

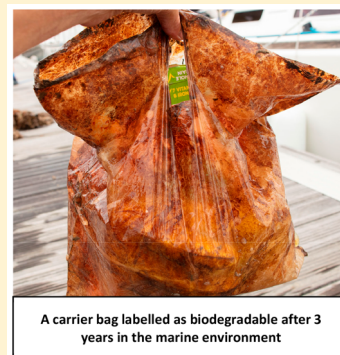
# Environmental Deterioration of Biodegradable, Oxo-biodegradable, Compostable, and Conventional Plastic Carrier Bags in the Sea, Soil, and Open-Air Over a 3-Year Period

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## **S** Supporting Information

**ABSTRACT:** There is clear evidence that discarded single-use carrier bags are accumulating in the environment. As a result, various plastic formulations have been developed which state they deteriorate faster and/or have fewer impacts on the environment because their persistence is shorter. This study examined biodegradable, oxo-biodegradable, compostable, and high-density polyethylene (i.e., a conventional plastic carrier bag) materials over a 3 year period. These materials were exposed in three natural environments; open-air, buried in soil, and submersed in seawater, as well as in controlled laboratory conditions. In the marine environment, the compostable bag completely disappeared within 3 months. However, the same compostable bag type was still present in the soil environment after 27 months but could no longer hold weight without tearing. After 9 months exposure in the open-air, all bag materials had disintegrated into fragments. Collectively, our results showed that none of the bags could be relied upon to show any substantial deterioration over a 3 year period in all of the environments. It is therefore not clear that the oxo-biodegradable or biodegradable formulations provide sufficiently advanced rates of deterioration to be advantageous in the context of reducing marine litter, compared to conventional bags.



A carrier bag labelled as biodegradable after 3 years in the marine environment

## 1. INTRODUCTION

Plastics are lightweight, strong, durable, and corrosion-resistant materials which have become an integral part of daily life worldwide.<sup>1</sup> The versatility of plastic, together with its low cost, has resulted in annual worldwide production exceeding 335 million tonnes.<sup>2</sup>

Approximately 50% of plastics are discarded after a single-use.<sup>3,4</sup> This creates a major waste management problem, with plastics accounting for approximately 8–10% of all the waste generated in the U.K.<sup>3,5</sup> Considerable quantities of end of life plastics also escape to the environment as litter, and single-use items constitute a large proportion of the litter found in marine and terrestrial environments.

There is evidence that plastic debris can harm maritime industries, tourism, and human wellbeing.<sup>6–8</sup> In the marine environment the accumulation of plastic debris has been identified as a major global issue by the United Nations Environment Assembly and in the G7 Leaders' declaration 2015.<sup>9–11</sup>

Plastic debris is widespread in terrestrial and freshwater environments. However, much of the existing information about the presence of plastics in these environments is focused on sources and transportation pathways to the oceans. Given that the majority of all plastics will be used and disposed of on land, terrestrial environments will themselves be subject to extensive pollution by plastics of all sizes, based on large amounts of anthropogenic litter from both point (e.g., landfill) and diffuse (e.g., general littering) sources. As such it is highly

likely that soils may also act as long-term sinks for plastic debris.<sup>12,13</sup>

Since their introduction in the 1970s, plastic carrier bags have become widespread in daily life worldwide.<sup>14</sup> They are typically considered as single-use items and are commonly made from polyethylene.<sup>15</sup> These bags are an iconic symbol of our “throw-away” society and their waste is often viewed as a very visible nuisance. In 2010, it was estimated that 98.6 billion plastic carrier bags were placed on the European Union (EU) market and about 100 billion plastic bags have been placed additionally every year since.<sup>16</sup>

Plastic carrier bags are often supplied free or for a low charge and used in high volumes. Consumption figures vary greatly between countries, with annual use per capita exceeding 450 bags in some EU countries.<sup>16</sup> Interventions to reduce the use of plastic bags have been varied in range and scope. Governments in many nations have strategies to either ban the sale of lightweight bags, charge customers for their use, and/or generate taxes from stores who sell them.<sup>17</sup> Several countries have already included bans or taxes, which have resulted in substantial reductions in use.<sup>18</sup> However, there is no consistency between countries.

