



Degradation and environmental assessment of compostable packaging mixed with biowaste in full-scale industrial composting conditions

Emmanuelle Gastaldi^{a,b}, Felipe Buendia^{a,c}, Paul Greuet^{a,b}, Zineb Benbrahim Bouchou^{a,c}, Anir Benihya^{a,c}, Guy Cesar^d, Sandra Domenek^{a,c,*}

^a Fondation AgroParisTech, Chaire CoPack, 91120 Palaiseau, France

^b UMR IATE, Université Montpellier, INRAE, L'Institut Agro Montpellier, 34000 Montpellier, France

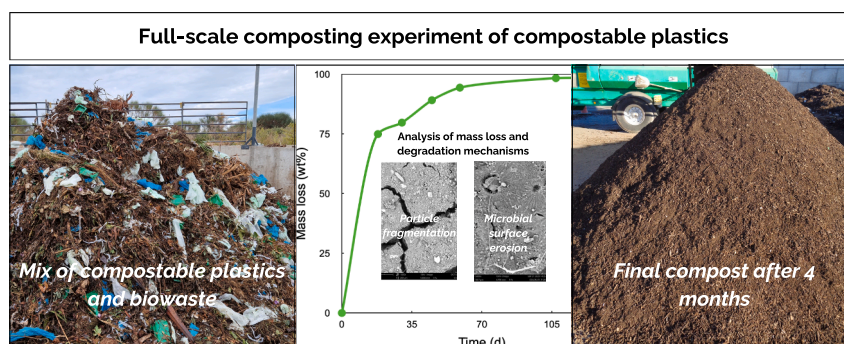
^c Université Paris-Saclay, INRAE, AgroParisTech, UMR SayFood, 91120, Palaiseau, France

^d Serpbio, 64240-La Bastide Clairence, France

HIGHLIGHTS

- Compostable plastics show no impact on full scale composting process.
- Safety and agronomic requirements of organic farming met.
- Composting plastics with biowaste has less environmental impacts than incineration.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Biodegradable plastics
Agronomic quality
Ecotoxicology
Polymer morphology
Life Cycle Assessment

ABSTRACT

The incorporation of representative commercial compostable materials into a full-scale open-air windrow composting process in an industrial site using household-separated biowaste was investigated. Two batches out of the same initial biowaste mixture were studied, one as control and the other containing initially 1.28 wt% of certified compostable plastics. No significant differences in the composting process were revealed. Compostable plastics exhibited a 98 wt% mass loss after 4 months, aligning with industrial composting times. The evolution of the morphology of the materials unveiled polymer specific degradation mechanisms. Both Safety requirements for organic farming were met. Ecotoxicity tests showed no adverse effects, agronomic fertilizing and amending quality was high, the materials compost even enhancing barley growth. The ecological impact assessment demonstrated an advantage for composting over incineration for seven of the eight indicators. In conclusion, this study shows the successful integration of compostable materials into industrial composting, upholding product safety and quality.

* Corresponding author at: UMR SayFood, 22 place de l'Agronomie, 91120 Palaiseau, France (S. Domenek).

E-mail address: sandra.domenek@agroparistech.fr (S. Domenek).

<https://doi.org/10.1016/j.biortech.2024.130670>

Received 8 January 2024; Received in revised form 26 March 2024; Accepted 4 April 2024

Available online 5 April 2024

0960-8524/© 2024 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

The management of municipal waste is a pivotal focus of ambitious policies aimed at mitigating climate change and environmental pollution. In the European Union, total waste generation reached approximately 233 million tons in 2021 (Statistica, 2023), with biowaste accounting for 37 % of this volume (European Environment Agency, 2023) and around 3 % of the EU's greenhouse gas emissions (European Environment Agency, 2020). Sorting biowaste at the household level is a powerful means to reduce the volume of solid municipal waste (Bassi et al., 2017; Policastro & Cesaro, 2023), and the aerobic composting process, a readily available low-tech process, produces compost, a soil conditioner and fertilizer (Vigoroso et al., 2021). Compostable plastics were developed with the intention of mitigating the adverse impacts of non-substitutable single-use plastic products by offering an alternative route to waste management. They can help the citizen in sorting biowaste at home and storing it without nuisances like odors, leaks, or flies (Puyuelo et al., 2013). Disposal alongside biowaste can help to avoiding presence of improper materials (Zhang et al., 2017), which reduce compost quality (Rodrigues et al., 2020). However, there is limited adoption of compostable plastics in waste management practices because of a lack of harmonized regulations (Kedzia et al., 2022) and limited data on the safety of the final products. Indeed, organic soil amendments/conditioners, such as compost, can introduce pollutants like microplastics into the soil because of the collection of biowaste polluted with non-compostable plastics (Edo et al., 2022; Porterfield et al., 2023). Microplastics, particularly at high concentrations, can have inhibitory effects on microbial communities (Zhou et al., 2023) and adverse effects on mesofauna in the soil and plants (Iqbal et al., 2020).

To evaluate the performance and safety of compostable plastics, lab-scale experiments have to be complemented by field trials that mimic real-world conditions (Haider et al., 2019). In contrast to lab-scale biodegradation studies based on standardized methodologies (Briasoulis et al., 2021), full-scale facilities offer less well-standardized environmental conditions, requiring careful documentation. Studies in full-scale composting sites are exceedingly rare in open literature due to their experimental complexity, the need for collaboration with municipal waste treatment facilities, and the substantial quantity of samples required for testing. Some studies exist (Greene, 2007; Klauss & Bidlingmaier, 2004; Zhang et al., 2017; Kale et al., 2006; Musiol et al., 2016a, b; Leppanen et al., 2020; Kawashima et al., 2021). The experiments mainly focused on polylactide (PLA) (Kale et al., 2006; Musiol et al., 2016a, b; Kawashima et al., 2021). Three utilized different types of plastics, making them more representative of real-world conditions (Greene, 2007; Klauss & Bidlingmaier, 2004; Zhang et al., 2017). Generally, tests were carried out by placing the materials in a gasket which was buried in the compost pile. While this setup makes it easier to recover fragments, it does not allow the materials to mix thoroughly with the bulk of the biowaste or provide significant mechanical shear. An in-depth description of the morphological changes of the polymer during the full-scale composting experiment is only available on PLA (Kale et al., 2006; Musiol et al., 2016a,b). Based on these experiments, the present work describes a large-scale experiment conducted at an industrial composting station using open-air windrow technology and receiving source-separated biowaste. To simulate a real-world scenario as closely as possible, different commercially available plastics were directly mixed with the biowaste at the facility and the results of compost quality and safety compared to a same batch without plastics. The degradation mechanism was studied using colored references of each major family of compostable materials (e.g., starch blends, PLA, polyhydroxyalkanoates (PHAs)). The data generated by this experiment were used for a Life Cycle Assessment (LCA) following the quality rules of the European Union Environmental Footprint standard. The environmental impacts of composting process were evaluated versus incineration of the plastics.

2. Materials and methods

2.1. Characteristics of industrial composting site

The composting experiment took place at an industrial composting facility (Syndicat Centre Hérault (SCH), Aspiran 34800, France) serving a sparsely populated rural region of 1,100 square kilometers and a population of 80,000 inhabitants. The SCH platform handles two main types of waste: biowaste and green waste. The biowaste stream originates from households and restaurants. It was separated at the source and collected door-to-door. The platform receives approximately 3,000 tons biowaste and 6,000 tons green waste per year. It uses the turned windrow technology on an impermeable concrete floor. Leachates are recovered and drained for reuse to moisten the organic waste during the first step of the composting process (active phase). The platform produces composts that comply with all the requirements of NF U44-051:2006 standard and are also certified as “fertilizing materials and growing media usable in Organic Agriculture” in accordance with the requirements of the reference framework I302.

2.2. Experimental setup of industrial composting experiment

Commercial certified compostable materials (322.5 kg), including flexible and rigid packaging items such as bags, films, trays, and coffee capsules (Table 1), were provided by various companies affiliated with the French Association of Biobased Compostable materials and members of the philanthropic partnership Chair CoPack (Foundation AgroParisTech).

Biowaste (food waste, leftovers from meals, etc.) and green waste (garden waste) were collected over the course of one week and stored in piles as received. The total mass of collected waste amounted to 40 t. The household biowaste contained green compostable plastic bags, which are used in this region for the collection of food waste. Before starting the experiment, most of the non-compostable items were manually removed from the biowaste, but some probably remained, given the large volume of very heterogeneous biowaste to be cleaned (40 tons). About 550 kg of different types of non-compostable waste (conventional plastics, tin cans, bottles, textiles, sanitary products, diapers, etc.) were removed and transported to the nearest landfill station (ISDND, Soumont 34700, France). After this cleaning process, the biowaste was divided into two batches of 20 t labeled “Materials” and “Control.” To each batch, 2.5 t of plant shreds were added to serve as a source of carbon that could be rapidly assimilated by microorganisms. Additionally, and 2.5 t of structuring agent were incorporated into each batch to allow aeration of pile. This structuring agent was obtained from the screening residues of compost derived from green waste treated at the same site. The composition of both batches was as follows: 80 % biowaste, 10 % plant shreds, and 10 % plant structuring agent.

The compostable plastics were introduced to the Materials batch using a wheel loader to achieve an incorporation rate of 1.28 wt%. The Control batch was not mixed with plastics. Both batches were placed in two individual open-air compartments with three concrete walls each (3 m wide, 6.5 m long and 1.5 m high) corresponding to two piles of 30 m³. The initial temperature, humidity, and pH of the two piles was 45 ± 5 °C, 50 ± 5 % and 8.5 ± 0.5, respectively. All these operations were carried out on the 26th of September 2022.

