#### **REVIEW ARTICLE**



# Bioplastic from Renewable Biomass: A Facile Solution for a Greener Environment

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## Abstract

Environmental pollutions are increasing day by day due to more plastic application. The plastic material is going in our food chain as well as the environment employing microplastic and other plastic-based contaminants. From this point, bio-based plastic research is taking attention for a sustainable and greener environment with a lower footprint on the environment. This evaluation should be made considering the whole life cycle assessment of the proposed technologies to make a whole range of biomaterials. Bio-based and biodegradable bioplastics can have similar features as conventional plastics while providing extra returns because of their low carbon footprint as long as additional features in waste management, like composting. Interest in competitive biodegradable materials is growing to limit environmental pollution and waste management problems. Bioplastics are defined as plastics deriving from biological sources and formed from renewable feedstocks or by a variation of microbes, owing to the ability to reduce the environmental effect. The research and development in this field of bio-renewable resources can seriously lead to the adoption of a low-carbon economy in medical, packaging, structural and automotive engineering, just to mention a few. This review aims to give a clear insight into the research, application opportunities, sourcing and sustainability, and environmental footprint of bioplastics production and various applications. Bioplastics are manufactured from polysaccharides, mainly starch-based, proteins, and other alternative carbon sources, such as algae or even wastewater treatment byproducts. The most known bioplastic today is thermoplastic starch, mainly as a result of enzymatic bioreactions. In this work, the main applications of bioplastics are accounted. One of them being food applications, where bioplastics seem to meet the food industry concerns about many the packaging-related issues and appear to play an important part for the whole food industry sustainability, helping to maintain high-quality standards throughout the whole production and transport steps, translating into cleaner and smarter delivery chains and waste management. High perspectives resides in agricultural and medical applications, while the number of fields of applications grows constantly, for example, structural engineering and electrical applications. As an example, bio-composites, even from vegetable oil sources, have been developed as fibers with biodegradable features and are constantly under research.

Keywords Bioplastic · Biomaterials · Environmental Pollutiion · Biopolymer · Biodegradable polymers

# 1 Introduction

Today, bioplastic materials represent a valid alternative to the conventional plastics and their applications. Actually, the bioplastics market share is around 1% of the 370 million tons of total global plastic produced. But their annual growth rates hover around 30% until 2025. European Bioplastics

Sudip Chakraborty sudip.chakraborty@unical.it (EUBP)—the association representing the bio-plastics industry's defined "bio-plastic" as the biodegradable plastic materials and plastics produced from renewable resources. IUPAC defined bioplastic as a derivative of "biomass or monomers with plant origin, at some point of processing can be designed" (Vert et al. 2012; Plastics Europe 2021).

Plastic materials comprise polymers with relatively high molecular weight. They are typically produced by chemical synthesis processes. The term bioplastics is used to distinguish polymers that originate from renewable sources as biomass. The synthetic polymers are made from monomers by polycondensation, or polyaddition or polymerization, and

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most of them have a simpler structure than natural ones. They can be classified into four different groups: elastomers, thermosets, thermoplastics and synthetic fibers. The most communal synthetic polymers are polypropylene (PP); polyethylene (PE), acrylonitrile–butadiene–styrene (ABS), polycarbonate (PC), polyamides (PAs), polystyrene (PS), polyethylene terephthalate; polyvinyl chloride (PVC), polytetrafluoroethylene (Teflon), poly(methyl methacrylate) (PMMA), acrylic polyurethane (PU, PUR). Some of their applications are shown in Fig. 1, where the size of bubbles shows the relative importance. These plastics are traditionally petrochemically derived, but the demand for their production from renewable feedstocks is growing.

Theoretically, all usual plastics are generally degradable, but they have a slow breakdown, hence considered non-(bio) degradable.

Biodegradation of bioplastics depends on their physical and chemical structures in terms of polymer chains, functional groups and crystallinity, but also on the natural environment in which they are placed (i.e., moisture, oxygen, temperature and pH). Biodegradation is an enzymatic reaction catalysed in different ecosystems by microorganisms, such as actinobacteria (Amycolatopsis, Streptomyces), bacteria (Paenibacillus, Pseudomonas, Bacillus, Bulkholderia) and fungi (Aspergillus, Fusarium, Penicillium) (Emadian et al. 2017). There are different concepts of biodegradation. One very common degradation process is called hydrolysis. The hydrolysis mechanisms are exaggerated by diffusion of water through polymer matrix. Time duration for the degradation may vary for different material, such as polylactic acid, has very slow degradation which is about 11 months (Thakur et al. 2018). Moreover, the biodegradation rate be contingent on the end-of-life decisions and the physicochemical conditions, such as moisture, oxygen, temperature, presence of a specific microorganism, presence of light. The main end-of-life choices for biodegradable plastics include recycling and reprocessing, incineration and other recovery

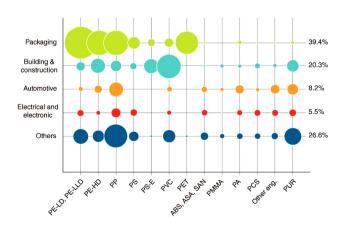


Fig. 1 Typical applications of polymers (Plastics Europe 2021)

options, biological waste treatments, such as composting, anaerobic digestion and landfill (Mugdal et al 2012; Song et al. 2009). The composting process represents the final disposition most favourable from an environmental point of view. The presence of ester, amide, or hydrolyzable carbonate increases biodegradation's susceptibility.

Bioplastics also do produce less greenhouse gases than that of usual plastics over their period. Therefore, bioplastics contribute to a more sustainable society.

Therefore, there are bioplastic alternatives to conventional plastic materials. It already plays a vital part in different fields of application. Bioplastics that are bio-based, have the same properties as general plastics and offer added advantages because they have a lesser carbon footprint on environment. Nevertheless, their low mechanical strength limits their application. Glass and carbon fibers are synthetic fibers commonly used to reinforce bioplastics, but they are not biodegradable. For this reason, they can be replaced by more environmentally friendly, abundant, and low-cost materials, such as lignocellulosic fibers and lignin (Yang et al. 2019). Other physical strengthening methods are the mold temperature increase, dehydrothermal treatment, and ultrasounds application. When applied to soy protein-based bioplastics, the thermal treatment enhanced the mechanical properties, the dehydrothermal treatment increased the superabsorbent capacity and ultrasounds lead to a structure with smaller pores. As a consequence, the treated bioplastics could be used in different applications (Jiménez-Rosado et al. 2020).

A new green one-step water-based process was proposed to convert vegetable wastes into biodegradable bioplastic films having similar mechanical properties with other bioplastics (Perotto 2018).

