1 The role of waste management in reducing bioplastics' leakage into the

environment: a review 2 3 Mirko Cucina^{1*}, Patrizia de Nisi¹, Fulvia Tambone¹, Fabrizio Adani¹ 4 5 ¹Gruppo Ricicla Lab. – DiSAA – Università degli Studi di Milano, Via Celoria 2, 20133 6 Milano, Italy. 7 8 *Corresponding author: 9 Mirko Cucina, PhD 10 mirko.cucina@unimi.it 11 +393338579190 12 Via Celoria 2, 20133, Milano (IT) 13 14

Abstract

Bioplastics are becoming more and more widespread as substitutes for petroleum-derived plastics due to their biodegradability. Bioplastics degradation under different environments has been described and reported to depend mainly on bioplastics' compositions and the environmental conditions. Incomplete degradation during waste management processes and leakage of bioplastics into the environment are becoming major concerns that need to be further investigated. In this context, the present paper aimed to review recent literature dealing with biodegradation of bioplastics under industrial (e.g. anaerobic digestion and composting) and natural (e.g. soil and water) environments, and to link it to the potential bioplastics' leakage into the environment. Reviewed data were used to estimate the potential role of waste management processes in decreasing the potential leakage of bioplastics. Depending on bioplastics' type and processing conditions, waste management can effectively reduce bioplastics' potential leakage, decreasing the concentration of these materials that can reach the natural environments.

Keywords

Anaerobic digestion; Bioplastic; Biodegradation; Composting; Leakage.

1. Introduction

41

42 About 60 million tonnes of petroleum-derived plastics were produced in 2019 in Europe, whereas the worldwide production accounted for 368 million tonnes 43 (PlasticsEurope, 2020). Europe's plastics demand in 2019 (about 50 million tonnes) was 44 45 mainly related to packaging (39.6 %), building and construction (20.4 %), and automotive (9.6 %) purposes. Nowadays, about 32 % and 43 % of plastic wastes are 46 47 recycled or processed for energy recovery in Europe, respectively, with the residual plastic being landfilled (PlasticsEurope, 2020). Although the quantity of plastics wastes 48 sent to recycling has increased by 92 % in Europe since 2006 (PlasticsEurope, 2020), 49 concerns about the sustainability of petroleum-derived plastics have arisen in recent 50 decades. Indeed, besides being produced from a non-renewable raw material (oil), 51 52 plastic wastes have been recognized as one of the most dangerous pollutants for the 53 environment due to their poor degradability and to their tendency to fragmentation and the production of microplastics that accumulate in the environment (Bellasi et al., 54 55 2020). Oxo-degradable plastics, which were first introduced as eco-friendly substitute 56 of petroleum-derived plastics, were recently recognized to give rise to serious 57 environmental issues, i.e. fragmentation into non-degradable microplastics, potential toxic effects of the oxidising additives (Abdelmoez et al., 2021, European Commission, 58 59 2018). In addition, polymers produced by renewable biomass such as bio-polyethylene 60 have been reported to be non-biodegradable (Mendieta et al., 2019). In this context, bioplastics have been introduced in recent decades as environmentally 61 62 friendly and sustainable alternative materials to petroleum-derived plastics. 63 The global bioplastics production capacity is foreseen to increase from around 2.1 64 million tonnes in 2020 to 2.8 million tonnes in 2025 (European bioplastic, 2020).

65 The rapid increase in bioplastics use is mainly related to policy changes and to a 66 positive perception of bioplastics by the end-users. From the policy side, the European Union planned to ban the most common single-use plastic items by 2021 (European 67 Parliament, 2019) as one of the strategies to achieve the 17 Sustainable Development 68 69 Goals described by United Nations (Fonseca et al., 2020). On the other side, besides a 70 generally poor knowledge of bioplastics, the general public's perception of bioplastics 71 and their biodegradability is positive (Dilkes-Hoffman et al., 2019, Taufik et al., 2020). 72 Bioplastics are a wide family of compounds that comprise (i) biodegradable and bio-73 based materials (e.g. starch-based polymers, polylactic acid – PLA, polyhydroxyalkanoates - PHAs), (ii) biodegradable and petroleum-derived materials 74 75 (e.g. polycaprolactone – PCL, polybutylene adipate terephthalate - PBAT) and (iii) nonbiodegradable bio-based materials (e.g. bio-polyethylene) (Abraham et al., 2021, Bátori 76 et al., 2018). Within all the bioplastics, PHAs, PLA and starch-based bioplastics 77 represent about 70 % of the entire bio-based biodegradable bioplastics produced and 78 79 used in 2020 (European bioplastic, 2020). Sources, main applications and production 80 capacity of these three bioplastics are reported in Table 1. Commonly, bioplastics are 81 composite material made of one main component (e.g. starch, PLA) blended with other components and additives. Due to that, in the present review the three bioplastics 82 83 studied (PHAs, PLA and starch-based bioplastics) were reported as *blends*, meaning 84 that they were made of composite materials. However, in this review only bioplastics blends made of biodegradable components have been considered. 85 86 PHAs biopolymers are obtained through microbial synthesis from carbohydrates and 87 then they are accumulated in the cell (Ganesh Saratale et al., 2021). PHAs are biodegradable polyesters characterized by physico-chemical properties not far from 88

89 those of petroleum-derived plastics (Papa et al., 2020), and their main applications are represented by packaging and agricultural/medical goods production. Although PHAs 90 91 production is actually limited by the high costs of substrates and the use of pure microbial cultures, several studies are focusing on the use of mixed microbial cultures 92 93 to reduce costs, which should permit enhanced PHAs usage (Villegas Calvo et al., 94 2018). Since PHAs blends entered the market, the share of this biopolymer family 95 continues to grow and production capacities are set to increase successfully almost 96 seven times by 2025 (Table 1) (European bioplastic, 2020). 97 PLA is obtained through the microbial fermentation of carbohydrates and subsequent polymerization (Folino et al., 2020). PLA is mainly used for the production of durable 98 99 and disposable goods (cutlery, glasses, dishes, and packaging) but is also used in building and construction, agricultural, medical applications and fibres production. The 100 101 production of PLA blends is also expected to grow due to new investments in PLA 102 production sites in China, the US, and in Europe and it is expected that these 103 biopolymers will represent the most produced bioplastics in 2025 (Table 1). 104 Starch-based blends are made of starch obtained from different crops (e.g. corn, rice, 105 potato) processed with plasticizers, and find their main applications in the production of 106 films, carrier bags and waste-collection bags (Abraham et al., 2021). Starch-based 107 blends were the first bioplastics to be widely used in common applications and they still 108 represent the most produced bioplastics, representing about 20 % of the bioplastics 109 produced in 2020. 110 Bio-based biodegradable bioplastics present different advantages, with the 111 biodegradability being the most important (Pilla, 2011). Biodegradation of bioplastics involves biotic and abiotic factors that lead to a complete degradation of the material 112

(Emadian et al., 2017). Generally, abiotic factors (e.g. temperature, water and sunlight) 114 lead to an initial chain scission in the bioplastic polymer, producing shorter oligomers that can pass through the cell walls of microorganisms. Microbial processes then complete the biodegradation of these shorter units into other compounds, depending on 116 the metabolic pathway of the microorganism (aerobic or anaerobic) (Bátori et al., 2018). Bioplastics biodegradation can be assessed by using international standard methods. The ISO 14855 (2018), ISO 17556 (2019), ISO 14851 (2019) and ISO 13975 (2019) 120 can be used to define whether a bioplastic is biodegradable or not in composting, soil, 121 aqueous and anaerobic digestion environments, as reviewed by Arikan and Ozsov (2015). Bioplastics usually showed a linear biodegradation and this was an indication that reaction rates were obeying a pseudo-zero order kinetic and that biodegradation rates were constant and independent of the bioplastic amount (Barbale et al., 2021, Chamas et al., 2020, Chinaglia et al., 2018, Degli Innocenti and Breton, 2020). The constant rate of biodegradation could be explained by considering that the biodegradation rate depended on the available C-polymer at the surface and not on the total C-polymer (Chinaglia et al., 2018). Similar considerations were reported by Modelli et al. (1999) who studied the degradation kinetic of PHA sheets. Whilst the substitution of petroleum-derived plastics represents a major goal to be 131 achieved, the fate of bioplastics in natural and industrial environments has still to be explored. In fact, some recent papers have pointed out that degradation kinetics of bioplastics are often incompatible with waste management systems (Battista et al., 133 134 2021, Folino et al., 2020, Zhang et al., 2018). Moreover, degradation kinetics appeared to be really slow in natural soils and aquatic environments, leading to possible 136 environmental concerns (i.e. bioplastic accumulation and fragmentation into

113

115

117

118

119

122

123

124

125

126

127

128

129

130

132

137 microplastics) (Bátori et al., 2018, Bhagwat et al., 2020, Emadian et al., 2017, Folino et al., 2020, Shruti and Kutralam-Muniasamy, 2019, Thakur et al., 2018). 138 139 In this context, the aim of the present paper is to review recent knowledge of biodegradation of bioplastics under controlled (e.g. waste management) and natural 140 141 environments, paying particular attention to the effect of waste management in reducing the potential bioplastics' leakage into the environment. 142 143 144 2. Bioplastics in waste management 145 Bioplastics enter the so-called *technosphere* after their production, through their collection and subsequent treatments in ad hoc plants (Degli Innocenti and Breton, 146 147 2020). Bioplastics degradation in the technosphere is crucial since bioplastics that are not processed and degraded can reach the biosphere. 148 149 150 2.1 Bioplastics collection 151 Separate collection of bioplastics is mandatory to allow for a better recycling of these 152 products after usage. In Europe, separate collection of bioplastics with bio-waste, i.e. 153 with the organic fraction of municipal solid wastes (OFMSW), was prescribed initially in 1994 (Pagga, 1998) and it is actually recommended (Dubois et al., 2020). 154 155 Nevertheless, the importance of a better system of certification and clearer instructions 156 for handling of bioplastics is needed. Indeed, it was recently pointed out that, beside the 157 positive appeal of bioplastics packages for consumers, lower correct disposal rates for 158 bioplastics packaging were recognized if compared to petroleum-derived packaging 159 (Taufik et al., 2020). This was probably related to the fact that consumers positively evaluate the biodegradability of bioplastics packaging (Magnier and Crié, 2015), 160

leading to a less careful collection and disposal of these products. However, an adequate management of such bioplastics by consumers is mandatory to capitalize on the environmental benefits of bioplastics. Due to the general recommendation of discharging bioplastics with OFMSW, the concentration of these products in the biowaste stream is rapidly increasing and is expected to reach high values in the coming years. For instance, bioplastics in Italy represented about 1 -2 % and 3 - 4 % (weight basis) of OFMSW in 2017 and 2019, respectively (ISPRA, 2020), and this value is expected to increase rapidly in future years, possibly reaching a concentration of 8-10 % (weight basis, forecast for 2030). Therefore, the knowledge of the fate of bioplastics during OFMWS management, i.e. through anaerobic digestion (AD) and composting, is becoming a major issue.

