



# Evaluating the properties of deinking paper sludge from the Mediterranean area for recycling in local areas as a soil amendment and to enhance growth substrates

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## Abstract

Recycling of paper generates large quantities of paper sludge that is rich in organic matter and can be exploited to counteract organic matter loss in agricultural soils, especially in some geographical areas, such as the Mediterranean. In order for deinking paper sludge (DPS) to be recycled, chemical and physical characterizations are required along with an analysis of possible contaminants. DPS from Italy and Tunisia, resulting from various deinking processes, were analyzed and compared in terms of their chemical properties (pH, OM, total N, C/N ratio, mineral nutrients, and trace metals), surface chemistry [diffuse reflectance infrared Fourier transform (DRIFT) spectroscopy and Fourier-transform infrared (FTIR) spectroscopy], and thermal stability [thermogravimetric analysis (TGA)]. The aim was to improve their use as amendments in soils or growth substrates. Elemental analysis revealed higher carbon concentrations in the Tunisian DPS and similar nitrogen contents, high mineral nutrient levels, and potentially toxic trace element levels below the limits allowed for soil amendment. No evidence was found for the presence of organic pollutants, pathogens, or toxic heavy metals. Surface chemical analysis revealed comparable materials rich in aromatic, phenolic, aliphatic, and polysaccharide structures along with clay minerals and carbonates. The thermal analysis revealed similar decomposition temperatures for the organic matter. Practical implications of the results are discussed, highlighting the similar properties of sludge following ink removal by different processes and the need to integrate the N content in order to be able to employ sludge as an amendment in soil and growing media. Given the similarity of the compositions of the materials, appropriate DPS management is key to improving soil fertility, reducing paper waste disposal, and implementing a circular economy in the Mediterranean, where climate change is having a significant impact.

**Keywords** Chemical properties · Deinking process · DRIFT mode · Nitrogen and organic matter content · Surface chemistry · Thermal stability

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## Introduction

Wastepaper is an important raw material that is used as a feedstock for the paper industry, and its recycling is strongly promoted to implement the fundamental principles of the circular economy. The Confederation of European Paper Industries (CEPI 2023) revealed that 70.5% of all paper and cardboard consumed in Europe was recycled in 2022, the world's highest paper recycling rate, with a commitment to recycle 76% of all paper consumed in Europe by 2030. However, the production of recycled paper generates a large amount of solid waste, which is mainly produced from pulping, deinking, and wastewater treatment. This waste is referred to here as deinking paper sludge (DPS). In the 2000s, the waste generated from recycled papermaking was estimated to be as high as 150 kg of dry solids/tonnes of manufactured recycled paper (Dahl et al. 2008). However, depending on the type of new paper produced, the extent of the generated solid residual waste varies, ranging from 170 to 600 kg (on a dry weight basis) for transforming 1 tonne of newsprint into tissue paper (Bajpai 2015). Although wastepaper recycling has a positive effect on the circular economy, the solid waste sludge generated represents an environmental and economic burden for recycled paper mills. The environmental concern is related to the leaching of toxic substances into the ground and the emission of greenhouse gases into the atmosphere (Faubert et al. 2016). In addition, landfilling or incineration of DPSs results in the loss of material with economic value and the ability to deliver ecosystem services (Zhang et al. 2015). The characteristics of the DPS and the media where it could act as an amendment are of paramount importance.

The Kyoto Protocol indicates that waste management and the use of residual fertilizing matter (RFM) are ways to reduce greenhouse gases and mitigate climate change (United Nations Climate Change—Kyoto Protocol). Back-to-Earth Alternatives, an integrative approach to return to the environment and close the material cycle in the context of a circular economy, support, for example, the use of DPS in the restoration of degraded mine sites (Fiero et al. 1999) or the use of mixed treatments of DPS with N fertilizer in agricultural and horticultural crops (Poornima et al. 2022). Results of abandoned sand pits treated with DPS revealed an increase in very low pH values, as well as improvements in water retention, cation exchange capacity, and bulk density (Fiero et al. 1999). The use of DPS as a soil amendment in cereal cultivation enhances grain yield and increases root biomass in agricultural soils (Marouani et al. 2021). Using DPS in calcareous soils at 60 Mg ha<sup>-1</sup> was found to mitigate CO<sub>2</sub> emissions, at 30 Mg ha<sup>-1</sup> to improve soil fertility (Marouani et al. 2020), and at 72 Mg ha<sup>-1</sup> to reduce erosion in agricultural soils with low organic matter (Rasa et al. 2021).

However, to maintain safety in terms of the environment and human health, some countries now regulate the use of DPSs as soil amendments (Gibbs et al. 2005). In fact, the properties of DPS may differ in their intrinsic composition, depending on the deinking process involved (mechanical or chemical pulping, with or without bleaching), the kind of wastepaper used (e.g., a printer, tissue paper, office paper, magazine paper, etc.) and, consequently, the ink types to be removed from fibers (such as oil-based and water-based inks, photocopy and laser-print toners, etc.), as well as paper coatings and fillers already present in the wastepaper, such as kaolinite (an aluminum silicate clay), talc (a magnesium silicate clay), calcium carbonate and titanium dioxide, and the final quality of the paper product (bleaching, fillers, and coatings) (Turner et al. 2022; Kaur et al. 2020; Simão et al. 2018; Hubbe and Gill 2016; Nie et al. 1998). Clay and carbonates are the most common fillers used in papermaking, together with kaolin, talc, and TiO<sub>2</sub>. These materials reduce the shrinkage of the paper pulp during drying (Gill 1995). Kaolin is used to improve the appearance of the paper and printability by producing high-quality printable paper in which the clay fills the gaps between paper fibers, improving smoothness, opacity, brightness, and ink receptivity (Shen et al. 2009). These paper components are present in high amounts in sludge (DPS), both when wastepaper is used in recycled mills and when wastepaper is added to the process as a filler or coating to increase the quality of the new paper produced.

