DEPARTMENT OF CIVIL AND ENVIRONMENTAL ENGINEERING

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ADVANCES IN LIFE CYCLE ASSESSMENT OF PLASTICS WITH MINERAL FILLERS IN PACKAGING APPLICATIONS

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Dedicated with all my love,
to my husband Emre
for standing next to me in this journey
and to my parents
for giving me the courage to start this journey.

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PREFACE

The work presented in this thesis was funded by GCR Group (Tarragona, Spain), producer of mineral masterbatches, and performed at UNESCO Chair in Life Cycle and Climate Change - ESCI-UPF (Barcelona, Spain), within the PhD Programme in Environmental Engineering at the Department of Civil and Environmental Engineering at Polytechnic University of Catalonia (UPC). The main objective of the project was to investigate the environmental profile of plastics with mineral fillers in packaging applications by using Life Cycle Assessment (LCA) methodology.

All the research was performed under the supervision of Dr. Pere Fullana i Palmer (Director of the UNESCO Chair in Life Cycle and Climate Change and Prof. at ESCI-UPF) and Dr. Rita Puig (Assoc. Prof. at Department of Informatics and Industrial Engineering, Universitat de Lleida). The data used in this work was provided by GCR Group or its customers.

This PhD thesis is mainly formed through the compilation of four scientific papers. Two of them (Chapter 2 and 3) have already been accepted for publication in peer-reviewed international scientific journals, which are defined as first quartile by Journal Citation Reports. The other two (Chapter 4 and 5) have already sent for publication and currently under review. The list of the papers developed is as follows:

I. Published papers:

- **Didem Civancik-Uslu,** Laura Ferrer, Rita Puig and Pere Fullana-i-Palmer (2018). Are functional fillers improving environmental behavior of plastics? A review on LCA studies. *Science of the Total Environment*. 626, 927-940. https://doi.org/10.1016/j.scitotenv.2018.01.149

- **Didem Civancik-Uslu,** Rita Puig, Laura Ferrer and Pere Fullana-i-Palmer (2019). Influence of end-of-life allocation, credits and other methodological issues in LCA of compounds: An in-company circular economy case study on packaging. *Journal of Cleaner Production*. *Special issue on product lifetimes*. 212, 925-940. https://doi.org/10.1016/j.jclepro.2018.12.076

II. Submitted papers:

- **Didem Civancik-Uslu,** Rita Puig, Stephan Voigt, Dieter Walter and Pere Fullana-i-Palmer. Improving the production chain with LCA and Eco-design: Application to cosmetic packaging. *Submitted to Journal of Resources, Conservation and Recycling. Special virtual issue of sustainable cycles and management of plastics.* Under Review.

- **Didem Civancik-Uslu,** Rita Puig, Michael Hauschild and Pere Fullana-i-Palmer, Life cycle assessment of carrier bags and development of an indicator of littering. *Submitted to Science of the Total Environment.* Under Review.

In addition, an oral presentation was done in an international congress:

- **Didem Civancik-Uslu**, Rita Puig, Laura Ferrer, Pere Fullana-i-Palmer. Functional fillers as a part of green chemistry: LCA Review *(Oral presentation)*, 7th International Conference on Green and Sustainable Innovation (ICGSI), Bangkok, Thailand, 17-19 October 2018.

In addition to the work forming the main body of this thesis, some additional work was also performed as a part of the GCR project. The following presentations were done:

- **Didem Civancik-Uslu**, Laura Ferrer, Rita Puig, Pere Fullana-i-Palmer. Environmental improvement of a polyethylene refuse bag by adding a mineral filler (granic 1522): a life cycle study *(Oral presentation)*, 10th World Congress of Chemical Engineering (WCCE10), Barcelona, Spain, 1th-5th October 2017.
- **Didem Civancik-Uslu,** Rita Puig, Mark Falcó, Pere Fullana-i-Palmer. Food trays with less environmental impact *(Poster presentation)*, 11th International Conference of Life Cycle Assessment of Food, Bangkok, Thailand, 17-19 October 2018.
- **Didem Civancik-Uslu**, Rita Puig, Mark Falco, Pere Fullana-i-Palmer. Impactos ambientales de las bandejas con rellenos minerales. Conama 2018. Congreso Nacional del Medio Ambiente.26-29 November 2018.

Finally, the following publications, dealing with carbon footprint and LCA were also developed during the PhD in addition to the ones forming the thesis:

- Maryam Yuli, Rita Puig, Miguel Angel Fuentes, **Didem Civancik-Uslu** and Marc Capilla (2018). Eco-innovation in garden irrigation tools and carbon footprint assessment. *International Journal of Environmental Science and Technology*, 1–14. https://doi.org/10.1007/s13762-018-1937-y
- Jaume Alberti, **Didem Civancik-Uslu**, Davide Contessotto, Alejandra Balaguera, Pere Fullana-i-Palmer (2019). Does a Life Cycle Assessment remain valid after 20 years? Scenario analysis with a Bus Stop study. *Journal of Resources, Conservation and Recycling,* 144, 169-179. https://doi.org/10.1016/j.resconrec.2019.01.041

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LIST OF ACYRONYMS

ADP	Resource depletion, mineral, fossils and renewables
AP	Acidification potential
CF	Carbon fiber
CL	Close-loop
EoL	End-of-life
EP freshwater	Eutrophication potential in freshwater
EP marine	Eutrophication potential in marine water
EVA	Ethylene vinyl acetate
EVOH	Ethylene vinyl alcohol
FU	Functional unit
GCC	Ground calcium carbonate
GF	Glass fiber
GHG	Greenhouse gas
GWP	Global warming potential
HDPE	High density polyethylene
ILCD	International reference life cycle data system
ISO	The international organization for standardization
LCA	Life cycle assessment
LCI	Life cycle inventory

LCIA	Life cycle impact assessment
LDPE	Low density polyethylene
LLDPE	Linear low-density polyethylene
NFs	Natural fibers
ODP	Ozone depletion potential
OL	Open-loop
PCOF	Photochemical ozone formation, human health
PCR	Post-consumer recycled
PE	Polyethylene
PE_non ren	Primary energy from non-renewable resources
PE_ren	Primary energy from renewable resources
PEF	Product environmental footprint
PET	Polyethylene terephthalate
PP	Polypropylene
PVC	Polyvinylchloride
Qp	Quality factor of primary material
Qs	Quality factor of secondary material
UNEP	United Nations environment programme

SUMMARY

Mineral masterbatches, in other words mineral fillers, are concentrates of calcium carbonate or talc in a polymer base. In addition to their optimal cost, their use improves mechanical properties of materials and reduce energy consumption during the processing. Finally, their use may help to reduce the carbon footprint of the final product, since they include (at some percentages) naturally existing minerals (calcium carbonate and talc) instead of virgin petrochemicals.

Due to the increasing environmental pollution that the world is recently facing, concerns over using plastic have also increased. Plastic packaging, origin of 30% of the total annual plastic production, is one of the major areas which requires special attention. Therefore, the main focus of this thesis is the environmental assessment of the use of masterbatches, with calcium carbonate and talc, in different plastic packaging applications by using life cycle assessment (LCA) methodology, as being the commonly accepted tool in estimating the potential environmental impacts of products or systems. While doing this, it is also aimed to provide advancements in the LCA methodology of plastic compound materials. In this thesis, through a collaboration with a producer of mineral masterbatches, different studies were performed based on real market cases.

The outcomes of this thesis are: i) a deep and extended literature review on environmental impacts of functional fillers (including masterbatches) used in the industry; ii) two LCA case studies ("plastic compound vs eucalyptus wood storage sheets" and "virgin polyethylene (PE) vs eco-designed cosmetic tubes with mineral fillers and recycled content"), in which the use of plastics with mineral fillers is compared to virgin plastic alternatives; iii) introduction of methods, within the inventory phase, to calculate the quality loss (also known as Qs/Qp factors) of compound materials in open-loop and closed-loop recycling; and iv) introduction of a littering indicator, within the impact assessment phase, which estimates the probability of a supermarket bag contributing to the littering problem (in land and marine environments).

RESUM

Els masterbatch minerals, en altres paraules, les càrregues minerals, són concentrats de carbonat de calci o talc en una base de polímer. A més dels seus avantatges econòmics, el seu ús millora les propietats mecàniques dels materials i redueix el consum d'energia durant la seva fabricació. Finalment, la seva utilització com a matèria primera per a la fabricació de productes pot ajudar a reduir la petjada de carboni dels mateixos. Això és així perquè inclouen (en diferents percentatges) minerals naturals (carbonat de calci i talc) en lloc de petroquímics verges.

A causa de la creixent contaminació ambiental a la qual el món s'enfronta recentment, també ha augmentat la preocupació per l'ús del plàstic. L'envàs de plàstic, origen del 30% de la producció total anual de plàstic, és una de les àrees principals que requereix atenció especial. Per tant, l'enfocament principal d'aquesta tesi és l'avaluació ambiental de l'ús de masterbatch, amb carbonat de calci i talc, en diferents aplicacions d'envasos de plàstic mitjançant l'ús de la metodologia d'avaluació del cicle de vida (ACV). Aquesta eina és comunament acceptada per estimar els impactes ambientals de productes o sistemes. A part de l'avaluació mediambiental, també es té com a objectiu proporcionar avenços en la metodologia de l'ACV de materials compostos de plàstic. En aquesta tesi, a través d'una col·laboració amb un productor de masterbatch minerals, es van realitzar diferents estudis de cas reals de mercat.

Els resultats d'aquesta tesi són: i) una revisió bibliogràfica profunda i extensa sobre els impactes ambientals de les càrregues funcionals (incloent masterbatch) utilitzats en la indústria; iii) dos estudis de cas d'ACV ("làmines d'emmagatzematge de plàstic contra de fusta d'eucaliptus" i "tubs de cosmètics de polietilè (PE) verge versus de disseny ecològic amb càrregues minerals i contingut reciclat"), en els quals l'ús de plàstics amb càrregues minerals es compara amb l'ús alternatiu de plàstics verges; iii) la introducció de mètodes, dins de la fase d'inventari, per a calcular la pèrdua de qualitat (també coneguda com a factors Qs / Qp) dels materials compostos en cicles obert i tancat de reciclatge; i iv) introducció d'un indicador d'escombraries en ambients terrestre i marí, dins de la fase d'avaluació d'impacte, que estima la probabilitat que una bossa de supermercat contribueixi a aquesta problema.

RESUMEN

Los masterbatches minerales, en otras palabras, las cargas minerales, son concentrados de carbonato de calcio o talco en una base de polímero. Además de sus ventajas económicas, su uso mejora las propiedades mecánicas de los materiales y reduce el consumo de energía durante su fabricación. Finalmente, su utilización como materia prima para la fabricación de productos puede ayudar a reducir la huella de carbono de los mismos. Esto es así porque incluyen (en algunos porcentajes) minerales naturales (carbonato de calcio y talco) en lugar de petroquímicos vírgenes.

Debido a la creciente contaminación ambiental a la que el mundo se enfrenta recientemente, también ha aumentado la preocupación por el uso del plástico. El envase de plástico, origen del 30% de la producción total anual de plástico, es una de las áreas principales que requiere atención especial. Por lo tanto, el enfoque principal de esta tesis es la evaluación ambiental del uso de masterbatches, con carbonato de calcio y talco, en diferentes aplicaciones de envases de plástico mediante el uso de la metodología de evaluación del ciclo de vida (ACV). Esta herramienta es comúnmente aceptada para estimar los impactos ambientales potenciales de productos o sistemas. A parte de la evaluación medioambiental, también se tiene como objetivo proporcionar avances en la metodología del ACV de materiales compuestos de plástico. En esta tesis, a través de una colaboración con un productor de masterbatches minerales, se realizaron diferentes estudios casos reales de mercado.

Los resultados de esta tesis son: i) una revisión bibliográfica profunda y extensa sobre los impactos ambientales de los cargas funcionales (incluyendo masterbatches) utilizados en la industria; iii) dos estudios de caso de ACV ("láminas de almacenamiento de plástico contra de madera de eucalipto" y "tubos de cosméticos de polietileno (PE) virgen versus de diseño ecológico con cargas minerales y contenido reciclado"), en los cuales el uso de plásticos con cargas minerales se compara conel uso alternativo de plásticos virgenes; iii) la introducción de métodos, dentro de la fase de inventario, para calcular la pérdida de calidad (también conocida como factores Qs / Qp) de los materiales compuestos en ciclos abierto y cerrado de reciclaje; y iv) introducción de un indicador de basura en ambientes terrestre y marino, dentro de la fase de evaluación de impacto, que estima la probabilidad de que una bolsa de supermercado contribuya a esa problema.

Chapter 1. INTRODUCTION

1.1. Background

This thesis is funded by GCR Group through its sub-division Granic, which is located in Tarragona, Spain[1]. The company produces mineral masterbatches with calcium carbonate and talc, called with the trademark Granic, to be used as fillers in plastic applications (see Figure 1.1). The company offers environmentally friendly plastic solutions for thermoplastics manufacturing industry such as film, blow molding, injection, raffia, rope, extrusion coating, sheets, thermoforming, pipes, profiles and foam.

Figure 1.1. Plastic masterbatches (Granic) (GCR Group, 2018)

The environmental commitment of the company is focused on minimizing the carbon footprint of plastic products. The company had already performed life cycle assessment (LCA) of some of their intermediate products, but this time they were interested in knowing the environmental profile of their application in final packaging products, since it is one of the company's biggest market. As a result, this thesis will be focusing on plastics with calcium carbonate and talc as fillers applied to plastic packaging.

In the following sections an introduction to plastic fillers, LCA methodology and LCA studies related to packaging will be presented.

1.1.1. Plastic fillers

Thanks to their optimal cost and high performance, thermoplastics have been used in many kinds of applications during the last few decades; and they have been even replacing other conventional

[1] https://www.gcrgroup.es/en/gcr/home

materials like glass, metal, and wood (DeArmitt, 2011). Because of the increasing demand for thermoplastics, people started to look for ways to reduce their cost. That was the initial reason behind the introduction of fillers to plastics. Primarily, the term "fillers" corresponded to cheap diluents introduced into plastics to reduce the overall cost. However, they were found to be more than this. Recently, the term "functional fillers" is being used, because they can provide other properties in addition to cost reduction (Rothon, 2002). The addition of fillers creates multiphase systems composed of micro/macrostructures giving characteristics to the material (DeArmitt, 2011). Improvement in processing, density, thermal expansion, thermal conductivity, flame retardancy, optical changes, electrical and magnetic properties, and mechanical properties like stiffness are examples of properties that can be changed through the addition of functional fillers to plastics (DeArmitt, 2011; Rothon, 2002).

In 2003, the global demand for fillers in plastics industry was predicted to be 15 million tons and their main markets were transportation and construction, later consumer products like furniture, industry and machinery, electrical appliances and electronics, and packaging were also important markets (Xanthos, 2010). In 2015, the global polymer filler market was more than USD 45 billion (Grand View Research, 2016). According to DeArmitt (2011), carbon black, $CaCO_3$, silica, $Al(OH)_3$, talc and kaolin are the major fillers contributing to a multi-billion euro/year market. Recently, an increased interest in environmental protection has led to using fillers to reduce environmental impacts of products by replacing petrochemical materials (Murphy, 2001).

Any particulate material added to plastics would serve as a filler (DeArmitt, 2011). Polymer composites with fillers are defined as mixtures of polymers with inorganic or organic additives with certain geometries; thus, consist of two or more components and phases (Xanthos, 2010).

Although other groupings are possible, fillers can be grouped into two main categories: inorganic and organic ones. Then, they can be even further subdivided based on their chemical family as shown in Table *1.1*, which includes some commonly known examples, as well. According to the market research performed, in 2015 inorganic fillers were found to lead the filler market with a 78.9% share (Grand View Research, 2016).

Table 1.1. Chemical grouping of fillers (Xanthos, 2010)

Groups	**Examples**
Inorganics	
Oxides	Glass, SiO_2, ZnO, Al_2O_3, Sb_2O_3 and MgO
Hydroxides	$Mg(OH)_2$ and $Al(OH)_3$
Salts	$CaCO_3$, $CaSO_4$, $BaSO_4$, hydrotalcite and phosphates
Silicates	Talc, kaolin, mica, montmorillonite, wollastonite, asbestos and feldspar
Metals	Steel and boron
Organics	
Carbon, graphite	Carbon fibers and nanotubes, carbon black, graphite fibers and flakes
Natural polymers	Cellulose and wood fibers, starch, cotton, sisal and flax,
Synthetic polymers	Polyester, aramid, polyamide and polyvinyl alcohol fibers

Another type of filler grouping based on physical shape would be fibrous fillers, which include both organic and inorganic fillers. Among them the most used one is; glass fiber (GF) and, more recently, natural fibers (NFs). Fibrous fillers can be used to change mechanical properties, electrical and magnetic properties of composites (Xanthos, 2010). In fiber-reinforced composites, the mechanical behavior depends on the type of fiber and on the fiber/matrix bonding interface. However, the higher cost of fibers can be a limiting factor to use them (Sathishkumar and Naveen, 2014).

With the increased interest in environment and sustainability, bio-composites have been developed significantly in the last century (Faruk et al., 2012). Plastics reinforced with NFs like sisal, flax, jute, and wood-fibers have become more and more popular (Xu et al., 2008). In addition to their biodegradability and/or renewable nature, they also offer low cost, low relative density and high specific strength (Faruk et al., 2012). They are even considered as the oldest fillers added to plastic composites (Zah et al., 2007). They have gained importance during the last years, as the replacement of fibrous fillers like glass or carbon, due to their above-mentioned properties (Ku et al., 2011; La Rosa et al., 2013). They have been especially exploited by the European car manufacturers (Holbery and Houston, 2006). In Table 1.2, examples of NFs used as fillers in plastics are given classified in different categories.

Table 1.2. NFs as fillers (Bos, 2004)

Natural Fibers	Examples
Straw Fibers	Wheat, corn, and rice
Bast	Hemp, jute, kenaf, lax
Leaf	Sisal, henequen, pineapple leaf fibers
Seed/fruit	Cotton, coir
Grass Fibers	China reed, bamboo, grass
Wood Fiber	

Ground calcium carbonate (GCC) is easily found on the earth, mostly in the form of limestone and chalk, which are formed from fossils (Maier and Calafut, 1998). With a market share of 34%, GCC is the most commonly used filler on the global market because it is a common and inexpensive material with superior functions like increasing stiffness, impact strength and flexural modulus of the plastic to which it is added. The demand is even expected to increase by 2.7% between 2015 and 2023 (Ceresana, 2016).

Hydrated magnesium silicate, known as talc, provides better rigidity and impact strength to plastics, especially to polypropylene (PP), when it is added. Thanks to the advanced milling technology, higher purity provides thermal stability; therefore, it is a good choice to use it in packaging (Murphy, 2001). Mineral masterbatch, which is the main focus of this thesis, is defined as a concentrate of calcium carbonate or talc in a polymer base. It helps to introduce calcium carbonate and talc into thermoplastics in a suitable way. Since it has good dispersion and optimization of flow, it guarantees easy processing (QY Research PVT LTD, 2018).

1.1.2. Life cycle assessment (LCA)

United Nations Environment Programme (UNEP) defines Life Cycle Assessment (LCA) as a tool used for evaluation of environmental aspects of a product or a system through all its life cycle stages (UNEP, 2003). The International Organization for Standardization (ISO) standardizes LCA methodology within the series starting with ISO 14040 (2006a). ISO defines LCA as “a compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product or system throughout its life cycle” and offers a methodological framework for performing it. According to ISO 14040 (2006a) an LCA shall be performed through the phases of goal and scope definition, life cycle inventory analysis (LCI), life cycle impact assessment (LCIA) and life cycle interpretation. The relation between the life cycle stages and direct applications of the results of LCA or LCI studies are presented in Figure 1.2.

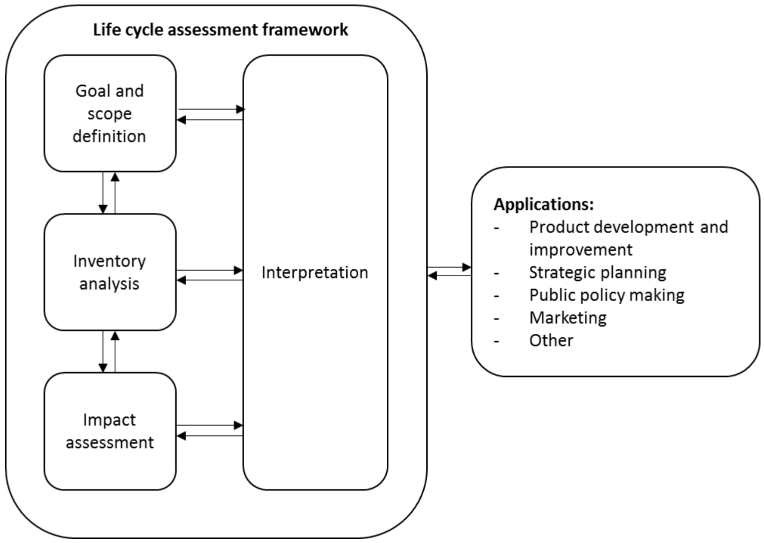


Figure 1.2. Phases of LCA (ISO, 2006a)

In the goal definition phase, the reasons for performing the study and the audience to which the results will be communicated are introduced. Following that, for scope definition, the product or service to be studied is described and agreed. Many other items regarding modeling of the system are also defined in this phase (ISO, 2006a), like: function, functional unit, system boundaries, allocation procedures, environmental impact categories selected, required data, assumptions and limitations.

In the inventory analysis phase, the system model, which is a flow model of a technical system with defined system boundaries, is built based on the goal and scope definition. According to Baumann and Tillman (2004), the activities of the inventory analysis phase include: 1) development of flowchart of the flow model, showing the activities and flows included in the system boundaries; 2) data collection, all the inputs and outputs (raw materials, energy, products and waste and emissions to the environment); and 3) calculation of the resource use and environmental emissions in relation to the functional unit.

Later, in the impact assessment phase, the environmental loads identified in the LCI are turned into environmentally relevant impact information elements. The first one is the classification, which is the assignment of inventory results to the different types of environmental impacts. The next step is characterization, in which the relative contributions of the emissions to impact categories are defined. If no impact assessment is carried out, the study is named as LCI. At the end of an LCA, sometimes the results are needed to be further interpreted or aggregated. For this, commonly accepted weighting methods, normalization or grouping may be used. For LCIA, classification and characterization are the mandatory steps, while weighing, normalization and grouping are optional (Baumann and Tillman, 2004), and weighting is usually avoided, as “there is no scientific basis for reducing LCA results to a single overall score or number, since weighting requires value choices” (ISO, 2006a).

Finally, in the interpretation phase, the results of LCI or LCA are presented in a way that it is consistent with the defined goal and scope. In this part, the conclusions are drawn by explaining the limitations and providing further recommendations (ISO, 2006a), According to ISO (2006a), the interpretation of results should state that the LCIA estimates potential environmental impacts, but not the actual impacts.

1.1.3. LCA of materials in packaging

As materials have a significant role in defining the environmental performance of services and products, special attention must be given to choosing correct materials for a specific function, in order to assure sustainability (Civancik-Uslu et al., 2018). In the literature, there are examples of studies where LCA is used to choose environmentally better materials (Broeren et al., 2016; Calado et al., 2018; Chang et al., 2018; Gazulla et al., 2008).

For example, a debate has been undergoing in the pallet industry for years regarding which type of pallet is better: wood or plastic? The answer to that question was to consider the whole life cycle of the pallet, from cradle-to-grave (Lacefield, 2008). An example of this is the LCA study of pallet alternatives by Bengtsson and Logie (2015). In the study, environmental impacts of plastic, cardboard, softwood and hardwood pallet alternatives were evaluated; for one-way use or pooled and manufactured in Australia or China. Results showed that pooled hardwood and softwood pallets were better than one-way and plastic pooled ones. The main reason for that is the energy needed to produce plastic pallets. In another study, the impact of packaging alternatives for fruit and vegetable transportation in Europe was studied (Albrecht et al., 2013). Single-use wooden and cardboard boxes were compared with multi-use plastic ones. One-way cardboard boxes were found to have the highest environmental impacts. On the other hand, one-way wooden and multi-use plastic ones produced similar results for global warming, acidification, and photochemical ozone creation, while plastic ones had better results in eutrophication and abiotic resource depletion categories.

In the literature, there are also LCA studies of composite materials. Sommerhuber et al. (2017) performed an LCA of wood-plastic composites made from virgin materials versus secondary materials (recycled), in addition to find the best end-of-life (EoL) scenario for the composite material. Wood-plastic composites made from secondary materials were found more environmentally friendly and recycling was the best EoL option.

Based on the above examples and many others in other sectors, there cannot be a simple answer to which material is environmental better; however, LCA may indeed be the most adequate tool to compare different material possibilities for a specific function.

The packaging industry looks for packaging solutions that are strong, hygienic, easy to handle, lightweight, and, most importantly, sustainable (Varun et al., 2016). The increased concern for the environment has attracted attention to the environmental burdens of packaging (Flanigan et al., 2013; Herbes et al., 2018). As a result of this, in the recent years, production of packaging in a more sustainable way has become more of an issue, not only to improve the environmental performance of products but also to meet with the requirements of the growing green product market (González-García et al., 2016).

There are many studies in the literature in which LCA is used to identify the environmental profile of packaging (Albrecht et al., 2013; Balaguera et al., 2019; Civancik-Uslu et al., 2019; Cleary, 2013). LCA is also used to help policy-makers to understand the environmental impacts of packaging (Lewis et al., 2010). De Koeijer et al. (2017) defines LCA as the major tool to evaluate the environmental profile of a product throughout its life cycle. Luz et al. (2018) offered a methodological approach to integrate LCA in product development and applied it to a softener packaging in their study. Some of them stress that the production stage of packaging is the major contributor to its environmental impacts (Navarro et al., 2018; Raugei et al., 2009; Roy et al., 2009).

It is commonly known that most manufacturing processes and products have negative impacts on the environment (Alves et al., 2009)and it is very important for companies to be aware of the environmental impacts of their processes and materials in order to improve their environmental strategy (Cinar, 2005). Materials normally have a significant part of the environmental impacts of a product. Therefore, to provide sustainability, special attention must be paid to choosing them when designing it. To this end, the search of alternative materials has increased to comply with the trending sustainability goals. However, does their use always promise environmental benefits?

For example, some trending natural fibers (NFs) applied to plastic composites have recently become very common, even replacing the use of inorganic fillers like glass fiber (GF) and talc in automotive or aviation applications (Alves et al., 2010; Boland et al., 2015; Luz et al., 2010; Scelsi et al., 2011). However, using NFs in composites may not guarantee that the material is more environmentally friendly. A thorough LCA of the resulting composite should be performed in order to face this uncertainty. To this end, LCA is defined as a useful tool to assess environmental impacts of newly developed materials throughout their life cycle (Wang et al., 2012; Xu et al., 2008). However, other more simplified applications of LCA may also be acceptable (Bala et al., 2010) and, even a life cycle perspective might be useful when quantification is difficult (Fullana-i-Palmer et al., 2011; Theng et al., 2017).

Sometimes, applying LCA to newly developed materials can be challenging, as performed by Theng et al. (2017) for a fiberboard made from corn stalk and kraft lignin as a green adhesive. One could face some difficulties; especially if the material is still in research and development or in pilot scale. Lack of data about process parameters, materials formulation, and material properties, etc., may result in some uncertainties when developing the model. In those cases, a preliminary LCA with the available data could be performed and further developed when more or better data are available (Hesser, 2015). According to ISO 14044 (2006b) "The depth and the breadth of LCA can differ considerably depending on the goal of a particular LCA". For instance, Delgado-Aguilar et al. (2015) mentioned in their study on cellulose nanofibers that they did not perform a fully-fledged LCA, but only a life cycle approach was used to find out from which stages the main impacts were coming from.

1.2. Research rationale

Thanks to their reasonable price and performance, plastics have been used very commonly in a wide range of applications like packaging, building & construction, automotive, electrical & electronic, household, leisure, sports, and agriculture and form a significant part of our lives. According to

Plastics Europe, in 2016, 335 million tons of plastic (including thermoplastics, polyurethanes and other plastics like thermosets, adhesives, coating and sealing) were produced in the world, and 60 million tons of those were in Europe (PlasticsEurope, 2017). Around 30% of the total annual plastic production is used for packaging purposes (UN Environment, 2018). In Spain, 47.93% of plastics (1,418.487 MT) were used for domestic, industrial and commercial packaging in 2014 (Cicloplast, 2014).

Due to the increasing environmental pollution that the world is facing, the concerns over the use of plastics have raised; governments are creating laws to restrict and control their use; and companies care much more about their environmental image. Plastic packaging is perceived as polluting by the public because of the large volume of solid waste produced, energy and material consumed, and emissions to different environmental compartments (Sonneveld, 2000). Recently, they have been said to be the major source for marine littering and, due to the continuous market growth, the risk of plastics reaching to the marine environment is increasing (Law, 2017).

Mineral fillers are commonly used in the plastics industry for different applications. As stated in section 1.1.1, the use of mineral fillers provides additional properties to the materials in which they are mixed with. The use of plastics with mineral fillers instead of virgin plastics may help to reduce environmental impacts of a product, since they naturally exist on the earth and replace virgin petrochemicals at some portions; therefore, they may propose a partial solution to the increasing concerns about the use of plastics.

Being this said, the focus of this thesis is to investigate, with the help of the LCA methodology, the environmental savings which can be achieved by using plastics with mineral fillers, specifically calcium carbonate and talc, as they are the most commonly used ones, in packaging applications.

Two major issues in LCA methodology will be also discussed in the thesis:

The first issue to solve is how to make the allocation of burdens and credits for solving multifunctionality in the case of compound materials both in an open-loop and a closed-loop recycling. There is an ongoing discussion on how to allocate the burdens and credits of recycling processes between different stages of product cascade systems (Figure *1.3*).The problem is to distribute burdens from the production of virgin materials and recycling, and the credits from the avoided production of virgin material to all the products within the cascade system. This distribution of burdens and credits must represent, as much as possible, the physical reality and avoid double counting of burdens or over-crediting of the systems. According to ISO 14044 (ISO, 2006b), system expansion is preferred over allocation, whenever possible, but in product-cascade systems where the information of one single product is needed (ie., for ecolabeling purposes), allocation has to be used.

