# Highlights

- A mechanically separated residual organic fraction (ms-OFRMW) was investigated
- A biostabilised residual organic fraction (bios-OFRMW) was investigated
- A biodried residual fine fraction (biod-FFRMW) was investigated
- ms-OFRMW could be suitable for bioenergy recovery through anaerobic digestion
- bios-OFRMW and biod-FFRMW may still contribute to landfill methane generation

## **CRediT** author statement

Manuela Carchesio: Methodology, Investigation, Formal Analysis. Martina Di Addario: Methodology, Investigation, Formal Analysis, Visualisation, Writing - Original Draft. Fabio Tatàno: Conceptualisation, Methodology, Resources, Formal Analysis, Visualisation, Writing -Original Draft, Writing - Review & Editing, Supervision, Project administration. Sandro de Rosa: Conceptualisation, Supervision, Project administration. Alma Gambioli: Resources, Supervision.

# Evaluation of the biochemical methane potential of residual organic fraction and mechanically-biologically treated organic outputs intended for landfilling

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#### Abstract

Mechanical biological treatment (MBT) approaches are being adopted to manage residual municipal waste (RMW) to promote the prevention or reduction of potential environmental impacts of landfilling. From this perspective, the present study aimed to increase the knowledge of the biological (anaerobic) stability of different MBT organic outputs and, conversely, initial methane generation from residual organic waste. Biochemical methane potential (BMP) tests, along with initial and final characterisations of substrates and digestates, were conducted on: a mechanically separated organic fraction from RMW (ms-OFRMW); a first MBT organic output represented by a biostabilised organic fraction from RMW (bios-OFRMW); and a different MBT organic output represented by a biodried fine fraction from RMW (biod-FFRMW). The ms-OFRMW had a BMP of 445.6 Nml CH<sub>4</sub> g VS<sup>-1</sup>, which was comparable or even higher than those from separately collected and source-sorted organic fractions. The fibre and liquor fractions of the digestate from ms-OFRMW with inoculum showed potential profiles of P-rich amendment and N-rich fluid phase, respectively, even satisfying environmental limits (with the exclusion only of Cu and Zn contents in fibre fraction that, however, remained within typical ranges for agricultural digestates). The BMPs for bios-OFRMW and biod-FFRMW were 143.4 and 261.0 Nml CH<sub>4</sub> g VS<sup>-1</sup>, respectively, indicating that these streams may still contribute to landfill methane generation. The BMPs for bios-OFRMW, biod-FFRMW, and ms-OFRMW were positively associated with the degrees of conversion of the substrates (17, 32, and 55%, respectively) and the potential dynamic respiration indexes (955, 3126, and 6062 mg O<sub>2</sub> kg VS<sup>-1</sup> h<sup>-1</sup>, respectively).

# Keywords:

Biochemical methane potential (BMP)

Biodrying

Biostabilisation

Mechanical biological treatment (MBT)

Organic output

Residual organic fraction

#### 1. Introduction

Landfills are known for their potential environmental impacts on air, soil, and water through the uncontrolled release of biogas and leachate, mainly generated from the organic matter in the deposited waste (Christensen et al., 2010a). In particular, landfills represent a significant source of anthropogenic emissions of methane, which is a strong greenhouse gas contributing to global warming (Capaccioni et al., 2011). To promote the prevention or reduction of the potential environmental impacts from landfills, the European landfill directive 1999/31/EC defined the following measures: (1) to ensure treatment (physical, thermal, chemical or biological, including sorting) of waste before landfilling; and (2) to progressively reduce the amount of biodegradable municipal waste being landfilled. At least at the European level, in dealing with these measures, the mechanical biological treatment (MBT) concept has increasingly been considered as an option for preliminary treatment of the organic fraction of residual municipal waste (RMW) prior to landfilling (Stegmann, 2010; Pantini et al., 2015; Trulli et al., 2018).

In the available range of technological variations, MBT plants are being used with the aim of producing either a biostabilised organic material destined for landfilling or an added-value waste-derived fuel as the main output (APAT, 2007; CITEC, 2008; Stegmann, 2010; Grilli et al., 2012). In the case of the MBT approach targeted at biostabilising organic waste, referred to as biostabilisation MBT, typically mechanical selection is first performed on the RMW with the aim of separating the organic fraction in the undersize stream, which is further partially biodegraded (mainly with aerobic systems) to obtain a stabilised organic output destined for landfilling (APAT, 2007; CITEC, 2008; Di Lonardo et al., 2012; Pantini et al., 2015; Cesaro et al., 2016). In the alternative case of the relatively new MBT approach optimised for waste-derived fuel production, referred to as biodrying MBT, the RMW undergoes aerobic biological pretreatment degrading part of the contained biodegradable

organic matter and acting as a waste drying process; further mechanical selection is implemented to obtain the final main output of processed solid fuel and a secondary output of biodried fine fraction enriched in organic material, which is intended for landfilling (CITEC, 2008; Velis et al., 2009; Bilitewski et al., 2010; Grilli et al., 2012).

To meet the aforementioned measures of landfill directive 1999/31/EC, which has been implemented in Italy with legislative decree 36/2003, many local waste authorities and landfill operators were encouraged to dispose of MBT organic outputs. However, these outputs are likely to still contain a certain amount of degradable organic compounds, whose biodegradation can be reactivated in the landfill body (Grilli et al., 2012; Heyer et al., 2013; Pantini et al., 2015). In view of these concerns, it is necessary to increase the knowledge of the biological stability conditions of different MBT organic outputs, with stability defined as the extent to which biodegradable organic matter has decomposed (Ponsá et al., 2008; Barrena et al., 2009). In particular, stability evaluation should provide an indirect indication of the actual presence of biodegradable organic matter, and consequently, it should represent a gradation on a scale of values, which thus possibly enables comparison of the levels of decomposition in different organic residues (Barrena et al., 2009; Tambone et al., 2011). Clearly, there exists a contextual need to investigate the potential of residual methane generation expected from different MBT organic outputs intended for landfilling (Heyer et al., 2013; Pantini et al., 2015). For a comprehensive research evaluation, the above needs should be associated with an investigation of the potential of initial methane generation expected from the residual organic fraction mechanically separated from the RMW.

Anaerobic digestion tests, performed under optimised operating conditions, are usually adopted to assess the biochemical methane potential (BMP) of biomass resources and organic waste (Caramiello et al. 2013; Carchesio et al., 2014). In addition, these tests can be seen as a possible approach to evaluate the biological stability of pretreated organic waste (APAT,

2003; Cossu and Raga, 2008; Ponsà et al., 2008; Grilli et al., 2012). In particular, determining the biological stability under anaerobic conditions appears to be consistent with the prevalent anaerobic environment that is expected when landfilling is the final destination of pretreated organic waste (Christensen et al., 2010b).

