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Bioenergy, material, and nutrients recovery from household waste: advanced material, substance, energy, and cost flow analysis of a waste refinery process

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Abstract

Energy, materials, and resource recovery from mixed household waste may contribute to reductions in fossil fuel and resource consumption. For this purpose, legislation has been enforced to promote energy recovery and recycling. Potential solutions for separating biogenic and recyclable materials are offered by waste refineries where a bioliquid is produced from enzymatic treatment of mixed waste. In this study, potential flows of materials, energy, and substances within a waste refinery were investigated by combining sampling, analyses, and modelling. Existing material, substance, and energy flow analysis was further advanced by development of a mathematical optimization model for determination of the theoretical recovery potential. The results highlighted that the waste refinery may recover ca. 56% of the dry matter input as bioliquid, yielding 6.2 GJ biogas-energy. The potential for nitrogen, phosphorous, potassium, and biogenic carbon recovery was estimated to be between 81% and 89% of the input. Biogenic and fossil carbon in the mixed household waste input was determined to 63% and 37% of total carbon based on ^{14}C analyses. Additional recovery of metals and plastic was possible based on further process optimization. A challenge for the process may be digestate quality, as digestate may represent an emission pathway when applied on land. Considering the potential variability of local revenues for energy outputs, the costs for the waste refinery solution appeared comparable with alternatives such as direct incineration.

Keywords: Bioenergy; Waste refinery; Material flow analysis (MFA); Biogenic carbon; Chemical composition; Cost analysis.

1. Introduction

Within the recent decade, focus on recovery of materials, resources, and energy from solid waste has increased significantly in the endeavor of reducing fossil fuel consumptions and resources depletion [1,2]. Particularly, separation and recovery of the biodegradable fraction of the municipal solid waste (MSW) is encouraged in [1,2] as a mean to produce bioenergy and recycle the nutrients (phosphorous, nitrogen, and potassium as organic fertilizers) on land. In addition, in the regions where landfilling (instead of, for example, incineration) is the most common disposal method, separation of the biodegradable organics (e.g. kitchen waste, tissues, etc.) becomes a necessary priority in order to minimize landfilling and comply with political targets (e.g. [3]). However, although technologies exist for sorting selected waste material fractions, an efficient separation of organic materials, for bioenergy and nutrients recovery, and recyclables, to reduce resource consumption, is difficult with mixed household waste.

Organic waste source-segregation at the household may contribute to this goal; yet, recent studies have highlighted that such a strategy may end up being inefficient (mass- and energy-wise) as a consequence of the losses occurring at the household and during the pretreatments [4]. Therefore, the development of technologies for separating the biodegradable fraction of the municipal waste and optimizing its energy conversion becomes very important. For instance, mechanical-biological treatment (MBT) plants typically use a combination of mechanical operations to separate the organic fraction of the incoming mixed waste from the remaining materials (e.g. plastic, metals, and paper), which are partly recovered (and sent for recycling) and partly mixed to produce refuse-derived fuel (RDF). The separated organic fraction could be anaerobically digested to produce biogas-energy or aerobically stabilized and landfilled [5].

Emerging waste refining technologies provide potential solutions for organic separation and promise improved energy and materials recovery [6,7]. For example, the waste refinery investigated in [6,7] uses enzymatic treatment to produce two outputs from the incoming waste: a bioliquid (liquefied paper, cardboard, and organics) and a solid fraction (undegraded materials). Many of these plants are, however, still in the pilot testing stage, and obtaining a sufficiently high quality of recovered materials is difficult. For instance, in the pilot plant described in [6,7] the post-treatment to separate recyclables needs further development. The post-treatment aims at maximizing the recovery of bioliquid and at sorting recyclables from the solid fraction ex-enzymatic treatment. In the ideal post-treatment all the biomass (and associated biogenic carbon) would be diverted to the bioliquid flow; in other words, all the biomass would be recovered. A number of unit processes may be useful for this purpose; for example, washing, pressing, and sieving with recirculation of the washing liquid into the enzymatic vessel. However, 100% efficiency is not realistic and some biomass would still be found in the solid fraction as undegraded organics (e.g. shells), paper, and textiles.

With regard to documenting the development potential, simple sampling at such preliminary facilities cannot provide data appropriate for full-scale implementation of the technologies. From this perspective, material-, substance-, and energy-flow analysis (MFA, SFA, and EFA) are useful techniques to assess mass, energy, and substance flows in a range of different urban systems (e.g. waste management, bioenergy, urban metabolism, etc.), including evaluation of the quality of the recovered resources [8]. In the specific context of waste management MFA and SFA are often utilized to highlight the fate of valuable materials and substances and to further suggest system

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improvements on the basis of the results. Further, the results of MFA and SFA are often used as a basis for life-cycle assessment (LCA). From this perspective, MFA, SFA, and LCA represent complementary tools for environmental management [9]. For instance, [10] used MFA to identify the relevant waste flows in a waste-emergency area and to suggest management solutions; [11] combined MFA and LCA to assess the performance of a garden waste composting plant; [12] and [13] used MFA and SFA to estimate flows and recycling efficiencies for electronic waste; [14] modelled the energy content of solid recovered fuel (SRF) based on MFA. [15]) combined SFA and LCA to assess the performance of bioenergy scenarios. However, in addition to mass and substance flow analyses, in order to address the theoretical performance of pilot-scale waste refineries, mathematical modelling needs to be applied to determine the potential optimum recovery of bioliquid, materials, and nutrients, thereby providing a target for further technological development. Mathematical optimization has been extensively used in studies about waste, bioenergy, and waste-to-energy in order to evaluate potential technology performances, limitations, and associated improvement potentials. Among the others, mathematical optimization modeling was applied to evaluate potential performances and limitations of waste- and biomass-to-energy systems (both thermal and biological) [16-23] and also to evaluate potential optimal solutions for maximizing energy and environmental savings in wastewater treatment [24,25], industrial production [26-28] and waste management strategies [29-31].

This study used an advanced MFA, SFA, and EFA approach based on a mathematical optimization model to evaluate the potential flows of materials, substances (e.g. carbon, nutrients, and selected metals), and energy within a waste refinery including downstream energy conversion processes. The objectives of the study

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were: i) a detailed sampling and characterization of the outputs of a pilot-scale waste refinery process (materials flow and chemical composition) with particular focus on the bioliquid; ii) the development of a mathematical optimization model to evaluate the potential for recovery of bioliquid, materials, and nutrients with a 'virtual' post-treatment phase; iii) the development of MFA, SFA, and EFA models based on the mathematical model outputs to illustrate the potential flows of materials, energy, carbon (including fossil carbon, i.e. C_{foss}), nutrients, and selected metals (Al and Fe); iv) the evaluation of the quality of the digestate left after anaerobic digestion of the bioliquid in order to assess the load of nutrients and metals in the scenario of application on land; v) the estimation of the costs of the waste refinery solution compared with alternative waste management systems.

2. Materials and methods

The study involved five major phases: 1) On-field sampling of the pilot-scale waste refinery outputs (bioliquid, fluff, and solid fraction ex-enzymatic treatment); hand-sorting of the solid fraction was also performed at this point: six individual waste material fractions were sorted and separated (see 2.2). Thus, in total, eight waste material fractions were collected (six from the solid fraction ex-enzymatic treatment plus bioliquid and fluff). 2) Preparation of the eight individual samples for chemical analyses (shredding, mixing, splitting, etc.). 3) Chemical composition analyses (including calorific value). 4) Elaboration of a mathematical optimization model to estimate the potential for bioliquid, materials, and nutrients recovery with a 'virtual' post-treatment. 5) Elaboration of MFA, SFA, and EFA to illustrate material, substance, and energy flows within the waste refinery process including virtual post-treatment and

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downstream energy and materials recovery processes. These flows were also used as basis for the cost analysis. Table 1 summarizes the five phases of the study with the associated methods applied.

Table 1

2.1 The waste refinery process

The study was based on the operation of a pilot-scale plant (0.5–1 Mg wet waste (ww) h^{-1}), where the waste was processed (heating and enzymatic treatment) without further post-treatment. The pilot-scale plant treated residual municipal solid waste (rMSW) collected from a residential district of Copenhagen (Denmark) where a vacuum-collection system is established. The waste was sampled and characterized within this study (as the output of the waste refining process; see section 3).

The waste refinery aims at producing two products from the incoming MSW: i) a bioliquid (i.e. slurry composed of enzymatically liquefied organics, paper, and cardboard) and a solid fraction (i.e. non-degradable waste materials). The refinery process consisted of two reactors: in the first reactor the waste was heated to about 75 °C for approximately 0.5–1 hour and then cooled to about 50–55 °C before entering the second reactor. In the second reactor selected enzymes were added (ca. 5 kg enzymes Mg^{-1} wet waste corresponding to ca. 7 kg enzymes Mg^{-1} dry waste), resulting in hydrolysis and breakdown of bonds in the organic materials, thereby essentially suspending organic materials in a liquid phase. The retention time was about 10–16 hours. A detailed description of the enzymatic processing can be found in [32]. After the second reactor, the liquid phase was separated from the remaining solid fraction by a

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vibrating sieve. Later, another vibrating sieve separated the liquid phase into a bioliquid and a solid "fluff" (phase containing materials such as cotton and textiles but also glass pieces, plastics, etc.). The bioliquid consisted primarily of suspended organic matter (food waste, paper, and cardboard), while the solid fraction mainly consisted of non-degradable materials such as plastic, metals, textiles, soil, ceramics, and glass pieces mixed with unseparated bioliquid. The solid fraction and the fluff require post-treatment to recover additional bioliquid (through, for example, washing and pressing).

The bioliquid can be exploited for biogas production (an option considered in this study), co-combusted in a coal-fired power plant, or utilized for producing ethanol [6,7]. Biogas production, as compared with direct incineration, provides additional flexibility to the energy system as the energy production can be regulated and storage is possible. This is crucial from the perspective of energy systems with high penetration of wind and other fluctuating renewables as illustrated in previous studies [33,34]. The solid fraction can undergo further sorting to recover valuable materials such as metals and plastic. The remaining residual solids (mainly non-recyclable plastic, textiles, yard waste, other undegraded organics and paper, and glass pieces) can be combusted for energy recovery. A modeling-based characterization of the residual solids fraction is reported in [7] based on typical Danish waste composition data. The present study also reports data on the composition and heating value of the waste materials constituting the residual solids (section 3) for the specific case of the MSW treated at the pilot-plant under assessment. The electricity and heat consumption for the operations of heating, enzymatic treatment, and sieving were about 20 kWh Mg^{-1} ww and 490 MJ Mg^{-1} ww, respectively. Additional information about the process and its environmental performance can be found in[6,7].

2.2 On-field sampling of the waste refinery outputs (phase 1)

The three outputs from the pilot-scale waste refinery (Fig. 1), that is, 1) bioliquid (named BL*), 2) fluff (named FF*), and 3) solid fraction (named SF*) were sampled during four days. Individual samples were collected regularly from each output flow at 20 minutes intervals for a nine-hour duration on each day of the sampling campaign. All individual samples during a single day were combined to one sample representing this day. In total, four samples for each output flow were obtained during the sampling campaign.

The following individual waste material fractions were then hand-sorted from the daily samples of SF*: i) hard plastic (e.g. HDPE), plastic containers, plastic bottles, and so on (named HP*); ii) soft plastic and plastic bags (named SP*); iii) textiles (named TXT*); iv) ferrous metals (named FE*); v) non-ferrous metals (named NFE*); vi) residue (named RES*) containing other plastic items, glass scraps, partly degraded and non-degraded organics (e.g. shells, peels), paper, and cardboard, and other unidentifiable material. The individual waste material fraction samples were weighted and subsequently combined with corresponding samples from the other days in order to represent the entire sampling period of four days. A similar procedure was followed for the other two output flows (BL* and FF*). The samples obtained for chemical analysis then represented an average over the full sampling campaign.

It has to be noticed that enzymes were added during the process (ca. 7 kg enzymes Mg^{-1} dry waste corresponding to ca. 3.9 kg enzymes dry matter added); therefore the sampled and analyzed outputs also included the mass contribution from the enzymes added. However, this additional contribution was very small compared with

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the total mass flow (i.e. 0.4%), and it can therefore be reasonably neglected when discussing the overall mass flows and chemical composition.

Fig. 1

2.3 Samples preparation (phase 2)

The hand-sorted waste material fractions were sub-sampled using the method described by [11,35] in order to obtain about 10 kg of representative samples. These were dried at 105 °C for 24 hours to establish the moisture content. For the bioliquid, drying was performed at 70 °C for ca. 72 hours to avoid losses. Comminution of the waste material fractions was done with a number of appropriate instruments depending on the specific waste material fraction whose particle size was to be reduced: cutter mill (Retsch SM 2000, Haan, Germany), shredder (ARP CS 2000 shredder), and rotary disc mill (Siebtechnik IS100A, Mülheim an der Ruhr, D). The overall procedure was similar to the one described by [35,36]). In detail:

- Dried bioliquid (BL) was shredded with the Retsch SM 2000 cutter mill down to a size of 1×1 mm.

- Dried hard and soft plastic and textiles (HP*, SP*, and TXT*) were first manually cut into smaller pieces and further treated in the Retch SM 2000 cutter mill down to a size of less than 1×1 mm.

- Dried ferrous and non-ferrous (FE* and NFE*) materials were first shredded with the ARP CS 2000 shredder, pressed into cubes, and mass-reduced by titanium drilling and the obtained scraps were further mass-reduced with the disc-mill (Siebtechnik IS100A, Mülheim an der Ruhr), down to a size of less than 1×1 mm. This laborious method was chosen because these fractions

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could not be directly cut, shredded, or crushed due to their strength.

- Dried residue (RES*) was shredded with the Retsch SM 2000 cutter mill down to a size of less than 1×1 mm.

