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ORIGINAL PAPER

Evaluation of selective clay minerals and biochar as materials for sewage sludge stabilization

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Abstract
The objectives of this study were to evaluate sewage sludge's stabilization with untested until now materials, such as selective clay minerals or biochar in comparison with liming, for enhancing sludge's fertilization capacity. Dewatered sewage sludge was mixed with bentonite, vermiculite, zeolite, biochar or lime at rates of 0, 15 and 30%, air-dried and analyzed for pathogens and chemical properties. Almost all fecal indicators of treated sludge with 15% bentonite, vermiculite or biochar were reduced by at least one-logarithmic unit ($\log_{10}$) (indicative value of sludge's stabilization), whereas those of limed sludge were undetectable. Electrical conductivity of all treatments significantly increased, and the highest values were obtained for untreated (6.1 dS m^{-1}) and limed sludge (above 7.0 dS m^{-1} for both addition rates). The untreated sludge had the significantly highest water-soluble ammonium-nitrogen (2817 mg kg^{-1}) and phosphorus (263 mg kg^{-1}) concentrations followed by sludge treated with bentonite, vermiculite or biochar, whereas limed sludge had the lowest content. Boron concentration of the untreated sludge was similar to the treated sludge. Total concentrations of heavy metals were far below the legislative permissible levels for sludge's agronomic use. Nutrients' total content of treated sludge ranged at levels of similar magnitude to the untreated sludge, except for certain cases where they were increased because of the materials' composition. Consequently, sewage sludge treated with 15% bentonite, vermiculite or biochar seems to be stabilized, retain bioavailable nitrogen and serve as a fertilizer of macro- and micronutrients. However, potential risks of agronomic use, i.e., soil salinization and boron phytotoxicity, should be considered.

Keywords Bentonite · Biochar · Pathogens · Plant nutrients · Sewage sludge · Vermiculite

Introduction

Sewage sludge is the organic by-product of domestic wastewater treatment, which contains pathogens, pollutants, heavy metals and other constituents (Wu et al. 2020). Agronomic application of sewage sludge is an emerging valorization strategy compared to land filling and incineration. Sewage sludge's organic matter can improve physicochemical properties of soils and supply them with macro- and micronutrients for plants, through mineralization process (Singh and Agrawal 2008). Due to its binary nature, sewage sludge is considered not only a waste that poses public health and environmental safety risks (European Commission 2015), but also a valuable resource (Christodoulou and Stamatelatou 2014). From a circular economy perspective, agronomic use of sewage sludge can recover organics and nutrients, reduce greenhouse gas emissions and mitigate environmental loading characterized by wasteful landfills and incinerations practices (Ye et al. 2022). It is therefore of outmost importance to find novel treatment methods to improve sewage sludge's fertilizing capacity, especially by preserving its nitrogen (N) content, and control undesirable parameters such as pathogens and heavy metals.

The European Council Directive 86/278/EEC (1986) defines treated sewage sludge as having undergone "biological, chemical or heat treatment, long-term storage or any other appropriate process so as significantly to reduce its fermentability and the health hazards resulting from its use."

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The same directive established upper permissible limits for heavy metal total concentrations in sewage sludge and soils (Table 1) and correlated sludge's addition rate with the needs of plants for nutrients. In addition to the Directive 86/278/EEC (1986), the regulation of the US Environmental Protection Agency (USEPA 1995) emphasizes, to some extent, on requirements for reduction in pathogens by classifying treated sludge into two categories, namely Class A and Class B. Class A sludge can be applied on land with no restrictions because pathogens have been reduced below detectable levels, whereas Class B sludge subjects to less stringent stabilization requirements, but also can be applied on land under specific conditions. Nowadays, agronomic use of sewage sludge subjects to different requirements in every country, as far as heavy metals, pathogens and organic contaminants is concerned (Collivignarelli et al. 2019).

