



Review

Biodegradation of oil-based plastics in the environment: Existing knowledge and needs of research and innovation

Noura Raddadi *, Fabio Fava

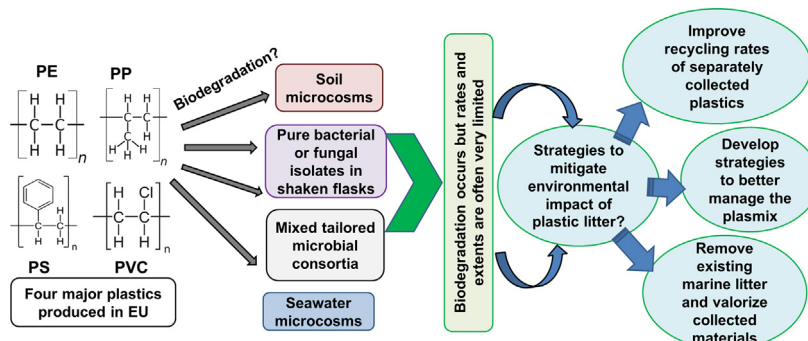
Department of Civil, Chemical, Environmental and Materials Engineering; Alma Mater Studiorum-University of Bologna, Italy



HIGHLIGHTS

- PS; PP; PVC and PEs biodegradation in terrestrial/aquatic/insect larval gut systems is reviewed.
- Microbial degradation by mixed communities and pure strains occurs under lab and field conditions.
- Biodegradation rates/extents are often very limited, very few data available on marine habitats.
- Microplastics biodegradation can very slightly contribute to reducing *in situ* marine litter.
- Need for strategies to prevent/remove existing marine litter and to valorize collected materials.

GRAPHICAL ABSTRACT



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ABSTRACT

The production of synthetic oil-based plastics has led to the accumulation of huge amounts of the plastic waste in the environment, especially in the marine system, very often the final sink for many types of conventional wasted plastics. In particular, (micro)plastics account for the majority of litter items in the marine environment and a high percentage of such litter is originating from land sources. Attempts to mitigate the harmful effects of conventional plastics such as the development of novel management strategies together with the gradual substitution of them with biodegradable (bio)plastics are representing future solutions. However, high amounts of conventional plastics have been accumulating in the environment since several years. Although many studies reported on their potential biodegradation by microbes in and from terrestrial environments, very little is known about the biodegradability of these plastics in freshwater systems and only recently more reports on their biodegradation by marine microorganisms/in marine environment were made available.

In this review, we first provide a summary of the approaches applied for monitoring and assessing conventional plastics biodegradation under defined conditions. Then, we reviewed historical and recent findings related to biodegradation of four major plastics produced in European Union (EU), i.e. Polyethylene, Polyvinyl Chloride, Polypropylene and Polystyrene, in terrestrial and aquatic environments and by pure and mixed microbial cultures obtained from them.

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* Corresponding author at: Department of Civil, Chemical, Environmental and Materials Engineering (DICAM), Alma Mater Studiorum-Università di Bologna, via U. Terracini 28, 40131 Bologna, Italy.

E-mail address: noura.raddadi@unibo.it (N. Raddadi).

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1. Introduction

The worldwide plastic production was about 350 million tons in 2017. The EU contributed for 64.4 million tons to such a global production (PlasticsEurope, 2018). Owing to their excellent mechanical properties, low cost, light weight, stability, durability and chemical inertness, these plastics are extensively used in several sectors and hence their production increased dramatically during the years. Considering their very low biodegradability in the environment, there have been public concerns about a potentially huge environmental accumulation and pollution problem that could persist for centuries. Currently, although different plastic waste management strategies have been developed, still a relatively large fraction of post-consumer plastic waste, including the residual mixture of plastics arising from the mechanical selection/treatment process of separately collected plastics and commonly defined as PLASMIX in Italy, is disposed of in landfills (Cossu et al., 2017; PlasticsEurope, 2018). Besides of the post-consumer plastic waste, also a large fraction of plastic/polymer byproducts generated in several production plants (and among which only a small amount can be reprocessed) is disposed of in landfill. The plastics disposed of in landfill are very often subjected to photooxidation leading to the production of small fragments of less than 5 mm in diameter, *i.e.* microplastics, which can reach the aquatic eco-systems. Besides landfill, microplastics in the marine environment can also derive from the fragmentation of large plastic fragments and many other land sources including tyre and road wear particles, laundry, household dust and personal care products (Li et al., 2016; Siegfried et al., 2017). In the marine environment, microplastics can absorb and concentrate persistent organic pollutants and together with plasticizers/additives enter the food chains once ingested by marine wildlife, potentially exerting toxic effects (Tanaka et al., 2013; Baztan et al., 2014; O'Connor et al., 2016; Hermabessiere et al., 2017). Indeed, microplastics occurrence/accumulation in the aquatic systems is raising health issues besides the environmental ones (Eriksen et al., 2014; Eerkes-Medrano et al., 2015; Suaria et al., 2016; The Lancet Planetary Health, 2017; Hermabessiere et al., 2017; Carbery et al., 2018). Marine litter is a major societal

challenge for many basins given its remarkable environmental, economic, social, political and cultural implications. It is adversely affecting the healthy status and the productivity of the sea and related activities of the area. Furthermore, microplastics accumulation is also a threat to terrestrial ecosystems (de Souza Machado et al., 2018). Hence, development of innovative plastic wastes management strategies as well as the progressive switch towards the production and use of the biodegradable/compostable (bio)plastics are being considered as potential strategies to mitigate the future impacts of plastic pollution. However, petroleum-derived synthetic plastics have been produced and accumulating in the environment during several decades, and there are still knowledge gaps about their rates of biodegradation in terrestrial and in particular in aquatic systems. Indeed, although different review papers reporting on the biodegradation of plastics have been published recently, they dealt either mainly with only one type of synthetic polymer/plastic (Ho et al., 2018), with one environmental type (Urbanek et al., 2018) or with bio-based plastics (Emadian et al., 2017; Ahmed et al., 2018) without providing a complete overview on all major recent findings and needs in the subject. Moreover, the most updated wide review on the microbial biodegradation of the main fossil-based plastics dates to 2019 (Moharir and Kumar, 2019) but the paper summarized the literature reports up to 2017 and did not include those recently published. Finally, there are a few and fragmented evidences about biodegradation of oil based plastics in aquatic systems and by microbes coming from such environments and such evidences merit to be reviewed because those are the habitats where recalcitrant plastics are accumulating (Suaria et al., 2016; Sighicelli et al., 2018).

The scope of the present review is to provide an overview i) on the analytical procedures to monitor synthetic polymers biodegradation and the challenges for a reliable assessment of the biodegradative capability of microbial catalysts, and ii) on all major recent findings related to microbial degradation of four key oil-based synthetic polymers and plastics produced in EU namely Polyethylenes (PEs), Polyvinyl Chloride (PVC), Polypropylene (PP) and Polystyrene (PS) in terrestrial habitats and in marine systems.