Deinking processes are designed to remove inks and other impurities from wastepaper using mechanical or chemical techniques or in combination, allowing the production of high-quality paper products (Tsatsis et al. 2017). Mechanical deinking involves physical forces acting on wastepaper during the processes of repulping, flotation, washing, screening, and centrifugal cleaning. Froth flotation is the most commonly used physical deinking technique and is particularly sustainable since air is injected into a mixture of wastepaper and water, creating a froth where ink molecules adhere to air bubbles and permitting the mechanical removal of inks and other contaminants (Bajpai 2014a; Tsatsis et al. 2017; Yang et al. 2022). Chemical deinking techniques use chemicals with specific functions, such as fiber wetting and swelling, ink removal, anti-redeposition, dispersion, flocculation, agglomeration, and oxidation and/or reduction of chromophores (Bajpai 2014b; Tsatsis et al. 2017; Yang et al. 2022). For example, alkali agents (sodium hydroxide and sodium silicate) have been used to break down inks by saponification or hydrolysis and detach them from paper fibers; enzymes, such as carbohydrate hydrolases used for cellulose and hemicellulose hydrolysis; solvent and chelating agents, which can form soluble complexes with heavy metal ions; surfactants, which help break ink–particle bonds and facilitate separation from fibers; and calcium salts to aid the agglomeration and

the formation of large hydrophobic clusters for ink separation, arriving from hydrophobic ink particles derived from ink collectors. After ink removal, the deinked fiber may be bleached with oxidative agents (usually hydrogen peroxide and pressurized O<sub>2</sub>) to increase the whiteness and purity, and to prevent yellowing of the newly formed paper by the addition of sodium hydroxide. Reductive agents, typically sodium hydrosulfite or formamidine sulfinic acid, are also used for brightening and color stripping of secondary fibers, although oxidative agents are more efficient at bleaching deinked pulps (Bajpai 2014a, b, c; Flicker 2007). Finally, the generated DPS is dewatered, resulting in a more concentrated and manageable sludge. Thus, the composition of DPS could reflect not only the chemicals and additives used in the deinking process but also the kind of wastepaper used, where additives or impurities will end up, to different extents, in the wastewaters and the solid waste generated. Consequently, DPS has a hybrid composition. It is made up of short cellulosic fibers and inorganic materials, both of which are sources of recalcitrant organic matter and mineral nutrients and help improve soil quality and plant development.

DPS thus has a high potential as an amendment. It improves soil health by increasing soil porosity and aggregation stability in agricultural soils (Camberato et al. 2006; Marouani et al. 2021); decreasing the water holding capacity, cation exchange capacity, and pH in an abandoned sandpit (Fiero et al. 1999); decreasing the soil bulk density; and increasing the soil fertility in combination with or composted with other high-nutrient byproducts (Camberato et al. 2006). In Italy, DPSs are permitted for environmental restoration at a maximum of 30%, as stated by Italian and EU regulations (DM 22/1998 (1998); EU 2014). However, there are constraints related to the low content of nitrogen (<0.5 mg kg<sup>-1</sup>). Moreover, when DPS is spread in crop fields, it can affect growth and yield in the first year of spreading, owing to N mineralization. Co-composting (Camberato et al. 2006) or composting with green manure (Marouani et al. 2019) is one way of overcoming such constraints.

Using DPS as a soil amendment in the Mediterranean area would be particularly beneficial. In fact, Mediterranean soils undergo drought, salinization, erosion, and desertification in response to the concomitant overexploitation of agricultural soils and higher temperatures (Ferreira et al. 2022). Global change model projections (Boulet 2016) indicated the Mediterranean area as one of the most affected by climate change: longer droughts and more frequent extreme precipitation events are likely to occur in this region, thus increasing the risk of soil organic matter losses. Understanding the deinking process and determining contaminant levels exceeding the limits set for RFM are thus challenging when managing sludge from the recycling paper industry. However, similarities in their main characteristics, irrespective of their origin/production process, should help international

strategies for DPS management and safe use in a circular economy. Within this concept, the use of DPS should consider the use of waste in neighboring areas both to limit transport and landfill disposal costs and to reduce other transport costs, such as redirecting waste to further processing areas (composting areas) and/or agricultural areas (as a soil improver) (Turner et al. 2022).

The novelty of the present study is in combining these two important environmental aspects, i.e., contextualizing the study in the Mediterranean area, where it is challenging to counteract the OM loss in soils affected by climate change, and to recycle DPS close to where the sludge was generated to reduce energy-related CO<sub>2</sub> emissions. Our aim was thus to characterize two different DPSs from recycled paper mills in the Mediterranean area—in Tunisia (TUN-DPS) and Italy (IT-DPS). Thus, the functional properties of both DPSs were evaluated for subsequent use as an organic improver in soils and growth substrates particularly exposed to climate change as is the Mediterranean area.

## Materials and methods

### Origin of paper mill sludge

Tunisian DPS (TUN-DPS) was obtained from a paper and tissue manufacturer in Sousse (Tunisie Ouate) and generated by a chemical deinking process. Tunisie Ouate was founded in 1982. The company provides manufacturing, conversion, and marketing services mainly for tissue paper made from pure cellulose and recycled paper, thus combining pulps from wood, raw, and nonrecycled (mainly from Canada) with those from wastepaper. It has a production capacity of for more than 40,000 tons a year, supplying first-class tissue paper products to the regions of Maghreb (Libya, Tunisia, Algeria, and Morocco), the Middle East, and Western Europe ([www.tunisieouate.com/](http://www.tunisieouate.com/), assessed in June 2023). An Italian DPS (IT-DPS), provided by a recycled paper mill (Lucart S.p.A.) located in Lucca, Tuscany, was generated by a mechanical deinking process. Lucart was founded in 1953 and in 1966 began building a series of plants specifically designed for the deinking of wastepaper and the purification of wastewater. The company produces 60,000 tons of recycled and reclaimed paper annually (<https://www.lucartgroup.com/en/>, assessed in June 2023).