Methods of sewage sludge's treatment include biological (e.g., composting), non-biological processes (e.g., liming) or a combination of them (Li et al. 2022). Composting involves mixing various bulking agents like clay minerals (Awasthi et al. 2018), biochar (Liu et al. 2017; Guo et al. 2020), sawdust, wood chips and agricultural wastes (e.g., wheat straw, plant leaves, sunflower stalk) (Nafez et al. 2015) for the stabilization, volume reduction and increase in fertilization capacity of the treated organic waste. Bulking agents affect composting parameters like temperature, pH and moisture content, all of which regulate microbial activity, elemental speciation and greenhouse gasses emissions (Barthod et al. 2018). Liming materials also affect the aforementioned parameters. Their strongly alkaline reaction results in high pH, temperature increase (because of lime's exothermic reaction with water) and ammonia (NH_3) emission, all of which are lethal for pathogens (Arthurson 2008). In addition, high pH and content of calcium (Ca) compounds lead also to immobilization of heavy metals. Several researchers investigated stabilization of sewage sludge employing alkaline industrial by-products, such as steelmaking slag (Papastergiadis et al. 2015; Samara et al. 2017), sugar beet factory lime (Shaheen et al. 2014) and coal fly ash (Su and Wong 2002). Co-composting sewage sludge with alkaline materials has also been attempted (Wong and Fang 2000).

Table 1 Maximum permissible concentrations for heavy metals in sewage sludge and soils, according to the European Council Directive 86/278/EEC (1986), which was adopted by the Greek law

Heavy metal	Sewage sludge (mg kg^{-1})	Soil
Cd	20–40	1–3
Cu	1000–1750	50–140
Ni	300–400	30–75
Pb	750–1200	50–300
Zn	2500–4000	150–300
Hg	16–25	1–1.5
Cr[#]	1000	60

Proposed Cr concentrations according to the 3rd Draft of the "Working Document on Sludge" are also included[#] (European Commission 2000)

During stabilization of sewage sludge employing liming or composting, N loss, mainly through NH_3 emission, considerably reduces the fertilizing value of the final product (Arthurson 2008). Based on this, Samara et al. (2019) investigated nine clay minerals, in respect to sewage sludge's stabilization and N retention, in comparison with liming. Their work outlined a novel, promising low-cost stabilization method of sludge, which could increase the fertilization value of the treated sludge. As a continuation of this work, in the current study, new materials were tested (i.e., vermiculite and biochar) and lower addition rates of already tested materials (i.e., bentonite and zeolite) for the stabilization of sewage sludge, in the perspective of using the treated sludge in agriculture. Sewage sludge's treatment with clay minerals seems promising because clay minerals exhibit reactive surfaces that interact with moisture, nutrients, heavy metals and pathogens. The same is evidenced for biochar, a significant factor in climate change mitigation through carbon (C) sequestration.

Bentonite, vermiculite and zeolite are all clay minerals of different structure, the two former are 2:1 phyllosilicates, whereas the latter is a tectosilicate mineral, with high permanent negative charge, due to extensive isomorphous substitution (in tetrahedral and octahedral structural sheets of bentonite and vermiculite and tetrahedral structural framework of zeolite) (Bish 2013). Biochar is the product of thermal degradation of organic materials in the absence of air (pyrolysis) (Lehman et al. 2011), which contains surface functional groups of variable pH-dependent charge (Tan et al. 2020). In general, total composition of biochar varies substantially mainly because of the total elemental composition of the feedstock and to a much lesser extent due to the pyrolysis conditions (Ippolito et al. 2015). Biochar is produced through various thermochemical conversion technologies such as hydrothermal carbonization, pyrolysis, gasification and torrefaction (Uday et al. 2022). Biochar can also be engineered by using pyrolysis with chemical, physical or biological modification techniques to improve its adsorption capacity and overall properties (Mahari et al. 2022). In the last few decades, biochar has been used in wastewater treatment, water contaminant removal, C capture, power generation, storage of energy, soil fertility enhancement and carbon sequestration as well as in the construction industry (Uday et al. 2022).

