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RESEARCH ARTICLE

REVISED Lab-scale and full-scale industrial composting of biodegradable plastic blends for packaging

[version 3; peer review: 2 approved]

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Abstract

Background

The acceptance of compostable plastic packaging in industrial composting plants is not universal despite available certification due to the persistence of plastic residues after composting. To better understand this discrepancy, this study compared the disintegration rates of two blends designed for rigid packaging (polylactic acid based) and soft packaging (polybutylene succinate based) in lab-scale composting tests and in an industrial composting plant.

Methods

A lab-scale composting test was conducted in triplicates according to ISO 20200 for 4, 8 and 12 weeks to check the disintegration potential of the blends. Duplicate test material were then exposed in the compost pile of an industrial composting plant for a duration of 3 weeks and compared with a supplementary lab-scale test of the same duration.

Results

The rigid packaging samples (1 mm thickness) retained on average 76.4%, 59.0% and 55.7% of its mass after 4, 8 and 12 weeks respectively in the lab-scale. In the plant, the average remaining mass was 98.3%, much higher compared to the average of 68.9% after 3

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weeks in the supplementary lab-scale test. The soft packaging samples (109±9 µm sample thickness) retained on average 45.4%, 10.9% and 0.3% of its mass after 4, 8 and 12 weeks respectively in the lab-scale. In the plant, a high average remaining mass was also observed (93.9%). The supplementary lab-scale test showed similar remaining mass but higher fragmentation after 3 weeks.

Conclusions

The results show that the samples achieved significant disintegration in the lab-scale but not in the plant. The difference between the tests that might further contribute to the differing degradation rates is the composition and heterogeneity of the composting substrate. Therefore, the substrate composition and thermophilic composting duration of individual plants are important considerations to determine the suitability of treating compostable plastic in real-world conditions.

Plain language summary

There is resistance from industrial composting plants to the treatment of compostable biodegradable plastics. This study aims to compare the disintegration rates of two new biodegradable plastic blends developed for rigid and soft packaging applications in controlled industrial composting conditions in the lab as well as under actual industrial composting conditions in a composting plant. Results show significant disintegration of the material in the lab-scale up to 12 weeks. However, there were notable differences in the degree of disintegration of the samples in the lab compared to real-world conditions after 3 weeks. The 1 mm thick polylactic acid-based blend for rigid packaging experienced much higher disintegration in the lab with 68.9% remaining mass after 3 weeks compared to 97.2% remaining mass in the industrial composting plant. The 109 µm thick polybutylene succinate-based blend for soft packaging had similar remaining masses comparing the lab and full-scale tests. However, the lab-test showed higher fragmentation after 3 weeks. The characteristics of the organic waste inputs are potential causes of the lower disintegration rates. The study highlights the need to better understand the real-world industrial composting conditions and their variations when evaluating composting as a treatment method for biodegradable plastics.

Keywords

Biodegradable plastic, compostable plastic, compostable packaging, industrial composting, polylactic acid, polybutylene succinate

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REVISED **Amendments from Version 2**

The revisions made in this version dealt mainly with the reformulation of sentences to improve clarity. Most of the improvements of the text were made in the introduction section. In addition, the term "on-field" was removed and replace with "full-scale" throughout the manuscript. The aim was to clarify that the "full-scale" experiment was done in a full-scale industrial composting facility and not "on-field", i.e. on agricultural land. Figure 3 was also updated by combining the individual data points of the full-scale tests into an average value with standard deviation to improve clarity. The text in the abstract, results as well as conclusion was updated to reflect this change. However, the main conclusions were not affected. The order of the superscript numbering in Table 1 and Table 2 was also altered to improve readability.

Any further responses from the reviewers can be found at the end of the article

Introduction

The prevalence of plastic waste and micro-plastic in the environment has spurred research and development of biodegradable plastics as alternatives to conventional fossil-based non-biodegradable plastics. Biodegradable plastics include polymers that can be broken down and mineralized by microbial action. Commonly available bio-based biodegradable plastics include polylactic acid (PLA) and polybutylene succinate (PBS) (IfBB, 2020). They can be produced from both fossil-based and bio-based resources. It is important to note the difference between the terms biodegradation and disintegration. In EN 13432, biodegradability is to be proved via the decomposition of the polymer structure and the subsequent release of the carbon in the plastic material as carbon dioxide. Disintegration on the other hand, refers to the fragmentation into very small pieces, i.e. measured by the loss of mass above a size threshold, > 2 mm in the case of EN 13432. Compostable plastics are a subcategory of biodegradable plastics that disintegrate significantly in specific composting conditions, but not necessarily in general environmental conditions on land or sea. Thus, they need to be kept in controlled closed systems.

Industrial compostable products are produced from biodegradable material and designed to disintegrate within a reasonable period under industrial composting conditions. The major characteristic of industrial composting is the ability to achieve thermophilic temperatures (Barrena *et al.*, 2014; Sundberg *et al.*, 2004), in the range from 55°C to 75°C. Composting might be an alternative for the treatment of plastic waste, for example when mechanical recycling is difficult, such as in the case of multilayer packaging (de Mello Soares *et al.*, 2022; Trinh *et al.*, 2021). However, industrial composting primarily aims to treat and stabilize organic waste and produce compost, an agricultural substrate. Thus, the feasibility of the treatment of biodegradable plastics within these plants is not a given.

For instance, there is no widespread acceptance and treatment of compostable plastics in industrial composting plants in Germany (ANS e.V. (ANS) *et al.*, 2019). One of the major issues quoted is the insufficient decomposition of biodegradable plastic products within the normal operating conditions of the plants. This occurs even with products certified as industrially compostable via international standards such as EN 13432. The reason is the wide range of different conditions in which industrial composting operators run their plants, which are sometimes quite distinct from the compostability certification conditions (Di Bartolo *et al.*, 2021; Hann *et al.*, 2020). The disintegration condition in EN 13432 requires that the product disintegrates under industrial composting conditions and leaves behind less than 10% of the material (> 2 mm) by mass after a maximum duration of 12 weeks in a "controlled pilot-scale test" or in industrial composting plants. The industrial composting plants in Germany on the other hand, have an active composting duration usually within a range of 4 to 8 weeks (Stadtreinigung Hamburg, 2019). In addition, there are differences in the composition of the input material fed into the composting process. Some facilities focus on garden waste while others are hybrid biogas and composting plants that predominantly run aerobic composting on the digestate from the anaerobic digestion process.

In literature, the behavior of biodegradable plastics in the composting process has been studied in both lab- and pilot scales. However, the duration and conditions of the composting process as well as the design and composition of the plastic vary. Ruggero *et al.*, 2021 simulated an industrial composting process in the lab with 20 days of thermophilic composting (58°C) and 40 days of compost maturation (37°C) with mature compost as the composting medium. They tested a starch and polybutylene adipate terephthalate (PBAT) composite film (50 µm thick), a PBAT based film (90 µm thick) and PLA pressed plate (500 µm). It was found that the starch and PBAT film underwent the highest mass loss (45%), whilst the PBAT film and PLA plate underwent low mass losses, of 8% and 3% respectively. The authors attributed the low mass loss of PLA plates to the short thermophilic phase as well as the higher thickness compared to other studies. Other studies focused on the design factors affecting the speed and degree of degradation. For example, microfibrillated cellulose was found to increase the degradation of PLA composites while cellulose nanocrystals retard degradation (Manzano *et al.*, 2021). The material thickness was also found to correlate with the disintegration speed of PLA and PBS blends (Tolga *et al.*, 2020). In the same study, the disintegration rate of a 7:3 (w/w) PLA:PBS blend was in a range of 25% to 35% after 12 weeks, depending on the thickness from 1 mm to 2 mm. Chalk was found to promote degradation possibly through facilitating water penetration while talc had the opposite effect. Researchers also studied the effect of treating compostable plastics in composting plants on the composting process itself. Cucina *et al.*, 2021 researched the effects of the high loading of commercially available compostable plastic products in organic waste (10 wt%) on compost quality. In the mentioned study, certified compostable starch-based shopping bags and PLA-based cutlery degraded by 48 wt% and 15 wt% respectively after a combination of a pilot-scale mesophilic dry

anaerobic digestion phase (35 days), active composting phase (15 days) and compost maturation phase (40 days). A final concentration of compostable plastic in compost was found to be around 18 wt%. Gadaleta *et al.* in turn found no effect of a 2% loading of cellulose acetate films on lab-scale anaerobic digestion and composting processes but noted also the presence of non-degraded material at the end of the process (Gadaleta *et al.*, 2022).

In a field study, 0.3 mm PLA rigid films were exposed in static compost piles comprising garden and food waste at a temperature range of 52°C to 59°C (Sikorska *et al.*, 2015). After 21 days, a decrease in the average molecular weight of the material from 105000 (Da) to 55000 (Da) was observed without significant mass loss. Another study exposed commercial PLA bottles to a mixture of cow manure, feed and wood shavings in a compost pile at 65°C (Kale *et al.*, 2007). Complete disintegration within 30 days was observed visually. In addition, the molecular weight dropped from an initial 230000 Da to below 50000 Da within 10 days. Similarly, a composting test in a static aerated compost pile for PLA/PHA mulch films reported high disintegration, with a 92% loss of surface area after two weeks (Sintim *et al.*, 2019).

Overall, the availability of published papers about full-scale composting tests in industrial plants is limited with varying outcomes. A study exposed various commercially available products made from compostable plastics to source separated municipal organic waste in an industrial composting plant for a total of 22 days with a temperature range of 30°C to 70°C throughout the run (van der Zee & Molenveld, 2020). A wide range of disintegration from 6% to 100% was obtained for different types of biodegradable polymers in different forms with a size cutoff of 2 mm. For PLA-based rigid materials, duplicate samples of plant pot cuttings achieved almost 100% disintegration, while the duplicate coffee capsule samples had a disintegration of 9% and 41% respectively. A research group exposed PLA/PBAT blend films from 0.02 mm to 0.1 mm thickness in "KNEER" container composting systems with an average temperature of 60°C (Musioł *et al.*, 2018). Though not explicitly measured, it was observed that significant disintegration only started after 21 days of incubation. In another study, certified compostable PLA tableware exposed to in-vessel composting using a mixture of bio-solids, food waste and woody material for an active phase of up to 14 days at 60°C (with recirculation of sample pieces >2 cm) and further curing for around 4 months (Zhang *et al.*, 2017). Complete disintegration was reported for all samples with a size cut-off of 3.2 mm.

Chemically, the disintegration of PLA in industrial composting conditions can be understood mainly through two mechanisms: the abiotic hydrolysis of the polymer chains in the presence of water and heightened temperatures resulting in shorter chains as well as microbiological enzymatic attack on the shorter chains (Karamanlioglu *et al.*, 2017). Above the glass transition temperature, higher chain mobility facilitates chemical and biological degradation (Karamanlioglu & Robson, 2013). Hydrolysis of PLA in aqueous media is accelerated with increased temperature, especially above the glass transition temperature reported around 57°C to 61°C (Rodriguez *et al.*, 2016). In addition, the mass loss trend with respect to composting time was reported to follow an S-shape curve, with an incubation phase at the beginning in which a rapid decrease in molecular weight was observed with no mass losses followed by a rapid decrease in mass after the incubation period. A lag phase was also observed for biodegradable plastics in some studies at the start of the composting process (Anstey *et al.*, 2014; Zhao *et al.*, 2005). This trend was also observed in industrial composting conditions for PLA/PBS blends (Tolga *et al.*, 2020).