Recent trends indicate the biocompatible and biodegradable polyhydroxyalkanoates (PHAs) as alternatives to conventional plastics which has wide variety of thermal and mechanical characteristics (Khatami et al. 2021). PHAs are linear polyesters, produced by microbiological, enzymatic, or chemical processes, but their industrial production is still not cost-competitive (Medeiros Garcia Alcântara et al. 2020). Renewable and inexpensive carbon sources-such as macroalgae, peanut oil, crude glycerol, and whey-have been studied to reduce production costs (El-malek et al. 2020). Innovative research proposed the production of PHAs by a three-step process consisting of CO<sub>2</sub> reduction to acetate and butyrate by microbial electrosynthesis, extraction/ concentration of acetate and butyrate, and PHAs production from volatile fatty acids. This process meets the demand to decrease CO<sub>2</sub> emissions and convert a greenhouse gas to bioplastics (Pepè Sciarria et al. 2018).

Currently, researchers pay great attention to the production of biomass-derived next-generation advanced polymer, such as poly(ethylene 2,5-furandicarboxlate) (PEF) (Hwang et al. 2020; Algieri et al. 2013, 2012; Iben Nasser et al. 2016). Moreover, another very new trend investigates green microalgae cells as raw materials for the production of cell plastics (Nakanishi et al. 2020).

# 2 Bioplastic Materials

Plastics are polymeric chains composed of repetitive units or monomers linked together. These macromolecules are conventionally synthesized by polymerization, polycondensation or polyaddition reactions from fossil sources. Interest in competitive biodegradable materials is growing to limit environmental pollution and waste management problems. Bioplastics are a new plastic generation, defined as plastics originating from a biological system and produced from renewable feedstocks or by a range of microorganisms. Since they significantly reduce the environmental impact in terms of greenhouse effect and energy consumption, they are a challenge for a greener future.

Having different properties, bioplastic materials are classified in three main groups, as shown in Fig. 2:

- Bio-based or partially bio-based plastics;
- Bio-based and biodegradable Plastics;
- Fossil resources and biodegradable Plastics,

#### 2.1 Non-biodegradable

Most of the current bioplastic market is non-biodegradable which makes problem for waste management (Algieri et al. 2012, 2017). Bio-based /partially bio-based plastics include bio-based drop-in PE and PP, polyethylene terephthalate (PET), and technical performance bio-based polymers, such as polytrimethylene terephthalate (PTT) or Thermoplastic polyester elastomers (TPC-ET), as well as bio-based PAs.

These non-biodegradable bioplastics are from renewable natural resources, that is from biomass without having the bio-degradation characteristics (Rahman and Bhoi 2021). This last is formed in a major part in Brazil, where they produce bioethanol from sugarcane by a fermentation route. The biopolyethylene is also produced from bioethanol, as other common bioplastics: polyethylene terephthalate (bio-PET), bio-PP or polypropylene (bio-PVC, polyvinyl chloride (bio-PVC),bio-PET, (Rujnić-Sokele and Pilipović 2017).

Bio-PE, bio-PET, and bio-PAs currently represent 40% around 0.8 million tonnes of global bioplastic production capacities (The bioplastics global market to grow by 36% within the next five years 2021). In these last years, the focus has shifted on polyethylene furoate (PEF), a novel polymer that is anticipated to enter the commercial market by 2023. This new polymer is comparable to PET, but it is completely bio-based and has superior barrier properties, which makes it an optimal material for beverage bottles.

Renewable r	aw materials	
Biopolymers e.g. Bio-PE (PP/PVS), biobased PET, PTT	Biopolymers e.g. PLA, PHA, Starch blends	
Not biodegradable		Biodegradable
Conventional polymers	Biopolymers	
Nearly all conventional plastics e.g. PE, PP, PET	e.g. PBAT, PBS, PCL	
Petrochemica	l raw materials	

Fig. 2 Types of bioplastics (Philp et al. 2013)

#### 2.2 Biodegradable

Plastics that are both biodegradable and bio-based, come from renewable natural resources, show the biodegradation property at some stage. This group includes the thermoplastically modified starch as well as other bio-degradable polymers like polyhydroxyalkanoates (PHA), polylactide (PLA), and polybutylene succinate (PBS).

Besides petrochemicals, PLA can be found from planned Escherichia coli (Jung and Lee 2011) or with woven bamboo fabric (Porras and Maranon2012).

Instead, PHAs in Fig. 3 shown a general structure are a varied cluster of biopolymers, but typically denote to poly(3hydroxybutyrate) and poly(3-hydroxybutyrate-co-3-hydroxy valerate) (PHBV). They are mostly produced from sugar or lipids by bacteria because PHAs represent an intracellular product of bacteria. Around 250 types of bacteria help to yield PHA. So, these bioplastics are collected with the demolition of bacteria and then disconnected from the microbial cell matter. Moreover, PHAs have good barrier characteristic and attractive in different biomedical applications. They also have the standard specification from marine degradability, which is ASTM D7081.

PHAs have different attributes: fully bio-degradable either in water or even in soil (Meereboer et al. 2020); good resistance as well as printability to oil and grease; until a temperature of 120 °C (Philp et al. 2013).

Moreover, PHAs came from agro- and food wastes, such as wheat bran, rice husk, potato peel, mango peel, straw and bagasse and (Gowda and Shivakumar 2014). They degrade in different rate in different media. Thus, as seen as in the case of PHAs, in general, the property of biodegradability can be directly related to the structure of the polymer and can thus be benefited with specific applications, particularly in case of packaging. PCL is a bio-degradable polyester which has very low melting point (~60 °C). It has general application in biomedical, which includes the surgical structure.

To state that a biodegradable material is necessary to have a standard specification and some material about the timeframe, the amount of biodegradation, as well as environmental conditions. Thus, EUBP focuses on more explicit claim of composability and the corresponding standard references as shown in Fig. 4.

If a product is classified as compostable, it has another advantage besides biodegradability, it differs from the oxo-biodegradable products. These lasts do not fulfill the standard EN 13,432 about compostability, because the oxofragmentation is not biodegradation. "Oxo-degradable" or "oxo-biodegradable" is made with conventional plastics including some additives to replicate biodegradation, with a small fragmentation remain in the environment.

## 2.3 Bio-Based Certification Standards

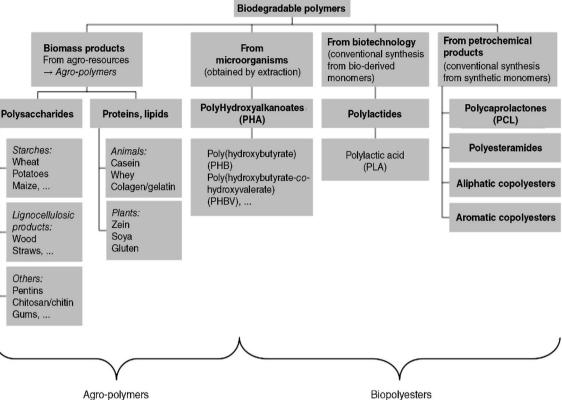
The term "bio-based" refers to material derived from biomass. The most common biomass for bioplastic uses is, for example, corn, sugarcane, and cellulose.

