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Quantifying the separation complexity of mixed plastic waste streams with statistical entropy: a plastic packaging waste case study in Belgium

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Abstract

Mixed plastic waste streams are to date present in nearly all societies. Depending on the source of the plastic waste stream, the complexity and difficulty to separate and recycle the waste stream differs. In this paper, the concept of statistical entropy is used to quantify the separation complexity of mixed plastic waste streams. To this end, the recently proposed multilevel statistical entropy analysis method is extended by adding a *multi-product system level*. Furthermore, an overview is presented of the research questions that can be addressed by different statistical entropy definitions. The proposed extended method is applied to a plastic packaging waste case study in Belgium for which the data are available in literature. The results indicate that the method based on statistical entropy allows to analyze the separation complexity of real-life mixed plastic waste streams. More specifically, the multilayer films contribute the most to the separation complexity of the studied plastic packaging waste stream. In addition, it is illustrated how the method can be used to identify key contributors to the separation complexity of mixed plastic waste streams and to evaluate measures to reduce the separation complexity of mixed plastic waste streams.

Keywords: Statistical entropy analysis, Recycling, Plastic waste, Waste management, Resource efficiency, Circular economy

Introduction

Plastics are an integral part of our daily lives and so is plastic waste. Although plastics are versatile materials which have been amply used and studied throughout the past century, their recyclability remains an issue. Plastics consist of diverse polymers which are mixed in different ways, at different scales and in addition a wide diversity of chemical additives exists. Exactly those properties that make plastics so desired as functional materials, make them difficult to recycle.

The potential of mechanical recycling hinges on the purity of waste plastic fractions, which in turn is subject to a tradeoff – higher purities come with lower yields and an inflation of small waste fractions.[1] Impurities are further propagated through a next life cycle of the plastics. Chemical recycling (pyrolysis for polyolefins, solvolysis for polycondensates) might be more lenient, yet is associated with lower yields, higher costs and higher energy consumption.[2,3] Moreover, downstream purification by mechanical methods is no longer possible (thus strictly requiring upstream mechanical refining), while downstream chemical separations following chemical recycling typically have a higher energy demand.

Nevertheless, in the search for new and exciting properties and appearance, the diversity of plastic materials is still increasing, despite calls to reduce the complexity of plastics.[4] The fact that mechanical recyclates with a quality similar to virgin materials have a favorable economic and environmental profile,[5,6] indicates that the plastics recyclability challenge is not enthalpic but rather entropic in nature.[7] In other words, wherever a clean plastic waste feedstock, i.e. having a low *entropy*, is present in sufficient quantity, mechanical recycling will be economically and environmentally favorable, given the valuable recyclates that can be obtained. However, attaining such clean waste feedstock may be unrealistic.

As the recycling of plastics plays an important role in the transition towards a more circular economy, generic methodologies and metrics are required to quantify the recyclability of plastics. Defining such metric remains a challenging task, due to the different factors that influence the recyclability of plastics,[8,9] as for instance the presence of different plastic types in mixed plastic waste streams,[10] degradation of plastics during the recycling process,[11 12] and presence of contaminations or compounds with potentially adverse health effects in the recyclate.[13] Yet, there is no methodology to quantify the complexity of plastics and its recyclability. Techno-economic assessment (TEA) and life cycle assessment (LCA), two state-of-the-art methodologies for quantitative sustainability assessment, have their limitations as these both rely on a background system for which detailed knowledge on the unit operations is available. This is typically not the case for new materials or products that are not yet on the market and for which there are no current systems are available, rendering methods as TEA and LCA practically not applicable. Moreover, given the high volumes of plastic waste global trade flows, it is a priori unknown to which kind of waste management system these materials will be subjected.[14]

Therefore, in this article, we propose a methodology that only relies on the innate material, component or product properties to quantify and predict recyclability and which is independent of the separation method used. More specifically, we hypothesize that the concept of statistical entropy can be used to quantify the separation complexity of mixed plastic waste streams and as such the recyclability in a generic manner. This is also motivated by the entropic nature of the plastics recyclability challenge. Statistical entropy analysis

(SEA) has initially been developed by Rechberger and Brunner[15] to study the distribution of individual substances in waste-to-energy plants and to investigate whether a waste treatment operation is either a diluting or concentrating operation. Later, this method was adopted for predominantly inorganic/metallic materials streams.[16–18] Parchomenko et al.[19] extended SEA to multilevel SEA, in which statistical entropies at the component (consisting of multiple substances, e.g. a multilayer drink carton) and product (consisting of multiple components, e.g. mixtures of different drink cartons) levels are considered. Multilevel SEA allows to analyze multi-component systems and the recyclability of components or products instead of being limited to only substances as in SEA. In our recent positioning paper,[20] we theoretically extended the multilevel SEA method to predict and/or assess the recyclability of plastics by including energy-related requirements (by a coupling with energy balances from generic transportation, sorting and refining technologies). However, as we show in this paper, the number of levels considered in the current multilevel SEA framework is insufficient to properly assess the mechanical and chemical recyclability of mixed plastic waste streams.

