

**Assessing the anaerobic degradability and the potential
recovery of biomethane from different biodegradable
bioplastics by a full-scale approach**

Mirko Cucina¹, Gabriele Soggia¹, Patrizia De Nisi¹, Andrea Giordano², Fabrizio
Adani^{1*}

¹Gruppo Ricicla Lab. – DiSAA – Università degli Studi di Milano, Via Celoria 2, 20133

Milano, Italy

²Acqua & Sole Srl - Via Giulio Natta, 27010 Vellezzo Bellini (PV), Italy

*Corresponding author email address: fabrizio.adani@unimi.it

Abstract

The aim of the present study was to evaluate the anaerobic degradability and the potential recovery of biomethane from different bioplastics using a full-scale approach. Bioplastics were placed inside a real anaerobic digestion plant working under thermophilic conditions and quantitative and qualitative degradation of bioplastics was evaluated. Laboratory-scale experiments were used to determine the amount of biomethane produced by anaerobic degradation of bioplastics. Polylactic acid-based items may degrade completely using retention times compatible with anaerobic digestion plants contributing positively to biomethane production, i.e., in 90 days $397 \pm 8 \text{ NL CH}_4 \text{ kg}_{\text{volatile solids}}^{-1}$ were produced by polylactic acid-based cutlery. Starch-based shoppers showed a quick degradation of the starch component in the first month of anaerobic digestion, followed by a slow degradation of the polyester component. Anaerobic digestion and/or anaerobic digestion coupled to digestate composting may represent the best strategy to dispose these wastes meeting the principles of Circular Economy.

Keywords

Biopolymers; Circular economy; Polylactic acid; Starch-based bioplastics; Waste management

1. Introduction

Plastic is a category of materials that comprises several synthetic polymers. Despite the usefulness and convenience of these materials, their production and end-of-life management pose a serious threat to the environment and living organisms, mainly due to green-house gases emission and environmental pollution (Bellasi et al., 2020; Cazaudehore et al., 2021; Emadian et al., 2017; Matthews et al., 2021; RameshKumar et al., 2020). Geyer et al. (2017) estimated that of the total plastic produced from its invention to 2015, 79 % was accumulated in landfill or reached the environment (mainly the oceans), 12 % was incinerated, while only 9 % was recycled. In this context, bioplastics are gaining interest due to policies that impose bans on conventional single-use plastic items (European Parliament, 2019), and to the perception of being more environmental-friendly, making them the best alternative to conventional plastics (Taufik et al., 2020). For instance, in 2020, a global capacity of bioplastics' production of about 2 million tonnes of bioplastics was reported (European bioplastic, 2020). Bioplastics may be defined as biodegradable and/or bio-based materials (European bioplastic, 2020). Overall, the advantages of biodegradable bio-based bioplastics are the lower carbon footprint, the reduction in oil dependence, the renewability of the source (biomass), and the possibility to valorise the bioplastics' end-of-life through proper waste management (Abraham et al., 2021; RameshKumar et al., 2020; Rosenboom et al., 2022). Nevertheless, the claims of biodegradability can be ambiguous if further information is not provided, i.e., timeframe, conditions necessary for biodegradation, and the level of biodegradation (Bátori et al., 2018; Cucina et al., 2021a; Ruggero et al., 2020; Zhang et al., 2018). Today, bioplastics certified as biodegradable and compostable are collected within biowaste and thus, their disposal follows waste management strategies used for organic wastes (i.e., anaerobic digestion, composting or coupled anaerobic digestion and composting). Composting represents a good approach

in degrading bioplastics, limiting their leakage into the environment. Nevertheless, because it requires energy to be performed (Zhao et al., 2020), it contributes positively to greenhouse gas emissions. Because of this fact and in the light of the application of Circular Economy rules, new strategies for treating bioplastics, recovering material and energy, appear mandatory (Cucina et al., 2021a; Rosenboom et al., 2022). Anaerobic digestion (AD) appears to be a promising strategy for bioplastics recovery, allowing energy production (biomethane), reducing C footprint, and making the circularity of these products real (Cazaudehore et al., 2022; Kakadellis et al., 2022; Rosenboom et al., 2022). Anaerobic degradability of bioplastics depends mainly upon both bioplastics' composition and the operative parameters of AD (i.e., temperature and retention time) (Abraham et al., 2021; Cucina et al., 2021a). For instance, thermophilic temperatures of 55-58 °C are necessary to obtain an effective degradation of bioplastics during AD due to the achievement of the glass transition temperature (Cazaudehore et al., 2021, 2022; Cucina et al., 2021a, 2021b; Yagi et al., 2013). In addition, it was highlighted that a long retention time of bioplastics inside the anaerobic reactor is required, even under thermophilic conditions, to achieve an effective degradation (Battista et al., 2021; Cazaudehore et al., 2022, 2021; Cucina et al., 2021a).

Bioplastics are expected to contribute substantially to the organic fraction of municipal solid (OFMSW) waste in the coming years (e.g., about 7-8 % on a weight basis in Italy by 2030) (Cucina et al., 2021b), so that they can play an important role in producing biogas when the OFMSW is anaerobically treated. According to Ebrahimzade et al. (2022) and García-Depraet et al. (2022), elucidating bioplastics degradation during AD is required to promote a practical and enhanced end-of-life management of bioplastics. Anyway, knowledge about the real degradation of bioplastics during AD and their conversion into biogas are still scarce, and all scientific studies reported were performed at laboratory-scale and using batch conditions (Cucina et al., 2021b; Ebrahimzade et al.,

2022; Yagi et al., 2013), completely forgetting the full-scale approach. This lack of knowledge was due to the difficulty of testing the degradability of bioplastic in full-scale reactor and under continuous condition, that better describe bioplastic degradability than batch lab-scale conditions (Ebrahimzade et al., 2022; García-Depraet et al., 2022). In this context, the purpose of this work was to assess the anaerobic degradability of three different bioplastic items using a full-scale approach. Moreover, to overcome the problem associated with the complexity of full-scale study, paper proposes an ingenious experimental design that could be replicated simplifying this kind of study (see Section 2.2.2). The bioplastics were co-digested with organic wastes in a full-scale thermophilic AD facility using a long retention time for bioplastics (90 days). At the same time, the experiment was replicated at laboratory-scale to evaluate the potential for biomethane recovery from bioplastics degradation.

2. Materials and methods

2.1 Bioplastics

The bioplastics studied were three different biodegradable items available at Italian supermarkets: starch-based shoppers (SBS) and PLA-based cutlery and dishes. Bioplastics were characterised by FT-IR analysis and results were reported in Section 3.1. The bioplastic items sampled were certified as compostable by both TÜV Austria (Austria) and the Italian Consortium of Composters (CIC) (Italy), according to standards (EN 13432, 2002). Prior to the anaerobic digestion experiments, the size of bioplastic items was reduced to pieces of about 25 x 25 mm, which represents the size used in the standard test to assess anaerobic degradability of bioplastics (ISO 14853, 2018).

