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

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17
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26

27 *Graphical Abstract:*



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

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34
35 *like to thank the*
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Centre (MBERC)
Electron
for their help
project.

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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 3 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.

Keywords:

Plastics, Carrier bags, Biodegradability, Waste Management

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1. Introduction

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

Approximately 50% of plastics are discarded after a single-use ^{3,4}. This creates a major waste management problem, with plastics accounting for approximately 8–10% of all the waste generated in the U.K. ^{3,5}. 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 ^{6–8}. 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 Leader’s declaration 2015 ^{9–11}.

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 ^{12,13}.

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Since their introduction in the 1970s, plastic carrier bags have become widespread in daily life worldwide ¹⁴. They are typically considered as single-use items and are commonly made from polyethylene ¹⁵. 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 ¹⁶.

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 ¹⁶. 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 ¹⁷. Several countries have already included bans or taxes, which have resulted in substantial reductions in use ¹⁸. However, there is no consistency between countries.

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 ^{19,20} and subtidal benthos ²¹. Even if properly discarded, lightweight bags can unintentionally be transferred away from landfill sites or other areas by wind or heavy rain ⁵.

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. 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

138 services they provide ²³. Additionally, Hodgson et al. (2018) used laboratory
139 experiments with carrier bags and showed that amphipods can shred plastic carrier
140 bags, generating numerous microplastic fragments.

141
142 The hydrophobicity and long carbon chain molecular structure of polyethylene, which
143 is widely used for plastic bags, makes it resistant to biodegradation under normal
144 conditions. The timeframe for the complete mineralisation is unknown, creating a
145 major waste management issue.

146
147 Awareness of the accumulation of end of life plastic and its impact on the environment
148 has, in part led, to interest in the development of degradable polymers. Biodegradable,
149 oxo-biodegradable and compostable plastics are often regarded as potential solutions
150 to the accumulation of plastic litter and waste. Some of these products are marketed
151 accompanied by statements indicating they can be '*recycled back into nature much*
152 *more quickly than ordinary plastic*' ²⁵ or '*plant-based alternatives to plastic*' ²⁶.

153
154 These materials are widely used for the production of carrier bags and some are also
155 used to make a variety of other items, including single-use cutlery, water bottles and
156 straws.

157
158 Biodegradation takes place through the action of enzymes and/or chemical
159 deterioration associated with living organisms, bacteria, fungi and algae. This occurs
160 in two steps; the first is the fragmentation of the polymers into sections of lower
161 molecular mass by means of either abiotic reactions (i.e. oxidation, photodegradation,
162 hydrolysis), or biotic reactions (i.e. degradation by microorganisms). This is followed
163 by bio-assimilation of the polymer fragments by microorganisms and its mineralisation
164 ²⁷.

165
166 A material may be labelled as 'biodegradable' if it conforms to certain national or
167 regional standards ^{28,29}. Such standards could include: ISO, European Norm – EN and
168 American Society for Testing and Materials (ASTM) International. Some standards are
169 appropriate for conditions that occur in an industrial composter, in which temperatures
170 are expected to reach 70 °C. Other standards, focus on laboratory-based
171 biodegradation tests using measurements of oxygen demand or CO₂ evolution; for

172 example, ISO 19679:2016 (2016) tests for the aerobic biodegradation of plastics at
173 the interface between seawater and sandy marine sediment. Oxo-biodegradable
174 plastics (oxo-plastics) are reported to contain an additive (pro-oxidant) which is
175 intended to break the molecular chain within the polymer which will then lead to its
176 biodegradation ³¹. However, there is typically no clearly defined timeframe given for
177 the breakdown of oxo-/biodegradable plastics ³².

178

179 In this context the term 'Composting' relates to enhanced biodegradation under
180 managed conditions, predominantly characterised by forced aeration and natural heat
181 production resulting from biological activity decomposing the material. The resulting
182 output material, *compost*, contains nutrients and can be used as a soil improver ³³.
183 Therefore, compostable plastics should biodegrade in a managed composting process
184 through the action of naturally occurring micro-organisms and typically do so in relation
185 to a specified timeframe ³². However, this can only occur if there is a specific waste
186 stream dedicated to compostable waste.