The subject of this contribution is to (*i*) extend the recently proposed multilevel statistical entropy analysis method by adding additional levels and (*ii*) to illustrate how the method can be used to identify and quantify which products contribute the most to the separation complexity of mixed plastic waste streams, in this case a plastic packaging waste case study in Belgium, based on real data from Roosen et al.[21] Note that this method can be applied to any type of waste mixture, as long as information about the composition is available. Despite the calls to increase the recyclability of plastics by reducing their complexity,[4] no tool exists at present to quantify and monitor the complexity of plastics. The proposed method can be used to steer, monitor and make policies to reduce the complexity and as such optimize the recyclability of plastics or plastic products. Moreover, the methodology can also be used to guide the design of a recycling system or a material recovery facility by quantifying the sorting efficiency of the different unit operations. As such, the statistical entropy based methodology allows to define a quantified basis for design for recycling, rather than the rules of thumb that are currently used.

Methodology

Need for a multi-product system level in multilevel SEA

The multilevel SEA method[19] divides a studied economic good into four different hierarchical levels: (*i*) substances, (*ii*) materials, (*iii*) components and (*iv*) products. Statistical entropies at the component and product level are defined to address different research questions (see[19,20] for more details). In the component level entropy, the material level is skipped and only substances are considered. The issue related to this is illustrated in the following example.

Consider the fictitious plastic waste collection bag filled with plastic bottles illustrated in Figure 1(A), similar to the case study presented in our earlier work.[20] The example studied in this paper consists of a plastic waste collection bag (product), in which two types of bottles (components) can be present: type 1 and type 2. Bottles of type 1 consist of a bottle cap, a monolayer PLA label and PET bottle material (materials). Bottles of type 2 consist of a bottle cap, a multilayer PLA+PE label and PET bottle material. These components consist of different substances: PET, PE, PLA and X. The levels that are defined in the multilevel SEA method are illustrated in Figure 1(B).

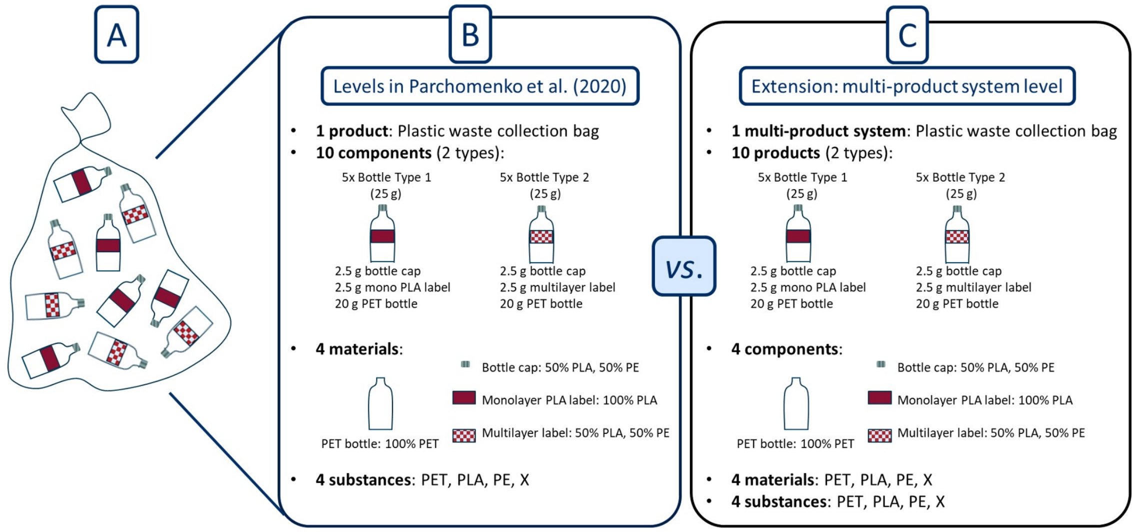


Figure 1: Illustration of the conceptual fictitious plastic waste collection bag filled with plastic bottles (A) using the levels defined in the multilevel SEA method (B) and using the extension with a multi-product system level (C).

As the existing multilevel SEA method[19] shortcuts the material level, the bottle caps, multilayer labels, monolayer labels and bottle material are not considered as levels to which the product can be recycled. Hence, the number of levels considered in the current method limits its applicability to evaluate the separation complexity or recyclability (by means of mechanical recycling) of the plastic waste collection bag towards bottle caps, bottle material, the monolayer and multilayer labels, but not towards its substances (PET, PLA, PE and X). Thus, it cannot distinguish the separation complexity in terms of bottle caps, bottle material, the monolayer and multilayer labels. Therefore, there is a need to extend the multilevel SEA method to additional levels, depending on the complexity of the case study.

By adding a multi-product system level as illustrated on the right side of Figure 1(C), the plastic waste collection bag is considered as a multi-product system, the plastic bottles are considered as products, consisting of different components (i.e., the bottle caps, bottle material, monolayer and multilayer labels) which in turn consist of materials which correspond with the different substances (PET, PLA, PE and X) in this specific case study. Consequently, statistical entropies can be computed for the bottle caps, PET bottle material, monolayer labels and multilayer labels. Two modifications are needed in the multilevel SEA methodology: (*i*) the product level statistical entropy needs to be redefined, as this is no longer the highest hierarchical level and (*ii*) a novel multi-product system level statistical entropy should be defined. Now, the method can predict the separation complexity of the plastic-waste collection bag towards the bottle caps, bottle material, monolayer and multilayer labels.

