



## Research Paper

# Monitoring of greenhouse gas emissions and compost quality during olive mill waste co-composting at industrial scale: The effect of N and C sources



Ana García-Rández<sup>a</sup>, Luciano Orden<sup>a,b</sup>, Evan A.N. Marks<sup>a</sup>, Javier Andreu-Rodríguez<sup>a</sup>, Samuel Franco-Luesma<sup>c</sup>, Encarnación Martínez-Sabater<sup>a</sup>, José Antonio Saéz-Tovar<sup>a</sup>, María Dolores Pérez-Murcia<sup>a</sup>, Enrique Agulló<sup>a</sup>, María Ángeles Bustamante<sup>a,\*</sup>, Maite Cháfer<sup>d</sup>, Raúl Moral<sup>a</sup>

<sup>a</sup> Centro de Investigación e Innovación Agroalimentaria y Agroambiental (CIAGRO-UMH), Universidad Miguel Hernández, Carretera de Beniel Km 3,2, Orihuela, Alicante 03312, Spain

<sup>b</sup> Dpto. de Agronomía, Universidad Nacional del Sur (UNS), San Andrés 800, Bahía Blanca 8000, Argentina

<sup>c</sup> Dpto. de Suelo y Agua, Estación Experimental Aula Dei (EEAD), Consejo Superior de Investigaciones Científicas (CSIC), 50059, Avda. de Montañana 1005, Zaragoza, Spain

<sup>d</sup> Instituto de Ingeniería de Alimentos Para El Desarrollo, Departamento de Tecnología de Alimentos, Universitat Politècnica de València, Valencia 46022, Spain

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## ABSTRACT

Olive mill wastes (OMW) management by composting allows to obtain valuable fertilizing products, but also implies significant fluxes of greenhouse gases (GHG). For a proper OMW composting, high C- and N co-substrates are necessary, but little is known concerning their effect on GHG emissions in OMW-industrial scale composting. In this study, different co-composting agents (cattle manure (CM), poultry manure (PM), sheep manure (SM) and pig slurry solid fraction (PSSF) as N sources and olive leaves (OLW) and urban pruning residues (UPR) as bulking agents and C sources) were used for OMW composting at industrial scale. Physico-chemical and chemical properties in the composting samples, and GHG (CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O) fluxes were monitored in 12 industrial-scale windrows. GHG emissions were firstly influenced by N source, with the highest accumulated global warming potential (GWP) associated with PM (512 kg CO<sub>2</sub>eq pile<sup>-1</sup>), since PM composts were associated with the greatest N<sub>2</sub>O (0.33 kg pile<sup>-1</sup>) and CH<sub>4</sub> emissions (15.67 kg pile<sup>-1</sup>). Meanwhile, PSSF was associated with the highest CO<sub>2</sub> emissions (1113 kg pile<sup>-1</sup>). UPR as a bulking agent facilitated 10 % greater mineralization of the biomass than OLW, however this C-source was not associated with higher GHG emissions. The results showed that while mineralization dynamics may be impacted by C sources, GHG emissions were mainly conditioned by the characteristics of nutrient-heavy feedstocks (PM and SM). Moreover, manures as nitrogen-laden co-substrates had widely differing effects on total GWP, and that of individual gases, but further research is necessary to understand the mechanisms explaining such differences.

## 1. Introduction

Olive oil production is key to the agro-industrial production in the Mediterranean basin, whose countries produce nearly 90 % of the world's olive oil (Galliou et al., 2018). Spain is the world leader in olive oil production, with 2,768,267 ha of olive orchard land and 5,170,373 t yr<sup>-1</sup> of olive oil (ESYRCE, (2022)). This activity generates large amounts of a pulpy, two-phase olive mill waste (OMW) by-product, called “alperujo.” Due to its high contents of phenols, OMW can be a potential pollutant, phytotoxic and antimicrobial agent (Medina et al., 2011;

Pinho et al., 2017). Thus, this organic waste must be properly managed and valorized to 1) avoid a potential environmental impact derived from an improper disposal 2) and adapt production to a circular economy model through the production of fertilizing materials, namely compost (Carmona et al., 2023).

Composting is the most common organic waste management technique worldwide, including the recycling OMW and transforming it into a fertilizing product (Sánchez-Monedero et al., 2010). Composting has been shown to be an efficient and low-cost technology for OMW treatment, and the composts produced are often used as organic amendments

\* Corresponding author.

E-mail address: [marian.bustamante@umh.es](mailto:marian.bustamante@umh.es) (M. Ángeles Bustamante).

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and fertilizers in agricultural soil (Tortosa et al., 2012; Chowdhury et al., 2013). As a microbiologically-driven process whose effectiveness and efficiency is conditioned by the chemical properties of the feedstocks involved (namely C and N contents), an ideal final C/N ratio lies between 10–20. OMW has very low N contents for an organic waste, requiring the addition of high-N sources, typically animal manures. Also, since OMW has high moisture contents and small particle size, bulking agents must be added to improve air flow during composting, whereas reduced airflow (anoxia) leads to increased emissions of CH<sub>4</sub> and N<sub>2</sub>O, GHG of primary concern.

Given this context, one of the environmental challenges related to composting of these materials is the minimization of GHG emissions during the process. In addition to GWP, GHG emissions are an important pathway for nutrient loss, reducing the fertilizing value of the compost (Jiang et al., 2015). As such, the reduction of these sources of pollution goes in hand with the production of composts with high agronomic value (Hao et al., 2004). Previous studies have shown that between 14 and 51 % of the total organic carbon (TOC) content of feedstock during composting can be released as CH<sub>4</sub> and CO<sub>2</sub> to the atmosphere (Wang et al., 2015; Awasthi, 2023). Carbon dioxide losses account for most of the total C mass loss, while CH<sub>4</sub> emissions can account for less than 10 % (Hao et al., 2004; Mulbry and Ahn, 2014). Similarly, the losses of N contents (based on raw material characteristics) can range from 16 to 74 %, specifically as NH<sub>3</sub> and N<sub>2</sub>O emissions (Yin et al., 2021). It has been found that nitrous oxide losses can represent 10 % of the initial N content (Maeda et al., 2011).

There are many factors that influence GHG emissions during composting, summarized in a systematic review by Pardo et al. (2015) that include: treatment type, treatment duration, climatic conditions (temperature, rainfall) and, especially the starting materials and bulking material and the experimental scale. Thus, different organic wastes have been used as co-composting agents for OMW composting: wool waste and wheat straw (Altieri and Esposito, 2010), olive leaves, wood chips and rice by-products (Komilis and Tziouvaras, 2009), poultry manure (Sellami et al., 2008; Hachicha et al., 2009a), olive leaves (Manios et al., 2006; Alfano et al., 2008; Michailides et al., 2011), and sewage sludge (Sánchez-Arias et al., 2008). Despite the recognition of material effects on GHG, very few studies are available concerning the quantification of GHG emissions and the effect of the co-composting agents used in the composting mixture. Manios et al. (2007) reported a decrease in methane production when increasing the proportion of bulking agent in the initial mixture of OMW composting piles in a preliminary study on CO<sub>2</sub> and CH<sub>4</sub> emissions during composting of two-phase olive oil mill sludge. Sánchez-Monedero et al. (2010, 2011) also studied GHG emissions during composting of two-phase olive mill wastes using different agro-industrial by-products and during four consecutive years, to evaluate the effect of the variability of the raw material characteristics and the performance of the composting plant on GHG emission. These authors reported a clear effect of the composition of the starting mixtures and the OM degradation rate on both CH<sub>4</sub> and N<sub>2</sub>O emissions, as well as the reduction of these emissions with the improvement of the composting operations, such the pile moisture and aeration. However, scarce information is available regarding the effect of the pile composition on GHG emissions at an industrial scale in real conditions.