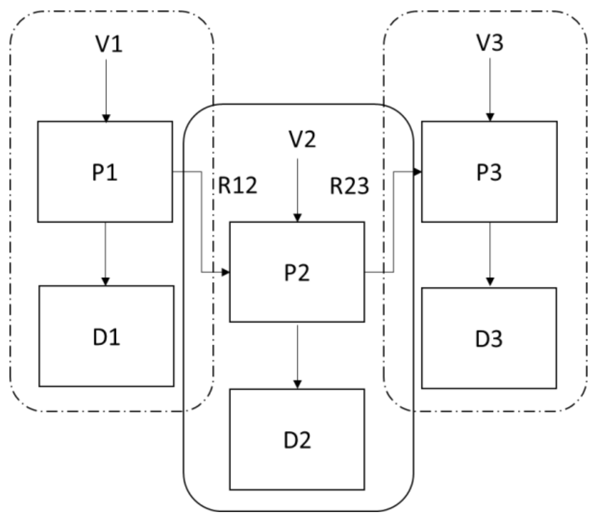


Figure 1.3 Product cascade system (Pi: products production processes; Di: disposal processes; Vi: virgin materials; R(i,i+1): recycled material between consecutive life cycles; and i: number of the loop in the life cycle of the material)

While there is an ongoing discussion on how to perform EoL allocation for one of the products in a product cascade system (100:0 approach; 100:100; etc.), within some of these methods, another question is how to calculate the credits that should be given to a product due to its recyclability, as the recycled material loses some of its properties during the recycling process (downcycling). Although there are many studies discussing and applying different EoL allocation methods, it is rare to find literature to normalize the calculation of quality loss of the recycled material despite of the fact that the necessity to calculate Qs/Qp quality factors (Qs, quality of the secondary material; Qp, quality of the primary material) was somehow addressed during the Product Environmental Footprint (PEF)/ Organizational Environmental Footprint (OEF) pilot projects of the EU Single Market of Green Products Initiative (EC, 2018).

The second issue is the development of a quantitative model which may help to integrate marine littering impact in LCA results. The Medellin declaration on marine litter in LCA and Management acknowledged that impacts related to marine debris, plastics and macroplastics are not properly addressed in LCA, despite of the fact that LCA is one of the most commonly used tools for environmental assessment (Sonnemann and Valdivia, 2017). Very recently, in an international workshop on marine littering, it was agreed that addressing marine littering within LCA methodology would be meaningful and feasible; however, the methodology needs to be further developed (Strothmann et al., 2018).

1.3. Research objectives

The specific research objectives of the thesis are:

1. To make a literature research on the environmental impacts of plastics with functional fillers, focusing on calcium carbonate and talc as fillers, to understand if their use can bring environmental advantages to plastic products. Also, to review the state of the art of LCA applied to plastics with fillers and identify the gaps in the literature.
2. To identify packaging products where plastics with mineral fillers can be of technical and economic interest and to perform real market based LCA studies comparing this new proposed material with the conventional ones, in order to quantify the environmental improvement, if any.
3. To improve the Life Cycle Inventory Analysis methodology by discussing how to credit the system when recycling in the case of plastic compounds, tackling issues such as end-of-life allocation and quality loss during recycling.
4. To improve the Life Cycle Impact Assessment methodology by defining a new impact indicator related to littering to be used in LCA studies, with the help of a case study on carrier bags.

1.4. Structure of the thesis

The research objectives of this thesis, given in the previous section 1.2, will be achieved through the present document as follows:

In chapter 2, an extended literature review on LCA of functional fillers is presented. As not many studies were available in the literature, the literature research has been performed for both inorganic and organic fillers, although the focus of this thesis is on mineral fillers.

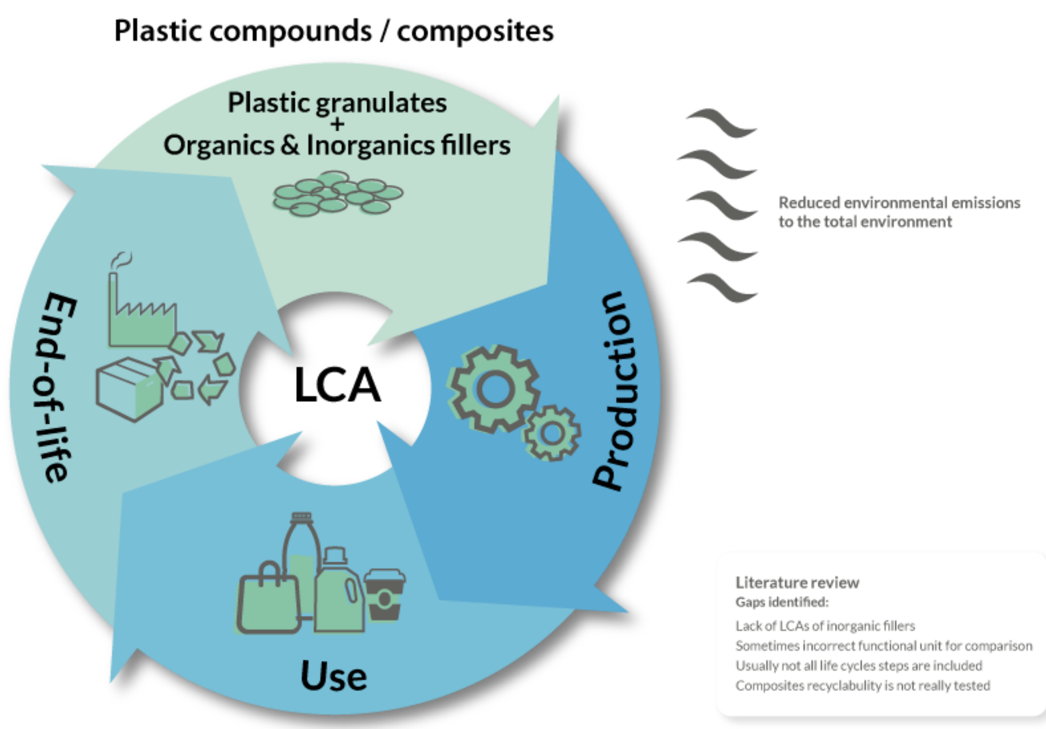
In chapter 3, an LCA methodological discussion on End-of-Life (EoL) allocation and definition of the quality factors for crediting the system in open-loop and closed-loop recycling is done, through a real case study comparing the use of plastic compound sheets with eucalyptus wood sheets as secondary packaging during storage at the participating company.

In chapter 4, an eco-design strategy is used to produce a cosmetic packaging with less environmental impact. The environmental profile of the eco-designed tube is compared to the original one by using LCA methodology.

In chapter 5, a quantitative impact indicator, estimating the risk of a carrier bag becoming marine litter, is introduced based on an LCA study comparing the environmental impacts of carrier bags made of different materials.

This thesis is formed out of four major parts: chapter 2 is the literature review, chapter 3 provides advancements in LCI, chapter 4 is an eco-design case study and chapter 5 provides advancements in LCIA. It offers a full cover of the LCA methodology, with an in-depth literature review and market case studies.

Chapter 2. ARE FUNCTIONAL FILLERS IMPROVING ENVIRONMENTAL BEHAVIOR OF PLASTICS? A REVIEW ON LCA STUDIES



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Abstract

The use of functional fillers can be advantageous in terms of cost reduction and improved properties in plastics. There are many types of fillers used in industry, organic and inorganic, with a wide application area. As a response to the growing concerns about environmental damage that plastics cause, recently fillers have started to be considered as a way to reduce it by decreasing the need for petrochemical resources. Life cycle assessment (LCA) is identified as a proper tool to evaluate potential environmental impacts of products or systems. Therefore, in this chapter, the literature regarding LCA of plastics with functional fillers was reviewed in order to see if the use of fillers in plastics could be environmentally helpful. It was interesting to find out that environmental impacts of functional fillers in plastics had not been studied too often, especially in the case of inorganic fillers. Therefore, a gap in the literature was identified for the future works. Results of the study showed that, although there were not many and some differences exist among the LCA studies, the use of fillers in plastics industry may help to reduce environmental emissions. In addition, how LCA methodology was applied to these materials was also investigated.

2.1. Methodology

Special interest was given to the environmental impacts of functional fillers used in polymer composites/compounds; both inorganic and organic fillers. In the studies considered, the final product (polymer composite/compound with filler) and LCA methodology used were investigated. The following sections were structured based on the final product and for each group, LCA studies reviewed were summarized. When possible, some numeric values were presented in the related sections. Global warming potential (GWP) was chosen as impact indicator, because it is one of the most known and used impact categories and it was difficult to compare impacts using other impact categories due to the different methodologies and units used.

In the first step of the study, environmental advantages of fillers compared to conventional materials (like virgin plastics and steel) were summarized, by reviewing different LCA studies. In the second part, different fillers, which can be used instead of a very common one (like GF, calcium carbonate or talc) were compared through existing LCA studies. In the study, 19 LCA studies comparing different alternative materials were reviewed in detail.

After collecting the related references from the literature, results were examined from two different perspectives. The first one was to identify how environmental impacts were affected with the use of functional fillers in plastic composites, as a common message about these materials is their supposed "greener" nature, and we wanted to confirm it. And the second one was to see how LCA was being approached by reviewing methodological issues, like boundary conditions, inventory development, methodologies for environmental impact assessment, and EoL scenarios.

2.2. Results and discussion

Although plastics with functional fillers have been in use for many years in the industry, their environmental impacts have been seldom investigated. There is only small amount of studies available in the literature; therefore, in this part, they were more deeply investigated in terms of both their goal and scope and methodological issues. 19 LCA studies were reviewed in the study. However, since some of them fall into different groups at the same time, they were counted in two different groupings. Distribution of the studies based on the material type is presented in Figure 2.1 Nearly half of the studies reviewed (43%) was comparing the use of NFs against GFs, followed by the second largest group of studies, which were LCA studies comparing the use of fillers against virgin plastics formed (24%). Following that, studies considering the use of talc against organic and inorganic fillers were studied (19%). Finally, the minority of LCA studies reviewed was considering the comparison of use of fillers against conventional materials like steel and aluminum (14%).

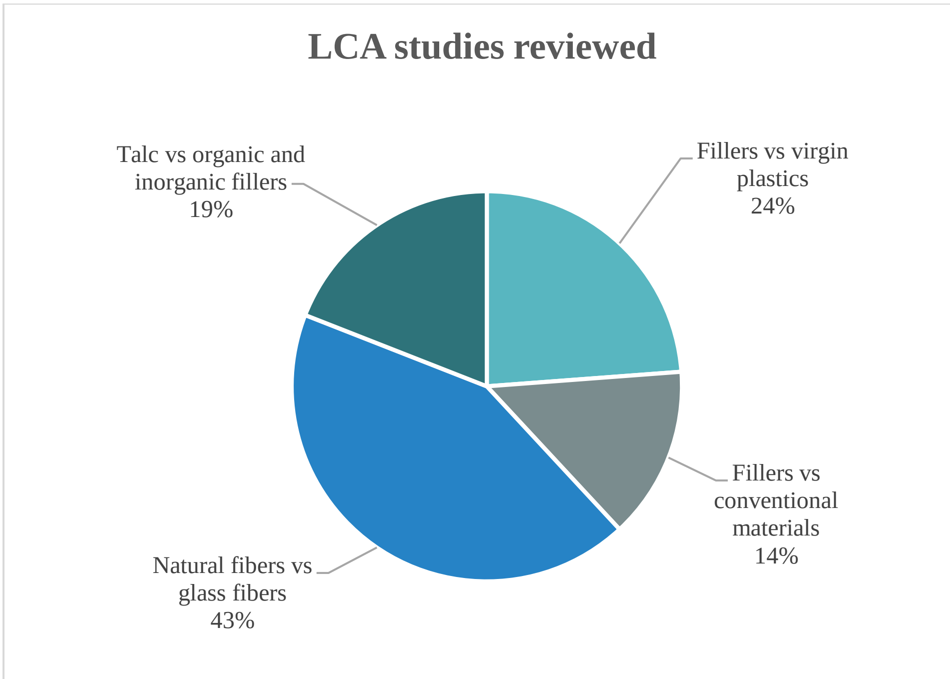


Figure 2.1: Distribution of LCA case studies reviewed

Three review studies regarding plastics with fillers were found in the literature before the present one; however, they were quite different in terms of their goal and scope (only one type of filler or no environmental impact included). La Mantia et al. (2011) made a review study limited to polymer-based materials filled with only natural-organic fillers which are renewable and biodegradable, including some environmental sustainability and impacts information. Weiss et al. (2012) made a review on the environmental impacts of bio-based materials like wood, paper, textile, rubber, insulation materials, and composites based on bio-plastics, which are actually out of the scope of this study. A more recent review was done by Thakur et al. (2014) regarding the use of raw NF-based polymer composites with a specific focus on their mechanical properties. Environmental advantages of using them were mentioned; however, not very deeply. To conclude, according to the research that was performed, there are no reviews of LCA studies of plastics with functional fillers yet, especially of inorganic ones.

2.2.1. Plastics with fillers vs virgin materials

2.2.1.1. Fillers vs virgin plastics

Although not many, there are some studies in the literature, in which LCA methodology was used with the purpose of identifying how the addition of functional fillers affects the environmental impacts of plastics in different applications (Roes et al., 2007; Vidal et al., 2009; Xu et al., 2008).

In one of them, environmental advantage of using PP-silicate nanocomposite was investigated on three different applications: PP based packaging film, polyethylene (PE) based agricultural film and GF reinforced PP based automotive panels (Roes et al., 2007). In each application, some portion of the base materials was replaced by a PP nanocomposite, resulting in the use of less material while guaranteeing the same functionality. Even though higher changes in environmental impacts were expected, in some cases, smaller changes were observed as it can be seen in Table 2.1. The reason for smaller changes in the results could be because of the small percentages of silicates in the polymers and the uncertainties in the nanoclay production process. According to the authors, effects of nanoclay production process must be less than expected. Nevertheless, in the case of agricultural film, results of the study showed some clear environmental benefits in GWP and all other impact categories evaluated, as well as some economic advantages; mainly due to higher weight reduction (36.5%) in agricultural film application than the other applications.

In another study, the changes in environmental impacts of virgin petrochemicals with the addition of NFs as fillers were investigated (Vidal et al., 2009). Three new different composites; PP + cotton liners, PP + rice husks, and high-density polyethylene (HDPE) + cotton liners were studied by comparing them with the corresponding virgin petrochemicals; PP or HDPE. According to the LCA results, composites showed better environmental impacts in terms of all impact categories, except eutrophication for the rice husk because of the fertilizers used for the cultivation of rice. Results for GWP are presented in Table 2.1 (Vidal et al., 2009).

Fillers may also be used as reinforcements to produce composites based on recycled thermoplastics with reduced environmental impacts. Al-Ma'adeed et al. (2011) studied composites formed by recycled PP and polyethylene (PE) with talc and GF to see how the environmental impacts change when fillers were used as reinforcement in recycled thermoplastics. When the environmental impacts of the recycled PP and PE composites with fillers were compared with virgin polymers by using LCA methodology, it was concluded that the former had lower GWP, except in the case of recycled PE with talc. For example, for the functional unit (FU) of 1 kg of material, GWP of virgin PP was estimated around 2.1 kg CO2 eq., while it was around 0.12 kg CO2 eq. for recycled PP. In addition, talc and GF were added to the recycled PP, and the results were around 0.75 kg CO2 eq. and 0.09 kg CO2 eq., respectively. In a similar way, GWP of virgin low-density polyethylene (LDPE) and recycled LDPE was found around 2.18 and 0.6 kg CO2 eq. And the results were around 3.55 kg and 2.15 kg CO2 eq., respectively when talc and GF were added. However, it is important to note that those results were read from the graphs in the study, thus they must be treated as approximate values.

The LCA studies were also reviewed in terms of the type of filler used, functional unit, boundary conditions, software and impact categories used for the calculation of environmental impacts, inventory development, EoL scenario and sensitivity analyses (see Table 2.2). As it can be seen from the table, functional unit was defined usually as 1 kg of polymer or a specific application like film or sheet. Cradle-to-grave LCA studies including different EoL scenarios like landfilling and incineration were common. GaBi and SimaPro were found to be commonly used (only one study from 1999 using Umberto software was found). Inventory data was collected from LCA databases, primary data from the producers (private sector) and other national or international databases.

Based on the studies reviewed, although the types of fillers used in each case were different and number of the studies considered were not that high, it was observed that the higher the proportion of functional fillers, the lower the environmental impacts.; just in parallel to what is said in a study by Xu et al. (2008). They studied wood-fiber-reinforced PP composites for different fiber contents: 10%, 30% and 50% by mass (Xu et al., 2008). Results clearly indicated that the addition of wood fibers reduced the environmental impacts proportionally due to the fact that an increasing number of virgin petrochemicals used was replaced with NFs. In the same way, Michaud et al. (2009) agreed that the reductions in environmental impacts from the life cycle of the product were parallel to the amount of wood fibers added to the HDPE/wood flour composite.

Table 2.1. Global warming potential (GWP) of the investigated studies

Reference	Methodology	Polymer	GWP (kg CO_2-eq)	
			Conventional	**Nanocomposite**
(Roes et al., 2007)	CML 2001	PP + silicate in packaging film	15.9	15.7
		PE + silicate in agricultural film	9242	5642
		GF + PP in automotive industry	569	570
(Vidal et al., 2009)	GWP [79]	HDPE + cotton	1.88	0.61
		PP + cotton	1.99	0.70
		PP + husks	1.99	0.71

Table 2.2. Methodological analysis of LCA studies (Fillers vs virgin plastics)

Reference	Polymer	Functional unit (FU)	System boundaries/ Software	Impact assessment	Inventory sources	End of life	Data quality assurance
(Al-Ma'adeed et al., 2011)	Recycled PP and PE filled with talc and GF	"1 kg of material"	Cradle-to-grave/GaBi	CML 2001	Ecoinvent and Buwal 250 (with some modifications for Qatar)	PE and PP were recycled, and, in both cases, landfill was assumed to be disposal method	-
(Roes et al., 2007)	PP packaging film PE agricultural film GF + PP in automotive industry	FU of packaging film: "amount of packaging film needed for 1000 bags of 200 g 'Fruitfante'' candies produced by Schuttelaar B.V. (Wad- dinxsveen, The Netherlands)"	Cradle-to-grave/SimaPro	CML 2001 NREU	PP from APME Eco profile Production of Nano clay and nanocomposite by the Institute for	Incineration with energy recovery	-

Reference	Polymer	Functional unit (FU)	System boundaries/ Software	Impact assessment	Inventory sources	End of life	Data quality assurance
		FU of agricultural film: "the amount of plastic film needed to cover a standard tomato greenhouse with a volume of approximately 650 m^3" FU of automotive panel: "Body panels of a low-weight family car that runs 150000 km during its entire lifetime"			Polymer Research (IPF, Dresden, Germany) Pilot plant data Energy Efficiency Office SimaPro database		
(Wötzel et al., 1999)	Hemp fiber composite vs ABS in automotive industry	"a side panel of the AUDI A3"	Cradle-to-grave/Umberto	Eco-indicator 95	Hemp production is representative of Central Europe Industrial Data for Audi A3.ABS injection molding from Association of Plastic Manufactures Europe	Recycling	SA* on production planning and cultivation scenarios
(Xu et al., 2008)	Wood-fiber reinforced PP	"three-layered structure of 2-mm sheets (127mm×127mm) i.e. one layer of fiber sandwiched between two sheets of PP"	Cradle-to-gate/SimaPro	Eco-indicator 99	Australian LCA database	Out of system boundaries	-
(Vidal et al., 2009)	Recycled PP and PE filled with cotton and rice husk	"1 kg of material"	Cradle-to-grave/SimaPro v7	GWP [79] NED [80] AP [81] EP [82]	Private companies in Spain Plastics Europe Several databases	Landfilling and incineration	-

** SA: Sensitivity Analysis*

2.2.1.2. Fillers vs other conventional materials

Carbon fiber (CF) reinforced polymers were found to be studied more often than conventional materials. CF reinforced polymers are considered to be important due to the weight reduction requirements in the automotive industry because they provide weight reduction while keeping the same strength and stiffness provided by steel (Das, 2011). However, the production of CF results fifteen times higher CO2 emissions than steel (Murphy, 2008). For this reason, greenhouse gas (GHG) emissions from the vehicle production stage may increase when steel is replaced with CF reinforced polymer due to the increase in fossil fuel consumption (Timmis et al., 2015). On the other hand, some benefits may be gained from fuel consumption during the use stage thanks to its lighter weight and, globally, lower GHG emissions can be achieved (Kelly et al., 2015).

Partial results of an LCA study by Das (2011) on CF reinforced polymer are presented in Table 2.3, together with its methodological analysis in Table 2.4. As it can be seen from Table 2.3, although the lignin-based CF has higher CO2 emission for the stages of raw material and manufacturing, it has less emission throughout its whole life cycle when compared with stamped steel's life cycle (Das, 2011). In addition to this, in a review study done by harmonizing 43 LCA results of light-weighted automobiles, it was concluded that the replacement of conventional materials like steel and iron with CF reinforced polymer reduces the GHG emissions of the vehicle and will be more likely to be used in the future in automotive industry (Kim and Wallington, 2013a).

CF reinforced polymer has been perceived as a 'next generation' composite material in aircraft due to its reduced weight in comparison to aluminum (Timmis et al., 2015). In one of the studies, the environmental impacts of aluminum alloy 2024, CF reinforced polymer and Glass fiber/Al (GLARE) used in aerospace panels were investigated with the help of LCA methodology (see Table 2.4). The results showed that despite the high energy requirement for their production and difficulties in disposal, the use of composites which are CF reinforced polymer and GLARE provides better environmental results thanks to the savings of fuel consumption due to the reduced weight (Scelsi et al., 2011). In a very similar way, an LCA was performed to compare the use of CF reinforced polymer with conventional aluminum structure in a Boeing-787 Dreamliner plane (see Table 2.4). It was observed that reductions in CO2 and NOx emissions were gained due to less fuel consumption by the plane thanks to its reduced weight. On an aircraft base, 20% reduction in CO2 emissions were gained (Timmis et al., 2015).

Under this section, the methodologies of LCA studies were reviewed in the same way that it was done in the previous section (see Table 2.4). The number of studies reviewed was even fewer than the previous section and all of them were in the area of the transportation industry, since CF reinforced polymer is a promising composite material thanks to its lightweight. Although the 3 LCA studies are very few to make a general conclusion, we would like to point out the potential of the use of CF reinforced polymer to reduce environmental impacts against conventional materials like aluminum or steel mainly because of its light-weight. Based on the LCA studies reviewed under this section, it is also important to note that making a cradle-to-grave assessment is crucial when comparing alternative materials in order not to miss any environmental advantages or disadvantages created in any life cycle stage of the product of concern.

To be commercially competitive on the market, composites must be technically and economically feasible, as well as being greener (Jiménez et al., 2016). Despite the fact that there are many studies in the literature investigating mechanical properties of plastic composites with fillers, few studies could be found focusing on the change in environmental impacts when conventional materials are replaced with fillers. However, according to the LCA studies available in the literature, despite the differences in the studied systems, the use of plastics with fillers as a replacement of conventional materials looks like promising to reduce environmental impacts of a product through its life cycle.

Table 2.3. GHG emissions for materials (Das, 2011)

Material/technology	**CO_2 emissions (kg CO_2 eq.)**	**Comments**
Per kg of manufactured part		Emissions for only material and production stages
Lignin CF P4 part	12.5	The part is produced by lignin and CF
Stamped steel part	4.4	The part is produced by stamped steel
Life cycle of part		Emissions for total life cycle stages; material, production, use and EoL
Life cycle lignin CF P4	1.338	The part is produced by lignin and CF
Life cycle stamped steel	1.478	The part is produced by stamped steel

Table 2.4: Methodological analysis of LCA studies (Fillers vs other conventional materials)

Reference	Polymer	Functional unit (FU)	System boundaries/Software	Impact assessment	Inventory sources	End of life	Data quality assurance
(Das, 2011)	CF reinforced polymer with four different precursors (for each precursor there are two possible production technologies) and production technology vs conventional steel	"The floor pan for a large rear wheel drive vehicle such as the Cadillac CTS under consideration by the United States Automotive Materials Partnership Multi-Material Vehicle (MMV)"	Cradle-to-grave/SimaPro	Ecoinvent version 1.01 and expanded by Pre-Consultants IPCC for GWP_{100}	SimaPro/ Ecoinvent databases GREET model	Recycling (in addition to conventional recycling system for steel, there is thermal treatment for the separation of CF reinforced polymer)	SA* on content of fiber SA* on energy requirement for lignin production
(Scelsi et al., 2011)	CF reinforced polymer vs aluminum based (GF/Al laminate) GLARE	"an aerospace panel"	Cradle-to-grave/SimaPro 7.1	Eco-indicator 99 (E) V2.05	Ecoinvent v2.0	Landfill is assumed (Due to the lack of data)	-
(Timmis et al., 2015)	CF reinforced polymer vs aluminum-based structure in aircraft	"section 46 of Boeing 787 fuselage" "Boeing 787 airframe consists of several one-piece CF reinforced polymer tube sections. Section 46 is the one of these tube sections"	Cradle-to-grave/SimaPro 7.2	Eco-indicator 99 (E) V2.05	Ecoinvent	Landfill (Due to lack of data)	-

2.2.2. One filler vs other filler in plastic application

This research has led to the finding that the comparative LCAs among different kinds of fillers applied to plastics is a more commonly studied topic. In fact, most of the environmental assessment studies of plastic composites with fillers are based on the comparison of the use of different filler alternatives with the aim of looking for a better composite material in terms of environmental, mechanical and physical properties. Therefore, the following sections were formed based on their availability in the literature. For example, environmental comparison of NFs against GFs in different applications by using LCA, especially in automotive industry, was identified as the most commonly studied topic. Therefore, this was defined as one sub-section. Later on, LCA studies comparing environmental impacts of the use of talc as an alternative to both inorganic and organic fillers in the literature were grouped as one-section; because it is a widely used filler and there were relatively more LCA studies available. Finally, information on $CaCO_3$; as being one of the most commonly available and used fillers by the industry, was given as one separate part, since there were no LCA studies available on it. Nevertheless, there are many studies regarding mechanical properties of use of $CaCO_3$.

2.2.2.1. Natural fibers (NFs) vs glass fibers (GFs)

Recently, there has been a growing interest in the use of NFs composites, due to the fact that they may be advantageous in terms of cost and environmental emissions and applicable to many sectors (Pickering et al., 2016). They can be used in the building and construction industry, for the production of door and window frames, decking materials, and furniture parts; and also in the automotive industry for the production of doors, seats, dashboards and many other applications (Xu et al., 2008). In a recent study by Jimenez et al. (2016), mechanical properties of starch-based biodegradable polymer reinforced with sugarcane bagasse were investigated. It was found out that 30% in weight of NFs added to the plastics provided more than 50% of the strength of the whole composite.

There are many types of NFs applicable to plastics composites (Bos, 2004; Joshi et al., 2004; Xanthos, 2010). Korol et al. (2016) compared the environmental impacts of different NFs (cotton, jute, and kenaf) applied to PP, by using LCA methodology. Results of the study for climate change midpoint are presented in Figure 2.2. It was found out that, among the PP-based plastic composites with NFs analyzed, cotton fibers were found to have the highest environmental impacts due to the industrial cultivation of cotton at a large scale (Czaplicka-Kolarz et al., 2013; Korol et al., 2016). Czaplicka-Kolarz et al. (2013) also concluded that, according to an LCA study in which cotton, cellulose, jute fiber, kenaf and GFs reinforced PP composites were compared, PP reinforced with cellulose fiber was found to be the one with the lowest environmental impact. Bamboo has also attracted attention to be used as a reinforcement to create more environmentally friendly composite materials, due to the fact that it grows very quickly and has high strength and stiffness (Kinoshota et al., 2008; Ogawa et al., 2010).

In addition, GFs are known to have advantageous properties like strength, flexibility, stiffness, and resistance and have been used in many applications in the form of plastic composites (Sathishkumar and Naveen, 2014). NF composites have been introduced as alternatives to mineral fiber reinforced composites because of their competitive mechanical properties like tensile strength and for being renewable (Espinach et al., 2016).

Comparing the environmental impacts of using GF as fillers in the automotive industry by using LCA, is one of the most commonly studied topics, especially their replacement with NFs (Alves et al., 2010; Boland et al., 2015; Corbière-Nicollier et al., 2001; Hesser, 2015; Joshi et al., 2004; Korol et al., 2016; La Rosa et al., 2013; Roes et al., 2007; Song et al., 2009). According to Joshi et al. (2004), NFs have been considered as alternatives to GF reinforced composites since the 1990s.

Due to the pressure from the fuel economy and the strict emission regulations, recently, car manufacturers are being forced to come up with new technologies or designs which will help them to adapt to these new requirements. Weight reduction is often considered as one of the most important ways to help fuel economy (Dhingra and Das, 2014; Kim and Wallington, 2013a; Penciuc et al., 2016). According to Kim and Wallington's (2013b) study, replacement of conventional materials like steel and iron by lighter alternatives like composites minimizes the GHG emissions during the use phase of the vehicle but increases the emissions from production phase. In some of the LCA studies conducted, it was pointed out that environmental savings from the life cycle of a product can be achieved through weight-lightening (Corbière-Nicollier et al., 2001; Schmidt and Beyer, 1998; Wötzel et al., 1999; Zah et al., 2007). However, Witik et al. (2011) showed that weight reduction may not always bring a better environmental performance. In their study, LCA was performed to environmentally compare light-weight polymer composites with conventional materials like steel or magnesium. But results showed that lighter materials may not always lead to better environmental impacts from the total life cycle of a product, because of the burdens caused by their production stage. Therefore, concluding that pressures from fuel economy and strict emission regulations are not enough to provide sustainability in the transportation sector.

There are several LCA studies showing that, in order to achieve a better environmental emission profile through light-weight design, NFs may be preferred in comparison to GF (Alves et al., 2010; Boland et al., 2015; Corbière-Nicollier et al., 2001; Hansen et al., 2000; Hesser, 2015; Korol et al., 2016; La Rosa et al., 2013; Schmidt and Beyer, 1998; Wang et al., 2012; Zah et al., 2007). In Table 2.5, methodological issues of the LCAs performed were summarized; together with their environmental evaluation in Table 2.6. According to the data collected, it was observed that the use of NFs is advantageous in terms of environmental emissions when compared with GF. In parallel to this, La Mantia et al. (2011), in their review on green composites, also claim that plastics with natural-organic fillers tend to have better environmental results compared to the ones with mineral-inorganic fillers.