- Dried fluff (FF*) was shredded with the Retsch SM 2000 cutter mill down to a size of less than 1×1 mm.

During all operations, large amounts of pulverized dry ice ($CO_{2,S}$) were added during handling to ensure sufficient cooling capacity and to facilitate the shredding/milling. Mixing and fractional mass reduction were done in the same way for all fractions by repeated mixing in a mechanical mixer (or by hand) and then mass-reduced with a riffle splitter (Rationel Kornservice RK12, Esbjerg, Denmark) until the mass required for chemical analysis was obtained.

2.4 Chemical composition analyses (phase 3)

Volatile solid (VS) content, chemical composition, and calorific value (higher heating value, HHV) were determined by standard analyses following the approach described in [36] specifically for solid waste material fractions. Volatile solid content was measured by incinerating the dried ground samples at 550 ℃ in a muffle. The elements C, H, N, and S were analysed by elemental analysis (GE-MA M-7-1); Cl and F were analysed in conformance with DIN 51727 B; P was analysed by inductively coupled plasma optical emission spectrometry (ICP-OES) in conformance with DIN EN ISO 11885 (E22). The remaining metals (i.e. Fe, Al, As, Cd, Cr, Cu, Fe, Hg, Mn, Mg, Ni, Pb, Sb, and Sr) were analysed by ICP-MS in conformance with DIN EN ISO 17294-2 (E29). The HHV_{db} (dry basis) was determined with a calorimetric bomb in conformance with DIN 51900. The content of biogenic carbon (C_{biog}, represented by the ^{14}C content in ^{12}C) was

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analysed by accelerated mass spectrometry (AMS) in conformance with CEN/TS 15747:2008. The theoretical methane potential of the bioliquid ($B_{o,th}$) was estimated by applying Buswell's formula (Eq. 1), which provided the composition of the bioliquid in terms of *carbohydrates*, *proteins*, *lipids*, volatile fatty acids (*VFA*), and ethanol (*EtOH*) determined with high performance liquid chromatography (HPLC). In addition, batch assays were also conducted according to the procedure described in [37]. The batch assays were conducted in 500 mL serum bottles inoculated with digested manure from thermophilic digestion plants and fresh bioliquid. Tests were run in triplicates for 52 days at 55 ºC [38].

$$B_{0,th} = 0.415 \cdot carbohydrates + 0.496 \cdot proteins + 1.014 \cdot lipids + 0.373 \cdot VFA + 0.73 \cdot EtOH$$
(1)

2.4.1 Chemical composition of the digestate

The composition of the digestate was determined through i) SFA modelling and ii) chemical analyses of actual samples from full-scale anaerobic digestion of the bioliquid. The first approach was based on the chemical composition of the sampled bioliquid (BL*), assuming that heavy metals (as well as phosphorous and potassium) were entirely transferred to the digestate, and modelling expected dry matter degradation occurring during the digestion (see section 2.6 and Supporting Information, SI); the second approach provided instead a 'snap-shot' of the chemical composition based on actual digestate samples representing a specific point in time. Selected parameters and substances (e.g. dry matter (DM), volatile solids (VS), C, H, N, P, K, other metals, etc.) were analysed. The substances C, N, and H were analysed by Isotope Ratio Mass Spectrometry (ANCA-SL-GSL). The metals (including P and K) were analysed by ICP OES in conformance with EN 13346:2000. The measured VS, C, and N contents were

used to calculate the expected DM, C, and N degradation occurring during the anaerobic digestion process (see section 2.6 and SI). In addition, the analyses of the metals provided a set of concentration values that were compared with the SFA modelling results and further used as a basis for evaluating the quality of the digestate in the scenario of application on land (see section 3.4).

2.5 Mathematical optimization model (phase 4)

2.5.1 Application of the mathematical model

The results of the chemical analyses provided the composition of the waste material fractions (BL*, HP*, SP*, TXT*, FE*, NFE*, RES*, and FF*) ex-enzymatic treatment. This did not include post-treatment of the solid fraction (which includes HP*, SP*, TXT*, FE*, NFE*, and RES*) and the fluff (FF*) to maximize bioliquid recovery. An optimization model has been elaborated to model the post-treatment phase; the 'Post-treatment' box in Fig. 1 is thus a virtual process representing the optimum recovery rates (in other words, the theoretically obtainable separation between bioliquid and solid materials). The mathematical modelling was facilitated by the software MATLAB. The results of the chemical analyses (phase 3 of Table 1), along with the measured (dry) mass flows of BL* and SF*, were used as input to the optimization model. For the modelling, it was assumed that all the bioliquid (BL) mixed with the solid fraction (which includes HP*, SP*, TXT*, FE*, NFE*, and RES*) and with the fluff (FF*) should in principle end up in the dedicated bioliquid flow (BL) after post-treatment. The model assumed that the bioliquid composition did not change with the post-treatment (it is envisaged that cold water will be used and no chemicals will be added). As mentioned above, ideally the biomass (indicated by the biogenic carbon) should only be found in

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the bioliquid flow. However, 100% efficiency is unrealistic as part of the biomass does not degrade. This translates into imposing that the concentration of C_{biog} 'tends' towards zero in materials other than bioliquid (see 2.5.1 and SI); this provided a realistic estimate of the potential for bioliquid recovery (i.e. overall amount recoverable) along with mass flows and chemical composition data for the waste material fractions which are undergoing 'post-treatment' (i.e. HP, SP, TXT, FE, NFE, RES, and FF) and have thus been 'cleaned' from the bioliquid. Fig. 2 illustrates the principles of the optimization model.

Figure 2

2.5.2 Formulation of the mathematical model

The mathematical formulation consisted of two parts: i) mass balance and ii) uncertainty handling. In the second part (ii) the uncertainty in the model input variables is first characterized by means of Probability Distribution Functions, and then it is propagated onto the results. A detailed description of the approach used to elaborate the uncertainties is reported in the Supporting Information (SI). The mass balance (i) consists in breaking down the total dry mass flow into bioliquid and material flow and determining the concentration of chemicals in the material flow. Input data are: i) the total (dry) waste materials flow, ii) the total chemicals flow, and iii) the bioliquid chemicals flow. All flows are known from sampling and analyses (Table 1). First, the chemical concentrations in the material flow are determined by solving, for each flow, a simple optimization problem. Then, the bioliquid and material component of the flow is calculated immediately from the chemicals (an overview of the mathematical model is presented in Fig. S1 of the SI).

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The model considered $N=8$ waste material fractions (BL*, HP*, SP*, TXT*, FE*, NFE*, RES*, FF*) and $K=13$ major chemical elements. The major chemicals considered were C_{foss}, C_{biog}, H, S, N, F, P, Cl, K, Fe, Al, Mg and $O_\&$. The parameter $O_\&$ aggregated oxygen and the remaining minor elements (e.g. heavy metals in traces) so that the sum of the twelve major chemicals (just listed) and $O_\&$ added up to 100%. The minor elements (e.g. heavy metals in traces) were aggregated with oxygen in one parameter as their overall contribution to the total mass was very small (about 1%). The dry mass flows (named P_i) of the $i=1,..,N$ waste material fractions were the sum of two components: the bioliquid component BL_i and the 'cleaned' waste material fraction M_i (i.e. cleaned from bioliquid, under some constraints, which are: HP, SP, TXT, FE, NFE, RES, and FF as shown in Fig. 2):

$$P_i = BL_i + M_i \quad i = 1,\ldots,N \tag{2}$$

The flows P_i represented fractions of the total dry mass. Hence they are positive, with unitary sum:

$$\sum_{i=1}^{N} P_i = 1$$
$$BL_i \geq 0, M_i \geq 0 \quad i = 1,\ldots,N \tag{3}$$

The variables CP_{ji}, CBL_j, CM_{ji} represented the concentration of the chemical $j=1,..K$, respectively within: 1) total (dry) mass flow P_i (i.e. bioliquid plus material, ex-enzymatic treatment), 2) (dry) bioliquid flow BL, 3) and (dry) material flow M_i (cleaned from bioliquid). The mass balance of the j-th element in the i-th waste material was described as follows:

$$CP_{ji}P_i = CBL_j BL_i + CM_{ji}M_i \tag{4}$$

As concentrations (or 'fractions'), they fulfilled the conditions similar to Eq. (3), i.e.:

$$\sum_{j'=1}^{K} CP_{j'i} = 1 \quad CP_{ji} \geq 0 \quad \forall i, j$$

$$\sum_{j'=1}^{K} CBL_{j'} = 1 \quad CBL_{j} \geq 0 \quad \forall i, j$$

$$\sum_{j'=1}^{K} CM_{j'i} = 1 \quad CM_{ji} \geq 0 \quad \forall i, j$$

(5)

Notice that CBL_{j} was denoted without i-index, as it was assumed that the chemical composition of the bioliquid was the same in all waste materials N=1,...,8.

In what follows, it is presented a methodology to determine BL_{i}, M_{i}, CM_{ji}, given P_{i}, CP_{ji}, CBL_{j}, for all i, j. The total number of unknowns was $N(K+2)$, whereas the conditions were N linear of the type of Eq. (2), and NK non-linear of the type of Eq. (4), for a total of $N(K+1)$ conditions. The problem was lacking N equations, therefore it was underdetermined. The missing conditions were formulated based on a priori knowledge of the distribution of specific chemicals within the materials. In order to apply the earlier described methodology to the mathematical problem, selected chemicals j must be in common to all waste material flows. One of these chemicals was the biogenic carbon (C_{biog}). The biogenic carbon was always among the K considered elements, and in theory it was expected to be found only within the bioliquid component (BL_{i}) of the flows. This, in theory, should provide the N missing conditions: let $CM_{biog,i}$ be the share of the biogenic carbon within the i-th waste material flow, then i.e. $CM_{biog,i} = 0$ for all materials i. In reality, however, small concentrations of biogenic carbon in the material flows are possible, as afore mentioned. Imposing strict equality conditions may not reflect the reality, and therefore it is likely to result in an unfeasible system of equations (e.g. substance balances not respected). A more robust approach would be to let $CM_{biog,i}$ be positive, *but small*, namely to solve the optimization problem:

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$$CM_{biog,i} \rightarrow \min_{P_i, CP_{ji}, CBL_j} \tag{6}$$

Subject to the constraints of Eqs. (2-5).

Such problem is of simple solution, as it can be broken down into N independent sub-problems, by introducing a term $0 \le \lambda_i \le 1$ such that:

$$BL_i = (1 - \lambda_i)P_i \quad M_i = \lambda_i P_i \tag{7}$$

And by combining Eq. (2), Eq. (4), and Eq. (7), to obtain:

$$\lambda_i CM_{ji} + (1 - \lambda_i)CBL_j = CP_{ji} \tag{8}$$

For each material i, the λ_i term and the share of the chemicals CM_{ji} for all $j = 1, \dots, K$, were obtained by solving the problem:

$$
\begin{aligned}
CM_{biog,i} &\rightarrow \min_{0 \le \lambda_i \le 1} \\
&\text{subject to:} \\
CM_{ji} &= \frac{CP_{ji} + (\lambda_i - 1)CBL_j}{\lambda_i} \qquad j = 1, \dots, K \\
CM_{ji} &\ge 0
\end{aligned}
\tag{9}
$$

which was a single-variable optimization framework, and it could be efficiently computed numerically, e.g. using golden section search and parabolic interpolation [39]. The terms BL_i, M_i were then immediately derived using Eq. (7). The entire analysis was performed by solving Eq. (9) for every material $i = 1, \dots, N$.

The determination of the material fraction flows M_i allows calculating the higher heating value, dry basis, (HHV$_{db}$) of the waste material fractions M_i as of post-treatment (i.e. 'cleaned' from bioliquid) according to Eq. (10).

$$P_i \cdot HHV_{db,P_i} = M_i \cdot HHV_{db,M_i} + BL_i \cdot HHV_{db,BL}$$

(10)

Where the only unknown is $HHV_{db,Mi}$ (MJ kg DM^{-1}). Both $HHV_{db,Pi}$ and $HHV_{db,BL}$ are known from analyses (Table 2).

2.6 Material, substance, and energy flow analysis (phase 5)

The results of the mathematical optimization model (BL_i, M_i, and CM_{ji}) were used as inventory for the MFA, SFA, and EFA of the waste refinery process. This was facilitated by the software STAN [40]. The MFA encompasses the waste refinery process itself (including the modelled post-treatment) and downstream material separation and energy conversion (i.e. anaerobic digestion of the bioliquid and incineration of the residual solids). A thorough description of the assumptions and input data used in the MFA, SFA, and EFA (in addition to the results of the mathematical model) can be found in the SI (i.e. transfer coefficients of DM and chemicals to flue gas and biogas, methane yield and energy conversion efficiencies for biogas conversion and incineration).

2.7 Cost analysis

A cost analysis was performed to compare the waste refinery-based system with direct incineration. The geographical and temporal scope of the analysis was Denmark, year 2013. The analysis included costs for collection, treatment, and disposal of residues, including eventual revenues associated with sales of electricity, heat, and recyclables as well as national taxes and subsidies. Externalities were not included.

The following two scenarios were assessed: A) collection of all municipal household waste without source segregation and waste refinery treatment (CHP production from the bioliquid-biogas and incineration of the residual solids fraction for

CHP production; separation and recycling of hard plastic and metals). B) Collection of all municipal household waste without source segregation and direct incineration for CHP production.

The price for electricity and heat was based on current Danish sale market prices (0.035 and 0.042 € kWh^{-1}, respectively). For recyclables and enzymes, they were based on recent World market trends (see SI). Notice that the Danish national CO_2 tax on incineration (ca. 22.3 € Mg^{-1} CO_2 emitted), on heat produced from incineration (ca. 8 € GJ^{-1} heat sold), and the national subsidy for biogas-electricity (ca. 0.047 € kWh^{-1}) were included in the analysis.