Extension with a multi-product system level - statistical entropy definitions and their interpretation

In Table 1 a summary of the different research questions and applicable statistical entropy definitions is presented. A more detailed description of the statistical entropy definitions is presented in the Supporting Information. The statistical entropy definitions corresponding with research questions 5, 6, 9 and 10 in Table 1 are novel compared to the existing multilevel SEA method. The redefined product level statistical entropy definition (research question 5 in Table 1) depends explicitly on $q_{n,p}$ (number of entities of component n in a unit of product p), contrary to the product level entropy in the existing multilevel SEA method[19] (research question 4 in Table 1). As the product level is no longer the highest hierarchical level, the multiplication of $q_{n,p}$ with m_n^c (i.e., the mass fraction of component n in the studied system) (see component level entropy definition, research question 2 in Table 1) equals the mass fraction of products in the system and allows to capture the contribution of the diversity of the products to the statistical entropy of the multi-product system. The multi-product system level entropy is the entropy at the highest hierarchical level in the considered case studies in this article. A similar reasoning as in multilevel SEA[19] is followed to define the statistical entropy at the highest hierarchical level as a function of the lower levels (in multilevel SEA the product level entropy).

Note that only mathematical formulations for absolute statistical entropies are presented in Table 1. Relative statistical entropies (i.e., the statistical entropy value divided by a maximum statistical entropy value) could also be used to answer the research questions in Table 1, except for research questions 8, 9 and 10. In these cases different systems are compared to each other, which requires the use of absolute statistical entropies as a different basis (i.e. maximum statistical entropy) is used for the calculation of relative statistical entropies. It would not be a fair to compare the entropy of two different systems in a relative manner, as typically also the maximum statistical entropy of these systems differs. These maximum statistical entropy values at the different levels correspond with the situation in which the underlying units are diluted maximally.[15,19]

Table 1: Summary of research questions and corresponding statistical entropy definitions that can offer an answer to these, together with their mathematical formulations.

Research question	**Statistical entropy**	**Mathematical formulation**
1. How are the substances distributed over a material flow system?	Substance level entropy (SEA[15])	$H^i(c_{i,f}, m_{i,f}) = -\sum_{f=1}^{F} m_{i,f} c_{i,f} \log_2(c_{i,f})$ $c_{i,f} = \frac{X_{i,f}}{M_f},\ m_{i,f} = \frac{M_f}{\sum_{f=1}^{F} X_{i,f}}$
2. How are the substances distributed over the components in the system and how do these contribute to the entropy of the system?	Component level[19] entropy	$H_n^c(c_{i,n}, m_n^c) = -\sum_{i=1}^{I} m_n^c c_{i,n} \log_2(c_{i,n})$
3. How are the substances distributed over the components (so components themselves, independent of the system)?	Individual component level entropy	$H_{n,indiv}^c(c_{i,n}) = -\sum_{i=1}^{I} c_{i,n} \log_2(c_{i,n})$
4. How are the substances and components distributed over the product (product as highest hierarchical level)?	Product level entropy[19]	$H^p(c_{n,p}, H_{n,rel}^c(c_{i,n}, m_n^c)) =$ $-\sum_{n=1}^{N} \log_2(c_{n,p})\, H_{n,rel}^c(c_{i,n}, m_n^c)$
5. How are the substances and components distributed over the products in the multi-product system (i.e., the highest hierarchical level)	Redefined product level entropy[1]	$H_p^{prod}(q_{n,p}, c_{n,p}, H_{n,rel}^c(c_{i,n}, m_n^c)) =$ $-\sum_{n=1}^{N} \frac{q_{n,p} q_{p,sys}}{q_{n,sys}} \log_2(c_{n,p})\, H_{n,rel}^c(c_{i,n}, m_n^c)$
6. How diluted/complex is the multi-product system under study?	Multi-product system level entropy	$H^{sys}(c_{p,sys}, H_{p,rel}^{prod}(c_{n,p}, H_{n,rel}^c(c_{i,n}, m_n^c))) =$ $-\sum_{p=1}^{P} \log_2(c_{p,sys})\, H_{p,rel}^{prod}(c_{n,p}, H_{n,rel}^c(c_{i,n}, m_n^c))$
7. How are the substances and components distributed over the products, independent of the system?	Individual product level entropy	$H_{p,indiv}^{prod}(c_{n,p}, H_{n,indiv}^c(c_{i,n}, m_n^c)) =$ $-\sum_{n=1}^{N} q_{n,p} \log_2(c_{n,p})\, H_{n,indiv}^c(c_{i,n}, m_n^c)$
8. When comparing different systems (e.g. S systems s): Which system has the greatest entropy in terms of components/is the most complex to separate in terms of components into its substances?	Absolute component level entropy	$H_n^{c,abs}(c_{i,n}, m_n^c) =$ $\left(-m_n^c \sum_{i=1}^{I} c_{i,n} \log_2(c_{i,n})\right) \left(\frac{M_{sys,s}}{\sum_{s=1}^{S} M_{sys,s}}\right)$
9. When comparing different systems (e.g. S systems s): Which system has the greatest entropy in terms of products/is the most complex to separate in terms of product into its components and substances ?	Absolute Redefined product level entropy	$H_p^{prod,abs}(q_{p,sys}, q_{n,p}, c_{n,p}, H_n^c(c_{i,n}, m_n^c)) =$ $\left(-\sum_{n=1}^{N} \frac{q_{n,p} q_{p,sys}}{q_{n,sys}} \log_2(c_{n,p})\, H_n^c(c_{i,n}, m_n^c)\right) \left(\frac{M_{sys,s}}{\sum_{s=1}^{S} M_{sys,s}}\right)$
10. When comparing different systems (e.g. S systems s): Which system has the greatest entropy/is the most complex to separate into its products, components and substances?	Absolute Multi-product system level entropy	$H^{sys,abs}(c_{p,s}, H_p^{prod}(c_{n,p}, H_n^c(c_{i,n}, m_n^c))) =$ $\left(-\sum_{p=1}^{P} \log_2(c_{p,s})\, H_p^{prod}(c_{n,p}, H_n^c(c_{i,n}, m_n^c))\right) \left(\frac{M_{sys,s}}{\sum_{s=1}^{S} M_{sys,s}}\right)$