2.2 Anaerobic digestion

AD anaerobically degrades organic wastes to biogas and digestate through four successive phases, (1) hydrolysis, (2) acidogenesis, (3) acetogenesis and (4) methanogenesis (Wang et al., 2018). Biogas is a gas mixture mainly composed by methane and carbon dioxide (55-70 % and 30-45 %, v/v), as well as small amounts of other gases (oxygen, sulphuric acid, hydrogen) (Liu et al., 2019). Biogas can be used as an alternative energy source through its combustion in boilers or combined heat and power units; however, the interest in biogas conversion to high-value products has been increasing recently (Patel et al., 2020, Wu et al., 2016). Digestate is widely considered as a potential organic fertilizer, being rich in plant macronutrients (N, P and K) and organic matter (Castro et al., 2017, Peng et al., 2020, Tambone et al., 2017). AD can be operated at psychrophilic (18-20 °C), mesophilic (35-40 °C) and thermophilic (50-60

185 °C) temperature regimes (Hupfauf et al., 2018), with the last two conditions being the 186 more effective for organic matter degradation and biogas production. 187 Recent literature concerning bioplastics degradation during AD was reviewed and summarized in Table 2. In addition, the kinetic constant of biodegradation and the time 188 189 for complete degradation under anaerobic conditions were estimated using a pseudozero order kinetic model (Chinaglia et al., 2018). PHAs blends were recognized to 190 degrade faster than the other bioplastics during AD, and their biodegradation was not 191 192 strongly influenced by the temperature regime (Yagi et al., 2014, Yagi et al., 2013). 193 Indeed, the estimated time for complete degradation of PHAs blends during AD was found to be 31 \pm 20 days under mesophilic conditions and 36 \pm 28 days under 194 195 thermophilic conditions (Table 2). These results were in accordance with Noda et al. (2010) and Siracusa et al. (2008) who reported that PHAs can completely degrade under 196 anaerobic conditions in 5 - 6 weeks. PLA and starch-based blends showed a slower 197 degradation during AD in comparison with PHAs blends. PLA blends were found to 198 199 degrade completely in 423 ± 76 and 116 ± 48 days in mesophilic and thermophilic 200 conditions, respectively (Cazaudehore et al., 2021, Zhang et al., 2018). Similarly, 201 temperature also played a key role in starch-based blends' degradation during AD. 202 These bioplastics showed a significant reduction of time needed to complete 203 degradation when AD was performed under thermophilic instead of mesophilic conditions (- 60 %) (Calabro' et al., 2020, Cazaudehore et al., 2021). 204 205 Summarizing, the biobased biodegradable bioplastics studied showed a decreasing 206 degradation under anaerobic conditions following the order: PHAs blends > starch-207 based blends ≥ PLA. These differences can be related to the differences in chemical composition of the different bioplastics. Siracusa (2019) suggested that the degradation 208

process is more easy and natural for PHAs polymers and copolymers since they are produced directly from microorganisms. Conversely, the number of organisms able to degrade the chemical structure of synthetic biopolyesters (e.g. PLA) is limited. Therefore, the degradation of such synthetic biopolyesters depends on the environment and microbial population. In anaerobic conditions, fewer enzymes are present and the growth of microorganisms is slower, leading to a slow degradation of bioplastics. The role of temperature regimes in bioplastics degradation during AD was pointed out in recent literature (Calabro' et al., 2020, Cazaudehore et al., 2021, Folino et al., 2020). With the exception of PHAs blends, for which temperature did not influence anaerobic degradation, thermophilic temperatures (55 \pm 2 °C) significantly accelerated PLA and starch-based blends' degradation. This was probably due to changes in the mechanical properties of bioplastics that can occur only under thermophilic conditions. For example, PLA blends' degradation was enhanced by reaching their glass transition temperature (55-60 °C) that causes PLAs' mechanical properties to change, making them more hydrophilic and accessible for microbial hydrolysis (Marek and Verney, 2016). This was also in accordance with Hamad et al. (2015) who described that elevated temperature (58 °C) is needed to reduce the molecular weight of PLA blends and to start their biodegradation. Similarly, also the biodegradation of starch-based blends was enhanced by thermophilic temperatures. This was probably related to the fact that the polyester component of starch-based blends can be degraded only at elevated temperature, as described by Gil-Castell et al. (2020), who reported that polyesters such as poly(3)-hydroxybutyrate-co-3-hydroxyhexanoate (PHBH) biodegraded effectively only at temperatures higher than 58 °C. Taking into consideration both that AD of OFMSW is commonly performed with short hydraulic

209

210

211

212

213

214

215

216

217

218

219

220

221

222

223

224

225

226

227

228

229

230

231

retention times (HRT) (20-30 days) to optimise biogas yields and volumes of waste treated (Panigrahi and Dubey, 2019, Shrestha et al., 2020), and the reviewed literature concerning bioplastics degradation during AD, some of the following conclusions can be derived. Only PHAs blends were recognized to be compatible with AD processes conducted with conventional HRT. In fact, 80 % of degradation (weight basis) is usually considered a goal that should be achieved at the end of the HRT for a biomass suitable for AD (Ran et al., 2018), and PHAs blends can satisfy this requirement (Table 2). PLA and starch-based blends required about 10 - 15 times a conventional HRT of 30 days to degrade completely under mesophilic conditions. Moving to thermophilic regimes, the degradation kinetic increased but the objective of degrading 80 % (weight basis) of bioplastics in the HRT cannot apparently be achieved. Similar conclusions were reported by Battista et al. (2021), who observed only a partial degradation of single-use bioplastics under anaerobic conditions and the requirement of a very long retention time to achieve acceptable degradation rates. The high amount of bioplastics' residues in the digestate may also led to difficulties for its subsequent utilization, above all in new applications, i.e. nutrient recovery producing fertilizer-like material to be used in agriculture and /or the use as substrate for micro-algae cultivation. In particular, the effect of the presence of bioplastic residues in compost and digestate because of their use in agriculture is later described and discussed.

252

253

254

255

256

233

234

235

236

237

238

239

240

241

242

243

244

245

246

247

248

249

250

251

2.3 Composting

Composting aerobically degrades organic wastes to compost and two main by-products (heat and carbon dioxide) (Cerda et al., 2018, Wang and Zeng, 2018). Composting is a self-heating process that proceeds through three main phases, (1) mesophilic (25-40 °C),

(2) thermophilic (55-65 °C) and (3) maturation. Compost is a nutrient-rich organic amendment able to provide N, P, K and organic matter to the soil (Wang and Zeng, 2018). During composting, labile organic matter is mineralized and complex recalcitrant materials tend to concentrate, increasing the organic matter stabilization of the compost (Cucina et al., 2018). Performances of bioplastics degradation during composting taken from recent literature are summarized in Table 3. Assuming a pseudo-zero order kinetic model for the aerobic degradation of bioplastics (Chinaglia et al., 2018), the kinetic constant of biodegradation and the time for complete degradation under aerobic conditions were estimated and reported in the same Table. Data reported for bioplastics showed higher kinetic constants of degradation under thermophilic composting conditions than those reported for AD (Gómez and Michel, 2013). All the biobased biodegradable bioplastics studied showed a quick degradation, and the kinetic constant of degradation increased following the order: PLA blends < starch-based blends ≤ PHAs blends. Consequently, time estimated for complete degradation of bioplastics was 84 ± 47 days, 124 ± 83 days and 119 ± 43 days for PLA, PHAs and starch-based blends, respectively (Table 3). Several factors affect bioplastics degradation in composting environments, with temperature and bioplastics chemical composition being the most important (Emadian et al., 2017). The high temperatures of the active phase of composting (> 55 °C) allow reaching the glass transition temperature of the most common bioplastics, leading to the passage from the crystalline status of polymer to the amorphous one, and so increasing polymer hydrophilicity (Amin et al., 2019, Zhang et al., 2007), leading to higher hydrolyzation and enhancing the kinetic of bioplastics degradation during composting.

257

258

259

260

261

262

263

264

265

266

267

268

269

270

271

272

273

274

275

276

277

278

Values reported in Table 3 for bioplastics degradation under composting conditions showed that bioplastics can effectively degrade during a conventional composting process of OFMSW. Indeed, composting is a longer process than AD and usually lasts about 90 days (Ayilara et al., 2020), and most of the bioplastics studied were found to degrade in less than 90 days. Interestingly, a few papers investigated the behaviour of starch-based blends during mesophilic composting (23 – 25 °C) and reported that under aerobic conditions, temperature also played a key role in bioplastics degradation (Accinelli et al., 2012, Mohee et al., 2008). Indeed, starch-based blends degraded with a reduced kinetic under mesophilic conditions (-46 %) if compared to thermophilic temperatures. These results were in accordance with Rudnik and Briassoulis (2011) who observed a very slow biodegradation of PLA bioplastic under mesophilic composting. Certainly, composting of OFMSW is carried out at industrial facilities operating under thermophilic conditions (maximum temperature 55-65 °C), but home composting is becoming more and more popular in recent years, especially in rural areas (Vázquez and Soto, 2017). Differently from industrial composting, during home composting temperature peaks do not usually exceed 35 – 40 °C (Guidoni et al., 2018) and this could lead to issues of bioplastics accumulation in compost produced in this way. In recent years, the use of composite bioplastics to improve their degradation under composting conditions has been investigated (Ahn et al., 2011, Anstey et al., 2014, Sarasa et al., 2009). Blending of biofuel by-products with polybutylene succinate proved to enhance biodegradation of the composites during composting due to the increased content of soluble sugars in the composite itself (Anstey et al., 2014). Similarly, Ahn et al. (2011) and Sarasa et al. (2009) promoted PLA biodegradation

280

281

282

283

284

285

286

287

288

289

290

291

292

293

294

295

296

297

298

299

300

301

Although composting represents a valuable strategy to treat OFMSW and degrade bioplastics, it should be highlighted that composting is an energy-requiring process (Lin et al., 2018). As already discussed, bioplastics' environmental profile is strongly dependent on their end-of-life strategy. For instance, bioplastics have been proved to have a higher global warming potential if industrial composting and disposal transportation are considered as their end-of-life strategy during LCA analysis (Zhao et al., 2020). Taking this into consideration, AD or AD coupled to digestate composting should represent preferred treatments for OFMSW and bioplastics treatment (Edwards et al., 2018, Wainaina et al., 2020) and, thus, strategies to improve bioplastics degradation during anaerobic processes should be further investigated.

under composting conditions by blending it with poultry feather fibres and corn,

3. Bioplastics in the environment

The transfer or leakage of bioplastics from the *technosphere* to the environment can occur both accidentally (e.g. littering) and voluntarily (e.g. agronomic use of digestate or compost containing residues of bioplastics). In both case, bioplastics can reach soil and aquatic (freshwater and marine water) environments, posing a serious concern for their ability to degrade in these natural systems.