All four materials have been widely investigated as sorbents for NH_3, ammonium (NH_4^+) and nitrate (NO_3^-) ions and heavy metals. Bentonite reduced NH_3 emissions from organic wastes (Redding 2013) and bentonite, vermiculite and zeolite were used to sorb lead (Pb), cadmium (Cd), copper (Cu), nickel (Ni), and chromium (Cr) from wastewater and sewage sludge (Kosobucki et al. 2008; Gu et al. 2019). Moreover, biochar controlled greenhouses gasses and NH_3 emissions from composts and reduced bioavailability of Cu, zinc (Zn), Pb, Cd in organic wastes (Guo et al. 2020). Biochars have been used as low-cost and efficient sorbents to remove N, P, heavy metals and organic contaminants from wastewater, while their production and modification-treatment technologies continuously evolve to increase their sorption and remediation capacity (Xiang et al. 2020).

Sludge treatment with clay minerals or biochar could be a low-cost, efficient treatment method for areas with available minerals and biochar resources. Other low-cost treatment methods such as composting and vermicomposting can increase organic matter decomposition and pathogen load reduction during the sludge treatment process; however, they can be insufficient in controlling nutrient loss, organic contaminants, heavy metals and the emission of greenhouse gasses (Liew et al. 2022). Bentonite, vermiculite and biochars have already been used as additives in composting and vermicomposting to control various parameters, improve the efficiency of the process and enhance the soil fertilizing and ameliorating capacity of the final product (Zhou et al. 2022). In addition, clay minerals have bactericidal properties, through aluminum (Al) binding to phospholipids which can lead to osmotic imbalance and cell-lysis or through sorption of nutrients on clay and thus their removal away from bacteria (Williams 2019). Biochar's bactericidal capacity against wastewater and soil borne pathogens has also been documented (Muoghalu et al. 2023). Furthermore, clay minerals and biochar were proven effective sorbents of organic compounds (Keil and Mayer 2014), which means that sewage sludge's organic matter can be protected from degradation and organic pollutants can be sorbed (Accardi-Dey and Gschwend 2003).

Overall, treating sewage sludge with clay minerals and biochar could result in organo-mineral fertilizers and soil amendments, which could provide slow release of nutrients to soils and comparable crops yield to conventional fertilizers, minimizing at the same time the load of pathogens and heavy metals (Husek et al. 2022). Balidakis et al. (2022, 2023) studied the effect of soil application of treated sludge with bentonite, vermiculite or biochar, which was produced based on the results of the current study. The researchers reported that application of 2% treated sludge to an acid and an alkaline soil enhanced the pH of acid soil, improved fertility of both soils, reduced microbial stress caused by acidity and increased biomass yield and nutrient uptake by plants of perennial ryegrass (*Lolium perenne* L.) and white clover (*Trifolium repens* L.) grown in the treated soils.

In the perspective of finding new methods for treatment of sewage sludge, for its safe use as an amendment for arable soils, the objectives of this study were to evaluate sludge's stabilization, in respect to both microbial and heavy metals load, by mixing sludge with bentonite, vermiculite, zeolite or biochar in comparison with liming. Moreover, certain chemical properties of the treated sludge, which could affect in a beneficial or harmful manner the soil chemical properties and the growth of crops, were studied. The whole research work is presented briefly in Fig. 1; the experimental work started in the beginning of May 2020 and was conducted at the facilities of the Faculty of Agriculture of Aristotle University of Thessaloniki, Thessaloniki, Greece.