The primary objective of this work was to monitor CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O emissions during the industrial-scale co-composting of olive mill waste with various manures and bulking agents (C and N sources), and to identify the conditions that minimize GHG emissions and optimize the final compost quality. When proposing composting systems, it is crucial to understand the exothermic behavior of the constituents at the same time understanding potential greenhouse warming impact, and to our knowledge a study at this scale has not been carried out previously. This approach of comparing C and N sources helps manage the complexity and diversity of feedstocks used in composting olive mill waste (and other organic wastes), guiding both environmental impact reduction and the agricultural value of the final compost. We hypothesized that

higher total N in feedstocks would increase N<sub>2</sub>O emissions, while more labile C sources would lead to greater overall GHG emissions. Also, given the high polyphenol contents of the feedstocks, we expected that the concentrations of these microbial activity-inhibiting compounds would be reduced when a more labile C substrate was employed.

## 2. Materials and methods

### 2.1. Experimental design

The composting experiment was carried out at the waste treatment plant of Sant Mateu (Castellón, Spain; 40°28'19.2" N, 0°13' 17.8" W), from March to September 2022. The wastes used in the composting mixtures were plant-based wastes, including olive mill waste (OMW), olive leaf waste (OLW) and urban pruning residues (UPR), and four different manures from nearby farms: poultry manure (PM), cattle manure (CM), pig slurry solid fraction (PSSF) and sheep manure (SM). Physico-chemical properties of the raw materials used in the initial composting mix are shown in Table 1.

Twelve trapezoidal composting piles (7,800 kg, with basal dimensions of 3 m × 6 m and a height of 1.5 m) were prepared with 65 % OMW in all mixtures, with an addition of UPR, OLW, or UPR + OLW (mixed 1:1; w/w) as an additional C source (15 %) and manure as an additional N source (20 %) (Table S1). These mixture proportions were based on maximizing the amount of OMW which may be added so as not to completely impair the composting practice (knowledge based on substantial previous experiences with this organic waste) and the additions of C and N were dimensioned to optimize the C:N of the entire mixture (C:N in the initial mixtures ranged from 18 to 25). The mixtures were prepared to obtain optimal mixes considering final volume, fresh matter and dry matter, based on the moisture and bulk density of each component. The procedure used to prepare and manage the composting piles was the same as that used in the waste treatment plant. Briefly, the formulated ternary composting mixture was mechanically mixed prior to be dispensed in piles using a horizontal feed mixer machine (17 m<sup>3</sup>) with tractor power and later, the piles were managed using a large mechanical windrow turner (Topturn 300, Komptech<sup>TM</sup>). Each pile was turned seven times with a windrow turner machine at 15, 35, 57, 77, 99, 119 and 142 days, and at each turning the piles were watered.

**Table 1**  
Physico-chemical and chemical properties of the raw materials used in composting piles.

	OMW	PM	CM	PSSF	SM	UPR	OLW
Moisture (%)	58.9	34.1	43.1	70.3	64.7	29.4	28.5
BD (g cm <sup>-3</sup> )	0.866	0.294	0.242	0.605	0.466	0.183	0.097
pH	6.7	5.9	8.6	6.9	9.7	7.5	6.1
EC (dS m <sup>-1</sup> )	3.05	7.13	7.69	5.29	13.35	2.71	1.34
OM (%)	92.9	83.9	75.3	72.0	63.5	75.1	79.3
TOC (%)	50.6	38.5	39.4	37.8	36.5	37.1	45.5
TN (%)	0.81	4.00	2.71	2.43	2.25	1.22	1.21
TOC/TN	62.51	9.64	14.53	15.56	16.23	30.31	37.67
PPH (mg kg <sup>-1</sup> )	5757	7402	4760	2029	7158	2492	6295
<i>Total elements</i>							
P (g kg <sup>-1</sup> )	1.44	14.02	8.83	28.64	5.87	1.62	1.06
K (g kg <sup>-1</sup> )	12.62	23.80	25.58	11.08	47.73	10.50	4.06
Ca (g kg <sup>-1</sup> )	0.84	1.71	3.37	3.17	4.34	6.29	5.84
Mg (g kg <sup>-1</sup> )	0.08	0.63	0.69	1.60	0.48	0.50	0.12
Na (g kg <sup>-1</sup> )	0.08	2.59	7.24	2.33	4.18	0.91	0.09
Fe (mg kg <sup>-1</sup> )	421	570	1660	2218	1399	4893	1885
Mn (mg kg <sup>-1</sup> )	23.60	518	281	759	154	154	93.98
Cd (mg kg <sup>-1</sup> )	0.02	0.17	0.11	0.20	0.10	0.20	0.09
Cr (mg kg <sup>-1</sup> )	9.94	5.76	12.77	10.42	8.11	55.30	14.71
Cu (mg kg <sup>-1</sup> )	16.31	77.96	44.60	157	18.80	22.16	134
Ni (mg kg <sup>-1</sup> )	2.70	3.68	4.67	6.44	3.22	10.93	4.43
Pb (mg kg <sup>-1</sup> )	1.53	3.02	3.92	4.15	3.00	28.60	3.42
Zn (mg kg <sup>-1</sup> )	19.93	376	197	1326	116	114	33.93

BD: Bulk Density; EC: electrical conductivity; OM: organic matter; TOC: total organic carbon; TN: total nitrogen; PPH: polyphenols.

Temperature was measured daily at five different points along the pile profile using probes for automatic temperature monitoring. The exothermic index (EXI<sup>2</sup>) was calculated as the quadratic sum of the daily difference between the temperature inside the pile and that in the surrounding environment during the bio-oxidative phase of composting (Vico et al., 2018). The moisture content of the piles was maintained at values from 40–60 % throughout the process by a sprinkler system as necessary. The composting process consisted of a 150-day bio-oxidation phase due to operational restrictions of the composting facility, followed by 30-day maturity phase.

## 2.2. Sampling and analyses

Six samples were taken from the twelve compost piles during the composting process. Four subsamples were taken from different sites on the whole pile profile to make the final sample representative. This sampling was done at 0, 15, 30, 60, 150 and 180 days. In this way, the samples at days 0–150 corresponded to the bio-oxidative phase of composting, while the samples at 180 days corresponded to the maturity phase. All the collected samples were dried (60 °C), ground, and sieved to 0.5 mm prior to be analyzed.

The raw materials and the composting samples were analyzed according to the methods used by Bustamante et al. (2007). Briefly, EC and pH were determined in a 1:10 (w/v) water extract, while water-soluble polyphenols (PPH) were determined by the modified Folin–Ciocalteu method in a 1:20 (w/v) water extract. Total organic carbon (TOC) was assessed by loss-on-ignition at 430 °C for 24 h and total nitrogen (TN) was determined by automatic microanalysis according to Martínez-Sabater et al. (2022). After microwave acid digestion, total contents of P, K, Ca, Mg, Na and heavy metals were analyzed by atomic absorption spectrometry. Losses of organic matter and TN were calculated from the initial (day 0) and final (day 180) ash contents (Paredes et al., 2000).

## 2.3. GHG measurements during composting

The static opaque closed PVC chamber technique was used to measure CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O from the top of the composting piles (Sánchez-Monedero et al., 2010). At the top of each chamber (volume: 0.007 m<sup>3</sup>, area: 0.049 m<sup>2</sup>) a rubber plug with a three-way stopcock was placed in order to take gas samples. Samples were taken at 11 occasions, at: 0, 7, 15, 30, 50, 65, 80, 95, 120, 150 and 180 days. The chambers were inserted 10 cm into the compost piles and air samples were taken at 0, 15 and 30 min using disposable syringes before the operations of turning and watering of the piles. The gas streams were pumped several times before sampling to achieve homogeneous mixing of the air within the chamber space (Marín-Martínez et al., 2021), and a single aliquot was taken from the static chamber. Samples were transferred to 20 ml glass vials (fitted with rubber stoppers) for analysis by gas chromatography (Agilent<sup>TM</sup> 7890B). Measured concentrations of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O were converted to mg gas m<sup>2</sup> day<sup>-1</sup> (Chan et al., 2011). Cumulative gas emissions during the experimental period were calculated by multiplying the average flux of two consecutive determinations by the time elapsed between them, and this amount was added to the previous cumulative total (Menéndez et al., 2006). The calculation of cumulative emissions was made considering the total area (trapezoidal prism pile) of each composting heap (25.6 m<sup>2</sup>). The chambers were removed between sampling to allow turning and watering.