3.1 Soil environment

Since soil contamination by petroleum-based plastics disposal has emerged as a major issue in the last decades, several papers dealing with bioplastics degradation in soil can already be found. In this review, soil contamination due to bioplastic leakage from

327 waste management because of the use of compost and digestate was considered. 328 Nevertheless, it is important to keep in mind that soil can be polluted directly by using 329 agricultural wastes containing bioplastics (e.g. mulching). Within the natural environments (soil, aquatic and air), soil is usually recognized as the 330 331 one which can provide the fastest degradation of bioplastics, mainly due to the wide diversity of microorganisms that live in the soil (Emadian et al., 2017). Nonetheless, 332 333 bioplastics degradation rate is strictly related to the chemical composition of the 334 polymer, as well as to the soil characteristics (e.g. pH, clay and organic matter contents) 335 (Elsawy et al., 2017, Folino et al., 2020). Siracusa (2019) reported that ageing factors such as sunlight and temperature can also promote or slow down biodegradation rates of 336 bioplastics in soil. Consequently, biodegradation in soil can differ from place to place, 337 from season to season (Siracusa, 2019). These considerations can be the reason behind 338 339 the high variability of results found in literature concerning bioplastics degradation in soil, which are summarized in Table 4. Assuming a pseudo-zero order kinetic model for 340 341 bioplastics degradation in soil (Chinaglia et al., 2018), kinetic constants and times for 342 complete degradation in soil were also estimated (Table 4). 343 In soil, bioplastics showed a slow degradation if compared to both AD and composting. This fact could probably be due to the lower concentration of microorganisms in soil in 344 345 comparison with digestate and compost mixtures. Among the three blends studied, 346 PHAs' blends showed the highest kinetic constants of degradation in soil, followed by 347 starch-based blends and PLA blends. PLA blends showed the longest estimated time for 348 complete degradation, i.e. about 4-5 years, and this was probably due to the low 349 temperatures that characterize the soil environment in the experiments reviewed in the present paper, i.e. 20 - 30 °C (Kalita et al., 2021, Palsikowski et al., 2018). These 350

temperatures were far below the glass transition temperature of PLA blends, leading to such a slow degradation. PHAs blends and starch-based blends shared similar estimated times for complete degradation (1.2 and 1.6 years, respectively). The possible blending of bioplastics with other biodegradable materials was investigated in recent years in order to accelerate the biodegradation of bioplastics in soil. For instance, blending potato peel waste fermentation residues with PHBs led to a reduced crystallinity of the blend and to a faster degradation in soil compared to the pure PHBs (Wei et al., 2015). PLA degradation in soil has also been proved to be promoted by production of composites made of PLA and oil palm fibres (Harmaen et al., 2015). Although years are required for biodegradation of bioplastics in soil, this time is significantly shorter than that of the petroleum-derived plastics that require hundreds or thousands of years to degrade in soil (Chamas et al., 2020). These plastics (e.g. polypropylene, polyethylene, polyvinylchloride) are highly stable and cannot enter the degradation cycle of the biosphere, or show slow degradation rates that do not allow for a complete disintegration (Ahmed et al., 2018, Briassoulis et al., 2020). As a consequence of that, Chamas et al. (2020) reported that high-density polyethylene, polyvinylchloride and polystyrene buried in soil had half-life times of about 2,500 – 5,000 years that are much more than the times required to degrade bioplastics (Table 4).

369

370

371

372

373

374

351

352

353

354

355

356

357

358

359

360

361

362

363

364

365

366

367

368

3.2 Aquatic environment

Although the aquatic environment was recognized as the most susceptible to plastics contamination (Calabrò and Grosso, 2018), until now only a few papers dealing with the fate of bioplastics in fresh and seawater can be found. Results of literature study which are summarized in Table 5 including kinetic parameters of bioplastics degradation in the

aquatic environments, estimated assuming a pseudo-zero order model (Chinaglia et al., 2018). A large variability in bioplastics degradation in aquatic conditions was found but generally, degradation of bioplastics in fresh and seawater appeared to be slower than degradation in soil or under active waste management (AD and composting). This was mainly related to the characteristics of the aquatic environment that play a central role in bioplastics degradation. Besides bioplastics' characteristics, temperature, pH, nutrients content and microbial population density and diversity are the most important factors that affect bioplastics degradation in the aquatic environment (Harrison et al., 2018, Rana, 2019, Urbanek et al., 2018). Low temperature, nutrients and microbial population density might be responsible for the slow degradation of bioplastics in the aquatic environment. The role of temperature was highlighted by Volova et al. (2007) who studied the rate of PHAs degradation in seawater. The authors concluded that seasonal changes in water temperature were responsible for different rates of degradation. Sekiguchi et al. (2011) studied the degradation of PHBs bioplastics under three different seawater typologies, concluding that microbial community composition of the aquatic system can also influence significantly the degradation rate of bioplastics. The aquatic environment is really heterogeneous, i.e. it is possible to classify at least eight different liquid environments (Folino et al., 2020). Different aquatic environments host different microbial communities, and this could explain the differences observed in the degradation of the same bioplastics under different liquid environments (Emadian et al., 2017, Sekiguchi et al., 2011, Tosin et al., 2012). Due to all the described factors, PHAs blends showed a wide range of degradability in aquatic systems, and the estimated time for their complete degradation ranged from 50 days in seawater at 21 °C to 4,348 days in fresh and seawater at 25 °C (Bagheri et al.,

375

376

377

378

379

380

381

382

383

384

385

386

387

388

389

390

391

392

393

394

395

396

397

399 2017, Thellen et al., 2008). PLA blends presented the slowest degradation rate in aquatic environments (the average estimated time for complete degradation was over 10 400 401 years). Concerning starch-based blends, degradation in fresh and seawater, a high 402 variability was reported. Whilst Accinelli et al. (2012) obtained only a 1.5 % 403 degradation (weight basis) of starch-based bioplastics under both fresh and seawater 404 systems (25 °C, 90 days), other studies reported a significant degradation of these bioplastics. For instance, Tosin et al. (2012) obtained a 69 % degradation (weight basis) 405 of starch-based shoppers after 236 days and this quick degradation was probably related 406 407 to both characteristics of the tested materials and the environmental conditions (sea 408 water + sediment). 409 Blending bioplastics with biodegradable materials has been studied recently to enhance 410 their degradation in aquatic systems. Beltrán-Sanahuja et al.(2020) obtained a significant increase of the degradation kinetic in seawater (16 °C) by blending PLA with 411 cellulose (kinetic constant of degradation increased from 0.15 mg g⁻¹ d⁻¹, pure PLA, to 412 0.80 mg g⁻¹ d⁻¹, PLA and cellulose blend). 413 414 Summarising, times needed for complete degradation of bioplastics in aquatic 415 environments ranged from about 4 years (PHAs blends) to 12 years (PLA), but it must 416 be highlighted that, since the biodegradation of bioplastics in aquatic environments is 417 dependent upon several factors, this makes it very difficult to define reliable conditions. 418 Nevertheless, the values found are much lower than the values estimated by Chamas et 419 al. (2020) for the degradation of petroleum-derived plastics in marine environments. 420 Indeed, they reported that half-lives of high-density polyethylene, low-density 421 polyethylene and polypropylene varied from few decades to more than 2,500 years.

4. Assessment of potential bioplastics' leakage into the environment

423

424

425

426

427

428

429

430

431

432

433

434

435

436

437

438

439

440

441

442

443

444

445

446

Plastic pollution of natural environments has been thoroughly studied in the last decades with particular emphasis on microplastics accumulation and their toxic effects (He et al., 2018, Jung et al., 2021). In the last few years, concerns emerged regarding the partial degradation of bioplastics in natural and industrial environments, as well as the fate in the environment of bioplastics residues coming from waste management (Abraham et al., 2021, Emadian et al., 2017, Folino et al., 2020). The major concern regarded the possible leakage of bioplastics from the technosphere to the biosphere, both following accidental and voluntary pathways (e.g. littering and agronomic use of compost or digestate rich in bioplastics residues). The possible accumulation of bioplastics and their smallest fragments, as well as their eco-toxicological effects were recently reviewed by Shruti and Kutralam-Muniasamy (2019). They described the generation of microplastics from biobased biodegradable bioplastics (e.g. PHBs) and reported that several studies have identified that some biodegradable microplastics showed the same effects as those of petroleum-derived microplastics (e.g. transfer of chemical contaminants, increased stress in benthic communities) (Green, 2016, Hartmann et al., 2017). Nevertheless, the same authors identified different knowledge gaps in biodegradable microplastics effects, i.e. understanding the timeframe of disintegration and degradation of bioplastics and ensuring biodegradability and less persistence (Shruti and Kutralam-Muniasamy, 2019). Recently, Degli Innocenti and Breton (2020) defined bioplastics as intrinsically biodegradable materials due to their tendency to biodegrade similarly to natural polymers under different industrial and environmental conditions. The present paper confirms that intrinsic biodegradability can be ascribed to bioplastics since they were shown to be degradable in all the studied environments, even though with different

kinetics (Tables 2, 3, 4 and 5). In the contexts described, intrinsic biodegradability of bioplastics makes them always more sustainable than petroleum-derived plastics from an environmental point of view. Accumulation of bioplastics and their fragments in soil and aquatic environments after leakage appears to be unlikely in high amounts, because of the dynamic equilibrium that becomes established between bioplastics' leakage and biodegradation in the environment that, as discussed, requires years. This is in contrast with what happens with petroleum-derived non-degradable plastics that tend to accumulate in natural environments due to their long degradation time (centuries) (Chamas et al., 2020). Based on the data reviewed in the present paper, an attempt to estimate the potential bioplastics' leakage into the environment was carried out. Indeed, defining the potential bioplastics' leakage can be useful since any time a plastic or bioplastic item enters the biosphere from the technosphere, it can accumulate and, depending on their potential hazard for living organisms (Barbale et al., 2021), the result can be toxic. The potential bioplastic accumulation in the environment depends mainly on the concentration of the bioplastic leakage and on its residence time (biodegradability in the environment). This can be described by the following linear proportionality previously used by Degli Innocenti and Breton (2020) to describe the ecological risk of bioplastic, then adapted to describe plastic/bioplastics' leakage into the environment: $Potential\ leakage = Residence\ time\ x\ Concentration\ (Equation\ 1)$

447

448

449

450

451

452

453

454

455

456

457

458

459

460

461

462

463

464

465

466

467

468

469

470

Potential leakage = Residence time x Concentration (Equation 1)

It appears evident that the potential leakage derived from plastics and bioplastics can be reduced by decreasing one or both of the factors of Equation 1.

