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A review of biodegradable plastics from multidisciplinary perspectives

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1. Introduction

Plastics are lightweight, versatile, affordable and highly durable materials that have become ubiquitous in our everyday lives. Plastics have brought numerous societal benefits and are used for wide-ranging applications across sectors including in transportation, medicine, agriculture and within domestic households (Thompson et al., 2009, Napper and Thompson, 2020). However, many plastic objects have applications as single-use, disposable items such as packaging. Due to the short usage of these items and the increasing global production of plastics, which has seen a 7-fold increase since the mid-1970s (PlasticsEurope, 2019) plastics are a major component of waste which can pose challenges for solid waste management infrastructure (Kaza et al., 2018). A global analysis of the production, use and end-of-life fate of plastics concluded that out of the 8300 million tons (Mt) of plastics produced between 1905 to 2015, 2500 Mt are in use, 4900 Mt have been discarded either in landfills of the environment, 800 Mt have been incinerated and only 600 Mt have been recycled (Geyer et al., 2017). Numerous studies report the presence of plastics in aquatic and terrestrial environments (Courtene-Jones et al., 2017, Napper et al., 2020, Lusher et al., 2015, Hurley et al., 2020) and its accumulation in marine ecosystems over the last 70 years (Ostle et al., 2019, Courtene-Jones et al., 2019, Thompson et al., 2004, Law et al., 2014). Indeed, the accumulation of end-of-life plastics, both in managed waste systems and in the environment, is seen as a global environmental issue (GESAMP, 2016).

Similar to hydrocarbon-based polymers, biopolymers have also been available for over a century (Morris, 1986) but have remained largely undeveloped (Gross and Kalra, 2002). However, over recent decades biodegradable polymers have received increased attention (Babu et al., 2013, Philp et al., 2013). This is due to a myriad of reasons including increased economic opportunities and technological advances, attempts to move away from reliance on fossil-fuels and growing societal concern to the widespread environmental accumulation of conventional plastics and their associated impacts on wildlife, ecosystem services and human wellbeing (Beaumont et al., 2019, Lettner et al., 2017) together with the assumption that bio-based materials may be less harmful.

Biodegradable and bio-based plastics currently only make up a small volume of the total plastic market (see section 2 for definitions). In 2020, the global production capacity for biodegradable polymers was 1.227 million tonnes, with a further 0.884 million tonnes of bio-based polymers. This corresponds to a total of ~0.5% (0.3% biodegradable plastics, 0.2% bio-based polymers) of the 360 million tonnes of plastics produced (PlasticsEurope, 2019, European bioplastics, 2021). Demand for biodegradable and bio-based plastics has increased over the last two decades largely due to the reasons stated above (Philp et al., 2013, Jayanth et al., 2017), and future projections expect production capacity to increase further still to an estimated 2.87 million tonnes by 2025 (European bioplastics, 2021). Bio-based and biodegradable plastics are used for an increasing number of applications, including flexible and rigid packaging materials (wrapping, trash bags, food containers), textiles and hygiene products (fabrics, diapers, sanitary products), consumer goods (e.g. egg cartons, toys, tableware) and agricultural/horticulture items (mulch films, planters, plant ties) (Yin and Yang, 2020).

In 2018, the European Commission established its European Plastic Strategy in a Circular Economy (European Commission, 2018). Based on the principles of resource efficiency and the waste hierarchy, the circular economy is designed to keep materials in the loop as long as possible and prevent the depletion of natural resources (Prieto-Sandoval et al., 2018). A recent report highlighted the role of biodegradable plastics within a circular economy and recognises that in some applications where it is challenging to remove or collect a plastic product or its fragments from the environment after use (e.g. agricultural mulch films); or where it is difficult to separate plastic from organic material that is destined for a composting waste stream (e.g. organic waste bags (European Parliament, 2018)) or wastewater treatment, biodegradable plastics have the potential to bring advantages, compared with conventional plastics (SAPEA, 2020). As such, biodegradable polymers have been proposed as part of the solution to the environmental accumulation of plastics, with several European strategies and directives such as the Waste Framework Directive and the EU Directive on packaging and packaging waste, recognise the role of biodegradable plastics (European Commission, 2018, European Parliament, 1994, European Union, 2019, European Parliament, 2018). In addition, the development of biodegradable plastics is also an attempt to achieve some of the United Nations Sustainable Development Goals (SDGs), laid out in the 2030 Agenda for Sustainable Development, for example SDG 12: responsible consumption and production, SDG13: climate action, SDG 14: life below water and SDG 15: life on land (UNEP, 2016). However, it is important to emphasise that simply replacing conventional plastics with biodegradable alternatives is not a viable solution to address plastic pollution, poor waste management or the inappropriate disposal of waste, i.e. littering (SAPEA, 2020, UNEP, 2015, Viera et al., 2020, Haider et al., 2019).

Most plastics labelled as biodegradable require specific conditions (temperature, oxygen availability, microorganisms, humidity and light (see sections 2-4) in order to degrade, and are usually required to undergo biological decomposition within an industrial facility. The specific conditions required for complete biodegradation may not be present within natural environments and as such there is a potential that this group of plastics may still cause environmental harm in the same way as conventional plastics. Currently, there is limited information regarding the mechanisms and timescales for biodegradation of polymers and products in natural environments, and a lack of assessment regarding the ecological, societal and economic risks associated. Such information is required to advance the development of biodegradable materials and inform holistic impact assessments.

2. Classifications of biodegradable plastics

In an attempt to discuss plastics classified as biodegradable, bio-based or compostable, it is firstly necessary to clarify and specify the correct terminology to address such materials. A term that is often misused and which creates confusion is “bioplastic”. According to European Bioplastics “a plastic material is defined as a bioplastic if it is either bio-based, biodegradable, or features both properties”, so the term is used for general purposes (European Bioplastics, 2021). However, the International Union of Pure and Applied Chemistry (IUPAC) gives a different meaning, defining bioplastic as a “bio-based (composed or derived in whole or in part of biological products issued from the biomass) polymer that can be shaped by flow at some stage in its processing into finished products” (Vert et al., 2012). Since a bio-based polymer is not necessarily biodegradable or “environmentally friendly”, the IUPAC discourages the use of the term “bioplastic” and it will not be used for the purposes of the current chapter, preferring the term “bio-based” (Vert et al., 2012).