However, as it can be seen from Table 2.5, significant differences exist between the investigated LCA studies in terms of the studied systems. For example, there is a variety of NFs and their composition in the composite material differs. Although they tend to have better environmental performance, there are still some unclear points (like transportation and cultivation of NFs) that require attention when making a decision about their use (Alves et al., 2010; Boland et al., 2015; Corbière-Nicollier et al., 2001; Korol et al., 2016; Wötzel et al., 1999). Duflou et al. (2012) also stated that NFs have the potential of reducing environmental impacts by replacing GF composites, reminding that there are still a lot of issues to be investigated concerning both the mechanical and the environmental properties. For example, it is stated that cellulose fibers require more energy than GF during the production process (Boland et al., 2015). Environmental superiority of bio-composites over synthetic fiber composites should be analyzed carefully through LCA, because of the relatively more resource-intensive processing of bio-fibers (Yan et al., 2014). In addition, in the study by Zah et al. (2007) curauá (ananas) fibers were found to be slightly better in terms of environmental emissions; however, it was pointed out that curauá fibers do not have the mechanical properties of GF and thus few recommendations were done to make the NFs stronger. At this point, here comes the issue of functional unit. In their study, two different functional units were considered; 1)“1 kg of an interior car part made of GF composites” and 2) “the complete life cycle of a car was taken as functional unit” (Zah et al., 2007). For the first one, since the interior car part can be used for different purposes, three scenarios were considered; 1) equal stability, 2) equal weight, 3) equal volume. It was found out that climate change impact of the curauá composite is not that different from GF in the case of “equal stability”. However, in the case of “equal weight”, the curauá fiber had slightly better environmental impacts. And since the density of the curauá fiber is lower than GF, for the functional unit of “equal volume”, NFs caused less environmental impact in all impact categories. Therefore, it can be concluded from here that when performing comparative LCA studies for alternative materials, it is very important to choose the right functional unit allowing to make proper comparison depending on the function.

In LCA studies, environmental impact categories used can create some differences in results, as well. For example, renewable raw materials as fillers may be better in terms of fossil energy use and GHG emissions but they may have worse scores in LCA studies in relation to land use, ecotoxicity and eutrophication potential impact categories (Weiss et al., 2012). Therefore, special attention must be paid for the selection of environmental impact categories which are going to be studied.

On the other hand, even though in most of the comparative LCA studies NFs were found to be more environmentally friendly than GF reinforced composites, depending on the application, the use of GF reinforced composites can provide some environmental benefits as well. A good example to this is an LCA study by Taranu et al. (2015) about the application of GF reinforced polymers to timber beam in order to maximize strength. The results showed that, despite the negative influence of fiber reinforced polymers, GF reinforced polymers added to timber are able to reduce the environmental impacts by reducing the amount of timber used.

According to extended review by Joshi et al. (2004) on comparing the LCA studies investigating the environmental impacts of NFs against GF reinforced composites, despite the many existing differences in LCA studies like system boundaries, NF chosen, and so on; NFs are tend to be environmentally better as a result of four main reasons: (1) NF production is more environmentally friendly; (2) since more NFs are needed for the same performance, less amount of base polymer is needed; (3) the light-weight of NFs provides advantages in the use phase; and (4) incineration of NFs provides energy and CO2 credits (Joshi et al., 2004). Nevertheless, the correct choice of the functional unit in LCA studies plays an important role.

Table 2.5. Methodological analysis of LCA studies (NFs vs GFs)

Reference	Polymer	Functional unit (FU)	System boundaries/ Software	Impact assessment	Inventory sources	End of life	Data quality assurance
(Alves et al., 2010)	Jutes fiber/polyester composites vs GF/polyester composites in automotive industry	"frontal bonnet of the buggy" in other words "the engine cover of $0.35m^2$ which achieves the required mechanical and structural performance"	Cradle-to-grave/ SimaPro 7.0	Eco-indicator 99	Private companies Simapro database IDEMAT ECOINVENT Literature and governmental reports specific to Brazil Recycling data based on experiments	Mechanical recycling Incineration Landfill	-
(Boland et al., 2015)	Cellulose fibers Kenaf fibers GF in PP in vehicles	"The automotive component which is a semi-structural con-sole substrate with a fixed volume"	Cradle-to-grave/ GaBi 6	Life cycle energy demand GHG emissions by IPCC 2012	Literature GaBi database EPA database Private companies Ecoinvent database	All components are assumed to be dismantled and shredded	SA* on electricity used to compound the fiber and resin materials together SA on biogenic carbon storage within NFs
(Corbière-Nicollier et al., 2001)	China reed fiber vs GF in PP (transport pallets)	FU: "a standard transport pallet satisfying service requirements (transport of 1000 km per year) for 5 years"	Cradle-to-grave/ No information	Critical Surface-Time method (CST95) CML 92 Eco points Eco-indicator 95	Literature BUWAL database IATE-HYDRAM reports	Disposal Incineration Recycling	SA* on product lifetime, fiber content and transport distance

Reference	Polymer	Functional unit (FU)	System boundaries/ Software	Impact assessment	Inventory sources	End of life	Data quality assurance
(Hesser, 2015)	Kraft pulp fiber reinforced PP vs PP Talcum reinforced PP vs PP GF reinforced PP vs PP	FU1: comp. based on mass; MJ/kg material for NREU and kg CO2e/kg for GWP FU2: comp. based on volume; MJ/m3 material for NREU and kg CO2e/m3 for GWP FU3: comp. based on strength; MJ/panel for NREU and kg CO2e/panel for GWP FU4: comp. based on stiffness; MJ/panel material for NREU and kg CO2e/panel for GWP	Cradle-to-gate/ Non-valid	PAS 2050 for Global warming potential (GWP) Energy Requirement	Literature GEMIS database Site data collection Experimental data (Due to lack of data, simplified LCA is performed)	Out of system boundaries (Offered to be further investigated)	-
(Wötzel et al., 1999)	Hemp fiber composite vs ABS in automotive industry	FU: “a side panel of the Audi A3”	Cradle-to-grave/ Umberto	Eco-indicator 95	Hemp production is representative of Central Europe Industrial Data for Audi A3.ABS injection molding from Association of Plastic Manufacturers Europe	Recycling	SA* on production planning and cultivation scenarios
(Korol et al., 2016)	PP PP+GF PP Cotton fiber PP Jute fiber PP Kenaf fiber	F: “the production of one standard plastic pallet made from PP different composites with different shares and types of filler”	Cradle-to-gate/ SimaPro 8	ReCiPe Midpoint	Ecoinvent 3.1	Out of system boundaries	-

Reference	Polymer	Functional unit (FU)	System boundaries/ Software	Impact assessment	Inventory sources	End of life	Data quality assurance
(La Rosa et al., 2013)	GF /thermoset composite GF -hemp/ thermoset composite	FU: "one elbow fitting used in the seawater cooling pipeline of a Sicilian chemical plant, with an estimated life of 20 years"	Cradle-to-grave/ SimaPro	ReCiPe Midpoint	Primary data for manufacturing process Ecoinvent v2.2 for other parts	Landfilling (thermoset composites cannot be recycled, and incineration is not a valid option in Italy)	-
(Zah et al., 2007)	PP + Curauá fiber vs PP + GF in automotive industry	FU of the first part: "1 kg of an interior car part made of GF composites" FU of the second part: "the complete life cycle of a car was taken as functional unit".	Cradle-to-grave/ No information	CML 2001	Ecoinvent Primary data for the harvesting and processing of ananas fiber (curaua)	Incineration	-
(Ogawa et al., 2010)	Bamboo fiberboard vs GFR polymer	FU: "a 1250-cm^3 in volume self-bonding fiberboard" which corresponds 1 kg of bamboo	Cradle-to-grave/ Non-valid	GWP Energy Consumption	JEMAI-LCA Japan Environmental Management Association for Industry. Manufacturing data from field	GFRP product is assumed to be separated in GF and FBR resolvent	-

Table 2.6. Environmental evaluation of s NF to GF

Reference	**NF type in comparison to GF**	**Is NF found to be environmentally superior to GF?**
(Alves et al., 2010)	Jute fibers	Yes. It was observed that composites including jute fibers (untreated, dried, and bleached/dried) have 5-10% less emissions than GF.
(Boland et al., 2015)	Natural cellulose and kenaf fiber	Yes. Composites with cellulose and kenaf had 10-20% reductions in their GHG emissions compared to GF.
(Corbière-Nicollier et al., 2001)	China reed fiber	Yes. China reed fiber was provided 54% less GHG emissions than GF.
(Hesser, 2015)	Kraft pulp fiber	Yes. Kraft pulp fiber provided GHG reductions around 14-35%.
(Korol et al., 2016)	Kenaf, jute and cotton fiber	Kenaf and jute-based composites had lower impacts. However, cotton based one was found to have highest environmental impacts due its cultivation (Figure 2.2)
(La Rosa et al., 2013)	Hemp fiber added GF	Yes. Results showed that use of hemp fibers in GF composite reduced GWP around 20%.
(Wang et al., 2012)	Kenaf fiber and soy-based resin	Yes. It was concluded that GWP of kenaf is 80% less than GF.
(Zah et al., 2007)	Cruauá fiber	Depends. In the case of same strength, not different than GF. In the case of equal weight, slightly better than GF. Finally, in the case of same volume, less emissions than GF.

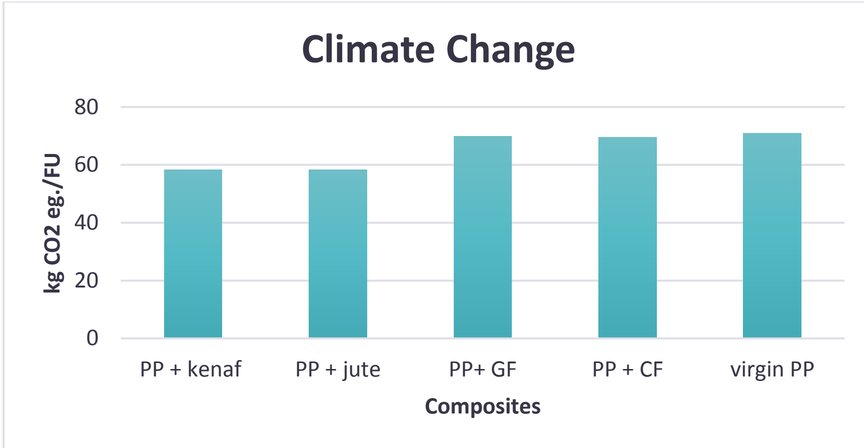


Figure 2.2. Results of climate change midpoint of composite materials for the FU of one plastic pallet (Korol et al., 2016)

2.2.2.2. Talc vs organic and inorganic fillers

Among the inorganic fillers added to plastics, talc is one of the most commonly used fillers in thermoplastics industry due to its high-performance functionalities. In spite of the fact that plastics filled with talc show high flexural modulus, impact resistance, Young's modulus, and yield strength, during the last decades, organic fillers have started to be seen as competitors of talc for some applications because of their better properties such as low density and biodegradability (Premalal et al., 2002). Since talc is defined as non-renewable natural resource and contributing to abiotic depletion (Caraschi and Leão, 2002), it is possible to find some literature looking for alternative materials (e.g. hollow glass microspheres, kraft pulp fiber and sugarcane bagasse fiber) as a replacement of talc (Delogu et al., 2016; Hesser, 2015; Luz et al., 2010; Munoz et al., 2006). For example, Luz et al. (2010) found that a bagasse-PP composite is environmentally superior to a talc-PP composite for automotive applications, despite of the similar mechanical properties (Table 2.7).

It is also possible to find some LCA studies investigating the environmental advantages of other inorganic fillers applied to plastics, like GF. Al-Ma'adeed et al. (2011) compared plastic filled with talc and with GF reinforced composites. The results of the LCA conducted showed that among the analyzed composite materials with 15% of filler (GF and talc) and 85% of virgin thermoplastics (PP and PE), talc was found to have higher environmental impacts than GF (Al-Ma'adeed et al., 2011).

In a recent study by Delogu et al. (2016), with the aim of designing a light-weight automotive component manufactured by Magneti Marelli, standard talc reinforced PP was replaced with an innovative hollow glass micro-sphere reinforced PP composite. PP reinforced with 23% of hollow glass microspheres was analyzed vs a PP reinforced with 25% of talc through LCA, together with their mechanical properties. The composite with talc had better flexural modulus, tensile strength, flexural strength and Izod impact strength than the one with hollow glass microspheres, while the hollow glass micro-sphere reinforced PP composite gave a lower environmental impact at the use stage, although at the material production stage it was worse in terms of environmental emissions However, in overall terms, hollow glass microsphere reinforced PP was advantageous in terms of environmental and economic reasons. This is a clear example (like others which are described in this review) of misuse of LCA. In order to compare two options, they must have the same functional unit and this would mean that the systems compared (reference flows) should have similar technical properties. This means that the amount of PP reinforced with talc was over-dimensioned and should have been reduced until the properties were as bad as those of the other composite.

Table 2.7. Methodological analysis of LCA studies (Talc vs organic and inorganic fillers)

Reference	Polymer	Functional unit (FU)	System boundaries/ Software	Impact assessment	Inventory sources	End of life	Data quality assurance
(Al-Ma'adeed et al., 2011)	Recycled PP and PE filled with talc and GF	"1 kg of material"	Cradle-to-grave/ GaBi	CML 2001	Ecoinvent and Buwal 250 (adapted to Qatar)	PE and PP will be recycled, and landfill was assumed to be disposal method	-
(Delogu et al., 2016)	Hollow GF-reinforced composite vs talc-reinforced composite	"an automotive dashboard panel, supporting and housing all the instrumentation for the vehicle use, to be mounted on Alfa Romeo Mito 955 diesel engine, with a life-distance of 150,000 km for 10 years"	Cradle-to-grave/ GaBi	CML 2001, Primary energy demand	Raw materials production from GaBi 6.3 and Ecoinvent 3.1 databases Manufacturing data from direct measurements.	Landfilling and incineration Recycling is not considered as an alternative; because of the difficulty in dismantling	SA* on production phase of hollow glass microspheres
(Luz et al., 2010)	PP + sugarcane bagasse vs PP + talc in automotive industry	"the surface area covered, i.e.,m^2"	Cradle-to-grave/ GaBi 4.3	CML 2001	Primary data from Brazilian industry GaBi database	Incineration Recycling Landfill	-
(Munoz et al., 2006)	PP-based composite with talc (New formulation for eco-design)	"A single panel"	Cradle-to-grave/ No information	ARD [80] GWP [83] AP [82] HTP [81] FATP [81] EP [82] POFP [84] Energy, water, landfill	Commercial databases Private companies Literature	Landfilling Energy recovery in MSW Incinerator or cement kiln Mechanical recycling	SA* on the quality of recycled plastic

2.2.2.3. Calcium carbonate filled plastics with fillers

Inorganic fillers have been used in plastic applications, mainly with the aim of improving heat distortion temperature, toughness, hardness, mold shrinkage and stiffness (Chan et al., 2002). Among them, calcium carbonate is a very abundant mineral on earth. It is mainly found in three forms: calcite, aragonite, and vaterite. It is widely used in the paper, rubber and plastic, as well as, adhesive and paint applications. Among thermoplastics, PP and polyvinyl chloride (PVC) are the main markets for calcium carbonate. For example, calcium carbonate is a filler used together with PP to increase the mechanical properties of its plastics, especially to enhance PP's rigidity for use in the automotive industry (Thenepalli et al., 2015).

There are many references in the literature studying the mechanical properties of plastics filled with calcium carbonate (Adeosun and Usman, 2014; Eiras and Pessan, 2009; Roussel et al., 2005). In their study, Roussel et al,(2005) say that it is possible to improve productivity in the process through the use of calcium carbonate because of its thermal conductivity, specific heat and thermal expansion characteristics. And in their study, they evaluated different case studies including blown film, extrusion coating, sheet extrusion/thermoforming, and extrusion blow molding with the aim of showing the importance of proper filler and resin combination (Roussel et al., 2005) In a study by Eiras and Pessan (2009), the change in tensile and impact properties of PP homopolymer with the addition of calcium carbonate minerals was studied at four different composition levels. The results showed an increase in elastic modulus while showing a little increase in yield stress (Eiras and Pessan, 2009). In another study, mechanical and physical properties of LDPE filled with calcium carbonate and fly ash were investigated. Flexural strength and crystallinity of composites were observed against the composite composition. The optimum combination of calcium carbonate and fly ash was determined to achieve optimum density (Adeosun and Usman, 2014). Despite the fact that studies investigating mechanical and physical properties of plastics filled with calcium carbonate, none has been found related to their environmental impact evaluation. For example, Thenepalli et al. (2013) investigated calcium carbonate as a new functional filler to PP for automotive applications. In another study, it is mentioned that PP and PVC are the main markets for calcium carbonate fillers and it is the best filler to enhance the mechanical properties of PP used in automobiles. It can provide the possibility of increased surface finishing, control of manufacturing, electric and impact resistance (Thenepalli et al., 2015). Other fillers like kaolin and clay minerals can also enhance the mechanical properties. However, they are related to asbestos and, thus, seen as not very environmentally friendly. Meanwhile, calcium carbonate is safe and abundant on the earth. Unfortunately, in none of these studies, the environmental impacts of the use of calcium carbonate as filler in plastics were covered in deep by using the LCA methodology.

2.2.3. LCA in new designs with plastics vs virgin plastics

Environmental impacts of products may come from any stage in their life cycle. Since the decisions regarding products are made during their design phase, this phase is very important for identifying environmental impacts and improvements. To this end, LCA can be an important tool to help eco-design by pointing out the critical points and comparing alternatives (Gazulla et al., 2008). The use of LCA in the automotive industry can provide the insights about how important is the choice of materials, the manufacturing process, and the fuel consumption to reduce the GHG of the vehicle (Boland et al., 2015). In Brazil, one of the largest agricultural sprayer machine companies used LCA methodology to compare environmental impacts of different fiber reinforcements against GF in an electronic-command panel of the sprayer machine. They investigated the environmental impacts of a new composite based on jute fibers as a replacement for traditional GF reinforced composite for the selected product. The results of the study were important to understand the importance of LCA as a tool to help eco-design (Alves et al., 2009).

LCA can be used for design-for-recycling purposes, as well. The increase of strict legislation on the EoL of vehicles has resulted in the use of LCA as a tool for comparison of current designs with new designs in the automotive industry. Munoz et al. (2006) used LCA to assess the environmental impacts of a “designed-for-recycling” plastic composite door panel for cars. They concluded that LCA is a very useful tool to validate new designs in terms of environmental impacts through their life cycle. Even if the study was focused on the EoL scenarios, LCA revealed some interesting points within other life cycle stages of the product.

2.2.4. LCA of end-of-life (EoL) scenarios of plastics with fillers

In some LCA studies, end of life scenarios of products/materials were not very deeply investigated. One of the reasons for that was the lack of data (Scelsi et al., 2011; Timmis et al., 2015). However, different end of life options will have different effects on the environment (Duflou et al., 2012; Väntsi and Kärki, 2015). The waste hierarchy is defined as waste prevention, reuse, recycling, energy recovery and disposal by European Commission (European Commission, 2008). Thus, the idea is to minimize disposal and incineration but to maximize recycling. For this purpose, Witik et al. (2013) investigated three possible EoL scenarios for CF reinforced plastic waste through LCA methodology: (1) recycling via pyrolysis; (2) incineration with energy recovery; and (3) disposal through landfilling. It was seen that, even if the waste hierarchy is a good rule to deal with waste, it may not always guarantee the lowest impacts for the treatment of CF reinforced polymer waste.

Type of the raw material used may determine the EoL scenario of each product. Muñoz et al. (2006) found that the little change in subcomponents of composite material used in a car door panel design, which is mainly composed of plastics like talc-filled PP, acrylonitrile butadiene styrene, PVC, polyoxymethylene, polyester, polyurethane and polyamide GF reinforced, reduces the impact of production by increasing the recyclability of the composite material. However, the environmental impacts in the use stage remain the same. It was also concluded that recycling is often the best case within the hierarchy of EoL scenarios; however, it is highly dependent on the substitution rate of polyolefin by more recyclable subcomponents. Delogu et al. (2016) still recommend to focus and further investigate the benefits of achieving a light-weight design over achieving recycling targets in the automotive sector, while Witik et al. (2011) had already concluded that the benefits of light weighting composites for automotive industry outweighs the fact of not being recyclable.

2.3. Conclusions

Because of the increasing demand for thermoplastics, a wide range of functional fillers, both organic and inorganic, is applied to plastics with different goals: cost reduction, process improvement, altering mechanical or physical properties or reducing environmental emissions. Since the new trend in the market is to look for more environmentally friendly materials in order to meet with certain sustainability targets, this paper focused on the literature to see if the use of fillers in plastics could be promising to reduce the environmental impacts. The results of this study may be interesting for the scientific community to attract attention to environmental advantages of the use of fillers in plastics industry. More specifically it could be interest of LCA experts, since how LCA methodology has been used in this area was reviewed; as well as it could be interesting for material experts because the types of fillers used in different applications were investigated in terms of their environmental impacts.

According to the studies reviewed and presented in this paper, as a response to the major objective of this chapter, it can be concluded that the environmental impacts of plastics can be reduced through the addition of functional fillers while maintaining or improving the required technical properties of the conventional material. In the reviewed studies, it was observed that plastics with functional fillers had smaller GWP than their virgin counterparts. Functional fillers tend to reduce the environmental impacts of these materials because they reduce the amount of virgin petrochemical materials used in the composite by replacing them with a material with a lower environmental impact.

Another objective of this chapter was to find out the gaps in the literature to provide guidance to the future work. Many studies can be found in the literature which is dealing with the mechanical and physical properties of plastics with different kinds of fillers. However, their environmental impacts are seldom studied, especially concerning the plastics with mineral fillers. It has been observed that organic fillers are more often studied than inorganic fillers in terms of their environmental profile. Specifically, it was nearly impossible to find studies about calcium carbonate, even if it is one of the most commonly used mineral fillers in many industrial applications. Therefore, an important research gap has been found.

Finally, the last objective of this chapter was to investigate LCA methodology used in the case studies reviewed. Based on the review done, it can be concluded that LCA is a good tool to make the environmental analysis of materials; however, the results are application specific and no general conclusions should be driven. Nevertheless, despite the differences between the LCA studies, three major conclusions can be obtained regarding LCA application to plastic composites:

- Since different materials may present different physical and mechanical properties, in the case of comparative LCA studies, it is very important to define a proper functional unit serving the same function, thus allowing to make a fair comparison between the material types. This is rarely done in the composite's LCA literature reviewed here.
- It is suggested to perform cradle-to-grave LCA, when evaluating the environmental impacts of a material, in order to avoid problem shifting in between the life cycle stages.
- EoL stage of plastic composites is rarely based in real specific data or experiments on the recyclability of the newly developed composite.

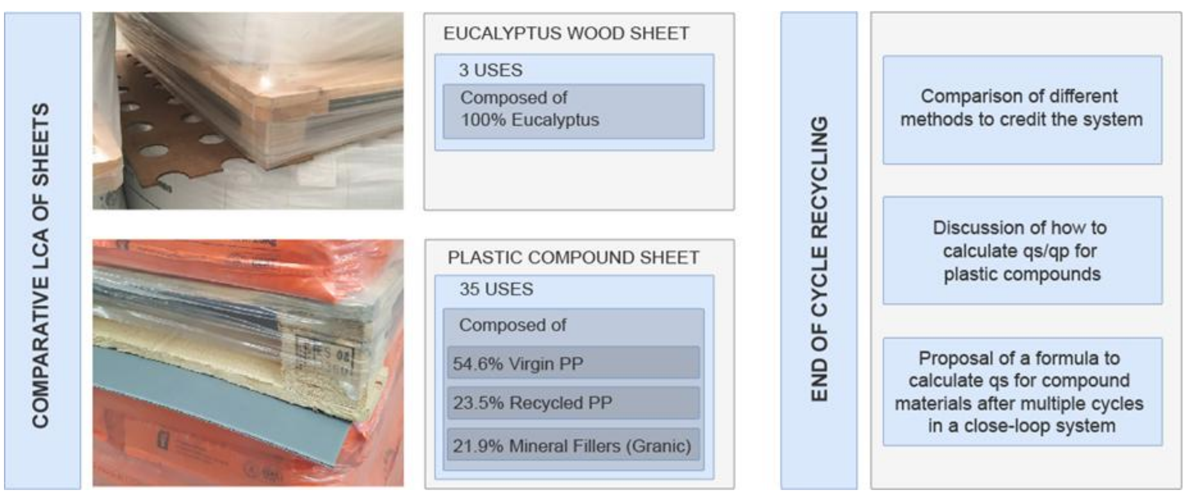
In the case of functional unit, for example, when applying LCA methodology to plastics, a clear application (a given product) was often the object of study, so the function and the functional unit of the LCA could be defined. However, many times a 1 kg of material was chosen as the functional unit (or reference flow). In order to compare different materials, it is essential that the options being compared fulfill the same amount of service or function. As the different materials have different physical properties, the amounts used as reference flows should be taken as those with equal properties; i.e. decreasing the amount of the composite with higher quality. This reasoning is not always followed and, therefore, the comparisons are not fair.

Related to life cycle stages, it was observed through the LCAs reviewed that the fillers may have advantages over conventional materials or other types of fillers for one specific life cycle stage, although it may have worse environmental results for another life cycle stage. Therefore, a cradle-to-grave LCA should be addressed to be able to say one filler is environmentally better than the other one.

And finally, according to EoL scenarios considered in the reviewed literature, landfilling, energy recovery and material recycling are always theoretical scenarios, not based on real experiments or real applied solutions for the newly developed composite. Therefore, much effort is needed on this subject to decide if the new composite is environmentally better, especially considering that circular economy is one of the main sustainability drivers nowadays.

Although the number of LCA studies is still very low within a quite important universe of technical studies of plastic composites, we can conclude that this study was needed to attract attention to the use of functional fillers in plastics and proper application of LCA methodology in order to understand their environmental advantages on the application base. Finally, it can be recommended for the future work to focus on performing environmental analysis for fillers which have not been studied too often and, while performing LCA studies, to choose the correct functional unit, and if possible, to make cradle-to-grave analysis and to collect more data about EoL of the materials used.

Chapter 3. INFLUENCE OF END-OF-LIFE ALLOCATION, CREDITS AND OTHER METHODOLOGICAL ISSUES IN LCA OF COMPOUNDS: AN IN-COMPANY CIRCULAR ECONOMY CASE STUDY ON PACKAGING



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Abstract

The aim of this article is to present a circular economy case study and investigate and discuss effects of end-of-life (EoL) allocation and crediting strategies on the results of this case study. In the case study, replacement of eucalyptus wood sheets, which are used to separate loaded pallets to prevent damaging each other during top storage in the company, by plastic compound alternatives composed of virgin PP, recycled PP and mineral fillers, is studied. When their life time is over, plastic compound sheets are sent to be recycled in the recycling facilities of the company. While performing this comparative LCA, a methodological discussion on how to credit the system in open-loop (OL) and close-loop (CL) recycling is performed. The use of Q factors (quality factors), instead of 1:1 substitution of virgin materials by recycled ones, is recommended and how to define these Q factors is discussed. The use of Q factors based on the mechanical properties of virgin and recycled materials, which is flexural modulus in this case, is recommended. Finally, a formula for the calculation of the Q factor of the compound material leaving the CL recycling after several recycling cycles, is proposed. Results show that, for this case study, plastic compound sheets are environmentally better alternative than eucalyptus wood sheets for most of the environmental impact categories evaluated due to the following reasons: higher number of uses, lower weight, use of recycled PP and mineral fillers, and longer lifetime. However, in two impact categories (resource depletion water and resource depletion mineral, fossils and renewables) eucalyptus wood sheets are found to have slightly better results. For the rest of the impact categories, the difference in the results are so high that different crediting methods do not affect the results in this case; however, they may in others. Among the scenarios evaluated OL recycling with market mix substitution is found to provide the highest impacts.

3.1. End-of-life (EoL) allocation in LCA

According to a recent literature (Allacker et al., 2017), for EoL allocation, at least 11 different formulas have been identified as widely used and accepted methodologies, and some of them are included in different standards.

One of these methodologies is the cut-off (or 100:0) approach, also known as the recycled content approach. It implies that 100% of virgin material production has to be assigned to the first product of the cascade, and the burdens of EoL recycling process are ascribed to the downstream product (Baumann and Tillman, 2004). Another methodology is the 100:100 approach, where burdens from the production of virgin material are 100% allocated to the product using this virgin material, and the burdens and credits from recycling at the EoL (Allacker et al., 2017). This last methodology will be the one used in this paper as it considers both the recycled content in the input and final recyclability of the output. The authors believe that the use of 100:100 formula is balanced as it benefits both, the products using recycled material and the products designed to be recyclable.

Therefore, the novelty of this chapter is to calculate quality loss during recycling for a compound material in a multi-loop recycling system and its use in closed-loop and open-loop recycling based on a real case study (using 100:100 approach for EoL modeling). Besides, it will be the first case study investigating the environmental profile of calcium carbonate as the filler in thermoplastics.

In this study, LCA will be used to environmentally compare different materials (eucalyptus wood and plastic compound) for sheets, which are used to separate pallets during storage. Therefore, the first aim of this chapter is to identify which option is environmentally better regarding the application of the circular economy in the company. And, the second aim is, by using the case study, to focus on the use of quality factors (Q factors) when crediting the system in open-loop and closed-loop recycling.

The methodology used to perform the LCA case study is explained in section 3.2 and the results are presented in section 3.3. Following that, in section 3.4, the use of Q factors for plastic recycling is discussed in the case of open-loop and closed-loop systems. Methodology for those alternatives is explained in the same section for clarity reasons

3.2. Methodology of the case study

3.2.1. Goal and scope

The goal of the LCA study is to evaluate environmental impact improvements, if achieved any, by changing the material of the sheets used for storage in a Spanish company (GCR Group, 2018). The current case, which is the use of eucalyptus-based particleboard as the sheet (system A), will be compared with an alternative scenario considering the use of plastic compound sheets (system B) by using LCA methodology, according to an attributional approach. At the EoL stage of the alternative scenario, plastic sheets will be recycled at the recycling facilities of the company in contribution to in-company circular economy, thus producing no waste. It certainly is a Spanish case study; however, the service given by the sheets is quite universal in non-humid conditions (for humid ones, the wood sheets may perform worse and be early broken). Environmental impacts are identified by using LCA methodology following the ISO 14040 (ISO, 2006a) and ISO 14044 (ISO, 2006b) standards.

3.2.1.1. Product description

The company produces mineral masterbatches for the plastic processing industry, which are called Granic. When this study was performed, eucalyptus wood sheets were the current practice at the storage of their products in the factory. Wooden sheets are produced out of eucalyptus hardwoods in Spain. The fibers are connected to each other by only heat and pressure without the addition of any artificial binder. Each wooden sheet has the dimensions of 1200 x 1000 x 6 mm, the density of 950 kg/m^3 and weights 5 kg.

In correspondence to the fewer number of uses and high cost of wooden sheets, an eco-design alternative by changing the sheets to plastic ones is proposed. These plastic sheets are composed of 20.1% Granic 1081 (talc-based), 1.8% Granic 1522 (calcium carbonate based), 23.5% recycled PP and 54.6% virgin PP, and are produced by one customer of the company. Produced plastic sheets are slightly smaller, with the dimensions of 1100 x 950 x 3 mm and the weight of 1.24 kg. Sheets are presented in Figure 3.1.

Figure 3.1. Eucalyptus wood sheet (on the left), plastic compound sheet (on the right)

Since the use of eucalyptus wood sheets was the current situation in the company, as a result of many years of use, it was already known that they get destroyed easily and are discarded after being used 3 times. On the other hand, based on the testing in the company, plastic compound sheets are used 35 times on average. This number is the average obtained with 3 sheets bought and tested by the company in normal logistic operation. Since more statistically representative data is not available, sensitivity analysis on the number of uses is performed in section 3.3.3. For the EoL, wood sheets are first sent to a nearby composting facility and, following that, they are placed in a landfill with energy recovery; while plastic sheets are recycled.