Remark:

[1] In the remainder of this paper, the redefined product level entropy is mentioned in the text as product level entropy.

Evaluation and interpretation of multilevel SEA results for the fictitious plastic waste collection bag example

Consider the following relevant research questions that can be analyzed with multilevel SEA for the fictitious plastic waste collection bag example:

- Which bottles contribute the most to the separation complexity of the plastic waste collection bag?
- What contributes most to the separation complexity of the system into its substances: the bottle cap, the bottle material or the labels?

The existing multilevel SEA,[19] with the levels defined in Figure 1(B), can only answer the first research question by calculating the component level entropy values (cfr. Table 1, research question 2), as there is no statistical entropy definition at the material level. The bottles of type 2 with a component level statistical entropy of 0.461 contribute more to the separation complexity than the bottles of type 1 with a lower statistical entropy value of 0.442 (see mathematical formulation for component level entropy in Table 1, research question 2).

The proposed extended multilevel SEA method, using the levels defined in Figure 1(C), allows to answer both research questions. The first research question is addressed by calculating the redefined product level statistical entropy values (see Table 1, research question 5). It is concluded that the bottles of type 2 contribute more to the separation complexity than those of type 1, due to the higher product level statistical entropy of type 2 bottles 0.260 compared with the redefined product level entropy of 0.174 of the type 1 bottles. The second research question, related to the contribution of the bottle cap, bottle material and labels, is addressed by calculating the component level entropies (see Table 1, research question 2). The separation complexity at the component level is caused by the bottle caps and the multilayer labels. From the component level statistical entropy values it is concluded that the bottle caps contribute twice (component level entropy of 0.10) as much to the separation complexity of the system than the multilayer labels (component level entropy of 0.5). This is due to the fact that there are twice as many bottle caps as multilayer labels. However, both components are equally diluted in their substances, such that if we would consider the complexity of the components themselves, the complexity would be comparable. More specifically, the individual component level entropies would be the same (cfr. Table 1, research question 3). The statistical entropies of the bottle material and monolayer label both equal zero as both the bottle material and the monolayer labels consist of a single substance. Therefore these do not contribute to the separation complexity of the components into its substances.

Results and discussion

A plastic packaging waste case study in Belgium

The case study is based on the data of Roosen et al.,[21] who characterized mixed waste plastics in the P+MD curbside collection bag system.[22] We consider the plastic packaging waste stream (multi-product system) as the highest hierarchical level in this case study. This plastic packaging waste stream, representing the average plastic composition of a plastic waste collection bag, consists of 102 products (of 8 product types): 24 PET bottles, 19 multilayer films, 14 PP trays, 12 PET trays, 11 monolayer films, 10 PE bottles, 9 (E)PS trays and 3 PP bottles. These products consist of 29 component types, such as (per bottle type and tray type) bottle residue, bottle caps, bottle labels, bottle material, tray residue, tray labels, tray lidding, tray material, monolayer film residue, monolayer film material, multilayer film residue, multilayer label and multilayer film material. In addition 12 materials are considered, assumed to correspond to the substances in our analysis of this case study: PET, PE, PP, EVOH, PA, PV(D)C, Al, PUR, EVA, (E)PS, Paper, X. More details about the different levels and the composition are summarized in Table 2. The bottle products have a mass of 20.0 g, the tray products have a mass of 15.0 g and the film products have a mass of 7.5 g.

For the plastic packaging waste case study following three research questions are formulated.

1. Which products are the most complex in terms of separation into constituting components and how do these products contribute to the overall separation complexity/entropy of the plastic packaging waste stream? See Table 1, research question 5.
2. Which components are the most complex in terms of separation into constituting substances and how do these components contribute to the overall entropy of the plastic packaging waste stream? See Table 1, research question 2.
3. What measures could be taken to reduce the separation complexity of the system? Does it make sense to collect some products separately? See Table 1, research question 10.

