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ORIGINAL PAPER

Effect of Paper vs. Bioplastic Bags on Food Waste Collection and Processing

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Abstract

Purpose The most abundant among the separately collected waste materials in Italy is food waste. This research aims to evaluate the influence of the type of collection bag on the food waste management chain. In Italy, the food waste collection is mainly based on bioplastic bags. As an alternative, a new type of recycled paper bag shows potential advantages.

Methods The two types of collection bag were compared evaluating the weight loss of food waste during the household storage, by means of an experimental assessment simulating the domestic dynamic bag filling. Moreover, the biomethane production of bags under anaerobic conditions was measured at the lab-scale level with Biochemical Methane Potential (BMP) tests.

Results During the household storage, the breathable fabric of the paper allows for higher weight losses, ranging on average between + 29 and + 44% compared to bioplastic. BMP tests, carried out under different conditions (temperature, inoculum), showed a 2–14 times higher generation of methane by paper bags compared to bioplastic bags, when referred to 1 kg of inserted food waste.

Conclusions Collecting the food waste inside paper bags shows advantages compared to the use of bioplastic bags. First, the waste collection is benefitted thanks to the lower weight of material to be transported to treatment plants, leading also to the possibility of decreasing the collection frequency. Moreover, paper resulted more compatible than bioplastic with the anaerobic digestion treatment, which is currently rapidly increasing as a food waste management option.

Graphic Abstract

Keywords Food waste · Collection bag · Paper · Bioplastic · Anaerobic digestion

Extended author information available on the last page of the article

Statement of Novelty

The organic fraction (mainly constituted by food waste) is generally the most important municipal waste stream separately collected. For this reason, the research aims to analyze the performances of the food waste management chain searching for potential optimization. The novelty is the focus on how different collection bag types interact with the food waste and influence the food waste treatments. Starting from the evaluation of the current Italian situation mainly based on bioplastic bags, a peculiar type of recycled paper bag, specifically manufactured for the food waste collection, was examined showing potential advantages during the food waste household storage and in the food waste treatment.

Introduction

The organic fraction is the most relevant among all the separately collected materials in the municipal solid waste in Italy: in the year 2019, 6.4 million tonnes of organic waste were separately collected and treated in composting or anaerobic digestion plants [14]. The organic fraction is mainly composed of food waste from households (more than 70%), as well as of green waste from gardens.

In Italy, the current collection systems of food waste from households are mainly based on the use of bioplastic bags, together with a very small amount of paper bags (< 1%). Most of the bioplastic bags currently employed for food waste collection is made of the starch-based Mater-Bi® polymer whose composition is 70% polybutylene adipate terephthalate, 20% starch, and 10% additives [10].

Regarding the treatment, in 2019 the organic waste was processed in 345 composting and anaerobic digestion plants [14], with the latter rapidly increasing, due to several advantages, such as a more favorable energy balance and the presence of economic incentives for biomethane production. Focusing on the sole food waste, in 2019 about 1.7 million tonnes were sent to composting, while 2.9 million tonnes were processed in both combined anaerobic/aerobic plants as well as in purely anaerobic ones, compared to just 1.6 million tonnes in the year 2015 [14].

The anaerobic treatment typically requires the removal of bioplastic bags before the digestion process, especially when wet or semi-dry technologies are employed, because they cannot be separated from plastic bags and their management can cause hydraulic problems in the plant. During such removal, a non-negligible amount of food waste remains inside the bags and is then not delivered to the anaerobic digestion. This leads to a lower production of biogas/biomethane and to an increase of the process residues that must be furtherly treated or disposed of.

This is particularly evident when comparing the waste composition analyses of the material entering the treatment plants with the actual amounts of residues that are generated by the same plant. The Italian Composting Consortium has carried out 850 analyses on 127 tonnes of food waste, finding an average amount of non-compostable materials lower than 5% in weight on a wet basis [9] but the actual amount of residues generated at the treatment plants is substantially higher. In several examined plants, characterized by different technologies (wet, semi-dry, and dry) and receiving food waste mainly inside bioplastic bags, residues account for more than 15% of the input waste on average, with maximum values close to 30%. They are largely constituted by bags, that accidentally drag a considerable amount of organic substance.

On the contrary, the above limitations are remarkably reduced when using paper bags for the collection. Paper is more compatible with the anaerobic digestion process and therefore it does not require prior removal. This would lead to a greater simplicity of the plant design in terms of a more basic pre-treatments stage and a consequent reduction of the generation of residues. As an example, the sole Italian plant that currently receives only food waste contained into paper bags generates < 10% of residues.

The aim of this research is to analyze the environmental and energy performances of the food waste treatment chain (storage at the household, collection, transport, treatment, and management of residues), with a particular focus on the impact of the different types of collection bags. This paper reports the first part of the study, focused on two aspects: (i) the behavior of the food waste during the household storage when collected inside of bioplastic and paper bags; (ii) the anaerobic degradability of both bag types by means of lab-scale Biochemical Methane Potential (BMP) tests.

With reference to the first objective, the weight loss of the food waste during the household storage was firstly evaluated to assess how it is affected by the type of bags used for the collection. The evaluation of the collection bag behavior during the household storage is important for its influence on the amount and potentially on the quality of food waste that is subsequently collected, transported, and sent to treatment plants. The present study aimed at investigating the progressive bag filling due to the daily food consumption, with a procedure similar to that adopted in [19]. On the contrary, most of the previous studies on this topic were carried out with the initial complete bag filling [4, 13, 24].

The behavior of the different collection bags in the anaerobic digestion process was then analyzed at the lab-scale by means of BMP tests, in order to evaluate the maximum amount of methane achievable under anaerobic conditions. This test measures the ultimate methane production from organic feedstocks in an optimized batch system, and it is the key analytical tool to characterize organic substrates with respect to their behavior under anaerobic degradation conditions. Literature delivers some recent publications related to the evaluation of the anaerobic degradability of starch-based bioplastics (such as Mater-Bi®) [2, 5, 6, 11, 25, 30]. Regarding paper bags, to the authors knowledge, there are no publications related to BMP assays performed on bags specifically manufactured for food waste collection, such that analyzed in the present research.

Materials and Methods

In both household storage assays and BMP tests, the following different typologies of food waste collection bags were compared: (i) bags made of recycled paper, especially manufactured for the food waste collection. They are provided with a separated cartonboard bottom to be inserted into the bag before its use; (ii) bioplastic bags (made of the starch-based Mater-Bi® polymer). Two typologies of bioplastic bags were tested: those specifically designed for the food waste collection (so-called dedicated) and the conventional shopping bags that, after being used for the purchase at the supermarket, can be re-used for the collection of food waste (shopper). Further information about differences in the chemical composition of the bags were not made available by the producers.

Household Storage Tests

The first comparison between bioplastic and paper bags was performed at the level of waste generation. In detail, the waste weight loss during the time occurring between the delivery in the bag by the user and the collection was analyzed by adopting a dynamic, progressive bag filling approach. Accordingly, during 2 years, 112 domestic tests were performed in parallel to compare paper and bioplastic bags behavior: 59 paper vs. bioplastic dedicated bags and 53 paper vs. bioplastic shopper bags.

