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Upcycling of printed plastic films: LCA analysis and effects on the Circular Economy

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Abstract

In this work, the environmental impacts caused by an innovative upcycling process of printed plastic scrap have been assessed through Life Cycle Analysis (LCA) methodology for the first time. The process consists of removing the inks from the plastic surface before extrusion, so that clear high quality pellets are obtained, suitable to be used in high added value applications (such as packaging). The upcycling technology is compared with two traditional waste treatments: conventional recycling (or downcycling) and incineration with energy recovery. Upcycling is considered to be better aligned with Circular Economy principles and its implementation in the industry requires a comprehensive analysis of environmental impacts. Despite the importance of this topic, only a few studies can be found in the literature. Furthermore, the lack of uniformity and consensus in LCA modelling can lead to the conclusion that upcycling causes the biggest environmental burdens. Therefore, downcycling or incineration are shown as preferable options, regardless of the irreversible loss of the plastics' potential to be recirculated. To avoid this error, we have emphasised the importance of including the market share for recycled products in the LCA modelling and establishing the virgin plastic substitution ratio correctly. Also, we have suggested that in the perspective of the Circular Economy, the energy produced during incineration cannot substitute the energy from fossil fuels.

Keywords

Circular economy, upcycling, LCA, plastic, waste treatment.

1. Introduction

mechanical recycling (31.1%), energy recovery (41.6%) and landfilling (27.3%) are the main waste treatment options (Plastics Europe, 2018). Nevertheless, adopted waste management strategies vary among countries, even among municipalities within the same country. For instance, Switzerland, Austria or Germany have implemented landfill restrictions aiming at prioritising recycling and energy recovery. On the contrary, other countries like Bulgaria, Greece or Malta still dump more than 70% of their waste in landfill sites. This is to say that there is yet no consensus on the waste management best practises, in part, because appropriateness and feasibility of the PSW treatments strongly depend on the waste characteristics (e.g. source, homogeneity, contamination). Therefore, although plastic waste management has been assessed in a large number of previous studies, there is still much room for discussion, especially considering that the plastic sector is expected to change following the Circular Economy principles.

The Circular Economy model, which implementation in companies, regions and countries is growing, defends that the plastic waste is a valuable resource with the potential to be recirculated in a new material cycle (Webster, 2017). It is considered that the potential of plastic products to be recirculated should be exploited. In this context, two types of recycling processes can be mentioned: Downcycling and Upcycling (Figure 1). Downcycling is a process of plastic waste recovery which results in a reduction in quality of the material (inferior physical properties, dark colours, disturbing odour, etc.). Plastic degradation leads to a reduction in circularity potential, i.e., the ability to recover the material in a closed-loop (Eriksen et al., 2019). Thus, recovered materials are intended for low added value applications (e.g., trash bags, pipelines, agricultural buckets, etc.). On the contrary, during upcycling, the quality is improved so that the material is suitable to be used in the same application as the original product (Sung, 2015). Therefore, upcycling of plastic waste must prevail over other treatment options in order to preserve the quality of the material and ensure the maximum number of material cycles. Moreover, upcycled pellets are suitable to be used in more demanding applications. This might satisfy the growing interest of the plastic sector in recycled materials. Plastic producers are more and more committed to green marketing, which is in turn
induced by social pressure. For instance, some laundry and home care products manufacturers have committed to using a certain per cent of recycled plastic of proper quality in their packages (Henkel, 2018).

Many research works have been focused on upcycling plastic waste into carbon nanomaterials. (Zhuo and Levendis, 2014) conducted an extensive literature review on this topic reporting that several value-added products such as carbon fibres, fullerenes, carbon nanotubes and graphene can be produced from plastic waste. For instance, (Gong et al., 2014) developed a novel method to produce graphene flakes using waste polypropylene (PP). (Choi et al., 2018) studied the fabrication of transparent conducting films derived from polyethylene (PE) thin films transformed into carbon nanosheets. (Aji et al., 2018) transformed PP plastic waste into photoluminescence polymer carbon dots with potential to be used for photocatalyst, bioimaging and as sensors in optoelectronic materials. In general, polymer breaking down processes such as carbonization or pyrolysis are intended for the treatment of the post-consumer waste fraction that cannot be recycled mechanically due to the presence of incompatible polymers, a high degree of degradation, organic contamination, etc. The vast majority of these processes have been developed only at laboratory scale, so that their environmental impacts cannot be fully evaluated. Only a few studies on plastic waste upcycling through mechanical processes can be found. (Ragaert et al., 2018) studied the
upcycling of post-industrial PP contaminated with polyethylene terephthalate (PET) using a compatibilization method (i.e. adding impact modification additives) for a high-impact consumer product manufacturing. In a recent study, Ragaert et al. also presented a new process for the upcycling of the currently non-recyclable fraction of post-consumer plastic waste. The process consists of eliminating problematic contaminants such as non-ferrous particles and polyvinyl chloride (PVC) and producing new products through injection moulding (Ragaert et al., 2020). Despite the potential of this process for the materials recovery, the environmental impacts were not reported.

