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Title page

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

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2 **Abstract**

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

18 **Keywords**

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

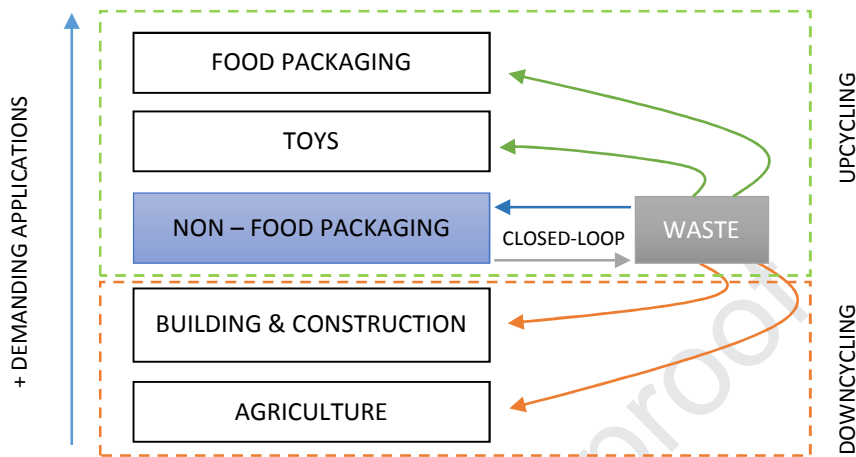
20 **1. Introduction**

21 Plastic solid waste (PSW) generation has become a global issue with 274 Mt released in 2010 (Geyer
22 et al., 2017). In an attempt to establish common rules for waste management, the European
23 Commission launched the Waste Framework Directive 2008/98/EC (Council Directive, 2008) based on
24 the following hierarchy: prevention, preparing for reuse, recycling, recovery and disposal. In Europe,

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

36 The Circular Economy model, which implementation in companies, regions and countries is growing,
37 defends that the plastic waste is a valuable resource with the potential to be recirculated in a new
38 material cycle (Webster, 2017). It is considered that the potential of plastic products to be
39 recirculated should be exploited. In this context, two types of recycling processes can be mentioned:
40 Downcycling and Upcycling (Figure 1). **Downcycling** is a process of plastic waste recovery which
41 results in a reduction in quality of the material (inferior physical properties, dark colours, disturbing
42 odour, etc.). Plastic degradation leads to a reduction in circularity potential, i.e., the ability to recover
43 the material in a closed-loop (Eriksen et al., 2019). Thus, recovered materials are intended for low
44 added value applications (e.g., trash bags, pipelines, agricultural buckets, etc.). On the contrary,
45 during **upcycling**, the quality is improved so that the material is suitable to be used in the same
46 application as the original product (Sung, 2015). Therefore, upcycling of plastic waste must prevail
47 over other treatment options in order to preserve the quality of the material and ensure the
48 maximum number of material cycles. Moreover, upcycled pellets are suitable to be used in more
49 demanding applications. This might satisfy the growing interest of the plastic sector in recycled
50 materials. Plastic producers are more and more committed to green marketing, which is in turn

51 induced by social pressure. For instance, some laundry and home care products manufacturers have
 52 committed to using a certain per cent of recycled plastic of proper quality in their packages (Henkel,
 53 2018).



54
 55 *Figure 1. Schematic representation of possible recycling options for post-industrial plastic waste. Source: own*
 56 *elaboration.*

57 Many research works have been focused on upcycling plastic waste into carbon nanomaterials. (Zhuo
 58 and Levendis, 2014) conducted an extensive literature review on this topic reporting that several
 59 value-added products such as carbon fibres, fullerenes, carbon nanotubes and graphene can be
 60 produced from plastic waste. For instance, (Gong et al., 2014) developed a novel method to produce
 61 graphene flakes using waste polypropylene (PP). (Choi et al., 2018) studied the fabrication of
 62 transparent conducting films derived from polyethylene (PE) thin films transformed into carbon
 63 nanosheets. (Aji et al., 2018) transformed PP plastic waste into photoluminescence polymer carbon
 64 dots with potential to be used for photocatalyst, bioimaging and as sensors in optoelectronic
 65 materials. In general, polymer breaking down processes such as carbonization or pyrolysis are
 66 intended for the treatment of the post-consumer waste fraction that cannot be recycled
 67 mechanically due to the presence of incompatible polymers, a high degree of degradation, organic
 68 contamination, etc. The vast majority of these processes have been developed only at laboratory
 69 scale, so that their environmental impacts cannot be fully evaluated. Only a few studies on plastic
 70 waste upcycling through mechanical processes can be found. (Ragaert et al., 2018) studied the

71 upcycling of post-industrial PP contaminated with polyethylene terephthalate (PET) using a
72 compatibilization method (i.e. adding impact modification additives) for a high-impact consumer
73 product manufacturing. In a recent study, Ragaert et al. also presented a new process for the
74 upcycling of the currently non-recyclable fraction of post-consumer plastic waste. The process
75 consists of eliminating problematic contaminants such as non-ferrous particles and polyvinyl chloride
76 (PVC) and producing new products through injection moulding (Ragaert et al., 2020). Despite the
77 potential of this process for the materials recovery, the environmental impacts were not reported.
78 Post-industrial waste compared with domestic waste has a bigger potential for upcycling since it is
79 generally clean and homogeneous. Therefore, it can largely contribute to the implementation of the
80 Circular Economy model. In this paper, an innovative upcycling process for the recovery of printed
81 plastic films from post-industrial source has been presented. Unlike the recycling methods described
82 above, this process has been validated at industrial scale.

83 Plastic film converting companies generate between 8-12% of printed scrap during their production
84 processes. This waste must be managed properly to ensure the sustainable development of the
85 plastic sector. Flexible plastics present some technical issues during recycling that reduce the
86 recycling rates (e.g. low bulk density, multilayer structures). In addition, the majority of plastic films
87 have been printed on the surface. The presence of inks worsens the quality of recycled pellets since
88 they volatilize during extrusion due to high temperature, increasing the chance for defects to occur.
89 Additional technologies, such as venting or degassing, are required for the reprocessing of printed
90 films. Moreover, the recycled product usually has a dark colour which is less attractive for the
91 consumer. For this reason, conventional recycling methods based on re-extrusion of plastic waste
92 with the inks are considered downcycling processes. The upcycling of printed scrap can be achieved
93 through the implementation of deinking technologies intended to remove the ink from the plastic
94 surfaces. Different deinking processes available on the market have been described in a previous
95 study (Horodytska et al., 2018). In this work, the focus has been put on the processes where
96 detergents or surfactants are used (Fullana and Lozano, 2015). The other methods are based on the

97 use of solvents, which is less convenient from both economic and environmental point of view.
98 Reprocessing of plastics without the ink maintains the quality of the original material. Hence, the
99 recycled product can be used to produce high added value products. Despite the potential of post-
100 industrial waste to be recirculated, it is frequently sent to incineration for electricity and heat
101 production due to its high calorific value (Zevenhoven et al., 1997, Sahlin et al., 2007).