The resulting cumulative fluxes of N<sub>2</sub>O and CH<sub>4</sub> were used to assess the net contribution, which was converted into carbon dioxide equivalents (CO<sub>2</sub>eq) with global warming potential (GWP) values of 273 and 27.2, respectively (IPCC (2021)). CO<sub>2</sub> emissions from the composting process were not included in the GWP as they are considered to be of biogenic origin (Christensen et al., 2009). Therefore, these emissions do not contribute to the GWP as they are assumed to be compensated by the CO<sub>2</sub> production from OM loss, which was considered as part of the short C cycle (IPCC (2006)). The GWP for N<sub>2</sub>O and CH<sub>4</sub> were also calculated

using the emission factors proposed by the IPCC for “Biological treatment of solid waste” (IPCC (2006)). The estimate for the different composting treatments was made on the basis on the fresh weight of each pile.

The economic value of the nutrients in the composts obtained was calculated based on the value of the N, P<sub>2</sub>O<sub>5</sub> and K<sub>2</sub>O fertilizing units of commercial mineral fertilizers urea (46–0–0), triple super phosphate (TSP) (0–46–0) and potassium chloride (KCl) (0–0–60) (Jara-Samaniego et al., 2017). A compost moisture content of 30 % was considered, being this the mean value of the range established by the Spanish normative for compost (RD 999/2017). The mean value of these fertilizers was 350, 418, and 330 € ton<sup>-1</sup> for urea, TSP and KCl, respectively (Agriculture and Rural Development UE, 2023). Thus, the values of the N, P<sub>2</sub>O<sub>5</sub> and K<sub>2</sub>O fertilizing units were estimated to be 760, 900, and 550 € ton<sup>-1</sup>.

## 2.4. Statistical analyses

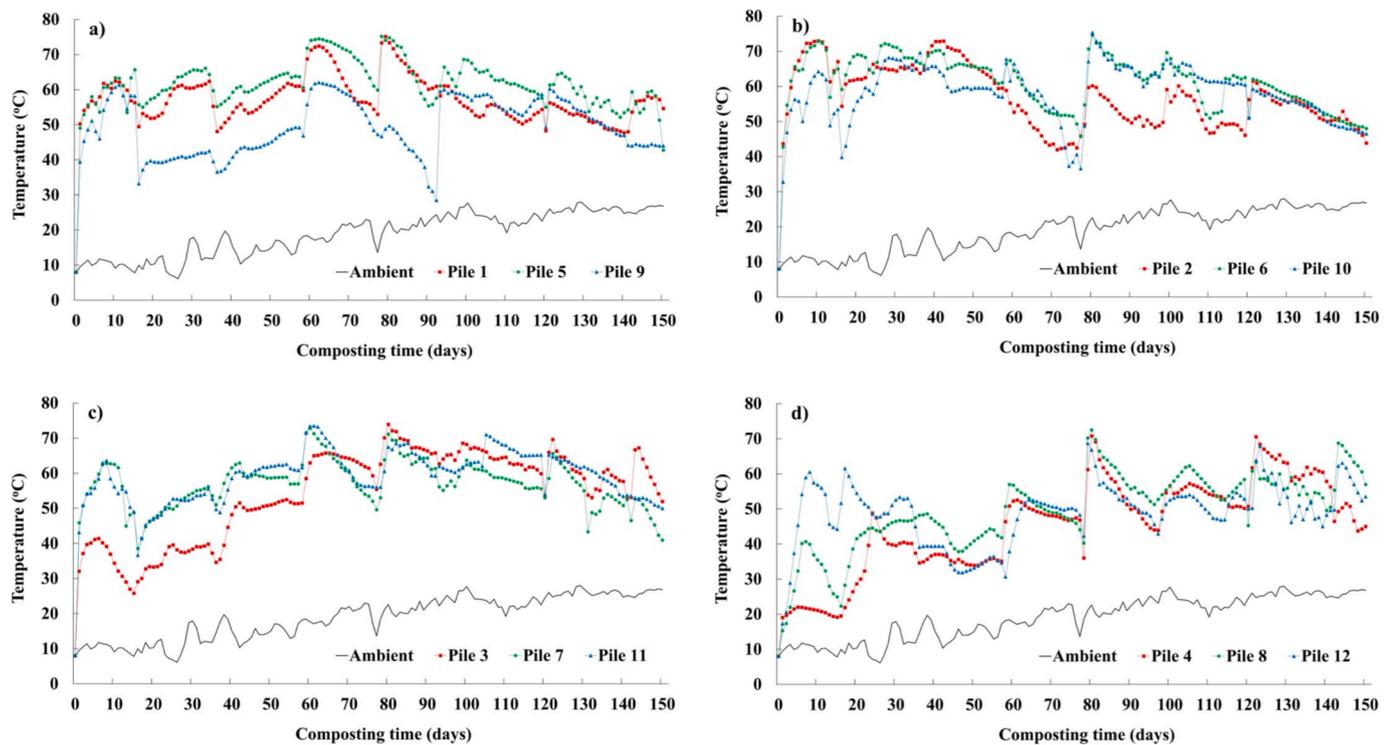
Data analysis was performed using Infostat® (v.2020), a statistical software package linked to the R programming environment (Di Rienzo et al., 2020). One-way analysis of variance (ANOVA) and least significant difference (LSD) test at  $p < 0.05$  were used to assess the statistical significance of differences between values of each parameter (manure and bulking agents) studied during composting. SigmaPlot 13 (Systat Software Inc.) was used to calculate the loss of OM during composting, according to the first-order kinetic function. Adjusted R-squared ( $R^2_{adj}$ ) and  $F$ -values were used to fit the curve to the function and to indicate the statistical significance of the curve fitting. Daily greenhouse gas fluxes were also analyzed via one- and two-way ANOVAs as above. To understand the similarities between final products based on chemical properties,  $k$ -means clustering and hierarchical clustering analysis (HCA) were used. For this, a new dataset was created based on the average values ( $n = 3$ ) for the final products (sampling at 180 days), resulting in one observation per compost. For  $k$ -means clustering, the optimum cluster number was found using the silhouette method (*factoextra::fviz\_nbclust*). Secondly, an agglomerative hierarchical clustering analysis (*stats::hclust*) was applied, in which cluster-observations are joined iteratively based on dissimilarities until only one cluster remains. Here, Ward's squared dissimilarity criterion was used for the clustering algorithm.

## 3. Results

### 3.1. Temperature evolution during the composting process

The composting piles containing PM, CM, and PSSF in the mixtures showed a typical composting temperature trend for these materials (Sánchez-Monedero et al., 2010), reaching thermophilic temperatures (>55 °C) during the first days (up to 15 days) of composting and maintaining the thermophilic phase 120 days in all the mixtures (Fig. 1). All composting piles achieved temperatures > 70 °C, assuring sanitization, and validating the use of these co-substrates. The SM pile reached thermophilic temperatures more slowly (60 days) and had a shorter thermophilic phase than the other piles. In the piles, temperatures slowly decreased until sharp increases immediately after turning (which occurred regularly, approximately every 20 days), which can be attributed to the improved oxygenation and homogenization of the mixture (Valverde-Orozco et al., 2023). As such, the gradually decreasing temperatures beginning a few days after turning are owing to the depletion of oxygen. Examining the temperature profiles (Fig. 1), the lowest-performing piles for N sources PSSF and SM were those with UPR as a C source (piles 3 and 4), and lowest and second lowest for PM were UPR + OLW and UPR (piles 1 and 9). In the case of CM, temperature profiles were quite similar until the maturation phase, where again UPR (pile 2) had the lowest temperatures.

