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# Research article

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# Suitability of renewable organic materials for the synthesis of organo-mineral fertilizers: Driving factors and replacement of peat

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#### ABSTRACT

In organo-mineral fertilizers (OMFs) with low organic carbon ( $C_{org}$ ) final content, the organic fraction enhances the mineral fraction efficiency. Therefore, a high-quality organic fraction is crucial. While geogenic materials like peat have been used extensively for producing high-quality OMFs, exploring alternative organic sources such as biowastes can add circular value to these fertilizers. However, since biowastes vary significantly based on origin, processing, season, or collection area, each material must be analyzed separately for suitability in OMFs. We propose a set of physicochemical parameters impacting OMF formulation, manufacture, and potential use to facilitate this analysis.

Our study involved the collection of 16 organic materials across Italy, categorizing them into geogenic materials (peat and leonardite), wood biochar (BC), green compost (GC), farmyard manure compost (MC), municipal solid waste compost (MSWC), and vermicompost (VC). After characterization, we analyzed the contribution of each organic material to an OMF with 7.5 %  $C_{org}$ , in which a low amount of nutrients derives from the organic material. Most parameters showed high variability among groups; no material matched peat and leonardite across all parameters. However, the  $C_{org}$  stability in composted biowastes was generally acceptable for OMF use. Granulometry (>5 mm), pH (>8), and formula space (>90 %) obligate blending with another organic fraction, while P and K in the raw material are insignificant for low  $C_{org}$  OMFs. Most examined materials had potential for OMF production, though adjustments are necessary to enhance their quality. Based on the proposed parameters, MSWC and VC samples stood out as potential high-quality organic matrices for OMF production, offering a promising alternative to peat. The prospect of replacing peat in OMF manufacturing with biowastes holds promise, mainly when industries can search for local substitutes.

# 1. Introduction

An Organo-Mineral Fertilizer (OMF) is a fertilizer resulting from combining inorganic fertilizers (MFs) with high nutrient contents with organic matrices or soil improvers (OMs) [1,2,3]. Based on the current European regulation for fertilizers [1], OMFs are a mixture

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List of a	bbreviations
В	Boron
С	Carbon
Са	Calcium
Cd	Cadmium
Corg	Organic carbon
Cr	Chromium
Cu	Copper
DM	Dry matter
FA	Fulvic acid
Fe	Iron
GC	Green compost
HA	Humic acid
HD	Humification degree
К	Potassium
K <sub>2</sub> O	Potassium oxide
MC	Manure mixed compost
MF	Mineral fertilizers
Mg	Magnesium
Mn	Manganese
MSWC	Municipal solid waste compost
Ν	Nitrogen
Ni	Nickel
N-NH <sub>4</sub>	Ammonium N
N-NO <sub>3</sub>	Nitrate N
Norg	Organic nitrogen
OF	Organic fraction
OFprop:	
OMF	Organo-mineral fertilizer
Р	Phosphorus
$P_2O_5$	Phosphorus pentoxide
Pb	Lead
PTE	Potentially toxic elements
S	Sulfur
TEC	Total extractable carbon
VC	Vermicompost
Zn	Zinc

of one or more organic materials with one or more mineral fertilizers that require a minimum of 3 or 7.5 % organic C (C<sub>org</sub>) if it is liquid or solid, respectively. The potential benefits will depend on the quality of the organic material, its proportion in the OMF, and physical and chemical interactions between the organic and mineral fractions [4]. Therefore, selecting appropriate organic materials for OMFs is a key point in OMF production because their nature will influence the final quality and the sustainability of the OMF. Among the general considerations also organic material availability, cost, transportation have to be included. With a local, abundant, and cheap organic material, economic and energetic sustainability of OMFs would increase [5].

The European law [1] sets chemical and microbiological limitations to the final OMF composition, but not to the organic matrices that can be used in its formulation. However, organic materials require microbiological, physical, and chemical analyses to ensure the material can be used in granular or pelletized OMFs, fostering beneficial interactions with the mineral fraction. The physical aspects, including particle size distribution and moisture content, for their impact in the granulation or pelletization process of the OMF [6]. Chemical aspects, such as the quality and content of organic carbon ( $C_{org}$ ) and nutrients, are crucial in defining the proportions of organic material and mineral fertilizer required for the OMF and its stability [4,7]. Additionally, the content of potentially toxic elements (PTE) [8], salts [9], and the pH [10] of the organic material can serve as limiting parameters for OMF production since high values can limit crop growth and health.

In addition, organic material should be chemically and biochemically stable to produce a homogeneous and storable final product and to maintain constant interactions with the mineral component over time [3,11]. An organic material can be considered stable if it is resistant to fast mineralization [12] and undergoes slow degradation and consequent nutrient release [13].

Traditionally, organic geogenic materials, such as peat and leonardite, have been used in OMF production because of their uniformity, stability, and affordable price [14]. Their use in OMFs is allowed in the current EU regulation [1], together with lignite. However, using peat and leonardite is environmentally unsustainable since their extraction reintroduces geologically stored C into the C cycle. Therefore, the replacement of organic geogenic materials with renewable and local alternatives that do not require transport over long distances is highly desirable [5]. The utilization of biowaste materials such as pig slurry has been proposed as a potential candidate for inclusion in organo-mineral fertilizers [15]. Biowaste materials are readily available locally, and their integration into OMFs fosters a circular value by establishing a closed-loop system that repurposes and recycles waste materials for agricultural benefits [16–18]. In addition, biowaste materials can increase their stability with relatively cheap and straightforward technologies such as composting [19], anaerobic digestion [20], vermicomposting [21], and pyrolysis [22]. This approach reduces waste, diverting materials from landfills or incineration [23]. Although the pyrolysis process modifies the chemical structure of biowaste with respect to compost or digestion processes [24], it offers a faster method to process residues of pruning and timber industries. The use of wood biochar has been studied in OMFs, showing the potential to slow down the release of N and K [25], and P in acid soils [26].

In addition to the organic material quality, the proportion between the organic and mineral fractions can influence the OMF properties. One group of OMFs consists of products in which the organic material is used as the primary source of one or more nutrients, and a mineral fertilizer is added only to correct the nutrient ratio [2,27,28]. This type of OMF, in addition to supplying nutrients, can work as an amendment in improving soil's physical properties [29–32]. However, as with organic fertilizers, this type of OMF requires a large volume of application, in the order of tons per hectare [2,33], thus increasing application costs.

A second group of OMFs includes products in which the organic material is used in a low proportion to the mineral fertilizer; therefore, its contribution to nutrient supply is scarce. In OMFs with 7.5 %  $C_{org}$ , the organic fraction represents 30–50 % of the total OMF granule [34,35]. The benefits of this type of OMF are that they should increase the efficiency of the mineral fertilizer by slowing down the nutrient release [4,34,36,37]. Some examples of the good performance of these OMFs are reported in the literature. For example, in a pot experiment with *Lolium perenne*, low  $C_{org}$  OMFs made with peat showed a slight increase in yield after five months and a 15 % decrease in N losses through leaching [38]. Low  $C_{org}$  OMFs made with biochar showed a slowdown in the mineral P release into the soil solution, reducing the nutrient fixation to the soil and increasing the P uptake and P recovery by 10–20 % in a greenhouse experiment with millet (*Pennisetum glaucum*) grown for 60 days and followed by maize (*Zea mays*) for further 30 days (26). The increase in use efficiency of the P mineral fertilizers, even with the addition of low quantities of  $C_{org}$ , is of interest because it can reduce the depletion of phosphate reserves and the cost of energy to produce fertilizers [2].