102 The selection of the best waste treatment option is a complex task and requires a deep
103 understanding of all processes. Among the different waste treatment methods, upcycling is better
104 aligned with the circular economy principles. Nevertheless, the benefits of upcycling are occasionally
105 not evident since more complex and resources consuming operations are required. As a
106 consequence, downcycling methods are frequently implemented for post-industrial waste treatment
107 owing to their lower complexity and costs, regardless of the irreversible and meaningful loss of
108 quality (Singh et al., 2017). Therefore, the environmental impacts associated with the different waste
109 treatment options must be thoroughly studied for the appropriate decision-making. Life Cycle
110 Assessment (LCA) methodology has been widely used for comparison of environmental impacts of
111 plastic waste management scenarios (Perugini et al., 2005, Lazarevic et al., 2010, Merrild et al.,
112 2012). However, the results are sometimes questioned since they are strongly influenced by the
113 assumptions made during the analysis and the quality of the data used (Ayres, 1995). The majority of
114 studies put the focus on mixed waste (mainly hard plastics) from domestic sources (Bovea et al.,
115 2010, Song et al., 2013, Fernández-Nava et al., 2014, Erses Yay, 2015). Gu et al. have assessed
116 mechanical recycling of several plastic materials made of PE including film scraps, agricultural films
117 and shopping bags from an environmental point of view. The environmental impacts of different
118 process stages, such as washing, sorting, shredding, extrusion and re-granulation were assessed. The
119 results showed that extrusion has the largest impact on the environment (Gu et al., 2017). In a study
120 published by Hou et al., several waste management options for plastic films from post-consumer
121 sources have been compared. Unlike post-industrial scrap, the sorting of domestic waste films is a
122 challenging task and must be included in the computation (Hou et al., 2018). Huysman et al. studied

123 the recycling of post-industrial plastic film scrap generated before converting steps such as printing.
124 Therefore, the plastic waste was not contaminated with inks. The authors considered the quality of
125 the recovered material by analysing the compatibility between polymers (Huysman et al., 2017).
126 In this work, for the first time, the environmental impacts associated with the upcycling process of
127 the printed plastic films from post-industrial source have been assessed and compared with the
128 traditional waste management options such as re-extrusion (downcycling) and incineration with
129 energy recovery. The influence of assumptions made in LCA has been evaluated, and two
130 modifications have been suggested to include when plastic waste management options are assessed.
131 In addition, the system boundaries have been extended to the end-of-life stage of the secondary
132 plastics produced with the upcycled post-industrial waste. The environmental impacts of two
133 material cycles were computed considering the current post-consumer waste management scenario
134 and then compared with two possible scenarios. The results show the direction in which the waste
135 management strategy should be developed and the real potential of current plastic waste
136 management options to fulfil the requirements of the circular economy model.

137 **2. Materials and methods**

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

141 **2.1. Goal and scope definition**

142 The goal of this study is to compare the environmental impacts of three waste management options:
143 upcycling process, downcycling and incineration with energy recovery. It is a 'gate to grave' approach
144 focusing on the end-of-life of printed plastic scrap from converting industry, including waste
145 treatment operations. This study does not consider the whole life cycle of the product as in a 'cradle
146 to grave' approach. The upstream life cycle stages of plastic products (production phases) are not

147 included since a) plastic scrap is considered as a waste, b) they are similar between the compared
148 scenarios and would not provide any insights for the analysis.

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

155 **2.2. Life cycle inventory analysis (LCI)**

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

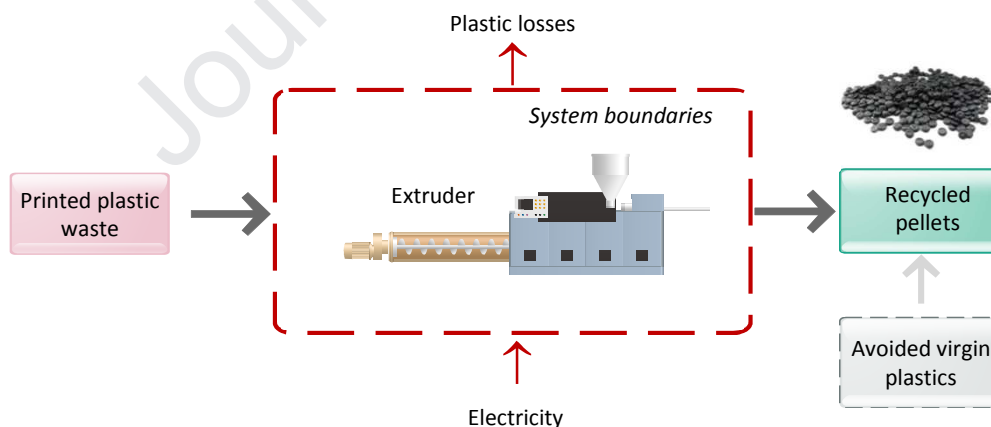
168 **2.2.1. Description of scenarios and data inventory**

169 The study focuses on three waste management scenarios.

170 **Scenario 1: Downcycling**

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

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



185
 186 *Figure 2. Modelled waste treatment scenario and overview of system flows: Scenario 1: Downcycling of plastic*
 187 *waste. Source: own elaboration (made with Edraw Max).*