The term “biodegradable” has also been debated, depending on the definition of “biodegradation”. Both the IUPAC and the American Society for Testing and Materials (ASTM) provide similar definitions: “degradation

of a polymeric item due to cell-mediated phenomena" – IUPAC (Vert et al., 2012); "degradation resulting from the action of naturally-occurring micro-organisms such as bacteria, fungi, and algae" – ASTM (Lambert and Wagner, 2017). Additional terms were also suggested by the Organisation for Economic Co-operation and Development (OECD) which distinguish between "primary" and "ultimate" biodegradation (OECD, 1992). "Primary" biodegradation is a chemical alteration of a substance due to biological activity and causes the loss of one of the specific properties of the substance. Instead, "ultimate" biodegradation refers to a substance that, under aerobic conditions, is completely utilized by microorganisms, resulting in the production of carbon dioxide (CO_2) water, mineral salts and biomass.

Recently, a working group of the Science Advice for Policy by European Academies (SAPEA) produced a report on "Biodegradability of plastics in the open environment" and proposed a more comprehensive definition of biodegradation. In detail, they introduced the concept of plastic biodegradation not as a material property but as a system one, that includes not only the specific material properties that make it potentially biodegradable, but also the specific conditions of the environment that receives such material (SAPEA, 2020). This is a more complete way to consider and define biodegradation, that takes into account the fact that a specific plastic material cannot biodegrade the same in all the different types of environments (i.e. marine, riverine, terrestrial etc.) and even in the same environment as conditions may be very different (i.e. humidity, pH, microbial community etc.). The SAPEA definition of "plastic biodegradation" is reported in Table 1, along with the main terminology related to the topic.

It is important to highlight that a bio-based polymer can be either biodegradable or not; a biodegradable polymer can be bio-based but also fossil-based; a compostable polymer is also biodegradable but not vice-versa. Figure 1 summarizes the distinctions and interconnections among the different categories. A term that is not discussed within this chapter but it is worth considering briefly, is "oxo-degradable", which has been used for a category of plastics that have been at the centre of different debates. These materials are conventional fossil-based polymers (i.e. LDPE) that contain additives to accelerate their oxidation and fragmentation by the action of ultra-violet (UV) light or heat and oxygen. In 2019 they were banned by the European Union due to concerns about their actual impact on the environment (Manfra et al., 2021).

The lack of consensus on the classification of biodegradable plastics adds more confusion and challenges to a scientific field that still requires more research. Further investigations are needed to fully understand the behaviour of such materials in the environment and if they can effectively be part of the solution for plastic pollution. Therefore, a common agreement and harmonization on the accepted definition of "biodegradation" is of striking importance.

Table 1. The main terminology related to biodegradable polymers and those used within this chapter *.

Term	***Definition***
Bio-based polymer	A polymer composed or derived in whole, or in part of biological products issued from the biomass (including plants, animals and marine or forestry materials).
Biodegradable plastic	Plastic is considered biodegradable when it breaks down to basic elemental components (water, biomass and gas) with the aid of microorganisms.

Biodegradation	Microbial conversion of all the organic constituents of a material to carbon dioxide, new microbial biomass and mineral salts under oxic conditions or to carbon dioxide, methane, new microbial biomass and mineral salts, under anoxic conditions.
Compostable plastic	Plastic capable of undergoing biological decomposition in a compost site as part of an available program, such that the material is not visually distinguishable and breaks down into carbon dioxide, water, inorganic compounds, and biomass, at a rate consistent with known compostable materials.
Degradable polymer	Polymer in which macromolecules are able to undergo chain scissions, resulting in a decrease of molar mass.
Degradation	Degradation that results in desired changes in the values of in-use properties of the material because of macromolecule cleavage and molar mass decrease.
Fossil-based/conventional polymer	A polymer derived from petrochemicals.
Macromolecule	Molecule of high relative molecular mass, the structure of which essentially comprises the multiple repetition units derived, actually or conceptually, from molecules of low relative molecular mass.
Plastic	A material that contains, as an essential ingredient, one or more organic polymeric substances of large molecular weight (i.e. polymers). A plastic is solid in its finished form but can be shaped by flow during manufacturing or finishing into finished articles.
Polymer	Substance composed of macromolecules.

*Based on Kyrikou & Briassoulis 2007; Vert et al. 2012; Lambert et al. 2017; SAPEA 2020.

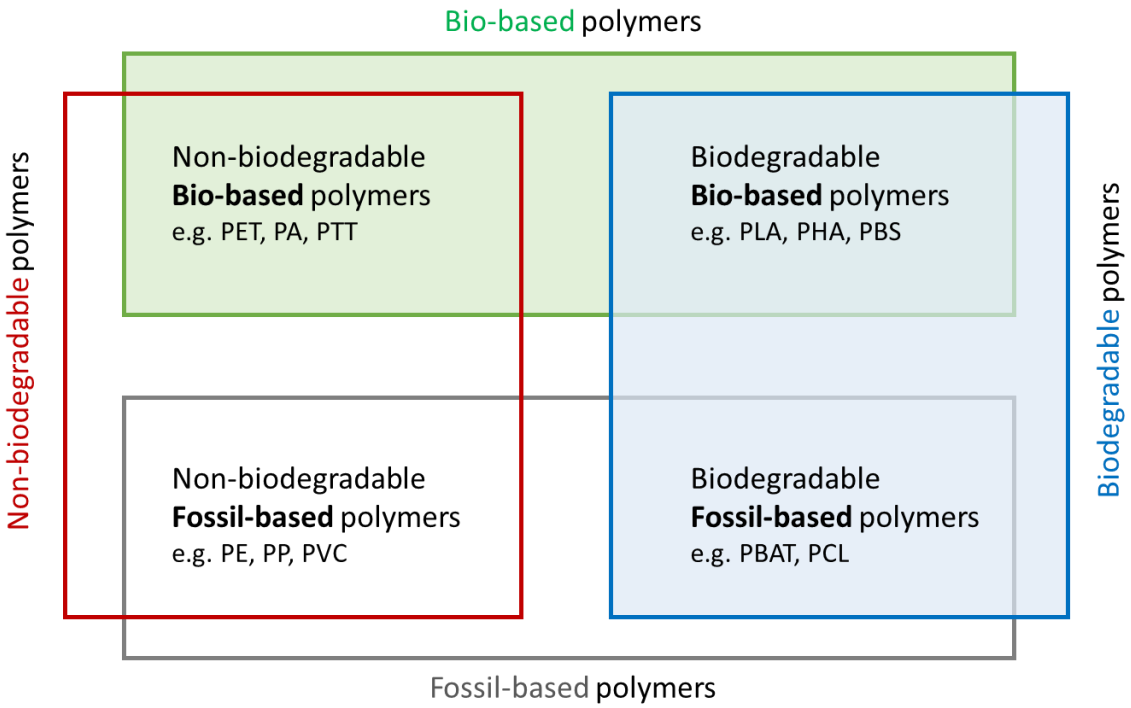

Figure 1. Categories of biodegradable and non-biodegradable polymers. This schematic was adapted from European Bioplastics (2021).