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There are concerns that littering of plastic carrier bags presents a substantial source of contamination in the oceans. They have been found to be one of the most common items in the intertidal<sup>19,20</sup> and subtidal benthos.<sup>21</sup> Even if properly discarded, lightweight bags can unintentionally be transferred away from landfill sites or other areas by wind or heavy rain.<sup>5</sup>

The presence of carrier bags in the marine environment can have a number of effects; for example, previous research by Bugoni et al. (2001), found that out of 50 stranded dead sea turtles, plastic carrier bags were the main debris ingested.<sup>22</sup> Additionally, Green et al. (2015) found that within 9 weeks in the marine environment, plastic carrier bags can create anoxic conditions within the sediment, and that their presence can significantly lower abundances of infaunal invertebrates. This indicates carrier bags can rapidly alter marine assemblages and the ecosystem services they provide.<sup>23</sup> Additionally, Hodgson et al. (2018) used laboratory experiments with carrier bags and showed that amphipods can shred plastic carrier bags, generating numerous microplastic fragments.

The hydrophobicity and long carbon chain molecular structure of polyethylene, which is widely used for plastic bags, makes it resistant to biodegradation under normal conditions. The time frame for the complete mineralization is unknown, creating a major waste management issue.

Awareness of the accumulation of end of life plastic and its impact on the environment has, in part led, to interest in the development of degradable polymers. Biodegradable, oxo-biodegradable and compostable plastics are often regarded as potential solutions to the accumulation of plastic litter and waste. Some of these products are marketed accompanied by statements indicating they can be “recycled back into nature much more quickly than ordinary plastic”<sup>25</sup> or “plant-based alternatives to plastic”.<sup>26</sup>

These materials are widely used for the production of carrier bags and some are also used to make a variety of other items, including single-use cutlery, water bottles, and straws.

Biodegradation takes place through the action of enzymes and/or chemical deterioration associated with living organisms, bacteria, fungi, and algae. This occurs in two steps; the first is the fragmentation of the polymers into sections of lower molecular mass by means of either abiotic reactions (i.e., oxidation, photodegradation, hydrolysis), or biotic reactions (i.e., degradation by microorganisms). This is followed by bioassimilation of the polymer fragments by microorganisms and its mineralization.<sup>27</sup>

A material may be labeled as “biodegradable” if it conforms to certain national or regional standards.<sup>28,29</sup> Such standards could include: ISO, European Norm – EN, and American Society for Testing and Materials (ASTM) International. Some standards are appropriate for conditions that occur in an industrial composter, in which temperatures are expected to reach 70 °C. Other standards, focus on laboratory-based biodegradation tests using measurements of oxygen demand or CO<sub>2</sub> evolution; for example, ISO 19679:2016 (2016) tests for the aerobic biodegradation of plastics at the interface between seawater and sandy marine sediment.<sup>30</sup> Oxo-biodegradable plastics (oxo-plastics) are reported to contain an additive (pro-oxidant) which is intended to break the molecular chain within the polymer which will then lead to its biodegradation.<sup>31</sup> However, there is typically no clearly defined time frame given for the breakdown of oxo-/biodegradable plastics.<sup>32</sup>

In this context the term ‘Composting’ relates to enhanced biodegradation under managed conditions, predominantly

characterized by forced aeration and natural heat production resulting from biological activity decomposing the material. The resulting output material, *compost*, contains nutrients and can be used as a soil improver.<sup>33</sup> Therefore, compostable plastics should biodegrade in a managed composting process through the action of naturally occurring micro-organisms and typically do so in relation to a specified time frame.<sup>32</sup> However, this can only occur if there is a specific waste stream dedicated to compostable waste.

There is a lack of clear evidence that biodegradable, oxo-biodegradable, and compostable materials offer an environmental advantage over conventional plastics, and the potential for fragmentation into microplastics causes additional concern.<sup>28,34</sup> To date, studies focusing on the deterioration of different types of degradable plastics in the environment give varying results and are shorter in time frame.

The EU is proposing a process to restrict the use of oxo-plastics,<sup>33</sup> because of the lack of consistent evidence about rates of deterioration in the environment, allegedly misleading claims to consumers and risks that labeling products as biodegradable may inadvertently promote littering behavior.

The present study describes the deterioration in different natural environments of bags, which were stated to have biodegradable, oxo-biodegradable or compostable properties. We do not specifically attempt to quantify biodegradation performance in relation to any specific standard such as degradability in a commercial composting facility. Rather we assess whether or not there has been any meaningful deterioration in the context of reducing marine litter; for example, had the bag remained intact or deteriorated into visible fragments? A conventional polyethylene plastic carrier bag was also examined for comparison. All bags were available at the point of sale in U.K. high-street retailers. These materials were exposed in various environments that discarded carrier bags could encounter; in open-air, buried in soil, and submersed in the marine environment. This is the first research where plastic deterioration has been examined simultaneously across these three natural environments, together with controlled conditions in the laboratory. Five different plastic carrier bag formulations were considered, and their deterioration was evaluated over a 3-year period. Deterioration was considered in terms of visible loss in surface area, as well as approaches to detect more subtle changes in tensile stress, surface texture, and chemical structure.