TUN-DPS and IT-DPS were supplied in a dry form as raw materials without any other pretreatment and were classified with a nondangerous waste code. Waste codes were established by the European Commission Decision 2000/532/EC1 to categorize waste types in accordance with a standard framework, including level of health and environmental hazards, and based on a combination of composition and production processes (EU Commission 2014). The European

waste code 03 03 05 (Deinking sludges from paper recycling) and the waste code 03 03 10 (Fibre rejects, fibre-, filler- and coating-sludges from mechanical separation) were attributed to the TUN-DPS and IT-DPS, respectively. Code 03 03 10 permits use in environmental restoration or soil amendment at a certain limit, fixed in Italy at a 30% maximum. In Tunisia, the application of biosolids on agricultural land is limited by law to only experimental plots conducted as pilot demonstration projects (Normes Tunisiennes (NT) 106.20-2002). DPS has been used in Canada as a residual fertilizing material (RFM) in agriculture, soil restoration, and land management. In Quebec, the use of DPS follows a “Guide sur le Recyclage des Matières Résiduelles Fertilisantes” (Hébert 2015) for recycling and using RFM in the agricultural, forestry, and ornamental horticulture sectors, and sets out applicable criteria and standards. For this reason, we adopted the Canadian limits as a reference for the TUN-DPS.

### Quantitative analysis—physical and chemical characterization

The main chemical and physical properties [bulk density (BD), pH, and electrical conductivity (EC)] of the air-dried IT-DPS and TUN-DPS were determined following European standard methods for soil improvement and growing media. Accordingly, the laboratory BD (CEN 13041:2012 method) was determined by the ratio of the mass of DPS filling the test cylinder with a nominal capacity of 1 L. The pH (CEN 13037:2012 method) was measured after 1 h of shaking in grade 2 water, i.e., with a specific conductivity not higher than  $0.2 \text{ mS m}^{-1}$  at  $25 \text{ }^\circ\text{C}$  and a  $\text{pH} > 5.6$  at a ratio of 1:5 v:v. A pH meter (pH 211, Hanna Instruments) previously calibrated using Hanna pH standard buffer solutions (pH 7.00 and pH 4.01) was used. EC measurements (CEN 13038:2012 method) were carried out after filtration of the same extract prepared for pH analysis using a Crison GLP31 previously calibrated with standard solutions ( $1413 \text{ mS cm}^{-1}$  and  $12.88 \text{ mS cm}^{-1}$ ) and a temperature correction set at  $25 \text{ }^\circ\text{C}$ .

Total carbon, nitrogen, hydrogen, and sulfur ( $C_{\text{tot}}$ ,  $N_{\text{tot}}$ ,  $H_{\text{tot}}$ , and  $S_{\text{tot}}$ ) as well as organic carbon ( $C_{\text{org}}$ ) were determined through the dry combustion method (ASTM Method D5373-21) using a Flash Smart Elemental Analyzer (Thermo Fisher Scientific) with a detection limit of 0.01%. In addition, the pseudototal concentrations of nutrient elements (P, K, Ca, Mg, Na, Fe, and Mn) and trace heavy metals (As, Cu, Cd, Cr, Hg, Ni, Pb, and Zn) were determined according to EPA Method 3051A (U.S. EPA 3051A 1997) via modification with the addition of hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) to aid in the decomposition of organic matter. DPS samples were acid digested with  $\text{H}_2\text{O}_2$  (30%, v/v) and  $\text{HNO}_3$  (65%, ratio 1:2.5) in closed Teflon vessels for microwave digestion using a laboratory station

microwave oven model ETHOS 900 (Milestone, USA). Elements in the filtrated extracts were analyzed by inductively coupled plasma emission spectroscopy (ICP–OES 5900, Agilent Scientific Instruments, USA) according to EPA Method 6010C (US EPA 6010C 2007). The detection limit of ICP–OES was  $0.02 \text{ mg L}^{-1}$  for K, Ca, Mg, Na, Fe, and Mn;  $0.01 \text{ mg L}^{-1}$  for As, Cd, Cu, Cr, Ni, and Pb; and  $0.01 \text{ } \mu\text{g L}^{-1}$  for Hg. The pseudototal phosphorus concentration in the same extract was determined colorimetrically according to Murphy and Riley (1962) and using a UV–Vis 1900i spectrophotometer (Shimadzu, Kyoto-Japan) at 720 nm, with a detection limit of  $0.02 \text{ } \mu\text{g L}^{-1}$ . All analyses were carried out in triplicate. Statistical analyses were performed using R version 4.2.2 (R version 4.2.2 Copyright (C) 2022) to calculate the average mean and standard deviation and determine the significant differences between means using a *t*-test ( $p < 0.05$ ).

Organic contaminants such as polychloride biphenyls (PCBs), dioxins, and furans were extracted from DPS samples following EPA Methods 3545A and 1613B (US EPA 3545A 2007; US EPA 1613B 1994) using a Soxhlet extractor system in a 1:1 hexane:acetone solvent. Extracts were concentrated using a stream of  $\text{N}_2$  and analyzed by gas chromatography-electron capture detector (GC/ECD) according to EPA 8082A (US EPA 8082A 2007) using the Varian GC model CP-3800 equipped with a 63Ni electron capture detector of 15 mCi activity with an autosampler. In addition, biological characterization through the analysis of fecal coliforms and *Escherichia coli* using a membrane filtration method (*E. coli*  $\text{g}^{-1}$ ) and confirmation of the presence or absence of *Salmonella* sp. were performed on TUN-DPS according to the most likely number (MLN) method (Hébert 2015).

### Qualitative analysis

#### Fourier-transform infrared (FTIR) spectroscopy

The functional groups in both DPSs were identified through FTIR, which was conducted in diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) mode using an infrared spectrometer (IR Tracer-100; Shimadzu, Kyoto-Japan). DPS samples were previously dispersed in KBr (ratio 1:200) to produce pellets under vacuum using pure KBr as a reference material. The samples and KBr were dried at  $75 \text{ }^\circ\text{C}$  before the pellet preparation to limit moisture interference. The spectra were recorded for each sample by collecting 128 scans in the wavenumber range of  $4000$  to  $400 \text{ cm}^{-1}$  at  $4 \text{ cm}^{-1}$  resolution. For comparison, the spectra of both DPS samples were baseline corrected and normalized in absorbance mode.