While the replacement of organic geogenic materials with stabilized biowastes is desirable in OMFs, and peat-free OMFs typically utilize locally sourced organic materials, there remains uncertainty regarding specific parameters of organic materials that can influence positively or negatively the OMFs and to which extent those parameters will influence specific types of OMF. These uncertainties are linked to the high variability among organic materials depending on their origin and degradation process [38]. Because the concept of OMF is widely open, it allows different fertilizer formulations, combinations of materials, and physical structures [1]. A complete understanding of what constitutes a high-quality organic material for an OMF is essential for assessing whether biowaste materials can effectively replace organic geogenic materials. Before investing in the formulation, granulation, or pelletization of the OMF and forwarding an agronomic test, a set of physical and chemical parameters must be defined to accept or reject an organic material as a potentially suitable matrix.

The objective of this study was to determine if standard chemical and physical analysis methods can be used as a first filter to discard organic materials for their use as the organic matrix for low  $C_{org}$  OMF. A summary of analyses required for evaluating an organic material for use in a commercial OMF is presented in Fig. S1. Then, additional analyses, including law compliance and agronomic and environmental tests regarding nutrient release, losses, and crop growth, should be conducted on the final OMF product before commercialization [39].

Materials categorized as high-quality matrixes for OMF can be considered potential substitutes for organic geogenic materials. To achieve this aim, this paper proposes a list of physical and chemical parameters based on the European regulation for fertilizers and other unregulated parameters to describe i) the quality of the organic matter of the organic matrix, ii) the limitations for the OMF elaboration and application, iii) the organic matrix nutrient contribution to the OMF, and iv) the space for creating OMF mixtures. We discuss this list of parameters using real cases of potential organic matrixes for a solid OMF containing 7.5 % C<sub>org</sub>. We excluded microbial characteristics from our analyses because the absence of potentially harmful microorganisms should be tested in the final product [1,40], and the granulation or pelletization process are expected to modify the bacterial load of the OMF components [41]. Therefore, 14 renewable organic materials from various locations across Italy were collected and characterized physically and chemically and produced using different local raw materials and stabilizing processes. Additionally, peat and leonardite samples were included as references.

Researchers or industries can use the proposed list of parameters to expand the search for potential organic matrixes. By harnessing compost and biochar materials as viable organic matrixes, OMF industries can reduce reliance on non-renewable organic geogenic materials while contributing to sustainable waste management practices.

# 2. Materials and methods

# 2.1. Organic materials collection

Organic materials were mainly collected from industries across Italian regions. Different organic materials were gathered according to different typical supply chains. Two were organic geogenic materials (Peat and Leon), and two were wood biochar (BC). The other materials had gone through various composting periods ranging from three to nine months: green compost based on pruning residues (GC); compost containing livestock manure sometimes mixed with agricultural wastes (MC); municipal solid waste compost (MSWC); and manure-based vermicompost (VC) (Table 1). Materials were collected between February and April 2021 and stored in plastic bags

until analyses. An aliquot was then air-dried and sieved at 0.5 cm to separate large woody or stone parts. The fine fraction was subsequently mechanically milled to <2 mm to increase the uniformity of the materials, except GC-prun, which was first sieved at <2 mm due to the presence of stones that did not allow the milling of the sample.

# 2.2. Physico-chemical characterization

Organic materials were characterized following mainly a conventional protocol used in Italy for compost analyses [42]. The analyzed parameters determine how an organic material will be processed to create an OMF granule or pellet, such as the granulometry or the amount of C<sub>org</sub>. Parameters of an organic material that will affect an OMF in the field were also included, such as the content of total nutrients or heavy metals. However, parameters related to the final OMF itself have been left out: for example, pathogen colonies may vary during the drying and heat dehydration process of the OMF mixture; the organic matter and nutrient complexes in the OMF, as well as the N-P-K release dynamics, will change depending on the type of mineral fertilizer used - which in turn will also depend on the desired nutrient concentration in the final OMF. Therefore, their study escapes the scope of this paper and requires specific attention. Our research group has covered the nutrient dynamics of N and P in biowaste OMFs and studied potential mechanisms of interactions between the organic and mineral fractions in previous papers [35,43,44].

The parameters have been grouped and based on how they will influence an OMF with 7.5 %  $C_{org}$  into 1) limiting parameters for OMF manufacturing, which can cause the discarding of an organic fraction; 2) parameters linked to the organic matter characterization, which will affect the stability of the OMF, as well as potential interactions between organic and mineral fractions; and 3) macronutrient and micronutrient parameters, which will serve as a supplement to the nutrients provided by a mineral fraction.

Analyses were conducted in triplicate to evaluate the analytical variability, except for some determinations where the chemical variability was low (humidity, particle size, salinity, pH, EC, carbonates and bulk density) and those characterized by high costs or long analytical procedure (NMR, organic matter fractionation).

# 2.2.1. Limiting parameters for OMF manufacturing

The limiting parameters included humidity, particle size, salinity, pH, and potentially toxic elements (PTE). This group includes parameters that can strongly impact the final product stability, availability, and toxicity.

The residual humidity of air-dried samples was calculated after drying samples at 105 °C for 24h. The particle size of air-dried samples was measured before sieving at 0.5 cm and milling, as this initial characteristic indicates the suitability of an organic material to be used in a pelletizer or a granulator for OMF production. The particle size was determined by sieving samples at >20, 20–10, 10–2, 2–1, 1–0.5, and <0.5 mm. The salinity was measured by determining the electrical conductivity in a solid:water suspension (1:10 w:v). The pH was determined in the same solution with an inoLab<sup>TM</sup> 7110 benchtop pH meter (WTW, Fisher Scientific Inc, USA). The total contents of heavy metals - cadmium (Cd), chromium (Cr), nickel (Ni), lead (Pb), zinc (Zn), and copper (Cu) - were determined by flame atomic absorption spectrometry (FAAS, PerkinElmer AAnalyst 400, USA), after heating the organic material at 450 °C for 4 h and dissolving the residue with 1 M HCl/HNO<sub>3</sub> for 2 h.

# 2.2.2. Organic matter characterization

The organic materials were analyzed for their elemental composition, fractionated into base-extractable C, humic and fulvic acids, and characterized by <sup>13</sup>C CP NMR spectroscopy.

Total C was determined by elemental analysis (UNICUBE, Elementar Analyses System GmbH, Langensbold, Germany). Carbonate content was measured by adding hydrochloric acid to the sample and measuring the volume of carbon dioxide gas produced (ISO 10693:1995). Organic C (C<sub>org</sub>) was calculated as the difference between total C and inorganic C, assuming the latter to be derived from carbonates.

The organic material was then subjected to chemical fractionation with 0.5 M sodium hydroxide and 0.1 M sodium pyrophosphate under  $N_2$ . The suspension was shaken for 48 h at 65 °C and then centrifuged at 3000 rpm for 20 min. The supernatant was filtered using cellulose acetate filters of 0.8  $\mu$ m and its total C content (Extractable organic carbon, TEC) was determined. Humic acids (HA) and fulvic acids (FA) were then separated by precipitation with 1 M sulfuric acid, centrifuged, and HA separated as the precipitate. The remaining supernatant was passed through a column filled with polyvinylpyrrolidone to separate the FA from the non-humic fraction. The TEC, HA, and FA were characterized for C and N content by elemental analysis. TEC and Humic C were not determined for biochar samples as these parameters are specific for materials that undergo a microbially-driven mineralization process.