Three sensitivity analyses were performed to test: S1) the influence of heat and electricity prices (and related revenues) on the total cost; S2) the influence of hard plastic management on the total cost; S3) the influence of the electricity recovery efficiency at the incinerator. A detailed description of the background data used for the analysis, of the calculation of treatment and collection costs, and of the sensitivity analyses is reported in the SI. Results are discussed in section 3.6.

3. Results and discussion

3.1 Chemical composition

The chemical composition of the input waste to the facility was determined based on substance balance calculation from the chemical composition of the individual waste material fractions sampled (Table 2). For the purpose of comparison, the chemical composition of the rMSW as determined by [35] for Danish households is also reported. The concentration of organic substances such as carbon, hydrogen, nitrogen, phosphorous, potassium, fluoride, chloride, iron, aluminium, and magnesium was very

close to the values reported in the literature. This was also true for the higher and lower

heating values on a dry basis (HHV_{db} and LHV_{db}). The content of fossil carbon (37% of

the total C, i.e. 63% of the total C was of biogenic origin) was in accordance with the

measurements carried out at a range of Danish full-scale waste incinerators treating

similar waste [41]. However, except for nickel, the concentration of the other heavy

metals in the input waste was found to be significantly lower than in [42] and in similar

studies on MSW [43]. The most likely reasons for these differences are: i) differences in

waste composition (the analysed waste is only representative of a specific residential

area with a specific collection system) and ii) the intrinsic difficulties in representative

sampling of heavy metals. In this respect, findings from [44] suggest that analyses of

heavy metal contents in waste can be more accurate if the grain size of the samples is

reduced to below 0.5 mm. It is likely that the concentrations of these metals were

therefore underestimated. However, this did not affect the modelling results as, overall,

these chemicals contribute only a small quantity (about 1–2%) to the total mass. The

composition of the waste material fractions as calculated by the mathematical model

(i.e. after 'virtual' post-treatment) is reported in Table 2. This clearly differed from the

chemical composition of the waste materials as sampled since it was calculated

assuming a 'virtual cleaning' of the bioliquid fraction (Fig. 2). The major differences lie

in the content of organic substances (C, H, N, P, K, Cl, F, Al, Fe, Mg) as their presence

in the sampled waste materials was due to the contamination with unseparated bioliquid.

The results of the batch assays indicated an average methane potential of the

bioliquid of 443 NL CH_4 kg^{-1} VS (±21 NL CH_4 kg^{-1} VS) [38]. This is in accordance

with the theoretical value (445 NL CH_4 kg^{-1} VS) calculated with Buswell's formula (Eq.

1) based on the measured content of carbohydrates, proteins, lipids, VFA, and ethanol in

the bioliquid (see values in the footnote of Table 2). Notice that the CH_4 production

measured in full-scale tests (365 NL kg^{-1} VS, as mentioned earlier) corresponded to a

yield of about 82%. Yet, based on Buswell's formula, the stoichiometric ratio of CH_4 to

CO_2 in the biogas can be quantified (0.52, volume basis).

Table 2

3.2 Material and energy flows

The material and energy flows are reported in Fig. 3. Uncertainties related to relevant

flows are mentioned in brackets (e.g. ± 'value') as standard deviations of the mean

values. Notice that the values reported in the text are expressed per 1 Mg dw (dry

weight), unless otherwise specified.

All in all, more than half of the dry matter (56%) can potentially be recovered

into the bioliquid (BL) flow as liquefied organic and paper material (see Fig. 3). This

embedded 8.9 GJ of primary energy (based on LHV_{db}), of which about 70% could be

transferred to methane (6.2 GJ) based on a yield of 82% relative to the measured

methane potential. This resulted in an ultimate recovery of 690 kWh (±120) of

electricity and 3,100 (±540) MJ of heat (Fig. 3). If the focus is on optimizing biogas

production from a unit-input of mixed MSW, then the overall potential is significantly

increased compared with other pre-treatment technologies that only separate the organic

fraction such as MBT. The reasons for this are: 1) not only organic waste, but also paper

and cardboard are used (in the form of bioliquid) for biogas production, thus increasing

the total production per unit of waste input; 2) the higher methane potential and

associated yield of the liquefied paper and cardboard in the bioliquid (ca. 365 NL CH_4

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kg^{-1} VS corresponding to 310 NL CH$_4$ kg^{-1} DM) compared with typical yields (ca. 150

NL CH$_4$ kg^{-1} VS) for non pre-treated paper and cardboard [45]; 3) The biogas-energy

production in MBT may be limited by low quality of the organic substrate separated as

illustrated in [46,47]. Notice that our results concerning dry matter recovery into the

bioliquid are largely in agreement with the findings of [32].

The hard plastic and soft plastic items (HP and SP) constituted about 8% and

9%, respectively, of the total dry weight. The energy content equalled 2.8 and 2.6 GJ,

respectively. Textiles (TXT) represented 7.6% of the dry weight, carrying 1.3 GJ of

primary energy. Ferrous (FE) and non-ferrous (NFE) materials made up 4.1% and 2.4%

of the total dry weight, respectively, adding up to ca. 0.95 GJ of primary energy. The

residue represented about 12% of the overall dry matter, embedding about 2 GJ. Lastly,

the amount of fluff (FF) screened prior to digestion of the bioliquid was ca. 0.4% of the

total dry weight, containing about 0.0042 GJ. Based on the results, the total amount of

primary energy contained in the residual solids (consisting of SP, TXT, and RES) and

potentially available for combustion was 5.9 GJ, corresponding to a LHV$_{db}$ of about 21

GJ Mg^{-1} DM. This resulted in a gross recovery of 410 ($\pm$88) kWh Mg^{-1} DM of

electricity and 4,400 ($\pm$950) MJ Mg^{-1} DM of heat. If HP was incinerated (instead of

separated and recovered), the primary energy available for combustion would be raised

to ca. 8.7 GJ, corresponding to a LHV$_{db}$ of about 24 GJ Mg^{-1} DM. This would equal a

production of ca. 600 ($\pm$100) kWh Mg^{-1} DM of electricity and 6,500 ($\pm$1,100) MJ Mg^{-1}

DM of heat, and these values are 46% higher compared with the scenario involving HP

separation. However, incineration of this fraction would also induce additional direct

CO$_2$ emissions at the stack (see section 3.3).

As previously discussed (section 2.2), the contribution of the enzymes to the

total mass flow was ca. 0.4% (i.e. negligible compared with the total waste flow).

Fig. 3

3.3 Substance flows

The substance flows for C, C_{foss}, N, P, K, Fe, and Al are illustrated in Figs. 4–6. These are the most important substances with respect to biogas (C), nutrients (N, P, and K), and materials recovery (Fe, Al, and C_{foss} for plastic). Heavy metals flows (e.g. Cd, Ni, Pb, Cu, etc.) are also important for the use on land of the digestate left from anaerobic digestion. In this respect, *ad hoc* characterization of the digestate was performed (see section 3.4). Notice that the values reported in the text are expressed per 1 Mg dw, unless otherwise specified.

Most of the carbon (Fig. 4) was found in the bioliquid flow (240 kg), of which almost 100% was biogenic carbon (Fig. 4). Based on the mass balances detailed in the SI (Eqs. S2–S5), about 75% of this carbon was biogasified during anaerobic digestion, which reduced the carbon left in the digestate to about 60 kg. As discussed earlier, ideally, all the biogenic carbon should be found in the bioliquid flow. However, this is unrealistic as part of the biomass does not liquefy. Based on the results (Fig. 4), it can be estimated that the technical potential recovery (i.e. biogenic carbon in the bioliquid flow over the total biogenic carbon in the waste input) may be as high as ca. 81% (±11%) of the theoretical maximum (i.e. the total biogenic carbon found in the input waste), and that the remaining biomass can mainly be found in the flows of textiles, residue, soft plastic, and fluff. The contamination of these materials from the biomass can be seen from Table 2 based on the presence of biogenic carbon. In fact, it is largely expectable that part of the textiles, even if of biogenic origin, is not entirely liquefied. The residue flow, as defined in section 2.2, contains partially- and non-degraded organic

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materials (e.g. shells, peels). Further, part of the biomass may still be stuck to plastic bags or found into unopened bags (there was no shredding in the process). Finally, fluff contains, among the others, cotton and textiles (thus partly composed of undegraded biomass).

Significant fossil carbon flows (Fig. 4) were embedded into HP (65 kg), SP (49 kg), and residue (40 kg). Overall, under the assumption that hard plastic was separated and recovered, 95 kg of 'residual' fossil carbon were incinerated, corresponding to a direct emission of ca. 350 kg CO_2. In other words, sorting and recovery of HP avoided ca. 240 kg of direct CO_2 emission. Additional direct CO_2 emission savings may be achieved by separating and recovering SP (potential: 180 kg CO_2) and eventually other plastic items found in the residue.

As mentioned in section 2.2, enzymes were added in the process and therefore the analyzed carbon flows also included the contribution from the enzymes added. However, as reported in section 2.2, this contribution can be reasonably neglected as it is very small compared with the total waste flow. In terms of carbon, the contribution from the added enzymes corresponded to ca. 0.5% of the total carbon and to ca. 0.83% of the total biogenic carbon (based on a carbon content of the enzymes equal to 62%, dry basis).

About 85% of the nitrogen (i.e. 11 kg) was found in the bioliquid flow (Fig. 5), of which 87% (i.e. 9.7 kg) ended up in the digestate. A minor amount of nitrogen was also found in the residue, textiles, and soft plastic sent for incineration (overall ca. 0.8 kg) and in the biogas (1.4 kg). The phosphorous (Fig. 5) was primarily collected in the bioliquid flow (about 1.4 kg, that is, 83% of the total), although a share was incinerated through textiles, residue, and SP (overall 0.33 kg). As for N and P, the K (Fig. 5) was

also recovered mainly in the bioliquid flow (about 3.9 kg, i.e. 89% of the total).

As expected, almost all of the iron (Fe) and aluminium (Al) were found, respectively, in the ferrous (about 33 kg, i.e. 85% of the total) and non-ferrous (about 17 kg, i.e. 77% of the total) material flows (Fig. 6). Both ferrous and non-ferrous materials contained Fe and Al. Iron and aluminium were also found to a minor extent in residue and bioliquid flows.

Fig. 4

Fig. 5

Fig. 6

3.4 Characterization of the digestate

The quality of the digestate left after anaerobic digestion is of particular concern because of the opportunity to apply this product on agricultural land, thereby recycling the nutrients (N, P, and K). This management strategy, however, is strongly dependent on the content of heavy metals and other hazardous chemicals (e.g. PAH, phthalates, etc.), which might constitute an impediment to use on land. Table 3 illustrates the results of the two approaches used to characterize the digestate (section 2.4.1). In addition, the Danish limit values for application of digestate on land [48] and selected literature values illustrating the typical composition of digestate obtained from processing source-segregated organic waste [49,50] are reported for the purpose of comparison.

The analytical concentrations of most of the assessed chemicals (C, N, P, K, Fe, Al, Sr, Mn, Mg, Cr, Ni, Hg, Pb) were similar to the corresponding modelled SFA results. Some differences were found for Fe, Cu, and Cd, whose modelled

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concentrations were slightly lower than the analysed values. For heavy metals, the results of the analyses showed that the concentrations of Cd and Ni exceeded the selected limit values (mg per kg of total P). For Ni, this was also confirmed by the SFA results. The concentration of the other heavy metals (e.g. Cr, Cu, Pb, etc.) was within the limits. However, as highlighted in Table 3, these were generally in the upper end of the range of literature values for source-segregated household waste. This was an expected result as the assessed digestate originated from treatment of household rMSW where the biogenic fractions (e.g. organics, paper, and cardboard) were enzymatically and mechanically separated from the non-degradable solids; this process may intuitively lead to a higher metal contamination of the bioliquid substrate compared with processing source-segregated organic and paper. In addition, the concentration of DEHP (diethylhexyl phthalate) exceeded the corresponding limit and the sum of nonylphenols was close to the relative limit value. These results suggest that the quality of the digestate may not comply with selected criteria for application on land. This, however, depends on the specific legislation and on the type of waste treated. Notice that these results find a large correspondence in the environmental impacts associated with digestate on land application evaluated by parallel life-cycle assessment studies performed on the same waste refinery process [6]. Alternative solutions to direct on land application may be: i) co-digestion of the bioliquid with raw manure with further application of the digestate on land, ii) incineration of the digestate, and iii) post-composting of the digestate and disposal in landfill. The first allows for recycling of the nutrients, although the decreased concentration of metals and DEHP would only be an effect of dilution and the overall load would remain unchanged. The second and third prevent loading of the agricultural soil with metals and DEHP; however, this would

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induce losses of nutrients (even though it is envisioned that P will potentially be recovered from bottom ash) and additional energy consumption (for dewatering and drying).

Table 3

3.5 Uncertainties

As thoroughly detailed in the SI, the estimation of the uncertainties focused on the observed daily variation of the waste material fraction flows and on the uncertainty associated with the chemical analyses. Generally, the first was significantly higher than the second, thus determining the overall uncertainty of most of the assessed flows. The observed variations associated with the waste material fractions ranged from ±38% (of the mean value) for textile to ±56% for ferrous materials. These daily variations most likely were a consequence of variations in operational conditions of plant and thereby represent a "natural" variability in the output flows. The variations were included in the material, substance, and energy flow modelling as uncertainties related to the flows and thereby also applied to the energy layer (as no uncertainty was considered for the heating value). At the substance level, most uncertainties were affected by the variation in the waste material fraction flows, as mentioned above. As shown in Figs. 3–6, the largest uncertainties were generally associated with the minor flows (e.g. P non-ferrous materials and soft plastic, K in residue, N in textiles, carbon in air pollution control residue, i.e. APC). S Significant uncertainties were found in the liquid fraction recirculation flow of fossil C (Fig. 4), Fe and Al (Fig. 6). The remaining uncertainties were of the order of ±6 to ±69% of the mean value. The uncertainty associated with the energy flows of biogas and flue gas was close to ±20%. For carbon flows, the

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uncertainties ranged from about ±16% (C in the biogas flow) to ±68% (C in the ferrous materials). For phosphorous the uncertainty ranged between ±13% (P in the digestate) and ±58% (P in ferrous materials and hard plastic). The slightly lower flow of P in the digestate compared with the other nutrients (N and K) may be the result of sampling and modelling uncertainties.

