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Using Statistical Entropy Analysis to Predict the Degree of Circularity of Plastic Recycling Technologies

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Abstract

The outstanding growth of plastic production and consumption has brought to light the importance of recycling as one of the main pathways in the transition towards Circular Economy (CE). Plastic wastes are characterized by different chemical composition and contamination levels that prevent them from being processed via a single recycling technology. Today, plastic wastes are primarily recycled via mechanical or chemical recycling, based on energy absorption, capital expenditure, plastic structure's complexity prerequisites and value retention. However, scientific literature lacks a method able to predict the recycling technology to employ case by case. To this end, Statistical Entropy Analysis (SEA) is discussed in this dissertation as a tool to cover this gap, by introducing a recyclability indicator that can guide industrial actors in the identification of the best recycling technology based on the circularity degree of waste materials. The method is applied to the plastic municipal solid wastes produced in Lombardy in 2019, which are investigated at different aggregation levels: substances (polymers), components (plastic objects) and products (total wastes gathered). Entropy values are calculated at each level to describe the complexity of the plastic's structures, and then, coupled with information about recycling processes' energy requirements to generate the recyclability indicator. Based on this thesis' results, the SEA's recyclability indicators are then combined with ad hoc economic utility criteria to understand which recycling process is more circular based on the plastic structure's complexity of each case. In conclusion, the predictive behaviour of the SEA method is compared with the other existing circularity assessment methods, and future possible implications of the SEA are set forth.

Keywords: Statistical Entropy Analysis, Circular Economy, Plastics, Recycling, Recyclability Indicator, Lombardy

Sommario

L'incredibile crescita della produzione e del consumo della plastica ha portato alla luce l'importanza del riciclo come una delle vie principali nella transizione verso l'economia circolare (EC). I rifiuti di plastica sono caratterizzati da differenti composizioni chimiche e livelli di contaminazione che impediscono loro di essere processati tramite una singola tecnologia di riciclo. Al giorno d'oggi, i rifiuti plastica vengono principalmente riciclati tramite riciclo meccanico o chimico, la cui differenza dipende dall'energia assorbita, dall'ammontare di capitale necessario, dai prerequisiti riguardo la complessità della struttura delle plastiche e dal valore trattenuto durante processo. Tuttavia, nella letteratura scientifica manca un metodo capace di predire quale tecnologia di riciclo impiegare caso per caso. A questo fine, l'analisi di entropia statistica (AES), viene discussa in questa dissertazione come strumento capace di riempire questa mancanza, tramite l'introduzione di un indicatore di riciclabilità che può guidare gli attori industriali nell'identificazione della miglior tecnologia di riciclo sulla base del grado di circolarità dei rifiuti. Il metodo è applicato ai rifiuti solidi urbani di plastica generati in Lombardia nel 2019, che vengono analizzati su differenti livelli di aggregazione: sostanze (polimeri), componenti (oggetti di plastica) e prodotto (totale dei rifiuti raccolti). I valori di entropia vengono calcolati ad ogni livello per descrivere la complessità della struttura delle plastiche e, successivamente, accoppiati con le informazioni riguardanti la richiesta energetica dei processi di riciclo per generare l'indicatore di riciclabilità. Sulla base dei risultati di questa tesi, gli indicatori di riciclabilità della AES vengono uniti con dei criteri ad hoc di valutazione dell'utilità economica per capire quale processo di riciclo sia più circolare, tenendo conto della complessità della struttura plastica di ogni caso. In conclusione, il carattere predittivo del metodo AES vene paragonato con gli altri metodi esistenti di verifica della circolarità, e possibili implicazioni future del metodo AES vengono promosse.

Parole chiave: Analisi di entropia statistica, Economia circolare, Plastica, Riciclo, Indicatore di riciclabilità, Lombardia

Executive Summary

Plastics have become the most employed product in almost any industrial sector, increasingly replacing other materials. Besides a complete restructuring of the whole industrial sector, though, the plastic advent has brought one of the biggest problems of human history: its production, consumption and disposal generate pollution and environmental damages that have proved to be particularly struggling to cope with. Many initiatives and projects carried on by accredited authorities are gaining momentum to deal with the plastic issue over time, trying to find a solution to the extremely high amount of littering and landfilling that harm not only the planet but public health itself. The most promising modern-day philosophy is represented by the concept of Circular Economy (CE). Stemming from the wider concept of Industrial Ecology (IE), CE tries to assimilate the industrial pathways of products, services and processes as if they were natural processes, where the wastes become seeds for a new industrial cycle. CE aims to overcome the traditional linear economic approach of take-make-dispose, in favour of a new circular approach where the wastes are minimized and products are recovered at the end of their useful life. The CE is a broad concept that embeds several distinct definitions (reason why it is often referred as an "umbrella concept") and has its root in multiple dimensions, like the environmental, social and economic. Furthermore its employment can be assessed at different levels of applicability, depending on what is the scale of the initiatives that are thought to be implemented: they can focus on the single industrial context (micro level), on the pending relationship between more than one industry trying to study the existence of viable synergies (meso level) or on the applicability of circular statements at regional/national levels in order to create a basis for ad hoc policies generation (macro level).

In order to pursue the total CE penetration throughout all the levels, many methods to assess the circularity of a product/process have been created. The most acknowledged one is the Life Cycle Assessment (LCA) that investigates the circularity concept by analyzing the impact on the environment of industrial processes by performing a comparison between some alternatives, with the

aim of identifying which is the best from a CE perspective. Another vastly employed method is the Material Flow Analysis (MFA) that provides a perspective on the energy and material consumption of a reference scenario, with the aim of easing the identification of the critical spots where to intervene. None of the existing methods, though, takes into consideration that, each time a product undergoes a recycling cycle, its quality downgrades, reducing its economic utility. In other words, they neglect the thermodynamic perspective of the issue, focusing solely on the economic/environmental impacts of existing procedures, without any clue about how to proceed or which technological process to involve according to the specific material. The Statistical Entropy Analysis (SEA) emerges as a tool to cover this gap as it allows for computing a recyclability metric that can be used as a scoreboard to address each single waste item analyzed and the recycling technique that best fits the specific case, in accordance with the CE principles. Practically, the SEA method takes well-defined waste flows (within a given geographic area and time frame) and computes a recyclability indicator that takes into account both the complexity of the wastes' plastic structure and the energies needed to recycle them, where, the first, is reflected by the entropy that it is possible to attribute to each single plastic structure, while, the second, is the sum of the energies required to go through the entire recycling process. The analysis is targeted to understand how the recyclability indicator can help predict the degree of circularity of the plastic recycling technologies under investigation. In fact, different technologies can be described through a dimension termed "degree of circularity" that highlights the value that is possible to retain processing the plastic wastes. The higher the amount of value recovered during the transformation, the higher the degree of circularity of the technological intervention and vice versa. SEA's output, according to the entropy level of the plastic structure, includes the recyclability indicator of the waste material under analysis, and, leaning on this value, guides the selection of the recycling process based on the degree of circularity, namely high recyclability index allows high degree of circularity measures, while lower recyclability suggests a lower degree of circularity techniques.

The geographical scope of the analysis is the Lombardy region, in Italy, because it presents the highest level of plastic waste production, the highest number of plastic recycling facilities, and the highest plastic recycling rate (55%) in the entire national territory. The objects of analysis are the municipal plastic wastes produced in 2019 in the reference scenario. The selection of this particular type of products is guided by the fact that municipal solid waste (MSW) are the ones for which there is the majority of separate collection programs, thus it is easier to access the data regarding their amount. Within this scenario, mechanical and chemical recycling processes are reckoned as the two possible ways to recover value from plastics, where the first is associated with a higher degree of circularity and the second with a lower degree. The whole analysis is performed with the final intent to direct the plastic wastes to either mechanical recycling technologies, from which it is possible to obtain recycled finite products that can be reintroduced in a market; or chemical treatments that, instead, have the objective of producing substitute raw materials by breaking the plastic wastes into their elementary monomers/polymers.

In this thesis, the SEA method is developed on a multilevel structure, the entropies are computed progressively following a hierarchical scheme starting with the substance level, representing the elementary building blocks of the plastic structures (polymers/resins); then at the component level, where distinct structured plastic items are taken into consideration; finally, at the product level where the amount of plastic waste is considered as a whole. The reason why distinct levels of aggregation are used in the analysis is due to the need of providing the most comprehensive information about how the recyclability indicators is computed, which factors concur to its final value, and, therefore, to understand potential sources of criticalities. Following a thorough mathematical scheme, the multilevel analysis ends with the identification of the relative entropy values of (in order) substances, components and products; where the entropy computed at lower levels contributes to obtaining the entropy for higher levels. The SEA method is, then, extended in order to introduce the reference to the recycling technologies into the recyclability indicator computation. The decomposition energies are introduced to link the entropies with the recycling processes to assess. Different objects could

have a similar plastic's structure complexity (similar entropy value), but they can significantly differ in terms of the energy required to treat them; for this reason, the final recyclability indicator is built upon both the dimensions. In this dissertation, data regarding mainly the components' level of aggregation has been considered: the decomposition energies fed into the calculation refer to the energy expense needed to transform a component into the substances and the recyclability indicator gives information about the distinct plastic components. This is because the component level is the one providing the most interesting insights from a managerial point of view since they are the ones undergoing the recycling process and they are the ones that can, then, generate new market opportunities as recycled plastic items.

For data collection, we reviewed existing literature to understand what are the main plastic's resins that can be found in the MSW, and what are the distinct plastic objects that compose the plastic waste gathered through separate collection (together with their proportion of occurrence). Then, through the identification of the pieces that concur in forming the components (in SEA vocabulary termed materials), we linked the substances to the components in order to get useful information about the way the substances are distributed among the plastic components (e.g., a plastic bottle represents the plastic component; its lid, label and plastic body are the materials; thus, knowing the substances' percentages that shape each material, we got the structure's complexity of the component). The subsequent calculation of the relative entropy can explain how the objects from a lower level of aggregation distribute into a higher level of aggregation (substances into components and components into product). The substance-level entropy is the ground zero analysis, it gives information about the dilution/concentration of the substances in general terms: if one resin is present in many components with similar proportion its entropy is high, while if it is present into few components with unbalanced percentages it is low. The component-level entropy is focused on the substances' dilution/concentration within each distinct plastic component: a component has high entropy if it is made by many substances with similar percentages, and low entropy if it is mainly made by one or few substances. Finally, the product-level entropy provides insights about the structural complexity

of the whole amount of MSW gathered according to the heterogeneity of components that can be found in the wastes. The decomposition energies we leaned on to build, together with the entropy values, the recyclability indicators refer to the energy expenditure needed to go through all the steps of mechanical recycling, since it is the procedure with the highest degree of circularity. It is important to say that the development of the SEA has been characterized by the introduction of some assumptions that have been required in order to overcome the problem of complex data accessibility. Among all the assumptions taken, the most relevant one is that we performed all the calculations considering just the first loop of recycling. It means that we treated the waste as if they had become wastes for the first time, so, neglecting almost entirely the impact of the repeated processing contamination that unavoidably characterizes the plastic structure of recycled products. Thus, the purer the plastic structure, the lower the entropy, the higher the recyclability indicator expected and so the higher the odds to be able to reprocess the wastes through the highest degree of circularity method, namely the mechanical recycling. In fact, the recyclability indicators obtained in the reference scenario show how all the plastic items could theoretically be recovered through mechanical recycling when the assumption of the first loop of recycling stands. In the case under consideration in this thesis, according to the results obtained, the mechanical recycling has proved to be the most circular recycling process, based on the higher recyclability values for all the plastic components considered.

To demonstrate the robustness of the results obtained, we carried out a sensitivity analysis, variating some specific input data to see whether and how the final results get modified accordingly. The first variation considers an increment of the number of components that comprehend in their assembly more than one material, with a consequent reduction of the number of the components made by one single material structure, with the aim to investigate whether the number of the components over total could have an impact on the relative entropy. The second variation is about the decomposition energies: in the reference case they have been assumed to be equal for all the components considered, through the sensitivity analysis we investigated how the recyclability indicator changed when,

instead, each component gets associated to its specific decomposition energy, variable according to their particular plastic structure. Both the variations have provided evidence of a change from the reference scenario's results: the first showing how the relative entropy depends not only on the chemical structure of the plastic items but also on their number, and so does also for the recyclability indicator; while the second variation has put the lights on the fact that the entropy itself is not sufficient to create a recyclability metric, since objects having the same structural complexity but involving different substances can have very different decomposition energies, hence different recyclability indicators. Then, in the last scenario considered, the decomposition energies referring to chemical recycling have been put in the calculations and the recyclability index of all the substances happened to decrease. It can be read as a sign of the fact that, due to higher energy expenditures, chemical recycling is considered as an option to recover value just when the recyclability indicator touches low values, as it is associated with a lower degree of circularity.

According to the insights obtained from results and sensitivity analysis, it is possible to conceptualize the implications of the SEA recyclability indicator as a tool to support the choice of recycling technologies: when the indicator is high, its complexity is low, therefore mechanical recycling can be performed and a sensible amount of value can be retained by the process. This procedure keeps going on until when the repeated reprocessing entails the purity of the plastic structure so highly that a further SEA performed would show a low value of the recyclability indicator, comporting the choice of the chemical recycling technique as the best suitable way despite its lower degree of circularity. In addition, it could be possible to couple the SEA's output with market information about the marginal revenue achievable from recycled products. Every recycling process reduces the embedded quality (and so the value) of the recycled plastics, in order to decide whether it is convenient to perform one other loop, the marginal cost of the recycling process gets compared to the marginal revenue that such quality-level could assure on the market. If there is economic sense in performing one other loop, then the recycling keeps going, otherwise a different option of value recovery gets selected. In this way the SEA method can be used to get in advance all the circularity interventions that can be selected

for the plastic wastes. Its predictive behavior defines its novelty among all the existing circularity assessing methods, and its capability to give the a priori perspective of how the issues should be tackled case by case, allows to use this method to design CE strategies on all the assessment levels of analysis (micro, meso and macro), and to consider future expansions of the method also to embrace other value chains.

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1 Introduction

Since its discovery, plastics have been widely employed in almost all the industrial sectors and aspects of everyday life, thanks to the mechanical and chemical features that make plastics exceptionally versatile for various purposes (Hsu et al., 2020). However, plastics are nowadays considered at the center of some of the most challenging and worrying problems of modern society: waste is piling up, collection struggles to keep up and recycling is troublesome and costly (EMF, 2016).

Plastics were discovered at the beginning of the 20th century, that is when the term "plastic material" was used for the first time; however, it was just with the coming of world-war II that their production and employment started an outstanding growth that continues today. From mid 20th century, plastic materials have overcome their military prevailing use and have exponentially permeated a multitude of roles in everyday life. Plastic packaging is a brilliant example of this shift from wood-, paper- or glass-made items that have been replaced by the more economically convenient plastics. On the downside, this market shift posed constraints related to the ecological impacts associated with production, consumption and end-of-life management of plastics (EMF, 2016).

Over the past years, a consistent body of literature has addressed the importance and urgency to transition from a linear "take – make – dispose" economic model towards a Circular Economy (CE) where products are retained and reused within material closed-loops (EMF, 2017). There is no univocal definition of CE, however several main traits are identifiable:

- reduction of the waste generation;
- decoupling from virgin material extraction;
- extension of a product's useful life through recycling, repairing, refurbish and remanufacture

In this perspective, the transition to a CE involves a rethinking of the way the plastic materials are produced, used and disposed of. In traditional linear economies, plastic products are used just one time (single-use plastics) before being thrown away. This trend results in almost 95% loss of their

value and material loss to landfills and incinerators, or, in the worst scenario, abandonment into the environment. In 2018, approximately 359 million metric tons of plastics were produced worldwide (Bassetti, 2020). The conductive thread that drives for a change is the unsustainability of the current situation, indeed, future projections are worrying. The International Energy Agency (IEA) states that plastic production will continue to grow in the next twenty years, indicating that it could reach 540 million metric tons by 2040, and the environment will be irreversibly damaged unless urgent actions take immediately place (Bassetti, 2020).

Several institutions take care of the plastics matter worldwide, trying to pave the way and set the guidelines for a CE to thrive, one of the most influential is the Ellen Macarthur Foundation's "New Plastic Economy Initiative" (EMF, 2017). It gives a vision of a circular economy for plastic in which it never becomes waste, developed in six key points:

1. Elimination of problematic or unnecessary plastic packaging through redesign, innovation, and new delivery models is a priority.
2. Reuse models are applied where relevant, reducing the need for single-use packaging.
3. All plastic packaging is 100% reusable, recyclable, or compostable.
4. All plastic packaging is reused, recycled, or composted in practice.
5. The use of plastic is fully decoupled from the consumption of finite resources.
6. All plastic packaging is free of hazardous chemicals, and the health, safety, and rights of all people involved are respected.

Several methodologies exist to compute and assess the level of circularity of a particular analyzed output, the most famous is the life cycle assessment (LCA): "*a science-based technique for assessing the impacts associated with entire product life cycles. LCA can provide technical support to CE decision-makers, to assess trade-offs of impacts on a variety of environmental impact indicators*" (Rebitzer, et al., 2004; Pennington, et al., 2004). So, in a CE optic, LCA provides a scheme that tells

which, and to what extent, is the impact of a product/process to the environment, shedding light on where and how it is possible to intervene for advancing the degree of circularity in the system (Boucher et al., 2020). Inside LCA, a relevant mention should be about the material flow analysis (MFA), that is focused on the quantification of the mass flow that crosses a well-defined reference system. This method tracks the number of materials that enter, stay and leave the system, hence providing information about the total consumption, the accumulation within a determined time interval and the quantity that leaves the system as waste (Rechberger & Brunner, 2016; OECD, 2008).

These methods address the objective of controlling a system and detecting possible areas of intervention to reach closed-loop systems. There is one question that these methods do not completely tackle, though, the so-called "thermodynamic limit" of those loops (Nimmegeers, et al., 2021; Martinez et al., 2019) . Linear economy states that, after the use, the product should be disposed of, thus concluding its path with the wastefulness of the embedded value (Sariatli, 2017). In a CE perspective, instead, after the use phase, a product should get back to its starting point and begin a new lifecycle, with a theoretical recovery of its residual value and mass. The problem that arises is that these cycles cannot go on in eternity due to a physical limit that stems from the intersection of the second law of thermodynamics and the industrial process (Walter, 2019). Considering a general product-making process in simple terms: energy is required to generate work, and work to produce goods. It is ought to specify that, in order to get to the final product, it must be considered not just the exact amount of energy needed to produce the good, but a higher amount. This is because real processes inevitably happen at a "lower than 1" efficiency, thus part of energy will be dissipated to fulfil the first and second thermodynamics' laws (Walter, 2019).

According to physics principles, therefore, the energy and the resources employed to get the result cannot be fully recovered, but part of them get unavoidably lost every time a process takes place. Every loop creates dissipation and entropy, attributed to losses in quantity (physical material losses, by-products) and quality (mixing, downgrading) (Walter, 2019). The "increase of entropy" principle

states that no matter how efficient a process is, the entropy will always become higher, in other words, the value that a product can offer drops.

Literature lacks a well-defined structured method to evaluate the level of entropy associated to a specific process or product, that would help understand whether going for a recycling loop would be feasible or not based on its entropy level. In principle, if the entropy level is rather low a new recycling loop is justified because the product's structure complexity is not so high as it hinders the recovery process (Nimmegeers, et al., 2021). Statistical entropy analysis (SEA) is a methodology that has shown initial success in filling this literature gap, evaluating CE strategies on the material, component and product levels to identify critical stages of resource and functionality losses (Nimmegeers, et al., 2021).

This dissertation aims to explore the potential of using SEA to measure the system's circularity via assessment of material and energy flows across the plastic's value chain in the Lombardy region, Italy. Accordingly, the following research question is set forth:

RQ: How can statistical entropy analysis help predict the degree of circularity of technological downstream interventions?