2. Analytical procedure for the evaluation of oil-based polymer/plastic (bio)degradation and their limitations

Different techniques can be applied in combination in order to quantify the biodegradation activities towards oil based synthetic polymeric materials. They include both microbiological as well as physical-chemical analyses. These techniques can be summarized below.

2.1. Gravimetric determination of weight loss

Evaluation of the gravimetric weight loss is a widely used method in polymer degradation assays. This method does not constitute a direct proof of the degradation and especially in the case of plastics where high quantities of additives are used in the formulations, such as for example PVC based plastics, for which a weight loss can be due to the release of additives or their soluble components. Moreover, weight losses should be interpreted with caution as they can be also due to chemical hydrolysis and fragmentation/disintegration of plastics, in particular in the case of weathered polymeric materials or to a loss of the material when powder polymer is used as substrate, leading to an overestimation of the weight loss percentage. Finally, in any case, the weight loss is often very low (due to very slow and limited biodegradation extents) and the data might be affected by a high inaccuracy. Hence, the gravimetric technique should always be associated with other techniques in order to assess and characterize putative biodegradation activities.

2.2. Thermogravimetric analysis (TGA)

TGA measures the thermal stability of a polymer. A decrease in such stability is an indicator of polymer degradation. A particular attention should be kept if the plastic subjected to TGA contains high amount of additives in its formulation, since also the additive components such as heat stabilizers participate to the final thermal stability of the plastic. This for example is the case of PVC plastic film, which can have up to 35% (w/w) of additive components. In this case, biodegradation of the polymer chains could be considered if the plastic film subjected to biodegradation assay exhibits a lower thermal stability compared to its corresponding abiotic control film (Giacomucci et al., 2019a; Giacomucci et al., 2019b unpublished).

2.3. Differential scanning calorimetric (DSC) analysis

DSC is a useful technique for the assessment of different thermal properties of materials, and to study the thermal transition of synthetic polymers, such as glass transition (T_g). The shifting of T_g to lower temperatures is often related to a decrease in the stability of the polymer as a result of its degradation (Lucas et al., 2008).

2.4. Gel permeation chromatography (GPC)

GPC provides inputs on the number average molecular weight (Mn %) and the molecular weight distribution of the polymer. A decrease in the Mn of the polymer is a proof of chain scission and, in turn, of microbial attack (Ali et al., 2014; Yang et al., 2015; Tian et al., 2017; Giacomucci et al., 2019a). The drawbacks of this technique is that it is not highly sensitive since the analysis is performed on the bulk polymer and may not allow the detection of biodegradation of the polymer in the initial steps of the process since such biodegradation occurs firstly on the surface of the polymer. Hence, this technique could be useful when combined with other techniques for the evaluation of synthetic plastics biodegradation.

2.5. Fourier transform infrared spectroscopy (FTIR)

FTIR is frequently used to reveal chemical modifications of the polymer structure, and for monitoring chemical changes in polymeric film. It

allows the detection of the formation of functional groups as a result of microbial attack (Lucas et al., 2008). However, this technique is not highly informative if the plastic film formulation has a high amount of additives; which makes very complicated the identification of the functional groups formed due to microbial attack to the polymer chains. Furthermore, complete removal of the biofilm developed on the plastic surface is mandatory to avoid misleading in the interpretation/identification of the functional groups i.e. in order to be sure that the observed functional groups correspond to the structural changes of the polymer and not to the cell debris.

2.6. Microscopy observations of the surface

The evaluation of polymer biodegradation is based on the evaluation of the changes in the physical properties like for example the formation of cracks and holes or the formation of biofilm, which reflects the microbial colonization (Esmaeili et al., 2013; Peixoto et al., 2017; Mohanrasu et al., 2018). Stereomicroscopy observations could be considered in a first step and can be followed by the use of higher magnification microscopes like Scanning Electron Microscopy (SEM) observations. Using SEM observations, it is possible to detect the microbial colonization of the polymer/plastic surface. The colonization of the polymer surface cannot be a sufficient proof of the biodegradation ability of the microorganism since the polymer surface could be used by the microbe as a support for biofilm formation. Hence, further analyses need to be performed to confirm the biodegradation activity.

2.7. Radiolabeling

This technique consists in labelling the carbon in the polymer to be used as substrate for microbial growth with carbon isotope ^{14}C . The mineralization is then confirmed by measuring the radioactive gas produced ($^{14}\text{CO}_2$, $^{14}\text{CH}_4$) (Federle et al., 2002; Tian et al., 2017). Although this technique is not destructive and highly precise for the evaluation of polymer biodegradation, its application is limited by the difficulties and cost of preparing the radioactive polymer as well as by the necessity of specific measures for management and disposal of the radiolabeled samples.

2.8. Standard methods

Different standard tests have been developed by different organizations such as, among others, ISO (International Organization for Standardization), CEN (European Committee for Standardization) and ASTM (American Society for Testing and Materials) for the evaluation of the biodegradation of plastics/polymers under aerobic or anaerobic conditions (Ho et al., 2018 and references therein). These tests are based mainly on the measurement of gas produced after a specific incubation period. However, in the case of petroleum-derived plastics where a very little amount of gas could be released, this method could not be reliable for its very low sensitivity and thus it should be applied in conjunction with others. Moreover, the production of gas could be also associated with the degradation of other compounds present in the matrix in which the test is performed (compost, soil etc...). Finally, no robust standard methods for the evaluation of plastic biodegradability in aquatic systems have been developed so far (Harrison et al., 2018).

2.9. Other recently reported analytical techniques

In addition to the above mentioned analytical techniques commonly used for evaluating conventional plastics/polymers biodegradation, other techniques have been reported (also as preliminary studies) and were applied in few cases/or for biodegradable polymers but not yet optimized/standardized for regular use to monitor biodegradation of fossil-based conventional plastics/polymers. These include for example: i) Reflectometric interference spectroscopy (RIfS), a useful technique for

the evaluation of variation in the physical thickness of a biodegradable polymer and which has been used for monitoring enzymatic biodegradation of thin polycaprolactone (PCL) polymer film but not yet applied in the case of conventional plastics/polymers (Ooya et al., 2016) and ii) Elemental analyser/isotope ratio mass spectrometry (EA/IRMS), a technique based on the estimation of carbon stable isotopes ($\delta^{13}\text{C}$) which could reflect (bio)degradation of plastic material as revealed by an increase of $\delta^{13}\text{C}$ values (Berto et al., 2017).

3. Biodegradation of synthetic plastics under different environmental conditions and microorganisms involved in the processes

Petroleum-deriving synthetic polymers and plastics can be divided into two main groups in relation to their potential biodegradability. The first includes those biodegradable in soils, such as aliphatic polyesters, polycaprolactone, polyethylene succinate, polyurethane and acrylate (Shah et al., 2014; Ghosh et al., 2013; Ahmed et al., 2018); while the second comprises those known to be highly recalcitrant to biodegradation including PE, PP, PS and PVC. The knowledge about biodegradation of the latter group of plastics under terrestrial and aquatic environmental conditions as well as by particular microbes isolated from insect larval gut is reviewed here.