3. Chemical composition of biodegradable polymers

Biodegradable polymers can be found in nature or synthesised from natural or fossil sources. Natural polymers are created during the growth cycles of all organisms and they comprise polysaccharides (i.e. cellulose, starch, chitosan, etc.), polypeptides (i.e. gelatin) and bacterial polyesters (i.e. polyhydroxyalkanoates) (Chandra and Rustgi, 1998). Among natural polysaccharides, one of the most used is starch which is easily biosynthesized by plants (i.e. corn, rice, potato etc.) and composed of linear amylose polymer and highly branched amylopectin polymer (Fink, 2020). Starch can be easily modified to obtain a thermoplastic polymer but its fast degradation via hydrolysis limits its applications. For this reason, it is often blended with other synthetic polymers for longevity, creating materials like Mater-bi® produced by Novamont, which is now widely used in applications such as packaging, agricultural mulch films, disposable cutlery or consumer goods (Moeini et al., 2020).

Polyhydroxyalkanoates (PHAs) are a class of polymers first discovered in 1926, synthesized and intracellularly accumulated in most bacteria under unfavourable growth conditions. In general, PHAs are composed of R(-)-3-hydroxyalkanoic acid monomers (carbon atoms in the range C3-C14), with a variety of saturated/unsaturated and straight/branched chains containing aliphatic or aromatic side groups (Ojumu et al., 2004). The most common PHAs are polyhydroxybutyrate (PHB) that is synthesised by the polymerization of 3-hydroxybutyrate monomer, and the copolymer polyhydroxybutyrate-valerate (PHBV) used in packaging applications (Siracusa et al., 2008).

Beyond natural occurring polymers, biodegradable plastics can be also artificially synthesised using conventional fossil-based resources or renewable biological ones. Among the polymers considered

biodegradable and bio-based, the one that has received great attention and is used in numerous applications is poly(lactic acid) (PLA), an aliphatic polyester considered compostable. The building block of PLA is lactic acid that can be derived from renewable resources like starch and sugar through fermentation. PLA has been mainly used in biomedical applications like tissue scaffolds, implant devices etc. However, recent advancements in its manufacturing have broadened its applications that now range from disposable food service ware, to packaging and textiles (Lim et al., 2008).

Another interesting bio-based biodegradable polyester is represented by poly(butylene succinate) (PBS) and its copolymers, that are used as food packaging, bags, mulch films, flushable hygiene products, etc. PBS is synthesized by polycondensation of succinic acid and 1,4-butanediol, that can be both derived from fossil or renewable resources (Xu and Guo, 2010). One of its most promising copolymers is poly(butylene succinate-co-butylene adipate) (PBSA), characterised by a lower crystallinity than PBS and greater flexibility of the polymer chains, features that make PBSA more susceptible to biodegradation as we further discuss below (Ray et al., 2007).

Finally, biodegradable polymers can also be obtained from petrochemical resources, for instance poly(butylene adipate-co-butylene terephthalate) (PBAT) and poly(ε-caprolactone) (PCL, Fig. 1). The first is a copolymer of terephthalic acid, butanediol, and adipic acid, mainly used in agricultural applications also in blends with PLA and starch (Agarwal, 2020). PCL is an aliphatic polyester synthesized through ring-opening polymerization of the cyclic ester ε-caprolactone, applied in medicine and sustainable packaging (Bartnikowski et al., 2019).

Abbreviations of the polymers referred to within this chapter are summarised in Box 1.

HDPE	High density polyethylene
LDPE	Low density polyethylene
PA	Polyamide
PBAT	Poly(butylene adipate-co-butylene terephthalate)
PBS	Poly(butylene succinate)
PBSA	Poly(butylene succinate-co-butylene adipate)
PBSe	Polybutylene sebacate
PBSeT	Polybutylene sebacate-co-terephtalate
PCL	Poly(ε-caprolactone)
PE	Polyethylene
PET	Polyethylene terephthalate
PHA	Polyhydroxyalkanoates
PHB	Polyhydroxybutyrate
PHBV	Polyhydroxybutyrate-valerate

PLA	Poly (lactide acid)
PLGA	Poly(lactic-co-glycolic acid)
PLLA	Poly(l-lactide acid)
PP	Polypropylene
PTT	Polytrimethylene terephthalate
PVC	Polyvinyl chloride

Box 1. Abbreviations and definitions of polymers referred to in this chapter

4. Biodegradation of biodegradable polymers

In recent years several reviews have summarized the data available on the degradation of some of the most widely used biodegradable polymers (i.e. see Lambert et al. 2017, Haider et al. 2019, Agarwal 2020, Manfra et al. 2021). Different aspects of polymer chemistry affect biodegradation. In general, biodegradation comprises two steps (Agarwal, 2020). The first involves the breakdown of macromolecular chains into molecules of lower molecular weight like oligomers and monomers. This fragmentation can be caused by hydrolysis (in presence or absence of enzymes), oxidation or other mechanisms mainly depending on both the polymer chemical structure and the receiving environment characteristics. The second step consists of the microbial uptake of these low molar mass molecules, resulting in the formation of CO_2, CH_4, biomass and water. The biodegradability of synthetic polymers can depend on the presence in their polymer chains of characteristics similar to those of natural occurring polymers (Chandra and Rustgi, 1998, Kijchavengkul and Auras, 2008). As natural polymers like starch generally biodegrade thorough hydrolysis followed by oxidation (Chandra and Rustgi, 1998), aliphatic polyesters like PLA, PHB and PCL can biodegrade especially thanks to the hydrolysable ester bond in the main chain that is sensitive to microbial attack (Tserki et al., 2006).