3.2.1.2. Function, functional unit and reference flow

Sheets serve to separate two wooden pallets carrying the final packaged products (Granic) and they protect the final product packaging of being damaged during on-top storage. After this storage, the final product is loaded to the trucks with wooden pallets, leaving the sheet in the company to be re-used for the same purpose. Therefore, to be able to have comparable results, the functional unit of LCA is defined as 105 uses of the sheet, which is able to separate and protect the final product during storage, either by eucalyptus wood or plastic compound sheet. In order to fulfill the functional unit, a reference flow of 35 wood sheets are needed to be bought for the current scenario. For the alternative scenario, since the plastic sheets can be reused 35 times, 3 plastic sheets would be enough as reference flow. Technical details regarding evaluated scenarios and calculated reference flow to the corresponding functional unit are presented in Table 3.1. Although the dimensions of both sheets are different, the function is the same and the reduction of the plastic sheet dimensions is an additional eco-design measure to better fit the dimensions of the pallets used.

Table 3.1. Technical details of the scenarios

	Eucalyptus wood sheets (current scenario)	**Plastic compound sheets (alternative scenario)**
Materials	Eucalyptus wood	Virgin PP (54.6%) Recycled PP (23.5%) Granic 1081 (20.1%) Granic 1522 (1.8%)
Number of uses	3	35
Weight	5 kg	1.3 kg
Dimensions	1200 x 1000 x 6 cm	1100 x 950 x 3 cm
Functional Unit	105 uses	105 uses
Reference Flow	175 kg of sheet	3.9 kg of sheet

3.2.1.3. Impact categories used

For the calculation of environmental impacts, ILCD/PEF recommendations v1.09 are used and the following environmental impact categories and indicators are evaluated: acidification [Mole of H+ eq.] (AP), climate change (excluding biogenic carbon) [kg CO2 eq.] (GWP), eutrophication freshwater [kg P eq.] (EP freshwater), eutrophication marine [kg N eq.] (EP marine), ozone depletion [kg CFC-11 eq.] (ODP), photochemical ozone formation, human health [kg NMVOC] (PCOF), resource depletion, mineral, fossils and renewables [kg Sb eq.] (ADP), resource depletion water [m3 eq.], primary energy from non-renewable resources (net cal. value) [MJ] (PE_non ren) and primary energy from renewable resources (net cal. value) [MJ] (PE_ren).

3.2.2. System boundaries

System boundary is defined as cradle-to-grave; from raw material extraction to the EoL of the sheets. Figure 3.2 illustrates the system boundary overviews for eucalyptus wood and plastic compound sheets. System A corresponds to the current situation in which eucalyptus-based particle boards are being used. According to the company, when their lifetime is over, they are transported to the nearby composting facility and, later, to the landfill with energy recovery.

In system B, plastic compound sheets (produced using Granic products) are considered. The company buys the sheets from a provider, which is their customer as well, and uses them in the factory for storage purposes. In this scenario, at the EoL, they are recycled at the recycling facilities of the company and sold to be used for other purposes.

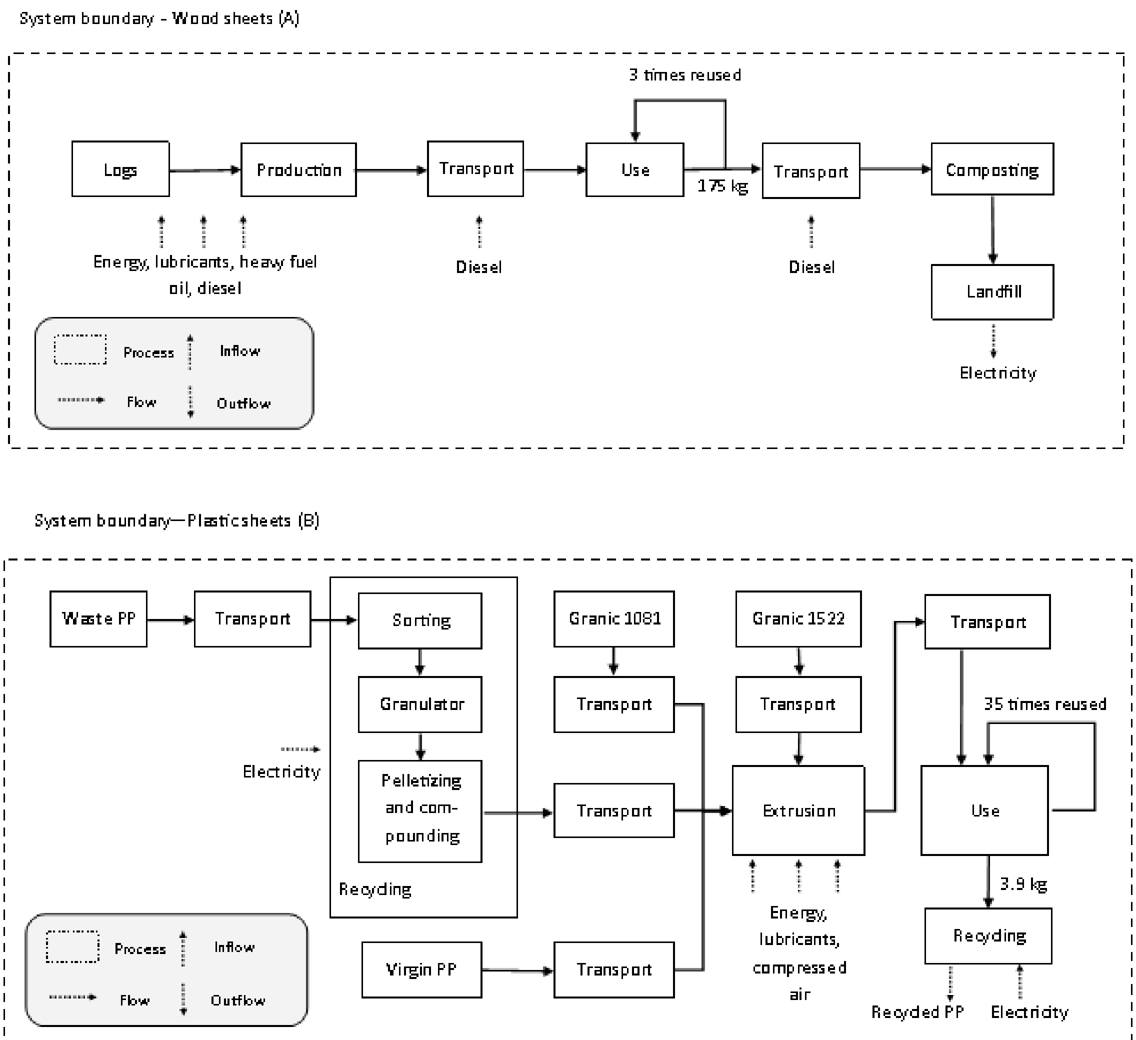


Figure 3.2. System boundaries of eucalyptus wood (System A) and plastic compound sheets (System B)

3.2.3. Data collection

LCA is performed by using the GaBi 8 software (Thinkstep, 2016). Data on masterbatches (Granic) produced by the company and transportation means and distances are gathered from the company (Table 3.2). For rest of the processes, Ecoinvent (Wernet et al., 2016) or GaBi databases (Thinkstep,

2016) is used. For transportation, 22-ton diesel truck is chosen from GaBi database. For the production of plastic compound sheets, plastic extrusion process; and for the production of eucalyptus wood sheets, particle board production process is considered. In the case of particle board production, the process is taken from the Ecoinvent 3.3 database and adapted to the case study by doing the following:

- Inputs of electricity from different sources are summed up and replaced by the Spanish Electricity Grid Mix because the company producing those sheets is in Spain.
- Since, in this case study, particle boards are produced from the eucalyptus tree, all different waste wood output flows are summed up and replaced by a single "waste wood, untreated waste" flow, presenting eucalyptus wood.
- Inputs of wood and bark chips are summed up as eucalyptus due to the same reasoning given above.

Table 3.2. Distances between the facilities

	Distance (km)
Eucalyptus wood sheets	
From the sheet production facility to the Company	75
From the Company to the treatment facility	68
Plastic compound sheets	
From the waste PP supplier (Nantes, France) to the Company (Tarragona, Spain)	1035
From the Company to the sheet production facility	287
From the virgin PP supplier to the sheet production facility	250

At the EoL, for eucalyptus wood sheets, since the composting process is not available in the databases, direct landfilling with energy recovery from biogas production is assumed. This may cause reduction in the overall impacts of the system since the composting process uses electricity. It may also affect the water emissions from the landfill since a residue with higher water content is assumed. On the other hand, for plastic compound sheets, the recycling process, composed of the granulator, pelletizing and compounding, is considered.

IDs of the processes that are used in the modeling are presented in Table 3.3 in detail. In addition, other required data on energy consumption, lubricants, compressed air, heavy fuel oil, diesel consumption, and truck are taken from GaBi database.

Table 3.4 shows the major inputs of raw materials and electricity consumption for each LCA scenario evaluated. In addition to that, avoided impacts from EoL of the sheets (landfilling with energy recovery and recycling) are presented in the table with a negative sign.

Table 3.3. IDs of processes used in modeling

Life cycle stage	**The ID of the process**	**Database source**
Raw materials (Wood sheets)	RoW: hardwood forestry, eucalyptus ssp., sustainable forest management	Ecoinvent 3.3 SP33
Production process (Wood sheets)	RoW: particle board production, for indoor use, from virgin wood	Ecoinvent 3.3 SP 33
EoL (Wood sheets)	EU-28: Wood products (OSB, particle board) on landfill ts <p-agg>	GaBi professional + extensions 2017 (SP33)
Raw materials (Plastic compound sheets)	DE: Polypropylene granulate (PP) mix ts	GaBi professional + extensions 2017 (SP33)
Production process (Plastic compound sheets)	GLO: Plastic extrusion profile (unspecific) ts <u-so>	GaBi professional + extensions 2017 (SP33)
EoL (Plastic compound sheets)	DE: Granulator ts <u-so> DE: Pelletizing and compounding ts <u-so>	GaBi professional + extensions 2017 (SP33)

Table 3.4. Inputs per reference flow of each scenario

Scenario	**Ref. flow (kg)**	**Round wood (m^3)**	**Virgin PP (kg)**	**Recycled PP (kg)**	**Granic (kg)**	**Electricity (MJ)**	**Avoided electricity from EoL (MJ)**	**Avoided PP from EoL (kg)**	**Avoided Granic from EoL (kg)**
Wood sheets	175	0.2483	-	-	-	93.39	110.8	-	-
Plastic lamina sheets	3.9	-	2.14	0.9211	0.86	10.9	-	2.024	0.81

3.2.4. 100:100 EoL allocation approach with credits from recycling

The 100:100 EoL allocation method (Allacker et al., 2017) is one of the most commonly used methods to include both: recycling content in the input material and recyclability at the EoL. According to Allacker et al. (2017) the formula to obtain the environmental footprint (EF) of a material, using 100:100 approach with crediting for avoided virgin production, would be:

$$EF = (1 - R_1) * E_v + R_1 * E_{recycled} + R_2 * E_{recycling,EoL} - min\,(abs(R_2 - R_1), R_2) * E_v^* * \frac{Q_s}{Q_p} + (1 - R_2) * E_D$$

Where:

EF: emissions and resources consumed (per unit of analysis) arising from the production and the EoL stages of the product life cycle.

E_V: emissions and resources consumed (per unit of analysis) arising from the acquisition and pre-processing of virgin material.

E^*_V : emissions and resources consumed (per unit of analysis) arising from the acquisition and pre-processing of virgin material assumed to be substituted by recyclable materials.

$E_{recycled}$: emissions and resources consumed (per unit of analysis) arising from the production process of the recycled material, including collection, sorting and transportation processes.

$E_{recycling,EoL}$: emissions and resources consumed (per unit of analysis) arising from the recycling process at the EoL, including collection, sorting, transportation and recycled material production processes

E_D: emissions and resources consumed (per unit of analysis) arising from disposal of waste material (e.g. landfilling, incineration and pyrolysis).

R_1: “recycled content of material”, is the proportion of material in the input to the production that has been recycled in a previous system (0 = <R1 <=1).

R_2: “recycling fraction of material”, is the proportion of the material in the product that will be recycled in a subsequent system, i.e. the rate between recycled output and virgin material input. R2 shall, therefore, take into account the inefficiencies in the collection and recycling processes (0 = <R2 =<1).

Q_S: quality of the secondary material, i.e. the quality of the recycled material.

Q_P: quality of the primary material, i.e. the quality of the virgin material.

Therefore, in the case of plastic compound sheets, above defined 100:100 allocation approach is used by considering only transport and recycling burdens to the waste PP coming from Nantes, France. And, for the EoL recycling of the sheets, credits from substitution are considered. However, at the EoL, credits are only given for the corresponding portion of virgin material (virgin PP + Granic) used in the composition of plastic sheet, not for the recycled content. In addition, when no disposal of laminas at their EoL is considered (R_2=1), the previous formula is adapted as follows:

$$EF = (1 - R_1) * E_v + R_1 * E_{recycled} + (1 - R_1) * E_{recycling,EoL} - (1 - R_1) * E_v^* * \frac{Q_s}{Q_p}$$

3.3. Results and discussion

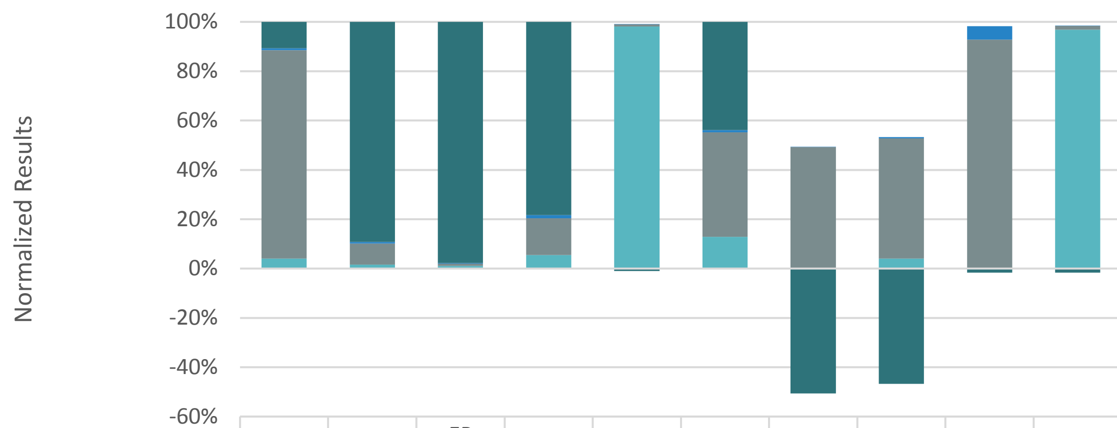
3.3.1. Current scenario (wood sheets)

The results of each life cycle stage of wood sheets are presented in Figure 3.3. In the figure, the total life cycle is presented as 100% and divided into four main subsystems: raw materials preparation, production, transport, and EoL. Since no impacts are expected from the use phase, it is not included in the table. Raw materials preparation stage includes hardwood forestry from eucalyptus. Production represents the particle board production process and all the necessary consumptions like electricity, lubricants, heavy fuel oil and diesel. Transportation includes all the truck transports from the producer of sheets to the company, where they will be used, and from the company to the composting facility and the corresponding diesel consumption. Finally, the EoL stage is the summation of the impacts caused by landfilling and the credits gained from energy recovery by electricity production at the landfill. In Figure 3.3, negative values shown for water consumption and ADP for EoL represent the avoided burdens that are caused by electricity production in landfilling of that product.

For all of the impact categories, transportation is identified as the least contributing stage. On the other hand, results show that production process and EoL stages are the major contributors to the all evaluated impact categories, except for ozone depletion and primary energy from renewables. For those, the raw materials stage is found to have the largest impact, due to the forestry of eucalyptus trees.

For the impact category of resource depletion mineral, fossils and renewables, electricity consumption is identified as the major contributor; however, its contribution is nearly balanced by the credits gained from electricity production at landfilling (the overall result is 5.66 x 10-6 kg Sb eq). For the impact category of resource depletion water, a negative global value of -0.0275 m3 eq is found. Negative results have no physical meaning and they are due to inconsistencies. The reason for this negative value is identified as the use of different databases for the modeling of life cycle of wood sheets, as Thinkstep considers different type of water flows and localize them according to the regions, while Ecoinvent considers one single type of water flow characterization for the estimation of the impact. Nevertheless, the impact results would be no better for wood (but on the contrary), if the same database could have been used for all the processes involved, thus avoiding inconsistencies. So, we consider the calculations acceptable for the aim of the study.

In addition, for renewable energy sources, like wood, part of the energy consumed in the production process comes from the sun (so, no impacts are considered for this input energy). This fact makes it possible to recover more energy in the EoL than the one consumed from the technosphere as input, thus obtaining more credits than burdens. As said before, this is the case for all renewable energy sources.



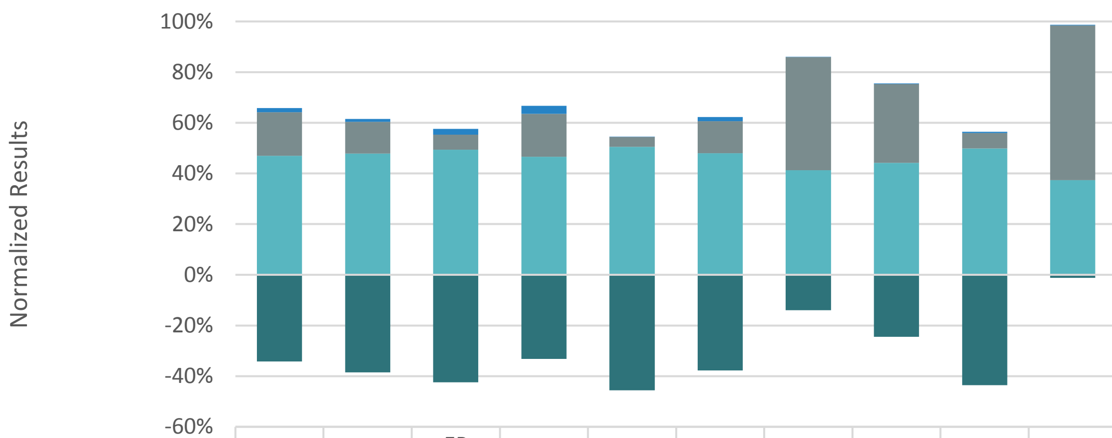
	AP [Mole of H+ eq.]	GWP [kg CO2 eq.]	EP freshwater [kg P eq.]	EP marine [kg N eq.]	ODP [kg CFC-11 eq.]	PCOF [kg NMVOC]	Water [m3 eq.]	ADP [kg Sb eq.]	PE_non ren [MJ]	PE_ren [MJ]
End-of-life	4,78E-02	1,98E+02	2,31E-03	1,09E-01	-4,25E-0	1,72E-01	-1,14E+0	-3,97E-0	-5,27E+0	-1,20E+0
Transport	3,94E-03	1,26E+00	5,83E-06	1,79E-03	4,13E-13	3,39E-03	1,48E-03	4,57E-07	1,71E+01	8,57E-01
Production process	3,79E-01	1,90E+01	1,88E-05	2,06E-02	3,61E-09	1,66E-01	1,11E+00	4,14E-05	2,90E+02	1,14E+02
Raw materials	1,83E-02	3,62E+00	2,67E-05	7,69E-03	4,13E-07	5,05E-02		3,49E-06	2,29E-03	7,24E+03

Figure 3.3. LCA results for eucalyptus wood sheets

3.3.2. Alternative scenario (plastic compound sheets) compared to current scenario (wood sheets)

In the case of plastic compound sheets, a similar life cycle grouping is done for the presentation of LCA results. Four main life cycle phases are considered: raw materials preparation, production, transport, and EoL. The group of raw materials preparation includes the following processes: production of virgin PP, production of two different types of mineral filled plastic granulates (Granic 1522 and Granic 1081), and waste PP coming from a company in Nantes (France) and it's onsite recycling at the subject company. On the other hand, production refers to all the production processes of plastic sheets which are: mixing of plastic granulates, extrusion, and all related consumptions like electricity, thermal energy, compressed air, and lubricants during extrusion. Transportation includes the transportation of the raw materials to the production facility and then to the company where the final product will be used. Finally, as EoL scenario, onsite recycling of the plastic sheets at GCR Group is considered. Recycling process consists of granulating, pelletizing and compounding processes. In the EoL, the system is credited only for the recyclability of the virgin fraction; the amount of virgin PP and Granic (which are made from virgin PP and mineral fillers) used as input.

Results of the LCA of plastic compound sheets are presented in Figure 3.4. The impacts from total life cycle are normalized to 100% and then divided into the life cycle stages explained above. Negative values on the graph represent the avoided burdens from recycling at the EoL. As it can be seen from the figure, impacts from the transportation stage are very few compared to other life cycle stages. Most of the impacts come from the raw materials production stage, mainly due to virgin PP production, with the exception of water resource depletion and primary energy demand from renewables, because they are more or less balanced by the credits gained from the production of recycled PP and Granic at EoL. Therefore, in the case of water depletion and primary energy demand from renewables, the production of sheets is the major stage where emissions come from, mainly due to the consumption of electricity.



	AP [Mole of H+ eq.]	GWP [kg CO2 eq.]	EP freshwater [kg P eq.]	EP marine [kg N eq.]	ODP [kg CFC-11 eq.]	PCOF [kg NMVOC]	Water [m3 eq.]	ADP [kg Sb eq.]	PE_non ren [MJ]	PE_ren [MJ]
End of life	-6,75E-0	-3,53E+0	-9,63E-0	-1,57E-0	-4,80E-0	-6,72E-0	-4,00E-0	-3,58E-0	-1,46E+0	-2,73E-0
Transport	3,29E-04	1,06E-01	4,92E-07	1,49E-04	3,49E-14	2,83E-04	1,25E-04	3,85E-08	1,44E+00	7,22E-02
Production process	3,40E-03	1,14E+00	1,36E-06	8,02E-04	4,18E-10	2,26E-03	1,28E-01	4,56E-06	2,06E+01	1,31E+01
Raw materials	9,26E-03	4,39E+00	1,12E-05	2,20E-03	5,32E-09	8,53E-03	1,18E-01	6,48E-06	1,67E+02	8,01E+00

Figure 3.4. LCA results for plastic compound sheets (System B)

In Figure 3.5, the results of the plastic compound sheets are compared to results of wood sheets. Results show that the use of plastic sheets has less impact on most of the environmental impact categories evaluated. The impacts of plastic sheets vary between 0.1 - 1.4 % of the impacts of wood sheets, except in the case of primary energy from nonrenewable sources, it reaches to 14% of the wood's impact. On the other hand, in the case of resource depletion water and resource depletion mineral, fossils and renewables, wood sheets show better emissions.

For the resource depletion water impact category, for wood and plastic, the values obtained are -0.0275 m3 eq and 0.206 m3 eq, respectively. As it was explained above, in the case of wood sheets, a negative value is reached due to inconsistencies from the different databases used for the production of wood laminas (Ecoinvent) and the energy recovery from landfilling (Thinkstep). On the other hand, in the case of plastic compound sheets, the major water consumption occurs due to the electricity consumption during the extrusion of sheets, since the impacts of virgin material production are neutralized by the credits from recycling.

In the case of resource depletion, mineral, fossils and renewable, a slight difference is identified between the scenarios of wood and plastic. Results are calculated as 5.66x10-6 for system A and 7.49x10-6 kg Sb eq for system B. Therefore, it can be assumed that both options have more or less the same impact for this impact category.

In the circular economy context, it is also important to note that environmental impacts from transportation stage reduced by one digit in the case of plastic compound laminas. The reasons for that are identified as; only 3 times reuse of wood sheets increases the amount of waste produced and thus transportation to landfill, and on-site recycling of plastic compounds helps to reduce the impacts from the transportation of waste even though recycled PP is transferred from France to Spain.

To conclude, by checking the results for all evaluated impact categories, it can be said that, for this specific case study, the use of plastic compound sheets is significantly advantageous.

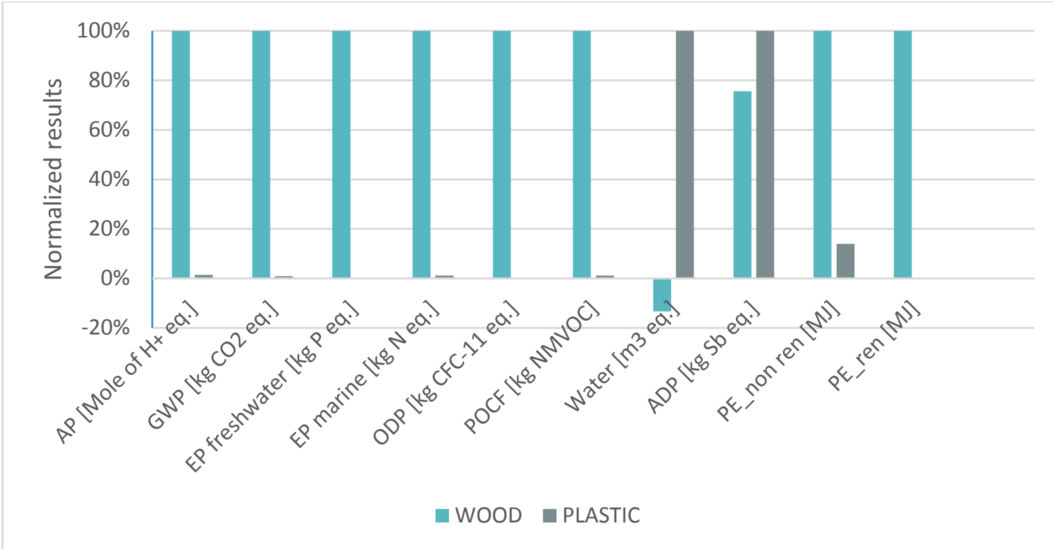


Figure 3.5. LCA results of eucalyptus wood vs plastic compound sheets (base scenario)

3.3.3. Sensitivity analysis on "number of uses"

Based on the testing of plastic compound sheets in the company, their number of uses is defined as 35 times for the specified function. Since plastic sheets have a significantly higher number of uses than wood ones, a sensitivity analysis is used to identify its importance on the results. Results of the analysis for some impact categories are presented in Figure 3.6.

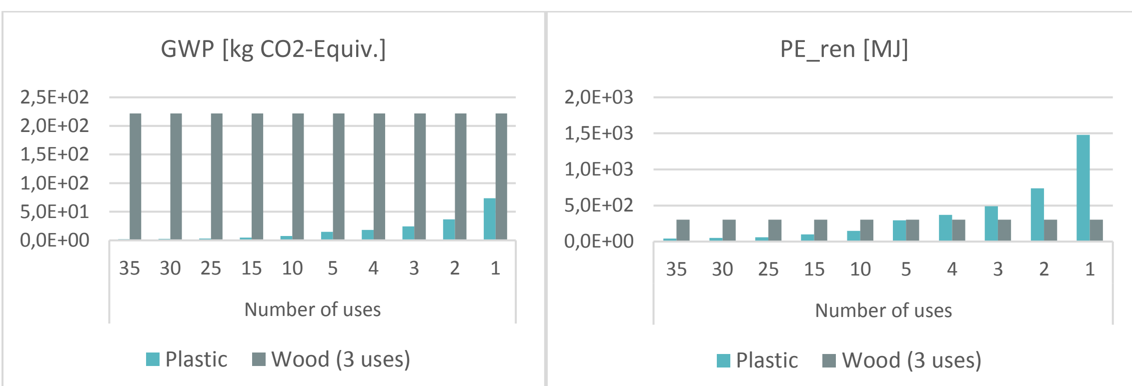


Figure 3.6. Sensitivity analysis on "number of uses" for plastic compound sheets (the categories not presented here follow the same pattern as GWP)

Results show that plastic compound sheets present better environmental results than wood sheets even when they are only used 1 time for most impact categories (e.g. GWP in Figure 3.6). The exceptions are only three impact categories: resource depletion water and resource depletion, mineral, fossils and renewable, as explained in chapter 3.3.2, give already better results for wood when plastic sheets are used 35 times; therefore, any decrease in the number of uses only worsen the situation for plastic. The third exception is primary energy from non-renewable resources, which has a breaking point at 5 uses as it can be seen in Figure 3.6.

3.4. Discussion on different alternatives to crediting the system from recycling of plastics

In LCA studies, as stated by Bala-Gala et al. (2015) it is a very common practice to credit the system when recycling a material considering that this recycled material will substitute virgin material (1:1 substitution ratio of virgin material by recycled one). Our belief is that using 1:1 substitution ratio can lead to over-crediting the system. Therefore, quality loss of recycled materials should be considered when giving credits to the system as stated also by other authors (Allacker et al., 2017; Koffler and Finkbeiner, 2018).

In addition to the aforementioned idea, closed-loop (CL) and open-loop (OL) recycling scenarios could lead to different results when modeling the system.

Therefore, in the following sections, system B is reevaluated considering CL and OL recycling scenarios with quality factors (which will be referred as Q factors from now on). A Q factor is the ratio between the qualities of the recycled material to the virgin with respect to a specific property which is important for the function of the product. This is called as Qs/Qp ratio by EC for PEF (Allacker et al., 2017; EC, 2012).

3.4.1. Open-loop recycling

3.4.1.1. Description of the open-loop recycling model with Q factors

In system B, which is an OL system, recycled PP granulates are produced at the end of recycling of plastic sheets to be used in another application. In section 3.3.2., environmental credits are given to the system from the production of recycled PP without considering the quality loss, however, in this section quality loss of recycled PP will be considered through a Q factor. Again, as explained in section 3.2.4, environmental credits are only given for the portion of virgin materials (virgin PP and mineral fillers) used in the sheets.

However, there is a lack of literature on values of Q factors for plastics. Rigamonti et al. (2009) as cited in Bala-Gala et al. (2015) propose a Q factor of 0.9 for plastics, but this is a very general value which could be more precisely calculated. According to Bala-Gala et al. (2015), Q factors should be calculated based on technical properties and contamination levels of plastics.

In the present study, Q factors for PP in OL recycling will be calculated based on; 1) technical properties 2) price 3) composition and price.

In the case of plastic sheet application, the most important technical property of the material is the flexural modulus. In other words, it is the capacity of the material to resist bending without breaking. Some values for flexural modulus of virgin and recycled PP are found in different sources in the literature (Raj et al., 2003; Srebrenkoska et al., 2014; Yin et al., 2013) but, curiously, in some cases, flexural modulus is higher for recycled PP than for virgin PP. However, in this study, primary data for flexural modulus of recycled PP was measured as 1005 MPa in the laboratory of the subject company using Instron-Ceast 3366 equipment and following the ISO 178 standard. Since, according to our market search, the flexural modulus of virgin PP changes between 950-1200 MPa, for the estimation of Q factor the middle value of that range is taken (1075 MPa) and, therefore, the Q factor is estimated as 0.94. This means that, with one recycling loop, the PP loses a value of 6% of this property.

