5 L diesel/Mg of bottom ash after Fe separation, 58 kW h/Mg of Non-Fe scrap; given as mean values of normal distributions with relative standard deviations of 20%).

The metal sorting facility (MSF) splits the source-segregated metals into pure Fe, Al, and Cu scraps and residues as outputs. The recovery efficiency for Fe is given by a triangular distribution with a mode of 95% and a range from 70% to 98% (cf. Møller et al., 2013). The recovery efficiencies for Al and Cu are defined as triangular distributions with modes of 90% and ranges from 70% to 95%. The energy consumption for sorting one tonne of metal input is also based on Møller et al. (2013) and given by 73 kW h of electricity (triangular distribution with a range from 40 to 90 kW h and a mode of 73 kW h) and 1 ± 0.2 L of diesel (normal distribution with mean 1 and a relative standard deviation of 20%).

The plastics sorting facility (PSF) splits the input of source-segregated plastics into PET bottles (baled), HDPE flakes (hard), and mixed plastics residues, which are used as a residue-derived fuel (RDF) in industrial combustion processes. The recovery efficiencies for PET bottles and HDPE are defined by triangular distributions with modes of 85% (cf. Møller et al., 2013) and ranges from 60% to 90%. Based on Møller et al. (2013), the energy consumption for sorting one tonne of plastics is similar to treating one tonne of metals in the MSF.

The anaerobic digestion facility (ADF) receives the organic waste fraction (food waste) and converts it to biogas and digestate. The biogas is used in a combined heat and power plant to produce heat and electricity. The ADF is modeled based on data reported by Banks et al., 2011 for anaerobic digestion of source-segregated domestic food waste. The reported methane yield of 98 m³ per tonne of wet food waste input was defined as the mode of triangular distribution ranging from 50 to 100 m³ per tonne of input (skewed shape to reflect lower methane yields often reported in other studies, cf. Davidsson et al., 2007). Afterwards the lower heating value of the biogas is converted into electricity (16% of LHV) and into heat (27% of LHV) (net conversion efficiencies based on Banks et al., 2011). In addition, 1 ± 0.2 L diesel per tonne of input is consumed from external sources to operate the ADF.

Waste collection is modeled via fuel intensities depending on the type of collection system using Danish data extracted from the EASETECH database (cf. Clavreul et al., 2014). For residual waste, diesel consumption is 1.6 ± 0.32 L per tonne of waste collected. For source-segregated waste materials, diesel consumption is $3.5\pm0.7\,\mathrm{L}$ per tonne of waste collected. All transport is covered by truck transport on the road $(0.03\pm0.003\,\mathrm{L}$ diesel per kilometer and tonne of waste) with the distances given in Table SI-5 of the SI. The relative standard deviation of fuel consumption for waste collection is set to be double the relative standard deviation of fuel consumption for truck transport (10%), because collection activities may vary to a larger degree (settlement structure, collection routes, collection trucks) than road transport with trucks.

2.3. Exergy analysis

Exergy analysis measures the loss of available work due to the transformation of matter and energy. As exergy analysis is based on the Second Law of thermodynamics, it can be used to describe the quality of energy. Exergy was defined as the maximum work output attainable in the natural environment, or a minimum work input necessary to realize the reverse process (Rant 1964 cited in Szargut, 2005). Exergy does not satisfy a law of conservation, because every irreversible process causes exergy destruction.

2.3.1. Exergy flow analysis

In the absence of magnetism, surface tension, and nuclear reaction, exergy can be divided into physical exergy, chemical exergy (Ex,ch), potential exergy, kinetic exergy, and exergy in electricity (cf. Hepbasli, 2008). For the exergy flow calculations in this study only physical and chemical exergy and electricity are relevant and included in the balances. The physical exergy accounts for systems at pressure and temperature conditions different from their natural environment. For the exergy balances of the energy recovery facilities (WtE plant and CHP unit of the ADF), the exergy of heat Ex_{Heat} is calculated according to the formula given in Eq. (1) (see Hepbasli, 2008), where Q_k is the heat transfer rate through the boundary at temperature T_k at location k, the subscript zero indicates properties at conditions of the ambient environment (for all calculations standard conditions with T_0 = 298,15 K and P_0 = 1 bar are used). Electricity has an exergy conversion factor of 1, as electricity can be completely transformed into work. For the exergy content of the produced heat, 368 K is used as the water temperature of the district heating system. The temperature of the off gas released through the stack is set to 403 K (cf. Table SI-4 of the SI).

$$\mathsf{Ex}_{\mathsf{Heat}} = \left(1 - \frac{T_0}{T_k}\right) \cdot \mathsf{Q}_k \tag{1}$$

The chemical exergy of a compound is due to the exergy stored in chemical bonds with a composition different from the surroundings (reference environment). In this work all the specific chemical exergy values for compounds are taken from Szargut (2005) with some additional exergy values for organic compounds extracted from Ignatenko et al. (2007) and Alvarenga et al. (2013).

The chemical exergy of a substance in a mixture is lower than the exergy of the substance in its pure state, as work is required to separate a mixture into its pure constituents. The loss of exergy due to mixing occurs as pure constituents are spontaneously diluted in a mixture. The minimum exergy required to separate a substance from a matrix is equal to the exergy of mixing. Based on Ignatenko et al. (2007) the exergy of a material stream is calculated as the sum of its components' chemical exergies (first part of Eq. (2)) and the exergy loss due to the mixing of the different components (second part of Eq. (2)). In Eq. (3), Ex_{ch} is the total chemical exergy of the material flow (J), n_i is the number of moles of component i in the material flow, n is the total number of moles in the mixture, $e_{ch,i}$ is the specific chemical exergy of component i (J/mol), i is the gas constant (8.31 J/mol K), i is the standard

temperature (K), and x_i is the molar fraction of the component i in the mixture of the material flow.

$$\operatorname{Ex}_{\operatorname{ch}} = \sum_{i} n_{i} e_{\operatorname{ch},i} + R T_{0} n \sum_{i} x_{i} \ln(x_{i})$$
 (2)

Waste materials are mixtures of multiple compounds which are associated with each other in many different ways. Hence, to calculate the exergy of material flows in a waste system, simplifications and assumptions are necessary. The composition of each flow is approximated by a mixture of several representative compounds (cf. Table 3). The waste input is defined via 17 material fractions

Table 3Compounds considered for calculating the chemical exergies of the material flows in the waste treatment scenarios.