Post-industrial waste compared with domestic waste has a bigger potential for upcycling since it is generally clean and homogeneous. Therefore, it can largely contribute to the implementation of the Circular Economy model. In this paper, an innovative upcycling process for the recovery of printed plastic films from post-industrial source has been presented. Unlike the recycling methods described above, this process has been validated at industrial scale.

Plastic film converting companies generate between 8-12% of printed scrap during their production processes. This waste must be managed properly to ensure the sustainable development of the plastic sector. Flexible plastics present some technical issues during recycling that reduce the recycling rates (e.g. low bulk density, multilayer structures). In addition, the majority of plastic films have been printed on the surface. The presence of inks worsens the quality of recycled pellets since they volatilize during extrusion due to high temperature, increasing the chance for defects to occur.

Additional technologies, such as venting or degassing, are required for the reprocessing of printed films. Moreover, the recycled product usually has a dark colour which is less attractive for the consumer. For this reason, conventional recycling methods based on re-extrusion of plastic waste with the inks are considered downcycling processes. The upcycling of printed scrap can be achieved through the implementation of deinking technologies intended to remove the ink from the plastic surfaces. Different deinking processes available on the market have been described in a previous study (Horodytska et al., 2018). In this work, the focus has been put on the processes where detergents or surfactants are used (Fullana and Lozano, 2015). The other methods are based on the
use of solvents, which is less convenient from both economic and environmental point of view.

Reprocessing of plastics without the ink maintains the quality of the original material. Hence, the recycled product can be used to produce high added value products. Despite the potential of post-industrial waste to be recirculated, it is frequently sent to incineration for electricity and heat production due to its high calorific value (Zevenhoven et al., 1997, Sahlin et al., 2007).

The selection of the best waste treatment option is a complex task and requires a deep understanding of all processes. Among the different waste treatment methods, upcycling is better aligned with the circular economy principles. Nevertheless, the benefits of upcycling are occasionally not evident since more complex and resources consuming operations are required. As a consequence, downcycling methods are frequently implemented for post-industrial waste treatment owing to their lower complexity and costs, regardless of the irreversible and meaningful loss of quality (Singh et al., 2017). Therefore, the environmental impacts associated with the different waste treatment options must be thoroughly studied for the appropriate decision-making. Life Cycle Assessment (LCA) methodology has been widely used for comparison of environmental impacts of plastic waste management scenarios (Perugini et al., 2005, Lazarevic et al., 2010, Merrild et al., 2012). However, the results are sometimes questioned since they are strongly influenced by the assumptions made during the analysis and the quality of the data used (Ayres, 1995). The majority of studies put the focus on mixed waste (mainly hard plastics) from domestic sources (Bovea et al., 2010, Song et al., 2013, Fernández-Nava et al., 2014, Erses Yay, 2015). Gu et al. have assessed mechanical recycling of several plastic materials made of PE including film scraps, agricultural films and shopping bags from an environmental point of view. The environmental impacts of different process stages, such as washing, sorting, shredding, extrusion and re-granulation were assessed. The results showed that extrusion has the largest impact on the environment (Gu et al., 2017). In a study published by Hou et al., several waste management options for plastic films from post-consumer sources have been compared. Unlike post-industrial scrap, the sorting of domestic waste films is a challenging task and must be included in the computation (Hou et al., 2018). Huysman et al. studied
the recycling of post-industrial plastic film scrap generated before converting steps such as printing. Therefore, the plastic waste was not contaminated with inks. The authors considered the quality of the recovered material by analysing the compatibility between polymers (Huysman et al., 2017).

In this work, for the first time, the environmental impacts associated with the upcycling process of the printed plastic films from post-industrial source have been assessed and compared with the traditional waste management options such as re-extrusion (downcycling) and incineration with energy recovery. The influence of assumptions made in LCA has been evaluated, and two modifications have been suggested to include when plastic waste management options are assessed. In addition, the system boundaries have been extended to the end-of-life stage of the secondary plastics produced with the upcycled post-industrial waste. The environmental impacts of two material cycles were computed considering the current post-consumer waste management scenario and then compared with two possible scenarios. The results show the direction in which the waste management strategy should be developed and the real potential of current plastic waste management options to fulfil the requirements of the circular economy model.

2. Materials and methods

The LCA analysis has been performed following the ISO 14040:2006 Standard (ISO-Norm, 2006). The LCA software Quantis Suite 2.0 and Ecoinvent 2.2 has been used for computing the impacts of the studied processes.

2.1. Goal and scope definition

The goal of this study is to compare the environmental impacts of three waste management options: upcycling process, downcycling and incineration with energy recovery. It is a 'gate to grave' approach focusing on the end-of-life of printed plastic scrap from converting industry, including waste treatment operations. This study does not consider the whole life cycle of the product as in a ‘cradle to grave’ approach. The upstream life cycle stages of plastic products (production phases) are not
included since a) plastic scrap is considered as a waste, b) they are similar between the compared scenarios and would not provide any insights for the analysis.