Both acidic and basic pH can catalyze the abiotic hydrolysis of polyesters (Woodard & Grunlan, 2018). There are studies that reported that basic media (pH 12 to >13) facilitated the surface erosion of polylactic acid (von Burkersroda *et al.*, 2002) and the weight loss of poly(butylene adipate-co-butylene furandicarboxylate)s (PBAFs) and poly(butylene succinate-co-butylene furandicarboxylate)s (PBSF) (Peng *et al.*, 2017). Although there are various studies reporting the ability of microorganisms to biodegrade PLA (Al Hosni *et al.*, 2019; Kawai, 2010), there is still a debate on the effect of microorganisms compared to abiotic factors in terms of macro-scale material disintegration. PBS is also susceptible to both abiotic hydrolytic degradation (Lindström *et al.*, 2004) and enzymatic degradation (Xu & Guo, 2010).

Due to the push back from some industrial composting plants on the composting of certified compostable plastics as well as mixed results from full-scale tests reported in scientific literature, there is still a need to better understand the factors affecting disintegration rates in actual industrial composting conditions (full-scale) as well as potential differences in disintegration compared to controlled lab-scale tests. This work thus assessed the degradation rate, measured by mass loss, of two biodegradable plastic prototypes developed for rigid (PLA-based) and soft packaging (PBS-based) respectively within the BIO-PLASTICS EUROPE research project in both lab-scale simulated (DIN EN ISO 20200) and actual industrial composting conditions. The controlled, lab-scale test represented the disintegration potential of the prototypes and was conducted for a duration up to 12 weeks, the maximum allowed composting duration based on EN 13432. On the other hand, the test in the composting plant represented a real-life scenario. The composting medium of both cases was characterized for comparison. The results outlined the difference of disintegration rates in the lab and in a plant and explored the possible reasons for the differences.

Methods

Biodegradable plastic test samples

The biodegradable plastic materials tested were: (I) a PLA-based biodegradable plastic blend (BPE-RP-PLA) developed for rigid packaging and (II) a PBS-based biodegradable plastic blend (BPE-SP-PBS) developed for soft packaging. Table 1 lists the main characteristics of each blend and the sample thicknessused in this study.

Table 1. The main characteristics of the biodegradable plastic blends tested.

Blend code	BPE-RP-PLA	BPE-SP-PBS
Target application	Rigid packaging	Soft packaging
Base polymer	PLA	PBS
Main Filler	Calcium Silicate	Talc
Ash content	28.7%	9.7%
Sample thickness	1 mm plates	109±9 µm films[1]
Specific surface area	14 cm^2/g	130±11 cm^2/g[1]
Glass transition temperature (T_g)	59°C[2]	Around -30°C[3]
Mass average molecular weight	237000 g/mol[2]	N/A

[1] ± denotes standard deviation, relevant due to the variation in thickness of the prototype films; [2] Data from the producer; [3] Typical range for PBS from (Su *et al.*, 2019; Wang *et al.*, 2019);

The blends were developed based on general product requirements for rigid and soft packaging within the H2020 Research Project BIO-PLASTICS EUROPE (BPE). In addition, the prototypes (sheets and films) were also produced within the project. The main filler in BPE-RP-PLA is calcium silicate with an overall ash content of 28.7%. Injection molded plates of 1 mm thickness were used to represent rigid packaging. On the other hand, BPE-SP-PBS was tested in the form of films, with a thickness of 109 µm. The main filler is talc and the blend has an ash content of 9.7%. The thickness tested represents an approximation of the thickness that the final products of the target application would have.

Lab-scale industrial composting

The lab-scale test served as a baseline test for ideal industrial composting conditions as well as to check the disintegration potential of the blends. The lab-scale test was carried out based on ISO 20200, which simulates the thermophilic industrial composting conditions. The composting medium was synthetic bio-waste composed of 40% sawdust, 30% rabbit feed, 10% fresh compost, 5% sucrose, 4% corn seed oil and 1% urea by dry mass. Brief descriptions of the material sources are given in *Extended data* (Chong *et al.*, 2022b). Fresh compost was taken from the Bützberg Biogas and Composting plant as inoculum. The water content was adjusted to 55 wt% at the start of the experiment.

For each reactor system, 1.1 kg of composting medium was mixed with around 10 g of plastic sample (~1 wt% sample loading) in polypropylene boxes with lids (dimensions 34 cm X 20 cm X 12.5 cm). The initial mass of the dry plastic samples was weighed and recorded before adding to the reactor. Each box had two holes of 5 mm diameter on the sides for aeration. For BPE-RP-PLA, samples cut into 2.5 cm X 2.5 cm pieces were used following ISO 20200. For BPE-SP-PBS films, individual pieces cut into 5 cm X 5 cm were used instead to make them more manageable. The reactor systems were placed in a convection oven maintained at a temperature of 58°C throughout the experiment, simulating the thermophilic composting phase.

The reactor contents were weighed and the moisture content was replenished based on the schedule defined in ISO 20200 to ensure sufficient moisture content. The contents were gently mixed in specific intervals according the schedule. The mass loss of the plastic samples was measured after 4, 8 and 12 weeks. Triplicate reactor systems were set up for each time point. In addition, 3 reactor systems without the plastic samples were set up as control. Throughout the test, it was ensured that >80% of the sample surface was covered by the substrate medium. For the film prototypes with a high surface area, a 100% coverage was not practically possible at the beginning of the disintegration process. However, the mixing step ensured that in time, all sample surfaces had contact with the composting substrate.

At the end of the defined composting duration, the process was terminated by drying the reactor contents in the oven after removing the lid until constant weight was achieved. The mass before and after drying was used to estimate the final moisture content. The remaining plastic samples were recovered by sieving the reactor contents with a 2 mm mesh analytical sieve (RETSCH GmbH, Haan, Germany). The sample pieces were cleaned gently under running water and oven-dried at 58°C before final weighing. The initial and final weight was used to calculate the remaining mass using Equation 1. The remaining dried compost medium was then milled into powder and further analyzed for pH, C/N ratio and volatile organic solids content using standard methods. Further details of the materials and methodologies used are described in *Extended data* (Chong *et al.*, 2022b).

$$Remaining\ mass\,(\%) = \frac{Mass_{final} > 2\,mm}{Mass_{initial}} \times 100\% \qquad \text{Equation 1}$$

To facilitate direct comparison with the full-scale test in an actual plant, a second lab-scale tests based on ISO 20200 was conducted using an exposure duration of three weeks as well as identical sample dimensions used for the full-scale test. This test was done to check the potential difference in

disintegration rates between the lab-scale conditions (temperature and substrate mix) compared to the full-scale test.

Full-scale industrial composting test

To study the behavior of biodegradable plastic samples in real-world conditions, the samples were subjected to industrial composting in the Bützberg Biogas and Composting Plant located in Tangstedt, Germany. The plant receives separately collected bio-waste from households in the region. Before undergoing biological treatment, the waste was shredded (< 8 cm), sieved and sent through magnetic separation to remove impurities such as plastics and metals. The plant then ran a dry anaerobic digestion process on the pre-treated organic waste for biogas production followed by in-vessel composting of the digestate material. Hybrid fermentation chambers 24 m x 5 m x 4.5 m were used for the anaerobic digestion phase. For this study, the composting phase was carried out in the hybrid fermenters with aeration after the anaerobic digestion phase. The industrial composting outputs were finally sieved to produce compost for sale.

For each test instance, around 10 g of sample and 1 kg of fresh digestate material from anaerobic digestion were mixed and placed into polyethylene terephthalate (PET) mesh bags with a mesh size of 1–2 mm. The initial mass of the dry plastic samples was recorded before adding to the PET mesh bags. This enabled recovery of sample pieces > 2 mm at the end of the experiment for mass loss measurements. For BPE-RP-PLA, pieces of 5 cm X 5 cm were used according to DIN EN ISO 16929, the standard for pilot scale disintegration tests. For BPE-SP-PBS, individual film pieces of 10 cm X 10 cm were used. The bags were then sealed and placed into specially constructed metal sample cages (25 cm X 25 cm X 50 cm) with stainless steel grates with a mesh size of 10–12 mm. The cages were filled with more digestate material and placed in the middle of the composting pile together with two probes for temperature measurements. The experimental setup is shown in Figure 1. Each biodegradable plastic blend was tested in duplicate.

The exposure was carried out for three weeks, which is the composting duration for this batch of digestate material. After the first and second week, the composting pile was mixed in which the entire mass was transferred via a wheel loader from one hybrid fermentation chamber to another. In these phases, the sample cages were extracted, gently rotated to encourage mixing and reinserted into the composting pile. At the end of the composting process, the sample bags were extracted and dried in an oven at 105°C until a constant weight was reached. The plastic pieces were then extracted through sieving at 2 mm, washed and oven-dried at 58°C before final weighing. The initial and final weight was used to calculate the remaining mass using Equation 1. Samples of the composting medium were taken at the start and end of the experiment, oven-dried at 105°C to measure moisture content. The biomass

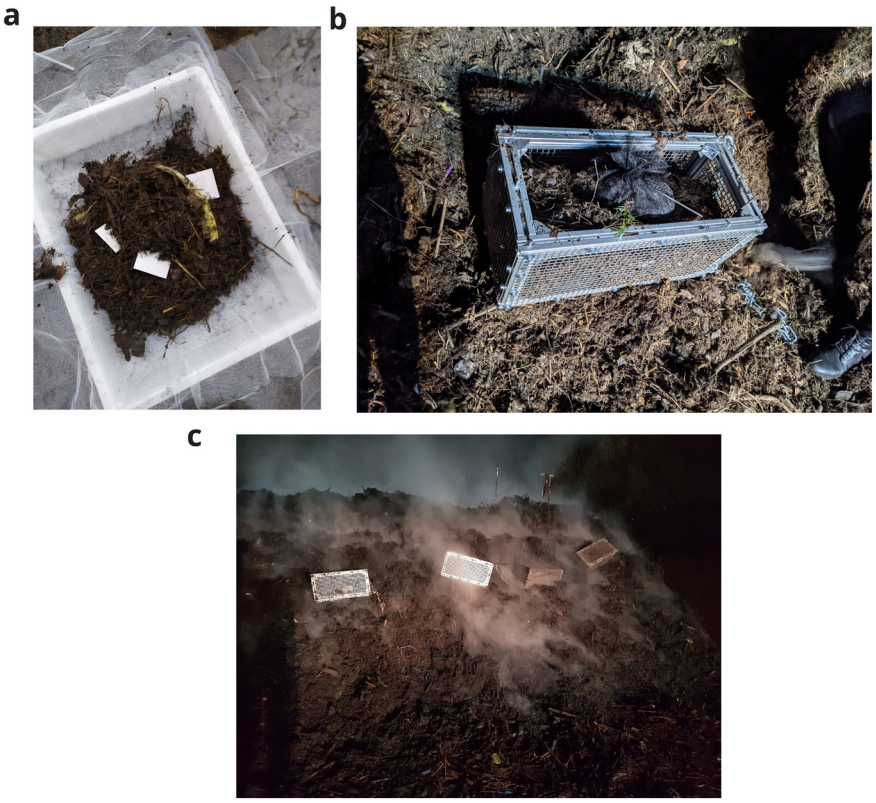

Figure 1. Experimental setup in the industrial composting plant. (**a**) biodegradable plastic samples with digestate material in sample nets. (**b**) sample nets in a metal cage. (**c**) position of the cages in the composting pile.

was then ground into powder for further characterization of pH, C/N ratio and volatile solids content using standard methods described in *Extended data* (Chong *et al*., 2022b).