On the other hand, according to a market search done by the company, the price rate of recycled PP to virgin PP is found to be 0.6. If the market price is related to the quality of the material, a Q factor of 0.6 should be considered for recycled PP.

Finally, considering the composition of the recycled sheet (78.1 % recycled PP, 20.1% talc-based filler and 1.8% calcium carbonate-based filler) and the prices of each material (see Table 3.5), a Q factor of 0.66 is calculated as follows.

$$Q = (0.781 \times 0.6) + (0.201 \times 0.9) + (0.018 \times 0.65) = 0.66$$

Estimated Q factors for recycled PP are summarized in Table 3.6.

Table 3.5. Price and composition of materials in recycled sheets

Material	Price in proportion to virgin PP (market avg.)	Composition in recycled sheet (%)
Virgin PP	1	0
Recycled PP	0.6	78.1
Talc based filler	0.9	20.1
Calcium carbonate-based filler	0.65	1.8

Table 3.6. Calculated Q factors for recycled sheets

Method	Q factor calculated for recycled PP
Technical property (Flexural modulus)	0.94
Price	0.6
Composition	0.66

Now, the Q factors must be applied to credit the system. One possibility is to consider that the recycled material obtained will substitute virgin material at the same rate as the Q factor (i.e., Q factor of 0.6 means that 1 kg of recycled material will substitute – it is worth - 0.6 kg of virgin material, thus the credits to the system will come from the 0.6 kg of virgin material avoided).

Another possibility, suggested by Bala-Gala et al., 2015, is to consider that the recycled material will substitute the average material found in the market (market mix substitution approach). In this case, the formula proposed by those authors to calculate the credits of 1 kg of recycled material to the system is as follows:

$$\text{Environmental credit} = x*REC + (1-x)*Q*VIR \qquad (1)$$

Where:

x is the proportion of recycled material in the average market mix

(1-x) is the proportion of virgin material in the average market mix

Q is the quality factor of recycled material vs. virgin material ($Q \leq 1$)

REC is the environmental load of the recycling process (1 kg of recycled material in output)

VIR is the environmental load of the production process of the virgin material (1 kg in output).

For the calculation of REC virgin PP production, and for REC, recycling processes given in Table 3.3 are used.

3.4.1.2. Results for open-loop recycling model with Q factor

The results of the OL recycling model are presented in Figure 3.7 for some of the impact categories, considering that the recycled material will substitute a fraction of virgin one (the fraction in accordance with its Q factor). Q factors of 1 (system B), 0.94, 0.66 and 0.6 are evaluated. As expected, when Q factor decreases, results of each impact category increase and follow the same trend (since environmental credits given to the system decrease). It can be concluded that the use of Q factors can cause a significant change in the environmental impacts results (see Figure 3.7).

Figure 3.7. Results of OL recycling of plastic sheets by using different Q factors

Environmental credits given to the system are also calculated by using the formula proposed by Bala-Gala et al. 2015 (market-mix substitution approach). The proportion of the recycled PP in the average market mix is found as 12.5% for the year of 2016 (Plastics Europe, 2017). In Figure 3.8, results of the application of the two different methods (assuming replacement of the material by using the same Q factor of 0.94 and considering the market mix substitution approach) are given in comparison to baseline scenario (system B). In the graph, results are given normalized to system B for the clarity of comparison. According to the results, system B has the lowest environmental impacts, while the market mix substitution approach resulted in the highest environmental impacts.

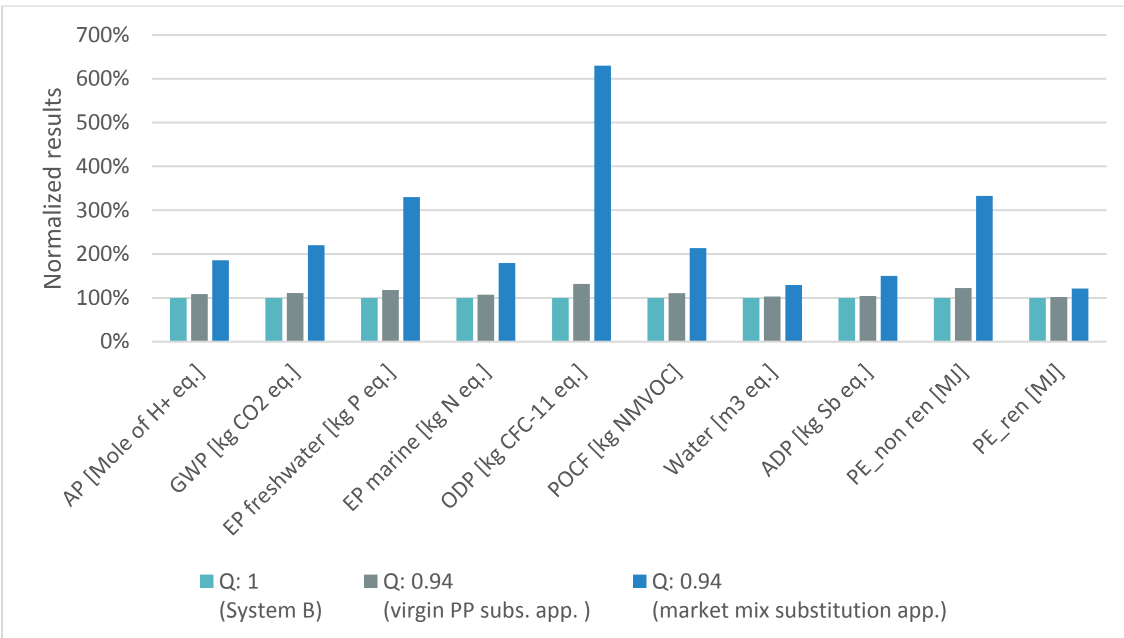


Figure 3.8. Comparison of two Q factor application methods with base scenario (system B) in OL recycling

In addition, the market share of recycled PP is expected to exceed 25% by the year of 2025 (Plastics Europe, 2017). In that case, if the market mix approach proposed by Bala-Gala et al. (2015) is used to calculate environmental credits of recycling PP, the results show that environmental credits given to the system will be slightly lower. For example, in the case of GWP, credits calculated are decreased from 1.45 to 1.26 kg CO2 eq. from 2016 to 2025. The reason for this is that environmental impacts of recycling of PP are lower than those of the production of virgin PP.

3.4.2. Closed-loop recycling

3.4.2.1. Description of a closed loop recycling model

In this section, the closed-loop (CL) recycling scenario in which it is assumed that plastic sheets are used in the same application (sheets) after they are recycled is investigated. This is presently not the real case in the base scenario (system B), but it is interesting for the discussion of results. This hypothetical CL recycling scenario is presented in Figure 3.9.

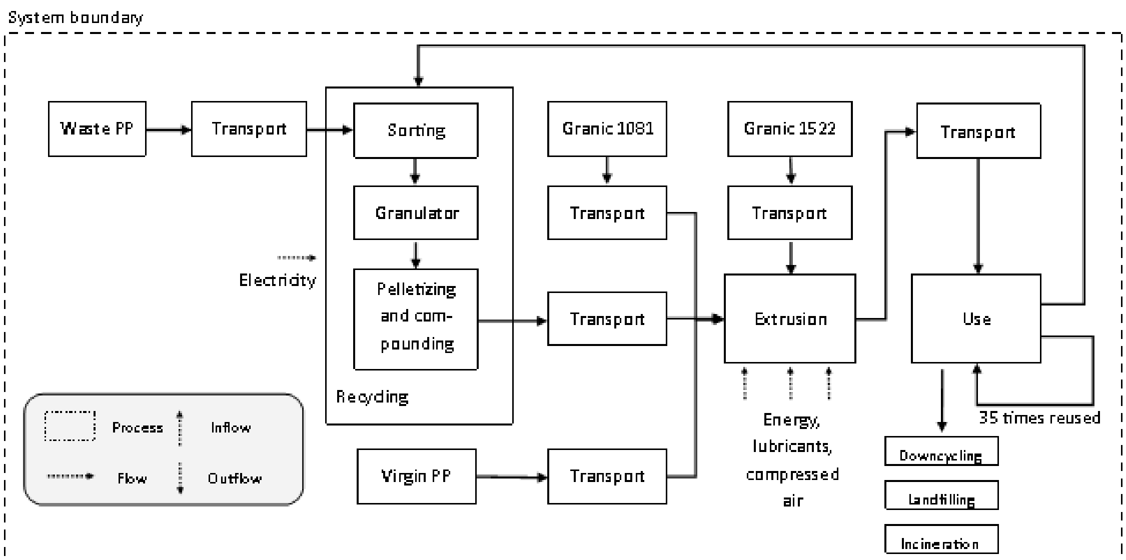


Figure 3.9. Closed-loop recycling model for plastic sheets

In the CL recycling scenario, it is assumed that recycled sheets are only able to replace the portion of recycled PP used in the composition of sheets. In order to meet with the requirements of the sheets, in each cycle, 54.6% of virgin PP is added. In addition, as recycled sheets in the CL system already contain some amount of minerals, in the model, the amount of both mineral fillers added are rearranged in each loop based on the amount coming from the recycled PP.

Since the percentage of recycled PP in the plastic sheets is defined as 23.5%, in the case of CL recycling it is not necessary to recycle all the sheets. Therefore, for the rest of the sheets, which are not CL-recycled, different EoL scenarios are considered: cut-off rule, downcycling, landfilling and incineration. In Table 3.7, investigated scenarios are briefly explained together with the assumptions done.

Table 3.7. EoL scenarios considered in closed-loop (CL) recycling

Scenarios	**Assumptions**
Closed-loop recycling with cut-off to outgoing waste (CL with cut-off)	• The portion, which is not going to be recycled inside the system, goes out of the system with neither burdens nor credits (according to cut-off-rule).
Closed-loop recycling with downcycling (CL downcycling)	• The portion which is not going to be recycled as sheets is assumed to be sent to downcycling to produce recycled PP granulates. • This scenario considers a reduction of material's quality and consequently a change in Q factor. For that purpose, a formula is proposed.
Closed-loop recycling and EoL landfilling (CL landfilling)	• The portion, which is not going to be recycled inside the system, is assumed to go to landfill.
Closed-loop recycling and EoL incineration with energy recovery (CL incineration)	• The portion, which is not going to be recycled inside the system, is assumed to go to incineration with energy recovery.

Proposed formula for identification of Q factor of plastic compound sheets for downcycling in closed-loop

In this study, since the sheets are composed of both recycled and virgin PP, it is assumed that CL recycled sheets are only used to replace a recycled portion of the sheet. Therefore, in each cycle, there is the same portion of virgin PP entering the system and a portion of sheets exiting the system to produce other products (Figure 3.9). Therefore, different than the common assumption of "in CL recycling, properties of recycled material are not very different from those of virgin" (Huysman et al., 2015), we believe that, in this case, quality loss should be considered for the portion which is leaving the system to go to downcycling.

Depending on the number of cycles taking place (n) in the CL system, the sheet will be composed of a mixture of the same amount of virgin PP, mineral fillers and n, n-1, n-2… times recycled PP. For example, after the first cycle, it will have recycled PP of two types: PP recycled once and PP recycled twice, but after the second cycle it will also have PP recycled 3 times, in addition to virgin PP and mineral fillers. Therefore, assuming that the quality loss is the same in each recycling cycle, to first time recycled PP a Q factor of 0.94 has to be applied, and to third time recycled PP a Q factor of 0.83 (multiplying three times 0.94). However, in this specific application, as the sheets are a mixture of different materials, Q factor of the flow leaving the CL system for downcycling can be calculated as follows:

If:

V: % virgin plastic in the product expressed on a per unit basis

G: % of Granic in the product expressed on a per unit basis.

R: % of recycled plastic in the product expressed on a per unit basis

Q: Quality factor of the recycled plastic

Q_n: Quality of the plastic after n times of recycling

t: Proportion of the recycled plastic to total plastic (recycled plus virgin) in the product (R/(V+R))

Then, after the first recycling of lamina, each component will become one more time recycled material. For example, the virgin material will become one-time recycled and recycled material used as raw material will be two-timed recycled material. In each cycle, the components will lose their property, except Granic (G). It is assumed that Granic does not lose its property since it includes a high percentage of minerals. Therefore, the Q factor of the sheet leaving the system after the first recycling in CL scenario can be written as follows:

$$Q_1 = G + VQ + RQ^2 \qquad (2)$$

After the second time recycling in CL, again virgin material entering the loop will become one-time recycled (VQ), the one-time recycled portion from the previous cycle (VQ) will go through its second recycling (tVQ^2) and two-times recycled material (RQ^2) will go to third recycling (tRQ^3). And the formula becomes:

$$Q_2 = G + VQ + tVQ^2 + tRQ^2 \qquad (3)$$

The same applies to the third recycling in CL. After the third recycling, in addition to what is explained before, three-times recycled material will turn into four-time recycled material. And the formula can be written as:

$$Q_3 = G + VQ + tVQ^2 + t^2VQ^3 + t^2RQ^4 \qquad (4)$$

The same continues to happen until the end of recycling cycles. Therefore, after the n[th] recycling:

$$Q_n = G + VQ + tVQ^2 + \cdots + t^{n-1}VQ^n + t^{n-1}RQ^{n+1} \qquad (5)$$

According to the Eq. (5) it is observed that Q_n follows a mathematical series. Therefore, the following Eq. (6) is proposed to estimate the Q factor of the recycled material, which is purged in a CL system with downcycling, depending on the number of cycles it undergoes. It assumes that the Q factor for the Granic fraction is 1 and it is only valid if the product is composed of both recycled and virgin parts (if no virgin material was used, Q_n would be equal to G).

$$Q_n = G + V\sum_{i=1}^{n}\left(\frac{R}{R+V}\right)^{i-1} Q^i + \frac{R^n}{(R+V)^{n-1}} \quad Q^{n+1} \qquad (6)$$

If $n \to \infty$, meaning that CL recycling system is in balance, the Q factor (Q_∞) of the CL recycling system can be estimated by taking the limit of the Eq. (6) as follows;

$$Q_\infty = \lim_{n\to\infty} Q_n = \lim_{n\to\infty}\left(G + V\sum_{i=1}^{n}\left(\frac{R}{R+V}\right)^{i-1} Q^i + \underbrace{\frac{R^n}{(R+V)^{n-1}}Q^{i+1}}_{\to 0}\right) \qquad (7)$$

While $n \to \infty$, the last part of the Eq (7) goes to 0.

The general rule to find the number that the above series converges is as follows, for any x complying with |x|<1:

$$\sum_{i=1}^{\infty} x^i = \frac{1}{1-x} - 1 \qquad (8)$$

By slightly modifying the series in Eq. (7) to deliver Eq. (9) and using the general rule given in Eq. (8) to deliver Eq. (10), the sum of the series can be calculated as Eq. (11):

$$\sum_{i=1}^{n}\left(\frac{R}{R+V}\right)^{i-1} Q^i = \left(\frac{R+V}{R}\right)\sum_{i=1}^{n}\left(\frac{R}{R+V}\right)^{i} Q^i = \left(\frac{R+V}{R}\right)\sum_{i=1}^{n}\left(\frac{RQ}{R+V}\right)^{i} \qquad (9)$$

$$\sum_{i=1}^{\infty}\left(\frac{RQ}{R+V}\right)^{i} = \frac{1}{1-\frac{RQ}{R+V}} - 1 \qquad (10)$$

$$\left(\frac{R+V}{R}\right)\sum_{i=1}^{\infty}\left(\frac{RQ}{R+V}\right)^{i} = \left(\frac{R+V}{R}\right)\left(\frac{1}{1-\frac{RQ}{R+V}} - 1\right) \qquad (11)$$

Therefore, if $n \rightarrow \infty$, the general formula for the estimation of Q_{∞} can be defined as in Eq. (12) or Eq. (13):

$$Q_{\infty} = G + V\left(\left(\frac{R+V}{R}\right)\left(\frac{1}{1-\frac{RQ}{R+V}} - 1\right)\right) \qquad (12)$$

$$Q_{\infty} = G + \left(\frac{QV\,(R+V)}{R+V-RQ}\right) \qquad (13)$$

By using Eq. (13), Q_{∞} was calculated for the fraction of plastic compound sheets leaving the system (using Q = 0.94 and the given composition of the plastic laminas (in section 2.1.1)). A $Q_{\infty} = 0.935$ was found.

3.4.2.2. Results for the different recycling models

Results of the CL recycling scenarios (Table 3.7) compared with system B (base-scenario) are presented in Figure 3.11.The results are given normalized to system B, which is OL recycling.

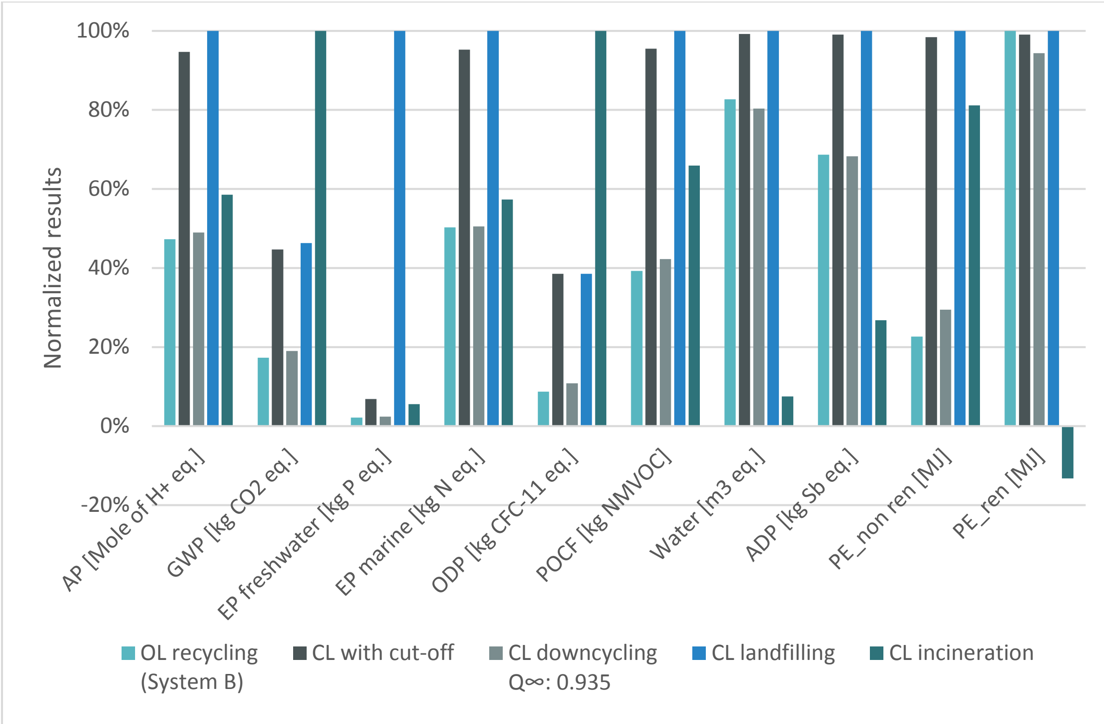


Figure 3.10. Comparison between base scenario (System B) and CL scenarios

Among the CL compared recycling scenarios, for most impact categories CL landfilling is identified as the scenario with the highest impacts.

The exceptions are ozone depletion and climate change. For those impact categories, CL incineration scenario is found to have the highest impacts. When plastics are incinerated, high amounts of halon derivatives (contributing to ozone layer depletion) are released together with high amounts of carbon dioxide (contributing to climate change).

CL incineration scenario has also been observed to have significantly lower impacts than the other scenarios for water depletion, resource depletion and primary energy demand from renewables. This lower impact is due to the credits gained from the production of electricity from plastic incineration.

CL with the cut-off is found to have the second highest environmental emissions since this scenario does not account for any credits from the waste getting out of the system. This comparison helps to point out the significant contribution of EoL crediting to the results.

CL downcycling and OL recycling scenarios (both without losing materials in landfills or incineration and with credits from materials leaving the system) have the lowest impacts for acidification, climate change, eutrophication (fresh and marine water), ozone depletion, photochemical ozone formation and energy demand from non-renewables, as they maximize the use of materials.

3.4.3. Eco-design of sheets in CL downcycling

In this section of the study, eco-design alternatives of sheets, containing less virgin material, are investigated. While doing this, it is assumed that the eco-designed options would be theoretically possible because the quality loss of the recycled PP was calculated as 6% (Q factor was estimated to be 0.94).

As shown in Figure 3.11, the fraction of virgin PP is changed from 0% to 50% by the interval of 5, in addition to 55.4%, which was the original percentage of virgin PP. The amount of recycled PP used in the composition of sheets is changed according to the amount of virgin PP, while keeping the amount of mineral fillers coming from Granic constant. For example, in the case of 10% virgin PP, the portion of recycled PP is defined as 68.1% and Granic is kept as 21.9%, as it was in the original design. For each point given in, by using Eq. (8), Q_{∞} for the material leaving the system is calculated and found as follows; 0.219, 0.610, 0.740, 0.805, 0.844, 0.870, 0.889, 0.903, 0.913, 0.922, 0.929 and 0.935, respectively. The number of uses, which is 35 in the original design, is assumed to be the same for all alternatives.

Results show that when the amount of virgin PP increases in the composition of the lamina, environmental impacts increase (Figure 3.11). However, it is observed that this increase does not present a linear trend. Although the impacts from virgin PP increase, the reason for this non-linear increase is identified as the increased amount of credits taken from the recycling of virgin PP due to its higher composition in the laminas. In addition, when the amount of CL recycled PP in the system increases, the amount of mineral fillers coming within the recycled PP increases, and this results in the less need for entrance of mineral fillers as raw materials to the CL system. Therefore, savings are achieved.

On the other hand, the smallest decrease can be achieved for primary energy demand from renewable sources. The main reason for this is the high dependency of both processes; recycling and production of PP, to electricity. For the rest of the impact categories, it is observed that each has a reduction trend following the difference between recycling and production of PP processes.

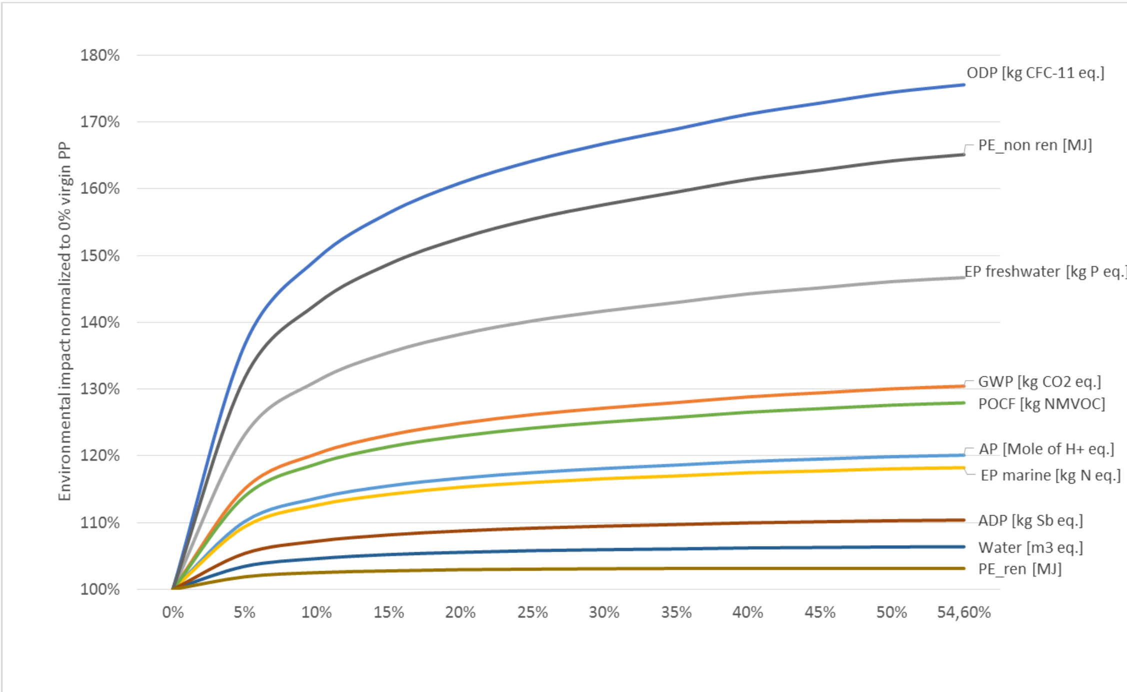


Figure 3.11. Results of eco-designed vs original sheets in CL downcycling scenario

3.4.4. Final discussion of results

The results of two baseline scenarios, wood sheet (system A) and plastic sheet (system B), are compared to OL recycling market mix substitution approach and CL downcycling, in Figure 3.12. CL downcycling represents the originally designed sheets (54.6% virgin PP), while in the case of eco-designed sheet nearly half amount of virgin PP (25%) is considered. In the figure, for comparison purposes, each scenario is represented as percentages in contribution to 100%. According to the results, for most impact categories wood sheet has higher environmental impacts than all other scenarios.

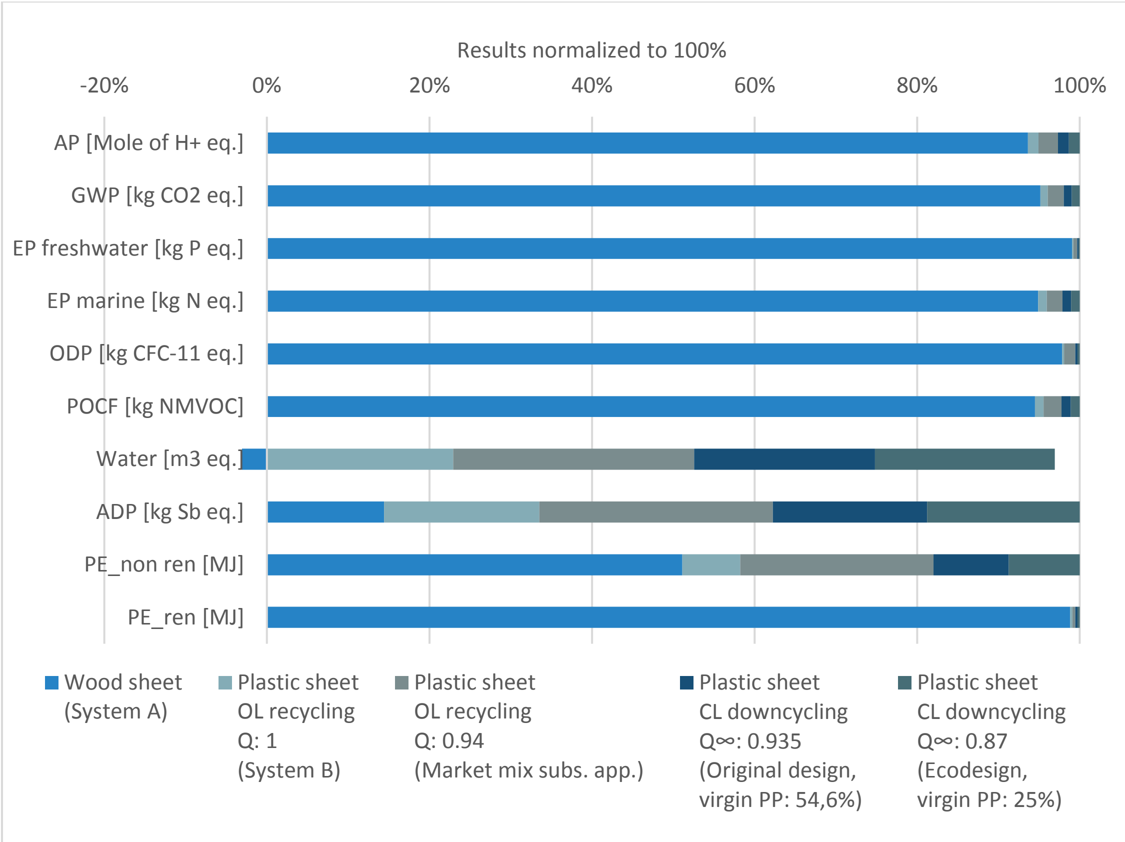


Figure 3.12. Comparison of alternative scenarios

In Figure 3.13, recycling scenarios, the results of the impact categories where eucalyptus wood sheets have much more higher impacts than plastic compound ones, are presented separately for better visualization. It can be seen that OL recycling with market mix substitution approach has the highest impacts for all of the impact categories. As this approach uses of Q factor of 0.94 instead of 1:1 substitution (System B) and considers the market share of recycled material, it gives fewer credits to the system and, therefore, more impacts.

On the other hand, system B has more or less the same impacts as CL downcycling with Q factor of 0.935 (less than 10% difference). This small difference is the result of the use of Q factor and avoidance of waste PP transportation in CL scenario. In addition, further impact reductions can be achieved through the eco-design of plastic sheets with Q factor of 0.87 (with 25% virgin PP).

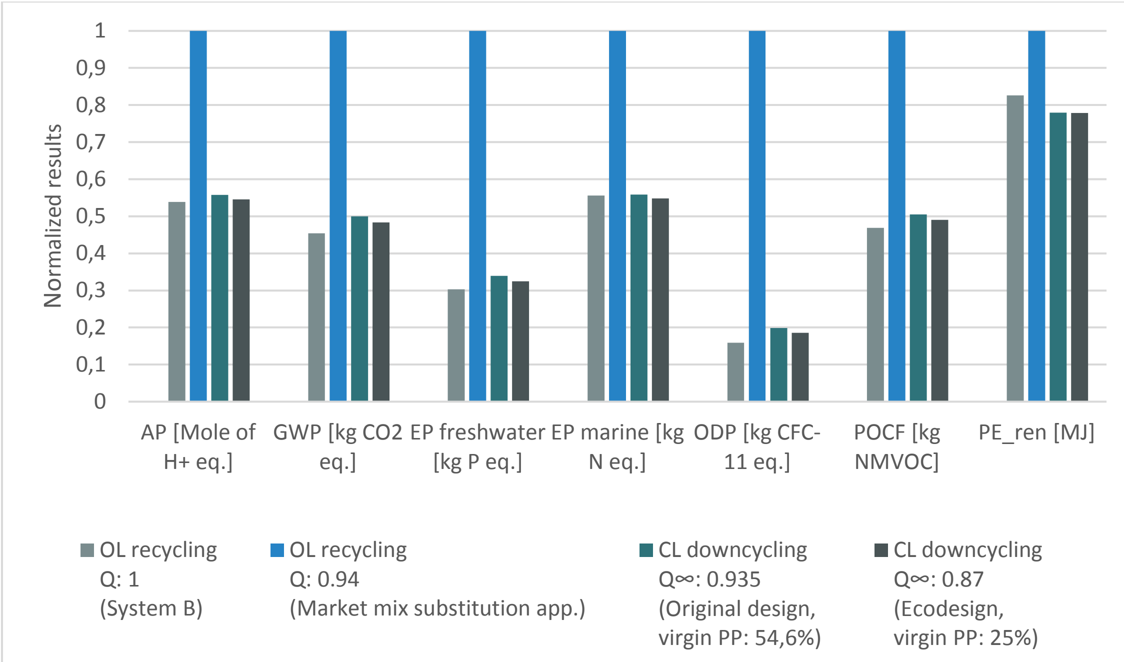


Figure 3.13. Comparison of recycling scenarios for the impact categories where wood sheets dominate

3.4.5. Methodological discussion

3.4.5.1. About the meaning of 100:100 EoL allocation method and its implications in decision-making process

In this study, we used the 100:100 EoL modeling approach because it favors both: the recyclability of products (by giving credits to the products that are recyclable) and the use of recycled material (by considering only the collection, transport and recycling burdens for the production of these recycled materials). Thus, emissions are estimated by considering the virgin and recycled content of the product and excluding the EoL credits from the recycled content (only credits from the EoL recycling of the virgin content are considered).