For the summary temperature profile information (Table S2), no differences were observed regarding the maximum temperature (TMax),



**Fig. 1.** Temperature profiles of compost heaps during the process. a) PM + C sources (UPR, OLW, UPR + OLW); b) CM + C sources (UPR, OLW, UPR + OLW); c) PSSF + C sources (UPR, OLW, UPR + OLW); d) SM + C sources (UPR, OLW, UPR + OLW). See [Table S1](#) for pile numbers and acronyms.

but statistically significant differences ( $p < 0.05$ ) were observed in the average temperature (Tave), observing the lowest values for SM among the N sources (48 °C) and the highest for OLW among the C sources (57 °C; [Table S2](#)). The EXI<sup>2</sup> parameter showed that there were significant differences in the exothermic behavior of the piles. The composting mixtures containing CM had the highest EXI<sup>2</sup>, followed by the composting piles made with PM and PSSF, which had intermediate values, and finally the piles with SM had the lowest index. Since the composts made with OLW alone had the highest average temperatures and a more intense and longer exothermic phase, it therefore had a higher EXI<sup>2</sup> index compared to the other C source materials (OLW > UPR + OLW ≥ UPR), probably due to its contents of cellulose and hemicellulose, which are the main components of this bulking agent ([Espeso et al., 2021](#)). For N sources, SM had the lowest EXI<sup>2</sup>, as can be seen from the physico-chemical parameters of the initial characterization.

### 3.2. Physico-chemical and chemical parameters evolution during the composting process

Concerning compost properties, initial pH values in the composting

piles were generally < 6.7, except for the piles with SM whose pH ranged 7.36–8.00. All the piles showed an increase in pH, which was most notable up to the third sampling (30 days), where the pH values ranged from 8.0 to 8.6. During the bio-oxidative phase all the piles experienced a decrease in EC, while in the maturity phase, a reduction in the content of soluble salts for all piles was observed. When comparing material properties ([Table 2](#)), no significant differences in pH and EC were observed in relation to the C source combined with manures. The treatments grouped by N source showed maximum pH and EC values for SM (8.27 and 4.5 dS m<sup>-1</sup> respectively). TOC and TN had significant differences for both C and N sources. SM had the lowest TOC values (41.65 %), while TN was the highest for PM (2.34 %). The C source with the highest TOC values was UPR + OLW and that with highest TN was OLW. Considering C source, TOC/TN was lowest in blends with OLW (21.82 %) and highest with UPR + OLW (24.31 %). Significant differences were found for macronutrient contents in based on the N source used, with the highest concentrations of N in PM, P in PM and PSSF, and K in SM ([Table 2](#)). Similarly, the composts with these types of raw materials also showed the highest NPK contents. The initial OM concentrations in the SM composting piles were the lowest of the N sources,

**Table 2**

Effect of the N and C sources used in the olive oil mill waste composting processes on selected parameters (GLM repeated measures, n = 216, dry matter basis).

		Moisture (%)	BD (g cm <sup>-3</sup> )	pH	EC (dS m <sup>-1</sup> )	OM(%)	TOC(%)	TN(%)	TOC/TN	P (g kg <sup>-1</sup> )	K (g kg <sup>-1</sup> )
N source	PM	45.13	0.56	7.76	3.53b	79.77b	43.55 ab	2.34b	18.76 a	5.63b	18.07c
	CM	43.85	0.55	7.93	3.34 ab	79.90b	44.40b	1.81 a	25.10b	3.65 a	15.76b
	PSSF	46.16	0.54	7.81	2.54 a	80.11b	44.45b	1.78 a	25.86b	5.88b	12.87 a
	SM	48.81	0.62	8.27	4.51c	74.41 a	41.65 a	1.99 a	21.34 a	3.37 a	21.98 d
	<i>F-anova</i>	0.23 ns	0.7 ns	1.3 ns	8.2 ***	6.5 **	3.48 *	11.5 ***	8.98 ***	20.05 ***	35.1 ***
C source	UPR	46.77	0.59	7.99 a	3.47	77.85	42.83 a	1.99b	22.17 a	4.94c	17.79b
	OLW	46.42	0.55	7.92 a	3.55	78.36	43.61b	2.04b	21.82 a	4.70b	17.12 a
	UPR + OLW	44.76	0.56	7.91 a	3.42	79.43	44.09c	1.91 a	24.31b	4.25 a	16.60 a
	<i>F-anova</i>	3.6 ns	3.3 ns	2.5 ns	3.0 ns	5.0 *	67.7 ***	9.7 **	31.2 ***	19.57 ***	11.6 ***

The values shown represent the result of the generalized linear repeated measures model estimate, considering the entire composting period. \*, \*\*, \*\*\*: significant difference between treatments at  $p < 0.01$ ,  $p < 0.001$  and  $p < 0.0001$ , respectively. Different letters within a column indicate significant differences between treatments ( $p < 0.05$ ).

with an average of 79.6 %, compared to averages of approximately 85 % for the remaining N sources. Among C sources, UPR caused the greatest absolute mineralization rates of the piles, with final average OM losses of 61 %, as compared to 50.1 % for OLW or 49.5 % for OLW + UPR.

Fig. 2 shows the curve fitting with the kinetic model used for OM losses. OM losses were generally greatest up to 100 days, whereas the highest temperatures were generally achieved between 60 and 100 days (Fig. 1), which is usual for a composting process. The lowest OM mineralization was observed between 150–180 days (maturity phase), indicating stability. Thus, the OM model during composting followed a first-order kinetic equation:  $OM_{loss} = A(1 - e^{-kt})$ . Table S3 shows the curve fitting for the model parameter values for each treatment. All equations were significant at  $p < 0.001$ . The C sources did not show significant differences for the parameters  $A$  (maximum mineralization) and  $k$  (rate constant). On the other hand, for the N sources, significant differences were found for the parameter  $A$ , following  $SM > PSSF \geq CM = PM$ . Fig. 2 shows the dynamics of OM mineralization for N sources during the composting process adapted to the proposed equation. Whereas the parameter  $A$  is indicative of the maximum rates of mineralization, based on the kinetic model and the differences observed in the parameter  $A$  between the different N sources, there was a more progressive evolution of OM for the SM composts. Also, though not significant in the statistical test, comparing C sources the highest parameter  $A$  is observed in the mixtures with UPR.

### 3.3. Greenhouse gas emissions during composting

The  $N_2O$  fluxes from the compost piles were low at the beginning of the experiment for most treatments, with the exception of PM composts (Fig. 3a and b). The greatest peaks in  $N_2O$  emissions were observed

between days 65 and 120, and the highest  $N_2O$  emissions were observed in PM composts ( $160 \text{ mg } N_2O-N \text{ m}^{-2} \text{ day}^{-1}$ ; Fig. 3 a), while OLW was the C source with the highest  $N_2O$  peak at 120 days (Fig. 3 b). Methane fluxes ranged between 0 to  $10 \text{ g } CH_4-C \text{ m}^{-2} \text{ day}^{-1}$  for all treatments, the PM and UPR composts having the greatest  $CH_4$  fluxes. Carbon dioxide fluxes ranged between 30 to  $120 \text{ kg } CO_2-C \text{ m}^{-2} \text{ day}^{-1}$  from the beginning of the composting process until day 65. Two major emission peaks were observed on days 80 and 120, reaching maximum  $CO_2$  fluxes values of  $270 \text{ kg } CO_2-C \text{ m}^{-2} \text{ day}^{-1}$  for the PM treatment (Fig. 3 e).

Table 3 shows the cumulative GHG emissions and GWP estimates for the different compost blends. The N sources induced significant differences ( $p < 0.05$ ) on total GHG emissions, while the C sources did not (Table 3). The greatest GWP was associated with PM composts ( $512 \text{ kg } CO_2eq \text{ pile}^{-1}$ ), followed by SM composts ( $437 \text{ kg } CO_2eq \text{ pile}^{-1}$ ). Composts with PM caused the highest cumulative  $N_2O$  emissions ( $0.33 \text{ kg } \text{pile}^{-1}$ ) as well as  $CH_4$  cumulative emissions ( $15.6 \text{ kg } \text{pile}^{-1}$ ). On the other hand, CM composts provoked the least overall GWP. Behavior of CM and PSSF was generally similar except in terms of  $CH_4$  emissions, which were much greater in PSSF. PSSF composts had the highest cumulative  $CO_2$  emissions of all the N sources ( $1113 \text{ kg } \text{pile}^{-1}$ ; Table 3), but this gas contributes relatively little to overall GWP.