| Material flow | Representative chemical compounds |
|---|---|
| Household waste input Vegetable & food waste | Cellulose ($C_6H_{10}O_5$), Alanine ($C_3H_7NO_2$), Oleic acid ($C_{18}H_{34}O_2$), H_2O |
| Aluminum foil & | Al, Cellulose ($C_6H_{10}O_5$), H_2O |
| containers | |
| Aluminum cans Diapers & hygiene products | Al, Fe, H ₂ O Polyethylene ((C ₂ H ₄) _n), H ₂ O |
| Glass | SiO ₂ , H ₂ O |
| Paper and cardboard Paper-plastic- composites | Cellulose ($C_6H_{10}O_5$), SiO ₂ , H_2O Cellulose ($C_6H_{10}O_5$), Polypropylene ((C_3H_6) _n) |
| Plastics (other) Plastics (hard) Cat litter | Polypropylene $((C_3H_6)_n)$, PVC $((C_2H_3CI)_n)$, H_2O Polyethylene $((C_2H_4)_n)$, H_2O SiO ₂ , H_2O |
| Stones & other non- comb. | SiO ₂ , H ₂ O |
| Fe metals | Fe, Al, H ₂ O |
| Other metals | Fe, Al, Cu, SiO ₂ , H ₂ O |
| Textiles, shoes, rubber | Cellulose (C ₆ H ₁₀ O ₅), Rubber ((C ₇₈ H ₁₂₉ S) _n), H ₂ O |
| Garden waste & wood | Cellulose ($C_6H_{10}O_5$), Lignin ($C_{10}H_{11}O_3$) |
| Other combustibles | Average of combustible fractions, H ₂ O |
| Hazardous waste | Zn, Fe, Mn, SiO ₂ , H ₂ O |
| (batteries) | |
| Air (WtE input) | N_2 , O_2 |
| Water (WtE input) | H_2O |
| Auxiliary material | Hydrated lime (CaOH ₂), Limestone (CaCO ₃), |
| (WtE) | Sodium hydroxide (NaOH) |
| Bottom ash (WtE | SiO ₂ , CaO, MgO, Fe, Fe ₂ O ₃ , Sl, Al, Al ₂ O ₃ , Cu, CuO, |
| output) | H ₂ O |
| Fly ash (WtE output) | NaCl(s), ZnCl(s), CaSiO ₃ , Fe ₂ O ₃ , SiO ₂ |
| Filter cake (WtE output) | CaSO ₄ , H ₂ O |
| Effluent (WtE output) | NaCl(1), H ₂ O |
| Off gas (WtE output) | N ₂ , O ₂ , H ₂ O, CO ₂ |
| Fe scrap (BAT output) | Fe, Fe ₂ O ₃ , SiO ₂ |
| Al scrap (BAT output) | Al, Al ₂ O ₃ , SiO ₂ |
| Cu scrap (BAT output) | Cu, CuO, SiO ₂ |
| Bottom ash residue (BAT output) | SiO ₂ , CaO, MgO, Fe, Fe ₂ O ₃ , Sl, Al ₂ O ₃ , Cu, CuO, H ₂ O |
| Fe scrap (MSF output) | Fe |
| Al scrap (MSF output) | Al |
| Cu scrap (MSF output) | Cu |
| Residue (MSF output) | Fe, Al, SiO2, Cu, Cellulose (C ₆ H ₁₀ O ₅), H ₂ O |
| PET granulate (PSF | Polyethylene $((C_2H_4)_n)$ |
| output) HDPE granulate (PSF | Polyethylene $((C_2H_4)_n)$ |
| output) | 1 oryethyrene ((C2114)n) |
| Mixed plastics (PSF | Polyethylene $((C_2H_4)_n)$, Polyprop. $((C_3H_6)_n)$, PVC |
| output) | ((C ₂ H ₃ Cl) _n), H ₂ O |
| Wash water (ADF | H_2O |
| input) | |
| Biogas (ADF output) | CH_4 , CO_2 , H_2O |
| Digestate (ADF output) | Cellulose (C ₆ H ₁₀ O ₅), Alanine (C ₃ H ₇ NO ₂), Oleic acid |
| T00 | $(C_{18}H_{34}O_2), H_2O$ |
| Effluent (ADF output) | H_2O |

Abbreviations: WtE = Waste-to-Energy, BAT = Bottom Ash Treatment, MSF = Metal Sorting Facility, PSF = Plastics Sorting Facility, ADF = Anaerobic Digestion Facility.

that are again composed of two to five relevant compounds for representation of the chemical composition. The compounds considered in each flow are defined to correspond to the elemental concentrations of the individual material fractions. For instance, the material fraction "Aluminium foil and containers" consists of 81 g dry matter/100 g wet weight and 19 g water/100 g wet weight. 75% of the dry matter is Al, 12% is biogenic C, 6% is O, 2% is H, and 5% are other elements. Based on the major constituents, metallic Al, cellulose (organic matter), and water were defined as representative constituents of this material fraction (see Table 3). The mass fractions of each chemical compound considered for each material flow and the data to calculate the flow exergies are shown in Tables SI-6 to SI-10 of the SI. However, though the compounds are chosen to account for the largest share of elements present in each fraction, the chosen number of compounds is arbitrary to some degree. Therefore, the effect of considering a higher number of material constituents for the exergy flow calculations is evaluated in a sensitivity analysis. For this, the number of compounds constituting the dry mass of each material flow is doubled and the resulting changes of the exergy flows as well as of the resource efficiency calculations are analyzed (cf. Chapter 4.1 and Table SI-14

In case of the exergy flow analysis, the value of a resource is expressed by its exergy content (i.e. the specific exergy of a material is used as a proxy for resource quality). Therefore, the resource recovery efficiency is calculated by dividing the exergy content of the useful outputs (i.e. waste-derived materials which form an input to (secondary) production processes) by all the exergy inputs of the waste treatment scenario (see Eq. (3)). The efficiency evaluation highlights losses of exergy in the waste system. The resource efficiencies are necessarily below 100% for the scenarios, because in real systems irreversible processes cause exergy to be destroyed.

$$Resource\ recovery\ efficiency = \frac{Exergy\ (useful\ outputs)}{Exergy\ (inputs)} \tag{3}$$