2 Background choice

2.1 Geographical scope

The dissertation's purpose is to investigate the transition toward a regional model of CE of plastics in the Lombardy region, in northern Italy. The choice of this geographical scope is due to several factors. First, this region presents a well-developed plastic industry. Its edge over the other regions is represented by the wide shared awareness about CE practices and European legislation about plastic recycling, not only companies but people themselves have more and more internalized the required actions contributing to a more sustainable future. Second, separate solid waste collection schemes have already been adopted by 99,8% of the municipalities (Regione Lombardia, 2019) with an outstanding efficiency: more than 72% of the urban waste is sent to separate collection (Regione Lombardia, 2019), explanatory if compared to data at national level that stand at 61,3% (ISPRA, 2020). Third, 90% of all collected plastic waste in Lombardy is diverted from landfills and recovered, this amount can be further split in recovery of raw (recycle), covering roughly two third of the total, and recovery of energy (incineration) which claims the remaining third (ARPA Lombardia, 2020).

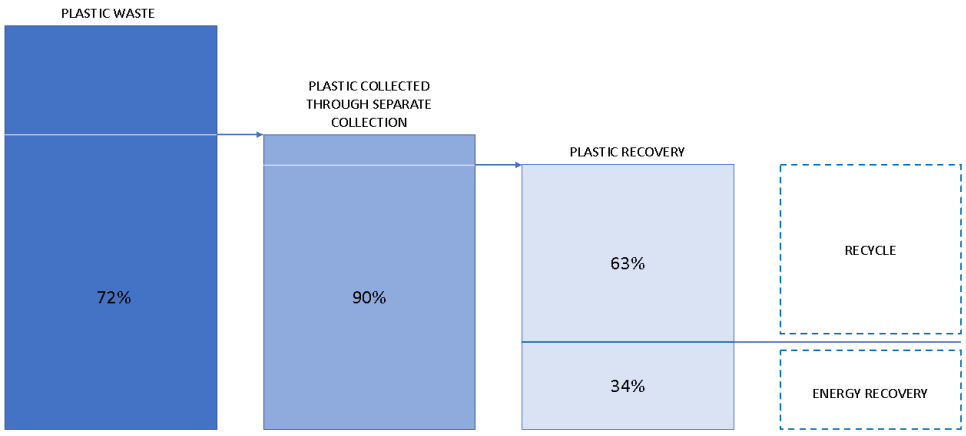

Figure 1 - Plastic waste management in Lombardy

Fourth, the Lombardy region well represents a good scenario where to perform this type of analysis also because of its availability and accessibility of data. Thanks to the "Osservatorio Rifiuti Sovraregionale (O.R.So)[1]"(ARPA Lombardia, "Produzione e gestione rifiuti in Lombardia", 2020), a database gathering all the information about the plastic wastes' production and flows.

Fifth, Lombardy's strength about recycling is not only due to the numbers, but also to the way it is performed. In fact, it is possible to find not only plants performing traditional mechanical recycling, but there is also initial evidence of thermo-chemical treatments, that can open a wide set of new possibilities in the recycling of plastics. This last concept paves the way to investigate a higher-level additional stream of recycling for plastics, including also those items that are cut off from traditional mechanical recycling processes.

2.2 Technological scope

Four main types of plastic recycling techniques exist which can be described by means of their degree of circularity (Al-Salem et al., 2009; Singh et al., 2017). The highest degree of circularity is obtained when the recycling process "makes a perfect loop", namely when the recycling process returns the same value starting product. It is represented by the primary recycling (or closed-loop mechanical recycling) and allows to retain the maximum amount of the embedded value (Al-Salem et al., 2010). Decreasing the degree of circularity there is the secondary recycling, which still belongs to the mechanical group of recycling procedures but goes under the name of open-loop recycling, since the output product is different from the starting one (Al-Salem et al., 2010). The recovery process downgrades the product's value causing a drop of its quality content. Tertiary recycling refers to all

[1] This database provides support to regional authorities in planning future projects and initiatives with the aim of achieving an effective environmental sustainability throughout the regional territory (ARPA, "Economia circolare in Lombardia", 2019). Its relevance is given by the fact that, to have at hand a reliable set of input waste data is crucial to provide with the analysis, information about the possible directions to go through to purchase a deeper penetration of CE practices in the reference context.

the chemical recovery procedures that, despite having promising future CE potentials, are associated to an even lower degree of recyclability (Al-Salem et al., 2010). It is because of the main current industrial applications of chemical recycling, which are destined to the generation of fuels and gases from the plastic waste. It is possible to lean on chemical recycling also to produce elementary monomers and polymeric chains that can act as substitutes of the virgin raw materials for plastic production, but this procedure is still under development and does not reach the commercial scale yet (Ragaert et al., 2017). Quaternary recycling refers to the energy recovery from incineration and has the lowest degree of circularity (Al-Salem et al., 2010).

Considering that only mechanical and chemical recycling involve the generation of a loop, they are the ones to be investigated in order to promote the widest CE penetration. The first consists of gathering and sorting plastic waste into similar polymers streams, which are then washed and reduced into flakes (or better, granulates) that will be then used as raw materials to produce new plastic products; the second, instead, sees the plastic waste streams to be depolymerized into constituting monomers and/or polymeric chains, through complex chemical processes (pyrolysis, gasification, hydrogenation), that can be used as a feedstock for the production of new petrochemicals and plastics (Ragaert et al., 2017; Dogu, et al., 2021). While mechanical recycling is suitable for only a limited portion of plastic waste (PE, PET, PP, PS) depending on their polymeric structure and on the contamination level, chemical recycling is theoretically able to recover value from all the polymers regardless the type (PVC, PS, PE, PP, PET, PU, PA, PLA, PC, PHA, PEF, PMMA, plasmix), including mixed materials plastics that are currently cut off from the existing recycling programs[2] (Hopewell et al., 2009). However, although it has a huge potential toward a CE penetration and a wide room for further developments, chemical recycling is still a too high capital-intensive practice: it requires more effort to decompose existing polymers than getting virgin ones, thus, it is way less employed in the industry than traditional mechanical technologies (Ragaert et al., 2017). Specifically,

[2] Mentioned recycling programs refer to mechanical recycle, which does not take into consideration such type of mixed plastics in the recovery process.

in Europe, less than 2% of collected plastic wastes are treated with chemical recycling technologies (PlasticsEurope, 2018). In addition, also the pollution footprint of chemical recycling is a matter of concern: if compared to the mechanical, chemical has a higher impact in terms of byproducts generation (Jeswani et al., 2020). Due to all these issues, it could likely happen that it results in being less economically appealing than mechanical recycling or even than producing new virgin materials. For this reason, in the analysis, mechanical recycling is taken as the reference way to assess the CE practices, while chemical recycling will be deemed as a viable option, when possible, acknowledging the lower presence of plants on the territory.

3 Literature review on existing metrics for measuring circular economy

The importance that the CE has gained through time is represented by the fact that there are many methods and frameworks to measure it (e.g., EMF, 2016, 2017; EC, 2015, 2020). The birth of the CE philosophy is a consequence of the aim of taking distance from the traditional economic approach in favor of a model able to reduce the harmful impact on the environment and social welfare. Therefore, measuring CE means understanding how far we are from the linear model, and whether the new industrial solutions will enhance more sustainable and circular scenarios (Bonacorsi, 2020). Having a good measurement system is crucial to continuously improve the achievements and to optimize the benefits, because it gives a way to visualize whether there are bottlenecks within a studied system, and, if so, to quantify their negative impacts. CE can be thought as an "*umbrella concept*" covering several aspects of economic and industrial processes and having roots into different dimensions, such as the technological, social and environmental spheres just to cite few (Moraga, et al., 2019). Indeed, literature still lacks a shared definition of it, each

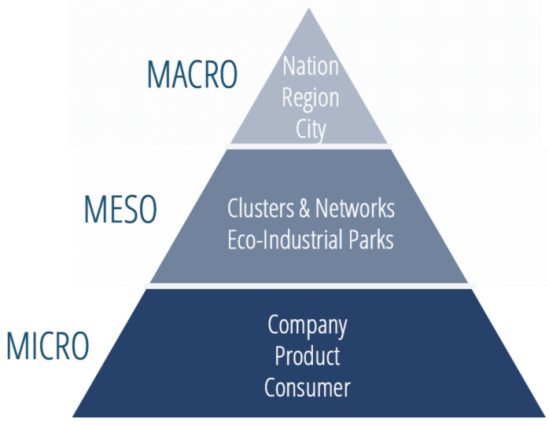

Figure 2 - Levels of CE analysis, Circular Regions – "What is Circular Economy"

formulation focuses only on few aspects, or, on some applicability fields (Kirchherr et al., 2017). Consequently, its heterogeneity is the reason why many different metrics to assess circularity exist; different indicators serve different purposes and some tools could be better in one situation, while others for different scopes (e.g., LCA focuses on the environmental impact of industrial practices, while MFA addresses the question more on resources management) (Saidani et al., 2017). Despite this variety, each metric used is, and must be, aligned with the ultimate goal of the analysis: achieving circular economy gains (Corona et al., 2019). A further perspective that is relevant to mention when it comes to talk about CE's assessment metrics, is the level of analysis of CE measurements (figure 2): activities referring to regions, cities and municipalities belong to the macro level; actions on eco-industrial networks and linkages between industries compose the meso level; and company- or even single industry-related activities form the micro level (Barreiro-Gen & Lozano, 2020; Ghisellini et al., 2015). According to the purpose of this study, the level of interest refers to the micro one, where the recyclability of plastic waste stream is analyzed.

3.1 Life cycle assessment

Among the frameworks employed to investigate the circularity a relevant role is represented by the life cycle assessment (LCA). It helps to quantify the environmental pressures related to goods and services (products), the environmental benefits, the trade-offs and areas for achieving improvements considering the full life cycle of the product (EC, 2022): from raw material extraction through processing, manufacturing, distribution, use, disposal, and, to a limited degree,

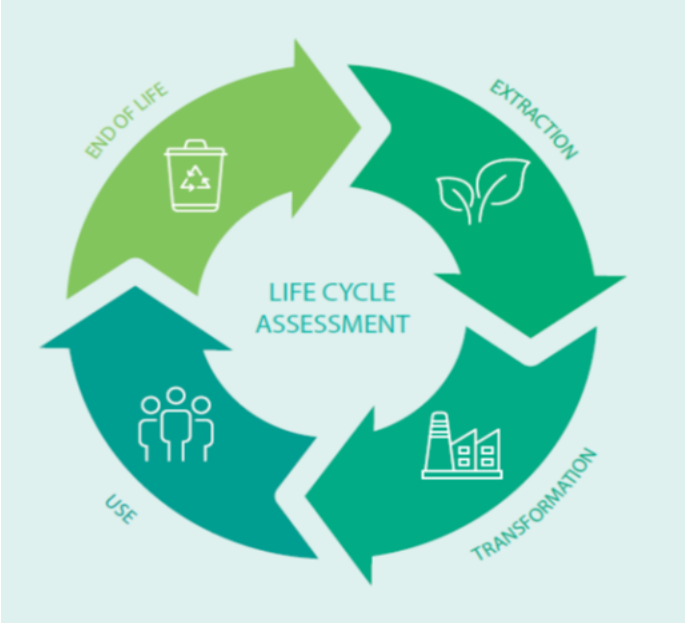

Figure 3 - Life Cycle Assessment, FibreNet "LCA: Benefit and Limitations", 2018

recycling (figure 3) (Shen et al., 2010; Gu et al., 2017). LCA definition is standardized from the international organization for standardization (ISO), as "*a technique for assessing the environmental aspects and potential impacts associated with a product by: compiling an inventory of relevant inputs and outputs of a system; evaluating the potential environmental impacts associated with those inputs and outputs; and interpreting the results of the inventory and impact phases in relation to the objectives of the study*" (UNI EN ISO 14040). A functional unit, that can be seen as a quantifiable measure of the properties and required characteristics that a system should fulfill, is used as a term of comparison to investigate what and where are the points of criticality in the whole system, shedding light on the most environmentally sustainable option to take (Arzoumanidis et al., 2020). The contribution given to the CE is reflected in the informative content embedded in such analysis, as it gives to the decision makers a clear view on the actions to be taken when it comes to pursue the sustainability of a system and allows for the comparison between different scenarios. Despite its several advantages, the complex structure of the LCA also embodies its main weaknesses.

The amount of data and time required to come up with a reliable assessment is vast: detailed information about the processes' inputs and outputs, and indicators to bridge the aspects to the impacts require a not negligible commitment (Van Stijn et al., 2021). This issue leads to a LCA's high degree of complexity when it comes to assess the required actions to enhance CE penetration, the vast amount of data increases the odds of information loss. In order to minimize it, it is common use to address the LCA on just one or few aspects of the process (those that are expected to be the most interesting from a CE point of view) (Van Stijn et al., 2021). If on one side the partial point of view requires less data and computations compared to the general system's perspective, on the other it could lead to the burden shifting issue. A process is composed of many jointly contributing steps, addressing the analysis about CE interventions on just some of them could create side effects that prevent the implementation of CE actions on the others that are not under the direct analysis's spotlight (Van der Meer, 2018).

This trade-off between adopting the general perspective in the LCA (high computational complexity but less probability of undesired side effects) and focusing the analysis on few process's steps (easier to implement but less reliable) leads this method to be difficult to implement and to use.

Although its disadvantages, the LCA method is widely employed as a tool to investigate CE as it always comes with crucial assumptions and simplifications of the reference scenario of applicability. In fact, a simpler system offers easiness in spotting the bottlenecks and planning the CE intervention, though their real-world applicability is hindered by the fact that some frictions get neglected (or simplified) (Van Stijn et al., 2021). Therefore, the LCA is extremely powerful when the intent is to understand where and how to intervene, but when it happens to practically pursue the CE measures, they often need to be further assessed to see whether they fit in a real world scenario, comporting additional computations.

One other LCA's drawback is its objective-dependence: according to the scope of the study, specific environmental impacts are associated to one aspect because of the context of the analysis, while other contexts would have different aspect-impact associations. The point is that different LCA's outcomes, coming from different objectives sought, give birth to a span of distinct possible CE actions that could be even in contrast one to the other. It raises the question of which is the best to be followed, additionally increasing the method's complexity of use (Van Stijn et al., 2021).

3.2 Material Flow Analysis

A further methodology often used to evaluate the circularity of a system is the Material Flow Analysis (MFA), a systematic approach to assess the flows and stocks of material through a system within a defined spatial and temporal boundary (Rechberger & Brunner, 2016). Differently from the LCA, there is no standardized definition of the MFA, though, it is possible to rely on the directive given by the European agency for the environment, which describes it as: "*an evaluation method which assesses the efficiency of use of materials using information from material flow accounting" (*EEA

Glossary*)*. MFA helps identify waste from natural resources and other materials in the economy which would otherwise go unnoticed in conventional economic monitoring systems (EEA, 2001). MFA points the focus on the process itself and helps spot what and where are the inefficiencies considering all the activities and conversions that happen. From a CE perspective, the advantage lies in the capability to see which (and how much) materials are used, and whether there is the need to opt for less impacting ones; to see how many by-products and waste get generated during the transformation process; and what is the amount of material that leaves the system (Hsu et al, 2020). All these information together offer a way to take corrective measures of resource recovery and environmental pollution wherever there is space and need for them.

Although useful for analytical purposes, the MFA is not suitable for comparisons: it focuses on just one material (or substance) flow without providing a "what-if" scenario, since a diverse object would require a completely new ad-hoc analysis with its own assumptions and limitations (Condeixa, 2016). In a CE optic the main drawback is represented by the fact that each MFA's implementation juts assess action of resources' substitution, implying that theoretically any resource could be replaced by one or more other ones according to environmental pressure they provide, ignoring the quality differences among materials (Zhang, 2019). Considering that the objective is to enhance the circularity of the processes while keeping the value of the product, the absence of a compelling way to compare the scenarios decreases the power of the MFA method.

Furthermore, it is a static tool, it provides all the knowledge needed within the reference time frame, but no information can be extracted from outside the time boundaries of the system (Condeixa, 2016). Therefore, the CE measures coming from the investigation of the method's results are stuck within the method's time boundaries, namely blind to future developments of the process.

MFA explicitly does not provide environmental assessment but analyzes the material and energy balance of a process, usually the two metrics are combined considering the MFA as a component of the LCA in order to come up with a higher degree of reliability on the results.

3.3 Material Circularity Indicator

Even if widely known and acknowledged, MFA and LCA are not the only methods employed to scientifically assess to what extent companies or products correspond to the CE. In addition, it could be countered that the two metrics above deal more with the identification, quantification and mitigation of externalities within industrial processes rather than providing a direct way to compute the circularity level of a product. To cover this gap, it is possible to opt for one other famous and relatable methodology: the material circularity indicator (MCI) metric, developed specifically by the Ellen MacArthur foundation for capturing the circularity of products based on facts and numbers (EMF, 2015). Its objective is to label one material flow on a scale from 0 to 1, where the higher the score the higher is the circularity or recyclability level. The MCI focuses neither on the environmental impact of a product, nor on the specific process technologies it goes through, but puts its interest on verifying what is the source of the composing materials and substances of a product (Rechberger & Brunner, 2002). In order to get a result of 1, all raw materials are reused or recycled, where there is no (or minimum) loss in their recycling process. The opposite case, where all raw materials are virgin, the value of the indicator is set[3] at 0,1 (EMF, 2015). This scale provides an easy-to-read tool for the decision maker to understand the level of circularity of the products and gives a fast picture of how distant the results are from the ideal case. The output of the study is an idea of how urgent the need for a corrective measure is. A relevant characteristic of the MCI metric is that it attempts to take product durability into account, considering that one of the pillars of the CE is the creation of value through material retention, the inclusion of the material duration inside the cycle is crucial (Rocchi et al., 2021). However, also the MCI framework is not exempt from limitations: according to EMF (2015), the measurement of an indicator specifically for each component increases the complexity of

[3] When the indicator assumes values lower than 0,1 it means that the production process of such product has been worse than linear.

the method (high data requirement), posing a burden for its applicability since it could be difficult to accommodate the variety of reuse and recovery rates for each resource.

3.4 Towards a new CE measurement tool: Statistical Entropy Analysis

Considering its dominant role at the center of the CE issue, all the methodologies explained have been employed for plastic waste management, as it represents one of the most interesting fields where to look for corrective measures' applicability. In this direction, the use of all these assessing methodologies implies, usually, the necessity to introduce some harsh assumptions like ideal (thermodynamic) working systems or the presence of perfect technology transfers without which would be challenging to gather the required data. Sticking to plastic recycling, problems could appear in meeting the complexity of the post-consumer plastic waste management through the "design-for-recycling" aspirations of such methods, and the simplifying assumptions enhance the gap. Furthermore, of the tools listed so far to compute the circularity, both LCA and MFA had the characteristics of being ex-post calculation strictly bounded to a rigid background: almost all sustainability assessments used for plastics cycles start by establishing mass and energy balances, which can further be translated into a life cycle inventory or into economic data (Nimmegeers et al., 2021). The issue is that innovations based on disruptive technologies may fail to demonstrate their environmental and/or economic potential, as the background technological system is usually considered (Nimmegeers et al, 2021). Aiming to get a more generic method, Rechberger and Brunner (2002) developed the statistical entropy analysis (SEA) to assess the capability of a system to preserve the functionality of the resources employed at the highest level possible. Every time a recycling loop takes place, the resources undergo a new cycle of decomposition, processing and transformations that unavoidably change their intrinsic composition, SEA aims at investigating the portion of the value that the system can keep cycle by cycle labelling each object of analysis with an entropy value (Rechberger & Brunner, 2002). The higher the entropy level, the lower the value content of the

product analyzed as the modifications in the resources' structure excessively entailed their value and this would be reflected in the final output of the next cycle (Rechberger & Brunner, 2002). As SEA only assesses if a specific state is achieved, and not how it is achieved, it is independent of any background system. However, in order to get to a comprehensive informative content, it is ought to consider the SEA on a multilevel structure: where entropy values are computed at different aggregation stages, from elementary substances to components (made of substances) and final product (made of components), in this way it is possible to investigate CE alternatives such as reuse, repair, remanufacturing or combination of these (Parchomenko et al., 2020). The model has, though, a relevant limitation: it could happen that two different waste streams end up having a similar relative statistical entropy, but they would significantly differ in the way these waste streams can be separated or further recycled. This is explained by the fact that the entropy value gives information about the concentration (or dilution) state of a waste stream but does not take into consideration the type of chemical substances involved (Nimmegeers et al., 2021). Considering, therefore, that the objective is to use the SEA to predict the recyclability of plastics in a generic context, a solution can be to couple the method with the energy required for generic transportation, sorting and refining processes. In this way the numeric indicators that the SEA provides can be read as "easiness of recycling" from industrial players, whose can take decision on the recycling actions to put in place for them. This is because, despite the similar entropy level, the energy required to treat two waste streams may significantly differ one to the other according to their chemical composition, and so would do also the recyclability (Nimmegeers et al, 2021).