2.3. Monitoring of industrial composting process

The materials and control batch underwent identical treatment using the usual procedure employed by the composting platform. The temperature of the two batches was recorded three days a week during the first three weeks at 8 points distributed in the composting pile at 0.5 m and 1.5 m depth using large scale thermocouples (Hanna instruments, France). Watering and turning were carried out simultaneously twice a week for the first 3 weeks of the active thermophilic phase. Then

Table 1
Characteristics and amounts of the compostable materials included in the materials batch.

Material	Application	Polymer composition	TUV Austria label	Thickness max–min (µm)	Amount added (kg)	Initial concentration in biowaste (%)	color
E1	Waste bin Bag	Polyester/starch	OKH	93–17	50	0.20	green
E2	Fruit & Veg. Bag	Polyester/starch	OKH	45–12	10	0.04	transparent
E3	Carrier Bag	Polyester/starch	OKi	174–48	65	0.26	blue
E4	Fruit & Veg. Bag	Polyesters blend	OKi	37–9	30	0.12	white
E5	Film	Polyester	OKH	180–22	30	0.12	transparent
E6	Tray	PLA	OKH	429–381	65	0.26	green
E7	Tray	PLA	OKi	406–325	20	0.077	transparent
E8	Tray	Cellulose + polyesters	OKi	450–430	30	0.12	white
E9	Coffee capsule	PHA	OKi	1200–720	20	0.079	pink
E10	Coffee capsule	Polyesters blend	OKi	3089–850	2.5	0.01	white

OKH and Oki are labels delivered by TUV Austria for home composting and industrial composting conditions, respectively; concentration in 20 t of biowaste mixed with plant shred and structuring agent.

frequency reduced to once a week. To monitor the moisture content of the piles, a representative sample was taken once per week and dried in an oven at 103 ± 1 °C until constant weight. When necessary (i.e. in absence of rainfall), the moisture content was adjusted to approximately 50 wt% by adding an appropriate amount of leachate.

The switch from the thermophilic active phase to the maturation phase was determined on site using the self-heating test “Rottegrad” (EN 16087–2:2011), which monitors the temperature increase of a compost sample in a Dewar for 7 days. Monitoring started in from week 4 and was repeated weekly until the sample temperature increase was below 40 °C, which corresponds to grade IV of the Official European stability classification designating finished compost (Brinton et al., 1995). The self-heating test was validated at day 49, i.e. 8 weeks after starting the composting process. The frequency of pile turning and watering was once a week for the first two weeks of the maturation phase. After that, operations were stopped. At the end of the maturation phase (i.e. 19 weeks after the start of the composting), both batches were subjected to sieving to produce the final compost. Three different fractions based on grain size were collected: fine (<12 mm), medium (12–90 mm), and coarse (>90 mm). The water content of each fraction was determined by drying at 103 ± 1 °C to calculate composting yields. The finest fraction was stored in an open-air pile for a ripening period of at least 5 weeks, awaiting the results of the laboratory analysis on compost quality, before being sold as soil improver.

2.4. Assessment of compost quality and ecotoxicity

2.4.1. Assessment of compost quality according to criteria of NFU 44–051 standard

The fine fraction resulting from the sieving (≤ 12 mm) of the two batches of compost was sent to an independent accredited laboratory (SADEF, France) to analyze the safety and agronomic quality according to the NF U44-051:2006 standard dedicated to organic soil improvers. The safety requirements cover the determination and quantification of inert materials and impurities (XPU 44–164:2004), pathogen microorganisms as viable helminth eggs (XP X33-017:2004) and Salmonella (NF EN ISO 6579:2002), HAP trace organic compounds (XP X 33–012:2000) and trace elements (NF EN 13650:2002, NF EN ISO 17294–2:2016). The agronomic assessment of the two composts covers the determination of total solid, organic matter, organic carbon, total nitrogen C/N ratio, pH, density, phosphorus, potassium, calcium, magnesium, total Sulphur content according to standardized methods (NF EN 13040:2007, NF EN 13039:2011, NF EN 13037:2012, NF EN 13654–2:2002, NF EN ISO 17294–2:2016). The ISMO indicator corresponding to the percentage of stable organic matter related to the total organic matter content was calculated as follows:

$$\text{ISMO} = 44.5 + 0.5 \cdot \text{SOL} - 0.2 \cdot \text{CEL} + 0.7 \cdot \text{LIC} - 2.3 \cdot \text{MinC3} \quad (1)$$

with SOL, CEL and LIC being respectively assigned to the biochemical

soluble fraction, the cellulose fraction and the lignin fraction, all expressed in % of organic matter and determined using the Van Soest's fractioning method (Soest and Wine, 1967). MinC3 corresponds to the percentage of organic carbon mineralized after 3 days of incubation (XPU44-162: 2009).

2.4.2. Ecotoxicological analysis

2.4.2.1. Bioassay for assessment of emergence and growth of higher plants. A phytotoxicity test was carried out on both compost batches (NF U44-167:2016) to assess the risk of inhibiting the emergence and growth of two higher plants (barley, *Hordeum vulgare*, variety Etincel and watercress, *Lepidium sativum*, type Alénois). Three doses of compost were tested: 10 t/ha (Dose 1), 20 t/ha (Dose 2) and 100 t/ha (Dose 10) and compared to a control with no input. Four replicates were performed for each treatment. Bioassays took place in a regulated greenhouse, under controlled conditions of temperature, light, and humidity. Plant emergence was measured by counting the germinated seedlings of 20 seeds sown per pot. Measurements were carried out 48 h after at least 70 % of the seedlings in the control pots had emerged (i.e. at a maximum of 5 and 6 days for watercress and barley, respectively). Growth was studied on 10 seedlings per pot (supernumerary seedlings were discarded after the germination was scored). Growth was studied for a maximum of 20 days, at the end of which the plants were harvested, weighed (rm), dried in an oven at 75 °C and then weighed again (dm).

2.4.2.2. Determination of acute toxicity on earthworms. The acute toxicity test on earthworms (*Eisenia fetida*) was carried out in accordance with standard NF ISO 11268–1:2012 using an ISO soil (a mixture of Fontainebleau sand/kaolin/sphagnum with a dry matter ratio of 7/2/1) containing compost at 3 doses: 10, 20 and 100 t/ha, (i.e., 0.33, 0.66 and 3.33 wt% of compost per soil) and a control corresponding to ISO soil without compost addition. Earthworm mortality was determined on 10 individuals after 14 days of incubation at 20 ± 2 °C with a light/dark cycle of 16/8h (approximately 500 lx) without food intake. Each dose was tested with 4 replicates.

2.4.2.3. Determination of acute toxicity on microcrustaceans. The immobilization test of microcrustaceans was carried out on *Daphnia magna* (ISO 6341:2012) using an eluate of the two composts obtained with a liquid/solid mass ratio of 10 (calculated as dry matter equivalent) as described in the leaching protocol of standard EN 12457–2:2002. The test is based on determining the concentration that immobilizes 50 % of the *Daphnia* tested within 24 h and referred to as EC 50 %-24hr. The test was performed in two stages. A preliminary of the effective concentration of compost eluate causing an effect on 50 % of the population after 24 h (EC 50–24 h) was carried out to determine the concentration range for the definitive test. Then, the immobilization test was conducted by selecting a range of concentrations (usually in geometric progression) to

cover the concentration interval, within which in the preliminary test caused the immobilization percentage to rise from 0 to 100 %. Statistical testing was carried out by variance analysis with $P = 0.05$ using Fischer's test.

2.5. Compost sampling and plastic extraction

Representative samples were collected from the Materials and Control batch on days 18, 30, 45, 59, 107, and 128. For batches of compost up to 580 m³, as was the case here, the [NF EN 12579:2013](#) standard recommends taking 12 elementary samples, each weighing 1 kg, from various locations within the batch. The quartering method ([ASTM C702/C702M \(2018\)](#)) was used to reduce the sample size without creating a systematic bias due to product heterogeneity and obtain a representative final sample of around 10 kg per batch and per sampling time. Subsequently, all the compost samples were air-dried. They were then passed through a series of sieves with decreasing mesh sizes to obtain eight fractions ranging from 10 mm to 0.2 mm. Each fraction was weighed, and its water content was determined. For all fractions greater than or equal to 1 mm in size, all the fragments of compostable plastic were manually extracted using fine tweezers, categorized by size and color, and weighed.

2.6. Lab-scale testing of biodegradation of compostable plastic test materials

Respirometry biodegradation tests monitoring CO₂ release from the test materials were conducted in industrial composting conditions using a compost made of biowaste as inoculum (SCH, 34,800 Aspiran, France). For that, the virgin samples were cut in pieces of 5 x 5 mm. Exactly 50 mg of equivalent carbon were introduced into 3 g of compost previously sieved at 2 mm. A volume of 2.7 mL of deionized water was added to obtain a humidity of 50 wt%. The carbon content of all the materials was measured by elemental analysis (ThermoQuest NA 2500, CE Instruments Ltd, Wigan, UK).

