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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 study, 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.

Keywords: Sustainability, environmental impacts, LCA, composite, fiber, mineral

1. INTRODUCTION

1.1. Plastics with fillers

Because of their optimal cost and high performance, thermoplastics have been used in many different kinds of applications during the last few decades; and they have been even replacing other conventional 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 used, because they can provide other properties in addition to cost reduction (Rothon, 2001). 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, 2001).

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, $\text{Al}(\text{OH})_3$, 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). In this paper, in addition to polymer composites, polymer systems which are the mixture of polymer and additives will be subject to research. This kind of mixtures is referred as compounds.

Fillers can be grouped into two main categories: inorganic or organic ones. Then, they can be even further subdivided based on their chemical family as shown in Table 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 78.9% share (Grand View Research, 2016).

Table 1: Chemical grouping of fillers (Xanthos, 2010)

Groups	Examples
Inorganics	
Oxides	Glass, SiO ₂ , ZnO, Al ₂ O ₃ , Sb ₂ O 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

Ground calcium carbonate (GCC) is easily found on 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 inorganic filler in plastics 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 in packaging (Murphy, 2001). Among other inorganic fillers, silicates like mica, kaolin, and wollastonite can modify some mechanical properties of plastics. For example, the use of kaolin (mainly from kaolinite origin) in plastics has increased due to its coupling characteristics (Xanthos, 2010).

On the other hand, the mostly used fibrous fillers are 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 2, examples of NFs used as fillers in plastics are given classified in different categories.

Table 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 fibres
Seed/fruit	Cotton, coir
Grass Fibers	China reed, bamboo, grass
Wood Fiber	

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 its all 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 puts a methodological framework for doing this. 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 interpretation of results. In the goal and scope definition phase, the product or service to be studied is described and agreed, together with the purpose of the study. Many other choices regarding modeling of the system like system boundaries, environmental impacts to be considered and level of detail are also made in this phase. In the inventory analysis phase, the flow model is developed, data on resources and emissions from the system are collected proportionally to a functional unit taken as a reference (Baumann and Tillman, 2004). Later, in the LCIA phase, the environmental loads identified in the LCI are represented in terms of environmentally relevant impact information. At the end of the LCA, the results are assessed in order to analyze the quality of data, assumptions, and results to draw conclusions (life cycle interpretation phase).

1.3. New Materials and LCA

It is known that most manufacturing processes and products have negative impacts on the environment (Alves et al., 2009). 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 a product. To this end, the development of new

materials has increased to comply with trending sustainability goals. However, does their use always promise environmental benefits?

For example, some trending NFs applied to plastic composites have recently become very common, even replacing the use of inorganic fillers like 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 environmentally friendly. A thorough LCA of the resulting composite should be performed in order to decrease 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 to be developed when more data is 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.

On the other hand, plastics have become a major concern for marine pollution and based on the continuous growth of the market the risk of plastics reaching to the marine environment is increasing too (Law, 2017). Plastics in the marine environment are persistent and

transportable. The authors believe that the use of fillers in plastics may contribute positively to plastic marine littering problem since they reduce the amount of petrochemical used and they are natural. However, no studies were found in the literature on plastics with fillers in the marine environment. Recently, during the “Conferencia Internacional de Análisis de Ciclo de Vida en Latinoamérica” which took place in Medellin, Colombia in June 2017, The Medellin Declaration on Marine Litter in LCA was signed to encourage studies on LCA methodology to work on the marine litter (Sonnemann and Valdivia, 2017).

This paper aimed at identifying what has been published so far about the environmental impacts of plastic composites and compounds with functional fillers and reviewing them to understand if the use of fillers tends to decrease the environmental impacts. By doing this, it was also aimed to point out the gaps in the literature to put light to future research and provide life cycle approach to the researchers/industries who are working with plastic composites/compounds with fillers. Finally, we wanted to investigate how the LCA methodology has been applied to plastic composites.