The main factors affecting biodegradability can be distinguished in two categories: polymer characteristics and exposure conditions (Kijchavengkul and Auras, 2008). Considering the first one, factors like mechanical properties, glass transition temperature, morphology, hydrophilicity and crystallinity, all have an influence on biodegradation (Eubeler et al., 2010). The nature of the chemical bonds present in the polymer chain is very important to determine biodegradation. For example, conventional polyolefins (i.e. PE, PP) have carbon-carbon single bonds that makes them hydrophobic and resistant to degradation (Kyrikou and Briassoulis, 2007). To be susceptible to mechanisms like hydrolysis, polymer chains need to contain hydrolysable covalent bonds as in ester, anhydride, amide groups; whereas, other characteristics of the polymer structure (i.e. branched chains, unsaturated bond) may influence oxidation (Lucas et al., 2008). The presence of the functional groups previously mentioned have an additional positive effect on biodegradation, since they increase the flexibility of the chain, an aspect discussed in detail below (Kijchavengkul and Auras, 2008). The molecular weight of the polymer also influences the biodegradation rate, since a high molecular weight implies longer chains with more bonds to cleave to breakdown the polymer in oligomers and monomers (Kyrikou and Briassoulis, 2007). Moreover, by increasing the molecular weight, the glass transition temperature (temperature at which the polymer passes from the glassy to the rubbery state) increases and the polymer become less flexible (Kijchavengkul and Auras, 2008). The accessibility of the polymer chains to

degradation agents like microorganisms, water etc., is an important aspect that is linked to characteristics like the polymer flexibility and its morphology. Regarding the flexibility, for instance the presence of bulky side groups can limit the movements of the polymer chains, whereas the presence of carbon double bonds increase flexibility by facilitating the rotation around contiguous single bonds (Kijchavengkul and Auras, 2008). For the morphology, many of the widely used biodegradable polymers are semi-crystalline (i.e. PLA, PBS, etc.); the crystalline regions not only reduce the flexibility of the polymer increasing its stiffness and density, but are also less accessible to water, oxygen, microbes etc, than the amorphous ones (Kijchavengkul and Auras, 2008, Kyrikou and Briassoulis, 2007, Laycock et al., 2017, Lucas et al., 2008). Since they are impermeable to water, the crystalline regions slow hydrolytic degradation and, in addition, reduce transport processes like solvent and gas diffusion due to their well organised cross-linked crystalline domains (Laycock et al., 2017, Lucas et al., 2008). The disorganised amorphous domains are more flexible and accessible to degradation agents and water can penetrate these regions more easily causing a faster hydrolysis (Laycock et al., 2017).

Passing to the exposure conditions, they can be further discriminated in abiotic and biotic factors (Kijchavengkul and Auras, 2008). Temperature, pH and moisture are among the abiotic factors that can affect the hydrolysis reaction and also the microbial activity (Haider et al., 2019). By increasing temperature and moisture, both hydrolysis and microbial action increases, especially if the temperature is above the glass transition of the polymer and so it is more flexible and accessible to water and microbes (Kijchavengkul and Auras, 2008). In biotic conditions, enzymes (proteins with a complex 3D structure) are the most important contributors and their degradation activity can be influenced by oxygen levels, pH, nutrients and microbial population (Laycock et al., 2017).

Another important aspect to consider is that a plastic item is not only composed of polymers, but also included are additives that have their specific influence on biodegradation and must be taken into account. Additives are used because they are chemically and physically active, modifying the polymer chemical structure, mechanical properties, optical characteristics etc. (Kyrikou and Briassoulis, 2007). They can be divided in functional additives (e.g. plasticizers, stabilisers, flame retardants), colorants (e.g. dyes, pigments), fillers (e.g. calcium carbonate, clay) and reinforcements (e.g. glass and carbon fibres) (Hahladakis et al., 2018). Depending on the types of additives, their presence can accelerate or hinder biodegradation (Kijchavengkul and Auras, 2008). In the framework of biodegradable plastics, additives should be considered to have their dedicated regulatory assessment and be included in the testing of the material as a whole, and those certified as biodegradable should consider the polymer as well as the additives that are incorporated (SAPEA, 2020).

5. Standardisation and certification of biodegradable plastics

Most of the research performed on the biodegradation of polymeric materials have been focused on soil or compost environments, with little attention to the aquatic systems (Eubeler et al., 2009, Kijchavengkul and Auras, 2008, Kyrikou and Briassoulis, 2007).

Since the 1980s, different efforts have been made to develop standard tests for biodegradation, starting with guidelines developed by the Organization for Economic Cooperation and Development (OECD) (OECD, 1992). The American Society for Testing and Materials (ASTM), the European Committee for Standardization (CEN) and International Organization for Standardization (ISO) have also been working on standards that are

available by paying a fee, but difficult to compare due to the different definitions of biodegradability adopted (Kyrikou and Briassuoilis 2007; Harrison et al., 2018; Filiciotto and Rothenberg, 2021).

In general, all these methods refer to specific environments with specific characteristics and microorganism population, and are often a balance between real time conditions and feasible testing time (Eubeler et al., 2009; Filiciotto and Rothenberg, 2021). Their main criticisms are related to the lack of representability of real environmental conditions and on the analytical procedure applied to assess and quantify the biodegradation (Haider et al., 2019). Currently, the main procedures applied involve measurements of oxygen consumption or CO2 evolution, polymer weight loss, molecular weight evolution, tensile properties, enzyme assays, extent of fragmentation and ecotoxicity (Kyrikou and Briassoulis, 2007; Lott et al., 2020). It is important not to use a single analytical procedure to assess the biodegradation, since it can cause misleading interpretations (Albertsson and Hakkarainen, 2017).

The recent SAPEA report has tried to provide some clarity and new guidelines for a realistic assessment of plastic biodegradation (SAPEA, 2020). Considering regulatory aspects, the definition of plastic biodegradation reported in Table 1 needs to be accompanied with some specifications that include the type of open environment in which the biodegradation occurs, and the extent of biodegradation reached in a pre-defined timeframe. The report introduces a new approach for biodegradation standard testing, based on a three-tier testing scheme that is composed of the following steps: (1) the plastic material is tested at laboratory scale possibly using real inoculums, not prepared ad-hoc; (2) the material undergoes field testing in real natural conditions; (3) the plastic material in tested in tanks (mesocosms), simulating real environmental conditions in larger volumes than compared to laboratory tests. By combining the data obtained from these three types of tests it is possible to obtain a full assessment of the biodegradation of the tested material (SAPEA, 2020).

Standards test methods give the means to compare the biodegradability of a material worldwide but do not provide certifications, that are instead given by accredited bodies following payment (Filiciotto and Rothenberg, 2021). Usually, independent accredited testing facilities test the material following standard procedures and pass the results to the certifier that review them and eventually give the certification to the client (SAPEA, 2020). Currently, the main institutions that provide certifications are: TÜV Austria, DIN CERTCO (Germany), Biodegradable Products Institute (BPI; USA), Japan BioPlastics Association (JBPA), Australasian Bioplastics Association (ABA) and the European Union (Filiciotto and Rothenberg, 2021, SAPEA, 2020). TFurther efforts and harmonization actions are needed to create effective certification and labelling systems that are based on environmentally realistic test methods to prevent false claims by the industry and misleading of consumers.