2.2 Full-scale anaerobic digestion

2.2.1 Anaerobic digestion plant

The AD plant used for the full-scale experiment is located in the Lombardy Region, northern Italy, (Pigoli et al., 2021). In brief, the plant transforms organic wastes (mainly sewage sludge) into biogas (used for energy production through a combined heat and power unit) and two fertilizers (digestate and ammonium sulphate). The plant consists of three thermophilic digesters in series operating in thermophilic conditions (55 °C) treating a high-solid feedstock, mainly composed of municipal sewage sludge (see e-supplementary materials). The main operational parameters of the plant are reported in Table 1 and more details can be found in Pigoli et al. (2021). Prior to the beginning of the experiments, feedstock and digestate were sampled and characterized to evaluate their variability with time (one sample per month, for a total of three samplings) (Table 1) (see e-supplementary materials). Being the variability of feedstock and digestate very low, it was assumed that their composition did not affect bioplastics' degradation.

2.2.2 Experimental design

Bioplastics were placed in cube shaped boxes made of steel (length of the wall: 10 cm; volume of the boxes: 1 L) (Figure 1a) after being cut up as described in Section 2.1. To allow the entrance of the digestate into the boxes and ensure a proper contact between digestate and bioplastics, the walls of the boxes were perforated (2 mm mesh). The size of the mesh was selected by taking into consideration that (1) bioplastics are considered disintegrated when the size of the residues is under 2 mm (EN 13432, 2002) and that (2) inert materials (i.e., plastic, glass, metal, and bioplastic) with a size under 2 mm are not considered to negatively affect fertilizers' quality (EC, 2019). Twelve boxes were used for the full-scale experiment, six of them filled with 10 g of SBS and the remaining ones filled with 10 g of PLA-based cutlery and 10 g of PLA-

based dishes. SBS were tested alone since they completely filled the boxes at the beginning of the experiment. Conversely, PLA-based items were tested together since they were easily recognizable. The boxes were inserted inside the first digester using a system of chains and they were grouped to allow three samplings (2 SBS boxes + 2 PLA-based items boxes = 4 boxes per sampling x 3 = 12 boxes), as shown in Figure 1b. Samplings were performed at the 30th, 60th and 90th days, in order to evaluate the effect of prolonged bioplastics retention time (BRT) on their degradation. At the sampling time, boxes were recovered, and bioplastics were washed with rinse water to remove the digestate. Bioplastics were then dried in a ventilated oven at 40 °C to constant weight. Bioplastics degradation (%) was determined after weighing dried bioplastics, according to the equation:

$$\% degradation = \left(\frac{BP\ t_0 - BP\ t}{BP\ t_0} \right) \times 100$$

Where $BP\ t_0$ is the mass of bioplastics at the beginning of the experiment and $BP\ t$ is the mass of bioplastics recovered after sampling.

2.3 Laboratory-scale anaerobic digestion

Biochemical methane potential (BMP) of bioplastics was evaluated to assess the potential production of biomethane from bioplastics' anaerobic degradation. BMPs were determined following the method reported by Schievano et al. (2008) with small modifications. Digestate collected from the AD plant described in Section 2.2.1 was used as inoculum and the three bioplastic items were tested separately using 500-mL serum bottles. Three g of bioplastic were added to 300 mL of inoculum, leaving 200 mL of headspace in the bottles. Control blanks and positive control were prepared using 300 mL of inoculum without bioplastics and 300 mL of inoculum with 3 g of cellulose, respectively. Bottles were hermetically closed after flushing N₂ and placed in an oven at 55 ± 2 °C for 90 days. Bottles were periodically analysed for both quantitative and

qualitative determination of biogas production by means of withdrawing extra-pressure gas with a 100-mL syringe and gas chromatography (Carlo Erba Megaserie 5300, capillary column 25 m x 0.32 mm diameter and flame ionization detector), respectively. Biogas production of blank control was subtracted from biogas production of every sample.

Bioplastics' degradation in laboratory-scale experiments at 30, 60 and 90 days was evaluated through mass loss determination using destructive samples prepared using the same procedures as in the bottles used for BMPs assays. At sampling times, bottles were opened, and bioplastics were removed, rinsed, sieved to pass a 2 mm sieve, dried, and weighed to quantify bioplastics' degradation following the same method described in Section 2.2.2. Finally, stoichiometric calculations were used to estimate the bioplastics' degradation during BMP assays. Briefly, starting from the composition and amount of biogas produced at 30, 60 and 90 days, and assuming normal conditions (20 °C and 1 atm), the molar volume of gas (22.414 L mol⁻¹) was used to determine the mass of methane and carbon dioxide produced. The results were compared to the degradation determined through mass loss to understand if mass loss was due to biochemical processes (i.e., fermentation of substrates to CH₄ and CO₂) or to physical processes (i.e., fragmentation).

2.4 Composting experiment

Due to the different behaviour shown by SBS and PLA-based items under anaerobic conditions that will be discussed in Section 3, only SBS was tested for aerobic degradation during composting. SBS recovered after 30 days of full-scale thermophilic anaerobic digestion (SBS-TAD) and non-treated SBS (SBS-NT) were studied, in order to evaluate the effect of the anaerobic treatment on the aerobic degradability of SBS. Samples recovered after 30 days of AD were chosen taking into consideration that 30

days is a common HRT in AD plant treating OFMSW through AD coupled to digestate composting.

The active phase of composting was carried out in a 100 L adiabatic reactor equipped with temperature and oxygen probes; oxygen was kept around 10 % (v/v) by air insufflation controlled by feedback modality. The reactor was filled with about 80 kg of a mixture made of organic fraction of municipal solid wastes (OFMSW) and other organic materials. Bulking agent (wood chip) was added to the mixture to ensure both proper oxygenation and avoid excessive compaction (see e-supplementary materials).

Both OFMSW and wood chips were collected from a local composting plant. Ten g of SBS-TAD were carefully mixed with about 1 kg of the composting mixture and placed in one of the perforated steel boxes used in the full-scale AD experiment (Figure 1a).

About 1 kg of the mixture was the amount of material needed to fill the box without varying the mixture's bulk density, leading to a bioplastic concentration of 1 % w/w.

This concentration of bioplastic is the range recommended in bioplastics standard methods to assess biodegradability and disintegrability (EN13432, 2002). Two other boxes were filled with SBS-TAD and composting mixture, and another three boxes were filled with SBS-NT (10 g) mixed with composting mixture (about 1 kg), for a total of six boxes. The boxes were placed in the middle of the composting reactor at the beginning of the active phase. After 30 days, the active phase was considered

concluded, and the fresh compost (including the steel boxes) was placed in an adiabatic reactor (volume 70 L) for the maturation phase that lasted for another 30 days (for a total composting duration of 60 days). The fresh compost was turned periodically to ensure a proper oxygenation during the maturation phase, as well as biomass contained in the boxes. At the end of maturation, a representative sample of mature compost was collected for analytical determination, and the steel boxes were recovered and opened.