3.1. Terrestrial environments

3.1.1. Polyethylene biodegradation

The majority of studies on the biodegradation of synthetic polymers and plastics by terrestrial deriving pure and mixed aerobic cultures have been performed on PE films (Roy et al., 2011; Restrepo-Flórez et al., 2014) (Table 1). LLDPE has been reported to be the more susceptible to microbial attack. Its biodegradation has been suggested to proceed through the release of oxidases that cleave the PE chains to low MW compounds that are transported within the cell and subjected to mineralization through alkane biodegradation pathway (Santo et al., 2013; Gravouil et al., 2017). PE was shown to be subjected to biodegradation by bacteria such as the actinomycete *Rhodococcus ruber* (Sivan et al., 2006). Peixoto et al. (2017) isolated, from plastic debris found in soil, untreated LDPE (191.000 g/mol)-degrading bacteria from the genera *Comamonas*, *Delftia*, and *Stenotrophomonas* that showed metabolic activity and cellular viability after a 90-day incubation with PE as the sole carbon source. The biodegradation was confirmed by FTIR analysis that revealed the formation of functional groups, by microscopic observations that showed surface roughness and presence of deep cavities, by phase imaging which revealed a 46.7% decrease in the viscous area of biodegraded PE, and by Raman spectroscopy, which confirmed a loss in crystalline content of the biodegraded PE. Tribedi and Dey (2017) reported on gravimetric weight loss of 6% and 3.5% for UV-treated and untreated LDPE films incubated for 28 days in soil microcosms. Muhonja et al. (2018) reported on different bacterial (genera *Pseudomonas*, *Bacillus*, *Brevibacillus*, *Cellulosimicrobium*, *Lysinibacillus*) and fungal (genus *Aspergillus*) isolates from dumpsite that showed the ability to degrade LDPE films after 16 weeks incubation under aerobic conditions. Specifically, gravimetric weight reductions of up to $36.4 \pm 5.53\%$ and $35.72 \pm 4.01\%$ were recorded for *Aspergillus* sp. and *Bacillus* sp. isolates, respectively. Furthermore, the authors reported that FTIR analyses performed on the incubated films revealed the detection of aldehyde, carboxyl and ether functional groups. The authors did not specify, however, whether additives or other copolymers were present in the film composition.

Skariyachan et al. (2016, 2017) reported on very high weight loss percentages reaching up to $81 \pm 4\%$ and up to $75 \pm 2\%$ of LDPE strips after an incubation period of 120 days with tailored consortia composed of bacterial isolates obtained from plastic garbage processing areas (Skariyachan et al., 2016) or plastic-contaminated cow dung (Skariyachan et al., 2017), respectively. In the same studies, a very high degradation of LDPE pellet, HDPE pellet and strips was also reported (Table 1). However, the authors did not specify if the plastic

strips have additives among their components and did not mention if they observed plastics fragmentation (which could result in a loss of the material) and no GPC analysis was performed to ascertain a molecular weight decrease of the polymer, which is the most solid proof for a polymer (bio)degradation.

Gajendiran et al. (2016) reported on the biodegradation of LDPE by the fungus *Aspergillus clavatus* isolated from landfill soil. LDPE biodegradation was monitored for 90 days and confirmed based on 35% gravimetric weight loss, 2.32 g l⁻¹ CO₂ production based on Strum test as well as the surface changes observed by FTIR and microscopy analyses. *Lysinibacillus xylanilyticus* and *Aspergillus niger* isolated from landfill soils were tested in a study by Esmaeili et al. (2013) for their ability to degrade LDPE (UV-irradiated and non irradiated). After an incubation period of 126 days in soil, biodegradation percentages of 29.5% and 15.8% for the UV-irradiated and non-UV-irradiated films, respectively, were recorded in the presence of a co-culture of the selected microorganisms according to ASTM D5988–03 standard method. Moreover, SEM observations showed the formation of pits and cavities on the surface of the film while FTIR and XRD showed structural and morphological changes. In another study, Skariyachan et al. (2018) reported on high degradation capability of thermophilic bacterial consortia of *Brevibacillus* spp. and *Aneurinibacillus* sp. screened from waste management landfills and sewage treatment plants towards LDPE and HDPE pellets and strips. Recently, biodegradation of PE microplastic granules by mixed bacterial cultures composed of *Bacillus* sp. and *Paenibacillus* sp. isolates obtained from soil samples collected from a decommissioned landfill site in Korea has been reported (Park and Kim, 2019). After 60 days incubation, reductions of the gravimetric weight (14.7% versus 5% for abiotic control) and mean diameter (22.8%) of PE particles were recorded.

Oxidized PE wax (obtained from PE with an average Mw ~4000 and an average Mn ~1700) was used in a study by Radecka et al. (2016) as carbon source for growth and polyhydroxyalkanoates (PHAs) production by *Ralstonia eutropha* which could be an interesting strategy for the management and the valorization of PE waste into biopolymers.

3.1.2. Polypropylene biodegradation

Until now, a little is known about the biodegradation of PP polymers and plastics. The first attempts on the evaluation of microbial biodegradation of PP have been performed by Cacciari et al. (1993), who reported that microbial communities were able to degrade isotactic PP. After 175 days incubation in the presence of the bacterial consortia obtained from different soil samples that were rich in plastic wastes, the polymer had 40% methylene chloride extractable compounds and a mixture of hydrocarbons (between C₁₀H₂₂ and C₃₁H₆₄) were detected and identified in the extract (Cacciari et al., 1993). Few other reports dealing with the biodegradation of pretreated and/or pro-oxidant additives-containing PPs have also been published. Biodegradation of thermally pretreated 0.05 mm thick PP films by a soil microbial consortium after one year incubation under aerobic conditions has been reported; in that case a 10.7% gravimetric weight loss was observed for the thermally pretreated PP compared to only 0.4% for the untreated polymer (Arkatkar et al., 2009). Arkatkar et al. (2010), studied the biodegradation of unpretreated (PPUT), chemically (Aquaregia pretreated or Fenton's pretreated) and physically pretreated (thermal and UV pretreated) PP films with four bacterial species, namely *Pseudomonas azotoformans* MTCC 7616 and three isolates enriched from one soil sample recovered from a plastic dumping site (identified as *Pseudomonas stutzeri*, *Bacillus subtilis* and *Bacillus flexus*). After one year of monitoring, although the three strains were able to form biofilm on the surface of all polymeric films, biodegradation was mainly detected on the sole UV-pretreated PP inoculated with *B. flexus* where a 2.5% weight loss of the polymeric film was recorded. UV pretreated pro-oxidant blended-PP incubated for 1 year with *Phanerochaete chrysosporium* NCIM 1170 and *Engyodontium album* MTP091 fungal strains in shake flasks showed 18.8% and 9.42% gravimetric weight losses, respectively (Jeyakumar

Table 1

Main reports on biodegradation of PEs, PP, PS and PVC under terrestrial environments and by isolates obtained from them or from the gut of some insect larvae.

