Field test on the biodegradation of poly(butylene adipate-*co*-terephthalate) based mulch films in soil

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ABSTRACT

The use of plastic much films has been fundamental to promoting food production in many regions of the world. However, concern is growing about the progressive accumulation of plastic residues in soil after crop harvest and its subsequent impact on soil health and potential to enter the food chain. Although biodegradable films have been developed to prevent these problems, it is still unclear whether they are environmentally benign. Here we evaluated the physical and chemical breakdown of four commercial poly(butylene adipate-*co*-terephthalate) based biodegradable mulch films (BMF1, BMF2, BMF3 and BMF4) in an agricultural soil over a 26-month period. Based on visual examination, degradation followed the series BMF4 > BMF1, BMF2 > BMF3. Importantly, microplastic residues (fragments <5 mm) still remained in the soil of all 4 plastic types after 2 years, suggesting that they are likely to accumulate over time if used on an annual basis. Viscosimetry, Fourier transform infrared (FTIR) spectroscopy and Thermogravimetric Analysis (TGA) were used to characterise the breakdown process. Our results indicated that the degradation of the mulch film after burial in agricultural soil may be linked to the nature of the polymer but also to its manufacturing formulation. Although the peak changes of polyester in the infrared spectrum were not distinct, the plastic films showed other signs of degradation including a reduction in intrinsic viscosity after burial in soil. The different degradation rates of BMF1 and BMF2 at the molecular level may be due to the different $CaCO_3$ contents. In conclusion, under field conditions, we show that slight variations in the formulations of commercial biodegradable mulch films leads to very different persistence rates in soil. Further, we conclude that their slow rate of degradation will ultimately lead to their progressive accumulation in soil if used repeatedly.

1. Introduction

Plastic film mulches are routinely used in China to promote food security, with an annual use of nearly 1.5 million tons, covering a total land area of 18.4 million ha [1]. These films increase crop yields by promoting water and nutrient use efficiency as well as reducing soil erosion and disease incidence, allowing the more efficient use of pesticides [2,3]. Due to the persistence of plastic film residues in soil and their progressive accumulation (i.e. "white pollution"), their long-term use is not viewed as sustainable [1]. In addition, there are increasing concerns about the potential negative effects of microplastic residues in soil due to their potential to adversely affect soil health and enter the

human and marine food chain [4–6]. One of the key debates is therefore whether the short-term gains in food production due to plastic film use are enough to outweigh the potential long-term risks to soil functioning and organism health. These problems are compounded by the practical difficulties in recovering plastic films from fields and the lack of recycling facilities capable of handling plastics contaminated by soil [7]. In addition, improper recycling or disposal of waste agricultural plastic has also been shown to cause secondary environmental pollution [2,8].

One potential solution to help reduce plastic residue pollution at source, is the use of biodegradable mulch films which can degrade within years [9,10], rather than centuries [3,11,12]. The production of biodegradable films, which partially or completely degrade through the

action of sunlight and soil microorganisms, has grown in importance in recent years [9,11]. These materials are often synthesized from renewable materials and eliminate the need for both recovery and final disposal, since they are designed to degrade *in situ* [13]. However, whether these products are environmentally benign remains unknown. For example, it is unknown whether they also produce microplastics, how long these persist and whether they pose an environmental risk? It is therefore necessary to determine the factors regulating the rate of degradation of different biodegradable plastics in agricultural soils to allow their environmental impact to be critically evaluated and also to select the optimal type of biodegradable plastics for different cropping regimes.

The degradability of biodegradable films depends on a range of factors including: (i) the physical structure and chemistry of the plastic polymer, (ii) soil type and quality, (iii) agricultural management regime, and (iv) prevailing climate [13,14]. The degradability of biodegradable plastic is normally measured by burying the materials in soil for known times, followed by recovery and measurement of mass loss [15]. In addition, the mechanistic basis underpinning the breakdown process can be evaluated using FTIR and viscosimetry to determine changes in polymer molecular weight and chemical structure [16–18]. The aim of this study was therefore to determine the breakdown of 4 different commercial biodegradable mulch films. We hypothesized that fundamental differences in their formulation would lead to significant variation in their degradation rate determined visually and using viscometry, FTIR and thermogravimetric analysis (TGA).