187

188 There is a lack of clear evidence that biodegradable, oxo-biodegradable and
189 compostable materials offer an environmental advantage over conventional plastics,
190 and the potential for fragmentation into microplastics causes additional concern ^{28,34}.
191 To date, studies focusing on the deterioration of different types of degradable plastics
192 in the environment give varying results and are shorter in timeframe.

193

194 The EU is proposing a process to restrict the use of oxo-plastics ³³, because of the
195 lack of consistent evidence about rates of deterioration in the environment, allegedly
196 misleading claims to consumers and risks that labelling products as biodegradable
197 may inadvertently promote littering behaviour.

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201

202 The present study describes the deterioration in different natural environments of
203 bags, which were stated to have biodegradable, oxo-biodegradable or compostable
204 properties. We do not specifically attempt to quantify biodegradation performance in
205 relation to any specific standard such as degradability in a commercial composting

206 facility. Rather we assess whether or not there has been any meaningful
207 deterioration in the context of reducing marine litter; for example, had the bag
208 remained intact or deteriorated into visible fragments? A conventional polyethylene
209 plastic carrier bag was also examined for comparison. All bags were available at the
210 point of sale in U.K. high-street retailers. These materials were exposed in various
211 environments that discarded carrier bags could encounter; in open-air, buried in soil
212 and submersed in the marine environment. This is the first research where plastic
213 deterioration has been examined simultaneously across these three natural
214 environments, together with controlled conditions in the laboratory. Five different
215 plastic carrier bag formulations were considered, and their deterioration was
216 evaluated over a 3-year period. Deterioration was considered in terms of visible loss
217 in surface area, as well as approaches to detect more subtle changes in tensile
218 stress, surface texture and chemical structure.

219

220 **2. Methodology**

221

222 **2.1 Sample Preparation**

223

224 Five different types of plastic carrier bag were compared (Table 1): these included
225 two types of oxo-biodegradable bag (labelled here as Oxobio1 and Oxiobio2), one
226 biodegradable bag, one compostable bag, and a high-density polyethylene (HDPE)
227 carrier bag (labelled in this research as a conventional carrier bag), which was not
228 stated to have any particular deterioration/compostable properties. Deterioration in
229 this study is used to describe the process of a becoming a lower quality or condition.

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Designated label for testing	Degradation properties (as stated on bag)	Information stated on websites linked to the product	Disposal/anti-littering information (as stated on bag)
Oxobio1	Degradable Plastics (D ₂ W trademark, logo)	Oxo-biodegradable (https://www.symphonyenvironmental.com/d2w/)	No information
Oxobio2	Planet safe plastic; incorporating EPI's totally degradable plastic additives (EPI trademark, logo)	Oxo-biodegradable (http://www.epi-global.com/en)	No information
Biodegradable	Biodegradable bag (exo plastics logo, sustainable bioplastic; Biodegradable ISO 14855)	No claims about biodegradability on exo plastics website (https://www.exoplastics.com/) ISO 14855 is an international standard covering aerobic biodegradability of plastic materials under controlled composting conditions	Recyclable (no numerical category for recycling type stated)
Compostable	Completely compostable, recycle me with food	Plant-based compostable foodservice packaging (https://www.vegware.com/about/info_1.html) Compostable packaging is designed to be recycled together with food waste. https://www.vegware.com/close-the-loop/info_50.html EN13432 is the packaging waste directive and standards for compostability http://www.bpf.co.uk/topics/standards_for_compostability.aspx	Reuse me first for shopping, and then as a food waste caddy liner! This completely compostable bag complies with standard EN13432. Suitable for industrial food waste recycling – visit www.foodwastenetwork.org.uk . Recycling category '7 - other'
Conventional	High Density Polyethylene; No degradation properties stated	No manufacturer given	Reuse at home or recycle. Recycling category '2 – HDPE'

235

236

237 **Table 1.** *Information on the tested carrier bags and the properties as stated on the*
238 *manufacturer's website. All bags were opaque and obtained based on their*
239 *prevalence in retail stores in and around Plymouth, U.K.*

240

241

242 The bags were chosen as they were all opaque and were obtained based on their
243 prevalence in retail stores in and around Plymouth, U.K. Sixteen samples of each
244 bag were obtained. In order to obtain a representative sample of each bag type, a
245 maximum of two bags were sourced from any one store on any single occasion.
246 Where repeat visits to the same store were necessary to obtain sufficient
247 independent samples, these visits were separated by at least 2 weeks. Hence our
248 experiment was designed to contain a range of products and production batches so
249 as to be as representative as possible. Since the specific retail stores from which the

250 carrier bags were obtained is not of particular relevance, bags will only be described
251 based on their formulation (Table 1).