## 2. MATERIALS AND METHODS

**2.1. Sample Preparation.** Five different types of plastic carrier bag were compared (Table 1): these included two types of oxo-biodegradable bag (labeled here as Oxobio1 and Oxobio2), one biodegradable bag, one compostable bag, and a high-density polyethylene (HDPE) carrier bag (labeled in this research as a conventional carrier bag), which was not stated to have any particular deterioration/compostable properties. Deterioration in this study is used to describe the process of a becoming a lower quality or condition.

The bags were chosen as they were all opaque and were obtained based on their prevalence in retail stores in and around Plymouth, U.K. Sixteen samples of each bag were obtained. In order to obtain a representative sample of each bag type, a maximum of two bags were sourced from any one store on any single occasion. Where repeat visits to the same store were necessary to obtain sufficient independent samples, these visits were separated by at least 2 weeks. Hence our

Table 1. Information on the Tested Carrier Bags and the Properties As Stated on the Manufacturer's Website<sup>a</sup>

designated label for testing	degradation properties (as stated on bag)	information stated on web sites linked to the product	disposal/antilittering information (as stated on bag)
Oxobio1	degradable plastics (D <sub>2</sub> W trademark, logo)	oxo-biodegradable ( <a href="https://www.symphonyenvironmental.com/d2w/">https://www.symphonyenvironmental.com/d2w/</a> )	no information
Oxobio2	planet safe plastic; incorporating EPI's totally degradable plastic additives (EPI trademark, logo)	oxo-biodegradable ( <a href="http://www.epi-global.com/en">http://www.epi-global.com/en</a> )	no information
biodegradable	biodegradable bag (exo plastics logo, sustainable bioplastic; biodegradable ISO 14855)	no claims about biodegradability on exo plastics web site ( <a href="https://www.exoplastics.com/">https://www.exoplastics.com/</a> )	recyclable (no numerical category for recycling type stated)
compostable	completely compostable, recycle me with food	ISO 14855 is an international standard covering aerobic biodegradability of plastic materials under controlled composting conditions plant-based compostable foodservice packaging ( <a href="https://www.vegware.com/about/info_1.html">https://www.vegware.com/about/info_1.html</a> ) compostable packaging is designed to be recycled together with food waste. <a href="https://www.vegware.com/close-the-loop/info_50.html">https://www.vegware.com/close-the-loop/info_50.html</a> EN13432 is the packaging waste directive and standards for compostability <a href="http://www.bpf.co.uk/topics/standards_for_compostability.aspx">http://www.bpf.co.uk/topics/standards_for_compostability.aspx</a>	reuse me first for shopping, and then as a food waste caddy liner! this completely compostable bag complies with standard EN13432
conventional	high density polyethylene; no degradation properties stated	no manufacturer given	suitable for industrial food waste recycling—visit <a href="http://www.foodwastenetwork.org.uk">www.foodwastenetwork.org.uk</a> , Recycling category "7 - other"
			reuse at home or recycle, recycling category "2 - HDPE"

<sup>a</sup>All bags were opaque and obtained based on their prevalence in retail stores in and around Plymouth, U.K.

experiment was designed to contain a range of products and production batches so as to be as representative as possible. Since the specific retail stores from which the carrier bags were obtained is not of particular relevance, bags will only be described based on their formulation (Table 1).

Each carrier bag type was cut into strips; 15 × 25 mm. The strip samples were taken from the main body of the carrier bag (not the handles or the sides), to provide areas of similar structure. A strip of each plastic carrier bag type was then placed into a pouch made of high-density polyethylene (HDPE) mesh and sewn secure using nylon fishing twine. Each pouch structure (150 × 200 mm) was sewn together to provide five equally spaced separated compartments. These compartments were then used to house an individual strip of each bag type (Supporting Information (SI) Figure S1). The HDPE mesh (1 × 1 mm) allowed exposure to external environments and each compartment was sewn so as to allow the bag samples to move relatively freely. Each pouch structure was attached to a permanent panel to aid removal.