Table 1 shows a summary of the advantages and disadvantages, according to a CE optic, of the methods described above:

Table 1 - Pros and cons of CE assessment methods

Circularity assessing method	Advantages	Disadvantages	Source
LCA	· Provide comprehensive view on the environmental impacts · Quantify the environmental impacts · Allow comparisons · Recognize inefficiencies across life cycle phases	· Strong dependency on assumptions · Strong Objective-dependency · Complexity in data gathering and assumptions · Difficult to quantify the environmental impacts · Ex-post method	· Shen et al., 2010 · Gu et al, 2017 · Arzoumanidis et al, 2020 · Van Stijn et al, 2021 · FibreNet, "*Life Cycle Assessment: Benefits and Limitations*", 2018
MFA	· Streamline material/energy flows · Spots critical material/energy flows · Provides quantitative data	· Not suitable for comparisons · Neglects quality differences among the materials · Static tool · Complexity in data gathering and implementation · Ex-post method	· Condeixa, 2016 · Zhang, 2019

MCI	· Provides quantitative measure of recyclability · Provides a scale for the comparison with ideal cases · Considers materials duration in the cycle	· High level of difficulty in getting the data · Computational complexity · Ex-post method	· EMF, 2015 · Rechberger & Brunner, 2002 · Rocchi et al, 2021
SEA	· Provides quantitative measure of recyclability · Assess whether a result is reached, not how · The analysis is carried on more aggregation levels · Allows the comparison	· High difficulty in getting the data · Complexity of computation · Every recycling loop needs a new analysis	· Nimmegeers et al, 2021 · Rechberger et al, 2002 · Parchomenko et al, 2020

4 Statistical Entropy Analysis

The first definition of entropy was given by the physicist Rudolf Clausius in 1850, who looked for a way to characterize the fact that every spontaneous natural process is irreversible, meaning that there is no way to perfectly get back to the initial conditions once the transformations take place because of unavoidable energy losses. In 1870, Ludwig Boltzmann introduced another way to look at it, mathematically reading entropy as the superimposition of a certain number of microstates, composing a whole macrostate, paving the way for the statistical entropy to thrive. The higher the number of microstates, the higher the disorder into the system, the higher the statistical entropy (Balibrea, 2016).

Boltzmann's mathematical formulation inspired, then, Claude Shannon to propose, in 1948, a new perspective to consider statistical entropy, outside the sole physics, as the loss or gain of information about a system, high entropy level means high informative content (Balibrea, 2016).

The strength of the concept of entropy lies in its capability of being declined in many different dimensions because of its very adaptable inner meaning. Looking at the different progressive definitions is possible to extract a common thread: the statistical entropy gives an idea of how complex and structured an object under analysis could be, regardless of the specific lens used to look at it.

Considering the whole plastics industry, and particularly its ongoing shift from a traditional linear model to a sustainable circular way of performing the processes, statistical entropy can be used as a lever to unlock innovative and useful ways to assess the degree of recyclability of plastic waste, and therefore contributing to the assessment of their circularity. Practically it can be used to describe the concentration of an element or compound as it undergoes transformations in a system (Bhavik et al., 2011). Any plastic product is obtained after a process of transformation, modification and assembly of several raw materials in input, polymers in this case, that consume energy and work. Once they go through those steps, input materials are not the same as they were at the beginning, both in terms of quality (energy consumption) and quantity (industrial waste), in other terms, they have increased their entropy level (Nimmegeers et al., 2021).

In their first formulation of statistical entropy analysis (SEA), Rechberger & Brunner (2002), applied the calculation of the statistical entropy deriving it as an evolution from the MFA: the backbone of a process is the flow of goods which are, in turn, made of substances (elements or compounds). While different strategies exist to increase the circularity of industrial processes (e.g., recycling, remanufacturing, reuse, etc.), the assessment of their effectiveness at keeping resources in closed loops is still challenging (Parchomenko et al., 2020). In their SEA formulation, the authors stressed the difference between goods and substances' flows because: "*Traditional economic data comprises*

mainly quantities for goods, while decisions regarding the management of resources and the environment require data on substances", showing that relevant resource efficiency decisions have to pass through substances data analyses (Rechberger & Brunner, 2002). Each system could, indeed, be seen as a unit that concentrates, dilutes or leaves unchanged the throughput of a substance: SEA was thought exactly to investigate this issue (Rechberger & Brunner, 2002). Taking as example a plastic bottle's manufacturing process, substances are represented by the bottle's constituent polymers, while the good is the bottle itself. SEA enables to assess whether the bottle's substance concentration changed with respect to their initial one, and to what extent. This concept is crucial since, according to the CE principles, when the bottle becomes waste, the mass and value of the substances should be recovered to produce new plastic goods, without/limiting injection of new virgin polymers. Statistical entropy's indicators about the substance's concentration gives clarity not only on the feasibility of such activity (whether it is possible to get enough quantity from wastes), but also on the required effort to extract them as secondary raw materials (Rechberger & Brunner, 2002).

When investigating the CE transition, SEA appears as a robust way for assessing CE strategies and their combination to minimize resource functionality loss. It follows the process of materials transition step by step, evaluating and keeping track of the distribution pattern of a substance, allowing also to identify and compare the different process's "*metabolic systems*" (Rechberger & Greiner, 2002). Literature provides several examples[4] of how SEA can lead to relevant insights in the matter of circularity of industrial processes. Although widely recognized and still in development, many SEA publications focus on just the individual substance level, not taking into consideration that CE practices apply also to components and products (Velazquez Martinez, et al., 2019; Tong et al.,

[4] The examples include the assessment of system performance of different municipal solid waste incinerator technologies (Rechberger and Brunner, 2002), wastewater treatment plants (WWTP) (Sobantka & Rechberger, 2013), the evaluation of the Austrian phosphorus cycle (Laner et al., 2017) and the European copper cycle (Rechberger & Graedel, 2002). Other studies applied the method to the copper cycle in China (Yue et al., 2009), to the process of lead smelting of a large state-owned enterprise (Bai, et al., 2015), or to the results of a sieving experiment of a crushed lithium-ion battery on a lab- scale (Velázquez-Martinez et al., 2019).

2021; Busu & Busu, 2018). The substance limited perspective has the drawback of neglecting those CE strategies belonging to higher aggregation levels (e.g., reuse, repair, remanufacturing, or their combinations), missing a whole group of contributions (e.g., how the substances sort themselves to structure a plastic item, and which implications it could have on the final recyclability) toward increasing circularity (Parchomenko et al., 2020). Therefore, in order to cover the most compelling analysis on CE activities implementation, Parchomenko et al. (2020), proposed the adoption of a "*multilevel SEA*" where, besides the assessment on the resource efficiency at the substance level, the method considers, first, various separate substances and materials assembled to form the component level, and then the combination of such components at the product level.

In their work, Nimmegeers et al. (2021), raised the point that, although efficient, the multilevel SEA by itself has some limitations. Adopting a CE assessment optic, especially with regards to plastic waste recycling, not all the statistical entropies are equal in meaning: two different waste streams could theoretically have very similar statistical entropy but can strongly differ in the way they are separated and recycled (or reduced in relative statistical entropy) (Nimmegeers et al., 2021). Looking for a way to overcome the issue, the authors have expanded the method by considering a coupling with energy balances from post collection phases (e.g., generic transportation, sorting and refining technologies), to show how the energy required to treat waste streams with similar statistical entropy can differ. In their study, the recyclability of plastics is then defined by a metric encompassing both relative statistical entropy and relative decomposition energy of waste streams.

In this work, we adopt the extended multilevel SEA as a tool to investigate the degree of recyclability of plastic packaging waste streams in the Lombardy region. Formulas, nomenclature and classification are faithful to the work of Nimmegeers et al. (2021), as well as the structure of the analysis.

4.1 Mathematical formulation of extended multilevel SEA

In order to face the multilevel SEA mathematical formulation, it is important to have in mind the hierarchical structure of the system considered (figure 4). The inputs of the system are the substances (*i*), which can be seen as the elementary building blocks, while their assembly forms the materials (and material flows) (*f*). Different combinations of substances form different typologies of material, and the aggregation of different typologies of materials constitute the components (*n*). At the highest level of the pyramid, the components are furtherly gathered to form the product (*p*).

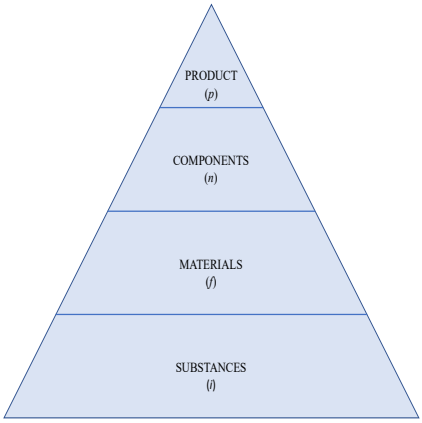

Figure 4 - Pyramid scheme of SEA method's structure

4.1.1 Substance-level entropy

Consider a plastic manufacture process involving a material flow *f* (which can be a component or a good, defined in mass and time) and a substance *i*. It is possible to define a flow rate M_f of the generic material flow *f*, and the dimensionless mass fraction $c_{i,f}$ of a particular substance *i* inside *f*. The flow rate gives the total quantity of a specific material in output of the process, while the mass fraction provides what percentage of a substance is present inside the material flow. In order to get what is the total percentage of a substance present in the final output of the process it is possible to compute the substance flow rate $X_{i,f}$ (in mass per time), given by:

$$X_{i,f} = M_f c_{i,f} \qquad (1)$$

Additionally, the standardized mass fraction per time can be computed by dividing the material flow rate M_f by the total flow of a substance *i* over the *F* material flows:

$$m_{i,f} = \frac{M_f}{\sum_{f=1}^{F} X_{i,f}}$$

(2)

Table 2 provides a summary of the main dimensions that will be employed throughout the entire development of the SEA method.

Table 2 - List of SEA's main dimensions

Number	Dimension	Definition
1	i	Substance flow
2	f	Material flow
3	M_f	Material flow rate
4	$c_{i,f}$	Mass fraction of *i* inside *f*
5	$m_{i,f}$	Standardized mass fraction of *i* inside *f*
6	$X_{i,f}$	Substance flow rate of *i* inside *f*

Given these dimensions is possible to compute what is the statistical entropy[5] $H^i(c_{i,f}, m_{i,f})$ of a substance *i*, defined by the equation:

[5] It is defined through the letter H in order not to confusing it with the thermodynamic entropy S.

$$H^i(c_{i,f}, m_{i,f}) = -\sum_{f=1}^{F} m_{i,f} c_{i,f} \log_2(c_{i,f})$$

(3)

The value of the statistical entropy varies according to the distribution of the substances inside the materials: if a substance is present in all the materials of the process with the same quantity, the entropy will be maximum, otherwise, if the substance is concentrated in just one (or few) material its value will be close to zero. To assess the results, in SEA method the relative statistical entropy, expressed as: $H^i_{rel}(c_{i,f}, m_{i,f})$, is used since it has the advantage, with respect to the absolute statistical entropy, to allow the comparison among different streams.

$$H^i_{rel}(c_{i,f}, m_{i,f}) = \frac{H^i(c_{i,f}, m_{i,f})}{H^i_{max}}$$

(4)

Where:

$$H^i_{max} = (\sum_{f=1}^{F} m_{i,f}) \, .$$

(5)

The relative statistical entropy formula sees the ratio of the statistical entropy value and the maximum[6] statistical entropy value. This latter is reached when the substance is equally distributed among the material flows and the concentrations in all material flows are the same for substance i.

[6] The formulation described refers to closed systems, since they are the ones considered in the work, it is different if the system is open.

4.1.2 Component-level entropy

The formula of the relative statistical entropy of a component *n* is mathematically similar to the one involving the substances, with the only relevant difference that the dimensions refer to concentration and fraction of substances at the component level, not in the materials as it was for the substance level. The normalized component mass (m_n^c) describes the mass fraction of a component *n* compared with all components at a stage in the system, while the concentration of substance *i* in the component *n* is defined as $c_{i,n}$.

$$H_n^c\left(c_{i,n}, m_n^c\right) = -\sum_{i=1}^{I} m_n^c c_{i,n} \log_2(c_{i,n})$$

(6)

One other difference between the two levels of statistical entropy described is that, at the component level, the summation is performed over the different substances, while at the substance level it is over the material flows, reflecting the objective sought at each level of analysis. At the component level, like the previous one, the relative formulation of the statistical entropy is used.

$$H_{n,rel}^c\left(c_{i,n}, m_n^c\right) = \frac{H_n^c\left(c_{i,n}, m_n^c\right)}{H_{max}^c}$$

(7)

Where:

$$H_{max}^c = -\sum_{i=1}^{I} c_{i,tot} \log_2(c_{i,tot})$$

(8)

In this case, $c_{i,tot}$ refers to the concentration of the substance *i* in the total system stage. The maximum component-level entropy corresponds with either the case in which all substances are present in one material flow or equally distributed.

4.1.3 Product-level entropy

The highest degree of analysis refers to the product level, in this case the statistical entropy is defined as:

$$H^p\left(c_{n,p}, H^c_{n,rel}\left(c_{i,n}, m^c_n\right)\right) = -\sum_{n=1}^{N} \log_2(c_{n,p})\, H^c_{n,rel}\left(c_{i,n}, m^c_n\right) \quad (9)$$

$$c_{n,p} = \frac{q_n}{N_{tot}} \quad (10)$$

$c_{n,p}$ gives the level of concentration of the component *n* in the product *p*, it is defined by the ratio of the number of units of component *n* (q_n) over the total number of components present in the system (N_{tot}). The two terms that compose the statistical entropy at this level of analysis depend on both the distribution of components and the distribution of substances over the components, respectively represented by the component concentration and the relative statistical entropy of the components. For this reason, a high value of statistical entropy at product level could be due to either the presence of many distinct components, or because of a high dilution, or heterogeneous distribution, of the substances over the product's components. The main dimension used for the analysis is the relative statistical entropy:

$$H^p_{rel}\left(c_{n,p}, H^c_{n,rel}\left(c_{i,n}, m^c_n\right)\right) = \frac{H^p\left(c_{n,p}, H^c_{n,rel}\left(c_{i,n}, m^c_n\right)\right)}{H^p_{max}}$$

(11)

Where:

$$H^p_{max} = \log_2(N_{tot})$$

(12)

In this case the maximum value of entropy corresponds to the case in which every substance is uniformly distributed over the product's components, indeed, the maximum degree of dilution is given by the total number of components.

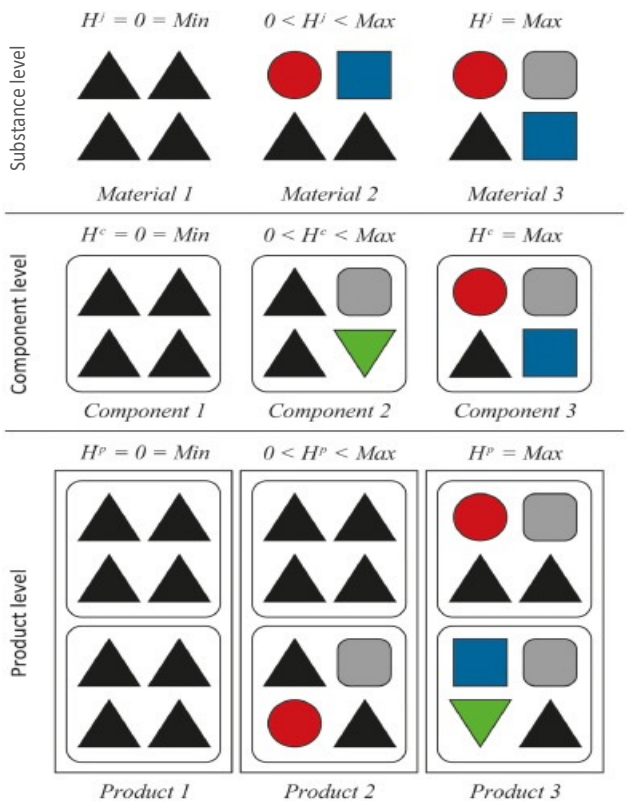

Figure 5 - Conceptual representation of the material, component and product levels and the influence of substance and component distribution on statistical entropy values (Parchomenko et al, 2020)

To recap, the statistical entropy value varies according to the distribution of substances according to the level considered in the analysis. In figure 5 there is a conceptual representation of the statistical entropy value that helps in understanding what characterizes its maximum and minimum values.

At the substance level, the minimum value of entropy is obtained when the material is highly concentrated on just one substance, while the maximum is reached with the highest degree of heterogeneity. When it comes to performing the analysis on the upper levels (component and

product), besides the concentration of substances in the components and the products respectively, the entropy value is affected also by the number of materials (at the component level) and of components (at the product level) that form the analyzed product. At the component level maximum statistical entropy means to have a component formed by many materials which in turn have also high embedded heterogeneity. The same concept is valid also for the product level: products made of many diluted components give high value of statistical entropy.

4.2 Multilevel SEA with decomposition energies

Adopting the CE perspective, the multilevel SEA is useful but not compelling in order to evaluate the recycling strategies. This chapter proposes an extension of the methodology based just on entropy values by considering the energy consumption aspects to come up with a definitive metric to investigate the recyclability of plastic waste from packaging. The SEA method offers a clear view of the mix of substances that form, progressively, materials, components and the final product; however, it does not provide any insight about the often-distinct way in which such materials and/or substances are linked. The relevance of this final perspective is given by the fact that, depending on the way the linkages are created, different processes have to be performed to decompose the components in their constituting materials and/or in their constituting substances (and the same for products and materials). It is possible to define the decomposition energies, in unit per mass (J/kg), as the overall energy expenditure required to decompose an object from a more aggregate state to a lower one (e.g., components to materials, materials to substances, …), through the definition of the specific process required (π) and the efficiency of decomposition (η):

- Energy to decompose a component into substances: $E_n^c(\pi, \eta_i^c)$
- Energy to decompose a component into materials: $E_n^c(\pi, \eta_m^c)$
- Energy to decompose a product into components: $E^p(\pi, \eta_n^p)$

These are the most relevant decomposition energies to be considered, but not the only that could be used in the analysis. Figure 6 gives a snapshot of all the possible energies of a process with their meaning.