In the comparative tests, one paper bag and one bioplastic bag were placed inside aerated bins. Before each bag filling (twice a day, after lunch and dinner), the food waste was homogenized, split into two portions with the same weight discharged respectively in the paper bag and in the bioplastic bag. Each comparative test lasted 120 h (5 days). At the end of the test, the two bags were removed and weighed with a scale (readability of 1 g). The weight loss (WL) with respect to the total inserted waste was subsequently calculated, for both bags, according to Eq. (1) where *WL* is the weight loss, *WI* is the weight of waste inserted into the bag, and *WF* is the final weight of the waste (after 120 h).

$$WL = (WI - WF)/WI \tag{1}$$

Moreover, in 73 of the performed tests (38 paper vs. bioplastic dedicated bags and 35 paper vs. bioplastic shopper bags), the weight loss was evaluated not only at the end of the test but also before each bag filling.

In addition to the evaluation of the weight loss, observations on the resistance of the bags were performed at the end of the tests.

Six different commercial types of bags (three dedicated and three shopper bags) from six different producers were tested. The analyses were performed during the different seasons, with the aim to consider the variations of both the environmental conditions (temperature, humidity) and the composition and characteristics of food waste. The following temperature ranges were observed: 19–21 °C (winter), 20–26 °C (spring), 25–29 °C (summer), and 19–23 °C (autumn). The tests were conducted by different households to consider various eating habits and therefore different amounts and characteristics of the generated food waste.

Statistical Analysis

The differences between bioplastic dedicated and paper bags and between bioplastic shopper and paper bags were statistically tested using the software SPSS v.25. First, the normality of the weight loss distribution, for each bag typology, was numerically verified by applying the Kolmogorov–Smirnov and the Shapiro–Wilk tests (with a 0.05 significance level). Normality conditions were not always satisfied and therefore the non-parametric Mann–Whitney U test (with a 0.05 significance level) was selected for the evaluation of the differences between the two materials.

The differences in the weight loss distributions among the seasons were also statistically tested for each bag typology. As stated by the Shapiro–Wilk test (more reliable according to the small samples size), normality conditions were not always satisfied and therefore the non-parametric Kruskal–Wallis test (with a 0.05 significance level) was applied for the evaluation of differences among seasons.

BMP Tests

The anaerobic degradability and the corresponding biomethane yield of the three bag typologies were evaluated in batch at the laboratory scale, by means of BMP tests.

Experimental Plan of BMP Tests

Tests were carried out with three different types of digestate, serving as inoculum, sampled from full scale anaerobic digestion plants processing food waste: (i) *inoculum 1*—test series with a digestate from a wet mesophilic plant where food waste is delivered exclusively inside paper bags; (ii) *inoculum 2*—test series with a digestate from a wet mesophilic plant where food waste is mainly delivered inside bioplastic bags, the most common situation in Italy; (iii) *inoculum 3*—test series with a digestate from a wet thermophilic plant where food waste is mainly delivered inside bioplastic bags.

Inoculum 1 and *inoculum 2* were selected to verify different behaviors between digestates having a different acclimatization to the examined bag typologies. Thereafter, *inoculum 2* and *inoculum 3* were used for comparing results under mesophilic and thermophilic conditions. Total solids (TS), volatile solids (VS), and pH were measured for all the three inocula (Table 1).

During the first test series (*inoculum 1*), bags were manually cut in square pieces with 0.5 cm and 2 cm sides to check if differences in the size of the bag pieces at the macro level can affect the anaerobic digestion process and the repeatability of BMP test replicates. Since the robustness of results slightly improved (lower standard deviation) with the smallest size (0.5 cm), the subsequent series of tests were carried out only with this size.

BMP Test Setup

Each test was performed in duplicate using 600 mL bottles. Before all tests, the digestate was preincubated for 5–7 days at test temperature to decrease the endogenous methane production. The tests with *inoculum 1* and *inoculum 2* were performed at mesophilic conditions, while thermophilic conditions were adopted for *inoculum 3*. An inoculum to substrate ratio (I/S) equal to 2 $VS_{inoculum}/VS_{substrate}$ was adopted for all tests. A mineral medium containing macro and micronutrients was also dosed and tap water was added to reach a 480 mL working volume. Two bottles serving as a blank were prepared for each test series dosing the same amount of inoculum used for testing the substrate, the mineral medium and tap water. Before the tests, pH was measured resulting in the range 7.5–8.3 for all the tests and N_2 was flushed for 5 headspace volume exchanges to ensure initial anaerobic conditions [12, 26]. Table 1 summarizes the conditions adopted for all the tests and the main characteristics of the inocula.

The methane was measured by means of a volumetric system (Automatic Methane Potential Test System II, Bioprocess Control®). At the end of the tests, pH was measured, resulting in the range 7.1–8.0 for all tests. The methane yield for each bottle was calculated as the difference between the accumulated methane volume at standard conditions (273 K, 1 atm) from the *i*-th bottle with the substrate and the average accumulated methane volume at standard conditions from the blank samples; the result was divided by the mass of substrate as VS dosed in the *i*-th bottle. The BMP of each substrate was defined as the average of the test replicates when the daily net production of each of the last 3 days resulted lower than 1% of the cumulative net production until that day [17].

Lastly, at the end of each test, the digestate from each bottle was sieved at 2 mm to recover the undigested bag pieces, which were washed and then dried at 30 °C for mass balance calculations. A 500 µm sieve was further used for the samples of the *3B* test to recover smaller bag pieces. Microscope observations of the recovered materials were also performed.

Focusing on the role of the examined food waste collection bags, the biomethane productions per gram of bag, evaluated according to VS and TS contents, and subsequently per kilogram of discarded food waste were calculated. They depend on the tested bags weight (paper bag = 22.4 g/bag; bioplastic dedicated bag = 7.4 g/bag; bioplastic shopper bag = 13.9 g/bag) and on the quantity of waste that can be carried in it. The capacity of the three examined bags, evaluated with experimental tests, resulted 2.3 L for the paper and the bioplastic dedicated bag, and 2.4 L for the bioplastic

Table 1 BMP test conditions and inocula characteristics

Substrate			Test conditions		Inoculum characteristics		
Bag typology	Test ID	Size (cm)	Temperature (°C)	Inoculum	pH	TS (g/kg)	VS (g/kg)
Paper	*1A-2 cm*	2 × 2	35 ± 0.5	*Inoculum 1*	7.5	16.6 ± 0.1	10.2 ± 0.1
	1A-0.5 cm	0.5 × 0.5					
Bioplastic dedicated	*1B-2 cm*	2 × 2					
	1B-0.5 cm	0.5 × 0.5					
Bioplastic shopper	*1C-2 cm*	2 × 2					
	1C-0.5 cm	0.5 × 0.5					
Paper	*2A*	0.5 × 0.5	35 ± 0.5	*Inoculum 2*	8.1	25.9 ± 0.7	14.8 ± 0.7
Bioplastic dedicated	*2B*	0.5 × 0.5					
Bioplastic shopper	*2C*	0.5 × 0.5					
Paper	*3A*	0.5 × 0.5	50 ± 0.5	*Inoculum 3*	8.1	26.7 ± 0.1	12.0 ± 0.01
Bioplastic dedicated	*3B*	0.5 × 0.5					
Bioplastic shopper	*3C*	0.5 × 0.5					

shopper bag (limited by the capacity of the bin). Anyway, generally this capacity is not totally utilized, since food waste is typically collected twice a week. For this reason, a filling level equal to 2 kg food waste/bag was assumed for all the examined bags.