The humification degree (HD), i.e. the amount of base-extractable C that has been transformed into fulvic and humic acids, was calculated as follows:

$$HD = \frac{(Humic Acid C + Fulvic Acid C)}{Extractable organic C}$$

Furthermore, the organic materials were characterized by high-resolution solid-state <sup>13</sup>C cross-polarization magic-angle-spinning nuclear magnetic resonance (<sup>13</sup>C CPMAS NMR) spectroscopy. The spectra were obtained using a JEOL ECZR 600Hz at a frequency of 150.91 MHz. Samples were packed to a volume of 60  $\mu$ l into 3.2 mm cylindrical zirconia rotors and analyzed at room temperature. The RAM-CPMAS pulse sequence (1H 90° pulse = 2.0  $\mu$ s; contact time = 1 ms; optimized relaxation delays of 2 s; 35,000 scans) with the TPPM 1H decoupling (rf field = 112 kHz) during the acquisition period was used for all treatments. Glycine was used as an external standard, and its resonance was used to refer to the <sup>13</sup>C chemical shift scale. The <sup>13</sup>C NMR spectra were divided into six regions (0–45

Table 1Organic materials (OM) description.

OM	Description	Processing	Stabilization time (months)		
Peat	Peat.	Organic geological formation			
Leon	Fossil leonardite from lignite deposits.	Organic geological formation			
BC-1	Wood biochar.	Pyrolisis			
BC-2	Wood biochar.	Pyrolisis			
GC-fung	Mushroom substrate (55 %), almond hust (10 %), wood chip (15 %), and vegetable residues (10 %).	Composting	3		
GC-hort	Shredded wood and forest leaves (30 %) with horticultural residues low in solids.	Composting	5 to 6		
GC-prun	Pruning	Composting	6		
GC-zeo	Pruning with added zeolite	Composting	6 to 9		
MC-man1	Bovine and equine manure.	Composting	6 to 9		
MC-man2	Solid pig manure.	Composting	6 to 9		
MC-pom	Manure (60 %), olive pomace (20 %), and olive tree pruning (20 %).	Composting	5 to 6		
MC-sol	Solid manure (75 %) and wood chips (25 %).	Composting	6		
MSWC-1	Municipal solid waste organic fraction and digestate sludge.	Composting/Digestion	6		
MSWC-2	Municipal solid waste organic fraction and digestate sludge.	Composting/Digestion	6		
MSWC-3	Municipal solid waste organic fraction and digestate sludge.	Composting/Digestion	3 to 5		
VC-man	Vermicompost of bovine manure compost.	Composting/Vermicomposting	6 to 9		

ppm, 46–60 ppm, 61–110 ppm, 111–155 ppm, 156–165 ppm, and 166–190 ppm) and quantified with the Delta v5.3.1 software (Jeol Ltd., Japan).

# 2.2.3. Nutrient characterization

Macronutrient (N, P, and K), meso- and micronutrient contents were determined in the organic materials. Total N was determined by the Kjeldahl distillation method after digesting 5 g of fresh sample with concentrated sulfuric acid and selenium catalysis [45]. Ammonium N (N-NH<sub>4</sub>) and nitrate N (N-NO<sub>3</sub>) were extracted from 10 g with 100 ml of 1 M KCl under mechanical shaking for 60 min and further centrifugation at 3000 rpm for 10 min. The N-NH<sub>4</sub> was determined by the Kjeldahl method on the extract. N-NO<sub>3</sub> concentration was determined spectrophotometrically (U-2000 spectrophotometer, Hitachi, Tokyo, Japan) using the Griess reaction [46]. Organic N (N<sub>org</sub>) was obtained by difference Total N – (N-NO<sub>3</sub> + N-NH<sub>4</sub>). Total P content was determined spectrophotometrically (UV-1900i, Shimadzu Corp, Japan) using a solution of ammonium molybdate as reported by Murphy & Riley (1962), after acid digestion with nitric and perchloric acid. Potassium (K), calcium (Ca), Magnesium (Mg), iron (Fe), and manganese (Mn) were determined after nitric acid digestion using atomic absorption spectrometry (FAAS, PerkinElmer AAnalyst 400, USA). Total sulfur (S) was determined by elemental analysis (UNICUBE, Elementar Analyses System GmbH, Langensbold, Germany). Total boron (B) was determined by UV–vis spectrophotometry at 410 nm after adding calcium oxide to the sample and heating at 600 °C for 1 h (Keren, 1996). The samples were then redissolved with 2N sulfuric acid.

# 2.3. Formula space

Given an organic material and a desired C<sub>org</sub> concentration target, the formula space is defined as the proportion of space in an OMF that can be "filled" with mineral fertilizer, organic fertilizer, or the so-called fillers, such as granular limestone.

The formula space is related to the proportion of the organic material (OFprop) in the OMF, which depends on its concentration of  $C_{org}$  and the  $C_{org}$  target in the OMF itself.

$$OFprop~(\%) = \frac{Corg~OMF~target~(\%)*100}{Corg~DM~(\%)}$$

Formula space (%) = 100 - OFprop(%)

Where  $C_{org}$  OMF target refers to the desired  $C_{org}$  content in the OMF, in our work, 7.5 % according to the EU regulation [1];  $C_{org}$  DM refers to the  $C_{org}$  content in the organic material expressed in % over the dry matter. If OFprop is >100 %, the organic material does not have enough  $C_{org}$  to be used in an OMF with 7.5 %  $C_{org}$ , making it unsuitable for OMF production.

When the minimum  $C_{org}$  OMF target is satisfied, the rest of the formula space can be used to complete OMF, considering the mineral fertilizer and other materials as fillers that improve the agglomeration between fractions or additional organic materials to provide extra N<sub>org</sub>.

The OFprop defines the contribution of the organic material in humic C, macro and micronutrients, PTE, and salinity, among other parameters, to the targeted OMF to the OMF in macronutrients. Therefore, it has been utilized to calculate the contribution of each organic material to the theoretical OMF containing 7.5 %  $C_{org}$ , considering macro, meso, and microelements, as well as heavy metals and salinity. The latter is described as oxides, per European regulations governing OMFs, to distinguish between the characterization values of the organic materials and their contribution to the OMFs. For the same regulation, the values of the contribution of the organic material to the OMF for  $C_{org}$ , TEC, Humic C, and nutrients are expressed in % of the total OMFs.

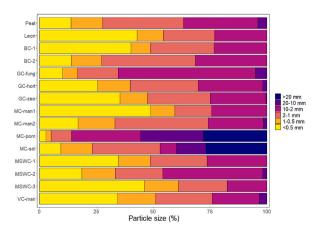


Fig. 1. Particle size fraction of organic materials previous milling.

#### 3. Results

# 3.1. Limiting parameters for OMF manufacturing

There was high variability in the particle size among samples and groups (Fig. 1). GC-fung, MC-pom, MC-sol, and MSWC-2 contained a significant fraction of coarse materials (>2 mm). Conversely, Peat, Leon, BC-1, BC-2, GC-hort, GC-zeo, MC-man1, MC-man2, MSWC-1, MSWC-3, and VC-man had 63.3–82.5 % of the particles finer than 2 mm.

The residual humidity was highly variable among samples and groups (Table 2). The highest values were found in MC-man2, MC-sol, MC-pom, and BC-2 (50.1–59.9 %), followed by materials like Peat, GC-zeo, VC-man, MSWC-2, and GC-fung (35.9–43.9 %), while the rest of the samples had a residual humidity between 6.9 and 29.8 %.

The electrical conductivity, used as a salinity proxy, was also very variable between and within groups. VC-man ( $1.5 \text{ mS cm}^{-1}$ ), Peat ( $1.5 \text{ mS cm}^{-1}$ ), and Leon ( $2.0 \text{ mS cm}^{-1}$ ), and MSWC group ( $1.6-2.2 \text{ mS cm}^{-1}$ ) had similar electric conductivity, while GC and MC showed high variability in their groups. MC-man1 ( $4.8 \text{ mS cm}^{-1}$ ) and GC-hort ( $4.0 \text{ mS cm}^{-1}$ ) had approximately double the electrical conductivity of Peat, while BC samples had the lowest values ( $<0.3 \text{ mS cm}^{-1}$ ).