2.3.2. Exergetic life cycle assessment (LCA)

The functional unit of the assessment is the treatment of 1000 kg of household waste. In addition to the material and energy flows of the waste treatment scenarios, life-cycle inventory (LCI) data (e.g. primary and secondary metal production, electricity production, fuel provision, etc.) are retrieved from the ecoinvent database (version 3.1), which includes several thousands of industrial life cycle inventory datasets (Swiss Centre for Life-Cycle Inventories, 2014). CEENE is used as an LCIA method to evaluate the impact on natural resources, as it provides a single measure to quantify the consumption of all kinds of resources to provide a specific service or product (Dewulf et al., 2007). The total CEENE score is composed of eight categories, namely atmospheric resources, land resources, water resources, minerals, metal ores, nuclear energy, fossil fuels, and renewable resources. Thus, CEENE evaluates energy carriers, non-energetic resources, and land occupation. Solar irradiation available for photosynthesis is used as a proxy for land occupation, since this solar exergy is no longer available to nature. With respect to land occupation and the biotic portion of renewable resources, the CEENE method is adapted according to Alvarenga et al., 2013, to account for land resources using spatially differentiated characterization factors. The datasets used for quantifying the total resource impacts of the waste treatment scenarios are shown in Tables SI-11 and SI-12 of the SI. Scenario inputs (waste input, fuel & electricity inputs, and auxiliary materials) as well as direct land occupation by the waste treatment plants are considered as burdens in the assessment. Savings are credited to the scenarios based on the substitution of products by waste-derived products via material recycling or energy

recovery processes. System expansion is applied following a consequential approach using marginal technologies to account for multi-functionality (EC-JRC-IES, 2010). Hence, functionally equivalent products are identified for the waste-derived products (e.g. primary aluminum is replaced by cast aluminum produced from aluminum scraps) and substitution ratios (between 0 (no substitution) and 1 (full substitution)) are applied to account for the savings achieved by resource recovery from waste by the avoided alternative production route.

In order to be consistent with the exergy efficiencies for the exergy flow analysis, the upstream burden of the input waste streams (i.e. the effort required to produce the materials contained in the waste) is included in the assessment and compared to the recovery of materials from the waste for secondary production. Due to the consideration of the resource potential of the waste input, the efficiency evaluation allows for the identification of resource losses, offering a means to optimize waste treatment in an objective way. Therefore, the zero burden assumption is abandoned for the waste input in the present analysis. Hence, for each of the originally 48 material fractions of the waste input one or more representative product datasets are identified and the upstream burden of the waste input is considered via the respective CEENE scores (cf. Table SI-11 of the SI). The products constituting the waste input are chosen on the level of primary production (e.g. low-alloyed steel for ferrous metals in waste, paper (newsprint) for waste magazines, grass (organic) for yard waste, etc.) in most cases, though in some cases manufactured products (e.g. NiMH batteries for waste batteries, sanitary ceramics for waste ceramics, etc.) are used.

The benefits achieved by waste-derived products are determined for each scenario based on defined recycling processes and substitution pathways (cf. Table SI-13 of the SI). The recovered scrap metals are directed to secondary metal production with different yields for Fe, Al, and Cu. Fe recycling is modeled using the ecoinvent process "Steel, low-alloyed, electric arc furnace", with a yield of 90% (90% of the Fe in the input is directed to the secondary product) for scrap from bottom ashes and a yield of 100% for scrap from the source segregated metal stream. Secondary steel is considered to fully substitute primary steel (factor 1), which is modeled by the ecoinvent process "Steel, low-alloyed, converter". Al recycling is described by the process "Aluminium, cast alloy, treatment of aluminium scrap, post-consumer, at refiner" with only the metallic Al in the scrap directed to the secondary Al product (oxidized Al ends up in the slag). Full substitution is assumed for secondary Al with respect to primary Al (process "Alumnium, primary, production"). For Cu too, all the metallic Cu is directed to the secondary product in the recycling process ("Copper, treatment of scrap by electrolytic refining"), with secondary Cu being a full substitute for primary Cu ("Copper, production, primary"). Due to a lack of inventory data for recycling processes, the material recycling of plastics is not explicitly modeled, but the benefit from plastics recycling is determined based on market price-based substitution ratios for the plastics outputs of the sorting facility. Thus, instead of modeling secondary production efficiencies, the savings are assumed to be directly proportional to the average market prices of primary vs. secondary plastics raw materials. In view of the market prices of PET bottle bales and HDPE flakes compared to the primary plastic flakes (cf. www.plasticker.de), a substitution ratio of 0.35 was used in the analysis (cf. Table SI-15 of the SI). The residual mixed plastics are used as a residue derived fuel (RDF) and substitute natural gas based on their heating value ("Natural gas, high pressure, market, Denmark"). The benefit from digestate application on agricultural land is determined based on its phosphate content, which substitutes the equivalent amount of phosphate from mineral fertilizer (ecoinvent process "Ammonium nitrate phosphate, as P₂O₅, at regional storehouse"). Full substitution of the marginal technologies is assumed for the produced electricity and heat based on their energy contents. These are "Electricity, high voltage, production, hard coal, Denmark" and "Heat production, natural gas, at industrial furnace >100 kW, Europe", respectively. Because the accounting of material recycling from waste is typically associated with large uncertainty (cf. Brogaard et al., 2014) and has a major impact on the results of life cycle assessments of waste systems (cf. Laurent et al., 2014), the effect of choosing different substitution ratios as well as recycling processes and substituted products on the modeling results, is investigated in a sensitivity analysis. Therefore, break-even substitution ratios (ratio when one scenario becomes better or worse than another one) are determined and the effect of different product substitution pathways on resource efficiency is analyzed (cf. Chapter 4.1 and Table SI-19 of the SI).

The resource recovery efficiency from a cumulative resource accounting perspective is calculated by dividing the overall benefit achieved by resource recovery from waste by the overall burden of the waste treatment scenario (see Eq. (5)). The CEENE scores are used to express the value of a resource, thus the higher the cumulative exergy extraction to provide a specific resource, the higher the value of the respective flow. This way of calculating recovery efficiencies focuses on the identification of resource losses, with a 100% efficiency representing a theoretical optimum that will not be achieved in real systems.

$$Resource\ recovery\ efficiency = \frac{CEENE(overall\ benefit)}{CEENE(overall\ consumption)} \end{(5)}$$

3. Results

3.1. Material and energy flows

Material and energy flows are determined for each scenario based on the inputs to the waste treatment scenario and transfer coefficients to model the partitioning of inputs into the various outputs. The material flow diagram of Scenario D is shown in Fig. 2 in Sankey style (width of an arrow is directly proportional to flow value). Only material flows are included in the diagram. Each flow is given by its most probable value without a specification of the corresponding uncertainty range (uncertainty ranges are shown in Table 4). In case of Scenario D, around 73% of the household waste is directly incinerated in the WtE plant. The bottom ash residues amount for 20% of the waste input or 140 kg, out of which 10 kg of Fe scrap, 6.1 kg of Al scrap, and 0.2 kg of Cu scrap are recovered. One percent of the household waste input is source-segregated as metals, 2.5% are collected as hard plastics, and around 24% is collected as organic waste. These sourcesegregated waste materials are transported to processing facilities, where materials and energy are recovered (cf. Fig. 2 and Table 4).