The biodegradation tests were carried out in a miniaturized set-up as previously described ([Salomez et al., 2019](#)) using the standardized conditions described in [NF ISO 14855-1:2012](#) standard for composting in industrial environment. Experiments were carried out at 58 ± 1 °C in hermetically sealed glass jars (1 L, Le Parfait, Villeurbanne, France) containing three open polypropylene vials (60 mL each); one with 3 g of wet compost mixed with the sample, a second with 15 mL of NaOH (0.2 M), which traps the CO₂ released by the microorganisms, and a third one with distilled water to keep the relative humidity inside the jar close to 100 %. At regular time intervals, the glass jars were opened to determine the amount of CO₂ trapped by the NaOH solution by back titration of the carbonate ions. The latter were then precipitated by the addition of 5 mL of barium chloride solution (20 wt% in water), in each flask, in the presence of thymophthaleine (0.10 % in ethanol) and titrated by a HCl solution (0.1 M). The vials containing the compost were weighted and, if necessary, the appropriate amount of water was added to maintain 50 % of humidity. The flask containing the NaOH solution was taken out for titration and replaced by new vial containing a NaOH solution. The glass jars were left open for 2 min in order to be aerated. Then, each glass jar was closed and put back in the incubator. The biodegradation tests included a positive control and a blank. A powder of pure cellulose (BE 600–10 TG grade) provided by Arboce (Rosenberg, Germany), with a median apparent diameter (d₅₀) of 18 µm, was used as positive control. For the blank, the experiment was conducted without adding any sample, to be able to measure the CO₂ naturally produced by the compost and the CO₂ present in the air of the glass jar. All experiments were performed in triplicate. The CO₂ production of the blank was subtracted from the measurements. The percentage of biodegradation (%) was calculated using Eq. (2):

$$\text{Biodegradation} = \frac{CO_{2,\text{sample}} - CO_{2,\text{blank}}}{CO_{2,\text{theoretical}}} \cdot 100 \quad (2)$$

where CO_{2,sample} and CO_{2,blank} are the amounts of CO₂ (mg) released in the test jar and in the blank control jar, respectively. CO_{2,theoretical} is the theoretical amount of carbon dioxide (mg) produced by the total oxidation of the carbon added by the test material. As required by the [NF ISO 14855-1: 2012](#) standard, the validity of the test is obtained when the positive control reached a biodegradation percentage higher than 70 % in less than 45 days.

2.7. Physical-chemical characterization of sampled materials during composting

2.7.1. Macromolecular mass averages by size-exclusion chromatography

Size-exclusion chromatography (SEC) was performed using a Gilson pump (France) coupled to a Waters autosampler and refractometric index detector (Waters, France). The separation was carried out on a system consisting of a guard column (PLgel 5 µm) and three columns (two columns PLGel 5 µm MIXED-C and one column PLGel 3 µm MIXED E, Agilent Technologies) maintained at 40 °C in a column oven (Waters, France). The flow rate of the CHCl₃ was 1 mL/min. The calibration curve was established using three standard kits (EasiVials, 2 mL) containing each four narrow polystyrene standards of molecular weight between 4.69•10³ and 5.68•10⁶ g/mol (Agilent Technologies). Data treatment was done using Empower 3 software (Waters). Samples were prepared by dissolution of the uncleaned solids in dried CHCl₃ (app. 20 mg/mL) at room temperature or after heating at 40 °C without stirring at least 4 h. The supernatant was sampled after sedimentation of the insoluble phase and filtered before analysis with the help of 0.45 µm Teflon syringe filters. If enough sample available, the preparations were done in duplicate.

2.7.2. Thermal properties by differential scanning calorimetry

Thermal properties of compostable plastic materials were assessed by differential scanning calorimetry (DSC) (DSC1, Mettler Toledo, Switzerland) under N₂ (flow rate 50 mL/min) using at minimum 1 mg non cleaned solid in 40 µL aluminum pans. Calibration was carried out with Indium and Zinc standards. The samples were scanned with heat-cool-heat program between - 60 and + 180 °C at 10 °C/min heating or cooling rate. Melting enthalpy (ΔH_m) and glass transition temperature (T_g) shown here were analyzed at the first heating scan. If enough material was available, measurements were done in duplicate. Because of the unknown formulation of the samples, the crystallinity degree could not be determined.

2.7.3. Thermal stability by thermo-gravimetric analysis

The thermal stability of the materials was assessed by Thermo-Gravimetric Analysis (TGA) using a Mettler TGA2 apparatus (Mettler Toledo, USA). Thermal analyses were performed from 25 °C up to 800 °C at 5 °C/min under nitrogen flow (20 mL/min). An average weight of 10 mg was used each time, if possible, with regards to the available quantities, measurements were duplicated.

2.7.4. Surface analysis by scanning electron microscopy

The compostable materials surfaces were observed by scanning electron microscopy (SEM). SEM observations were performed using a Desktop SEM (Phenom ProX, Fondis Bioritech, France) with an acceleration voltage ranging between 5 and 10 kV secondary electrons. Samples were directly mounted on stub using carbon conductive tape and then coated with Gold/Palladium during 45 s at 20 mA (4 nm thick) by ion sputtering (Mini Sputter Coater SC7620, Quorum Technologies Ltd, UK).

2.8. Life cycle assessment of the composting process

2.8.1. Goals, scope and context of the life cycle assessment study

The life-cycle assessment (LCA) methodology was used to assess and compare the environmental performance of biodegradable packaging in two end-of-life processing treatments: industrial composting and industrial incineration. It was conducted according to the guidelines ISO 14040:2006 which comprises four stages that will be developed below: i) goal and scope definition, ii) life cycle inventory, iii) life cycle impact and assessment and iv) interpretation and analysis. The objective was to compare the environmental impact of the end-of-life treatment by composting to incineration, which is the main actual treatment of packaging waste in France. Therefore, the system boundary is defined as gate-to-factory-gate (e-supplementary information) taking into account the initial point as the collection of the household waste, the composting process for the production of soil fertilizers and amendments, and industrial incineration for the production of electricity.

2.8.2. Functional unit

The functional unit (FU) chosen was 1 t of packaging waste in France formulated with 70 wt% of biobased origin and 30 wt% fossil origin. This value corresponds to the average quantity of fossil material present in the test materials which were not all 100 % biobased. The packaging waste collection is supposed to be door-to-door for both cases.

2.8.3. Life cycle inventory assessment

The conceptual modelling of the industrial composting scenario (e-supplementary information) was conducted mostly with real processing data collected by the authors from an industrial scale composting process thanks to the collaboration with the enterprise “Syndicat Centre Hérault”, completed with data found in the literature for the ammonia and carbon monoxide emissions (Al-Rumaihi et al., 2020). The modelling of the industrial incineration scenario (e-supplementary information), was carried out with data from the literature, which is very detailed on this subject (Beylot et al., 2018; Chin et al., 2023).

2.8.4. Life cycle impact assessment

The life cycle impact assessment was done with the software OpenLCA v2 and the Ecoinvent® 3.0.1 database. The methodology used was CML-baseline and for the environmental performance profile, the categories assessed were: abiotic depletion potential (ADP), acidification potential (AP), eutrophication potential (EP), global warming potential at 100 years (GWP₁₀₀), human toxicity potential (HTP), ozone layer depletion potential (OLDP), photochemical oxidation potential (POP), and terrestrial ecotoxicity potential (TEP). Both the methodology and categories were selected in accordance with the most representative in biowaste treatment studies.

3. Results

3.1. Monitoring of composting process and compost safety and quality

3.1.1. Impact of compostable materials on composting process

The parameters of the composting piles were monitored weekly before turning the pile for aeration. The target humidity of 50 % was maintained by adding leachate water to the piles when necessary. The average temperature during the active composting phase was 69.7 °C ± 2.5 and 69.7 °C ± 3.6 in the Control and Materials batches, respectively (e-supplementary information). These conditions are adequate to eliminate pathogens in the compost pile, indicating the successful hygienization treatment of both batches. The compost maturity index was measured at weekly intervals from the 30th day to pilot the composting process. After 45 days, the self-heating test carried out on-site on both composts indicated a temperature below 40 °C marking the end of the active phase. Starting with that time, the frequency of turning and watering has been reduced, resulting in a drop in temperature for both

batches throughout the 12-week maturation phase. No significant differences in temperature or duration of the thermophilic and maturation phases were observed between the two batches (e-supplementary information).

The two batches were screened after the maturation phase to obtain three granulometric fractions, where only the fine fraction (<12 mm) constitutes a marketable product. The medium and coarse fractions were found to be too contaminated by conventional plastics for recycling as structuring agent in a new composting process, and as a result, they were sent to a landfill. The total mass of the compost after 128 days was 9.78 t and 10.11 t for the Control and the Materials batches, respectively. After screening, 5.46 and 6.01 t of marketable compost were obtained, corresponding to a composting yield of 27 % for the control batch, and 29 % for the materials batch. Within the limits of measurement uncertainty, no significant difference was observed between the two batches.

3.1.2. Impact of compostable materials on compost safety

Commercial composts are required to comply with the specific quality and safety criteria, as outlined in the NF U44-051:2006 standard. Stricter limits are imposed for organic farming, as specified in the I3024 guidelines for “fertilizing materials and growing media usable in organic agriculture.” The assessment of compost safety is detailed in Table 2. Metal and glass debris remained well below established limit values. No pathogenic microorganisms were detected. Quantities of heavy metals were consistently measured at levels significantly lower (about 10 times) than the detection limit. Organic pollutants were mostly non-measurable, except for a single instance of fluoranthene, reaching the detection limit of the applied methodology. The presence of inert plastics was generally negligible due to their manual elimination during the bio waste preparation at the beginning of the test. In summary, the resulting compost met the NF U44-051:2006 standard requirements and could be labeled as suitable for organic farming according to the I3024 reference document. Notably, the quantity of inert plastics in the materials batch was smaller than in the control batch. The overall number of inert materials was significantly lower than observed in comparable composting experiments (e.g., up to 3.4 %, as measured by Rodrigues et al., 2020). This reduction was attributed to the effective cleaning of biowaste on their arrival, with approximately 1.4 wt% of improper materials being discarded at that stage.