2. Materials and methods

2.1. Site description and experimental material

The experimental site was located at the Shunyi Station of the Chinese Academy of Agricultural Sciences, Beijing, China (40°09′N, 116°52′E). The site is characterized by a warm, temperate semi-humid monsoon climate, controlled by Mongolian high pressure in winter (cold and dry), and southeast monsoon in the summer (hot and wet). The site is located 20 m above sea level with an annual average temperature of 11.5 °C. The average monthly temperature in January is −4.9 °C (minimum −19.1 °C) while in July it is 25.7 °C (maximum 40.5 °C), with 195 frost-free days per annum and sunshine of 2750 h year^{-1}. The average annual rainfall is 625 mm, of which 75% falls from June to August, and the mean annual relative humidity is 50%. The weather conditions during the trial are presented in Fig. S1. The soil at the site is classified as a meadow cinnamon soil (silt 64.2%, sand 28.7%, and clay 7.1%) used for the production of winter wheat (*Triticum aestivum* L.) and summer corn (*Zea mays* L.). The initial properties of the soil (0–25 cm depth) at the start of the experiment were as follows: total organic C of 14.1 g kg^{-1}; total N of 0.86 g kg^{-1}; available N, P, and K of 158.3, 46.1, and 137.2 mg kg^{-1}, respectively; bulk density of 1.20 g cm^{-3} and a pH value of 7.89 (methods of analysis are listed in Table S1). The four types of degradable plastic mulch films used in the experiment were BMF1 (BASF, China), BMF2 (Guangdong Kingfa Sci. and Tech. Co., Ltd.), BMF3 (Jiangsu Zhongke Jinlong-CAS Chemical Co. Ltd.), BMF4 (Shandong Yikeman Tech. Co., Ltd.). Information and testing methods for

mechanical properties of the four mulch films in this study are provided in Table 1 and Table 2, respectively.

2.2. Experimental design

The experiment consisted of four treatments each with three replicates. Samples of each biodegradable film were cut into strips (30 × 45 cm), placed into marked polypropylene mesh bags (35 × 55 cm) and buried at a soil depth of 10 cm in replicate plots measuring 2.2 × 2.5 m in August 2015 (Fig. S2). The trial lasted a total of 26 months, with replicate groups of samples collected at 2, 7 and 26 months after burial. The samples were then transferred to the laboratory to evaluate film degradation, structural degradation and changes in viscosity.

2.3. Effect of burial time on the degradation of plastic films

Visual scoring was used to record changes in color, morphology and the surface integrity of the plastic films. The breakdown of the plastic mulch film was categorized into five phases as follows: Stage 1 (Crack initiation phase) when small cracks (ca. < 2 cm wide) start to appear in the film; Stage 2 (Macro-cracking phase) when large cracks (ca. 2–20 cm wide) are clearly visible; Stage 3 (Fragmentation phase) when the film has cracked and broken into large pieces; Stage 4 (Disintegration phase) when only small fragments remains (i.e. microplastics), and Stage 5 (Disappearance phase) when the mulch film is no longer visible to the naked eye.

After retrieval from the soil, any soil adhering to the film surface was removed by washing with distilled water. The weight of mulch film remaining was then determined by air-drying at room temperature. Film weight loss (WL) was used as an indicator of degradation using the following formula: WL (%) = $(M_0-M_1)/M_0 \times 100$, where M_0 (g) and M_1 (g) is the dry weight of the mulch film before and after burial. These measurements were only possible for the 2-month burial time point, as it proved impossible to recover the fragmented plastic film after this time.

Changes in intrinsic viscosity were used to estimate the reduction in

Table 2
Methods used for the determination of the thickness and mechanical properties of the biodegradable films according to the standard GB/T 35795-2017 [19]. All tests were repeated on 5 separate samples.