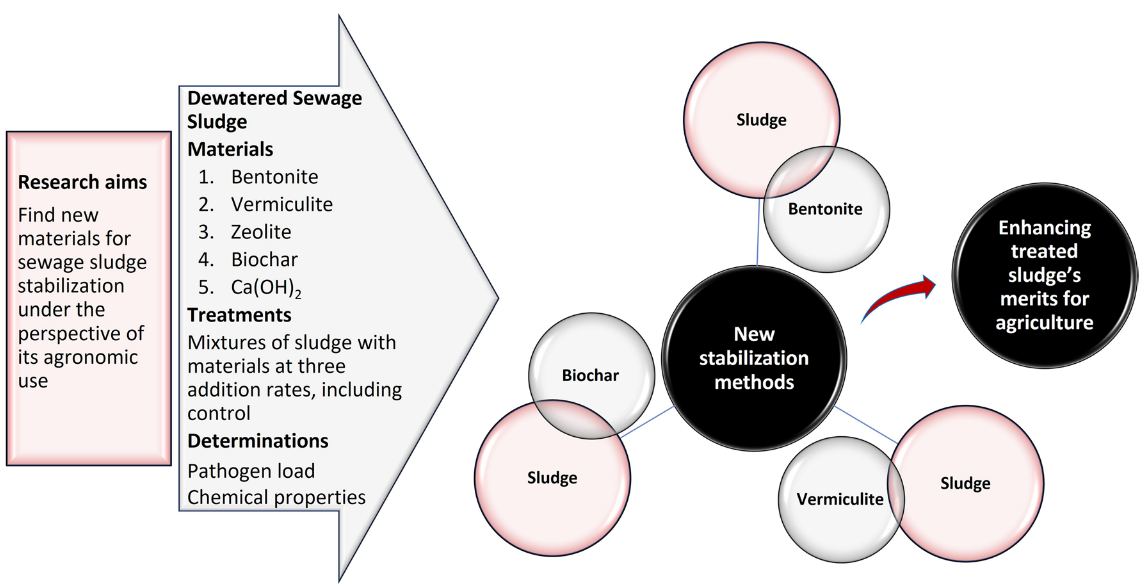

Fig. 1 Schematic representation of the whole research work

Materials and methods

Sewage sludge, minerals and biochar—preparation and laboratory analyses

Dewatered sewage sludge was collected from the wastewater treatment plant of Thessaloniki, Greece, analyzed for total *coliforms*, *Escherichia coli* and total *enterococci*, using the most probable number (MPN) methods (ISO 7251 2005; ISO 4831 2006), and tested for *Salmonella spp.* (ISO 6579 2002).

Moreover, pH was measured at a 1:5 (w/v) suspension with water (H_2O), dry matter was determined after drying at 105 °C until constant weight and organic matter was estimated by loss on ignition (LOI) at 500 °C for at least four hours. Water-soluble nitrate-nitrogen (NO_3-N), ammonium-nitrogen (NH_4-N), phosphorus (P), potassium (K), sodium (Na), Ca, magnesium (Mg), boron (B), Cu, Zn, iron (Fe), manganese (Mn), Ni, Cr, Cd, Pd and mercury (Hg) were extracted with water (1:10 w/v ratio) after equilibration for one hour and electrical conductivity (EC) was measured in the filtrate. Kjeldahl-N was determined (Bremner 1996) and the total concentrations of P, K, Na, Ca, Mg, Cu, Zn, Fe, Mn, Ni, Cr, Cd, Pd and Hg were measured after digestion with aqua regia (ISO 11466 1995) and that of B after dry ashing at 500 °C for at least four hours.

As far as the methods used for the analytical determinations of the chemical species are concerned, NO_3-N and NH_4-N were determined by ultraviolet spectrometry and the sodium salicylate-sodium nitroprusside method (Mulvaney 1996), respectively, P by the molybdenum blue-ascorbic acid (Kuo 1996), B by the azomethine-H method (Keren 1996), K and Na by flame photometry and the rest of the metals were determined by atomic absorption spectrometry. In addition, the sodium adsorption ratio (SAR) was calculated from the water-soluble concentrations of Na, Ca and Mg.

Regarding clay minerals, bentonite was provided from Milos Island and vermiculite along with zeolite were provided from northern Greece, while biochar was produced from pine tree residues through flame curtain pyrolysis at 600 °C for one hour (Kalderis et al. 2020). All materials passed through a 2 mm sieve and were analyzed for pH at a 1:10 (w/v) suspension with H_2O and cation exchange capacity (CEC) (Ming and Dixon 1987; ISO 23470 2007). In addition, EC, water-soluble Na, Ca, and Mg were determined, and SAR was calculated, as well as the total concentrations of macro- and microelements mentioned above were determined, employing the same methods.