Residence time factor depends mainly on the type of materials and environment. As reviewed in this work, traditional plastics are not biodegradable in industrial and natural

environments and the first factor of Equation 1 can almost be assumed to be infinite. This makes it clear why potential leakage of plastics is always higher than the potential leakage of bioplastics, which are intrinsically biodegradable in years. Therefore, the only way to reduce petroleum-derived plastics' leakage is to reduce the concentration factor, i.e. by substituting plastic items with bioplastic ones and increasing the separate collection and recycling of petroleum-derived plastics. As reviewed in sections 4.1 and 4.2, residence time of bioplastics in natural environments (soil, water) varies according to the type of bioplastics and the environmental conditions, but it can be assumed to range between 1 and 10 years. Assuming 1,000 years residence time for petroleumderived plastics, it is possible to estimate that bioplastics reduce the potential leakage with respect to traditional plastics by a 1/100 - 1/1000 factor (considering the same concentration of material). In this context, waste management can play a role in reducing the potential leakage of bioplastics since it can enhance bioplastics' degradation under industrial controlled conditions, decreasing the *concentration* factor in Equation 1. The potential role of waste management in decreasing the potential bioplastics' leakage following their usage and disposal was assessed by simulating different scenarios described in Table 6. The other assumptions made for the potential leakage assessment were that bioplastics that were not treated in waste management directly leaked into natural environment, bioplastics residues in digestate and compost reached the natural environment after waste management, and that all bioplastics divided equally between soil and aquatic environments. The AD process was assumed to have 25 days HRT, whereas 90 days treatment were considered for thermophilic composting (Cerda et al., 2018, Panigrahi and Dubey, 2019). For the integrated AD and composting process, the timing used in

471

472

473

474

475

476

477

478

479

480

481

482

483

484

485

486

487

488

489

490

491

492

493

the assessment were 25 days HRT of AD and 65 days of thermophilic composting of the digestate. Based on these considerations and on the degradation rates reviewed in this paper (Tables 2, 3, 4 and 5), the potential leakage derived by the disposal of 100 kg of bioplastics was calculated following Equation 2:

495

496

497

498

499

500

501

502

503

504

505

506

507

508

509

510

511

512

513

514

515

516

517

518

Potential leakage = $(C \times RT)_{soil} + (C \times RT)_{aquatic\ env.}$ (Equation 2) where C is the concentration of bioplastic and RT is the residence time of bioplastic. The potential leakage of each scenario was estimated in a range from 0 (minimum leakage) to 1 (maximum leakage, absence of waste management) by dividing the result for the potential leakage calculated for the worst scenario (Scenario 0, absence of waste management and leakage of all the bioplastics into the environment). The results of potential leakage assessment for the three bioplastics considered in the present review (PHAs, PLA and starch-based blends) are reported in Table 6. PHAs blends showed a high reduction of potential leakage as a consequence of waste management and the leakage was reduced to zero in two scenarios (100 % bioplastics treated through AD and digestate composting). Interestingly, no differences were observed between mesophilic and thermophilic ADs, as expected from these kinds of bioplastics that were easily biodegradable in anaerobic conditions. PLA blends showed a small reduction of potential leakage after AD processes due to the slow degradation of these materials under anaerobic conditions. Conversely, PLA blends showed the highest benefits from composting and the potential leakage was reduced to 0.5 and 0 when 50 % and 100 % (weight basis) of PLA blends were managed through thermophilic composting. The integration of AD and digestate composting also led to strong reduction of the potential leakage associated with PLA blends (90 % and 100 % reduction for mesophilic and thermophilic AD coupled to composting, respectively).

The key role of temperature in degradation of PLA blends was confirmed in the present potential leakage assessment, and it appeared evident that a stage of thermophilic composting is mandatory to degrade PLA bioplastics quickly. Waste management also reduced the potential leakage associated with starch-based blends, even if not one of the studied scenarios allowed reducing the leakage to zero. The best results were obtained from scenarios 6 (100 % bioplastics in composting), scenario 8 (100 % bioplastics in mesophilic AD + composting) and scenario 10 (100 % bioplastics in thermophilic AD + composting). Summarising, the proposed potential leakage assessment highlighted the potential role of waste management in reducing the concentration of bioplastics that can reach the environment. Indeed, the best results were obtained when 100 % (weight basis) of the bioplastics were supposed to be treated in waste management. Separate collection of bioplastics within OFMSW appears as the most suitable solution to avoid leakage of bioplastics into the environment and to reduce their concentration in soil and water. The integration of AD and digestate composting appeared as the most promising treatment to degrade bioplastics, and this was an interesting result since coupled AD and composting allows recovering energy and nutrients from organic waste making the whole process sustainable from an energetic point of view (Ma et al., 2018).

537

538

539

540

541

542

536

519

520

521

522

523

524

525

526

527

528

529

530

531

532

533

534

535

5. Future challenges

The present review highlighted how waste management can reduce the presence of bioplastics in the environment, minimizing the potential leakage of these materials, because of their biodegradation under controlled conditions. Therefore, research dealing with bioplastics biodegradation during waste management with particular attention on

how to enhance bioplastics degradation under AD and/or composting conditions is becoming necessary and mandatory in the near future. Reaching a temperature higher than 55 °C is mandatory to improve the degradation kinetic of most of the bioplastics commonly used (e.g. PLA and starch-based bioplastics) due to the chemical and mechanical transformations that occur after reaching the glass transition temperatures. Nowadays, mesophilic AD is more widespread than thermophilic AD for OFMSW treatment, mainly due to higher stability and lower investments and energy requirements (Kumar and Samadder, 2020). Moving to thermophilic AD facilities may improve bioplastics degradation (Cazaudehore et al., 2021). On the other hand, mesophilic AD can be considered if it is coupled with digestate composting, since the thermophilic conditions of the active phase of composting can enhance bioplastics degradation. Nevertheless, sometimes composting after mesophilic AD is not capable of achieving high process temperatures because the major part of the readily degradable organic matter has been degraded during the AD (Tambone et al., 2015). In this case, AD-HRT plays an important role in determining residual organic matter to be degraded during composting. Other strategies to enhance bioplastics degradation under anaerobic conditions are the material pre-treatments before biological processes. Actually, this opportunity is still almost unexplored and the few papers dealing with this topic have reported controversial results. Acidic and basic chemical pre-treatments of starch-based and PLA bioplastics carried out at room temperature did not affect bioplastics degradation under AD (Battista et al., 2021). Conversely, Calabro' et al. (2020) reported that basic pretreatment of starch-based bags using NaOH 5 % w/v for 24 h, increased by 344 % and 283 % the degradation under mesophilic and thermophilic AD, respectively. In the same

543

544

545

546

547

548

549

550

551

552

553

554

555

556

557

558

559

560

561

562

563

564

565

paper, mechanical pre-treatment of starch-based bags did not produce significant results. These data agreed with those of Benn and Zitomer (2018) who reported that basic and thermal pre-treatment of PHBs and PLA increased the bioplastics conversion to biogas up to over 100 %. Since these are the first reported results of bioplastics pre-treatments enhancing degradation under anaerobic conditions, the need for more investigation has emerged. In fact, improving the degradation of bioplastics in AD can act positively in reducing the potential leakage of bioplastics but can also affect positively the biomethane production of the digester. Bioplastics have the potential to increase significantly the biomethane potential of OFMSW, and it is reasonable to suppose that in 2030, because bioplastic content in the OFMSW could reach 10 % (w/w), the contribution of bioplastics to total biomethane production of OFMSW could reach about 40 %. Unmixed AD may represent an interesting treatment for bioplastics' disposal because in this particular case, the solid retention time (SRT) is much longer than HRT allowing for a significant biomethane recovery from bioplastics even under mesophilic conditions. Surely, this research field need to be further explored and better addressed. Another research field that needs to be further investigated in order to improve degradation of bioplastics under industrial and natural environments is the use of bioplastics composites blended with easily biodegradable materials. The present paper already reported some examples of how the presence of soluble sugars, proteins and/or cellulose in the bioplastics structure may promote their degradation under composting, soil and aquatic environments (Beltrán-Sanahuja et al., 2020, Harmaen et al., 2015, Sarasa et al., 2009). In this context, future research should consider the findings indicated above on enhancing bioplastic biodegradability (Kalita et al., 2020). Plant

567

568

569

570

571

572

573

574

575

576

577

578

579

580

581

582

583

584

585

586

587

588

residues and organic wastes may represent low-cost suitable sources of easily biodegradable molecules to be blended with bioplastics (Ncube et al., 2020). Recycling bioplastics to recover valuable monomers is actually an unexplored field. Only PLA has been recently studied to recover lactic acid or lactate esters through chemical recycling (hydrolysis and alcoholysis processes, respectively), showing the need for more research on this topic (Lamberti et al., 2020). Conversely, the recovery of functional chemicals (e.g. sugars, volatile fatty acids, biofuels) from petroleum-derived plastics has also been investigated in recent years (Al Rayaan, 2021, Bäckström et al., 2017). Although bioplastics were introduced as eco-friendly substitutes for single-use plastic items and they were always intended to be collected within the bio-waste, the possible separate collection of bioplastics should be encouraged in a scenario of increased use of these products. Separate collection and subsequent conversion of bioplastics into valuable products through chemical processes would be possible in this way. Bioplastics management may move from the traditional degradation paradigm to a new one named bioplastics as feedstock, where bioplastics are no longer intended as single-use products but are projected to be recovered in a circular economy perspective. In a future perspective of separate collection of bioplastics' wastes, incineration coupled to energy recovery and bioreactor landfill with biomethane recovery could also be considered for the disposal of these materials. Although a preliminary study by Piemonte (2011) reported incineration to be lower performing than mechanical recycling, the waste-to energy strategy for bioplastics' waste might deserve more attention in future research due to energy recovery and the reduction of leakage's risk. The increasing use of bioplastics and their disposal within the OFMSW also challenges the existing regulations concerning compost quality. Actual regulations do not

590

591

592

593

594

595

596

597

598

599

600

601

602

603

604

605

606

607

608

609

610

611

612

discriminate between petroleum-derived plastics and bioplastics and, consequently, it is reasonable to expect an increase in the quantity of composts that do not meet quality requirements, thereby increasing the costs and environmental impacts of OFMSW management. For instance, Italian legislation concerning high quality compost production prescribes a 0.5 % w/w limit for the presence of *inert materials* (e.g. plastics, metals and glass particles with particle size > 2 mm), without any distinction between petroleum-derived plastics and bioplastics (Decreto Legislativo 29 Aprile 2010, 2010). The same limits were proposed by the European Commission as end-ofwaste criteria for biodegradable waste subjected to biological treatments (i.e. compost and digestate) (Saveyn and Eder, 2014). Bioplastics residues in compost should not be taken to count as *inert materials* (plastics, metals, glass) since they have been proved to degrade in natural environments (e.g. soil, water) (Emadian et al., 2017, Calabro' et al., 2020). Conversely, they might be considered together with the other biodegradable polymers constituting compost (e.g. cellulose, hemicellulose, lignin etc.) and thus regulations might be adapted to the new framework by excluding bioplastics from the fraction of *inert materials*. Nevertheless, the development of a standardized procedure to distinguish bioplastics and plastics residues in compost is needed before adapting the regulations dealing with compost quality. In summary, although more research to enhance bioplastic degradation under engineered environments is needed, the environmental benefits of bioplastics in comparison with petroleum-derived plastics are unequivocal because of their complete biodegradation in reasonable time.

636

637

614

615

616

617

618

619

620

621

622

623

624

625

626

627

628

629

630

631

632

633

634

635

6. Conclusions

The present review paper summarizes the current knowledge on bioplastics degradation under different environments (industrial and natural) and highlights the potential relationship between degradation of bioplastics during waste management and their potential leakage. Specifically, enhancing bioplastics degradation during anaerobic digestion and composting may reduce the concentration of bioplastics leaking to the soil and water environments, minimizing the potential environmental impact of these materials. Further investigation is needed to improve biodegradation of bioplastics in waste management through different strategies (e.g. thermophilic processes, pretreatments for anaerobic digestion, introduction of easily degradable blends).

647

648

638

639

640

641

642

643

644

645

646

Funding

- This work was supported by Gruppo Ricicla labs., University of Milan (UNIMI), Italy,
- project N. 14135 RV_PRO_RIC16FADAN02_M.