These permanent panels were placed in one of four different conditions; buried in soil, exposed outdoors in the air, submerged in the marine environment and placed in a blacked-out box in the laboratory as a control. The buried samples were situated at the University of Plymouth's Skardon Garden (50°22'38.4"N, -4°08'11.9"W) and were buried to a depth of approximately 0.25 m (SI Figure S1a). The samples that were exposed in open-air were also situated in Skardon Garden and were placed on a south facing wall (SI Figure S1b). Samples placed in the marine environment were submerged on a beam at Queen Anne's Battery Marina (50°36'48.4"N, -4°12'96.5"W) at a depth of approximately 1 m (SI Figure S1c). Three kg weights were connected on each side of the beam to maintain depth. Control samples were placed in a blacked-out box (kept at room temperature) in a laboratory at the University of Plymouth.

All samples were deployed on the 10th July 2015. There were three subsequent sampling dates; 6th April 2016 (9 months), 6th January 2017 (18 months), 6th October 2017 (27 months). Additionally, whole bags of each material were also deployed in polypropylene mesh in each environment at the same time and used for visual inspection over the 3-year period (23rd August 2018).

Over this period, the samples would have been exposed to sea (8.8–18.8 °C; United Kingdom Sea Temperatures, 2019) and air (1.5–21.5 °C; Met Office, 2016) temperatures, typical of those in a temperate environment. The soil type in the South West of the U.K. is freely draining and slightly acidic.<sup>35</sup>

Before deployment, four subsample strips from each carrier bag type were tested to provide a comparison starting point. After deployment, four replicate samples of strips from differing bag replicates were collected from each environment on each sampling date. Samples were removed from the mesh structure, gently cleaned using distilled water, air-dried (30 °C), and tested (see below) within 48 h.

**2.2. Visual Inspection.** The first step on each sampling date was to visually inspect the samples to check for surface area loss, holes or disintegration. Random samples of each carrier bag type were then also visualized by scanning electron microscopy (JEOL, 7001F) prior to deployment, and then from each environment at 27 months.

Measurements of tensile stress and molecular structure using Fourier transform infrared spectroscopy (FT-IR) were made in order to detect any more subtle changes.



**Figure 1.** Oxo-biodegradable bags (oxobio2) which had either been submerged in the marine environment (left) or buried in soil (right) for over three years. Each bag is holding 2.25 kg of typical groceries.

**2.3. Tensile Stress Testing.** The thickness of each strip was measured using an electronic micrometer (Sealey; AK9635D). Each strip was measured at 10, 50, 100, and 140 mm from a central point. This produced four reference points for each sample and the mean was then calculated. The maximum load (N) for each strip was then measured using a tensile testing machine at a rate of 100 mm min<sup>-1</sup> (Instron, system ID 3345 k1669, Norwood, MA, force transducer model 2519-104, capacity 500 N). Then, the maximum tensile stress of each strip was calculated using the following equations:

$$(i) A = bh \quad (ii) \sigma = \frac{F}{A}$$

where  $b$  is the width (25 mm),  $h$  is the height (mean thickness), and  $F$  (maximum load, N) is the force for each extracted strip. For each strip eq (i) allowed calculation of the cross-sectional area ( $A$ ) and eq (ii) allowed calculation of the tensile stress ( $\sigma$ , MPa). The maximum tensile stress of a material is also termed as its ultimate strength (and referred to as the rate of disintegration within this research).

Normality of the data was confirmed by using QQ plots to examine distribution. One-way analysis of variance (ANOVA) was used to compare the maximum tensile stress difference between the different bag types before being exposed in any environment. The effects of bag type, environment and time on the maximum tensile stress was then examined. This was compared using the percentage change of tensile stress from 0 to 9 months and 9 to 27 months using a three-way ANOVA; the three factors were (bag type, environment, time). Time had two levels (0–9 and 9–27 months), bag type had five levels (Oxobio1, Oxobio2, biodegradable, compostable, and conventional) and environment consisted of four levels (control, open-air, marine, soil). Posthoc Tukey tests were then used to identify the significant effects. Any samples which were too brittle to test or were no longer visible were omitted from the analysis. All statistical tests were performed in R ver. 3.4.1 (R Core Team 2017).

**2.4. Molecular Composition Analysis (FT–IR) and Image Analysis.** In order to assess any subtle deterioration effects on the molecular composition of the materials, samples were analyzed by FT–IR microscopy in transmission mode with a Hyperion 1000 microscope coupled to a Vertex 70 spectrometer (Bruker). For each sample, the spectra was recorded with 32 scans in the region of 4000–600 cm. Prior to FT–IR, samples were cleaned with absolute ethanol to remove any residues. The spectra obtained were compared against a spectral database of synthetic polymers (BPAD polymer and synthetic fibers ATR).