SBS residues were recovered from each box and washed with rinse water to remove the

compost. Bioplastics were then sieved to pass a 2 mm sieve and dried in a ventilated oven at 40 °C to constant weight. Bioplastics' degradation after composting was then determined using the equation reported in Section 2.2.2.

2.5 Chemical and spectroscopic analysis

2.5.1 Chemical analysis

Chemical analyses on feedstock and digestate from AD experiments, and on composting mixture and mature compost from composting experiments were carried out on fresh samples following standard methods. Total solids (TS) and volatile solids (VS) contents were determined according to standard procedures of the American Public Health Association, as well as total organic carbon (TOC), Total N and ammonium-N (APHA, 2017). pH was determined in aqueous solution using a 1:2.5 sample:water ratio and a pH-meter.

2.5.2 FT-IR spectroscopic analysis

Qualitative modifications of bioplastics items after biological treatments were investigated by spectroscopic analysis, using the Fourier Transform InfraRed (FT-IR) spectra that were collected in total reflectance mode (ATR) using a Shimadzu IRAffinity-1S equipped with a Miracle Pike ATR device (Shimadzu Italia Srl, Milano, Italy) (wavenumber range of 4,000–500 cm⁻¹ and resolution of 2 cm⁻¹). Non-treated bioplastics were dried at 37 °C before spectroscopic analysis, whereas bioplastics' samples recovered from biomasses (i.e., digestate, compost) were dried at 37 °C, cleaned and gently brushed by toothbrush to remove all the deposits formed on their surfaces before spectroscopic analysis. Shimadzu LabSolutions IR Peak Software (Shimadzu Italia srl, Milano, Italy) was used to determine peaks' areas.

2.6 Statistics

All experiments and analyses were replicated three times except for the full-scale anaerobic digestion, which was duplicated. Mean and standard deviation values were calculated according to standard procedures (Microsoft Excel Software). Determination of significant differences among the parameters analyzed at a level of significance of $P < 0.05$ was carried out by analysis of variance (ANOVA) and Tukey's test, whereas linear regression analysis was carried out to determine significant correlations between selected parameters at a level of significance of $P < 0.05$ (Microsoft Excel Solver 2013).

3. Results and discussion

3.1 Bioplastics characterization

To better understand the results from the degradation experiments, bioplastics were initially characterized by means of FT-IR spectroscopic analysis (see e-supplementary materials). Diagnostic peaks were assigned according to literature (Elfehri Borchani et al., 2015; Ruggero et al., 2020; Totaro et al., 2019; Zhang and Zhang, 2016). For SBS, four diagnostic peaks were highlighted, being two of them attributed to functional groups of starch ($3,400\text{ cm}^{-1}$ and $1,050\text{ cm}^{-1}$) and the other two bands assigned to a polyester component ($1,750\text{ cm}^{-1}$ and 730 cm^{-1}) (Elfehri Borchani et al., 2015; Ruggero et al., 2020). The spectrum of PLA-based cutlery was characterized by four diagnostic peaks that are commonly referred to polylactic acid ($3,000$, $1,750$, $1,400$ and $1,050\text{ cm}^{-1}$), whereas PLA-based dishes showed an intense peak at around 1050 cm^{-1} that may have covered the diagnostic peak of the ether group of PLA. According to literature, PLA-based dishes may be blended with poly(butylene succinate) (PBS) or cellulose which are polymers that absorb IR in that region (de Matos Costa et al., 2020; Totaro et al., 2019).

3.2 Bioplastics' anaerobic degradation

3.2.1 Full-scale experiment

Visual inspection of bioplastics after each sampling (30, 60 and 90 days) was carried out to evaluate changes in terms of consistency, thickness, discolouring and erosion of the material as suggested by Ruggero et al. (2019). Figure 2 shows recovered bioplastics after 90 days of thermophilic AD as examples. Size reduction was evident for all bioplastics items, as well as loss of transparency. This might be due to loss of crystallinity during AD such as recently reported by Ruggero et al. (2021) during the investigation of degradation of film and rigid bioplastics during simulated composting. Similarly, Zhang et al. (2018) reported that starch-based and PLA-based bioplastics showed few changes after mesophilic anaerobic digestion, i.e., slight changes in color, deformation and/or fragmentation. Table 2 shows the degradation of bioplastic items during the full-scale AD experiment. At the end of the experiment, PLA-based cutlery degraded by 70.8 ± 0.2 % on a weight basis (wb), followed by PLA-based dishes and SBS (60 ± 0.2 % and 39.6 ± 0.2 % wb, respectively). At this date, it was possible to compare the results from this full-scale experiment only with literature data coming from laboratory-scale experiments, but this comparison gave interesting results. In fact, SBS degradation obtained in the full-scale experiment at the 90th day was similar to the degradation reported for this bioplastic under thermophilic AD, that ranged from 35 % to 45 % wb (Calabro' et al., 2020; Cazaudehore et al., 2021; Vardar et al., 2022). Similarly, PLA-based items' degradation found in the full-scale experiment was in line with those reported in the literature (Cazaudehore et al., 2021; Vardar et al., 2022; Yagi et al., 2013, 2010). For instance, Cazaudehore et al., (2021), found that PLA-based coffee capsules degraded by about 60 % after 100 days of thermophilic AD. The kinetic of degradation during thermophilic AD at full-scale was evaluated considering the results obtained after the three samplings (30th, 60th and 90th days)

(Table 2). SBS degradation mainly occurred during the first 30 days, after which the degradation rate decreased, reaching a plateau. Since SBS is made up of two components (starch and polyesters), it can be hypothesized that the starch was quickly degraded in the first 30 days and then the other component slowly degraded during the experiment (Cucina et al., 2021). This hypothesis was supported by literature, where it was reported that SBS are generally made of starch ranging from 20 % to 40 % on a wb, and that starch was preferentially degraded during biological treatment with respect to the polyester component (Calabro' et al., 2020; Ruggero et al., 2020). Conversely, PLA-based items showed a linear kinetic of degradation during the full-scale AD experiment ($y = 1.5x$, $R^2 = 0.99$ and $y = 1.1x$, $R^2 = 0.99$ for PLA-based cutlery and dishes, respectively), according to the literature (Chamas et al., 2020; Chinaglia et al., 2018; Cucina, et al., 2021a, 2021b). Interestingly, bioplastics' degradation observed in the present work was higher than results reported in literature dealing with bioplastics' degradation during mesophilic AD. For instance, Vardar et al. (2022) reported an average degradation of PLA-based and starch-based bioplastics near to 30 % and 25 % wb during mesophilic AD. This was probably due to the thermophilic treatment that reached the glass transition temperature of bioplastics (55-60 °C), which is known to change the structure of the polymer, promoting the modification from the crystalline structure to the amorphous one, the latter being more biodegradable (Camacho-Muñoz et al., 2020).