651

652

References

- 653 1. Abdelmoez, W., Dahab, I., Ragab, E. M., Abdelsalam, O. A., Mustafa, A., 2021.
- Bio-and oxo-degradable plastics: Insights on facts and challenges. Polym. Adv.
- 655 Technol. 32(5), 1981-1996.https://doi.org/10.1002/pat.5253
- 656 2. Abraham, A., Park, H., Choi, O., Sang, B.I., 2021. Anaerobic co-digestion of bioplastics
- as a sustainable mode of waste management with improved energy production -A
- review. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2020.124537
- 659 3. Accinelli, C., Saccà, M.L., Mencarelli, M., Vicari, A., 2012. Deterioration of bioplastic
- carrier bags in the environment and assessment of a new recycling alternative.
- 661 Chemosphere 89. https://doi.org/10.1016/j.chemosphere.2012.05.028

- 662 4. Ahmed, T., Shahid, M., Azeem, F., Rasul, I., Shah, A.A., Noman, M., Hameed, A.,
- Manzoor, N., Manzoor, I., Muhammad, S., 2018. Biodegradation of plastics: current
- scenario and future prospects for environmental safety. Environ. Sci. Pollut. Res.
- 665 https://doi.org/10.1007/s11356-018-1234-9
- 666 5. Ahn, H.K., Huda, M.S., Smith, M.C., Mulbry, W., Schmidt, W.F., Reeves, J.B., 2011.
- Biodegradability of injection molded bioplastic pots containing polylactic acid and
- poultry feather fiber. Bioresour. Technol. 102.
- 669 https://doi.org/10.1016/j.biortech.2011.01.042
- 670 6. Al Rayaan, M.B., 2021. Recent advancements of thermochemical conversion of plastic
- waste to biofuel-A review. Clean. Eng. Technol. 2.
- 672 https://doi.org/10.1016/j.clet.2021.100062
- 673 7. Amin, M.R., Chowdhury, M.A., Kowser, M.A., 2019. Characterization and
- 674 performance analysis of composite bioplastics synthesized using titanium dioxide
- 675 nanoparticles with corn starch. Heliyon 5. https://doi.org/10.1016/j.heliyon.2019.e02009
- 676 8. Anstey, A., Muniyasamy, S., Reddy, M.M., Misra, M., Mohanty, A., 2014.
- 677 Processability and Biodegradability Evaluation of Composites from Poly(butylene
- succinate) (PBS) Bioplastic and Biofuel Co-products from Ontario. J. Polym. Environ.
- 679 22. https://doi.org/10.1007/s10924-013-0633-8
- 680 9. Arikan, E. B., Ozsoy, H. D., 2015. A review: investigation of bioplastics. J. Civ. Eng.
- 681 Arch, 9, 188-192. https://doi: 10.17265/1934-7359/2015.02.007
- 682 10. Ayilara, M.S., Olanrewaju, O.S., Babalola, O.O., Odeyemi, O., 2020. Waste
- management through composting: Challenges and potentials. Sustain.
- 684 https://doi.org/10.3390/su12114456

- 685 11. Bäckström, E., Odelius, K., Hakkarainen, M., 2017. Trash to Treasure: Microwave-
- Assisted Conversion of Polyethylene to Functional Chemicals. Ind. Eng. Chem. Res.
- 56. https://doi.org/10.1021/acs.iecr.7b04091
- 688 12. Bagheri, A.R., Laforsch, C., Greiner, A., Agarwal, S., 2017. Fate of So-Called
- Biodegradable Polymers in Seawater and Freshwater. Glob. Challenges 1.
- 690 https://doi.org/10.1002/gch2.201700048
- 691 13. Barbale, M., Chinaglia, S., Gazzilli, A., Pischedda, A., Pognani, M., Tosin, M., Degli-
- Innocenti, F., 2021. Hazard profiling of compostable shopping bags. Towards an
- 693 ecological risk assessment of littering. Polym. Degrad. Stab. 188.
- 694 https://doi.org/10.1016/j.polymdegradstab.2021.109592
- 695 14. Báreková, A., Demovičová, M., Tátošová, L., Danišová, L., Medlenová, E.,
- Hlaváčiková, S., 2021. Decomposition of single-use products made of bioplastic under
- real conditions of urban composting facility. J. Ecol. Eng. 22(4), 265-272.
- 698 https://doi.org/10.12911/22998993/134040
- 699 15. Bátori, V., Åkesson, D., Zamani, A., Taherzadeh, M.J., Sárvári Horváth, I., 2018.
- Anaerobic degradation of bioplastics: A review. Waste Manag. 80.
- 701 https://doi.org/10.1016/j.wasman.2018.09.040
- 702 16. Battista, F., Frison, N., Bolzonella, D., 2021. Can bioplastics be treated in
- conventional anaerobic digesters for food waste treatment? Environ. Technol. Innov.
- 704 22. https://doi.org/10.1016/j.eti.2021.101393
- 705 17. Bellasi, A., Binda, G., Pozzi, A., Galafassi, S., Volta, P., Bettinetti, R., 2020.
- Microplastic contamination in freshwater environments: A review, focusing on
- interactions with sediments and benthic organisms. Environ. MDPI.

- 708 https://doi.org/10.3390/environments7040030
- 709 18. Beltrán-Sanahuja, A., Casado-Coy, N., Simó-Cabrera, L., Sanz-Lázaro, C., 2020.
- Monitoring polymer degradation under different conditions in the marine environment.
- 711 Environ. Pollut. 259. https://doi.org/10.1016/j.envpol.2019.113836
- 712 19. Benn, N., Zitomer, D., 2018. Pretreatment and anaerobic co-digestion of selected PHB
- and PLA bioplastics. Front. Environ. Sci. 5. https://doi.org/10.3389/fenvs.2017.00093
- 714 20. Bhagwat, G., Gray, K., Wilson, S.P., Muniyasamy, S., Vincent, S.G.T., Bush, R.,
- Palanisami, T., 2020. Benchmarking Bioplastics: A Natural Step Towards a
- Sustainable Future. J. Polym. Environ. https://doi.org/10.1007/s10924-020-01830-8
- 717 21. Briassoulis, D., Mistriotis, A., Mortier, N., Tosin, M., 2020. A horizontal test method
- for biodegradation in soil of bio-based and conventional plastics and lubricants. J.
- 719 Clean. Prod. 242. https://doi.org/10.1016/j.jclepro.2019.118392
- 720 22. Cafiero, L. M., Canditelli, M., Musmeci, F., Sagnotti, G., Tuffi, R., 2021. Assessment
- of disintegration of compostable bioplastic bags by management of electromechanical
- and static home composters. Sustainability 13(1), 263.
- 723 https://doi.org/10.3390/su13010263
- 724 23. Calabro', P.S., Folino, A., Fazzino, F., Komilis, D., 2020. Preliminary evaluation of
- the anaerobic biodegradability of three biobased materials used for the production of
- disposable plastics. J. Hazard. Mater. 390.
- 727 https://doi.org/10.1016/j.jhazmat.2019.121653
- 728 24. Calabrò, P.S., Grosso, M., 2018. Bioplastics and waste management. Waste Manag.
- 729 https://doi.org/10.1016/j.wasman.2018.06.054

- 730 25. Castro, L., Escalante, H., Jaimes-Estévez, J., Díaz, L.J., Vecino, K., Rojas, G.,
- Mantilla, L., 2017. Low cost digester monitoring under realistic conditions: Rural use
- of biogas and digestate quality. Bioresour. Technol. 239.
- 733 https://doi.org/10.1016/j.biortech.2017.05.035
- 734 26. Cazaudehore, G., Monlau, F., Gassie, C., Lallement, A., Guyoneaud, R., 2021.
- 735 Methane production and active microbial communities during anaerobic digestion of
- three commercial biodegradable coffee capsules under mesophilic and thermophilic
- conditions. Sci. Total Environ. 784. https://doi.org/10.1016/j.scitotenv.2021.146972
- 738 27. Cerda, A., Artola, A., Font, X., Barrena, R., Gea, T., Sánchez, A., 2018. Composting
- of food wastes: Status and challenges. Bioresour. Technol.
- 740 https://doi.org/10.1016/j.biortech.2017.06.133
- 741 28. Chamas, A., Moon, H., Zheng, J., Qiu, Y., Tabassum, T., Jang, J.H., Abu-Omar, M.,
- Scott, S.L., Suh, S., 2020. Degradation Rates of Plastics in the Environment. ACS
- Sustain. Chem. Eng. 8. https://doi.org/10.1021/acssuschemeng.9b06635
- 744 29. Chinaglia, S., Tosin, M., Degli-Innocenti, F., 2018. Biodegradation rate of
- biodegradable plastics at molecular level. Polym. Degrad. Stab. 147.
- 746 https://doi.org/10.1016/j.polymdegradstab.2017.12.011
- 747 30. Cucina, M., Tacconi, C., Sordi, S., Pezzolla, D., Gigliotti, G., Zadra, C., 2018.
- 748 Valorization of a pharmaceutical organic sludge through different composting
- 749 treatments. Waste Manag. 74. https://doi.org/10.1016/j.wasman.2017.12.017
- 750 31. Decreto Legislativo 29 Aprile 2010, n. 75, 2010. Riordino e revisione della disciplina
- in materia di fertilizzanti, a norma dell'articolo 13 della Legge 7 Luglio 2009 n. 88.
- 752 Gazz. Uff. n. 121 Suppl. Ordin. n.106, Roma.