Plastic	Type of microorganism	Biodegradation assay conditions	Biodegradability method	Reference
PE films	<i>Rhodococcus ruber</i> strain C208	8 weeks incubation in shaken flasks under aerobic conditions	Gravimetric weight reduction of 7.5%	Sivan et al. (2006)
UV-irradiated and non-UV-irradiated LDPE films	Coculture of <i>Lysinibacillus xylanilyticus</i> and <i>Aspergillus niger</i>	Incubation for 126 days in soil	Biodegradation percentages of 29.5% (UV-irradiated) and 15.8% (non-UV-irradiated) films in the presence of the coculture according to ASTM D5988–03 standard method. Formation of pits and cavities on the surface of the film detected by SEM observations while FTIR and XRD showed structural and morphological changes of the films surfaces.	Esmaeili et al. (2013)
UV-treated and UV-untreated LDPE films	Soil microcosms	Incubation for 28 days	Gravimetric weight loss of 6% (UV-treated) and 3.5% (UV-untreated) LDPE films.	Tribedi and Dey (2017)
LDPE film and pellet	2 <i>Enterobacter</i> sp. and 1 <i>Pantoea</i> sp. obtained from plastic garbage processing areas used individually or as an artificial consortium.	Incubation for 120 days at 37 °C with artificial consortia	Gravimetric weight loss percentages of 70 ± 4, 68 ± 4, and 64 ± 4% weight reduction for LDPE films and 21 ± 2, 28 ± 2, 24 ± 2% weight reduction for LDPE pellets incubated in the presence of individual isolates	Skariyachan et al. (2016)
LDPE and HDPE film and pellet	Artificial thermophilic bacterial consortium composed of bacterial isolates (<i>Bacillus vallismortis</i> , <i>Pseudomonas protegens</i> , <i>Stenotrophomonas</i> sp. and <i>Paenibacillus</i> sp.) obtained from dung of cows fed off plastic-contaminated spots	Incubation for 120 days at 55 °C	Gravimetric weight loss percentages of 75 ± 2, 55 ± 2, 60 ± 3, and 43 ± 3% for LDPE film, pellets, HDPE film and pellets, respectively	Skariyachan et al. (2017)
Unpretreated LDPE films	<i>Comamonas</i> , <i>Delftia</i> , and <i>Stenotrophomonas</i> isolated from degraded plastic debris	Shaken flasks incubated for 90 days at 28 °C.	Metabolic activity and cellular viability until the end of the experiment; formation of functional groups revealed by FTIR; deep cavities on the film surface detected by microscopy, 46.7% decrease in the viscous area and loss in crystalline content of the biodegraded PE	Peixoto et al. (2017)
PE films	Bacterial (<i>Pseudomonas</i> , <i>Bacillus</i> , <i>Brevibacillus</i> , <i>Cellulosimicrobium</i> , <i>Lysinibacillus</i>) and fungal (genus <i>Aspergillus</i>) strains from Dandora dumpsite Nairobi-Kenya.	16 weeks incubation in shaken flasks at 37 °C and 28 °C	Gravimetric weight reductions of up to 36.4 ± 5.53% and 35.72 ± 4.01% recorded for <i>Aspergillus</i> sp. and <i>Bacillus</i> sp. isolates, respectively. Detection of aldehyde, carboxyl and ether functional groups by FTIR.	Muhonja et al. (2018)
LDPE films (bags)	<i>Aspergillus clavatus</i> strain JASK1 isolated from landfill soil.	90 days incubation in shaken flasks	Gravimetric weight reductions of 35%, 2.32 g l ⁻¹ CO ₂ production based on Strum test and surface changes observed by FTIR and microscopy	Gajendiran et al. (2016)
LLDPE film	<i>Enterobacter asburiae</i> YT1 and <i>Bacillus</i> sp. YP1 isolated from the guts of plastic-eating waxworms.	60-day incubation period in shaken flasks at 30 °C under aerobic conditions	Biofilm formation, gravimetric weight losses of 6.1 ± 0.3% and 10.7 ± 0.2% after incubation with <i>E. asburiae</i> YT1 and <i>Bacillus</i> sp. YP1, respectively. Molecular weights reduction ~6–13% of the residual PE films, and release of 12 water-soluble daughter products detected.	Yang et al. (2014)
HDPE, LDPE, PP films and pellets	Thermophilic artificial bacterial consortia constructed based on <i>Brevibacillus</i> sps. And <i>Aneurinibacillus</i> sp. isolates obtained from waste management landfills and sewage treatment plants	140 days incubation in test tubes at 50 °C under aerobic conditions	Biofilm formation; fatty-acids end products identified by GC–MS; gravimetric weight reduction for LDPE, HDPE and PP strips were 58.21 ± 2, 46.6 ± 3 and 56.3 ± 2% respectively; those for LDPE, HDPE and PP pellets weight reductions were 45.7 ± 3, 37.2 ± 3 and 44.2 ± 3% respectively	Skariyachan et al. (2018)
PP powder	<i>Stenotrophomonas panacihumi</i> PA3–2 isolated from a soil sample recovered from an open storage yard for municipal solid waste	90 days incubation under composting conditions at 37 °C	KS M3100–1:2002; MOD ISO 14855:1999 method and GPC analysis	Jeon and Kim (2016)
UV or thermally-pretreated PP films	Co-culture of <i>Bacillus</i> and <i>Pseudomonas</i>	Flasks incubated at 28 ± 2 °C and 180 rpm for 12 months	Contact angle measurements revealed hydrophilicity of the polymers; biofilm formation of plastic surface; 27.1% weight reduction revealed by TGA, and gravimetric weight loss percentages of up to 1.95 ± 0.18% recorded	Aravinthan et al. (2016)
PE microplastic granules	mixed bacterial cultures composed of <i>Bacillus</i> sp. and <i>Paenibacillus</i> sp. isolates obtained from soil samples collected from a decommissioned landfill site	60 days incubation in shaken flasks at 30 °C under aerobic conditions	Reductions of the gravimetric weight (14.7% versus 5% for abiotic control) and mean diameter (22.8%) of PE particles were recorded.	Park and Kim (2019)
Pretreated (100 °C or UV for 10 days) and blended	<i>Phanerochaete chrysosporium</i> NCIM 1170; <i>Engyodontium album</i> MTP091	12 months incubation in shaken flasks	Gravimetric weight losses of 18.8% and 9.42%, with <i>P. chrysosporium</i> and <i>E. album</i> ,	Jeyakumar et al. (2013)

Table 1 (continued)