While the uncertainties associated with the analytical procedures were generally very low compared with the observed variations in material flows, the sampling procedure was designed to minimize the uncertainties associated with the material composition by involving a large number of small sub-samples which were then combined before analysis. This procedure follows standard practices within sampling of solid materials such as ash waste (e.g.[51]). Further reduction of uncertainties associated with the output flows could be reached by extending the sampling period, i.e. involving more than the four days included in this study. While this would have been preferable, extending the sampling period should however be balanced by the need for a period of "stable" operation and comparable conditions. In this case, the four days were selected as a reasonable compromise between these two aspects. It should be noted that the recirculation flow (and associated chemical concentrations) was calculated by the STAN software based on the mass balance of the output flows. The uncertainties related to this flow could potentially be reduced by direct chemical analysis of the flow. However, as the recirculation flow represented a further optimization of the process, including additional washing and post-treatment, this flow could only be modelled and not analysed.

3.6 Cost analysis results

The results of the cost analysis for the Danish case study (Table 4) highlighted that the cost of the waste refinery solution (ca. 51 € Mg^{-1} ww) appeared slightly higher but, overall, comparable with the (direct) incineration alternative (ca. 48 € Mg^{-1} ww). The collection cost (ca. 36 € t^{-1} ww) would be exactly the same as for incineration if no source segregation was implemented in the system (which is the case for scenarios A and B). The total cost of the waste management systems was estimated to ca. 84 (waste refinery scenario) - 87 (incineration scenario) € Mg^{-1} ww.

As documented in the SI, the costs are only slightly affected by heat and electricity price (sensitivity S1): lower heat revenues (e.g. 10-20 € MWh^{-1}) would increase the cost of incineration (and thus of scenario B) making the total cost of the waste refinery solution (scenario A) equal to that of the alternative (see Table 4 and Table S9). In other words, the total costs for the waste refinery would be decreased in a context where the revenues for the electricity sold are higher than those for the heat sold (as the total electricity recovered is higher than that of direct incineration, but the heat recovered slightly lower). With this respect, the Danish market represents a particular case as heat revenues for incineration are significantly high compared with other European Countries.

Lower electricity recovery efficiency at the incinerator may also change the final cost making the waste refinery scenario overall preferable to the alternative (Table 4, sensitivity S3). In addition, hard plastic incineration (instead of recycling) also brought slightly higher revenues for the waste refinery solution (Table 4, sensitivity S2) indicating that, economically, incineration appeared preferable to recycling.

It has to be noted that when including the national subsidy for biogas-electricity,

the net cost for organic waste digestion was negative (i.e. net gaining) under the assumptions taken for electricity and heat recovery (see SI). This does not mean the plant actually gains money: en fact, these plants typically co-digest organic waste with manure which represents the bigger portion (ca. 80-90% of the total input, mass basis) and which dramatically decreases the overall energy production (thus raising costs) due to its extremely low dry matter content.

3.7 Perspectives on technology optimization potentials

Overall, the modelling results showed that about 81-89% of the nutrients could potentially be recovered in the bioliquid flow with the current technology set-up. However, still 11-19% of the total biomass input (i.e. organic waste, paper, cardboard, biogenic textiles, etc.) was found in the solids flow (mainly in the residue namely "RES"). This lost portion of biomass, although partly not liquefiable (e.g. fruit shells) and thus not recoverable as bioliquid, could be further decreased by improved enzymatic treatment (longer retention time, or optimized enzymes mix) in order to optimize the overall degradation of the biomass and to facilitate its further separation from the remaining solids. To this respect, the role of the post-treatment (here only modelled) will also be fundamental in order to maximize the separation bioliquid-solids and thus the total recovery of bioenergy. Further, in the broader perspective of maximizing the overall environmental performance of the technology, separation and recovery of ferrous, non ferrous, and eventually plastic materials for recycling and substitution of virgin products will also be important as documented in other studies [6,7]. Additional energy recovery through combustion of the non-recyclable materials such as residue, textile, fluff, and (eventually) soft plastic may also contribute to further

increase the overall environmental and energy savings by avoiding energy production from more polluting fossil sources (e.g. coal or oil), especially with respect to the GHG and SO_x emissions.

All in all, the integration of materials, bioenergy (from bioliquid), and energy (from residual solids) recovery could make the performance of this technology comparable (or better) to the best available state-of-the-art waste technologies [6,7].

4. Conclusion

A waste refinery may recover ca. 56% of the initial DM as bioliquid, yielding 6.2 GJ of biogas-energy from 1 Mg MSW dw, corresponding to 690 kWh electricity and 3,100 MJ heat. The residual solids contained about 5.9 GJ to be incinerated for additional energy recovery. Metals and plastic recovery may provide additional environmental benefits. The potential for recovery of nutrients and carbon in the digestate was estimated to be between 81% and 89% of the input. However, the quality of the digestate may be critical for use on land. Alternatives such as incineration, landfilling, or co-digestion may be possible solutions. Considering the potential variability of local revenues for energy outputs, the costs for the waste refinery solution appeared comparable with alternatives such as direct incineration.

Supporting Information (SI)

Additional information on mathematical elaboration of the uncertainties, MFA, SFA, EFA modelling, and cost analysis is available free of charge via the Internet at http://www.sciencedirect.com.

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Table 1. Overview of the methods (with associated inputs and outputs) used in the 5 phases that constitute the basis of this study. BL: bioliquid; EFA: energy flow analysis; FE: ferrous material; FF: fluff; HP: hard plastic; MFA: material flow analysis; MSW: municipal solid waste; NFE: non-ferrous material; RES: solid residue; SFA: substance flow analysis; SP: soft plastic; TXT: textiles; *sampled waste material fractions.

Phase	Input	Method	Output
1	MSW undergoing heating, enzymatic treatment and seaving at the waste refinery pilot-scale plant.	On-field sampling (BL*, FF*, SF*) and hand-sorting of 6 selected material fractions from the sampled SF*: HP*, SP*, TXT*, FE*, NFE*, RES*.	Representative samples of 8 waste material fractions: BL*, HP*, SP*, TXT*, FE*, NFE*, FF*, RES*. Related mass flows were also quantified.
2	Representative samples of 8 waste material fractions: BL*, HP*, SP*, TXT*, FE*, NFE*, FF*, RES*.	Samples preparation: drying, shredding, mixing, splitting, etc.	Dried powdered samples prepared for chemical analyses.
3	Dried powdered samples prepared for chemical analyses.	Chemical analyses.	Results of chemical analyses for BL*, HP*, SP*, TXT*, FE*, NFE*, FF*, RES*.
4	Mass flows and results of chemical analyses for BL*, HP*, SP*, TXT*, FE*, NFE*, FF*, RES*.	Mathematical optimization to model 'virtual' post-treatment.	Re-calculated mass flows and chemical composition of HP, SP, TXT, FE, NFE, FF and RES. Estimation of potential for BL recovery.
5	Results of the mathematical optimization model and of laboratory analyses (e.g. methane potential, digestate composition$^{\alpha}$, etc.).	MFA, SFA, and EFA.	Mass, substance, and energy flows within the waste refinery process including post-treatment and downstream energy and materials recovery processes. Cost analysis based on MFA/EFA flows.

α Digestate samples representing a specific point in time were collected from a full-scale digester to evaluate the quality of the digestate obtained from bioliquid digestion.

Table 2. Chemical composition of the sampled (*) and modeled waste material fractions (kg kg^{-1} DM except for HHV$_{db}$ and LHV$_{db}$ that are expressed as MJ kg^{-1} DM). BL: bioliquid; FE: ferrous material; FF: fluff; HP: hard plastic; NFE: non-ferrous material; RES: solid residue; SP: soft plastic; TXT: textiles; T/R: ratio between the total amount of the selected chemical in this study and in [18]; μ: share of the total DM. σ: standard deviation of μ (e.g. the share of HP* on the total DM is 10% ±2.8%). O$_{\&}$: sum of oxygen and (aggregated) heavy metals in traces (see SI); agg.: aggregated in O$_{\&}$; n.r.: not reported. Eventual inconsistencies are due to rounding (values rounded to 2 significant digits).

	BL* (BL)	HP*	HP	SP*	SP	TXT*	TXT	FE*	FE	NFE*	NFE	RES*	RES	FF*	FF	TOTAL	T/R
μ	9.0E-02^{α}	1.0E-01	8.2E-02	2.1E-01	9.0E-02	1.7E-01	7.6E-02	5.0E-02	4.1E-02	2.8E-02	2.4E-02	3.5E-01	1.2E-01	9.1E-03	4.0E-03	1.0E+00	1.0
σ	4.1E-02	2.8E-02	2.3E-02	7.6E-02	3.4E-02	4.4E-02	2.1E-02	2.8E-02	2.3E-02	1.4E-02	1.2E-02	1.3E-01	4.7E-02	4.1E-04	2.2E-04	-	n.r.
C$_{biog}$	4.2E-01	9.4E-02	2.0E-02	3.0E-01	1.4E-01	4.0E-01	3.7E-01	8.7E-02	1.7E-02	1.2E-01	2.0E-05	3.1E-01	1.1E-01	3.7E-01	3.0E-01	2.9E-01	1.0
C$_{foss}$	8.3E-03	6.4E-01	7.8E-01	2.4E-01	5.4E-01	3.9E-02	7.6E-02	5.3E-02	6.2E-02	2.9E-02	1.8E-02	1.2E-01	3.2E-01	7.0E-02	1.5E-01	1.7E-01	1.3
H	5.1E-02	1.1E-01	1.2E-01	8.1E-02	1.2E-01	6.2E-02	7.5E-02	4.5E-02	4.4E-02	3.2E-02	2.8E-02	6.0E-02	7.6E-02	6.7E-02	8.7E-02	6.7E-02	1.1
S	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	0.72
N	2.0E-02	3.7E-03	2.3E-06	1.2E-02	1.9E-03	1.1E-02	3.9E-04	5.0E-03	1.9E-03	2.3E-02	2.4E-02	1.4E-02	3.1E-03	3.6E-02	5.6E-02	1.3E-02	1.0
F	1.8E-04	7.0E-05	4.5E-05	1.2E-04	4.4E-05	1.1E-04	2.8E-05	9.0E-05	7.1E-05	1.2E-04	1.1E-04	1.2E-04	1.1E-05	1.5E-04	1.1E-04	1.2E-04	0.9
P	2.5E-03	4.8E-04	2.2E-05	1.4E-03	1.1E-05	1.7E-03	7.6E-04	5.1E-04	9.4E-05	6.9E-04	3.5E-04	2.4E-03	2.2E-03	1.9E-03	1.2E-03	1.7E-03	0.84
Cl	1.1E-02	2.2E-03	2.1E-04	1.6E-02	2.2E-02	6.0E-03	1.4E-04	1.9E-03	8.9E-07	2.2E-03	5.3E-04	8.5E-03	4.0E-03	6.1E-03	1.5E-06	8.7E-03	1.0
K	7.0E-03	1.5E-03	2.5E-04	5.0E-03	2.5E-03	5.1E-03	2.9E-03	1.3E-03	1.3E-04	1.7E-03	7.0E-04	4.5E-03	4.7E-05	6.0E-03	4.7E-03	4.4E-03	1.0
Fe	1.7E-03	1.7E-03	1.7E-03	2.1E-03	2.5E-03	1.9E-03	2.2E-03	6.7E-01	8.1E-01	1.1E-01	1.3E-01	3.4E-03	6.5E-03	3.8E-03	6.4E-03	3.9E-02	1.4
Al	3.0E-03	1.2E-03	8.1E-04	2.1E-03	8.8E-04	2.4E-03	1.7E-03	2.3E-02	2.8E-02	6.0E-01	7.1E-01	7.0E-03	1.4E-02	3.7E-03	4.6E-03	2.2E-02	0.94
Cd	1.8E-07	1.0E-07	agg.	1.1E-07	agg.	1.7E-07	agg.	1.0E-07	agg.	1.1E-07	agg.	1.1E-07	agg.	1.3E-07	agg.	1.2E-07	0.02
Cr	1.9E-05	1.3E-05	agg.	1.3E-05	agg.	8.3E-06	agg.	2.3E-04	agg.	1.7E-04	agg.	1.7E-05	agg.	1.2E-05	agg.	2.9E-05	0.3
Cu	3.5E-05	1.2E-05	agg.	7.5E-05	agg.	3.6E-05	agg.	1.3E-04	agg.	6.7E-04	agg.	4.8E-05	agg.	2.7E-05	agg.	6.8E-05	0.1
Ni	1.2E-05	5.7E-06	agg.	4.7E-06	agg.	4.2E-06	agg.	3.3E-04	agg.	2.2E-04	agg.	2.4E-04	agg.	5.3E-06	agg.	3.4E-05	0.71
Sr	1.0E-04	2.0E-05	agg.	6.0E-05	agg.	6.2E-05	agg.	1.2E-04	agg.	2.1E-05	agg.	6.7E-05	agg.	1.0E-04	agg.	6.5E-05	n.r.
Mn	8.0E-05	3.4E-05	agg.	4.6E-05	agg.	6.4E-05	agg.	1.9E-03	agg.	5.4E-03	agg.	5.6E-03	agg.	7.3E-05	agg.	2.3E-03	6.9
Mg	2.2E-03	7.1E-04	3.6E-04	1.3E-03	5.7E-05	1.3E-03	1.6E-04	1.3E-03	1.1E-03	9.6E-03	1.1E-02	1.8E-03	1.0E-03	2.5E-03	2.8E-03	1.7E-03	1.1
As	1.4E-06	1.0E-06	agg.	1.0E-06	agg.	1.0E-06	agg.	1.1E-05	agg.	2.5E-06	agg.	1.0E-06	agg.	1.4E-06	agg.	1.6E-06	0.2
Hg	1.0E-07	1.0E-07	agg.	1.0E-07	agg.	1.0E-07	agg.	1.0E-07	agg.	1.0E-07	agg.	1.0E-07	agg.	5.0E-07	agg.	1.0E-07	0.04
Pb	1.2E-05	2.2E-05	agg.	7.6E-06	agg.	8.0E-06	agg.	4.7E-06	agg.	1.3E-05	agg.	1.2E-05	agg.	2.3E-04	agg.	1.3E-05	0.06
Sb	1.4E-06	3.5E-05	agg.	1.0E-06	agg.	6.7E-06	agg.	3.8E-06	agg.	1.0E-06	agg.	1.5E-06	agg.	1.0E-06	agg.	5.7E-06	n.r.
O$_{\&}$	4.7E-01	1.4E-01	6.9E-02	3.4E-01	1.7E-01	4.7E-01	4.7E-01	1.1E-01	3.8E-02	6.8E-02	7.0E-02	4.6E-01	4.6E-01	4.3E-01	3.8E-01	3.8E-01	n.r.
HHV$_{db}$	17	33	37	23	31	18	19	11	9.3	25	26	17	18	15	13	20	1.1
LHV$_{db}$	16	31	34	21	28	17	18	9.6	8.3	24	26	16	16	14	11	19	1.0