BMP Kinetic Interpretation

The kinetic behavior was firstly investigated, by evaluating the time to achieve 50% and 90% of the final BMP. Then, in order to strengthen such results, the Levenberg–Marquardt algorithm was used for least squares curve fitting on all data available (irrespective to the termination criterion), adopting different model equations depending on the type of bag. The estimates and standard errors of model's parameters, the 95% confidence interval and the adjusted coefficient of determination of the model's fitting were also determined.

Easily hydrolysable substrates or substrates with low potential of inhibition are normally well described by reverse L-shape curves such as the first-order kinetic and the Chen and Hashimoto models [7]. The first-order kinetic model (Eq. 2) is commonly applied to simulate the anaerobic biodegradation since it describes the BMP results when disintegration/hydrolysis is the rate-limiting step [15], no lag phase is observed, and G_0 (the maximum methane yield achievable at infinite time) represents the total yield of hydrolysable VS at the beginning of the test. In detail, $G(t)$ is the cumulative methane yield at time t (NmL CH_4/gVS), G_0 is the methane potential of the feedstock (NmL CH_4/gVS), k is the first-order disintegration rate constant as well as methane production rate constant (L/day), and t is the anaerobic digestion time (day).

$$G(t) = G_0 \times \left[1 - exp(-k \times t)\right] \quad (2)$$

Complex substrates containing high level of slowly degradable patterns result in curves more related to the S-shape (sigmoidal) or the stepped curve such as modified Gompertz and Logistic models [15, 29]: complex organic compounds are converted to less complex soluble organic compounds by enzymatic hydrolysis, becoming available to bacteria for the subsequent phases. A lag phase at the beginning of the batch tests can be observed due to the initial breakdown of complex substrates [31]. Despite it has no clear biochemical interpretation based on reaction kinetics, the modified Gompertz model (Eq. 3) includes the estimate of the duration of the lag phase (λ), then matching the S-type curve model. In Eq. (3), the maximum rate of methane production (μ_m) is the inflection point and G_0 is the horizontal asymptote of the curve.

$$G(t) = G_0 \times exp\left\{-exp\left[\frac{\mu_m \times exp(1)}{G_0} \times (\lambda - t) + 1\right]\right\} \quad (3)$$

As explained in "BMP Tests" section, stepped trends can be easily recognized in some of the experimental curves obtained for bioplastics: however, as an in-depth study pertaining the kinetics of bioplastics degradation is not the main target of this study, the first-order kinetic model was used to fit BMP data to get a general overview on the kinetic behavior and allow for a comparison among different bioplastics. The Gompertz model, common when dealing with cellulose substrates [18], was instead adopted for fitting paper bag data, then accounting for the lag phase needed to enzymes to hydrolyze complex carbohydrates (cellulose) before producing biogas.

Analytical Methods

TS and VS were determined in duplicate according to Standard Methods 2540 [1]. The pH was measured by means of a portable multi-probe meter (Hach-Lange, HQ40D). The chemical oxygen demand (COD) was determined according to Standard Methods 5220 [1]. Microscope observations were performed using an optical microscope (BM60 Optech, Exacta + Optech Labcenter S.p.A., Italy).

Results and Discussion

Household Storage

The results of the evaluation of the domestic weight loss are reported in Fig. 1a for the 59 tests performed to compare paper and bioplastic dedicated bags. On average, the weight loss of the waste inserted into the bioplastic dedicated bags is 23% lower compared to that of waste collected inside the paper bags. As regards the statistical analysis, the results of the non-parametric Mann–Whitney U test (U = 905; asymptotic significance = 7.00E−6; mean rank: paper = 73.66, bioplastic dedicated = 45.34) show a statistically significant difference between paper and bioplastic dedicated bags (higher weight loss for paper). This aspect is related to the breathable fabric of paper that allows for a relevant evaporation of moisture, giving a higher weight loss. Moreover, a better oxygenation of food waste could allow for a faster activation of an aerobic degradation process, that in turn will favor the evaporation of moisture because of the temperature increase. According to [3], the use of paper bags for the food waste collection will lead to important weight reduction (up to more than 25%), mainly due to the evaporation of water.

The results of the 53 comparative paper vs bioplastic shopper tests are shown in Fig. 1b. On average, the weight loss of the waste inserted into the bioplastic shopper bags is 31% lower compared to that of waste collected inside the paper bags. The difference between paper and bioplastic shopper bags is higher compared to that observed between

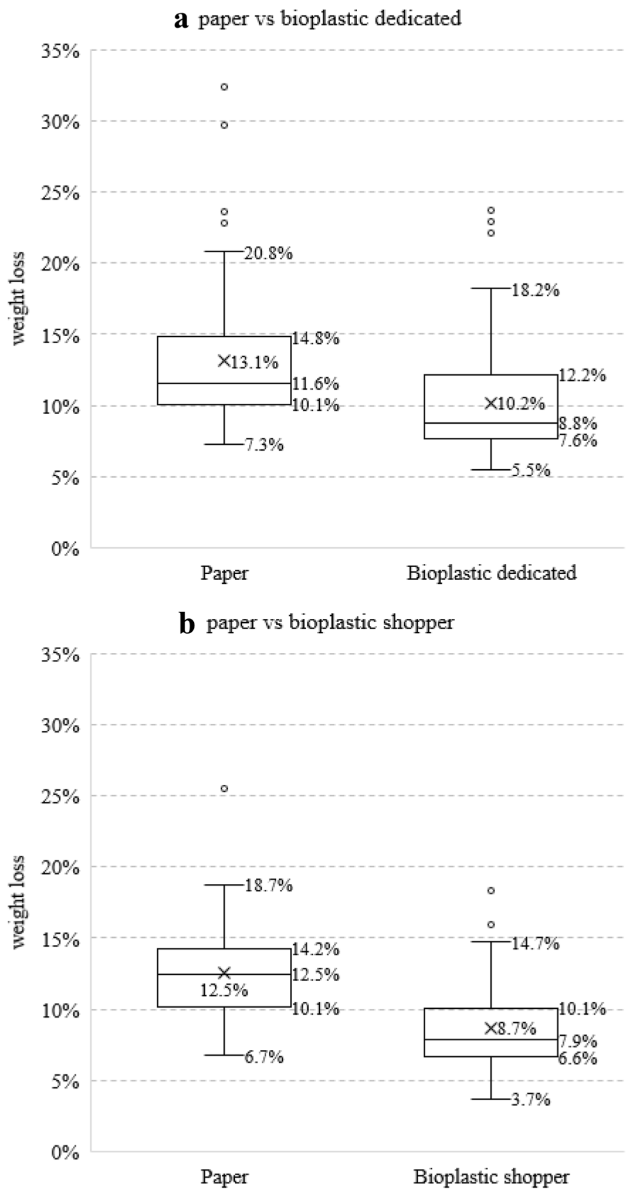

Fig. 1 Household storage tests results: food waste weight losses in the time between the delivery in the bag by the user and the collection for the 59 comparative tests paper vs bioplastic dedicated bags (**a**) and the 53 comparative tests paper vs bioplastic shopper (**b**)

paper and bioplastic dedicated bags: this is likely because shoppers are thicker compared to dedicated bags, since they must guarantee the mechanical properties required for their primary use (carrying the purchased goods at the supermarket).