The results of the ecotoxicity tests are detailed in Table 3. The phytotoxicity test, evaluating the potential inhibition of emergence and growth in two higher plants, was conducted on both compost batches at three different doses following FDU 44-167. The results revealed no significant differences in germination rates for watercress and barley across different conditions, irrespective of the batch (Control or Materials). The Materials batch at dose 1 exhibited a statistically significant increase in biomass production for watercress compared to other conditions (Table 3). Similarly, barley showed significantly higher biomass production at dose 2 for the Materials batch and at dose 10 for both batches compared to other conditions, including the control (Table 3). The literature on phytotoxicity tests of composts produced with compostable plastics is scarce. One only study reported negative impacts (Greene, 2007), but in this case, the methodology applied was not suitable for assessing the ecotoxicity of a compost used as soil improver, given the very high concentrations of compost used (50 % vs. 0.33–3.33 % here), which could explain the negative results obtained. In line with the present experiments, those of studies using similar methodology (Klauss & Bidlingmaier, 2004; Kawashima et al., 2021), did not observe adverse effects of composts obtained from compostable materials on plant growth. Regarding the search for acute toxicity on terrestrial organisms, results in Table 3 demonstrated the absence of acute toxicity characteristics on earthworms (*Eisenia fetida*). Neither batch caused significant earthworm mortality in after 14 days, irrespective of the compost doses. Similar conclusions were drawn from the immobilization test conducted on microcrustaceans using a concentrated eluate of both composts. This eluate had a liquid/solid mass ratio of 10, which is three

Table 2
Safety assessment of the control and materials batches according to criteria of NFU 44–051 standard and organic agriculture.

	Control batch	Materials batch	NF U44-051 Criteria ¹	Organic Agriculture Criteria
Inerts and impurities (%) ²				
$d < 1.1 < d < 1.35$	Film and PSE > 5 mm	0.00	0.00	< 0.3
	Other plastics > 5 mm	0.03	0.00	< 0.8
	Total synthetic 2–5 mm	0.00	0.00	
	Total synthetic > 2 mm	0.03	0.00	
$d > 1.35$	Glass and Metal > 5 mm	0.05	0.02	
	Glass and Metal 2–5 mm	0.04	0.09	< 2
	Stone and calcareous > 5 mm	9.3	8.7	
	Stone and calcareous 2–5 mm	5.3	5.3	
	Fines < 2 mm	9.7	9.3	
	Total inerts and impurities	24.4	23.4	
Pathogen microorganisms				
	Viable Helminth eggs ³	absence	absence	absence
	Salmonella spp ⁴	absence	absence	absence
Trace organic compound (mg/kg db) ⁵				
	Fluoranthene (HAP)	< 0.05	0.06	< 4
	Benzo(b)fluoranthene (HAP)	< 0.05	< 0.05	< 2.5
	Benzo(a)pyrene (HAP)	< 0.05	< 0.05	< 1.5
Trace elements and metal trace elements (mg/kg db) ⁶				
	As	5.19	5.20	< 18
	B	39.4	42.3	
	Cd	0.37	0.36	< 3
	Cr	24.9	22.1	< 120
	Cu	53.8	53.3	< 300
	Hg	< 0.2	< 0.2	< 2
	Fe	8117	8331	
	Ni	15.8	15.8	< 60
	Pb	19.2	21.4	< 180
	Se	<1.5	<1.5	< 12
	Mn	230	244	
	Zn	122	129	< 600
	Mo	<2	<2	< 200

¹ Regulatory safety criteria required by NFU 44–051 – 2006 – (Organic soil improvers — Designations, specifications and marking.).

² Values expressed related to dry matter content according to XP U 44–164:2004 (Amendements organiques et supports de culture - Méthode d'analyse des composants inertes dans un compost).

³ Value expressed for 1.5 g of raw matter according to XP X33-017:2004, Caractérisation des boues-Dénombrement et viabilité des oeufs d'helminthes parasites - Méthode par une technique de triple flottation.

⁴ Value expressed for 25 g of raw matter according to NF EN ISO 6579:2002, Microbiologie des aliments - Méthode horizontale pour la recherche des Salmonella spp.

⁵ Value expressed related to dry matter according to XP X 33–012:2000 - Caractérisation des boues - Dosage des hydrocarbures aromatiques polycycliques (HAP) et des polychlorobiphényles (PCB).

⁶ Value expressed related to dry matter according to NF EN 13650:2002, Amendements du sol et supports de culture — Extraction d'éléments solubles dans l'eau régale NF EN 16174 and NF EN ISO 17294 (ICP MS) excepted for Hg which is dosed by atomic spectrometry of cold vapor.

times higher than the dose 10 used in ecotoxicity tests on higher plants and earthworms. The concentration causing immobilization of 50 % of the *Daphnia* tested in 24 h was not significantly different between both batches.

3.1.3. Impact of compostable plastic materials on agronomic compost quality

Table 4 summarizes the parameters related to the agronomic quality of the composts. The agronomic effectiveness of an organic amendment relies on both its fertilizing value (N, P, K, Ca, Mg) and its amending value, which is linked to the stability of organic matter. The amending value determines the behavior of the compost once it has been incorporated into the soil, based on the resistance of its organic matter to biodegradation. Minimum or maximum requirements for these parameters are stipulated in NF U44-51:2006.

Composts obtained from both the Control and Materials batch met all

the requirements, showing the high agronomic quality of the resulting product. Table 4 compares furthermore the obtained values to those generally observed by the composting platform. No significant difference in quality of the Control and Materials batch with respect to usual data was observed. The compost fertilizing parameters (N, P, K, Ca, Mg) and its amending value (C/N, organic matter, ISMO) in Table 4 were moreover compared to those obtained by Rodrigues et al., 2020. These authors collected data on the fertilizing value of compost made out of source-separated biowaste from 20 plants with annual capacities ranging from > 18,500 to < 4875 t in Catalonia (Spain). Mean fertilizing values were C/N = 9.9, P = 0.99 %, K = 1.6 %, Ca = 8.8 %, and Mg = 0.7 %. The measured characteristics of the Control and Materials compost aligned with average values of this dataset. In summary, no significant impact of including plastics in the compost was observed, neither concerning the characteristics of the compost from the industrial platform, or with respect to the literature data.

Table 3
Ecotoxicity assessment of the control and materials batch.

Phytotoxicity ¹ Emerging seedlings at day 5–6 (%)	Watercress		Barley	
	Control batch	Materials batch	Control batch	Materials batch
Dose 0 = Control	85.00 ± 5.77 ^a	85.00 ± 5.77 ^a	80.00 ± 13.54 ^a	80.00 ± 13.54 ^a
Dose 1 = 0.33 % (10 t/ha)	75.00 ± 13.54 ^a	73.75 ± 6.29 ^a	82.50 ± 15.00 ^a	86.25 ± 4.79 ^a
Dose 2 = 0.66 % (20 t/ha)	75.00 ± 12.25 ^a	85.00 ± 7.07 ^a	87.50 ± 12.58 ^a	81.25 ± 7.50 ^a
Dose 10 = 3.33 % (100 t/ha)	85.00 ± 10.80 ^a	76.25 ± 4.79 ^a	78.75 ± 13.15 ^a	82.50 ± 6.45 ^a
Growth of aerial parts at day 20 (g dry biomass/plant)				
Dose 0 = Control	0.06 ± 0.02 ^a	0.06 ± 0.02 ^a	0.34 ± 0.04 ^a	0.34 ± 0.04 ^a
Dose 1 = 0.33 % (10 t/ha)	0.06 ± 0.01 ^a	0.13 ± 0.04 ^b	0.34 ± 0.04 ^a	0.32 ± 0.02 ^b
Dose 2 = 0.66 % (20 t/ha)	0.07 ± 0.02 ^a	0.09 ± 0.01 ^a	0.33 ± 0.03 ^a	0.38 ± 0.03 ^b
Dose 10 = 3.33 % (100 t/ha)	0.08 ± 0.02 ^a	0.09 ± 0.02 ^a	0.41 ± 0.02 ^{b,c}	0.44 ± 0.02 ^c
Acute toxicity test on earthworm (<i>Eisenia fetida</i>) ²				
	Earthworm surviving (n/10)		Mortality after 14 days (%)	
	Control batch	Materials batch	Control batch	Materials batch
Dose 0 = Control	9.50 ± 0.58 ^a	10.00 ± 0.00 ^a	5.0	0.0
Dose 1 = 0.33 % (10 t/ha)	9.75 ± 0.50 ^a	9.67 ± 0.58 ^a	2.5	2.5
Dose 2 = 0.66 % (20 t/ha)	9.25 ± 0.50 ^a	10.00 ± 0.00 ^a	7.5	0.0
Dose 10 = 3.33 % (100 t/ha)	9.50 ± 0.58 ^a	9.67 ± 0.58 ^a	5.0	2.5
Acute toxicity on microcrustaceans ^{3,4}				
	Inhibition of the mobility of <i>Daphnia magna</i>			
	Control batch	Materials batch		
EC 50 % – 24 hr (%) ⁵	53.6 ± 6.4 ^a		53.9 ± 3.8 ^a	

All subscripts are determined by variance analysis with $p = 0.05$ (LSD Fisher test) for each data set in the column.