In this study, BMP test series (appropriately associated with the initial substrate and final digestate characterisations) were conducted to investigate: (1) a mechanically separated organic fraction from RMW (ms-OFRMW); (2) a first MBT organic output represented by a biostabilised organic fraction from RMW (bios-OFRMW), directly derived from the aerobic biostabilisation of ms-OFRMW; and (3) a different MBT organic output represented by a biodried fine fraction from RMW (biod-FFRMW) generated by an alternative MBT plant aimed at the primary production of waste-derived fuel. The investigated MBT organic outputs have both been disposed of at the controlled landfill for non-hazardous waste of the Fano town district (Marche Region, Central Italy, Adriatic Sea side), referred to as the reference landfill site hereafter, which is operated by the "ASET" public multi-utility group (Capaccioni et al., 2011). Table S1 of the Supplementary material reports representative data on the management of municipal waste (MW) in the regional territories where the two different MBT configurations generating the investigated substrates are located.

The objectives of the experimental test series conducted in this study were: (1) to evaluate the bioenergy recovery potential of ms-OFRMW through anaerobic digestion; (2) to evaluate the anaerobic biological stability of two alternative MBT organic outputs destined for landfilling (bios-OFRMW and biod-FFRMW) based on the resulting BMPs and their comparison with ms-OFRMW; (3) to examine the association of the obtained BMPs with both the resulting degrees of conversion of the substrates and alternative stability measures based on the aerobic respirometric approach (Ponsá et al., 2008); (4) to consider initial and final characterisations of the substrates and digestates, respectively, in the overall evaluation of the tested substrates; and (5) to obtain basic methane yield data to be used for the estimation via modelling (Willumsen and Barlaz, 2010) of the contributions of the deposited bios-OFRMW and biod-FFRMW to the overall methane generation at the reference landfill site.

#### 2. Materials and methods

#### 2.1. Substrates and inocula

The left side of Fig. 1 shows a flow chart of the overall biostabilisation MBT process that generates the ms-OFRMW as an intermediate stream and the bios-OFRMW as a final output. The collected and received RMW first undergoes mechanical separation involving bland shredding (to tear plastic bags) and sieving by a rotating trommel with a screen size of 40 mm. The resulting oversize fraction is directly disposed of at the reference landfill site, while the undersize fraction consists of the investigated ms-OFRMW. The ms-OFRMW is directly collected in movable biocontainers with an individual volume of 25 m<sup>3</sup>, which are arranged for the aerobic biostabilisation process. In particular, the biocontainers are connected to an air intake system and undergo a first biostabilisation phase consisting of intensive bio-oxidation. Then, the material is removed from the biocontainers and placed for a further curing phase in biocells with concrete walls, which are covered with breathable cloths and aerated from the bottom. The whole aerobic biostabilisation process lasts at least 14 days. The resulting output, which represents the investigated bios-OFRMW, is finally disposed of at the reference landfill site.

The right side of Fig. 1 shows a flow chart of the alternative biodrying MBT plant that generates the biod-FFRMW as a secondary output. After mechanical pre-processing of the received RMW consisting of pre-sorting of possible bulky, non-treatable materials and shredding (to tear plastic bags), the main stream is sent to the biodrying process. In particular,

biological pretreatment is conducted on contained piles of approximately 5 m in height, where forced aeration is realised through air intake systems placed at the bottom for a retention time of at least 14 days. Then, mechanical post-treatment or refining occurs, which consists of size separation via rotating trommel (with a small screen size of 20 mm) and a processing phase of the oversize material (removal of metals and inert fraction, fine shredding, and possibly pressing) to finally obtain the primary output of waste-derived fuel. The resulting secondary output of the undersize, fine fraction (< 20 mm), which represents the investigated biod-FFRMW, is generally destined for landfilling, including disposal at the reference landfill site.

The considered substrates were sampled by quartering method (UNI, 2013). Then, both ms-OFRMW and bios-OFRMW were manually fragmented simply using scissors at the laboratory prior to the subsequent characterisation and BMP test series. Instead, the biod-FFRMW was characterised and tested at the laboratory without any pretreatment, since it consisted of a visibly more homogeneous and fine material compared with ms-OFRMW and bios-OFRMW.

The inocula used for the BMP test series were anaerobic sludge samples obtained from the anaerobic digestion treatment stage at a wastewater treatment plant located in Central Italy.

Both the substrates and inocula were characterised in terms of moisture, total solids (TS), volatile solids (VS), total Kjeldahl nitrogen (TKN), total ammoniacal nitrogen (TAN), total phosphorus (TP), total potassium (TK), and heavy metals (Cd, Cr, Cu, Ni, Pb, and Zn). All the previous analytical determinations were conducted at least in triplicate. In addition, the ultimate analysis was conducted on a sample of each substrate. Finally, the aerobic stability of each substrate was measured using the potential dynamic respirometric index (PDRI) (Tambone et al., 2011; Grilli et al., 2012). The adopted analytical methods are summarised in Section 2.6.

#### 2.2. Lab-scale system

The lab-scale system (Fig. 2), specifically used for the BMP test series, is based on a volumetric approach for measuring methane production (Angelidaki and Batstone, 2010) and represents the development of an essential system applied in previous experimental studies (Caramiello et al., 2013; Carchesio et al., 2014). One-litre glass bottles equipped with proper inlets were used as batch digesters with a working volume of approximately 500 ml. The digesters were placed on heating magnetic stirrers connected with digital thermoregulators to maintain the internal temperature, measured with temperature probes inserted through pertaining inlets, at the set point. The headspaces of the digesters were connected via gasimpermeable Tygon tubes to the upper inlets of eudiometers for the periodical measurement of methane production through the volumetric liquid displacement principle (Guwy, 2004; Polettini et al., 2009; Ezebuiro et al., 2018). The eudiometers consisted of 1-litre vertical glass cylinders with a 2 ml progressive graduation from top to bottom; moreover, the lower outlets of the cylinders were connected via additional gas-impermeable Tygon tubes to respective glass reservoir tanks. The graduated cylinders and related interconnecting tubes and lower portions of the reservoir tanks were filled with a liquid chemical barrier consisting of an alkaline solution (NaOH) (Esposito et al., 2011; Carchesio et al., 2014). During the periodical gas measurements, the volumes of alkaline solution displaced from the upper parts of the graduated cylinders into the reservoir tanks were assumed to be equivalent to the corresponding volumetric methane productions (Esposito et al., 2011; Carchesio et al., 2014; Escalante et al., 2018). The accumulation of displaced liquid volumes yielded the temporal evolution of specific methane production (related to the initial total VS content of the feed) after normalising the gas volumes to 0 °C and 1 atm (Carchesio et al., 2014).

Before closing the digesters, the pH of each feed was measured with a portable pH-meter and adjusted to a level of approximately 8 with a Na<sub>2</sub>CO<sub>3</sub> buffer solution to limit excessive pH decreases during the early phase of anaerobic digestion (Esposito et al., 2011; Caramiello et al., 2013). In each BMP test series, the temporal evolution of pH was monitored in one digester containing the mixture of substrate with inoculum by a permanently inserted electrode of a portable pH-meter through the silicone septum of the corresponding inlet.