6. Environmental considerations of biodegradable plastics

Biodegradable plastics have a wide range of applications (European bioplastics, 2021) which can lead to their introduction or leakage into the natural environment. For example, biodegradable plastics are used directly in the environment as agricultural mulch films (Kasirajan and Ngouajio, 2012). Other uses are as fibres used in wet wipes and sanitary products which when disposed of via the sewage system can shed fibres; these can be discharged into aquatic environments or be introduced to land via the application of sewage sludge as a fertiliser (O Briain et al., 2020, Zubris and Richards, 2005, Daria et al., 2020). Research to understand the environmental risks associated with biodegradable plastics is currently in the early stages, and further work is therefore required to evaluate their impacts and consider these in relation to conventional plastics.

Ecotoxicological data for biodegradable polymers is scarce. In contrast to many other chemical substances, the environmental impact assessment of biodegradable polymers is not covered by the European legislation on chemicals (REACH). A recent proposal calls for more stringent criteria for biodegradable polymers (ECHA, 2020), which may increase future testing of these materials. When considering the potential ecological impact of plastic polymers either petrochemical or biodegradable, the exposure time and concentration can influence their ecotoxicity. Currently biodegradable plastics only constitute ~0.3% of overall plastic production, consequently the emissions to the environment are considerably lower than for conventional plastics. As the quantities of biodegradable plastics are expected to increase (European bioplastics, 2021) so too may the quantities in the environment (i.e. the concentration to which an organism is exposed). The exposure time is linked to persistence, i.e. the biodegradability of the polymer in a particular environment. Studies indicate that biodegradation rates can vary widely between environments (terrestrial or aquatic), exposure conditions and seasons (e.g. temperature, oxygen availability, pH) and the microbial communities within the environment (Sintim et al., 2020, Napper and Thompson, 2019, Dilkes-Hoffman et al., 2019a, Haider et al., 2019, Narancic et al., 2018), in addition to the properties of the polymer itself (chemical composition, crystallinity, thickness and shape) (Volova et al., 2010). Where conditions for biodegradation are not favourable, biodegradable plastics may persist in the environment for substantial periods of time (see sections 6.1 and 6.2).

A further concern is related to additives contained within biodegradable plastics (SAPEA, 2020). As with conventional plastics, bio-based plastics contain additives used to enhance functionality or convey certain desirable properties. While bio-polymers are often promoted as a more sustainable alternative to petrochemical plastics, it is not known whether they represent a safer alternative with regards to the chemicals they contain. Several chemicals, such as phthalates and per- and polyfluoroalkyl substances (PFAS) which are used in conventional plastic products have also been detected in biodegradable or bio-based items (Anderson and Shenkar, 2021, Timshina et al., 2021). Additionally the behaviour of chemical additives such as their potential to leach into the environment and their persistence after the polymer has completely biodegraded, has not been evaluated.

Recent studies indicate that products made from biodegradable polymers may have similar toxicity to conventional plastics (Zimmermann et al., 2019, Zimmermann et al., 2020). Non-targeted chemical screening of 42 samples of biodegradable and bio-based plastic products, many of which were used as food contact material, indicated that those composed of starch and cellulose contained the greatest number of chemical features (typically >10,000) (Zimmermann et al., 2020). Of the 42 products tested, 29 contained chemicals which induced baseline toxicity, 18 which induced oxidative stress, 10 antiandrogenicity and one estrogenicity in the bacterium *Aliivibrio fischeri.* While the authors did not identify every chemical signature, they found that certain chemicals were present in all of the plastics tested (bio-based and conventional plastics), and others were specific to polymers/products. The study also indicated that across all of these endpoints, more commercial products induced toxicity than raw materials (Zimmermann et al., 2020) emphasising the need for testing of final products.

6.1. Biodegradable plastics in terrestrial environments

To date, the majority of research into the degradation of biodegradable plastics has been focussed on terrestrial ecosystems, and in particular studies have considered their use in agricultural applications. Modern agriculture relies heavily on the use of plastic materials. In some cases, these are applied directly to

the open environment, for example mulch films, which are challenging and costly to recover and separate from soil (Kasirajan and Ngouajio, 2012). Plastic mulch films are used globally to increase crop yields while lowering the consumption of water, fertilizers and herbicides, and minimising soil erosion (Kasirajan and Ngouajio, 2012) and thus convey clear societal and economic advantage in terms of food production and security.

Most mulch films are composed of polyethylene and residual fragments in soils can accumulate reducing water infiltration, altering soil ecosystems and ultimately plant germination and growth (Liu et al., 2014, Gao et al., 2019). Biodegradable mulch films offer similar physical barrier properties to conventional plastic films (Bandopadhyay et al., 2018), but are designed to be ploughed into the soil over which they were used, where it is anticipated they will biodegrade (Hayes and Flury, 2018). As such they are proposed to offer a promising advantage over conventional polymers (SAPEA, 2020). Biodegradable mulch films (PLA, PHA, PBAT, PBS) have been used since the 1980s (Sintim and Flury, 2017) and presently account for ~ 5% of the market share (APE Europe, 2020).

The standards for soil degradation (ISO 17556:2019 or ASTM D5988) specify that at least 90% of the organic carbon must be converted to carbon dioxide within two years. The deterioration of mulch film varies with soil type and climate (temperature, moisture and solar radiation), meaning biodegradation rates can vary geographically as well as seasonally (Miles et al., 2017, Li et al., 2014b, Sintim et al., 2020). Field studies conducted in two regions of Vietnam reported that under natural soil conditions PHA films lost 98% and 47% of their weight (Boyandin et al., 2013). At three test sites in the USA, commercial PCL mulch films buried at a depth of 8-12cm in natural soil for 24 months lost between 11 – 98 % of their area (Li et al., 2014b). In many agricultural systems, mulch films are used for a single growing cycle and may be applied to the same field year after year. Hence there is the potential for localised accumulation (Miles et al., 2017, Bandopadhyay et al., 2018).