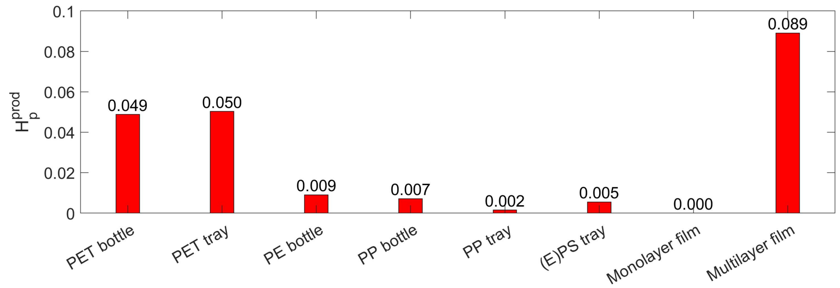
The first two questions are answered by computing the redefined product level entropy and the component level entropy. The third question is addressed by evaluating the absolute system level entropy and simulating different scenarios in which the plastic packaging waste stream is separated into different streams. Note that the levels considered in research questions 1 and 2 roughly correspond with energy classes for separations. In this context, mechanical recycling refers to the separation of a mixed plastic waste stream and its products into its constituting components by mechanical operations and chemical recycling refers to the separation and extraction of the different substances which is done chemically.[1] More specifically, the redefined product level entropy (research question 1) corresponds with an energy level required for mechanical recycling, while the component level entropy (research question 2) corresponds with an energy level required for chemical recycling. As illustration based on literature values, the energy level required for chemical recycling (e.g., selective dissolution of multilayer labels corresponding with an energy consumption of 1.23 MJ/kg[23] is approximately 2.7 times higher than the energy level for mechanical recycling (e.g., energy consumption of 0.32 MJ/kg to get mechanically recycled pellets[23]).

Table 2: Overview of the different levels considered for the plastic packaging waste case study,[21] together with the number of products in the system, number of components in the different products and substance composition of the components.

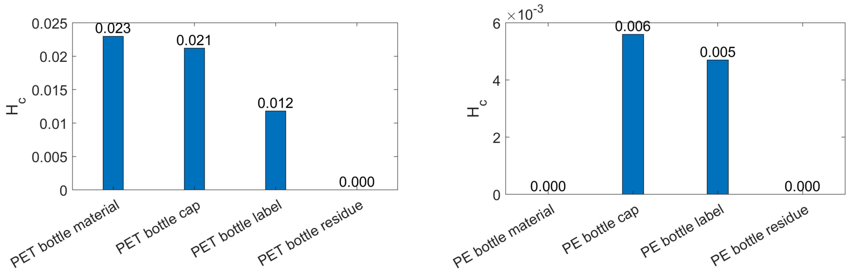
$q_{p,sys}$	**Product**	**Components** ($q_{n,p}$)	**Mass fraction substances in components (same as materials in this case study) (%)**											
			PET	PE	PP	EVOH	PA	PV(D)C	Al	PUR	EVA	(E)PS	Paper	X
24	PET bottles (20.0 g)	PET bottle residue (1)												100
		PET bottle cap (1)		80.65	19.35									
		PET bottle label (1)	25.93		59.26								14.81	
		PET bottle material (1)	98.90								1.10			
12	PET trays (15.0 g)	PET tray residue (1)												100
		PET Tray label (1)											100	
		PET tray lidding (1)	39.20	56.00		4.80								
		PET tray material (1)	91.67	8.33										
10	PE bottles (20.0 g)	PE bottle residue (1)												100
		PE bottle cap (1)		6.90	93.10									
		PE bottle label (1)	61.54	15.38									23.08	
		PE bottle material (1)		100										
3	PP bottles (20.0 g)	PP bottle residue (1)												100
		PP bottle cap (1)		62.77	37.23									
		PP bottle label (1)	30.77										69.23	
		PP bottle material (1)			100									
14	PP trays (15.0 g)	PP tray residue (1)												100
		PP tray label (1)	50										50	
		PP tray lidding (1)		50	50									
		PP tray material (1)			100									
9	(E)PS trays (15.0 g)	(E)PS tray residue (1)												100
		(E)PS tray label (1)	8.82									8.82	82.35	
		(E)PS tray lidding (1)	8.70	47.83	21.74	21.74								
		(E)PS tray material (1)										100		
11	Monolayer films (7.5 g)	Monolayer film residue (1)												100
		Monolayer film material (1)		100										
19	Multilayer films (7.5 g)	Multilayer film residue (1)												100
		Multilayer label (1)											100	
		Multilayer film material (1)	10.58	69.00	4.31	2.58	7.26	3.69	1.48	0.98	0.12			