Item	Method
Thickness	Measured using a Millimar C1200 Thickness Gauge (Mahr Ltd., Germany).
Tensile load	Evaluated using an Intelligent Electronic Tensile Testing Machine (Labthink Co. Ltd., China).
Tensile strength	Tensile strength (TS) calculated according to the tensile load (F), sample width (L) and thickness (T) using the formula TS = F/(L × T)
Nominal tensile strain at break	Initial distance between the fixtures set at 50 mm, stretch speed 500 mm/min with sample stretched to break point.
Right angle tearing load	Test speed set at 200 mm/min with each sample tested at vertical/horizontal positions.
Average tear strength	Tear strength (δ) was calculated according to the formula δ = P/D (P indicated tearing load and D indicated sample thickness).

Table 1
Information of the four different types of commercial plastic mulch films used in the field study.

Item	Main composition provided by the manufacturer	Color	Thickness (μm)	Tensile load (N)	Tensile strength (MPa)	Nominal tensile strain at break (%)	Right Angle tearing load (N)	Average tear strength (kN/m)
BMF1	PBAT (60%)	White	10.5	1.90/1.70	21.8/15.5	260/275	1.2/1.1	130/125
BMF2	PBAT (70%)	White	11.2	1.60/1.55	20.5/14.2	270/280	1.1/1.0	128/120
BMF3	PBAT(70%)+PPC(20%)	White	11.5	1.55/1.40	18.3/13.8	230/260	0.9/0.8	100/95
BMF4	PBAT(60%)+PHA(20%)	White	10.5	1.50/1.40	19.5/13.4	234/265	0.8/0.7	115/93

Notes: BMF: Biodegradable mulch film; PBAT, Poly(butylene adipate-*co*-terephthalate); PPC, Polypropylene carbonate; PHA, Polyhydroxyalkanoate. Data before and after "/" means the vertical/horizontal mechanical property.

molecular weight (M) of the biodegradable plastics [20,21]. The relationship between intrinsic viscosity and molecular weight conforms to the Mark-Houwink equation:

$$[\eta] = KM_\eta^\alpha$$

where $[\eta]$ is intrinsic viscosity (unit is dL/g), the specific viscosity as the polymer solution concentration approaches zero; K and α were constants independent of molecular weight. The intrinsic viscosity of the plastic mulch films was determined by an Ubbelohde viscometer with para-xylene dissolved plastic materials at 125 °C. The intrinsic viscosity of BMF3 could not be determined by this method due to the chemical nature of the product.

Fourier transform infrared (FTIR) spectroscopy was used to identify changes in the types and amounts of chemical bonds present in the plastic film after burial in soil. The FTIR spectra of the plastic mulch films (ca. 2 mg) was determined using a Nicolet IS10 spectrometer (Thermo-Fisher, Waltham, MA, USA). The FTIR spectra were recorded between 4000 and 400 cm^{-1} wavenumber, taking the average of 32 scans and operating with a 4 cm^{-1} resolution.

Thermogravimetric Analysis (TGA) was used to measure and record the sample mass change with temperature during the thermal degradation reaction [22]. TGA of different samples in our study was carried out using a thermal analyzer (STA 449 F3 Jupiter®; Netzsch-Gerätebau GmbH, Selb, Germany). Samples of about 4 mg were heated at a heating rate of 20 °C min^{-1} from room temperature up to 800 °C under a N$_2$ atmosphere.

3. Results

3.1. Effect of soil burial time on biodegradable film breakdown

Our results showed that the disintegration of BMF4 was fastest, BMF3 was slowest, and the other two formulation films (BMF1 and BMF2) containing only PBAT polyester components were intermediate (Fig. 1). In this trial, BMF1 and BMF2 showed signs of cracking by month 2, with large cracks evident by month 7, and complete fragmentation apparent by month 26 (Table 3). In contrast, small amounts of cracking were only evident in BMF3 by month 26. The BMF4 film showed the fastest signs of crack propagation, with complete fragmentation evident by month 7. The corresponding mass losses of BMF2, BMF4, BMF1 and BMF3 after burial in soil for 2 months were 12.7%, 11.6%, 4.72% and 6.32% respectively.

Table 3
Visual score of the physical breakdown of four different commercial bioplastic mulch films after burial in soil for up to 26 months.

