Life cycle assessment of PE and PP multi film compared with PLA and PLA reinforced with nanoclays film

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1 LIFE CYCLE ASSESSMENT OF PE AND PP MULTI FILM COMPARED WITH PLA AND PLA

2 **REINFORCED WITH NANOCLAYS FILM.**

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10 Abstract

Plastic has become essential for our economy and the packaging industry. However, 11 12 plastic use is linked to environmental problems such as waste generation and loss of resources, since only 42% of the plastic used for packaging purposes is recycled. 13 Another problem associated with the use of plastic materials is caused by their 14 15 abandonment in the environment since they are non-degradable polymers. This paper 16 analyses the environmental performance of using biodegradable poly-lactic acid bags, 17 pure (PLA) and reinforced with nanoclays (PLA+NC), in comparison to conventional 18 alternatives made of polyethylene (PE) and polypropylene (PP) for being used to pack 19 fresh bakery products. The results reveal that for Climate Change and Fossil Resources 20 use, PLA+NC performs better than the alternatives. In the case of Climate Change, it has 45% less impact than low density polyethylene (LDPE), 39% less than PP, and 2% 21 22 less than PLA. However, the use of PLA+NC, results in higher impacts on Land Use and 23 Water Use, because this is produced from crops. Compared with PLA, PLA+NC has 5% less impact on these impact categories, but between 99-100% more impact on Land Use 24 25 and between 79-81% more impact on Water use than PP and LDPE. Thus, poly-lactic 26 acid bags reinforced with nanoclays are shown as an alternative for fossil-based 27 polymers (PE and PP) for certainty type of applications when we focus on Climate 28 Change and Fossil resources use reduction. In this sense, the results also reveal that the most environmentally friendly end-of-life for PLA and PLA+NC is incineration instead 29 30 of composting.

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32 Key Words

33 Bioplastic, Circular economy, Sustainable waste management, Poly-Lactic Acid,

34 Composite materials.

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36 **1. Introduction**

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38 **1.1. Bio-based as an alternative to fossil-based plastic**

Plastics are a family of lightweight and versatile materials that have become essential for our economy, with many applications both at the macro and microeconomic levels. In 2019 plastic production worldwide reached 370 Mt, the vast majority of them: fossilbased, from which 58 Mt were produced in Europe (Plastics Europe, 2020). The packaging industry consumes the major amount of plastic produced in Europe, representing about 40% of the total consumption (Oliver-ortega et al., 2021; Plastics Europe, 2020).

46 The plastic sector plays a crucial role in the maintenance of food safety, extending the 47 life of products as well as minimizing food waste (Matthews et al., 2021; World Economic Forum, 2016). However, plastic packaging has its proper drawbacks and environmental 48 49 related problems. Only in Europe, about 23 Mt of plastic packaging is produced every 50 year. This can be translated into a generation of 174 kg of packaging waste per inhabitant 51 in the EU (Eurostat, 2018), from which only 42 % is recycled, 39.5% is incinerated with 52 energy recovery and 18.5% is landfilled (Plastics Europe, 2020). The accumulation of 53 plastic in our natural environment and oceans, linked to its non-biodegradability, is another problem to be addressed (Borrelle et al., 2020; Cordier and Uehara, 2019). 54

All of the aforementioned issues lead the European Commission to adopt the European strategy for plastics in a circular economy (European Commission, 2018), as part of the European action plan for a circular economy (European Commission, 2020). This strategy aims to develop a new plastics economy, where the production of plastic and the design of plastic products fully respect reuse, repair, and recycling needs, and more sustainable materials are developed and promoted.

Particular attention should be given to the use of multi-layer packaging solutions. They allow reducing the packaging at the minimum expression while offering maximum protection to the food although. As a counterpart, and as pointed out by Dilkes-Hoffman et al., (2018), they pose big challenges for recycling due to the high costs and technological difficulties for separating the different layers or the inability to recycle mixed polymers.