### 3. RESULTS

Prior to exposure in different environments, the maximum tensile stress and thickness of the bags were measured. Oxobio2 had the highest tensile stress and thickness ( $28.82 \pm 1.55$  MPa and 0.04 mm), the compostable bag had the lowest tensile stress ( $10.47 \pm 1.23$  MPa) and the biodegradable, conventional and Oxobio1 bag had the lowest thickness (0.02 mm) (SI Table S1). All bag types had relatively consistent thickness.

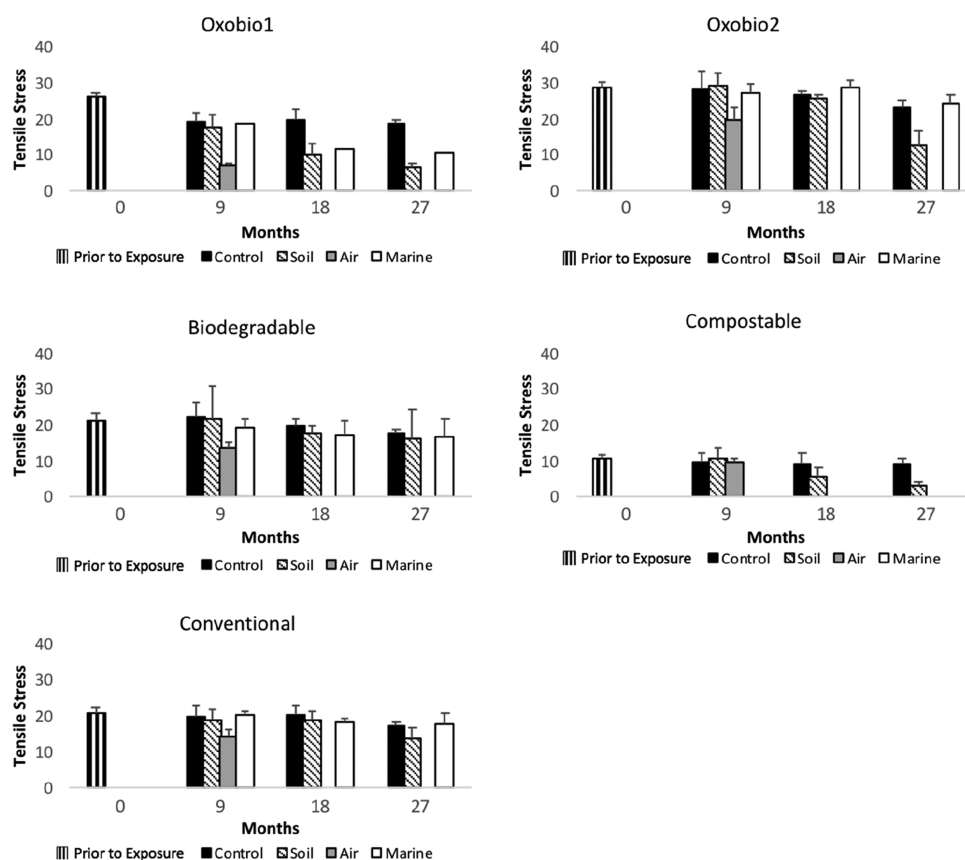
Before commencing the experiment there were significant differences in mean maximum tensile stress ( $F_{(4, 15)} = 12.94$ ,  $p = < 0.01$ ) between the carrier bag materials (SI Table S2). Posthoc Tukey's HSD tests showed that the compostable bag had a significantly lower maximum tensile stress when compared against all other bag types. All other comparisons were not significant.

After the various exposure periods, all pouch structures were successfully recovered from all environments. The strips and whole bags were then analyzed visually.

For plastic bag strips in both the control and soil environment, no surface area loss was measurable over the period of 27 months. Within the marine environment, a microbial biofilm was visible on the surface of all bags after 1-month. However, the compostable bag samples (including whole bags) were no longer visible by the first sampling date of 9 months.

After 9 months, in the open-air environment all bag types (including conventional polyethylene) were too brittle to test and had or were disintegrating into pieces. Most of the pieces were in the microplastic size range (<5 mm); therefore, they could not be examined for tensile stress. The whole bags were also found to have disintegrated into microplastic pieces. Substantial quantities of the fragments that formed were visible to the naked eye on the ground beneath the test rig and in the pouches. While disintegration into microplastic was apparent it was not clear whether this fragmentation could have altered the potential for the plastic to biodegrade and more work would be needed to establish this together with the associated time scale.

Scanning electron images were obtained before environmental exposure and then again after 27 months. Minor changes were noticeable within the open-air environment for sample fragments from both conventional and compostable bag types. Cracks and holes were present in the conventional bag material suggesting deterioration (SI Figure S2;1b). For the compostable material, solid deposits that looked like filamentous bacteria were visible on the surface; however, no cracks or holes were present nearby (SI Figure S2;2b).