Treatment	Time since burial in soil (months)		
	2	7	26
BMF1	2	3	4
BMF2	2	3	4
BMF3	0	0	1
BMF4	3	4	4

Notes: Visual scoring schema: 1, Crack initiation phase; 2, Macro-cracking phase; 3, Fragmentation phase; 4, Disintegration phase; 5, Disappearance phase.

3.2. Change in biodegradable film intrinsic viscosity after different burial times in soil

Intrinsic viscosity varied depending on the type of plastic mulch film and decreased with increasing burial time in soil (Table 4). From month 2–7, the change in intrinsic viscosity of BMF4 was the smallest, decreasing from 235.0 dL/g to 174.8 dL/g, a reduction of 25.6%. The reduction for BMF1 was greater than for BMF2, decreasing by 51.2% and 31.1% respectively over the same period, while the intrinsic viscosity of BMF2 decreased by a further 34.4% from month 7–26. As the degraded film residues could not be collected, the intrinsic viscosity of BMF1 and BMF4 at month 26 could not be determined.

3.3. Chemical changes in biodegradable film after different burial times in soil made by FTIR

The FTIR fingerprint (Fig. 2) of BMF1 and BMF2 showed distinct peaks at 2960, 1710, 1578, 1504, 1410, 1390, 1100, 1020, 873, and 727 cm^{-1}. The peak assignments in FTIR spectra are described in Table 5. In particular, the peaks at 1450 cm^{-1} (trans O–CH$_2$ deformation vibration in the plane) and 1270 cm^{-1} (C–O stretching vibration in aromatic ester)
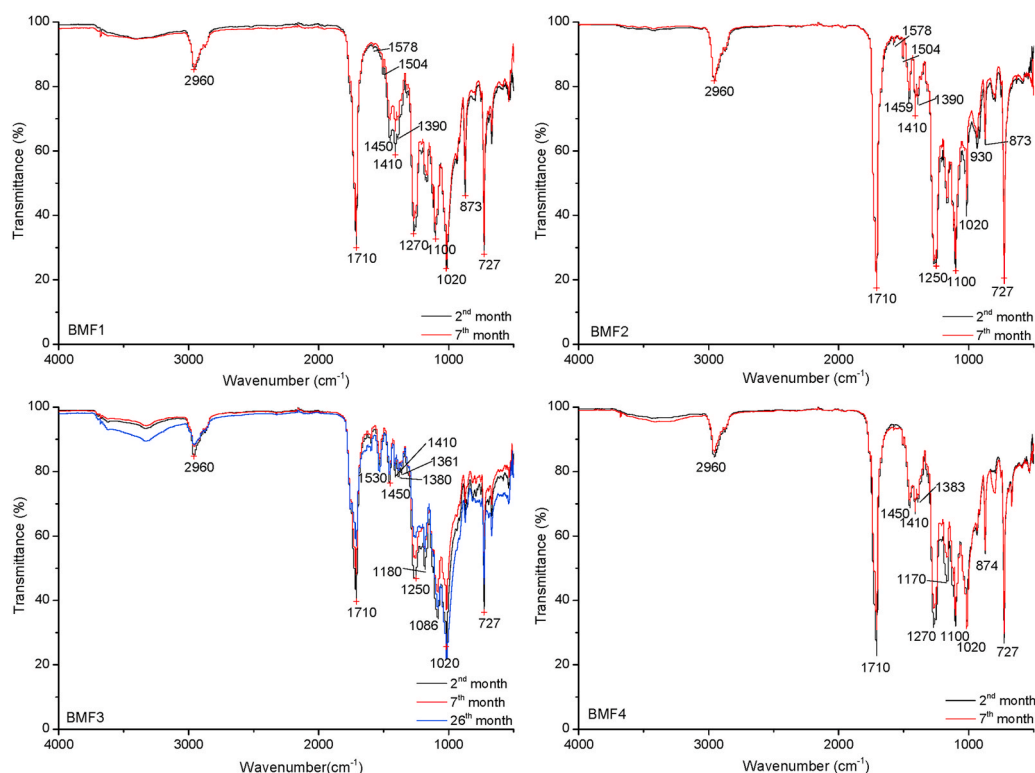
Table 4
Changes on intrinsic viscosity of four different bioplastic mulch films after burial in soil for up to 26 months.