According to the results of the Mann–Whitney U test (U = 517; asymptotic significance = 2.05E−8; mean rank: paper = 70.25, bioplastic shopper = 36.75) the difference between paper and bioplastic shopper bags (bigger weight loss for paper) is statistically significant.

Table 2 summarizes the main literature findings of similar evaluations. During almost all the tests, the bags were completely filled from the beginning, and the weight losses were evaluated in the following days (up to 7 days on average). Firstly, when paper and bioplastic bags are compared, results always show higher losses for the paper. Excluding the tests performed with closed bins, all the examined bags show a rapid increase of the losses in a few days. This aspect is strictly related to the predominant methodology of carrying out the test with the initial complete bag filling. The only "dynamic" study, based on a progressive bag filling, shows substantially lower losses after 4 days from the beginning of the test [19].

Figure 1 indicates the average losses after 5 days of test (13.1% and 12.5% for paper bags, 10.2% for dedicated bioplastic bags and 8.7% for bioplastic shoppers). Comparing these results to those reported in the examined literature, the average losses of waste contained into bioplastic bags are about twice those obtained in [19] after 4 days of dynamic filling. On the contrary, losses of the performed test on Mater-Bi® bags are up to 73% lower when compared to 7 days losses of tests with a static initial filling (Table 2) but also generally lower than losses measured 3 days after the initial filling, up to 46% when compared to [24]. Similarly, for paper bags, losses of the performed test are about 64% lower when compared to losses after 7 days of tests performed in [24] but also about 45% lower than losses 3 days after the initial filling. This comparison indicates that tests with a static initial filling reported in literature do not represent the real domestic situation.

The observations of the physical status of bags at the end of tests performed to compare paper and bioplastic dedicated bags highlighted some breakings of dedicated bioplastic bags with some release of leachate at the bottom of the collection bin, mainly during spring and summer when wet food waste was inserted (e.g. melon seeds, watermelon rinds). On the contrary, a relevant wetting of paper bags was observed in the same tests but without any breaking (Fig. S1 to S3 of the Supplementary Information).

As regards the comparative paper vs bioplastic shopper tests no breakings were observed.

In addition to the differences between paper and bioplastic bags, the study focused on the evaluation of the main factors affecting the losses. Figure 2 reports the results of all 112 tests, for each bag typology, split among the four seasons.

Considering the samples means, for paper bags, the weight loss in summer is 65% higher compared to that of autumn (the season with the lower mean), while for bioplastic dedicated and shopper bags the weight loss in summer is respectively 83% and 75% higher compared to that of winter. As regards the differences in the weight loss distributions among seasons, the following

Table 2 Summary of tests for the evaluation of the weight loss of food waste contained into different bag typologies; examined bags and main results for bioplastic and paper bags

References	Examined bags and bins	Test typology	Waste weight losses during the storage into bioplastic and paper bags
[19]	Compostable bag + aerated bin Polyethylene bag + aerated bin Compostable bag + closed bin Polyethylene bag + closed bin	Dynamic test (1/4 bag filling per day)	Compostable bag + aerated bin: 4.7% losses after 4 days
[4]	Polyethylene bag Mater-Bi® bag Paper bag	Static lab test (initial complete filling)	Mater-Bi® bag: 4% loss after 1 day – 7% loss after 3 days Paper bag: 9% loss after 1 day – 19% loss after 3 days
[24]	Combination of 3 bag typologies (polyethylene, Mater-Bi®, paper) and 3 bin typologies (open, aerated, closed)	Static lab test (initial complete filling)	Mater-Bi® bag + aerated bin: 12% loss after 2 days –16% loss after 3 days – 26% loss after 7 days Paper bag + open bin: 17% loss after 2 days – 23% loss after 3 days – 35% loss after 7 days Closed bins after 7 days: 4% (Mater-Bi® bag) – 5% (paper bag)
[13]	Mater-Bi® bag + open bin Mater-Bi® bag + closed bin Polyethylene bag + closed bin	Static lab test (initial complete filling)	Mater-Bi® bag + open bin: 15% loss after 3 days – 32% loss after 7 days Mater-Bi® bag + closed bin: <4% loss after 7 days

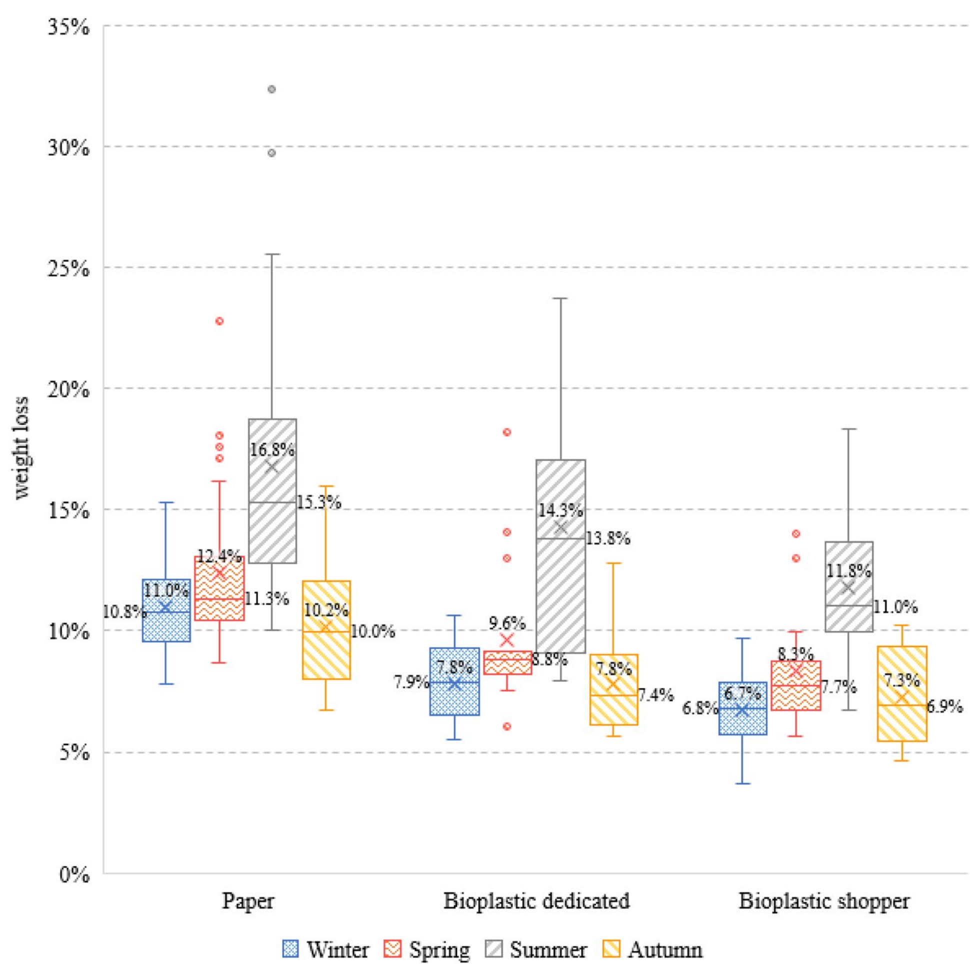

Fig. 2 Household storage tests results: food waste weight losses in the time between the delivery in the bag by the user and the collection for all the 112 performed tests. Results for the three examined bag typologies (paper, bioplastic dedicated, bioplastic shopper) are split among the four seasons (the mean and the median are reported respectively inside and close to each box)