Regarding pH, the analyzed Peat was alkaline (8.0), with Leon (6.1) and MC-man1 (7.5) being relatively neutral. All other analyzed materials were more alkaline than Peat (9.3–10.8).

PTE concentrations (Table 2 and S1) in studied materials were variable for most elements apart from Cd, found in relatively similar concentrations in all materials. Pb, Zn, and Ni showed higher variability in their contents. Total Cr and Ni were found in relatively high concentrations in BC, GC, and MSWC and lower in MC, Peat, Leon, and VC. Pb was found in high concentrations only in BC-1 and GC-prun, while in the rest of the materials, it did not exceed 40 mg kg<sup>-1</sup> DM. Zn and Cu also had high variability among groups. Zn was low in biochar, GC-fung, MC (except for MC-man1), Peat, Leon, and VC. In contrast, Cu had the lowest concentrations in Peat, Leon, and GC-fung. Cu and Zn can also be considered nutrients, but their values are presented only once in Table 2.

# 3.2. Organic matter characteristics

The organic carbon ( $C_{org}$ ) content varied widely among the materials and groups (Table 3 and Table S2): BC-1 and BC-2 had a high  $C_{org}$  content (>70 %); MC-sol, and MC-pom with medium  $C_{org}$  range (38.5 %–39.7 %); Peat, GC-zeo, GC-hort, VC-man, MSWC-1, MSWC-2, MSWC-3, MC-man2, Leon, and GC-prun with medium to low  $C_{org}$  range (16.7 %–28.2 %); and, MC-man1 and GC-fung with low  $C_{org}$  range (9.9 %–14.9 %).

The C/N ratio showed major differences among the samples. As expected, biochar samples showed very high values; Leon had intermediate values, while Peat and composted biowastes had lower values, typical of well-composted materials (Table 3), with the lowest values for the MSWC group.

The humic C also had a high variability between groups and samples. The humic carbon (Humic C) content was low in GC-hort, MC-man1, GC-fung, and GC-prun samples (3.2–6.6 %), while other samples contained a medium humic C content (8.0–15.2 %). The humic C content was correlated with the degree of humification. GC-zeo, MSWC-3, GC-fung, and GC-prun contained a humification degree below 60 %.

Solid-state <sup>13</sup>C CPMAS NMR spectra are shown in Fig. 2, while the relative C distribution in the OF is shown in Table 4. Leon had a strong signal in the 0–45 ppm region (long-chain alkyl moieties), accounting for 67.9 % of total C. In contrast, MC-man2, MC-sol, and MC-POM had a relatively low fraction (<15.9 %) compared to all other materials (20.3–33.9 %).

All treatments showed a 46-60 ppm signal, attributed to C-N or C-O groups. The signal was highest in VC-man (16.6 %), followed

# Table 2

Variables considered as limiting parameters for the manufacturing of OMFs are residual humidity (RH°), electrical conductivity (EC), pH, and potentially toxic element content (Cd, Cr, Ni, Pb, Cu, Zn). Cu and Zn are also considered nutrients.

OM	$RH^{\circ}$	EC	pH	Cd	Cr	Ni	Pb	Cu	Zn			
	%	$mS cm^{-1}$		$mg kg^{-1}$	mg kg <sup>-1</sup> DM							
Peat	37.6	1.54	8.0		79	36	10	33	105			
Leon	13.1	2.00	6.1		75	59	10	16	51			
BC-1	9.0	0.13	10.7		141	95	177	28	206			
BC-2	59.9	0.30	10.3		108	90	10	29	90			
GC-fung	43.9	0.60	10.8		124	89	10	28	45			
GC-hort	6.9	3.95	10.7		66	37	10	115	141			
GC-prun	24.6	1.11	9.5		199	129	48	193	434			
GC-zeo	38.4	1.86	9.3		192	109	22	125	228			
MC-man1	27.5	4.78	7.5	<6	57	33	10	174	162			
MC-man2	56.5	1.61	9.1		53	40	10	72	356			
MC-pom	50.9	1.39	9.9		20	10	10	33	169			
MC-sol	50.1	1.68	10.1		20	10	10	36	192			
MSWC-1	26.4	2.19	9.3		128	89	32	94	179			
MSWC-2	35.9	1.69	9.2		147	108	35	117	288			
MSWC-3	29.8	1.63	9.2		255	202	27	230	387			
VC-man	43.2	1.49	9.9		59	50	10	70	264			

#### Table 3

Organic matter characteristics and nutrient content: Humification degree (HD), Carbon to Nitrogen ratio (C/N), Organic C (C<sub>org</sub>), Total Extractable Carbon (TEC), Total Humic Carbon (Chum), total N (N<sub>total</sub>), organic N (N<sub>org</sub>), P, K, Ca, Mg, S, B, Fe, Mn. Cu and Zn are shown in Table 2.

ОМ	HD	C/N	Corg	TEC	C <sub>hum</sub>	N <sub>total</sub>	Norg	Р	К	Ca	Mg	S	В	Fe	Mn
			$mg g^{-1} DM$												
Peat	0.79	16	166.9	100.6	79.5	10.0	9.3	2.4	10.3	40.7	14.0	11.3	0.08	30.2	0.3
Leon	0.68	71	206.1	136.2	92.1	2.2	2.1	0.1	4.3	34.6	5.3	34.3	0.05	26.7	0.1
BC-1	nd	142	717.7	nd	nd	4.0	3.9	3.4	11.3	45.8	4.8	2.7	0.08	8.3	0.3
BC-2	nd	230	758.0	nd	nd	2.2	2.0	2.4	5.7	35.0	4.9	1.2	0.09	2.9	0.4
GC-fung	0.46	20	98.8	68.5	31.6	6.0	5.8	0.8	9.7	77.7	6.1	3.6	0.06	16.0	0.5
GC-hort	0.62	13	201.1	107.0	66.0	13.9	13.3	6.2	31.8	51.1	11.7	6.1	0.09	20.7	0.7
GC-prun	0.53	18	193.8	113.7	60.3	11.0	10.7	4.4	13.9	52.9	14.6	7.3	0.09	17.5	0.6
GC-zeo	0.56	13	280.4	143.7	80.9	22.2	20.7	7.2	11.7	44.8	12.8	5.8	0.06	16.8	0.4
MC-man1	0.63	11	149.1	80.6	51.0	11.6	9.3	4.8	11.1	90.0	27.9	17.0	0.05	12.3	0.4
MC-man2	0.95	20	228.8	131.0	124.7	14.7	13.9	20.7	14.4	79.3	16.6	7.9	0.06	6.8	0.5
MC-pom	0.74	19	396.5	205.2	152.3	22.6	22.3	6.6	12.3	34.8	7.2	5.6	0.09	3.6	0.3
MC-sol	0.70	17	384.7	200.1	139.5	26.3	26.1	7.5	14.1	32.7	7.3	5.9	0.09	3.8	0.3
MSWC-1	0.69	10	212.7	135.1	92.6	23.1	21.4	8.4	10.0	46.5	11.5	5.4	0.06	16.3	0.4
MSWC-2	0.65	13	276.8	136.7	89.0	22.3	21.0	7.3	10.9	41.3	10.7	5.9	0.07	16.3	0.5
MSWC-3	0.56	10	282.2	147.0	82.1	25.8	21.7	17.5	10.1	34.4	10.6	9.6	0.09	40.3	0.4
VC-man	0.72	15	198.1	135.3	97.6	14.8	14.1	8.7	13.4	40.5	12.0	7.7	0.07	15.0	0.7

nd = not determined.