67 One of the strategies followed up to improve the environmental performance of plastic 68 has been the addition of bio-based functional fillers to conventional plastics (Civancik-

Uslu et al., 2018; Hottle et al., 2013) that, in turn, may help to reduce production costs. 69 70 This is especially common in the food (Briassoulis and Giannoulis, 2018) and beverage 71 sector (Guo et al., 2021). Among the literature, we also find reviews of different 72 commercial biopolymers (La Rosa et al., 2014) and articles comparing reinforced 73 polymers with traditional plastics (Yates and Barlow, 2013). The most remarkable 74 tendency for reducing the environmental impact has been the development of bio-75 plastics, which comprise the groups of bio-based and biodegradable plastics, to 76 decouple plastic production from fossil resources consumption. The production of bio-77 based plastics is growing worldwide as an alternative to conventional plastics (Arkin et 78 al., 2019). It still only represents about one percent of the more than 368 Mt of plastic 79 produced annually, although the production capacities are set to increase from around 2.11 Mt in 2020 to approximately 2.87 Mt in 2025 (European Bioplastics, 2021). Among 80 all bio-based and biodegradable plastics, Poly-lactic acid (PLA) has become one of the 81 more promising options (Oliver-ortega et al., 2021), because it can be processed with 82 similar technologies and machinery as non-renewable based plastics, it has similar 83 84 mechanical properties to other thermoplastics. Regarding its gas barrier properties, and 85 according to the tests performed by Briassoulis and Giannoulis (2018), PLA has lower O₂ transmission rates and similar CO₂ transmission rates than conventional PP films. 86 Water permeability barriers of PLA are far from the ones of conventional fossil-based 87 88 plastics – about two orders of magnitude higher (Robertson, 1993). Especially for the 89 packaging sector, it is necessary to reduce the water permeability. One of the options to 90 improve them is by the addition of nanoclays that, properly dispersed in the PLA matrix, 91 produce difficulty in gas permeability, enhancing the barrier properties of the material. 92 The study performed by Rhim et al., (2009), revealed that the addition of different types 93 of nanoclays may decrease the water vapour permeability (WVP) between 6-33% in 94 relation to a control PLA film.

95 When focusing on food packaging, another important issue to be considered is related 96 to overpackaging. Reducing it is one of the measures highlighted in the New Circular 97 Economy Action Plan of the EU (European Commission, 2020). Finding the balance 98 between using packaging to reduce food waste without overpacking is crucial in this respect (see Figure 1). In a strict sense, overpacking can also be interpreted as using 99 100 materials with more properties than the ones needed for the application. Just to illustrate 101 it with an example, using PP film bags, with more than 200 days guarantee of water 102 barriers, to pack fresh bakery muffins does not make sense, since the durability of the 103 product is less than a week. It is in this context where alternatives such as PLA, or PLA 104 reinforced with nanoclay, can be presented as a suitable alternative to guarantee the

- 105 balance between food waste production and overpackaging. Taking into account that
- 106 PLA can be composted after its life use, which could even improve the environmental
- 107 benefits of this material in contrast to non-biodegradable materials.
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Figure 1: Sörås curve



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Source: Based on (Erlöv, 2000)

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115 **1.2. Life Cycle Assessment (LCA) approach**

116 According to the Life Cycle Initiative or the United Nations, Life Cycle Assessment (LCA) 117 is a time-tested assessment methodology to evaluate the environmental performance of a product or service throughout its life cycle (Life Cycle Initiative, 2021). LCA is 118 standardized by the standards ISO 14040 - defining the principles and framework - and 119 120 14044 – detailing the requirements and providing guidelines on how to apply them. Essentially, LCA can be described as a balance of materials and energy of the analysed 121 product system, combined with an assessment of the potential environmental impacts 122 123 associated with the inputs (consumption of materials and energy) and outputs (emissions to water, soil, and air) of the same system. Altogether, it provides a comprehensive and 124 holistic view of the environmental loads of the products or services under study, covering 125 126 a wide set of environmental performance indicators such as Global Warming Potential, Acidification Potential, Eutrophication Potential, Ozone Layer Depletion Potential, 127

Human Toxicity Potential or Ecotoxicity Potential (Baumann and Tillman, 2004; Muñoz,2006).