- 753 32. Degli Innocenti, F., Breton, T., 2020. Intrinsic Biodegradability of Plastics and
- Ecological Risk in the Case of Leakage. ACS Sustain. Chem. Eng.
- 755 https://doi.org/10.1021/acssuschemeng.0c01230
- 756 33. Dilkes-Hoffman, L., Ashworth, P., Laycock, B., Pratt, S., Lant, P., 2019. Public
- attitudes towards bioplastics knowledge, perception and end-of-life management.
- 758 Resour. Conserv. Recycl. 151. https://doi.org/10.1016/j.resconrec.2019.104479
- 759 34. Dubois, Sims, Moerman, Watson, Bauer, Bel, Mehlhart, 2020. Guidance for separate
- 760 collection of municipal waste.
- 761 35. Edwards, J., Othman, M., Crossin, E., Burn, S., 2018. Life cycle assessment to
- compare the environmental impact of seven contemporary food waste management
- systems. Bioresour. Technol. 248. https://doi.org/10.1016/j.biortech.2017.06.070
- 764 36. Elsawy, M.A., Kim, K.H., Park, J.W., Deep, A., 2017. Hydrolytic degradation of
- polylactic acid (PLA) and its composites. Renew. Sustain. Energy Rev.
- 766 https://doi.org/10.1016/j.rser.2017.05.143
- 767 37. Emadian, S.M., Onay, T.T., Demirel, B., 2017. Biodegradation of bioplastics in
- natural environments. Waste Manag. https://doi.org/10.1016/j.wasman.2016.10.006
- 769 38. European bioplastic, 2020. Market update 2020: Bioplastics continue to become
- mainstream as the global bioplastics market is set to grow by 36 percent over the next
- 5 years [WWW Document]. https://www.european-bioplastics.org/market-update-
- 772 2020-bioplastics-continue-to-become-mainstream-as-the-global-bioplastics-market-is-
- set-to-grow-by-36-percent-over-the-next-5-years/.
- 774 39. European Commission, 2018. Report from the commission to the european parliament

- and the council on the impact of the use of oxo-degradable plastic, including oxo-
- degradable plastic carrier bags, on the environment.
- 777 40. European Parliament, 2019. Directive (EU) 2019/904 of the European Parliament and
- of the Council of 5 June 2019 on the reduction of the impact of certain plastic products
- on the environment.
- 780 41. Folino, A., Karageorgiou, A., Calabrò, P.S., Komilis, D., 2020. Biodegradation of
- wasted bioplastics in natural and industrial environments: A review. Sustain.
- 782 https://doi.org/10.3390/su12156030
- 783 42. Fonseca, L.M., Domingues, J.P., Dima, A.M., 2020. Mapping the sustainable
- development goals relationships. Sustain. 12. https://doi.org/10.3390/SU12083359
- 785 43. Ganesh Saratale, R., Cho, S.K., Dattatraya Saratale, G., Kadam, A.A., Ghodake, G.S.,
- Kumar, M., Naresh Bharagava, R., Kumar, G., Su Kim, D., Mulla, S.I., Seung Shin,
- H., 2021. A comprehensive overview and recent advances on polyhydroxyalkanoates
- 788 (PHA) production using various organic waste streams. Bioresour. Technol.
- 789 https://doi.org/10.1016/j.biortech.2021.124685
- 790 44. Gil-Castell, O., Andres-Puche, R., Dominguez, E., Verdejo, E., Monreal, L., Ribes-
- Greus, A., 2020. Influence of substrate and temperature on the biodegradation of
- 792 polyester-based materials: Polylactide and poly(3-hydroxybutyrate-co-3-
- hydroxyhexanoate) as model cases. Polym. Degrad. Stab. 180.
- 794 https://doi.org/10.1016/j.polymdegradstab.2020.109288
- 795 45. Gómez, E.F., Michel, F.C., 2013. Biodegradability of conventional and bio-based
- plastics and natural fiber composites during composting, anaerobic digestion and long-
- 797 term soil incubation. Polym. Degrad. Stab. 98.

- 798 https://doi.org/10.1016/j.polymdegradstab.2013.09.018
- 799 46. Green, D.S., 2016. Effects of microplastics on European flat oysters, Ostrea edulis and
- their associated benthic communities. Environ. Pollut. 216.
- 801 https://doi.org/10.1016/j.envpol.2016.05.043
- 802 47. Guidoni, L.L.C., Marques, R.V., Moncks, R.B., Botelho, F.T., da Paz, M.F., Corrêa,
- L.B., Corrêa, É.K., 2018. Home composting using different ratios of bulking agent to
- food waste. J. Environ. Manage. 207. https://doi.org/10.1016/j.jenvman.2017.11.031
- 805 48. Hamad, K., Kaseem, M., Yang, H.W., Deri, F., Ko, Y.G., 2015. Properties and
- medical applications of polylactic acid: A review. Express Polym. Lett. 9.
- https://doi.org/10.3144/expresspolymlett.2015.42
- 808 49. Harmaen, A.S., Khalina, A., Azowa, I., Hassan, M.A., Tarmian, A., Jawaid, M., 2015.
- Thermal and biodegradation properties of poly(lactic acid)/fertilizer/oil palm fibers
- blends biocomposites. Polym. Compos. 36. https://doi.org/10.1002/pc.22974
- 811 50. Harrison, J.P., Boardman, C., O'Callaghan, K., Delort, A.M., Song, J., 2018.
- Biodegradability standards for carrier bags and plastic films in aquatic environments:
- A critical review. R. Soc. Open Sci. https://doi.org/10.1098/rsos.171792
- 814 51. Hartmann, N.B., Rist, S., Bodin, J., Jensen, L.H.S., Schmidt, S.N., Mayer, P.,
- Meibom, A., Baun, A., 2017. Microplastics as vectors for environmental
- contaminants: Exploring sorption, desorption, and transfer to biota. Integr. Environ.
- 817 Assess. Manag. https://doi.org/10.1002/ieam.1904
- 818 52. He, D., Luo, Y., Lu, S., Liu, M., Song, Y., Lei, L., 2018. Microplastics in soils:
- Analytical methods, pollution characteristics and ecological risks. TrAC Trends

- Anal. Chem. https://doi.org/10.1016/j.trac.2018.10.006
- 821 53. Hupfauf, S., Plattner, P., Wagner, A.O., Kaufmann, R., Insam, H., Podmirseg, S.M.,
- 2018. Temperature shapes the microbiota in anaerobic digestion and drives efficiency
- to a maximum at 45 °C. Bioresour. Technol. 269.
- 824 https://doi.org/10.1016/j.biortech.2018.08.106
- 825 54. ISO 14855, 2018. ISO 14855. Determ. Ultim. Aerob. Biodegrad. Plast. Mater. under
- 826 Control. Compost. Cond. Method by Anal. Evolved Carbon Dioxide Part 1 Gen.
- 827 Method.
- 828 55. ISO 13975, 2019. ISO 13975. Plastics Determination of the ultimate anaerobic
- biodegradation of plastic materials in controlled slurry digestion systems Method
- by measurement of biogas production.
- 831 56. ISO 14851, 2019. ISO 14851. Determination of the ultimate aerobic biodegradability
- of plastic materials in an aqueous medium Method by measuring the oxygen
- 833 demand in a closed respirometer.
- 834 57. ISO 17556, 2019. ISO 17556. Plastics Determination of the ultimate aerobic
- biodegradability of plastic materials in soil by measuring the oxygen demand in a
- respirometer or the amount of carbon dioxide evolved.
- 837 58. ISPRA, 2020. Rapporto rifiuti urbani (edizione 2020). ISPRA Area Comun.
- 838 59. Jung, J.W., Park, J.W., Eo, S., Choi, J., Song, Y.K., Cho, Y., Hong, S.H., Shim, W.J.,
- 2021. Ecological risk assessment of microplastics in coastal, shelf, and deep sea
- waters with a consideration of environmentally relevant size and shape. Environ.
- Pollut. 270. https://doi.org/10.1016/j.envpol.2020.116217

- 842 60. Kalita, N. K., Bhasney, S. M., Kalamdhad, A., Katiyar, V., 2020. Biodegradable
- kinetics and behavior of bio-based polyblends under simulated aerobic composting
- conditions. J. Environ. Manage. 261, 110211.
- https://doi.org/10.1016/j.jenvman.2020.110211
- 846 61. Kalita, N.K., Hazarika, D., Kalamdhad, A., Katiyar, V., 2021. Biodegradation of
- biopolymeric composites and blends under different environmental conditions:
- Approach towards end-of-life panacea for crop sustainability. Bioresour. Technol.
- Reports 15. https://doi.org/10.1016/j.biteb.2021.100705
- 850 62. Kumar, A., Samadder, S.R., 2020. Performance evaluation of anaerobic digestion
- technology for energy recovery from organic fraction of municipal solid waste: A
- review. Energy. https://doi.org/10.1016/j.energy.2020.117253
- 853 63. Lamberti, F. M., Román-Ramírez, L. A., Wood, J., 2020. Recycling of bioplastics:
- routes and benefits. J. Polym. Environ. 1-21. https://doi.org/10.1007/s10924-020-
- 855 01795-8
- 856 64. Lin, L., Xu, F., Ge, X., Li, Y., 2018. Improving the sustainability of organic waste
- management practices in the food-energy-water nexus: A comparative review of
- anaerobic digestion and composting. Renew. Sustain. Energy Rev.
- 859 https://doi.org/10.1016/j.rser.2018.03.025
- 860 65. Liu, T., Zhou, X., Li, Z., Wang, X., Sun, J., 2019. Effects of liquid digestate
- pretreatment on biogas production for anaerobic digestion of wheat straw. Bioresour.
- Technol. 280. https://doi.org/10.1016/j.biortech.2019.01.147
- 863 66. Ma, H., Guo, Y., Qin, Y., Li, Y.Y., 2018. Nutrient recovery technologies integrated
- with energy recovery by waste biomass anaerobic digestion. Bioresour. Technol.

- 865 https://doi.org/10.1016/j.biortech.2018.08.114
- 866 67. Magnier, L., Crié, D., 2015. Communicating packaging eco-friendliness: An
- exploration of consumers' perceptions of eco-designed packaging. Int. J. Retail
- 868 Distrib. Manag. 43. https://doi.org/10.1108/IJRDM-04-2014-0048
- 869 68. Marek, A.A., Verney, V., 2016. Photochemical reactivity of PLA at the vicinity of
- glass transition temperature. the photo-rheology method. Eur. Polym. J. 81.
- 871 https://doi.org/10.1016/j.eurpolymj.2016.06.016
- 872 69. Mendieta, C. M., Vallejos, M. E., Felissia, F. E., Chinga-Carrasco, G., Area, M. C.,
- 2020. Bio-polyethylene from wood wastes. J. Polym. Environ. 28(1), 1-16.
- 874 https://doi.org/10.1007/s10924-019-01582-0
- 875 70. Modelli, A., Calcagno, B., Scandola, M., 1999. Kinetics of aerobic polymer
- degradation in soil by means of the ASTM D 5988-96 standard method. J. Environ.
- Polym. Degrad. 7. https://doi.org/10.1023/A:1021864402395
- 878 71. Mohee, R., Unmar, G.D., Mudhoo, A., Khadoo, P., 2008. Biodegradability of
- biodegradable/degradable plastic materials under aerobic and anaerobic conditions.
- 880 Waste Manag. 28. https://doi.org/10.1016/j.wasman.2007.07.003
- 881 72. Ncube, L. K., Ude, A. U., Ogunmuyiwa, E. N., Zulkifli, R., Beas, I. N., 2020.
- 882 Environmental impact of food packaging materials: A review of contemporary
- development from conventional plastics to polylactic acid based materials. Materials,
- 884 13(21), 4994. https://doi.org/10.3390/ma13214994
- 885 73. Noda, I., Lindsey, S.B., Caraway, D., 2010. NodaxTM Class PHA Copolymers: Their
- Properties and Applications. https://doi.org/10.1007/978-3-642-03287-5 10