There are two approaches to EoL modeling at the policy level: one is to promote recyclability (through credits from avoided burdens) and the other to promote recycled materials (through cut-off rule). Depending on the society, one is preferable in front of the other. For example, in a well-trained community with high recycling rates, it is important to promote the use of recycled materials. In this case, the cut-off rule would be applied to secondary materials with the aim of promoting their use. On the other hand, for societies with low-quality secondary materials, it is more important to promote recyclability. In that case, cut-off may not be applied to the secondary materials, but the credits from the recycling of secondary materials would be given to the system. We think that it is better to benefit everyone to push industries to re-think their products to have a higher content of recycled material together with higher recyclability rates (and because you don't know how other products of the cascade will model the EoL). It is possible, using this approach, that some double counting occurs (ie. double counting of recycling processes, because their burdens will be allocated both to the product upstream of the cascade and to the product using the recycled material). But there is a precedent of double counting also in the carbon footprint scope 3 calculation described in PAS 2050 standard (BSI, 2011), where indirect emissions are allocated both, to the companies using raw materials and the companies producing those raw materials.

Therefore, the question to be asked is if we (the scientific community) want to contribute to a specific change in society then, among the internationally accepted EoL models, we have to choose our approach according to the change that we want to contribute to.

3.4.5.2. About the calculation of Q factors in open-loop and closed-loop recycling

Until now, in most of the studies, a Q factor of 1 is very commonly used by LCA practitioners. However, this is not what matches best with reality and the use of Qs/Qp factors is preferred whenever possible, thus, probably affecting the environmental results of a system. Results show that impacts have a trend to change opposite to the Q factor (Figure 3.7). However, knowing how to define the Q factor and how to apply it to the system is essential.

In this study, three different methods for the calculation of Q factors are analyzed. Among them, the authors believe that a Q factor based on mechanical properties represents better the reality, also in accordance with ISO 14044 standard, which gives priority to physical properties, whenever possible, instead of market values. In this case, the flexural modulus is taken as a reference mechanical property (the most representative property for the function of the product). Depending on the application of the materials, other mechanical properties can be taken into consideration. Q factors are important to more accurately estimate the environmental credits to the system. As it is discussed before, the use of a Q factor is vital to avoid over-crediting the system and to represent the real quality of materials. On the other hand, the use of a market mix substitution approach, in OL recycling, results in higher environmental impacts due to the fact that the environmental impact of the production of virgin PP is higher than the production of recycled PP. The use of this approach may result in giving even fewer credits to the systems when the proportion of recycled PP in the market increases. However, the authors believe that not only the use of Q factors but also the use of this approach may be useful to get more representative results of the systems with recycling.

In the case of CL recycling, a formula is proposed to calculate the Q factor of the flow leaving the CL system for downcycling. In this specific application, as the sheet is a mixture of different materials (virgin PP, recycled PP and mineral fillers), Q factor depends on the composition of each material in the mixture. If it was a single material sheet, in each cycle Q factor would decrease rapidly. However, in this case, since there is the entrance of virgin PP and mineral fillers, Q factor decays relatively slowly. As calculated in section 3.4.3, when the amount of virgin material decreases in the composition, the Q factor decreases. However; never reaches to zero as the mineral fillers are assumed not to lose their quality during recycling.

3.5. Conclusions and recommendations

The use of eucalyptus wood and plastic compound sheets as auxiliary storage material is environmentally compared by using the LCA methodology in the concept of circular economy, based on a Spanish case study. While doing this, taking the plastic compound sheets as an example and the 100:100 EoL allocation modeling, different methodologies for crediting recycling within the system are investigated: OL and CL recycling with the use of Q factors.

The results of the case study showed that plastic compound sheets (composed by 54.6% virgin PP, 23.5% recycled PP and 21.9% PP with mineral fillers) have fewer impacts than eucalyptus wood sheets in most of the evaluated impact categories, except resource depletion water and resource depletion mineral, fossils and renewables, whatever methodology is used for crediting the system. Even though wood is perceived to be more environmentally friendly when compared to plastics due to its renewability, this study showed some factors which help to reduce the environmental impacts of the plastic compound alternatives up to the point where, for the present Spanish application, they are better than conventional eucalyptus wood. There are three major reasons for this:

- The number of uses and lower weight:

Plastic compound sheets are used more times (35 times) than eucalyptus wood sheets (3 times) thanks to their better mechanical properties. Therefore, for the defined functional unit, fewer plastic sheets were required since they have a longer service life than wood sheets. Results of the sensitivity analysis show that plastic compound sheets present better results than eucalyptus wood sheets for most of the impact categories even if only used 1 time, except for 3 impact categories: resource depletion water; resource depletion, mineral, fossils and renewables and primary energy from non-renewable resources. This is mainly due to the lighter weight of plastic sheets (1.3 kg vs 5 kg).

- The use of recycled PP and mineral reinforced PP granulates:

Plastic compound sheets are composed of virgin PP, PP reinforced with talc, PP reinforced with calcium carbonate and recycled PP. The use of recycled PP and minerals like talc and calcium carbonate helps to reduce the environmental impacts of plastic sheets by reducing the amount of virgin PP used. Based on the information provided by the company, the addition of minerals to the plastics also provides better mechanical properties extending their service life.

- Recycling of plastic sheets when their lifetime is over:

Finally, onsite recycling of plastic sheets after being used at the facility of the same company provides environmental credits to the system and provides clear advantages to the company in terms of the application of circular economy in in-company borders. Onsite recycling assures 100% recycling and eliminates the transportation of waste plastics to a recycling facility, opposite to what occurs with wood sheets; which are nowadays landfilled. In addition, this option results in no waste production in the company and keeps the value of the resources.

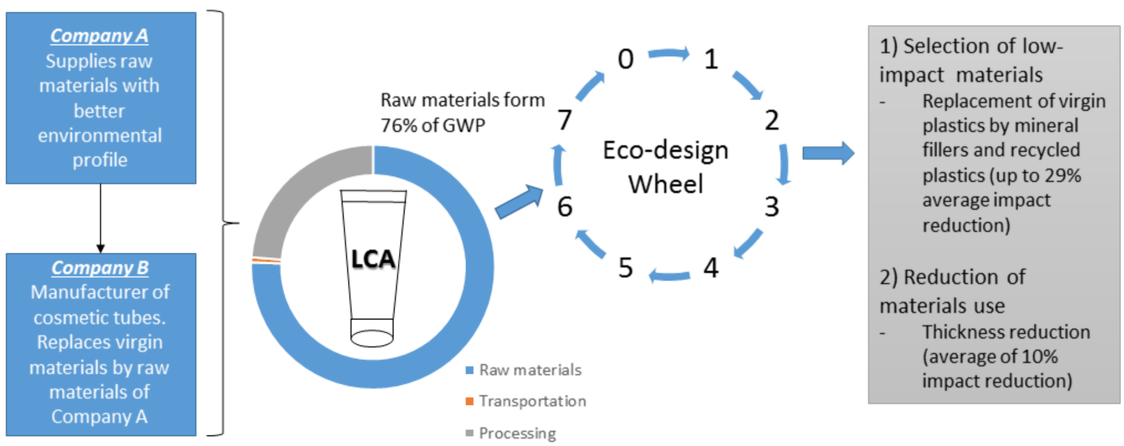
Methodological discussion on how to credit the system in LCA studies with recycling show that an assumption of 1:1 substitution of virgin PP by recycled PP may result in over-crediting the system. Therefore, the use of Q factors may represent the reality more accurately. Among the proposed methods, a calculation of the Q factor based on the mechanical properties of the material is defined as the most convenient one because it is based on physical properties and not in market prices (in accordance with ISO 14044).

Results show that methodologies for crediting the system of plastic compound sheets (OL and CL recycling with a Q factor) do not change the results, since the quality loss when recycling is low in this case study and there is a significant difference in the compared alternatives (wood and plastic compound). However, among the methodologies investigated, the OL recycling with a market mix substitution approach gives the highest environmental impacts, since it considers both the use of Q factor and market share of recycled PP.

When diminishing the portion of virgin material in the product (in an eco-design alternative), its environmental impact doesn't diminish linearly because, although the impact of virgin input is reduced, the credits from the recycling of this virgin portion are also reduced, thus compensating somehow the improvement.

The calculation of a Q factor for the recycled material leaving the system is proposed for the closed-loop option. And to this end, a formula is developed and offered to calculate the Q factor in compound materials for multi-loop recycling cycles.

Chapter 4. IMPROVING THE PRODUCTION CHAIN WITH LCA AND ECO-DESIGN: APPLICATION TO COSMETIC PACKAGING



This chapter has been submitted as:

Didem Civancik-Uslu, Rita Puig, Stephan Voigt, Dieter Walter and Pere Fullana-i-Palmer. Improving the production chain with LCA and Eco-design: Application to cosmetic packaging. *Submitted to Journal of Resources, Conservation and Recycling. Special Virtual Issue of Sustainable Cycles and Management of Plastics.* Under Review.

Abstract

One of the main drivers for companies to incorporate environmental improvements is economic benefit, either by obtaining a more valuable product or gaining new customers. Circular economy aligns environmental improvements with these drivers to achieve higher and quicker environmental benefits. In this paper, a case study of packaging eco-design aligned with a circular economy strategy along the production chain is presented. Life cycle assessment (LCA) was used to identify the product life cycle stages where eco-design would be more efficient. The results showed that, for the studied product, this life cycle stage was raw materials' production (virgin petrochemicals).

Virgin petrochemicals used in the composition of cosmetic tubes were therefore partially replaced by mineral fillers (calcium carbonate based) and post-consumer recycled plastics, to improve the environmental profile of this packaging. Samples of different options for the same cosmetic tubes were produced by collaboration between a company producing the plastic granulates and a company producing the cosmetic tubes. Cradle-to-gate LCA were performed on three different cosmetic tubes made of: 1) virgin high-density polyethylene (HDPE) and linear low-density polyethylene (LLDPE); 2) virgin LLDPE and mineral filler; and 3) post-consumer recycled HDPE and mineral filler. The replacement of virgin petrochemicals by mineral fillers helped to reduce the environmental impacts by an average of 12% and the use of post-consumer recycled plastic further decreased the emissions up to 29% for 6 out of the 9 evaluated impact categories. The technical requirements were fulfilled for all three compared samples and the option with better environmental performance was, in this case, also the one with lower economic costs. In addition, sensitivity analysis were performed on 1) end-of-life allocation method used and 2) thickness of the tubes.

This project promoted the circular economy downstream the production chain and helped to strengthen the bonds between supplier and customer.

4.1. Introduction

If environmental concerns are to be widely and quickly spread among companies, in addition to public awareness and consumers' willingness-to-buy drivers, corporate strategies must be encouraged to adopt a more efficient and sustainable use of natural resources within the company (Rieckhof et al., 2015) and all along the supply-chain (Geissdoerfer et al., 2018), following the principles of life cycle management (Fullana i Palmer et al., 2011). This goal must be implemented at strategic company level.

Usually, the relationship along the supply chain begins when a customer asks some kind of environmental improvement to its supplier. To reduce efforts (and to ensure quality) associated with this collaborative relationship, third-party product sustainability certifications appeared (Chkanikova and Kogg, 2018). This is an instrument for the transfer of significant life cycle information along the production chain and a tool to facilitate corporate life cycle management.

This type of client-provider relationship can otherwise begin with the interest of the provider to gain a new customer thanks to the best environmental profile of its product or material. In this last case, third-party product environmental certification may not be enough to produce the change, and a closer collaborative relation may be needed in the beginning; thus, including environmental, as well as technical and economic aspects of the new product, demonstrating its eco-efficiency (Laso et al., 2018). In this chapter (and up to the knowledge of the authors for the first time in the literature), a case study is presented on cosmetic packaging where a plastic granulate supplier starts a collaboration with a new customer by spreading its circular economy strategy downstream the production chain.

Cosmetic tubes are a type of packaging used for semi-liquid cosmetics, which preserves its content for longer time, prevents drying and helps correct dosing. This type of tubes is difficult to recycle because, when collected as waste, they still contain some cosmetic and, in the packaging selection plant, they are so small that they end up in the plastic-mix fraction. Thus, they are commonly landfilled or incinerated with the consequent emissions and resource losses.

In the present case study, cosmetic tubes were re-designed according to circular economy principles through the collaborative relation between a plastic-granulate provider and a cosmetic tube manufacturer, through the transmission of circular economy from the first company to the second, downstream in the production chain (opposite to the most common upstream transmission: from client to provider). LCA was used in combination with eco-design, as suggested by several authors (Muñoz et al., 2009; Navajas et al., 2017), to identify the life cycle stage environmentally most significant, where application of eco-design strategies would be more efficient.

The goal of this chapter is to see if it is possible to achieve a better environmental profile of a cosmetic tube by applying LCA and eco-design strategies. The alternatively proposed cosmetic tubes were produced, and their technical properties and costs were evaluated. LCA methodology was also used to calculate the environmental profile of each tube with the aim of identifying the one with least environmental emissions. The project took place with a close collaboration between supplier and customer-to-be.

Finally, some LCAs of cosmetic products focusing on the ingredients exist in the literature (Glew and Lovett, 2014; Martinez et al., 2017; Secchi et al., 2016) and evaluates packaging as a part of the full life cycle of a cosmetic product (Borghi et al., 2004, 2003; Fullana and Puig, 1997; Golsteijn et al., 2018; Molander et al., 2004; Vallés et al., 2001). Thus, this will be the first LCA of cosmetic tubes packaging in the literature, despite of its growing market (Future Market Insight, 2017).

4.2. Methodology

The circular economy principle applied in this case study is the one dealing with resource conservation and recycling of materials. The methodology used was based on the application of the eco-design strategy wheel (Brezet and van Hemel, 1997) taking into consideration the most impacting life cycle stage (according to LCA results), to see which strategies were applicable to cosmetic tubes. Then, we selected the strategies the manufacturer felt comfortable with, to produce new cosmetic tubes (described in section 4.2.1). Finally, the LCA evaluation of the newly obtained cosmetic tubes compared with the previous one was performed.

The LCA follows the ISO Standards 14040 (ISO, 2006a) and 14044 (ISO, 2006b). Methodological details on the LCA study are given in sections from 4.2.2 to 4.2.6.

4.2.1. Eco-design methodology

The eco-design strategy wheel was checked to find which strategies were applicable to the present product (Figure 4.1). According to the LCA results for cosmetic tubes (Figure 4.4) strategies related to raw materials production were identified as the most relevant ones. Thus, although other strategies in the wheel could be also applicable, the cosmetic tubes manufacturer decided to apply strategy 1 (the use of materials with lower impact) and later on, as a new improvement option, strategy 2 (reducing the amount of material) due to the improved mechanical properties obtained with mineral fillers.

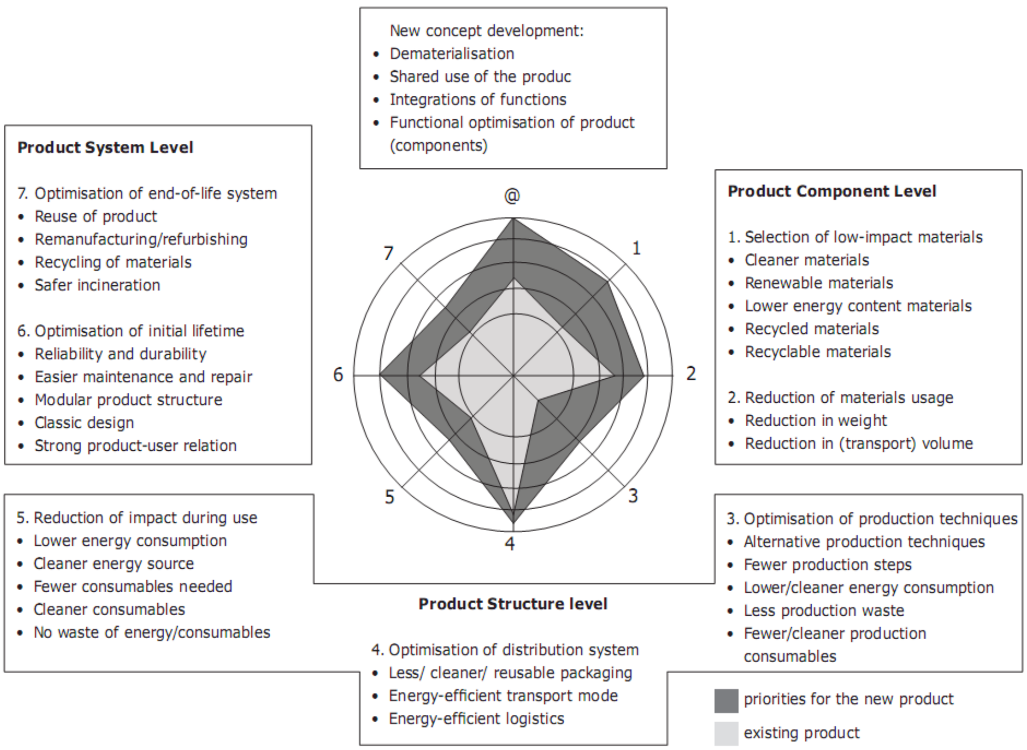


Figure 4.1. The eco-design strategy wheel (Brezet and van Hemel, 1997)

Materials with lower impact that can replace virgin petrochemical plastics are mineral fillers (based on calcium carbonate), which are present in nature in large amounts and are constantly being produced from CO2, and recycled plastics. Therefore, the virgin petrochemicals, which are high-density polyethylene (HDPE) and linear low-density polyethylene (LLDPE), were replaced by: a) mineral fillers, which are Granic products produced by GCR, a company in Tarragona, Spain; and b) post-consumer recycled HDPE at some percentages. The proposed replacements intend to reduce resource loss (as said, mineral fillers are very common in nature and recycled plastic is re-used in the techno-sphere). Therefore, three types of cosmetic tubes were produced, and their environmental impacts were evaluated

4.2.2. Goal and scope

The goal of the present LCA study is to calculate the environmental impacts of three different cosmetic tubes knowing that all of them are technically possible based on the information provided by the producer of the tubes, Linhardt (Linhardt, 2019), a packaging company placed in Hambrücken, Germany. All of the tubes have the same appearance (Figure 4.2) and the differences in weight and composition of the tubes are presented in Table 4.1.

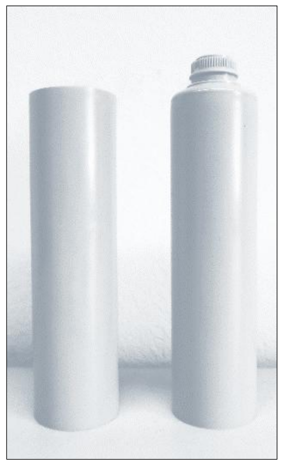


Figure 4.2. Cosmetic tubes without/with shoulder injected

Table 4.1. Weight and composition of tubes

	Weight	Body	Shoulder
Tube 1 (original)	10.4 g	HDPE (40%) LLDPE (40%) EVOH (8%) Adhesive (12%)	HDPE (100%)
Tube 2 (with mineral filler)	11.6 g	Granic 742 (45%) LLDPE (36.7%) EVOH (7.3%) Adhesive (11%)	HDPE (70%) Granic 742 (30%)
Tube 3 (with mineral filler and recycled content)	11.6 g	Granic 742 (45%) PCR-HDPE (36.7%) EVOH (7.3%) Adhesive (11%)	PCR-HDPE (70%) Granic 742 (30%)
**Granic 742: calcium carbonate-based filler* **PCR: post-consumer-recycled*			

The original tube, tube 1, was mainly composed of virgin plastics. In tube 2, HDPE was replaced by Granic 742, which is a mineral filler based on calcium carbonate (produced by GCR Group in Tarragona, Spain (GCR Group, 2018)). In addition to this change, tube 3 was made by also replacing LLDPE by post-consumer recycled (PCR) HDPE.

The functional unit of the study is: “Production of a single cosmetic tube having a 35 mm diameter and 120 mm length, and able to carry 75 mL of cosmetics”.

The corresponding reference flows relate to 10.4 g, 11.6 g, and 11.6 g of tube weight, respectively.

The system boundary of the study is defined as “cradle-to-gate” (Figure 4.3), from the extraction of raw materials to the gate of the production facility, to obtain the environmental profile of the three alternative tubes. For tube 1 and 2, each component is transported from different suppliers to the tube production facility in Germany. They are fed to the screw machine and the body part of the tube is co-extruded. Following that, the shoulder of the tube is produced by injection molding and combined with the body of the tube.

In the case of tube 3, for the production of recycled HDPE as raw material, washing, granulator, pelletizing, and compounding processes are considered. In each of the recycling processes, electricity is consumed. During the washing of the plastic waste before granulating, water and thermal energy are also used.

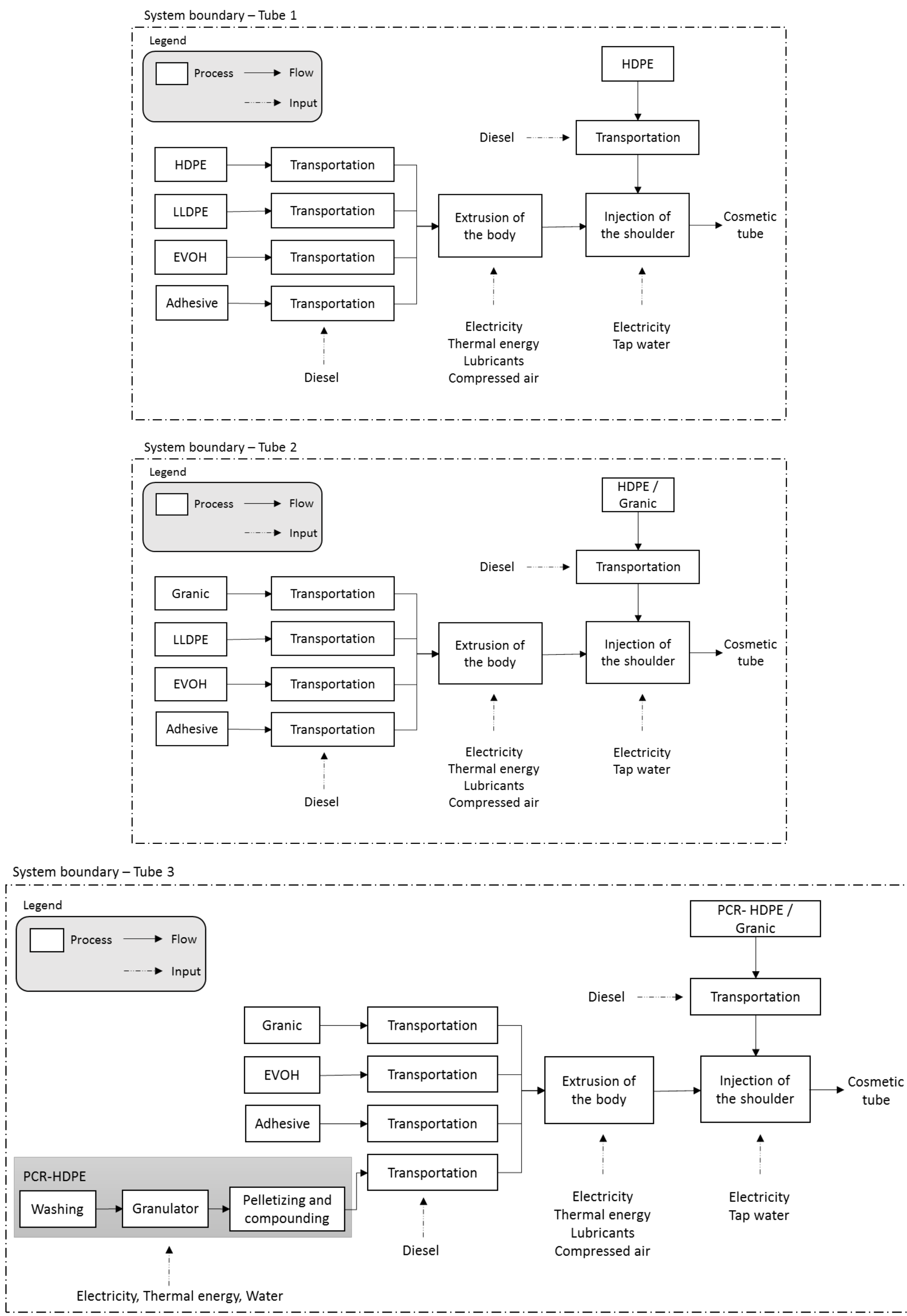


Figure 4.3. System boundaries of the tubes

4.2.3. Data collection

LCAs of the tubes were performed by using GaBi 8 software. The required data was intended to be collected from the producers of raw materials and manufacturer of the tubes. However, when data were not available, they were taken from life cycle databases.

Raw materials production

The life cycle inventory (LCI) data for HDPE and LLDPE were taken from Thinkstep Professional Database SP33 (Thinkstep, 2017), while the data for post-consumer recycled HDPE was taken from the producer.

Since ethylene vinyl alcohol (EVOH) production process is not available in the LCI databases, instead ethylene vinyl acetate (EVA) copolymer process was used as a proxy (Thinkstep, 2017), as EVOH is synthesized from EVA and they have similar profile (ETH Sustainability Summer School, 2011). In addition, Humbert et al. (2009) say in their study that the use of EVA as a proxy of EVOH shows minor changes in the results, which are below the defined cut-off criteria. For the adhesive, due to the lack of specific data, the closest adhesive available in the database, simple 2-component epoxy adhesive, was used as an approximation. Finally, for the mineral fillers, LCA results were taken directly from the producer

Transportation

Distances between the supplier of each raw material and the production facility, which is located in Germany, were identified and presented in Table 4.2.

Table 4.2. Distances between the raw material suppliers and the production facility

Raw material	**Distance (km)**
HDPE	361
LLDPE	256
Mineral filler (Granic)	1280
PCR HDPE	1175
EVOH	471
Adhesive	338

The transportation of the raw materials to the production facility was done by truck. The truck data were taken from Thinkstep Professional Database for a 32 tons capacity Euro 6 truck with 85% utilization rate.

Production processes

There were no primary data for the extrusion and injection molding processes. Therefore, LCI data were taken from Thinkstep Professional Database. The specific data for Germany were available for injection molding process, while not for extrusion. Therefore, the process used for extrusion was a global means. However, electricity, thermal energy, and lubricants consumed during the extrusion process were selected for Germany. Only compressed air consumption was the representative of global consumption as it was not available specifically for Germany.

For the recycling of HDPE as raw material, washing, granulator, pelletizing and compounding processes were considered. The processes were taken from Thinkstep Professional database. For the consumption of thermal energy and electricity, country specific data was chosen. At the washing process, European process water was used, while municipal wastewater treatment data for Germany was used.

4.2.4. Assumptions

The printing and final packaging processes of the tubes were excluded from the system boundary, as they are the same for all alternative tubes.

The end-of-life modeling of the tubes was not included in the LCA, as the aim of this study is to communicate the environmental profile of the tubes to the client (in this case, the cosmetics producer). Depending on the specific markets the tubes may be sold in, the waste management of the tubes will be different.

4.2.5. Impact assessment

For the estimation of the environmental impacts, International Reference Life Cycle Data System (ILCD)/Product Environmental Footprint (PEF) recommendation (v1.09) was used. The following midpoint impact categories were estimated:

- Acidification (AP) [mole H+ eq.]

- Climate change, excluding biogenic carbon (GWP) [kg CO2 eq.]
- Eutrophication freshwater (EP freshwater) [kg P eq.]
- Eutrophication marine (EP marine) [kg N-eq.]
- Photochemical ozone formation, human health (PCOF) [kg NMVOC]
- Resource depletion water (Water) [m3 eq.]
- Resource depletion, mineral, fossils and renewables (ADP) [kg Sb eq.]

In addition, the following indicators were also calculated:

- Primary energy from non-renewable sources (PE_non_ren) [MJ]
- Primary energy from renewable sources (PE_ ren) [MJ]

4.2.6. Allocation procedure

In the case of tube 3, the 100:0 allocation method (also known as cut-off approach), was used (Baumann and Tillman, 2004). Thus, the burdens of HDPE recycling were allocated to the tube which uses PCR-HDPE as raw material. On the other hand, knowing that end-of-life (EoL) assumptions may have important influence on the results (Civancik-Uslu, et al. 2018; Sandin et al., 2014), the use of 50:50 allocation, as being selected for the PEF methods (Allacker et al., 2017), was also considered as a sensitivity analysis in order to see the importance of the allocation method on the results. In 50:50 allocation, impacts of virgin material production and recycling process are allocated equally between the two products of a product cascade (A. L. Nicholson et al., 2009).

4.3. Results and discussion

LCA combined with eco-design, as a circular economy strategy aimed at reduction of resource use, was applied downstream in the production chain. Two new cosmetic tubes (tube 2 and 3) were produced and environmentally compared with the previous one (tube 1), through LCA methodology.