**Figure 2.** Mean maximum tensile stress of plastic carrier bag samples, shown as maximum stress before breakage displayed (mean + S.D.) over a 27-month exposure period in four different environments (control, marine, soil, open-air). Open-air is labeled as air in this graph. If bag type is not shown in relation to an environment, it denotes complete disintegration/fragmentation and hence samples were not testable.

After 3 years, photographs were taken of the whole bags from both the soil and marine environment (Figure 1). As a qualitative assessment of functionality, the bags were loaded with typical groceries from a local supermarket (weight 2.25 kg). Oxobio1, Oxobio2, biodegradable, and conventional were still functional and retained the items with no breakages. However, the compostable bag type (which was only present in the soil environment for 27 months) was unable to hold any weight without tearing.

The maximum tensile stress of all plastic types decreased in all environments over time, but at different rates (Figure 2). This testing involved destructive sampling of 262 samples, with each measurement taken from a previously untested strip.

As the compostable bag samples had completely disappeared from the mesh in the marine environment this gave an unbalanced data set, and so this bag type was examined using a separate analysis just considering the remaining environments and sampling dates. Additionally, all bag types after 9 months in the open-air environment could not be tested due to being too brittle; these were subsequently omitted from statistical testing from 9 to 27 months.

From the perspective of litter and potential interactions with biota, most of the bag samples remained intact. However, subtle changes in tensile stress were apparent in all the bag materials indicating some degree of deterioration; the factors time, bag type and environment showed significant differences between 0–9 months exposure (SI Table S3). Post hoc comparisons found that Oxobio1 lost strength at a significantly faster rate than the other bags between 0–9 months ( $p = <$

0.01). There were also differences between the two Oxobiodegradable samples; Oxobio1 lost strength significantly faster than Oxobio2 ( $p = 0.01$ ). Additionally, bags exposed in the open-air environment lost strength more rapidly when compared to the other environments: control ( $p = < 0.01$ ), marine ( $p = < 0.01$ ) and soil ( $p = < 0.01$ ).

A second ANOVA was conducted which included the compostable bag type. This bag type needed a separate analysis as all its samples had completely disappeared within the marine environment after 3 months. This ANOVA showed that the compostable bag material had a significant difference in tensile stress to Oxobio1 ( $p = < 0.01$ ). The compostable bag material exposed in the open-air environment also lost its strength more rapidly when compared to bags exposed to both control ( $p = < 0.01$ ) and soil ( $p = < 0.01$ ) environments.

Exposure from 9–27 months produced similar relative changes in tensile stress patterns as 0–9 months. However, Oxobio1 and the Conventional bag type were also found to differ in tensile stress ( $p = < 0.01$ ). As samples in the open-air were too brittle to test after 9 months, the only significant difference between environments was between soil and control ( $p = < 0.01$ ). Furthermore, the specific order of tensile stress between the bags was largely unchanged throughout 27 months, whereas environment type seemed to have a greater effect (SI Table S4).

When comparing bag types (and ignoring any samples that had deteriorated to such an extent tensile stress could not be tested), Oxobio1 had the greatest loss in tensile stress over 27 months for all environments; soil (75% loss), marine (60%

loss), and control (29% loss). Conventional plastic had the least reduction in tensile stress for both soil (34% loss) and the marine environment (14% loss). Compostable plastic had the lowest change in tensile stress within the control environment (11% loss), but samples within open-air and marine environment showed total disintegration (SI Table S5).

Subtle changes in chemical composition were indicated by FT-IR analysis. Some samples developed a small poorly defined carbonyl stretch at a wavenumber of approximately  $1715\text{ cm}^{-1}$ ; this is indicative of oxidation which is a sign of deterioration and was more evident for samples exposed in the open-air. However, this varied between materials and environments, with no clear pattern being evident.

#### 4.0. DISCUSSION

Here we report the deterioration of several plastic carrier bag materials after exposure in the marine, soil, open-air, and control environment over a period of 3 years. All bags were obtained from mainstream retail shops and four of the materials were promoted as having some level of enhanced degradability or composability presumably in relation to conventional polyethylene. Apart from the compostable bag material deployed in the marine environment, fragments or whole samples of each bag material type were present in all environments after 27 months and some of the whole bag samples were still functional as plastic bags after 3 years in the natural environment.