3.2. Exergy flow analysis

The exergy flows are the result of the specific chemical and physical exergies calculated for each material or energy flow and the flow amount (cf. Table 1). The exergy flows of Scenario D are shown in Fig. 3. Most of the exergy is contained in the household waste input, of which almost 77% are directed to the WtE plant. In the WtE plant around 7000 MJ_{ex} are lost (not shown explicitly in Fig. 3), resulting in an exergy recovery efficiency via electricity and heat of 16%. During the subsequent treatment of the bottom ash, 54% of the exergy input in the ashes is recovered via the metal scraps. With respect to the source-segregated fractions, plastics and organic waste constitute the main exergy flows, with a

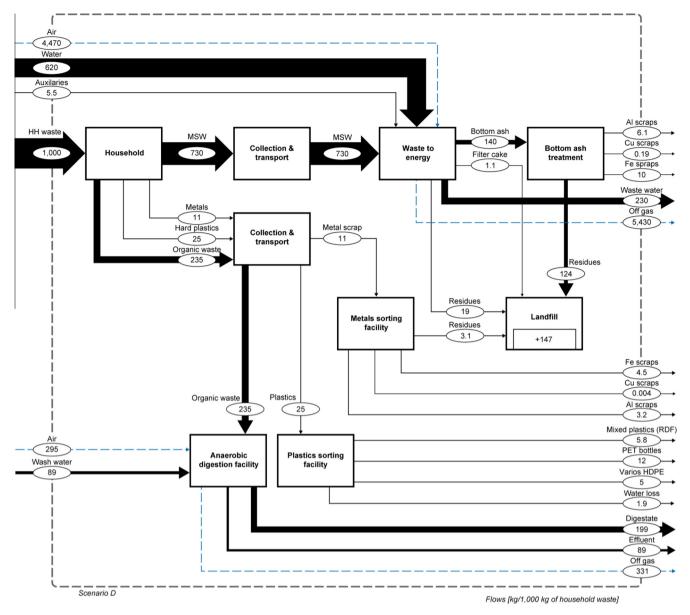


Fig. 2. Material flows of Scenario D in kg per 1000 kg of household waste input (mass balances may not add up exactly in the diagram due to rounding; gaseous flows are shown as blue dashed lines not in Sankey style; energy flows are not shown).

particularly high specific exergy of plastics (44 $M_{\rm Jex}/kg$) compared to metals (13.4 $M_{\rm Jex}/kg$) and organic waste (6.5 $M_{\rm Jex}/kg$)). The highest recovery efficiency is achieved for plastics in the PSF (99%), which is due to the residues being considered as a product and the subsequent utilization of the residue-derived fuel not being considered within the exergy flow analysis (large losses occur during incineration which takes place outside the investigated system). As a consequence of the high separation efficiencies as well as the (assumed) purity of the metal input, the recovery efficiency for metals in the MSF is 83%. Finally, the lowest efficiency among the source-segregated wastes is achieved for organic waste, because only 25% of the exergy input is directed to electricity and heat and digestate. Again, the major losses in the ADF are associated with biogas combustion and energy recovery.

The consumed and recovered exergies in the various flows are shown for each scenario in Fig. 4. In any scenario, the exergy input via the household waste is by far the largest flow, constituting around 98% of the total exergy consumption of the scenarios (cf. Fig. 4). The most important materials for the exergy content of

the waste input are the organic waste (31%) and other combustible waste fractions (e.g. plastics (25%), paper (22%), textiles and hygiene products (16%)), with metals and other inert materials only amounting for 3% of the total exergy of the waste input. Among the benefits, the production of electricity and heat accounts for 50% (Scenario D) to 89% (Scenario A) of the total exergy recovered in products. Apart from energy recovery, the recovery of plastics is associated with large exergy benefits (37% of total exergy recovered in Scenario C and 34% in Scenario D), due to the high specific exergy content of these fractions. Consequently, the metal recovery contributes only little to the total exergy of the recovered resources (between 8% for Scenario D and 13% for Scenario B). From Fig. 4, it is apparent that the exergy recovered increases from Scenario A toward Scenario D. This is due to the increasing diversion of waste fractions to treatment pathways different from waste incineration, where large exergy losses occur in the system. However, in comparison to Scenario C, the amount of exergy recovered in Scenario D only increases due to the consideration of digestate as a product, as the energy recovery from food

Table 4Selected material and energy flows for each scenario per 1000 kg of household waste input.

| Material flows | Scenario A (kg) | | Scenario B (kg) | | Scenario C (kg) | | Scenario D (kg) | |
|-----------------------------------|-----------------|--------------------|-----------------|--------------------|-----------------|--------------------|-----------------|--------------------|
| | Mean | Range ^a |
| Waste to WtE | 1000 | n.a. | 989 | n.a. | 964 | n.a. | 729 | n.a. |
| Fe scrap (BAT) ^b | 15.5 | 13.3-16.8 | 10.2 | 8.8-11.1 | 10.2 | 8.8-11.1 | 10.2 | 8.7-11.1 |
| Al scrap (BAT) ^b | 9.5 | 8.0-10.5 | 6.9 | 5.9-7.6 | 6.2 | 5.2-6.8 | 6.1 | 5.2-6.7 |
| Cu scrap (BAT) ^b | 0.2 | 0.1-0.3 | 0.2 | 0.1-0.3 | 0.2 | 0.1-0.2 | 0.2 | 0.1-0.2 |
| Metals to MSF | n.a. | n.a. | 10.8 | n.a. | 10.8 | n.a. | 10.8 | n.a. |
| Fe scrap (MSF) | n.a. | n.a. | 4.5 | 3.5-4.6 | 4.5 | 3.5-4.6 | 4.5 | 3.5-4.6 |
| Al scrap (MSF) | n.a. | n.a. | 3.2 | 2.6-3.3 | 3.2 | 2.6-3.3 | 3.2 | 2.6-3.3 |
| Cu scrap (MSF) | n.a. | n.a. | 0.004 | 0.0-0.0 | 0.004 | 0.0-0.0 | 0.004 | 0.0-0.0 |
| Plastics to PSF | n.a. | n.a. | n.a. | n.a. | 24.9 | n.a. | 24.9 | n.a. |
| PET bottles | n.a. | n.a. | n.a. | n.a. | 12.3 | 9.3-12.7 | 12.3 | 9.3-12.7 |
| Various HDPE | n.a. | n.a. | n.a. | n.a. | 5.0 | 3.8-5.2 | 5.0 | 3.8-5.2 |
| Mixed plastics | n.a. | n.a. | n.a. | n.a. | 5.8 | 5.5-9.3 | 5.8 | 5.5-9.3 |
| Organic waste to ADF | n.a. | n.a. | n.a. | n.a. | n.a. | n.a. | 235 | n.a. |
| Digestate (ADF) | n.a. | n.a. | n.a. | n.a. | n.a. | n.a. | 199 | n.a. |
| Residues to landfill ^c | 162 | 160-164 | 162 | 161-165 | 161 | 160-163 | 147 | 147-150 |
| Energy flows | (MJ) | Range ^a |
| Electricity from WtE | 1230 | 593-1870 | 1230 | 593-1870 | 1110 | 533-1680 | 975 | 470-1480 |
| Heat from WtE | 3040 | 1210-4980 | 3040 | 1180-4960 | 2750 | 1030-4470 | 2410 | 933-3920 |
| Electricity from ADF | n.a. | n.a. | n.a. | n.a. | n.a. | n.a. | 133 | 79-134 |
| Heat from ADF | n.a. | n.a. | n.a. | n.a. | n.a. | n.a. | 220 | 130-222 |
| Electricity consumed | 2.2 | 1.3-3.0 | 4.4 | 3.2-5.2 | 10.8 | 8.0-12.0 | 10.8 | 8.0-11.9 |
| Diesel consumed | 118 | 89.5-145 | 120 | 91.2-146 | 127 | 100-153 | 157 | 130-183 |