**Table 1** Main chemical and physical properties of the Italian and Tunisian DPS

Parameter	IT-DPS	TUN-DPS
Bulk density (kg L <sup>-1</sup> )*	0.34 ± 0.01	0.43 ± 0.01
pH (H <sub>2</sub> O)	8.0 ± 0.1	7.9 ± 0.2
EC (mS m <sup>-1</sup> )	191.0 ± 4.5	204.3 ± 7.9
C <sub>tot</sub> (%)*	21.76 ± 0.40	34.90 ± 0.71
C <sub>org</sub> (%)*	19.00 ± 0.80	31.50 ± 0.65
N <sub>tot</sub> (%)	0.39 ± 0.06	0.37 ± 0.08
S <sub>tot</sub> (%)	0.57 ± 0.06	0.66 ± 0.11
H <sub>tot</sub> (%)	1.85 ± 0.18	2.17 ± 0.27
C/N ratio	48.72	85.13

Asterisks denote significant difference of the analyzed parameter between the two DPS, according to the *t*-test ( $p < 0.05$ )

**Table 2** Pseudototal concentration of mineral nutrients in the Italian and Tunisian DPS

Element	IT-DPS	TUN-DPS
P (mg kg <sup>-1</sup> )*	220 ± 14.1	350 ± 20.0
K (g kg <sup>-1</sup> )	0.324 ± 0.06	0.400 ± 0.09
Ca (g kg <sup>-1</sup> )*	207 ± 6.30	270 ± 19.0
Mg (g kg <sup>-1</sup> )	3.99 ± 0.19	4.49 ± 0.40
Na (mg kg <sup>-1</sup> )	174 ± 7.80	180 ± 30.0
Fe (mg kg <sup>-1</sup> )*	537 ± 38.0	1060 ± 100
Mn (mg kg <sup>-1</sup> )*	48.1 ± 2.69	520 ± 99.0

Asterisks indicate a significant difference among means following *t*-test analysis ( $p < 0.05$ )

**Table 3** Potentially toxic elements, organic contaminants, and pathogens in IT- and TUN-DPS and legal limits in Italy and Canada

Contaminants (d.w. basis)	IT-DPS	TUN-DPS	Limits and Regulations	
			IT DM 22/1998 EU Commission 2014a	CAN <sup>a</sup> (Hébert 2015)
As <sub>tot</sub> (mg kg <sup>-1</sup> )	< 5	< 1.5	5.0	13
Cd <sub>tot</sub> (mg kg <sup>-1</sup> )	0.2	0.08	1.5	3.0
Cr <sub>tot</sub> (mg kg <sup>-1</sup> )	< 5	8.05	0.5 (Cr <sup>6+</sup> )	210
Cu <sub>tot</sub> (mg kg <sup>-1</sup> )	50.0	51.4	150	400
Hg <sub>tot</sub> (mg kg <sup>-1</sup> )	< 0.1	< 0.2	1.5	0.8
Ni <sub>tot</sub> (mg kg <sup>-1</sup> )	< 10	2.01	30	62
Pb <sub>tot</sub> (mg kg <sup>-1</sup> )	< 0.5	< 10	40	120
Zn <sub>tot</sub> (mg kg <sup>-1</sup> )	38.0	101	500	700
PCBs (mg kg <sup>-1</sup> )	< 5	nd	5	5
Dioxins; Furans (ng EQT kg <sup>-1</sup> )	< 5; < 50	3.3; nd	5; 50	17
<i>Escherichia coli</i> (UFC g <sup>-1</sup> , dry matter)	< 10	< 10	–	< 1000
<i>Salmonella</i> sp. (fresh matter)	Absent for 10 g	Absent for 10 g	–	< 3

<sup>a</sup>Unrestricted use, category C1

## Thermal analysis

The thermal behavior of the DPS was determined via thermogravimetric analysis (TGA) and differential thermogravimetry (DTG) using platinum crucibles in air injection (combustion) on a Q600 analyzer (TA Instruments). Heating was performed from 25 to 900 °C at a rate of 5 °C min<sup>-1</sup> and with an isothermal stage of 5 min at 800 °C. An airflow rate of 100 cm<sup>3</sup> min<sup>-1</sup> was maintained during the analyses.

## Results and discussion

### Characterization

The physical and chemical properties of the DPSs, based on the dry weight of the materials, are presented in Tables 1, 2, and 3. Table 1 presents the main basic properties of IT-DPS and TUN-DPS and reveals significantly higher values of BD and total and organic C in the TUN-DPS. No significant differences were found in pH, EC, or total N, S or H. Although significantly different, the BD of both materials was low, characterizing a loose, highly porous, and fibrous organic-rich material, reflecting the particular mixture of fibers, fillers, inks, and chemicals used to separate fibers and remove the ink from wastepaper. A greater amount of inorganic and/or nonfibrous materials used in the chemical deinking process could give rise to a sludge with a higher bulk density (Jele et al. 2022; Migneault et al. 2010). On the other hand, the mechanical action of repulping wastepaper could promote a greater proportion of shorter fibers and denser structures, decreasing the bulk density of the sludge (Jele et al. 2022; Migneault et al. 2010). Thus, a valid explanation for

the slightly higher BD of TUN-DPS (Table 1) could result from the nonfibrous content, i.e., compounds used to help in wastepaper repulping and deinking operations. Clays and calcium salts (used as fillers or coatings) in the wastepaper and higher in TUN-DPS could also be the origin of a higher value of the BD. The higher BD values are thus unrelated to the specific deinking process.