¹ according to FD U44-167 (2016) – test launched on 20 seeds per bloc with 4 repetitions per modality - requirement for the test validity: >70 % emerged seedling at Dose 0.

² according to NF ISO 11268-1 (2012) – test launched on 10 earthworms with 4 repetitions per modality - requirement for the test validity: <10 % mortality at Dose 0.

³ Tests carried out on an eluate of the two composts using a liquid/solid mass ratio of 10 (calculated as dry matter equivalent) according to the leaching protocol EN 12457-2 (2002).

⁴ according to ISO 6341 (2012) - requirement for the test validity: percentage of immobilization ≤ 10 % after 48hr in control container.

⁵ EC 50 %-24hr: Effective concentration causing an effect on 50 % of the population after 24 h (determined by Log-Probit statistical model, Toxcalc software).

Table 4

Agronomic assessment of the control and materials batches according to criteria of NFU44-051 standard and organic farming, and comparison with results of other characterizations of compost made of biowaste.

Agronomic value	Control batch	Materials batch	NF U44-051 Criteria	OF Criteria	Usual compost parameters on the composting site (n = 32) ¹¹		
					Mean ± SD	min	max
Solid content ¹ (%)	67.0	57.6	≥ 30	≥ 30			
Compact density ¹ (Kg/l)	0.442	0.497					
Organic matter ² (%)	49.6	47.9	≥ 20	≥ 20	50.8 ± 4.4	38.4	58.0
Organic carbon ³ (%)	27.8	28.2			26.6 ± 3.1	15.4	34.4
C/N ⁴	10	11	> 8	> 8	12.1 ± 1.5	9.8	15.0
pH ⁵	8.4	8.3					
Total Nitrogen ^{6,7} (N) (%)	2.55	2.37	< 3	< 3	2.0 ± 0.2	1.6	2.4
Phosphorus ^{7,8} (P ₂ O ₅) (%)	1.13	1.10	< 3	< 3	0.93 ± 0.09	0.7	1.1
Potassium ^{7,8} (K ₂ O) (%)	2.02	2.06	< 3	< 3	1.6 ± 0.2	1.0	2.1
Calcium ⁸ (CaO) (%)	12.30	11.90			10.9 ± 1.3	6.7	14.3
Magnesium ⁸ (MgO) (%)	1.30	1.44			1.3 ± 0.1	0.9	1.6
Total Sulphur ⁸ (SO ₃) (%)	0.77	0.67					
Humigenous potential ⁹ (%)	73.0	76.8			76.62 ± 5.7	68.2	83.5
Soluble fraction (% MO)	29.0	35.2					
Cellulose fraction (% MO)	8.6	21.0					
Lignin fraction (%MO)	45.1	44.4					
3-day mineralization (%) ¹⁰	6.0	5.3			3.88 ± 1.8	1.8	6.7
Stable MO (kg/t db)	362	368			396.8 ± 53.7	311	443

¹Value expressed related to raw matter, according to NF EN 13040:2007. ²Value obtained by incineration related to dry matter content according to NF EN 13039:2011. ³Value expressed related to dry matter content according to NF ISO 14235:1998. ⁴ Value deduced by calculation (C organic/N Total). ⁵ Determined according to NF EN 13037:2012. ⁶Value expressed related to dry matter content according to NF EN 13654-2:2002. ⁷Soil improvers should respect $N + P_2O_5 + K_2O < 7\%$ related to raw matter content. ⁸Value expressed related to dry matter content according to NF EN ISO 17294-2:2016- Part 2. ⁹Humigenous potential also called ISMO represents the percentage of stable organic matter in relation to the total organic matter content according to XP U44-162: 2009. ¹⁰Percentage of organic carbon mineralized after 3-days incubation according to XPU44-162. ¹¹Characteristics of compost made of biowaste between 2017 and 2023 in the plant selected for the study.

The Organic Matter Stability Index (ISMO) (as per XPU 44-162 standard) represents the percentage of stable organic matter in relation to the total organic matter content, providing an indication of the amount of organic matter contributing to maintaining soil organic matter stocks or needed to reconstitute soil humus stocks. Higher ISMO values indicate greater organic matter stability in the soil (Lashermes et al., 2009). Values can range between 20 to almost 100 %. Composts

originating from biowaste generally have ISMO values in the range of 75 % to 85 % (Lashermes et al., 2009). The composts obtained in this full-scale experiment had high ISMO values, in the same range as those made from biowaste on the composting site (Table 4). These values indicate the high agronomic quality of the compost, independently of the inclusion of compostable plastics, as evidenced by their bio-assimilation during the composting process.

3.2. Monitoring of materials degradation during composting

3.2.1. Biodegradability and mass loss in composting conditions

In the full-scale composting experiment, the degradation of the compostable plastics was monitored by measuring their mass loss over time, although this alone does not provide definitive evidence for biodegradation. As the compost pile is an open environment, it is not possible to measure the CO₂ released throughout the composting process. To confirm ultimate biodegradation, laboratory-scale respirometry tests were conducted using an inoculum of the Aspiran composting platform in accordance with EN 13432:2000 guidelines on all materials. It should be noted that the standardized test temperature at the laboratory scale (58 °C ± 1) is lower than that encountered in the industrial composting facility (around 70 °C), making it a conservative estimate of actual composting conditions. The mineralization curves of the test materials are shown in the e-supplementary information. The comparison of the half-times ($t_{1/2}$) of biodegradation showed that all flexible compostable materials degraded faster than cellulose ($t_{1/2} = 33 \pm 7$ d). The half-times of the starch/polyester blends increased with the thickness of the material (E1: 5 ± 1 d, E3: 9 ± 1 d, E4: 10 ± 1 d), the half-time of the polyester material was longer (E5: 12 ± 1 d). The time to reach the ultimate biodegradation of all the flexible films was smaller compared to the cellulose control material, surpassing the 90 % limit of ultimate biodegradation in 85 days, well ahead of the requirements of EN 13432:2000 (6 months) (e-supplementary information). The colored tracer material of starch/polyester blends (2.5 times thicker than the others; 50 vs. 20 μ m) demonstrated a time to ultimate biodegradation comparable (within the experimental uncertainty) to those of the other materials. All materials were cut into small pieces for the respirometry test. This simulated the disintegration phase and minimized the impact of the sample geometry on the biodegradation kinetics. This could be the reason why no significant differences were observed in the time to reach 90 % ultimate biodegradation. The time to reach the ultimate biodegradation of rigid materials was even faster than the one of flexible films. All rigid materials reached it before 60 days. Precisely, the $t_{1/2}$ of cellulose of the assay was 20 ± 1 d. The $t_{1/2}$ of E8 and E9 were 10 ± 1 d. The PLA references had very different kinetics. E6 showed 5 ± 1 d. E7 displayed a longer lag-time ($t_{1/2} = 22 \pm 1$ d), but degraded fast, once the process was started. It reached the plateau of 100 % biodegradation before 60 days, as all other rigid materials. In conclusion, the lab-scale test evidenced the ultimate biodegradability of all tested materials under industrial composting conditions (e-supplementary information).

The degradation of biodegradable plastics was tracked through regular sampling during compost pile turn over, and their mass loss was analyzed by measuring the remaining material passed through a 1 mm sieve. The mass loss curve of the colored materials, which were easily identifiable, showed a sharp increase right from the start of the composting process (Fig. 1a). This behavior was consistent with the biodegradation curves obtained by the laboratory-scale respirometry tests (e-supplementary information).

At the end of the active thermophilic phase (49 days), approximately 85 wt% of colored plastics had disintegrated, and nearly complete biodegradation was measured (98 wt%) at the end of the maturation phase (128 days). Differences were observed among families of materials, with a recovery of less than 1 wt% for starch/polyester blends and PLA-based materials. On the other hand, approximately 32 wt% of the PHA coffee capsules remained in the final compost, which was unexpected given that PHAs are known to be easily biodegradable polymers, as confirmed by numerous studies (Briassoulis et al., 2021). It is noteworthy that these capsules were new and empty. The observed persistence of these materials suggests that the high thickness (3 mm) and narrow shape of the coffee capsule hindered efficient disintegration. This highlights the importance of taking geometrical aspects into account in biodegradation studies, which are often not fully predicted by lab-scale disintegration and biodegradation tests. Transparent plastics were tedious to recover because the smaller fragments were hardly