Results and discussion

Disintegration rate in lab-scale industrial composting

Figure 2 shows the average measured remaining mass of the samples larger than the 2 mm limit as defined by ISO 20200, as well as the standard deviation of the triplicate measurements. The remaining mass of BPE-SP-PBS is 45.4%, 10.9% and 0.3% by weeks 4, 8 and 12 respectively. BPE-SP-PBS in the current form will thus fulfil the 90% disintegration condition (10% remaining mass) set by ISO 20200 after a little over 8 weeks. In contrast, 55.7% of BPE-RP-PLA remained larger than 2 mm after 12 weeks with only a small decrease between week 8 and week 12. BPE-RP-PLA in its current form thus does not fulfil the 90% disintegration condition based on ISO 20200. For both materials, the decrease in volatile solids of the composting medium (denoted R in ISO 20200) met the minimum requirement of the standard set at 30% from week 4 onward. The R-values and mass loss data are tabulated in *Underlying data* (Chong *et al*., 2022a). The large standard deviation at week 4 for BPE-SP-PBS can be attributed to uneven contact of the films with the composting medium, as it was observed that the pieces above the composting medium experienced little disintegration. After week 4, the pieces reduced in size and thus could be evenly distributed within the composting medium.

The lab-scale results indicate that BPE-SP-PBS of a thickness ≤ 109 µm would meet the disintegration requirement in an industrial composting plant as long as the residence time of thermophilic composting is longer than 8 weeks. On the other hand, BPE-RP-PLA in its current form would not disintegrate sufficiently in industrial composting plants with an active thermophilic composting time of fewer than 12 weeks. A reduction in thickness to achieve a higher surface area to volume ratio or changes to the blend composition are potential approaches to increase the disintegration rate from the material design point of view. In addition, it should be noted that the composting temperature of 58°C is slightly lower than the glass transition temperature of 59°C for BPE-RP-PLA. An increase in the composting temperature should also facilitate faster disintegration.

Comparing PLA and PBS, PLA/PBS blends with a higher ratio of PLA were reported to achieve higher disintegration rates in industrial composting conditions by Tolga *et al.* (Tolga *et al.*, 2020). The authors attributed this to a higher autocatalytic degradation phenomenon in PLA and showed that thickness has a strong effect on the disintegration speed. A lower abiotic degradation rate of PBS compared to PLA in an alkali solution of pH 13 at 37°C was proposed from the results of a study (Wang *et al.*, 2016). In addition, the filler content and type differed for the two blends. Talc used in BPE-SP-PBS was reported in a paper to retard disintegration of PLA/PBS blends (Tolga *et al.*, 2020). The effect of calcium silicate on the biodegradability of bioplastic was not yet directly studied in research. The much lower thickness and thus much higher specific surface area of the BPE-SP-PBS (130 cm^2/g) samples likely contributed to a higher disintegration rate compared to the thicker BPE-RP-PLA (14 cm^2/g) in the timeframe studied.

Disintegration rate in an industrial composting plant

The degradation rates after exposure in an industrial composting plant are shown in Figure 3. In addition, the results from the second round of lab-scale test using identical sample dimensions and exposure duration are shown for comparison. After 3 weeks of exposure to industrial composting of the digestate material from anaerobic digestion, the plastic samples experienced only minor mass losses. For BPE-RP-PLA, the average remaining mass was 98.3%. BPE-SP-PBS samples experienced slightly higher mass loss with an average remaining mass of 93.9%. After 3 weeks of exposure based on ISO 20200 in the lab, the remaining mass of BPE-RP-PLA

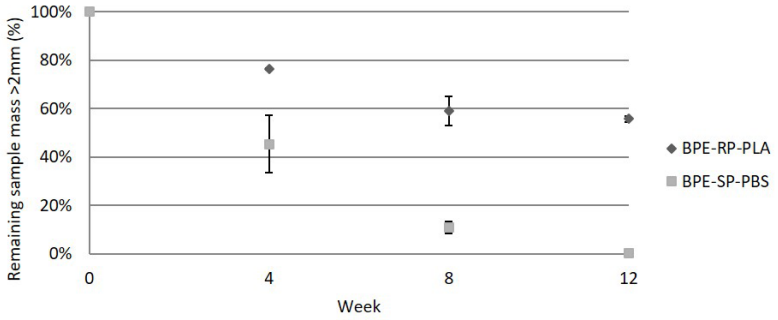

Figure 2. The remaining mass of samples in the lab-scale simulated industrial composting. Error bars denote standard deviation.

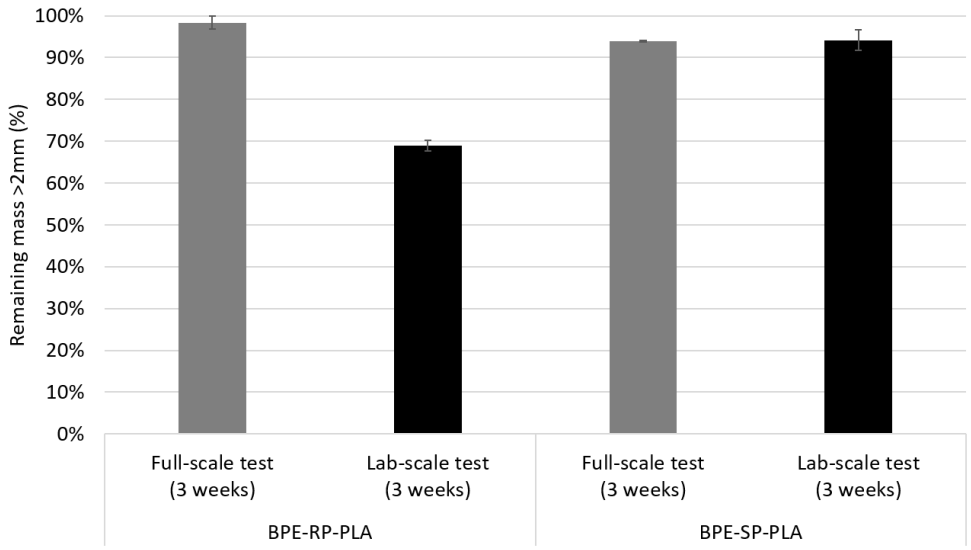

Figure 3. The remaining mass of samples after industrial composting full-scale (in grey) in comparison to the lab-scale composting test (in black) for a duration of 3 weeks. Error bars denote standard deviation.

and BPE-SP-PBS were on average 68.9% and 94.2%, respectively.

For BPE-RP-PLA, the resulting mass loss after 3 weeks of full-scale industrial composting was much lower compared to the lab-scale equivalent. Although the reduction in mass is not significant at full-scale, BPE-RP-PLA had significant visual changes including yellowing and unevenness on the surface. Figure 4 depicts the full-scale samples before and after full-scale industrial composting. In addition, the samples became more brittle and prone to fragmentation. This signifies a structural degradation of the polymer matrix and indicates that the material was still within the incubation phase, theoretically representing a reduction in molecular weight before the start of mass loss due to structural disintegration.

For BPE-SP-PBS, the resulting mass loss after 3 weeks of full-scale industrial composting was similar to the lab-scale equivalent, yet with a lower fragmentation of the samples. An average of 14% of the remaining mass after 3 weeks of composting in the lab was having a particle size between 2 mm and 10 mm, the fragmentation of the pieces > 10 mm was also much higher (see Figure 4 (e)). >99% for the full-scale samples remained > 10 mm as most of the pieces remained intact except for small holes found in some pieces (see Figure 4 (d)). Data of the particle size distribution is given in *Underlying data* (Chong *et al.*, 2022a). Additionally, BPE-SP-PBS full-scale samples experienced yellowing and warping with noticeable holes and thus were also in a more advanced stage of the disintegration process compared with the full-scale test in this aspect.

These observations indicate that the rate of mass loss of biodegradable plastics will only be significant if the duration of the thermophilic composting phase exceeds the incubation phase, as degradation of the material occurs at the beginning within the polymer matrix without causing structural disintegration. The extent of disintegration must also cross the size cutoff threshold of 2 mm before mass loss will be captured using the applied methodology. This indicates that even small differences in the duration of composting can result in large differences in remaining sample mass. The difference between the lab-scale and full-scale test could be due to the differences in the compositing medium and conditions, which are further discussed in Differences in composting conditions between the lab-scale and full-scale tests.

The results indicate that the tested blends in the current shape and form are not suitable for treatment in the industrial composting process where the test was carried out. Although degradation of the material was observed, the disintegration rate is very low within the active composting duration of the plant. It should be noted that the use of mesh bags might have negatively affected the disintegration rate by reducing the plastic-substrate contact area (Sintim *et al.*, 2019). To minimize this risk, around 1 kg of organic waste (similar to the amount used in the lab-scale tests) was placed together with the samples in the mesh bags to facilitate direct contact. In addition, the thickness of the material play a deciding role in achieving sufficient disintegration. The suitability to treat compostable plastics in industrial composting plants in general is further discussed in Feasibility of treating compostable plastic packaging in industrial composting plants.

A parallel study using cellulose acetate-based films also showed low disintegration (<20%) after the industrial composting process (Gadaleta *et al.*, 2023). In contrast, degradation rates up to 100% were found after 22 days of industrial composting

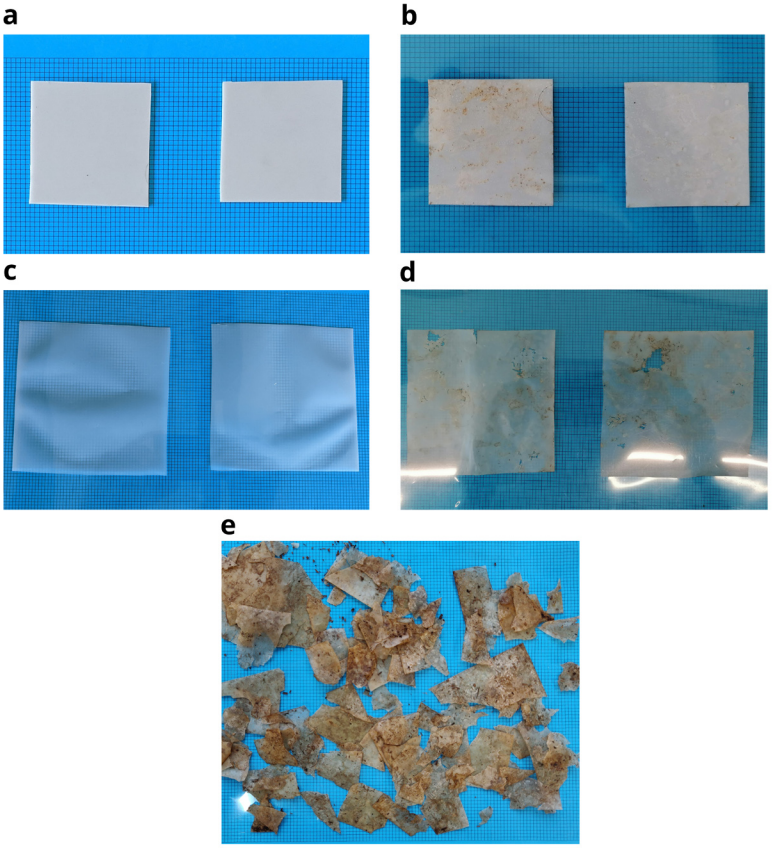

Figure 4. Samples before and after industrial composting. (**a**, **b**) BPE-RP-PLA before and after 3 weeks of full-scale composting. (**c**, **d**) BPE-SP-PBS before and after 3 weeks of full-scale composting. (**e**) BPE-SP-PBS samples after 3 weeks of lab-scale composting.

of commercially available PLA based plant pot cuttings (van der Zee & Molenveld, 2020). The study also reported a high variance of the disintegration rate between the PLA coffee capsule sample duplicates and attributed it to environmental variation. However, since exact dimensions of the samples were not given, a more detailed comparison is not possible. Another study reported that PLA/PBAT films only started to disintegrate after 21 days of exposure to industrial composting conditions (Musioł *et al.*, 2018). It should be noted that comparison between full-scale and pilot scale studies are difficult because of the different sample dimensions, substrate composition as well as general environmental conditions. In general, high variability can be seen from the outcomes of industrial composting.