Treatment	Time since burial in soil (months)		
	2	7	26
BMF1	383.3	187.2	–
BMF2	371.7	256.1	168.0
BMF3	–	–	–
BMF4	235.0	174.8	–
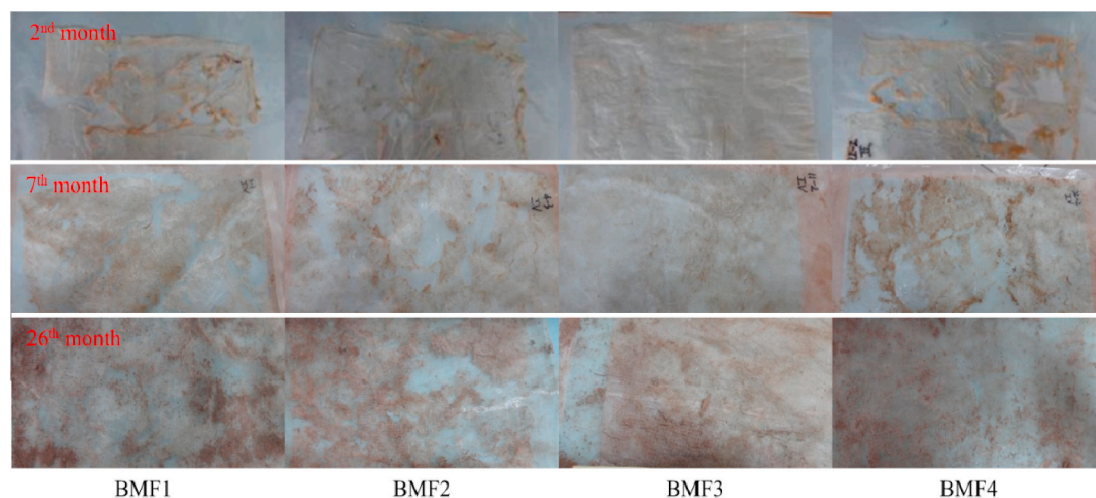
Fig. 1. Photographs showing the influence of soil burial time on the degradation of four commercial biodegradable mulch films (BMF1-4).

Fig. 2. FTIR-based determination of structural changes of four different commercial formulations of biodegradable mulch film (BMF1, BMF2, BMF3, BMF4) after burial in an agricultural soil for either 2, 7 or 26 months. In some cases, recovery was not possible at month 26 due to excessive film degradation preventing recovery.

Table 5

Peak band assignments in Fourier transform infrared (FTIR) spectra for four different types of commercial biodegradable plastic film.

Wavenumbers (cm^{-1})	Assignment	Vibration	Reference
2960	C–H	asymmetric stretching vibration	[23]
1710	C=O	symmetrical stretching vibration	[24]
1578	benzene	skeleton vibration of benzene ring	[25,26]
1504	benzene	skeleton vibration of benzene ring	[14,25, 26]
1459	CH$_2$	in-plane deformation vibration	[14,26]
1450	O–CH$_2$	trans deformation vibration in the plane	[27]
1410	C–H	in-plane deformation vibration	[14]
1390	C–H	wagging vibration	[28]
1380	CH$_3$	symmetrical deformation vibration	[27,29]
1361	C–H	superposition of C–H deformation vibration in the plane and CH$_2$ sway vibration out the plane	[29]
1270	C–O	stretching vibration in aromatic ester	[26,29]
1250	C–O–C	antisymmetric stretching vibration in aliphatic eater	[29]
1100	C–O–C	stretching vibration in aliphatic	[28–30]
1020	=C–H	in-plane deformation vibration of two adjacent surfaces on the p-substituted benzene ring	[14,26, 31]
930	C–O	trans telescopic vibration	[26]
873	C–H	off-plane deformation vibration	[26]
727	C–H	off-plane deformation vibration	[14,26]

were attributable to the presence of BMF1, however, the peaks for BMF2 were found to be associated with bands at 1459 cm^{-1}, 1250 cm^{-1}, and 930 cm^{-1}. In the infrared spectra of BMF3, the characteristic peaks at 1380 cm^{-1} and 1361 cm^{-1} were also found. In the BMF4 spectra, 1450 cm^{-1} is the superposition of CH$_3$ symmetrical deformation vibration and

CH$_2$ in-plane deformation vibration, 1100–1270 cm^{-1} is C–O–C stretching vibration and CH$_2$ sway vibration out of the plane.