For the sampled bioliquid (BL*), the DM content was 20%. The VS was 85% (of DM). Carbohydrates, lipids, proteins, VFA and ethanol were (% VS): 85, 2.8, 1.9 and 2.2.

α The share of BL on the total dry mass flow equaled 5.6E-01 after summing the individual contributions from the remaining waste material fractions undergoing post-treatment (Figs. 2-3).

Table 3. Modeled and analysed concentrations of selected chemicals in the digestate from bioliquid digestion and in selected digests from source-separated organic waste processing. The values are expressed as mg kg^{-1} DM (except for DM, VS, C and N which are expressed as % DM). * Dewatered sample. DG: digestate; n.d.: not detected; n.m.: not modelled; n.a.: not analysed; n.r.: not reported. PAH: sum of polyaromatic hydrocarbons; DEHP: diethylhexyl phthalate; LAS: linear alkylbenzene sulfonate; NPs: sum of nonylphenols. Eventual inconsistencies are due to rounding (values rounded to 2 significant digits).

Chemical	DG (model)	DG (analysis)	DG (analysis)*	[29]	[29]	[30]	[28][η]	[28][θ]
DM	n.m.	4.2 ±0.1	16	44	2.7	1-2	n.r.	n.r.
VS	n.m.	59	59	n.r.	n.r.	65	n.r.	n.r.
C	28 ±8	28	29	n.r.	n.r.	39	n.r.	n.r.
H	n.m.	n.a.	n.a.	n.r.	n.r.	n.r.	n.r.	n.r.
S	2,600 ±700	5,600	6,000	n.r.	n.r.	5,000	n.r.	n.r.
N	4.6 ±1.2	4.6 ±0.14	n.a.	n.r.	n.r.	3.5	n.r.	n.r.
F	470 ±130	n.a.	n.a.	n.r.	n.r.	n.r.	n.r.	n.r.
P	6,600 ±1,700	6,400 ±900	6,000	n.r.	7,963	9,000	n.r.	n.r.
Cl	29,000 ±7,600	n.a.	n.a.	n.r.	n.r.	n.r.	n.r.	n.r.
K	18,000 ±5,000	21,000	7,800	n.r.	n.r.	n.r.	n.r.	n.r.
Fe	5,000 ±150	17,000	20,000	n.r.	n.r.	n.r.	n.r.	n.r.
Al	8,000 ±2,200	8,100	11,000	n.r.	n.r.	n.r.	n.r.	n.r.
Cd	0.47 ±0.12[ε]	0.75 ±0.029[α]	0.70[β]	0.76	0.95	0.3-0.7	0.8	100
Cr	50 ±13	53 ±2.5	57	n.r.	9.9	n.r.	100	n.r.
Cu	92 ±24	120 ±5.9	130	n.r.	76	45-125	1,000	n.r.
Ni	32 ±8.3[ζ]	30 ±2.7[γ]	30[δ]	16	10.7	8-28	30	2,500
Sr	260 ±70	270	270	n.r.	n.r.	n.r.	n.r.	n.r.
Mn	210 ±55	370	400	n.r.	n.r.	n.r.	n.r.	n.r.
Mg	5,800 ±1,500	6,300	4,000	n.r.	n.r.	8,000-11,000	n.r.	n.r.
As	3.7 ±1.0	9.3	10	n.r.	n.r.	n.r.	n.r.	n.r.
Hg	0.26 ±0.069	0.3	n.a.	0.42	0.24	n.r.	0.8	200
Pb	32 ±8.3	27 ±3.3	18	54	17	10-60	120	10,000
Sb	n.d.	n.d.	n.d.	n.r.	n.r.	n.r.	n.r.	n.r.
Zn	n.m.	880 ±110	850	205	339	150-300	4,000	n.r.
Co	n.m.	3.9	4.6	n.r.	n.r.	n.r.	n.r.	n.r.
PAH	n.m.	1.5	n.a.	n.r.	0.47	n.r.	3	n.r.
DEHP	n.m.	75 ±1	n.a.	n.r.	61	n.r.	50	n.r.
LAS	n.m.	130	n.a.	n.r.	100	n.r.	1,300	n.r.
NPs	n.m.	9.6 ±0.35	n.a.	n.r.	5.3	n.r.	10	n.r.

α Corresponding to 120 mg kg^{-1} P (limit 100, see [28][θ]). β Corresponding to 110 mg kg^{-1} P (limit 100, see [28][θ]). γ Corresponding to 4,700 mg kg^{-1} P (limit 2,500, see [28][θ]). δ Corresponding to 5,000 mg kg^{-1} P (limit 2,500, see [28][θ]). ε Corresponding to 78 ±53 mg kg^{-1} P (limit 100, see [28][θ]). ζ Corresponding to 5,300 ±2,700 mg kg^{-1} P (limit 2,500, see [28][θ]). η Limit from [28] expressed as mg kg^{-1} DM. θ Limit from [28] expressed as mg kg^{-1} P.

Table 4. Total cost (€ Mg^{-1} ww) of the waste management system for scenarios A and B. AD: anaerobic digestion; INC: incineration; S1: sensitivity 1 (heat price set to 10 € GJ^{-1} heat, as an example); S2: sensitivity 2 (hard plastic HP incinerated instead of recycled); S3: sensitivity 3 (incineration electricity recovery efficiency set to 20%, instead of 25%, for both scenario B and A). Values rounded to 2 significant digits.

Scenario	Collection	Treatment			Total
		WR	AD	INC	
A	36	55	-11	7	87
B	36			48	84
A (S1)	36	50	4	13	103
B (S1)	36			67	103
A (S2)	36	58	-11	2	85
B (S2)	36			48	84
A (S3)	36	54	-11	9	88
B (S3)	36			53	89

Fig. 1. Illustration of the pilot-plant process (within the boundary) and associated outputs. Boxes and flows indicated by grey dashed lines were modelled using the optimization model. Flows in grey lines were modelled using MFA based on the results of the optimization model. Flows in black dashed lines were modelled using MFA based on the results from the optimization model and analyses of the digestate. APC: air pollution control residue; BA: bottom ash; BG: biogas; BL: bioliquid; DG: digestate; FE: ferrous material; FF: fluff; HP: hard plastic; LF: liquid fraction; MSW: municipal solid waste input; MSW': MSW after heating and enzymatic treatment; NFE: non-ferrous material; RES: residue; SP: soft plastic; TXT: textiles; *sampled waste material fractions.

Fig. 2. Illustration of the principles of the mathematical optimization model. Nomenclature as in Fig. 1.

Fig. 3. Mass and energy flows within the waste refinery and downstream energy conversion processes (kg DM and GJ). APC: air pollution control residue; BA: bottom ash; BG: biogas; BL: bioliquid; DG: digestate; FE: ferrous material; FF: fluff; FG: flue gas; HP: hard plastic; LF: liquid fraction; MSW: municipal solid waste input; MSW': MSW after heating and enzymatic treatment; NFE: non-ferrous material; RES: residue; SP: soft plastic; TXT: textiles; *sampled waste material fractions.

Fig. 4. Total and fossil carbon flows within the waste refinery and downstream energy conversion processes (kg). Nomenclature as in Fig. 3.

Fig. 5. N, P, and K flows within the waste refinery and downstream energy conversion processes (kg, g, and kg). Nomenclature as in Fig. 3.

Fig. 6. Fe and Al flows within the waste refinery and downstream energy conversion processes (kg). Nomenclature as in Fig. 3.

Mass (kg DM)

- MSW: 1,000
- MSW': 1,500±12%
- LF recirculated: 460±39%
- Sieving I outputs:
  - NFE*: 28±49%
  - FE*: 50±56%
  - HP*: 100±27%
  - SP*: 200±37%
  - TXT*: 170±27%
  - RES*: 350±38%
- LF: 560±13%
- FF*: 0.91±44%
- Post-treatment outputs:
  - NFE: 24±51% → Recycling
  - FE: 41±56% → Recycling
  - HP: 82±28% → Recycling
  - SP: 90±38%
  - TXT: 76±28%
  - RES: 120±38%
  - FF: 4±6%
- Incineration outputs:
  - FG: 210±21%
  - BA: 69±21%
  - APC: 13±25%
  - → Treatment Disposal
- BL: 560±13%
- Anaerobic digestion outputs:
  - BG: 350±21%
  - DG: 210±29%
  - → CHP

Energy (GJ)

- MSW: 19±3%
- MSW': 26±12%
- LF recirculated: 7.4±45%
- Sieving I outputs:
  - NFE*: 0.68±49%
  - FE*: 0.53±56%
  - HP*: 3.1±27%
  - SP*: 4.4±37%
  - TXT*: 2.8±27%
  - RES*: 5.6±38%
- LF: 8.9±13%
- FF*: 0.012±44%
- Post-treatment outputs:
  - NFE: 0.61±51%
  - FE: 0.34±56%
  - HP: 2.8±28%
  - SP: 2.6±38%
  - TXT: 1.3±28%
  - RES: 2±38%
  - FF: 0.042±6%
- Incineration outputs:
  - FG: 5.6±22% } 410±88 kWh / 4400±950 MJ
  - BA: 0.15±22%
  - APC: 0.15±22%
- BL: 8.9±13%
- Anaerobic digestion outputs:
  - BG: 6.2±17% } 690±120 kWh / 3100±540 MJ
  - DG: 2.7±30%

Total carbon (kg)

MSW 460±6% → Heating & Enzymatic treatment → MSW' 660±13% → Sieving I

From Sieving I:
- NFE* 4.1±50%
- FE* 7±57%
- HP* 74±29%
- SP* 110±38%
- TXT* 73±29%
- RES* 150±39%

→ Post-treatment

200±44% LF recirculated
240±16% LF
FF* 0.4±45%

Post-treatment outputs:
- 0.43±51% → NFE
- 3.3±68% → FE
- 66±28% → HP
- SP 61±38%
- TXT 34±28%
- RES 54±38%
- FF 1.8±6%

→ Incineration
- 150±21% FG
- 1.8±27% BA
- 0.26±287% APC

Sieving II → BL 240±16% → Anaerobic digestion
- 180±16% BG
- 60±17% DG

Fossil carbon (kg)

MSW 170±18% → Heating & Enzymatic treatment → MSW' 170±18% → Sieving I

From Sieving I:
- NFE* 0.81±49%
- FE* 2.6±56%
- HP* 65±29%
- SP* 50±37%
- TXT* 6.4±27%
- RES* 42±38%

→ Post-treatment

3±1446% LF recirculated
4.9±13% LF
FF* 0.064±45%

Post-treatment outputs:
- 0.43±51% → NFE
- 2.6±56% → FE
- 65±28% → HP
- SP 49±38%
- TXT 5.7±42%
- RES 40±38%
- FF 0.59±6%

→ Incineration
- 94±25% FG
- 1.1±30% BA
- 0.17±287% APC

Sieving II → BL 4.8±14% → Anaerobic digestion
- 3.6±14% BG
- 1.2±14% DG

Nitrogen (kg)

Phosphorous (g)

Potassium (kg)

Iron (kg)

Aluminium (kg)

Supporting Information (SI) for:

Bioenergy, material, and nutrients recovery from household waste: advanced material, substance, energy, and cost flow analysis of a waste refinery process

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This SI document includes text, figures and tables with details on the mathematical modelling and on the background data, equations, and calculations performed for the material, substance, energy, and cost flow analysis.

Table of Contents

1. Formulation of the mathematical optimization model

1.2 Elaboration of the uncertainties

Two sources of uncertainties were considered in the study: i) uncertainty associated with the sampled waste material fraction flows and ii) uncertainty associated with the analytical concentration of the chemicals. The first refers to the observed daily variation of the waste material fraction flows (see Table 2 of the manuscript, parameter σ). The second is the uncertainty of the chemical analyses. These were: 10% for C, H, N, S, 7.5% for Cl and F, 2.5% for P, and 6.2% for the metals (including K). The uncertainties are here intended as those values corresponding to 68% of the confidence interval around the mean (this also applies to Figs. 3-6 and Tables 2-3 of the manuscript).