- 887 74. Pagga, U., 1998. Biodegradability and compostability of polymeric materials in the
- context of the European packaging regulation. Polym. Degrad. Stab. 59.
- https://doi.org/10.1016/s0141-3910(97)00192-4
- 890 75. Palsikowski, P.A., Kuchnier, C.N., Pinheiro, I.F., Morales, A.R., 2018. Biodegradation
- in Soil of PLA/PBAT Blends Compatibilized with Chain Extender. J. Polym. Environ.
- 892 26. https://doi.org/10.1007/s10924-017-0951-3
- 893 76. Panigrahi, S., Dubey, B.K., 2019. A critical review on operating parameters and
- strategies to improve the biogas yield from anaerobic digestion of organic fraction of
- municipal solid waste. Renew. Energy. https://doi.org/10.1016/j.renene.2019.05.040
- 896 77. Papa, G., Pepè Sciarria, T., Carrara, A., Scaglia, B., D'Imporzano, G., Adani, F., 2020.
- 897 Implementing polyhydroxyalkanoates production to anaerobic digestion of organic
- fraction of municipal solid waste to diversify products and increase total energy
- recovery. Bioresour. Technol. 318. https://doi.org/10.1016/j.biortech.2020.124270
- 900 78. Patel, S.K.S., Gupta, R.K., Kondaveeti, S., Otari, S. V., Kumar, A., Kalia, V.C., Lee,
- 901 J.K., 2020. Conversion of biogas to methanol by methanotrophs immobilized on
- chemically modified chitosan. Bioresour. Technol. 315.
- 903 https://doi.org/10.1016/j.biortech.2020.123791
- 904 79. Peng, W., Lü, F., Hao, L., Zhang, H., Shao, L., He, P., 2020. Digestate management
- for high-solid anaerobic digestion of organic wastes: A review. Bioresour. Technol.
- 906 https://doi.org/10.1016/j.biortech.2019.122485
- 907 80. Piemonte, V., 2011. Bioplastic wastes: the best final disposition for energy saving. J.
- 908 Polym. Environ. 19(4), 988-994. https://doi.org/10.1007/s10924-011-0343-z

- 909 81. Pilla, S., 2011. Handbook of Bioplastics and Biocomposites Engineering Applications,
- Handbook of Bioplastics and Biocomposites Engineering Applications.
- 911 https://doi.org/10.1002/9781118203699
- 912 82. PlasticsEurope, 2020. Plastics the Facts 2020. An analysis of European plastics
- 913 production, demand and waste data.
- 914 83. Ran, G., Li, D., Zheng, T., Liu, X., Chen, L., Cao, Q., Yan, Z., 2018. Hydrothermal
- pretreatment on the anaerobic digestion of washed vinegar residue. Bioresour.
- 916 Technol. 248. https://doi.org/10.1016/j.biortech.2017.06.068
- 917 84. Rana, K.I., 2019. Usage of Potential Micro-organisms for Degradation of Plastics.
- 918 Open J. Environ. Biol. https://doi.org/10.17352/ojeb.000010
- 919 85. Ren, Y., Yu, M., Wu, C., Wang, Q., Gao, M., Huang, Q., Liu, Y., 2018. A
- omprehensive review on food waste anaerobic digestion: Research updates and
- tendencies. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.09.109
- 922 86. Rudnik, E., Briassoulis, D., 2011. Degradation behaviour of poly(lactic acid) films and
- fibres in soil under Mediterranean field conditions and laboratory simulations testing.
- 924 Ind. Crops Prod. 33. https://doi.org/10.1016/j.indcrop.2010.12.031
- 925 87. Ruggero, F., Onderwater, R. C., Carretti, E., Roosa, S., Benali, S., Raquez, J. M., ...,
- Wattiez, R., 2021. Degradation of film and rigid bioplastics during the thermophilic
- phase and the maturation phase of simulated composting. J. Polym. Environ. 1-14.
- 928 https://doi.org/10.1007/s10924-021-02098-2
- 929 88. Sarasa, J., Gracia, J.M., Javierre, C., 2009. Study of the biodisintegration of a
- bioplastic material waste. Bioresour. Technol. 100.

- 931 https://doi.org/10.1016/j.biortech.2008.11.049
- 932 89. Saveyn, H., Eder, P., 2014. European Commission, Joint Research Centre Institute
- 933 for Prospective Technological Studies End-of-waste criteria for biodegradable waste
- subjected to biological treatment (compost & digestate): Technical proposals, Joint
- 935 Research Center Scintific and Policy Reports.
- 936 90. Sekiguchi, T., Saika, A., Nomura, K., Watanabe, Toshihiro, Watanabe, Toru,
- Fujimoto, Y., Enoki, M., Sato, T., Kato, C., Kanehiro, H., 2011. Biodegradation of
- aliphatic polyesters soaked in deep seawaters and isolation of poly(ε-caprolactone)-
- 939 degrading bacteria. Polym. Degrad. Stab. 96.
- 940 https://doi.org/10.1016/j.polymdegradstab.2011.03.004
- 941 91. Shrestha, A., van-Eerten Jansen, M.C.A.A., Acharya, B., 2020. Biodegradation of
- bioplastic using anaerobic digestion at retention time as per industrial biogas plant and
- international norms. Sustain. 12. https://doi.org/10.3390/su12104231
- 944 92. Shruti, V.C., Kutralam-Muniasamy, G., 2019. Bioplastics: Missing link in the era of
- 945 Microplastics. Sci. Total Environ. https://doi.org/10.1016/j.scitotenv.2019.134139
- 946 93. Siracusa, V., 2019. Microbial degradation of synthetic biopolymers waste. Polymers
- 947 (Basel). https://doi.org/10.3390/polym11061066
- 948 94. Siracusa, V., Rocculi, P., Romani, S., Rosa, M.D., 2008. Biodegradable polymers for
- food packaging: a review. Trends Food Sci. Technol.
- 950 https://doi.org/10.1016/j.tifs.2008.07.003
- 951 95. Tambone, F., Orzi, V., D'Imporzano, G., Adani, F., 2017. Solid and liquid
- 952 fractionation of digestate: Mass balance, chemical characterization, and agronomic

- and environmental value. Bioresour. Technol. 243.
- 954 https://doi.org/10.1016/j.biortech.2017.07.130
- 955 96. Tambone, F., Terruzzi, L., Scaglia, B., Adani, F., 2015. Composting of the solid
- 956 fraction of digestate derived from pig slurry: Biological processes and compost
- properties. Waste Manag. 35. https://doi.org/10.1016/j.wasman.2014.10.014
- 958 97. Taufik, D., Reinders, M.J., Molenveld, K., Onwezen, M.C., 2020. The paradox
- between the environmental appeal of bio-based plastic packaging for consumers and
- their disposal behaviour. Sci. Total Environ. 705.
- 961 https://doi.org/10.1016/j.scitotenv.2019.135820
- 962 98. Thakur, S., Chaudhary, J., Sharma, B., Verma, A., Tamulevicius, S., Thakur, V.K.,
- 2018. Sustainability of bioplastics: Opportunities and challenges. Curr. Opin. Green
- 964 Sustain. Chem. https://doi.org/10.1016/j.cogsc.2018.04.013
- 965 99. Thellen, C., Coyne, M., Froio, D., Auerbach, M., Wirsen, C., Ratto, J.A., 2008. A
- Processing, Characterization and Marine Biodegradation Study of Melt-Extruded
- Polyhydroxyalkanoate (PHA) Films. J. Polym. Environ. 16.
- 968 https://doi.org/10.1007/s10924-008-0079-6
- 969 100. Tosin, M., Weber, M., Siotto, M., Lott, C., Degli Innocenti, F., 2012. Laboratory Test
- 970 Methods to Determine the Degradation of Plastics in Marine Environmental
- Conditions. Front. Microbiol. 3. https://doi.org/10.3389/fmicb.2012.00225
- 972 101. Urbanek, A.K., Rymowicz, W., Mirończuk, A.M., 2018. Degradation of plastics and
- plastic-degrading bacteria in cold marine habitats. Appl. Microbiol. Biotechnol.
- 974 https://doi.org/10.1007/s00253-018-9195-y

- 975 102. Vázquez, M.A., Soto, M., 2017. The efficiency of home composting programmes and
- compost quality. Waste Manag. 64. https://doi.org/10.1016/j.wasman.2017.03.022
- 977 103. Villegas Calvo, M., Colombo, B., Corno, L., Eisele, G., Cosentino, C., Papa, G.,
- 978 Scaglia, B., Pilu, R., Simmons, B., Adani, F., 2018. Bioconversion of Giant Cane for
- Integrated Production of Biohydrogen, Carboxylic Acids, and Polyhydroxyalkanoates
- 980 (PHAs) in a Multistage Biorefinery Approach. ACS Sustain. Chem. Eng. 6.
- 981 https://doi.org/10.1021/acssuschemeng.8b03794
- 982 104. Volova, T.G., Gladyshev, M.I., Trusova, M.Y., Zhila, N.O., 2007. Degradation of
- polyhydroxyalkanoates in eutrophic reservoir. Polym. Degrad. Stab. 92.
- 984 https://doi.org/10.1016/j.polymdegradstab.2007.01.011
- 985 105. Wainaina, S., Awasthi, M.K., Sarsaiya, S., Chen, H., Singh, E., Kumar, A., Ravindran,
- 986 B., Awasthi, S.K., Liu, T., Duan, Y., Kumar, S., Zhang, Z., Taherzadeh, M.J., 2020.
- 987 Resource recovery and circular economy from organic solid waste using aerobic and
- anaerobic digestion technologies. Bioresour. Technol.
- 989 https://doi.org/10.1016/j.biortech.2020.122778
- 990 106. Wang, P., Wang, H., Qiu, Y., Ren, L., Jiang, B., 2018. Microbial characteristics in
- anaerobic digestion process of food waste for methane production—A review.
- Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.06.152
- 993 107. Wang, S., Zeng, Y., 2018. Ammonia emission mitigation in food waste composting: A
- review. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.07.050
- 995 108. Wei, L., Liang, S., McDonald, A.G., 2015. Thermophysical properties and
- biodegradation behavior of green composites made from polyhydroxybutyrate and
- potato peel waste fermentation residue. Ind. Crops Prod. 69.

- 998 https://doi.org/10.1016/j.indcrop.2015.02.011
- 999 109. Wu, B., Zhang, X., Shang, D., Bao, D., Zhang, S., Zheng, T., 2016. Energetic-
- environmental-economic assessment of the biogas system with three utilization
- pathways: Combined heat and power, biomethane and fuel cell. Bioresour. Technol.
- 1002 214. https://doi.org/10.1016/j.biortech.2016.05.026
- 1003 110. Yagi, H., Ninomiya, F., Funabashi, M., Kunioka, M., 2014. Mesophilic anaerobic
- biodegradation test and analysis of eubacteria and archaea involved in anaerobic
- biodegradation of four specified biodegradable polyesters. Polym. Degrad. Stab. 110.
- 1006 https://doi.org/10.1016/j.polymdegradstab.2014.08.031
- 1007 111. Yagi, H., Ninomiya, F., Funabashi, M., Kunioka, M., 2013. Thermophilic anaerobic
- biodegradation test and analysis of eubacteria involved in anaerobic biodegradation of
- four specified biodegradable polyesters. Polym. Degrad. Stab. 98.
- 1010 https://doi.org/10.1016/j.polymdegradstab.2013.03.010
- 1011 112. Zhang, J., Liang, Y., Yan, J., Lou, J., 2007. Study of the molecular weight dependence
- of glass transition temperature for amorphous poly(l-lactide) by molecular dynamics
- simulation. Polymer (Guildf). 48. https://doi.org/10.1016/j.polymer.2007.06.030
- 1014 113. Zhang, W., Heaven, S., Banks, C.J., 2018. Degradation of some EN13432 compliant
- plastics in simulated mesophilic anaerobic digestion of food waste. Polym. Degrad.
- 1016 Stab. 147. https://doi.org/10.1016/j.polymdegradstab.2017.11.005
- 1017 114. Zhao, X., Cornish, K., Vodovotz, Y., 2020. Narrowing the Gap for Bioplastic Use in
- 1018 Food Packaging: An Update. Environ. Sci. Technol.
- 1019 https://doi.org/10.1021/acs.est.9b03755

1020 115. Table 1. Main characteristics of selected bio-based biodegradable
1021 bioplastics and their global production in 2020 and 2025.