Abbreviations: WtE = waste-to-energy, BAT = bottom ash treatment, MSF = metal sorting facility, PSF = plastics sorting facility, ADF = anaerobic digestion facility, PET = polyethylene terephthalate, HDPE = high-density polyethylene, n.a. = not applicable.

- ^a Uncertainty range is defined as the interval including 95% of the values (from the 2.5 percentile to the 97.5 percentile).
- ^b The scrap flow consists of the respective pure metal (75%) and other elements (25%).
- ^c Sum of the material flows F8, F10, F15, and F21 (cf. Fig. 1).

waste digestion does not appear to be more efficient than energy recovery in the WtE plant. The exergy results including their uncertainty ranges are shown for the major waste flows and the recovered resources in Table SI-16 of the SI.

3.3. Exergetic life cycle assessment (LCA)

The CEENE consumed in the scenarios is dominated by the upstream burden related to production of the materials in the waste input, which accounts for 99.6-99.7% of the total CEENE consumed by each scenario. The consumption of fuels and electricity accounts for up to 0.17% (Scenario D), the consumption of auxiliaries and landfilling constitute 0.22% (Scenario A) at the maximum, and the direct land occupation caused by the treatment plants amounts is less than 0.03% of the CEENE consumed in any scenario (see Fig. 5 and Table SI-17 of the SI for detailed results including uncertainty ranges). The burden associated with the waste input represents the major input to the treatment system, with a lot of variation in the contributions to the overall CEENE score from the individual material fractions in the waste. Waste paper accounts for 42% of the waste's upstream burden. This is due to the large impact associated with virgin paper production (due to consumption of land and biotic resources) as well as because of paper and cardboard comprising almost 20% of the waste (cf. Table 2). 21% of the upstream burden is associated with the organic waste fraction consisting of meat products (pork, chicken), vegetables, grains, grass, and wood. Further, textiles and hygiene products and paper-plastic composites add up to 20% of the total burden, again due to the large impacts associated with bio-based materials in the CEENE method and due to the consideration of carton-plastic-containers on a higher product refinement stage (as liquid packaging board). All the other fractions together account for 17% of the waste's CEENE, with the share of metals only being 4%. Compared to the overall burden of the scenarios (between 63,450 and 63,510 M_{ex}-eq.), the savings achieved by material and energy recovery are relatively small and amount to 8841 MJ at the minimum (Scenario A) and 9185 MJex-eq. at the maximum (Scenario B), respectively. Most of the savings are due to the production of electricity and heat (between 77% (Scenario D) and 89% (Scenario A)), metal recovery accounts for up to 15% (Scenario B) of the savings, and plastics recovery is responsible for approximately 8% of the CEENE benefit. The digestate utilization as a fertilizer results in relatively little benefits and constitutes only 0.1% of the overall savings achieved in Scenario D. From Fig. 5 it is also visible that the CEENE savings do not increase gradually from Scenario A toward D, but the savings are largest for Scenario B. The material recovery of source-segregated plastics does not result in greater savings than incineration, mainly because waste-derived plastics do not substitute primary plastics on a 1:1 basis, but with a ratio of 0.35. The anaerobic digestion of food waste does not perform better in terms of resource recovery than the incineration in the WtE plant. However, it should be emphasized that the differences in the savings are comparatively small and that the associated uncertainty ranges largely overlap (cf. Tables 5 and SI-17). Therefore and because of the effect of methodological choices on the results, which will be discussed based on sensitivity analyses below, the differences between the scenarios should not be over-stressed.

3.4. Resource efficiencies according to the alternative exergy analysis approaches

The recovery efficiency of a scenario is calculated by dividing the total product output (representing potential savings) by the total input (representing burdens in the system). In Table 5 the efficiencies are shown for each scenario for the exergy flow analysis as well as for the exergetic LCA.

With respect to the exergy flow analysis the recovery efficiency continuously increases from Scenario A (17.1%) toward Scenario D (26.9%). In view of the associated uncertainty ranges, the recovery efficiencies of Scenario C and D are significantly higher than those of Scenario A and B. On the one hand, this is due to the high losses

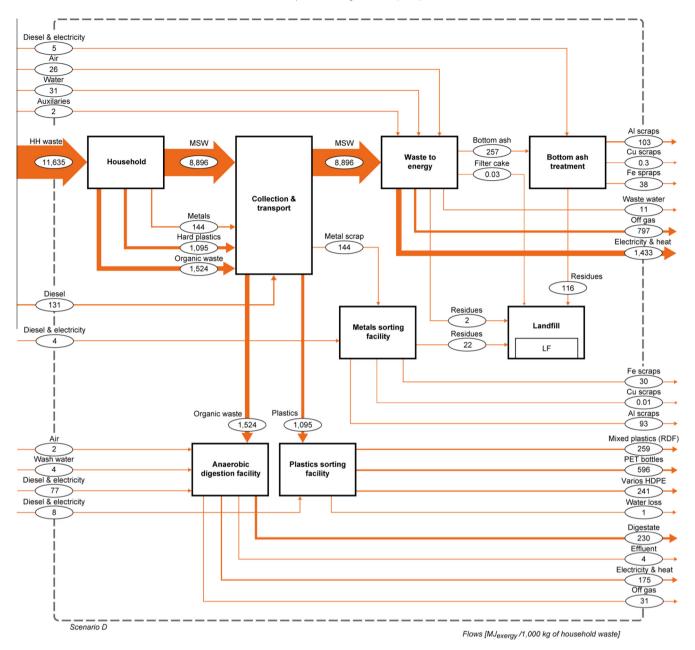


Fig. 3. Exergy flows of Scenario D in MJ_{exergy} per 1000 kg of household waste input (exergy losses are not explicitly shown for the process balances). All exergy flows due to transport and collection are aggregated in one process to avoid individual collection and/or transport processes between each treatment plant.