1. METHODOLOGY

In this study, 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 mostly 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 end-of-life scenarios.

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 are only small amount of studies available in the literature; therefore, in this paper, 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 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%).

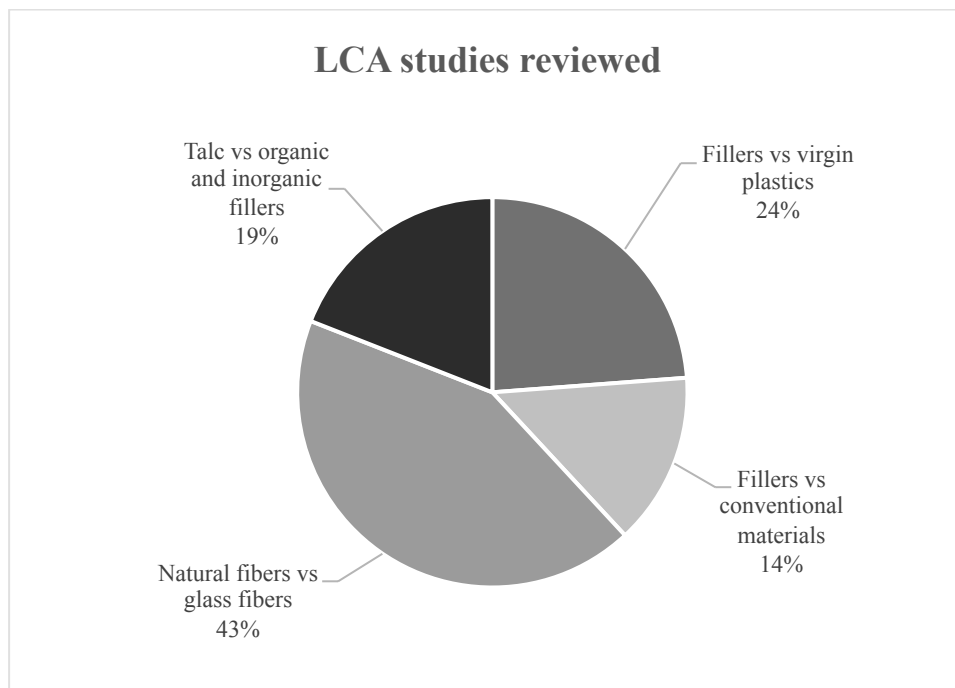


Figure 1: Distribution of LCA case studies reviewed

Three review studies regarding plastics with fillers were found in the literature previous to 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 bioplastics, 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.

3.1. Plastics with fillers vs virgin materials (comparative LCAs)

3.1.1. 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, 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 3. 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 3 (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 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, end-of-life scenario and sensitivity analyses (see Table 4). 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 end-of-life 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 amount 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 3: 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 4: Methodological analysis of LCA studies (Fillers vs virgin plastics)

Reference	Polymer	Functional unit (FU)	System boundaries/S 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-dinxveen, The Netherlands)" FU of agricultural film: "the amount of plastic film needed to	Cradle-to-grave/SimaPro	CML 2001 NREU	PP from APME Eco profile Production of Nano clay and nanocomposite by the Institute for Polymer Research (IPF, Dresden,	Incineration with energy recovery	-

		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"			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/Umber to	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

3.1.2. 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 5, together with its methodological analysis in Table 6. As it can be seen from Table 5, 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 aircrafts 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 fibre/Al (GLARE) used in aerospace panels were investigated with the help of LCA methodology (see Table 6). 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 6). It was observed that reductions in CO₂ and NO_x 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 6). 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 lightweight. 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 5: 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 end-of-life
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 6: 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 requiremeent 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)	-

* SA: Sensitivity Analysis

3.2. One filler vs other fillers 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 .

3.2.1. 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. 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 (Kinoshita 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; Penciu 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 7, methodological issues of the LCAs performed were summarized; together with their environmental evaluation in Table 8. 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 7, 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 7: Methodological analysis of LCA studies (NFs vs GFs)