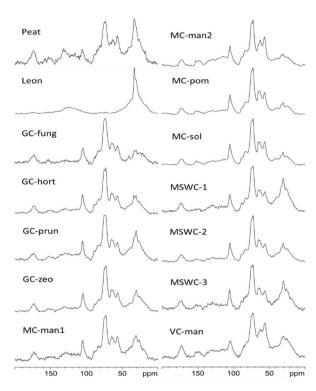


Fig. 2. Nuclear Magnetic Resonance (NMR) spectra from the analyzed organic materials.

by MC-man1, MC-man2-MC-sol, and MC-pom (12.4–14.8 %). Another group was formed by Peat, GC-zeo, GC-hort, MSWC-1, MSWC-2, MSWC-3, and GC-fung (11.6–12.2 %), while the lowest signal was in GC-prun (9.9 %) and Leon (5.9 %).

The 61–110 ppm O-C region, mainly attributed to saccharidic C, had strong signals in all samples except for Leon, representing only 4.7 % of total C. For Peat, MC-man1, VC-man, MSWC-1, MSWC-3, and GC-prun, this region contributed between 36.2 and 47.5 %, while for GC-zeo, GC-hort, MSWC-2, MC-man2, MC-sol, MC-pom, GC-fung, it represented between 50.5 and 58.6 %. This strong signal is probably caused by cellulose and hemicellulose [47].

The 111–155 ppm aromatic C region presented a high variability, too. Leon presented the highest value (19.9 %), followed by Peat, MC-man1, VC-man, MC-man2, MC-sol, and GC-prun (10.0–15.5 %), then by GC-zeo, GC-hort, MSWC-1, MSWC-2, MSWC-3, and MC-pom (8.0–8.9 %), and finally by GC-fung (4.6 %). The 156–165 ppm region (phenolic C) had a low signal (<1.1 %); Peat, MC-man1, VC-man, MSWC-1, and Leon had the highest phenolic C fraction (0.6–1.1 %), while the other compounds had on average only 0.1 %.

#### Table 4

Integration areas	for th	e maior	C-types in	1 the 13	SC CPMAS	NMR spectra.

OM	Alkyl C	Aliphatic C	O-alkyl C	Aromatic C	Phenolic C	Carboxyl C	Alkyl C/O Alkyl C		
	0-45 <sup>a</sup>	46–60	61–110	111–155	156–165	166–190			
	%								
Peat	31.6	12.2	36.2	14.4	0.8	4.8	0.9		
Leon	67.9	5.9	4.7	19.9	0.6	1.0	14.4		
GC-fung	20.3	11.6	57.4	4.6	0.1	6.0	0.4		
GC-hort	22.5	11.6	54.5	8.1	0.1	3.2	0.4		
GC-prun	28.2	9.9	47.5	11.4	0.1	2.9	0.6		
GC-zeo	25.3	12.0	50.5	8.0	0.1	4.1	0.5		
MC-man1	25.4	12.4	44.3	12.6	1.1	4.2	0.6		
MC-man2	14.3	14.8	52.8	15.5	0.1	2.5	0.3		
MC-pom	15.9	14.4	58.6	8.3	0.1	2.7	0.3		
MC-sol	15.1	14.1	57.5	10.0	0.1	3.2	0.3		
MSWC-1	33.9	12.1	43.9	7.2	0.1	2.8	0.8		
MSWC-2	23.2	11.9	51.8	8.2	0.6	4.3	0.4		
MSWC-3	29.2	11.2	46.8	8.9	0.1	3.8	0.6		
VC-man	23.3	16.6	41.7	12.9	0.6	4.9	0.6		

<sup>a</sup> All values reported in the row refer to the range value expressed in ppm.

The 166–190 ppm region C (C in carboxyl, amide, and ester groups) ranged from 1 % (Leon) to 6 % (GC-fun); however, no apparent differences between groups were found.

#### 3.3. Nutrient contents

Peat was richer than Leon in total and organic N, P, K, and Mg, while Leon was richer in S (Table 3 and Table S2). Vermicompost (VC-man) was generally similar to Peat in all macronutrients. BC was poor in macronutrients and richer than Peat in Ca. MSWC samples were richer in N, N<sub>org</sub>, and P than Peat. MC and GC groups had a high variability; MC-sol, MC-pom, and GC-zeo were richer in N than Peat, while others showed similar values. Except for GC-hort, all GC and MC were richer in P than Peat. Some matrices, particularly those of groups MC and MSWC, were high in nutrients if compared with the European legislation thresholds that set limits to the final OMF product, although not to the OM fraction.

All analyzed groups generally showed high variability in microelement contents unrelated to groups (Table 3). Mn was found in moderate concentrations in all analyzed elements. Peat and MSWC-3 had the higher Fe concentration. Compared to GC, MSWC, and VC, Peat was poorer in Mn, while only MC-sol was richer in Mn than Peat in the MC group. Compared to most composts, Peat was poor in Zn.

Further chemical characteristics of the OMs are reported in Table S3.

# 3.4. Formula space

When targeting an OMF with 7.5 % C<sub>org</sub>, the calculated OFprop was highly variable among samples and groups. GC-fung had an OFprop of 76.0 % (Fig. 3), making this material unsuitable for OMFs with a high concentration of mineral fertilizers. The lowest

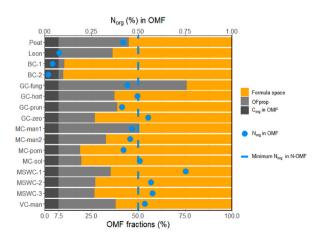


Fig. 3. Proportions of mineral fertilizer and organic material necessary to provide 7.5 % C<sub>org</sub> to an OMF reported in bottom X-axis. The Norg contribution to the OMF is indicated as % in the top X-axis.

OFprop was found in BC-1 (9.9 %) and BC-2 (10.5 %). GC-zeo, MSWC-2, MC-sol, and MC-pom would cover between 18.9 and 27.1 %. The other materials ranged between 32.8 and 50.3 %. Although all organic materials contained nutrients, particularly N, the contribution of N from the organic fraction ( $N_{org}$ ) in the theoretical OMF with 7.5 %  $C_{org}$  was minimal, less than 0.75 % (Fig. 3).

#### 4. Discussion

Deciding if a recycled organic material can be used to produce a low  $C_{org}$  OMF—and therefore effectively replace peat and leonardite—requires considering different physical and chemical aspects related to the industrial manufacturing requirements, the stability and nutrient supply of the OMF, and the by-characteristics that negatively impact plants or soil after fertilization.

# 4.1. Particle size and water content

From an industrial point of view, the homogeneity and uniformity of the organic material is a discriminating parameter, as it ensures an even distribution of organic fraction and, subsequently, mineral nutrients in the unit of pellets and grains of the OMF [1]. The particle size helps understand and measure the homogeneity of organic materials [48] and the likelihood of materials being granulated during the industrial production process [49]. There is scarce information about raw material particle size distribution and their attitude to produce homogeneous granules with an even nutrient distribution. It has been proposed that the ideal granule size for OMF is between 1.1 and 5.5 mm for conventional twin-disc spreaders [2,50]. In granular OMFs, solid granules of mineral fertilizers, such as TSP or DAP, are used as the nucleus of the final OMF; therefore, the organic materials should have a fine particle size so as not to increase excessively the size of the new OMF granule and surpass the proposed ideal OMF granule diameter. Materials like MSWC-2, MC-sol, MC-pom, and GC-fung contain around 50 % or more of particles above 2 mm, and therefore, removing or milling the coarse fraction to make them suitable for granular OMFs is necessary. Therefore, those materials are less desirable because the manipulation cost for OMF producers would increase.