Plastic	Type of microorganism	Biodegradation assay conditions	Biodegradability method	Reference
(metal or starch) PP. Pro-oxidant (Mn, Mn/Fe or Co) additives-containing PP film subjected to artificial photooxidation and thermooxidation	<i>Rhodococcus rhodochrous</i> ATCC 29672	180-day incubation period	respectively Biodegradability of PP films containing Mn/Fe or Mn additive that were able to support bacterial growth evaluated by ATP measurements	Fontanella et al. (2013)
Commercial additive-containing unpretreated (PP-UT) and thermally pretreated (PP-TT) PP films	Soil consortium from a plastic dumping site	<i>In vitro</i> biodegradation, one year incubation under aerobic conditions	Gravimetric weight loss of 10.7% observed for the thermally pretreated PP compared to only 0.4% for the untreated film	Arkatkar et al., 2009
UV-pretreated PP film	<i>Bacillus flexus</i> isolated from a soil consortium enriched from plastic dumping site	One year incubation period	2.5% gravimetric weight loss	Arkatkar et al. (2010)
Isotactic PP films	Mixed soil community obtained from different soil samples that were rich in plastic wastes	5 months incubation, anaerobic conditions	The film had 40% methylene chloride extractable compounds and a mixture of hydrocarbons (between C ₁₀ H ₂₂ and C ₃₁ H ₆₄) were detected and identified in the extract	Cacciari et al. (1993)
¹⁴ C-PS polymer films with molecular weight of 15 and 29 kDa, subjected to ozonation pre-treatment	<i>Penicillium variable</i> CCF3219	16 weeks incubation in liquid medium (pH 7.5, without additional carbon substrate)	Mineralization of labelled polymers detected (0.16 ± 0.03% and 0.04 ± 0.01% for the low and high MW PS, respectively.; decrease of molecular weight of the polymer detected by GPC).	Tian et al. (2017)
Styrofoam PS films	<i>Exiguobacterium</i> sp. strain YT2 isolated from the guts of the larvae of <i>Tenebrio molitor</i> Linnaeus	60 day incubation period	Biofilm formation on PS film over a 28 day incubation period, pits and cavities on PS film surfaces; Gravimetric weight reduction of 7.4 ± 0.4% after 60 days incubation; decrease of molecular weight of the residual PS by 11%, and release of water-soluble daughter products.	Yang et al. (2015)
Pure standard (Sigma) PS flakes (mixture of polymers of two molecular weights, 4000 and 200,000)	<i>Rhodococcus ruber</i> C208	Shaken flasks incubated for 8 weeks at 28 °C.	Gravimetric weight loss of 0.8%; growth of the bacterium in mineral medium with PS powder as unique carbon source	Mor and Sivan (2008)
PVC film	Fungal isolates (<i>Phanerochaete chrysosporium</i> , <i>Lentinus tigrinus</i> , <i>Aspergillus niger</i> , and <i>Aspergillus sydowii</i>) isolated from the surface of thin pure PVC films buried in sterilized soil soaked with sewage sludge for 10 months.	7 weeks incubation in shaken flasks at 30 °C	All isolates were able to grow in mineral salt medium and degrade PVC polymer; highest rate observed by <i>P. chrysosporium</i> which showed reduction of the polymer molecular weight (from 200,000 to 178,292 Da); changes on polymer surface and surface and NMR and FTIR profiles were also observed	Ali et al. (2014)
Thermostable PVC films containing 75% (w/w) epoxidized vegetable oil	<i>Pseudomonas aeruginosa</i> and <i>Achromobacter</i> sp. isolated from a mixed community, enriched from crude petroleum-oil contaminated soil samples collected from petroleum oil fields, in the presence of the modified PVC films	180 days incubation in shaken flasks at 37 °C	Decrease in tensile strength of the plastic by about 53% and 43% in the presence of <i>P. aeruginosa</i> and <i>Achromobacter</i> sp., respectively; surface erosion and structural change of the matrix recorded by FTIR and SEM observation.	Das et al. (2012)
PVC films containing about 35% (w/w) of additives and PVC film wastes (used for fruits and vegetables packaging)	<i>Pseudomonas citronellolis</i> DSM 50332 and <i>Bacillus flexus</i> DSM 1320	30 or 90 days incubation in shaken flasks at 30 °C	PVC films brittle fracturing recorded after 45 days with both strains; GPC measurements showed a reduction of the Mn (%) by around 10 and 7% in the presence of <i>P. citronellolis</i> and <i>B. flexus</i> , respectively; after 90 days; Gravimetric weight losses between 13.07 ± 0.36% and 18.58 ± 0.01% for PVC films wastes incubated with <i>P. citronellolis</i> , after 30 days incubation.	Giacomucci et al. (2019a)
Plasticized PVC (pPVC) film	Several fungal isolates (<i>Aureobasidium</i> , <i>Alternaria</i> , <i>Aspergillus</i> , <i>Phaeoocomyces</i> sp.; <i>Epicoccum</i> ; <i>Taphrina</i> , <i>Cladoporium</i> ; <i>Paecilomyces</i> ; <i>Penicillium</i> ; <i>Thanatephorous</i> ; <i>Emericella</i> ; <i>Rhodotorula</i>) obtained from the surface of pPVC exposed to the atmosphere (Blackley, Manchester, UK) for up to 95 weeks	PVC film pieces placed on the surface of agar in Petri dish, inoculated with single fungal (yeast or mould) isolate and incubated at 25 °C for 4 weeks.	Growth with the intact pPVC film as the sole carbon source, up to 7% gravimetric weight loss of the film recorded	Webb et al. (2000)

et al., 2013). Fontanella et al. (2013) compared the biodegradability of different PP films (51 to 63 µm) containing various pro-oxidant additives (based on Mn/Fe, Co and Mn) and the additive-free films using *Rhodococcus rhodochrous* ATCC 29672. After 6 months incubation, they reported the biodegradability of PP films containing Mn + Fe or Mn

additive that were able to support bacterial growth, while no biodegradation was observed in the case of additives-free polymers.

Jeon and Kim (2016) isolated, from a soil sample recovered from an open storage yard for municipal solid waste, a bacterium identified as *Stenotrophomonas panacihumi* PA3–2 that showed the ability to degrade

PP. The assay was performed for 90 days under composting conditions at 37 °C and the biodegradation activity was evaluated according to KS M3100-1:2002; MOD ISO 14855:1999 method and GPC analysis. Biodegradation of UV and thermally pretreated PP by a co-culture of *Bacillus* and *Pseudomonas* was also reported by Aravinthan et al. (2016). Throughout 12 months incubation, high bacterial growth rate was recorded in the presence of the pretreated PP as major carbon source. At the end of the experiment, UV-treated PP exposed to the co-culture exhibited 22.7% thermogravimetric (TG) weight loss and a gravimetric weight loss percentage of $1.95 \pm 0.18\%$. Skariyachan et al. (2018) reported on the biodegradation of PP pellet and strips by aerobic thermophilic artificial bacterial consortia constructed based on *Brevibacillus* spp. and *Aneurinibacillus* sp. isolates obtained from waste management landfills and sewage treatment plants. After 140 days incubation, biofilm formation was observed on the surface of the plastic and a gravimetric weight loss of up to $56.3 \pm 2\%$ was recorded. Biodegradation was also supported by FTIR, microscopy observations as well as by gas chromatography-mass spectrometry and NMR analysis of end products.