130 LCA has been widely used for research on packaging solutions. Many studies in the 131 literature compare different material alternatives in the food packaging industry 132 (Verghese et al., 2012), including reusable options as reusable plastic containers for food 133 (Accorsi et al., 2014; Humbert et al., 2009), packages for juices (Banar and Cokaygil, 134 2008), and coffee (De Monte et al., 2005). In other studies, the LCA methodology is used for the comparison of beverage packaging made of different materials like aluminum, 135 wood, PET, PLA and glass (Saleh, 2016; Van der Harst and Potting, 2013). For instance, 136 137 Blanca-Alcubilla et al., (2020) compared these materials in the aviation catering sector. Humbert et al., (2009) compared glass and plastic alternatives for baby food packages 138 139 and Papong et al., (2014) compared drinking water bottles from different polymers. Some 140 life cycle studies of mineral-filled composite plastic materials are already found in the literature (Civancik-Uslu et al., 2018; Guo et al., 2021), although very few have yet been 141 142 found in which the addition of materials to the polymer matrix provided an increase in function (Lorite et al., 2017; Oliver-ortega et al., 2021). 143

The present article aims to assess the environmental impacts of PLA reinforced with nanoclays in comparison to PLA, PE, and PP when used as plastic film for packing fresh bakery products, using the LCA methodology. PLA reinforced with nanoclays is assessed to be a substitute to multi-layer films of PE and PP offering enough protection and barrier properties for the product packed and, the literature comparing these materials under a life cycle perspective is scarce.

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- 151 2. Materials and methods
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153 2.1 Goal and scope definition

This study aims to compare the environmental performance of film made of PLA reinforced with nanoclay with alternative films made of PLA, PP, and PE for packaging bakery fresh products. The main characteristics of the films compared are shown in Table 1.

The functional Unit (FU) defined to describe and compare the function of the product is
"one plastic bag of 20 x 30 cm and 25 µm thick to be used to pack and maintain fresh
bakery products during 4 days".

The reference flow to fulfill the FU in each case is presented in Table 2. The analysis performed corresponds to a "cradle-to-grave" LCA, covering all relevant processes from raw materials production to the final waste treatment. The modeling has been done using GaBi professional software and databases.

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Table 1. The main properties of the films compared

	Density	WVTR
	(g/cm ³)	(g H2O ⋅ m ⁻² ⋅ d ⁻¹)*
Film PLA	1.24	185
Film PLA + 4% nanoclay	1.21	145
Film PP	0.90	3
Film LDPE	0.95	8
*Water vapour transmissior	n rate	

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Table 2. Reference flow of the films compared

	Materials	Weight (g)
Film PLA	PLA	1.86
Film PLA + 4% nanoclay	PLA	1.74
-	Nanoclay	0.073
Film PP	PP	1.35
Film PE	PE	1.42

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171 2.2 Life cycle inventory analysis (LCIA)

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173 2.2.1. Raw material production

174 2.2.1.1 PLA

To produce PLA, a life cycle inventory of PLA/starch blend production from the GaBi database has been used (Sphera, n.d.). The production of PLA is based on data provided to produce Ingeo Polylactide production by Natureworks LCC. It represents the fictive route for production in Europe and it is representative of the period 2020-2023. The inventory includes all relevant processes needed for the production of the material. The inventory includes up to the production of granulates ready to be extruded.

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182 2.2.1.2 PLA with nanoclay

The PLA reinforced with nanoclay used in this study was the one developed by the Group LEPAMAP-PRODIS, Department of Chemical Engineering, University of Girona, and described by Oliver-ortega et al., (2021). However, to make it comparable with data to produce PE and PP at the industrial level, instead of using the energy consumption of

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the machinery at the lab scale, the energy consumption of equivalent equipment suitableto be used at the industrial scale has been used.

189 PLA reinforced with nanoclay is a nanocomposite material made of a PLA polymer matrix 190 and the addition of hydrophilic bentonite and surface modified nanoclays containing 191 trimethyl stearyl ammonium, in a proportion in weight of 96:4. The inventory includes the 192 production of the raw materials needed, the transportation from the producers and the 193 production process. The production of PLA granulates was taken from the GaBi professional database and is the same used for the PLA inventory. For the production of 194 195 nanoclays, the life cycle inventory for the production of kaolinitic clay, granular, or 196 powder, with a moisture content of 0 to 14% from the GaBi Database has been used. 197 Data is representative for the period 2015-2025 and represents an average production in Europe (Europe 28+Turkey and Ukraine). 198

The methodology to produce the nanocomposite material is the masterbatch (see Figure 200 2). It starts with a concentrated blend of PLA and nanoclays (22% wt of nanoclays) that 201 is, afterward, diluted with more PLA in a high-intensity mixer. The material is milled to 202 obtain pellets ready to be extruded.