Differences in composting conditions between the lab-scale and full-scale tests

The lab-scale tests based on ISO 20200 simulates a controlled thermophilic environment with a well-defined composting medium with small particle sizes (95% < 1 cm). On the other hand, the digestate material with which industrial composting was carried out had a larger and broader particle size distribution (shredded to ≤ 8 cm). The composition would also change seasonally depending on separately collected organic waste from households. The measured pH, moisture content, volatile solids content and C/N ratio of the composting medium in both tests are listed in Table 2. The standard deviation of the total volatile solid measurements reflects the heterogeneity of the full-scale composting medium, which had higher variation compared to the composting medium used in the lab.

The average volatile solid content of the lab-scale composting medium was higher compared to the full-scale medium. Since the full-scale medium stems from organic waste, it is expected that the inorganic content, i.e. sand and dust, will be higher due to the mixed collection of food and garden waste. The pH of the lab-scale composting medium was slightly acidic while the pH of the full-scale medium was neutral; yet both in the accepted range for bacteria and fungi (Chen *et al.*, 2011). The C/N ratios of the lab-scale and full-scale composting medium were close to the optimal for composting, quoted at 25 to 30 (Kumar *et al.*, 2010). Generally, it could be argued that the lab-scale composting medium with its high organic solids content, smaller particle sizes and thus homogeneity is more conducive to biotic disintegration as it facilitates a better sample-medium contact and higher biological activity. This could explain the better disintegration rates of the samples in the lab. In addition, the composting medium in the lab-scale test is mixed more frequently (based on the schedule in ISO 20200) compared

Table 2. Lab-scale and full-scale composting medium at the start of the experiment.

Test type	Lab-scale (3 weeks)	Full-scale
Brief description	A well-defined mix of materials based on ISO 20200.	A mixture of food and garden waste after dry anaerobic digestion. The composition depends on separately collected organic waste in the region.
Particle size	95% < 1 cm	≤ 8 cm
Total dry solids[1]	45.0% (Start)[2] 29.0%±1.0% (End)[3]	37.1%±5.0% (Start) 47.6%±1.9% (End)
Total volatile solids[4]	90.6%±0.6%	37.8%±2.6%
pH[5]	5.8±0.1	6.9±0.1
C/N ratio[5]	33±2	19±1

1 Based on wet weight; 2 The moisture content was adjusted at the start of the experiment and thus not measured; 3 After three weeks of composting BPE-SP-PBS (Round 2); 4 Based on total dry solids; 5 Measured on oven-dried and ground samples;

to the full-scale composting medium (once a week). The substrate particle size affects composting performance, shredding increases the surface area to volume ratio, homogeneity, and also allows sufficient aeration and heating as long as the resulting substrate size is within an optimal range (Amuah *et al.*, 2022; Reyes-Torres *et al.*, 2018). A higher particle size distribution and heterogeneity of the industrial composting medium might contribute to differing composting rates in different zones of the composting pile. In this study, the metal cages and mesh bag might have amplified the effects of heterogeneity by preventing the thorough mixing of the substrate within the cages with the rest of the substrate.

Factors affecting abiotic disintegration include temperature, pH and moisture. The temperature profiles of the full-scale test differed slightly to the lab-scale tests. In the lab-scale experiments, the temperature was kept constant at 58°C via a convection oven. In contrast, the temperature of the full-scale composting medium depended on self-heating and went through multiple cycles corresponding to the mixing schedules. The profiles of the temperature measured by the two probes are shown in Figure 5. The two minima in the middle correspond to the mixing phase, where the composting chambers are open and the composting medium is transferred from one chamber to another to facilitate mixing. The temperature ranged from 40°C to 80°C with an approximate average of 64°C. In both cases, the temperatures were well within the thermophilic range with the full-scale composting having a higher average temperature.

Indicated by the measurements at the start and end of the experiments, the lab-scale tests environment had a lower moisture content compared to the full-scale test at the start but a higher moisture content at the end. A wetter environment might facilitate better abiotic disintegration of PLA and PBS. The disintegration of PLA initially relies on chemical hydrolysis to break down long-chain polymers (Mochizuki & Hirami, 1997) and thus benefits from higher humidity (Karamanlioglu *et al.*, 2017). The degradation of PBS relies on enzymatic activity on the surface (Su *et al.*, 2019) as well as abiotic hydrolysis (Muthuraj *et al.*, 2015) and thus would also benefit from increased moisture. Though, the slightly higher starting pH of the synthetic organic waste (pH 6) in comparison to the full-scale substrate (pH 7) in the lab-scale could have positively contributed to the higher hydrolysis rates. On the other hand, a more basic pH was reported to accelerate surface erosion (von Burkersroda *et al.*, 2002). The difference in terms of moisture content and pH is not a clear-cut in this study. Overall, the full-scale test had better abiotic degradation conditions in terms of temperature. This did not however translate to higher disintegration compared to the lab-scale.

Feasibility of treating compostable plastic packaging in industrial composting plants

Due to the complexity of the interaction between product design and composting parameters, the feasibility of treating compostable plastics in industrial composting plants depends on many factors in addition to the certification of the material. This study indicates that both blends in their current form do not meet the 90% disintegration requirement for compostable packaging according to EN 13432 after exposure to the process at the Bützberg plant. Although, BPE-SP-PBS did show sufficient disintegration in the lab-scale test after 12 weeks. A downside to the full-scale tests is the use the sample nets and metal cages, which could have minimized the exposure to mechanical stress during the composting process, i.e. during mixing. In addition, the cages prevented thorough mixing of the substrate.

The disintegration of BPE-SP-PBS in other industrial composting plants might meet the requirement if the thermophilic composting phase is longer than 8 weeks, as indicated by the lab-scale test results. However, the thermophilic composting phase of most industrial processes does not exceed 3 weeks (Ruggero *et al.*, 2021). For BPE-RP-PLA, the thermophilic composting phase would need to be longer than 12 weeks to achieve sufficient degradation according to the lab-scale test results.

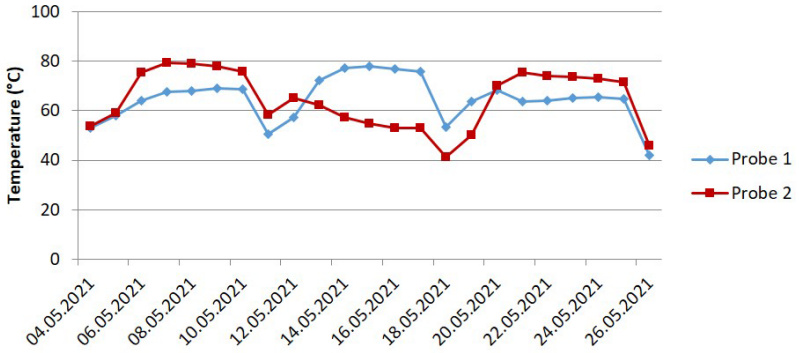

Figure 5. The temperature profile of the full-scale industrial composting (Bützberg Biogas and Composting Plant, 2021).

The conditions defined for certified compostable packaging, for example, those with their disintegration tested according to the pilot-scale disintegration test according to ISO 16929, are 4 weeks of composting above 55°C, 4 weeks of composting above 50°C and 4 weeks of composting below 45°C using fresh organic waste as inputs. These conditions also deviate significantly from those in Bützburg and similar plants with a shorter active composting phase. The German Ordinance on Biowastes requires hygienation temperatures of at least 55°C over two weeks and 60°C over 6 days during thermophilic composting (BioAbfV, 2013). A sufficient degradation of the certified materials are thus not guaranteed solely based on these parameters.

Another important determining factor for disintegration is the shape and form of the biodegradable packaging articles, i.e. Tolga *et al.* (2020) found that the degree of disintegration linearly correlated with the thickness of PLA/PBS blends. Especially for rigid packaging, the thickness presents an extra obstacle as seen for BPE-RP-PLA. Extra pre-processing steps, i.e. shredding, would likely be needed to increase the disintegration of biodegradable plastics in industrial composting plants. Alternatively, thinner samples will theoretically allow for faster disintegration. However, research on the effects of differing substrate composition, particle size distribution and other parameters such as total volatile solids is lacking and important to understand the requirements for sufficient disintegration in actual plants as well as to understand the potential differences between plants.

It should be noted that the disintegration rate calculated considering a size cutoff of 2 mm is not sufficient when considering the feasibility of treating compostable plastic in industrial composting plants. The behavior and ultimate biodegradation of the smaller compostable plastic particles in the environments in which they might enter, i.e. agricultural or garden soil, as well as their toxicity, should be determined (Fojt *et al.*, 2020; Karamanlioglu & Alkan, 2020; Sintim *et al.*, 2019). Lastly, integration with the wider waste management system needs to be considered. For one, the motivation of the industrial composting plants to sanitize organic waste and produce compost might not align with the treatment of compostable plastics, which might complicate the process without added benefit to the plant operator. In addition, the acceptance of biodegradable plastics in the organic waste stream increases the risk of contamination of conventional plastics, which are proven to be stable in industrial composting as well as anaerobic digestion (Alassali *et al.*, 2018).

Conclusion

Heeding the resistance from some waste management authorities to treat compostable plastics in composting plants, the behavior of biodegradable and compostable plastics under actual industrial composting conditions should be thoroughly examined to assess their environmental impact. Therefore, this study investigated the disintegration rate of a PLA blend and a PBS blend designed for rigid and soft packaging respectively under simulated lab-scale and on-site industrial composting conditions.

While under simulated industrial conditions according to ISO 20200, BPE-RP-PLA and BPE-SP-PBS samples achieved significant disintegration with averages of 55.7% and 0.3% respectively after 12 weeks, showing good disintegration potential. However, the mass loss was low after full-scale composting with an average remaining mass of 98.3% for the BPE-RP-PLA samples and 93.9% for the BPE-SP-PBS samples after 3 weeks. This stark difference is not solely due to the shorter composting duration of the full-scale test, as the supplementary lab-scale test of the same duration showed higher mass loss for BPE-RP-PLA with an average of 68.9% remaining mass. BPE-SP-PBS achieved similar mass loss after 3 weeks in the lab but showed more fragmentation with 14% of its remaining mass under 10 mm particle size compared to <1% for the full-scale samples. Based on the data in this study, the abiotic conditions such temperature, moisture content and pH in the lab-scale was not notably better compared to the full-scale test. The methods used also ensured that at least 1 kg of composting substrate was in direct contact with the plastic samples in all cases. This point towards the potential role of other composting parameters

such as the process scale, substrate composition and particle size distribution in the rate of disintegration. These factors may vary for different plants and seasonally. In addition, the presence and diversity of microorganisms play a role. More research is needed in order to understand their effects on biodegradable plastic products.

Although BPE-SP-PBS samples in their current thickness almost disintegrated entirely after 12 weeks, a minimum thermophilic composting duration of 8 weeks would be necessary to reach the 90% disintegration (10% remaining mass) required by EN 13432, which exceeds the thermophilic phase of most composting plants. Biological waste treatment plants including composting plants are generally designed to treat and sanitize biological waste and are not optimized to treat compostable plastics, especially concerning the retention time. Henceforth, realistic certification requirements that reflect differing retention times and substrate composition which occur in real-life applications should be considered. Furthermore, systemic integration considering the organic waste value chain and the motivations of industrial composting plants is critical.