3.1.3. Polystyrene biodegradation

Most of the studies evaluating the biodegradation of PS have been performed using pure microbial cultures. Mor and Sivan (2008) reported on biofilm formation on pure PS flakes by the actinomycete *Rhodococcus ruber* isolate C208. Incubation of the biofilm under shaken flask for up to 8 weeks resulted in 0.8% gravimetric weight loss. Mineralization of ozone-pretreated PS by the fungal strain *Penicillium variable* has been reported (Tian et al., 2017). An extensive review on biodegradation of PS and modified/blended PS has been published recently by Ho et al. (2018).

In a recent study by Johnston et al. (2018), an interesting approach coupling biodegradation of prodegraded high impact and general purpose PS flakes to the production of PHAs was reported. The bacterial strain *Cupriavidus necator* H16 was indeed shown to be able to grow in the presence of oxidized (by thermal and ozone pretreatment) Mn-myristate and Co-stearate containing prodegraded PS as carbon source and produce PHAs.

3.1.4. Poly-vinyl-chloride biodegradation

There are only few reports on the biodegradation of PVC polymers and plastics. Webb et al. (2000) reported that tested fungal strains obtained from the surface of plasticized PVC films exposed to the atmosphere (Blackley, Manchester, UK) were able to grow with the intact plasticized PVC (pPVC) (0.5 mm thick films) as the sole source of carbon, degrade the plasticizer dioctyl adipate (DOA), produce extracellular esterase, and cause weight loss (max of about 7%) of the substratum during growth *in vitro*. Ali et al. (2014) reported on the isolation of 4 fungal strains after burial of thin pure PVC films in sterilized soil soaked with sewage sludge for 10 months. The isolates identified as *Phanerochaete chrysosporium*, *Lentinus tigrinus*, *Aspergillus niger* and *Aspergillus sydowii* were able to grow in mineral salt medium and degrade PVC polymer after 7 weeks of incubation. In specific, in the presence of *Phanerochaete chrysosporium* reduction of the polymer molecular weight; changes on polymer surface and NMR and FTIR profiles were observed. Biodegradation of plasticized PVC by *Pseudomonas aeruginosa* and *Achromobacter* sp. bacterial strains isolated from hydrocarbon-contaminated soil samples (Das et al., 2012) has also been reported. Finally, in a recent study performed by our group (Giacomucci et al., 2019a), pure strains of *Bacillus* and *Pseudomonas* were able to degrade PVC films. GPC measurements showed a reduction of the average molecular weight M_n (%) by up to 10% after 90 days of PVC film incubation with the bacterial strains; suggesting that both strains were able to attack the PVC polymer and cause some chains scission leading to the formation of smaller fragments (the films were completely fragmented at this incubation time). Moreover, *P. citronellolis* was able to degrade PVC wastes used for fruits and vegetables packaging; after 30 days incubation, as shown by gravimetric weight losses between $13.07 \pm 0.36\%$

and $18.58 \pm 0.01\%$ for PVC films incubated with the bacterium, compared to a maximum loss of $8.39 \pm 1.10\%$ in the case of the abiotic controls.

3.2. Aquatic systems

Currently, there are several reports on the occurrence of microplastics in freshwater systems including lakes, rivers as well as in wastewater treatment plants (Li et al., 2018; Leslie et al., 2017; Peng et al., 2018; Anderson et al., 2016; Xiong et al., 2018; Sruthy and Ramasamy, 2017; Ziajahromi et al., 2017; Lasee et al., 2017; Prata, 2018; Horton et al., 2017; Su et al., 2016; Anderson et al., 2017; Sighicelli et al., 2018; Wang et al., 2018; Free et al., 2014; Klein et al., 2015; Kapp and Yeatman, 2018; Blettler et al., 2017). Metagenomic studies have also been performed for the characterization of microbial communities colonizing surfaces of microplastics in freshwater systems (Arias-Andres et al., 2018; Miao et al., 2019). However, few data are available on plastics degradation under these environmental conditions (Naz et al., 2013; Sarmah and Rout, 2018). Indeed, investigation of (micro)plastics occurrence in freshwater systems has been performed mainly with the aim of studying the role of these systems in the transport of terrestrial -derived (micro)plastic pollutants to marine environment.

Regarding the marine habitat, several studies have focused on the assessment and characterization of microbial communities on the surface of the plastics sampled from the marine environment using culture-independent approaches. Such studies did not however provide in most cases clear evidences about the actual plastic biodegradation ability of the colonizers. A metagenomic approach for deciphering the putative genes expressed in plastic surface-associated biofilm was used (Bryant et al., 2016). In a recent study, Debroas et al. (2017) defined the microbial core (eukaryotes, bacteria and Archaea) with their main metabolic pathways and the putative hitchhikers associated with various plastic polymers in the North Atlantic Gyre. Microbial biofilm communities associated to ocean plastics such as PE, PP, polyethylene terephthalate (PET) or to un-characterized microplastics has been described using 16 S ribosomal RNA gene analysis (Zettler et al., 2013; Oberbeckmann et al., 2014; Harrison et al., 2014; De Tender et al., 2015; Oberbeckmann et al., 2016, 2017; Jiang et al., 2018). In addition, some studies reported on the weathering of microplastics in the ocean (Ter Halle et al., 2017), while to date a few reports refer about the biodegradation of the conventional plastics under marine environmental conditions by pure isolates or mixed microbial consortia (Table 2). They have been performed mostly under aerobic conditions.

3.2.1. Polyethylene biodegradation

Sudhakar et al. (2007) reported on a maximum gravimetric weight loss of 2.5% for low LDPE and 0.8% for HDPE after 6 months incubation *in situ* in ocean waters. HDPE incubated at 30 °C for 30 days in shaken flasks containing synthetic medium in the presence of *Arthrobacter* sp. or *Pseudomonas* sp. isolated from the plastic waste dumped sites of the Gulf of Mannar region (India) exhibited nearly 12% and 15% of gravimetric weight loss respectively (Balasubramanian et al., 2010). Harshvardhan and Jha (2013) reported on a gravimetric weight loss of up to 1.75% of LDPE incubated for 30 days in the presence of marine bacterial isolates in Bushnell-Haas medium under shaken flasks. Syranidou et al. (2017a) collected naturally weathered PE films from two coastal sites in Chania (Greece) and used them for biodegradation experiments under simulated marine conditions. Specifically, a two-phase microcosms experiment was set up and includes an initial phase during which microbial biofilms (indigenous or bioaugmented by 10^8 CFU mL⁻¹ *Lysinibacillus* sp. and *Salinibacterium* sp. isolates selected from marine environment in the presence of LLDPE as main carbon source) were let to develop on the surface of linear LDPE (LLDPE) films for 6 months and then harvested and used as inoculum in biodegradation assay of naturally weathered PE films. The results showed the

Table 2

Biodegradation of PE, PP, PS and PVC under aquatic systems.