### 3.4. Characteristics of the final composts

At the end of composting (180 days), the final composts had significant differences in the physico-chemical and chemical properties depending on the N source used (Table 4). The pH of the N-source composts was slightly alkaline with values between 8.3–8.6, with relatively high electrical conductivity (around 2–3  $\text{dS m}^{-1}$ ), high TN (2 %) and low TOC/TN ( $< 20$ ), except for PSSF, which did not reach optimal

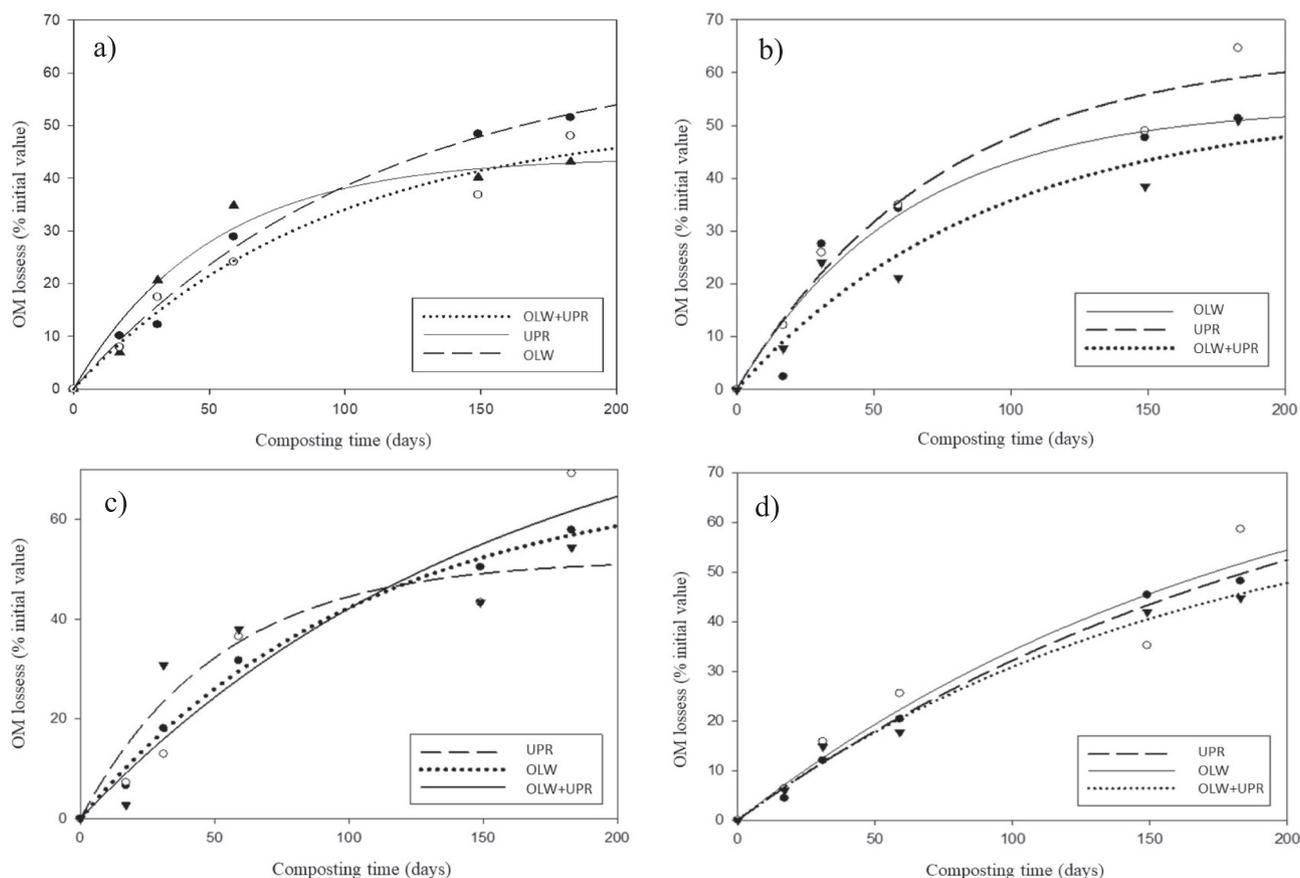


Fig. 2. Organic matter (OM) losses during heap composting by N. Source: a) PM; b) CM; c) PSSF; d) SM

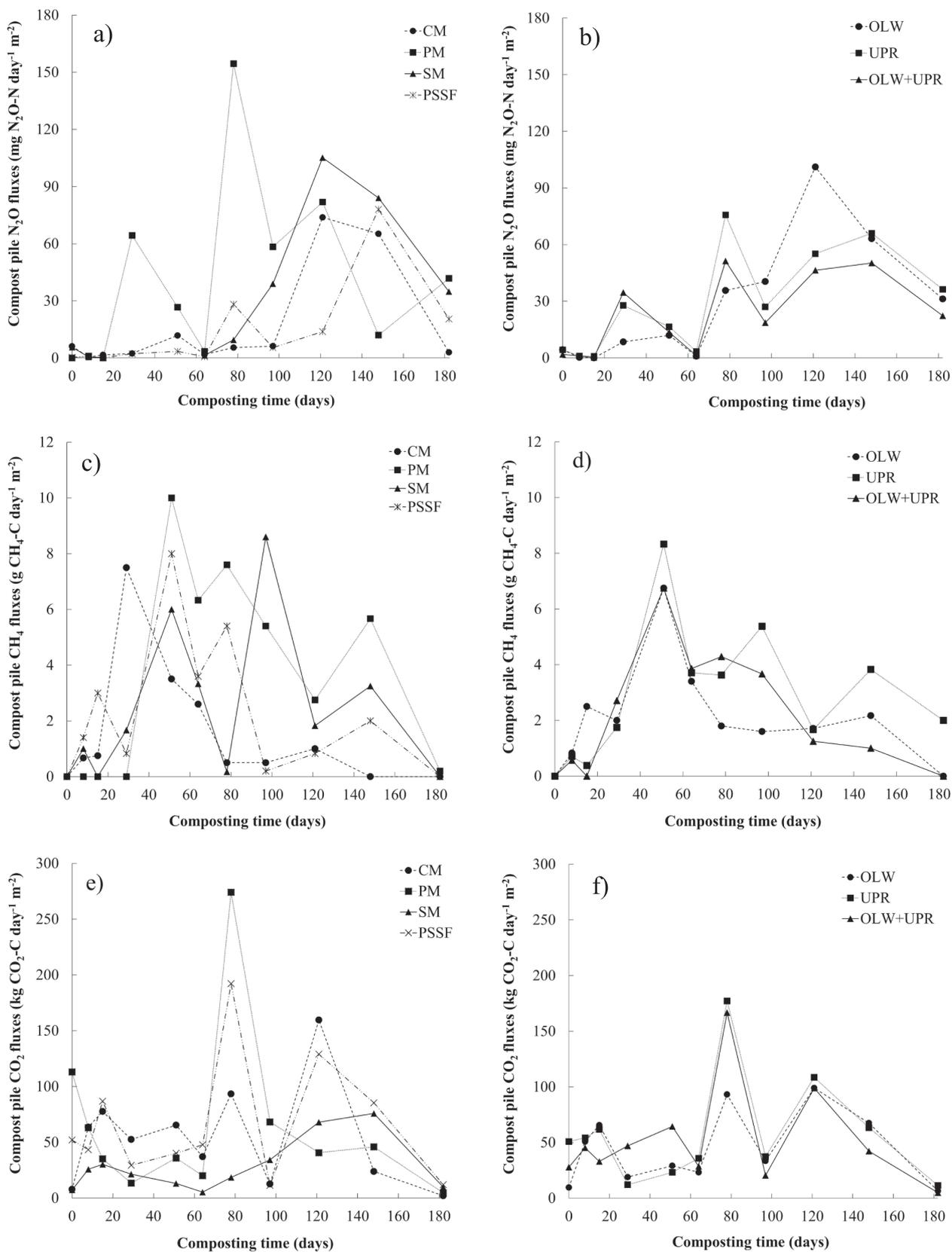


Fig. 3. Daily fluxes GHG emissions measured during composting over the 180-day measurement period. In these graphs, fluxes are expressed as elemental contents of each gas (N, C).