Data availability

Underlying data

B2Share: Underlying data_Lab-scale and on-field industrial composting of biodegradable plastic blends for packaging_Chong *et al*., 2023

https://doi.org/10.23728/b2share.f1e07485392443cfa455c9745f4d95ec (Chong *et al*., 2022a)

This project contains the following underlying data:

- CN ratio_Industrial test.xlsx
- CN ratio_lab-scale.xlsx
- Data list.docx
- Moisture content.xlsx
- Particle size distribution after composting_BPE-SP-PBS.xlsx
- pH.xlsx
- Remaining mass industrial test.xlsx
- Remaining mass lab-scale_BPE-RP-PLA.xlsx
- Remaining mass lab-scale_BPE-SP-PBS.xlsx
- R value lab-scale test reactors_BPE-RP-PLA.xlsx
- R value lab-scale test reactors_BPE-SP-PBS.xlsx
- Total volatile solids.xlsx

Extended data

B2Share: Extended data_Lab-scale and on-field industrial composting of biodegradable plastic blends for packaging_Chong *et al*., 2022

http://doi.org/10.23728/b2share.5356b112d6bd424aa79b10d397ee1b81 (Chong *et al*., 2022b)

This project contains the following extended data:

- Extended data_Methodology.docx

Data are available under the terms of the Creative Commons Attribution 4.0 International license (CC-BY 4.0).

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Version 3

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✔ **Mehlika Karamanlioglu**
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I have no further comments since the responses are satisfactory.

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Biotechnology, biopolymers, environmental degradation of plastics.

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Reviewer Report 12 February 2024

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✔ **Douglas G. Hayes**
Department of Biosystems Engineering & Soil Science, University of Tennessee, Knoxville, TN, USA

The authors have addressed all of the issues I raised satisfactorily. I therefore recommend that this manuscript be accepted for indexing.

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Biobased and biodegradable agricultural plastics, micro- and nanoplastics,

colloidal systems

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Version 2

Reviewer Report 20 October 2023

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Mehlika Karamanlioglu
Faculty of Engineering and Architecture, Department of Biomedical Engineering, Istanbul Gelisim University, İstanbul, Turkey

There are some significant improvements in the revised paper. However, there are still some parts that need more explanation. Further comments are made below:

1 - The main aim of this study should be clearly presented especially in the background and in the introduction. What do you aim to compare? The degradation rate of 2 plastics, or only the degradation rates in the lab scale and in the composting site? If it is the latter one, the comparison on the 3rd week can enable to achieve this aim.

Therefore, why did you conduct the tests on the 4th, 8th and 12th week in lab scale? It should be written and explained clearly in relevant sections.

2 - The term 'on-field' is confusing. Industrial composting vs. lab scale sounds more accurate.

3 - The supplementary lab-scale test for 3 weeks should be written in lab-scale industrial composting part in the method with more detail. This is actually an important test so it should be emphasized more in the methods part rather than being just a supplementary test.

Also it is written for the supplementary test that 'This test was done in triplicate'. Could you elaborate on that?

4 - Burial of samples in lab scale test should be given in more detail. Were they directly buried in the pile?

5 - Although molecular weight and Tg are given, still more detail about the plastics used in this study should be given. What does BPE stand for, how did you obtain these plastics, did you make them or buy them commercially, i.e., were they commercial packages such as deli containers, etc.?

It is mentioned that '...two biodegradable plastic prototypes developed for rigid (PLA-based) and soft packaging (PBS-based) respectively within the BIO-PLASTICS EUROPE research Project...' How did you develop them? It should be added to methods parts as materials used.

6 - In the industrial composting test, why do you have 2 results for the same plastic (Field sample 1 and 2)? It is written that: 'In the plant, the remaining mass was 97.2% and 99.5%', Do these plastics have any difference? If there is no difference, if they are only duplicates, you should present the average of them with standard deviation or standard error of the mean.

7 - Cite the source after these sentences: 'The authors attributed this to a higher autocatalytic degradation phenomenon in PLA. The same study also showed that thickness has a strong effect on the disintegration speed.'

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Biotechnology, biopolymers, environmental degradation of plastics.

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard, however I have significant reservations, as outlined above.

Author Response 22 Jan 2024

Zhi Kai Chong

The authors would like to thank Mehlika Karamanlioglu for the helpful review and comments to improve the structure. The edits made in response to each comment as well as explanations are given point-by-point below in italics.

1 - The main aim of this study should be clearly presented especially in the background and in the introduction. What do you aim to compare? The degradation rate of 2 plastics, or only the degradation rates in the lab scale and in the composting site? If it is the latter one, the comparison on the 3rd week can enable to achieve this aim.
Therefore, why did you conduct the tests on the 4th, 8th and 12th week in lab scale? It should be written and explained clearly in relevant sections. *Thank you for the comment. To improve the description of the study's aim, the last paragraph of the introduction was revised to "The controlled, lab-scale test represented the disintegration potential of the prototypes and was conducted for a duration up to 12 weeks, the maximum allowed composting duration based on EN 13432. On the other hand, the test in the composting plant represented a real-life scenario. The composting medium of both cases was characterized for comparison. The results outlined the difference of disintegration rates in the lab and in a plant and explored the possible reasons for the differences.". The main aim of the study was to investigate the behavior of compostable plastic packaging prototypes in real-life composting conditions as well as the relation to their behavior in a controlled, lab-scale test. The composting test in the lab was necessary for up to 12 weeks to determine the disintegration potential of the material (ISO 20200 and EN 13432 allows*

for composting up to 12 weeks). In addition, this allows us to make inferences on the behavior of the material in other composting plants that might have a longer thermophilic composting duration compared to the one presented in this study.
2 - The term 'on-field' is confusing. Industrial composting vs. lab scale sounds more accurate. *"On-field" was replaced by "full-scale". The description "full-scale" is still needed because both "lab-scale" and "full-scale" tests refer to industrial composting conditions, which is characterized as thermophilic composting.*

3 - The supplementary lab-scale test for 3 weeks should be written in lab-scale industrial composting part in the method with more detail. This is actually an important test so it should be emphasized more in the methods part rather than being just a supplementary test.
Also it is written for the supplementary test that 'This test was done in triplicate'. Could you elaborate on that? *The text mentioned was moved to the Section "Lab-scale industrial composting" to facilitate easier understanding. The text was also revised to make it clear that it is relevant for comparison between the lab-scale and full-scale composting conditions (temperature and substrate mix). "Triplicate" refered to three separate composting instances (boxes) for each time point. The results were used to calculate the average and standard deviation for each time point.*

4 - Burial of samples in lab scale test should be given in more detail. Were they directly buried in the pile? *We ensured that >80% of the samples were covered by the substrate in the lab-scale test. Due to the high surface area to mass ratio, a 100*% coverage was not *possible. Further explanations were added as suggested to paragraph 3 of the Section "Lab-scale industrial composting".*

5 - Although molecular weight and Tg are given, still more detail about the plastics used in this study should be given. What does BPE stand for, how did you obtain these plastics, did you make them or buy them commercially, i.e., were they commercial packages such as deli containers, etc.? It is mentioned that '...two biodegradable plastic prototypes developed for rigid (PLA-based) and soft packaging (PBS-based) respectively within the BIO-PLASTICS EUROPE research Project...' How did you develop them? It should be added to methods parts as materials used. *The last paragraph of the section "Biodegradable plastic test samples" was revised to make the source of the samples clearer. The plastic prototypes were not obtained from commercial sources but produced by other partners within the BIO-PLASTICS EUROPE project. Though the formulations (blends) were developed within the project, the formulations and their effects on disintegration was not part of this study. This study focused on the potential disintegration behaviour of compostable plastic packaging products in industrial composting conditions.*

6 - In the industrial composting test, why do you have 2 results for the same plastic (Field sample 1 and 2)? It is written that: 'In the plant, the remaining mass was 97.2% and 99.5%', Do these plastics have any difference? If there is no difference, if they are only duplicates, you should present the average of them with standard deviation or standard error of the mean. *Thank you for the suggestion, we have updated the figure as suggested. The full-scale test data was presented as a mean with standard deviation. The text were also updated to reflect the figures presented.*

7 - Cite the source after these sentences: 'The authors attributed this to a higher autocatalytic degradation phenomenon in PLA. The same study also showed that thickness has a strong effect on the disintegration speed.'` *The text was revised to make it clear that these sentences are also cited from the study cited in the previous sentence "Comparing PLA and PBS, PLA/PBS blends with a higher ratio of PLA were reported to achieve higher disintegration rates in industrial composting conditions by Tolga et al. (Tolga* et al.*, 2020)."*

Competing Interests: No competing interests were disclosed.

Reviewer Report 11 October 2023

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Douglas G. Hayes
Department of Biosystems Engineering & Soil Science, University of Tennessee, Knoxville, TN, USA

Assessment of revised version of "Lab-scale and on-field industrial composting of biodegradable plastic blends for packaging", Chong ZK, Hofmann A, Haye M, Wilson S, Sohoo I, Alassali A and Kuchta K

Assessor: Douglas G. Hayes, Biosystems Engineering, University of Tennessee (USA)

I find the revised version of the manuscript is greatly improved. I recommend acceptance after the following minor revisions are addressed.

1. Why is the term "on-field" used for the industrial composting test? Industrial composting took place in a composting facility in Tangstedt, Germany, not in an agricultural field. I find this term to be confusing and perhaps misleading, and recommend that it be replaced with "industrial composting" (or equivalent) throughout the manuscript, including figures and tables.

2. Introduction, paragr 1, "development of alternatives" to what? Unclear.

3. Introduction, paragr 1, "decomposition of carbon" is awkward, unless a nuclear fission reaction is being referred to.

4. Introduction, paragr 1, "Compostable plastics .. that disintegrate significantly". This statement is of concern to me, since oxodegradables can "disintegrate significantly" but will not disappear. If the term "compostable" is employed with regard to a standard such as ISO 14855, the plastic material needs to fulfill several criteria pertaining not only to disintegration, but also biodegradation and heavy metal content. This sentence should be revised.

5. Introduction, paragr 2, “Industrially compostable products are designed to disintegrate ..” For the same rationale as provided above for Remark 4, this sentence needs to be revised. Oxodegradables are designed to “disintegrate” but are not compostable because of their inability to fully biodegrade.

6. Introduction, paragr 4, “has been studied in the literature in the lab ..” is awkward. The sentence would suggest that the scientists have conducted their literature searches in their lab rather than their office, or elsewhere.

7. Introduction, paragr 4: Please provide units for the ratio “7:3”, such as “w/w”, “mol/mol”, or “v/v”.

8. Introduction, paragr 7, “Chemically, the disintegration of PLA .. microbiological enzymatic attack”. Are abiotic reactions limited to hydrolysis? Can PLA undergo photodegradative reactions in the presence of sunlight that differ from hydrolysis?

9. For Table 1, why are there error bars for only some, but not all, of the entries?

10. On-field industrial composting test” section, paragr 1. Please describe greater detail on the “magnetic separation” procedure used to prepare the compost. Can a reference be cited?

11. Results and Discussion section, paragr 3; Grammar error (verb tense): “Comparing PLA and PBS ..blends with a higher ratio .. was reported”.

12. “Disintegration rate in an industrial composting plant” subsection, paragr 3: “Though not captured by the remaining ..”: “mass > 2 mm measure” is awkward. Also, what is meant by “second round lab-test”? I do not recall seeing anything related in the Experimental section.

13. “Disintegration rate in an industrial composting plant” subsection, paragr 4, “significant if the incubation phase is exceeded” : “exceeded” is awkward / unclear.