Stabilization experiment—details and laboratory analyses

In the beginning of May 2020, bentonite (B), vermiculite (V), zeolite (Z), biochar (BC) and reagent grade calcium hydroxide [$Ca(OH)_2$] (RG), were mixed with dewatered sewage sludge at rates equal to 0% (control, i.e., untreated sewage sludge), 15% (B15, V15, Z15, BC15 and RG15) and 30% (B30, V30, BC30 and RG30) (wet weight basis) (treatments), in three replications. The treatments were placed in plastic pots and left for air-drying in an unheated greenhouse of the Farm of Aristotle University of Thessaloniki, with periodic mixing for 60 days. The experimental design was the completely randomized (CRD) and randomization was repeated every 15 days. In the middle of the air-drying period, sub-samples of the treatments were analyzed for pH, dry matter content and total C and N, by an elementary analyzer, whereas at the end of the air-drying period, the treatments passed through a 4-mm sieve and were analyzed for the aforementioned four properties, as well as for the properties reported for the dewatered sewage sludge, employing the same methods.

All analyses, reported in the current and the previous subsection, were conducted in triplicate at the Soil Science Laboratory of the Faculty of Agriculture of Aristotle University of Thessaloniki.

Table 2 Fecal indicators and *Salmonella spp*. of the dewatered sewage sludge

Total *coliforms*	*E. coli*	Total *Enterococci*	*Salmonella spp.*
(MPN g^{-1} of wet weight)			(25^{-1} g^{-1} of wet weight)
5.7 $10^4 \pm 5.1$ 10^4	2.7 $10^4 \pm 2.3$ 10^4	4.5 $10^4 \pm 4.2$ 10^4	Detectable

MPN most probable number

Values are presented as means ± standard error

Statistical analysis

For each property of all treatments, one-way ANOVA was conducted, using the statistical package SPSS, version 26, and the protected LSD test at $p = 0.05$ was used for mean comparisons.

Results and discussion

Microbial and chemical properties of the dewatered sewage sludge

The dewatered sewage sludge had low microbial load (Table 2) compared to values (of 10^6 magnitude) reported in the literature (Carrington 2001; Samara et al. 2019). This could be attributed to the good hygienic state of public health and hospitals, tanneries and meat-processing factories operating in the area the wastewater treatment plant serves (Dumontet et al. 1999). The pH of dewatered sewage sludge was almost neutral and both EC and SAR were low, compared to values reported in literature, especially for EC (1.0–31.0 dS m^{-1}) (Samara et al. 2019) (Table 3). However, the pH of sludge can decrease upon decomposition of its organic matter and EC can increase depending on the sludge's degree of dewatering-condensation (Merrington et al. 2003).

The water-soluble (readily available) concentrations of the three main essential macronutrients for plant growth (i.e., NO_3-N, NH_4-N, P and K) were considerable (Table 3), whereas the water-soluble micronutrients (Cu, Zn, Fe and Mn) were not detectable, except for B. In fact, total B of sludge was in water-soluble form (Table 3) and this is somewhat concerning, because B has a unique characteristic; its deficiency-sufficiency-toxicity ranges for crops are very close to each other (< 0.5, 0.5–2, > 5 mg kg^{-1}, respectively) (Johnson and Fixen 1990). Total concentrations of macro- and microelements were also considerable; it is worth noting that the water-soluble concentrations of N (both NO_3-N and NH_4-N), P and K were 10%, 7% and 65% of their total concentrations, respectively. This was attributed to the different form and chemical behavior of the elements, i.e., N is mainly in organic form in sewage sludge and P and K tend to form insoluble and soluble substances, respectively. The total concentrations of heavy metals, which regulate the agronomic use of sewage sludge according to legislation were either far below the permissible levels (Table 3 and compare with Table 1) or not detectable in the case of Cd and Hg. This is probably because Thessaloniki's wastewater treatment plant does not receive industrial waste effluents and heavy metal load builds up only from runoff and domestic wastewaters (European Commission 2001).

Chemical properties of the clay minerals and biochar

All four materials had alkaline to strongly alkaline pH values and low EC (Table 4). The three clay minerals had high CEC in comparison with biochar. This was attributed to the high permanent negative charge of the minerals due to extensive isomorphous substitution (Bish 2013). On the other hand, the low CEC of biochar was attributed to the relatively small amount of surface functional groups (Kalderis et al. 2020). Although the high CEC values of the particular clay minerals indicate high exchange capacity and retention in respect to cations, these are not necessarily related with their full capacity to sorb heavy metals, as far as alleviating heavy metal load of sewage sludge is concerned. This is because retention of heavy metals by clays is not only a function of CEC, but also a function of surface complexation and precipitation reactions (Churchman et al. 2006).