results of the Kruskal–Wallis test were obtained for the three bag typologies: (i) paper bag (H = 42.37; asymptotic significance = 3.34E−9; mean rank: winter = 42.63, spring = 55.07, summer = 85.78, autumn = 32.60); (ii) bioplastic dedicated bag (H = 22.57; asymptotic significance = 5.00E−5; mean rank: winter = 20.31, spring = 31.19, summer = 44.88, autumn = 18.30); (iii) bioplastic shopper bag (H = 21.31; asymptotic significance = 9.10E−5; mean rank: winter = 17.43, spring = 25.71, summer = 41.80, autumn = 20.00). According to these results, for all the examined bags, the weight loss distribution is statistically different in at least one pair of seasons.

Consequently, to test the different pairs, a post hoc procedure for the pairwise multiple comparison was performed. Mann–Whitney U pairwise tests were carried out (the considered significance level was 0.05/3 = 0.017 according to the Bonferroni correction that allows to adjust the rejection level on the total number of tests). Results of the pairwise tests are reported in Table S1 of the Supplementary Information for all bags. The pairwise comparison showed, for all bags, a weight loss statistically different (higher) in summer with respect to the other seasons. On the contrary, the weight loss is statistically the same in winter, spring, and autumn for all bags, with the exception of spring and autumn for the paper bag. Results are in accordance with the temperatures at which tests were performed, that were very similar in winter, autumn, and spring (excluding some hot spikes) and sensibly higher in summer (always > 25 °C).

The seasonal variability in summer compared to the other seasons is evident also observing the weight loss behavior during the 73 tests in which it was evaluated not only at the end of the test but also before each bag filling, as shown in Fig. 3. In winter, the loss trend shows a more consistent increase in the first hours, followed by a stabilization. On the contrary, during summer, the increase is more constant until the end of the test. As an example, for paper, very similar average losses were observed after the first 32 h: 5% and 6% respectively in winter and summer. The difference between the two seasons increases after 80 h (8% and 12%) and achieves its maximum value at the end of the test (12% and 18%).

BMP Tests

Substrates characterization in terms of TS, VS, and COD is hereafter reported: (i) dedicated bioplastic bag: TS = 988 ± 2.2 g/kg; VS = 959 ± 7.3 g/kg; COD = 1345 ± 344 g/kg; (ii) shopper bioplastic bag: TS = 989 ± 1.9 g/kg; VS = 875 ± 0.5 g/kg; COD = 1506 ± 2 g/kg; (iii) paper bag: in this case, TS, VS, and COD were defined according to the proportion between its two components: main paper bag (78% by weight–wet basis, TS = 977 ± 1.0 g/kg; VS = 899 ± 1.3 g/kg; COD = 1094 ± 27 g/kg) and cartonboard bottom (22%, TS = 973 ± 0.3 g/kg; VS = 830 ± 1.0 g/kg; COD = 1100 ± 88 g/kg).

For each test, Table 3 reports the mean and the standard deviation of the final BMP value, the coefficient of variation of the test, and the anaerobic degradability evaluated considering a theoretical methane production of 330 NmL

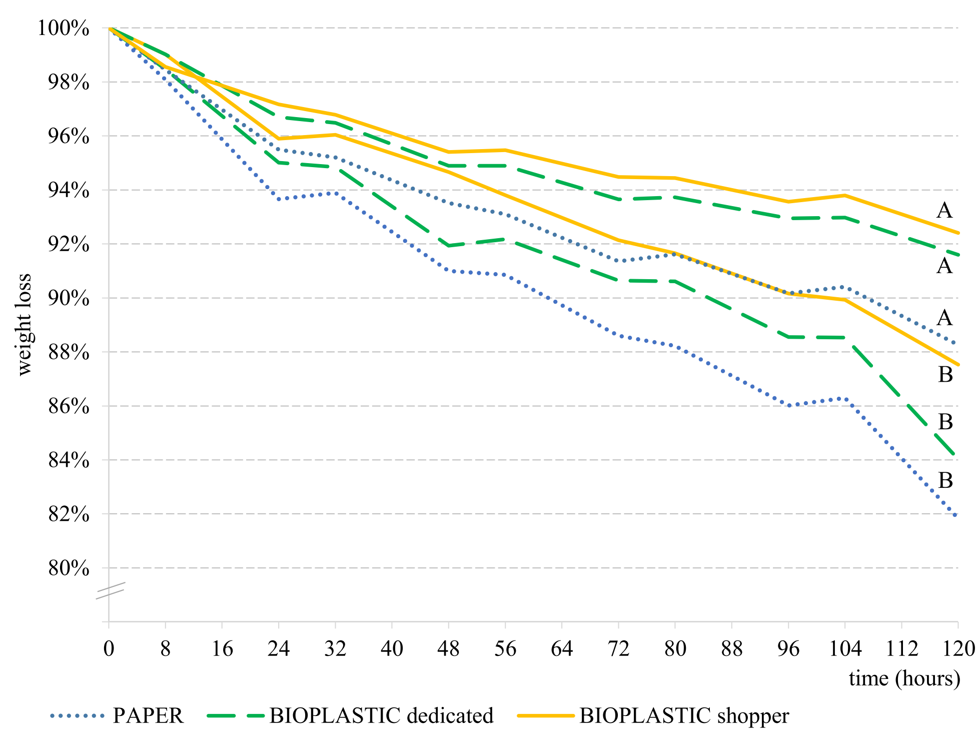

Fig. 3 Household storage tests results: change in weight losses during the tests performed in winter (A) and summer (B). For each bag typology the mean of performed tests is reported

Table 3 BMP test results: final BMP mean ± standard deviation calculated on VS basis (BMP); coefficient of variation of the test (CV); anaerobic degradability on COD basis (AD-COD); comparison between paper and bioplastic bags; final mean BMP calculated per gram of bag (BMP-bag); final mean BMP per kg of waste inserted into the bag (BMP-waste)