In addition, dry organic materials lack adherence and cohesive properties to form granules with the mineral fertilizer. Therefore, adding water or solutions with additives to the organic material is necessary during the granulation or pelletizing process to ensure the particles can bind together effectively [2]. On the contrary, organic materials with high humidity clump and cake during OMF pelletization worsen the OMF processing line. The water content in the organic material may dilute the concentration of  $C_{org}$  or nutrients; however, the OMF granulation process usually contains a drying step to reduce the final humidity [2]. Therefore, the humidity content in the raw material needs to be considered because of the energy it will take to dry the final OMF. After one month of air drying, several materials contained high water contents (50–60 %), such as BC-2, MC-man2, MC-pom, and MC-sol, that would increase their cost of drying or manipulation. Taking Peat (40 %) as the reference material, most materials are close to their residual humidity, which shows that an air-drying process can reduce the water content of raw materials before OMF granulation.

#### 4.2. Organic matter and nutrients

In the second step, the OMF design implies the correct balance between  $C_{org}$  and nutrient elements, for which the formula space calculation becomes a predominant aspect. The organic material  $C_{org}$  concentration and the OMF  $C_{org}$  target –minimum 7.5 %  $C_{org}$  in solid OMFs [1] – define the OFprop. Low  $C_{org}$  concentration in organic materials implies an increase of the OFprop in the OMF, reducing the space for mineral fertilizer [15]. Consequently, materials such as MC-man1 ( $C_{org} = 14.9$  %) and GC-fung ( $C_{org} = 9.9$  %) leave poor space for adding mineral fertilizers, forcing the production of OMFs with low nutrient concentrations. On the other hand, high  $C_{org}$  concentration implies that the organic material is not required in large quantities in the OMF; this is the case of BC-1 ( $C_{org} = 71.8$  %) and BC-2 ( $C_{org} = 75.8$  %), which would represent less than 11 % of OFprop in an OMF. The reduction of the OFprop affects the physical properties of the OMFs, decreasing its density and increasing abrasion fragility and crushing strength [15] or not providing enough material to encapsulate the mineral fertilizer. Fixing a low OFprop would require the use of fillers, such as sand, granular limestone, or sawdust, to fulfill the formula space [51], causing additional interactions between soil and fertilizer [52].

More difficult is processing an OMF where  $C_{org}$  and one or more macronutrients are targeted [1]. If the OMF provides only N (N-OMF), the OMF needs to contain 6 % of total N, and 1 % needs to be  $N_{org}$ . On the other hand, if the OMF contains N with another macronutrient (NPK/NP/NK OMFs), the minimum content for total N,  $P_2O_5$ , or  $K_2O$  is 2 %, but the macronutrients have to sum 8 % of the total OMF, while the  $N_{org}$  content needs to be at least 0.5 % [1]. For an OMF with 7.5 %  $C_{org}$ , none of the materials could provide 1 % of  $N_{org}$ . Biochar and leonardite samples provide less than 0.1 % of  $N_{org}$  in the OMF, making those materials unsuitable for an N-OMF alone. On the contrary, most analyzed materials are near or above 0.5 % of  $N_{org}$  (Table 5) and, therefore, suitable for NPK/NP/NK OMFs. Considering the formula space, organic materials that have a wide space for mineral fertilizers are still suitable matrixes for N-OMFs since the targeted  $N_{org}$  content can be achieved by adding materials rich in  $N_{org}$  like blood meal, feather meal, or seabird guano [53]. None of the analyzed organic materials would contribute the minimum total N,  $P_2O_5$ , or  $K_2O$  content to a 7.5 %  $C_{org}$  OMF; therefore, mineral fertilizers are required to supply each nutrient.

The formula space must also be considered when choosing the type of mineral macronutrient source. For example, targeting high N concentrations may require using urea (N = 46 %) instead of ammonium sulfate (N = 21 %), or if the OMF contains P, using diamonium phosphate (N = 18 %) instead of triple superphosphate to provide extra N. With this consideration, materials such as MC-man1 ( $C_{org} = 14.9$ ) and GC-fung ( $C_{org} = 9.9$ ) are suitable only for OMF with low N-P<sub>2</sub>O<sub>5</sub>-K<sub>2</sub>O ratios.

An OMF could also be registered as a source of meso and micronutrients [1]. However, none of the analyzed materials would contribute significantly to mesonutrients as Mg and S in a 7.5 % C<sub>org</sub> OMF (Table 5) and, therefore, should not be considered as an Mg

# Table 5

11

Organic materials (OMs) contribution to theoretical OMFs with 7.5 C<sub>org</sub>. Each OM parameter needs to be evaluated with the effect of "dilution" once mixed with the mineral fertilizer rather than with the direct values of its characterization. The minimum (min) and maximum (max) values can be used to determine if an OM requires minimum or major adjustment to fit ideal values or if the OMF needs to be discarded. Values are expressed as oxides as this form is used in OMF regulation (EC, 2019).

OM	Corg	Chum	N <sub>total</sub>	Norg	$P_2O_5$	K <sub>2</sub> O	CaO	MgO	$SO_3$	Fe	Cd	Cr	Ni	Pb	Cu <sup>3</sup>	Zn <sup>3</sup>
	%										mg kg	$^{-1}$ DM				
Peat	7.5	3.6	0.4	0.4	0.2	0.6	1.3	0.4	0.2	1.357	2.1	27.8	12.7	3.5	11.5	36.9
Leon	7.5	3.4	0.1	0.1	0.0	0.2	0.9	0.1	0.5	0.973	2.0	25.6	20.2	3.4	5.6	17.2
BC-1	7.5	nm	0.0	0.0	0.1	0.1	0.3	0.0	0.0	0.086	0.6	13.4	9.0	16.8	2.7	19.6
BC-2	7.5	nm	0.0	0.0	0.1	0.1	0.2	0.0	0.0	0.029	0.6	10.4	8.6	1.0	2.8	8.7
GC-fung	7.5	2.4	0.5	0.4	0.1	0.9	4.2	0.3	0.1	1.211	4.0	82.6	59.0	6.7	18.4	29.8
GC-hort	7.5	2.5	0.5	0.5	0.5	1.4	1.4	0.3	0.1	0.774	2.1	23.5	13.3	3.6	40.8	50.0
GC-prun	7.5	2.3	0.4	0.4	0.4	0.7	1.5	0.3	0.1	0.676	1.8	61.1	39.5	14.7	59.3	133.3
GC-zeo	7.5	2.2	0.6	0.6	0.4	0.4	0.9	0.2	0.1	0.449	1.4	45.4	25.8	5.3	29.6	54.1
MC-man1	7.5	2.6	0.6	0.5	0.6	0.7	3.2	0.8	0.3	0.619	2.7	25.8	15.3	4.6	79.5	74.1
MC-man2	7.5	4.1	0.5	0.5	1.6	0.6	1.9	0.3	0.1	0.224	1.4	12.5	9.3	2.3	16.8	83.4
MC-pom	7.5	2.9	0.4	0.4	0.3	0.3	0.5	0.1	0.0	0.068	1.0	3.2	1.6	1.6	5.3	27.0
MC-sol	7.5	2.7	0.5	0.5	0.3	0.3	0.5	0.1	0.0	0.075	1.0	3.4	1.7	1.7	6.2	32.8
MSWC-1	7.5	3.3	0.8	0.8	0.7	0.4	1.2	0.2	0.1	0.576	1.9	41.0	28.5	10.1	30.2	57.2
MSWC-2	7.5	2.4	0.6	0.6	0.5	0.4	0.8	0.2	0.1	0.443	1.5	35.9	26.3	8.5	28.6	70.3
MSWC-3	7.5	2.2	0.7	0.6	1.1	0.3	0.7	0.2	0.1	1.070	1.4	60.5	48.0	6.4	54.6	91.9
VC-man	7.5	3.7	0.6	0.5	0.8	0.6	1.1	0.3	0.1	0.570	1.8	18.1	15.2	3.1	21.2	80.6
<sup>1</sup> Min	7.50		2.0 (2.5)	0.5 (1.0)	2.00					0.2 (0.5)					10(100)	10(100)
<sup>2</sup> Max											3.0	$2.0^{4}$	50.0	120.0	1500.0	600.0