Bio-based plastics have the exceptional advantage over general plastics materials which can reduce the dependency on fossil resources, resulting lesser amount of emission of greenhouse gas. Consequently help the EU achieving the goals of CO<sub>2</sub> emission in 2020 (Bioplastics-Facts and Figures 2021).

Usually, companies indicate their bio-based products with the wording "bio-based carbon content" or with "biobased mass content", but some other standard certifications exist to individuate them. A methodology to measure the bio-based carbon content in materials exists which is called the 14C-method. Thanks to this method, the European standard, and the corresponding USA standards exist. They are CEN7TS 16,137 and ASTM 6866, respectively, for EU standard and US standard. Moreover, a method to individuate a bio-based mass content was introduced by the French Association Chimie du Vegetal (ACDV) with

Fig. 3 The general structure of polyhydroxyalkanoates (Ojumu et al. 2004)

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			/100-30000
<i>n</i> =1	R =	hydrogen	poly(3-hydroxypropionate)
		methyl	poly(3-hydroxybutyrate)
		ethyl	poly(3-horoxyvalerate)
		propyl	poly(3-hydroxyhexanoate)
		pentyl	poly(3-hydroxyoctanoate)
		nonyl	poly(3-hydroxydodecanoate)
<i>n</i> = 2	R =	hydrogen	poly(4-hydroxybutyrate)
<i>n</i> = 3	R =	hydrogen	poly(5-hydroxyvalerate)



Agro-polymers

Fig. 4 Different biodegradable polymers and corresponding their raw materials used (Vilpoux and Averous 2014)

a corresponding certification scheme. It consists to take chemical elements-such as oxygen, nitrogen, and hydrogen-into account, besides the bio-based carbon.

## **3** Bioplastics Applications

### 3.1 Food Packaging

One of the main recent focuses of the food industry concerns packaging-related issues, which defines a whole industry by itself. This kind of industry is constantly following the needs and criteria of the food production world, and its focus on the development of new biopolymer-based packaging is crucial for the whole food industry sustainability as well as its quality standards, leading to more clean and sustainable delivery chains from the production facilities and their internal storage systems, to transport facilities, to market places to consumer houses.

The need for high-standard storage features and the urge for packaging with high economic, low ecological impact, ease of customization, and low encumbrance can be answered by compostable or degradable bioplastics (Jabeen et al. 2015).

Still, the effective applications of packaging in the food industry are few in respect to other fields and need to be enlarged; but nowadays, the biggest food distribution organizations are sensitive to the problem and seem willing to convert to bioplastics as much as possible.

One important aspect to consider when developing this kind of material is that diverse food products need different packaging features, resulting in the need for the development of many technologies, such as multi-layer films, modified atmosphere packaging, and smart and active packaging. One of the main requested features for food packaging is the shielding from water and oxygen. While it is not difficult to develop bio-based multicomponent synthetic coatings to act as a barrier, this arises as a downside, the difficulty for a recycling option, as long as the recycling itself is practicable for single-component materials.

To have a quick view as shown in the Table 1 below (Pilla 2011), the main features required in food packaging are moisture and oxygen permeability and mechanical properties. The Table 1 below compares the main materials, both bio-based and synthetic, used in the field (see Table 2).

The main issues of bio-based polymers in the food industry field are their relative high price than conventional plastic and the less than ideal water barrier features, but to mention the most widely applied materials in this field, starch-based

Table 1	Comparison bet	ween main polymers	used in the food industry
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Polymers	Moisture permeability	Oxygen permeability	Mechanical propertie
Bio-based			
Cellulose (CA) acetate	Moderate	High	Moderate
Starch/polyvinyl alcohol	High	Low	Satisfactory
Proteins	High-medium	Low	Satisfactory
Cellulose/cellophane	High-medium	Very High	Satisfactory
Polyhydroxyalkanoates (PHA) Polyhydroxybutyrate /valerate (PHBA)	Low	Low	Satisfactory
Polylactate	Moderate	High-moderate	Satisfactory
Synthetic			
Low density polyethylene	Low	Very High	Moderate-good
Polystyrene	High	Very High	Poor-moderate

 Table 2
 Main bioplastics applications in the food industry

Application	Biopolymer	Company or users	References
PLA			
Coffe and other bevarages	Cardboard and cups with PLA coating	KLM	Jager (2010)
Beverages	Cups made with PLA	Mosburger (JP)	Sudesh and Iwata (2008)
Fresh salads	bowls made with PLA	MCDonald's	Haugaard et al. (2001)
Carbonted water, juices and dairy drinks	bottles Cups made with PLA	Biota, noble	Auras et al. (2004)
Fresh cut fruits, vegetables, bakery goods	trays and packs made with PLA	Asda (retailer)	Jager (2010), Koide and Shi (2007)
Organic pretzels, potato chips	bags made with PLA	Snyder's of Hanover, PepsiCo's Frito-lay	Weston (2012)
Bread	Paper bags with PLA window	Delhaize (retailer)	Delhaize (2007)
Organic poultry	bowls made with PLA, absorb pads	Delhaize (retailer)	
Starch based			
Milk chocolate	Corn starch trays	Cadbury food group, Marks and Spencer	Highlights in Bioplastics, Website European bioplastics (2021)
Organic tomatoes	Packaging based on Corn	Iper supermarkets (Italy), Coop in Italy	
Cellulose-based			
Kiwi	Bio-based trays wrapped whit cellulose film	Wal-Mart	Blakistone and Sand 2008)
Potato chips	Metalized cellulose film	Boulder Canyon	Highlights in Bioplastics, Website
Organic pasta	Cellulose based packaging	Birkel	European bioplastics (2021)
Sweets	Metalized cellulose film	Quality street, Thorn ton	

films are mostly used for fruit and vegetable packaging and transportation. Here, this materials' main positive feature is the high breathability, a key element for preserving the shelf life of the fresh products (Bastioli 2001).

Wolf et al. (2005), in 2005, mentioned a price range for modified starch polymers from  $\notin 1.50$  to  $\notin 4.50$  per kg, the cheaper mostly being injection molding foams, so that an average price would sit around  $\notin 2.50-3.00$  per kg.

As different types of food require diverse features, a distinction by food typology is hereby adopted to give a comprehensive view.