In all scenarios, the following aspects have been considered: (a) the manufacture of the auxiliary inputs of the recycling process, e.g. deinking reagents, (b) the operation of the plant, (c) the management of the remaining waste from the recycling process, (d) the use phase of the outputs. To get the most comprehensive perspective, the production chain (when it can be identified) of each direct flow has been considered in the computation. The functional unit was defined as the treatment of 1000 kg of post-industrial printed plastic waste.

2.2. Life cycle inventory analysis (LCI)

The main assumptions and data used to quantify the potential impacts of the inputs and outputs of each scenario are described here. A converting company transforms the input material (plastic pellets, additives, inks, etc.) into new products through different processes. For instance, the fabrication of plastic shopping bags encompasses processes such as blown film extrusion, printing and bag-making. It is precisely during printing and bag-making steps that the printed scrap is generated. In this study, the waste management of a medium-size converting company from the Valencian Community (Spain) has been assessed. This company produces mainly polyethylene flexible packaging for personal and home care products. Around 8% of their annual production become printed plastic waste that the company sends to conventional recycling facilities. The recycled pellets are used to replace virgin plastics in new products manufacturing. The virgin plastic substitution rate is defined as the amount of recycled plastic that can substitute virgin resins over the total amount of plastic necessary for the manufacturing of a product.

2.2.1. Description of scenarios and data inventory

The study focuses on three waste management scenarios.

Scenario 1: Downcycling
Post-industrial plastic waste is usually managed separately from post-consumer streams to avoid contamination. Since the material is homogeneous and its origin is well known, no sorting or separating technologies are required (just some manual sorting). Also, washing is not necessary because the input waste is clean enough for existing technologies. Therefore, the printed waste after shredding is directly sent to extrusion (Figure 2). Extrusion machines must be properly conditioned to process heavily printed material. For instance, ultrafine filtration, homogenization and degassing stages are required to ensure the highest quality of the recycled pellets (EREMA, 2016).

Virgin plastic substitution rate depends on the final application. For example, ordinary garbage bags can contain up to 100% of recycled material. However, when some specific requirement must be met (strength or impermeability) then the substitution rate decreases. In this study, 80% substitution rate has been considered, representing a broad range of possible applications. The energy consumption is limited to the extrusion equipment and it is around 750 kWh. European electricity mix has been considered to determine the burdens associated with energy production. The recycling efficiency varies between 90-97%. The highest 97% was established for the study.

Figure 2. Modelled waste treatment scenario and overview of system flows: Scenario 1: Downcycling of plastic waste. Source: own elaboration (made with Edraw Max).

Scenario 2: Upcycling (Recycling with deinking)
Upcycling of plastic scrap is a recycling process with a deinking stage. This is an innovative technology that removes the ink from plastics surface before extrusion. As a result, mechanical and thermal properties of the recycled plastics are similar to the original material owing to minimal degradation during reprocessing. Also, the aesthetical properties are improved since the material has a clear white colour or it is transparent. The ink is removed during a washing stage where some washing agents (detergents) in water-based solution and temperature are used. The mechanical operations included in this scenario are shredding, washing with deinking, drying through centrifugation, extrusion and wastewater treatment.

The data related to the deinking plant were provided by Cadel Deinking Company which has developed this innovative technology. The production capacity of this plant is 500 kg/h. The electricity consumption of the deinking process (including shredding, washing, water heating, drying and wastewater treatment) is around 600 kWh per tonne of input plastic. An average energy need for the extrusion machine is 750 kWh per tonne of input plastic. European electricity mix is also considered in this scenario. As mentioned before, non-hazardous reagents are used for deinking. Also, during the wastewater treatment, specific chemicals are added. In total, the process requires 46 kg of reagents per tonne of input plastic. Despite the recirculation of treated water, it is necessary to add a small quantity of tap water to cover the losses originated during the process (1200 L per tonne of input plastic). The secondary outputs of the plant are wastewater not collected for treatment (44 kg per tonne of input plastic) and an aqueous sludge containing inks (90 kg per tonne of input plastic). The wastewater is purified in a medium-size municipal wastewater treatment plant. And the sludge is managed through municipal incineration as non-hazardous waste. The recycling efficiency is about 97%. The remaining part is collected and sent to landfill (Figure 3).

In this case, the converting company uses the recycled pellets for high added value products manufacturing. The quality requirements are higher and, therefore, the recycled content should be
lower than in the downcycling scenario. In this case, the selected virgin plastic substitution rate is 20%.

Figure 3. Modeled waste treatment scenario and overview of system flows: Scenario 2: Upcycling of plastic waste. Source: own elaboration (made with Edraw Max).

Scenario 3: Incineration

Flexible plastic waste is usually sent to incineration plants along with the municipal solid waste stream. This operation is well described in the Ecoinvent 2.2 database (disposal, polyethylene, 0.4% water, to municipal incineration [kg]), which contain the data required for the LCA analysis. The calorific value of plastic films is reported to be 41.41 MJ/kg (Asamany et al., 2017). And the energy obtained is used to produce electricity (21% of efficiency) and heat (74% of efficiency) (Merrild et al., 2012).