- 203 It has been considered that, at an industrial scale, only one final extrusion to get the 204 pellets would be needed. Thus, and because the extrusion of the PLA was already 205 included in the inventory used for PLA raw material, no additional energy consumption 206 is considered. Only PLA and nanoclay production as well as their transportation to the facility is considered. For the transportation, a Truck with 10-20 t of gross weight with 207 208 11.4 t of payload, fulfilling Euro VI requirements is used. The distance considered is 100 209 km with an average utilization rate per weight of 0.53. The production of diesel needed for transportation is already included. It is representative of a diesel mix production at a 210 211 refinery in Europe-28 for the period 2017-2023.
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231 2.2.2. Film production

For modeling this stage, the same LCI process from the GaBi database has been used. It includes the extrusion of the film and it is non-product-specific. It considers an energy consumption of 1.6MJ per kg of material extruded and 4% of losses in the process. The LCIs used for the electricity mix production as well as for the thermal energy from natural gas are representative of Spain for the period 2017-2023. The LCI for lubricants production at the refinery is representative of Europe-28 for the period 2017-2023.

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239 2.2.3. Transport

The transport of the film from the producer to the packed and the correspondent transport to the selling points have been modelled considering a truck trailer Euro VI 14-20 t of Gross Weight (11.4 t payload capacity) from the GaBi database. The production of diesel mix at refinery representative for Europe-28 for the period 2017-2023 has already been used.

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246 2.2.4. Use stage

247 Due to the singularity of the products analysed, this stage has been neglected.

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249 2.2.5. End of life

Plastic film is recovered in packaging sorting plants within the mixed plastic fraction, in which there is no distinction between the different types of polymers. Its main treatment in the European Union is incineration with energy recovery (Plastics Europe, 2020). This has been the end-of-life treatment analysed in the baseline case study. The LCIs used is representative of Spain for the period 2020-2023 and include the recovery of electricity, as well as the correspondent credits due to the electricity that has not been produced using the Spanish average electricity production mix.

However, as PLA can also be composted, a sensitivity analysis to visualize the effects
of a different end-of-life process for PLA and PLA+NC bags is also performed.

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260 2.3 Impact categories and characterization factors

The environmental impact categories included in this study have been selected from the ones recommended by the Product Environmental Footprint (PEF) of the European

263 Commission (Manfredi et al., 2012). In particular, the ones included in Table 3 have been

- selected for being considered the most relevant impact categories related to the type of
- 265 products analysed in this study.
- 266

Table 3. Environmental impact categories considered

EF Impact Category	EF Impact Assessment Model	EF Impact Category indicators	Source
Climate Change	Bern model- Global Warming Potentials (GWP) over 100 years time horizon	Kg CO₂ equivalent	(IPPC, 2007)
Resource Use fossils	Abiotic resource depletion fossil fuels (ADP-fossil) based on the lower heating value	[MJ]	Based on (van Oers et al., 2002) as implemented in CML, v. 4.8 (2016)
Land Use	Soil Quality Index	[Pt]	Re-Calculated by JRC starting from LANCA® v 2.2 as a baseline model.
Water Use	Swiss Ecoscarcity model	m ³ water used related to local scarcity of water	(Frischknecht et al., 2006)

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271 **3. Results and discussion**

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273 3.1 Shelf-life days calculation

To guarantee that the functional unit of the 4 alternatives analysed is the same, which means that they can maintain in perfect conditions the properties of the fresh bakery products packed during a period of 4 days, the shelf life in days of each one of them has been calculated.

The absorption studies of Cervenka et al. (2008) applied to gingerbread have been used to estimate the shelf life (θ_s) (Eq 1) of the gingerbreads stored in different types of bags (100g of gingerbread with initial moisture of 2.5% H₂O, inside 20x30cm bags with a thickness of 25µm) at room temperature, taking into account the specific packaging material used.