Fruits and vegetables have a high respiration rate, which can lead to a fast decaying of optimal conditions, besides, they are highly susceptible to water, carbon dioxide, and ethylene concentration. So as the main features, a package should provide a good carbon oxide/oxygen ratio in the atmosphere around the product, a good barrier against light, good mechanical properties, and a barrier to odors.

Raw meat is highly susceptible to spoilage bacteria and pathogens growth. High oxygen concentration in the packaging is requested to preserve the fresh meat's color, so high oxygen permeability is required. So vacuum packaging is often considered a good choice, while adding oxygenadsorbing layers, resulting in active packaging, can better preserve cured meat (Andersen and Rasmussen 1992).

Dairy products need low oxygen permeability materials to avoid oxidation and microbial growth. In addition to that, a good barrier to light can preserve fats' oxidation. Other main features are the water evaporation factor and the avoiding of odor absorption from the exteriors. These features can reside in some forms of polysaccharides as pectins, which are mainly produced by extraction from fruit and vegetable sources and could act as a safety barrier for food products (Baldino et al. 2018). For example, the study of Cerqueira et al. (The bioplastics global market to grow by 36% within the next five years 2021) on polysaccharide edible coatings to preserve cheese showed good results in terms of the lower ratio of superficial mold growth compared to uncoated cheese.

The following Table 2 (Kumar and Thakur 2017) is a collection of the main current applications of bioplastics in the food industry.

## 3.2 Agricultural Applications

Agricultural applications of PHAs-based bioplastics are limited to nets, grow bags, and mulch films. Bioplastics-based nets are alternatives to high-density polyethylene, traditionally used to increase the crop's quality and yield and protect it from birds, insects, and winds. Grow bags, known also as planter bags or seedling bags, are commonly made of low-density polyethylene. Instead, PHAs-based grow bags would be biodegradable, root-friendly, and non-toxic to the surrounding water bodies. Finally, bioplastics in mulch films are essential to uphold exceptional soil structure, moisture retention, control weeds, and prevent contamination, in substitution of fossil-based plastics (El-malek et al. 2020).

## 3.3 Medical Applications

Advancements in biomedical applications of biodegradable plastics lead to the development of drug delivery systems and therapeutic devices for tissue engineering, such as implants and scaffolds (Narancic et al. 2020).

Polymers play a crucial role in many medical and biomedical application (Parisi 2015, 2018). These fields can take advantage of cellulose as main green bioplastic. Thanks to its nontoxicity, non-mutagenicity, and biocompatibility, cellulose has been deeply studied for implants, tissue, and neural engineering, and pharmaceutical fields, as shown in Fig. 5 (Picheth 2017).

Cellulose is organized in a fibrillar structure, with fibrils being the elementary structural unit with a cell diameter of 10 nm organized to macroscopically form fibers.

Bacterial cellulose is used in the development of cellulosic membranes to be applied for tissue repair scopes. These membranes exhibit pores in a range of  $60-300 \mu m$ . Also, modified cellulose matrix and bacterial nano-networks have been studied (Verma et al. 2008; Li et al. 2009; Liu et al. 2013).

Nanocelluloses and their composites are the main sources for any green plastic studies about the fabrication of medical implants, either in dental, orthopedic, and biomedical fields. More recent studies are developing 3D printing and magnetically responsive nanocellulose-based materials (Gumrah Dumanli 2016).

Another application worth mentioning is wound dressing nano-cellulosic membranes, with features as wound pain reduction, extruding retention reepithelialization acceleration and of infection reduction. Patented products of this kind are already available on the market, such as Bioprocess®, XCell®, and Biofill® (Magnocavallo et al. 1993; Fontana 1990).

Also, the biocompatibility of PHAs makes them ideal for medical applications, such as cancer detection, wound healing dressings, post-surgical ulcer treatment, bone tissue engineering, heart valves, artificial blood vessels, artificial nerve conduits and drug delivery matrices (El-malek et al. 2020).

## 3.4 Novel Industrial Applications

PHAs-based wood-plastic composites are novel industrial applications of bioplastics. They are very interesting for

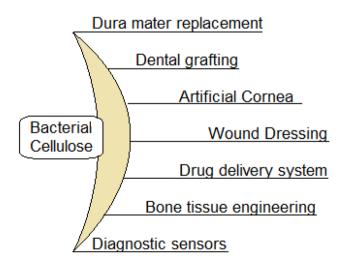


Fig. 5 Biomedical applications of bacterial cellulose (Picheth 2017)

their low cost, biodegradability, mechanical and physical properties that can be enhanced by suitable pre-treatments. PHAs-based lignin composites are recently applied as films in 3D printing, thank their shear-thinning profile that helped in the layer adhesion and reduced the warpage (El-malek et al. 2020).

## 3.5 Other Applications

Bioplastics applications are constantly researched in many other fields, such as structural and electrical engineering. Although relying on biopolymers can result in less than ideal features, in respect to conventional plastics, bio-composite materials are crucial for research developments and for widening the application fields (Luca et al. 2017). Polymer composites are produced combining natural textile (basalt, carbon), natural fibers (jute, kenaf, hemp and sisal),-fillers (clays, zeolite, graphene) commonly used in many traditional application (Candamano et al. 2021, 2020), with polymers (Mohammed et al. 2015; Candamano et al. 2017), which can be chosen to be biodegradable (Rouf and Kokini 2016; Díez-Pascual 2019). Re-inforced biocomposites include recycled wood fibers or by-products from food crops harvesting. Even regenerated cellulose fibers from renewable sources like vegetal by-products or bacterial (Reddy et al. 2015) are included in this field, as sourcing *nanofibrils* of cellulose and chetin (Roy et al. 2014).

As an example, starches, which are considered one of the main resources in this field, can be used in a multitude of applications, which are collected in the Fig. 6 below.

Civil engineering applications include the utilization of foam composite made from vegetable oil sources. Their main features are generally low weight, acceptable physical properties, and good thermal insulation features. They are mostly used in composite-layers panels, in addition to metal or polymeric panels for construction. Some developments were brought to re-inforce rigid foam composites using fillers, short fibers, and long fibers. Bio foams obtained from vegetable oils are mainly produced from soybean, palm, and rapeseed oils (Lu and Larock 2009), and they derive from a chemical modification of the oils: -OH groups are added to an unsaturated triglyceride through hydroxylation of doublebonded carbons or triglyceride alcoholysis or by the esterification of the fatty acids and glycerol molecules contained in the oils, thus producing a monoglyceride utilizing a catalytic reaction (Pilla 2011). The mechanical and thermo-acoustical properties of bio foams are dependent on the cell structure and size. As an example, closed-cell foams are best suited for high compressive strength and impact robustness, while open-cell structures are a good choice for acoustic insulation means.