The most relevant LCI data have been summarized in Table 1.
Table 1. LCI data for the three studied scenarios.

<table>
<thead>
<tr>
<th></th>
<th>Downcycling</th>
<th>Upcycling</th>
<th>Incineration</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Energy (kWh/tonne plastic)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Deinking</td>
<td>-</td>
<td>600</td>
<td>-</td>
</tr>
<tr>
<td>Extrusion</td>
<td>750</td>
<td>750</td>
<td>-</td>
</tr>
<tr>
<td><strong>Reagents (kg/tonne plastic)</strong></td>
<td>-</td>
<td>46</td>
<td>-</td>
</tr>
<tr>
<td>Water (L/tonne plastic)</td>
<td>-</td>
<td>1200</td>
<td>-</td>
</tr>
<tr>
<td><strong>Residues (kg/tonne plastic)</strong></td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Wastewater</td>
<td>-</td>
<td>44</td>
<td>-</td>
</tr>
<tr>
<td>Sludge</td>
<td>-</td>
<td>90</td>
<td>-</td>
</tr>
<tr>
<td><strong>Substitution ratio (%)</strong></td>
<td>80</td>
<td>20</td>
<td>-</td>
</tr>
<tr>
<td>Process efficiency (%)</td>
<td>97</td>
<td>97</td>
<td>-</td>
</tr>
<tr>
<td><strong>Electricity</strong></td>
<td>-</td>
<td>-</td>
<td>21</td>
</tr>
<tr>
<td>Heat</td>
<td>-</td>
<td>-</td>
<td>74</td>
</tr>
<tr>
<td><strong>Calorific value (MJ/kg)</strong></td>
<td>-</td>
<td>-</td>
<td>41.41</td>
</tr>
</tbody>
</table>

2.2.2. Modelling of the end-of-life stage and allocation rules for co-products

The system can generate several products, called co-products. In this situation, the environmental impact of a specific co-product is thus only a part of the impacts of the whole system. Several approaches do exist to compute the impact of a specific product in a multi-products system. In this study, the methodology called 'extension of boundaries' has been adopted to consider all the impacts avoided thanks to the production of the co-products.

The conventional recycling by means of re-extrusion produces dark colour pellets for non-demanding applications (e.g., trash bags, pipelines, pots). Meanwhile, the innovative deinking technology makes possible the production of high-quality pellets suitable for the same application as the original
Both processes make virgin plastic production decrease. The difference is the value of the recycled product. Finally, the energy obtained during incineration replaces the European energy mix.

2.3. Life cycle impact assessment (LCIA)

LCIA methodology IMPACT 2002+ vQ2.2 (version adapted by Quantis) has been used to connect the LCI results to the corresponding environmental impacts (Humbert et al., 2012). This methodology is a combination of the classical impact assessment and the damage oriented methodologies. The evaluation of the potential environmental impacts is performed in two steps. First, the elementary flows identified during the LCI analysis are associated with a number of impact categories at midpoint level. Some midpoint categories are human toxicity, aquatic ecotoxicity, aquatic eutrophication, global warming, non-renewable energy, etc. The impact on each category is obtained through a characterization factor expressed in kg-equivalents of a studied substance compared to a reference substance. Secondly, all the midpoint categories are grouped into four damage categories (the end-point in the cause-effect chain).

The environmental burdens of the recycling processes have been calculated as a difference between the impacts associated with the recycling operations and the avoided impacts associated with the production of virgin plastic. The avoided consumption of virgin plastics at a converting company depends on the recycling efficiency and the substitution ratio ($S$). The impacts of the incineration treatment have been calculated using the municipal waste incineration data and subtracting the avoided impacts of using the European energy mix from renewable and non-renewable sources for electricity production and natural gas, burned in cogeneration, for district heating. Fossil fuels are still the major source of energy representing more than 70% of the gross inland consumption (Directorate-General for Energy, European Commission, 2018).

2.3.1. Impact categories

The impacts on four categories have been evaluated and compared in this work. These are Human Health, Ecosystem Quality, Climate Change and Resource conservation. All the categories are
environmentally relevant and included in the ISO 14040:2006 (ISO-Norm, 2006). Different units are used to express the impact in selected categories. Disability-Adjusted Life Years (DALY) is used in the human health category and represents the disease severity, considering both mortality and morbidity. In other words, the number of DALYs represents the number of years of life lost over the overall population (not per person). The midpoint categories included in the computation are human toxicity, respiratory effects, ionizing radiation, ozone layer depletion and photochemical oxidation. The ecosystem quality is expressed in Potentially Disappeared Fraction of species over a certain amount of square metre during a certain amount of year (PDF·m$^2$·y). It is the sum of aquatic ecotoxicity, terrestrial ecotoxicity, terrestrial acidification/nutrification, land occupation, and, aquatic acidification, aquatic eutrophication and water turbined. The unit used in the climate change category is kg equivalent of carbon dioxide, which is used as a reference substance. The midpoint category used is the global warming potential. Finally, in the resources category, MJ is used to express the energy extracted or needed to extract the resource. Two midpoint categories are considered here: non-renewable energy consumption and mineral extraction. Normalization and weighting of the impacts were not performed in this study.