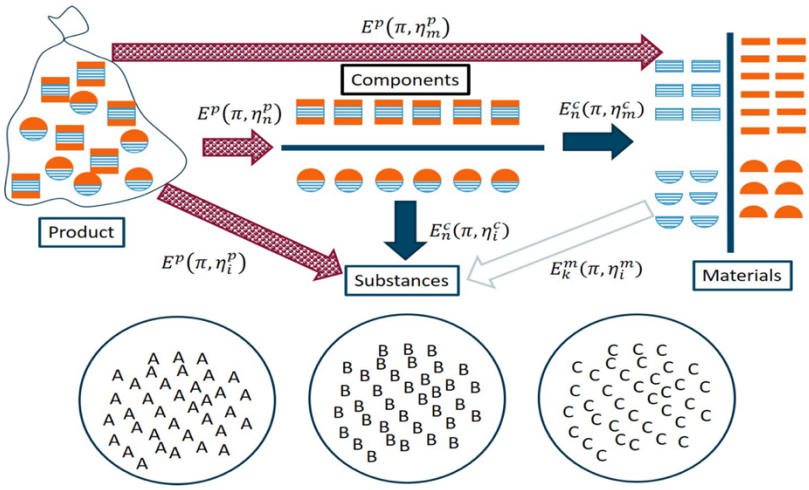

Figure 6 - conceptual representation of the decomposition energies for plastic waste. (Nimmegeers et al., 2021)

In order to exploit the decomposition energy to evaluate recycling, a relative metric has to be built for the energies there is the need of defining the maximum reference value. The maximum decomposition energy (in J/kg) is defined as the energy needed to produce the virgin substances present at the highest hierarchical stage (the product level). This allows the comparison of the relative decomposition energies also at the lower levels, as the same maximum decomposition energy is used.

$$E_{max} = \frac{1}{M_{tot}} \sum_{n=1}^{N} \sum_{i=1}^{I} q_n c_{i,n} M_n e_i^{production} \quad (13)$$

where M_{tot} is the total mass of the studied system, q_n is the number of components n in the product, $c_{i,n}$ is the mass fraction of substance i in component n, M_n is the mass of component n (in kg) and

$e_i^{production}$ is the energy that is needed to produce 1 kg of virgin substance *i* (expressed in J/kg). The value of the maximum decomposition energy can be used to compute the relative decomposition energies of a component and of the product:

$$E_{n,rel}^{c}(\pi, \eta_m^c, \eta_i^c) = \frac{E_{m,i}^{c}(\pi, \eta_m^c, \eta_i^c)}{E_{max}}$$

(14)

$$E_{rel}^{p}(\pi, \eta_n^p, \eta_m^p, \eta_i^p) = \frac{E_{n,m,i}^{p}(\pi, \eta_n^p, \eta_m^p, \eta_i^p)}{E_{max}}$$

(15)

According to the meanings of the obtained relative statistical entropy and relative decomposition energy, a recyclability metric can be defined considering the contribution of both. It is reasonable that such metric assumes the minimum value when either the entropy or the energy is maximal, which corresponds to minimum recyclability l. On the opposite side, it is possible to get to the maximum value of recyclability when energy and entropy assume values close to zero. There are two possible mathematical formulations of the recyclability metric, the first gives a value between 0 and 1, where 0 means low recyclability and 1 means high recyclability.

$$R_n^{(1)} = \left(1 - H_{n,rel}^{c}(c_{i,n}, m_n^c)\right)\left(1 - E_{n,rel}^{c}(\pi, \eta_m^c, \eta_i^c)\right)$$

(16)

$$R_p^{(1)} = (1 - H_{rel}^{p}\left(c_{n,p}, H_{n,rel}^{c}(c_{i,n}, m_n^c)\right))(1 - E_{rel}^{p}(\pi, \eta_n^p, \eta_m^p, \eta_i^p))$$

(17)

The second formulation of the metric provides a wider span of values that the indicator can assume. In this way it is also easier to compare indicators from different components/products, since it is easier to get a more comprehensive estimate of the analyzed object's recyclability value.

$$R_n^{(2)} = \frac{\left(1 - H_{n,rel}^c\left(c_{i,n}, m_n^c\right)\right)}{\left(1 - E_{n,rel}^c(\pi, \eta_m^c, \eta_i^c)\right)}$$

(18)

$$R_p^{(2)} = \frac{(1 - H_{rel}^p\left(c_{n,p}, H_{n,rel}^c\left(c_{i,n}, m_n^c\right)\right))}{(1 - E_{rel}^p(\pi, \eta_n^p, \eta_m^p, \eta_i^p))}$$

(19)

In this dissertation, both recyclability metrics have been employed.

5 Methodology

The work is focused on the application of the extended multilevel SEA to assess the energy potential embedded in plastic waste flows and to estimate the recyclability potential of collected plastic waste. In order to do so, a thorough methodology is needed to comprehend which flows to consider and what data to analyze. In general, the term "plastic waste" embeds a wide variety of plastic materials with distinct characteristics and different end-of-life treatment methods. Figure 7 shows the SEA's methodology flow chart, highlighting all the steps we went through in order to develop the analysis.

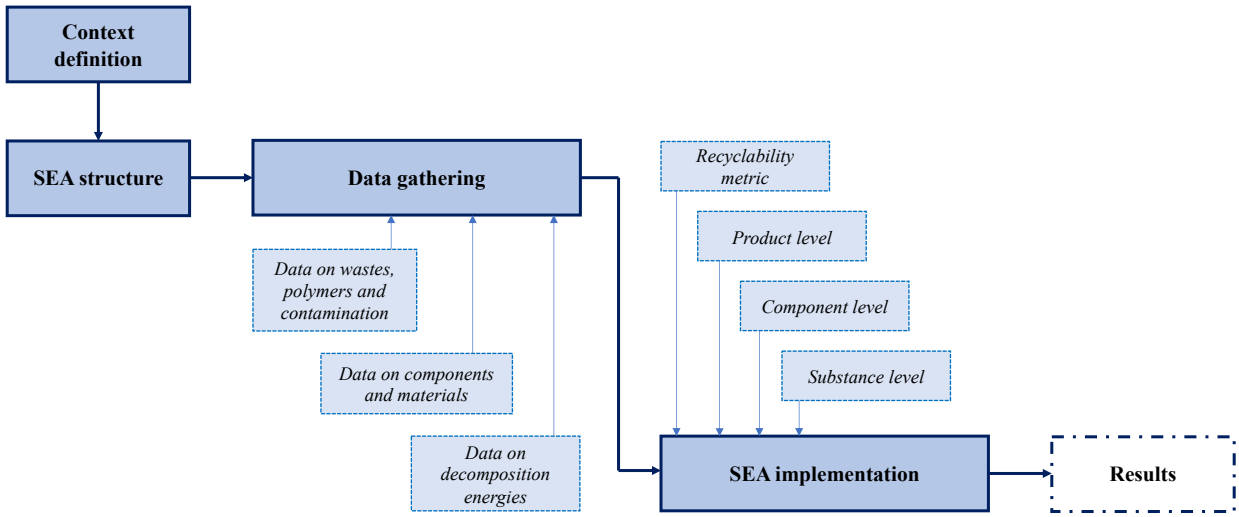

Figure 7 - SEA methodology flow chart

5.1 Context definition

The first step to prepare the SEA analysis is to identify the players involved in the computations, namely what plastics waste to consider, why and how. During an initial screening phase, only municipal solid wastes (MSW) are selected[7] for the analysis. This is because the regard of other types of waste, like industrial scraps or special wastes, would compromise the computation because of the

[7] According to ISPRA (2019), it is specified that the wastes' amount provided belongs to MSWs collection

likely presence of hazardous materials requiring treatment. Furthermore, this typology of plastic wastes eases the connection with the specific recycling technology employable (according to the considerations of section 2.2), since they represent the highest share of wastes being gathered though collection programs and, of consequence, of wastes sent to recovery facilities. Besides the constraints set by the technical limits of recycling processes, there is the major consideration that every country has different rules for recycling programs: there are some relevant differences regarding which typologies of plastic waste get collected. In order to perform a compelling analysis, it is crucial to have in mind the plastic waste collection directives that apply in the geographical scenario under consideration (OECD, 2018). According to Corepla (2020), in the Italian context only the plastic waste belonging to the packaging sector is gathered through separate collection, and subsequently sorted and processed for recycling. In addition, it is worth to point out that the packaging sector accounts for the highest share of plastic production (roughly 40% of the whole European production), and thereby it translates into the highest share of waste generated (PlasticsEurope, 2020). Plastic packaging materials are mainly made of thermoplastics: open-chain polymeric materials which can be melted and molded repeatedly without incurring irreversible chemical changes (PlasticsEurope, 2020). It is precisely thanks to this feature of modifying their structure when exposed to heat, that it is possible to investigate recycling solutions for them. Different thermoplastics compose different plastic products, the resin identification coding system (RIC) (figure 8), an international standardization aiming at providing "*a consistent system to facilitate recycling of post-consumer plastics*", allows to clarify which polymers[8] are contained within the product considered (ASTM International, 2016).

[8] The meaning of the number inside the RIC is to provide the ease of recycling, considering the actual technologies, of the product: PET is the easiest polymer to recycle while PP and PS are more challenging. The identification code 7 symbolizes all the products made of mixes of different polymers, very difficult to recycle

Figure 8 - ASTM plastic resin coding system

Therefore, the perimeter of the SEA analysis conducted in this dissertation (investigating the potentials of CE activities on plastic waste) comprises the post-consumer plastic waste from packaging, including all plastic-containing materials which can be used for the containment, protection, handling, delivery and presentation of other products.

The packaging plastic wastes are analyzed starting from their constituent polymers, which represent the center of gravity of the recycling activity, in fact the objective is to get from collected waste to raw polymers (presenting characteristics and performance similar to those of their virgin counterparts) as efficiently as possible.

5.2 The model

The entire structure of the method is based on the work by Nimmegeers et al. (2020). As regard the SEA performed on packaging waste, the polymers have been assumed to represent the substance level, namely the ground level of the analysis. It could have been possible to further deepen the analysis investigating the inner chemical compound of each, and so getting to the real "zero level" of analysis, however it would have been a worthless increment of the computation complexity since, for the scope of the work, reaching the polymers level is enough to get interesting insights about CE

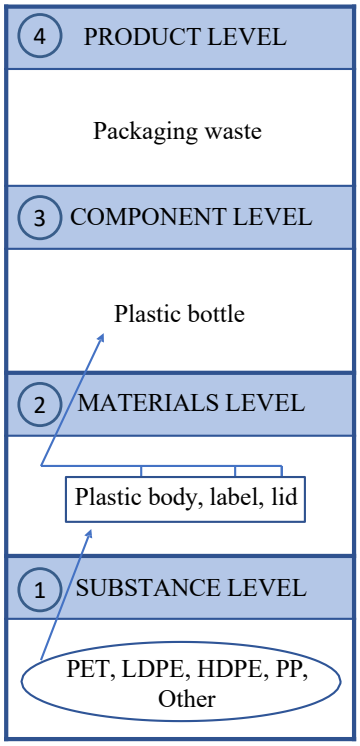

Figure 9 - Example of the SEA's structure on a plastic bottle

promoting actions. As discussed in section 4.1, the analysis is performed on different levels of aggregation (figure 9): it starts at the substance level, which represent the building block of the analysis, and get to the final product, passing through the materials (aggregation of substances) and the components (aggregation of materials). In order to understand the analysis performed on the packaging plastic waste, it is possible to visualize the final product (top level of analysis) as a "huge bag" containing all the Lombardy's waste from packaging. The figurative bag is assumed to be a bundle of plastic items which represent the component level of analysis (plastic product, e.g. plastic bottles, glasses, wraps, containers, etc.). Going down another step, each component is seen as an assembly of parts (e.g., a bottle embeds the plastic body, the label and the lid) which form the SEA's materials level of the pyramid scheme.

Finally, each material is directly analyzed in terms of its constituent polymers (substance level). Therefore, the SEA lineup considers first the polymers and their dilution/concentration percentages within the materials, namely how they are distributed, giving the value of the substance-level relative statistical entropy. Second, it puts together the materials generating the components, investigating, at this point, what is the level of polymers' concentration/dilution but within the components, providing the component-level relative statistical entropy. The entropy content at this level embeds information of two levels of turbulence, since it is created considering not only the polymers' dispersion within the materials, but also the dispersion of the materials that form the components. Finally, the indicators found at the component level are employed to get to the unique product-level relative statistical entropy, which keeps track of the overall recyclability content in the most general terms. In the very

last step of analysis the entropies at the component level get coupled with the energies of decomposition to get to a recyclability indicator. This last procedure is performed only at the component level because, considering what the components are assumed to be for the analysis, they are the active interested parties in recycling processes.

5.3 Data gathering

One of the major issues that hindered the data gathering activity has been the almost entire absence of information available at regional level, all the papers and databases scanned are full of country-level records. Therefore, in order to provide a coherent and accurate analysis for the Lombardy region, some data assumptions were made (which will be discussed later), keeping in mind that wherever there is an assumption there is also the generation of unavoidable biases entailing the overall reliability. For this reason, every time that a datum has been obtained with the support of one or more assumptions, also a comment about the new flawed reliability is set forth.

5.3.1 Wastes, polymers and contamination

The first step of data gathering activity has been organized according to a rigorous schedule according to which we identified:

- The total amount of plastic wastes generated in Lombardy in 2019
- The main polymers composing those wastes and the percentage of each type of polymer over the total amount
- The contamination level (if present) of polymers

Lombardy generated a total of 255315,34 tons of plastic packaging waste in 2019. This value was obtained from the "*catasto rifiuti 2019*" of ISPRA's, (2019) database and refers to the amount of plastic wastes picked up by separate collection, which, given the Lombardy region's directive identifying in packaging wastes the only to be eligible for separate collection, are assumed to

correspond to the packaging plastic waste amount. However, in order not to totally rely on the assumption that each separate collected item directly belongs to the packaging sector, we considered that over the total amount collected the packaging wastes account for 90%[9]:

Packaging waste collected in Lombardy: 255315,34 * 90% = 229783,8 tons.

The research of the polymers' composition of the collected plastic wastes was a challenging task since there is paucity of comprehensive estimates. Therefore, in order to get to this information, we considered specific assumptions and computations. According to Geyer et al. (2017), it is possible to get that the average lifetime of plastic packaging is particularly short in comparison to other sectors' plastic products.

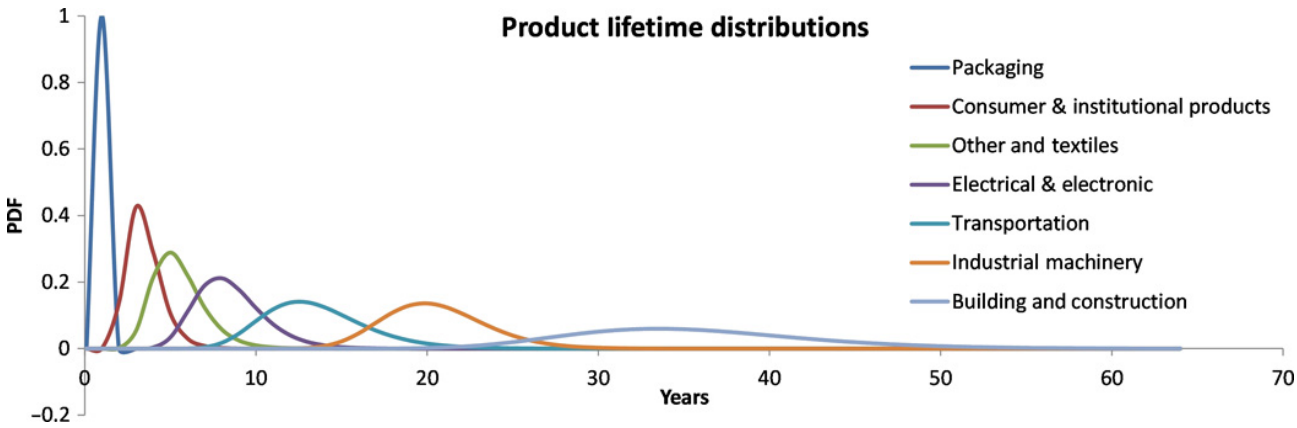

Figure 10 - Probability density functions of plastic products lifetime from different sectors (Geyer et al., 2017)

As it is noticeable from figure 10, the average life length of packaging plastic is no longer than a single year. The reason is given by the fact that most packaging products are designed for single uses only, therefore it is reasonable to deem that the packaging products produced in 2019 become waste within a one-year period. This led to the assumption that it is possible to leverage on the 2019 plastic

[9] This assumption takes into account the errors that can be committed in collecting and producing the wastes, it intends to avoid the consideration that the 100% of the collected items belong to the packaging sector. This assumption increases the data reliability.

demand amount to get insights about the packaging wastes, in fact, the probability to find them as waste within the same year of production is rather high. The advantage of using the demand amount to derive the wastes generated is that there is plenty of information available about the polymers produced and their percentage over the total which we could rely on to feed the SEA method. According to PlasticsEurope (2020), the overall amount of plastic produced in Europe in 2019 was 50,7 million tons, 39,6% of which represented by packaging.

2019 European plastic packaging production: 50,7 M tons * 39,6% = 20,077 M tons.

In order to scale this datum from the European level to the Lombardy level, we employed an allocation quota proportional to the population[10]:

$$allocation\ quota = \frac{Lombardy\ 2019\ population}{EU\ 2019\ population} = \frac{10027602}{747182815} = 0{,}0134 \quad (20)$$

At this point we employed the allocation quota to get the amount of packaging items produced at the Lombardy region's level: $20{,}077\ M\ tons * 0{,}0134 = 269447{,}004$ tons. The allocation quota computed above the population embeds the limitation to neglect that the industrial production of Lombardy has a higher weight on the European total production, nevertheless, for the scope of the work there is no consideration of import/export but the focus is on the plastic waste generated by the consumption within the region's boundaries, so the allocation quota on the population represents a valid proxy. Table 1 sums up the information about the wastes produced (and derived) that will be employed in the SEA's computations.

[10] The information about the EU population is taken from PopulationPyramid.net; while as regard the population of Lombardy the source is ISTAT

Table 3 - Plastic production at different geographic levels

Data	Amount	Unit of measure	Geographic level	Year	Source	Reliability[11]
Plastic produced	50700	ktons	EU	2019	PlasticsEurope (2020)	High
Plastic for packaging produced	20080	ktons	EU	2019	PlasticsEurope (2020)	High
Plastic for packaging produced	269,45	ktons	Lombardy	2019	Computed	Medium

Once we collected the information about the 2019's plastic for packaging produced in Lombardy, the following step was to find how this production is divided according to the different plastic polymers and what is the percentage of each stream among the total. In doing so, we leveraged on the plastic demand by resin type of 2019, by PlasticsEurope (2020), that splits the overall European production into the corresponding polymers' percentages presented in Table 4.

Table 4 - Plastic demand by resin type 2019 (PlasticsEurope 2020)

RESIN	%	TONS
PET	7,90%	21286,31
LDPE	17,40%	46883,78
HDPE	12,40%	33411,43
PP	19,40%	52272,72
PVC	10,00%	26944,70
PS	6,20%	16705,71
PUR	7,90%	21286,31
Other	18,80%	50656,04
TOTAL	**100%**	**269447,00**

[11] The column "reliability" embeds the following information:
- High: data directly coming from verified sources
- Medium: data obtained through computations based on verified sources
- Low: data obtained after numerous assumptions

Relying on the percentage values (table 4) we were able to find the amount (in tons) of each polymer produced at the regional level. The row "other" refers to particular typologies of plastic which are composed of mixes of the other resins or that come from special processes and represent the portion of contaminated polymers which can be found in the waste collected and that somehow thwart the recycling processes (PlasticsEurope, 2020). The critical point is posed by PVC (Polyvinyl Chloride) and PUR (Polyurethane) because their chemical and physical characteristics make them suitable for application sectors[12] different from packaging. It is particularly rare to find these two resins into plastic products for packaging, therefore we accounted for this issue by removing the amounts of production of these two polymers from the total and recomputed the percentages and values accordingly, as presented in Table 5.

Table 5 - Adjusted plastic demand for packaging 2019

RESIN	%	TON
PET	9,62%	21286,31
LDPE	21,19%	46883,78
HDPE	15,10%	33411,43
PP	23,63%	52272,72
PS	7,55%	16705,71
Other	22,90%	50656,04
TOTAL	**100%**	**221215,99**

The result we got as a proxy for the total amount of plastic waste from packaging in 2019 is 221215,99 tons (table 5), that embeds all the assumptions and calculations described so far. A comparison between such value and the one taken directly from ISPRA's database, related to the wastes gathered

[12] PVC is the main plastic employed in the building sector, while PUR is the main component of insulation products

through separate collection, shows that the deviation between the two is rather low, as shown in Table 6.

Table 6 - Summary and comparison of collected and computed data

Data	Amount	Unit of measure	Geographic level	year	source
Packaging waste collected in Lombardy (90%)	229783,8	tons	Lombardy	2019	ISPRA 2019
Plastic for packaging produced	221215,99	tons	Lombardy	2019	Computed
Deviation (%)	3,73%				

For this reason, we opted to use the amount of production as a proxy for the waste generated, so that the percentage values of polymers are coherent.