252

253 Each carrier bag type was cut into strips; 15 x 25 mm. The strip samples were taken
254 from the main body of the carrier bag (not the handles or the sides), to provide areas
255 of similar structure. A strip of each plastic carrier bag type was then placed into a
256 pouch made of high-density polyethylene (HDPE) mesh and sewn secure using
257 nylon fishing twine. Each pouch structure (150 x 200 mm) was sewn together to
258 provide 5 equally spaced separated compartments. These compartments were then
259 used to house an individual strip of each bag type (Fig.S1). The HDPE mesh (1 mm
260 x 1 mm) allowed exposure to external environments and each compartment was
261 sewn so as to allow the bag samples to move relatively freely. Each pouch structure
262 was attached to a permanent panel to aid removal.

263

264 These permanent panels were placed in one of four different conditions; buried in
265 soil, exposed outdoors in the air, submerged in the marine environment and placed
266 in a blacked-out box in the laboratory as a control. The buried samples were situated
267 at the University of Plymouth's Skardon Garden (50°22'38.4"N, -4°08'11.9"W) and
268 were buried to a depth of approximately 0.25 m (Fig.S1a). The samples that were
269 exposed in open-air were also situated in Skardon Garden and were placed on a
270 south facing wall (Fig.S1b). Samples placed in the marine environment were
271 submerged on a beam at Queen Anne's Battery Marina (50°36'48.4"N, -
272 4°12'96.5"W) at a depth of approximately 1 m (Fig.S1c). 3 kg weights were
273 connected on each side of the beam to maintain depth. Control samples were placed
274 in a blacked-out box (kept at room temperature) in a laboratory at the University of
275 Plymouth.

276

277 All samples were deployed on the 10th July 2015. There were 3 subsequent
278 sampling dates; 6th April 2016 (9 months), 6th Jan 2017 (18 months), 6th October
279 2017 (27 months). Additionally, whole bags of each material were also deployed in
280 polypropylene mesh in each environment at the same time and used for visual
281 inspection over the 3-year period (23rd August 2018).

282

283 Over this period, the samples would have been exposed to sea (8.8 °C - 18.8 °C;
284 United Kingdom Sea Temperatures, 2019) and air (1.5 °C - 21.5 °C; Met Office,
285 2016) temperatures, typical of those in a temperate environment. The soil type in the
286 South West of the U.K. is freely draining and slightly acidic ³⁵.

287

288 Before deployment, 4 subsample strips from each carrier bag type were tested to
289 provide a comparison starting point. After deployment, four replicate samples of
290 strips from differing bag replicates were collected from each environment on each
291 sampling date. Samples were removed from the mesh structure, gently cleaned
292 using distilled water, air dried (30 °C) and tested (see below) within 48 hours.

293

294

295 2.2 Visual Inspection

296

297

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

302

303 Measurements of tensile stress and molecular structure using Fourier transform
304 infra-red spectroscopy (FTIR) were made in order to detect any more subtle
305 changes.

306

307

308 2.3 Tensile Stress Testing

309

310 The thickness of each strip was measured using an electronic micrometer (Sealey;
311 AK9635D). Each strip was measured at 10,50,100 and 140 mm from a central point.
312 This produced 4 reference points for each sample and the mean was then
313 calculated. The maximum load (N) for each strip was then measured using a tensile
314 testing machine at a rate of 100 mm min⁻¹ (Instron, system ID 3345 k1669 - USA,
315 force transducer model 2519-104, capacity 500 N). Then, the maximum tensile
316 stress of each strip was calculated using the following equations:

317

318

$$\text{i) } A = bh$$

$$\text{ii) } \sigma = \frac{F}{A}$$

319

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

325

326 Normality of the data was confirmed by using QQ plots to examine distribution. One-
327 way analysis of variance (ANOVA) was used to compare the maximum tensile stress
328 difference between the different bag types before being exposed in any environment.
329 The effects of bag type, environment and time on the maximum tensile stress was
330 then examined. This was compared using the percentage change of tensile stress
331 from 0 to 9 months and 9 to 27 months using a three-way ANOVA; the three factors
332 were (bag type, environment, time). Time had two levels (0-9 and 9-27 months), bag
333 type had 5 levels (Oxobio1, Oxobio2, biodegradable, compostable and conventional)
334 and environment consisted of 4 levels (control, open-air, marine, soil). Post-hoc
335 Tukey tests were then used to identify the significant effects. Any samples which
336 were too brittle to test or were no longer visible were omitted from the analysis. All
337 statistical tests were performed in R ver. 3.4.1 (R Core Team 2017).

338

339 2.4 Molecular Composition Analysis (FTIR) and Image Analysis

340

341 In order to assess any subtle deterioration effects on the molecular composition of
342 the materials, samples were analysed by FTIR microscopy in transmission mode
343 with a Hyperion 1000 microscope coupled to a Vertex 70 spectrometer (Bruker). For
344 each sample, the spectra was recorded with 32 scans in the region of 4000 to 600
345 cm. Prior to FTIR, samples were cleaned with absolute ethanol to remove any
346 residues. The spectra obtained were compared against a spectral database of
347 synthetic polymers (BPAD polymer & synthetic fibres ATR).

348

349

350 **3. Results**

351

352 Prior to exposure in different environments, the maximum tensile stress and
353 thickness of the bags were measured. Oxobio2 had the highest tensile stress and
354 thickness (28.82 ± 1.55 MPa and 0.04 mm), the compostable bag had the lowest
355 tensile stress (10.47 ± 1.23 MPa) and the biodegradable, conventional and Oxobio1
356 bag had the lowest thickness (0.02 mm) (Table S1). All bag types had relatively
357 consistent thickness.

358

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

364

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

367

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

373

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

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

390

391 After 3 years, photographs were taken of the whole bags from both the soil and
392 marine environment (Fig. 1). As a qualitative assessment of functionality, the bags
393 were loaded with typical groceries from a local supermarket (weight 2.25 kg).

394 Oxobio1, Oxobio2, biodegradable and conventional were still functional and retained
395 the items with no breakages. However, the compostable bag type (which was only
396 present in the soil environment for 27 months) was unable to hold any weight without
397 tearing.