<sup>1</sup> minimum value to be claimed as an OMF (final product), according to EC (2019); in the case of N, higher N and N<sub>org</sub> values correspond to OMF containing only N as macronutrient; in the case of Ca, Mg, Na and S, the reported threshold refers to Compound inorganic fertilizers, as there is no limitation on OMFs; in the case of Na, the value reported in brackets refers to the maximum allowed concentration.

<sup>2</sup> maximum value admissible in an OMF (final product), according to EC (2019).

<sup>3</sup> Cu and Zn can be considered both contaminants and micronutrients. The EU regulation 2019 provides limits in mg kg<sup>-1</sup> DM for contaminants while expressing minimums in % by mass for microelements.

<sup>4</sup> as Cr VI, while here the analysis was made on total Cr.

or S source. Peat, GC-hort, MC-man1, VC-man, MSWC-1, MC-man2, GC-fung, and GC-prun contain more than 1 % of total CaO that may help regulate soil pH. While for micronutrients, GC-hort, MC-man1, VC-man, and MSWC-3 would contribute with Fe (0.5 %), Cu (0.01 %), and Zn (0.01 %) to an OMF and could be used for grassland fertilization, while almost all the rest of materials would provide minimum Fe (0.02 %), Cu (0.001 %), and Zn (0.001 %) values for horticultural use. Biochar and leonardite materials do not provide Fe concentrations for grassland or horticulture. Depending on the target OMF use, most organic materials can supply microelements; however, not for a full range of microelements, nor in high concentrations.

Generally, OMF processing pays attention to organic matter content laid out by C<sub>org</sub> concentration in the organic material. However, no less importance must be attributed to organic matter quality, as indicated by the humic C content and the humification degree. Humic-related substances have been reported to enhance crop growth by influencing microbial and physicochemical properties of soil [54], stimulating plant metabolism [55,56], and helping remediate soil contaminants owing to a high sorption capacity [57,58]. Therefore, OMFs with high humic C are desirable, and, as Peat tends to have a high content of humic C and a high humification degree [59], it has been claimed as a high-quality material in OMFs [34], even preferable to other organic alternatives [60]. In addition, when humic C represents more than 50 % of total organic C, the material has achieved a good humification process and, therefore, high chemical stability. However, no references indicate what humic C value or humification degree needs an organic material to generate an effect in an OMF. Among all the analyzed materials, VC-man, MSWC-1, MC-man2, and Leon would provide the highest humic C content (3.3–4.1 %) and could replace peat in OMF (Table 3, Table 4 and Table S4).

Another important characteristic of the organic fraction is its chemical stability, i.e., the capability to resist bio-driven changes over time. There is no unique method to measure the stability of the materials, and the humification degree and the C/N ratio have been used as proxies [13,61,62]. The humification degree, which indicates the transformation of the base-extractable fraction into stable humic and fulvic acids, shows that except for GC-fung, all tested organic materials have values > 50 %, and Peat, MSWC-1, and MC-pom, even >70 %, indicating the presence of a low amount of fermentable organic compounds (TEC – (HA-C + FA-C)) such as sugars and oligopeptides [3,11]. The C/N ratio is for all samples  $\leq 20$ , confirming the good degree of decomposition for biowastes. The humification degree and C/N ratio cannot be used to establish the degree of decomposition for biochar samples since they are a product of pyrolysis and thus characterized by intrinsic stability due to the formation of polycondensation aromatic rings during pyrolysis [63, 64]. Therefore, BC-1 and BC-2 fulfill the stability required for OMF production.

The alkyl C/O-alkyl C ratio (Table 4) is used as a marker of compost maturity [65,66], as during composting, alkyl C increases due to the selective enrichment of the most recalcitrant alkyl components, while O-alkyl C decreases due to polysaccharides hydrolysis and sugar oxidation. The alkyl C/O-alkyl C ratio confirms that Leon is a more stable material (14.4) than Peat (0.9) due to the dominance of highly recalcitrant alkyl moieties and low presence of saccharide compounds [14]. Among the other materials, the alkyl C/O-alkyl C ratio ranged between 0.3 and 0.8, independent of the raw material and class of organic materials. Differences can also be observed in the 111–155 ppm region, which represents the aromatic-C and may be related to the presence of lignin resistant to microbial decomposition [67]. These differences were expected due to the diversity of the raw materials used for composting, composting time, and conditions [68,69]. Moreover, MSWCs are materials that, before composting, underwent a digestion process that increased their chemical stability [70–72], as confirmed by the alkyl C/O-alkyl C ratio of MSWC-1 and MSWC-3 compared to composted materials with a similar particle size distribution in the fraction above 2 mm (GC-zeo, GC-hor, MC-man1).

Although organic matter stability -measured as compost maturity-is desirable to provide OMF uniformity over time, the organic matter must be able to react with the mineral fertilizer to enhance mineral nutrient protection. A higher polarity in organic matter could produce a higher concentration of complexing of N and P chemical species [73]. Organic structures such as O-alkyl C, phenolic C, and carboxyl C present high polarity [74]. In Tables 4 and it is observed that the polarity of the organic materials will be highly influenced by the O-alkyl C, and in lower degree by the carboxyl C. Biowastes materials had a higher content of O-alkyl C + phenolic C compared to Peat. Still, in biowastes with high maturity, the difference was only 6–10 %, while the content of Leon was 30 % lower than Peat. Selecting materials that balance both values is necessary rather than looking only for polarity or stability.

# 4.3. Contaminants and chemical properties

Contaminants were left as the last section to discuss since their content in the OMF will depend on the proportion of organic material used. Only after the OMF is designed can it be understood how much salinity, pH, or contaminants will be in the OMF.

An OMF should be free from harmful substances such as PTE, pathogens, and other contaminants that could threaten human or environmental health [1]. An organic material that produces an OMF can have concentrations above the contaminants set thresholds because its concentration is diluted once mixed with the mineral fertilizer and potential fillers. An OMF will be safe to use in agriculture if the sum of the contaminants of each fraction is below the final product's threshold, being more relevant if the OMF contains P-fertilizers as those are generally a significant source of trace metals [75]. Therefore, it is better to consider the contaminant contribution to the target OMF, which is exemplified as a 7.5 %  $C_{org}$ -OMF (Table 5).

Some analyzed organic materials had a high content of heavy metals. GC-fung would provide 4 mg Cd kg<sup>-1</sup> DM and 59.0 mg Ni kg<sup>-1</sup> DM to the target OMF, thus making this material unsuitable. Also, P fertilizers in Europe can contain between 0.7 and 42 mg Cd kg<sup>-1</sup> and up to 195 mg Ni kg<sup>-1</sup>, although the mean concentrations are 7.4 mg Cd kg<sup>-1</sup> and 14.8 mg Ni kg<sup>-1</sup> DM [76]. Therefore, materials like MC-man1, MSWC-3, and GC-prun, which would contribute 2.7 mg Cd kg<sup>-1</sup> DM, 48.0 and 39.5 mg Ni kg<sup>-1</sup> DM, should not be considered to produce OMFs containing phosphate fertilizers. BC-1 is the only material above the threshold of Pb; however, its use could be safe if OMF contains less than 66 % of BC-1 (47 %% of C<sub>org</sub>). All analyzed materials have at least four times less Cu and three times less Zn than the limits in OMFs. Phosphate fertilizers cannot be considered a significant Pb, Cu, or Zn source [76,77]; therefore, there is no risk of contamination with Pb, Cu, and Zn through the P fertilizer for any of the analyzed organic materials.