In a CE optic, in order to assess recyclability, it is crucial to know the level of degradation and contamination of wastes when they reach the recycling centres, because the higher the contamination of waste, the lower the efficiency exploitable to recover the value. In this dissertation, "level of contamination" is intended as the presence of impurities in the structure of resins represented by percentages of other polymers. For our analysis, we explored cross-contamination between the polymers' structure, caused by their reprocessing once they have been gathered as waste, therefore, it is likely to find contaminants in already recycled plastics. Specifically, this work focuses on analyzing the first-round loop of recycling, which means considering those plastic wastes that are collected to be recycled for the first time (as it will be discussed in the paragraph about the limitations of the SEA method). Nonetheless, we conveyed that it is still quite likely that some of the plastics gathered have already been reprocessed at least once, allowing us to take into consideration the presence of possible contaminants, especially within the most widespread polymers in the packaging sector (above all LD/HD-PE and PP). Along with the results from Juan et al. (2021), we found

significant information about cross-contamination between PP and HDPE, detected through the Fourier transformation infrared technique (FTIR). The study found that PP contains around 4-5% of recycled HDPE, and HDPE contains between 8-10% of recycled PP. In order to investigate the behavior of the SEA in presence of levels of contamination, we considered in the computation these amounts of cross-contamination every time PP and HDPE are the objects of analysis.

5.3.2 Components and materials

The next step has been to identify the plastic items collected through separate collection, that translated in SEA dictionary, it means to identify which are the components. All the information gathered about the resins and their contamination levels create the backbone of the analysis, since they represent the knowledge needed to perform the substance level investigation, however, in order to scale the method through the higher levels, the identification of the components (and materials) is required. The intent was to assess the most common packaging items (bottles, glasses, cups, etc.) that compose the collected waste stream. The majority of information about wastes tend to identify the packaging wastes as an indistinct whole, without coping with the problem of identifying the different items that concur in shaping it or the proportions of each item over the total. In order to come up with a list of components to feed the SEA, we decided to screen papers and publications about the most environmentally damaging plastic products polluting landfills and seas and to filter them up to consider the ones that belong to the packaging sector. Then we included in our analysis the items that appeared with the highest frequency across different publications, assuming that these could be a good proxy to describe also the ones collected through separate collection in the reference scenario (Table 7).

Table 7 - List of plastic items gathered to identify plastic packaging items

Number	List of items	Source
1	· Food containers · Cups for beverages · Beverage containers · Plastic bags · Packets and wrappers	European commission, directive on single-use plastic (2019)
2	· Coffee cups · Plastic lids · Plastic cups · Plastic containers · Plastic plates	WWF, "*10 worst single-use plastics*", (2022)
3	· Plastic bottles · Food wrappers · Food takeaway containers · Cups · Plastic bags	City-to-Sea, "*The most polluting single-use plastic items*", (2020)
4	· Plastic bags · Plastic bottles · Food containers/cutlery · Wrappers · Plastic caps/lids	Carmen Morales-Caselles et al., (2021)
5	· Plastic beverage bottles · Food wrappers · Plastic bottle caps · Plastic grocery bags · Plastic take-out/away containers	STATISTA, "*Most common items found during the international coastal cleanup 2020*", (2021)
6	· Food wrappers · Plastic bottle caps · Plastic beverage bottles · Plastic grocery bags · Other plastic/foam packaging	Willona Sloan, "*Plastic products take over top 10 list during international coastal cleanup*" - Waste360, (2018)
7	· Plastic shopping bags · Food wrappers · Plastic bottles and lids · Plastic takeaway containers · Polystyrene containers and products · Plastic cups and lids · Disposable plastic cups, plates and bowls	Shane Cucow, "*The most dangerous plastic products polluting our oceans*" – Australian marine conservation agency, (2020)

Based on the results of these studies, Table 8 reports SEA components employed in our analysis.

Table 8 - List of SEA's components

Number	SEA Components[13]
1	Drinking bottles
2	Cleaning product bottles
3	Plastic (grocery) bags
4	Takeaway containers
5	Packets/wraps
6	cups

Six items - Drinking bottles, Cleaning product bottles, Plastic bags, Takeaway containers, Packets/Wraps, Cups - are assumed to represent the majority of plastic packaging waste from Lombardy's separate collection for the year 2019.

In order to close the chain of the extended multilevel SEA's levels, the last step was to define what are the materials that fill the gap between the substances and the components. Following the definition of SEA's components, the materials' identification was performed by means of visual inspection of the selected packaging items (e.g., a bottle has a plastic body, a lid and a label). The materials chosen to describe the components are: bottle body 1 (mainly made of PET), bottle body 2 (different structure more unbalanced through PP and HDPE), lid, label, bag body, pack body 1 (PP related), pack body

[13] Even though there is no direct distinction between drinking and cleaning product's bottles in table 5's directories, the decision to take them as separate items is due to the different substances they are composed of.

2 (PS oriented). The presence of many substances that could contribute to shaping the components impacted on the identification of the materials, that is the reason why there are some of them very similar to each other, like bottle body 1 and 2 or pack body 1 and 2: they serve the same scope, but are composed of different substances (polymers), so it is crucial to keep them separated in the analysis. Looking at the way the materials bind together assembling the components, in table 9, half of the plastic items have a multi-material structure while the other half have a mono-material structure. This characteristic has been used to verify which are the impacts that a more (or less) complex components' structure has on the final relative statistical entropy value.

Table 9 - Connection between components and materials

	Plastic body 1	**Plastic body 2**	**lid**	**label**	**Bag body**	**Pack body 1**	**Pack body 2**
Drinking bottles	*x*		*x*	*x*			
Cleaning product bottles		*x*	*x*	*x*			
Plastic (grocery) bags					*x*		
Takeaway containers							*x*
Packets/wraps				*x*		*x*	
cups			*x*				*x*

The detecting process of the materials' composition in terms of polymers has proved itself to be rather challenging, as well as crucial for the development of the method. Without a compelling description about which polymers contribute to shape a component (and relative proportions) all the SEA's attempts to assess circularity and CE actions would be useless (Nimmegeers et al., 2021). The polymeric structure of the materials considered is described in table 8. According to various internet databases, the same material could be made of different polymeric structures as well as mixes of

distinct resins. In order to take into consideration this issue, for those materials, a proportional subdivision of the polymeric structure has been considered. For example, cleaning product bottles' plastic bodies could be mainly made of PP or HDPE, according to the chemical characteristics of the liquid they are supposed to bear. Thus, aiming at keeping track of this heterogeneity, and considering that the wastes get to sorting centers as a mixture of either products made of one polymer or products made of one another (or even mixes of them), we decided to design a chemical reference structure of the cleaning product bottles containing all the possible polymers that could be found inside them in coherent proportions. This assumption has the duty to create a reliable proxy of the polymeric structure, without which it would be too difficult to perform the computation. It is important to say that the proportions considered in such structures come from simple statistical calculations made upon the information gathered through the various sources, they don't claim to provide real world exact data but rather estimates that allowed us to go through SEA's calculation.

Table 10 - List of SEA materials with their reference polymeric structure

Number	Material	Polymeric structure	Sources
1	Plastic body 1	· 99% PET · 1% Other	PlasticFinder.it – "PET, Polietilene teraftalato", (2021) WebArchive.org – "Polyethilene Teraphtalate (PET) uses and market data"(2007)
2	Plastic body 2	· 37% PP · 53% HDPE · 9% LDPE · 1% Other	PlasticFinder.it – LDPE, polietilene a bassa densità", 2021 PlasticFinder.it – "HDPE, Polietilene ad alta densità", 2021 PlasticFinder.it, "PP, Polipropilene", 2021

			Sciencing.com, “The properties of Polyethylene”, (2017) (Malpass & Band, 2012)
3	Lid	· 55% PP · 40% HDPE · 5% Other	PlasticFinder.it – LDPE, polietilene a bassa densità”, 2021 PlasticFinder.it – “HDPE, Polietilene ad alta densità”, 2021 Sciencing.com, “The properties of Polyethylene”, 2021 Malpass et al., 2012
4	Label	· 30% PP · 30% LDPE · 30% HDPE · 10% Other	PlasticFinder.it – LDPE, polietilene a bassa densità”, 2021 PlasticFinder.it – “HDPE, Polietilene ad alta densità”, 2021 PlasticFinder.it, “PP, Polipropilene”, 2021 Nimmegeers et al., 2021 Malpass et al., 2012
5	Bag body	· 60% LDPE · 26% HDPE (4% PP contamination) · 10% Other	PlasticFinder.it – LDPE, polietilene a bassa densità”, 2021 Sciencing.com, “The properties of Polyethylene”, 2021

6	Pack body 1	· 82% PP (8% HDPE contamination) · 10% Other	PlasticFinder.it, "PP, Polipropilene", 2021 Malpass et al., 2012
7	Pack body 2	· 70% PS · 12% PP (8% HDPE contamination) · 10% Other	ChemicalSafetyFacts.org, "Polystirene", (2021)

For the computation, we considered some degree of contamination in each polymeric structure, identified by the term "other" in Table 10. It is because, in this way, we can take track of some likely cross contamination percentages that could be found in the wastes. As already said, that voice embeds all those polymeric structures that do not fit into a unique category but that are composed by mixes of different resins, therefore it well suits its duty as an estimate of the structures' contamination. One other point to be clarified is the variable proportion of the voice "other" according to different materials, these amounts are driven by the number of distinct resins present in the structure. For those materials that have more than one polymer in their structure, it is less likely to find (at least at the first recycling cycle) other typologies of contaminants. In these circumstances, the presence of other plastics is reduced to minimum (1%), while for those materials that are mainly composed of a single resin the presence of other plastics is more probable (5-10%). Furthermore, in order to stay true to the results from Juan et al. (2021) in the computation, whenever the polymeric structure presents either only HDPE or only PP, the amount of cross-contamination between the two has been indicated.

Another important set of data we looked for is the weight of each material identified, useful to compute the various flow dimensions in the SEA analysis. In fact, we used these data to obtain the components' mass as the result of the sum over its materials', then, the knowledge about each material's' weight is employed to evaluate further relevant data such as each component's weight within the whole product and the SEA's mass flows. In order to get the desired amounts, we relied on the way described by Nimmegeers et al. (2021) in their work on the "plastic bag example", where they estimate the weight of a plastic bottle (component) as the sum of the single weights of its body, cap and label (materials), equivalent to 20g, 2,5g and 2,5g respectively. The weights are summarized in table 11.

Table 11 - Weights of the SEA's materials

Number	Material	Weight	Unit of measure
1	Plastic body 1	20	g
2	Plastic body 2	25	g
3	Lid	2,5	g
4	Label	2,5	g
5	Bag body	5	g
6	Pack body 1	10	g
7	Pack body 2	10	g

5.3.3 Decomposition energies

The last step of data gathering is about the energy data needed to compute the final recyclability metric. The objective is to find the amount of energy (per unit) spent during the entire process of waste gathering, sorting, pre-treatment and recycling. The energy value related to the whole recycling process of the selected components (decomposition energy) has been taken from the work of Nimmegeers et al. (2021). Considering the similarity between the steps through which all the plastic items have to pass in order to complete their recycling round loop, we decided to rely on such value

also for the other SEA's components (beyond the sole plastic bottles). In their study, the decomposition energy is obtained considering that the sorting process, together with the polymers' pretreatment activities (washing, grinding, sink-floating and near-infrared separation), account altogether for 0,36 MJ of energy consumption per kg of plastic. In addition, a 0,87 MJ/kg of energy is spent to recycle pellets via mechanical recycling, leading to a total expense of 1,23 MJ/kg (Nimmegeers et al., 2021). Subsequently, we collected the values regarding the maximum decomposition energy of each resin, defined as the energy required to produce virgin polymers from scratch, present per kg of product. The values are obtained from literature and are summed up in table 12.

Table 12 - List of maximum decomposition energies of SEA's substances

Resin	**Maximum decomposition energy**	**Unit of measure**	**Source**
PET	83	MJ/kg	Nimmegeers et al. 2021
LDPE	76	MJ/kg	Nimmegeers et al. 2021
HDPE	76	MJ/kg	Nimmegeers et al. 2021
PP	73	MJ/kg	PlasticsEurope – "*Ecoprofile of the European plastic industry: Polypropylene (PP)*", (Boustead, 2005)
PS	83	MJ/kg	EPA.gov – "*Expanded polystyrene life cycle analysis literature review*", (2021)
Other	78,2	MJ/kg	Computed

For what concerns the value of maximum decomposition energy of "other" plastics it has been estimated performing an average on the other polymers' energies. With this data on hand, we have all data needed to compute the other energy dimensions for the recycling metrics and, in general, to perform all the calculations required by the SEA.

6 *Analysis and results*

The first step to ignite the SEA development is to define what the percentages that split the whole amount of collected plastic waste into the components are, namely what is the proportion of plastic bottles over the total, what is the proportion of plastic bags over the total, and so on. This computation is required because it allows to obtain data about the components' mass flows M_f (tons), and these are then used to find the number of how many distinct items of each type is present in the wastes, by dividing each component's mass flow by the weight of that very single component (value that is given by the sum over the materials' weight that form each component). Identifying the real proportion of plastic items is impossible since such information is not available. Therefore, we decided to estimate those proportions taking as reference the lists of items described in table 7 (section 5.1, pag 40) (top 10 items found during the international coastal clean-up). The reason for this decision is double-fold: first, it is the only list that puts the items in decreasing order according to the amounts collected; second, we supposed that the plastic products gathered from the ocean clean-up[14] are likely to be the same which are daily thrown away as postconsumer waste, and so collected, thus the list is expected to give us a proxy of the proportions composing the total amount of waste analyzed. The resulting subdivision is depicted in table 11. It is ought to specify that these results may be biased due to uncertainties in data estimation. To overcome this issue, we carried on the SEA analysis for distinct

[14] There is no evidence of a correlation between the wastes gathered through separate collection in Lombardy and the ones obtained through coastal clean-up; however, the correlation described stems from the assumption that the plastic litters crowding the coasts are also the most likely items to be commonly thrown away. Therefore we supposed that they are the same, in terms of distinct plastic objects themselves, that it is possible to gather as postconsumer waste.

scenarios which can mitigate the problem of unreliability. In fact, the analysis of more than one outcome provides investigation insights coming from different perspectives and helps spotting points and relative addressing solutions from multiple sides of the problem. As it is explained in section 7 there are different possible outcomes whose variation have impacts on the final results, thus more than one combination of plastic items is considered to describe the collected plastic waste composition. The action of considering multiple scenarios increases the reliability of our results because it allows to absorb possible biases on the final results. table 13 below summarizes the reference scenario's proportions and represents the term of comparison for the following altered scenario.

Table 13 - Proportion of SEA's components in the reference scenario

Number	Component	Proportion over the total	Amount (M_f)	Unit of measure
1	Drinking bottle	20,27%	44840,481	Tons
2	Cleaning product bottle	14,09%	31169,333	Tons
3	Plastic bag	19,42%	42960,145	Tons
4	Takeaway containers	15,67%	34664,546	Tons
5	Packet/wrap	22,47%	49707,233	Tons
6	Cups	8,07%	17852,130	Tons
	TOTAL	**100%**	**221215,990**	**Tons**

The objective of the SEA is to compute the value of the statistical entropy at the substances, components and products levels. Having at our disposal the data regarding the redistribution of plastic items throughout the collected wastes, we are now ready to deepen the multilevel computation step by step.

6.1 Substance level

In order to provide a clear description of the procedure to get to the substance-level statistical entropy, it is important to proceed progressively introducing how the dimensions are computed. Recalling the statistical entropy formula, it is a function of two dimensions: the substance mass fractions ($c_{i,f}$), and the standardized substance mass fraction ($m_{i,f}$) (equation 2) . The first describes the percentage of a substance *i* present inside the component *f*. For example, in order to compute the PET mass fraction of the drinking bottle component, it has to be considered that such component is given by the assemble of three materials (plastic body 1, the lid and the label), which could (or could not) be composed of a percentage of PET substance. The mass fraction dimension aims to find what is the proportion of PET substance that concurs in shaping the component, passing through the percentage of that very same substance into the drinking bottle's constituting materials:

$$c_{PET,drinking\ bottle} = \frac{PET_{\%}^{material\ 1} * m_{drinking\ bottle}^{material\ 1}\ [g] + \cdots + PET_{\%}^{material\ 7} * m_{drinking\ bottle}^{material\ 7}\ [g]}{component's\ mass\ \ [g]} \quad (21)$$

This formula is then extended in order to cover all the substances *i* and the components *f*, in table 14 all the mass fractions computed are summarized. Given its formulation, such dimension assumes value between 0 and 1, where the first means that there is no presence of substance *i* inside *f* and the second meaning that the substance *i* accounts for the 100% of the component *f*.

Table 14 - SEA's substance level mass fractions

Substance mass fraction ($c_{i,f}$)	Drinking bottle	Cleaning product bottle	Plastic bag	Takeaway containers	Packet/wrap	cups
PET	0,792	0,000	0,000	0,000	0,000	0,000
LDPE	0,030	0,100	0,600	0,050	0,060	0,100

HDPE	0,070	0,500	0,260	0,080	0,124	0,124
PP	0,085	0,379	0,040	0,120	0,716	0,156
PS	0,000	0,000	0,000	0,700	0,000	0,560
Other	0,023	0,021	0,100	0,050	0,100	0,060

In order to define the standardized mass fraction it is prior required to compute the substance flow rate ($X_{i,f}$), describing the actual amount of a substance i inside the component f (in tons). The substance flow rate is given by the product between the substance mass fraction and the component flow rate (M_f), as it is summarized in table 15.

Table 15 - SEA's substance flow rates

Substance flow rate ($\boldsymbol{X_{i,f}}$)	Drinking bottle	Cleaning product bottle	Plastic bag	Takeaway containers	Packet/wrap	cups
PET	35513,66	0,00	0,00	0,00	0,00	0,00
LDPE	1345,21	3116,93	25776,09	1733,23	2982,43	1785,21
HDPE	3138,83	15584,67	11169,64	2773,16	6163,70	2213,66
PP	3811,44	11818,37	1718,41	4159,75	35590,38	2784,93
PS	0,00	0,00	0,00	24265,18	0,00	9997,19
Other	1031,33	649,36	4296,01	1733,23	4970,72	1071,13

The standardized mass fraction evens the component's flow rate (M_f) by dividing it over the sum of the components' substance flow rate ($\sum_{f=1}^{F} X_{i,f}$), summarized in table 16.

Table 16 - SEA's standardized substance mass fractions

Standardized substance mass fraction ($\boldsymbol{m_{i,f}}$)	Drinking bottle	Cleaning product bottle	Plastic bag	Takeaway containers	Packet/wrap	cups
PET	1,263	0,878	1,210	0,976	1,400	0,503

LDPE	1,221	0,848	1,169	0,944	1,353	0,486
HDPE	1,093	0,759	1,047	0,845	1,211	0,435
PP	0,749	0,521	0,717	0,579	0,830	0,298
PS	1,309	0,910	1,254	1,012	1,451	0,521
Other	3,261	2,267	3,124	2,521	3,615	1,298

The standardized substance mass fraction has the advantage to link the flow rate of a component to the flow rate of a substance, and so, it opens the possibility to investigate what is the dilution/concentration level of the second inside the first. This bound between the two flow rates is what allows the computation of the statistical entropy at the substance level. Yet there is one issue to tackle: although statistical entropy gives clarity about the (more or less distributed) presence of a substance within a component, substances present themselves inside the components' materials in different proportions and quantities. It means that also the maximum entropy value (equation 5, section 4.1.1, pag. 37) changes for each substance, since that is a function of the highest degree of dilution the substance could reach. For this reason, it is impossible to compare the values of statistical entropies and draw conclusions on which one is bigger and why, since a higher value does not necessarily mean higher entropy if the maximum value does not coincide. In order to come up with an indicator that would allow us to make a comparison between the values, we relied mainly on the relative statistical entropy (equation 4, section 4.1.1, pag. 37). Entropy's outputs are summarized in table 17.