of exergy during combustion processes, which is the case for all plastic during the residual waste incineration in the WtE plant in A and B compared to the material recycling of PET and HDPE plastics from source segregated plastics waste in C and D. On the other hand, the exergy value of metals is rather low in comparison to combustible materials, which is why the small the slight improvement in metal recovery from A to B (in Scenario A 84% of the Fe input and 55% of the Al input is recovered; in Scenario B 87% of the Fe input and 65% of the Al input is recovered) has only a minor effect. Ultimately, for the exergy flow analysis the recovery efficiencies increase, as a consequence of increasing amounts of recovered material not being directed to combustion processes (within the system). Because the exergy of a flow is a cumulative measure, the value of a flow is higher if more materials are directed toward this flow.

Whereas significant differences in the scenario efficiencies can be observed for the exergy flow analysis, the exergetic LCA results in very similar efficiencies with large overlaps of the associated uncertainty ranges (see Table 5). Therefore, the scenario rankings are rather tentative, with Scenario A exhibiting the lowest recovery efficiency (13.9%) and Scenario B having the highest one (14.5%). For Scenario B, the CEENE scores of the products and services substituted by the metals, electricity and heat are higher than the saved burdens associated with the recycling of source segregated plastics (Scenario C) and the anaerobic digestion of food waste (Scenario D). As noted above, it is clear from the uncertainty analysis, that the differences are very small and not robust given the uncertainty ranges of the scenarios' recovery efficiencies (see Table 5). For all the scenarios the efficiencies are low compared to the flow analysis results, which is primarily due to the huge upstream burden of the waste input in the accounting-level analysis. In terms of CEENE, the resource recovery achieved by the scenarios is limited, because the waste-derive products substitute to a large degree fossil energy intensive processes, but no products

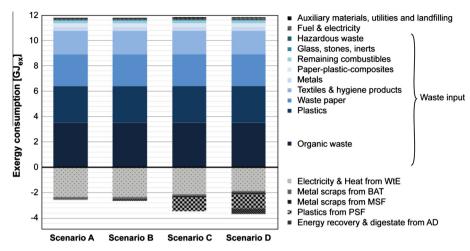


Fig. 4. Comparison of the exergy consumed (positive values) and the exergy recovered (negative values) in the flows of each scenario.

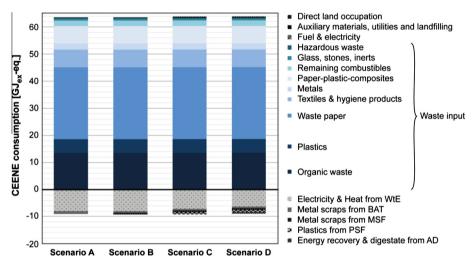


Fig. 5. Comparison of the cumulative exergy consumed (positive CEENE values) and the exergy recovered (negative CEENE values) for each scenario.

Table 5Exergy efficiencies of the scenarios based on the exergy flow analysis (process-level) and the exergetic LCA (accounting-level).

| Range ^a (%) |
|------------------------|
| |
| 12.4-15.5 |
| 12.9-16.0 |
| 12.8-15.7 12.6-15.2 |
| |

^a Uncertainty range is defined as the interval including 95% of the values (from the 2.5 percentile to the 97.5 percentile).

with bio-based production chains. Thus, the burden associated with bio-based products in the input can only be compensated by savings from resource recovery to a small degree, given the substitution pathways of the scenarios.

4. Discussion

4.1. Sensitivity analysis

The uncertainty analysis addressed the energy consumption of waste collection, treatment, and transport, as well as the material

and energy recovery efficiencies of the individual waste treatment plants. Step-wise regression analysis is performed to investigate the effect of uncertain model parameters on the variation of the model results for the exergy flow analysis and for the exergetic LCA (see Fig. SI-2 of the SI). In both cases, the overall energy efficiency of the WtE is the critical parameter with respect to the resource efficiency achieved by an individual scenario. The higher the energy efficiency, the higher the benefits from resource recovery are. In case of the exergy flow analysis, electricity production is much more important for the achieved recovery efficiency than heat production (cf. Fig. SI-2). The higher the share of electricity production, the higher the overall recovery efficiency is. In the exergetic LCA, this is not the case because the CEENE savings achieved by heat production (which reflect the fuels and technologies being off-set) are relatively high compared to the exergy content of the heat flow. Apart from the energy efficiencies, the biogas yield during anaerobic digestion (for Scenario D) and the sorting efficiencies for metal separation, particularly for Al, are also influential parameters. The energy consumption of transport and treatment processes plays a minor role for the recovery efficiencies of the scenarios (cf. Figs. 2, 3 and SI-2). In summary, a scenario's resource recovery efficiency is strongly influenced by the model parameters on energy efficiency of electricity and heat production.

Therefore, these parameters have to be carefully defined in any assessment of waste systems involving energy recovery.

4.1.1. Compounds considered in the exergy flow analysis

A specific focus is put on the sensitivity of the results with respect to methodological choices concerning the representation of resource quality. In terms of the exergy flow analysis, the number of representative compounds constituting the dry matter is doubled for each material flow (see Table SI-14 of the SI). Thereby, the exergy of mixing increases and the chemical exergy content of the compounds changes depending on the specific exergy of the newly considered constituents and the average specific exergy of the original flow. For most flows, the consideration of more compounds has a very limited effect and the flow exergy changes by less than 5%. Only for the solid residues from air pollution control of the WtE plant, exergy increases by around 60% and 120% in case of fly ash and filter cake, respectively (see Table SI-18 of the SI). These increases are the consequence of bulk compounds (SiO₂, CaSO₄) being replaced by compounds with substantially higher specific exergies (e.g. Cd(OH)2, CaCl2). Apart from these compounds, the exergies of most flows slightly decrease because of the higher losses due to mixing and, in some cases, the lower average specific exergy of the flows (e.g. the exergies of plastics flows decrease mainly due to some minor inorganic constituents being considered). Consequently, like for the flow exergies, the overall effect of considering more constituents for each material flow on the scenarios' exergy efficiencies is relatively small (Table 6). For Scenario A and B the efficiencies remain practically the same as in the base case. For Scenario C and D the exergy efficiencies decrease by 0.2% and 0.3%, respectively, because of the lower exergies of the source-segregated organic and plastics waste flows.