14. “Disintegration rate in an industrial composting plant” subsection, paragr 6, “only started to disintegrated” (grammar).

15. “Differences in composting conditions between ..” subsection, first paragr: Please insert a space between “8” and “cm”. Please also insert spaces between the numerical values and the units in Table 2, “particle size’ row.

16. For Table 2, I see that the numbering scheme for the footnotes is down the columns moving left to right. For many journals, the preference is to number from left to right across a column, and ten move down the rows. Please check with the journal on the preferred format.

17. “Differences in composting conditions between ..” subsection,, paragr 2: “optimal size after shredding increases the surface area to volume ratio”. Actually, the trend of increasing SA/V with decreasing size occurs both above and below an optimal size. Please reword this sentence.

18. “Differences in composting conditions between ..” subsection, paragr 3: “dips” is slang. Suggest replacing with “minima”.

19. “Differences in composting conditions between ..” subsection, paragr 4: “The degradation of PBS relies on enzymatic activity .. as well as hydrolysis” Since hydrolysis can be abiotic or biotic, this sentence is a little awkward. I suggest using “as well as abiotic hydrolysis” to clarify.

20. Paragraph above Conclusions: “scope” is a little awkward. (“..the disintegration rate with a scope above 2 mm..”)

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Biobased and biodegradable agricultural plastics, micro- and nanoplastics, colloidal systems

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard, however I have significant reservations, as outlined above.

Author Response 22 Jan 2024

Zhi Kai Chong

The authors would like to thank Douglas G. Hayes for the review. We appreciate the comments and the attention to detail. The edits made in response to each comment as well as explanation are given point-by-point below in italics.

1. Why is the term “on-field” used for the industrial composting test? Industrial composting took place in a composting facility in Tangstedt, Germany, not in an agricultural field. I find this term to be confusing and perhaps misleading, and recommend that it be replaced with “industrial composting” (or equivalent) throughout the manuscript, including figures and tables.

“On-field” was replaced by “full-scale”. The description “full-scale” is still needed because both “lab-scale” and “full-scale” tests refer to industrial composting conditions, which is characterized as thermophilic composting.

1. Introduction, paragr 1, “development of alternatives” to what? Unclear.

The sentence was revised to “The prevalence of plastic waste and micro-plastic in the environment has spurred research and development of biodegradable plastics as alternatives to conventional fossil-based non-biodegradable plastics.”.

1. Introduction, paragr 1, “decomposition of carbon” is awkward, unless a nuclear fission reaction is being referred to.

The sentence was revised to “It is important to note the difference between the terms biodegradation and disintegration. In EN 13432, biodegradability is to be proved via the decomposition of the polymer structure and the subsequent release of the carbon in the plastic material ultimately as carbon dioxide.”

1. Introduction, paragr 1, “Compostable plastics .. that disintegrate significantly”. This

statement is of concern to me, since oxodegradables can “disintegrate significantly” but will not disappear. If the term “compostable” is employed with regard to a standard such as ISO 14855, the plastic material needs to fulfill several criteria pertaining not only to disintegration, but also biodegradation and heavy metal content. This sentence should be revised.

We appreciate the comment. We regard the disintegration ability as a property of plastics that are inherently compostable without referring to a specific standard. The heavy metal content would be part of a certification and/or standard. We thus differentiate here between (generally) compostable plastics and certified compostable plastics, the latter which follows a certain certification. In this context, we are referring to the former and thus prefer to keep the sentence as it is. The ultimate biodegradability is implied with the use of the term “biodegradable plastics”.

1. Introduction, paragr 2, “Industrially compostable products are designed to disintegrate ..” For the same rationale as provided above for Remark 4, this sentence needs to be revised. Oxodegradables are designed to “disintegrate” but are not compostable because of their inability to fully biodegrade.

The sentence was revised to “Industrial compostable products are produced from biodegradable material and designed to disintegrate within a reasonable period under industrial composting conditions.”

1. Introduction, paragr 4, “has been studied in the literature in the lab ..” is awkward. The sentence would suggest that the scientists have conducted their literature searches in their lab rather than their office, or elsewhere.

The sentence was revised to “In literature, the behavior of biodegradable plastics in the composting process has been studied in both lab- and pilot scales.”

1. Introduction, paragr 4: Please provide units for the ratio “7:3”, such as “w/w”, “mol/mol”, or “v/v”.

“w/w” was added to the sentence.

1. Introduction, paragr 7, “Chemically, the disintegration of PLA .. microbiological enzymatic attack”. Are abiotic reactions limited to hydrolysis? Can PLA undergo photodegradative reactions in the presence of sunlight that differ from hydrolysis?

We agree with the comment, PLA may also degrade via other abiotic pathways such as thermal-degradation and photodegradation. However, biodegradable polyesters were reported to be particularly susceptible to hydrolysis (Woodard und Grunlan 2018; Speranza et al. 2014) and thus can be regarded as the main abiotic degradation mechanism in industrial composting conditions. With the absence of sunlight, the photodegradative pathway should be less relevant.

1. For Table 1, why are there error bars for only some, but not all, of the entries?

That is a good question. We added the standard deviation for the film thickness because it has significant variability due to the fact that it was a prototype produced in the lab. We added an explanation for the standard deviation in the captions.

1. On-field industrial composting test” section, paragr 1. Please describe greater detail on the “magnetic separation” procedure used to prepare the compost. Can a reference be cited?

The sentence was revised to: “The plant receives separately collected bio-waste from households in the region. Before undergoing biological treatment, the waste was shredded (< 8 cm), sieved and sent through magnetic separation to remove impurities such as plastics and metals.”. The magnetic separation process is carried out on the organic waste inputs before composing. We received this information through personal communication with the industrial plant and thus can

not provide a reference. However, impressions of the plant can be seen on this website: https://www.stadtreinigung.hamburg/ueber-uns/muellverwertung-anlagen/biogas-und-kompostwerk-buetzberg/

1. Results and Discussion section, paragr 3; Grammar error (verb tense): "Comparing PLA and PBS ..blends with a higher ratio .. was reported".

The sentence was corrected.

1. "Disintegration rate in an industrial composting plant" subsection, paragr 3: "Though not captured by the remaining ..": "mass > 2 mm measure" is awkward. Also, what is meant by "second round lab-test"? I do not recall seeing anything related in the Experimental section.

The sentence was revised to: "Additionally, BPE-SP-PBS full-scale samples experienced yellowing and warping with noticeable holes and thus were also in a more advance stage of the disintegration process compared with the full-scale test in this aspect." to reduce confusion.

1. "Disintegration rate in an industrial composting plant" subsection, paragr 4, "significant if the incubation phase is exceeded" : "exceeded" is awkward / unclear.

The sentence was revised to: "These observations indicate that the rate of mass loss of biodegradable plastics will only be significant if the duration of the thermophilic composting phase exceeds the incubation phase, as degradation of the material occurs at the beginning within the polymer matrix without causing structural disintegration.".

1. "Disintegration rate in an industrial composting plant" subsection, paragr 6, "only started to disintegrated" (grammar).

The sentence was corrected.

1. "Differences in composting conditions between .." subsection, first paragr: Please insert a space between "8" and "cm". Please also insert spaces between the numerical values and the units in Table 2, "particle size' row.

The text was corrected.

1. For Table 2, I see that the numbering scheme for the footnotes is down the columns moving left to right. For many journals, the preference is to number from left to right across a column, and ten move down the rows. Please check with the journal on the preferred format.

We checked this with the editors and made the necessary corrections.

1. "Differences in composting conditions between .." subsection,, paragr 2: "optimal size after shredding increases the surface area to volume ratio". Actually, the trend of increasing SA/V with decreasing size occurs both above and below an optimal size. Please reword this sentence.

The sentence was revised to: "The substrate particle size affects composting performance, shredding increases the surface area to volume ratio, homogeneity, and also allows sufficient aeration and heating as long as the resulting substrate size is within an optimal range (Amuah et al., 2022; Reyes-Torres et al., 2018)."

1. "Differences in composting conditions between .." subsection, paragr 3: "dips" is slang. Suggest replacing with "minima".

The text was corrected.

1. "Differences in composting conditions between .." subsection, paragr 4: "The degradation of PBS relies on enzymatic activity .. as well as hydrolysis" Since hydrolysis can be abiotic or biotic, this sentence is a little awkward. I suggest using "as well as abiotic hydrolysis" to clarify.

"abiotic" was added to the sentence.

1. Paragraph above Conclusions: “scope” is a little awkward. (“..the disintegration rate with a scope above 2 mm..”)

The sentence was revised to: “It should be noted that the disintegration rate calculated considering a size cutoff of 2 mm is not sufficient when considering the feasibility of treating compostable plastic in industrial composting plants.” Literature Speranza, V.; Meo, A. de; Pantani, R. (2014): Thermal and hydrolytic degradation kinetics of PLA in the molten state. In: *Polymer Degradation and Stability* 100, S. 37–41. DOI: 10.1016/j.polymdegradstab.2013.12.031. Woodard, Lindsay N.; Grunlan, Melissa A. (2018): Hydrolytic Degradation and Erosion of Polyester Biomaterials. In: *ACS macro letters* 7 (8), S. 976–982. DOI: 10.1021/acsmacrolett.8b00424.

Competing Interests: No competing interests were disclosed.

Version 1

Reviewer Report 01 November 2022

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Douglas G. Hayes
Department of Biosystems Engineering & Soil Science, University of Tennessee, Knoxville, TN, USA

I find this manuscript to be publishable after addressing minor revisions. The experimental work is reliable and was performed correctly, per ISO 16929 and 20200 standardized testing conditions for measuring disintegration. The interpretation of the data is mostly correct, in my opinion. However, there are a few places where I believe interpretation should be reconsidered. First, one must be careful on extending results achieved using standardized tests and specifications achieved per a standard to actual performance in an industrial composting facility. As the authors likely know, composting is truly an art and its performance can be affected by several different parameters. Second, biodegradation is directly controlled by the surface area per volume of the plastic material. This is relevant in two contexts: A) The PLA test material has a 10-fold higher thickness compared to PBS, which will lead to a lower biodegradation rate for PLA, and B) The mesh bags used in the study will likely introduce a resistance to mass transfer of nutrients and gases across the mesh bag matrix. In addition, the presentation can be improved in a few places in the manuscript.

Regarding the mass transfer inhibition of biodegradation (disintegration) caused by the mesh bag, I would recommend performing a side-by-side laboratory test of a standard plastic testing material (e.g., a thin PLA film) directly in compost vs in a mesh bag placed in compost. Such an experiment does not need to be carried out for the full duration of the experiments described in

your paper; perhaps a few weeks would suffice. We did this: H.Y. Sintim, et al., 2019[1] (within Supplementary Materials) and found for our systems that the mesh bags significantly slowed the biodegradation rate in compost.

My specific comments are given below.

1. Introduction, first paragraph: I would recommend defining “disintegration” and “composting” carefully. EN 13432 or one of the ISO disintegration testing methods should have these definitions. I disagree with to the use of “only” (“..would ONLY disintegrate significantly..”). A substance’s ability to readily biodegrade under composting conditions does not preclude the substance’s ability to readily biodegrade in a different environment (e.g., soil, activated sludge, or seawater).

2. Introduction, 3rd paragraph: “The disintegration condition in EN 13432 .. after a maximum of 12 weeks”. This sentence needs to be qualified: “..according to the _____ standardized testing conditions”

3. More information should be provided on the properties of the polymeric materials, such as weight-averaged molecular weight, glass transition temperature, apparent density (grams per square meter), etc.

4. Methods, Lab-scale composting. Were the composting ingredients or the compost itself treated with sieving prior to the addition of plastic?