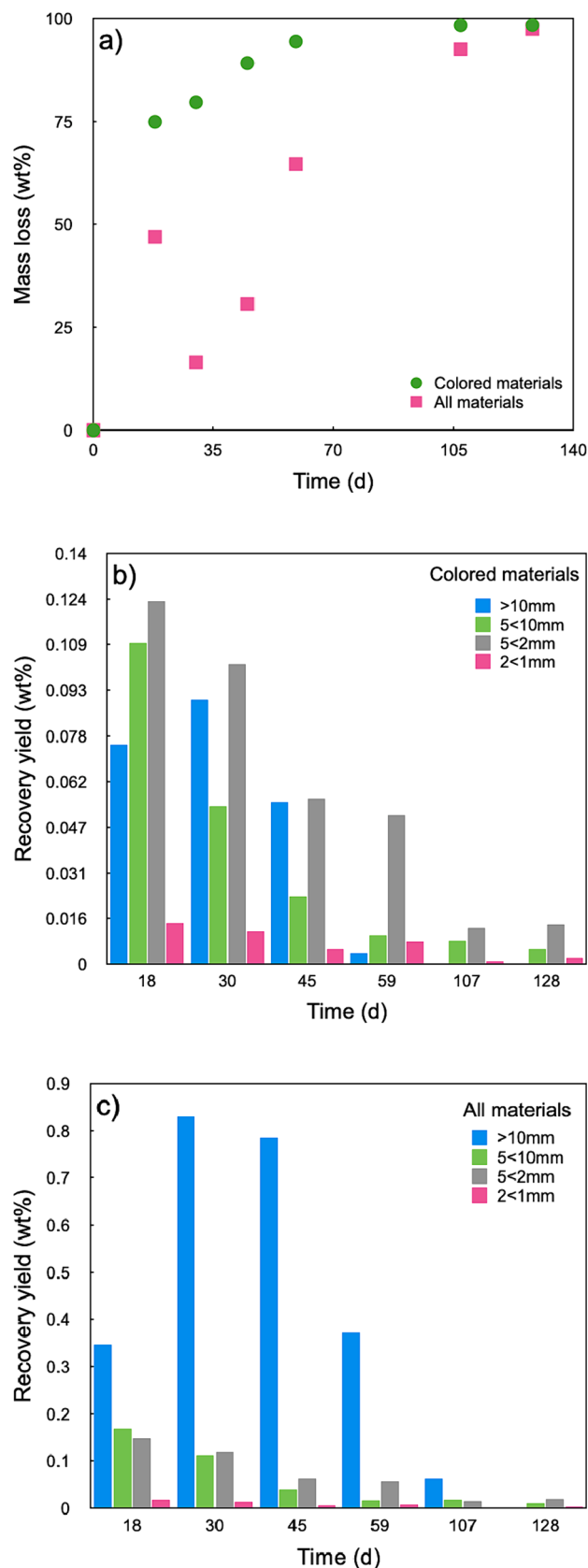


Fig. 1. Quantification of mass loss and recovered fragments during the composting process: a) mass loss of plastics calculated from the sum of the recovered materials, b) granulometric distribution of colored fragments at different sampling times, c) granulometric distribution of all recovered fragments at different sampling times.

visible or could be confused with other particles because transparent and white materials became brown over time. The mass loss curve was noisier and seemingly slower than that of colored plastics (Fig. 1a). Conventional plastics are often transparent bags or trays and may be present in the biowaste due to sorting errors that were not identified during the initial clean-up of the waste pile. Because of these two issues, the evaluation of the mass loss of the transparent and white fragments had higher uncertainty in comparison to that of colored particles.

In order to evaluate the disintegration kinetics and the possible presence of sorting errors, the granulometric distribution of the recovered fragments was analyzed. It revealed rapid disintegration of colored materials into small particles (Fig. 1b). Most of particles were smaller than 10 mm after 18 days, shifting to reduced size over time. The particles recovered after 59 days were all smaller than 5 mm. In the case of the transparent particles, large fragments persisted throughout the process (Fig. 1c). It should be noted that the analysis at 128 days was carried out on the compost, sieved with 12 mm mesh size, which explains the absence of large particles. To evaluate the risk of confusion of compostable plastics with non-compostable impurities, the fragments which were larger than 12 mm were manually recovered from the sieves for identification. Fourier-Transformation Infra-Red spectroscopy (FT-IR) was used to identify the particles. The transparent fragments fraction contained in addition to the compostable materials polyethylene terephthalate, nylon, and polyethylene (e-supplementary information). The presence of contamination would also be the reason why the percentage of inert plastics was higher in the Control batch than in the Materials batch (Table 2). The evolution of the granulometric distribution of the smaller fractions of the transparent and white materials was consistent with that of the colored ones. After the final screening, roughly 2 wt% of transparent films was recovered, but this included the non-compostable plastic that constituted contamination.

3.2.2. Morphological changes of compostable plastics during composting

The structural and morphological changes of the colored polymers were analyzed to trace the degradation of the fragments collected in the compost. SEM surface images of the fragments sampled during the composting process are presented in the e-supplementary information. Over time, the surfaces became rough and cracked. Microbial attachment was observed on all surfaces, with the presence of both bacteria and fungi. The surfaces of the starch blends displayed increasing cracks but retained a relatively smooth overall surface over time, suggesting a process of surface erosion. PHA fragments exhibited relatively smooth surfaces with only a few cracks, evolving into holes and craters with longer composting times, indicating local surface degradation. PLA showed deep cracks evolving into deep fragmentation, with microbial colonization observed inside the large fractures. This material rapidly disintegrated into small fragments.

All data on the evolution of the structural and morphological characteristics of the materials are gathered in Fig. 2. The indicators of polymer degradation are the decrease in macromolecular chain length (Fig. 2a,b), and the thermal stability (Fig. 2c), resulting from the shortening of the macromolecules. Fig. 2c shows the evolution of the degradation temperature of the colored samples as a function of composting time. Due to the limited quantity of fragments available, repeated experiments were only carried out on the initial materials. It is interesting to note that the degradation temperatures of the starch/polyester blend only decreased by 3 °C, possibly due to the surface erosion mechanism that primarily affects the outer layers without significantly altering the material's bulk properties. Similarly, the thermal stability of PHA decreased by 15 °C, which is consistent with the surface erosion process shown by the SEM images (e-supplementary information) and already documented in the literature (Salomez et al., 2019; Weng et al., 2011). Thus, the thickness (2 mm) of PHA particles might have preserved intact the core of the material. In contrast, the thermal stability of PLA decreased significantly, dropping by 74 °C. The extensive degradation observed is supported by SEM images (e-

supplementary information) revealing deep and wide surface cracks. PLA generally requires chemical hydrolysis prior to biodegradation of hydrolysis products (Kale et al., 2007b). This hydrolysis can be catalyzed by the acidity of the small oligomers or the generated lactic acid, leading to accelerated degradation within the material due to the local accumulation of acid molecules (Gerometta et al., 2019; Musiol et al., 2016b). The formation of deep cracks (e-supplementary information) facilitating efficient disintegration and the substantial decrease in thermal degradation temperature (Fig. 2c) align with this mechanism.

The changes in macromolecular chain length were also analyzed. The decrease in the average macromolecular weight (M_w) of the polymer chains over time is depicted in Fig. 2a,b. This indicates chemical or biochemical degradation of the polymer (Kale et al., 2006; Kale et al., 2007a; Musiol et al., 2016b). The M_w decreased in fragments found on the surface of the pile (Fig. 2a), indicating degradation, even though the location outside the composting pile is not an ideal situation. This is a consequence of the choice to mix the compostable plastics directly with biowaste to mimic a real situation. During the turning operations, some specimens are ejected from the pile, while others remain inside, resulting in different individual process histories. SEC analysis proved to be very material-intensive, given the very small quantities of fragments recovered for each granulometric fraction. In addition, for the fragments recovered after 60 days, no signal was obtained due to the degradation of a significant part of the sample below the 3000 Da, which was the lower limit of detection. Within the sample set, no correlation between M_w and fragment size was observed. Consequently, all the data are presented on a single graph without distinction of the granulometric fraction Fig. 2a,b. Not surprisingly, the decrease in M_w of the fragments recovered from inside the pile was faster than that of the specimens taken from outside (Fig. 2b). PLA degraded faster than the polyester fraction of starch/polyester blends or even PHA. Note that the starch component of the starch polyester/blend could not be analyzed, as starch is insoluble in CHCl_3 . Results are consistent with the surface erosion mechanism of the biodegradation of starch/polyester blends and PHA, which allows the material at the core of the fragment to remain relatively intact. PLA, on the other hand, undergoes bulk degradation and most of the macromolecules are therefore cut.

DSC analysis of the fragments required less quantity than SEC analysis, allowing the few fragments recovered to be analyzed as a function of time and granulometric fraction (Fig. 2d-h). The melting enthalpy of PHA showed no significant change over the first 59 days, independent of the granulometric fraction (Fig. 2d). The signal of the glass transition was below the detection limit. After 128 days, the amount of crystallites present decreased slightly, although this was not significant for all size classes. This is again consistent with the surface erosion degradation process (Weng et al., 2010), where crystalline and amorphous phases degrade with similar kinetics (Salomez et al., 2019). Fig. 2e,f illustrates the alterations in the melting enthalpy (Fig. 2e) and T_g (Fig. 2f) of the colored starch-polyester blend. The data in Fig. 2f specifically represents the polyester part no distinct signal for the starch glass transition was found. Generally, the starch fraction in these blends degrades faster than the polyester component (Bulatovic et al., 2019). No significant change in the polyester fraction was recorded, and no correlation with particle size was observed. The minimal morphological change suggests a surface erosion process, like the behavior of PHA. Fig. 2g,h illustrates the morphological changes in PLA during composting. Unlike the other materials, PLA was initially amorphous and quickly crystallized during the composting process (Fig. 2g). PLA disintegrated faster than PHBV and starch-polyester blends. No fragments larger than 10 mm were recovered after 18 days, and none larger than 5 mm after 59 days. Crystallization of PLA during composting is well-documented (Kale et al., 2007a; Kale et al., 2007b; Musiol et al., 2016a; Musiol et al., 2016b). This phenomenon may be due to the fact the composting temperature exceeds T_g , which allows cold crystallization, or to the selective degradation of amorphous phases compared to crystalline phases. The interplay between PLA morphology and hydrolysis has recently been

discussed in detail (Limsukon et al., 2023). Selective degradation of the amorphous phase can lead to an increase in surface roughness and fragmentation into small particles (Gaillard et al., 2019). Fig. 2h shows the evolution of T_g over time during composting. The dispersion of T_g values observed during the first sampling could not be explored further due to the limited quantity of material recovered. After 18 days, the T_g decreased. This may be due to plasticization, as PLA can be plasticized by its oligomers and lactic acid (Martin & Averous, 2001), which are hydrolysis products generated during its abiotic degradation. After leaching out of the plasticizers, the longer-term decrease in T_g could be associated with the reduction in macromolecular chain length (Saeidlou et al., 2012). In conclusion, the detailed examination of the structural changes occurring in colored polymers during composting demonstrated the degradation of all polymers and pointed to the different degradation dynamics of compostable plastics.