Suppose that for all i, j, the total (dry) flows P_i, the chemicals concentration CP_{ji}, and the concentration of the chemicals CBL_j in the bioliquid are observed for K times, each time being an independent measurement. Let $P_i^k, CB_{ji}^k, CBL_{ji}^k$ be the k-th observation. Due to several sources of uncertainty affecting the measurements, such as errors, simplifications, inherent limited representativeness of the samples, etc., the K observations are expected to differ from each others. More specifically, they are expected to be *similar* but not identical. It is said that, the more is the diversity, the more is the level of uncertainty. Clearly, even the material flow chemicals CM_{ij}^k and the coefficient λ_i^k, obtained by solving problem of Eq. (9) (see manuscript), depends on the observation k. Same concept applies to λ_i^k when it is combined with P_i^k in Eq. (7), to obtain the bioliquid flow BL_i^k, and the material flow M_i^k. In general, it is said that the uncertainty in P_i, CB_{ji}, CBL_{ji} is *propagated* onto CM_{ji}, BL_i, M_i, through Eqs. (7-9) of the main manuscript. In order to handle the uncertainty within the MFA, the uncertainty in the input data of the presented methodology must be quantified, all along with the propagated mass balance uncertainty. This study proposes a classical statistical

approach, where the measurement errors are characterized by means of Probability Distribution Function (PDF), and the uncertainty propagation is performed using Monte Carlo simulation (Fig. S1).

The uncertainty of the flows $P_1, \ldots, P_N$, was characterized as the conjugate prior of the parameters of the multinomial distribution, also known as the Dirichlet distribution, see e.g. [1]. The Dirichlet is a multivariate distribution generating random discrete PDFs, which are nothing but vectors of positive elements whose sum equals to one, hence the same properties as for the flows $P_1, \ldots, P_N$ (Eq. (3)). Formally the vector $\mathbf{P} = (P_1, \ldots, P_N)$ is said to have a Dirichlet PDF:

$$f(\mathbf{P}|\boldsymbol{\alpha}) = \frac{\Gamma(\alpha_0)}{\prod_{i=1}^{N} \Gamma(\alpha_i)} \prod_{i=1}^{N} P_i^{\alpha_i - 1} \tag{S1}$$

And denoted with $\mathbf{P} \sim \mathrm{Dir}(\alpha_0, \alpha_1, \ldots, \alpha_N)$, with a set of parameters $\alpha_0, \alpha_1, \ldots, \alpha_N$ such that $\alpha_i > 0$ and $\alpha_0 = \sum_{i=1}^{N} \alpha_i$, where $\Gamma(\cdot)$ is the Gamma function. The parameters are estimated from K observations using a maximum likelihood procedure, described in [2].

The uncertainty of the concentrations CB_{ji}, CBL_{ji} was characterized and quantified directly within the laboratory analysis, as a Gaussian noise having means $\mathrm{E}[CB_{ji}], \mathrm{E}[CBL_{ji}]$ and variances $\mathrm{Var}[CB_{ji}], \mathrm{Var}[CBL_{ji}]$ for all i, j. Note that in this case a dataset of K observations was not available, therefore statistical independence was assumed. This means that, for example, the probability for the carbon concentration to have a certain value in one flow is influenced neither by the carbon concentration in another flow, nor by the concentration of any other element in all flows. Such assumption may be seen not entirely realistic, as considering correlations between chemicals in different flows may reduce the level of uncertainty in the MFA. However, even if the uncertainty *may* be reduced, it is also true that it cannot be increased. Therefore, in a situation such as in the current study, where

no information about correlations was available, assuming independence could be considered as a worst case scenario approach, hence it is safe.

The uncertainty is propagated using MonteCarlo Sampling. This consists in using Random Number Generators to produce K samples $P_i^k, CB_{ji}^k, CBL_{ji}^k$ for all i, j, distributed according to the PDFs discussed above. Then the corresponding CM_{ji}^k, BL_i^k, M_i^k are determined using Eqs. (7-9) as already discussed. With K Monte Carlo simulation iterations, the modelling results were estimated by means of expected values $\mathrm{E}[P_i], \mathrm{E}[BL_i], \mathrm{E}[M_i] \mathrm{E}[CM_{ji}]$, where $\mathrm{E}[x] = 1/n \sum_{k=1}^{n} x_k$. Similarily, the level of uncertainty was estimated by means of standard deviation from the expected value, i.e. $(\mathrm{Var}[P_i])^{1/2}, (\mathrm{Var}[BL_i])^{1/2}, (\mathrm{Var}[M_i])^{1/2}, (\mathrm{Var}[CM_{ji}])^{1/2}$, where $\mathrm{Var}[x] = \mathrm{E}[x^2] - \mathrm{E}^2[x]$. The larger the standard deviation, the larger the level of uncertainty.
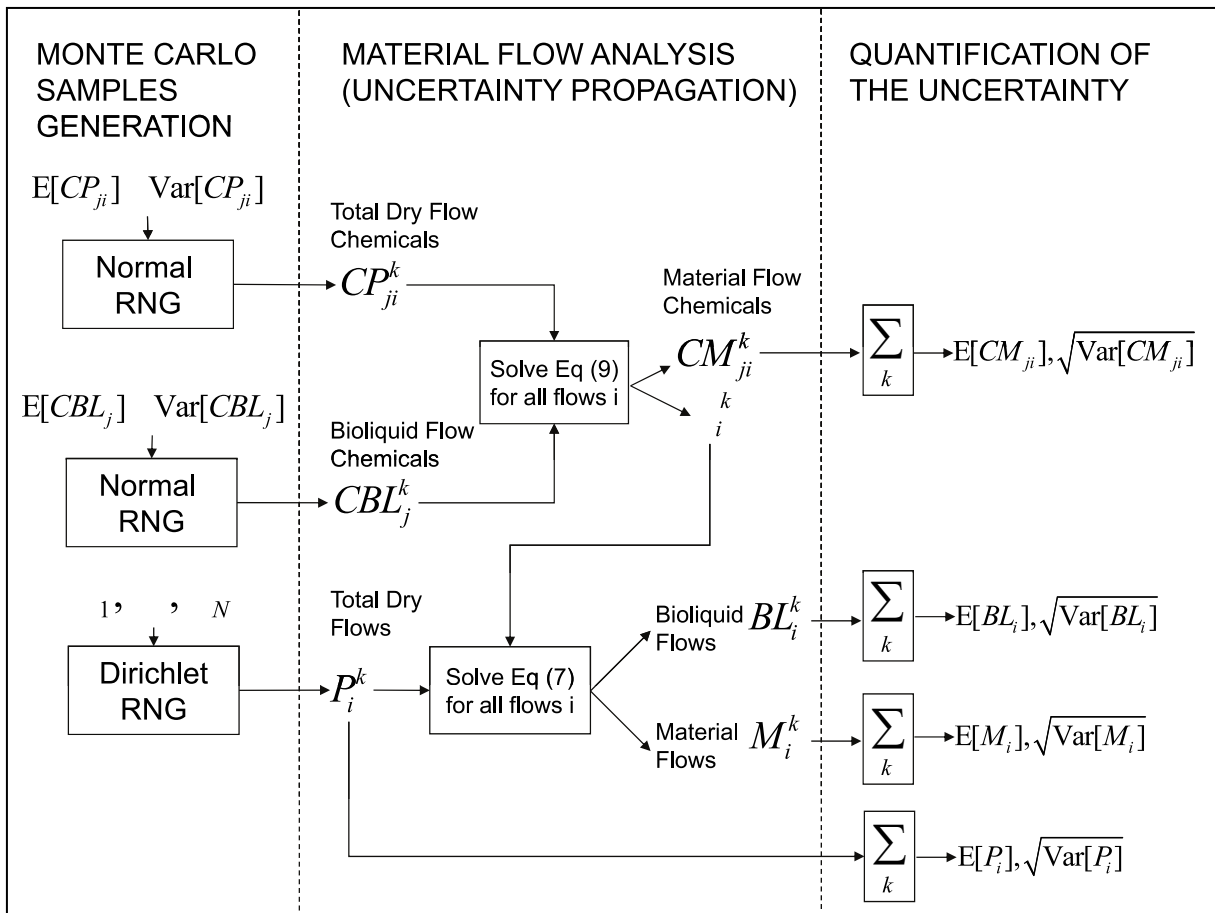
2. Material, substance, and energy flow analysis: input data and modelling

The results of the mathematical optimization model (BL_i, M_i and CM_{ji}) were used as inventory for the MFA, SFA and EFA of the waste refinery process. This was facilitated by the software STAN [3]. The MFA encompasses the waste refinery process itself (including the modelled post-treatment) and downstream material separation and energy conversion (i.e. anaerobic digestion of the bioliquid and incineration of the residual solids).

The transfer coefficients (to bottom ash, air pollution control residue, i.e. APC, and to flue gas) of the waste material fractions incinerated were based on a literature review of a number of five Danish incinerators (namely *Amaforbrænding, Vesterforbrænding, Taastrup* incinerator, *Aahrus* incinerator, and *Herning* incinerator). Based on this, the transfer coefficients for DM to bottom ash and APC were, respectively, 23% (±0.6%) and 4.3% (±0.57%). The remaining was thus transferred to flue gas (intended as after flue gas cleaning). The transfer coefficients of C, N, P, K, Fe and Al to flue gas, bottom ash and APC were based on the findings of [4] for household waste. For example, the transfer coefficient for Fe was assumed 99% (± 0.2%) to bottom ash and 0.8% to APC (± 0.1%); the transfer coefficient for Al was 88% (± 2%) to bottom ash and 12% (± 2%) to APC.

The actual methane production from the bioliquid undergoing anaerobic digestion was set to 365 NL kg^{-1} VS based on preliminary results from tests operated at a full-scale anaerobic digestion plant [5]. The share of DM, C, and N transferred to the digestate and to the biogas (i.e. transfer coefficients) were calculated based on mass balances (Eq. (S2-S5)) using the experimental data available about digestate composition (Table 3) and assuming ash conservation. It was assumed that P, K, Fe, and Al found in the bioliquid were not transferred to the biogas.

The lower heating value, on dry basis (LHV_{db}), of the waste material fractions was recalculated from the HHV_{db} (see section 2.5.2 of the manuscript) using Eq. (S6). With

respect to later combined heat and power (CHP) efficiency for gas engine (biogas combustion) and incineration (combustion of the residual solids that includes soft plastic, textiles and residue), electricity and heat efficiencies of 40% and 50% (relative to the energy content of the biogas) were assumed for the gas engine based on [6] and of 25% and 75% for the incinerator [7]. The latter are gross efficiencies relative to the LHV_{db} for state of the art incinerators combusting high-energy content materials and provided with flue-gas condensation (based on [7]).

$$VS_{BL} \cdot BL = VS_{DG} \cdot DG + \Delta VS_{BG} \tag{S2}$$

$$\Delta VS_{BG} = \Delta DM_{BG} = BL - DG \tag{S3}$$

Where:

VS_{BL}: volatile solids content of the bioliquid   (kg VS kg^{-1} DM)

VS_{DG}: volatile solids content of the digestate   (kg VS kg^{-1} DM)

ΔVS_{BG}: amount of VS converted into biogas (BG)   (kg VS kg^{-1} ww)

ΔDM_{BG}: amount of DM converted into biogas (BG)   (kg DM kg^{-1} ww)

BL: dry matter content of the bioliquid   (kg DM kg^{-1} ww)

DG: dry matter content of the digestate   (kg DM kg^{-1} ww)

$$C_{BL} \cdot BL = C_{DG} \cdot DG + \Delta C_{BG} \tag{S4}$$

$$N_{BL} \cdot BL = N_{DG} \cdot DG + \Delta N_{BG} \tag{S5}$$

C_{BL}: carbon content of the bioliquid   (kg C kg^{-1} DM)

C_{DG}: carbon content of the digestate   (kg C kg^{-1} DM)

N_{BL}: nitrogen content of the bioliquid   (kg N kg^{-1} DM)

N_{DG}: nitrogen content of the digestate   (kg N kg^{-1} DM)

ΔC_{BG}: amount of C converted into biogas (BG)   (kg C kg^{-1} ww)

ΔN_{BG}: amount of N converted into biogas (BG)   (kg N kg^{-1} ww)

$$LHV_{db} = HHV_{db} - 2.442 \cdot 8.936 \cdot H/100 \tag{S6}$$

LHV_{db}: higher heating value, dry basis   (MJ kg^{-1} DM)

HHV_{db}: higher heating value, dry basis   (MJ kg^{-1} DM)

H: hydrogen content, dry basis   (kg H kg^{-1} DM)

8.936 is the conversion factor of H to H_2O (kg of H_2O formed by combustion of 1 kg H).

2.442 is the latent heat of vaporization of H_2O at 25 ºC (MJ kg^{-1}).

3. Cost analysis

3.1 Waste management scenarios and prices: assumptions

An economic cost analysis including costs for collection, treatment (including revenues from energy and materials sold), and disposal of residues was performed on the following scenarios:

A. Collection of all municipal household waste without source segregation and waste refinery treatment (CHP production from the bioliquid-biogas and incineration of the solid fraction for CHP production; separation and recycling of hard plastic and metals).