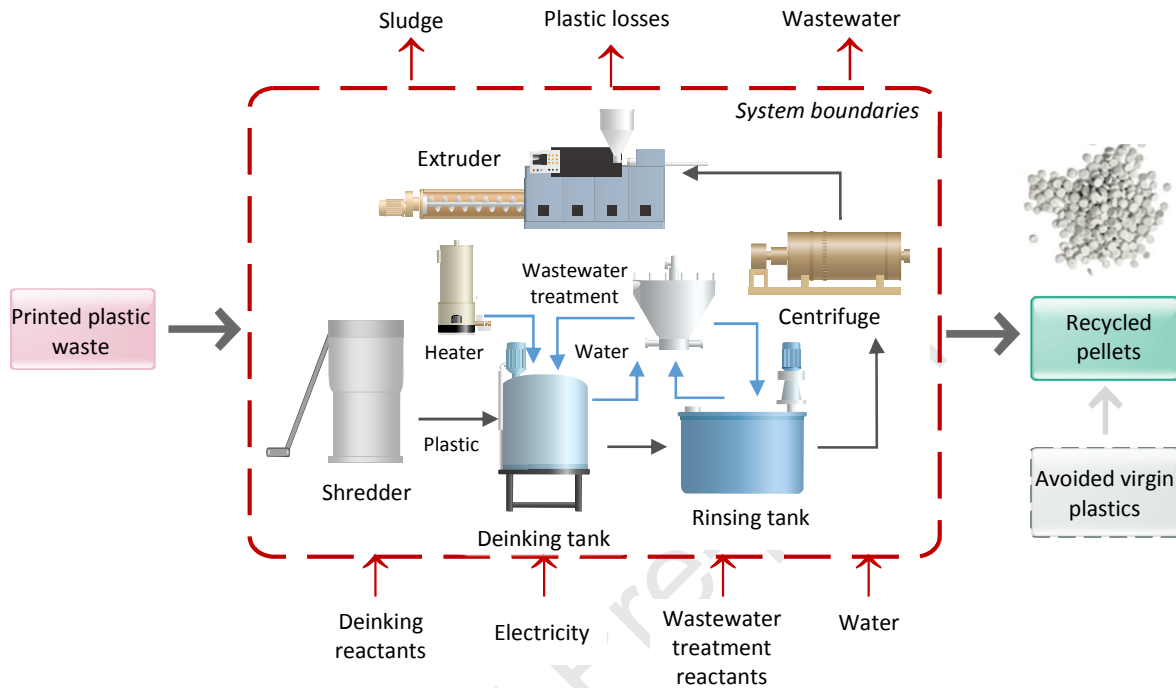
188 **Scenario 2: Upcycling (Recycling with deinking)**

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

197 The data related to the deinking plant were provided by Cadel Deinking Company which has
198 developed this innovative technology. The production capacity of this plant is 500 kg/h. The
199 electricity consumption of the deinking process (including shredding, washing, water heating, drying
200 and wastewater treatment) is around 600 kWh per tonne of input plastic. An average energy need
201 for the extrusion machine is 750 kWh per tonne of input plastic. European electricity mix is also
202 considered in this scenario. As mentioned before, non-hazardous reagents are used for deinking.
203 Also, during the wastewater treatment, specific chemicals are added. In total, the process requires
204 46 kg of reagents per tonne of input plastic. Despite the recirculation of treated water, it is necessary
205 to add a small quantity of tap water to cover the losses originated during the process (1200 L per
206 tonne of input plastic). The secondary outputs of the plant are wastewater not collected for
207 treatment (44 kg per tonne of input plastic) and an aqueous sludge containing inks (90 kg per tonne
208 of input plastic). The wastewater is purified in a medium-size municipal wastewater treatment plant.
209 And the sludge is managed through municipal incineration as non-hazardous waste. The recycling
210 efficiency is about 97%. The remaining part is collected and sent to landfill (Figure 3).

211 In this case, the converting company uses the recycled pellets for high added value products
212 manufacturing. The quality requirements are higher and, therefore, the recycled content should be

213 lower than in the downcycling scenario. In this case, the selected virgin plastic substitution rate is
 214 20%.



215

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

218 Scenario 3: Incineration

219 Flexible plastic waste is usually sent to incineration plants along with the municipal solid waste
 220 stream. This operation is well described in the Ecoinvent 2.2 database (disposal, polyethylene, 0.4%
 221 water, to municipal incineration [kg]), which contain the data required for the LCA analysis. The
 222 calorific value of plastic films is reported to be 41.41 MJ/kg (Asamany et al., 2017). And the energy
 223 obtained is used to produce electricity (21% of efficiency) and heat (74% of efficiency) (Merrild et al.,
 224 2012).

225 The most relevant LCI data have been summarized in Table 1.

226

227

228

229

230 *Table 1. LCI data for the three studied scenarios.*

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

231

232

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

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

238 The conventional recycling by means of re-extrusion produces dark colour pellets for non-demanding
 239 applications (e.g., trash bags, pipelines, pots). Meanwhile, the innovative deinking technology makes
 240 possible the production of high-quality pellets suitable for the same application as the original

241 material. Both processes make virgin plastic production decrease. The difference is the value of the
242 recycled product. Finally, the energy obtained during incineration replaces the European energy mix.

243 **2.3. Life cycle impact assessment (LCIA)**

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

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

263 **2.3.1. Impact categories**

264 The impacts on four categories have been evaluated and compared in this work. These are Human
265 Health, Ecosystem Quality, Climate Change and Resource conservation. All the categories are

266 environmentally relevant and included in the ISO 14040:2006 (ISO-Norm, 2006). Different units are
267 used to express the impact in selected categories. Disability-Adjusted Life Years (DALY) is used in the
268 human health category and represents the disease severity, considering both mortality and
269 morbidity. In other words, the number of DALYs represents the number of years of life lost over the
270 overall population (not per person). The midpoint categories included in the computation are human
271 toxicity, respiratory effects, ionizing radiation, ozone layer depletion and photochemical oxidation.
272 The ecosystem quality is expressed in Potentially Disappeared Fraction of species over a certain
273 amount of square metre during a certain amount of year ($\text{PDF}\cdot\text{m}^2\cdot\text{y}$). It is the sum of aquatic
274 ecotoxicity, terrestrial ecotoxicity, terrestrial acidification/nitrification, land occupation, and, aquatic
275 acidification, aquatic eutrophication and water turbidity. The unit used in the climate change
276 category is kg equivalent of carbon dioxide, which is used as a reference substance. The midpoint
277 category used is the global warming potential. Finally, in the resources category, MJ is used to
278 express the energy extracted or needed to extract the resource. Two midpoint categories are
279 considered here: non-renewable energy consumption and mineral extraction. Normalization and
280 weighting of the impacts were not performed in this study.