FT-IR analysis was used to evaluate qualitative changes in bioplastics' composition during AD (see e-supplementary materials). A preferential consumption of the starch with respect to the polyester component was found on observing the evolution of SBS spectra. For example, in the spectrum of bioplastic sampled at 30th days the diagnostic peak of starch at 3400 cm⁻¹ almost disappeared (see e-supplementary materials). This supported the hypothesis that starch was preferentially consumed by microorganisms in

the first 30 days of AD, leading to the concentration of the other component.

Composition of PLA-based items was not affected by AD, and FT-IR spectra of both PLA-based dishes and cutlery did not vary in time (see e-supplementary materials).

Considering that PLA-based items degraded by 60 ± 0.2 % and 70.8 ± 0.2 % wb (dishes and cutlery, respectively), it may be suggested that PLA-based bioplastics degraded through hydrolyzation into monomers that enter the microbial cell and can be metabolized, without significant modifications to the polymer structure (Cucina et al., 2021b).

With regard to PLA-based dishes, which were probably made of a blend of PLA with other polymers (i.e., PBS), it is likely that the different components degraded at the same rate since no significant variations were observed in the spectra of these items during AD. The different mechanisms of anaerobic degradation proposed for SBS and PLA-based items was validated by calculating the ratio between the areas of the diagnostic peaks as suggested by Cucina et al. (2021b) (Table 3). In the first 30 days, the ratio between starch (1050 cm^{-1}) and polyester (1750 cm^{-1}) peaks area increased from 1.6 to 3.8, confirming the preferential consumption of starch. After 60 and 90 days the ratio remained unvaried (ranging from 3.5 to 3.7), according to what was reported by Cucina et al. (2021b) while studying degradation of starch-based shoppers under mesophilic AD, composting and soil incubation. No significant differences were observed when calculating the ratio between diagnostic peaks' areas for PLA-based items. The ratios remained constant over time, ranging from 0.76 to 0.92 for dishes (ratio $1750\text{ cm}^{-1} / 1050\text{ cm}^{-1}$) and from 2.67 to 3.24 for cutlery (ratio $1750\text{ cm}^{-1} / 1400\text{ cm}^{-1}$). These results confirmed that PLA degradation followed a “take away” mechanism (Cucina et al., 2021b) and that other possible additives, (i.e., PBS or cellulose), degraded with a similar kinetic and mechanism.

3.2.2 Laboratory-scale experiment

Regarding bioplastics' degradation, results during the laboratory-scale experiment confirmed the full-scale data (Table 2). At the end of the laboratory-scale experiment, SBS, PLA-based cutlery and PLA-based dishes degraded by $37.8 \pm 1.0 \%$, $72.1 \pm 1.4 \%$ and $61.1 \pm 0.7 \%$ on a wb, respectively, and statistical analysis confirmed that no significant differences were found between quantitative degradation in full-scale and laboratory-scale experiments ($P < 0.05$). As already described for the degradation assessment at full-scale, the results from laboratory-scale were in accordance with recent literature dealing with bioplastics degradation in the laboratory under thermophilic anaerobic conditions. For instance, PLA degraded by about 24 % in 30 days, 43 % in 40 days and 68 % in 60 days during an experiment of thermophilic anaerobic digestion (Yagi et al., 2013), whereas SBS degraded by about 30 % after 60 days of thermophilic AD (Calabro' et al., 2020). From a qualitative point of view, FT-IR investigation of bioplastics during laboratory-scale thermophilic AD showed that bioplastics' degradation in laboratory followed the same mechanisms observed in the full-scale trial (Table 3) (see e-supplementary materials). All these data support the idea that full-scale degradation can be well reproduced at laboratory-scale and that the biomethane potentials obtained at laboratory-scale can be assumed also for a full-scale plant.

3.3 Potential for biomethane production from bioplastics' degradation

At the end of the thermophilic AD (90th days), SBS produced 224 ± 16 NL CH₄ kg VS⁻¹, while PLA-based cutlery and PLA-based dishes produced 397 ± 8 NL CH₄ kg VS⁻¹ and 330 ± 26 NL CH₄ kg VS⁻¹, respectively (Table 4). Overall, the results of biomethane production were in line with literature dealing with thermophilic AD of bioplastics (Cazaudehore et al., 2021; Kakadellis et al., 2022; Yagi et al., 2013, 2010), and the results

were higher than those obtained from mesophilic AD experiments, as expected. Indeed, not only bioplastics' degradation but also biomethane production were reported to be enhanced by thermophilic conditions (Gil-Castell et al., 2020). Temperatures higher than 55 °C are often reported to reduce the molecular weight of bioplastics and promote the modification of the biopolymer from the crystalline structure to the amorphous one, favouring their biodegradation.

With reference to SBS, it should be highlighted that the kinetics of biomethane production during the 90 days of AD reflected the kinetics of degradation determined through mass loss. Indeed, the largest part of the biomethane was produced during the first 30 days of incubation (about 65 % of the total biomethane produced) and this can be attributed to the quick mineralization of starch that was suggested, considering qualitative and quantitative degradation of SBS. Assuming that starch contributes about 30 % by weight to the total SBS and considering a BMP of 450 NL CH₄ VS⁻¹ (Cucina, et al., 2021c; Elfehri Borchani et al., 2015), it was calculated that there was a total theoretical contribution of starch to SBS's BMP of 135 NL CH₄ VS⁻¹, which was similar to that measured during first 30 days of the experiment (142 ± 24 NL CH₄ kg VS⁻¹). After the degradation of starch, it is likely that biomethane production from SBS can be attributed to the slow degradation of the polyester component. Clearly, obtaining the complete degradation of SBS and the production of its ultimate biomethane potential would require long time, probably not compatible with common HRT used in AD plants. For instance, Cazaudehore et al. (2021) reported that after 100 days of AD, starch-based coffee capsules degraded by about 50 %, allowing an estimate that more than 200 days are needed to complete the degradation (Cucina et al., 2021a). The incomplete degradation of SBS during AD may lead to the presence of bioplastics' residues in the digestate, making its agricultural reuse problematic; indeed, digestate may act as a source of bioplastics' leakage into the soil. Although it was highlighted that

bioplastics require a short time to degrade in soil in comparison with plastics (Chamas et al., 2020; Cucina et al., 2021a), the possible contamination of the environment through bioplastics and micro-bioplastics is an emerging issue that deserves more attention (Fojt et al., 2020; Qin et al., 2021). For instance, little is known about bioplastics' degradation in soil in the presence of digestate or compost, as well as the potential toxicity of micro-bioplastics, showing the need for further studies on this topic. Moreover, European Regulation 1009/2019 (EC, 2019) set a maximum content of inert materials (plastics, glass, and metals > 2 mm) in component materials categories, i.e., digestate, of 0.5 %, and this value will be decreased to 0.25 % by 2026 (EC, 2019). Considering this, it is evident that AD of SBS may be a suitable strategy to recover energy from the starch component of the bioplastic, but that also other management strategies should be investigated to enhance the degradation of the polyester component, avoiding its leakage into the environment.