The carbon content in IT-DPS and TUN-DPS is relatively high (approximately 22% and 35%, respectively), owing to their organic nature, i.e., both materials have a high content of lignocellulosic fibers. In contrast, the inorganic carbon in the DPSs (approximately 2.76% and 3.40%, respectively, for IT-DPS and TUN-DPS) probably relates to the presence of carbonate (usually  $\text{CaCO}_3$ ) (Bajpai 2014b; Hubbe and Gill 2016). The higher content of C in the TUN-DPS could be attributed to two factors: (1) the higher content of fibrous material characteristic of the sludge generated in the chemical process than in the mechanical process, and (2) the possibly greater presence of  $\text{CaCO}_3$  as a filler or coating from the wastepaper, which is not attributable to a specific deinking process (chemical or mechanical).

The alkaline pH values in the sludge (Table 1) were comparable to those of other deinking sludges in the range of 7.6–7.8 (Beauchamp et al. 2002). However, neither mechanical nor chemical processes had a greater effect on the pH of the DPSs. In general, sodium hydroxide, sodium silicate, and hydrogen peroxide are responsible for the alkaline pH of sludge (Jele et al. 2022; Bajpai 2014b; Nasnar et al. 2021). Sodium hydroxide is used to swell fibers and separate ink from fibers or to promote a saponification reaction of ink binders; sodium silicate is used to prevent ink redeposition on fibers; and hydrogen peroxide acts in the processes of fiber deinking and bleaching (Jele et al. 2022; Bajpai 2014b; Nasnar et al. 2021).

The high EC found in IT-DPS and TUN-DPS (Table 1) reflects the soluble salts/dissociated additives used in the processes of pulping, deinking, and bleaching of wastepaper and can be considered a proxy of potential fertility in terms of the presence of mineral nutrients available to plants and microorganisms (Vannucchi et al. 2018). Nitrogen and S are generally very low in deinking paper sludge (Deviatkin et al. 2016; Manirakiza et al. 2019), and the DPS investigated here reflects this, giving rise to high C/N ratio (Table 1), which in turn indicates a need for N supply if DPS is to be applied to agricultural lands (Turner et al. 2022) or used as an amendment in nursery substrates (Vannucchi et al. 2021). The optimal C/N ratio for cultivation is approximately 10, but when the ratio goes above 30, plants perish due to N deficiency/mineralization, as N could be unavailable to plants owing to its preferential use by microbial organisms to break down DPSs (Hagemann et al. 2016). Vannucchi et al. (2021) described the sustainability of a substrate composed of DPS pellets and mature green waste compost for the growth of

trees in pots in nurseries and for fostering their capacity to counteract transplant stress. The photosynthetic performance of trees on this substrate was improved, although it was stated that N needs to be supplied regularly owing to an increased C/N ratio.

Table 2 presents the pseudototal concentrations of the elements that are considered vital for determining the nutritional status of the plants (P, K, Ca, Mg, Na, Fe, Mn, Cu, and Zn). The contents of Cu and Zn are subject to legal restrictions in waste, and these elements are considered contaminants when the concentrations are above these limits. The pseudototal concentrations of P, Ca, Fe, and Mn were significantly greater in the TUN-DPS than in the IT-DPS, while no significant differences were found for the other elements. The high Ca concentration in the DPS could be due to the presence of calcium compounds (carbonates or hydroxides) used in the paper finishing process and/or in coating-like materials such as paper brighteners/opacifiers and could already be present in wastepaper (Jele et al. 2022). Consequently, the higher Ca content in TUN-DPS could be due to both the addition of calcium salt in the chemical deinking process and to its greater presence in the wastepaper used in the Tunisian mill. Higher concentrations of Fe, Mn, and Zn were found in the TUN-DPS than in the IT-DPS, probably owing to the greater presence of inorganic material such as clay, which is likely the result of the different sources of paper to be recycled (Tunisia, Libya, Algeria, etc.), which is highly variable in the Tunisian mill. The presence of these elements could be linked to the inks or impurities from kaolin coating paper (Bajpai 2014b; Jele et al. 2022) or to other inorganic materials used, particularly during chemical processing, thus explaining their significantly greater content in TUN-DPS. Despite the differences in the contents of some of the elements in DPSs, they are nevertheless important. For example, Zn and Cu are microelements essential for plant growth and thus contribute to nutrient input in poor-quality soils (Gibbs et al. 2005). However, ink removal processes could also result in high and toxic levels of Cu, Zn, and other metals (e.g., Cr), thus subject to content thresholds for use in environmental purposes (EU 2014; DM 22/1998 (1998); Hébert 2015). Sodium values in both DPSs were similar and not a cause for concern. However, in the case of repeated applications in soils at high DPS doses, there may be a risk of salinity and sodicity, particularly in dry and hot climates such as the Mediterranean, thus potentially influencing plant development and increasing the risk of desertification (Turner et al. 2022). Consequently, under these conditions, the Na content of treated soils should be monitored.

Table 3 presents the contents of potential contaminants in both DPSs in terms of heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn), and organic contaminants such as polychloride biphenyls (PCBs), dioxins, furans, and pathogens. All

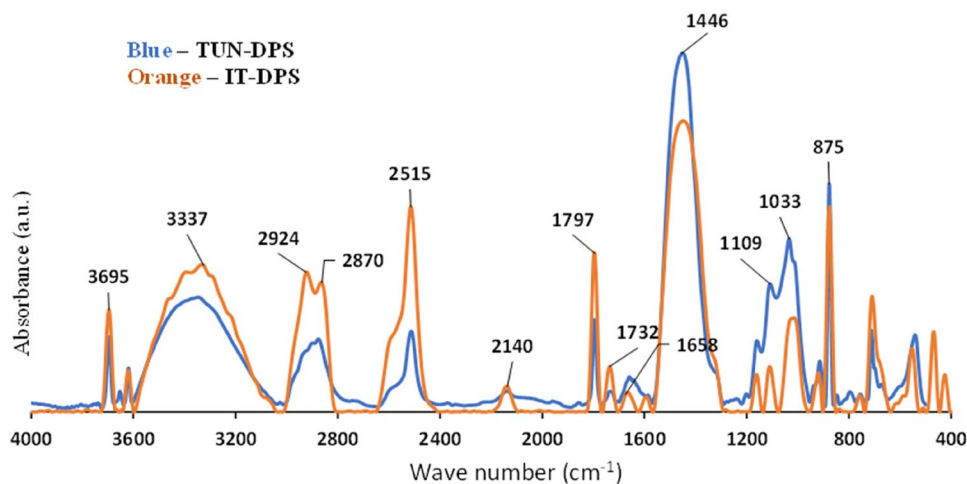
potential pollutants investigated were below the legal limit for their use in soils in accordance with the European and Canadian regulatory standards for RFM uses (EU 2014; DM 22/1998 (1998); Hébert 2015). Therefore, the evaluation of toxic elements, organic contaminants, and pathogens in both DPSs did not reveal the presence of contamination in these waste materials.