281 **2.3.2. Sensitivity analysis and model simulations**

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

287 **3. Results**

288 **3.1. LCA of the studied waste management options.**

289 The results are shown as positive and negative potentials (positive and negative values in the
290 graphs). A positive potential indicates a burden to the environment (negative environmental impact

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

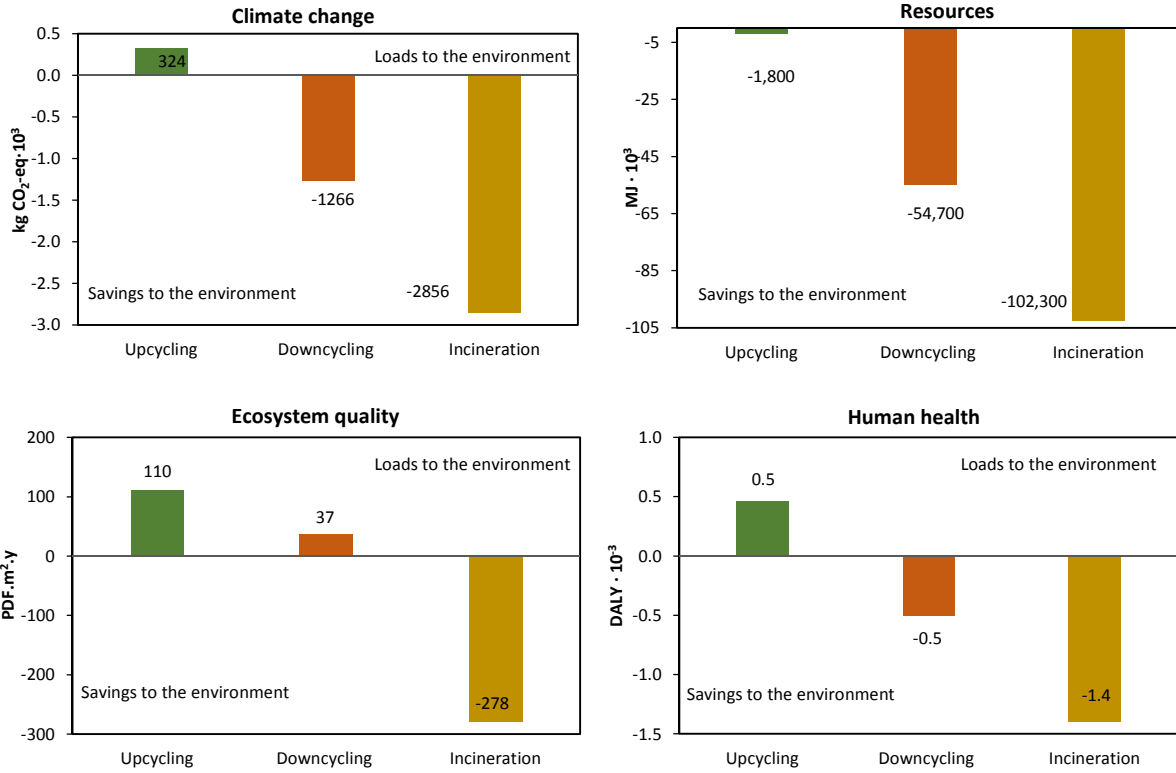
305 This approach also shows the benefits of plastic waste incineration over recycling processes. The high
306 heating value of polymeric materials makes them attractive for electricity or heating production
307 avoiding the use of such environmental pollutants as fossil fuels. Therefore, it generates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

313 If the LCA analysis is based on the virgin plastic substitution rate, important parameters such as the
314 quality of the recycled pellets and the intended applications are not considered. As a result, the
315 recycling option in which poor quality pellets suitable only for low demanding (usually single-use)

316 products are obtained seems to be more environmentally beneficial than the recycling process that
317 produces high quality recycled resins. The acceptable recycled content is higher since the
318 requirements of the non-demanding applications are lower. Thus, the avoided production of virgin
319 plastic increases.

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

332 The assumptions made to obtain the results shown in Figure 4 and Figure 5 lead one to make
333 decisions which go against the circular economy principles and the EU waste hierarchy. The quality of
334 the recycled pellets and the target market should be considered. Also, fairer energy substitution
335 criteria should be implemented.

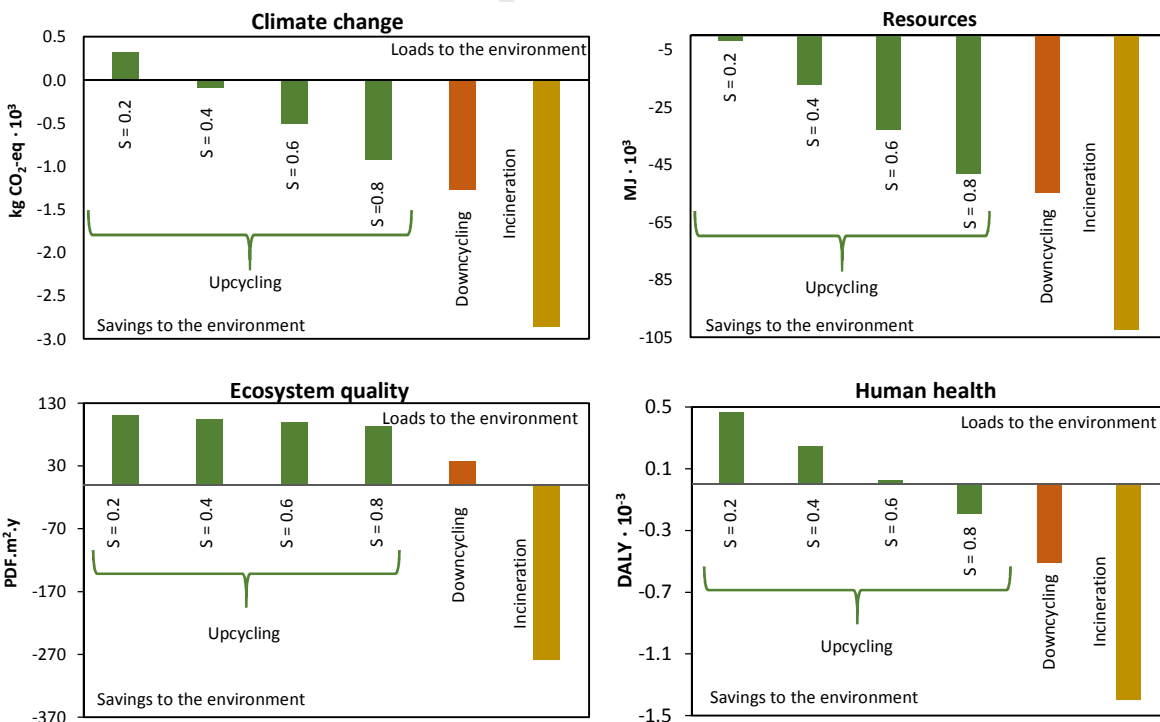


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338

Figure 4. Environmental impacts of each plastic scrap treatment scenario.