#### 2.3. BMP test series

Three BMP test series (Table 1) were conducted in batch mode to investigate the considered substrates ms-OFRMW (test series  $T_{ms-OFRMW}$ ), bios-OFRMW (test series  $T_{bios-OFRMW}$ ), and biod-FFRMW (test series  $T_{biod-FFRMW}$ ). In each test series, the reaction lines, each comprising the components described in Section 2.2, were arranged in triplicate for the mixture of substrate with inoculum and the inoculum as a blank. The substrate to inoculum ratio was fixed at 2:1 on a VS basis (Caramiello et al., 2013; Carchesio et al., 2014). The operating temperature was set to mesophilic conditions (38 °C) (Weiland, 2010; Ware and Power, 2017). The average laboratory ambient air temperatures were 27.6, 23.7, and 26.1 °C in test series  $T_{ms-OFRMW}$ ,  $T_{bios-OFRMW}$ , and  $T_{biod-FFRMW}$ , respectively.

For the mixtures of substrate with inoculum in the performed test series, the experimental measures of cumulative specific methane production were simulated using either the exponential rise to maximum model or the modified Gompertz model (Lo et al., 2010; Carchesio et al., 2014), the mathematical details of which are summarised in Table S2 of the Supplementary material. In particular, the modified Gompertz model considers a sigmoidal curve with the possible presence of an initial phase in which the methane production rate increases to a maximal value ( $\mu_m$ ), followed by a phase of progressive decrease until an upper horizontal asymptote (A) is reached; the time-axis intercept of the tangent in the inflection point of the curve represents the lag time ( $\lambda$ ) (Zwietering et al., 1990). Specifically, the best-fit curve for each test series was selected based on which model showed a better combination

of a high coefficient of determination  $(R^2)$  and low relative difference between the predicted upper horizontal asymptote (A) and measured final specific methane production (calculated as [(A - final specific methane production)/(final specific methane production)] \* 100), further confirmed by visual inspection of a suitable fit (Ware and Power, 2017).

#### 2.4. Determination of the resulting BMP and degree of conversion

The resulting BMP, regarded as the net specific methane yield directly and only attributable to each substrate (ms-OFRMW, bios-OFRMW, or biod-FFRMW) investigated in a given test series (Owen et al., 1979; Angelidaki and Batstone, 2010), was determined in accordance with the procedure indicated by Carchesio et al. (2014) and summarised in Table S3 of the Supplementary material.

Mass conversion analysis of the tested substrates was performed on a chemical oxygen demand (COD) basis expressing the resulting degree of conversion as the mass ratio between the COD converted to methane and the initial COD of the substrate (Labatut et al., 2011; Moody et al., 2011; Ghasimi et al., 2016). The denominator and numerator in calculating the degree of conversion were determined in accordance with the procedure indicated by Carchesio et al. (2014) and summarised in Table S4 of the Supplementary material.

#### 2.5. Digestate characterisation

In addition to pH temporal evolution monitoring in one digester (see Section 2.2), the pH was measured in the remaining digesters at the end of the respective BMP test series with a portable pH-meter. Primary sampling of the digestates consisted of pooled samples collected from respective digesters. Moisture and TS determinations were performed in triplicate on the whole digestates. Then, following solid-liquid separation by centrifugation, both the separated fibre (or solid) and liquor (or liquid) fractions of the whole digestates (Zhang et al., 2012;

Fuchs and Drosg, 2013; Nkoa, 2014) were characterised in terms of TS, VS, TKN, TAN, TP, TK, and heavy metals. These determinations were conducted at least in triplicate. The adopted analytical methods are summarised in Section 2.6.

#### 2.6. Analytical methods

The related moisture and TS contents were determined by drying at 105 °C. The VS content was determined by loss-on-ignition at 550 °C. The TKN and TAN contents were determined following the analytical method established for wastewater sludge (IRSA, 1985). The TP content was determined via inductively coupled plasma optical emission spectrometry (Vista-MPX, Varian) (UNI, 2004, 2009). The TK and heavy metal contents were determined through pretreatment based on microwave digestion (ETHOS 1, Milestone) (UNI, 2004), followed by analysis via inductively coupled plasma mass spectrometry (XSERIES 2 ICP-MS, Thermo Fisher Scientific) (USEPA, 2014). The ultimate analysis was conducted by using an elemental analyser (EA1110, Carlo Erba) (UNI, 2011). Finally, the PDRI was determined by using an adiabatic respirometer (ECHO, Emme3) (UNI, 2016).

#### 2.7. Statistical analysis

The means of the physico-chemical characteristics of the investigated substrates were statistically compared based on analysis of variance (ANOVA) with related Tukey HSD test. The means of the physico-chemical characteristics of the mutual fibre and liquor fractions of the whole digestate obtained from each BMP test series were statistically compared based on the *t*-test. All these statistical elaborations, as well as the nonlinear regression analysis functional to the curve-fitting procedure mentioned in Section 2.3, were conducted using KaleidaGraph (version 4.5.4, Synergy Software).

#### 3. Results and discussion

#### 3.1. Representative physico-chemical characteristics of the substrates and inocula

The analytical and statistical data on the physico-chemical characteristics of the investigated substrates are reported in Table 2. As outlined in Table S5 of the Supplementary material, the mean contents of physico-chemical characteristics of the investigated ms-OFRMW and bios-OFRMW from the biostabilisation MBT (Fig. 1, left), shown in Table 2, were mostly in line with or close to literature data on mechanically sorted and aerobically stabilised organic fractions, respectively. Regarding the investigated biod-FFRMW from the biodrying MBT (Fig. 1, right), the resulting mean moisture and VS contents (Table 2) fell within the overall ranges of 14.86-37% fresh matter (FM) and 44.53-50.3% TS, respectively, derivable in the literature for four samples of mechanically sieved fine fractions from biodried RMW (APAT, 2003; Grilli et al., 2012).

Interestingly, the direct comparison between ms-OFRMW and bios-OFRMW (Table 2) revealed mutual decreases in mean moisture and VS contents and mutual increases in mean TKN and heavy metal contents. Except for VS and Pb, the mentioned mutual differences were statistically significant. In general, decreases in moisture and VS contents (with VS representative of the organic matter: ANPA, 2001) and increases in nitrogen and heavy metal contents could be indicative of progress in a typical aerobic biodegradation process (Tatàno et al., 2015), although the influence of heterogeneity in materials such as the organic fractions mechanically separated from RMW could also be expected (van Praagh et al., 2009; Di Lonardo et al., 2015). In particular, the relative reduction in the VS content between ms-OFRMW and bios-OFRMW (Table 2), calculated as [(VS<sub>ms-OFRMW</sub> – VS<sub>bios-OFRMW</sub>)/(VS<sub>ms-OFRMW</sub>)] \* 100, was limited to 10.1%, which fell within the range of 5.2-16.1% derivable in the literature from previous mutual characterisations of untreated and biologically treated undersize fractions from RMW (Barrena et al., 2009; Salati et al., 2013; Di Lonardo et al.,

2015). Indeed, it should be pointed out that the presence of non-biodegradable volatile matter (i.e., plastic and rubber) generally occurring in heterogeneous materials, such as the organic fractions mechanically separated from RMW, may mask the actual biogenic VS reduction attained by the aerobic biological pretreatment (APAT, 2003, 2007; Ponsá et al., 2008; Grilli et al., 2012; Salati et al., 2013).