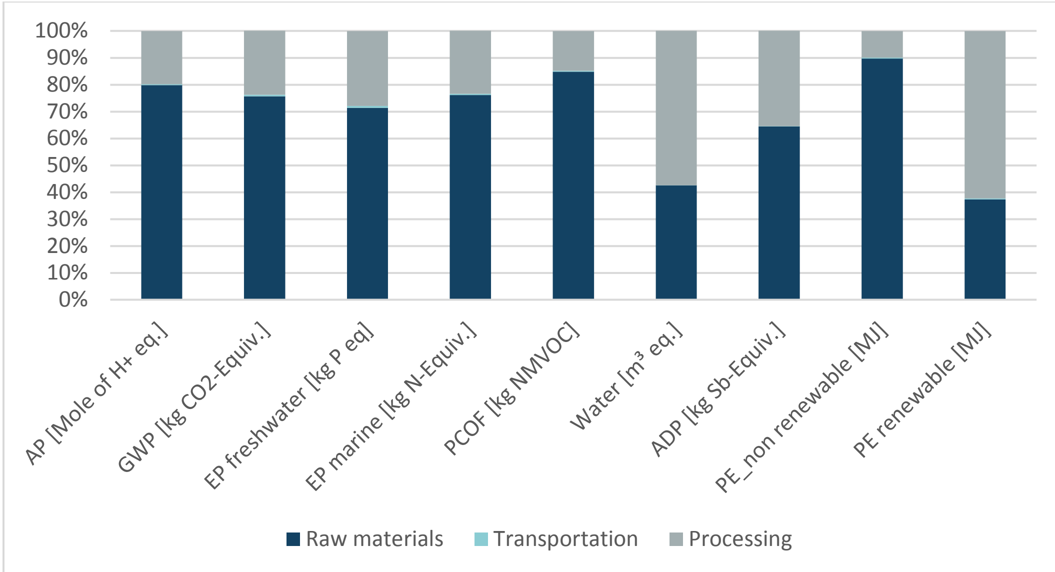


Figure 4.4. LCA results for tube 1

In Figure 4.4, the LCA results of tube 1 are presented in percentages for life cycle stages: raw materials, transport, and processing. “Raw materials” stage includes all the emissions from the activities related to the raw materials preparation (HDPE, LLDPE, PCR-HDPE, Granic, EVOH and adhesive). For example, in the case of PCR-HDPE, all the activities related to the production of recycled HDPE were included in the raw materials stage: washing, granulator, pelletizing and related electricity, and water consumption. The “Transportation” stage covers the emissions from the truck and diesel production. Finally, all the emissions occurring during the extrusion and injection molding processes are presented as “Processing” in the figure.

As seen in Figure 4.4, for most of the impact categories raw materials form more than 50% of the total impact. However, in the case of water depletion and primary energy from renewable sources, the most contributing part is the manufacturing of tubes, mainly due to the consumption of electricity.

Although the amounts of adhesive and EVOH used in the composition of the tubes are relatively small, their contribution to the impacts was identified as important. For example, in the case of freshwater eutrophication, their contribution reached to 46% of the total impact for tube 1. On the other hand, the impacts from the transportation of raw materials were found to be very low (up to 1 % of the total life cycle).

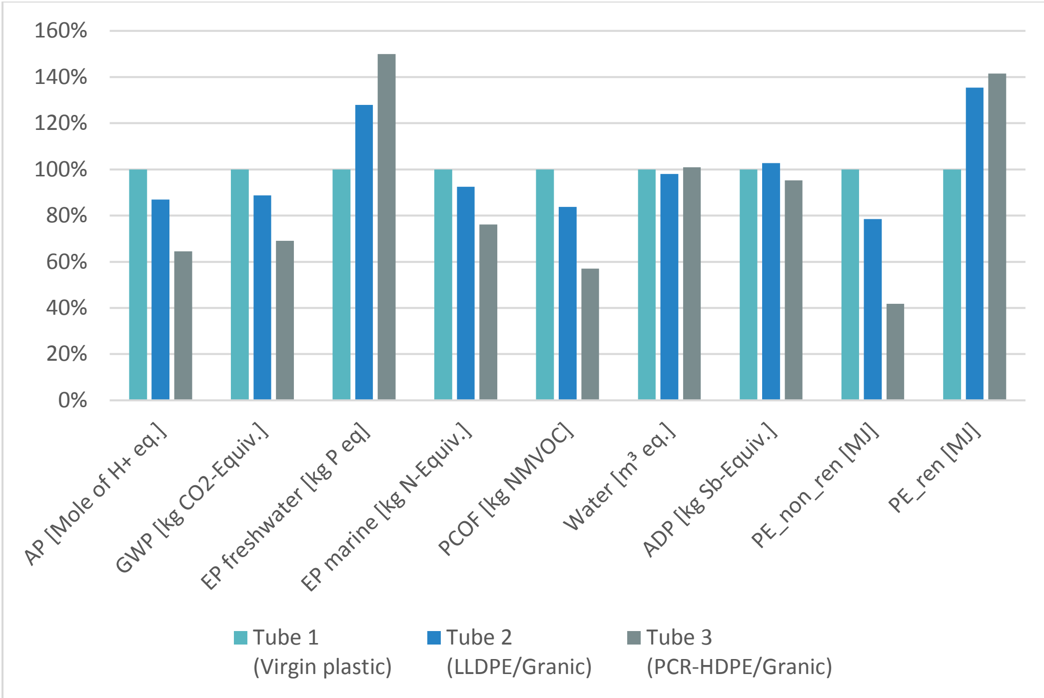


Figure 4.5. LCA results of cosmetic tubes

The results for the original tube (tube 1) are presented in Figure 4.5, in comparison to the proposed alternatives with mineral fillers and recycled content (tube 2 and tube 3, respectively). Comparing the results of tube 1 and 2, it can be observed that, when HDPE content of the original tube was replaced with mineral fillers (which is Granic 742 in this case), 6 out of the 9 evaluated environmental impacts decreased by an average of 12%. The reason for the improvement is the replacement of petrochemical-based raw materials by minerals, specifically calcium carbonate. On the other hand, 3 impact categories increased: EP freshwater (by 28%), ADP (by 3%). and PE renewable (by 35%). For EP freshwater and PE renewable, the reason for the increase was the mining of calcium carbonate, which significantly contributes to freshwater eutrophication and energy consumption. However, in the case of ADP, the slight increase happened during the manufacturing of the tubes. It is important to note that tube 1 weighs 10.4 g, while tube 2 weighs 11.6 g, due to the addition of the denser mineral fillers. This increase in weight causes higher consumption during the processing of the tube (electricity, lubricants, etc.), as the model proportionally relates electricity consumption to mass being extruded or injected, without taking into account that the filler may behave differently to heat and pressure than the plastic.

In the case of tube 3, in which LLDPE is replaced by post-consumer-recycled HDPE (PCR-HDPE) and the virgin HDPE by mineral fillers, even further reductions were achieved. In this case, for 6 of the impacts, a 29% reduction was gathered in average compared to the original tube. The use of recycled HDPE helped to reduce the emissions further compared to the tube 2. On the other hand, the results for EP freshwater, PE renewables, and water consumption increased (by 50%, 41% and 3% respectively). The reason for the increase in EP freshwater is the washing of post-consumer plastics during recycling and the production of calcium carbonate (used as filler). The last aspect also contributes to the PE renewables increase.

4.3.1. Sensitivity analysis

4.3.1.1. Weight reduction

The use of mineral fillers instead of virgin plastics results in the increase of weight of the tubes from 10.4 g to 11.6 g (see Table 4.1). Because of previous experiences in other products, it was known that, thanks to the improved physical properties of the polymer produced with mineral fillers, a reduction of the weight for tubes 2 and 3 was possible by reducing the thickness of the tube. Therefore, a sensitivity analysis was performed to see how the environmental profile of the tubes would change by reducing the weight from 11.6 g to the original 10.4 g (see this sensitivity analysis for tube 2 in Figure 4.6).

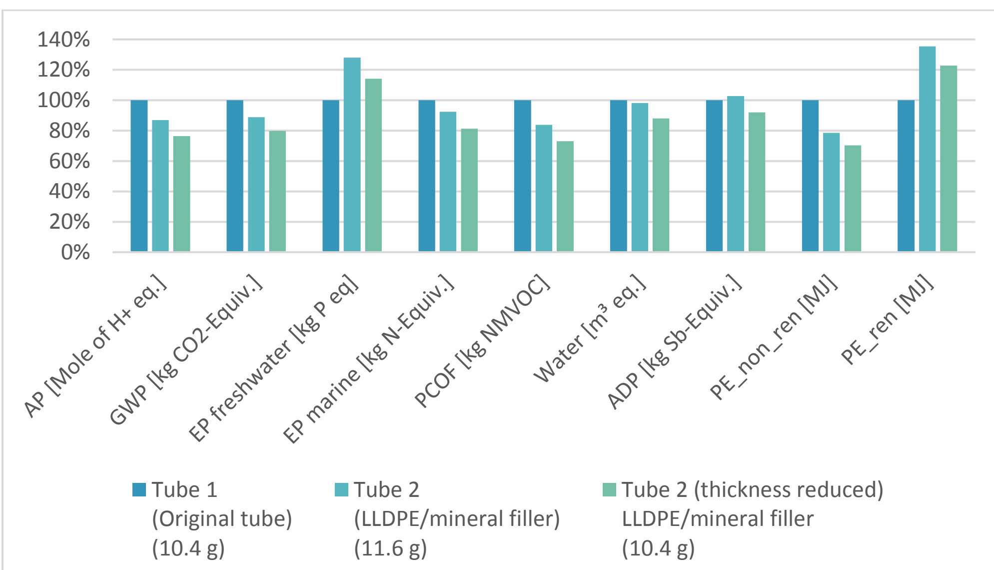


Figure 4.6. Sensitivity analysis on tube 2 weight

The LCA results showed that the decrease in the weight caused further reductions in the overall impact results (average of 10%). Using LCA to study different possible scenarios, is a good eco-design tool for packaging. For example, the tube-weight required to have all impact categories smaller than the ones of the original design could be determined with LCA, as well.

4.3.1.2. EoL Allocation

For tube 3, a sensitivity analysis was performed to see what would happen if the 50:50 allocation method was used instead of 100:0 for the modeling of PCR-HDPE as raw material. It means that 50% of the burdens due to the production of virgin HDPE and 50% of the burdens due to its recycling were allocated to the PCR-HDPE used as raw material in tube 3. This would also mean that, if the final user of the tube with the cosmetics inside would want to add the gate-to-grave life cycle stages, the same method should be applied to the percentage of tubes being recycled. As this is totally unknown to the tube manufacturer, at this point it seems more adequate to use the 100:0 rule as baseline scenario.

Results of the study are shown in Figure 4.7 together with the other tubes in order to see the effect of the allocation method. As expected, due to the emissions coming from the production of virgin HDPE, the results increased between 6% and 20% when the allocation method was changed to 50:50. PE from non-renewables impact category showed the greatest increase with a 20%, since it is an impact category highly affected by the consumption of virgin petrochemicals. On the other hand, despite of the smaller increase in EP marine and ADP, the change of EoL allocation made tube 3 to have more or less the same impact as tube 2. In this study, it did not cause a significant change on the overall results; however, it may in other cases. Even though one single study is not enough to make a general conclusion, it can be a good example as the change of EoL method caused 6- 20% difference in the results. As said above, if the tubes were recycled once used up, then the following system in the open-loop recycling will take the 50:50 rule as well, decreasing somehow the impacts of the tubes.

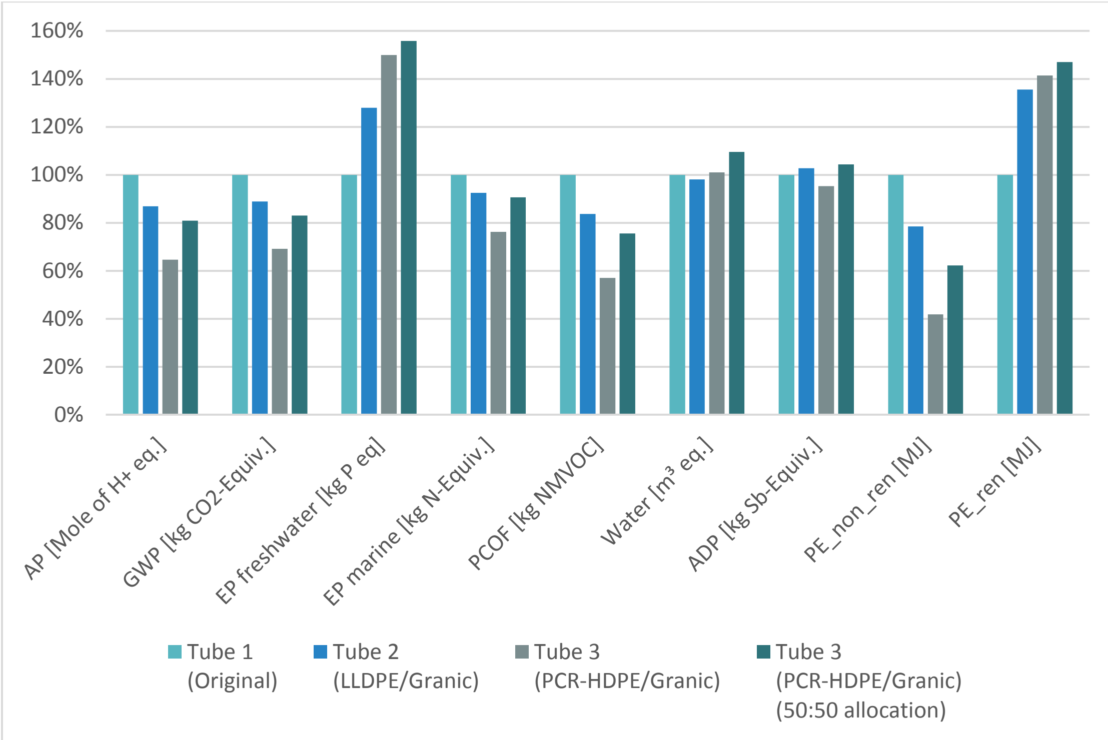


Figure 4.7. Results of the sensitivity analysis on EoL allocation method

4.4. Conclusions

Circular economy based on resource conservation was applied, not in-company, but along the production chain, by close-collaboration between a raw material supplier (with a strong internal circular economy strategy) and a possible-to-be customer. The product of the customer, a cosmetic tube usually landfilled or incinerated at the end of his life, was eco-designed by using both mineral fillers and recycled material in order to reduce the use of virgin petrochemical plastics. The results of the study showed that cosmetic tubes with less environmental emissions can be obtained by changing these raw materials at some portions and maintaining its technical feasibility and reducing costs.

The use of mineral fillers instead of virgin petrochemicals showed a clear advantage, reducing the environmental impacts by an average of 12%. In addition to that, if LLDPE was replaced by PCR-HDPE, environmental impacts further decreased by an average of 29% for most of the impact categories.

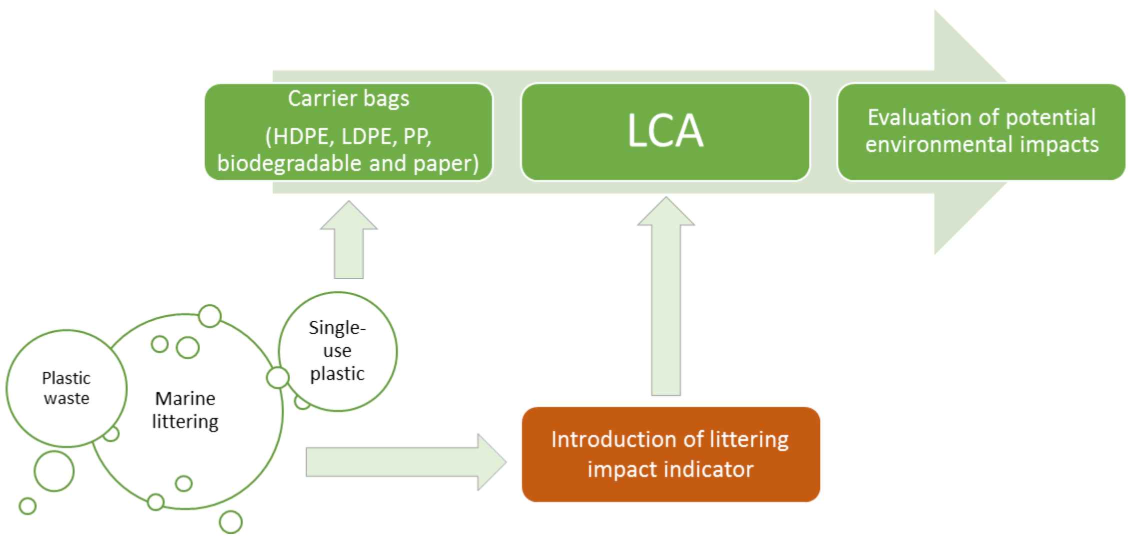
This study also showed the importance of raw material selection in the environmental profile of a product, based on a case study about cosmetic tube packaging. In most impact categories, the major contribution to the impact comes from the production of raw materials followed by electricity consumption from extrusion and injection molding processes. The study also showed that the contribution of EVOH and adhesives to the results is high, despite of their small amount in the composition of the tubes. Therefore, it is advisable to obtain better environmental data on EVOH and adhesives to improve the accuracy of the LCA model.

Using mineral fillers, due to their higher density, may result in higher weight of the tubes. Nevertheless, thanks to its superior properties, weight reduction was technically possible by reducing the thickness of the tubes; which brought a 10% further reduction in the emissions.

Two different EoL allocation methods were tested in this study, the 100:0 and the 50:50, to modeling the PCR-HDPE in tube 3. Impact results varied between 6% and 20%. In this case study, it did not affect the comparison, as tube 3 remained the best option for most impacts. However, it showed the importance of EoL allocation methods in product profiles.

LCA was used to identify the most contributing life cycle stages, and eco-design solutions were used where they could be more efficient. This showed the importance of combining LCA with eco-design. The project was a clear example on how the circular economy strategy can be transferred downstream in the production chain (from provider to customer) in a collaborative relationship, when the provider is the one pushing for the improvement instead of being the client, as it usually happens.

Chapter 5. LIFE CYCLE ASSESSMENT OF CARRIER BAGS AND DEVELOPMENT OF A LITTERING INDICATOR



This chapter has submitted as:

Didem Civancik-Uslu, Rita Puig, Michael Hauschild and Pere Fullana-i-Palmer, Life cycle assessment of carrier bags and development of an indicator of littering. *Submitted to Science of the Total Environment.* Under Review.

Abstract

Increased plastic consumption has resulted in high amounts of plastic waste ending up in the environment. Recently, the European Commission (EC) has identified a list of single-use plastics, including plastic bags, most commonly found in the European beaches. As a response, alternatives for plastic carrier bags have been more of a concern. Many LCAs have been performed to evaluate the environmental profile of different carrier bags; however, without considering the possibility of contribution to the littering problem. Therefore, in this study, an indicator has been introduced, based on an LCA study of carrier bags which was performed in Spain. The indicator is influenced by parameters such as: number of bags to fulfill the functional unit, weight, surface, fee, and biodegradability. In this paper, a comparative LCA of HDPE, LDPE, PP, paper and biodegradable plastic bags is presented. Following that, a littering indicator is introduced to allow a comparison of the risk of littering of the different carrier bags in marine environment. The results given by the Littering Potential indicator rank the bags oppositely to the results given by the LCA as usual. Further research is needed to refine the model and include additional contributing variables.

5.1. Introduction

The increase in the global plastic production and consumption has led to the accumulation of plastic litter in the environment, especially in the marine waters. Marine littering is defined as "any persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment" (UNEP, 2009).

A recent study (Jambeck et al., 2015) showed that, among 275 million tons of plastic waste generated in 192 coastal countries in 2010, 4.8 to 12.7 million tons of them entered the ocean. It is predicted that this number may double, if no improvements are done in waste management systems. Mismanaged plastic waste is identified as one of the main hotspots for macroplastic littering, especially in the coastal zones (UN Environment, 2018).

Marine littering can cause major impacts on ecological, social and economic values. In the case of ecological impacts, the individual organisms like marine mammals, reptiles, birds and fish, may entangle or ingest floating litter. Marine litter can also damage their habitats, like coral reefs, and extend the lifetime of rafting of organisms due to the longer lifetime of plastics. Seafood safety and related human health issues (microplastics), and loss of pleasure due to the environmental degradation (macroplastics) are the major social impacts of marine littering. Finally, marine littering is expected to have some economic impacts on fisheries and tourism activities, and an additional cost due to clean-up activities (UNEP, 2016).

Single-use plastics, including plastic bags, have been recently identified as one of the major contributors to marine litter (Steensgaard et al., 2017; Xanthos and Walker, 2017), in addition to other environmental impacts. In the EU, 100 billion plastic bags per year are currently used (EC Environment, 2017). Specifically, LDPE plastic bags are considered as one of the major sources of marine pollution (Steensgaard et al., 2017), as they are the most commonly used ones (Singh and Cooper, 2017).

A recent review study (Xanthos and Walker, 2017) showed that different measures are being used to reduce plastic marine pollution from plastic bags at legislation level: to ban the sale, to charge customers or to charge the stores which sell them. In Europe, Germany and Denmark pioneered the application of a ban in 1991 and 1994. Following that, since 2002, the other European countries have introduced different levies. In 1994, the Packaging and Packaging Waste Directive 94/62/EC was introduced, specifying reuse targets for plastic packaging. Following that, the European Commission suggested higher recycling targets for plastic packaging: 45% by 2020 and 60% by 2025 (European Commission, 2014). Later, in 2015, the amending Directive 2015/720 was introduced to reduce the consumption of lightweight plastic carrier bags. According to this Directive, members states are required to take measures to reduce the consumption of plastic bags to 40 bags per person annually by 2025 and conduct LCAs of bags (Steensgaard et al., 2017). Although the lightweight plastic bags are identified among the most commonly found items on European beaches, since there is already existing legislation on bags, the Directive on reduction of the impact of certain plastic products on the environment only envisions some measures regarding the extended producer responsibility and raising awareness for lightweight plastic carrier bags (European Commission, 2018).

Paper bags, biodegradable bags, reusable plastic bags, raffia bags and cotton bags are the alternatives to conventional one-use plastic bags. However, they pollute too. Moreover, they may have more impacts than conventional single-use plastic bags, depending on how they are being used. For example, a recent study done by the Danish Environmental Protection Agency (2018) showed that reusable low density polyethylene (LDPE) carrier bags, which are commonly available in Danish supermarkets, provide the lowest impacts for most of the environmental impact categories with regards to production and disposal among the other alternatives (PE, recycled LDPE, polyethylene (PP), recycled polyethylene terephthalate (PET), polyester, biopolymer, paper, cotton and composite bags). The study identified the reuse of LDPE bag as waste bin liner as the preferable end-of-life scenario. Finally, it was recommended that all bags should be reused as many times as possible. Another study, conducted in 2014 in United States, concluded the same: reusable LDPE bags have a better environmental profile than: a) a single-use HDPE bag with 30% recycled content (if reused 6.2 times), and b) a 100% recycled paper bag (if reused 1.7 times). However, the study claims that most of the users do not use LDPE bags a sufficient number of times (Kimmel, 2014). According to Greene (2011), in order to have less greenhouse gas emissions, in addition to be reusable, a plastic bag should contain some recycled plastics. In this study, a PE-based reusable bag with 40% of post-consumer recycled plastic was identified as the one with the lowest impacts. An LCA review study, conducted in 2006 and published in 2011 due to increasing debate on supermarket carrier bags, also identified high density polyethylene (HDPE) carrier bags as the most environmentally friendly option, basically thanks to their lighter weight. As the other studies reviewed, this study also points out the importance of a high number of reuses and its secondary use as waste bin liner (Edwards and Fry, 2011).

Recently, due to the increased debate on environmental impacts of single-use plastics (including plastic bags) and the marine littering problem, impacts of alternative carrier bags are being discussed. Although governments from all over the world performed LCA studies of carrier bags, none of the studies considered the effects of marine littering. Therefore, the aim of this paper is:

- To use a case study on plastic bags carried out in Spain to add some LCA methodological issues to this topic, including a fate model for a marine littering impact category.

The case study to be used is an LCA on supermarket bags conducted in Spain in 2008 by our research group with the aim of providing strategies to Spanish market about the use of supermarket bags (Fullana-i-Palmer and Gazulla Santos, 2008). Although performed 10 years ago, the results were very similar to those of the recent above mentioned Danish study (The Danish Environmental Protection Agency, 2018); therefore, it seems adequate to use it. In our study, an index for the qualitative comparison of littering risk of different types of bags was also developed, following the ISO 14044 clause 4.4.2.2.1: "… However, in some cases, existing impact categories, category indicators or characterization models are not sufficient to fulfil the defined goal and scope of the LCA, and new ones have to be defined." In addition, following the ISO 14044 clause 6, a critical review was performed (ISO, 2006b).

In this chapter, the littering model is shown and further developed, considering information more recently available, and the new strategies being defined by international organizations.

5.2. LCA case study on supermarket bags

5.2.1. Goal and scope

The goal of this study was defined as the identification of environmental impacts of the supermarket bags available in Spain through their life cycle. The following types of bags were investigated:

- Single use high density polyethylene (HDPE) bag
- Reusable low-density polyethylene (LDPE) bag (reusable for the same function)
- Reusable polypropylene (PP) woven bag

- Single use recycled paper bag
- Single use biodegradable Mater-Bi bag

The environmental analysis was carried out following the LCA methodology as defined in ISO 14040 (ISO, 2006a) and ISO 14044 (ISO, 2006b) standards. In addition, LCA results were supported by the definition of a qualitative indicator representing the littering impact of the bags. Although there was no consensus on the identification of the littering impact and knowing that it was considered as a difficult topic, the authors already believed at that time that a study on supermarket bags without considering this factor would not have enough credibility. The definition of the indicator will be explained in chapter 5.3.

5.2.1.1. Function, functional unit and reference flow

The primary function of supermarket bags is to help the customer of the supermarket to transport the purchased goods to the place of consumption. In addition, this function can be completed by the possible reuse of the same bag for the same or another purpose like collecting the domestic waste. It was known that, when this study was performed, 61% of the population was reusing the supermarket bags to collect domestic waste (Cicloplast, 2004). This second function was only considered for HDPE and biodegradable bags, since LLDPE and PP based bags were broken after reuse and the properties of paper bags are not suitable for this function.

The characteristics of the five analyzed supermarket bags are given in Table 5.1. When the study was conducted in 2008, plastic bags were available at the supermarkets in Spain; however, the paper and biodegradable bags were not, and the data about them were obtained from Canada and France, respectively.

Table 5.1. Characteristics of the analyzed supermarket bags

High density polyethylene bag (HDPE)	
Weight (g)	7.62
Dimensions (cm)	25 x 40
Volume (L)	13.75
Maximum load (kg)	9.25
Thickness (µm)	23.44

Reuse	Single use
Composition	10% recycled
Source	Samples from Spanish producers
Low density polyethylene bag (LDPE)	
Weight (g)	43.2
Dimensions (cm)	46.5 x 45.5
Volume (L)	29.3
Maximum load (kg)	-
Thickness (µm)	-
Reuse	Reusable
Composition	100% virgin
Source	A sample bag from Carrefour
Polypropylene woven bag (PP)	
Weight (g)	226
Dimensions (cm)	37 x 51.5
Volume (L)	43.3
Maximum load (kg)	-
Thickness (µm)	-
Reuse	Reusable
Composition	100% virgin PP, 100% recycled paper (carton layer)
Source	A sample bag from Carrefour
Paper bag	
Weight (g)	55
Dimensions (cm)	29.9 x 43 x 17.5
Volume (L)	22.5
Maximum load (kg)	-
Thickness (µm)	-
Reuse	Single use
Composition	100% recycled
Source	Sample bags from IGA and METRO (Canada)

Biodegradable bag	
Weight (g)	12
Dimensions (cm)	26.7 x 36.5
Volume (L)	14
Maximum load (kg)	-
Thickness (µm)	-
Reuse	Single use
Composition:	50% starch, 50% polycaprolactone
Source	A sample bag from Ecolobag (France) (ExcelPlas Australia, 2004)

The functional unit of the study was identified as: "To facilitate the transportation of purchased food and drinks to an average household for one year, from the point of sale to the place of consumption".

According to the Panel of Food Consumption, based on a sample of 8000 households, 644.1 kg of food and beverages were being consumed per person per year in Spain. Based on the data provided by the Panel, each household was formed by 2.71 persons and the yearly shopping was being done in 17 monthly purchases in a year. From this data, it was estimated that each year 1745.51 kg of food and beverages per household were acquired and 204 visits were made to supermarkets, hypermarkets, stores, markets, etc. It was considered that the average density of the purchased products was 0.45 kg / L[2], so that each purchase acquires 8.56 kg of products, with a volume of 19.01 liters.

For the determination of the corresponding reference flow, the transportation capacity of the bags (in terms of weight and volume) and the number of reuses were considered. The following Table 5.2 shows the results obtained when calculating the number of bags needed for 204 annual purchases of products, taking into account both the maximum transportable weight (6 kg)[3] recommended by

[2] This value is calculated from the following information: a transport container of 88 m3 capacity and containing different types of product to be sold in a supermarket, weighs 40 tons if it is fully loaded (Transportes José Luís Garucho, 2005).

[3] Although HDPE bags can easily carry 10 kg.

the Health and Safety National Institute (INSHT, 1998) and the volumetric capacity of the bag (considering an 85% use) and the number of reuses (Fullana-i-Palmer and Gazulla Santos, 2008).

Table 5.2. Reference flows corresponding to the functional unit defined

Material	**Volume**	**Used volume**	**Number of bags per purchase**		**Number of uses in purchasing**	**Number of bags per functional unit (204 purchases in a year)**
			Based on volume (19.01 L)	**Based on mass (8.56 kg)**		
HDPE	13.75	11.7	2	2	1	408
LDPE	29.3	24.9	1	2	1	408
					5	82
					10	41
PP	43.3	26.8	1	2	1	408
					10	41
					20	21
Paper	22.5	19.12	1	2	1	408
Biodegradable	14	11.90	2	2	1	408

For simplification purposes, the results were presented for 10 reuses for LLDPE bags and 20 reuses for PP bags.

5.2.1.2. System boundaries

In this study, the total life cycle of the bags, including extraction of raw materials, transportation, production, distribution to the consumers and end-of-life, was considered (see Figure 5.1).

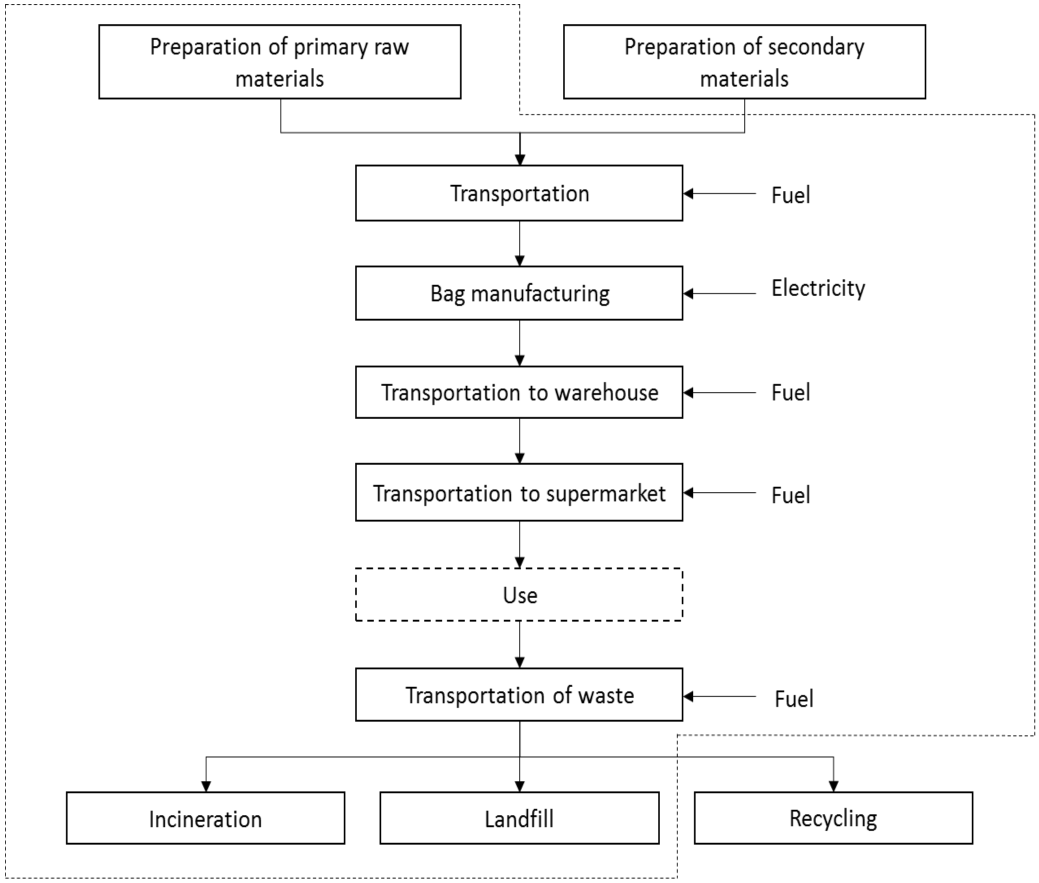


Figure 5.1. System boundaries

However, the following elements were decided to be kept outside the system boundary.