Table 17 - SEA's substance level statistical entropies

Substance	**Statistical entropy**	**Maximum entropy**	**Relative statistical entropy**
PET	0,3364	2,6389	0,1275
LDPE	1,6789	2,5899	0,6483
HDPE	2,0629	3,4301	0,8489

PP	1,2593	1,8851	0,6680
PS	0,6085	2,6906	0,2262
Other	3,7712	4,0076	0,9410

Looking at the relative statistical entropies, the PET shows the lowest value because its presence is bounded to just one single component (the drinking bottle) in massive proportion. Considering that the drinking bottle component is made of different materials and PET concurs just in shaping one, the statistical entropy is quite far from its maximum value (which reflects the perfect dilution of the substance). The PS has a quite low relative entropy value too, in fact looking at tables 14 and 15 it is possible to see that it appears just in two components' structure as they share the same material. All the other polymers are characterized by quite high values of relative statistical entropies, since they appear in almost all the components' materials and in variable proportion, meaning that their dilution level is quite high.

6.2 Component level

The statistical entropy computed at the component level gives information about the degree of dilution/dispersion that characterize the analyzed plastic item. Differently from the substance-level, statistical entropy that analyzes the single substance involvement throughout all the plastic items (for example PP has high entropy since it is present in many components with different proportions), the component level entropy focuses on the single component and takes the perspective of their chemical structure's composition, namely it highlights whether the structure is complex (heterogeneous mix) or homogeneous. To obtain this entropy, we relied on the mathematical formulation described by equation 6 (section 4.1.2, pag. 38). In doing so, no further computations are required to get to the aimed component level statistical entropy since we relied on the substance mass fractions already provided in table 14 and on the so-called "normalized component mass" which is represented by the proportions in table 13. The main computation of this section concern, instead, the maximum entropy

value (equation 8, section 4.1.2 pag. 38) that considers the overall concentration of substance *i* in the total system stage ($c_{i,tot}$), the results are listed in table 1. For example, in order to get to $c_{PET,tot}$, we multiplied the proportion of PET in each material by the total mass of such material in the system, then we divided all over the total system's mass:

$$c_{PET,tot} = \frac{PET_{\%}^{material\ 1} * M^{material\ 1}\ [tons] + \ \dots \ + \ PET_{\%}^{material\ 7} * M^{material\ 7}\ [tons]}{total\ system's\ mass\ [tons]}$$

(22)

Where:

$$M^{material\ 1}\ [tons] = m_{drinking\ bottle}^{material\ 1}\ [g] * \#drinking\ bottle + \cdots + \ m_{cups}^{material\ 1}\ [g] * \#cups$$

(23)

Table 18 - Overall concentrations of substances at system's stage

Substance	**c *i,tot***
PET	0,1606
LDPE	0,1661
HDPE	0,1856
PP	0,2707
PS	0,1549
Other	0,0622

The system stage point of view is required because the maximum component-level entropy corresponds to either the case in which all substances are present in one material flow or equally distributed. The availability of these data allowed us to compute the relative component-level statistical entropy, which is the relevant dimension sought that allows the comparison between the different items. The entropies at the component level are listed in table 19.

Table 19 - SEA's component level statistical entropies

Component	Statistical entropy	Maximum entropy	Relative statistical entropy
Drinking bottle	0,226	2,481	0,091
Cleaning product bottle	0,208	2,481	0,084
Plastic bag	0,285	2,481	0,115
Takeaway container	0,227	2,481	0,092
Packet/Wrap	0,291	2,481	0,117
Cups	0,148	2,481	0,060

Among all, the relative entropy at the component level is the more insightful dimension, because it shows the state of the distinct plastic items before they begin the whole recycling process. It stands out that all the relative entropies assume rather low values; this may be due to the fact that all the deemed components are assumed to have a low level of architectural complexity, in fact no more than three materials are needed per each. Therefore, when it comes to relate the statistical entropy to the maximum distribution that the substances could have among the system's structure, the relative entropy value happens to be low. Plastic bag and Packet/Wraps components have a slightly higher value compared to the others because they are, together with drinking bottles, the two most recurring plastic items among the wastes (proportions in table 13). Although the structure of drinking bottles is mostly composed of one single substance (PET), which constitutes almost totally its main material (99% of plastic body 1), the other two components (Plastic bags and Packet/Wraps) present a higher degree of material dispersion, which results in higher relative entropy. On the other hand, the explanation for the lower relative entropy value of the other components lies in their less dense proportions over the total. Although they are averagely composed of more materials (which have also a more diluted chemical compound), they are fewer in number and so their impact is reduced. In addition, it is important to recall that the analysis is based on the first recycling loop that furtherly explains the values obtained, in fact the lack of contamination due to reprocessing keeps them low.

6.3 Product level

The highest level of analysis considers the evaluation of the statistical entropy when all the components are seen as a whole. Considering the product-level entropy formula (equation 9, section 4.1.3 pag. 39), it is possible to note that it is a function of two main dimensions: the component-level statistical entropy and the component concentration ($c_{n,p}$). The first is presented in table 19 and accounts for the distribution of the substances within the wastes' distinct plastic items. As regards instead the component concentration, it accounts for the distribution of the components within the total waste amount, considering the percentage of each over the total. It is important to clarify that the component concentration, differently from the components' proportions (where the ratios are based on the mass, table 13), is computed as the number of each specific component over the total number of distinct items that form the wastes, hence it counts the items not their weight, listed in table 20.

Table 20 - SEA's product level component concentration

Component	$c_{n,p}$
Drinking bottle	0,0884
Cleaning product bottle	0,0512
Plastic bag	0,4234
Takeaway container	0,1708
Packet/Wrap	0,1959
Cups	0,0704

The maximum entropy (equation 12, section 4.1.3, pag. 40) corresponds with the situation where all the substances are uniformly distributed over the product's components. In this case, maximum value is determined by the total number of components (N_{tot}). Entropies at the product level are listed in table 21.

Table 21 - SEA's product level statistical entropies

Product	**Statistical entropy**	**Maximum entropy**	**Relative entropy**
Plastic waste	1,559	34,241	0,046

The product level relative entropy value is close to zero because of the concentrated chemical structure of the majority of components. Adopting the system's perspective all the components are considered to have a chemical structure that is (more or less) concentrated around one or few substances. The little variations in the components' chemical structure that provided the results about concentration/dilution at the lower levels of analysis become negligible when the product level perspective is adopted. Hence there is no component, among the ones considered, that stands out for a uniform distribution of substances as if each component's structure is approximated on account of its few main substances.

6.4 Recyclability metric

The last step of the multilevel SEA addresses the creation of a metric able to provide quantitative information on the plastic items' recyclability. The entropy values computed found at each level of analysis are crucial to understand the structure's complexity of the collected wastes, though it does not directly give information about their recyclability. In order to obtain the recyclability indicators, we computed the entropy values with the decomposition energies, as described in section 4.2. The

component level is the most relevant scenario to investigate when dealing with recycling, since information about distinct plastic items can be of particular interest for industrial purposes. The methodology to compute the recyclability metric requires the determination of the components' relative decomposition energies ($E^c_{n,rel}$) according to the equation 14 (section 4.2, pag. 43). In doing so, we relied on the energies of recycling process presented in section 4.2 (E^c_n = 1,230 MJ/kg) and on the energy values related to each substance's maximum decomposition energy (table 12), describing the requirements to produce virgin polymers ($e_i^{production}$). These energy values have been used to determine the unique value of the components' maximum decomposition energy (E_{max}) to be used as a leveler in the relative energy computation. From equation 13 (section 4.2, pag. 42) we obtained:

$$E_{max} = 77{,}533\ MJ/kg$$

(24)

Considering that the energy of recycling has been assumed to be the same for all the items, and that the components' maximum decomposition energy (equation 25) is a unique value obtained merging the substances' energies of production, also the relative decomposition energy results to be a unique value for all the components:

$$E^c_{n,rel} = \frac{E^c_n}{E_{max}} = 0{,}01586$$

(25)

At this point, we have all the dimensions required to compute the components' recyclability metrics ($R_n^{(1)}$, $R_n^{(2)}$) described in equations 16 and 18 (section 4.2, pag. 43, 44). The values obtained are shown in table 22.

Table 22 - SEA's recyclability metrics on the components

Component	R1	R2
Drinking bottle	0,895	57,296
Cleaning product bottle	0,901	57,740

Plastic bag	0,871	55,804
Takeaway containers	0,894	57,258
Packet/Wrap	0,869	55,645
Cups	0,925	59,271

The two recyclability indicators provide the same information; for the purpose of this dissertation, we employed: $R_n^{(1)}$ to show the gap with the maximum (ideal) recyclability, and $R_n^{(2)}$ because it provides insights on the distances between each other values, hence offering a good ground for comparison. The assumption that the energy of recycling is the same for all the components is introduced in order to stay true to the SEA development described by Nimmegeers et al. (2021) in their work. In addition, it reduces the complexity of the calculations since, in this way, the main contribution to the recyclability value is on behalf of just the components' relative entropy. In fact, it is possible to note how the most difficult items to recycle correspond to the ones with the highest entropy value (Plastic bags and Packet/Wraps). On the opposite side, the items which, according to the indicators, pose weaker barriers to recycling are the ones with the lower entropy value (and consequently higher recyclability index). Looking at the $R_n^{(1)}$ values in table 22, they are all close to the maximum value (equal to 1) showing how, considering that the reference recycling procedure is the mechanical recycling, the recyclability of all the components is quite high. Therefore, it is possible to theorize that, as expected and highlighted from the results, the mechanical recycling is the recycling way associated to the highest degree of circularity.

7 Sensitivity analysis

A sensitivity analysis was conducted to reduce potential biases from the data collection and computation processes. This analysis tests the robustness of the outputs by means of the significant variation of a specific set of input data, with the intent to simulate a set of scenarios that could happen. The aim of the analysis is to determine the externalities (indirect positive/negative effect of external events on the method's output), reflected by input data variations, whose impact can be interpreted by looking at the SEA's recyclability metric reaction.

According to equation 6 (section 4.1.2, pag. 38), the main factors that could impact on the component level relative entropy value are: the chemical structure (in terms of polymers distribution) ($c_{i,f}$), and the proportion of each component over the total. These two are, therefore, the most insightful dimensions to deem when it comes to assess the final outputs' reaction, however, considering that the scope of our analysis is to provide insights about CE implementation in the industrial context, we chose to vary only the components' material flows (M_f). In fact, recalling that we set the analysis on the first loop of recycling, we do not have access to data concerning the chemical contamination levels apported by the continued reprocessing, thus, whichever variation in this direction would not be based on existing data but on additional assumptions. For this reason, the first perspective of the sensitivity analysis is focused on spotting what are the implications of a change in the components' material flows. The second objective is to release the assumption according to which all plastic items are required to have the same energy of decomposition, in favour of the more realistic perspective where the energies depend on the specific case (and process) considered. However, it is ought to mention that univocal data linking each specific component to its own value of energy requirement (e.g., the energy requirement to recycle a drinking bottle) do not exist, all related information, if any, concerns the substance level. It leads to the necessity of introducing some additional assumptions allowing to scale the available energy data from the substance level to the single component's.

7.1 Scenario 1: variation of components' flow rate proportions

The components' material flows proportions of the reference scenario are listed in table 13 (section 6, pag. 65), they describe the recurring frequency of each one among the total waste. The sensitivity analysis allows us to assess whether an increase of those components that present a more structured composition provide significant variation on the output entropies in the SEA. In fact, in this way, we try to spot eventual entropy changes when we consider the combined effect of a complex structure and a dominant material flow. In order to do so, we introduced a 5% increase on the multi-materials components' proportions and a reduction of the mono-material items' proportions by 10% so to keep the same total amount (table 23).

Table 23 - Components' material flow proportions variation from reference to new scenario

Reference data		[tons]		New scenario		[tons]
drinking bottle	20,27%	44835,432	+5%	25,27%	55901,281
cleaning product bottle	14,09%	31176,574	+5%	19,09%	42230,133
plastic bag	19,42%	42966,707	-10%	9,42%	20838,546
takeaway containers	15,67%	34672,151	-10%	5,67%	12542,947
packet/wrap	22,47%	49710,273	+5%	27,47%	60768,033
Cups	8,07%	17854,853	+5%	13,07%	28912,930

The component level relative statistical entropy (equation 7, section 4.1.2, pag. 38) is a direct function of the proportions describing the material flow rate (percentages in table 13, section 6, pag. 65). It is reasonable to expect that an increase/decrease in the material flow rate proportions should have a strong impact on the final results, seeing the entropy of the new most recurring items grow and vice versa. For what concerns the substance level relative entropy, instead, the relationship with the altered material flows is indirect, by means of the standardized mass fraction ($m_{i,f}$) (equation 2, section 4.1.1, pag. 36), therefore the impact on the final entropy values is expected to be less significant. In other terms, what we expect to see is a worsening of the recyclability indexes of those components that have their proportions increased, suggesting that, regardless their polymerics structure that does not change, a higher amount of multi-material plastic items pose higher impediment to recycling since it

augments the efforts required to go through all the process's steps. The results of the sensitivity analysis are summarized in table 24.

Table 24 - Comparison on statistical entropies at substance and component level between the two scenarios

		Reference scenario	**New scenario** (+5% / -10%)	**Δ (%)**
SUBSTANCE LEVEL ENTROPY	PET	0,1275	0,145	13,71%
	LDPE	0,6483	0,739	14,05%
	HDPE	0,8489	0,828	-2,46%
	PP	0,6680	0,694	3,94%
	PS	0,2262	0,230	1,63%
	Other	0,9410	0,932	-0,94%
COMPONENT LEVEL ENTROPY	drinking bottle	0,0910	0,117	28,87%
	cleaning product bottle	0,0840	0,118	40,06%
	plastic bag	0,1147	0,058	-49,86%
	takeaway containers	0,0917	0,034	-62,60%
	packet/wrap	0,1172	0,148	26,38%
	Cups	0,0597	0,100	67,42%

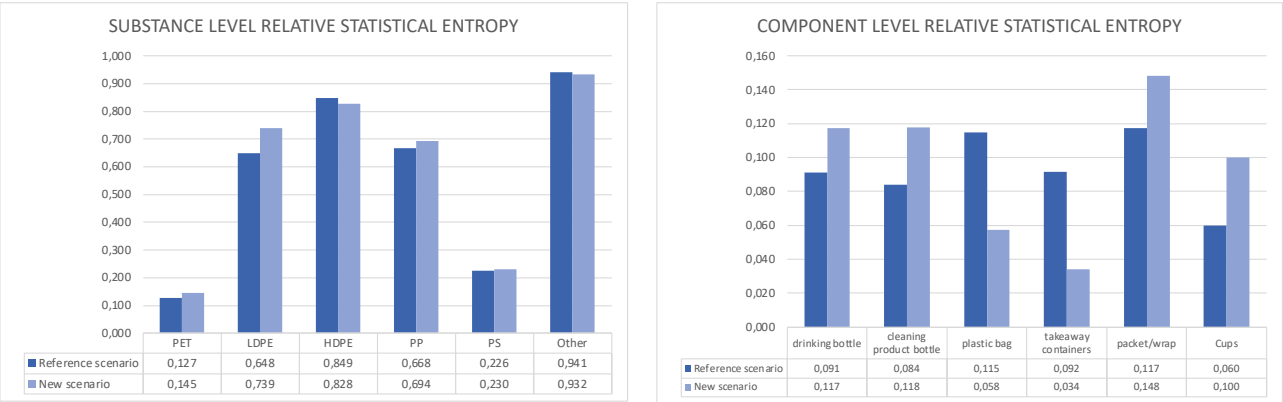

Figure 11 - Entropy comparison between the reference and new scenario

The results of the substance level entropy confirmed our expectations that the variations are generally small for substances, implying a notable robustness of such dimension to components' material flows changes. The first thing to highlight is that, according to the percentages (of components' material flows) that shape the two scenarios (table 23), the reference scenario results to be slightly more diluted

than the new one as it is showed by the two standard deviations: $\sigma_{reference\ scenario} = 0{,}0521$, $\sigma_{new\ scenario} = 0{,}0875$. This is the reason for the small changes noticeable in table 24. Positive variations reflect a lower densification of the substances present into the components' structure through the materials, that favor the dilution (increasing the complexity) as a consequence of the new higher recurrence of multi-material items. On the other side, negative variation describes the opposite phenomenon. However, the low deltas obtained suggest that the variation implemented does not have a sensible impact on the substance level entropies, which can be considered robust results.

For what concern the component level, the sensitivity analysis, provides much more evident variations in the relative entropies. The new absolute entropy values (0,117, 0,118, 0,058, 0,034, 0,148, 0,100) are still close to their minimum (0). However, given that only the first recycling loop has been considered, though, if we look at the deltas, the deviations from the reference case become much more significant. The first thing that comes to sight is that the components that see an entropy increase are exactly the ones whose percentage of recurrence have been increased by 5%, and the opposite for those that have been decreased. It is the straightforward consequence of the direct relationship pending between the components' relative entropy and the components' material flows and accounts for the numeric entity of the changes. Deepening the analysis is however possible to get further insights about the reasons that may comport such consistent variation of the entropies, in fact, it is possible to get information about how the outputs change by looking at the new configuration of the standardized mass fractions ($m_{i,f}$). As expressed in section 6, the standardized mass fractions links the substance flow rates within each component's material flow rate, providing a way to visualize the dilution/concentration of each component's structure. When the boxplot is narrow it means that the structure is more concentrated (the majority of the values are close to the average), when instead the boxplot is wide the substance dispersion is higher, meaning that the structure is

more diluted. As it is possible to see from the graphs in figure 12 (which are computed on the standardized mass fractions), the increase in the components'

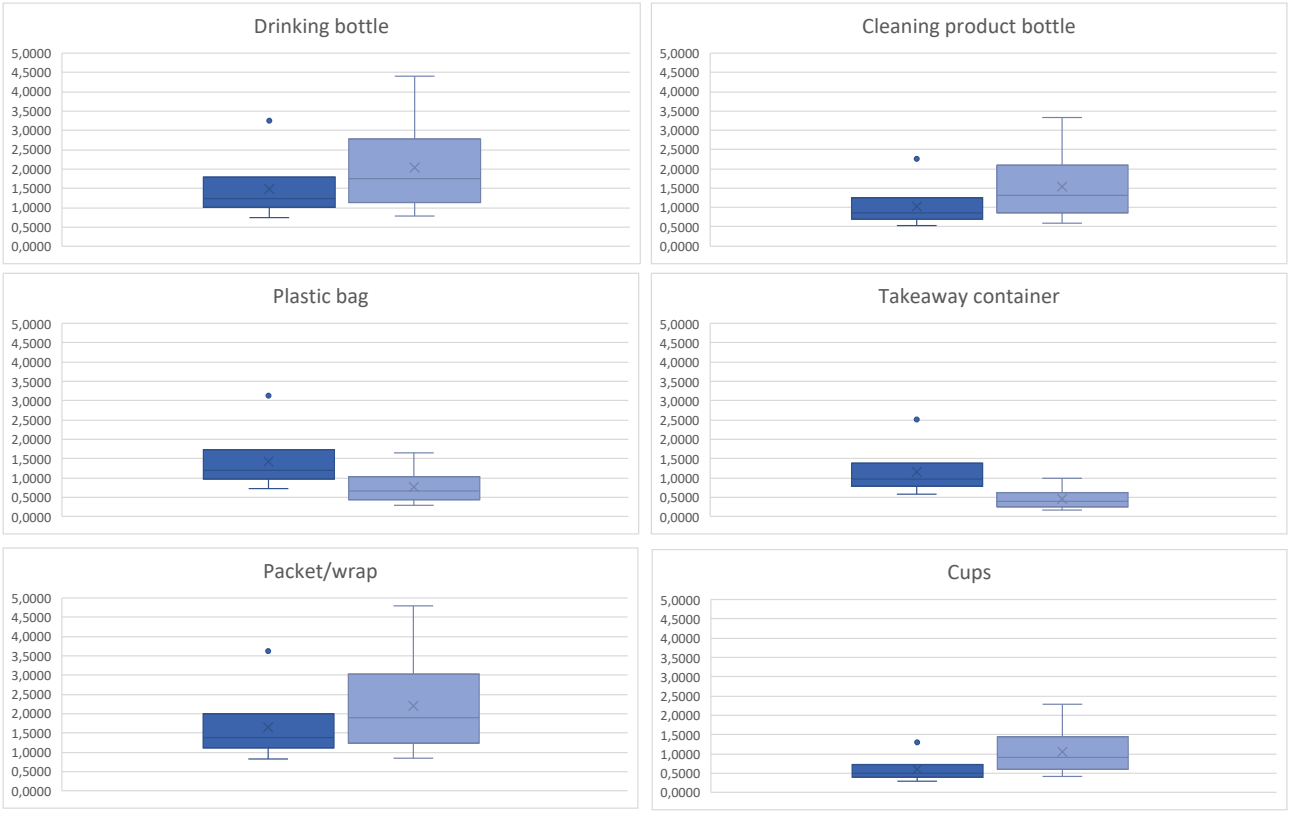

Figure 12 - Comparison on standardized mass fractions of substances within components between the two cases

material flow is assumed to be directly related to a higher dilution of the components' chemical structure, in fact, the boxplots of drinking bottles, cleaning product bottles, packets/wraps and cups show an enlargement going from the reference scenario to the new one. In parallel, the components whose proportion was decreased show also the tendency of their structure to become more concentrated, implying a reduction of their entropy and thus an increase of their recyclability. The analysis of the standardized mass fractions allows to give an explanation of the likely reason why the entropy of the components increases when the chemical structure of them is assumed to remain unchanged. The column charts in figure 11, are meant to show graphically the distances between the entropy values of the two cases.