339

340

Figure 5. Environmental impacts of upcycling with different substitution rates compared with downcycling and

incineration. At converting company level.

343

344

345 **3.2. Modified LCA analysis according to the circular economy needs.**

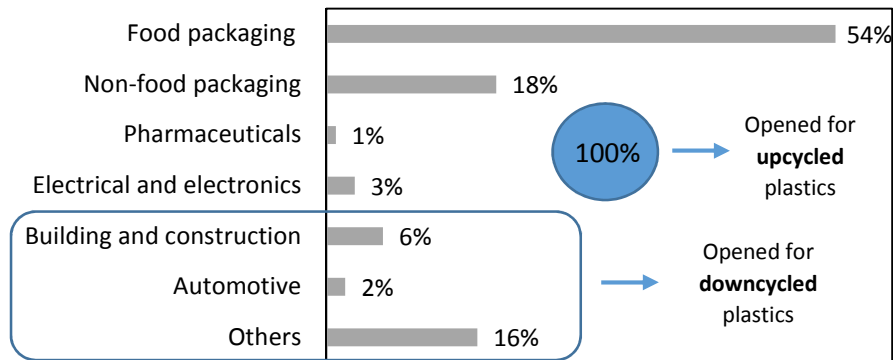
346 In this section, two simulations have been performed to consider the quality of the recycled material
347 and the source of energy.

348 **▪ Potential to substitute virgin plastics considering the target market**

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

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

363 European market of LDPE films can be divided into a number of sectors (Figure 6). Dark coloured
364 conventional pellets are suitable for building and construction, automotive and other less demanding
365 applications, which together represent 24% of the LDPE market. On the other hand, deinked pellets
366 owing to their higher quality can be used for all applications from food packaging to electronics or
367 building materials. So, its market share reaches 100%.



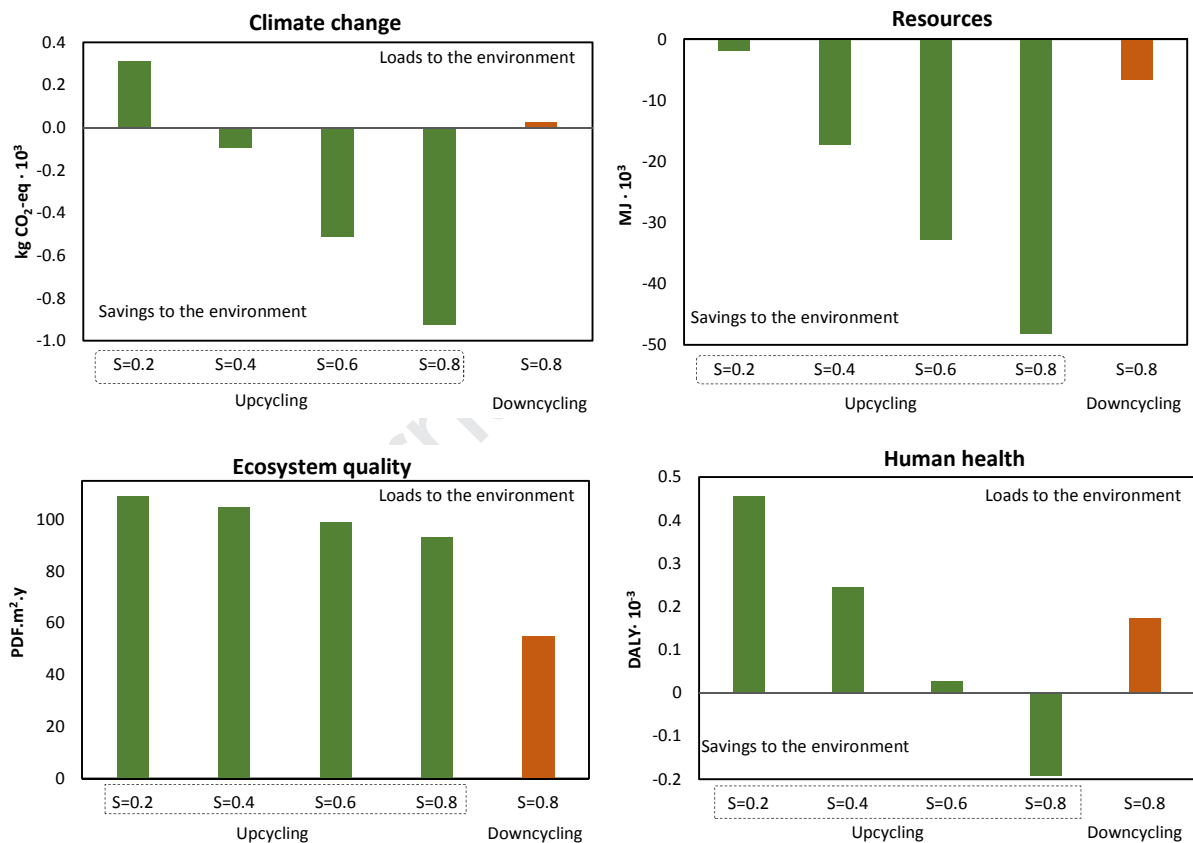
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Figure 6. European market share (%) of LDPE film. (Eriksen et al., 2019).