Differently from SBS, PLA-based items were converted to biomethane following a linear kinetic, as already observed considering the degradation at full- and laboratory-scale ($y = 3.6x$, $R^2 = 0.99$ and $y = 4.4x$, $R^2 = 0.99$ for PLA-based dishes and cutlery, respectively). Assuming a linear degradation of these bioplastics, it can be calculated that their complete degradation can occur in about 120-150 days. This would represent the bioplastics' retention time (BRT) required to reach the ultimate biomethane potential of PLA-based bioplastics (about 450 CH₄ VS⁻¹). Similar retention times were reported for complete degradation of PLA-based bioplastics and other bioplastics, i.e., polycaprolactone, which required 140 days to completely degrade and produce the ultimate biomethane potential under thermophilic AD (Cazaudehore et al., 2022; Cucina et al., 2021a). Although the BRT needed to obtain a complete anaerobic degradation of bioplastics may appear too long with respect to the HRT commonly applied in real AD plants, it should be highlighted that long HRT are common in AD plants treating agro-

industrial wastes and livestock effluents (Cazaudehore et al., 2022; Han et al., 2020). In addition, it can also be suggested to separate BRT from HRT of the AD plant, implementing physical barriers perforated with 2 mm mesh inside the reactor, similarly to what was done in the full-scale experiment described in this work. This may allow the recovery of a noticeable amount of biomethane from PLA-based items and the avoidance of the presence of undegraded bioplastics in the digestate, favouring its agricultural reuse.

Interestingly, degradation calculated from BMP data in laboratory experiments replicated the degradation determined from mass loss (both in full- and laboratory-scales) (Tables 2 and 4). At the end of the experiment (90 days), a degradation of 37.8 ± 1.0 % wb was found for SBS from mass loss in the laboratory experiment, compared to 40 % calculated from BMP. Similar results were obtained for PLA-based items. This result was important to prove that bioplastics degradation was mainly due to biochemical processes. Indeed, bioplastics were likely converted to biogas, resulting in the probable absence of micro-bioplastic residues in the digestate. From an environmental point of view, this can mean a reduction of bioplastic leakage into the environment following digestate application in agriculture.

3.4 Integrating anaerobic digestion and composting for starch-based shoppers management

According to the results presented in Sections 3.2 and 3.3, it appears evident that PLA-based items may be converted efficiently into biomethane in thermophilic AD plants by lengthening HRT or separating BRT from HRT of the digestate. This represents a clear advantage since energy recovered from bioplastics degradation can cover the energy requirements of other activities (i.e., production of new bioplastics) and reduce bioplastics leakage into the environment, enhancing circularity of these bioplastics.

Conversely, SBS showed a slow degradation following the first month of AD, when starch was quickly converted to biomethane, resulting in incomplete degradation and the presence of bioplastics residues in the digestate even when long HRTs are applied. Considering this, it was hypothesised to separate the management of SBS and PLA-based items. Since the amount of bioplastics' wastes is increasing and their separate collection is emerging as a possible strategy to collect them (Cucina et al., 2021a; Cucina et al., 2021b; Vardar et al., 2022; Vinci et al., 2021), the authors hypothesised a separate collection for PLA-based items (i.e., single use dishes, cutlery, drinking vessels and food-packaging) to be treated in AD plants. Conversely, SBS were thought suitable for treatment in OFMSW-AD plants where the digestate is post-treated by composting. This scenario was chosen for starch-based bioplastics since they are commonly used to collect the OFMSW and because integrated AD and composting is becoming a widespread technology to recover biomethane and fertilizers (i.e., compost) from OFMSW (le Pera et al., 2022). To evaluate the effect of integrating AD and composting on SBS degradation, SBS recovered after 30 days of full-scale thermophilic anaerobic digestion (SBS-TAD) were composted with OFMSW and other biowastes. Non-treated SBS (SBS-NT) were also studied to evaluate the effect of the anaerobic treatment on the aerobic degradability of SBS.

The temperature during the active phase of composting rapidly increased to values higher than 55 °C due to the oxidation of organic matter (see e-supplementary materials), ensuring the ideal conditions for SBS degradation throughout the experiment. During the maturation, temperature remained higher than 55 °C for about 15 days, and then decreased to room temperature. Once the maturation phase was ended, the boxes containing SBS were opened and the results of SBS degradation registered. No residues of SBS-TAD were found at the end of the maturation phase, whereas small residues of SBS-NT were still detectable, resulting in a degradation of

98.8 ± 0.8 % on wb. According to FT-IR spectra of the SBS-NT residues after composting (see e-supplementary materials), the residues were mainly composed of residues of polyester, since the starch-diagnostic peaks had almost disappeared (Table 3). These results may lead to interesting conclusions: i. firstly, this work confirmed that SBS effectively degraded under thermophilic composting conditions, and this was in accordance with the literature, i.e., compostable bioplastic bags degraded by 90 % in 60 days of composting according to Cafiero et al. (2021); ii. secondly, integration of AD and composting may represent a suitable and sustainable strategy to manage SBS following the treatments commonly used for OFMSW. In this way, biomethane potential of starch can be quickly recovered during AD using a common HRT of 20-30 days. Then, the degradation of the polyester component can be achieved with composting, minimising the leaching of bioplastics into the environment through compost utilization in agriculture. Coupling energy recovery and fertilizers production may enhance the circularity of SBS compared to the current methods of management. Indeed, SBS are commonly treated in composting facilities with OFMSW. Although composting may lead to SBS degradation, their mineralization to CO₂ may be positive to avoid their leakage into the environment, but from a sustainability point of view this strategy arouses concerns. LCA analysis of bioplastics life cycle, i.e., production, consumption, collection, and composting, proved that bioplastics have a high global warming potential if industrial composting and disposal transportation are considered as their end-of-life strategy (Zhao et al., 2020). Production of biomethane from SBS by AD and their sequential degradation during digestate composting may represent a positive contribution to the economic, energetic, and environmental assessment, improving the circularity of these important substitutes of fossil-based plastics.

4. Conclusions

Anaerobic digestion may be a suitable strategy to recover biomethane from polylactic acid-based bioplastics' wastes and to reduce their leakage into the environment. Starch-based shoppers showed a slower degradation in anaerobic conditions but coupling anaerobic digestion and composting may represent the best strategy to produce biomethane from the starch component and to completely degrade the polyester one. Considering the increasing amount of bioplastics' wastes expected to be produced in the coming years, anaerobic digestion and/or anaerobic digestion coupled to digestate composting may represent the best strategy to dispose these wastes while meeting the principles of Circular Economy.