As far as the total macro- and microelements are concerned, concentrations of the dominant structural elements of the materials were high, i.e., K in zeolite due to K of its structural channels (Koyama and Takeuchi 1977), Mg in vermiculite due to its interlayer Mg (Schultze 2005) and Fe in bentonite and vermiculite due to isomorphous substitution of Al by Fe in their octahedral structural sheets (Bish 2013) (Table 4). Biochar had considerable total concentration of B, probably due to its origin (Table 4), which is somewhat concerning, for the same aforementioned reasons reported for dewatered sewage sludge. Under the perspective

Table 3 Certain properties and water-soluble and total concentrations of nutrients and heavy metals of the dewatered sewage sludge, expressed on dry weight basis (except for dry matter)

Certain properties			Total concentrations		
Dry matter	(%)	13.8 ± 0.5	N	(%)	4.96 ± 1.18
LOI		67.9 ± 1.4	P	(mg kg^{-1})	14,858 ± 1896
pH	(1:10 H_2O)	7.1 ± 0.1	K		1117 ± 73
EC	dS m^{-1}	0.82 ± 0.05	Na		3467 ± 621
SAR		3.7 ± 1.1	Ca		8864 ± 1713
Water-soluble concentrations			Mg		4462 ± 1619
NO_3-N	(mg kg^{-1})	2265 ± 1100	B		23 ± 5
NH_4-N		2927 ± 432	Cu		151 ± 8
P		1002 ± 104	Zn		606 ± 38
K		725 ± 5	Fe		9201 ± 550
Na		5553 ± 144	Mn		152 ± 10
Ca		1280 ± 808	Ni		39 ± 2
Mg		665 ± 105	Cr		44 ± 0
B		26 ± 4	Pb		71 ± 15

LOI loss on ignition, *EC* electrical conductivity, *SAR* sodium adsorption ratio

Water-soluble Cu, Zn, Fe, Mn, Ni, Cr, Cd, Pb and Hg, and total Cd and Hg were not detectable

Values are presented as means ± standard error

Table 4 Certain chemical properties and total composition of the materials

Chemical properties		Bentonite (B)	Vermiculite (V)	Zeolite (Z)	Biochar (BC)
pH	(1:10 H_2O)	7.7 ± 0.0	9.0 ± 0.2	8.6 ± 0.0	9.2 ± 0.1
EC	(dS m^{-1})	1.48 ± 0.07	0.10 ± 0.00	0.06 ± 0.01	0.82 ± 0.04
SAR		8.2 ± 0.2	0.3 ± 0.0	ND	6.2 ± 0.2
CEC	($cmol_c$ kg^{-1})	71.2 ± 4.1	90.2 ± 0.4	129.8 ± 3.9	9.3 ± 0.4
Total concentrations		Bentonite (B)	Vermiculite (V)	Zeolite (Z)	Biochar (BC)
N	(%)	ND	ND	ND	0.81 ± 0.09
P	(mg kg^{-1})	ND	ND	ND	1073 ± 101
K		1122 ± 1	456 ± 41	20,939 ± 366	844 ± 51
Na		4237 ± 103	1352 ± 793	5823 ± 3	1832 ± 13
Ca		8895 ± 1522	945 ± 413	5550 ± 427	8714 ± 1187
Mg		4454 ± 327	17,119 ± 400	2642 ± 1332	902 ± 52
B		ND	ND	ND	24.1 ± 0.9
Cu		20.6 ± 0.3	42.5 ± 6.0	14.3 ± 8.1	6.0 ± 0.7
Zn		92.5 ± 5.0	32.3 ± 1.7	36.7 ± 0.9	24.8 ± 3.9
Fe		22,519 ± 2201	25,918 ± 3724	7226 ± 213	229 ± 59
Mn		386 ± 23	363 ± 47	487 ± 40	7.9 ± 0.9
Ni		11.2 ± 1.6	9.7 ± 1.0	ND	ND
Cr		6.1 ± 0.6	ND	ND	ND
Pb		14.2 ± 1.9	ND	42.8 ± 5.1	10.6 ± 0.2

EC electrical conductivity, *SAR* sodium adsorption ratio, *CEC* cation exchange capacity

ND: not detectable; the same stands for total Cd and Hg

Values are presented as means ± standard error

of applying the produced treated sludge to arable soils, the total concentrations of all heavy metals which should be considered according to legislation, were either far below the permissible levels or undetectable (Table 4 and compare with Table 1).