370 If the potential of the recycled material to substitute virgin plastic is calculated based on the entire
 371 market of a specific plastic, a different result is obtained (**Error! Reference source not found. 7**). The
 372 environmental savings of the downcycling process decrease: for climate change from 1266 kgCO₂eq
 373 to 24 kgCO₂eq and for resources from 54,700 MJ to 6615 MJ. The impact on human health changes
 374 from positive ($-5 \cdot 10^{-4}$ DALY) to negative environmental effect ($1.7 \cdot 10^{-4}$ DALY). Finally, the negative
 375 effect on ecosystem quality increases from 37 PDF.m².y to 55 PDF.m².y. Even so, the upcycling
 376 process with 20% of substitution appears to be less favourable than the downcycling. This scenario
 377 shows higher environmental burdens on climate change, ecosystem quality and human health. Also,
 378 the savings accomplished in resource conservation are lower. However, this perspective changes
 379 when the substitution ratio is higher than 40%. For instance, for climate change and resources
 380 categories, the use of 40% of recycled content produces a positive environmental effect (negative
 381 impact potential) and both values are above the levels of the downcycling process. Regarding
 382 ecosystem quality, the negative environmental effect decreases with the increase in the substitution
 383 ratio. Nevertheless, these values remain higher than in the downcycling scenario due to the use of
 384 chemical agents during the deinking operation. For human health category, 60% of recycled content
 385 is necessary to decrease the negative effects below the level of the downcycling. It is possible to
 386 produce savings to the environment if the substitution rate increases over 60%.

387 Therefore, the environmental benefits of the upcycling process are revealed when the avoided virgin
 388 plastic production is computed considering the global market. This approach takes account of the
 389 quality of the recycled plastic and the value of the products produced using recycling content. The
 390 additional effort that recycled materials upgrading requires is offset by the expansion of the target
 391 market. For instance, deinked clean pellets can be used for packaging manufacturing meanwhile
 392 conventional dark pellets are only suitable for less demanding applications. Moreover, if the more
 393 clean and homogeneous waste stream is diverted to new markets, then post-consumer plastic waste
 394 with lower quality can be introduced more easily for more forgiving applications.



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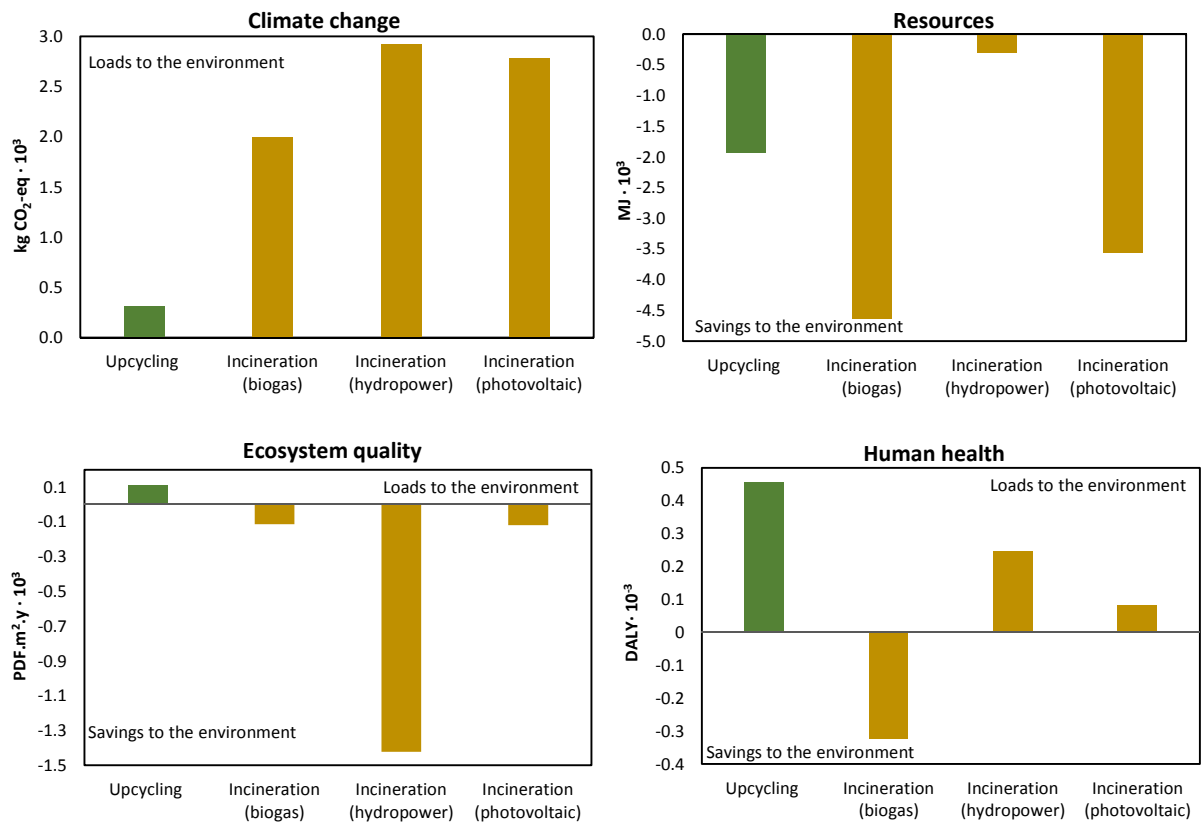
397 *Figure 7. Environmental impacts of upcycling and downcycling considering the market share ($MS_{upcycling}=1$;*
 398 *$MS_{downcycling}=0.24$). Influence of increasing substitution rates in the upcycling process included.*

399 ■ Potential to substitute fuels

400 As mentioned in section 3.1., the electricity and heat must be produced from renewable sources
 401 according to circular economy principles. Therefore, in LCA analysis the energy obtained during
 402 plastic waste incineration should substitute energy from non-fossil sources. In this study, three

403 examples of renewable energy have been assessed: biogas from agricultural plants, hydropower and
404 solar energy.