E-supplementary data

E-supplementary data for this work can be found in e-version of this paper online.

Funding

This study is part of the research project "POR FESR 2014-2020 - Call HUB Ricerca e Innovazione - Progetto BIOMASS HUB ID: 1165247", subcontractors with Acqua e Sole contract Numbers: CTE_NAZPR21FADAN_02.

References

1. Abraham, A., Park, H., Choi, O., Sang, B.I., 2021. Anaerobic co-digestion of bioplastics as a sustainable mode of waste management with improved energy production – A review. Bioresource Technology. <https://doi.org/10.1016/j.biortech.2020.124537>
2. APHA, 2017, 2017. APHA/AWWA/WEF (2017) Standard Methods for the Examination of Water and Wastewater.23rd Edition, American Public Health

- Association. American Water Works Association Water Environment Federation Stable.
3. Bátori, V., Åkesson, D., Zamani, A., Taherzadeh, M.J., Sárvári Horváth, I., 2018. Anaerobic degradation of bioplastics: A review. *Waste Management* 80. <https://doi.org/10.1016/j.wasman.2018.09.040>
 4. Battista, F., Frison, N., Bolzonella, D., 2021. Can bioplastics be treated in conventional anaerobic digesters for food waste treatment? *Environmental Technology and Innovation* 22. <https://doi.org/10.1016/j.eti.2021.101393>
 5. Bellasi, A., Binda, G., Pozzi, A., Galafassi, S., Volta, P., Bettinetti, R., 2020. Microplastic contamination in freshwater environments: A review, focusing on interactions with sediments and benthic organisms. *Environments - MDPI*. <https://doi.org/10.3390/environments7040030>
 6. Cafiero, L.M., Canditelli, M., Musmeci, F., Sagnotti, G., Tuffi, R., 2021. Assessment of disintegration of compostable bioplastic bags by management of electromechanical and static home composters. *Sustainability (Switzerland)* 13. <https://doi.org/10.3390/su13010263>
 7. Calabro', P.S., Folino, A., Fazzino, F., Komilis, D., 2020. Preliminary evaluation of the anaerobic biodegradability of three biobased materials used for the production of disposable plastics. *Journal of Hazardous Materials* 390. <https://doi.org/10.1016/j.jhazmat.2019.121653>
 8. Camacho-Muñoz, R., Villada-Castillo, H.S., Solanilla-Duque, J.F., 2020. Anaerobic biodegradation under slurry thermophilic conditions of poly(lactic acid)/starch blend compatibilized by maleic anhydride. *International Journal of Biological Macromolecules* 163. <https://doi.org/10.1016/j.ijbiomac.2020.09.183>
 9. Cazaudehore, G., Guyoneaud, R., Evon, P., Martin-Closas, L., Pelacho, A.M., Raynaud, C., Monlau, F., 2022. Can anaerobic digestion be a suitable end-of-life

scenario for biodegradable plastics? A critical review of the current situation,
hurdles, and challenges. *Biotechnology Advances* 56, 107916.
<https://doi.org/10.1016/j.biotechadv.2022.107916>

10. Cazaudehore, G., Monlau, F., Gassie, C., Lallement, A., Guyoneaud, R., 2021.
Methane production and active microbial communities during anaerobic digestion
of three commercial biodegradable coffee capsules under mesophilic and
thermophilic conditions. *Science of The Total Environment* 784.
<https://doi.org/10.1016/j.scitotenv.2021.146972>

11. Chamas, A., Moon, H., Zheng, J., Qiu, Y., Tabassum, T., Jang, J.H., Abu-Omar,
M., Scott, S.L., Suh, S., 2020. Degradation Rates of Plastics in the Environment.
ACS Sustainable Chemistry and Engineering 8.
<https://doi.org/10.1021/acssuschemeng.9b06635>

12. Chinaglia, S., Tosin, M., Degli-Innocenti, F., 2018. Biodegradation rate of
biodegradable plastics at molecular level. *Polymer Degradation and Stability* 147.
<https://doi.org/10.1016/j.polymdegradstab.2017.12.011>

13. Cucina, M., de Nisi, P., Tambone, F., Adani, F., 2021a. The role of waste
management in reducing bioplastics' leakage into the environment: A review.
Bioresource Technology. <https://doi.org/10.1016/j.biortech.2021.125459>

14. Cucina, M., de Nisi, P., Trombino, L., Tambone, F., Adani, F., 2021b. Degradation
of bioplastics in organic waste by mesophilic anaerobic digestion, composting and
soil incubation. *Waste Management* 134.
<https://doi.org/10.1016/j.wasman.2021.08.016>

15. Cucina, M., Pezzolla, D., Tacconi, C., Gigliotti, G., 2021c. Anaerobic co-digestion
of a lignocellulosic residue with different organic wastes: Relationship between
biomethane yield, soluble organic matter and process stability. *Biomass and
Bioenergy* 153. <https://doi.org/10.1016/j.biombioe.2021.106209>

16. de Matos Costa, A.R., Crocitti, A., de Carvalho, L.H., Carroccio, S.C., Cerruti, P., Santagata, G., 2020. Properties of biodegradable films based on poly(Butylene succinate) (pbs) and poly(butylene adipate-co-terephthalate) (pbat) blends. *Polymers* 12. <https://doi.org/10.3390/polym12102317>
17. Ebrahimzade, I., Ebrahimi-Nik, M., Rohani, A., Tedesco, S., 2022. Towards monitoring biodegradation of starch-based bioplastic in anaerobic condition: finding a proper kinetic model. *Bioresource Technology* 126661. <https://doi.org/10.1016/j.biortech.2021.126661>
18. EC, 2019. Regulation (EU) 2019/1009 Fertilizer Products. Official Journal of the European Union 2019.
19. Elfehri Borchani, K., Carrot, C., Jaziri, M., 2015. Biocomposites of Alfa fibers dispersed in the Mater-Bi® type bioplastic: Morphology, mechanical and thermal properties. *Composites Part A: Applied Science and Manufacturing* 78. <https://doi.org/10.1016/j.compositesa.2015.08.023>
20. Emadian, S.M., Onay, T.T., Demirel, B., 2017. Biodegradation of bioplastics in natural environments. *Waste Management*. <https://doi.org/10.1016/j.wasman.2016.10.006>
21. EN13432, 2002. EN13432. Packaging - Requirements for packaging recoverable through composting and biodegradation - test scheme and evaluation criteria for the final acceptance of packaging.
22. European bioplastic, 2020. Market update 2020: Bioplastics continue to become mainstream as the global bioplastics market is set to grow by 36 percent over the next 5 years [WWW Document]. <https://www.european-bioplastics.org/market-update-2020-bioplastics-continue-to-become-mainstream-as-the-global-bioplastics-market-is-set-to-grow-by-36-percent-over-the-next-5-years/>.