Based on their FTIR spectra, there were no distinct changes in the peaks at 1710 cm^{-1} of BMF1, BMF2, BMF3, and BMF4 at months 2 and 7. However, in the BMF3 film, the ester peak at 1710 cm^{-1} (aliphatic C=O) and 1250 cm^{-1} (aliphatic C–O) gradually decreased with increasing burial time up to 26 months. The structures of BMF1, BMF2, and BMF4 after burial for 26 months could not be measured due to the difficulty in recovering samples from the field.

3.4. Thermogravimetric analysis (TGA) of BMF1 and BMF2

According to the results presented above, the degradation trends of BMF1 and BMF2 were similar, with their degree of degradation dependent on burial time. We attributed this to the different formulations of the two plastic films which originated from different manufacturers. Thermogravimetric analysis was subsequently used to evaluate differences in their formulation chemistry (Fig. 3). Both samples showed a strong degradation peak at 400 °C, corresponding to the decomposition temperature for PBAT polymer. The percentage of PBAT is about 60% in BMF1 film and 70% in BMF2 films; the residual calcium carbonate (CaCO$_3$) and their decomposition products in BMF1 and BMF2 represented 21.2% and 13.3%, respectively. Based on the small amount of residual film components remaining after burning at 800 °C, we estimate that the two films contain about 30% and 20% CaCO$_3$, respectively. The small peak at 520 °C may be attributed to the degradation of other ingredients in the plastic film. These include trace filler auxiliaries for which the same thermal decomposition trend was observed in the two TGA spectrograms.

4. Discussion

In the new European Union (EU) standard for biodegradable mulch films (European Committee for Standardization, EN 17033:2018), it is
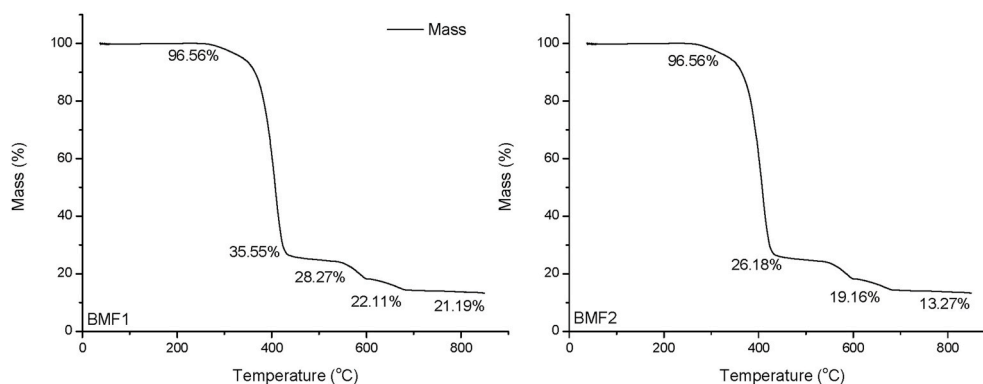
Fig. 3. Thermogravimetric Analysis of two different types of commercial biodegradable mulch film (BMF1 and BMF2) before burial in soil.