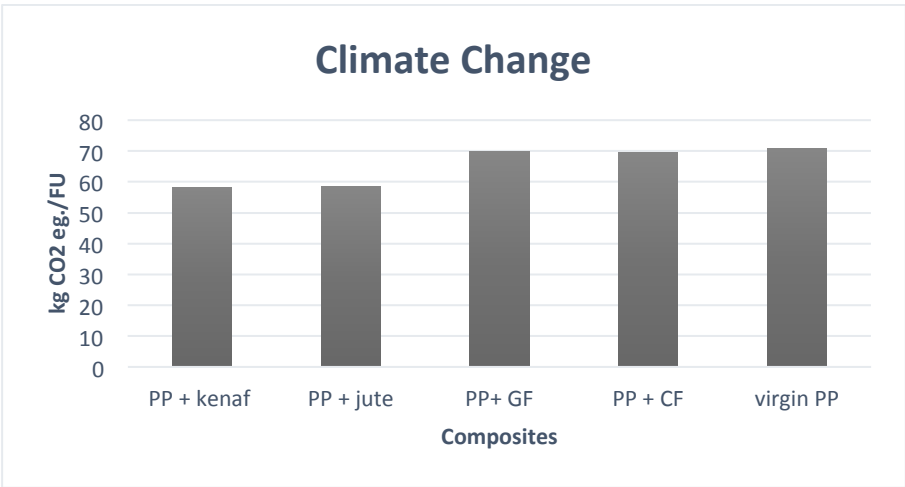
Reference	Polymer	Functional unit (FU)	System boundaries/ Software	Impact assessment	Inventory sources	End of life	Data quality assurance
(Alves et al., 2010)	Jutes fiber/polyester composites vs GF/polyester composites in automotive industry	“frontal bonnet of the buggy” in other words “the engine cover 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 console 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

(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 Manufactures 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	-

(La Rosa et al., 2013)	GF /thermoset composite GF -hemp/ thermoset composite	FU: "one elbow fitting used in the sea water 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 resolvable	-

Table 8: 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).
(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)	Cruaúá 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.



457 Figure 2: Results of climate change midpoint of composite materials for the FU of one plastic
458 pallet

3.2.2. 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 very similar mechanical properties.

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

485 environmental impact at the use stage, although at the material production stage it was worse
486 in terms of environmental emissions. However, in overall terms, hollow glass microsphere
487 reinforced PP was advantageous in terms of environmental and economic reasons. This is a
488 clear example (like others which are described in this review) of misuse of LCA. In order to
489 compare two options, they must have the same functional unit and this would mean that the
490 systems compared (reference flows) should have similar technical properties. This means that
491 the amount of PP reinforced with talc was over-dimensioned and should have been reduced
492 until the properties were as bad as those of the other composite.

Table 9: 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 (with some modifications for Qatar)	PE and PP will be recycled and in both cases, 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 Consumption water consumption and Landfill use	Commercial databases Private companies Literature	Landfilling Energy recovery in MSW Incinerator or cement kiln Mechanical recycling	SA* on the quality of recycled plastic

*SA: Sensitivity Analysis

3.2.3. Calcium carbonate filled plastics vs virgin plastics

Inorganic fillers have been used in plastic applications, mainly with the aim of improving heat distortion temperature, toughness, hardness, mould 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 polyvinylchloride (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.

3.3. LCA in new designs with plastics with fillers

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 GFR 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 end-of-life 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 end-of-life scenarios, LCA revealed some interesting points within other life cycle stages of the product.

3.4. LCA of end of life 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 end-of-life 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 end-of-life 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 (ABS), PVC, polyoxymethylene (POM), polyester (PES), polyurethane (PUR) 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 end-of-life scenarios; however, it is highly dependent on the substitution rate of polyolefins 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.

4. 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 study, 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 study 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 the study 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, four 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.
- End-of-life 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 end-of-life 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 end-of-life of the materials used.

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654

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