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

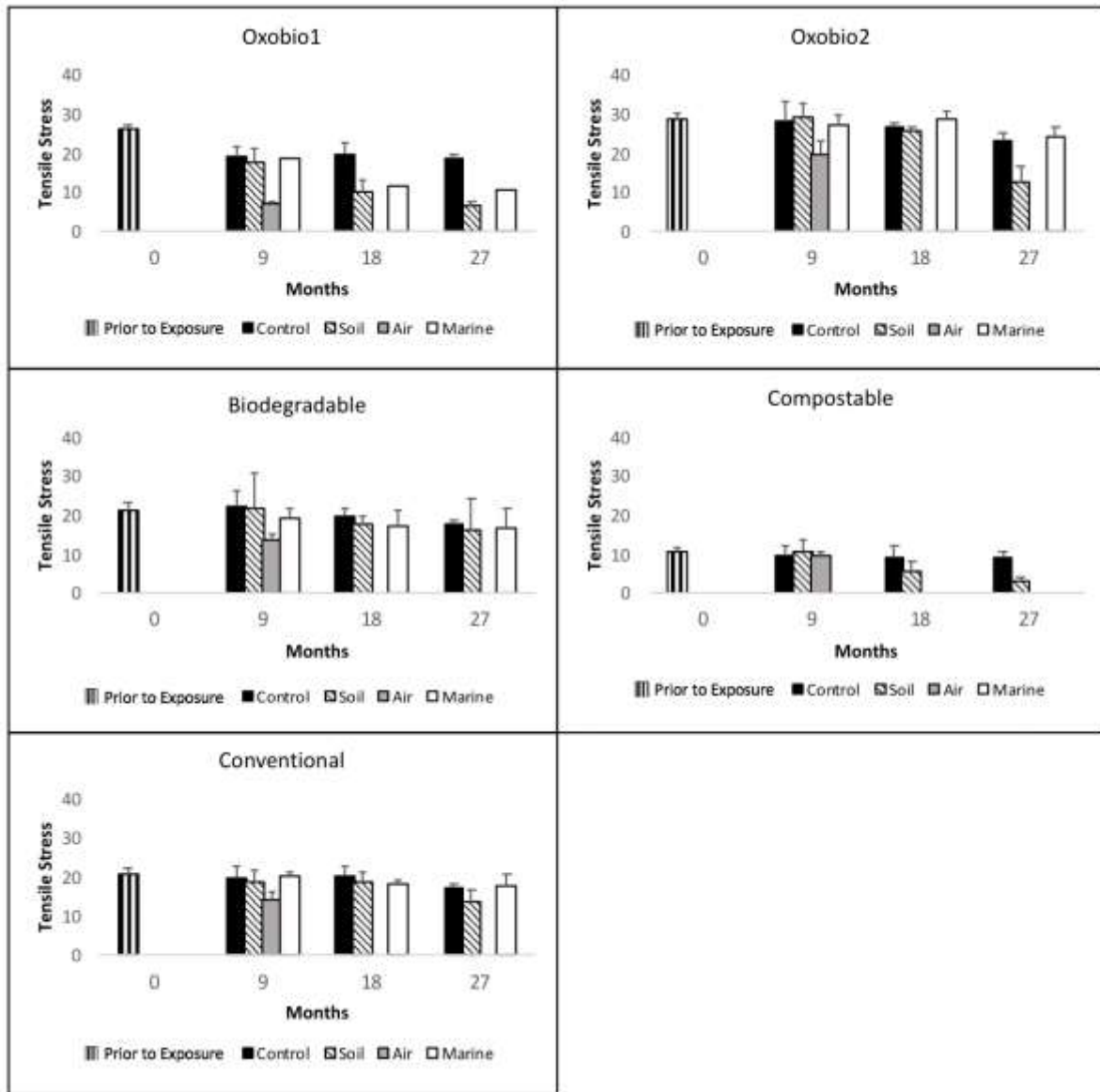
410

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

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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 4 different environments (control, marine, soil, open-air). Open-air is labelled 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.

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

436

437 From the perspective of litter and potential interactions with biota, most of the bag
438 samples remained intact. However, subtle changes in tensile stress were apparent in
439 all the bag materials indicating some degree of deterioration; the factors time, bag
440 type and environment showed significant differences between 0 - 9 months exposure
441 (Table S3). Post hoc comparisons found that Oxobio1 lost strength at a significantly
442 faster rate than the other bags between 0 - 9 months ($p = <0.01$). There were also
443 differences between the two Oxo-biodegradable samples; Oxobio1 lost strength
444 significantly faster than Oxobio2 ($p = 0.01$). Additionally, bags exposed in the open-
445 air environment lost strength more rapidly when compared to the other
446 environments: control ($p = <0.01$), marine ($p = <0.01$) and soil ($p = <0.01$).

447

448 A second ANOVA was conducted which included the compostable bag type. This
449 bag type needed a separate analysis as all its samples had completely disappeared
450 within the marine environment after 3 months. This ANOVA showed that the
451 compostable bag material had a significant difference in tensile stress to Oxobio1 (p
452 = <0.01). The compostable bag material exposed in the open-air environment also
453 lost its strength more rapidly when compared to bags exposed to both control ($p =$
454 <0.01) and soil ($p = <0.01$) environments.

455

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

463

464 When comparing bag types (and ignoring any samples that had deteriorated to such
465 an extent tensile stress could not be tested), Oxobio1 had the greatest loss in tensile
466 stress over 27 months for all environments; soil (75% loss), marine (60% loss) and
467 control (29% loss). Conventional plastic had the least reduction in tensile stress for
468 both soil (34% loss) and the marine environment (14% loss). Compostable plastic
469 had the lowest change in tensile stress within the control environment (11% loss),
470 but samples within open-air and marine environment showed total disintegration
471 (Table S5).