stated that mulch films should be considered compliant if they can demonstrate a satisfactory rate and level of biodegradation when buried in soil. Therefore, selecting a representative outdoor soil burial test is necessary to critically evaluate the plastic film biodegradation properties [32,33]. Preferably these should be done under realistic field conditions, rather than in the laboratory where conditions can be optimized (i.e. temperature and moisture) to promote faster degradation. Our view is that this should not be based on CO_2 evolution as this is subject to bias from both negative and positive soil organic matter priming and immobilization of mulch-derived C by the microbial biomass (which will not be recovered as CO_2). We do acknowledge that some of the uncertainty could be removed through the use of [14]C- or ^{13}C-labelled mulch films (allowing specific measurement of mulch-derived $^{13}CO_2$ or $^{14}CO_2$), however, these are not available at the present time. We therefore used alternative methods to evaluate degradation rate. For films buried in soil, the reduction in molecular weight is due to hydrolysis, particularly the progressive erosion of the mulch film surface [15]. Here intrinsic viscosity was used to capture changes of molecular weight in the different degradable plastic films. We showed that the intrinsic viscosity of each plastic mulch film decreased with increasing soil burial time, and that this corresponded well to the degree of degradation [21].

FTIR analysis produced a number of peaks reflecting the complex nature of the polymer film [34,35], and provides a complementary and sensitive analytical method to analyze the degradability of plastic mulch film at different soil burial times [15,29]. In our soil burial test, the FTIR changes of the three films (BMF1, BMF2 and BMF4) were not found to be distinct at either month 2 or 7. At this stage the FTIR spectrum still reflected the infrared structural fingerprint of the native polyester compounds, with little displacement in the absorption peak to other wavenumber associated with a reduction in molecular weight. For the BMF3 film, the absorbance peaks also exhibited no clear changes after burial in soil for 2 or 7 months, but the ester peak intensities at 1710 cm^{-1} (aliphatic C=O) and 1250 cm^{-1} (aliphatic C–O) decreased after 26 months, indicating the occurrence of random chain scission from hydrolysis [15]. This was consistent with the visual score we used to reflect the disintegration trend of BMF3. Although the four degradable mulch films underwent progressive disintegration and degradation, the burial time did not significantly change the functional groups in the polymers reflecting their slow degradation in soil.

Microplastic residues of all 4 plastic types still remained in agricultural soil after 2 years, suggesting that they are likely to accumulate over time if used on an annual basis. Therefore, research is needed to explore the ecological risks of biodegradable film residues in soil [36]. Once this is better understood, their potential to produce micro- and nano-plastic biodegradation products in soil should be incorporated into regulatory standards.

The degradation degree of the four plastic films generally increased in the order of BMF4 > BMF1 > BMF2 > BMF3. We attribute the faster degradation of BMF4 to the presence of PHA which is known to be

rapidly degraded in soil. PHA is an intracellular polyester synthesized by microorganisms and serve as a long-term energy storage source. Consequently, a very wide range of organisms in soil possess the enzymatic machinery necessary to hydrolyze and utilize PHA-derived C [37–39].

The new EU standard for bioplastics specifies the methods and evaluation criteria for evaluating biodegradable mulch films destined for use in agriculture (e.g. biodegradation, ecotoxicity, film properties, and constituents). It is therefore implicit that biodegradable mulch films need to not only meet the agronomic requirement, but also that they can be left on the field and ploughed into the soil at the end of the crop cycle without polluting the agricultural environment. In terms of degradation rate alone, BMF4 represents a promising biodegradable mulch film, possessing the fastest degradation rate in our study. However, it is also necessary to consider whether the rate of degradation matches the cropping cycle [40,41]. In the case of BMF3, it is likely that this will last throughout the entire cropping cycle relatively intact, however, its repeated use will lead to a net accumulation in the soil. It would therefore be desirable if we could control the breakdown rate of degradable mulch films such that mulch films could be better targeted to specific crops which may remain in the ground from a matter of weeks (e.g. salad vegetables), to months (e.g. beans, maize), to several years (e. g. vines, fruit trees) [41].