The elemental composition (in terms of C, O, H, and N) of each substrate, obtained from the ultimate analysis, and the representative chemical formulas consequently derived and needed to determine the resulting degrees of conversion (see Table S4 of the Supplementary material), are provided in Table S6 of the Supplementary material.

Finally, the resulting data from the similar physico-chemical characterisation conducted on the inocula used are provided in Table S7 of the Supplementary material. Moreover, a comparative evaluation of characteristics of the inocula used with literature indications is presented in Table S8 of the Supplementary material.

#### 3.2. Temporal evolutions of the BMP test series

Fig. 3 shows the resulting temporal evolutions of pH in individual digesters containing the mixtures of substrate with inoculum in test series  $T_{ms-OFRMW}$  (upper diagram),  $T_{bios-OFRMW}$  (central diagram), and  $T_{biod-FFRMW}$  (lower diagram). Starting from initial conditions, i.e., buffered maximum values (8.21, 8.13, and 8.08 in  $T_{ms-OFRMW}$ ,  $T_{bios-OFRMW}$ , and  $T_{biod-FFRMW}$ , respectively), the pH measures in Fig. 3 decreased to their respective minimum values at the initial phase of the anaerobic digestion. Interestingly, the lowest pH minimum value (5.90) occurred in  $T_{ms-OFRMW}$ , followed by that in  $T_{biod-FFRMW}$  (6.47), and  $T_{bios-OFRMW}$  exhibited the highest pH minimum value (6.55). Referring to the initial pH maximum values, the corresponding relative decreases (calculated as  $[(pH_{max} - pH_{min})/(pH_{max})] * 100)$  were 28.1, 19.9, and 19.4% in  $T_{ms-OFRMW}$ ,  $T_{biod-FFRMW}$ , and  $T_{bios-OFRMW}$ , respectively; thus, the relative difference in  $T_{ms-OFRMW}$  was larger than those in  $T_{biod-FFRMW}$  and  $T_{bios-OFRMW}$ . Indeed, the

extents of initial pH decreases, which are related to the primary production of volatile fatty acids and CO<sub>2</sub> at the beginning of the anaerobic digestion, can be comparatively seen as indicative of the respective amounts of readily and medium-degradable organic matter in the tested substrates (APAT, 2003; Christensen et al., 2010b; Pantini et al., 2015). Overall, the pH measures in each BMP test series in Fig. 3 evolved respecting the range of 6.0-8.3, where the anaerobic digestion is expected to occur (Angelidaki and Sanders, 2004); in particular, the pH minimum value in  $T_{ms-OFRMW}$  practically matched the lower limit of this range. In addition, following the increases after the minimum values, the pH measures in Fig. 3 also agreed (from the 7<sup>th</sup>, 5<sup>th</sup>, and 9<sup>th</sup> day on in  $T_{ms-OFRMW}$ ,  $T_{bios-OFRMW}$ , and  $T_{biod-FFRMW}$ , respectively) with the further restricted range of 7.0-8.0, which is reported in the literature as optimal for most methanogens (Angelidaki and Sanders, 2004; Weiland, 2010). Thus, the resulting evolutions of pH measures in Fig. 3 indicated that the pH-monitored digesters operated steadily during the respective test durations (Zheng et al., 2009).

Fig. 4 shows the resulting temporal evolutions of cumulative methane production in test series  $T_{ms-OFRMW}$  (upper diagram),  $T_{bios-OFRMW}$  (central diagram), and  $T_{biod-FFRMW}$  (lower diagram), relative to the initial total VS content in each mixture of substrate (ms-OFRMW, bios-OFRMW, and biod-FFRMW) with inoculum. The final specific methane productions from the mixtures of substrates with inocula were, in decreasing order, 305.0, 183.5, and 117.8 Nml CH<sub>4</sub> g VS<sup>-1</sup> for  $T_{ms-OFRMW}$ ,  $T_{biod-FFRMW}$ , and  $T_{bios-OFRMW}$ , respectively. For  $T_{ms-OFRMW}$  and  $T_{biod-FFRMW}$ , the selected modified Gompertz model shown in Fig. 4 (upper and lower) strongly fits the respective experimental measures, as indicated by the resulting high  $R^2$  values of 0.990 and 0.995, respectively (Hebel and McCarter, 2012); moreover, the relative differences between the respective predicted upper horizontal asymptotes (A) and measured final specific methane productions were negligible (as indicated in Table S9 of the Supplementary material). Regarding  $T_{bios-OFRMW}$ , the selected exponential rise to maximum

model shown in Fig. 4 (central) strongly fits the experimental measures according to the resulting high  $R^2$  value of 0.999 (Hebel and McCarter, 2012); similarly, as indicated in Table S9 of the Supplementary material, the relative difference between the predicted and measured final specific methane productions was negligible. Interestingly, the exponential rise to maximum modelling of the cumulative methane production measures for T<sub>bios-OFRMW</sub> in Fig. 4 (central) indicated the inherent absence of an initial lag phase (Sawyer et al., 2003; Ponsá et al., 2010). Moreover, the modified Gompertz modelling of the cumulative methane production measures for the remaining test series in Fig. 4 (upper and lower) revealed either the absence of an initial lag phase in T<sub>ms-OFRMW</sub> (mathematically due to a resulting negative  $\lambda$  value of -0.93 d: Ponsá et al., 2010) or a negligible initial lag phase in T<sub>biod-FFRMW</sub> (due to a resulting limited  $\lambda$  value of 0.74 d, accounting for only 1.3% of the overall anaerobic digestion duration). These conditions of an absent or negligible initial lag phase indicated that the performed BMP test series operated under optimal conditions (Pantini et al., 2015).

Finally, parametric comparison of the modified Gompertz modelling of the cumulative methane production measures for  $T_{ms-OFRMW}$  and  $T_{biod-FFRMW}$  (Fig. 4, upper and lower) also revealed a higher  $\mu_m$  value in  $T_{ms-OFRMW}$  (15.6 Nml CH<sub>4</sub> g VS<sup>-1</sup> d<sup>-1</sup>) than that in  $T_{biod-FFRMW}$  (9.0 Nml CH<sub>4</sub> g VS<sup>-1</sup> d<sup>-1</sup>), which indicated a more rapid anaerobic digestion in  $T_{ms-OFRMW}$  (Ware and Power, 2017).