5. Results and Discussion, first paragraph, “current form does not fulfil the 90% disintegration conditions” needs a qualifier, such as “specified in EN 13432”

6. Please consider merging Figs 2 and 3. The data of Fig 3 can be easily incorporated as datapoints into Fig 2. (Note, for the current Fig 3, y-axis numbers should have the zeroes to the right of the decimal place removed and the caption should indicate the duration.)

7. Fig 2: Do error bars represent standard deviation, standard error, or something else? Please elaborate (e.g., in the figure caption).

8. Results and discussion, “disintegration rate in an industrial composting plant” subsection, bottommost paragraph. This paragraph needs revision. First, as mentioned above, it is likely that the mesh bag promoted mass transfer limitations on the biodegradation reaction. Second, I believe that the non-compliance of PLA to the EN 13432 standards is at least partially attributable to the high, 1 mm, thickness of the films and perhaps the high content of calcium silicate nanofiller may play a role as inhibitor of biodegradation. Are the composting conditions and medium a factor? I find that the inclusion of a positive control into the experimental design, such as a standard plastic material (e.g., a thinner PLA film) improves in the interpretation of the results, although I realize that the ISO standardized tests for disintegration do not directly call for it. Please consider revising the discussion in this paragraph.

9. Feasibility of treating compostable plastic packaging .. “ subsection, first paragraph, “do not meet the 90% disintegration requirement”. I find this paragraph requires revision, noting first that the PBS material does appear to meet the requirement under laboratory

conditions, per Fig 2. The assessment given in this paragraph needs to take into account the high thickness of the PLA film and the mass transfer limitations imposed by the mesh bag for the data of Fig 3 (as discussed above)

References

1. Sintim HY, Bary AI, Hayes DG, English ME, et al.: Release of micro- and nanoparticles from biodegradable plastic during in situ composting.*Sci Total Environ*. 2019; **675**: 686-693 PubMed Abstract | Publisher Full Text

Is the work clearly and accurately presented and does it cite the current literature?
Yes

Is the study design appropriate and does the work have academic merit?
Yes

Are sufficient details of methods and analysis provided to allow replication by others?
Yes

If applicable, is the statistical analysis and its interpretation appropriate?
Partly

Are all the source data underlying the results available to ensure full reproducibility?
Yes

Are the conclusions drawn adequately supported by the results?
Partly

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Biobased and biodegradable agricultural plastics, micro- and nanoplastics, colloidal systems

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard, however I have significant reservations, as outlined above.

Author Response 25 Aug 2023

Zhi Kai Chong

The authors would like to thank Douglas G. Hayes for the detailed review and constructive comments. The table below summarises the authors' comments and enhancements made to the new version in response to the review. Review Section Authors' replies and comments I find this manuscript to be publishable after addressing minor revisions. The experimental work is reliable and was performed correctly, per ISO 16929 and 20200 standardized testing conditions for measuring disintegration. The interpretation of the data is mostly correct, in

my opinion. However, there are a few places where I believe interpretation should be reconsidered. First, one must be careful on extending results achieved using standardized tests and specifications achieved per a standard to actual performance in an industrial composting facility. As the authors likely know, composting is truly an art and its performance can be affected by several different parameters. Second, biodegradation is directly controlled by the surface area per volume of the plastic material. This is relevant in two contexts: A) The PLA test material has a 10-fold higher thickness compared to PBS, which will lead to a lower biodegradation rate for PLA, and B) The mesh bags used in the study will likely introduce a resistance to mass transfer of nutrients and gases across the mesh bag matrix. In addition, the presentation can be improved in a few places in the manuscript. The authors agree that extending results from standardized test to industrial composting plant is highly problematic. The difference between our results in the lab compared to the industrial composting plant is evidence of that. The possible reasons for the difference in disintegration rates were enhanced in the section “Differences in composting conditions between the lab-scale and on-field tests”, paragraph 2 to 4. The specific surface area of the samples were added in Table 1. In addition, the influence of the specific surface area on the results comparing the film and plate samples were added in the Section “Disintegration rate in lab-scale industrial composting”,paragraph 2 and 3. A discussion of the potential effect of the mesh bags was also included in the section “Disintegration rate in an industrial composting plant”,paragraph 5. Regarding the mass transfer inhibition of biodegradation (disintegration) caused by the mesh bag, I would recommend performing a side-by-side laboratory test of a standard plastic testing material (e.g., a thin PLA film) directly in compost vs in a mesh bag placed in compost. Such an experiment does not need to be carried out for the full duration of the experiments described in your paper; perhaps a few weeks would suffice. We did this: H.Y. Sintim, et al., 20191 (within Supplementary Materials) and found for our systems that the mesh bags significantly slowed the biodegradation rate in compost. Thank you for the helpful comments. A discussion of the potential effect of the mesh bags was added to the section “Disintegration rate in an industrial composting plant”,paragraph 5. In addition, the potential effect of using mesh bags coupled with the heterogeneity of the substrate is mentioned in the section “Differences in composting conditions between the lab-scale and on-field tests”,paragraph 2. Due to the presence of 1 kg of organic waste directly in contact with the samples in the on-field test, we believe the risk of reduced contact is minimal. However, the substrate within the mesh bags might have lower disintegration rates considering reduced mixing with the rest of the compost pile. 1. Introduction, first paragraph: I would recommend defining “disintegration” and “composting” carefully. EN 13432 or one of the ISO disintegration testing methods should have these definitions. I disagree with to the use of “only” (“..would ONLY disintegrate significantly..”). A substance’s ability to readily biodegrade under composting conditions does not preclude the substance’s ability to readily biodegrade in a different environment (e.g., soil, activated sludge, or seawater). Definitions for biodegradation and disintegration were added in section “Introduction”, paragraph 1. In addition, a sentence rephrased to “Compostable plastics are a subcategory of biodegradable plastics that disintegrate significantly in specific composting conditions, but not necessarily in general environmental conditions on land or sea.” 2. Introduction, 3rd paragraph: “The disintegration condition in EN 13432 .. after a maximum of 12 weeks”. This sentence needs to be qualified: “..according to the _____ standardized testing conditions” Qualifier added “in a “controlled pilot-scale test” or in

industrial composting plants" 3. More information should be provided on the properties of the polymeric materials, such as weight-averaged molecular weight, glass transition temperature, apparent density (grams per square meter), etc. The weight average molecular weight, glass transition temperature and specific surface area (square centimeter per gram) were added to Table 1. Due to the lack of data, the average molecular weight of BPE-SP-PBS is not available. The glass transition temperature typical for PBS was extracted from literature. The discussion in paragraph 2 and 3 of the section "Disintegration rate in lab-scale industrial composting" was also enhanced using the added information. 4. Methods, Lab-scale composting. Were the composting ingredients or the compost itself treated with sieving prior to the addition of plastic? The substrate mix was not treated with sieving prior to the test. This is because the components were based on ISO 20200 and were sourced from homogeneous sources. From our sieving analysis, at least 95% of the substrate had a particle size less than 1 cm. The compost itself was sieved in the production line of the composting plant. On visual inspection, we did not identify plastic particles that may affect the results of the experiment. 5. Results and Discussion, first paragraph, "current form does not fulfil the 90% disintegration conditions" needs a qualifier, such as "specified in EN 13432" Qualifier added "based on ISO 20200". 6. Please consider merging Figs 2 and 3. The data of Fig 3 can be easily incorporated as datapoints into Fig 2. (Note, for the current Fig 3, y-axis numbers should have the zeroes to the right of the decimal place removed and the caption should indicate the duration.) That is a good point, thank you. We added additional data for a lab-scale test for 3 weeks to Figure 3 reflecting the duration as well as material dimensions used in the on-field test in the industrial composting plant. We also edited the figure as suggested. Due to the differing dimensions of the samples used for the 1st lab-scale test, we prefer to keep the figures separate. 7. Fig 2: Do error bars represent standard deviation, standard error, or something else? Please elaborate (e.g., in the figure caption). Thank you for the reminder, the error bars represent the standard deviation. All figure captions were updated. 8. Results and discussion, "disintegration rate in an industrial composting plant" subsection, bottommost paragraph. This paragraph needs revision. First, as mentioned above, it is likely that the mesh bag promoted mass transfer limitations on the biodegradation reaction. Second, I believe that the non-compliance of PLA to the EN 13432 standards is at least partially attributable to the high, 1 mm, thickness of the films and perhaps the high content of calcium silicate nanofiller may play a role as inhibitor of biodegradation. Are the composting conditions and medium a factor? I find that the inclusion of a positive control into the experimental design, such as a standard plastic material (e.g., a thinner PLA film) improves in the interpretation of the results, although I realize that the ISO standardized tests for disintegration do not directly call for it. Please consider revising the discussion in this paragraph. We tested the blends in the current form as they were prototypes of soft as well as rigid packaging respectively. We acknowledge the role of the blend composition and shape on the results. We appended a note to that paragraph and discussed the potential effects of the sample thickness in the Section "Feasibility of treating compostable plastic packaging in industrial composting plants" , paragraph 4. A comparison considering the filler content was added in the section "Disintegration rate in lab-scale industrial composting", paragraph 3. 9. Feasibility of treating compostable plastic packaging .. " subsection, first paragraph, "do not meet the 90% disintegration requirement". I find this paragraph requires revision, noting first that the PBS material does appear to meet the requirement under laboratory conditions, per Fig 2. The assessment given in this paragraph

needs to take into account the high thickness of the PLA film and the mass transfer limitations imposed by the mesh bag for the data of Fig 3 (as discussed above) We understand and appreciate the comment. The section was revised to highlight the potential effects of sample thickness in paragraph 4. In addition, the wording was enhanced in order to make clear that the results do not represent an absolute negative evaluation but point out the many factors that needs to be additionally considered. A discussion of the potential effect of the mesh bags was also added to the section “Disintegration rate in an industrial composting plant”, paragraph 5 and reinterated in this section.

Competing Interests: No competing interests were disclosed.

Reviewer Report 10 October 2022

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This study compares degradation rate of compostable blends under lab conditions and in industrial composting plants. For this purpose, two different packaging blends, i.e., polylactic acid (PLA) based rigid packaging and polybutylene succinate (PBS) based soft packaging, were used. Lab-scale tests were carried out according to ISO 20200 and samples were recovered after 4, 8 and 12 weeks. On field test lasted for 3 weeks. Both were under thermophilic conditions. Mass loss of samples was calculated to determine degradation.

1. In order to make an accurate comparison of degradation rates between lab conditions and on field conditions, sample recovery time should be the same. Samples were recovered earlier from the industrial composting site compared to lab scale tests. PLA quickly degrades under thermophilic conditions, above its glass transition temperature (T_g), therefore one more week in an industrial composting site under thermophilic conditions would affect the results. It is mentioned that thermophilic phase does not exceed 3 weeks in industrial processes, therefore, samples could have been recovered during the 3rd week of incubation under lab conditions as well so that an accurate comparison between lab conditions and on field conditions can be conducted. Otherwise, there is a duration factor under thermophilic conditions which cannot be omitted. Actually, for better comparison and for research purposes, on field samples should have been collected after 4, 8 and 12 weeks as well.

2. Different dimensions of each blend are used for each degradation environment. To make an accurate comparison of degradation rates between lab conditions and on field conditions, sample dimensions should be the same.

3. Apart from mass change, another degradation assessment method is highly recommended such as a Gel Permeation Chromatography (GPC). Cleaning the debris without compromising the sample is not easy which may lead to inaccurate results. For instance, molecular weights can be compared after 3 weeks of incubation in both conditions.