Test ID	Inoculum	Substrate	BMP (NmL CH_4/gVS)	CV (%)	AD-COD (%)	Bioplastic vs paper (%)	BMP-bag (NmL CH_4/g bag)	BMP-waste (NmL CH_4/kg waste)
1A-2 cm	*1*	Paper	235 ± 12	5.2	58	–	208	2327
1A-0.5 cm			225 ± 38	17	55	–	199	2229
1B-2 cm		Bioplastic dedicated	62.1 ± 12.2	N.A.	N.A.	N.A.	N.A.	N.A.
1B-0.5 cm			42.0 ± 0.2	0.5	9.1	− 81	40.2	149
1C-2 cm		Bioplastic shopper	60.3 ± 13.5	22	11	− 74	52.8	367
1C-0.5 cm			62.4 ± 0.3	0.5	11	− 72	54.6	380
2A	*2*	Paper	272 ± 7	2.6	66	–	240	2687
2B		Biopl. dedicated	56.5 ± 0.4	0.7	12	− 79	54.2	201
2C		Biopl. shopper	86.1 ± 15.1	18	15	− 68	75.4	524
3A	*3*	Paper	262 ± 6	2.4	64	–	232	2597
3B		Biopl. dedicated	263 ± 10	3.8	57	+0.01	252	931
3C		Biopl. shopper	127 ± 4	2.9	22	− 52	111	774

Because of a technical issue *1B-2 cm* test result is not considered reliable. Further calculations on this data are omitted

N.A. not available

CH_4/gCOD (about 6% of COD used for growth). Because of a technical issue, the result of the *1B-2 cm* test is not considered reliable by the authors, then it is not further interpreted in the following. Despite this, the *1B-2 cm* test was not repeated since 0.5 cm size was then selected for the subsequent test series (*inoculum 2* and *inoculum 3*). Moreover, Table 3 reports the difference between the materials calculated according to Eq. (4).

$$Bioplastic\ vs\ paper = \left(BMP_{bioplastic} - BMP_{paper}\right)/BMP_{paper} \quad (4)$$

Under mesophilic conditions (*inoculum 1* and *inoculum 2*), the final BMP resulted 70–80% lower for both bioplastic types compared to that of the paper. Comparing the results for the same substrate, the digestate of the plant where food waste is delivered inside bioplastic bags (*inoculum 2*) allows for a higher final BMP (although obtained in a longer time) not only for the bioplastic bags but also for the paper bag: for both, an increase of about 20% is observed compared to the result of *inoculum 1* series. This is likely to be ascribed to the diversity of the biomass used as inoculum: the plant from which *inoculum 2* was sampled receives not only bioplastic bags but also cellulose, for example in the form of paper towels or napkins, while *inoculum 1* biomass is not used to deal with bioplastics.

With reference to the test series performed under thermophilic conditions (*inoculum 3*), the final BMP of paper and bioplastic dedicated bags is very similar, although obtained in a longer time for the latter. On the contrary, an important difference between paper and bioplastic shopper bags is still observed (52%), although lower than the difference found under mesophilic conditions.

As expected, paper bags were found to be highly biodegradable under all test conditions. Conversely, bioplastic bags resulted not much degradable under mesophilic conditions, while thermophilic conditions were more effective, with differences between dedicated and shopper bags. In particular, the percentage of conversion of COD into biogas for bioplastic dedicated bags increased from about 10% under mesophilic conditions to about 60%, while it remained within 22% for shopper bags. The different degradability of bioplastics is supposed to be related to a double beneficial effect of thermophilic conditions: the presence of different types of biomass, possibly more effective in the hydrolysis of bioplastics, and the weakening of the molecular structure of bioplastics at higher temperatures, likely improving its accessibility to microorganisms.

Results were then compared to findings from previous studies, of which those pertaining to bioplastics are summarized in Table 4. The final BMPs of bioplastic bags under mesophilic conditions are in the range 42.0–86.1 NmL CH_4/gVS, comparable to those reported in [6, 25] for Mater-Bi® and in [30] for two starch-based films. On the contrary, in [2] a significantly higher BMP was observed, although obtained in a very long time (250 days of test).

Considering thermophilic conditions, the result of bioplastic shopper bags (127 NmL CH_4/gVS) is very similar to those obtained in [25] for Mater-Bi® and in [11] for a film

Table 4 Publications related to BMP tests on Mater-Bi® and starch-based bioplastics; typologies of examined substrates and BMP tests results

References	Examined bioplastics	BMP test results for Mater-Bi® and starch-based bioplastics
[2]	Sugar cane cellulosic-fibres plate Starch bioplastics cutlery Starch bioplastics carrier bag Polylactic acid (PLA) items Polyhydroxyalkanoate (PHA) granules	Starch bioplastics carrier bag Mesophilic conditions: 201 mL CH_4/gVS
[5]	Mater-Bi® bag Bio-based wine cork Cellulosic plate	Mater-Bi® bag Extremely variable (no production in some replicates) Mesophilic conditions: up to 152 mL CH_4/gVS Thermophilic conditions: up to 186 mL CH_4/gVS
[6]	Coffee capsules made of three different bioplastics (Mater-Bi®, Vegemat®, Ecovio®)	Mater-Bi® capsule Mesophilic conditions: 68 mL CH_4/gVS Thermophilic conditions: 247 mL CH_4/gVS
[30]	4 cellulose-based films Cellulose diacetate film 2 starch-based films PLA film	Mesophilic conditions: 113 mL CH_4/gVS (starch-based film 1) 69 mL CH_4/gVS (starch-based film 2) In addition, the two examined starch-based bioplastics show little or no evidence of degradation in semi-continuous tests in which they are daily fed to the digester together with food waste
[25]	Mater-Bi® film Rigid PLA	Mater-Bi® Mesophilic conditions: 33 mL CH_4/gVS Thermophilic conditions: 113 mL CH_4/gVS
[11]	Film derived from corn starch PLA straw/cup PHA film	Film derived from corn starch Thermophilic conditions: 187 mL biogas/gVS

derived from corn starch. On the contrary, the BMP of dedicated bags (263 NmL CH_4/gVS) is sensibly higher, similar to that of Mater-Bi® items tested in [6]. Anyway, the high variability of results is also confirmed by [5]. This aspect suggests the need for further evaluation on other commercial types of bags, especially under thermophilic conditions.

Regarding paper, most of the analyses in the literature are related to cellulose-based items contained in the organic fraction of urban waste [11, 21] or in the overall domestic waste [16]. For dirty paper and dirty cardboard contained in the food waste [21], the BMP resulted respectively 372 and 271 mL CH_4/gVS. As regards cellulose-based plates and kraft paper, BMP tests performed in [11] resulted 507 and 133 mL CH_4/gVS. In [16], several paper waste types (miscellaneous paper, newspaper used for wrapping kitchen waste, wrapping paper, used paper, waste high-quality paper, and paper garbage bag) were analyzed, with BMP values in the range 260–570 mL CH_4/gVS. As a comparison, the BMP of mycrocristalline cellulose (CAS Number 9004-34-6) from an international interlaboratory study resulted 350 ± 29 mL CH_4/gVS [22]. Other evaluations of the BMP are instead related to pulp and paper mill residues, which largely contain cellulose [27, 28].