#### 3.3. BMPs and related associations with the degrees of conversion and PDRIs

The resulting BMPs were, in decreasing order, 445.6, 261.0, and 143.4 Nml  $CH_4$  g  $VS^{-1}$  for ms-OFRMW, biod-FFRMW, and bios-OFRMW, respectively. Thus, the ms-OFRMW had the highest BMP among the tested substrates, as expected since this residue was generated solely from mechanical pretreatment. Even the obtained BMP for ms-OFRMW was practically located in the middle of the range of 401-489 ml  $CH_4$  g  $VS^{-1}$  derivable in the

literature for separately collected and source-sorted organic waste (Cecchi et al., 2002; Zhang et al., 2012; Naroznova et al., 2016). The direct comparison of bios-OFRMW to ms-OFRMW revealed a relative BMP decrease of 68% (calculated as [(BMPms-OFRMW - BMPbios-OFRMW)/(BMP<sub>ms-OFRMW</sub>)] \* 100). Thus, based on BMP evaluation (Grilli et al., 2012), the bios-OFRMW exhibited an anaerobic biological stability increase of 68% as directly related to the ms-OFRMW. Instead, the indirect comparison of biod-FFRMW to ms-OFRMW revealed a smaller relative BMP decrease (or, conversely, a smaller anaerobic biological stability increase) that was limited to 41% (calculated as [(BMP<sub>ms-OFRMW</sub> - BMP<sub>biod-FFRMW</sub>)/(BMP<sub>ms-</sub> OFRMW)] \* 100). Interestingly, considering the two different MBT organic outputs, the bios-OFRMW exhibited a lower BMP than the biod-FFRMW. This could be attributed to the following: (1) in biostabilisation MBT (Fig. 1, left), the aerobic biological step was effectively directed towards the residual organic fraction only, while the whole main waste stream (including the organic fraction) was subjected to the aerobic biological step in biodrying MBT (Fig. 1, right); and (2) the biod-FFRMW consisted of a more homogeneous and fine material compared with the bios-OFRMW, thus promoting bacterial activity on the substrate during anaerobic digestion (Hajji and Rhachi, 2013). The resulting BMP for biod-FFRMW, which corresponded to 120.1 Nml CH<sub>4</sub> g TS<sup>-1</sup> assuming the substrate TS as the reference term, was higher than the methane yields of 93 and 98 Nml CH<sub>4</sub> g TS<sup>-1</sup> measured under mesophilic conditions (at lab and pilot scales, respectively) from a biodried fine fraction that was obtained, however, from a coarser (60 mm) final mechanical sieving (Grilli et al., 2012), thus confirming the possible positive effect of a finer substrate on the expected biomethane production. The resulting BMP for bios-OFRMW, equal to 71.2 Nml CH<sub>4</sub> g TS<sup>-1</sup> on a TS substrate basis, remained: (1) higher than the mean of 39.9 Nml CH<sub>4</sub> g TS<sup>-1</sup> obtained under mesophilic conditions from coarser aerobically biostabilised organic outputs (60 mm undersize fractions) (Scaglia et al., 2010); (2) slightly higher than the mean of 64.0 Nml CH<sub>4</sub> g TS<sup>-1</sup> derived under mesophilic conditions from two samples of coarser undersize fractions (50 mm) that were aerobically biostabilised for slightly shorter and longer periods, respectively, than the normal operation time of the biological step at a given MBT plant (Barrena et al., 2009); (3) but lower than the value of 121 Nml CH<sub>4</sub> g TS<sup>-1</sup> measured under thermophilic conditions from a fine aerobically biostabilised organic output (20 mm undersize fraction) (Pantini et al., 2015).

The resulting degrees of conversion of the tested substrates were, in decreasing order, 55, 32, and 17% for ms-OFRMW, biod-FFRMW, and bios-OFRMW, respectively. In general, a lower degree of conversion is indicative of a lower biodegradability of the substrate, most likely due to a greater presence of recalcitrant organic substances, less soluble/degradable compounds, or already humified organic matter (Labatut et al., 2011; Pantini et al., 2015; Tatàno et al., 2015). Overall, the resulting degrees of conversion of the tested substrates fell within the wide range of 9-78% reported on a COD basis for a large array of organic substrates evaluated in BMP tests (Labatut et al., 2011). In particular, the resulting degrees of conversions of bios-OFRMW and biod-FFRMW were within the further restricted range of degradability of 10-45% reported for MBT organic outputs (Pantini et al., 2015). Interestingly, a positive association was revealed, as shown in Fig. 5 (left-up), between the resulting BMPs and degrees of conversion for the tested substrates. By comparing both bios-OFRMW (directly) and biod-FFRMW (indirectly) to ms-OFRMW in terms of degree of conversion, the resulting relative decreases (calculated as [(degree<sub>ms-OFRMW</sub> - degree<sub>bios-</sub> OFRMW)/(degree<sub>ms-OFRMW</sub>)] \* 100 and [(degree<sub>ms-OFRMW</sub> - degree<sub>biod-FFRMW</sub>)/(degree<sub>ms-OFRMW</sub>)] \* 100, respectively) were equal to 69 and 42%, respectively, which practically matched the respective relative decreases in terms of BMP (Fig. 5, right-up).

The determined PDRIs for the tested substrates were, in decreasing order, 6062, 3126, and 955 mg  $O_2$  kg VS<sup>-1</sup> h<sup>-1</sup> for ms-OFRMW, biod-FFRMW, and bios-OFRMW, respectively. The

determined PDRI for ms-OFRMW was practically at the higher limit of the overall range of 3000-6000 mg O<sub>2</sub> kg VS<sup>-1</sup> h<sup>-1</sup> indicated in the literature for the residual organic fraction after mechanical screening (APAT, 2003; Di Maria et al., 2013; Salati et al., 2013; Cesaro et al., 2016; Trulli et al., 2018). The determined PDRI for biod-FFRMW was practically at the higher limit of the range of 889-3032 mg  $O_2$  kg VS<sup>-1</sup> h<sup>-1</sup> derivable in the literature for four samples of biodried fine fraction (APAT, 2003; Grilli et al., 2012). For bios-OFRMW, the corresponding PDRI was within the range of 780-985 mg O<sub>2</sub> kg VS<sup>-1</sup> h<sup>-1</sup> derivable in the literature for four samples of biostabilised organic fraction (APAT, 2003; Salati et al., 2013). Interestingly, as shown in Fig. 5 (left-down), a positive association was also revealed between the resulting BMPs and PDRIs for the tested substrates. However, by comparing both bios-OFRMW (directly) and biod-FFRMW (indirectly) to ms-OFRMW in terms of PDRI, the resulting relative decreases (calculated as [(PDRI<sub>ms-OFRMW</sub> - PDRI<sub>bios-OFRMW</sub>)/(PDRI<sub>ms-OFRMW</sub>)] \* 100 and [(PDRI<sub>ms-OFRMW</sub> - PDRI<sub>biod-FFRMW</sub>)/(PDRI<sub>ms-OFRMW</sub>)] \* 100, respectively) were equal to 84 and 48%, respectively, which were larger than the respective relative decreases in terms of BMP, particularly concerning the relation of bios-OFRMW to ms-OFRMW (Fig. 5, rightdown).