Rigid foam composites can be re-inforced with a wide range of fillers and fibers. Inorganic fillers, such as layered silicates, have considered the realization of synthetic polymer structures, while lignocellulosic fillers and fibers of vegetal sources, like soy or wood flours, fillers from paper and hemp fibers. Those kinds of re-inforcing materials can help the sustainability of the vegetable oil-derived rigid foams production and utilization.

## 4 Environmental Aspects of Bioplastics

### 4.1 Sustainability and Environmental Footprint

The sustainability of the whole family of bioplastics can be properly seen if all the stages of the materials, like sourcing, production, utilization, and disposal, are considered. In a more precise manner, the economic and environmental features of each of these stages are weighted. For example, the manufacture of biocomposites for construction applications gives direct benefits to the whole construction engineering industry's ecological impact.

Bio-based sustainable packaging aims to use renewable material sources and food and agricultural processing byproducts, which are sources that are not in competition with the food production chains (Reichert 2020). To classify the sources of materials used, we can utilize a biofuel classification, segregating first-, second-, and third-generation feedstocks. First-generation feedstock involves edible biomass like sugarcane, whey, and maize. The second generation comprises non-edible biomasses from lignocellulosic sources, ranging from agriculture, forest, and animal processing by-products, to municipal wastes. The most unconventional sources, listed as third-generation feedstock comprise biomass from algae (Naik et al. 2010).

The main biopolymer that seems to have good features and high versatility to compete with conventional plastics is polylactic acid (PLA) (Andreas Detzel 2006), made entirely from renewable sources. It exhibits mechanical properties similar to PET and PP. As a drawback, Andreas Detzel and Kauertz (2015) state how bioplastic bags are usually made with thicker films than conventional plastic bags, resulting in higher mass utilization. In addition to that, considering an average range, bioplastic films are made by 40% to 70%of fossil source components. The two features can lead to the conclusion that bioplastic bags can easily be the cause of a consistent environmental load in respect to conventional plastic bags. To have a better idea on how much the weight difference can be a problem for sustainability, we can consider that the weight per unit area of bioplastic-based bags exceeds by 30% circa the weight of PE films, this due to a higher density of the source materials (Andreas Detzel and Kauertz 2015).

Biodegradable plastics sources need high areas of farmland and vast volumes of water for their production, with

Adhesives		Constru	ction Industry
<ul> <li>Hot-melt glues</li> </ul>		<ul> <li>Concre</li> </ul>	te block binder
Stamps, bookbinding, envelopes		<ul> <li>Asbest</li> </ul>	tos, clay/limestone binder
<ul> <li>Labels (regular and waterproof)</li> </ul>		<ul> <li>Fire-res</li> </ul>	sistant wallboard
<ul> <li>Wood adhesives, laminations</li> </ul>		<ul> <li>Plywood</li> </ul>	d/chipboard adhesive
<ul> <li>Automotive, engineering</li> </ul>		<ul> <li>Gypsu</li> </ul>	n board binder
<ul> <li>Pressure sensitive adhesives corru</li> </ul>	igation paper	Paint fi	ller
Paper Industry	Cosmetic	and Pharm	aceutical Industry
<ul> <li>Internal sizing</li> </ul>	Dusting	powder	
Filler retention	• Make-u	-	
<ul> <li>Surface sizing</li> </ul>		er/extender	
• Paper coating (regular and color)	Face cre		
<ul> <li>Carbonless paper stilt material</li> </ul>	Pill coat	ing, dusting	g agent tablet binder/dispersing agent
<ul> <li>Disposable diapers</li> </ul>		<u> </u>	
<ul> <li>Feminine products sacks</li> </ul>			
Explosives Industry	Mining Indust	ry	Miscellaneous
<ul> <li>Wide range binding agent</li> </ul>	Ore flotation	•	<ul> <li>Biodegradable plastic film</li> </ul>
<ul> <li>Match-head binder</li> </ul>	• Ore sediment	ation	Dry cell batteries
	Oil well drillin	ıg mud	Printed circuit boards

Adhesive - Hot-melt glues - Stamps, bookbinding, envelopes - Labels (regular and waterproof) - Wood adhesive, laminations - Automotive, engineering - Pressure sensitive adhesives corrugation paper		Construction Industry - Concrete block binder - Asbestos, clay/limestone binder - Fire-resistant wallboard - Plywood/chipboard adhesive -Gypsum board binder -Paint filler
Paper Industry - Internal sizing - Filler retention -Surface sizing -Paper coating (regular and color) - Carbonless paper stilt material - Disposable diaspers - Feminine products sacks		Cosmetic and Pharmaceutical Industry - Dusting powder - Make-up - Soap filler/extender - Face creams - Pill coating, dusting agent tablet binder/dispersing agent
Explosives Industry - Wide range binding agent - Match-head binder	Mining Industry - Ore flotation - Ore sedimentation - Oil well drilling mud	Miscellaneous - Biodegradable plastic film - Dry cell batteries - Printed circuit boards - Leather finishing

#### Fig. 6 Non-food uses of starch

the consistent downside of using these resources otherwise allocated to food production. In addition to that, bioplastic production contributes to pollution because of the pesticides used for the crops and the chemicals used in the transformation processes, but here, the use of eco-friendly alternative methods can overcome the issue (Colwill et al. 2012).

As the last main drawback, bioplastic not composted after use may be trashed in landfills and consequently produce methane because of oxygen deprivation, resulting in a cause for greenhouse production. Even recycling brings up some issues: the recycling process. of these materials cannot be processed with conventional plastics and therefore need separate process streams.

## 4.2 Disposal Processes and Environmental Impact of Bioplastic Packaging

When considering packaging applications, market prices of bioplastics still result higher than the conventional plastic ones, so they access the market mainly for private consumption. This consideration leads us to the fact that bioplastic disposal routes mainly involve household consumption.

Figure 7 below reports the actual discarding processes followed for some bioplastic packaging types (Andreas Detzel and Kauertz 2015). As a result, composting is the main route end for disposal, but still a consistent fraction of the total mass reaches the residual waste and eventually be sent to incinerators, this because of mistakes in the disposal process or even separation by screening in the disposal plants (Ahamed et al. 2021).

Grundmann and Wonschik published a study on how bioplastic bags could interact in some fermentation disposal plants in Germany (Grundmann and Wonschik 2011). Anaerobic fermentation, as well as hydrolysis analysis, has been done to test this behaviour. Results show how thermophilic features are needed actually to act fermentation processes, while the higher degradation degrees fall around 20% values.

An extended life cycle assessment analyses have been addressed in the study of bio-PE systems by considering the steps below (Andreas Detzel and Kauertz 2015):

- Manufacture of the primary materials (bio-PE and PE-LD)
- Transport of the new product to processing
- Manufacture of the film products
- Transport of the film products
- Disposal of the films (WIP)
- Utilization of the films (recycling)
- Allocation of the use of secondary materials and secondary energy from recycling and disposal processes in the form of credits
- Accounting (credit) for the CO<sub>2</sub> bound in the bio-PE.