As regards the product level of analysis, the variation produced by the sensitivity analysis is almost negligible (-0,99%). According to table 25, there is a little decrement in the relative entropy that might be due to the more concentrated configuration of the new scenario (that has a higher components' material flow distribution standard deviation) against the reference one. Despite this, it is reasonable to think that the entity of the component variation is too small to provide significant impacts at the product level.

Table 25 - Comparison on statistical entropies at product level between the two scenarios

		Reference scenario	**New scenario** (+5% / -10%)	**Δ (%)**
PRODUCT LEVEL ENTROPY	Plastic waste	0,0455	0,0451	-0,99%

When it comes to analyzing the recyclability index, it is important to keep in mind that, according to formula 18 (section 4.2, pag. 44), it is a function of the combined effect of the components' relative entropy and the components' energy of recycling. However, both in the reference and in the new scenario, the assumption of equal energy of recycling for all the components stands; therefore, the only contribution given to the recyclability indicator is given by the variation of the components' entropies. It means that we should expect to see an increase of the recyclability for those components that have a negative variation of their entropy (lower complexity, higher recyclability) and a reduction of the indicator when, instead, the entropy increases. Table 26 summarized the obtained results.

Table 26 - Comparison on recyclability indicators between the two scenarios

R2	drinking bottle	57,30	55,51	-3,12%
	cleaning product bottle	57,74	55,49	-3,90%
	plastic bag	55,80	59,27	6,21%
	takeaway containers	57,26	60,73	6,07%

	packet/wrap	55,65	53,57	-3,73%
	Cups	59,27	56,60	-4,50%

Due to the fact that only the entropy value is able to affect the recyclability indicator, this latter does not represent a very informative dimension in this first sensitivity analysis developed.

7.2 Scenario 2: variation of components' decomposition energies

Maybe the strongest assumption set during the preparation and development of the SEA method concerns the identification of a unique value of required energy to recycle the different components. While it is reasonable to expect that some of these steps could be shared by different plastic items' flows, thus delivering the same energy requirement, it is quite strong to state that the entire process can be assumed to be equal for all the components' flows, since they present different characteristics that may need different ways to perform the activities. The objective is to release this constraining assumption in favour of the introduction of a specific decomposition energy requirement for each component analyzed and assess how the recyclability indicators react when they become subject to the combined effect of both the components' relative statistical entropy and a specific own energy of recycling.

The research of the specific energy values has brought to light that is not possible to obtain energy data referring directly to the whole component (e.g., energy required to recycle a drinking bottle or a plastic bag), but all the available information regards the energy required at the substance level (e.g., energy required to recycle PET or PE, etc.). In particular, from a deep literature review, it has been possible to get energy data only about the recycling of PET and HD/LD-PE resins, while no straightforward information has proven to be available about the other resins (PP, PS and Other). The reason lies in the higher involvement of PET and HD/LD-PE resins in the mechanical recycling

process rather than the others which are, in comparison, less common to be treated that way, therefore they do not have access to the same wide span of information as the firsts do. In order to solve this issue and set a decomposition energy value for each plastic resin, we decided to keep for PP, PS and Others, the same energy expense proxy taken from Nimmegeers et al., (2021) employed in the development of the reference scenario (1,23 MJ/kg). Table 27 summarizes all the energy of recycling gathered from literature for the different plastic resins. It is important to specify that each energy listed is the sum of the energy requirement of each step of the recycling process.

Table 27 - Substances' energies of recycling

Resin	Energy of recycling	unit of measure	Source
PET	2,16	MJ/kg	University of Cambridge – "*Energy balance calculation in recycling one PET bottle: Estimation and Qualitative Analysis*" (2021)
LDPE	3,77	MJ/kg	*Vlachopoulos (2009)*
HDPE	3,77	MJ/kg	*Vlachopoulos (2009)*
PP	1,23	MJ/kg	*Nimmegeers et al. (2021)*
PS	1,23	MJ/kg	*Nimmegeers et al. (2021)*
Other	1,23	MJ/kg	*Nimmegeers et al. (2021)*

Leveraging on these substance-related decomposition energy value, in order to get to the component-related energies, we performed a linear combination involving as weights the mass fractions ($c_{i,f}$) as follows:

$$E_f^{recycling}\ [MJ/kg] = c_{PET,f} * 2{,}16 + \left(c_{LDPE,f} + c_{HDPE,f}\right) * 3{,}77 + \left(c_{PP,f} + c_{PS,f} + c_{other,f}\right) * 1{,}23$$

In this way the decomposition energy of each plastic item is built based on the composition, in terms of substance configuration, of that single component. The obtained values are listed in table 28.

Table 28 - Components' decomposition energies

Component	Decomposition energy [MJ/kg]
Drinking bottle	2,221
Cleaning product bottle	2,755
Plastic bag	3,416
Takeaway containers	1,560
Packet/wrap	1,698
Cups	1,799

It is possible to see how in this new scenario the recycling energies vary one from the others, designing a more realistic scenario where to assess the embedded recycling content of each component. Lower energy requirement implies a higher recyclability content of the component, while a higher energy request causes the indicator to drop, this relationship can now be put together with the impact provided by the entropy to shape a more reliable value of the SEA's recyclability metrics. With this procedure, the final recyclability indicators do not take into considerations just the complexity of the components' structure in terms of dilution/concentration of the substances, but also what are the substances involved and how they contribute to their recycling. For example, looking just at the entropy values of plastic bags and packet/wraps, they present similar values (0,1147 and 0,1172 respectively), if then the energy required to recycle both is assumed to be the same it is reasonable to expect very similar values of their recyclability indicators. However, leaning on the values listed in table 28, we can note that their recycling energies are quite different, because of the different substances concurring in shaping them (associated to different efforts of recycling), so, despite their similar entropy, their recycling content is expected to be quite different.

Table 29 - Comparison on recyclability indicators between the two scenarios

		REFERENCE SCENARIO		NEW SCENARIO		Δ (%)
		R1	R2	R1	R2	
RECYCLABILITY INDICATORS	drinking bottle	0,895	57,30	0,883	31,73	-44,61%
	cleaning product bottle	0,901	57,74	0,883	25,78	-55,36%
	plastic bag	0,871	55,80	0,846	20,09	-63,99%
	takeaway containers	0,894	57,26	0,890	45,13	-21,18%
	packet/wrap	0,869	55,65	0,863	40,31	-27,55%
	Cups	0,925	59,27	0,918	40,51	-31,64%

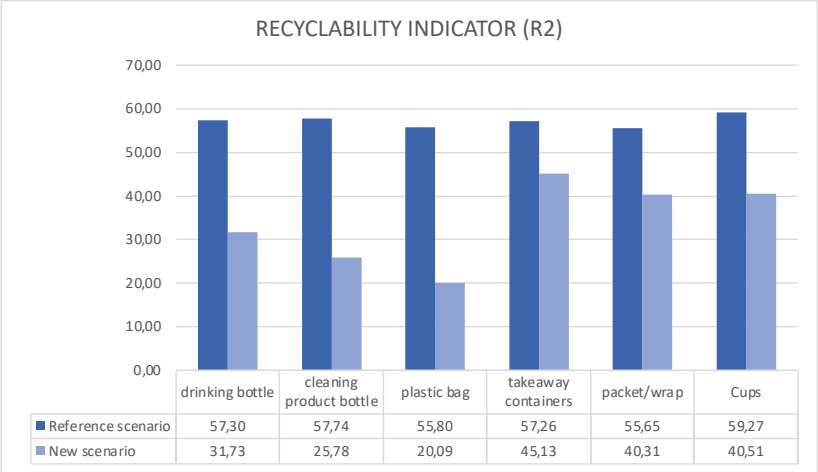

Figure 13 - Recycbality indicators comparison between the reference and new scenario

Table 29 shows the recyclability indicators both for the reference scenario and the new one (the deltas are computed using the $R_c^{(2)}$ since it better shows the differences between the different components, and so does the column chart graph in figure 13). As expected, the values obtained for the new scenario assume a more dispersed configuration due to the impact of the specific recycling energies introduced, in fact, if we compare the standard deviations of the two cases ($\sigma_{reference\ scenario} = 1{,}338$, $\sigma_{new\ scenario} = 9{,}270$) we can see that in the second case the dispersion is almost 7 times higher. The recyclability has decreased for all the components since the new energies introduced are all higher than the unique proxy employed in the reference case, in particular, it is worthy to look at

the distance, in the new scenario, between the recyclability indicators of plastic bags and packet/wraps. As previously said, we expected to see a differentiation between the two values and, in fact, it can be noted that now the second value is almost twice as high as the first, despite their entropy being very similar. According to the numeric results, therefore, it may be possible to state that not only the way the substances are distributed within the plastic structure impacts on the items' final recyclability but also the nature of the substances present in the configuration, since they are associated to particular energy requirements for the recycling process.

7.3 Comparison with chemical decomposition energy

The scope of this further analysis is to replace the recycling energies of the reference case (1,23 MJ/kg) concerning the mechanical recycling, with the energy requirement to recycle the components through chemical recycling, aiming to assess the new value of the recyclability indicators related to the new destination of the gathered waste. According to Jeswani et al. (2020), the energy requirement to pyrolyze 1 ton of plastic waste is 3260MJ (3,26 MJ/kg), this value has been used as a proxy for the recycling energies of all the components considered.

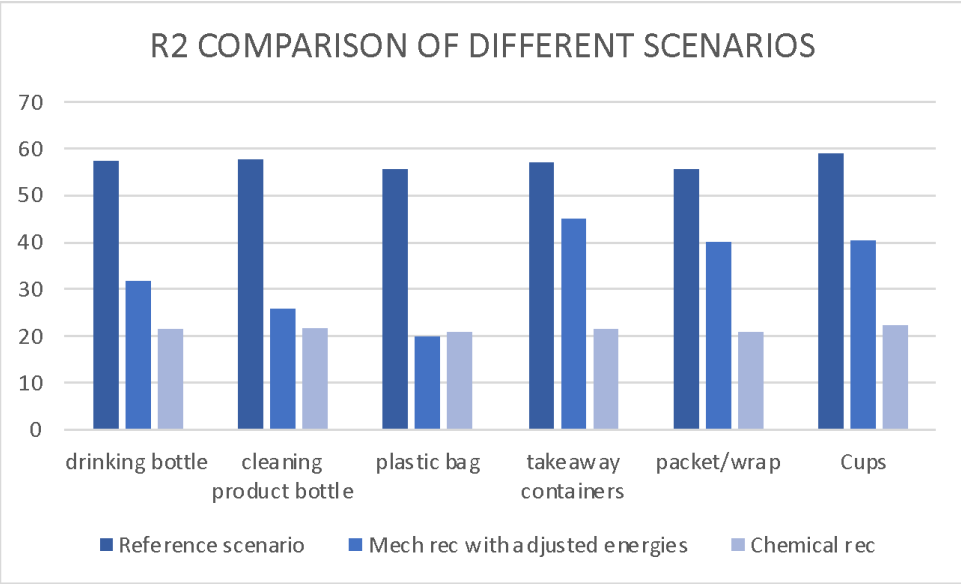

Figure 14 - Recyclability indicators comparison among different scenarios

The column chart in figure 14 graphically provides a comparison between the different scenarios: It is possible to see that the recycling indexes obtained with the chemical recycling energies are sensibly lower than the ones referring to the reference scenario (when the assumption of equal mechanical recycling energies stands). It proves that, because of the higher energy request to go through the CR, it is the second preferable choice after the MR. When we consider the scenario presented in figure 14 (with adjusted values of mechanical decomposition energy), though, the gaps between the indexes are found to be reduced. In particular, it is interesting to look at the case of the plastic bag component: here the combined effect of a high relative entropy value and of a high energy requirement to perform the recycling brings the recyclability index of the MR to be disadvantageous when compared to its CR counterpart. This seems to provide evidence that when the items' plastic structure becomes too complex (e.g., after many recycling loops), MR cannot be an advantage against CR anymore, but the second starts to represent the most sustainable solution for recycling.

Therefore, it is possible to summarize the results considering that the Mechanical recycling is the highest degree of circularity procedure between the two: it accepts pure-structure plastics and retains much of their embedded value. However, the effect of downgrading and contamination of reprocessing, could entail the conditions that make the mechanical the preferable one, leading the chemical recycling to become the best option.

8 Discussion

Based on our results, we were able to answer the dissertation research question on the relevance and pertinence of SEA to predict the degree of circularity of technological downstream interventions, with a focus on plastic recycling technologies. The dissertation results enabled us to identify the role of the SEA in advancing the CE of plastics, what is meant with the term "degree of circularity" and the SEA's contribution in measuring it, and the downstream interventions that could be derived by leaning on the results of such method. The presence of distinct dimensions where the CE implementation actions stem from, highlights the presence of various sources where to look for value extraction, sticking to the plastic waste management: the economic, environmental and social dimensions jointly offer different but intertwined domains to be explored by CE decision makers to set the right path (Iacovidou, et al., 2017). Literature offers a rather wide span of established methods for assessing CE initiatives that focus on few or even a single domain for their applicability, producing targeted results to be processed within their defined area of influence, and so does the SEA method. In order to identify which is the cutting-edge role of the SEA among the existing circularity assessment methods, it is crucial to recall that the CE concept is far from having a unique definition and it is often referred to as an umbrella concept (Kirchherr et al., 2017). The plurality of attributable definitions of CE together with the lack of standardized frameworks contributed to the emergence of different assessment methods, with each trying to bring solutions to a particular line of questioning and to cope with a higher or lower level of analysis, approaching the investigation differently (Mulrow & Santos, 2017; Corona et al., 2019). SEA positions itself in this multitude of assessment methods as a further tool able to enlarge the CE definition coverage, dealing with the gaps left by the others. If compared to LCA, SEA has the characteristic of providing an a priori assessment of the plastic waste's recyclability level, namely it sets the conditions to predict in advance the required actions to implement. The SEA's novel approach to circularity assessment lies in the very dichotomy between reaction and prediction: LCA is a method that spots the processes' criticalities (in terms of

environmental damage) and suggests the preferable option by means of comparative analysis upon a set of alternatives, telling, in this way, what is the most circular intervention to fix the system's inefficiencies. The point is that data needed to compare alternatives, with the aim of identifying the best way to CE enhancement, become available only after the process takes place, constraining the LCA to work as a reaction-based method. SEA moves the focus from the comparison of alternatives to the detailed description of the context, putting the lights on how the things are, and describing them by a recyclability metric which guides along the path of how they could be. Simplifying the procedure, wastes are taken and analyzed to obtain a recyclability indicator based on their amount and chemical composition. Leaning on such indicator we can predict, before the plastic waste go under the process, how complex the recovery would be and, so, which would be the preferable choice case by case. Similarly to SEA, also the MFA methodology provides the description of the state of the art of the process; the difference between the two methods is that

while MFA gives a static perspective of the material and energy balance in a resource management optic, SEA moves from the scenario description to the suggestion of guidelines about the best technology to use to get the highest circularity level possible. Therefore, it is possible to say that both methods are thought to give a systemic perspective: MFA on the resource management of the present state and SEA on the future recyclability routes to pursue.

When it comes to talk about CE's assessment metrics, another relevant topic is the level of analysis of CE measurements (described in section 3, pag. 8). The fact that circularity can be assessed at different levels further explains the existence of differences between the assessing methods: each of them has specific targets and, according to them, understanding the right level for the method employment is crucial to get the maximum yield on designing CE strategies (Saidani et al., 2019). The LCA method, given its goal of comparing different alternatives, provides its maximum potential when it is employed to look for CE intervention to be applied at the micro level, namely to one single

industry's product/process. If the surrounding context, where to look for CE strategies, adopts higher spatial scales (groups of industries, cities or even nations), the LCA contributions become weaker since there is no advantage in performing a comparative analysis. In these cases, in order to gauge the circularity of the activities to promote, it is more fruitful to rely on the perspective-based methods such as MFA and SEA itself, that can provide important insights on all the different levels of analysis (Linder et al., 2017). In this dissertation, SEA has been mainly applied at the micro level, assessing the recyclability of substances and components with reference to their possibility to be processed by mechanical recycling, however, the method offers the possibility to produce strategies for CE promotions also at higher levels. It has been described that, although it represents the preferable choice, the mechanical recycling is not the only recycling (or, in general, recovery) option to treat plastic waste; therefore, considering a framework where the different existing recycling routes are bound together working synergically, the recyclability metrics derived by SEA offers the possibility to be used as a scoreboard, addressing wastes to each (recycling) destination according to the recyclability value they have. In this way, the spatial scale of applicability moves from a scenario where the single recycling process is taken into consideration to a larger one where more processes (different recycling routes) are reckoned together to fully exploit the circularity potential of plastic wastes. This example of SEA applicability at the meso level can be further expanded to consider possible uses of such methodology at the macro level of analysis. The macro level application of the SEA method could be theorized as the possibility to create cross-sector strategies, involving the entire plastic value chain (forward and reverse chains). The global penetration of the CE philosophy involves the creation of an entire economic system that works synergically according to the industrial ecology rules. The SEA method could be coupled with both other methods' outputs and some macroeconomic indicators (such as GDP and population growth rate) to provide regional/national institutions with a comprehensive tool useful to plan and set policies and rules to link the efforts of multiple sectors and pave the way for such CE expansion.

In order to practically explain the role of the SEA methodology in the context of this dissertation and its pertinence to assess the CE plastic system, it is crucial to rely on the degree of circularity concept. By recalling the "technological scope" of section 2.2 it is possible to state that, in the spirit of promoting CE initiatives, primary recycling is the preferable option (as it keeps the majority of the embedded value) but it is constrained by its ability to treat only homogenous and uncontaminated plastic structures (Singh, et al., 2017). Otherwise, when the contamination downgrading effect of their structure is such to be not negligible, they have to be processed through secondary, or even further, recycling routes (Singh et al., 2017). From the SEA perspective, we have labelled the mechanical recycling with the highest degree of circularity thanks to its plastic-to-plastic conversion procedures (deeming the closed loop one step ahead the open loop), and the chemical recycling with a lower degree of circularity as, instead, is mainly employed for plastic-to-fuel applications (as it is graphically showed in figure 15).