In our FTIR spectrogram, all of the four biodegradable mulch films had no absorption peaks above 3000 cm^{-1}, indicating that there were no unsaturated bonds in their main chains; and the peak of 1710 cm^{-1} in the spectra indicated that O=C–O; the strong absorption peaks at 1400-1600 cm^{-1} indicated that there were benzene rings in their main chains. Thus, we can confirm that all of the four biodegradable mulch films used here contained PBAT polyester components. In addition, the peaks at 1410 cm^{-1} and 873 cm^{-1} indicated that the biodegradable films filling materials contained a certain amount of $CaCO_3$. Since different filling materials have specific decomposition peaks at different temperatures, TGA was used to further determine the composition of the degradable mulch film. Our FTIR spectra and TGA curve showed that neither BMF1 nor BMF2 had obvious polyethylene components, which were mainly composed by aliphatic polyester, $CaCO_3$, and other trace filler auxiliaries. The content of $CaCO_3$ in the two degradable films was about 30% and 20%, respectively, which might be the main factor leading to the different degradation degrees of these mulch films.

Some researchers have reported that $CaCO_3$ could affect the physical properties of plastic materials [42–44]. Al-Salem et al. [44] indicated that micrographs of plastic films with low $CaCO_3$ contents show a very smooth surface appearance and that these had minimal surface deterioration. The $CaCO_3$ filler, typically used to help reduce production costs, density and strength properties in commercial plastic materials, seemed to hinder surface cracking. However, Saowaroj et al. [42] reported that the inclusion of $CaCO_3$ creates voids which might promote microbial access to the interior of the film. Therefore, while we conclude that the

presence of $CaCO_3$ may directly impact on the degradability of plastic film, our understanding of this process remains uncertain and required further work.

Biodegradation in soil or compost depends on a complex synergy of biological and abiotic degradative processes [3,45]. The biodegradability of plastic film mainly depends on three factors: (l) the molecular structure and size of plastics; (2) the size and structure of the microbial community; (3) environmental factors, including humidity, temperature, pH, availability of nutrients, etc. [29,46,47]. Therefore, biodegradation is typically carried out in the dark (no UV exposure) at high humidity and in the presence of large amounts of inorganic salts and available carbon [28,48]. The degradation of the mulch film in soil burial is predominantly affected by soil microorganisms and soil moisture, with microbial activity directly related to soil moisture and temperature. Borrowman et al. [49] reported that moisture content was the most important environmental factor controlling polymer biodegradation rate. Although this is likely to be very important in semi-arid environments, extreme dryness was not a feature of the soils used here. In the initial stages, microorganisms, especially bacteria have been shown to be critical in the decomposition of biodegradable plastic film [45,49]. Both bacteria and fungi can use metabolize C contained in biodegradable film polymers [39], especially when they contain naturaly courring polymers such as in BMF4. Once the mulch films have fragmented, it is possible that direct ingestion by mesofauna (e.g. earthworms) plays an important role in the biodegradation process, however, little work has been undertaken on this to date. In addition, further work is needed to determine the rate of conversion of biodegradable mulch film fragments from micro-to nano-sized particles and the impact of this on their degradation rate, effect on soil organisms and potential for downward migration in the soil profile.

5. Conclusion

Here we evaluated the degradation of four commercial poly (butylene adipate-*co*-terephthalate) based biodegradable mulch films. We showed that they possessed very different degradation rates. We attribute this to differences in their chemistry (e.g. the presence of natural microbial polymers and $CaCO_3$ fillers). Although mulch film degradation was observed (i.e. reduction in molecular weight and physical fragmentation) no distinct changes in polyester functional groups were observed using FTIR. Visible microplastics residues (size < 5 mm) still remained in soil after a 2-year period suggesting that their repeated use will lead to their progressive accumulation of plastic in soil. To improve our understanding of mulch film behaviour in soil, further work is needed to identify the mechanisms and microorganisms involved in the process under field conditions. This will allow the better design of mulch films targeted at specific cropping applications. Additionally, long term studies are needed to gain a better understanding of the impacts of biodegradable plastic films on soil microbial activity and nutrient biogeochemistry.

CRediT authorship contribution statement

Ruimin Qi: Conceptualization, Investigation, Data curation, Formal analysis, Writing - original draft. **Davey L. Jones:** Writing - review & editing, Funding acquisition. **Qiuyun Liu:** Writing - review & editing. **Qin Liu:** Investigation. **Zhen Li:** Investigation. **Changrong Yan:** Conceptualization, Resources, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.polymertesting.2020.107009.

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