**Table 3**  
Cumulative GHG emissions during composting.

		kg N <sub>2</sub> O pile <sup>-1</sup>	kg CO <sub>2</sub> eq N <sub>2</sub> O pile <sup>-1</sup>	kg CH <sub>4</sub> pile <sup>-1</sup>	kg CO <sub>2</sub> eq CH <sub>4</sub> pile <sup>-1</sup>	kg CO <sub>2</sub> pile <sup>-1</sup>	GWP (kg CO <sub>2</sub> eq pile <sup>-1</sup> )
N source	PM	0.33c	88.33c	15.67b	423b	834 ab	512b
	CM	0.18 ab	48.50 ab	7.17 a	192 a	910b	240 a
	PSSF	0.16 a	43.33 a	11.50 ab	307 ab	1113b	351 ab
	SM	0.28 bc	75.50 bc	13.17 ab	361 ab	574 a	437 ab
	F-value	5.21 **	4.95 **	2.16 ns	2.32 ns	5.19 **	2.97 *
C source	UPR	0.25	69	14.88	402	921	471
	OLW	0.26	70.38	9.38	256	776	326
	UPR + OLW	0.19	52.38	11.38	306	876	359
	F-value	0.89 ns	0.94 ns	1.59 ns	1.56 ns	0.47 ns	1.36 ns

GWP: CO<sub>2</sub> equivalent from added N<sub>2</sub>O and CH<sub>4</sub> emissions (IPCC, 2021). \*, \*\*: significant difference between treatments at  $p < 0.01$  and  $p < 0.001$ , respectively. ns = no significant. Different letters within a column indicate significant differences between treatments ( $p < 0.05$ ).

**Table 4**  
Main characteristics of the mature compost (dry weight basis) grouped by N and C source, and corresponding statistical tests by group.

	PM	CM	PSSF	SM	F-value	OLW	UPR	OLW + UPR	F-value
<i>Physico-chemical</i>									
pH	8.45b	8.49 bc	8.26 a	8.60c	8 ***	8.52	8.40	8.43	1 ns
EC (dS m <sup>-1</sup> )	2.81b	2.56b	1.82 a	3.43c	38 ***	2.62	2.80	2.55	0 ns
TOC (%)	40.18 bc	39.36b	40.73c	36.20 a	26 ***	39.03 ab	38.15 a	40.18b	3 *
TOC/TN	17.73 a	18.67 a	25.36b	16.22 a	15 ***	18.75	17.80	21.93	2 ns
<i>Macroelements</i>									
TN (%)	2.33c	2.11b	1.65 a	2.24 bc	17 ***	2.11	2.19	1.94	1 ns
P (g kg <sup>-1</sup> )	7.82c	5.16 a	6.20b	4.76 a	15 ***	6.00	6.44	5.51	1 ns
K (g kg <sup>-1</sup> )	19.78c	17.43b	12.74 a	24.87 d	47 ***	18.74	18.95	18.43	0 ns
Na (g kg <sup>-1</sup> )	1.01b	1.83 d	0.59 a	1.33c	97 ***	1.31	1.16	1.11	0 ns
Ca (g kg <sup>-1</sup> )	4.14 ab	4.56b	3.97 a	6.46c	47 ***	4.86	5.00	4.50	0 ns
Mg (g kg <sup>-1</sup> )	0.51	0.47	0.46	0.44	0 ns	0.46	0.55	0.39	12 ***
<i>Microelements</i>									
Fe (mg kg <sup>-1</sup> )	2199 a	2804b	1995 a	3612c	15 ***	2479	3300	2179	8 **
Cu (mg kg <sup>-1</sup> )	92.78	77.44	80.56	75.89	1 ns	99b	72 a	73 a	16 ***
Mn (mg kg <sup>-1</sup> )	316c	162 ab	204b	143 a	20 ***	215	233	171	1 ns
Zn (mg kg <sup>-1</sup> )	232b	140 a	291c	104 a	25 ***	196	226	153	2 ns
<i>Maturity indices</i>									
PPH (mg kg <sup>-1</sup> )	2261c	1806b	1347 a	1901b	18 ***	2076b	1718 a	1691 a	3 *
Chum (%)	7.44c	6.03b	3.88 a	6.17b	23 ***	6.43b	6.19 ab	5.02 a	3 *
Cfulv (%)	2.10b	2.23b	1.88 a	2.30b	6 **	2.22b	2.22b	1.95 a	4 *
GI (%)	92b	81 a	93b	84 a	6 ***	90b	90b	82 a	4 *

Values shown are the averages for each measurement of the final compost (180 days) grouped by N-source and C-source. EC: electrical conductivity; OM: organic matter; TOC: total organic C; TN: total N; TOC/TN: total organic C/total N; PPH: polyphenols; Chum: humic acid-like carbon; Cfulv: fulvic acid-like carbon; GI: germination index. Associated with the reported F-value is the statistical significance: \*, \*\*, \*\*\*: significant difference between treatments at  $p < 0.01$ ,  $p < 0.001$  and  $p < 0.0001$ , respectively. ns = no significant. Different letters within a column indicate significant differences between treatments ( $p < 0.05$ ).

values, had abundant total P (around 5–8 g kg<sup>-1</sup>) and high values of total K (13–25 g kg<sup>-1</sup>). The GI values (%) were well differentiated by material, with 92 and 93 % for PM and PSSF composts, 81 and 84 % for CM and SM composts. Regarding other elements, no significant differences were observed for Mg, while for Ca and S, the SM had the highest concentrations (6.5 and 0.4 g kg<sup>-1</sup>, respectively). The analysis the composts based on C source (UPR, OLW, UPR + OLW) did not show significant differences in pH, EC, TN, TOC/TN, P or K (Table 4). Polyphenol contents showed differences both based on N and C source, in part reflecting original contents, mostly owing to manure properties (Table 1). However, it was seen that CM-based compost contents of polyphenols were actually relatively similar to those in PM and SM, despite the fact that these other feedstocks had much higher initial concentrations (Table 1). Also, concentrations of polyphenols in OLW composts were higher than those in UPR composts.

The *k*-means clustering analysis resulted in the definition of two principal groups: one cluster consisted of the SM-based composts and the PM and CM composts with UPR (cluster 1), while the second cluster (cluster 2) included all three PSSF composts, CM-OLW and CM-OLW + UPR, and PM-OLW PM-OLW + UPR. Based on the average values of the chemical parameters in each cluster, cluster 1 was generally characterized by higher nutrient contents, while cluster 2 was characterized by slightly higher organic matter contents and lower contents of nutrients and (Figure S1). The dendrogram analysis resulted in closeness (low

Euclidean distances) among composts of the same N source (Fig. 4). The exception was for PM-OLW + UPR, which was grouped adjacent to CM and PSSF composts. The greatest distances were found comparing SM and PSSF composts (Fig. 4).

In terms of economic value, the calculated values per nutrient are shown in Table S4. Based on the current market prices (which have fluctuated quite heavily in recent times due to world events), the nitrogen contents are generally of the highest value in the composts grouped by N-source, ranging from 12 to 17.7 € ton compost<sup>-1</sup>, followed by P<sub>2</sub>O<sub>5</sub>, and lastly K<sub>2</sub>O. Among the N-source composts, the range of nutrient value was not large, from 34 to 47 € ton compost<sup>-1</sup>.