At the end of the tests, the physical status of the substrates was visually inspected, with Fig. S4 (Supplementary Information) showing some bioplastic bags samples. Under mesophilic conditions, only slight changes in color brilliance were observed, without any appreciable size reduction compared to the input samples. In addition, the residual mass of undigested substrate at the end of the test was washed, dried, and weighed, getting the following results referred to the initial mass of tested sample, as average of replicates, in terms of percentage of substrate not converted: 92% (*1B-0.5 cm*); 83% (*1C-2 cm*); 83% (*1C-0.5 cm*); 91% (*2B*); 85% (*2C*).

The same calculations under thermophilic conditions show different behaviors between the two bioplastic typologies, in line with the test results. For the shopper bags samples (*3C* test), a 66% substrate weight reduction was observed together with a more significant change in color but with small changes in shape and dimension (Fig. S4). On the contrary, for the dedicated bags, a significant size reduction was observed with all the sample residues passing through a 2 mm sieve. Figure S4 shows the residues separated by a 500 µm sieve for the dedicated bioplastic bag. Microscope observations of dedicated bags samples after the tests under thermophilic conditions are reported in Fig. S5 to S7 of the Supplementary Information.

As expected, for all tests performed with paper bags, no sample residues were held by the 2 mm sieve.

Focusing on the role of the examined food waste collection bags, Table 3 reports the calculation of the biomethane production per gram of bag and per kilogram of inserted food waste. With reference to the mesophilic conditions, the specific methane production of paper bags is approximately one order of magnitude higher compared to that of bioplastic bags, with a more marked difference for dedicated bags whose weight is lower with respect to shopper bags. Under thermophilic conditions, the difference between the two examined materials is reduced but still appreciable. Comparing paper and bioplastic shopper bags, the latter still shows a 70% lower production. In the comparison between paper and bioplastic dedicated bags, the latter, even with a higher BMP per unit of fresh matter, shows a 64% lower methane production per kg of waste due to a lower bag weight.

Kinetic Interpretation

The cumulative biomethane production trends over time for all the tests are shown in Fig. 4. Besides the final BMP values, the kinetic behavior was also investigated, firstly by comparing the time to achieve 50% and 90% of the final BMP among different substrates and conditions: data are listed in Table 5, where the increase in the time necessary to go from 50 to 90% is also reported as Δt ($t_{90\%}$–$t_{50\%}$), indicating the extent of the slowing down of the process compared with the initial phase to get the half of the final BMP. The

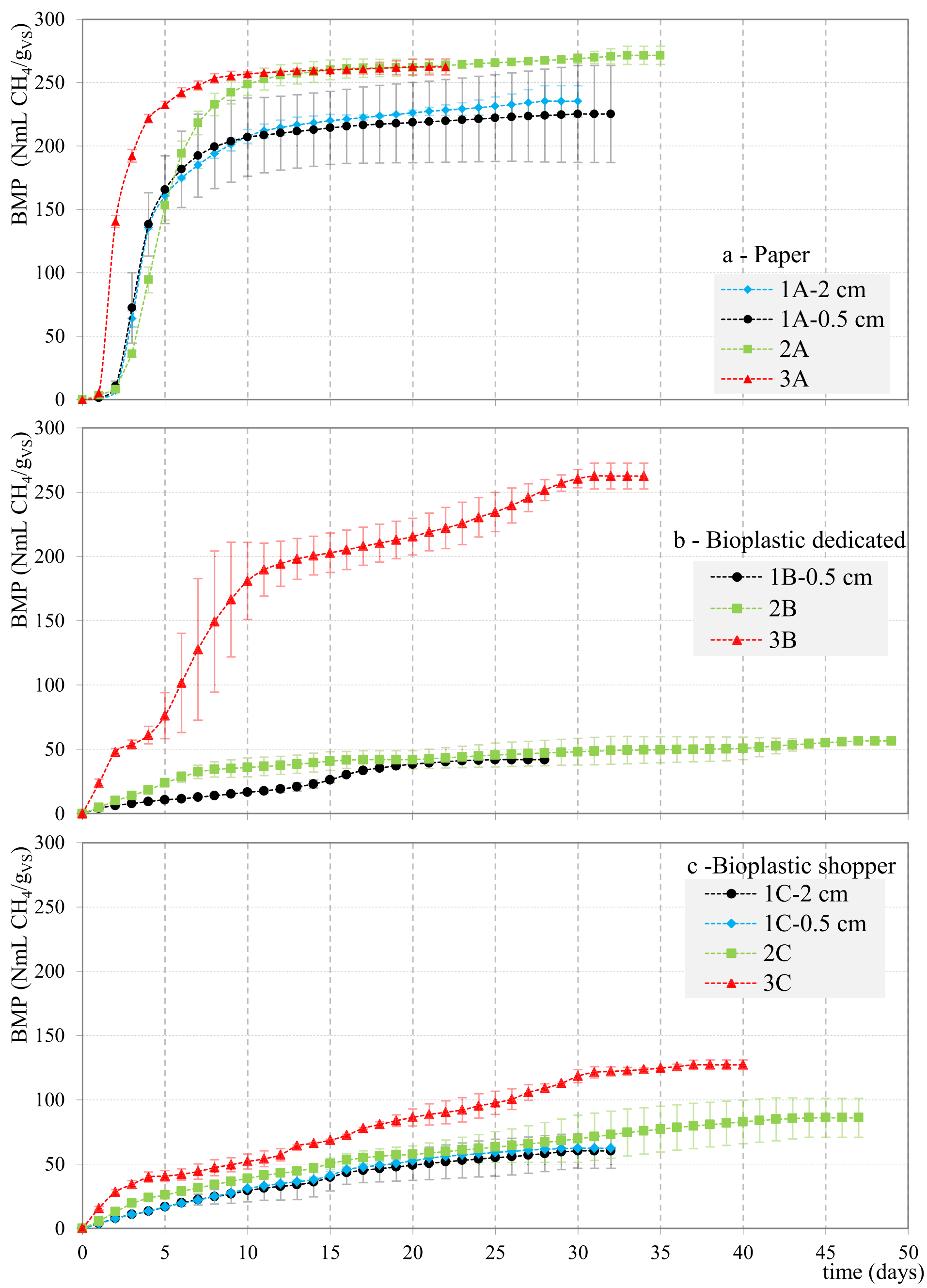

Fig. 4 Cumulative net specific methane production (at standard conditions—T = 0 °C and P = 1 atm) as a function of time, under different testing conditions, for the tested substrates: **a** paper bag, **b** bioplastic dedicated bag, **c** bioplastic shopper bag