When ploughed into soil, biodegradable mulch films provide an input of carbon, albeit a relatively small one. The growth of soil microbes in agricultural soils is usually carbon limited and studies have shown that microbes can respond to these small inputs of carbon, causing increases in biomass and enzyme activity (Li et al., 2014a), and changes in microbial community structure, for example enrichment of fungi (Li et al., 2014b, Moreno and Moreno, 2008, Ma et al., 2016, Muroi et al., 2016). Contrary to this, Bandopadhyay et al. (2020) found only minor site-specific effects on soil microbial communities and their function between biodegradable and conventional plastic mulch film treatments. It is not altogether clear what overall effect the changes in microbiota may have on the soil ecosystem and functioning as a whole, or how long-lasting these effects may persist.

Further studies have indicated that in the short-term (< 3 years), soil-biodegradable plastic mulch films may cause minor effects, and hence pose only minimal risk to soil physical, chemical and biological properties (Sintim et al., 2019, Li et al., 2014a). For instance, the soil quality index (based on the assessment of pH, electrical conductivity, total organic carbon, microbial biomass and β-glucosidase activity which measures metabolic capability) was shown to marginally reduce after an 18-month exposure to biodegradable mulch films compared to the control (no-mulch) treatment (Li et al., 2014a). Similar to polyethylene mulches, studies have reported that biodegradable mulch films have no measurable impact on the nitrification potential of soils (Ardisson et al., 2014, Kapanen et al., 2008) which is a further measure of soil health. A four-year study concluded that biodegradable mulches increased soil aggregate stability and water infiltration compared to conventional mulches, which were deemed as beneficial impacts to the soil (Sintim et al., 2021).

Further work is required to build upon these initial studies and examine whether the same observations occur in different geographic regions.

Tests where seeds were sown in soils which had previously been exposed to 1% (w/w) biodegradable plastic fragments for 6-7 months, did not find significant impacts on germination or the growth of cress, barley or sorghum (Rychter et al., 2006, Sforzini et al., 2016, Muroi et al., 2016). However, studies which have incorporated biodegradable fragments into soils have detected impacts on plant species. Following a 2-month exposure, a significant reduction in shoot biomass, total plant biomass and fruit biomass was observed for wheat plants grown in soils containing 1% (w/w) starch-based biodegradable plastic, compared to control groups (Qi et al., 2018). Ryegrass germination and growth were also significantly reduced when grown in soils containing PLA fragments (0.1% w/w) (Boots et al., 2019). While residues of conventional plastic-film mulches can also impact plant growth (Liu et al., 2021), biodegradable polymers have been demonstrated to have a greater negative effect on crop growth and yield than polyethylene (Qi et al., 2020, Qi et al., 2018, Boots et al., 2019).

Among terrestrial organisms, earthworms are key ecosystem engineers and important for soil health. Conventional plastic particles can reduce the growth rate, increase mortality rate and alter the metabolic activity and burrowing behaviour of earthworms (*Lumbricus terrestris, Eisenia foetida*, *Eisenia andrei*) (Cao et al., 2017, Huerta Lwanga et al., 2016, Rodriguez-Seijo et al., 2017, Prendergast-Miller et al., 2019). Studies on the impacts of biodegradable plastics on earthworms are limited. Boots et al. (2019) report that PLA fragments in soil (0.1% w/w) caused a reduction in worm biomass. Anecic species of earthworms such as *L. terrestris* construct permanent, long vertical burrows. They feed at the soil surface and drag organic matter into the burrows, this process has also been shown to redistribute conventional and biodegradable plastics within soils (Zhang et al., 2018, Rillig et al., 2017) and may enhance the degradation of biodegradable plastics (Sanchez-Hernandez et al., 2020).

As discussed above, studies show differing results when considering the effects of biodegradable plastics on i) microbial communities, ii) soil health and iii) terrestrial species. Evidence has indicated that biodegradable plastics may have little effect on the physical and chemical properties of the soil (Sintim et al., 2019, Li et al., 2014a) while the presence of fragments can cause detrimental effects on crop growth (Qi et al., 2020, Qi et al., 2018, Boots et al., 2019). International standards (ISO 17556:2019 or ASTM D5988) specify that 90% biodegradation must be achieved in two years, thus there is potential for localised accumulation if there is year-upon-year application of film; the effect of this remains currently unexplored.

6.2. Biodegradable plastics in aquatic environments

Aquatic environments span a diversity of habitats including rivers, lakes, estuaries and coastal ecosystems, the open-ocean and the deep sea, consequently there is substantial variability in temperature, oxygen availability, light and microorganism assemblages. The Vinçotte OK Biodegradable Water conformity mark (Vincotte, 2013) states that there should be 90% biodegradation in aquatic environments in 56 days; however, this assumes temperatures are between 20 to 25°C which are not representative of the majority of natural aquatic environments.

The biodegradation rates of polymers differ widely between aquatic environments and polymer types (Green et al., 2015, Volova et al., 2007, Nauendorf et al., 2016, Briassoulis et al., 2019, Bagheri et al., 2017), as also found for terrestrial studies. Based on available data, it was calculated that a PHA bottle may take between

1.5 to 3.5 years to biodegrade completely in the marine environment (Dilkes-Hoffman et al., 2019b). Bagheri et al. (2017) compared the biodegradation of a PLA film in artificial seawater and freshwater with PLGA, PCL, PHB and PBAT. PLGA had completely degraded in ≈270 days while PHB had lost only ~9% of its initial weight after 365 days and all other polymers showed no significant degradability in either treatment. These studies suggest that either intact items or fragments may remain in the aquatic environment for prolonged periods with the potential for ecological impacts.

Laboratory and field experiments comparing the deterioration of PHB at the sediment-water interface with PBS and Polybutylene sebacate-co-terephtalate (PBSeT) report all materials had biodegraded after 200 days (Briassoulis et al., 2020). Evidence suggests that the biodegradation rate, or the degree of disintegration, was greater for polymers (PHA, PHB, PBSe, PBSeT) in contact with the benthos than when in the water column (Briassoulis et al., 2019, Mayer, 1990). This may be due to the presence of diverse microorganisms within the benthic zone which could increase biodegradation, or due to bio-fouling the samples suspended in the photic zone may have reduced biodegradation rates; both of which require further investigation (Briassoulis et al., 2019, Lott et al., 2020).