## 4. Discussion

### 4.1. Feedstock and mixture properties influencing organic matter dynamics

As seen in Fig. 1, UPR composts generally promoted lower temperatures, either at the beginning or end of the composting process. However, thermal behavior of UPR depended on the particular N source used. For instance, effects in CM were less evident at the beginning of the process (probably due to its high OM contents and/or structural properties), with lower temperatures toward the end, while in the low-OM SM composts, temperatures were lower in the beginning, and

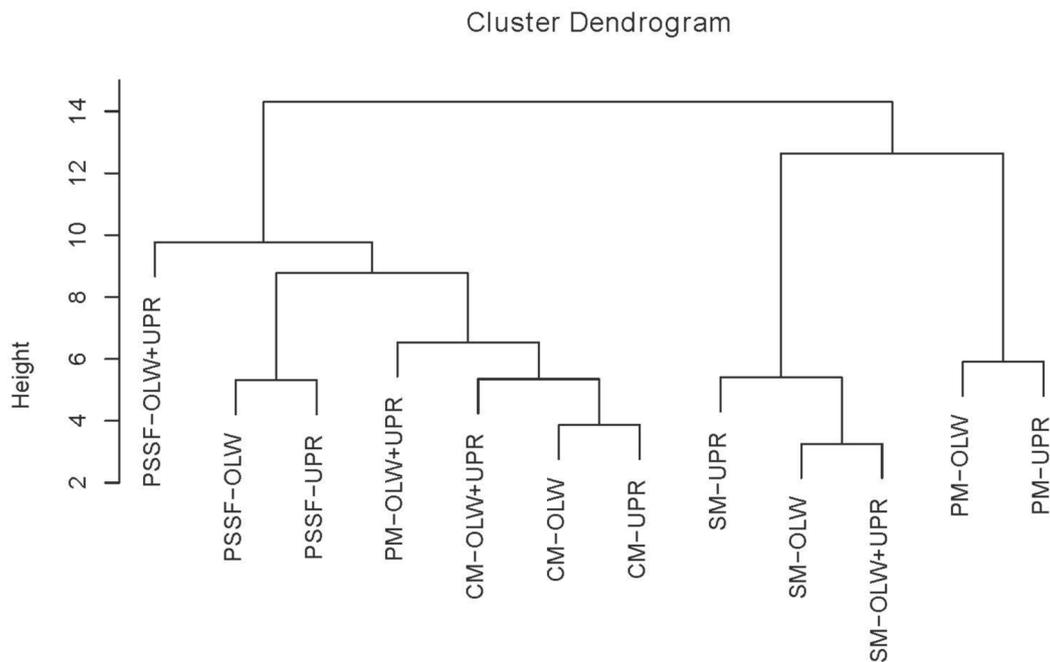


Fig. 4. Relationships between quality indicators and compost.

recovered towards the end, being indistinguishable from OLW. Interestingly, we have seen that UPR, as opposed to OLW, promoted greater absolute mineralization by experiment end. Pile temperature differences with UPR were more visible at the beginning of the composting process, with the exception of CM, which as stated previously may be due to its structural properties and particular composition of OM. Greater degradability of OLW likely promoted greater temperatures at the beginning, owing to chemical properties. However, it is possible that composting conditions may have been better throughout the experiment with UPR owing to structural properties (not chemical composition). PM and SM, with low OM contents and high mineral contents, exhibited mineralization dynamics which were characterized by slow increases (Fig. 2), likely owing to the low degradability. However, SM total mineralization rates were the highest (with highest estimated maximum mineralization of 87.2; Table S3), owing to the low C/N of this substrate (Table 2). Temperature increases at outset, highest in CM, in some cases reflect the particular N co-substrate, but we interpret that the total mineralization was more impacted by mixture C/N and the C source used (higher mineralization with UPR).

Putting the experiment into greater context, in general the losses of organic matter were similar to those reported by other experiments with *alperujo* OMW (50–60 % loss versus 55–68 % reported by García-Gomez et al. 2003). OM losses are known to be strongly influenced by shifting C/N ratios, which are key for compost degradability. For PM, CM and SM composts, the C/N ratios decreased by approximately 50 % during the composting process, which is similar to the results reported by Chowdhury et al. (2013), also using OMW. However, the PSSF compost showed a final ratio of 25.

#### 4.2. Greenhouse gas emissions

Considering cumulative GHG emissions (Table 3), only N source was associated with significant differences between the treatments. The highest cumulative N<sub>2</sub>O and CH<sub>4</sub> emissions were observed for PM and SM. N<sub>2</sub>O is the most potent GHG, with a very large effect on final GWP. However, CH<sub>4</sub> was the gas with the greatest total impact on GWP in this experiment due to large total emissions, of between 7–15 kg pile<sup>-1</sup> (depending on substrate; Table 3). Aeration, pile turning, and moisture management are known to be among the most important factors

influencing GHG emissions of manures (Liu et al. 2023), and despite regular and equal turning regimes in the experiment, the different physical characteristics of the materials likely impacted aeration. Gas peaks were likely influenced by the turning operations (every 20 days), however due to non-coincidence of gas sampling with pile turning (owing to the experimental design), it is difficult to discern exactly what influence these operations had.

N<sub>2</sub>O emissions are known to reflect N availabilities (Butterbach-Bahl et al., 2013). PM and SM were the materials with the lowest TOC/TN ratios and highest N<sub>2</sub>O emissions. PM is a very labile manure with N contents which are mainly in inorganic form (Rizzo et al., 2022). On the other hand, the lowest N<sub>2</sub>O emissions were observed for the PSSF treatment, which had the highest TOC/TN ratio (Table S3). N<sub>2</sub>O peaks are also known to be associated with the turnings due to the aeration of the piles, favoring N<sub>2</sub>O emissions first through nitrification and later denitrification (Maeda et al. 2011; Arriaga et al. 2017). Observing the N<sub>2</sub>O fluxes (Fig. 3), it seems that N<sub>2</sub>O peaks were greatest with PM during the bio-oxidative phase, and greatest with SM during the maturation phase. As such, N<sub>2</sub>O emissions may also reflect differences in material properties – either physical or chemical – which determine nitrogen dynamics during the composting process.

Carbon dioxide fluxes from the different compost piles showed fluxes of between 30 and 120 kg CO<sub>2</sub>-C m<sup>-2</sup> day<sup>-1</sup>, including two emission peaks on days 65 and 120, probably related to the turning operation, favoring the oxidation of the organic matter by microorganisms (Ahn et al., 2011; Arriaga et al., 2017). Periodic turning modifies the spatial and temporal distribution of O<sub>2</sub>, and aerobic microbial activity can oxidize a large proportion of the CH<sub>4</sub> to CO<sub>2</sub> before it is released to the atmosphere (Jäckel et al., 2005). Though cumulative CO<sub>2</sub> emissions were highest in PSSF, this had low overall impact on GWP.

Methane fluxes were characterized by increases in emissions after day 30 and a decrease after day 85. This pattern could be explained by the temperature of the piles, which varied between 30 and 50 °C between these dates, as these temperatures are considered the optimal temperature range for methane production (Arriaga et al., 2017; Sánchez-Monedero et al., 2010; Sánchez-Monedero et al. 2011). In addition, the mixing process can result in different CH<sub>4</sub> peaks, which could be related to the diffusion of CH<sub>4</sub> previously produced in the piles (Ahn et al., 2011). The cumulative CH<sub>4</sub> emissions from the piles were

greatest in the treatments with low C/N ratios, those with PM and CM as N sources. These greater emissions under PM and SM could be related to the inhibition of the methanotrophic activity by ammonium, which replaces CH<sub>4</sub> as substrate in the methanotrophic pathway (King and Schnell, 1994; Mancinelli, 1995).

Overall, the GWP results generally mirrored observations for individual N<sub>2</sub>O and CH<sub>4</sub> emissions, whereas the treatments with the highest N<sub>2</sub>O and CH<sub>4</sub> emissions were also the treatments with the highest CO<sub>2</sub>eq emissions. Within the N sources, the treatment with the highest CO<sub>2</sub>eq emissions was PM, which had the lowest C/N ratio, and also had the highest N<sub>2</sub>O emissions. It has been found previously that composts with low C/N ratios usually result in high greenhouse gas emissions (Sánchez-García et al., 2015).

The two tested bulking agents did not show strong differences in their influence on GHG. However, the potential importance of bulking agents should not be disregarded, as it is known that improving the structure of the heaps increases O<sub>2</sub> diffusion, with consequences for GHG production as described for the gases above. Pardo et al. (2015) also reported a reduction in GHG emissions during composting by increasing the proportion of bulking agents. Using the average of the emissions data from the 12 compost heaps of the experiment, we also carried out a theoretical calculation of the GWP (emission factors IPCC (2006)). This exercise resulted in a result of 1,500 kg CO<sub>2</sub>eq pile<sup>-1</sup>, compared to 300 kg CO<sub>2</sub>eq pile<sup>-1</sup> based on the actual data, a significant difference. However, it should be recognized that the IPCC guidelines do not distinguish between different composting methods (e.g. windrow composting, in-vessel composting, heated passive piles). A future objective of research supporting new IPCC standards would be to establish specific emission factors for different composting methods. In this way, for each process type, poorly managed composting processes can be more easily identified and excessive emissions diagnosed.