2.3.2. Sensitivity analysis and model simulations

Certain parameters described in data inventory section were assumed for this study but may change from application to application. The most important assumptions made are the virgin plastic substitution rate for the recycling scenarios, the market share of recycled pellets and the substituted material for energy production for the incineration scenario. Several simulations were carried out to determine the influence of these parameters on the LCA results.

3. Results

3.1. LCA of the studied waste management options.

The results are shown as positive and negative potentials (positive and negative values in the graphs). A positive potential indicates a burden to the environment (negative environmental impact...
or effect), while a negative potential indicates environmental savings (positive environmental impact or effect). At a converting company level, the upcycling process appears to be the worst waste management option for all impact categories assessed. It has a negative environmental impact on human health, ecosystem quality and climate change. It shows some benefits regarding resource conservation. However, the savings are around 30 times lower than for downcycling. This can be attributed to the difference in substitution ratio admissible for each application. The recycled material from the downcycling process substitutes a higher amount of virgin plastic due to more forgiving applications of the product. Therefore, it produces a positive effect on resource conservation, climate change and human health (Figure 4). The impacts of the upcycling process do not reach the level of the downcycling even if the substitution rate increases (Figure 5). The upcycling produces a positive environmental effect on climate change when the substitution ratio is higher than 0.4. Also, there is a change from negative to positive effect in the human health category when the substitution is more than 60%. Nonetheless, for the same substitution (80%) downcycling seems to be more environmentally beneficial than upcycling.

This approach also shows the benefits of plastic waste incineration over recycling processes. The high heating value of polymeric materials makes them attractive for electricity or heating production avoiding the use of such environmental pollutants as fossil fuels. Therefore, it generates a positive effect on resource conservation and climate change. Also, the absence of operations which consume energy and materials combined with efficient treatment of gas to remove air pollutants contributes to increasing the positive effect on human health and ecosystem quality. However, incineration is contrary to the circular economy principles and the quality and potential of plastic waste is not considered.

If the LCA analysis is based on the virgin plastic substitution rate, important parameters such as the quality of the recycled pellets and the intended applications are not considered. As a result, the recycling option in which poor quality pellets suitable only for low demanding (usually single-use)
products are obtained seems to be more environmentally beneficial than the recycling process that produces high quality recycled resins. The acceptable recycled content is higher since the requirements of the non-demanding applications are lower. Thus, the avoided production of virgin plastic increases.

Regarding incineration scenario modelling, if it is assumed that the recovered energy substitutes the energy from fossil fuels, then recycling will surely be a less favourable option. This is because fossil-based plastics have a high content of feedstock energy (i.e. heating value) since polyolefins are mainly produced from hydrocarbon feedstocks diverted from energy production. Moreover, the energy requirements for virgin PE production are usually lower. For instance, (Vlachopoulos, 2009) estimated the process energy requirements for LDPE at 28 MJ/kg, which is around 1.5 times lower than the heat value of LDPE (the value used in this study: 41.41 MJ/kg). Therefore, the energy saved by combustion is usually higher than the energy saved by avoiding virgin granulates production. Thus, the scenario with fossil fuels substitution will surely be more beneficial. However, the energy for electricity or heat production can be obtained from sources different from fossil fuels. The circular economy strategy promotes the use of renewable energy which should predominate in the near future.

The assumptions made to obtain the results shown in Figure 4 and Figure 5 lead one to make decisions which go against the circular economy principles and the EU waste hierarchy. The quality of the recycled pellets and the target market should be considered. Also, fairer energy substitution criteria should be implemented.
Figure 4. Environmental impacts of each plastic scrap treatment scenario.

Figure 5. Environmental impacts of upcycling with different substitution rates compared with downcycling and incineration. At converting company level.
3.2. Modified LCA analysis according to the circular economy needs.

In this section, two simulations have been performed to consider the quality of the recycled material and the source of energy.

- Potential to substitute virgin plastics considering the target market

The first modification is related to the boundaries of the study. In section 3.1., the avoided burdens have been calculated based on the virgin plastic substitution within the limits of a converting company. To determine the global environmental impacts, the boundaries should be extended to the entire market of plastic products. In this way, the quality of the recycled pellets plays a pivotal role in determining the global virgin plastic substitution potential. This is due to different quality requirements that vary depending on the intended applications. Therefore, the total avoided production of raw plastics has been calculated as a product of the amount of waste, the efficiency of the recycling process, the substitution rate (S) and the market share (MS).

So far, recycling companies have earmarked the post-industrial plastic recyclates for low demanding applications because this one was the only market which has been accepting a recycled content in its products. However, the current trend in the plastic sector is to introduce recycled content in high quality applications so that the target market for recovered materials expands. To achieve this, innovative recycling technologies are needed to preserve the quality of the plastic material regarding properties, appearance, odour, etc.