The FTIR/DRIFT spectra of DPS enabled us to characterize and compare the chemistry of their components. The results revealed the functional groups of the organic and inorganic molecules that make up the material (Fig. 1). In addition, other methods, such as thermogravimetric analysis and derivative curve analysis (TGA/DTA), help identify the temperature and degree of degradation of such components (Fig. 2), which is useful for characterizing and comparing their composition and behavior. Figure 1 shows the absorption spectra with the main characteristic DPS peaks. The main absorption peaks (Table 4) found in the IT-DPS at 3350, 2924 and 2870, 2140, 1797, 1732, 1650, 1446, 1109, and 1033  $\text{cm}^{-1}$  were attributed to specific functional groups

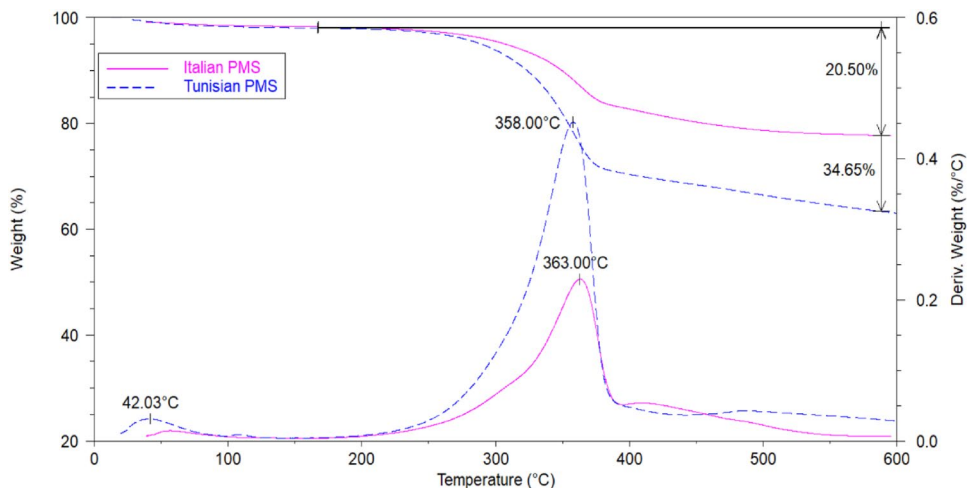
of organic molecules, indicating the presence of aromatic, phenolic, aliphatic, and polysaccharide structures (from highest to lowest wavelength), thus identifying the components of the OM in the DPS. These characteristic peaks corresponded to the FTIR spectra of TUN-DPS (3337, 2920 and 2850, 2140, 1797, 1732, 1658, 1446, 1058, and 1030  $\text{cm}^{-1}$ ), indicating the presence of similar functional groups and organic molecules. In addition, the presence of inorganic material in the spectra of IT-DPS and TUN-DPS were identified by the presence of peaks at 3695 and 3690  $\text{cm}^{-1}$ , respectively, which were attributed to OH groups in the kaolinite structures. On the other hand, the peaks at 2515 and 875  $\text{cm}^{-1}$  in IT-DPS and at 2520 and 875  $\text{cm}^{-1}$  in TUN-DPS were attributed to carbonates, while the peak at approximately 700  $\text{cm}^{-1}$  was attributed to quartz in both materials (Table 4).

Diffuse reflectance infrared Fourier transform (DRIFT) spectroscopy is particularly useful for analyzing solid samples, including soils and plant materials. For example, the effectiveness of soil amendments with DPS can be

**Fig. 1** Normalized Fourier-transform infrared spectra (DRIFT mode) of raw DPSs. The numbers on the spectra indicate the wavenumbers of the IR absorbance bands of the main functional groups



**Fig. 2** Thermogravimetric analysis (TGA) and derivative thermogravimetric (DTG) curves of the DPS in a nitrogen atmosphere. The temperature of the main degradation process and the percentage weight loss are indicated



**Table 4** Main Infrared absorbance bands in the spectra of IT-DPS and TUN-DPS

Wavenumber (cm <sup>-1</sup> ) for IT-DPS and TUN-DPS	Correspondence	Description
3695 and 3690	Stretching vibration of structural –OH groups of the kaolinite	Kaolinite structures
3350 and 3337	O–H stretching vibration (aliphatic and aromatic primary and secondary alcohols, carboxylic acids, absorbed water)	Lignin and carbohydrates
2924, 2870 and 2920	C–H stretching vibration of the aliphatic structure	Fatty acids, waxes, and various aliphatic components
2850	C–H symmetric stretch vibration in –CH <sub>2</sub>	Fatty acids and alkanes; fluorine, bromine, chlorine
2515 and 2520	Hydrochloride structure: hydrochloride amine C–NH <sub>3</sub> –Cl–; C=NH; C=N–C	Traces of hydrochloride amine C=NH; Carbonates
1797 and 1740	C=O stretching vibrations of acetyl groups in hemicelluloses and carbonyl aldehyde in lignin	Hemicelluloses, lignin
1658 and 1650	Aromatic skeletal (C=C)	Lignin
1446 and 1460	C–H deformation (asymmetric)	Carbohydrates and lignin
1158 1109 and 1158	C–O–C asymmetric stretch vibration in cellulose and hemicellulose	Cellulose and hemicellulose
1033 and 1030	C–O stretch in cellulose and hemicellulose	Cellulose and hemicellulose
875	C–H deformation in cellulose and =CH <sub>2</sub> in carbonate	Cellulose, carbonates
700	C–O–H out-of-plane bending in cellulose	Cellulose, quartz

monitored by investigating the composition, improvement, and persistence of organic matter through the formation of humic substances (Sonsri and Watanabe 2023; Zhu et al. 2023). Additionally, the possible liming effect and its persistence in acidic soils were monitored by this technique, given that calcium compounds (above all carbonates) are highly present in DPSs (Tinti et al. 2015). FTIR/DRIFT data provide valuable support for assessment-informed decision-making in agriculture, leading to improved soil management practices and enhanced crop productivity.