3.3. Environmental assessment

Environmental performance was assessed for two end-of-life treatments, industrial composting and industrial incineration. The biogenic carbon was treated neutral, as recommended by Van Roijen & Miller, 2022. The relative contribution to each impact category of both processes is shown in Fig. 3, the numerical data are presented in e-supplementary information. In seven out of eight impact categories composting performed better than incineration (ADP, AP, EP, GWP, HTP, OLDP, TEP). Nevertheless, in five impact categories (ADP, AP, EP, GWP, OLDP), composting presents relatively significant impacts.

Incineration exhibited a higher relative impact in global warming potential, contributing to 65 % of the total compared to composting which contributed by 35 %, although the 30 wt% of fossil carbon in the test materials were accounted for. This disparity stems from the straightforward conversion of organic compounds into CO₂ in incineration, leading to a more efficient yield. In contrast, composting involves complex metabolic pathways within microbial consortia, resulting in a less direct and complex transformation of organic compounds and the release also of other greenhouse gas emissions, such as methane and carbon monoxide. The electricity use of both processes generated the main impact in abiotic depletion potential, eutrophication potential and

ozone layer depletion potential. Their relative impacts were higher for incineration when compared to composting. Electricity consumption in incineration facilities is around 100 to 130 kWh per tonne of treated waste, whereas industrial composting facilities only require around 40 kWh per tonne of treated waste (mainly for the pumping of leachate). Analysing abiotic depletion potential, composting accounted for 42 % of the relative contribution, while incineration accounted for 58 %. In both cases, impacts were attributed to electricity consumption and the utilization of elements like uranium and chromium, linked to the prevalence of nuclear energy in the French energy mix. Furthermore, incineration presented a higher eutrophication potential at 59 %, while composting accounted for 41 %. This discrepancy is tied to potential emissions of nitrogen oxides (NO_x) and phosphates and nitrates into the air, water, or groundwater. The emissions, primarily phosphates and NO_x, are associated with fossil fuel usage. Incineration, with its high consumption during combustion initiation, contributed more significantly to this impact category compared to composting. Concerning ozone layer depletion, incineration showed 58 % of the relative impact in this category against 42 % of composting. Both were directly related to the emissions to the air generated during electricity production. In terms of human toxicity potential, the impacts were predominantly driven by metal emissions during electricity production, including Chromium VI, Vanadium, and Selenium. Notably, incineration introduced additional emissions of these compounds during combustion, resulting in a higher relative impact of 75 %, compared to 25 % for composting. Acidification and terrestrial ecotoxicity potential of the incineration process accounted for 62 and 98 % respectively and stemmed from the use of fossil fuel and its associated sulphur oxides emissions. The photochemical oxidation potential was the one impact category, where the composting process performed less compared to incineration, accounting for 74 % of relative impact. The impact comes primarily from the fossil fuel utilization. In addition to that carbon monoxide emissions are generated during the processes. Composting generated higher quantities of CO (0.076 kg/t of waste) than incineration (0.013 kg/t of waste).

A rough assessment of avoided impacts was done considering electricity production for incineration and replacement of inorganic fertilizers for composting. The results are presented in the e-supplementary information. It showed that each process brought some benefit in different impact categories. In seven out of eight impacts, the estimated benefit of replacing inorganic fertilizers by compost was higher as compared to generating energy from waste by incineration. The most significant benefits of using compost would be by reduction in terrestrial ecotoxicity and abiotic depletion. The most significant one for incineration was ozone layer depletion. Calculation of the total environmental impact of the processes, which equals the generated impact minus the avoided one, showed that in all categories, except ozone layer depletion, the composting process was less impacting. The numerical values are given in the e-supplementary information. The results underscore the high interest of comprehensive assessment of environmental impacts, as forces and weaknesses of different processes are not always observable in the same impact category. Independently of the overall environmental performance of one process compared to the other, every category of environmental impact holds individual significance as it reflects different facets of its ecological footprint (Bishop et al., 2021). Considering diverse processes in waste management allows the system to benefit from different environmental gains, preventing the over-emphasis of certain impacts and allowing sustainable development under a balanced and flexible framework that considers the broader environmental implications of both processes, composting and incineration.

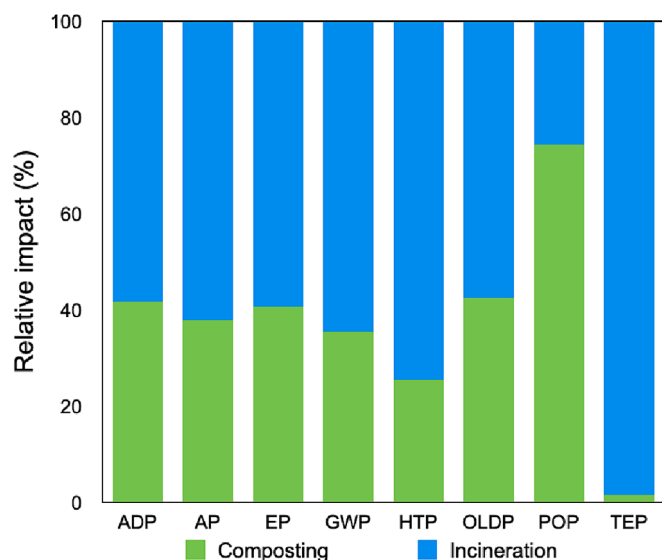


Fig. 3. Relative environmental profile for industrial-scale composting and incineration of packaging mixed with biowaste for the categories: abiotic depletion potential (ADP), acidification potential (AP), eutrophication potential (EP), global warming potential at 100 years (GWP₁₀₀), human toxicity potential (HTP), ozone layer depletion potential (OLDP), photochemical oxidation potential (POP), and terrestrial ecotoxicity potential (TEP).

4. Conclusion

This large-scale composting study evidenced that incorporating compostable materials with biowaste had no impact on the composting process. Compost obtained met safety and agronomic requirements of

standard NF U44-051 and organic farming criteria. No ecotoxicity was revealed on plants, earthworms or daphnia. The compostable materials showed rapid biodegradation after 4 months with around 2 wt% of very small residual fragments consisting mainly of PHA, which disintegrated more slowly due to geometric factors. The environmental assessment indicated that composting biodegradable plastics with biowaste had less impact than incineration. In short, compostable materials can be part of sustainable waste management strategies.

CRedit authorship contribution statement

Emmanuelle Gastaldi: Writing – review & editing, Writing – original draft, Validation, Supervision, Project administration, Methodology, Investigation, Data curation, Conceptualization. **Felipe Buendia:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Paul Greuet:** Data curation, Investigation, Methodology. **Zineb Benbrahim Bouchou:** Investigation, Methodology. **Anir Benihya:** Investigation, Methodology. **Guy Cesar:** Conceptualization, Data curation, Investigation, Methodology. **Sandra Domenek:** Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Validation, Writing – original draft, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgements

The project was funded by the Foundation of AgroParisTech, which hosts the philanthropic partnership Chair CoPack (www.chairecopack.fr). The authors wish to thank the team of the Syndicat Centre Hérault (Aspiran, France) for having hosted the full-scale experiment in their composting facility and for their assistance in the implementation of the overall composting process. Cheick Abou Coulibaly is also thank for providing FTIR analysis.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.biortech.2024.130670>.