Separation complexity of the products

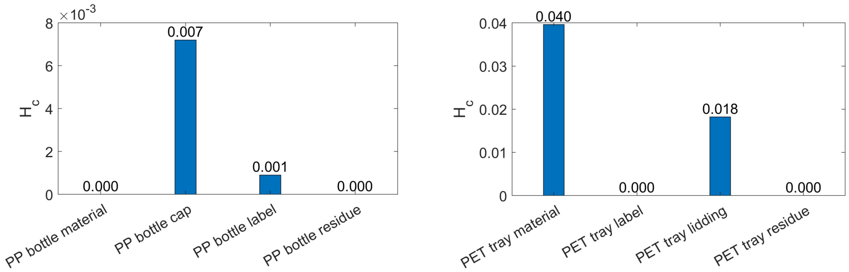
Figure 2(A) shows the redefined product level entropy H_p^{prod} values of the PET bottle, PE bottle, PP bottle, PET tray, PP tray, PP tray, (E)PS tray, monolayer film and multilayer film. Multilayer films contribute the most to the separation complexity of the plastic packaging waste stream as the product level statistical entropy is the greatest (H_p^{prod}=0.0891). This is due to the higher complexity of the multilayer components, i.e., the high number of distributed substances/materials and the high number of multilayer films in the plastic packaging waste stream (i.e., 19, second highest number after the PET bottles as indicated in Table 2). The PET trays (H_p^{prod}=0.0503) are the second largest contributor to the separation complexity of the plastic packaging waste stream, followed by the PET bottles (H_p^{prod}=0.0488). The monolayer films do not contribute to the separation complexity of the plastic packaging waste stream, as the product level entropy (see Figure 2(A)) and the related component level entropies (see Figure 2(H) equal zero. Both the monolayer film residue and the monolayer film material consist of an individual substance, leading to the minimum statistical entropy value of zero. This fact is also inherent to our definition of the hierarchical levels: defining a substance level that differs from the material level (e.g., materials consisting of a mix of substances) would increase the product level statistical entropy of the monolayer films. In practice, the number of monolayer films does have an effect on the separation complexity of the mixed plastic waste stream. If there are no monolayers present, the separation complexity is lower. Note that this analysis is purely done on the basis of statistical entropy, the enthalpic requirements for separations are not accounted for, as discussed in our earlier work.[20]



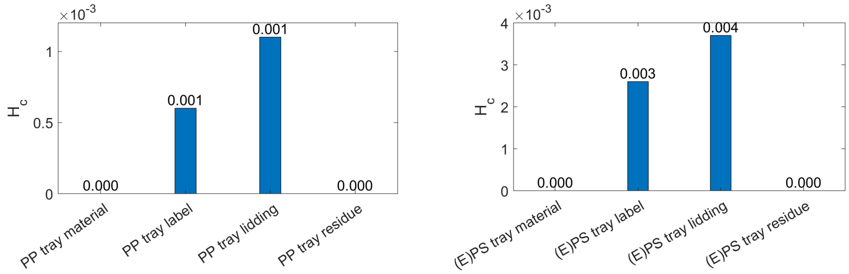
(A) Products



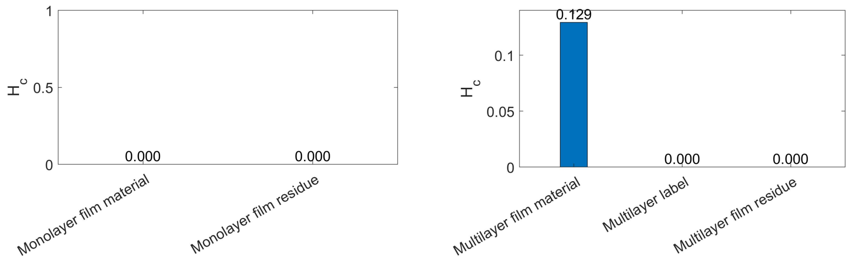
(B) PET bottle components **(C) PE bottle components**



(D) PP bottle components **(E) PET tray components**



(F) PP tray components **(G) (E)PS tray components**



(H) Monolayer film components **(I) Multilayer film components**

Figure 2: Statistical entropy values H_p^{prod} for the products (A) and statistical entropy values H_c for the components, grouped per product (B)-(I) in the plastic packaging waste stream case study.

Separation complexity of the components

The separation complexity of the products, discussed in the previous paragraph, is also closely related to the separation complexity of the components (for an overview of the components see third column in Table 2). The component level statistical entropies H_c are shown in the full bar charts in Figure 2. The multilayer film material has the highest separation complexity among the various materials (reflected by the highest component level entropy value H_c=0.1292), followed by the PET tray material (H_c=0.0396) and PET bottle material (H_c=0.0230). Note that all residue components are assumed to solely consist of the substance X, which is undetermined. In case these residue fractions consist of different substances, which is very likely, the results will be affected. In addition, the PE bottle material and PP bottle material are assumed to be pure, while the PET bottle material has some contaminants with EVA, influencing the contribution of the PET bottle material to the higher product level statistical entropy of the PET bottle, compared with the PP and PE bottles. Note that the presence of contaminations in the bottle material also increases the separation complexity, together with the number of components that are present in the plastic packaging waste stream. If one wants to reduce the statistical entropy, the multilayer film material would be a good first target to remove from the system.

Measures to reduce the separation complexity

In this paragraph different scenarios are studied as a thought experiment to avoid certain waste products in the plastic packaging waste stream and its impact to the entropy and separation complexity of the multi-product system. We want to stress that these scenarios are purely studied as demonstration of the potential of the methodology and do not reflect actual economically viable ideas. An overview of the studied scenarios is presented in Table 3, together with the absolute multi-product system level statistical entropy values. Contrary to the previous analyses, absolute mixed product system level statistical entropies are used to compare the different systems with each other (see also Table 1, research question 10).

Table 3: Overview of the scenarios of different multi-product waste streams together with their absolute multi-product system level entropy values $H^{sys,abs}$.