Research comparing the biogeochemical effects of conventional and biodegradable plastic bags found that the presence of both types of bags caused anoxic conditions within underlying sediment and the reduction of primary productivity and significantly reduced the abundance of infaunal invertebrates (Green et al., 2015). Sediment containing PLA had approximately half the concentration of ammonium in porewater as sediments containing conventional polymers (PVC, HDPE), which the authors hypothesise may be due to the chemical structure of PLA which can absorb cations from the environment and thus reduce concentrations in the porewater (Green et al., 2016, Green et al., 2017). A study investigating the effects of biodegradable plastic bags on seagrass meadows in the Mediterranean Sea demonstrated that the reduction in sediment oxygen and pH levels caused by the presence of the biodegradable bags also influenced the growth of seagrass leading to increased inter- and intraspecific competition between plants (Balestri et al., 2017). These studies indicate that the presence of biodegradable plastics have the potential to rapidly, within 1-2 months, alter marine assemblages and the ecosystem services they provide.

Research to examine the acute toxicity of PLLA particles (10 – 100 µg/L) on mussels (*Mytilus edulis*) demonstrated there was a dose dependent down-regulation of lipids, particularly glycerophospholipids which are important for the structure of biological membranes (Khalid et al., 2021). A reduction in the in-vitro fertilisation rates of solitary ascidians was found to correlate with higher exposure concentrations of PLA and PET control, but no statistical significance was identified between material types (Anderson and Shenkar, 2021). The authors attribute the effects to the presence of additives, such as phthalates, which were present in both materials.

Chronic exposures (30 – 60 days) to PLA induced stress responses in benthic invertebrates including oysters and lugworms (Green et al., 2016, Green et al., 2017, Green, 2016). Lugworms were also shown to exhibit behavioural responses, producing fewer burrows/casts when exposed to PLA, PE and PVC than in control treatments (Green et al., 2016). The process of bioturbation through the action of burrowing organisms, is important for the redistribution of oxygen and nutrients in sediments. The findings of this study therefore suggest potential ecological consequences through the reduction of bioturbation by lugworms. Following a 52-day exposure to PLA microplastics (25 mgL^{-1}), mussels were shown to produce fewer byssal threads which also had a lower strength of attachment, compared to control groups, (Green et al., 2019). Mussels are important ecosystem engineers, modifying the structural complexity of benthic habitats due to their

aggregation into beds. The authors highlight that both conventional and biodegradable microplastics could impact the functioning and structure of sedimentary habitats.

Biodegradable plastics have been shown to deteriorate into microplastic fragments under artificial freshwater and seawater experiments (Wei et al., 2021, González-Pleiter et al., 2019). PBAT fragmented more readily than polyethylene under aquatic conditions and the rate of PBAT fragmentation was faster in seawater than in freshwater, attributed to faster hydrolysis under the basic pH conditions in seawater (Wei et al., 2021). The effect of nano-sized PHB particles derived from the fragmentation of larger particles under simulated natural environmental conditions on three freshwater species was investigated. PHB-nanoplastics induced an increase in reactive oxygen species in all organisms and damage to cell membranes which caused a significant decrease in the growth of cyanobacteria (*Anabaena* sp.) and green algae (*Chlamydomonas reinhardtii*) and a significant immobilisation of *Daphnia magna* (González-Pleiter et al., 2019). While this study shows a potent acute toxicological effect, it is not clear what specific concentration of nano-PHB were used and whether these were environmentally relevant.

In summary, the rate of biodegradation varies considerably between different marine and freshwater environments. Exposure to biodegradable plastics has been shown to induce a range of deleterious effects on aquatic organisms including oxidative stress and behavioural responses such as reduced burrowing activity in lugworms (Green, 2016, Green et al., 2016, Green et al., 2017, González-Pleiter et al., 2019). Further work is required to elucidate the role of additives compared to the polymer itself on toxicity and observe a range of endpoints across a diversity of aquatic organisms. Further research is also needed to consider the wider biogeochemical effects of biodegradable plastics on aquatic ecosystems which is currently lacking.

7. Social considerations of biodegradable plastics

Widespread concern about plastic pollution in the environment has encouraged consumers to opt for alternatives to conventional plastic products. Over the last decade, partly driven by this shift in consumer demand, there has been substantial development of alternative plastics with higher biodegradable properties. Manufacturers have marketed these products using ecological strategies, known as green-marketing, to encourage consumers to buy or use them (Koenig-Lewis et al., 2014).

In green-marketing, ecological claims have been criticized for confusing or misleading buyers (Koenig-Lewis et al., 2014). This leaves consumers vulnerable to 'greenwashing', where some companies seek to make environmental claims about their products based on partial analysis of the underlying science in the expectation that a majority of consumers believe such claims (Belz and Peattie, 2012; Polonsky et al., 1997). This has led to calls for more rationality in promoting and evaluating 'green' products.

The UK's Competition and Markets Authority (CMA), a non-ministerial government department in the United Kingdom, has been concerned that the surge in demand for green products and services could incentivise some businesses to make vague, false or misleading claims about the sustainability or environmental impact of the products or services they sell (CMA, 2020). Examples of misleading behaviour could include (CMA, 2020):

- Exaggerating the positive environmental impact of a product or service
- Using complex or jargon-heavy language
- Implying that items are eco-friendly through packaging and logos when this is not true

Labels, logos and material type are the most important features for consumers in identifying environmentally-friendly products and packaging (Lindh et al., 2016; Scott and Vigar-Ellis, 2014). For example, a study conducted in South Africa (Scott and Vigar-Ellis, 2014), investigated how consumers identify environmentally-friendly packaging of food products. They reported that 45% of the participants looked for labels, while 30% looked for images or logos such as the recycling logo. The packaging material itself was used by 18% of the participants to judge if packaging was sustainable, and only 12% of the participants admitted to not knowing the difference between sustainable and other packaging.

The disposal of a product or packaging is another feature consumers use to identify sustainability. A study of consumers in Italy showed that the aspect of disposal (i.e. whether the packaging is recyclable, non-recyclable or biodegradable) of food packaging was most important compared to other packaging attributes. The participants regarded biodegradable packaging as having many advantages over recyclable and non-recyclable packaging and consumers relied heavily on labelling and logos to help them determine whether they viewed a product as sustainable (Arboretti Giancristofaro and Bordignon, 2016).

Consumers are also shown to be unfamiliar with and therefore confused by the term 'bio-based' plastic (Koutsimanis et al., 2012; Sijtsema et al., 2016). For example, research by Koutsimanis et al., (2012) found that only 55% of the participating US consumers answered correctly to the question: "Which raw materials are used to produce bio-based containers?" While the term bio-based plastic may imply that the material is fully biodegradable this is not the case (see section 4).