Figure 2 shows the weight loss curves of the samples (TGA) together with their derivative curves (DTG). The small weight loss before 200 °C in the TGA curve, highlighted by the small exothermic peak(s) in the derivative curve, was attributed to moisture vaporization for both DPSs. The DTG at these portions of the curves indicated a higher water content in the TUN-DPS than in the IT-DPS. During thermal degradation in the temperature range from 200 to 600 °C, two main thermal decomposition stages were identified in both DPSs, which could be correlated with cellulose/hemicellulose and lignin degradation. This behavior corresponds with previous findings on the characterization and pyrolysis of various paper mill waste materials (Jele et al. 2022; Lou et al. 2012; Mendez et al. 2009). In TUN-DPS and IT-DPS, the first and main weight losses of approximately 27% and 14%, respectively, were observed in the range of 200–390 °C, likely owing to the degradation of cellulose and hemicellulose (constituted by carbohydrates and aliphatic compounds). The greater weight loss in TUN-DPS suggested a greater cellulose content in this DPS, which was

consistent with sludge derived mostly from chemical deinking processes with respect to sludge coming mostly from mechanical separation process as is the case of IT-DPS.

During this thermal degradation process, TUN-DPS seems thermally less stable; i.e., the weight loss starts earlier, with a lower maximum decomposition rate temperature (358 °C) than that of IT-DPS (363 °C). The maximum degradation rate temperature for TUN-DPS corresponded with the study by Jele et al. (2022), which compared the thermal analysis of three different categories of DPS, including a deinking sludge. The second weight loss, occurring at a range of temperatures between 390 and 600 °C, is not well defined in the DTG curves. This value represents approximately 8% and 6% for TUN-DPS and IT-DPS, respectively, and could be due to the degradation of lignin and volatile inorganic compounds and further oxidation of organic components, as indicated by Assis and Chirwa (2021). However, in this second thermal process, IT-DPS degrades earlier than TUN-DPS, and the stability decreases during this second thermal process. The difference between the thermal stabilities of the two materials could be explained by several factors, including their chemical composition, e.g., the ink composition. Overall, the total weight loss for IT-DPS and TUN-DPS accounted for 20.5% and 34.7%, respectively, of the total weight loss. These findings indicate that the TUN-DPS had higher content of fibers (cellulose/hemicellulose and lignin) than the IT-DPS. Although in the TUN and IT samples the degradation processes have only been registered up to 600 °C, the TGA studies by Jele et al. (2022) also indicated a third significant degradation process at temperatures



higher than 700 °C, which was identified and attributed to the combustion of “residual carbon” and carbonate. The fact that most of the residual content at 600 °C was in TUN-DPS could indicate a greater content of inorganic C (Table 1), likely owing to the major presence of CaCO<sub>3</sub>, which is used as a filler or coating in papermaking. The higher content of Ca in the TUN-DPS (Table 2) could corroborate the hypothesis that CaCO<sub>3</sub> is the major component.

Thermogravimetric analysis (TGA) and derivative thermogravimetry (DTG) are useful techniques for obtaining information on the thermal decomposition of amendments of soil. The investigation of the effectiveness of soil amended with DPS provides insights into the quantity and thermal stability of soil organic matter (Jele et al. 2022; Méndez et al. 2010). These techniques can be used to determine the supply of organic matter in the soil, its transformation, the identification and quantification of mineral components from DPS, and the potential nutrient availability. The data collected contribute to improve soil management practices, nutrient cycling, and overall sustainability in agricultural systems. Together, the FTIR/DRIFT and TGA/DTG results helped identify the main components of DPS, confirming the presence of lignin, and cellulosic components of organic matter, as well as inorganic components to a lesser extent.

### DPS properties for application as an amendment in soil or nursery substrate

Significant differences in the basic characteristics of paper mill sludge have been reported in the literature and could depend on the kind of feedstock, the type of deinking process, and the phase paper is produced and the waste collected. Despite these differences, amending agricultural soils with DPS appears to have a positive impact (Gibbs et al. 2005; Turner et al. 2022) and is considered a sustainable option. The high content of organic matter and available nutrients for plant uptake, together with the ability of DPS to increase water retention and pH (limiting effect) and the formation of macroaggregates in poor-quality soils, improve both soil quality and plant health. However, monitoring is necessary when persistent land application occurs, particularly regarding N mineralization, salinity in terms of Na, and the accumulation or leaching of heavy metals in relation to soil properties, especially pH and texture.

In the DPS under study, a high C/N ratio (> 30), i.e., a high value of organic carbon (due to the high cellulosic content) compared with the low content of total nitrogen, suggested the need to supply N by adding a mineral fertilizer or mixing DPS with compost or manure (Baziramakenga et al. 2001; Vannucchi et al. 2021). Incorporating sludge into the soil can decrease the amount of inorganic N, particularly the N form most commonly used by plants (N-nitrate), through immobilization, which is a well-known

phenomenon attributed to N assimilation by microorganisms decomposing N-poor OM (Chen et al. 2014). DPS composting and vermicomposting have been carried out to reduce the high C/N ratio, with the side effects of concentrating the elements and reducing the waste mass (Turner et al. 2022). Thus, when DPS is applied to restore soil productivity, a complete characterization of the amendment as well as the receiving pedological environment and crop needs is necessary to highlight possible environmental concerns.