Scenario	Description	$H^{sys,abs}$
Scenario 0	Base case	0.431
Scenario 1	2 separate plastic packaging waste streams	
	(a) Plastic packaging waste stream without multilayer films	0.238
	(b) Multilayer films	0
Scenario 2	3 separate plastic packaging waste streams:	
	(a) PET bottles, PP bottles, PE bottles	0.0560
	(b) PET trays, PP trays, (E)PS trays	0.0693
	(c) Monolayer films and Multilayer films	0.0450

Note that a mixed plastic waste stream that consists of only one product type, as waste stream (b) in scenario 1 consisting only of multilayer films, has a multi-product system level entropy of zero, as $c_{p,sys} = 1$ leading to $\log_2(c_{p,sys}) = 0$. This means there is no complexity in separating the multi-product system into its different product types as it consists of only one product type. However, the separation complexity of the plastic packaging waste stream is also affected by the complexity of the constituting entities. Therefore, an aggregation of the product level entropies is defined in Equation (1), denoted here by $H^{sys,agg}$, as this reflects the separation complexity of the mixed plastic waste stream in terms of the number of products present in the waste stream and the complexity of these products in terms of their constituting components.

$$H_s^{sys,agg}(q_{p,s}, H_p^{prod}) = -\sum_{p=1}^{P} q_{p,s} H_p^{prod} \left(\frac{M_{sys,s}}{\sum_{s=1}^{S} M_{sys,s}} \right) \tag{1}$$

with $M_{sys,s}$ the mass of the multi-product system s, $q_{p,s}$ the number of products of product type p in system s.

$H^{sys,abs}$ and $H^{sys,agg}$ are 45% and 46% lower for the waste stream (a) in scenario 1 ($H^{sys,abs}$= 0.238, $H^{sys,agg}$=1.499) than in scenario 0 ($H^{sys,abs}$=0.431, $H^{sys,agg}$=2.795) as shown in Table 3, indicating that the separation complexity reduced when separating the multilayer films from the mixed-plastic waste stream. For the waste stream consisting of only multilayer films (scenario 1, waste stream (b)) $H^{sys,abs}$=0, which neglects the complexity of the products in its constituting components. The $H^{sys,agg}$ value however equals 1.297 indicating a more realistic complexity which is of a similar order of magnitude as the plastic packaging waste stream without multilayer films. In scenario 1 the multilayer films are collected and burned together with the residual municipal solid waste.

In scenario 2 we go one step further and evaluate bottles, trays and films that are separated into three different plastic waste streams. As expected, $H^{sys,abs}$, i.e. the separation complexity of the different systems, decreased substantially, compared to the base case (scenario 0). The waste stream consisting of the different types of bottles (a) has a $H^{sys,abs}$ or separation complexity that is 87% lower than the base case. The second waste stream consisting of the different types of trays (b) has a separation complexity that is 84% lower than the base case and the third waste stream consisting of the films (c) has a separation complexity of 89.6% lower than the base case. In addition it can be seen that the waste stream consisting of the trays has the highest absolute multi-product system level entropy and is the most diluted in its substances and components. Note that $H^{sys,agg}$ is not needed here, as it reflects the separation complexity of the different products present in the mixed-product waste stream in terms of its constituting components but does not account for the complexity in terms of separating the mixed-plastic waste stream into its constituting products. Therefore, we want to stress that $H^{sys,agg}$ should only be used in case one or more of the studied multi-product systems only contains products of the same type. In scenario 2 the energy burden of separating the different trays and films from the bottles is shifted to the households. To allow a full evaluation of the waste management systems, hierarchical levels should be extended to the geospatial distribution of these waste products within society.

Perspectives for future work

In the current framework, the material level (e.g. polymer blends) is not accounted for explicitly, which is a methodological limitation. In addition, plastics are complex polymeric substances built from monomers. The material properties of plastics and polymeric substances depend on distributions at the molecular level, e.g., the molar mass distribution, chain length distribution, degree of crystallinity, configuration, etc. These aspects, and thus the molecular level, need to be included when assessing the recyclability of plastics, together with statistical entropy definitions at the material and substance level. An additional challenge about recyclability is the way some materials are sprawled in society as this determines how efficient products can be collected at their end-of-life. Therefore, when designing new materials and products consisting of these materials optimally with respect to recyclability (design-for-recycling) or when designing more optimal collection schemes and evaluating different sorting and refining processes, also the anticipated geospatial distribution of the materials or products within society needs to be addressed. These levels will be addressed in future work.

In our recent positioning paper,[20] we highlighted the potential of the multilevel statistical entropy method (SEA) method to generically predict the recyclability of plastics and to this end proposed to extend multilevel SEA by a coupling with energy balances from generic transportation, sorting and refining technologies. This is also motivated by recycling technologies as reactive processing/extrusion in which entropy is not reduced, but less energy is needed with respect to separating the individual substances. These decomposition energies at the different levels are yet to be defined. A validation of such a method on real-life recycling technology case studies to which the present paper makes a first contribution, is imperative.

In the case study presented in this paper, the considered hierarchical levels for the statistical entropies correspond with energy classes to reduce the entropy of the plastic packaging waste stream. In future work, these levels should also be defined in this manner such that generic energy classes could be used and no explicit energy balances need to be established to evaluate and predict the recyclability of the waste stream.