- The production of machinery and industrial equipment. They were not considered due to the difficulty of data gathering, and that it was commonly believed that their allocated impacts are negligible compared to the other elements in the system.
- Impacts from the use phase. They are negligible. They are caused by the transportation of the bag from supermarket to the consumer's home, with is much shorter than the other transportation processes involved in the life cycle.
- Recycling of waste. In this study, for the allocation of impacts, the authors accepted to use the rule known as "cut-off", which considers that the recycling of waste belongs to the second life of the material. For this reason, recycling of waste was not considered in the system boundary (explained in section 5.2.1.4 in detail); however, the recycling of the incoming secondary material was considered indeed.

5.2.1.3. Multifunctionality

When the study was conducted, 61% of the Spanish population was reusing the supermarket bags as waste bin liners (Cicloplast, 2004). This fact brings a new function to the system. Therefore, following the recommendation of ISO Standards, in this study, allocation of impacts is avoided by system expansion. To know the allocated the impacts associated with the main function of the bags (the transportation of goods), the burdens of waste bin liner production are subtracted from the system (Figure 5.2).

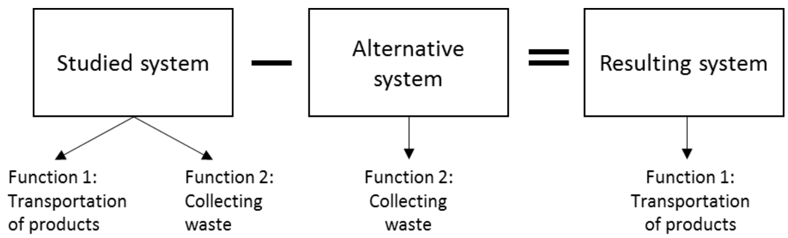


Figure 5.2. System expansion

5.2.1.4. Allocation

In this study, as the second life of the bags material was unknown in open-loop recycling, for the end-of-life allocation, the so-called “cut-off approach”, which assigns the recycling process to the next product cycle (Baumann and Tillman, 2004), was used.

Therefore, the burdens from the previous processes regarding the preparation of recycled material (transportation to the material recovery plant, separation and classification of recyclable materials, transportation to the recycling plant, washing and cutting to get recycled granulates) were allocated to the upstream system, while the credits from the EoL recycling of the bags were allocated to the downstream system. That’s the reason why, in the case of recycling, the system boundary was cut at the waste containers.

On the other hand, in the case of incineration and landfilling scenarios, waste management systems include energy recovery. In both scenarios, system expansion was used. The national energy profile of electricity production was not considered representative of the marginal changes that may occur in the demand. It is fundamental to correctly choose the technology displaced by the system, that is, the marginal technology of electricity production in Spain at that moment, which was identified as thermoelectric production from fuel/gas (MITYC, 2005; REE, 2006).

In the same way, within the production of paper, electricity and thermal energy are produced. Again, to avoid allocation, system expansion was performed as explained above.

5.2.1.5. Impact categories

The following environmental impact categories were decided to be used in the study among the ones recommended by a relevant operational guide to the ISO Standards at that time (Guinee et al., 2001):

- Abiotic resource depletion (ADP)
- Acidification potential (AP)
- Global warming potential (GWP)
- Eutrophication potential (EP)
- Photochemical ozone creation potential (POCP)

In addition, the following indicators were included in the analysis, as well:

- Primary energy consumption (PE)
- Water consumption
- Risk due to the abandonment of the waste bags on marine environment (explained in section 5.3)

5.2.1.6. Hypothesis and limitations of LCA

- It was a common behavior for the consumers not to fill the bags at their 100% capacity, especially if they were for free. Thus, it was assumed that the supermarket bags were filled at 85% of their capacity, both in terms of weight and volume.
- A theoretical number of uses was established for reusable bags, which was greater in the case of polypropylene (more resistant) than in that of LDPE.
- According to Cicloplast (2004), 61% of the population were using the supermarket bags to collect domestic waste. It was considered that only HDPE and biodegradable bags were

reused for this purpose, and at a maximum of 85% of its volume. This second function involves the proportional substitution of waste bin liners.

- In the case of PP bags, no environmental data was found on the production of braided PP. This material was assimilated to PP sheet. This was undoubtedly a hypothesis that benefits the results of the PP bag.
- In the case of the paper bag, it was considered that it was made of 100% recycled material based on Aspapel (2002).
- The location of bag producers, in the case of HDPE and LDPE was Spain, in the case of PP was China, and in the case of the biodegradable bag was Italy.
- For transportation in Europe, a Euro 3 truck with 17.3 tons of maximum load was used. For the distances above 700 km, it was assumed that the truck returned loaded with other products, thus it was out of the system. On the other hand, for the distances shorter than 700 km, a return trip was also included.
- For the end-of-life modelling, a generic Spanish waste management scenario, which includes recycling, incineration with energy recovery and landfilling with energy recovery from biogas production, was considered. Table 5.3 shows the percentage of different types of waste bags being treated through different options. In the case of bags having a secondary function of being used as waste bin liners, this waste management scenario applies after the second function. Thus, for the 39% of the bags, it applies after the first use and, for 61% after the second use, as waste bin liners.

Table 5.3. End-of-life treatment of carrier bags (Fullana-i-Palmer and Gazulla Santos, 2008)

Waste type	Recycling (%)	Incineration with energy recovery (%)	Landfill with energy recovery from biogas production (%)
HDPE bag	10.8	44.0	45.2
LDPE bag PP bag	19.9	13.7	66.4
Paper bag	57.3	4.4	38.3
Biodegradable bag	0	17.1	82.9
Wooden pallet	41.7	1.3	57.0
Cardboard boxes	57.3	4.4	38.3
Reused bags as waste bin liners	13.5	4.3	82.2

- For the transportation of the waste generated, three assumptions were taken: 1) all treatment plants were located at the same distance (50 km); 2) the packaging waste (wooden pallets and cardboard boxes) was transported directly from the supermarket to the corresponding treatment plant; and 3) the waste bags were transported by the consumer, on foot, to the city waste container.
- For the calculation of environmental loads from incineration with energy recovery and landfill scenarios, Ecoinvent database was used. In both cases, energy recovery was done through the gas or biogas produced from the incineration or decomposition of waste.
- Finally, it should be noted that, since no specific waste management models were found for the biodegradable material, the following hypotheses were adopted in terms of its behavior: the biodegradable material would behave like plastic in the case of energy recovery and like paper in the case of dumping (except for the different calorific value in both cases).

5.2.2. Inventory development

The data used in this study were collected from the GaBi database, installations or literature depending on the availability of the data. The inventories of five carrier bags are presented in Table 5.4.

For the raw materials, data regarding the production of HDPE, LDPE and PP granulates were taken from Gabi database 2005, while the paper production and granulate production data for biodegradable bags were provided by the producers. The composition of pigments, adhesives and dyes were gathered from the producers and modelled using the Gabi database.

In addition to the assumptions given in section 5.2.1 regarding the transportation, distances considered in the study are presented in Table 5.5.

The environmental impact associated with the manufacture of the bags was mainly due to the consumption of energy. The data on energy consumption during extrusion was gathered from the collaborating companies.

For the reutilization of the bags as waste bin liners, in the case of HDPE bags, it was assumed that for each 3.9 bags reused, the production of one garbage bag was avoided. The calculation was done as follows for the reference flow: 408 (number of bags) * 0.61 (reuse factor) * 13.75 L (volume) * 0.85 (fullness rate) = 2908.78 L. Assuming that each garbage bag has volume of 27.5 L, in total the production of 105 garbage bags was avoided. In the case of biodegradable bags, this number was found as 107 bags, doing the following calculations: 408 (number of bags) * 0.61 (reuse factor) * 14 L (volume) * 0.85 (fullness rate) = 2961.67 L and assuming the same capacity as above. The reused bags have the end-of-life scenarios presented in Table 5.3.

Table 5.4. Inventory for the carrier bags for functional unit

Raw materials	**HDPE bag**	**LDPE bag**	**PP bag**	**Paper bag**	**Biodegradable bag**
Virgin HDPE (g)	4.998				
Recycled HDPE (g)	0.762				
LDPE (g)		41.66			
PP (g)			135.6		
Pigments (g)	0.205	1.3	0.6		
Adhesives (g)	0.0054				
Dye (g)	1.65	0.24	1.8	2.1	0.2
Recycled paper (g)			88	49.7	
Glue (g)				3.2	
Biodegradable (g)					12
Packaging materials					
LDPE (g)	0.039	0.65	0.85		0.061
Cardboard (box) (g)	0.25	1.46	7.5	1.83	0.4
Wood (pallet) (g)	0.061	0.35	9.04	0.44	0.096
Energy consumption					
Electricity (kWh)	0.0044	0.020		0.0114	0.0055
End-of-life					
Landfill (g)	7.26	28.68	150	21.05	11.4
Incineration (g)	3.55	5.92	3	2.4	0.6
Recycling (g)	1.45	8.60	45	31.5	-

Table 5.5. Transport distances

	HDPE, LDPE, Paper bags (km)	PP bag (km)	Paper bag (km)	Biodegradable bag (km)
Transportation of raw materials				
HDPE	1500			
Recycled HDPE	400			
LDPE	1500			
PP		1500		
Pigments	530	1500		
Adhesives	530			
Dye	530	1500	530	500
Recycled paper		1500	750	
Biodegradable				500
Transportation of Packaging materials				
Cardboard (box)	200			
Wood (pallet)	200			
Transportation to supermarket	260	1773 (650 km truck in China + 16763 km ship + 360 km truck in Spain)	260	1500
Transportation to the treatment plant (End-of-life)	50	50	50	50

5.3. LCIA model on bags littering

In this section, a method for calculation of the littering indicator (land and marine) to identify the risk from abandoned bags on the environment is described. The model is proposed to be only used with comparison purposes. It is a simple approach to detect the bags that may have a greater risk of being abandoned and causing damage to the environment (impact on ecosystem and/or visual impact).

The first proposal of the littering indicator was done in 2008 as a part of the project of LCA of plastic bags in Spain (Fullana-i-Palmer and Gazulla Santos, 2008) based on different studies (ExcelPlas

Australia, 2004; Pwc-Ecobilan, 2004), and motivated by the critical review of the LCA study. In the present paper, that model has been revised and further developed also considering the latest proposal for European Legislation on reduction of plastics on the environment (European Commission, 2018).

It is assumed that littering is proportional: a) to the quantity of bags required for the same function (stated in LCA study); b) to the bags released to the environment c) to the dispersion of the bags on the environment and d) to the persistency of the bag's material.

Therefore, the characterization model is formed based on these four parameters (which are not usual LCI results), and their combination delivers the category indicator. The choice of the influencing parameters is based on the following reasons:

- P1- Quantity of bags. It refers to the number of bags which are required to meet the functional unit of the LCA study (the reference flow for each system being compared). It depends on the number of bags used and the surface area of one bag.
- P2 - Environmental release. It is the probability of the bag to being abandoned on the environment. For this parameter, the price of the bags at the supermarket was taken as the decisive contributor. For example, in the case of low-charge bags, the probability of abandonment by the consumer is expected to be higher than those of higher payment.
- P3 - Environmental dispersion. It is the bag floatability and the probability of flying out. For this parameter, the weight of the bag is the defining contributor. The lighter the bag, the higher the probability of flying.
- P4 - Environmental persistency. It is the persistency of the bag in the environment; in other words, for how long it will remain there after it is abandoned. For this parameter, biodegradability of the bag's material is chosen as related measure.

As said, the model was formed based on the four parameters above, which combined deliver the category indicator. The probability of the bags become litter is believed to be directly proportional to the number of bags required, while it is reversely proportional to the price, weight and biodegradability. Therefore, the index was defined as follows:

$$LP = \frac{P1^{f1}}{P2^{f2} x\ P3^{f3} x P4^{f4}}$$

Where:

LP = Indicator for assessing the littering potential on the environment

P1 = Quantity of residual bags

P2 = Environmental release

P3 = Environmental dispersion

P4 = Environmental persistency

f1, f2, f3, f4 = Weighting factors (all equal to 1, until further research inputs otherwise)

Where values are 0 < P1, P2, P3, P4 < 1.

The dimensionless parameters are calculated as follows:

$$P1 = \frac{(n\ x\ S)}{(n\ x\ S)_{max}}$$

Where:

n: Number of the bags corresponding to the functional unit

S: Surface area of one side of the bag (m^2)

$(n\ x\ S)_{max}$: Maximum result among the bags

- P2 can be calculated as;

$$P2 = \frac{p}{p\ max}$$

Where:

p: price of the bag (Euro)

$P_{\ max}$: Maximum price among the bags (Euro)

- P3 can be calculated as;

$$P3 = \frac{w}{w\ max}$$

Where:

w: weight of the bag (g)

$w_{\ max}$: Maximum weight among the bags (g)

- P4 can be calculated as;

$$P4 = \frac{d}{d_{max}}$$

Where:

d: environmental degradation rate of the material used in the bag (1/day)

d_{max}: maximum environmental degradation rate among the bags

The LP must be a number, so that results of different types of bags can be compared to each other. In the same way, the different influencing parameters and the mathematical operations combining them to get the category indicator must be numerical as well. The rules which were applied when defining the index are explained below:

- - All influencing parameters must be dimensionless for them to be combined. For that reason, while calculating each parameter (P1, P2, P3 and P4), each value is divided by the maximum result found.
- Therefore, no absolute impacts are pursued, but relative to one another. The introduced formula does not predict the impact on the final receivers, but it defines relative expressions.
- - Weighting factors (f1, f2, f3 and f4) exist to be able to include the importance of the parameters considered in the formula. In this study, all influencing parameters have an equal importance with a weighting factor of 1, as there is no proper research to assess these values so far.

5.4. Results and discussion

The results of the LCA study are presented in Table 5.6. Following that, the results calculated by using the LP impact indicator introduced in section 3 are presented in Table 5.7.

Table 5.6. LCA results of supermarket bags per functional unit

Impact	**Unit**	**Bag type**				
		HDPE	**LDPE**	**PP**	**Paper**	**Biodegradable**
ADP	kg Sb-eq.	9,67E-02	7,76E-02	1,54E-01	1,68E-01	5,34E-02
AP	kg SO2 eq.	2,89E-02	3,18E-02	2,01E-01	7,55E-02	4,84E-02
GWP	kg CO2 eq	9,32E+00	7,82E+00	2,42E+01	2,95E+01	1,45E+01
EP	kg PO4 eq.	2,90E-03	3,00E-03	1,36E-02	2,68E-02	1,12E-02
POCP	kg C2H4 eq.	4,70E-03	4,90E-03	1,55E-02	9,60E-03	5,40E-03
PE non-ren	MJ	2,26E+02	1,89E+02	3,55E+02	3,47E+02	1,35E+02
PE ren	MJ	1,13E+01	3,82E+00	7,21E+00	4,43E+01	1,64E+00
Water	kg	1,35E+01	1,56E+01	2,18E+01	1,30E+02	2,93E+02

Table 5.7. Results of the introduced littering indicator for the bags

Parameter	**HDPE bag**	**LDPE bag**	**PP bag**	**Paper bag**	**Biodegradable bag**
P1 – Quantity of bags	**0,77**	**0,16**	**0,08**	**1,00**	**0,77**
n	408,00	41,00	21,00	408,00	408,00
S (m^2)	0,10	0,21	0,19	0,13	0,10
n x S	40,80	8,61	3,99	53,04	40,80
P2 – Environmental release	**0,20**	**0,20**	**1,00**	**0,26**	**0,20**
p (Euro)	0,10	0,10	0,50	0,13	0,10
P3 – Environmental dispersion	**0,03**	**0,19**	**1,00**	**0,24**	**0,05**
w (g)	7,60	43,20	226	55,00	12
P4 – Environmental persistency	**0,01**	**0,01**	**0,01**	**1,00**	**0,99**
d (1/day)	0,1	0,1	0,1	13,60	13,40
LP (P1/(P2*P3*P4))	**15555**	**577**	**10,2**	**15,8**	**73,5**

For the calculation of the indicator, data was taken from the original LCA study (Fullana-i-Palmer and Gazulla Santos, 2008), except for the price, as there had been major policy changes, introducing mandatory fees. The average fee of PP bags was identified as 50 euro cents. HDPE, LDPE and biodegradable bags had the same fee of 10 euro cents. For paper bags, two different fees were found in the supermarkets, 10 and 15 euro cents. The average of them was taken as 12.5 euro cents.

For the calculation of P4, the study developed by California State University Chico Research Foundation, (2007), in which various laboratory tests were carried out following the protocols of ASTM D5338-98 (ASTM International, 2003), was used for the estimation of degradation rates of paper, plastic (PP) and biodegradable materials. In the case of a PP bag, having no data, the same rate as of PE bags was used, and, for the biodegradable bag the rate corresponding to "biodegradable materials based on corn" was used.

Table 5.7.shows the LP indicator results for the different bags. In the case of a bag being free of charge, P2 would equal to 0, which would make the indicator incalculable. To avoid this, it is recommended to use 1 cent instead of 0.

In Table 5.8 presents the LCIA results of the bags relative to the HDPE bag results, together with the results of the LP indicator.

Table 5.8. Comparison of the results of different bags (normalized to HDPE bag)

Impact	**Bag type**				
	HDPE	**LDPE**	**PP**	**Paper**	**Biodegradable**
ADP	1,0	0,8	1,6	**1,7**	0,6
AP	1,0	1,1	**6,9**	2,6	1,7
GWP	1,0	0,8	2,6	**3,2**	1,6
EP	1,0	1,0	4,7	**9,2**	3,9
POCP	1,0	1,0	**3,3**	2,1	1,2
PE total	1,0	0,8	**1,6**	1,5	0,6
Water	1,0	1,2	1,6	9,6	**21,7**
LP	**1,0**	0,0371	0,00070	0,0010	0,0047

LDPE and HDPE bags present better environmental results in most of the environmental impacts according to the scenarios considered for Spain. Both bags present equal impacts for EP and POCP and very similar result with AP (10% difference). On the other hand, LDPE bags present the best result in GWP, while biodegradable bags have the lowest impact in ADP and PE total. At the other extreme, PP bags and paper bags are those that generally have higher impact values.

Moreover, it is observed that the differences between the impacts of bags are remarkable. Depending on the impact category, the impact may change from 0.1 to 21.7 times. For example, in the case of water consumption, biodegradable bags have 27.1 times more consumption than the HDPE bags.

It should be noted that, if a smaller number of reuses for LDPE and PP bags were considered, since that would mean more units of bags to fulfill the functional unit, the results would be worse for them.

On the other hand, the risk indicator calculated for comparison purposes (LP) showed that single-use HDPE bags have the highest risk of littering, mainly as a result of being single-use, light-weight, cheap and non-biodegradable. It follows the LDPE bags. Since LDPE bags have higher number of reuses and weight, their risk of littering was estimated to be less than HDPE, but still higher than PP, paper and biodegradable bags. Although paper and biodegradable bags are single-use, thanks to their much higher biodegradability, the indicator gave small values for them. The best results were offered by the PP bag, because of the high weight and price compared to the other options, which makes it very difficult to be abandoned and dispersed on the environment.

While calculating the indicator, remarkable variations between the parameters of each bag were observed. For example, while calculating P1 (quantity of bags), although the surface area of the bags did not differ much, the number of bags required for the defined functional unit varied significantly depending on the number of reuses. In the case of P2 (environmental release), which considers the price, PP bags got the highest value compared to the others, as it has a relatively higher price than others. The weight of the bags showed a relevant variety while estimating P3 (environmental dispersion). For example, a PP bag weights 226 g while a HDPE bag weights 7.6 g. Finally, when calculating P4 (environmental persistency), biodegradability of materials showed difference between plastic bags (HDPE, LDPE, PP), and paper and biodegradable bags.

If the variation between the options for one specific parameter is high, this may also cause a wide variation in the final index calculated. For example, in the case of P2, if one of the bags has a very higher price compared to the others (PP bag), for the calculation of the final index, it is divided by 1 and result is not affected by this parameter. On the other hand, for other options which have relatively very lower price, the final index is divided by something very small, close to 0, and the indicator easily gets a very high value. The may be modified to avoid high influence by a specific parameter by giving values to the f factors representing the relevance of the parameter to the littering risk, which certainly is something to research in the future.

As the indicator does not calculate an absolute littering impact, but estimates relative risk between different bag options, the results can be interpreted as the proportion of bags contributing to littering problem in the environment. For example, according to the results shown in Table 5.7, one should expect to find 30 times more HDPE bags than LDPE bags on the environment or, for each 1000 units of HDPE bags found as littering, 7 units of PP bags should be found littered.

5.5. Conclusions

This study presents the results of an LCA of carrier bags which was conducted in Spain in 2008 by considering an indicator to assess the risk of marine littering of bags. Considering the usual impact categories, the results of the study showed that multiple-use LDPE bags were the ones that present the best environmental results in all impact categories, if they were used at least 10 times. Single-use HDPE bags, with a second use as waste bin liners, were the second best. Contrarily, multiple-use PP and single-use paper bags had the highest environmental impacts. On the one hand, the impact of the reusable bags (LDPE and PP) clearly depends on the number of used and the reuse of single-use bags (HDPE and biodegradable ones) to collect garbage represented an important saving of environmental impacts.

However, a feeling of incompleteness arises if no impact assessment on littering is performed. The littering indicator introduced in this study is a pragmatic formula to model littering impacts of carrier bags which are available at supermarkets. It calculates the relative risk between the different options, instead of assigning a final impact. It is a product specific indicator, since it calculates a value based on the properties of the bag. In the end, the bag with the lowest value gets the lowest probability of littering compared to the others.

For the calculation of the indicator, the presented formula considers the number of bags required to meet the functional unit of the study, surface area, weight, price and biodegradability of the materials applied to the bag. Among the bags within the LCA study, PP bags were the ones with lower risk of abandonment on the environment, while HDPE bags had the highest value. It was interesting to find out that, although the risk of littering for PP and paper bags was found to be very low, according to results of the LCA study, they had the highest impacts, while the opposite happened with the polyethylene bags. Thus, if policy making is focused on littering impacts, the decision to support a type of bag and ban another would be taken against science based and internationally agreed LCA results as usual.

Since the cleaning of environment from plastic is a difficult process, this method on littering assessment may provide a prevention-based solution in the life cycle of the bags. It may help to detect the one among the options with higher risk to end up in the environment.

5.6. Future research

In this study, the proposed littering indicator is based on the characteristics of the carrier bags. Therefore, one of the recommendations for future work would be to extend its application to the other types of packaging as well.

As a further step, in order to have a more sophisticated model, the inclusion of other physical and social parameters can be useful. For example, a plastic bag thrown away in a shoreline would have higher probability of contributing to marine littering than the one thrown kilometer away far from the marine environment. In addition, the regional waste management system in place would affect the littering risk differently than another. In places where a return-and-deposit system is applied, the probability of littering is expected to be somehow smaller than in those without, but it is uncertain how much this influences. Therefore, the question is knowing if other parameters would be relevant to introduce into the indicator, if the ones selected in this study are adequate, if weighting factors should be applied to give more importance to any of them, etc.

Finally, for the calculation of P4, the biodegradability of plastics under controlled composting was used, which may not represent the reality, e.g. in marine environments. Up to now, there is no standard defined to calculate the biodegradability of floating plastics in the marine environment and, due to the unavailability of data at the time when the LCA study was performed, biodegradability in composting was used for the calculation of the parameter.

Chapter 6. GENERAL CONCLUSIONS OF THE THESIS

6.1. Conclusions on methodological advancements in LCA

This thesis puts a light on some methodological issues in LCA, specifically in the case of plastic compound materials and packaging (meeting the objectives 3 and 4):

1. The 100:100 approach for open-loop end of life material recycling impact allocation was tested for intermediate products and its validity discussed.

2. Different methods to calculate the quality loss (Q, or Qs/Qp factor), in the case of compound material in an open-loop recycling system, have been introduced, instead of doing 1:1 substitution of primary by secondary material. Q factors should be linked to the most important property for the specific material application. In the case of packaging applications, this property was found to be the flexural modulus.

3. For closed-loop recycling, a formula has been developed for the calculation of the Q factor of material compounds, taking into account all components, alongside the material downcycling. In addition, other important LCA issues, such as end-of-life allocation and credits calculations, have been tackled.

4. The development of a quantitative impact indicator for littering within LCA impact assessment (using carrier bags as case study) has been performed. The use of a simple-to-use indicator can support the decision-making process of better eco-designing bags, considering the littering problem. Being a hot spot in LCA methodology development, this indicator intends to open a scientific debate about the introduction of an impact category on marine littering in LCA.

6.2. Specific conclusions on plastics with mineral fillers

A thorough literature review (Chapter 2) has been performed, delivering the following conclusions (meeting the objective 1):

5. The use of functional fillers has been identified as promising to reduce the environmental impacts of plastics, although each application is case specific and requires special research. The literature review found that plastics with functional fillers had smaller GWP results

compared to their virgin counterparts and reduced impact on resources, as fillers replace the virgin petrochemicals.

6. LCA has been identified as a good tool to analyze environmental impacts of plastics with fillers; as long as special attention is given to the following requirements:

   a) A proper functional unit, allowing to make proper comparisons between different materials, should be defined.

   b) Cradle-to-grave LCA should be performed to avoid problem shifting between different life cycle stages.

   c) Data on on EoL of plastic composites/compounds must be collected. Nowadays there is a data gap which makes the previous requirement important.

7. Although there were many studies about mechanical and physical properties, no published LCA studies of calcium carbonate as a plastic filler were found in the literature, despite of its common use in the industry. Therefore, the LCA case studies performed within this thesis work (specifically in Chapter 3 and Chapter 4) are the first ones (meeting objective 2).

8. A plastic compound was compared with eucalyptus wood for sheet application (Chapter 3). For this specific use, the former was found to be more environmentally friendly than the latter despite of its better environmental image. The strategies which contribute to achieve this result, following a circular economy perspective, are:

   a) A higher number of reuses;

   b) A lighter weight (less material) thanks to its superior physical properties.

   c) The use of upstream recycled PP and mineral filler reinforced PP, which have lower environmental impacts than virgin petrochemicals.

   d) Downstream closed-loop on-site recycling.

9. Cosmetic tubes were ecodesigned by following the eco-design wheel strategies (including the use of mineral fillers), and were compared with virgin PE tubes (Chapter 4), with a number of outcomes:

a) The raw materials stage was identified as the one with the highest contribution to the total impact of the virgin PP tube.

b) The replacement of virgin HDPE by HDPE with calcium carbonate-based mineral filler at some portions in the tube, reduced the environmental impacts by an average of 12% (meeting the objective 2).

c) In addition to that, the replacement of virgin LLDPE by post-consumer recycled (PCR)-HDPE provided a clear environmental impact reduction (up to 29%) (meeting the objective 2).

d) According to a sensitivity analysis, it was concluded that a thickness reduction applied to the tube would reduce up to an additional 10% of environmental impact.

e) Another sensitivity analysis showed that the choice of EoL allocation methods (100:0 and 50:50) causes a variation in the results (meeting the objective 3) from 6% to 20%.

f) Out of the whole working process with the production chain, it was clearly seen that a strong environmental commitment by the raw materials supplier can promote the development of new collaborations to produce products with better environmental profile (eco-designed products), as well as developing new business opportunities.

6.3. Limitations of the thesis and recommendations for future research

1. It is known by the production chain that, thanks to the better thermal conductivity of the mineral, plastics with mineral fillers get easily heated and cooled during processing; thus resulting in less energy consumption and higher production speed than conventional plastic processing. Unfortunately, no quantitative data on this fact was found or acquired during this research (in Chapter 3 and 4). If possible, in the future, specific consumption rates should be used when performing LCA.

2. In this thesis, only two mineral fillers have been studied: Calcium Carbonate and talc. In addition, for different applications, different physical properties may be used to define the FU. Only two applications (pallet separation sheets and cosmetic tubes) and one property (flexural modulus) have been investigated. More research is needed on this matter.

3. For the estimation of quality loss (Qs/Qp) of materials during recycling, the use of flexural modulus was recommended as it was the important property for this specific packaging application. For another application, another property may be used. While estimating the quality loss of the compound material, due to the lack of data, it was calculated based on the flexural modulus of PP.

4. Polypropylene (PP) and polyethylene (PE) with mineral fillers have mainly been investigated in the case of 2 packaging applications. In future research, the scope could be extended to biodegradable plastics with mineral fillers.

5. In this thesis (Chapter 5), the first quantitative littering impact indicator has been developed and sent for publication. However, it can be clearly improved in future research by solving several pending issues:

    a) Biodegradability in marine environments should be used instead the one from composting conditions.

    b) The fate of the bags from use stage to ocean littering may depend on other factors; for instance, the distance from home to the ocean or the type of waste management.

    c) The indicator doesn’t give an absolute number, like the usual impact categories, such GWP (kg CO2), etc., but a relative one to the bag with the higher impact.

    d) All parameters contributing to the indicator have been equally weighted, this having the same importance to the result, which is certainly not the case.

    e) The indicator is exclusive to supermarket bags and do not apply to other packaging, or beyond packaging (cutlery, fishing nets, cotton buds, straws, etc.).

6. Due to the lack of data for the parameters influencing the littering indicator of plastic bags with mineral fillers, this material was not considered in the case study. However, it can be assumed that plastics with mineral fillers would have a better biodegradability property than conventional plastic, thus promising better results in terms of marine littering. Future work can focus on estimating the risk for plastic bags filled with minerals, as well as investigating on how the fillers affect recyclability.

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