High salinity in fertilizers can cause plant stress, including reduced water uptake, nutrient imbalances, and seed germination inhibition [78]. In addition, high salt levels in fertilizers can contribute to soil salinization over time [79]. No specific salinity level in a fertilizer can be considered toxic, as the toxicity depends on each crop and growing conditions [78]. However, a recommended threshold is 4 mS cm<sup>-1</sup>, which is a tolerable value for crops with a medium sensitivity for salinity [80]. Organic materials can have a high content of salts depending on their source and processing and surpass this proposed threshold [81], thus representing a risk if used for direct fertilization. Among all the analyzed materials, GC-hort ( $4.0 \text{ mS cm}^{-1}$ ) and MC-man2 ( $4.8 \text{ mS cm}^{-1}$ ) would not be suitable for OMFs used in crops with salinity sensitivity. Additionally, those values could be a problem in OMFs made with mineral fertilizers like ammonium nitrate, ammonium thiosulfate, or potassium chloride, which have a salt index above 90 [82,83]. None of the other analyzed materials would contribute significantly to the OMF salinity, making them suitable for mixing with mineral fertilizers.

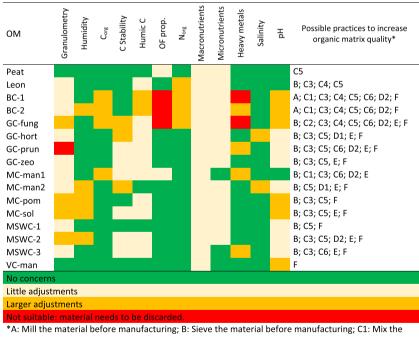
It is well known that a high pH in the microsites where the fertilizer is present in soil can increase N losses from fertilizers through NH<sub>3</sub> losses [84,85]. When applied together, biochars with neutral pH would reduce the NH<sub>3</sub> volatilization from mineral fertilizers compared to more alkaline fertilizers [86]. Therefore, organic materials with a pH closer to neutral should be preferred to strongly alkaline organic materials for OMF production. With this consideration, MC-man1 and Leon are ideal over other materials, with characteristics close to Peat.

Some chemical properties of the organic materials can be managed before adding them to OMFs. The pH of organic materials can be reduced by adding elemental sulfur, sulfuric acid, or large volumes of zeolite that also reduce the salinity by absorbing  $Na^+$  and  $Cl^-$ [87].

Only some of the analyzed samples can be used to manufacture OMFs with 7.5 % C<sub>org</sub>, however, the quality of the organic material can be improved for this type of OMF or others with higher C<sub>org</sub> content. Fig. 4 summarizes all the parameters discussed in the present work and proposes methods to increase the quality of organic materials.

# 5. Conclusions

By grouping physical and chemical characteristics into limiting parameters for OMF production or use, organic matter characteristics, and nutrient contribution, we proposed a method that gives criteria to select organic materials for the following steps of research for creating organo-mineral fertilizers. The possibility of discarding an unsuitable organic matrix early to produce OMFs



\*A: Mill the material before manufacturing; B: Sieve the material before manufacturing; C1: Mix the material with other organic materials to reduce C<sub>org</sub> content and increase proportion in OMF; C2: Mix the material with other organic materials to increase C<sub>org</sub> content and increase proportion in OMF; C3: Mix the material with other organic materials to increase C<sub>org</sub> content and increase proportion in OMF; C3: Mix the material with other organic materials rich in Humic C; C4: Mix the material with other organic materials rich in Humic C; C4: Mix the material with other organic materials rich in Humic C; C4: Mix the material with other organic materials rich in Humic C; C4: Mix the material with other organic materials with low heavy metals content; D1: Avoid mixing it with fertilizers with high salinity as ammonium nitrate, ammonium thiosulfate, or potassium chloride; D2: Avoid mixing it with P and K fertilizers with high heavy metals content; E: Prolong composting period to increase Corg stability and concentration; F: Acidify the material before manufacturing

Fig. 4. Summary of organic materials (OM) suitability for 7.5 % C<sub>org</sub> OMF production. Based on the analysis in Tables 2–4, each parameter received a color to indicate if the organic material requires adjustments or is discarded for OMF production.

reduces the formulation and testing costs and time, increasing the efficiency of the following characterization when real new OMFs are created.

Using standard physicochemical analyses, it is possible to determine the early suitability of organic materials for producing organomineral fertilizers (OMFs) with 7.5 % organic carbon ( $C_{org}$ ). Stability indexes such as the alkyl C/O alkyl C ratio or humification degree can adequately indicate biowaste maturity. High values in parameters such as granulometry, salinity, or pH can indicate that the organic material requires a greater manipulation of materials to be used directly in the OMFs. Values like the  $C_{org}$  or organic nitrogen ( $N_{org}$ ) help to analyze if the organic material requires a mixture with a secondary organic material to enhance its properties. The nutrient analysis showed that in low  $C_{org}$  OMFs, the organic material cannot provide a satisfactory quantity of macronutrients as P or K. Still, some micronutrients can be present in quantities that allow labeling the OMF as a micronutrient source. With the selected parameters, we can propose that only some organic materials from the analyzed samples are suitable for organo-mineral fertilizer (OMF) production. Among the analyzed waste materials, GC-zeo, VC-man, and MSWC-1 are the most promising to substitute organic geogenic materials in OMF production.

Among the studied materials, some seemed promising replacements for peat for most of the parameters. However, one or more variables (such as pH, salinity, and instability) had undesirable values. Different approaches can be chosen to correct some chemical and physical parameters, such as adding correctives, although they can create the inconvenience of reducing the formula space in OMFs.

After screening local materials, considering it the first step, it is necessary to create granular OMFs and evaluate nutrient release dynamics and agronomic performance. With the results of those tests, it will be possible to consider whether the list of parameters analyzed in this work can be correlated to the OMF performance test or, if necessary, to increase the number of parameters to be analyzed. Alternative parameters measured in the organic matrix such as pathogens, microplastic content, or antibiotic residues may require consideration, as the current regulations set strict limitations of the final OMF product quality based on these parameters.

# CRediT authorship contribution statement

**T.J. Sitzmann:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization, Writing – review & editing. **L. Celi:** Writing – review & editing, Methodology, Formal analysis, Conceptualization, Data curation, Supervision, Writing – original draft. **B. Moretti:** Writing – review & editing, Methodology, Formal analysis, Conceptualization, Data curation, Writing – original draft. **E. Padoan:** Writing – review & editing, Methodology, Formal analysis, Conceptualization. **S. Tagliavini:** Formal analysis, Conceptualization, Methodology. **L. Zavattaro:** Writing – review & editing, Supervision, Methodology, Conceptualization, Data curation, Data curation, Data curation, Project administration, Conceptualization, Data curation, Project administration, Methodology, Resources.

# Data availability statement

All data are included in the article and supplemental materials.

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# Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:The author S. Tagliavini works with one of the companies that provided one of the organic materials analyzed in this study; however, the authors declare that no financial or personal interests influence the results reported. If there are other authors, they 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 data

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