283 Eq. 1
$$ln\left(\frac{m_e-m_i}{m_e-m_c}\right) = \frac{P}{X} \cdot \frac{A}{W_s} \cdot \frac{p_0}{b} \cdot \theta_s$$

285 where me is the moisture value in the equilibrium of food exposed to external package relative humidity (RH) in the isotherm, mi is the initial moisture content (% weight) and 286 287 the m_c is the critical moisture content (% weight), considered the point in which the 288 product starts to lose properties. These values have been obtained from the isotherm absorption. P/X is the water vapour permanence ($g H_2O \cdot m^{-2} \cdot d^{-1} \cdot cmHg^{-1}$) of the packaging 289 material at 25°C and 50% of RH, A is the area of the package (m²), W_s is the dry food 290 291 content (g), p₀ is the vapour pressure at the storage conditions (2.376 cm Hg), and b is the slope from the isotherm of the curve moisture content versus water activity. 292

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Table 4. Shelf life of bakery products depends on the type of packaging

MATERIAL	Shelf life (days)
PLA	7.7
PLA+4% Nanoclays	9.8
PP	479.2
LDPE	178.8.0

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As can be seen in Table 4, all materials fulfill the defined FU of 4 days of durability. At this point, it is worth mentioning that the higher WVTR of PP and LDPE compared to PLA and PLA+NC result in much higher values of shelf life of the product, and also that the addition of nanoclays to PLA improves the shelf life up to 2 days (more than 25%) compared to PLA. However, although the results are far from common fossil-based plastics, moisture is not the only factor in bakery shelf-life products. Microbial growth, staling, and oxidative rancidity are other key factors for fresh bakery (Robertson, 2013).

303 When analysing the possible use of PLA and PLA+NC as PP and LDPE substitutes, it is 304 necessary to consider the life of the product packed itself. Cereals, pasta, and other dry 305 food are highly sensitive to water. The increase in the moisture content represents a loss 306 of the food properties and could damage irreversibly the product. Thus, the use of PLA 307 and its nanocomposites is inadequate. On the other hand, fresh pasta, fresh bakery, fruits, and horticultural products have limited life due their nature and components. In this 308 309 kind of product, their durability can be improved using packaging, and WVTR it is not so 310 important. It is in this context where the use of PLA or even better PLA+NC is recommended. 311

313 3.2 Environmental profile

The environmental results of the four bags are shown in Table 5. Those results are represented in the relative contribution in Figure 3. As can be observed, PP and LDPE solutions have a higher impact on climate change and fossil resources use indicators, whereas they have less impact and perform better for water and land use than the alternatives PLA and PLA+NC.

319 Regarding climate change, the option with more environmental impact is LDPE. PP, PLA, 320 and PLA+NC have correspondingly 6%, 43%, and 45% less impact on this impact 321 category than LDPE. When moving to the fossil resources use indicator, again LDPE is the most harmful option. PP has 9% less impact in this category followed by PLA with 322 323 44% less impact and PLA+NC with 46% less impact. However, since PLA is based on 324 plants, the corresponding need for land and water for the crops is included in the 325 accounting. This is the reason why these indicators are much higher for PLA and 326 PLA+NC than for the fossil-based alternatives. PP and LDPE have 98% and 99% less 327 environmental impact on land use and 79% and 81% less impact on water use than PLA. Regarding PLA+NC it is remarkable that the addition of nanoclays to the formulation 328 329 results in a decrease of 5% of the impact on these two impact categories in comparison 330 to pure PLA.

All in all, what can be stated is that there is not a clear best option in all impact categories. 331 332 Thus, the option should be selected bearing in mind the most relevant impact category to be addressed. If the purpose is to reduce global warming, then, the best option would 333 334 be PLA+NC. On the contrary, if the main problem to be addressed is water scarcity, then 335 LDPE would be the best option. Having a complete environmental profile of the analysed 336 options allow decision makers to make science-based choices, putting on the table the 337 fact that, usually, when dealing with environmental problems, no perfect solutions exist, 338 and that the choices we made trying to reduce one singular environmental problem may 339 produce more environmental impact in others.