Plastic	Type of microorganism	Biodegradation assay conditions	Biodegradability method	Reference
Naturally weathered linear LDPE films	Indigenous marine communities alone or bioaugmented with the marine pure isolates <i>Lysinibacillus</i> sp. and <i>Salinibacterium</i> sp. selected based on their ability to grow in the presence of LDPE as main carbon source	Seawater microcosms incubated at 25 °C on a stirring table at 120 rpm in darkness for up to 6 months.	19% and 4.2% mass loss in the presence of the bioaugmented and the indigenous community, respectively.	Syranidou et al. (2017a)
UV-treated PE, PS, PP microplastics	<i>Bacillus gottheilii</i> and <i>B. cereus</i> isolated from mangrove ecosystems in Peninsular Malaysia	Growth in mineral salt medium with 0.5 g of specific plastic polymers in shaken flasks for 40 days at 29 °C	Weight loss percentages by <i>B. cereus</i> were 1.6% and 7.4% for PE and PS, respectively. <i>B. gottheilii</i> recorded weight loss percentages of 6.2%, 3.6% and 5.8% for PE, PP, and PS, respectively.	Auta et al. (2017)
HDPE	<i>Arthrobacter</i> sp. or <i>Pseudomonas</i> sp. isolated from the plastic waste dumped sites of the Gulf of Mannar region (India)	HDPE incubated at 30 °C for 30 days in shaken flasks containing synthetic medium	Gravimetric weight losses of 12% (<i>Arthrobacter</i> sp.) and 15% (<i>Pseudomonas</i> sp.)	Balasubramanian et al. (2010)
Biodegradation of LDPE microplastics	Marine fungus <i>Zalerion maritimum</i> ATCC 34329	Shaken flasks with 0.130 g of microplastics incubated at 25 °C for 28 days	Mass and size decrease of the pellets as well as changes in FTIR spectra	Paço et al. (2017)
PE plastic bags	Microbial colonization and degradation of in temperate fine-grained organic-rich marine sediments.	Oxic and anoxic marine sediment slurry, incubation for 98 days at 10 °C in the dark	Plastics were colonized by different microbes but no sign of biodegradation was observed	Nauendorf et al. (2016)
HDPE films (bags)	<i>Brevibacillus borstelensis</i> isolated from marine sediment	Aerobic conditions, shaken flasks, 30 days incubation at 37 °C	11.4% weight loss, pits and cavities formation on the HDPE film surface observed by SEM.	Mohanrasu et al. (2018)
LDPE films (bags)	Marine <i>Kocuria palustris</i> M16, <i>Bacillus pumilus</i> M27 and <i>Bacillus subtilis</i> H1584 from pelagic waters, Arabian Sea, India.	Incubation for 30 days in Bushnell–Haas medium in shaken flasks at 37 °C	Gravimetric weight loss of up to 1.75%	Harshvardhan and Jha (2013)
Commercially available HDPE film (containing additives)	10 bacterial isolates belonging the genera <i>Bacillus</i> spp. and <i>Pseudomonas</i> spp. obtained from partially degraded PE material along with adhered soil samples recovered from the plastic waste dumped sites in the coastal region districts of Tamil Nadu (India)	4 weeks incubation in shaken flasks at 30 °C	Gravimetric weight loss percentages of up to 23.14 ± 0.24% and detection of oxidized groups by FTIR	Sangeetha Devi et al. (2019)
UV-treated PP microplastics	<i>Bacillus</i> sp. strain 27 and <i>Rhodococcus</i> sp. strain 36 isolated from mangrove sediment.	Growth in MSM with 0.5 g of microplastic in shaken flasks for 40 days at 29 °C	Weight loss percentages of 6.4% (<i>Rhodococcus</i> sp. strain 36) and 4% (<i>Bacillus</i> sp. strain 27) observed	Auta et al. (2018)
Naturally weathered PS films collected from coastal sites in Chania	Indigenous marine communities alone or bioaugmented with the marine pure isolates <i>Rhodococcus</i> sp, <i>Shewanella</i> sp and <i>Pseudomonas</i> sp. selected based on their ability to grow in the presence of PS as carbon source	Seawater microcosms, 6 months incubation	4.7% and 2.3% mass loss in the presence of the bioaugmented and the indigenous community, respectively.	Syranidou et al. (2017b)
PVC films	Enriched anaerobic marine consortia	Marine microcosms incubated at 20 °C for up to 24 month	Gravimetric weight loss of up to 11.67 ± 0.58%; decrease in the Mn% by 9% and a decrease in thermal stability in 3/16 consortia after 7 months incubation	Giacomucci et al., 2019b (Unpublished results)

development of bacterial biofilms on naturally weathered PE films surface composed of genera affiliated with polymer (PE, cellulose) and hydrocarbons degraders, and that exhibited biodegradation ability towards the naturally weathered PE film. Namely, after 6 months, the bioaugmented biofilm bacteria and the indigenous community decreased the gravimetric weight of weathered PE by 19% and 4.2%, respectively, while no weight reduction was observed for the abiotic controls. Auta et al. (2017) reported on the isolation of a *Bacillus gottheilii* strain from mangrove that was able to degrade commercial PE subjected to UV pretreatment for 25 days before being used in biodegradation assays. The microplastic exhibited a gravimetric weight loss of 6.2% as well as changes in the FTIR spectrum after 40 days incubation. Paço et al. (2017) evaluated the biodegradation of PE pellets by the marine fungus *Zalerion maritimum* in a minimum growth medium. Results showed that the fungus was capable of utilizing PE based on the mass and size decrease of the pellets. Eich et al. (2015) tested colonization of PE plastic bags in a field experiment in the Mediterranean Sea environment. The results showed that already after 15 days the film surface was colonized by different microorganisms, while no biodegradation activity was recorded at the end of the experiment (33 days incubation).

In a recent study by Karlsson et al. (2018), LDPE films (with no additives) thermally pre-degraded to different levels were placed in

stainless steel cages in the sea off the Swedish west coast for 12 summer weeks. The samples recovered at different time points and subjected to different analyses showed that all plastic films exhibited a continued oxidation in the field. Fragmentation of the pre-degraded and decrease in tensile strain of non-degraded films were also recorded. Both pre-degraded and non-degraded films were subjected to biofouling. All the observed processes were faster for pre-degraded material. No data on the polymers biodegradation were provided. Among 248 marine bacterial isolates obtained from partially degraded PE material along with adhered soil samples recovered from the plastic waste dumped sites in the coastal region districts of Tamil Nadu (India), 10 bacterial isolates belonging the genera *Bacillus* spp. and *Pseudomonas* spp. were considered to be high HDPE degraders based on gravimetric weight loss, biofilm formation on the surface of the plastic film and FT-IR after 30 days incubation period (Sangeetha Devi et al., 2019).