Bioplastic	Source	Main applications	Global production ^a (10 ³ tonnes year ⁻¹)		
			2020	Forecast 2025	
Starch-based blends	Starch from dedicated crops (e.g. corn, rice, potato) processed with plasticizers	Films, rigid materials (dishes, cutlery, glasses), packaging, medical products, mulching films, carrier bags, wastecollection bags	390	400	
PLA blends	Fermentation of carbohydrates from dedicated crops (e.g. corn, sugar cane, sugar beet, tapioca) to low MW ^b PLA and subsequent repolymerization to high MW PLA	Packaging, disposable goods, electronic applications, fibres, medical products, textiles and films, agricultural applications, building and constructions	390	560	
PHAs blends	Microbial synthesis and intracellular accumulation from carbohydrates (e.g. sugars, starch)	Packaging, agricultural applications, medical products	50	330	

118. **Table 2**. Degradation of bioplastics during anaerobic digestion and estimation of kinetic parameters.

1027 119.

1025

1026

Bioplastic	Temperature	Time (d)	Degradation (%)	k ^a (mg g ⁻	Time for complete degradation ^a (d)	Average time for complete degradation ± SD ^a (d)	Reference
		9	93	103.3	10		Yagi et al., 2014
	Mesophilic	42	86	20.4	49	31 ± 20	Ryan et al., 2017
PHAs blends		30	90	30	33		Reischwitz et al., 1997
	m 1:1:	14	90	64.3	16	26 . 20	Yagi et al., 2013
	Thermophilic	50	90	18	56	36 ± 28	Yagi et al., 2013
		60	12	2	500		Yagi et al., 2009
	Mesophilic	227	49	2.2	454	423 ± 76	Yagi et al., 2014
		65	20	3.1	322	423 ± 70	Zhang et al., 2018
		100	24	2.4	417		Cazaudehore et al., 2021
PLA blends		151	94	6.2	161		Tseng et al., 2019
bienus		60	90	15	67		Yagi et al., 2009
	Thermophilic	65	80	12.3	81	116 ± 48	Yagi et al., 2010
		75	75	10	100	110 ± 48	Yagi et al., 2013
		100	58	5.8	172		Cazaudehore et al., 2021
		65	18	2.8	357		Zhang et al., 2018
		50	26	5.2	192		Gómez and
	Mesophilic	30	24	8	125	376 ± 319	Michel, 2013 Calabro' et al.,
Starch- based	copiiiic	100	12	1.2	833	5.0 _ 517	2020 Cazaudehore et al., 2021
blends	m	30	37	12	83	140 00	Calabro' et al., 2020
	Thermophilic	100	47	4.7	213	148 ± 92	Cazaudehore et al., 2021

^{120. &}lt;sup>a</sup>Estimated in this work assuming a pseudo-zero order kinetic

1029 1030

Table 3. Degradation of bioplastics during composting and estimation of kinetic parameters.

121. 1031

Bioplastic	Temperature (°C)	Time (d)	Degradation (%)	k ^a (mg g ⁻¹ d ⁻¹)	Time for complete degradation ^a (d)	Average time for complete degradation \pm SD ^a (d)	Reference
	> 50	60	30	5	200		Sun et al., 2021
blends	58	110	80	7.3	137	124 ± 83	Weng et al., 201
	55	28	80	28.6	35		Tabasi and Ajji, 20
	n.a.	98	90	9.2	109		Kalita et al., 202
	58	130	75	5.8	172		Balaguer et al., 20
	65	58	84	14.5	69		Kale et al., 2007
	55	28	70	25	40	0.4 4=	Tabasi and Ajji, 20
blends	58	30	60	20	50	84 ± 47	Mihai et al., 201
	58	28	100	35.7	28		Arrieta et al., 20
	58	90	80	8.9	112		Sarasa et al., 200
	62	90	100	11.1	90		Báreková et al., 20
_	58	90	85	9.4	106		Javierre et al., 20
	55	85	50	5.9	169		Gómez and Michel,
	45 - 65	60	90	15	67	119 ± 43 (thermophilic)	Cafiero et al., 202
h-based blends	58	60	45	7.5	133		Ruggero et al., 20
	23	72	27	3.8	263		Mohee et al., 200
	25	90	43	4.8	208	220 ± 38 (mesophilic)	Accinelli et al., 20
	25 - 45	180	95	5.3	189		Cafiero et al., 202

122. Estimated in this work assuming a pseudo-zero order kinetic

1032 1033 123.bNot available

1034 124.

125. Table 4. Degradation of bioplastics in soil and estimation of kinetic 1036 parameters. 1037

126. 1038

Bioplastic	Temperature (°C)	Time (d)	Degradation (%)	$\begin{array}{c} k^{a} \\ (mg \ g^{\text{-}1} \ d^{\text{-}1}) \end{array}$	Time for complete degradation ^a (d)	Average time for complete degradation \pm SD ^a (d)	Reference
	n.a. ^b	180	64.3	3.6	278		Jain and Tiwari
	Room temperature	60	35	5.8	172		Wu, 2014
ends	20	280	48.5	1.7	588	435 ± 241	Gómez and Mich
	Natural conditions	365	98	2.7	370		Boyandin et al.
	Natural conditions	365	47	1.3	769		Boyandin et al.
	23	180	20	1.1	909		Kalita et al., 2
	20	98	10	1.0	1,000		Wu, 2012
ıds	25	180	16	0.9	1,111	$1,604 \pm 1,010$	Palsikowski et al
	n.a.	600	20	0.3	3,333		Urayama et al.,
	25	90	5	0.6	1,667		Unpublished
	20	110	14.2	1.3	769		Gómez and Mich
	25	90	37	4.1	244		Accinelli et al.,
ased blends	20	40	5	1.3	769	591 ± 313	Alvarez et al.,
	30	70	8	1.1	909		Rapisarda et al.
	25	90	34.3	3.8	263		Unpublished

127. ^aEstimated in this work assuming a pseudo-zero order kinetic 128. ^bNot available 129.

1039 1040 1041

1042 130.

1044 1045 131. **Table 5**. Degradation of bioplastics in aquatic environment and estimation of kinetic parameters.

1046

132.

Environment	Temperature (°C)		Degradation (%)	$\begin{array}{c} k^{a} \\ (mg \ g^{\text{-}1} \ d^{\text{-}1}) \end{array}$	Time for complete degradation ^a (d)	Average time for complete degradation \pm SD ^a (d)	Refer
Freshwater	25	365	8.5	0.23	4,348		Bagheri et
Sea water	25	365	8.5	0.23	4,348		Bagheri et
Freshwater	20	42	43.5	10.4	96		Volova et
Sea water	29	160	58	3.60	278	$1,570 \pm 2,154$	Volova et
Sea water	21	49	99	20.2	50		Thellen et
Sea water	12-22	90	30	3.30	303		Thellen et
Sea water Sea water	30 30	365 365	5.7 8.4	0.16 0.23	6,250 4,348		Greene. Greene
Freshwater	25	365	< 2	-	-	4 (20) 2 4(0	Bagheri et
Sea water	25	365	< 2	-	-	$4,629 \pm 2,468$	Bagheri et
Sea water	16	365	29.2	0.80	1,250		Beltrán-Sanahu
Sea water	16	365	5.6	0.15	6,667		Beltrán-Sanahu
Freshwater	25	90	1.5	0.17	5,882		Accinelli et
Sea water	25	90	1.5	0.17	5,882		Accinelli et
Sea water + Sediment	Room temperature	236	69	2.92	342	$3,068 \pm 3,249$	Tosin et a
Sea water	n.a. ^b	168	100	5.95	168		O'Brine and Th

1047

133. Estimated in this work assuming a pseudo-zero order kinetic

1048 134. Not available

1049 135.

136. **Table 6**. Ecological risk assessment for 100 kg of bioplastics under different waste management scenarios.

1053 137.

10511052

	PHAs blends				PLA blends				Starch-based blends			
Scena rio ^a	Degrad ation in WM ^b (%)	B P° so il (k g)	BP aquatic environ ment (kg)	Ri sk	Degrad ation in WM (%)	B P so il (k g)	BP aquatic environ ment (kg)	Ri sk	Degrad ation in WM (%)	B P so il (k g)	BP aquatic environ ment (kg)	Ri sk
0	-	50	50	1.0 0	-	50	50	1.0 0	-	50	50	1.0 0
1	75	31 .2	31.2	0.6 3	6	48 .5	48.5	0.9 7	7	48 .2	48.2	0.9 7
2	75	31 .2	31.2	0.2 5	6	44 .8	44.8	0.9 4	7	46 .5	46.5	0.9 3
3	75	12 .5	12.5	0.6 3	21	47	47	0.9 0	17	45 .8	45.8	0.9 2
4	75	12 .5	12.5	0.2 5	21	39 .5	39.5	0.7 9	17	41 .5	41.5	0.8 3
5	75	31 .2	31.2	0.6 3	100	25	25	0.5 0	65	33 .8	33.8	0.6 8
6	75	12 .5	12.5	0.2 5	100	0	0	0.0	65	17 .5	17.5	0.3 5
7	100	25	25	0.5 0	91	27 .2	27.2	0.5 5	62	34 .5	34.5	0.6 9
8	100	0	0	0.0	91	4. 5	4.5	0.0 9	62	19	19	0.3 8
9	100	25	25	0.5	100	25	25	0.5	72	32	32	0.6 4
10	100	0	0	0.0	100	0	0	0.0	72	14	14	0.2 8

138. aScenario description:

139. Scenario 0: no waste management foreseen. All the bioplastics leak into natural environments

140. Scenario 1: 50 % (weight basis) of bioplastics treated by AD under mesophilic conditions

141. Scenario 2: 100 % (weight basis) of bioplastics treated by AD under mesophilic conditions

142. Scenario 3: 50 % (weight basis) of bioplastics treated by AD under thermophilic conditions

143. Scenario 4: 100 % (weight basis) of bioplastics treated by AD under thermophilic conditions

144. Scenario 5: 50 % (weight basis) of bioplastics treated by thermophilic composting

145. Scenario 6: 100 % (weight basis) of bioplastics treated by thermophilic composting

146.Scenario 7: 50 % (weight basis) of bioplastics treated by mesophilic AD and digestate thermophilic composting

147. Scenario 8: 100 % (weight basis) of bioplastics treated by mesophilic AD and digestate thermophilic composting

148. Scenario 9: 50 % (weight basis) of bioplastics treated by thermophilic AD and digestate thermophilic composting

149. Scenario 10: 100 % (weight basis) of bioplastics treated by thermophilic AD and digestate thermophilic composting

150. bWaste management

151.°Bioplastic

50

1054

1055

1056

1057

1058

1059

1060

1061

1062

1063

1064

1065

1066

1067

1068

1069

1070

1071