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Table 5. Environmental profile of the analysed bags

	PLA	PLA+NC	PP	LDPE
EF 3.0 Climate Change - total [kg CO2 eq.]	4,27E-03	4,10E-03	6,93E-03	7,44E-03
EF 3.0 Resource use, fossils [MJ]	6,37E-02	6,10E-02	1,04E-01	1,13E-01
EF 3.0 Land Use [Pt]	3,00E-01	2,87E-01	4,02E-03	6,54E-03
EF 3.0 Water use [m ³ world equiv.]	3,23E-03	3,08E-03	6,88E-04	6,09E-04

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Figure 3: Comparison of the environmental profile of the analysed bags

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350 3.3 Sensitivity analysis

351 With the current technology, recycling PLA in a plastics mixed stream is not possible, due to high costs in the separation and processing, as well as issues related to 352 contamination and poor quality of the recycled materials (Benavides et al., 2020). This 353 354 is the reason why this material is commonly disposed in landfills in the United States or incinerated in the European Union (Plastics Europe, 2020), the latter the end of life 355 356 considered in the present study as a baseline. Even though, PLA is a biodegradable polymer that in the adequate conditions breaks down into CO₂, CH₄, and water (Castro-357 Aguirre et al., 2016; Lyu et al., 2007). 358

At mesophilic temperatures (21-35 °C), like those assumed to exist in landfills, PLA can be considered non-biodegradable (Kolstad et al., 2012; Krause and Townsend, 2016). However, it biodegrades 90% after 120 days at thermophilic conditions (50-65 °C) with those in an industrial composting facility (Itavaara et al., 2002). Unlike anaerobic degradation in landfills, composting consists of aerobic degradation of PLA, which is

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expected to fully oxidize the carbon to CO_2 and H_2O , reducing the emissions of CH_4 to less than 5% (Itavaara et al., 2002).

366 A sensitivity analysis has been performed to evaluate the environmental impacts of 367 composting this material at the end-of-life stage instead of incinerating it. The study has 368 been performed for a composting fraction of 0%, 50%, and 100%, meaning that the 369 remaining % is sent to incineration. The results presented in Figure 4 shows how the 370 environmental impacts of the overall system increase while the percentage of composting is increasing as well, both for PLA and PLA+NC. This is mainly due to the 371 372 methane emissions occurred during the composting process that does not occur, or 373 occur to at lesser extent, in incineration facilities. Although coming from biogenic 374 sources, CH₄ emissions have 34 times more global warming potential than CO₂.

In addition, the incineration of PLA produces electricity, with the corresponding environmental credits related to the production of electricity from the Spanish electricity production mix, whereas composting it in an industrial facility at 50-65 °C is energy intensive. Thus, can be stated that incineration of PLA and PLA+NC is most environmentally friendly than composting it for the analysed impact categories.

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Figure 4. Sensitivity analysis considering composting



383 4. Conclusions

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This paper reveals that the incorporation of nanoclays into PLA increases the barrier properties of the material in relation to PLA and results in less environmental impact for all the environmental impact categories analysed: climate change, fossil resources use, land use, and water use, in comparison to pure PLA. As far as known by the authors, the LCA developed in this paper is the second environmental study comparing reinforced-nanoclay with conventional polymers in a life cycle perspective after (Lorite et al., 2017).

From this paper can also be concluded that, from the environmental point of view, the 392 use of PLA+NC film or bags as an alternative packaging material to films of PP and LDPE 393 394 is recommendable if: a) we are trying to reduce our contribution to global warming 395 potential or on fossil resources use, and b) we are packing product for which the water 396 vapour transmission rate (WVTR) is not a critical factor for their conservation, such is the case of fresh pasta, fresh bakery, fruits or other horticultural products. However, the 397 398 preference for the use of PLA+NC can lead to more environmental impacts on land use 399 and water used that the fossil fuel alternatives, due to the plant base origin of PLA.

400 Consequently, another learning from this paper is the fact that when dealing with 401 environmental problems no unique or better solutions exist. Usually, when trying to 402 reduce an environmental problem, other environmental problems may arise. Using LCA 403 can help decision-makers to visualize those environmental shifts among different life 404 cycle stages, or from one product to another, and make more conscious and science-405 based decisions.

406

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HIGHLIGHTS

- Environmental assessment of different plastic bags to pack fresh bakery products.
- Comparison of fossil based (PP and PE) and plant-based polymers (PLA).
- Inclusion of the assessment of a new material: PLA reinforced with nanoclays (NC).
- Using Life Cycle Assessment to get the environmental profile of the alternatives.
- PLA+NC is recommended if water vapor transmission rate is not critical for packing.
- PLA and PLA+NC have more impact on land and water use than PP and PE.
- PLA and PLA+NC have less impact on climate change and fossil resources use.

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Declaration of interests

☑ 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.

□ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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