The study by Nauendorf et al. (2016) is among the very few reports available on the evaluation of plastics biodegradation under anoxic conditions. The authors monitored microbial colonization and degradation of PE carrier bags under oxic and anoxic marine conditions. Specifically, PE samples and biodegradable plastic carrier bags were incubated in natural sediments from Eckernförde Bay (Western Baltic Sea) for 98 days under both conditions. The results showed that, although

plastics were colonized by different microbes, no sign of biodegradation was observed.

3.2.2. Polypropylene biodegradation

Sudhakar et al. (2007) reported on a 0.5% gravimetric weight loss with unblended PP (1.5 mm thickness) after 6 months incubation *in situ* in ocean waters, although there have been the formation of a high-density biofilm. Auta et al. (2018) reported on a gravimetric weight loss of 6.4% by *Rhodococcus* sp. strain 36 and 4.0% by *Bacillus* sp. strain 27, isolated from mangrove systems (Peninsular Malaysia), of UV-pretreated PP after 40 days incubation. PP biodegradation was further confirmed using FTIR spectroscopy and SEM analyses, which revealed structural and morphological changes in the PP microplastics with microbial treatment. Mohanrasu et al. (2018) reported on the biodegradation of HDPE plastic bags by *Brevibacillus borstelensis* isolated from marine sediment. After 30 days incubation in shaken flasks, a weight loss of 11.4% was recorded and SEM observation showed pits and cavities formation on the HDPE film surface.

3.2.3. Polystyrene biodegradation

Syranidou et al. (2017b) reported that naturally weathered PS films collected from two coastal sites in Chania (Greece) were shown to be degraded in seawater microcosms based on Mn decrease of up to 32%, a gravimetric weight reduction of 4.7% and the observation of cracks/fissures on the surface of the films, after 6 months incubation. Biodegradation of UV-pretreated PS by *B. cereus* and *B. gottheilii* strains isolated from mangrove (Auta et al., 2017).

3.2.4. Poly-vinyl-chloride biodegradation

In a recent study performed by our group, different anaerobic marine consortia exhibiting the ability to degrade plasticized unpretreated PVC films were enriched and their associated microbial communities characterized (Giacomucci et al., 2019b). After 7 months incubation, 3/16 enriched consortia were able to degrade the films apparently acting against both the additives and the PVC polymer chains. For these consortia, gravimetric weight loss of up to $11.67 \pm 0.58\%$ was recorded, GPC showed a decrease in the Mn% from 100% up to ~91% suggesting that some polymer chain scission took place leading to the formation of smaller fragments, while TGA revealed a decrease in thermal stability. Prolongation of the incubation to 24 months allowed the selection of other three consortia able to break down the PVC polymer chains. To our knowledge, the study was the first reporting on biodegradation of plasticized PVC films under anoxic marine conditions and providing information about the composition of the marine anaerobic microbial communities potentially responsible for it. The PVC film-associated communities of the biodegrading microcosms was characterized and encompasses microbial phyla closely related to those previously reported from laboratory/field enrichments as able to degrade halogenated organic compounds and hydrocarbons, letting to hypothesize that dechlorination of PVC film could also have occurred. Hence, the study provided insights on the potential fate of PVC plastics introduced into marine environment.

3.3. Insect gut-associated microbes

Bacteria inhabiting the gastrointestinal tract of some insects larvae have been described as able to degrade petroleum-derived synthetic plastics such as PE and PS. Specifically, Yang et al. (2014) reported that waxworms, the larvae of *Plodia interpunctella*, were able of chewing and eating LLDPE films. They isolated from the worm's gut two bacterial strains, *Enterobacter asburiae* YT1 and *Bacillus* sp. YP1, capable of degrading non-pretreated LLDPE as supported by the ability of the two strains to form viable biofilms, to decrease the hydrophobicity and to cause damage on the surfaces of the LLDPE films. Furthermore, a gravimetric weight loss of up to $10.7 \pm 0.2\%$ and molecular weight

reduction up to 13% were recorded after 28 days incubation in the presence of the bacterial isolates used individually. Bombelli et al. (2017) reported on the bio-degradation of PE by larvae of the wax moth *Galleria mellonella*. The authors used commercially available PE bags but did not specify the characteristics of the plastic used *i.e.* the presence of additives or plasticizers.

In another study, microorganisms associated to the gut of mealworms, the larvae of *Tenebrio molitor* Linnaeus, have been reported to play a crucial role in the mineralization of polystyrene (Yang et al., 2015). Specifically, the authors isolated a bacterial strain *Exiguobacterium* sp. strain YT2 from the gut of the larvae that was able to grow on PS film. After 28 days incubation with strain YT2, changes in surface topography, decrease in hydrophobicity, and formation of carbonyl groups as well as reduction in molecular weight up to 11% and gravimetric weight loss of $7.4 \pm 0.4\%$ of the polymer, were recorded.

4. Conclusions and further perspectives

Research performed on the assessment of the biodegradation of the recalcitrant most produced petroleum-based polymers with carbon carbon backbones/plastics namely PS, PVC, PP and PEs, has proved that they can be attacked, under different lab and actual site conditions, by a variety of mixed microbial communities and pure fungal or bacterial strains from terrestrial or aquatic environment and from some insects larval gut. Most of the available reports have dealt, however, with PE biodegradation by terrestrial deriving microbes under aerobic conditions while data on other conditions and on the biodegradation of the other plastics are still very limited and especially in marine environment. The biodegradation rates and extents reported are often very limited and the available data prove that microbial adaptation to conventional plastics occurring in the environment is slowly occurring in both terrestrial and aquatic impacted environments.

Plastic wastes remarkably contribute to marine litter and its negative effects on the marine ecosystem is being demonstrated. However, the biodegradability of major types of plastics and plastic wastes has been evaluated mainly under terrestrial environments, based on the assumption that the fate of plastics is considered to be composting or landfilling, and thus efforts addressed to better determine the actual *in situ* biodegradation of wastes of conventional plastics in marine habitats are necessary. In any case, the evidences available allow to conclude that biodegradation of microplastics can very slightly contribute to mitigate environmental impact of plastic pollution or to reduce *in situ* marine litter, even through the *in situ* stimulation or bioaugmentation with plastic-degrading microorganisms.

Thus, strategies aiming at the improvement of recycling rates of separately collected plastics as well as to a better managing the plasmix are needed. Furthermore, we require site specific strategies for removing the marine litter already existing in the basin and to develop treatments/strategies for the on site valorization of the collected plastic materials. More importantly, we need to prevent marine litter problem by i) combining selective and efficient collection and recycling of the different plastics used in our everyday life, ii) eliminating the dump and landfill release of microplastics, iii) banning non-essential plastic products and restrict the use of plastic micro-granules in commercial products and in particular iv) gradually adopting biodegradable (bio)plastics, by starting from those used in marine habitats for preparing fishing gears, tubular net for marine aquaculture, mussel-culture socks or additives for painting and maintenance of ships and leisure boats.

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