4.1.2. Substitution in the exergetic LCA

Critical choices concerning substitution ratios and pathways are investigated for the exergetic LCA. In order to understand the importance of substitution ratios for the scenario rankings, break-even ratios are determined for each pair of scenarios. The break-even ratio is defined as the minimum substitution ratio for a resource flow of a scenario, in order for the scenario to be at least as good as the previous scenario following the order of A, B, C, and D. For the metal scraps at the MSF, the break-even ratio is 0.57, which is 43% lower than the ratio used in the base case. Thus, if 1 kg of metal from the MSF going to secondary production substitutes less than 0.57 kg of metal from primary production, Scenario B would perform worse than Scenario A. For PET and HDPE plastics from the PSF, the break-even ratio is 0.47, which is 34% higher than the substitution ratio of the base case Scenario C. However, the original ratio of 0.35 is already in the upper range of average market price ratios between primary plastics and sorted plastics (cf. Table SI-15 of the SI). For Scenario D, the biogas yield or the energy efficiency of biogas utilization needs to increase by at least 14.3% to outperform Scenario C. Given the relatively high biogas yields

defined for the ADF (cf. Banks et al., 2011; Davidsson et al., 2007), the assumption of such an increase may be overly positive in case of Scenario D. To further test the effect of varying substitution ratios on the results, market price-based substitution ratios are defined for all recovered material flows, except for RDF and digestate due to lack of price data (see Table SI-15 of the SI). Table 6 shows that the physically-based substitution ratios for metals are in good agreement with the market price-based ratios. For scraps from bottom ash treatment market prices point toward slightly higher benefits, whereas for scraps from the sorting of sourcesegregated metals market prices result in a little lower benefits. This may be attributed to the fact, that specific prices for different scrap qualities are not available and the ability of (generic) price data to reflect various scrap qualities is limited (cf. Koffler and Florin, 2013). For Scenario C and D, the differences are more pronounced, because the minimum ratios for PET bottles and HDPE flakes are substantially lower than the ratios used in the base case. Therefore, the minimum efficiencies for Scenario C and D in Table 6 (13.7% and 13.5%, respectively) are also below the efficiencies of all the base case scenarios (cf. Table 5).

Because the identification of marginal technologies to account for product substitution is not without ambiguity given complex and partly unknown market mechanisms, the effect of some alternative choices on the resulting scenario efficiencies is analyzed (see Table SI-19 of the SI for an overview of the alternative substitution pathways considered and their absolute effect on the exergy savings). For all the scenarios the alternative pathways result in substantially lower recovery efficiencies (Table 6, bottom). For Scenario A the alternative substitution pathways result in a recovery efficiency of 8.6% instead of 13.9% in the base case (difference: -5.3%). This is due to the produced electricity and heat replacing the average Danish mix instead of the marginal technologies, the use of Fe scrap to produce chromium steel, which causes Fe recovery to represent a burden instead of a benefit, and the cast Al from Al scrap replacing low-alloyed steel instead of primary Al (cf. Allegrini et al., 2015). The latter results in a lower benefit from Al recycling, because primary Al production is associated with much higher CEENE scores than the production of steel (functional substitution between cast aluminum and low alloyed steel is modeled based on volume, cf. Table SI-19 of the SI). Nevertheless, the use of the average Danish production mix for electricity and heat has the highest effect on the recovery efficiency of Scenario A, because it reduces the benefits from energy recovery by around 30%. For Scenarios C and D, where a part of the plastics are recovered as secondary raw materials, some alternative product substitutions for the plastics result in higher benefits (e.g. cotton fibers instead of PET granulate assuming that recovered PET flakes are used for textiles; global market instead of European production of primary HDPE granulate for secondary HDPE flakes) and others are associated with lower benefits (e.g. HDPE flakes do not substitute HDPE granulate but spruce wood). The minimum and maximum combinations of the alternatives are used to determine the efficiency

Table 6Effect of methodological choices on the scenarios' exergy efficiencies: exergy flow analysis – increase of the number of compounds considered for each material flow (top); exergetic LCA – variation of substitution ratios and pathways (bottom).

| | Scenario A (%) | Scenario B (%) | Scenario C ^a (%) | Scenario Da (%) |
|---|----------------|----------------|-----------------------------|-----------------|
| Exergy flow analysis | | | | |
| Change: higher number of compounds considered | 17.1 | 17.6 | 25.0 | 26.6 |
| Deviation from base case | +0.0 | +0.0 | -0.2 | -0.3 |
| Exergetic LCA | | | | |
| Change: market price-based substitution ratios | 14.0 | 14.2 | 13.7-14.1 | 13.5-13.9 |
| Relative deviation from base case | +0.1 | -0.2 | -0.6 to -0.2 | -0.6 to -0.2 |
| Change: different product substitution pathways | 8.6 | 8.6 | 8.1-9.1 | 7.6-9.1 |
| Deviation from base case | -5.3 | -5.9 | -6.2 to -5.2 | -6.5 to -5.0 |
| | | | | |

^a Minimum-maximum-ranges are given for some results of Scenario C and D, because more than one substitution ratio or pathway alternative was investigated.

Table 7Comparison of the two exergy analysis approaches for evaluating resource recovery from waste.