Over a 27-month period, little change in the chemical structure of any of the samples was revealed. Additionally, some subtle, but statistically testable and significant, changes in tensile stress were apparent overtime, but the extent of these changes varied among materials and environments. The rates of degradation of plastics in different environments will strongly depend on the local conditions to which they are exposed.<sup>36</sup> Physical and chemical changes in polymers can be caused by environmental factors including light (photo-oxidation), heat (photothermal oxidation), mechanical abrasion, moisture, chemical conditions, or biological activity (fungi, bacteria, yeasts, algae, and their enzymes).<sup>24,37,38</sup> For example, on the compostable bag samples in the open-air environment solid deposits that looked like filamentous bacteria were visible on the surface of the material (SI Figure S2;2b).

The tensile stress of bags exposed to sunlight outdoors (labeled as open-air) decreased faster than in the other environments. Between 9–18 months all of the samples exposed in the open air had fragmented and could no longer hold their original shape because they were too brittle. The faster rate of fragmentation in air may be due to greater levels of ultraviolet (UV) radiation and oxygen, in combination with higher temperatures than in the other environments.<sup>39,40</sup> The amount of exposure to UV would be decreased if plastics are buried in soil, landfill, or submerged in the marine environment and this may explain the slower rates of deterioration observed in these conditions during our study.

Samples buried in soil were found to lose tensile stress significantly faster than samples in the control environment possibly because of increased moisture content in the soil. Understanding the degradation of different plastic types in terrestrial environments is important as substantial quantities of plastic will end up in landfills.<sup>41</sup> Further, in the absence of a specific waste management pathway, for example to a commercial composter, all of these materials will, unless

littered be sent as residual waste to landfill or incineration. When plastic accumulates within the soil, it becomes part of a complex mixture of organic matter and mineral substituents. It has been suggested that within this environment microplastics could negatively impact organisms including earthworms.<sup>42,43</sup>

Our research showed that all carrier bag materials tested appeared intact after they were buried in soil conditions after 27 months. However, more subtle changes were detectable with a 25–69% reduction in tensile stress between the different bag types. These results are perhaps more realistic than the previous studies due to being exposed for a longer time period and being exposed to naturally fluctuating soil moisture or air temperature.<sup>34,44</sup>

Samples exposed in soil and open-air were, overall, found to lose tensile stress significantly quicker than in the marine environment. However, there were no significant differences between the marine environment and control samples suggesting that deterioration in the marine environment was slow. Reduced deterioration in seawater has been observed previously. Rutkowska et al., (2002) exposed polyethylene (PE) for 20 months in 2 m water depth in the Baltic sea and reported that there was no biodegradation.<sup>45</sup> Pegram and Andrady (1989) studied the weathering of several plastics typically found in beach debris using floating marine exposure tests over a 6-month period. They measured the rate of deterioration from the changes in tensile elongation at break (and, in some instances, by the force to rupture) and found it to be much slower (2%) for samples exposed in the sea compared to samples exposed in open-air (95%).<sup>46</sup> In the current experiment, after 9 months, conventional polyethylene was found to lose 31% in tensile stress in open-air, but only 2% in the marine environment.

Colonisation by micro- and macro-marine organisms (a process described as fouling), occurs in natural environments and will vary according to conditions (e.g., temperature). This fouling process may affect plastic in a variety of ways.<sup>47</sup> First, the biofilm may “shield” the plastic from UV light<sup>34</sup> thus reducing the rate of photodegradation. Within the marine environment, fouling can also make plastics negatively buoyant causing buoyant items to sink;<sup>48</sup> hence further reducing irradiance. In the current experiment, all samples in the marine environment readily acquired a coating of biofilm.

All samples of the compostable bag (Compost), including the whole bag, completely deteriorated within a 3-month period in the marine environment. Similarly, research by O’Brine and Thompson (2010) also found that a compostable bag type had 100% surface area loss between 16 and 24 weeks when deployed in the marine environment. This suggests that deterioration of compostable bags can be relatively rapid in seawater. However, more work would be needed to establish what the breakdown products of this deterioration are, such as microplastics or nanoplastics, and to consider any potential environmental consequences.

From the perspective of the remaining bag types, it might have been expected that the two oxo-biodegradable materials would degrade faster than both the biodegradable and conventional bag types as these bags have pro-oxidants which are incorporated into the polymer chains to accelerate photo- and thermo-oxidation.<sup>44</sup> However, throughout the 27 months of this experiment, Oxobio1 was the only bag type to lose tensile stress significantly faster compared to biodegradable, conventional and Oxobio2 bag types.