The following graphs present some of the results of the above-mentioned LCA analysis (see Table 3).

As a conclusion, it emerges that, compared to fossilbased plastics, bio-PE has better responses in Climate

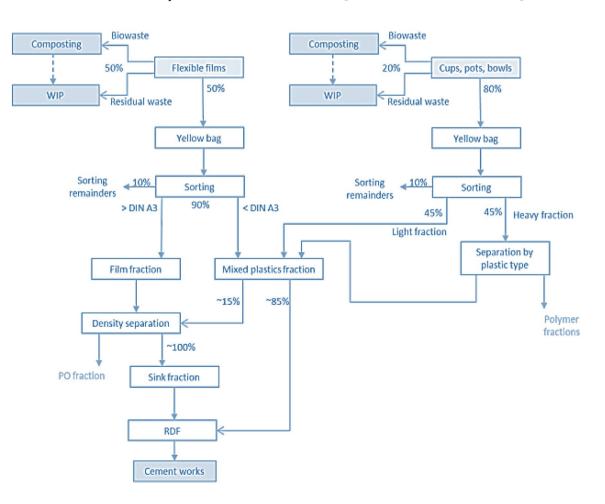


Fig. 7 Disposal flowchart of bioplastic packaging (Andreas Detzel and Kauertz 2015)

	Climate [PE_film m <sup>2</sup> ]	U	Fossil re [PE_film	esources n_30g/m <sup>2</sup> ]
	kg CO <sub>2</sub> lents pe film		kg crude equivale of film	e oil nts per m <sup>2</sup>
	bio-PE	fossil PE	bio-PE	fossil PE
Disposal in the 2nd LC	0.02	0.02	_	_
Recycling	0.005	0.005	0.001	_
Disposal in the 1st LC	0.05	0.05	-	-
Tansport of finished product	0.02	0.02	-	-
Processing	0.01	0.01	0.001	0.001
Transport of new goods	0.005	0.005	0.001	
Manufacture of primary materials	0.04	0.06	0.01	0.039
CO <sub>2</sub> uptake	-0.07	_	-	_
Secondary energy allocation LC1	- 0.02	- 0.02	- 0.005	- 0.005
Secondary energy allocation LC2	- 0.01	- 0.01	- 0.001	- 0.001
Secondary material alloca- tion	- 0.01	- 0.01	- 0.005	- 0.005

**Table 3** Climate Change and Consumption of Fossil Resources indi-<br/>cators, comparative LCA of film packaging made of fossil PE and<br/>bio-PE (Algieri et al. 2013)

Change and Consumption of Fossil Resources impact, but lacks in other features like Acidification, Eutrophication, and Human Toxicity impact factors.

## **5** Bioplastic Sources

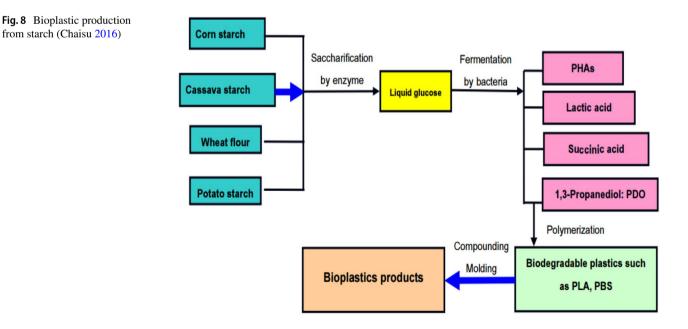
## 5.1 Agricultural Crops

Bioplastics can be produced from polysaccharides (e.g., starch, cellulose, chitosan/chitin), proteins (e.g. casein, gluten), and other carbon sources (Nachwachsende and Agency 2020).

Currently, the most used bioplastic is thermoplastic starch, obtained by enzymatic saccharification and microbial fermentation (Fig. 8) or by modifying starch with plasticizers with hydrophilic properties (Mojibayo et al. 2020).

Nevertheless, starch-based bioplastics treated with plasticizers and stored for long time face recrystallization and consequent deterioration of mechanical properties. To overcome this problem, starch-based bioplastics' performance may be improved by the addition of nanoparticles to obtain nanocomposite bioplastics used in automotive components, packaging materials, and drug delivery (Mose and Maranga 2011).

Starch is usually obtained from different terrestrial crops. Distilled water, glycerol, and vinegar were used to modify cassava starch for the production of bioplastic sheets (Mojibayo et al. 2020). Bioplastics from cassava starch were re-inforced also by coconut husk fibers (Babalola and Olorunnisola 2019). Condensation polymerization was performed to produce bioplastic from corn starch and glycerin to obtain nanocomposites for packaging applications (Ateş and Kuz 2020). Other starch sources are potatoes, wheat, and tapioca. The finest, smoothest, flexible and strong bioplastic was produced from tapioca starch (Gökçe 2018), but the potato-derived starch showed the best properties in terms



of extraction, ease of working, texture, and potential drying (Hamidon 2018). Composite bioplastics from tapioca starch and sugarcane bagasse fiber were recently investigated and ultrasounds treatment improved properties by enhancing the tensile strength and decreasing the moisture absorption rate (Asrofi et al. 2020).

Among proteins, wheat gluten can be processed to produce bioplastics (Rasheed 2011; Jiménez-Rosado et al. 2019).

Sugarcane is exploitable for bioplastic production by bacterial sugar assimilation (Pohare et al. 2017).

Finally, oil is a good carbon source for the production of bioplastic. Cottonseed oil (Magar et al. 2015), soybean oil (Park and Kim 2011), crude palm kernel oil, jatropha oil, crude palm oil, palm olein, corn oil, and coconut oil were typically investigated (Wong et al. 2012).

Lignocellulosic biomass is another promising resource for bioplastic production avoiding the consumption of food crops. Nevertheless, it requires suitable cost-effective pretreatments for decomposition into sugar monomers (Brodin et al. 2017; Govil 2020).

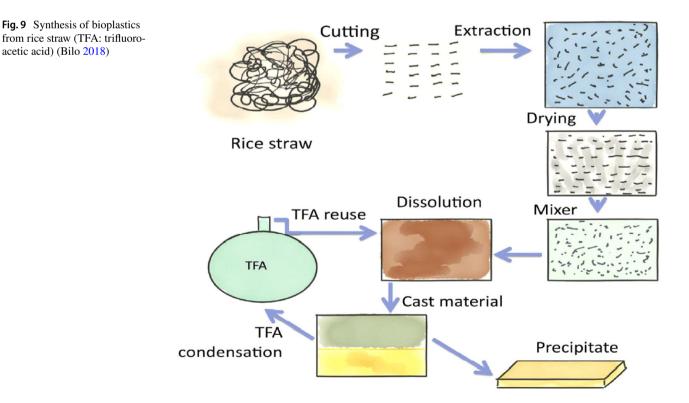
#### 5.2 Organic Waste Sources

Cassava and other crops require large land areas, water, and nutrients. Moreover, they compete with the food supply, and their use to produce bioplastics is not sustainable. Instead, it is interesting to consider the organic waste source to valorize a residue and turn a problem into an opportunity in a circular economy approach (Yadav et al. 2019).