European market of LDPE films can be divided into a number of sectors (Figure 6). Dark coloured conventional pellets are suitable for building and construction, automotive and other less demanding applications, which together represent 24% of the LDPE market. On the other hand, deinked pellets owing to their higher quality can be used for all applications from food packaging to electronics or building materials. So, its market share reaches 100%.
If the potential of the recycled material to substitute virgin plastic is calculated based on the entire market of a specific plastic, a different result is obtained (Error! Reference source not found. 7). The environmental savings of the downcycling process decrease: for climate change from 1266 kgCO$_2$ eq to 24 kgCO$_2$ eq and for resources from 54,700 MJ to 6615 MJ. The impact on human health changes from positive ($-5 \times 10^{-4}$ DALY) to negative environmental effect ($1.7 \times 10^{-4}$ DALY). Finally, the negative effect on ecosystem quality increases from 37 PDF.m$^2$.y to 55 PDF.m$^2$.y. Even so, the upcycling process with 20% of substitution appears to be less favourable than the downcycling. This scenario shows higher environmental burdens on climate change, ecosystem quality and human health. Also, the savings accomplished in resource conservation are lower. However, this perspective changes when the substitution ratio is higher than 40%. For instance, for climate change and resources categories, the use of 40% of recycled content produces a positive environmental effect (negative impact potential) and both values are above the levels of the downcycling process. Regarding ecosystem quality, the negative environmental effect decreases with the increase in the substitution ratio. Nevertheless, these values remain higher than in the downcycling scenario due to the use of chemical agents during the deinking operation. For human health category, 60% of recycled content is necessary to decrease the negative effects below the level of the downcycling. It is possible to produce savings to the environment if the substitution rate increases over 60%.
Therefore, the environmental benefits of the upcycling process are revealed when the avoided virgin plastic production is computed considering the global market. This approach takes account of the quality of the recycled plastic and the value of the products produced using recycling content. The additional effort that recycled materials upgrading requires is offset by the expansion of the target market. For instance, deinked clean pellets can be used for packaging manufacturing meanwhile conventional dark pellets are only suitable for less demanding applications. Moreover, if the more clean and homogeneous waste stream is diverted to new markets, then post-consumer plastic waste with lower quality can be introduced more easily for more forgiving applications.

Figure 7. Environmental impacts of upcycling and downcycling considering the market share (\(MS_{\text{upcycling}}=1; MS_{\text{downcycling}}=0.24\)). Influence of increasing substitution rates in the upcycling process included.

- **Potential to substitute fuels**

As mentioned in section 3.1., the electricity and heat must be produced from renewable sources according to circular economy principles. Therefore, in LCA analysis the energy obtained during plastic waste incineration should substitute energy from non-fossil sources. In this study, three...
examples of renewable energy have been assessed: biogas from agricultural plants, hydropower and solar energy.

This approach shows that regarding the climate change category, upcycling is the most beneficial scenario (Figure 8). Production of energy from renewable sources does not emit carbon dioxide to the environment. So, its substitution would be senseless. The effects on the other impact categories vary depending on the energy source. For instance, incineration with substitution of biogas and photovoltaic energy is more beneficial for natural resources conservation category. This can be explained with the fact that both sources require the use of extensive areas of land and, in the case of solar cells, exhaustible resources such as silica are also consumed. The use of land can also explain the positive effects of these energy sources substitution on ecosystem quality. Regarding substitution of hydropower energy, the environmental savings on this category are much higher probably due to the loss of aquatic habitat, harm to the fish population, deterioration of the landscape, etc. related to this source. Finally, three scenarios produce a negative effect on human health indicator: upcycling, incineration with hydropower energy and solar energy substitution. The burdens of the upcycling process are related to the use of electricity obtained from fossil fuels (European energy mix). And the savings due to hydropower and solar energy production are not enough to counter the emissions originated during plastic waste incineration. On the other hand, the production of energy in agricultural biogas plants affect negatively human health. This is possibly caused by the use of pesticides, fertilizers, etc., and also by the emissions from biogas combustion. As a result, its replacement by incineration produces a positive effect (Figure 8).
3.3. Upcycling of plastic waste and circular economy

The upcycling of post-industrial plastic waste brings savings to the environmental, especially on the impact categories such as climate change and resources conservation. In previous sections, the system boundaries have been drawn around the recycling plant. Nevertheless, the product’s higher quality leaves open the possibility for post-consumer plastic waste to be recycled after the use phase (Hahladakis and Iacovidou, 2018). Additional recycling cycles contribute to decreasing the virgin plastic production. Therefore, in this section, the system boundaries have been extended to the end-of-life stage of the products with recycled content. Landfilling has not been included in this study. So that the two post-consumer plastic waste treatment options are recycling and incineration (Figure 9). In the European countries where landfill restriction has been implemented, around 40% of plastic waste is recycled (recycling rate (R)) and 60% is incinerated (Plastics Europe, 2018). The energy produced during incineration is used to generate electricity and
The energy source replaced is biogas from agricultural plants. The post-consumer plastic waste is currently treated through downcycling processes since existing technologies are not prepared to remove all the contaminants and impurities. Hence, in the first or base scenario, the efficiency of the recycling process is 90%, the substitution rate is 80% and the market share is 24%. The substitution rate of virgin plastic by post-industrial recycled pellets (in the first material cycle) is 40%. The base scenario has been compared with two additional scenarios. In the second one, the recycling rate of post-consumer plastic waste has been increased to 80%. In the third one, the market share has been incremented to 100%. It was assumed that the upcycling process is necessary to increase the market share. So, the energy and resources consumption data for the post-consumer plastic waste recycling correspond to the deinking process.