The biological process was anaerobic in the BMP test series, which were conducted under steady and optimal conditions (as highlighted in Section 3.2), but the biological process was aerobic in the studied MBT configurations (Fig. 1) and in the PDRI determination, and the observed larger decreases in PDRI by relating both bios-OFRMW and biod-FFRMW to ms-OFRMW likely occurred because the anaerobic and aerobic biodegradations preferably affected specific, not completely overlapping pools of anaerobically or aerobically biodegradable organic matter, respectively (Ponsá et al., 2008; Barrena et al., 2009). Indeed, as a similar indication from the literature to the overestimation effect shown in Fig. 5 (right-down), also the relation of a large number of aerobically biostabilised organic-fraction

samples to untreated organic-fraction samples revealed that, on average, a larger decrease occurred in terms of PDRI (81%) compared to that (72%) in terms of anaerobic biogas potential (Scaglia et al., 2010).

#### 3.4. Representative physico-chemical characteristics of the digestates

The mean pH values of the digestates from the digesters with the original mixtures of substrates and inocula were 7.78, 7.37, and 7.89 in  $T_{ms-OFRMW}$ ,  $T_{bios-OFRMW}$ , and  $T_{biod-FFRMW}$ , respectively, which were in line with the aforementioned optimal range for most methanogens (see Section 3.2). The mean moisture contents of the whole digestates from the digesters with the original mixtures of substrates and inocula were 96.16, 96.09, and 94.31% FM in  $T_{ms-OFRMW}$ ,  $T_{bios-OFRMW}$ , and  $T_{biod-FFRMW}$ , respectively; these contents represented realistic values for full-scale anaerobic digestion, as confirmed, for instance, by the range of 90.4-98.3% obtained for digestates from the digesters with the original mixtures of substrates and inocula where soft substrates and inocula exhibited mean TS contents in a range from 3.84 (in  $T_{ms-OFRMW}$ ) to 5.69% FM (in  $T_{biod-FFRMW}$ ), which is consistent with a wet digestion process in which the digestate may contain up to 11.5% total solids (Monlau et al., 2015).

Table 3 lists the physico-chemical characteristics of the separated fibre and liquor fractions of the whole digestates from the digesters containing the original mixtures of substrates and inocula. Consistent with literature indications on the solid-liquid separation step of digestates in full-scale anaerobic digestion (Bauer et al., 2009; Fuchs and Drosg, 2013), the largest percentages by mass of the whole digestates were represented in Table 3 by the respective liquor fractions. Interestingly, the resulting smaller proportion by mass of the liquor fraction from  $T_{biod-FFRMW}$ , compared in Table 3 with the liquor fractions from the remaining BMP test series, was combined with the aforementioned higher TS content of the respective whole

digestate. Indeed, this inverse association is consistent with the inverse correlation that was reported between the TS contents of whole digestates and the mass proportions of the liquid fractions in a monitoring study of two full-scale biogas plants equipped with different solid-liquid separators (Bauer et al., 2009). The resulting TS contents of the fibre fractions in Table 3 were within the range of 9.92-41.6% FM reported for the solid fractions from mechanical separation of digestates in eleven full-scale anaerobic co-digestion plants (Akhiar et al., 2017). Moreover, the ratio of the TS content in the liquor fraction to that in the whole digestate, which can give an indication on the efficiency of solid-liquid separation (with a low ratio suggesting a good separation: Akhiar et al., 2017), was equal to 0.12, 0.14, and 0.16 in  $T_{ms-OFRMW}$ ,  $T_{biod-FFRMW}$ , and  $T_{bios-OFRMW}$ , respectively; these values were below (for  $T_{ms-OFRMW}$  and  $T_{biod-FFRMW}$ ) or practically at the lower limit (for  $T_{bios-OFRMW}$ ) of the range from 0.2 to over 0.8 characterising the digestates from the aforementioned full-scale anaerobic co-digestion plants (Akhiar et al., 2017).

With regard to the resulting nutrient contents, distinctive partitions with statistical significance were revealed in Table 3. In particular, the mean TKN, TAN, and TK contents in the liquor fractions were significantly higher (at the 0.05 level) than those in the respective fibre fractions; in contrast, the mean TP contents in the fibre fractions were significantly higher (at the 0.05 level) than those in the respective liquor fractions. These observed partitions qualitatively agreed with literature indications on the expected fractionations of N, P, and K contents between the separated liquid and solid fractions of digestates in full-scale anaerobic digestion (Möller and Müller, 2012; Fuchs and Drosg, 2013; Monlau et al., 2015).

As further revealed in Table 3, the mean heavy metal contents in the fibre fractions were always higher than those in the respective liquor fractions; these mutual differences were statistically significant (at the 0.05 level) with the exception of Ni in  $T_{biod-FFRMW}$ . Indeed, the production of liquids with distinctly reduced levels of heavy metals is mentioned in the

literature as one of the effects of digestate solid-liquid separation (Möller and Müller, 2012). In general, heavy metal solubilisation is limited by adsorption onto mineral and organic particles and under slightly alkaline conditions (Smith and Groudev, 2003; Carchesio et al., 2014), as verified by the above pH values of the whole digestates (Price, 2002).

Although the digestate composition is also influenced by the inoculum source (Monlau et al., 2015), in an attempt to comprehensively evaluate the possible bioenergy recovery option of ms-OFRMW through anaerobic digestion, characteristic partitioning data in Table 3 of the whole digestate from T<sub>ms-OFRMW</sub> were interpreted in terms of agronomic and environmental properties. Due to TAN solubility, the TAN/TKN ratio in the liquor fraction from T<sub>ms-OFRMW</sub> was higher (69.5%) than that in the respective fibre fraction (11.1%), thus enriching the liquor fraction in plant-available N (Möller and Müller, 2012; Tambone et al., 2017). In contrast, the mean VS contents from T<sub>ms-OFRMW</sub> (Table 3), although limited due to the expected impact of biodegradation during anaerobic digestion (Drosg et al., 2015), exhibited a statistically significant partition (at the 0.05 level) with a higher content in the fibre fraction than that in the respective liquor fraction. Therefore, compared to the liquor fraction, the fibre fraction from T<sub>ms-OFRMW</sub> could more readily act as an amendment (Tambone et al., 2017), in which the residual VS content is expected to mainly contain recalcitrant or inert organic matter that can improve the soil structure and promote humus formation (Tambone et al., 2010; Drosg et al., 2015). In particular, due to the aforementioned distinctive partition of TP (Table 3), the fibre fraction from T<sub>ms-OFRMW</sub> could potentially be considered a P-rich amendment (Monlau et al., 2015; Tambone et al., 2017).

Finally, as shown in Table 10 of the Supplementary material, the mean heavy metal contents in both the fibre and liquor fractions from  $T_{ms-OFRMW}$  (Table 3) conformed to upper environmental limits, either introduced in Italy for agroindustrial digestate or proposed at the European level for sludge land use, except for Cu and Zn in the fibre fraction, which exceeded

only the more stringent Italian limits. Nevertheless, the mean Cu and Zn contents in the fibre fraction from  $T_{ms-OFRMW}$  (Table 3) remained within the ranges of 14-270 and 72-2200 mg kg TS<sup>-1</sup>, respectively, reported as characteristic for agricultural digestates (Monlau et al., 2015).