Wastes from the food-processing industry are an important potential source of bioplastics (Tsang 2019; Jõgi and Bhat 2020). Vegetable wastes used to produce novel bioplastic films were carrots, radicchio, parsley, and cauliflowers (Perotto 2018). Novel starch- and/or cellulose-based bioplastics were produced from rice straw (Fig. 9), an agricultural waste usually used for bioethanol production (Agustin et al. 2014; Bilo 2018), and other agricultural wastes (Chaisu 2016).

Extrusion of rice bran and kraft lignin—that are industrial by-products of brown rice production and wood pulping process, respectively—produced a bioplastic with good extrudability and mechanical properties (Klanwan et al. 2016).

A residual product of crude oil palm production is an empty fruit bunch, composed of cellulose, hemicellulose, and lignin. Having high cellulose content (36.67%), this abundant waste could be used to produce bioplastics (Isroi and Panji 2016; Isroi et al. 2017). Microcrystalline cellulose and glycerol were added to keratin from waste chicken feathers to produce biopolymeric films (Ramakrishnan et al. 2018; Sharma et al. 2018). Microcrystalline cellulose was a re-inforcing additive in bioplastic production also from avocado seeds (Sartika et al. 2018), jackfruit seeds (Lubis et al. 2018), and cassava peels (Maulida and Tarigan 2016). Waste cassava peels were investigated in combination with kaffir lime essential oil for future applications in industry and medicine (Masruri et al. 2019). Cocoa pod husk and sugarcane



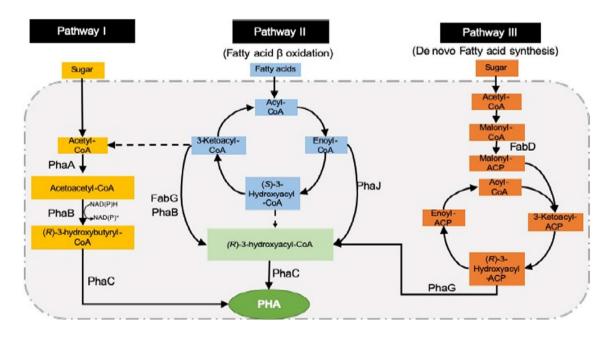
bagasse, which are wastes from the chocolate industry and the sugar industry, respectively, are promising for the production of biodegradable plastic films (Azmin et al. 2020). Bioplastics could be produced by injection molding from rapeseed oil production by-products, such as press cake or meal (Delgado et al. 2018). New bioplastics were prepared from potato peels and waste potato starch with eggshells and/or chitosan (from exoskeleton seafood wastes) as additives (Kasmuri and Zait 2018; Bezirhan Arikan and Bilgen 2019). Also, banana peels were used to produce a bioplastic with the addition of corn starch, potato starch, sage, and glycerol (Sultan and Johari 2017; Azieyanti et al. 2020). Bloodmeal is a low-value protein-rich by-product from meat processing, that is convertible into a bioplastic material (Low et al. 2014). Bioplastic fibers were fabricated also from gum arabic by electrospinning method (Padil et al. 2019).

Polyhydroxyalkanoates (PHA) is a group of biodegradable plastics produced by microorganisms from renewable sources (Shraddha et al. 2011) by the three pathways in Fig. 10.

Among PHAs sources, researchers investigated chicken feather hydrolysate (Benesova et al. 2017), animal fat waste (Riedel 2015), lignocellulosic biomass hydrolysate (Bhatia 2019), grass biomass (Davis 2013), fruit pomace, waste frying oils (Follonier 2014), olive oil mill pomace (Waller et al. 2012), saponified waste palm oil (Mozejko and Ciesielski 2013), low-quality sludge palm oil (Kang 2017), waste oil palm biomass (Hassan 2013), spent coffee grounds (Nielsen et al. 2017) and other carbon sources (rice straw, maltose, glucose, sugarcane liquor, corn steep liquor, corn stover liquor, cheese whey, waste potato starch, sugar beet molasses, etc.) (Khatami et al. 2021; Marjadi and Dharaiya 2010; Tripathi et al. 2012). Another interesting resource is the organic fraction of municipal solid wastes convertible into PHAs by acidogenic fermentation of pre-treated and hydrolyzed biomass (Ivanov et al. 2015; Ebrahimian et al. 2020).

Recent works investigated PHA production from volatile fatty acids, obtained by the anaerobic digestion of waste paper (Al-Battashi 2019; Al Battashi et al. 2020).

The most common PHA is polyhydroxybutyrate (PHB), produced from low-cost sugarcane molasses by *Bacillus cereus* (Suryawanshi et al. 2020) or *Staphylococcus epidermidis* (Sarkar et al. 2014), cheap agro-residues by *Bacillus* sp. (Getachew and Woldesenbet 2016), date syrup by *Pseudodonghicola xiamenensis* (Mostafa et al. 2020), non-food sugars from oil palm frond (Zahari et al. 2015) or biodiesel industry by-products (García 2013) or used cooking oil (Martino 2014) by *Cupriavidus necator*, wheat straw lignocellulosic hydrolysates by *Burkholderia sacchari* (Cesário et al. 2014), wheat bran hydrolysate by *Ralstonia eutropha* (Annamalai and Sivakumar 2016), bakery waste hydrolysate by *Halomonas boliviensis* (Pleissner 2014). An innovative approach consists of PHB production from landfill methane by methanotrophs (Chidambarampadmavathy et al. 2017).



**Fig. 10** The three metabolic pathways for PHA production (PhaA: b-ketothiolase; PhaB: acetoacetyl coenzyme A(CoA) reductase; PhaC: PHA synthase; FabG: 3- ketoacyl acyl carrier protein (ACP)

reductase; PhaG: acyl-ACP-CoA transacylase; PhaJ: enoyl-C ketoacyl acyl carrier protein (ACP) reductase; PhaG: acyl-ACP-CoA transacylase; PhaJ: enoyl-C) (Khatami et al. 2021)

## 5.3 Algae-Based Sources

Microalgae are a promising alternative source for bioplastics production because of their fast growth and no competition with food (Rahman and Miller 2017). Recently, several works investigated the synthesis of bioplastics from microalgae (Beckstrom et al. 2020; Simonic and Zemljic 2020). Microalgae could be used directly as biomass to produce bioplastics or indirectly by the extraction of PHBs and starch within microalgae cells. Other approaches include the production of microalgae-polymer blends through compression/ hot molding, melt mixing, solvent casting, injection molding, or twin-screw extrusion (Cinar et al. 2020).