This approach has been used to show the environmental effects of post-consumer plastic waste treatments (Figure 10). As has been discussed in section 3.2., incineration with renewable energy substitution increases the emissions of carbon dioxide. Therefore, the first scenario causes the biggest environmental burdens in the environment. The decrease of the incineration rate in scenario 2 results in negative net impact, which means saving to the environment. Although higher recycling rate increases the positive effect on the environment, the savings of post-consumer waste recycling are considerably higher (around 20 times) in scenario 3 when the target market expands. The same trend is observed on resources conservation category. The net impact is around 1.6 times higher in scenario 3 compared with scenario 1. These results show that it is good to recycle more. But it is more important to maintain the quality and the value of plastic products since upcycling of plastic waste produces the highest environmental savings.
Figure 9. Modelled scenarios with extended boundaries and overview of system flows for both post-industrial waste (PIW) and post-consumer waste (PCW) treatment. Source: own elaboration (made with Edraw Max).

To make possible the conditions established in scenario 3 (substitution rate and market share), post-consumer plastic waste recycling processes must be upgraded considerably. The quality of recycled post-consumer plastics is currently quite poor owing to the high level of contaminants, odours, non-intentionally added substances (NIAS), etc. Innovative decontamination (such as deodorization) technologies are needed to prepare the recycled content for more demanding applications.
Figure 10. Environmental impact on climate change and resources conservation of the upcycling process considering two material cycles. Three scenarios studied: current post-consumer waste management scenario (scenario 1) and simulated scenarios (scenarios 2 and 3). PCW (post-consumer waste). PIW (post-industrial waste).

4. Discussion

The computation of the environmental impacts associated with the recycling of plastic waste differs among published studies. Gu et al. studied the mechanical recycling of different plastic materials where the recycled pellets were used in high value and low value applications. The avoided virgin plastic production was calculated based only on the substitution ratio. The authors state that the substitution ratio decreases with the increasing value of the final product and attribute this to the inferior quality of the recyclates (Gu et al., 2017). Nevertheless, they do not mention that the quality requirements in high value applications are much stricter, which strongly influences the substitution ratio. Huysman et al. use the percentage of substituted virgin materials and the recycling rate in the computation of impacts produced during the recycling of post-industrial plastic waste. The authors compare two waste streams which undergo different recycling options and calculate a circularity indicator based on the polymer compatibility. Finally, they concluded that the recycling option where recovered plastics are used to produce low value garbage bags is more environmentally beneficial than the scenario where the recycled plastics are used in high added value applications (Huysman et al., 2017). This can be attributed to the difference in the substitution ratio (80% and 20%...
respectively) determined by the quality requirements of the final user. According to the results of the present paper, these conclusions could have changed if the authors had considered the market share of the intended applications. In a study presented by Hou et al., the recycling of plastic films from post-consumer waste (mixed and recyclable) has been assessed. The parameters used for the computations were the recycling rate, utilization rate (equal to the substitution ratio), composition of polymers in the film waste, and the mass fraction of films in the waste stream. The authors studied only one scenario with 66% of substitution ratio, but in the sensitivity analysis concluded that the results are strongly dependent on this parameter (Hou et al., 2018). In general, the lack of uniformity in the procedure adopted by different authors makes it difficult to compare and to determine the accuracy of the LCA results. In fact, Viau et al. recently published an article where 51 LCA studies on municipal waste management were reviewed to assess the discrepancies in the modelling of the raw materials substitution. Based on the study by Vadenbo et al. (2017), the authors state that the substitution potential must be calculated considering four parameters: the amount of potentially recoverable materials, the recycling efficiency, the substitution ratio, and the market response. The analysis showed that none of the revised articles mentions all four parameters. At the same time, 100% of selected articles take into account the substitution ratio. Nevertheless, 22% are not explicit and 65% were not justified by the authors of the studies. Therefore, there is a lack of rigour in LCA studies on municipal solid waste management (Viau et al., 2020). In the present paper, we have considered all four parameters and the substitution ratios have been established based on the information provided by a real plastic film converting company regarding quality requirements in different applications. Also, the importance of the market response parameter has been shown, especially when upcycling and downcycling processes are compared.