Table 5 Kinetic interpretation of BMP tests data

Test ID	Time to get the 50%/90% BMP and Δt ($t_{90\%}$–$t_{50\%}$) (days)		Model parameter estimation					
			Model	Parameter	Estimate	Standard error	95% Confidence interval	R^2 adjusted
1A-2 cm	$t_{50\%}$	4	Gompertz	G_0 (NmL CH_4/gVS)	226	2	222–230	0.979
	$t_{90\%}$	11		μ (NmL CH_4/gVS/d)	44.3	3.6	37.0–51.6	
	Δt	7		λ (d)	1.48	0.22	1.02–1.94	
1A-0.5 cm	$t_{50\%}$	4	Gompertz	G_0 (NmL CH_4/gVS)	218	1	215–221	0.989
	$t_{90\%}$	9		μ (NmL CH_4/gVS/d)	54.1	3.5	46.9–61.3	
	Δt	5		λ (d)	1.68	0.14	1.39–1.97	
2A	$t_{50\%}$	5	Gompertz	G_0 (NmL CH_4/gVS)	267	1	265–269	0.995
	$t_{90\%}$	10		μ (NmL CH_4/gVS/d)	53.5	1.8	49.8–57.2	
	Δt	5		λ (d)	2.25	0.09	2.06–2.43	
3A	$t_{50\%}$	2	Gompertz	G_0 (NmL CH_4/gVS)	260	1	257–262	0.984
	$t_{90\%}$	6		μ (NmL CH_4/gVS/d)	97.0	7.3	82.3–112	
	Δt	4		λ (d)	0.789	0.110	0.565–1.01	
1B-0.5 cm	$t_{50\%}$	14	1st order	G_0 (NmL CH_4/gVS)	67.3	7.7	51.7–83.0	0.957
	$t_{90\%}$	20		k (1/d)	0.0353	0.0062	0.0227–0.0480	
	Δt	6						
2B	$t_{50\%}$	6	1st order	G_0 (NmL CH_4/gVS)	51.8	0.6	50.5–53.1	0.960
	$t_{90\%}$	24		k (1/d)	0.106	0.005	0.0958–0.117	
	Δt	18						
3B	$t_{50\%}$	8	1st order	G_0 (NmL CH_4/gVS)	271	4.7	262–281	0.980
	$t_{90\%}$	26		k (1/d)	0.0900	0.0044	0.0811–0.0989	
	Δt	18						
1C-2 cm	$t_{50\%}$	11	1st order	G_0 (NmL CH_4/gVS)	78.8	1.5	75.7–81.8	0.997
	$t_{90\%}$	25		k (1/d)	0.0476	0.0016	0.0443–0.0508	
	Δt	14						
1C-0.5 cm	$t_{50\%}$	11	1st order	G_0 (NmL CH_4/gVS)	83.4	2.4	78.6–88.2	0.994
	$t_{90\%}$	23		k (1/d)	0.0472	0.0023	0.0425–0.0520	
	Δt	12						
2C	$t_{50\%}$	10	1st order	G_0 (NmL CH_4/gVS)	93.6	1.7	90.2–96.9	0.986
	$t_{90\%}$	29		k (1/d)	0.0503	0.0021	0.0462–0.0545	
	Δt	19						
3C	$t_{50\%}$	13	1st order	G_0 (NmL CH_4/gVS)	155	5.5	144–166	0.973
	$t_{90\%}$	29		k (1/d)	0.0430	0.0030	0.0370–0.0491	
	Δt	16						

degradation of paper is slightly affected by the inoculum type, while it is accelerated under thermophilic conditions. The degradation kinetics of the bioplastics dedicated bag is instead more influenced by the inoculum type than by the temperature: with *inoculum 2*, acclimated to bioplastic, the time to get 50% of the BMP is almost halved. However, compared to *inoculum 1*, the second part of the kinetics was found to be slowed down when using *inoculum 2*, then making the time to get the final BMP approximately the same, irrespective to the inoculum type. Neither the inoculum type nor the temperature conditions have proved to influence the degradation kinetic of bioplastic shopper bags.

Further information on the performance of the substrates under anaerobic conditions were gathered by fitting the cumulative methane production curves obtained from BMP tests with kinetic models. Table 5 reports model's statistics (estimates and standard errors of model's parameters, the 95% confidence interval, the adjusted R-Squared).

Regarding bioplastics, stepped trends are clearly recognizable from Fig. 4, suggesting the existence of diverse and progressive hydrolysis processes, most likely determined by the different accessibility of hydrolysable compounds within the polymer structure. As already found in previous studies [8, 20, 23], BMP curves with a stepped shape are typical for biodegradable polymers containing starch. With reference to model's selection, [8] adopted the modified Gompertz model for fitting BMP data; conversely, in the present study the first-order kinetics was identified as the best choice for data interpretation, based on visual inspection, and model's fitting evaluation (adjusted coefficient of determination). Further, the use of a common first-order kinetics instead of stepped models (that could be different depending on the type bioplastic and on the testing conditions), allows a direct comparison of the kinetic behavior as a whole. The estimates of the first-order hydrolysis constant confirm what already found from $t_{50\%}$ and $t_{90\%}$ data for bioplastics: for shopper bags, a small range of variation (0.043–0.050/day) is observed under all the various conditions tested. Degradation kinetics of dedicated bioplastic bags are conversely positively influenced by the acclimation of the inoculum, bringing an increase of about three times in the kinetic constant, from 0.035/day to 0.106/day; the thermophilic conditions tested on an acclimated biomass slightly influences the kinetic of the process but allows for G_0 values five times higher than those obtained at mesophilic conditions.

Finally, with reference to paper bags, the modified Gompertz model was used to account for the initial lag phase, which is typical for cellulose-based substrates, as already explained in "BMP Kinetic Interpretation" section. The increased kinetics observed at 50 °C suggests that thermophilic conditions both reduce the lag phase of the process (from 2.3 days to 0.79 day) and almost double the maximum rate of methane production. The first effect can be ascribed to the weakening of the chemical structure of paper at high temperatures as well as to the presence of a wider biodiversity of microorganisms; differently, the improvement observed in the maximum rate of methane production is likely to be more related to the different biomass developed at higher temperature.

Conclusions

The type of bag used for the separate collection of the food waste can play a fundamental role in determining the management both at the domestic level and during the processing at the treatment plants. First, bioplastic and paper collection bags have a different behavior during the household storage, with the paper allowing for higher weight losses, up to 44% on average. Currently, the most widespread food waste collection scheme in northern Italy is based on a biweekly curbside system. The reduction of the amount of waste to be collected and above all the lower odor and leachate release observed during their use at the household could pave the way to a potential decrease of the frequency of food waste collection, thus reducing costs and environmental impacts.

The BMP tests performed at the lab scale to evaluate the degradability of bags under anaerobic conditions showed a different behavior for the two materials under mesophilic conditions, with a limited degradation of bioplastic bags compared to an almost complete degradation of paper bags. The comparison showed, for paper bags, a production of methane per kilogram of discarded food waste approximately one order of magnitude higher compared to that of bioplastic bags. Under thermophilic conditions, the behavior of the different examined bioplastic bags is less homogeneous with a more marked difference between paper and bioplastic shopper bags. To further investigate this aspect, additional continuous lab scale tests for the evaluation of the co-digestion of bags together with food waste will be performed.

The performed tests showed the potential of paper bags to improve the system performances with multiple benefits. In order to confirm these results, further steps of the research could include the evaluation of the anaerobic digestion process at the full scale focusing on the response of plants to the different type of bags in terms of biogas production and amount of residues generated. Moreover, thanks to a comparative Life Cycle Assessment, the influence of the collection bag typology on the overall food waste management chain could be evaluated.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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