#### 4. Conclusions

The physico-chemical characteristics of the investigated ms-OFRMW and bios-OFRMW were mostly in line with or close to literature data on mechanically sorted and aerobically stabilised organic fractions, respectively. The detailed physico-chemical profile of the investigated biod-FFRMW could provide a useful reference for future characterisation studies on biodried fine fractions. By directly comparing ms-OFRMW and bios-OFRMW characteristics, the observed distinctive differences in moisture, TKN, and most heavy metals, which were statistically significant, qualitatively agreed with the expected evolution during an aerobic biodegradation process.

The BMP test series were conducted under steady and reliable conditions as concurrently confirmed by: (1) the resulting temporal evolutions of pH; (2) the strong fitting of cumulative methane productions with suitable modelling curves, along with the absence or negligible presence of an initial lag phase; and (3) the qualitative agreement of the distinctive partitions of mass proportions, nutrients, and heavy metals between the fibre and liquor fractions of the obtained digestates with expected characteristics of full-scale digestates.

The ms-OFRMW had a BMP (445.6 Nml  $CH_4$  g  $VS^{-1}$ ) that was comparable or even higher than those obtained from separately collected and source-sorted organic fractions. Thus, the mechanically separated organic fraction from RMW could be suitable for bioenergy recovery, and its diversion to a biomethanation step, still within the MBT concept, could be advantageous at a suitable territorial scale. Overall, for this management option, the fibre and liquor fractions of the whole digestate from the mixture of ms-OFRMW with inoculum exhibited agronomic profiles potentially indicating P-rich amendment and N-rich fluid phase, respectively, even satisfying upper environmental limits on heavy metals, except for only Cu and Zn in the fibre fraction; nevertheless, the attained Cu and Zn contents agreed with the typical ranges for agricultural digestates.

With regard to the investigated MBT organic outputs, the resulting BMPs (143.4 and 261.0 Nml CH<sub>4</sub> g VS<sup>-1</sup> for bios-OFRMW and biod-FFRMW, respectively) indicated that these types of streams destined for landfilling may still contribute to landfill methane generation (and possible emissions). By directly comparing bios-OFRMW to ms-OFRMW, a relative BMP decrease (or, conversely, an anaerobic stability increase) of 68% was observed. Instead, by indirectly comparing biod-FFRMW to ms-OFRMW, the relative BMP decrease (or anaerobic stability increase) was limited to 41%. The higher BMP for biod-FFRMW than that for bios-OFRMW could likely be related to the following differences between biodrying and biostabilisation MBT: (1) the total versus fractional waste stream affected by the biological step; and (2) the finer versus coarser size of the (final or initial) sieving step.

The increasing order of the BMPs for bios-OFRMW, biod-FFRMW, and ms-OFRMW was positively associated with both the resulting degrees of conversion of the substrates and the determined PDRIs. However, while the relative decreases observed by comparing both bios-OFRMW and biod-FFRMW to ms-OFRMW in terms of degree of conversion practically matched those in terms of BMP, the attained relative decreases (or, conversely, aerobic stability increases) in terms of PDRI appeared to be overestimated.

Complementary works should focus on (1) estimation of bios-OFRMW and biod-FFRMW contributions to the overall methane generation at the reference landfill site and (2) evaluation of an alternative management scenario, at the Marche regional scale, based on bioenergy recovery from ms-OFRMW through anaerobic digestion.

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# Table 1

Test series	Feed	Number of	S/I <sup>a</sup>	Barrier	Initial pH	Adjusted	Temperature	Duration
		digesters		solution	(mean)	pH (mean)	(°C)	(d)
T <sub>ms-OFRMW</sub>	ms-OFRMW + inoculum	3	2	NaOH	7.25	8.15	38	46
	Inoculum (blank)	3	-	NaOH	7.08	8.09	38	46
T <sub>bios-OFRMW</sub>	bios-OFRMW + inoculum	3	2	NaOH	7.10	8.16	38	45
	Inoculum (blank)	3	-	NaOH	7.17	8.10	38	45
$T_{biod\text{-}FFRMW}$	biod-FFMRW + inoculum	3	2	NaOH	7.22	8.08	38	56
	Inoculum (blank)	3	-	NaOH	7.48	8.03	38	56

Feeding and operating conditions of the BMP test series.

<sup>a</sup> S/I: substrate to inoculum ratio on a VS basis.

# Table 2

Representative physico-chemical characteristics of the substrates.

Characteristic <sup>a, b</sup>	ms-OFRMW	bios-OFRMW	biod-FFRMW
Moisture (% FM)			
Mean	46.81 b	25.19 a	25.40 a
Standard deviation	4.09	2.77	2.95
CV (%)	8.74	11.00	11.61
TS (%, FM)			
Mean	53.19 a	74.81 b	74.60 b
Standard deviation	4.09	2.77	2.95
CV (%)	7.69	3.70	3.95
VS (% TS)			
Mean	55.27 a	49.67 a	46.02 a
Standard deviation	7.61	5.39	3.93
CV (%)	13.77	10.85	8.54
TKN (g kg TS <sup>-1</sup> )			
Mean	8.71 a	17.15 b	13.26 b
Standard deviation	1.10	2.96	0.44
CV (%)	12.63	17.26	3.32
TAN (g kg TS <sup>-1</sup> )			
Mean	0.88 b	0.50 a	1.48 c
Standard deviation	0.08	0.08	0.13
CV (%)	9.09	16.00	8.78
$TP (g kg TS^{-1})$			
Mean	2.19 a	2.05 a	1.77 a
Standard deviation	0.32	0.15	0.18
CV (%)	14.61	7.32	10.17
TK $(g kg TS^{-1})$			
Mean	3.84 a	6.79 b	14.99 c
Standard deviation	0.51	1.11	0.14
CV (%)	13.28	16.35	0.93
$Cd (mg kg TS^{-1})$			
Mean	0 32 a	0.53 b	1 29 c
Standard deviation	0.06	0.05	0.19
CV (%)	18.75	9.43	14.73
$Cr(mg kg TS^{-1})$			
Mean	11.62 a	23 77 h	25 50 h
Standard deviation	2 51	5 24	0.27
CV (%)	21.60	22.04	1.06
$Cu (mg kg TS^{-1})$	21.00		1.00
Mean	49 08 a	123 98 h	143 31 b
Standard deviation	11 22	28.78	2 53
CV (%)	22.86	23.21	1 77
Ni (mg kg $TS^{-1}$ )			
Mean	10.78 a	17.08 b	19 24 b
Standard deviation	2 51	5 11	0.83
CV (%)	23.28	29.92	4 31
$Ph (mg kg TS^{-1})$	23.20	27.72	1.01
Mean	10.88 a	21.58 a	226 42 h
Standard deviation	10.00 a 1 74	21.30 a 8 62	7 10
CV(%)	15 99	39.94	3 14
$7n (mg kg TS^{-1})$	10.77	57.71	.11
Lii (iiig Kg 10) Mean	131 33 9	217.84 b	220.26 b
Stondard doviation	131.33 a 27 16	217.04 U 57.69	12 20
	27.40	J/.00 26.49	12.20
UV (%)	20.91	20.48	5.54

<sup>a</sup> Mean: average of at least three determinations; CV: coefficient of variation.