It is necessary to have clear definitions and product labelling to indicate appropriate usage and disposal of products and packaging (Thompson et al., 2009). With the development of new materials, such as biodegradable plastics, it may lead to confusion over which is the correct waste stream for disposal. For example, in Germany only 10% of consumers dispose of fossil fuel-based plastic packaging incorrectly, compared to 63% of consumers that dispose of compostable, bio-based plastic incorrectly (Taufik et al., 2020).

There needs to be the availability of a dedicated waste stream for the consumer to use and the appropriate infrastructure; such as an industrial composting facility. Plus, the consumer needs sufficient understanding to correctly separate their waste accordingly (Napper and Thompson, 2019). A lack of clarity and understanding from the consumer may lead to them disposing of an item incorrectly, e.g. via the recycling waste stream which could cause contamination of the recyclate. Other potential barriers for biodegradable plastics are higher prices, lack of availability, unfamiliarity and perceived lower quality (see section 8) (de Jonge and van Trijp, 2013; Hughner et al., 2007; Magnier and Crié, 2015; Stern, 2000).

We should not consider biodegradable plastics as a complete technical solution, thereby excusing our environmental responsibility, such as littering. It has been reported that the general public find it more acceptable to drop items composed of biodegradable plastics than non-biodegradable plastics because it is thought they are less harmful for the environment (Dilkes-Hoffman et al., 2019a). The importance of reducing or reusing plastic and biodegradable plastic items as much as possible throughout their lifespan and reducing reliance on single-use products needs to be communicated.

8. Economic considerations of biodegradable plastics

Bio-based plastics are derived from biomass, the majority of which originates from first-generation feedstock, defined as crops suitable for human or animal consumption (Brizga et al., 2020). With the projected expansion of the bio-based plastics market over the next decade (European bioplastics, 2021) and proposed substitutions of conventional polymers with biodegradable alternatives in some applications, it is predicted that there may be increased competition for land or resources to meet this demand (Posen et al., 2017, Brizga et al., 2020). A study by Brizga et al. (2020) estimated that at least 7.4 million hectares of land would be required to satisfy the land requirements to replace all petrochemical-based plastic packaging with bio-based alternatives in Europe. On a global scale, they estimate that 61 million hectares would be required to produce enough bio-based feedstock (Brizga et al., 2020). his may result in changes to land-use and present challenges in terms of natural resources and food security (Escobar et al., 2018) and as such demands for raw materials from bio-based plastics must be balanced against competing claims on land.

A number of economic barriers are frequently cited in relation to the slow adoption and development of bio-based and biodegradable polymers. Compared with conventional petrochemical-based plastics, the material price for bio-based polymers is high and the technical production processes are complex, making the final product more expensive than conventional plastics (Shen et al., 2020, Fahim et al., 2019). The expansion of the bio-based market may lead to the wider commercialisation of biodegradable plastics and thereby attain economies of scale (Ren, 2003).

The global interest in 'zero-waste' (which has principles based on the circular economy and the waste hierarchy (Franco-García et al., 2019)) has offered new business opportunities for biodegradable plastics (Brizga et al., 2020, Zaman, 2015). Biodegradable plastics may provide opportunities to move towards a more circular economy, through higher resource efficiency, but only if waste products can re-enter the economy though a biological cycle (Confente et al., 2020, SAPEA, 2020). Appropriate management and disposal practices are required for this to be achieved. In the absence of clear labelling (sections 5 and 7), there is confusion over how to appropriately dispose of biodegradable plastic products (Sijtsema et al., 2016) and as such they may enter inappropriate waste streams.

Contamination of recycling streams presents a problem. Biodegradable plastics may contaminate existing plastic recycling streams leading to low-quality recyclate, which may not be usable in the production of new products (Alaerts et al., 2018, Rujnić-Sokele and Pilipović, 2017) and therefore interrupt the circular flow of materials. For example, a study found that contamination of 0.1% PLA can cause structural problems to PET, rendering the material unusable for many applications (Alaerts et al., 2018). Alternatively, biodegradable plastics may enter organic recycling waste streams, where any residual material from incomplete degradation may compromise the quality of the resulting compost. As the compost derived from this process is largely intended to be returned to the natural environment, any plastic residues may also be introduced to the environment where there may be unintended environmental consequences (see section 6) (Hann et al., 2020). As such evaluating any unintended consequences arising from end-of-life disposal of biodegradable plastics is required.

9. Conclusion and outlook

The degradability of a polymer largely depends on its chemical structure, rather than its carbon source (i.e. bio-based or petrochemical-based). The potential benefits in terms of biodegradability will only be realised if end-of-life items reach an appropriate receiving environment for biodegradation to occur. Humidity,

temperature, oxygen availability and microorganisms vary across environments, resulting in different biodegradation rates. Additionally, manufacturers include additives at various concentrations and so materials made from the same polymer should not be considered chemically identical (Lambert and Wagner, 2017), placing emphasis on the need for testing (and certifying) biodegradability of the end-product rather than on the pure polymer. This calls for an effective framework of standards and certifications of biodegradable materials that take into account the plastic material as composed of polymers plus additives and the type of open environment where the biodegradation should occur.

The absence of clear labelling has led to consumer confusion when trying to choose 'sustainable' products and the appropriate way to dispose of items once they have reached the end of their life in service. This confusion can lead to an increase in biodegradable items become littered in the environment (Dilkes-Hoffman, et al 2019a), or disposed of via an uncorrected managed waste stream, e.g. with conventional plastic recycling, which may contaminate the resulting recyclate and rendering it unusable (Alaerts et al., 2018, Rujnić-Sokele and Pilipović, 2017).

Currently there are rather limited empirical data on the environmental effects of biodegradable plastics. The persistence of fragments from incomplete degradation along with residues of any chemical additives can induce a range of effects, including reduction of crop growth and yields, and in terrestrial and aquatic organisms a range of stress and behavioural responses have been reported, along with wider implications on ecosystem functioning.

Looking ahead, as biodegradable plastic consumption is projected to increase, economic opportunities may be presented. In certain applications biodegradable plastics can have a role as part of a circular economy (see Hann et al., 2020, SAPEA, 2020). Biodegradable polymers may offer advantages over conventional plastics where degradability is part of the product's function, for example as agricultural mulch films, bags to contain organic waste or drug delivery agents in biomedicine (SAPEA, 2020, Haider et al., 2019). However, substituting all commodity plastics with biodegradable alternatives may have unintended environmental and economic repercussions and are not a solution for plastic pollution (Narancic et al., 2018). Further product testing and research is required to holistically appraise their effect on society, economy and the environment.

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