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



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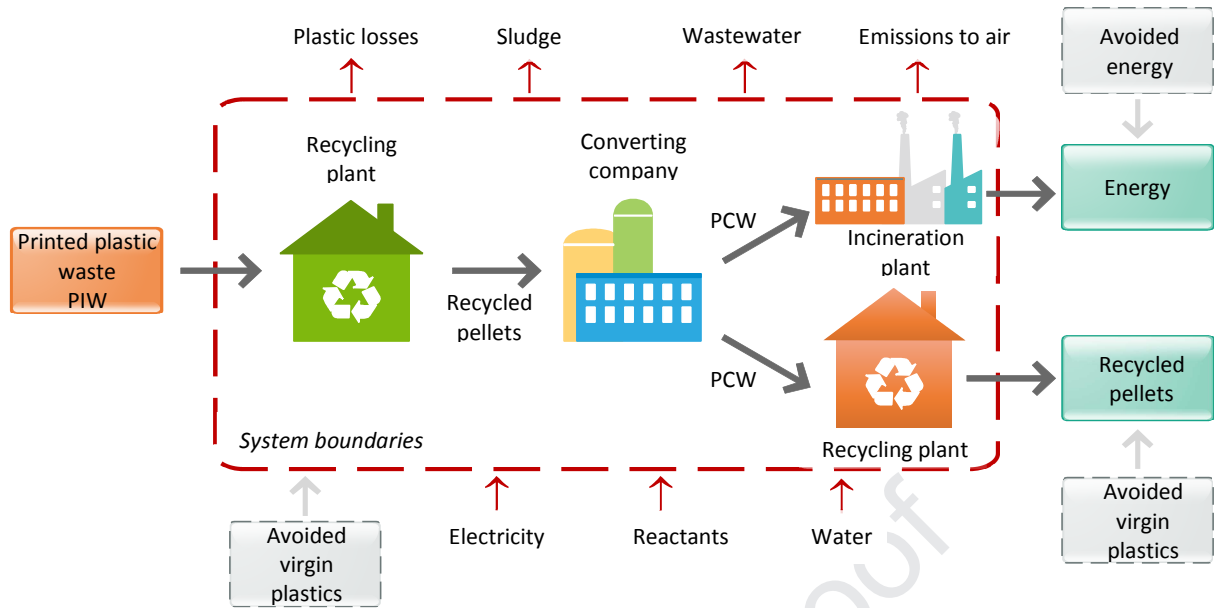
425 *Figure 8. Environmental impacts of upcycling considering the market share compared with incineration*
 426 *substituting renewable energy sources.*

427 3.3. Upcycling of plastic waste and circular economy

428 The upcycling of post-industrial plastic waste brings savings to the environmental, especially on the
 429 impact categories such as climate change and resources conservation. In previous sections, the
 430 system boundaries have been drawn around the recycling plant. Nevertheless, the product's higher
 431 quality leaves open the possibility for post-consumer plastic waste to be recycled after the use phase
 432 (Hahladakis and Iacovidou, 2018). Additional recycling cycles contribute to decreasing the virgin
 433 plastic production. Therefore, in this section, the system boundaries have been extended to the end-
 434 of-life stage of the products with recycled content. Landfilling has not been included in this study. So
 435 that the two post-consumer plastic waste treatment options are recycling and incineration (Figure
 436 9Error! Reference source not found.). In the European countries where landfill restriction has been
 437 implemented, around 40% of plastic waste is recycled (recycling rate (R)) and 60% is incinerated
 438 (Plastics Europe, 2018). The energy produced during incineration is used to generate electricity and

439 heat. The energy source replaced is biogas from agricultural plants. The post-consumer plastic waste
440 is currently treated through downcycling processes since existing technologies are not prepared to
441 remove all the contaminants and impurities. Hence, in the first or base scenario, the efficiency of the
442 recycling process is 90%, the substitution rate is 80% and the market share is 24%. The substitution
443 rate of virgin plastic by post-industrial recycled pellets (in the first material cycle) is 40%. The base
444 scenario has been compared with two additional scenarios. In the second one, the recycling rate of
445 post-consumer plastic waste has been increased to 80%. In the third one, the market share has been
446 incremented to 100%. It was assumed that the upcycling process is necessary to increase the market
447 share. So, the energy and resources consumption data for the post-consumer plastic waste recycling
448 correspond to the deinking process.

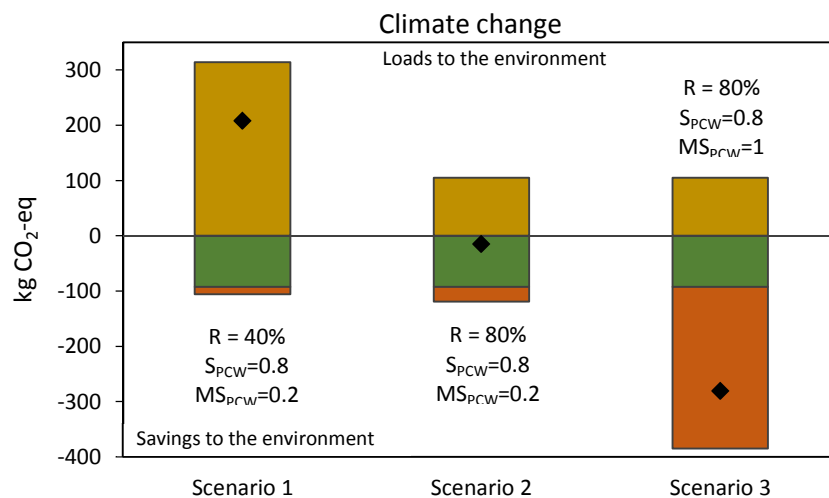
449 This approach has been used to show the environmental effects of post-consumer plastic waste
450 treatments (Figure 10). As has been discussed in section 3.2., incineration with renewable energy
451 substitution increases the emissions of carbon dioxide. Therefore, the first scenario causes the
452 biggest environmental burdens in the environment. The decrease of the incineration rate in scenario
453 2 results in negative net impact, which means saving to the environment. Although higher recycling
454 rate increases the positive effect on the environment, the savings of post-consumer waste recycling
455 are considerably higher (around 20 times) in scenario 3 when the target market expands. The same
456 trend is observed on resources conservation category. The net impact is around 1.6 times higher in
457 scenario 3 compared with scenario 1. These results show that it is good to recycle more. But it is
458 more important to maintain the quality and the value of plastic products since upcycling of plastic
459 waste produces the highest environmental savings.