4. In methods, more detail about the plastics used in this study should also be given in this study as well (Table 1). The information would help to explain the degradation rate of the plastics. For instance, PLA properties such as its molecular weight and T_g greatly affect its degradation rate.

5. A brief comparison between the difference in degradation rate of PLA and PBS can be discussed.

6. Page 3: Please add the reference after these sentences: 'Certified compostable starch-based shopping bags and PLA-based cutlery degraded by 48 wt% and 15 wt% respectively after a combination of a pilot-scale mesophilic dry anaerobic digestion phase (35 days), active composting phase (15 days) and compost maturation phase (40 days). A final concentration of compostable plastic in compost was found to be around 18 wt%.'

7. Page 6: Section 3.3 is mentioned but the section number cannot be found in the main text.

8. Page 8: 'However, due to the very low disintegration rates observed, i.e. all samples were lower than 7%, it is unlikely that the disintegration rates will improve above the 90% disintegration rate required by ISO 20200 and EN 13432.' The assumption drawn here is highly speculative since there is not enough data. No rate is determined, no interpolation is calculated and there are no mathematical models, etc. It is based on only one time sampling at the end of 3rd week. Sampling should have been carried out each week and for a longer period of time to make an estimation.

9. Page 9: Grammar should be revised in the following sentence: 'Further research on the effects of differing composting medium compositions, organic solids content, and particle size distributions are needed to understand the scenarios in which the disintegration rates will meet requirements'.

10. Page 10: In conclusion part, moisture content is another factor that affects degradation. Also, microorganisms and their diversity affect degradation in compost. They should be mentioned.

11. In conclusion part (Page 1), comparing 3rd week in the field and 4th week in the lab is not feasible. Why do the authors think the main reason of different degradation rate is the 'composition of the composting substrate'. The evidence should be provided.

    In the same part, the last sentence should be more clearly presented.

12. In general, this research can be a great contribution to the literature and to the process of industrial composting of certain plastics. The research is clearly and accurately presented but some more fundamental and also recent papers should be cited to better explain the

degradation of the plastics used in this study. Main factors that contribute to their degradation should be presented from the literature.

In literature, PBS and even high molecular PLA degrades fast under thermophilic conditions in compost (Kale et al., 2007). However, in this study less than 5% of PLA blend was degraded in 3 weeks. Weren't samples in full contact with the compost due to mesh bags? Weren't they buried in the pile? The reasons of slower degradation rate in this study should be discussed more in detail and compared with literature. Is there any other study that contained samples in mesh bags, how long did it take for them to degrade?

A significant revision and some additional data mentioned above are necessary for this study.

References

1. Kale G, Auras R, Singh S: Comparison of the degradability of poly(lactide) packages in composting and ambient exposure conditions. *Packaging Technology and Science*. 2007; **20** (1): 49-70 Publisher Full Text

Is the work clearly and accurately presented and does it cite the current literature?
Partly

Is the study design appropriate and does the work have academic merit?
Yes

Are sufficient details of methods and analysis provided to allow replication by others?
Yes

If applicable, is the statistical analysis and its interpretation appropriate?
Not applicable

Are all the source data underlying the results available to ensure full reproducibility?
Yes

Are the conclusions drawn adequately supported by the results?
Partly

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Biotechnology, biopolymers, environmental degradation of plastics.

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard, however I have significant reservations, as outlined above.

Author Response 25 Aug 2023

Zhi Kai Chong

The authors would like to thank Mehlika Karamanlioglu for the detailed review and

constructive comments. The table below summarises the authors' comments and enhancements made to the new version in response to the review. Review Section Authors' replies and comments (1) In order to make an accurate comparison of degradation rates between lab conditions and on field conditions, sample recovery time should be the same. Samples were recovered earlier from the industrial composting site compared to lab scale tests. PLA quickly degrades under thermophilic conditions, above its glass transition temperature (Tg), therefore one more week in an industrial composting site under thermophilic conditions would affect the results. It is mentioned that thermophilic phase does not exceed 3 weeks in industrial processes, therefore, samples could have been recovered during the 3rd week of incubation under lab conditions as well so that an accurate comparison between lab conditions and on field conditions can be conducted. Otherwise, there is a duration factor under thermophilic conditions which cannot be omitted. Actually, for better comparison and for research purposes, on field samples should have been collected after 4, 8 and 12 weeks as well. (2) Different dimensions of each blend are used for each degradation environment. To make an accurate comparison of degradation rates between lab conditions and on field conditions, sample dimensions should be the same. The lab-scale test was done mainly to check the disintegration potential of the samples in a controlled environment. However, we completely understand and agree with the downside to the duration mismatch for comparison between the two types of tests. To facilitate a better comparison between the industrial-scale and lab-scale tests, an additional test in the lab based on ISO 20200 was conducted with a duration of 3 weeks to match the duration exposed in the industrial plant and added to the new version of the paper. Sample dimensions identical to those in the field test were used. The results and discussions were reevaluated based on the new data and described in the Section "Disintegration rate in an industrial composting plant". It was observed that the lab-scale test according to ISO 20200 showed higher degradation and disintegration after 3 weeks. Conducting an experiment in the industrial plant for 4, 8 and 12 weeks was unfortunately not currently possible due to the high complexity of integrating the sample exposure method into the workflow of the plant. This is a potential point for future research. (3) Apart from mass change, another degradation assessment method is highly recommended such as a Gel Permeation Chromatography (GPC). Cleaning the debris without compromising the sample is not easy which may lead to inaccurate results. For instance, molecular weights can be compared after 3 weeks of incubation in both conditions. That is a really good point, however, a GPC test is not currently possible from our side. In our paper, the main measure of degradation is the mass loss with a cutoff threshold > 2 mm and we based our discussion on this measure. (4) In methods, more detail about the plastics used in this study should also be given in this study as well (Table 1). The information would help to explain the degradation rate of the plastics. For instance, PLA properties such as its molecular weight and Tg greatly affect its degradation rate. The average molecular weight was added for BPE-RP-PLA whilst the glass transition temperature was added for both BPE-RP-PLA and BPE-SP-PBS in Table 1. The average molecular weight for BPE-SP-PBS is not available. In addition, the effect of glass transition temperature on degradation was added to paragraph 7 of the Section "Introduction". The starting molecular weight of BPE-RP-PLA (237000 g/mol) was similar to a study (Kale et al., 2007) with 230000 g/mol, which reported complete disintegration within 30 days in a 65°C environment. However, the blend composition is different and the thickness was not mentioned in the study. Thus, making a direct comparison is not possible. In our study, the samples disintegrated significantly in the lab-

scale (58 °C). On the other hand, degradation was observed with minor disintegration (mass loss) after the on-field test (64 °C). We explained possible reasons for this discrepancy in the Section “Differences in composting conditions between the lab-scale and on-field tests”. (5) A brief comparison between the difference in degradation rate of PLA and PBS can be discussed. Thank you for the comment, a section discussing the contributing factors to the differing disintegration rate of BPE-RP-PLA and BPE-SP-PBS was added in Paragraph 3 of the Section “Disintegration rate in lab-scale industrial composting”. (6) Page 3: Please add the reference after these sentences: ‘Certified compostable starch-based shopping bags and PLA-based cutlery degraded by 48 wt% and 15 wt% respectively after a combination of a pilot-scale mesophilic dry anaerobic digestion phase (35 days), active composting phase (15 days) and compost maturation phase (40 days). A final concentration of compostable plastic in compost was found to be around 18 wt%.’ The sentence was rephrased mentioning the source, which was (Cucina et al., 2021). (7) Page 6: Section 3.3 is mentioned but the section number cannot be found in the main text. “Section 3.3” was corrected to “Differences in composting conditions between the lab-scale and on-field tests”. (8) Page 8: ‘However, due to the very low disintegration rates observed, i.e. all samples were lower than 7%, it is unlikely that the disintegration rates will improve above the 90% disintegration rate required by ISO 20200 and EN 13432.’ The assumption drawn here is highly speculative since there is not enough data. No rate is determined, no interpolation is calculated and there are no mathematical models, etc. It is based on only one time sampling at the end of 3rd week. Sampling should have been carried out each week and for a longer period of time to make an estimation. The statement was removed. (9) Page 9: Grammar should be revised in the following sentence: ‘Further research on the effects of differing composting medium compositions, organic solids content, and particle size distributions are needed to understand the scenarios in which the disintegration rates will meet requirements’. The statement was rephrased to “However, research on the effects of differing substrate composition, particle size distribution and other parameters such as total volatile solids is lacking and important to understand the requirements for sufficient disintegration in actual plants as well as to understand the potential differences between plants.” (10) Page 10: In conclusion part, moisture content is another factor that affects degradation. Also, microorganisms and their diversity affect degradation in compost. They should be mentioned. Thank you for the comment, the factors were mentioned as suggested and added in the conclusion. (11) In conclusion part (Page 1), comparing 3rd week in the field and 4th week in the lab is not feasible. Why do the authors think the main reason of different degradation rate is the ‘composition of the composting substrate’. The evidence should be provided. In the same part, the last sentence should be more clearly presented. The conclusion was revised and further elaborated addressing the comment with the new data from 3 weeks composting in the lab-scale: “Based on the data in this study, the abiotic conditions such temperature, moisture content and pH in the lab-scale was not notably better compared to on-field. The methods used also ensured that at least 1 kg of composting substrate was in direct contact with the plastic samples in all cases. This point towards the potential role of other composting parameters such as the process scale, substrate composition and particle size distribution in the rate of disintegration. These factors may vary for different plants and seasonally. In addition, the presence and diversity of microorganisms play a role.” (12a) In general, this research can be a great contribution to the literature and to the process of industrial composting of certain plastics. The research is clearly and accurately presented but some more fundamental and also recent papers

should be cited to better explain the degradation of the plastics used in this study. Main factors that contribute to their degradation should be presented from the literature. (12b) In literature, PBS and even high molecular PLA degrades fast under thermophilic conditions in compost (Kale et al., 2007). However, in this study less than 5% of PLA blend was degraded in 3 weeks. Weren't samples in full contact with the compost due to mesh bags? Weren't they buried in the pile? The reasons of slower degradation rate in this study should be discussed more in detail and compared with literature. Is there any other study that contained samples in mesh bags, how long did it take for them to degrade? Thank you for the comment. To enhance the new version, fundamental papers on degradation mechanics as well as more comparable composting studies were described and cited in the Section "Introduction": paragraph 5 to 8. In addition, paragraphs 2, 3 and 4 of the Section "Differences in composting conditions between the lab-scale and on-field tests" were enhanced. The authors think that this risk is minimal as the samples in this study were fully in contact with the organic waste as 1 kg of organic waste was enclosed together with the samples within the mesh bags. These bags were then placed in cages together with more organic waste. The cages were buried in the middle of the composting pile. A summary of other studies was added to Paragraph 5 and 6 on the introduction. In addition, a comparison with other studies of industrial composting was added to paragraph 6 of the Section "Disintegration rate in an industrial composting plant" though it should be noted that a direct comparison of on-field composting studies is difficult due to the differing sample dimensions, blend composition, substrate composition and general composting methods. References Cucina, M., Nisi, P. de, Trombino, L., Tambone, F., & Adani, F. (2021). Degradation of bioplastics in organic waste by mesophilic anaerobic digestion, composting and soil incubation. Waste Management (New York, N.Y.), 134, 67–77. https://doi.org/10.1016/j.wasman.2021.08.016 Kale, G., Auras, R., Singh, S. P., & Narayan, R. (2007). Biodegradability of polylactide bottles in real and simulated composting conditions. Polymer Testing, 26(8), 1049–1061. https://doi.org/10.1016/j.polymertesting.2007.07.006

Competing Interests: No competing interests were disclosed.