#### 4.3. Characteristics of the final compost

The cluster analysis resulted in the separation of materials which had high nutrient contents – with high EC, and low OM – and those with high OM matter contents. The analysis grouped SM and OLW composts, since plant leaves generally have higher concentrations of non-volatile elements and nutrients than woody parts. As such, the *k*-means cluster analysis was particularly sensitive to C-source. The dendrogram also reflected this dichotomy between nutrient and OM contents, but was more sensitive to N source: as seen in Fig. 4, the two main branches show a close association between PM and SM on one hand, and PSSF and CM on the other. Thus, the unsupervised grouping exercises coincided on the importance of organic matter and nutrients, however they differed on whether this was attributed to C or N source.

Concerning the particular chemical characteristics, among all the composts there were only small absolute differences in the range of pH values (8.2–8.7), total nitrogen (1.3–2.6 %), or total organic carbon (35.1–42.4 %). More differences were notable in the measured ranges of C/N (14.7–31.8), EC (1.5–3.9 dS m<sup>-1</sup>), ash contents (24.0–38.2 %), total K (10.5–27.5 g kg<sup>-1</sup>) and total P (4.2–9.3 g kg<sup>-1</sup>). From the process and maturity standpoint, the pH values of the final composts were optimal. According to de Bertoldi et al., (1983), pH values < 9 in the final stages of composting signify that the compost is mature, while different authors have stated that pH values within the range of 6.0–8.5 as adequate for the use of compost in agriculture (Bustamante et al., 2013; Hogg et al., 2002). Furthermore, feedstock did not significantly impact this parameter in a manner which would differentiate the materials. The N-source feedstocks are those which had the greatest influence on EC, whereas SM had the highest EC values, and PSSF the lowest, since EC reflects dissolved salts content and PSSF is a dewatered by-product. All composts obtained had values below 3.5 dS m<sup>-1</sup>, whereas EC > 4 dS m<sup>-1</sup> is considered to be potentially inhibiting for plant growth (Luo et al., 2018; Chang et al., 2023; Manu et al., 2019) and application to soil should be limited for salt-sensitive crops, especially if EC exceeds 5 dS m<sup>-1</sup>

(Albrecht, 2007). Product C/N affects both the process and the final quality of the compost (Zhu N., 2007). The total OM (%) of the obtained composts verified the specific EU standard for OLW compost OM > 45 % (RD 506/2013), and C/N was both adequate and quite similar among the composts, with the exception of PSSF, which had the lowest TN concentrations in the final product. This could be attributed to the lower N in this material (2.43 %) which could be easily solved by using a high proportion of animal manure as a co-composting material (Cegarra et al., 2000). Adequate C/N assures microbial degradation of organic matter, leading to the production of the so-called “humic substances,” associated with a high abundance of humic acids and fulvic acids, whereas highly humified OM increases the agricultural value of compost (Bernal et al., 2009).

According to the EU regulation regarding the content of heavy metals in OMW composts, the composts could be classified as “Type B” due to their Cu contents in CM, SM, and PM composts, and due to Zn contents in the PSSF composts. Foliar applications of Cu for disease control in olive trees may explain high Cu levels. The values of Zn in PSSF can be explained by its widespread use on pig farms to control diarrhea at weaning and to promote growth in piglets. However, as far as EU fertilizing product regulations are concerned (2019/1009), since the composts had acceptable organic matter contents and acceptable levels of heavy metals (and assuming removal of any pathogens), they would be suitable for marketing as organic amendments.

OMW composts produced with PM and CM typically have high nutrient contents (Ameziane et al., 2020) and accordingly their economic value was highest. According to the EU fertilizing product regulations cited above, for a fertilizing product to be sold as commercial fertilizer, one of the following conditions must be met: N contents equal or greater than 2.5 %, P<sub>2</sub>O<sub>5</sub> > 2 %, or K<sub>2</sub>O > 2 %. As seen in Table 4, the total N contents of the composts did not meet the N requirements in any case (grouped either by N source or C source), whereas the highest average N contents were found in PM composts (2.32 %). P<sub>2</sub>O<sub>5</sub> contents did not achieve the stated minimum, ranging from 1.09 % (sheep manure) to 1.79 % (poultry manure). Concerning K<sub>2</sub>O, only SM achieved sufficient concentrations, with 3.00 %. Overall, though the composts did not contain very high nutrient contents, the products could be considered as soil improvers or amendments with capacities to complement nutrient demands. Based on current market prices for fertilizing units, the composts had values which were above the Spanish median price of organic amendment composts (approximately 30 € t compost<sup>-1</sup>). Therefore, the calculated compost nutrient values – reaching 47 € t compost<sup>-1</sup> – could be competitively sold, depending on the costs of production (not considered in this study) and final selling price.

According to the EU standards (RD 503/2013), all the composts had PPH values below the maximum allowed concentration (< 0.8 %). The highest PPH concentrations were found in composts made with PM and SM, which had high initial concentrations in the raw materials. However, it was notable that final PPH concentrations in CM composts were similar to those in PM and SM, which probably indicates that polyphenols were relatively less degradable in this manure. Also, contrary to initial hypothesis, the composts based on OLW actually had higher PPH contents than mixtures with UPR. With this result, and considering the dynamics of temperature and organic matter mineralization described before, it seems that not the liability of the substrate, but rather the maintenance of microbial activities over the composting period (as described in section 3.2 above) may have been more crucial for the degradation of PPH. None of the composts obtained showed a phytotoxic effect, with GI values between 93–81 %, which is higher than that obtained by other authors for OMW composts (Cegarra et al., 2000; Baeta-Hall et al., 2005). It is generally considered that GI values above 80 % reflect a non-phytotoxic effect of mature composts (Hachicha et al., 2009b; Bargougui et al., 2020).

## 5. Conclusions

In this study, it is shown the feasibility of using multiple N sources and C sources to effectively manage OMW. As originally hypothesized, the results have shown that environmental sustainability of the composting process and environmental technology is highly impacted by the raw materials employed. The type of material impacts the dynamics of temperature, organic matter mineralization, and finally GHG emissions. In particular, the type and composition of the N-source raw materials were the main factors that influenced GHG emissions in OMW industrial co-composting using windrow composting systems. Furthermore, bulking agents had an important effect on final quality and also in the successful development of the composting process, intervening in the maintenance of microbial activities, and influencing the degradation of phytotoxic compounds. The results have shown that the high nutrient N sources generally have greater GWP impacts. However, further research concerning specific characteristics of the feedstocks (e.g. chemical characterization of recalcitrant compounds such as lignin, cellulose, etc.) and/or detailed enzymatic assays or microbiological community analyses uncover predominant biochemical processes in each case is necessary to explain this behaviour.

### CRedit authorship contribution statement

**Ana García Rández:** Writing – original draft, Validation, Resources, Methodology, Investigation. **Luciano Orden:** Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis. **Evan A.N. Marks:** Writing – review & editing, Writing – original draft, Visualization, Software, Conceptualization. **Javier Andreu-Rodríguez:** Validation, Resources, Methodology, Investigation. **Samuel Franco-Luesma:** Software, Formal analysis, Data curation. **Encarnación Martínez Sabater:** Validation, Resources, Investigation. **José Antonio Saéz-Tovar:** Writing – review & editing, Software, Resources, Investigation, Formal analysis. **María Dolores Pérez-Murcia:** Visualization, Resources, Methodology, Data curation. **Enrique Agulló:** Validation, Resources, Investigation. **María Ángeles Bustamante:** Writing – review & editing, Validation, Methodology, Conceptualization. **Maite Cháfer:** Validation, Supervision, Resources, Methodology. **Raúl Moral:** Writing – review & editing, Visualization, Project administration, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization.

### 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.

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### Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2024.11.039>.

## Data availability

Data will be made available on request.

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