The most investigated microalgae were Chlorella and Spirulina. Chlorella seems to have better bioplastic behavior, whereas Spirulina showed better blend performance (Zeller et al. 2013). Different species of Chlorella were used in biomass-polymer blends containing polymers and additives (Cinar et al. 2020). Moreover, bioplastic may be produced from Chlorella pyrenoidosa (Das et al. 2018) and Chlorella sorokiniana-derived starch granules (Gifuni et al. 2017). Similar to Chlorella, Spirulina was investigated for bioplastic production (Cinar et al. 2020). For example, a bioplastic-based film was produced from salt-rich Spirulina sp. residues with the addition of polyvinyl alcohol (Zhang et al. 2020). Another bioplastic was prepared from Spirulina platensis, showing good biodegradability (Maheshwari and Ahilandeswari 2011). Other microalgae or cyanobacteria used to produce bioplastics were Chlorogloea fritschii (Monshupanee et al. 2016), Calothrix scytonemicola (Johnsson and Steuer 2018), Neochloris oleoabundans (Johnsson and Steuer 2018), residual Nannochloropsis after oil extraction (Yan 2016), Nannocloropsis gaditana (Torres et al. 2015; Fabra et al. 2017), Phaeodactylum tricornutum (Hempel 2011), and Scenedesmus almeriensis (Johnsson and Steuer 2018). Ten green microalgae were screened for starch production and starch-based bioplastic development. C. reinhardtii 11-32A resulted in the most promising starchproducing strain with interesting plasticization properties with glycerol at 120 °C (Mathiot et al. 2019).

A microalgae consortium cultivated and harvested in a wastewater treatment plant was used as biomass to be mixed



Fig. 11 PHB production from microalgae (Cinar et al. 2020)

with glycerol as a plasticizer to obtain bioplastics (López Rocha et al. 2019).

New composites were formed by combination of microalgal biomass and petroleum. (Cinar et al. 2020; Chia et al. 2020). The PHB production is feasible in microalgae used as bioreactors by the introduction of bacterial pathways into microalgal cells (Hempel 2011) (Fig. 11).

Besides microalgae, macroalgae or seaweeds are aquatic plants rich in polysaccharides and potentially promising sources of bioplastics (Rajendran et al. 2012; Thiruchelvi et al. 2020). The whole red macroalga *Kappaphycus alvarezii* was recently investigated to produce a bioplastic film with the addition of polyethylene glycol as a plasticizer for food packaging applications (Sudhakar et al. 2020).

#### 5.4 Wastewater Sources

Wastewaters are rich in organic matter and salts and are an important resource to be reused for different applications (Hoek et al. 2016, Dasgupta et al. 2016). Casein-rich dairy wastewater is a possible substrate for the manufacturing of bioplastics (Fricke et al. 2019), but the physical properties of obtained brittle films were successfully improved by the addition of polysaccharides with proteins (Ryder et al. 2020). Starch-based bioplastic was developed from potato processing industry wastewater (Arikan and Ozsoy 2011). Activated sludge generated during the wastewater treatment is very abundant and could produce PHBs by thermal cracking (Liu et al. 2019). Mannina et al. (Mannina et al. 2019) recently implemented a new protocol to extract PHAs from mixed microbial cultures in a synthetic effluent simulating a fermented oil mill wastewater. PHAs were produced from municipal wastewater by a two-step process, consisting of anaerobic fermentation producing volatile fatty acids (VFA), and aerobic conversion of VFA to PHA by pure or mixed microorganisms (Pittmann et al. 2013). Moreover, a two-step process was recently suggested to produce PHAs from cheese whey agro-industrial wastewater (Carlozzi et al. 2020). Instead, a three-step process was proposed to accumulate PHAs in paper mill wastewater (Jiang et al. 2012).

Other wastewaters investigated for bioplastic production are wood mill effluents (Ben et al. 2011) and municipal sewage sludge (Bluemink et al. 2016).

The advantage and disadvantages of each source category are summarized in the following Table 4.

## 6 Conclusion

The research, application opportunities, sourcing and sustainability of bioplastics production have been discussed to clarify the field.

Bioplastic source category	Advantages		Disadvantages	
Agricultural crops	Renewability	Abundance Closed carbon cycle Mature processing technology in large scale	Threat to food security and eco-systems Use of large fertile land, water, and nutrients Negative contribute to the eco-balance Conflict food vs bioenergy/ biofuels/ biomateri- als Long-term unsustainability Necessary pre-treatment of lignocellulosic biomass Long time for production	High processing cost (mostly for PHAs micro- bial production). Required bioaugmentation, metabolic engineering and cost-effective down- stream processing in PHA production
Oganic wastes		Abundant low-cost/free sources Management of environmentally problematic wastes Conversion of wastes to valuable resources No competition with food and feed	Possible localization and/or seasonability of wastes Cost and complexity of logistic operations Necessary pre-treatment of lignocellulosic biomass	
Microalgae		Fast growth rates High productivity. Cultivation on non-arable land Utilization of degraded and saline water sources Integration with waste streams Wastewater remediation Broad environmental tolerance Reduced competition with food	Large volumes of water are required for indus- trial scale Expensive technology of cultivation, harvesting, extraction and fractionation of components in a large scale High energy costs of cultivation and processing	
Macroalgae		High biomass Cost-effective Easily cultivated in natural environment Able to grow in wide range of environments Harvested throughout the year	Biotechnological and genetic engineering tech- niques required Premature large-scale processing technology	
Wastewaters		High availability Abundant and cheap No competition with food and feed A waste becomes a resource	Complex new technologies, generating further wastes Low production yield	

 Table 4
 Advantages and disadvantages of different bioplastic source categories

To further advance the application of bioplastic, it is very necessary to manage carefully the waste disposal. Recycling appears the best solution from that point, for disposal of the bio-based product to maximize the environmental footprint as well as reduce the renewable resources consumption. Recycling of a bioplastic leads to an overall decrease of environmental impact which may associated with the production and disposal of the bioplastic itself. It is worth noting that due to the improper management and applications of bioplastics, the information reported in this paper can be useful for the environmental reliability. PHA materials are the main resource to substitute conventional plastic use in most of the engineering applications fields. Nowadays, the PHA costs of production are too high, but further research on technology and sourcing can reduce manufacturing costs for a versatility and heterogeneity and strengthen the applications of bioplastic.

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