Conclusions

In this paper the multilevel statistical entropy analysis method has been reviewed, extended and applied to quantify the separation complexity of mixed plastic waste streams. We have illustrated the opportunities and limitations of the current framework in assessing the separation complexity. In addition, we have clarified how mathematical formulations based on statistical entropy can be used to address different research questions related to the separation complexity (and recyclability) of mixed plastic waste streams. By applying the methodology on a real-life plastic packaging waste case study,[21] we have shown how multilayer materials introduce complexity in the separation system. We have illustrated how the multi-product system level and the statistical entropy functions defined at this level can help in assessing which products present in the plastic packaging waste stream contribute the most to the separation complexity. Perspectives for future work entail the definition of statistical entropy functions at the geospatial and molecular level and the definition of recyclability metrics in which decomposition energy aspects are also taken into account for which we already proposed possible avenues in our recent positioning paper.[20]

Author contributions Conceptualization, P.N and P.B.; methodology, P.N. and P.B.; software, P.N.; validation, P.N. and P.B.; formal analysis, P.N; investigation, P.N.; writing–original draft preparation, P.N.; writing–review and editing, P.N and P.B.; visualization, P.N.; supervision, P.B. All have read and agreed to the published version of the manuscript.

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Nomenclature

$c_{i,f}$	Mass fraction of substance i in material flow f.
$c_{i,n}$	Mass fraction of substance i in component n.
$c_{n,p}$	Number fraction of component n in product p.
$c_{p,s}$	Number fraction of products p in the multi-product system s.
$c_{p,sys}$	Number fraction of products p in the multi-product system.
f	Index running over the material flows.
F	Number of material flows.
H_n^c	Component level statistical entropy.
$H_n^{c,abs}$	Absolute component level statistical entropy.
$H_{n,indiv}^c$	Individual component level entropy.
$H_{n,rel}^c$	Relative component level statistical entropy.
H^i	Substance level statistical entropy.
H^p	Product level statistical entropy.[19]
H_p^{prod}	Redefined product level statistical entropy.
$H_{p,indiv}^{prod}$	Individual product level statistical entropy.
$H_{p,rel}^{prod}$	Relative product level statistical entropy.
$H_p^{prod,abs}$	Absolute refined product level statistical entropy.

H^{sys}	Multi-product system level statistical entropy.
$H^{sys,abs}$	Absolute multi-product system level statistical entropy.
$H_s^{sys,agg}$	Aggregation of the redefined product level statistical entropies.
i	Index running over the substances.
I	Number of different substances.
m_n^c	Mass fraction of component n in the system.
$m_{i,f}$	Standardized mass fraction of material flow f to the total flow of substance i.
M_f	Mass flow rate of material flow f.
$M_{sys,s}$	Mass of the multi-product system s.
n	Index running over the components.
N	Number of component types.
p	Index running over the products.
P	Number of product types.
$q_{n,p}$	Number of entities of component n,in product p.
$q_{n,sys}$	Number of entities of component n in the system.
$q_{p,sys}$	Number of units of product p.
s	Index running over multi-product systems.
S	Number of multi-product systems studied.
$X_{i,f}$	Mass flow rate of substance i in material flow f.

Supporting Information Available

The Supporting Information is available free of charge:

Nimmegeers and Billen 2021 Supporting info.pdf with a detailed description of the different statistical entropy definitions.

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Graphical abstract/Table of contents graphic and Synopsis

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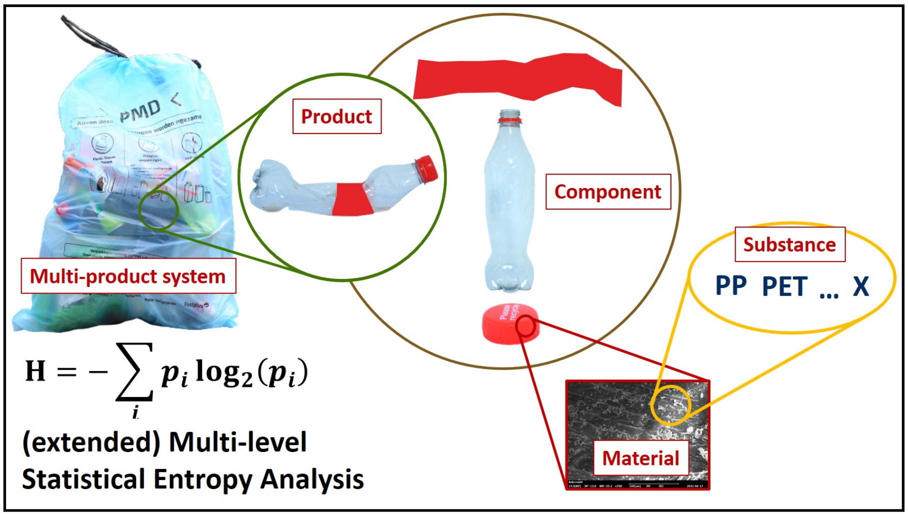

Synopsis: Statistical entropy definitions allow to quantify and propose measures to reduce the separation complexity of mixed plastic waste streams.