B. Collection of all municipal household waste without source segregation and direct incineration for CHP production.

The geographical scope of the analysis was Denmark. The temporal scope was 2013. The assumption was that the waste management system served a population of 100,000 inhabitants. The waste produced per capita equalled 512 kg person^{-1} y^{-1} based on average EU27 values for 2009 [8]. The collection scheme was based on a recent assessment (2013) for a Danish region of 100,000 household [9]. This represented a realistic multifamily (i.e. more households share a waste-container) collection scheme based on an existing household waste management system established in a region of Denmark. The frequency of collection for residual waste was one "emptying" per week. Prices for electricity, heat, recyclables, enzymes and man-work are listed in Table S1. The price for electricity was based on the Danish market (0.035 € kWh^{-1}). The price for heat (assumed as district heating) was based on the Danish heat market (0.042 € kWh^{-1}). For recyclables, they were based on 2013 market prices for these materials: however, the prices on the market refer to virgin materials. Therefore, the revenues for selling recycled waste materials were approximated as half of the corresponding market prices for virgin material in order to take into account the loss of quality and value (see Table S1). The collection cost of emptying one container (with volume

S10

assumed equal to 660 L) was assumed to be 3 € emptying^{-1} (this includes transportation costs to the point of unload, i.e. to the treatment plant) based on Danish conditions [9]. In addition, the Danish national tax on CO_2 (ca. 22.3 € Mg^{-1} CO_2 emitted), the national tax on heat from incineration (ca. 8 € GJ^{-1}), and the national subsidy for biogas-electricity (ca. 0.047 € kWh^{-1}) were considered in the analysis. The final results are presented with and without tax/subsidy.

Table S1. Prices used in the cost analysis (E: estimated based on unpublished data, e.g. bills from treatment plants, payslip, etc.). BA: bottom ash; APC: air pollution control residues.

Parameter	Price	Unit	Source	Note
Electricity	0.035	€ kWh^{-1}	E	Danish market price (http://www.nordpoolspot.com/)
Heat	0.042	€ kWh^{-1}	E	Average Danish market price
Enzymes	2	€ kg^{-1}	E	Specific for the enzymes used in the waste refinery
Water	0.67	€ Mg^{-1}	E	Danish price for water
Paper	52.5	€ Mg^{-1}	[10]	Average (Dec 2013 paper waste price)
Hard plastic	85/2	€ Mg^{-1}	[10]	Low quality (Dec 2013 plastic film price)
Ferrous metal	175/2	€ Mg^{-1}	[10]	Average of 170-180 (Dec 2013 steel price)
Non ferrous metal	800/2	€ Mg^{-1}	[10]	Dec 2013 aluminium price
Wage	33	€ h^{-1}	E	Average Danish wage for a plant with 30 employees
Emptying one container	3	€ emptying^{-1}	[9]	Average estimated Danish price
Capital cost incinerator	75.6	€ Mg ww^{-1}	[9]	€ per Mg of household wet waste treated
Capital cost waste ref.	42.9	€ Mg ww^{-1}	E	€ per Mg of household wet waste treated
Capital cost digester	25.7	€ Mg ww^{-1}	[9]	€ per Mg of household wet waste treated
Maintenance incinerator	7.3	€ Mg ww^{-1}	[9], [11]	€ per Mg of household wet waste treated
Maintenance waste ref.	7.3	€ Mg ww^{-1}	E	€ per Mg of household wet waste treated
Maintenance digester	7.3	€ Mg ww^{-1}	E	€ per Mg of household wet waste treated
Disposal BA	3.7	€ Mg BA^{-1}	[11]	€ per Mg of BA
Disposal APC	73	€ Mg APC^{-1}	[11]	€ per Mg of APC

3.2 Waste composition (household waste generated): assumptions

In order to calculate the cost for the collection and treatment of the waste material fractions, the waste composition presented in Table 2 in the manuscript was recalculated on a wet matter basis. In order to do so, the dry matter content of the material fractions (as generated at the household) was needed. This was taken from [12] where the composition of an average mixed household waste is detailed (see column DM in Table S2). Furthermore, the material fraction bioliquid (BL) was split into organic waste and paper, according to their estimated share in the bioliquid (ca. 50% each, based on [13]). The recalculated household waste composition (on a wet basis) is presented in Table S2. This (column 5 of Table S2) represents the composition of the household waste generated by the hypothetical region under assessment, and it was used for the calculation of the cost associated with collection and treatment. Notice that the dry matter content of the mixed household waste was equal to 52% (i.e. DM = 1000 kg DM/1928 kg ww = 52%).

Table S2. Estimation of the waste composition, wet matter basis (last column to the right). BL: bioliquid; DM: dry matter; HP: hard plastic; FE: ferrous metal; NFE: non ferrous metal; ww: wet waste. Residual solids are the sum of soft plastic, textiles, and residue (RES).

Waste material fraction	Content (% DM)	DM (% ww)	Total amount (kg ww)	Content (% ww)
Organic (from BL)	28%	0.42	672	35%
Paper (from BL)	28%	0.44	630	32.7%
Hard plastic (HP)	8.2%	0.83	99	5.1%
Soft plastic (HP)	9%	0.65	138	7.1%
Ferrous (FE)	4.1%	0.97	42	2.2%
Non ferrous (NFE)	2.4%	0.97	25	1.3%
Residual solids	20.3%	0.63	322	16.7%
Total	100%		1928	100%

Based on the LHV_{db} calculated for the household waste sampled in this study (ca. 19 MJ kg^{-1} ww, see Table 2 of the main manuscript), the LHV_{wb} (wet basis) would be then equal (assuming DM=52%) to 8.7 MJ kg^{-1} ww (i.e. 19 x 0.52 – (1-0.52) x 2.442).

Further, the amount of residual solids fraction produced by the waste refinery from 1 Mg ww would equal:

$$residual\ solids\ (wet) = \frac{290\ kg\ DM\ residual\ solids}{0.7\ kg\ DM\ kg^{-1}\ residual\ solids} \cdot 0.52\ kg\ DM\ kg^{-1}\ ww \qquad (S7)$$

$$= 0.215\ Mg\ wet\ residual\ solids\ Mg^{-1}\ ww$$

The DM content of the residual solids was assumed equal to 70% (generally between 65% and 75% according to [13]. Based on the energy flows reported in Fig. 3 of the manuscript, the LHV of the residual solids can be recalculated (assuming DM 70%) conformingly with Eq. (S6):

HHV = 5942 MJ / 290 kg = 20.5 MJ kg^{-1} DM

LHV = HHV x DM% - (100%-DM%) x 2.442 = 13.6 MJ kg^{-1} ww

3.3 Collection cost

3.3.1 Calculation method

Collection costs were calculated according to Eqs. (S8-S9).

$$\text{Collection cos}t = \text{emptying price} \times N \text{ containers} \tag{S8}$$

$$N \text{ containers} = \frac{\text{waste capita} \times \text{pop} \times \%\text{waste material} \times \text{segregation eff}}{\rho_{\text{waste material}}} \times \frac{1}{V_{\text{container}} \times \text{filling}} \tag{S9}$$

Where:

emptying price = 3 € emptying^{-1}

waste capita = 0. 512 Mg ww person^{-1} y^{-1}

$V_{\text{container}}$ = 660 L

filling = 80% (degree of filling of the container)

pop = 100,000 inhabitants

% waste material: share (wet basis) of the waste material in the mixed waste (Table S2)

$\rho_{\text{waste material}}$ = density of the waste material fraction (see below)

segregation eff. = source segregation efficiency for the individual waste material (see below)

The density of the waste in the container (Mg m^{-3}) was [14]:

- mixed household waste and residual waste: 0.16
- paper: 0.11
- plastic: 0.05
- organic: 0.3
- metals (ferrous, non ferrous): 0.1

3.3.2 Results

Results for the collection costs are reported in Table S3.

Table S3. Collection cost (€ Mg^{-1} ww) for the hypothetical region under assessment (100,000 inhabitants, multifamily houses, Denmark, 2013). The cost refers to one metric tonne of mixed household waste produced by the population.

Scenario	Collection (€ t^{-1} ww)					
	Organic	Paper	Plastic	Ferrous	Aluminium	Residual
A	-	-	-	-	-	36
B	-	-	-	-	-	36

3.4 Treatment cost

3.4.1 Calculation method

The treatment costs were calculated according to Eqs. (S10-S12). These represent a sort of future estimated gate-fee calculated with December 2013 prices.

For each parameter (e.g.: electricity, heat, materials, revenues, etc.) the total cost is obtained by multiplying the price (per unit) by the total amount consumed/produced. Table S4 details the main assumptions used for incineration, anaerobic digestion, and waste refinery (e.g. amount of electricity and heat produced/consumed, recovery efficiency for recyclable materials, etc.). The calculated amounts consumed/produced for the parameters involved in the economic analysis are listed in Table S5-S7.

$$Treatment\ \cos t\ =\ capital\ \cos t\ (amortisation)\ +\ ma\operatorname{int}enance\ +\ operation\ \cos t \qquad (S10)$$

$$Operation\ \cos t = manwork + chemicals + heat\ consumed + electricity\ consumed - heat\ prod - electricity\ prod$$
(S11)

$$Man - work = wage \times workhours \times working\ days \qquad (S12)$$

Where:

capital cost = see Table S1

maintenance = see Table S1

workhours = 8 hours d^{-1}

working days = 330 days y^{1}

wage = see Table S1

Table S4. Parameters used for the calculation of the treatment cost (gate fee): amount consumed/produced and corresponding source in this paper or in the literature. APC: air pollution control residues; BA: bottom ash; LHV: lower heating value of the mixed household waste (8.7 MJ kg^{-1} ww, see section 3.2 of this document).

Parameter	Unit	Incineration	Anaerobic digester	Waste refinery (Heat, enzymatic, seaving)
Water	kg Mg^{-1} ww	200 (wet flue gas cleaning)	-	400 (section 2.1 manuscript)
Electricity consumed	kWh Mg^{-1} ww	86 [13] [α]	40 [13] [β]	33 (section 2.1 manuscript)
Electricity produced	kWh Mg^{-1} ww	= LHV/3.6 x 25% = 2,416 x 25% [α]	= 4,486/3.6 x 40% [β]	= 690 x 0.52 (Fig. 3 and Table S3)
Electricity subsidy	kWh Mg^{-1} ww	-	-	-
Heat consumed	kWh Mg^{-1} ww	0	0	136 (section 2.1 manuscript)
Heat produced	kWh Mg^{-1} ww	= LHV/3.6 x 75% = 2,416 x 75% [α]	= 4,486/3.6 x 50% [β]	= 3,100 x 0.52 (Fig. 3 and Table S3)
Enzymes	kg Mg^{-1} ww	0	0	5 (section 2.1 manuscript)
Plastic recovered	kg Mg^{-1} ww	0	0	= Hard plastic input x 90% [σ]
Ferrous recovered	kg Mg^{-1} ww	= Ferrous input x 85% [γ]	0	= Ferrous input x 85% [σ]
Non ferrous recovered	kg Mg^{-1} ww	= Non ferrous input x 70% [γ]	0	= Non ferrous input x 90% [σ]
APC produced	kg APC Mg^{-1} ww	= 4.3% x DM$_{waste}$ = 4.3% 0.52 x 1000 [ε]	0	0
BA produced	kg BA Mg^{-1} ww	= 23% x DM$_{waste}$ = 4.3% 0.52 x 1000 [ε]	0	0
Maintenance	-	1	1	1
Man-work	man Mg^{-1} ww	30 (assumption)	8 (assumption)	8 (assumption)
National tax CO_2	Mg CO_2 Mg^{-1} ww	0.3 (based on C$_{foss}$ content, Fig. 4)	0	0.18 (based on C$_{foss}$ content, Fig. 4)
National tax heat	GJ heat	= Heat sold x 3.6/1000	= Heat sold x 3.6/1000	= Heat sold x 3.6/1000

α Gross electricity and heat efficiency at the incinerator was assumed 25% and 75% (see section 2.1 of this document) relative to the LHV$_{wb}$ of the waste. The electricity consumption was assumed 86 kWh Mg^{-1} ww conformingly with [13] (see SI of [13]).

β Gross electricity and heat efficiency at the engine was assumed 40% and 50% (see section 2.1 of this document) relative to the biogas-energy. The biogas-energy produced by a typical metric tonne of wet organic waste was assumed equal to 4,486 MJ Mg^{-1} ww corresponding to a practical methane production of 126 Nm^3 CH_4 Mg^{-1} ww conformingly with [13]. The electricity consumption for the digestion process was assumed 40 kWh Mg^{-1} ww conformingly with [13] (see SI of [13]).

γ Conformingly with the recovery efficiencies assumed in [13] (see SI of [13]).

σ Conformingly with the recovery efficiencies assumed in [13] (see SI of [13]).

ε The transfer coefficients of the dry matter to APC and BA were assumed 4.3% and 23%, respectively, as documented in section 2.1 of this document.

3.4.2 Results

Results for the treatment cost (i.e. estimated gate fee) are reported in Table S5-S7. Table S8 summarizes the total final cost of the waste management systems A and B.

Table S5. Calculation of the treatment cost for incineration of 1 Mg of wet household waste. BA: bottom ash; APC: air pollution control residues; ww: wet waste. Negative values indicate savings.