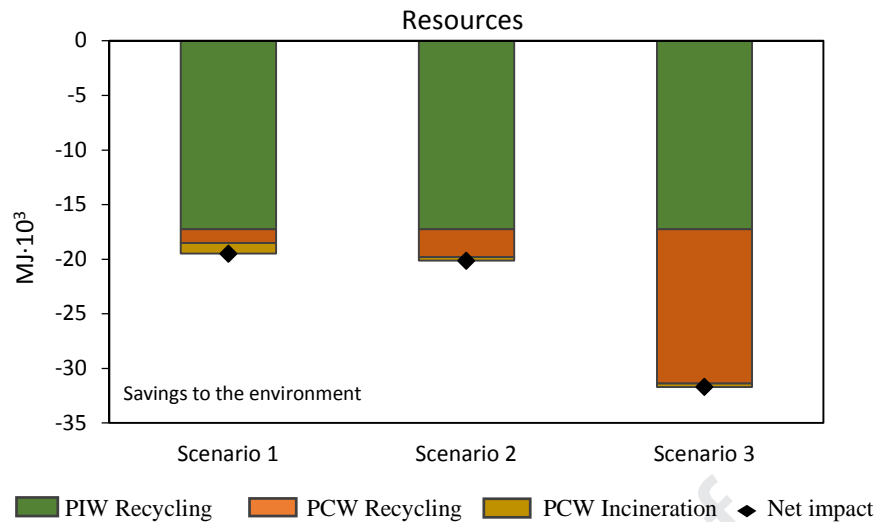
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461 *Figure 9. Modelled scenarios with extended boundaries and overview of system flows for both post-industrial*
 462 *waste (PIW) and post-consumer waste (PCW) treatment. Source: own elaboration (made with Edraw Max).*

463 To make possible the conditions established in scenario 3 (substitution rate and market share), post-
 464 consumer plastic waste recycling processes must be upgraded considerably. The quality of recycled
 465 post-consumer plastics is currently quite poor owing to the high level of contaminants, odours, non-
 466 intentionally added substances (NIAS), etc. Innovative decontamination (such as deodorization)
 467 technologies are needed to prepare the recycled content for more demanding applications.



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471 *Figure 10. Environmental impact on climate change and resources conservation of the upcycling process*
 472 *considering two material cycles. Three scenarios studied: current post-consumer waste management scenario*
 473 *(scenario 1) and simulated scenarios (scenarios 2 and 3). PCW (post-consumer waste). PIW (post-industrial*
 474 *waste).*

475 4. Discussion

476 The computation of the environmental impacts associated with the recycling of plastic waste differs
 477 among published studies. Gu et al. studied the mechanical recycling of different plastic materials
 478 where the recycled pellets were used in high value and low value applications. The avoided virgin
 479 plastic production was calculated based only on the substitution ratio. The authors state that the
 480 substitution ratio decreases with the increasing value of the final product and attribute this to the
 481 inferior quality of the recyclates (Gu et al., 2017). Nevertheless, they do not mention that the quality
 482 requirements in high value applications are much stricter, which strongly influences the substitution
 483 ratio. Huysman et al. use the percentage of substituted virgin materials and the recycling rate in the
 484 computation of impacts produced during the recycling of post-industrial plastic waste. The authors
 485 compare two waste streams which undergo different recycling options and calculate a circularity
 486 indicator based on the polymer compatibility. Finally, they concluded that the recycling option where
 487 recovered plastics are used to produce low value garbage bags is more environmentally beneficial
 488 than the scenario where the recycled plastics are used in high added value applications (Huysman et
 489 al., 2017). This can be attributed to the difference in the substitution ratio (80% and 20%

490 respectively) determined by the quality requirements of the final user. According to the results of the
491 present paper, these conclusions could have changed if the authors had considered the market share
492 of the intended applications. In a study presented by Hou et al., the recycling of plastic films from
493 post-consumer waste (mixed and recyclable) has been assessed. The parameters used for the
494 computations were the recycling rate, utilization rate (equal to the substitution ratio), composition of
495 polymers in the film waste, and the mass fraction of films in the waste stream. The authors studied
496 only one scenario with 66% of substitution ratio, but in the sensitivity analysis concluded that the
497 results are strongly dependent on this parameter (Hou et al., 2018). In general, the lack of uniformity
498 in the procedure adopted by different authors makes it difficult to compare and to determine the
499 accuracy of the LCA results. In fact, Viau et al. recently published an article where 51 LCA studies on
500 municipal waste management were reviewed to assess the discrepancies in the modelling of the raw
501 materials substitution. Based on the study by Vadenbo et al. (2017), the authors state that the
502 substitution potential must be calculated considering four parameters: the amount of potentially
503 recoverable materials, the recycling efficiency, the substitution ratio, and the market response. The
504 analysis showed that none of the revised articles mentions all four parameters. At the same time,
505 100% of selected articles take into account the substitution ratio. Nevertheless, 22% are not explicit
506 and 65% were not justified by the authors of the studies. Therefore, there is a lack of rigour in LCA
507 studies on municipal solid waste management (Viau et al., 2020). In the present paper, we have
508 considered all four parameters and the substitution ratios have been established based on the
509 information provided by a real plastic film converting company regarding quality requirements in
510 different applications. Also, the importance of the market response parameter has been shown,
511 especially when upcycling and downcycling processes are compared.

512 The incineration of plastic waste with energy recovery has been addressed in numerous studies and
513 the results vary depending on the waste composition, the heating value, the electricity or heat
514 production efficiency, and the source of substituted energy. Merrild et al. concluded that
515 incineration of municipal waste can be more beneficial than recycling when the level of energy

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

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

542 we have also presented the benefits of increasing the recycling rate in addition to the benefits
543 obtained from upgrading the quality of the recycled product.

544 **5. Conclusions**

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

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

559 In the second place, the energy produced during incineration should replace energy from renewable
560 sources instead of fossil fuels. The substitution of fossil fuels will surely provide more benefits than
561 recycling. This can be attributed to the fact that the heating value of plastics is higher than the
562 energy consumed during raw pellets production. This means that the energy recovered through
563 incineration is likely to be higher than the energy saved by avoiding virgin plastic consumption.
564 Moreover, according to circular economy principles, the energy has to come from renewable
565 sources. Therefore, if our society is moving forward to this new model, fossil fuels should not be
566 considered.

567 The use of upcycled pellets in new products manufacturing increases the value of these products,
568 thus, make them recyclable. Accordingly, a bigger number of material cycles is possible. The
569 influence of the recycling rate and the target market has been studied. The post-consumer waste
570 recycling rate slightly influences the environmental benefits of the process. The major savings are
571 produced when the target market for recycled plastics expands. For this to be possible, upcycling
572 processes for post-consumer waste must be implemented. Accordingly, innovative decontamination
573 technologies should be studied in future works to maintain the quality of the products and increase
574 the number of possible applications. The new technologies need to be accompanied by a transparent
575 and thorough LCA analysis considering all the relevant parameters and adapting the assumption
576 made to the Circular Economy principles.

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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.

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