| | Exergy flow analysis | Exergetic LCA using CEENE |
|---------------------------------------|--|--|
| Similarities | Calculation of exergy requires a reference environment, which cannot Exergy calculations need to be based on detailed data about the component cess characteristics Exergy-based resource evaluations inherently put more weight on fue | osition of flows and conditions (e.g. pressure, temperature) as well as pro- |
| Main strength | Mapping of exergy flows allows for the identification of losses in the investigated system | The cumulated exergy spent to provide a service or product (i.e. the effort in exergetic terms) is well suited to reflect the value of a resource |
| Limitations & challenges | Many assumptions and simplifications are required to express the complex mixture of compounds (and their association) in waste Exergy is cumulative for valuables and contaminants, making its use as a material quality indicator problematic The exergy of mixing is calculated based on highly theoretical assumptions (see Eq. (2)) and contributes very little to the total exergy of (solid) material flows System boundary choices are critical and data availability issues make the inclusion of all relevant processes hardly feasible for more complex waste systems | In the CEENE method bio-based production systems obtain high importance compared to systems mainly relying on fossil energy sources The quantification of upstream burdens of the waste input (necessary for efficiency evaluations) requires many assumptions on processes outside the waste system System boundary choices are crucial, but databases can be used to provide inventory data for processes outside the waste system The choice of substitution ratios and pathways is critical for the results, which is why avoided production needs to be carefully considered |
| Application for efficiency evaluation | For process-level studies: Well delineated systems with clearly defined input and output flows, where energy flows are of primary importance, e. g. evaluation of a waste-to-energy process | As a complimentary analysis to quantify resource consumption and resource efficiency in LCA of waste systems. In particular, for systems where bio-based products play an important role |

ranges of Scenario C and D in Table 6 (see also Table SI-19 of the SI). Finally, for Scenario D another alternative would be the purification of biogas to substitute natural gas in the grid. In the present analysis this alternative results in higher savings achieved by anaerobic digestion of organic waste (cf. Table SI-19 of the SI), but the achieved savings are highly sensitive to the energy needed for biogas purification. In summary, the choice of substitution pathways has to deal with complex markets and has a potentially large impact on the achieved resource recovery efficiency of a waste treatment system. In addition, one has to be careful to account for the functional equivalence between the waste-derived products and alternative production pathways (e.g. volume-based instead of mass-based substitution for construction materials). Therefore, it is important to analyze product substitution and its effect on the results to appropriately reflect the quality of the waste-derived resources in the exergetic LCA.

4.2. Comparison of the exergy analysis approaches used for resource recovery evaluation

The use of exergy as a proxy for resource quality appears to be problematic within the exergy flow analysis for several reasons. First, the lack of direct correspondence between the exergy content of a material and the functionality of the material (cf. Gaudreau et al., 2009). In the exergy flow analysis, this emphasizes the importance of fuels and energy flows, rather than material flows. Second, the cumulative nature of the exergy content of a flow does not allow distinction between contaminants and valuables. Thus, highly contaminated wastes may have exergy values very similar to purified secondary raw materials, although these materials have very different resource quality (cf. Table 7). In addition, the exergy of mixing plays a minor role for exergy flows of solid materials as these exergy values are based on ideal assumptions not correlating with the actual efforts required to separate the mixtures of single particles, composite materials, and alloys (cf. Valero Delgado, 2009). Finally, the delineation of the waste system is problematic, because the utilization of waste-derived products and their effects on the alternative production systems may not be part of the analysis (see Table 7). In such cases, the benefits of resource recovery are not determined based on the utilization of waste-derived products but solely based on their exergy content. For instance, incineration within the waste system is associated with high exergy losses, whereas the losses during energetic uses of waste-derived resources taking place outside the investigated system are not accounted for. In view of the limited knowledge and data about the fate of waste-derived products and recycling processes, the choice of appropriate system boundaries represents a very critical aspect for such a flow analysis.

In this study, the evaluation of resource recovery within the exergetic LCA is done solely based on the CEENE method, which builds on the exergy extracted from the natural environment. Using this method compared to resource indicators not explicitly accounting for land occupation (e.g. CExD, cf. Dewulf et al., 2007), bio-based production systems obtain greater importance relative to systems dominantly relying on fossil energy sources (cf. Table 7). Thus, in a full LCIA, the exergetic resource depletion assessment should be complemented with emission-oriented impact categories, such as climate change. Another challenge related to exergetic LCA, and within LCA in general, is the quantification of substitution ratios and substitution pathways. As these choices are typically not unambiguous, the effect of assumptions about substitution on the exergetic recovery efficiencies needs to be carefully evaluated when applied to concrete case studies. The quantification of the upstream burdens related to the waste input does not change the overall ranking of the scenarios with respect to resource efficiencies, but including these burdens allows the identification of resource losses in the system. Therefore, the assumptions associated with defining the products constituting the waste primarily affect the absolute efficiency levels, not the relative performance of the scenarios.

The discussion of the two exergy analysis approaches for expressing the resource efficiency of residual household waste treatment is summarized in Table 7, where similarities, strengths and weaknesses of the approaches are compared. Based on the comparison, exergy flow analysis turns out to be well suited for evaluating the efficiency of process-level waste studies rather than large-scale waste system evaluations. The exergetic LCA on the other hand, may prove valuable as a complimentary assessment in waste LCA, which can express the resource efficiency of complex waste treatment systems.

5. Conclusions

In this study, exergy analysis is applied to evaluate the resource efficiency of different household waste management scenarios. In case of the exergy flow analysis, scenario efficiencies are around

17–27%, with higher efficiencies going together with increasing amounts of material recycling. This is mainly due to high losses of exergy during waste incineration in the basic scenario. In case of the exergetic LCA, scenario efficiencies are around 14%. The major reasons for the low efficiencies are the high upstream burden of the waste with a large share of bio-based products, which are typically associated with high CEENE scores, and the fact that secondary products mainly substitute products with high fossil energy intensities, but lower CEENE scores. The recovery of metals appears to be beneficial in any case, but is much more important for the overall efficiency in the accounting-level analysis due to the avoided burden associated with primary metal production. Material recovery of plastics on the other hand, appears to be very beneficial in the exergy flow analyses (high losses during incineration are avoided), but adds only little to the resource efficiency in the exergetic LCA, where plastic incineration with high recovery efficiencies is preferable over material recycling. For the exergy flow analysis, this can be attributed to the high exergy content of the plastic wastes, whose (potential) contamination is not reflected by their exergies. Contrarily, the lower quality of waste plastics is reflected by the substitution ratios defined in the exergetic LCA based on average market prices. Thus, the consideration of material qualities is challenging in both types of analyses. For the exergy flow analysis, the main problem lies in the use of exergy content and exergy losses as a proxy for resource quality and resource losses, respectively. For the exergetic LCA, the major challenge is the definition of substitution ratios and pathways, which reflect the actual quality of the recovered resources.

The evaluation of the resource efficiency of complex waste systems based on the analyses of exergy flows within the waste treatment system is constrained by the limited information about secondary production processes and their interaction with primary or traditional production chains. Due to extended system boundaries, the benefits of recovering resources from waste can be better reflected in the exergetic LCA, because the effect of resource provision on production processes outside the waste system is accounted for. The consideration of the upstream burdens associated with the waste, as demonstrated in this study, may form a basis to optimize resource recovery in future waste LCAs. The use of cumulative exergy consumption measures in waste LCA should be complimented by other measures addressing different impacts of waste treatment.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.wasman.2015.09.006.

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