23. European Parliament, 2019. Directive (EU) 2019/904 of the European Parliament and of the Council of 5 June 2019 on the reduction of the impact of certain plastic products on the environment.
24. Fojt, J., David, J., Přikryl, R., Řezáčová, V., Kučerík, J., 2020. A critical review of the overlooked challenge of determining micro-bioplastics in soil. *Science of The Total Environment* 745, 140975. <https://doi.org/10.1016/j.scitotenv.2020.140975>
25. García-Depraect, O., Lebrero, R., Rodriguez-Vega, S., Bordel, S., Santos-Beneit, F., Martínez-Mendoza, L. J., Aragao Börner, R., Börner, T., Muñoz, R., 2022. Biodegradation of bioplastics under aerobic and anaerobic aqueous conditions: Kinetics, carbon fate and particle size effect. *Bioresource Technology* 344, 126265. <https://doi.org/10.1016/j.biortech.2021.126265>
26. Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made. *Science Advances* 3. <https://doi.org/10.1126/sciadv.1700782>
27. Gil-Castell, O., Andres-Puche, R., Dominguez, E., Verdejo, E., Monreal, L., Ribes-Greus, A., 2020. Influence of substrate and temperature on the biodegradation of polyester-based materials: Polylactide and poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) as model cases. *Polymer Degradation and Stability* 180. <https://doi.org/10.1016/j.polymdegradstab.2020.109288>
28. Han, Y., Yang, L., Chen, Xueming, Cai, Y., Zhang, X., Qian, M., Chen, Xingkui, Zhao, H., Sheng, M., Cao, G., Shen, G., 2020. Removal of veterinary antibiotics from swine wastewater using anaerobic and aerobic biodegradation. *Science of The Total Environment* 709, 136094. <https://doi.org/10.1016/j.scitotenv.2019.136094>
29. ISO, 2018. 14853: Plastics - Determination of the ultimate anaerobic biodegradation of plastic materials in an aqueous system - Method by measurement of biogas production.

30. Kakadellis, S., Lee, P.-H., Harris, Z.M., 2022. Two Birds with One Stone: Bioplastics and Food Waste Anaerobic Co-Digestion. *Environments* 9, 9. <https://doi.org/10.3390/environments9010009>
31. le Pera, A., Sellaro, M., Bencivenni, E., D'Amico, F., 2022. Environmental sustainability of an integrate anaerobic digestion-composting treatment of food waste: Analysis of an Italian plant in the circular bioeconomy strategy. *Waste Management* 139, 341–351. <https://doi.org/10.1016/j.wasman.2021.12.042>
32. Matthews, C., Moran, F., Jaiswal, A.K., 2021. A review on European Union's strategy for plastics in a circular economy and its impact on food safety. *Journal of Cleaner Production*. <https://doi.org/10.1016/j.jclepro.2020.125263>
33. Pigoli, A., Zilio, M., Tambone, F., Mazzini, S., Schepis, M., Meers, E., Schoumans, O., Giordano, A., Adani, F., 2021. Thermophilic anaerobic digestion as suitable bioprocess producing organic and chemical renewable fertilizers: A full-scale approach. *Waste Management* 124. <https://doi.org/10.1016/j.wasman.2021.02.028>
34. Qin, M., Chen, C., Song, B., Shen, M., Cao, W., Yang, H., Zeng, G., Gong, J., 2021. A review of biodegradable plastics to biodegradable microplastics: Another ecological threat to soil environments? *Journal of Cleaner Production* 312, 127816. <https://doi.org/10.1016/j.jclepro.2021.127816>
35. RameshKumar, S., Shaiju, P., O'Connor, K.E., P, R.B., 2020. Bio-based and biodegradable polymers - State-of-the-art, challenges and emerging trends. *Current Opinion in Green and Sustainable Chemistry*. <https://doi.org/10.1016/j.cogsc.2019.12.005>
36. Rosenboom, J.-G., Langer, R., Traverso, G., 2022. Bioplastics for a circular economy. *Nature Reviews Materials* 7, 117–137. <https://doi.org/10.1038/s41578-021-00407-8>

37. Ruggero, F., Carretti, E., Gori, R., Lotti, T., Lubello, C., 2020. Monitoring of degradation of starch-based biopolymer film under different composting conditions, using TGA, FTIR and SEM analysis. *Chemosphere* 246. <https://doi.org/10.1016/j.chemosphere.2019.125770>
38. Ruggero, F., Gori, R., Lubello, C., 2019. Methodologies to assess biodegradation of bioplastics during aerobic composting and anaerobic digestion: A review. *Waste Management and Research*. <https://doi.org/10.1177/0734242X19854127>
39. Ruggero, F., Onderwater, R.C.A., Carretti, E., Roosa, S., Benali, S., Raquez, J.M., Gori, R., Lubello, C., Wattiez, R., 2021. Degradation of Film and Rigid Bioplastics During the Thermophilic Phase and the Maturation Phase of Simulated Composting. *Journal of Polymers and the Environment* 29. <https://doi.org/10.1007/s10924-021-02098-2>
40. Schievano, A., Pognani, M., D'Imporzano, G., Adani, F., 2008. Predicting anaerobic biogasification potential of ingestates and digestates of a full-scale biogas plant using chemical and biological parameters. *Bioresource Technology* 99. <https://doi.org/10.1016/j.biortech.2008.03.030>
41. Taufik, D., Reinders, M.J., Molenveld, K., Onwezen, M.C., 2020. The paradox between the environmental appeal of bio-based plastic packaging for consumers and their disposal behaviour. *Science of the Total Environment* 705. <https://doi.org/10.1016/j.scitotenv.2019.135820>
42. Totaro, G., Sisti, L., Fiorini, M., Lancellotti, I., Andreola, F.N., Saccani, A., 2019. Formulation of Green Particulate Composites from PLA and PBS Matrix and Wastes Deriving from the Coffee Production. *Journal of Polymers and the Environment* 27. <https://doi.org/10.1007/s10924-019-01447-6>
43. Vardar, S., Demirel, B., Onay, T.T., 2022. Degradability of bioplastics in anaerobic digestion systems and their effects on biogas production: a review.

Reviews in Environmental Science and Bio/Technology 21, 205–223.

<https://doi.org/10.1007/s11157-021-09610-z>

44. Vinci, G., Ruggieri, R., Billi, A., Pagnozzi, C., di Loreto, M.V., Ruggeri, M., 2021.

Sustainable management of organic waste and recycling for bioplastics: A lca approach for the italian case study. Sustainability (Switzerland) 13.