<sup>b</sup> Means followed in the same line by the same lower-case letter are not significantly different at the 0.05 level according to ANOVA and related Tukey post hoc test.

# Table 3

Representative physico-chemical characteristics of the separated fibre and liquor fractions of the whole digestates from the digesters containing the original mixtures of substrates and inocula.

Characteristic <sup>a, b</sup>	T <sub>ms-OFRMW</sub>		T <sub>bios-OFRMW</sub>		T <sub>biod-FFRMW</sub>	
	Fibre	Liquor	Fibre	Liquor	Fibre	Liquor
Allocation by mass of whole digestate (%)	24.34	75.66	21.82	78.18	34.56	65.44
15 (% FM) Mean	14 88 b	0.45 a	18 07 b	0.61 a	13 26 h	0.78 a
Standard deviation	0.78	0.02	0.81	0.004	0.53	0.01
CV (%)	5.24	4.44	4.48	0.66	4.00	1.28
VS (% TS)						
Mean	17.83 b	8.40 a	53.48 b	33.73 a	53.01 b	35.09 a
Standard deviation	0.59	0.42	1.42	3.26	0.62	1.21
CV (%)	3.31	5.00	2.66	9.66	1.17	3.45
TKN (g kg TS <sup>-1</sup> )						
Mean	30.04 a	105.54 b	29.54 a	131.66 b	37.08 a	184.89 b
Standard deviation	0.26	1.26	2.07	2.26	1.59	3.39
CV (%)	0.87	1.19	7.01	1.72	4.29	1.83
TAN (g kg TS <sup>-1</sup> )						
Mean	3.34 a	73.32 b	3.11 a	130.39 b	3.53 a	150.83 b
Standard deviation	0.23	0.91	0.07	1.44	0.11	1.57
CV (%)	6.89	1.24	2.25	1.10	3.12	1.04
$TP (g kg TS^{-1})$						
Mean	21.10 b	2.63 a	20.52 b	2.03 a	35.19 b	3.54 a
Standard deviation	2.67	0.02	2.86	0.02	3.92	0.03
CV (%)	12.65	0.76	13.94	0.99	11.14	0.85
TK (g kg $TS^{-1}$ )						
Mean	3.44 a	85.42 b	3.00 a	52.53 b	1.28 a	55.68 b
Standard deviation	0.19	8.51	0.16	9.02	0.11	6.32
CV (%)	5.52	9.96	5.33	17.17	8.59	11.35
$Cd (mg kg TS^{-1})$						
Mean	1.01 b	0.43 a	0.77 b	0.00 a	3.50 b	0.58 a
Standard deviation	0.07	0.02	0.05	0.00	0.44	0.03
CV (%)	6.93	4.65	6.49	-	12.57	5.17
$Cr (mg kg TS^{-1})$						
Mean	47.55 b	2.56 a	41.17 b	2.31 a	77.54 b	21.78 a
Standard deviation	1.73	0.47	2.88	0.50	1.69	0.98
CV (%)	3.64	18.36	7.00	21.65	2.18	4.50
Cu (mg kg TS <sup>-1</sup> )						
Mean	255.35 b	8.55 a	268.78 b	1.91 a	315.77 b	57.58 a
Standard deviation	18.03	0.53	8.08	0.05	12.32	1.40
CV (%)	7.06	6.20	3.01	2.62	3.90	2.43
Ni (mg kg TS <sup>-1</sup> )						
Mean	32.29 b	13.99 a	22.61 b	7.81 a	35.40 a	32.49 a
Standard deviation	4.06	0.66	0.57	0.28	2.74	1.71
CV (%)	12.57	4.72	2.52	3.59	7.74	5.26
Pb (mg kg $TS^{-1}$ )						
Mean	34.45 b	3.22 a	35.05 b	0.26 a	344.13 b	45.10 a
Standard deviation	1.04	0.35	2.16	0.02	1.92	0.43
CV (%)	3.02	10.87	6.16	7.69	0.56	0.95
Zn (mg kg TS <sup>-1</sup> )						
Mean	900.42 b	35.56 a	811.50 b	14.28 a	920.28 b	224.63 a
Standard deviation	17.65	2.50	41.97	2.47	38.03	7.39
CV (%)	1.96	7.03	5.17	17.30	4.13	3.29

<sup>a</sup> Mean: average of at least three determinations; CV: coefficient of variation.

<sup>b</sup> Comparing the separated fibre and liquid fractions for each test series, means followed in the same line by the same lowercase letter are not significantly different at the 0.05 level according to the *t*-test.



**Fig. 1.** Flow charts of the technological configurations generating the investigated substrates: biostabilisation (left) and biodrying (right) MBT (legend: continuous line box = material stream; dotted line box = operation; box with grey background = investigated substrate).



**Fig. 2**. The lab-scale system developed and used for the BMP test series: overall visualisation (left); details of digester (right-down) and upper part of eudiometer with reservoir tank (right-up) (legend: A = digester; B = heating magnetic stirrer; C = pH electrode; D = portable pH-meter; E = digital thermoregulator; F = eudiometer; G = reservoir tank; H = gas-impermeable Tygon tube).



**Fig. 3.** Temporal evolutions of pH in individual digesters containing the mixtures of substrate with inoculum in BMP test series  $T_{ms-OFRMW}$  (upper),  $T_{bios-OFRMW}$  (central), and  $T_{biod-FFRMW}$  (lower).



**Fig. 4.** Temporal evolutions of cumulative methane production (relative to the initial total VS content) for the mixtures of substrate with inoculum in BMP test series  $T_{ms-OFRMW}$  (upper),  $T_{bios-OFRMW}$  (central), and  $T_{biod-FFRMW}$  (lower). The modelled curve fits are also reported. For diagram clearness, the vertical axes have different scales.



**Fig. 5.** Resulting associations of BMPs with degrees of conversion (left-up) and PDRIs (leftdown) for the tested substrates, and corresponding relative decreases by comparing bios-OFRMW and biod-FFRMW to ms-OFRMW in terms of BMP and degree of conversion (right-up) or PDRI (right-down). Legend: relative decreases calculated as [(BMP<sub>ms-OFRMW</sub> -BMP<sub>bios-OFRMW/biod-FFRMW</sub>)/(BMP<sub>ms-OFRMW</sub>)] \* 100 in terms of BMP, [(degree<sub>ms-OFRMW</sub> degree<sub>bios-OFRMW/biod-FFRMW</sub>)/(degree<sub>ms-OFRMW</sub>)] \* 100 in terms of degree of conversion, and [(PDRI<sub>ms-OFRMW</sub> - PDRI<sub>bios-OFRMW/biod-FFRMW</sub>)/(PDRI<sub>ms-OFRMW</sub>)] \* 100 in terms of PDRI.



Appendix A. Supplementary material Click here to download E-Component: Supplementary\_material\_OFRMW\_WM\_FIN.pdf