Soil parameters such as pH, EC, and macro- and micro-nutrients are also important. The DPSs from Italy and Tunisia investigated in our study had slightly alkaline pH values (approximately 8). It has been reported that the pH was increased from 0.1 to 0.7 by applying 100 t/ha DPS (Gibbs et al. 2005). Our approach could thus be a good option for neutral and acidic soils, although overliming should be monitored. In fact, soil reactions between 6.7 and 7.8 provide optimal conditions for nutrient uptake, while a pH higher than 7.8 reduces the mobility of important microelements such as Fe and Mg (Samson et al. 2017). Marouani et al. (2020) reported the efficient mitigation of cumulative CO<sub>2</sub> and a liming effect when an acidic clayey soil was amended with DPS from deinking paper sludge composted with manure and mixed with wood ash. In the same study, the effect of DPS on soil physical properties was also observed, particularly for clay soils in which the soil structure was changed, the bulk density was reduced, and the porosity and moisture retention were increased, thus facilitating root zone development. The low density of DPS allows a decrease in soil BD, which is desirable in heavy clayey soils or in urban green infrastructure where the acceptable weight of the substrate is limited (Vannucchi et al. 2018). Thanks to the fibers of lignin and cellulose, DPS as a soil amendment improves the physical properties of sandy soils, thereby increasing aggregate stability and, in terms of hydraulic capacity, improving infiltration and water storage (Turner et al. 2022). The values of EC, which are directly correlated with the presence of available ions, are important for determining the potential fertility of an amendment; therefore, DPS could increase soil nutrient levels. In fact, Mediterranean soils subject to drought are at risk of salinity and sodicity, clayey soils can accumulate toxic elements, and sandy soils are at risk of leaching to the water table. An accurate evaluation of the application rate and monitoring are thus necessary. In addition, some authors have reported limited effects on the communities of soil microorganisms (Gallardo et al. 2012), and an increase in microbial activity following the coapplication of biochar and paper mill biosolids has been found (Manirakiza et al. 2019). However, the effects on the soil biota and possible interactions are still under study.

The impact of DPS valorization and management pathways on mitigating GHG emissions compared with conventional disposal methods, mainly landfilling or incineration,

is also relevant to investigate. When DPS is landfilled, toxic chlorophenols can eventually reach the water table, while incineration can generate atmospheric pollution owing to the combustion of halogenated products, which produces dioxins and furans (SYLVIS 2009). Sludge incineration also emits GHG, particularly nitrous oxide (N<sub>2</sub>O) (Faubert et al. 2016). Evaluating the environmental impact of the different valorization pathways through life cycle analysis and measurement of GHG emissions during processing and in service will contribute to determining whether such practices could help achieve government targets for reducing GHG emissions and climate change attenuation. Consequently, exploiting paper sludge in agricultural soils could represent an effective solution for DPS recycling as a soil amendment or a component of the growth substrate, provided that monitoring and managing changes in soil properties and biological activities are carried out. The properties of the sludge generated during the deinking process play a crucial role in determining the potential uses of the sludge in different agricultural sectors. The environmental impact of DPS from mechanical processes (IT-DPS), in terms of toxicity and improvement in growth media, may be more effective than that of sludge from chemical deinking processes. This is because, although the C/N ratio in IT-DPS is high, it is approximately half of that in TUN-DPS: less intervention is thus needed for N fertilization. In addition, the slightly but significantly lower BD in the IT-DPS than in the other system means that DPS can be exploited for ameliorating structural and/or soil compactness. However, in addition to the previous speculation on the fertilization potential of DPSs, the specific properties of the matrix in which they will be incorporated, and the plant species involved should also be considered.

## Conclusions

The Mediterranean is a hotspot of biodiversity due to the complexity of its ecosystem, and it is greatly affected by climate change. The management of waste, especially incineration, increases the presence of climate-altering gases in the atmosphere, highlighting the need for waste recycling and reuse. On the other hand, desertification is a worrying emergency in Mediterranean soils owing to the loss of organic matter, which can be combated with organic carbon-rich waste. DPS can be used as a soil amendment and source of OC in combination with compost or manure to integrate N. The novelty of the present study is to combine these two important environmental aspects and contextualize the study in the Mediterranean, where sludge is produced and where there are significant environmental challenges.

The data on the properties of DPS materials from Italy and Tunisia corroborate the hypothesis that these materials

are suitable amendments for soil and plant-growth substrates. The DPS obtained with different processes of wastepaper recycling showed similar properties to those that could be exploited as soil amendments. The high content of carbon and nutrients in the investigated DPS confirms the necessity of recycling the material as an amendment in plant cultivation, providing organic fertilization. Mulching or incorporation can be used based on the land's requirements for soil enhancement, weed suppression, full-scale land remediation, and potential monitoring of soil and water contamination hazards. Material recycling policies should also address the quality improvement of secondary products by combining, for example, recycling initiatives with technological development and strategies for contaminant removal from these materials, thus supporting the maximum economic and environmental value recovery from DPSs. Given the importance and the large amounts of DPS produced, this study reinforces the need to develop international standards for DPS management independently of its origin/process. Countries with no existing management standards for DPS could thus be encouraged to adopt such international standards, considering site-specific conditions, soil characteristics, and on-site crop requirements. Future studies could focus on the effects of sludge application in agricultural and urban soils considering different soil characteristics, plants cultivated, and microbiological aspects. For agricultural soils, sludge should improve the crop production. In the case of urban infrastructure, the aim should be to improve the ecosystem services provided by urban areas. In general, studies on DPSs should demonstrate their possible role in adapting to climate change in the Mediterranean area. In addition, DPS could be tested as a component of a biocomposite with biodegradable polymers for producing a biofilm for mulching or nursery pots to be buried and degraded in the soil, providing OM and mineral nutrients.

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**Data availability** Not applicable.

## Declarations

**Conflict of interest** The authors have no competing interests to declare that are relevant to the content of this article.

**Ethical approval** The manuscript has only been submitted to this journal.

**Consent to participate** Not applicable.

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