<https://doi.org/10.3390/su13116385>

45. Yagi, H., Ninomiya, F., Funabashi, M., Kunioka, M., 2013. Thermophilic

anaerobic biodegradation test and analysis of eubacteria involved in anaerobic

biodegradation of four specified biodegradable polyesters. Polymer Degradation

and Stability 98. <https://doi.org/10.1016/j.polymdegradstab.2013.03.010>

46. Yagi, H., Ninomiya, F., Funabashi, M., Kunioka, M., 2010. Bioplastic

biodegradation activity of anaerobic sludge prepared by preincubation at 55°C for new anaerobic biodegradation test. Polymer Degradation and Stability 95.

<https://doi.org/10.1016/j.polymdegradstab.2010.01.023>

47. Zhang, W., Heaven, S., Banks, C.J., 2018. Degradation of some EN13432

compliant plastics in simulated mesophilic anaerobic digestion of food waste.

Polymer Degradation and Stability 147.

<https://doi.org/10.1016/j.polymdegradstab.2017.11.005>

48. Zhang, X., Zhang, Y., 2016. Reinforcement effect of poly(butylene succinate)

(PBS)-grafted cellulose nanocrystal on toughened PBS/polylactic acid blends.

Carbohydrate Polymers 140. <https://doi.org/10.1016/j.carbpol.2015.12.073>

49. Zhao, X., Cornish, K., Vodovotz, Y., 2020. Narrowing the Gap for Bioplastic Use

in Food Packaging: An Update. Environmental science & technology.

<https://doi.org/10.1021/acs.est.9b03755>

Figure captions

Figure 1. (a) Steel boxes used for sampling of bioplastics in the full-scale experiment;
(b) system used to introduce the steel boxes inside the anaerobic digester.

Figure 2. Visual inspection of non-treated (left) and digested bioplastic items after 90
days of thermophilic anaerobic digestion (right). (a) Starch-based shoppers, (b) PLA-
based dishes and (c) PLA-based cutlery.

717 **Table 1.** Main operative parameters of the anaerobic digestion plant and digestate
718 characteristics.

	Parameter	Unit	Value
<i>Thermophilic anaerobic digestion</i>	HRT ^a of the sludge	Days	30.1 ± 3.1
	OLR ^b	kgVS m ⁻³ d ⁻¹	3.4 ^c ± 0.3
	Temperature	°C	55 ± 2
<i>Digestate characteristics</i>	Total solids	%	11.2 ^e ± 0.3
	Volatile solids	% d.m. ^d	59.9 ± 0.9
	pH	pH unit	8.3 ± 0.1
	Total organic C	g kg ⁻¹ d.m.	308 ± 90
	Total N	g kg ⁻¹ d.m.	76 ± 2
	C/N		4.1
	Ammonium-N	g kg ⁻¹ d.m.	31 ± 2

719 ^aHydraulic retention time of the sludge

720 ^bOrganic loading rate

721 ^c Mean value ± Standard Deviation (n = 14)

722 ^dDry matter

723 ^eMean value ± Standard Deviation (n = 3)

724

Table 2. Degradation of bioplastics items during anaerobic digestion at full- and laboratory-scale.

	Starch-based shoppers		PLA^a-based dishes		PLA-based cutlery	
Time (d)	<i>Full-scale^b</i>	<i>Lab-scale^c</i>	<i>Full-scale</i>	<i>Lab-Scale</i>	<i>Full-scale</i>	<i>Lab-scale</i>
	(% weight basis)					
30	23.8 ± 0.4 (23.5; 24.1)	26.8 ± 2.9	16.2 ± 0.1 (16.1; 16.2)	17.9 ± 0.9	15.8 ± 0.2 (15.6; 15.9)	16.4 ± 0.6
60	34.6 ± 0.1 (34.5; 34.7)	35.0 ± 0.5	41.2 ± 0.1 (41.1; 41.2)	43.9 ± 1.4	51.6 ± 0.0 (51.6; 51.6)	52.0 ± 1.2
90	39.6 ± 0.2 (39.5; 39.8)	37.8 ± 1.0	60.0 ± 0.2 (59.8; 60.1)	61.1 ± 0.7	70.8 ± 0.2 (70.6; 70.9)	72.1 ± 1.4

^aPolylactic-acid

^bMean value ± SD, n = 2. The results of the two replicates are reported in brackets.

^cMean value ± SD, n = 3

729 **Table 3.** Ratio between diagnostic peaks areas during anaerobic digestion and
730 composting of bioplastics.

		Starch-based shoppers	PLA ^a -based dishes	PLA-based cutlery
<i>Time (days)</i>		<i>1750 cm⁻¹ / 1050 cm⁻¹</i>	<i>1750 cm⁻¹ / 1050 cm⁻¹</i>	<i>1750 cm⁻¹ / 1400 cm⁻¹</i>
<i>Full-scale AD</i> ^b	0	1.6 ± 0.1	0.79 ± 0.01	2.67 ± 0.13
	30	3.8 ± 0.2	0.76 ± 0.01	2.8 ± 0.04
	60	3.5 ± 0.1	0.83 ± 0.05	2.95 ± 0.02
	90	3.7 ± 0.1	0.92 ± 0.01	3.24 ± 0.03
<i>Lab-scale AD</i>	0	1.5 ± 0.1	0.82 ± 0.02	2.81 ± 0.03
	30	3.5 ± 0.1	0.74 ± 0.06	2.93 ± 0.07
	60	3.4 ± 0.2	0.86 ± 0.03	3.04 ± 0.11
	90	3.6 ± 0.2	0.84 ± 0.05	3.01 ± 0.05
<i>Composting</i> ^c	0	1.4 ± 0.2	n.d. ^d	n.d.
	60	3.9 ± 0.1	n.d.	n.d.

731 ^aPolylactic acid

732 ^bMean value ± Standard Deviation (n = 3)

733 ^cComposting of starch-based shoppers; ratio between diagnostic peaks areas at the end
734 of composting of starch-based shoppers treated anaerobically was not carried out since
735 no residues of bioplastics were found

736 ^dnot determined

737

738

739 **Table 4.** Biochemical methane potential (BMP) of bioplastics and degradation
740 calculated from BMP.

Parameter	Unit	Starch-based shoppers	PLA ^a -based dishes	PLA-based cutlery
<i>BMP – 30 days</i>	NL CH ₄ kg VS ⁻¹	142 ^b ± 24	81 ± 13	104 ± 12
<i>BMP – 60 days</i>	NL CH ₄ kg VS ⁻¹	194 ± 28	215 ± 11	279 ± 20
<i>BMP – 90 days</i>	NL CH ₄ kg VS ⁻¹	224 ± 16	330 ± 26	397 ± 8
<i>Degradation^c– 30 days</i>	%	25.1	14.3	18.4
<i>Degradation – 60 days</i>	%	34.4	38.1	49.4
<i>Degradation – 90 days</i>	%	40.0	58.4	70.3

741 ^aPolylactic acid

742 ^bMean value ± Standard Deviation (n = 3)

743 ^cCalculated from BMP

744

(a)



(b)



745 **Figure 1**

746

(a)



(b)



(c)



747 **Figure 2**