Koutny et al., (2006) studied the biodegradability of high-density polyethylene film (HDPE) and low-density polyethylene film (LDPE) containing pro-oxidants and antioxidants. These were tested against microbial strains (Koutny et al., 2006; Larkin et al., 2005). After an abiotic pretreatment consisting of photooxidation and unnaturally high thermo-oxidation (60 °C) which was intended to mimic around 3 years of outdoor weathering, the samples were inoculated, incubated up to 200 days (27 °C) and their metabolic activities were followed. An initial phase of *fast* microbial growth was observed, and the authors suggest this was probably caused by utilization of low molecular extractable compounds. This was followed by a long period of stabilized metabolic activity. Analysis performed at the end of incubation indicated that any biodegradation had probably only affected the surface layer of the materials.

The current study showed that, oxo-biodegradable, degradable, and conventional carrier bag materials did not degrade quickly in any of the natural environments examined and, in some cases, formulations merely disintegrated into small pieces (such as those in the open-air environment). There are considerable concerns about the accumulation of microplastics in the environment and it remains to be established whether fragmentation into microplastics presents greater environmental risks than the original intact items of litter. From the perspective of cleansing, fragments are certainly considerably harder, if not impossible, to remove from the environment compared to intact items.

It is of importance to understand the actual environmental degradability performance of materials which are claimed to have enhanced degradation properties as these could make consumers more relaxed about discarding, or even littering them, rather than reusing and recycling. Due to the growing interest in products which indicate enhanced environmental outcomes, we should be careful that such products do not inadvertently encourage littering or compromise alternative approaches to waste reduction such as recycling.

Designing products specifically to degrade in the environment is very challenging because of the natural variability between environment types, as illustrated by the present study. In addition, formulations that are designed to be less durable may compromise recyclability since they decrease the durability of the recycle. It is also important to set the benefits of the various formulations into a wider context since reducing the diversity of polymers that are widely used is likely to facilitate greater recycling. To gain the maximum benefit from materials with enhanced rates of degradability, it is essential to have clear definitions and product labeling to indicate appropriate usage and disposal.<sup>7</sup>

If products are designed or marketed with the intent to make a valuable contribution in reducing the impacts of plastic litter in the natural environment then it is imperative to have appropriate standard tests against which to assess performance. These standards would need to incorporate the variability of natural environmental conditions (e.g., temperature/pH/light) and an appropriate time scale of deterioration such that it is clear items are deteriorating sufficiently rapidly to make a difference and not leave any potentially harmful degradation products (chemicals or fragments). In addition to appropriate standards and tests, the relevant receiving environment in which breakdown is expected to occur also needs to be stated.

Clearly there may be drivers for the design of products with modified degradability other than deterioration in the natural

environment, but in order for any these potential benefits to be realized it is essential that such products have a high probability of actually reaching the appropriate waste stream. This will require availability of a dedicated waste stream, the appropriate infrastructure such as an industrial composting facility and sufficient understanding among consumers to correctly separate their waste accordingly. Some nations actively promote the use of carrier bags with biodegradable, degradable, or compostable formulations, for example using fiscal measures or other legislation. This includes some nations with relatively poor waste management infrastructure where the likelihood of these products reaching an appropriate waste stream seems low. Given the findings of this study, the benefits of such policy measures are unclear.

In conclusion, the current experiment has shown that biodegradable, oxo-biodegradable and conventional plastic formulations persist and remain functional in the soil and the marine environment for over 3 years. The compostable bag was the only material that completely disappeared from the experimental test rig within the marine environment and did this within a 3-month period, but this product remained intact in soil. Hence the current study indicated that over a 3-year period, none of the materials examined could be relied upon to deteriorate sufficiently enough to reduce the negative effects of littering on biota or aesthetics across all three environments. Moreover, it was not clear that materials which claimed to have enhanced degradation consistently deteriorated faster than conventional polyethylene. Deterioration was influenced by the receiving environment, but this was not consistent among material types. Hence, we suggest that statements about the degradation of products should be clearly linked to appropriate standards, made in conjunction with statements on the receiving environment (air, soil, water) and time scale to which those claims relate. Since degradable and compostable materials are typically not compatible with widely available recycling infrastructure, it is also important that the users are informed of the appropriate disposal route which in most circumstances will be disposal to the residual waste stream. It is only by providing accurate, unambiguous, and complete guidance to the user regarding disposal that the potential benefits of these novel materials can be realized without the negative consequences that could result in inappropriate disposal as well as unintended environmental consequences. For many applications in which plastic carrier bags are used, perhaps durability in the form of a bag that *can* and *is* reused many times presents a better alternative to degradability.

## ■ ASSOCIATED CONTENT

### 📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b06984.

Additional information as noted in the text (PDF)

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### Notes

The authors declare no competing financial interest.

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