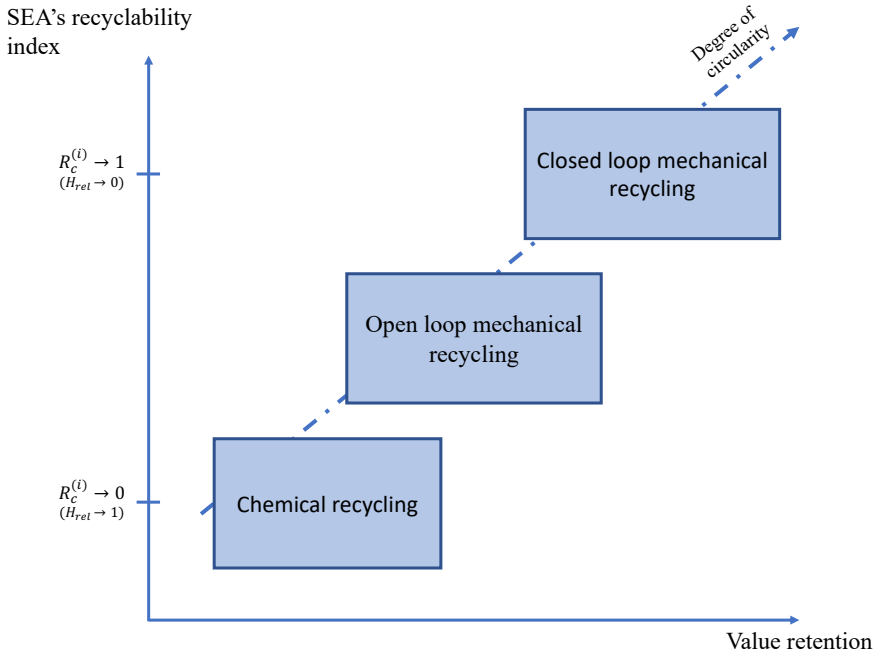

Figure 15 - Graphical representation of recycling technologies' degree of recyclability

In these terms, recalling that the employment of one method rather than the other is defined by the structural complexity of the plastic wastes (or in other terms by their entropy), the recyclability

metrics obtained from SEA can be used as a tool to properly address the plastic wastes to their suitable recycling route. When the recyclability indicator shows a high value, it means that the structure's complexity of the plastic items is low and therefore it could be processed through closed-loop mechanical recycling, allowing to preserve the highest share of value. On the contrary, when its value is low, the effect of the structure's contamination is such to determine for them lower degree of circularity, such as open-loop recycling or chemical recycling.[15]

Although it is not ready to be scaled up to a commercial level, the fact that through chemical recycling it is possible to obtain polymers and monomers chains paves the way for new circular recycling outputs. In fact, chemical recycling has the potential to provide a more sustainable alternative to the employment of virgin raw materials; its output materials have the same chemical structure and performance as the virgin polymers, and furthermore do not involve new extraction of fossil fuels. Hence, it could be theoretically used to decouple the plastic production from raw materials depletion (Davidson et al., 2021; Meys, et al., 2020). The maximum gain we can obtain, in terms of process circularity, is represented by the design of an industrial system that reckons the mechanical and chemical recycling as complementary parts of the same whole recovery process (Al-Salem et al., 2009). This model is termed

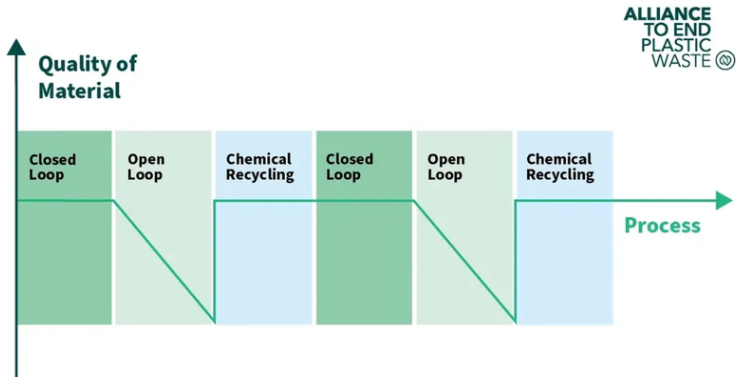

Figure 16 - The cascade recycling mode, "Thoughts from the alliance: The cascade recycling", Martyn Tickner 2021 – Alliance to end plastic waste

[15] One of the main end-markets for chemical recycling outputs from gasification and pyrolysis is crude diesel, making plastic-to-fuel a preferred route (ZWE, 2019). However, this option undermines the principles of CE and decarbonization agendas (e.g., EC, 2015a, 2018), as plastic-to-fuel does not reduce the global demand for plastic nor it produces a viable substitute for fossil-based plastics (ZWE, 2019). For this reason it is deemed as lower degree of circularity than the other methods

"cascade recycling" (figure 16) and involves the use of the closed-loop recycling as much as possible (until the quality of the feedstock allows it), then, it switches to open-loop recycling to keep exploiting the plastics' embedded value as long as market opportunities make economic sense. Once the materials' quality drop no longer justifies the rounding loops, chemical recycling is considered to lift what cannot be mechanically recycled back to the initial value in the form of new raw materials (Tickner, 2021).

The SEA results to be a very useful tool to study, design, assess and implement such recycling framework because it provides recyclability metrics built on the statistical entropy of the plastic products, and thus it keeps track of the downgrading quality of the products cycle-by-cycle. In order to clarify the SEA's role with respect to the other CE assessing methods, it is useful to recall that every time a recycling process (of whichever type) takes place, the entropy of recycled plastic is reduced, but never equal to zero. Indeed, when products start a new lifecycle, they begin with an initial entropy that is higher than zero. According to the output value of SEA's recyclability metric, after each cycle it is possible to decide whether the product can be sent to closed-loop recycling (highest degree of recyclability), or it requires to go for lower degree of recyclability recycling methods that entail more its quality. The decoupling point between closed and open loop recycling is represented by the technical possibility to perform one other closed loop, if the structure has become too complex (high entropy) closed loop would not be technologically possible anymore, then the switching to open loop recycling is required. Going further with the process, repeated open loop cycles do not have any technical constraint that could prevent the plastic product to go through one additional loop, but they entail the quality of the product each time a cycle is performed, thus reducing progressively its market value. As long as the marginal price of performing the additional open loop cycle remains lower than the marginal revenue that such quality-level product can get from the market, then the additional open loop recycling procedure is justified. When, though, it happens that

the marginal open loop cycle cost becomes higher than the marginal revenue, the quality-entailing effect of the repeated recycling has brought the product's value to drop up to a critical low amount, meaning that there are no market opportunities left for such low-quality plastic product that justify the continuation of the open loop cycles. At this point the only option is to make the product go for chemical recycling process to recover substitute raw materials to be employed in a brand-new overall procedure.

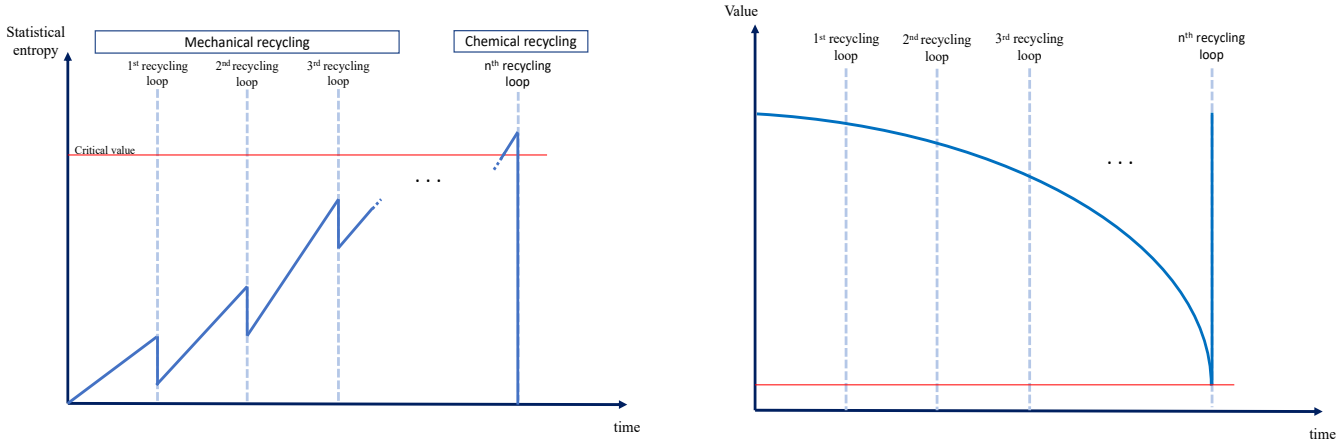

Figure 17 - graphical representation of the increase of entropy due to the repeated recycling (left); graphical representation of value decrease due to repeated recycling (right)

As it is possible to see from figure 17, along with the repetition of cycles, the entropy continuously raises while the quality value decreases. The value starts decreasing at a slower pace, symbolizing that the first recycling process/es are closed loop type, then its pace accelerates cycle after cycle because of the quality entailing characteristic of the lower degree of circularity recycle type. The high critical value of statistical entropy (graph on the left) is reflected by the low critical amount of product's value (graph on the right), its achievement comports the end of the cascade recycling procedure and the extraction of raw materials from chemical recycling as only feasible recycling option left.

The critical value represents the decoupling point beyond which going for chemical recycling becomes more convenient than the mechanical recycling. The massive energy and capital

consumption of the chemical recycling process, together with its early state of advancement, prevent it from being the reference option and confine it to be the backup rebound for when the conditions for the mechanical recycling do not stand anymore.

In this context, SEA is able to compute and estimate the recyclability level of the products through their lifetime and, therefore, also to predict the degree of circularity of the measures that should be put in place case by case in order to foster the transition toward the CE.

8.1 Limitations of the method

SEA methodology is not exempt from limitations, despite its strong contribution in promoting CE applications there are some critical issues that increase the complexity of its use. The first major complication is reflected by the massive amount of data needed to perform the calculations. Once the typology of plastic waste to be analyzed is chosen and the amount (in tons) obtained, in order to set the stage for a multilevel implementation, SEA requires information about: the different components (plastic items) present and their respective proportions over the total; the distinct materials that concur in the components' structure; the weights of each single material (in grams); the substances representing the chemical compounds of the selected plastics with their proportion inside each material; the likely contamination levels present inside the chemical compounds and finally the decomposition energies of each component with their maximum values. While some of those data are easily accessible and publicly available, a consistent part of them is barely possible, if not totally impossible, to obtain. Despite rigid municipality directives, separate collection of plastic waste is a task borne by the citizens at the domestic level, so, it is likely to expect within the wastes the presence of items that were not supposed to be gathered through plastic's collection programs. Whereby, to identify the exact composition of plastics items inside the regional waste streams is a rather difficult operation. So does also the process of obtaining information about the contamination of each

material's plastic structure, neither literature nor secondary sources are able to provide exactly the identity and the percentages of contaminants inside each plastic structure, for the simple primary reason that it differs from object to object. A utopic scanning system able to thoroughly describe each single item's chemical structure one by one would be needed and, even considering that such activity was technically feasible, it would require an excessive amount of time and resources. These considerations altogether raise the level of computational complexity of the SEA method.

The second main limitation of the model is represented by the fact that, in order to set the guidelines for the whole recyclability journey of wastes, in terms of suggesting which recycling technology to choose for each loop, a new development of the method is required to update the information every time a new material cycle occurs. For this reason, although the method is utterly useful for this implication, its implementation is associated with a sensible level of complexity that thwarts a generalized adoption.

9 Conclusion

This conclusion chapter summarizes the key research findings in relation to the research aim and research questions, as well as the value, contributions and implications thereof. It also reviews the limitations of the study and proposes opportunities for future research.

The research explored the implementation of the Statistical Entropy Analysis as a tool for predicting the degree of circularity of downstream interventions allowing to recover the maximum value possible from plastic wastes in the Italian Lombardy region scenario, according to the calculation of the relative entropy, which provides a picture of their chemical structure's complexity. Leaning on the SEA's relative entropies, the method allows for the computation of a recyclability indicator for each plastic item considered, that has been used as a metric to understand which was the right recyclability technology to select, that best fitted each specific case. According to the results obtained in this dissertation, we found a generalized low value for the plastic waste's entropies, that, in turn, means high values of the items' recyclability. It brought to the conclusion that mechanical recycling technologies appear to be the most employed to recover value from plastic waste. However, a sensitivity analysis applied to the results has put the light on the fact that a modification of the complexity of the plastic items' structure, likely due to the repeated reprocessing of the plastics, could comport a shift from mechanical to chemical recycling technologies as the preferable route to recover mass and value when mechanical recycling is no longer a viable route.

It is its very capability to straightforwardly tell whether a recycling technology is better than another, based on the numeric recyclability indicator that SEA gives in output when it is fed with plastic waste's data, represents the main possible implication from a managerial perspective. In this sense, the method should be employed to plan and design in advance the recyclability interventions required case by case, regarding the specific plastic waste gathered. In addition, a possible coupling of the SEA's indicators with market data about the recycled products' value is investigated, assuming that it could be useful to immediately understand whether there is economic viability to perform one

recycling technology or it is better to consider a different one according to the consultation of the marginal cost of recycling.

Despite the theorized implications and further applications of such CE assessing method, its development throughout the dissertation work has been characterized by some assumptions that unavoidably affected its overall reliability. The main limitation of our study is represented by the fact that only one recycling loop (the first recycling loop) has been considered in the calculations. This comported to consider, as much as possible, an almost uncontaminated structure of the plastic items shaping the wastes, given that they are supposed to have become waste for the first time, thus without being contaminated by previous reprocessing. The necessity to set such assumption stems from the difficulty we would have encountered if trying to enrich the analysis with also information about previous contamination in the structure and about future development of plastic structure's modification. Consequently, this had an impact on the whole reliability of the analysis, since the interpretation of the recyclability indicator had to neglect the real impact of a strong dimension such as the contamination, which instead has been introduced just as an assumed value. Other limitations of the study are reflected by a number of assumptions about the input data. SEA method itself brings the complexity factor of requiring information that is often impossible to obtain from literature or online databases.

To conclude, SEA methodology has proved to be a very promising tool to be considered when it comes to assess the circularity of CE actions, as it investigates circularity in an unprecedented way by considering the thermodynamics of recycling, and it links this dimension with a thorough method to choose the recycling technology to utilize. This paves the way for a new perspective when analyzing the CE, that could be coupled with other methodologies (like LCA or MFA) to provide a more comprehensive investigation of CE solutions. Furthermore, its well-fitting job in dealing with plastic waste opens up the possibility to use the SEA method also for other material streams (e.g., other recyclables) and therefore other value chains.

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APPENDIX I - *List of Dimensions*

i	Substance flow
f	Material flow
n	Component flow
p	Product
M_f	Material flow rate
$c_{i,f}$	Mass fraction of *i* inside *f*
$m_{i,f}$	Standardized mass fraction of *i* inside *f*
$X_{i,f}$	Substance flow rate of *i* inside *f*
H^i	Statistical entropy of substance *i*
H^i_{rel}	Relative statistical entropy of substance *i*
H^i_{max}	Maximum statistical entropy of substance *i*
H^c_n	Statistical entropy of component *n*
$H^c_{n,rel}$	Relative statistical entropy of component *n*
H^c_{max}	Maximum statistical entropy of components
H^p	Statistical entropy of product *p*
$c_{n,p}$	Concentration of the component *n* inside *p*
q_n	Ratio of the number of units of component *n*
N_{tot}	Total number of components present in the system
H^p_{rel}	Relative statistical entropy of product *p*

H_{max}^{p} Maximum statistical entropy of products

E_{n}^{c} Energy to decompose a component into substances

E_{n}^{c} Energy to decompose a component into materials

E^{p} Energy to decompose a product into components

E_{max} Maximum decomposition energy

$E_{n,rel}^{c}$ Relative decomposition energy of a component into substances

E_{rel}^{p} Relative decomposition energy of a product into components

$R_{n}^{(1)}$ Recyclability indicator 1 of components

$R_{p}^{(1)}$ Recyclability indicator 1 of a product

$R_{n}^{(2)}$ Recyclability indicator 2 of components

$R_{p}^{(2)}$ Recyclability indicator 2 of a product

APPENDIX II - *List of Equations*

Number	Equation
1	$X_{i,f} = M_f c_{i,f}$
2	$m_{i,f} = \frac{M_f}{\sum_{f=1}^{F} X_{i,f}}$
3	$H^i(c_{i,f}, m_{i,f}) = -\sum_{f=1}^{F} m_{i,f} c_{i,f} \log_2(c_{i,f})$
4	$H^i_{rel}(c_{i,f}, m_{i,f}) = \frac{H^i(c_{i,f}, m_{i,f})}{H^i_{max}}$
5	$H^i_{max} = (\sum_{f=1}^{F} m_{i,f})$
6	$H^c_n(c_{i,n}, m^c_n) = -\sum_{i=1}^{I} m^c_n c_{i,n} \log_2(c_{i,n})$
7	$H^c_{n,rel}(c_{i,n}, m^c_n) = \frac{H^c_n(c_{i,n}, m^c_n)}{H^c_{max}}$

8	$$H^c_{max} = -\sum_{i=1}^{I} c_{i,tot} \log_2(c_{i,tot})$$
9	$$H^p\left(c_{n,p}, H^c_{n,rel}\left(c_{i,n}, m^c_n\right)\right) = -\sum_{n=1}^{N} \log_2(c_{n,p})\, H^c_{n,rel}\left(c_{i,n}, m^c_n\right)$$
10	$$c_{n,p} = \frac{q_n}{N_{tot}}$$
11	$$H^p_{rel}\left(c_{n,p}, H^c_{n,rel}\left(c_{i,n}, m^c_n\right)\right) = \frac{H^p\left(c_{n,p}, H^c_{n,rel}\left(c_{i,n}, m^c_n\right)\right)}{H^p_{max}}$$
12	$$H^p_{max} = \log_2(N_{tot})$$
13	$$E_{max} = \frac{1}{M_{tot}} \sum_{n=1}^{N} \sum_{i=1}^{I} q_n c_{i,n} M_n e_i^{production}$$
14	$$E^c_{n,rel}(\pi, \eta^c_m, \eta^c_i) = \frac{E^c_{m,i}(\pi, \eta^c_m, \eta^c_i)}{E_{max}}$$
15	$$E^p_{rel}(\pi, \eta^p_n, \eta^p_m, \eta^p_i) = \frac{E^p_{n,m,i}(\pi, \eta^p_n, \eta^p_m, \eta^p_i)}{E_{max}}$$

16	$R_n^{(1)} = \left(1 - H_{n,rel}^c\left(c_{i,n}, m_n^c\right)\right)\left(1 - E_{n,rel}^c(\pi, \eta_m^c, \eta_i^c)\right)$
17	$R_p^{(1)} = (1 - H_{rel}^p\left(c_{n,p}, H_{n,rel}^c\left(c_{i,n}, m_n^c\right)\right))(1 - E_{rel}^p(\pi, \eta_n^p, \eta_m^p, \eta_i^p))$
18	$R_n^{(2)} = \frac{\left(1 - H_{n,rel}^c\left(c_{i,n}, m_n^c\right)\right)}{\left(1 - E_{n,rel}^c(\pi, \eta_m^c, \eta_i^c)\right)}$
19	$R_p^{(2)} = \frac{(1 - H_{rel}^p\left(c_{n,p}, H_{n,rel}^c\left(c_{i,n}, m_n^c\right)\right))}{(1 - E_{rel}^p(\pi, \eta_n^p, \eta_m^p, \eta_i^p))}$
20	$allocation\ quota = \frac{Lombardy\ 2019\ population}{EU\ 2019\ population}$
21	$c_{\ PET,drinking\ bottle} = \frac{PET_{\%}^{material\ 1} * m_{drinking\ bottle}^{material\ 1}[g] + \cdots + PET_{\%}^{material\ 7} * m_{drinking\ bottle}^{material\ 7}[g]}{component's\ mass\ [g]}$
22	$c_{PET,tot} = \frac{PET_{\%}^{material\ 1} * M^{material\ 1}[tons] + \ \ldots\ + PET_{\%}^{material\ 7} * M^{material\ 7}[tons]}{total\ system's\ mass\ [tons]}$
23	$M^{material\ 1} = m_{drinking\ bottle}^{material\ 1}[g] * \#drinking\ bottle + \cdots + m_{cups}^{material\ 1}[g] * \#cups$

24	$E_{max} = 77{,}533\ MJ/kg$
25	$E_{n,rel}^{c} = \frac{E_{n}^{c}}{E_{max}} = 0{,}01586$