472

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

478

479 4.0 Discussion

480

481 Here we report the deterioration of several plastic carrier bag materials after
482 exposure in the marine, soil, open-air and control environment over a period of 3
483 years. All bags were obtained from mainstream retail shops and 4 of the materials
484 were promoted as having some level of enhanced degradability or composability
485 presumably in relation to conventional polyethylene. Apart from the compostable bag
486 material deployed in the marine environment, fragments or whole samples of each
487 bag material type were present in all environments after 27 months and some of the
488 whole bag samples were still functional as plastic bags after 3 years in the natural
489 environment.

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

508

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

518

519 Samples buried in soil were found to lose tensile stress significantly faster than
520 samples in the control environment possibly because of increased moisture content
521 in the soil. Understanding the degradation of different plastic types in terrestrial
522 environments is important as substantial quantities of plastic will end up in landfills
523 ⁴¹. Further, in the absence of a specific waste management pathway, for example to
524 a commercial composter, all of these materials will, unless littered be sent as
525 residual waste to landfill or incineration. When plastic accumulates within the soil, it
526 becomes part of a complex mixture of organic matter and mineral substituents. It has
527 been suggested that within this environment microplastics could negatively impact
528 organisms including earthworms ^{42,43}.

529

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

536

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

551

552 Colonisation by micro- and macro-marine organisms (a process described as
553 fouling), occurs in natural environments and will vary according to conditions (e.g.
554 temperature). This fouling process may affect plastic in a variety of ways ⁴⁷. Firstly,
555 the biofilm may 'shield' the plastic from UV light ³⁴ thus reducing the rate of photo-
556 degradation. Within the marine environment, fouling can also make plastics
557 negatively buoyant causing buoyant items to sink ⁴⁸; hence further reducing
558 irradiance. In the current experiment, all samples in the marine environment readily
559 acquired a coating of biofilm.

560

561 All samples of the compostable bag (Compost), including the whole bag, completely
562 deteriorated within a 3-month period in the marine environment. Similarly, research
563 by O'Brine and Thompson (2010) also found that a compostable bag type had 100%

564 surface area loss between 16 and 24 weeks when deployed in the marine
565 environment. This suggests that deterioration of compostable bags can be relatively
566 rapid in seawater. However, more work would be needed to establish what the
567 breakdown products of this deterioration are, such as microplastics or nanoplastics,
568 and to consider any potential environmental consequences.

569

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

577

578 Koutny et al., (2006) studied the biodegradability of high-density polyethylene film
579 (HDPE) and low-density polyethylene film (LDPE) containing pro-oxidants and
580 antioxidants. These were tested against microbial strains (Koutny et al., 2006; Larkin
581 et al., 2005). After an abiotic pre-treatment consisting of photooxidation and
582 unnaturally high thermo-oxidation (60 °C) which was intended to mimic around 3
583 years of outdoor weathering, the samples were inoculated, incubated up to 200 days
584 (27 °C) and their metabolic activities were followed. An initial phase of *fast* microbial
585 growth was observed, and the authors suggest this was probably caused by
586 utilization of low molecular extractable compounds. This was followed by a long
587 period of stabilized metabolic activity. Analysis performed at the end of incubation
588 indicated that any biodegradation had probably only affected the surface layer of the
589 materials.

590

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

598 fragments are certainly considerably harder, if not impossible, to remove from the
599 environment compared to intact items.

600

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

608

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

618

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

628

629 Clearly there may be drivers for the design of products with modified degradability
630 other than deterioration in the natural environment, but in order for any these
631 potential benefits to be realised it is essential that such products have a high

632 probability of actually reaching the appropriate waste stream. This will require
633 availability of a dedicated waste stream, the appropriate infrastructure such as an
634 industrial composting facility and sufficient understanding amongst consumers to
635 correctly separate their waste accordingly. Some nations actively promote the use of
636 carrier bags with biodegradable, degradable or compostable formulations, for
637 example using fiscal measures or other legislation. This includes some nations with
638 relatively poor waste management infrastructure where the likelihood of these
639 products reaching an appropriate waste stream seems low. Given the findings of this
640 study, the benefits of such policy measures are unclear.

641

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

666

667

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