The incineration of plastic waste with energy recovery has been addressed in numerous studies and the results vary depending on the waste composition, the heating value, the electricity or heat production efficiency, and the source of substituted energy. Merrild et al. concluded that incineration of municipal waste can be more beneficial than recycling when the level of energy
recovery at the incineration plant and the plastic fraction within the waste stream is high (Merrild et al., 2012). On the contrary, Perugini et al. showed that plastic waste combustion is the less preferable option, which can be explained by the fact that the heating value used in this study is lower and the recovered energy is used only to produce electricity with an efficiency of 25% (Perugini et al., 2005). Similar results were obtained in a study by (Cossu et al., 2017), in which several scenarios of the treatment of the residues obtained during the selection process of plastic materials have been assessed. Both studies are based on Italian waste management scenarios. Therefore, it can be concluded that in Italy and other southern countries where the incineration plants are mainly used for the production of electricity (there is no need for district heating), the energy recovery scenarios produce burdens to the environment. Regarding the substitution of energy, in general, all the authors agree that only the displacement of coal-fired power produces environmental savings (for instance, (Hou et al., 2018). In this paper, we have built the most favourable incineration scenario considering a high energy recovery incineration plant with the production of both electricity and district heating. The composition of the waste stream is 100% polymeric with a high calorific value. In these conditions, incineration is more beneficial than recycling if the recovered energy substitutes the energy from fossil sources. Nevertheless, the substitution of energy from renewable resources is senseless, especially for the climate change category.

Regarding the upcycling of post-consumer plastic waste (section 3.3), no LCA studies covering both post-industrial and subsequent post-consumer waste treatment were found. Nevertheless, a few similar studies can be mentioned. For instance, Toniolo et al. compared the environmental impacts of using recycled plastics to produce potentially recyclable or non-recyclable products. The authors showed that assuring the recyclability of the final products produces the highest environmental savings, which is in line with the results obtained in this paper (Toniolo et al., 2013). Sevigné-Itoiz et al. conducted a comprehensive study of post-consumer plastic waste treatment in Spain. The authors studied the effects of increasing the amount of collected plastics sent to recycling and concluded that the environmental benefits could be significantly increased (Sevigné-Itoiz et al., 2015). In this paper,
we have also presented the benefits of increasing the recycling rate in addition to the benefits obtained from upgrading the quality of the recycled product.

5. Conclusions

Plastic waste upcycling is aligned with the circular economy objectives since the quality and the value of plastic products is maintained. It is considered real recycling since it makes possible to close the material loops. Downcycling processes are, in fact, closer to the Linear Economy model. The value of the products decreases significantly, so that the material has to be landfilled or incinerated after few cycles. Despite this, certain assumptions made in LCA analysis lead to a solution where upcycling apparently causes the highest environmental burdens. This happens when only the virgin plastic substitution ratio is considered in the recycling scenarios and when the energy produced during incineration replace the use of fossil fuels. Two modifications should be taken into account to obtain fairer results.

In the first place, the substitution rate restricts the avoided virgin plastic production to the product level. Nevertheless, the target market for recycled pellets should be included in the comparison among different recycling processes. The higher quality of the upcycled pellets makes them suitable for a broader range of applications, so that the avoided consumption of virgin plastic at the market level increases, and so does the environmental savings.

In the second place, the energy produced during incineration should replace energy from renewable sources instead of fossil fuels. The substitution of fossil fuels will surely provide more benefits than recycling. This can be attributed to the fact that the heating value of plastics is higher than the energy consumed during raw pellets production. This means that the energy recovered through incineration is likely to be higher than the energy saved by avoiding virgin plastic consumption. Moreover, according to circular economy principles, the energy has to come from renewable sources. Therefore, if our society is moving forward to this new model, fossil fuels should not be considered.
The use of upcycled pellets in new products manufacturing increases the value of these products, thus, make them recyclable. Accordingly, a bigger number of material cycles is possible. The influence of the recycling rate and the target market has been studied. The post-consumer waste recycling rate slightly influences the environmental benefits of the process. The major savings are produced when the target market for recycled plastics expands. For this to be possible, upcycling processes for post-consumer waste must be implemented. Accordingly, innovative decontamination technologies should be studied in future works to maintain the quality of the products and increase the number of possible applications. The new technologies need to be accompanied by a transparent and thorough LCA analysis considering all the relevant parameters and adapting the assumption made to the Circular Economy principles.

Acknowledgements

This work was supported with a Grant from the University of Alicante as part of the InnoUA Transfer of Knowledge programme (Industrial Doctorate). The author O.Horodytska kindly wishes to thank the University of Alicante for a pre-doctoral employee stays grant, which has been used to develop the research at École Polytechnique Fédérale de Lausanne (EPFL); and the ICT4SM group for their support during the research stay. Language help provided by Nathan Kavanagh from the University of Strathclyde is also gratefully acknowledged.

References


Choi, D., Yeo, J., Joh, H., Lee, S., 2018. Carbon Nanosheet from Polyethylene Thin Film as a Transparent Conducting Film: “Upcycling” of Waste to Organic Photovoltaics Application. ACS Sustainable Chemistry & Engineering. 9, 12463-12470. DOI: 10.1021/acssuschemeng.8b03066.


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• Removing the inks from plastic surfaces is beneficial for the environment.
• Target market for recycled pellets should be included in LCA analysis.
• Energy produced during incineration should not substitute fossil fuels combustion.
• Expansion of the target market causes the biggest environmental savings.
Declaration of interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.