References

- Al-Rumaihi, A., McKay, G., Mackey, H.R., Al-Ansari, T., 2020. Environmental Impact Assessment of Food Waste Management Using Two Composting Techniques. *Sustainability* 12 (4), 23.
- ASTM C702/C702M:2018. Standard practice for reducing samples of aggregate to testing size. West Conshohocken, PA: ASTM International.
- Bassi, S.A., Christensen, T.H., Damgaard, A., 2017. Environmental performance of household waste management in Europe - An example of 7 countries. *Waste Manag.* 69, 545–557.
- Beylot, A., Muller, S., Descat, M., Ménard, Y., Villeneuve, J., 2018. Life cycle assessment of the French municipal solid waste incineration sector. *Waste Manag.* 80, 144–153.
- Bishop, G., Styles, D., Lens, P.N.L., 2021. Environmental performance comparison of bioplastics and petrochemical plastics: A review of life cycle assessment (LCA) methodological decisions. *Resour. Conserv. Recycl.* 168, 105451.
- Briassoulis, D., Pikasi, A., Hiskakis, M., 2021. Organic recycling of post-consumer/ industrial bio-based plastics through industrial aerobic composting and anaerobic digestion - Techno-economic sustainability criteria and indicators. *Polym. Degrad. Stab.* 190.
- Brinton, W., Evans, E., Droffner, M.L., Brinton, R.B., 1995. Standardized test for evaluation of compost self-heating. *Biocycle* 36 (11), 64–69.
- Bulatovic, V.O., Grgic, D.K., Slouf, M., Ostafinska, A., Dybal, J., Jozinovic, A., 2019. Biodegradability of blends based on aliphatic polyester and thermoplastic starch. *Chem. Pap.* 73 (5), 1121–1134.
- Chin, M.Y., Lee, C.T., Woon, K.S., 2023. Developing Circular Waste Management Strategies Based on a Waste Eco-Park Concept: A Multiobjective Optimization with Environmental, Economic, and Social Trade-offs. *Ind. Eng. Chem. Res.* 62 (41), 16827–16840.
- Edo, C., Fernandez-Pinas, F., Rosal, R., 2022. Microplastics identification and quantification in the composted Organic Fraction of Municipal Solid Waste. *Sci. Total Environ.* 813.
- EN 16087-2: 2011. Soil improvers and growing media - Determination of the aerobic biological activity - Part 2: Self heating test for compost.
- EN 13432: 2000 - Requirements for packaging recoverable through composting and biodegradation - Test scheme and evaluation criteria for the final acceptance of packaging.
- EN 12579:2013. Soil improvers and growing media-sampling.
- European Environment Agency, 2020. EEA Report No 04/2020 Bio-waste in Europe — turning challenges into opportunities. Accessed 13/10/2023. <https://www.eea.europa.eu/publications/bio-waste-in-europe>.
- European Environment Agency, 2023. Coverage of the population with high convenience collection systems in EU Member States by waste stream in municipal waste, and average share of each material in municipal waste. <https://www.eea.europa.eu/data-and-maps/figures/coverage-of-the-population-with>. Accessed 13/10/2023.
- Gaillard, T., George, M., Gastaldi, E., Nallet, F., Fabre, P., 2019. An experimental and theoretical study of the erosion of semi-crystalline polymers and the subsequent generation of microparticles. *Soft Matter* 15 (41), 8302–8312.
- Gerometta, M., Rocca-Smith, J.R., Domenek, S., Karbowski, T., 2019. Physical and Chemical Stability of PLA in Food Packaging. in: *Reference Module in Food Science*, Elsevier. <https://doi.org/10.1016/B978-0-08-100596-5.22471-2>.
- Greene, J., 2007. Biodegradation of Compostable Plastics in Green Yard-Waste Compost Environment. *J. Polym. Environ.* 15 (4), 269–273.
- Haider, T.P., Volker, C., Kramm, J., Landfester, K., Wurm, F.R., 2019. Plastics of the Future? The Impact of Biodegradable Polymers on the Environment and on Society. *Angewandte Chemie-International Edition* 58 (1), 50–62.
- Iqbal, S., Xu, J.C., Allen, S.D., Khan, S., Nadir, S., Arif, M.S., Yasmeen, T., 2020. Unraveling consequences of soil micro- and nano-plastic pollution on soil-plant system: Implications for nitrogen (N) cycling and soil microbial activity. *Chemosphere* 260, 8.
- Kale, G., Auras, R., Singh, S.P., 2006. Degradation of Commercial Biodegradable Packages under Real Composting and Ambient Exposure Conditions. *J. Polym. Environ.* 14 (3), 317–334.
- Kale, G., Auras, R., Singh, S.P., Narayan, R., 2007a. Biodegradability of polylactide bottles in real and simulated composting conditions. *Polym. Test.* 26 (8), 1049–1061.
- Kale, G., Kijchavengkul, T., Auras, R., Rubino, M., Selke, S.E., Singh, S.P., 2007b. Compostability of bioplastic packaging materials: An overview. *Macromol. Biosci.* 7 (3), 255–277.
- Kawashima, N., Yagi, T., Kojima, K., 2021. Pilot-Scale Composting Test of Polylactic Acid for Social Implementation. *Sustainability* 13 (4).
- Kedzia, G., Ocicka, B., Pluta-Zaremba, A., Rzniewska, M., Turek, J., Wieteska-Rosiak, B., 2022. Social Innovations for Improving Compostable Packaging Waste Management in CE: A Multi-Solution Perspective. *Energies* 15 (23), 19.
- Klauss, M., Bidlingmaier, W., 2004. Pilot scale field test for compostable packaging materials in the City of Kassel, Germany. *Waste Management* 24 (1), 43–51.
- Lashermes, G., Nicolardot, B., Parnaudeau, V., Thuriès, L., Chaussod, R., Guillotin, M.L., Linères, M., Mary, B., Metzger, L., Morvan, T., Tricaud, A., Villette, C., Houot, S., 2009. Indicator of potential residual carbon in soils after exogenous organic matter application. *Eur. J. Soil Sci.* 60 (2), 297–310.
- Leppanen, I., Vikman, M., Harlin, A., Orelma, H., 2020. Enzymatic Degradation and Pilot-Scale Composting of Cellulose-Based Films with Different Chemical Structures. *J. Polym. Environ.* 28 (2), 458–470.
- Limsukon, W., Rubino, M., Rabnawaz, M., Lim, L.T., Auras, R., 2023. Hydrolytic degradation of poly(lactic acid): Unraveling correlations between temperature and the three phase structures. *Polym. Degrad. Stab.* 217, 10.
- Martin, O., Averous, L., 2001. Poly(lactic acid): plasticization and properties of biodegradable multiphase systems. *Polymer* 42 (14), 6209–6219.
- Musiol, M., Sikorska, W., Adamus, G., Janeczek, H., Kowalczyk, M., Rydz, J., 2016a. (Bio)degradable polymers as a potential material for food packaging: studies on the (bio)degradation process of PLA/(R, S)-PHB rigid foils under industrial composting conditions. *Eur. Food Res. Technol.* 242 (6), 815–823.
- Musiol, M., Sikorska, W., Adamus, G., Janeczek, H., Richert, J., Malinowski, R., Jiang, G. Z., Kowalczyk, M., 2016b. Forensic engineering of advanced polymeric materials. Part III - Biodegradation of thermoformed rigid PLA packaging under industrial composting conditions. *Waste Manag.* 52, 69–76.
- NF ISO 14855-1:2012. Determination of the ultimate aerobic biodegradability of plastic materials under controlled composting conditions - Method by analysis of evolved carbon dioxide - Part 1: General method.
- NF U44-051:2006. Organic soil improvers — Designations, specifications and marking.
- Policastro, G., Cesaro, A., 2023. Composting of Organic Solid Waste of Municipal Origin: The Role of Research in Enhancing Its Sustainability. *Int. J. Environ. Res. Public Health* 20 (1).
- Porterfield, K.K., Hobson, S.A., Neher, D.A., Niles, M.T., Roy, E.D., 2023. Microplastics in composts, digestates, and food wastes: A review. *J. Environ. Qual.* 52 (2), 225–240.
- Puyuelo, B., Colon, J., Martin, P., Sanchez, A., 2013. Comparison of compostable bags and aerated bins with conventional storage systems to collect the organic fraction of

- municipal solid waste from homes. A Catalonia Case Study. *Waste Management* 33 (6), 1381–1389.
- Rodrigues, L.C., Puig-Ventosa, I., López, M., Martínez, F.X., Ruiz, A.G., Bertrán, T.G., 2020. The impact of improper materials in biowaste on the quality of compost. *J. Clean. Prod.* 251.
- Saeidlou, S., Huneault, M.A., Li, H., Park, C.B., 2012. Poly(lactic acid) Crystallization. *Prog. Polym. Sci.* 37, 1657–1677.
- Salomez, M., George, M., Fabre, P., Touchaleaume, F., Cesar, G., Lajarrige, A., Gastaldi, E., 2019. A comparative study of degradation mechanisms of PHBV and PBSA under laboratory-scale composting conditions. *Polym. Degrad. Stab.* 167, 102–113.
- Soest, P.v., Wine, R. 1967. Use of detergents in the analysis of fibrous feeds. IV. Determination of plant cell-wall constituents. *Journal of the Association of Official Analytical Chemists*, 50(1), 50-55.
- Statista. 2023. Generation of municipal waste in the European Union (EU-27) from 2004 to 2021 <https://www.statista.com/statistics/983592/municipal-waste-generation-european-union-eu-28/>. Accessed 13/10/2023.
- Van Roijen, E.C., Miller, S.A., 2022. A review of bioplastics at end-of-life: Linking experimental biodegradation studies and life cycle impact assessments. *Resour. Conserv. Recycl.* 181, 106236.
- Vigoroso, L., Pampuro, N., Bagagiolo, G., Cavallo, E., 2021. Factors influencing adoption of compost made from organic fraction of municipal solid waste and purchasing pattern: A survey of Italian professional and hobbyist users. *Agronomy* 11 (6), 1262.
- Weng, Y.-X., Wang, Y., Wang, X.-L., Wang, Y.-Z., 2010. Biodegradation behavior of PHBV films in a pilot-scale composting condition. *Polym. Test.* 29 (5), 579–587.
- Weng, Y.-X., Wang, X.-L., Wang, Y.-Z., 2011. Biodegradation behavior of PHAs with different chemical structures under controlled composting conditions. *Polym. Test.* 30 (4), 372–380.
- Zhang, H., McGill, E., Gomez, C.O., Carson, S., Neufeld, K., Hawthorne, I., Smukler, S.M., 2017. Disintegration of compostable foodware and packaging and its effect on microbial activity and community composition in municipal composting. *Int. Biodeter. Biodegr.* 125, 157–165.
- Zhou, Y.T., Ren, X.A., Tsui, T.H., Barcelo, D., Wang, Q., Zhang, Z.Q., Ding, Y.Z., 2023. Microplastics as an underestimated emerging contaminant in solid organic waste and their biological products: Occurrence, fate and ecological risks. *J. Hazard. Mater.* 445.