Parameter	Price	Unit	amount	Unit	Cost ($€\,t^{-1}$ ww)
Capital cost	75.6	$€\,Mg^{-1}$ ww	1	-	75.6
Water	0.0007	$€\,kg^{-1}$	200	$kg\,Mg^{-1}$ ww	0.1
Electricity consumed*	0.035	$€\,kWh^{-1}$	86	$kWh\,Mg^{-1}$ ww	3
Electricity produced	0.035	$€\,kWh^{-1}$	-604	$kWh\,Mg^{-1}$ ww	-21
Electricity subsidy	-	$€\,kWh^{-1}$	-	$kWh\,Mg^{-1}$ ww	0
Heat consumed*	0.042	$€\,kWh^{-1}$	0	$kWh\,Mg^{-1}$ ww	0
Heat produced	0.042	$€\,kWh^{-1}$	-1812	$kWh\,Mg^{-1}$ ww	-77
Enzymes	2	$€\,kg^{-1}$	0	$kg\,Mg^{-1}$ ww	0
Sales of plastic	0.043	$€\,kg^{-1}$	0	$kg\,Mg^{-1}$ ww	0
Sales of ferrous	0.088	$€\,kg^{-1}$	-36	$kg\,Mg^{-1}$ ww	-3.1
Sales of non ferrous	0.4	$€\,kg^{-1}$	-17	$kg\,Mg^{-1}$ ww	-6.9
Disposal APC	73	$€\,Mg^{-1}$ APC	0.02	$kg\,APC\,Mg^{-1}$ ww	1.6
Disposal BA	3.7	$€\,Mg^{-1}$ BA	0.12	$kg\,BA\,Mg^{-1}$ ww	0.4
Maintenance	7.3	$€\,Mg^{-1}$ ww	1	-	7.3
Man-work	33	$€\,man^{-1}\,h^{-1}$	0.3	$man\,h\,Mg^{-1}$ ww	8.7
National tax CO_2	22.3	$€\,Mg^{-1}\,CO_2$	0.3	$Mg\,CO_2\,Mg^{-1}$ ww	7.2
National tax heat	8	$€\,GJ^{-1}$	6.5	$GJ\,Mg^{-1}$ ww	52.2
Total cost (without subsidy/tax)					**-11**
Total cost (with subsidy/tax)					**48**

*Assumed as own consumption (i.e. same price for produced/sold and consumed/bought).

Table S6. Calculation of the treatment cost for anaerobic digestion of 1 Mg wet organic waste. BA: bottom ash; APC: air pollution control residues; ww: wet waste. Negative values indicate savings.

Parameter	Price	Unit	amount	Unit	Cost ($€\,t^{-1}$ ww)
Capital cost	25.7	$€\,Mg^{-1}$ ww	1	-	25.7
Water	0.0007	$€\,kg^{-1}$	-	$kg\,Mg^{-1}$ ww	0
Electricity consumed*	0.035	$€\,kWh^{-1}$	40	$kWh\,Mg^{-1}$ ww	1.4
Electricity produced	0.035	$€\,kWh^{-1}$	-498.4	$kWh\,Mg^{-1}$ ww	-17
Electricity subsidy	0.047	$€\,kWh^{-1}$	-498.4	$kWh\,Mg^{-1}$ ww	-23.4
Heat consumed*	0.042	$€\,kWh^{-1}$	0	$kWh\,Mg^{-1}$ ww	0
Heat produced	0.042	$€\,kWh^{-1}$	-623	$kWh\,Mg^{-1}$ ww	-26
Enzymes	2	$€\,kg^{-1}$	0	$kg\,Mg^{-1}$ ww	0
Sales of plastic	0.043	$€\,kg^{-1}$	0	$kg\,Mg^{-1}$ ww	0
Sales of ferrous	0.088	$€\,kg^{-1}$	0	$kg\,Mg^{-1}$ ww	0
Sales of non ferrous	0.4	$€\,kg^{-1}$	0	$kg\,Mg^{-1}$ ww	0
Disposal APC	73	$€\,Mg^{-1}$ APC	0	$kg\,APC\,Mg^{-1}$ ww	0
Disposal BA	3.7	$€\,Mg^{-1}$ BA	0	$kg\,BA\,Mg^{-1}$ ww	0
Maintenance	7.3	$€\,Mg^{-1}$ ww	1	-	7.3
Man-work*	33	$€\,man^{-1}\,h^{-1}$	0.1	$man\,h\,Mg^{-1}$ ww	2.9
National tax CO_2	22.3	$€\,Mg^{-1}\,CO_2$	-	$Mg\,CO_2\,Mg^{-1}$ ww	-
National tax heat	8	$€\,GJ^{-1}$	-	$GJ\,Mg^{-1}$ ww	-
Total cost (without subsidy/tax)					**-6**
Total cost (with subsidy/tax)					**-30**

*Assumed as own consumption (i.e. same price for produced/sold and consumed/bought).

Table S7. Calculation of the treatment cost for the waste refinery including downstream energy recovery processes. BA: bottom ash; APC: air pollution control residues; ww: wet waste. Negative values indicate savings. The capital and maintenance costs for incineration were calculated from Table S5 knowing that 1 Mg ww treated in waste refinery produces 0.215 Mg wet residual solids (see section 3.2 of this document). The electricity and heat revenues were based on the amount incinerated and related LHV (13.6 MJ kg^{-1} ww). For anaerobic digestion of the bioliquid, it was assumed that 1 Mg bioliquid is equivalent to treating 1 Mg organic waste from a capital and maintenance cost perspective (the substrates are similar for properties and water content). It was also assumed that 1 Mg ww household waste would generate approximately 1 Mg wet bioliquid.

Parameter	Waste refinery (heat, enzymatic, seaving)					Anaerobic digestion (bioliquid)					Incineration (residual solids)				
	Price	Unit	amount	Unit	Cost (€ t^{-1} ww)	Price	Unit	amount	Unit	Cost (€ t^{-1} ww)	Price	Unit	amount	Unit	Cost (€ t^{-1} ww)
Capital cost & royalty	42.9	€ Mg^{-1} ww	1	-	42.9	25.7	€ Mg^{-1} ww	1.0	-	25.7	75.6	€ Mg^{-1} ww	0.215	-	16.2
Water	0.7	€ Mg^{-1}	400	kg Mg^{-1} ww	0.3	0.7	€ Mg^{-1}	-	kg Mg^{-1} ww	0.1	0.7	€ Mg^{-1}	43	kg Mg^{-1} ww	0.1
Electricity consumed*	0.035	€ kWh^{-1}	33	kWh Mg^{-1} ww	1.2	0.04	€ kWh^{-1}	40	kWh Mg^{-1} ww	1.4	0.035	€ kWh^{-1}	18.5	kWh Mg^{-1} ww	0.6
Electricity produced	0.035	€ kWh^{-1}	0	kWh Mg^{-1} ww	0	0.04	€ kWh^{-1}	-358	kWh Mg^{-1} ww	-12.5	0.035	€ kWh^{-1}	-203	kWh Mg^{-1} ww	-7.1
Electricity subsidy	0.047	€ kWh^{-1}	0	kWh Mg^{-1} ww	0	0.05	€ kWh^{-1}	-358	kWh Mg^{-1} ww	-16.8	0	€ kWh^{-1}	0	kWh Mg^{-1} ww	0
Heat consumed*	0.042	€ kWh^{-1}	136	kWh Mg^{-1} ww	5.7	0.042	€ kWh^{-1}	0	kWh Mg^{-1} ww	0	0.042	€ kWh^{-1}	0	kWh Mg^{-1} ww	0
Heat produced	0.042	€ kWh^{-1}	0	kWh Mg^{-1} ww	0	0.042	€ kWh^{-1}	-447	kWh Mg^{-1} ww	-19	0.042	€ kWh^{-1}	-609	kWh Mg^{-1} ww	-26
Enzymes	2	€ kg^{-1}	5	kg Mg^{-1} ww	10	2	€ kg^{-1}	0	kg Mg^{-1} ww	0	2	€ kg^{-1}	0	kg Mg^{-1} ww	0
Sales of plastic	0.043	€ kg^{-1}	-89.0	kg Mg^{-1} ww	-3.8	0.04	€ kg^{-1}	0	kg Mg^{-1} ww	0	0.043	€ kg^{-1}	0	kg Mg^{-1} ww	0
Sales of ferrous	0.088	€ kg^{-1}	-35.8	kg Mg^{-1} ww	-3.1	0.09	€ kg^{-1}	0	kg Mg^{-1} ww	0	0.088	€ kg^{-1}	-7.7	kg Mg^{-1} ww	-0.7
Sales of non ferrous	0.4	€ kg^{-1}	-22.2	kg Mg^{-1} ww	-8.9	0.4	€ kg^{-1}	0	kg Mg^{-1} ww	0	0.4	€ kg^{-1}	-3.7	kg Mg^{-1} ww	-1.5
Disposal APC	73	€ Mg^{-1} APC	0.0	kg APC Mg^{-1} ww	0	73	€ Mg^{-1} APC	0	kg APC Mg^{-1} ww	0	73	€ Mg^{-1} APC	0.005	kg APC Mg^{-1} ww	0.3
Disposal BA	3.7	€ Mg^{-1} BA	0.0	kg BA Mg^{-1} ww	0	3.7	€ Mg^{-1} BA	0	kg BA Mg^{-1} ww	0	3.7	€ Mg^{-1} BA	0.0026	kg BA Mg^{-1} ww	0.1
Maintenance	7.3	€ Mg^{-1} ww	1.0	-	7.3	7.3	€ Mg^{-1} ww	1	-	7.3	7.3	€ Mg^{-1} ww	0.215	-	1.6
Man-work	33	€ man^{-1} h^{-1}	0.088	man h Mg^{-1} ww	2.9	33	€ man^{-1} h^{-1}	0.1	man h Mg^{-1} ww	2.9	33	€ man^{-1} h^{-1}	0.057	man h Mg^{-1} ww	1.9
National tax CO$_2$	22.3	€ Mg^{-1} CO$_2$	-	Mg CO$_2$ Mg^{-1} ww	-	22.3	€ Mg^{-1} CO$_2$	-	Mg CO$_2$ Mg^{-1} ww	-	22.3	€ Mg^{-1} CO$_2$	0.18**	Mg CO$_2$ Mg^{-1} ww	4
National tax heat	8	€ GJ^{-1}	-	GJ Mg^{-1} ww	-	8	€ GJ^{-1}	-	GJ Mg^{-1} ww	-	8	€ GJ^{-1}	1.4	GJ Mg^{-1} ww	11.2
Total cost (without subsidy/tax)					**55**					**6**					**-14**
Total cost (with subsidy/tax)					**55**					**-11**					**7**

*Assumed as own consumption (i.e. same price for produced/sold and consumed/bought). **Based on the C$_{foss}$ content of the residual solids incinerated (Fig. 4: 94 kg C x 0.52 x 44/12 = 179 kg CO$_2$) where 0.52 is the dry matter content and 44/12 is the molar ratio CO$_2$ to C (Table S2 of this document).

3.5 Total cost of the waste management scenarios

Table S8. Total cost (€ Mg^{-1} ww) of the whole waste management system for the scenarios A and B. AD: anaerobic digestion; INC: incineration; WR: waste refinery. In brackets are the costs when tax/subsidy is not included in the analysis.

Scenario	Collection	Treatment					Total
	Residual	WR	AD	INC	Recycling		
A	36	55	-11 (6)	7 (-14)	Included in WR	87 (83)
B	36			48 (-11)	Included in INC	84 (25)

3.6 Sensitivity analysis: the influence of heat and electricity price, and energy recovery

A sensitivity analysis (S1) was performed to illustrate the importance of heat, electricity price, and energy recovery on the results. Table S9 shows the cost of the waste management system for different heat and electricity prices. These were compared with the baseline scenario used in the analysis. The Danish heat tax was maintained proportional to the total heat revenue quantified for the baseline (i.e. tax = 68% of heat revenue, rough assumption).

Another sensitivity analysis was performed to assess the influence of the energy recovery at the waste refinery (S2) and at the incineration (S3), as this affects the revenues from heat and electricity. For the waste refinery (S2) it was assumed that also hard plastic (i.e. HP, LHV=2800 MJ / 89 kg = 31.5 MJ kg^{-1} DM hard plastic, based on the flows reported in Fig. 3 of the manuscript) was incinerated for energy recovery instead of being recycled. This changed the amount of residual solids incinerated to 380 kg DM (i.e. 282 kg ww, recalculated per wet matter) and the LHV of the residual solids to 14.3 MJ kg^{-1} ww (HHV=21.5 MJ kg^{-1} DM residual solids with hard plastic, calculated based on flows reported in Fig.3 of the manuscript; water content assumed 70%). For incineration (S3), it was assumed a lower electricity recovery (20% of the LHV of the waste, instead of 25%). The results are reported in Table S10-S11.

The results highlight that the assumption regarding the revenues for heat and, also, electricity as well as the assumptions regarding the performance of incineration (i.e. electricity recovery) and the management of hard plastic (recycled or incinerated) may only slightly change the ranking of the scenarios. Overall, considered the uncertainties related to these parameters, the two alternatives appeared comparable from a cost perspective.

Table S9. Total cost (€ Mg^{-1} ww) of the whole waste management system under different heat and electricity prices (S1). Costs include taxes and subsidies. El: electricity.

Scenario (El. price 35 € MWh^{-1})	Heat price: 0 € MWh^{-1} (No heat recovery)	Heat price: 10 € MWh^{-1}	Heat price: 20 € MWh^{-1}	Heat price: 30 € MWh^{-1}	Baseline results (Heat price: 42 € MWh^{-1})
A	108	103	98	93	87
B	108	103	97	91	84

Scenario (El. price 50 € MWh^{-1})	Heat price: 0 € MWh^{-1} (No heat recovery)	Heat price: 10 € MWh^{-1}	Heat price: 20 € MWh^{-1}	Heat price: 30 € MWh^{-1}	Heat price: 42 € MWh^{-1}
A	101	96	91	86	80
B	101	95	89	83	76

Table S10. Total cost (€ Mg^{-1} ww) of the whole waste management system under different hard plastic management (S2). Costs include taxes and subsidies. El: electricity. INC: incineration. WR: waste refinery.

Scenario	S2 (Hard plastic to incineration)	Baseline results (Hard plastic to recycling)
A	85	87
B	84	84

Table S11. Total cost (€ Mg^{-1} ww) of the whole waste management system under different electricity recovery (S3). Costs include taxes and subsidies. El: electricity. INC: incineration. WR: waste refinery.

Scenario	S3 (El. recovery INC=20%)	Baseline results (El. recovery INC=25%)
A	88	87
B	89	84

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