Effect of the clay minerals and biochar on the microbial load of sewage sludge

After the air-drying period, although all fecal indicators reduced and *Salmonella spp.* was not detectable at all treatments, a reduction in above one-logarithmic unit (log_{10}) was observed only at the B15, V15, BC15 and B30 treatments, with the BC15 treatment showing the highest values (Table 5). In other words, microbial load was reduced by 70–98% in these cases and thus 15% addition of bentonite, vermiculite or biochar to dewatered sewage sludge can stabilize sewage sludge. Higher reduction in fecal indicators (one-two logarithmic units) and absence of *Salmonella spp.* after addition of clay minerals (i.e., bentonite, attapulgite, mixed clay of attapulgite-saponite and zeolite) to sludge is reported by Samara et al. (2019), probably due to differences in minerals and treatment process. Due to pH increase in the RG15 and RG30 treatments at the strongly alkaline range (i.e., 12.3 and 12.4 respectively, Fig. 2), but also because of the temperature increase and NH_3 release which are lethal for pathogens (Arthurson 2008), the microbial load of both limed sludge treatments was not detectable. A similar microbial load reduction was observed by Papastergiadis et al. (2015) and Samara et al. (2017) after adding strongly alkaline steelmaking slag to sludge at rates higher than 10%.

The survival of pathogens during sewage sludge's stabilization depends on several factors, such as moisture and nutrient content, decomposition of organic matter, microbial competition, pH, temperature, aeration and time (Li et al. 2022). In the current study, at the end of air-drying period the initial high moisture content of 86% of dewatered sewage sludge dropped below 10% at all treatments including control, whereas in the middle of that period, similar results were obtained for all treatments apart from control, which had 76% moisture content. During stabilization period, sludge treatments were thoroughly mixed to provide aeration and ensure homogenization. Aeration supplies microorganisms with oxygen for organic matter decomposition and removes moisture, which plays a significant role in reducing sludge's pathogen load, volume and leachate production upon its land application (Wu et al. 2020). Sludge's organic fraction is mainly composed of negatively charged polymers which form a complex structure with the biological cells of pathogens and other microorganisms, interlocking high

Table 5 Reduction in the fecal indicators [logarithmic scale ($\log_{10}$)] of the treatments with respect to dewatered sewage sludge, at the end of the air-drying period

Fecal indicator	Treatment				
	B15	V15	Z15	BC15	Control
Total *coliforms*	0.6	0.6	0.5	1.1	0.4
E. coli	1.0	1.3	0.7	1.5	0.7
Total *Enterococci*	1.1	1.1	0.9	1.6	1.0
Fecal indicator	Treatment				
	B30	V30	Z30	BC30	Control
Total *coliforms*	0.7	0.6	0.2	0.8	0.4
E. coli	1.3	0.9	0.7	1.0	0.7
Total *Enterococci*	1.4	0.9	0.5	0.5	1.0

B15, V15, Z15, BC15 and RG15: dewatered sewage sludge treated with 15% addition of bentonite, vermiculite, zeolite, biochar and $Ca(OH)_2$, respectively; B30, V30, Z30, BC30 and RG30: dewatered sewage sludge treated with 30% addition of bentonite, vermiculite, zeolite, biochar and $Ca(OH)_2$, respectively; Control: untreated, just air-dried dewatered sewage sludge

Salmonella spp. was not detectable at all treatments, including the $Ca(OH)_2$ treatments (RG15 and RG30) and control; in addition, all indicators were not detectable at both $Ca(OH)_2$ treatments

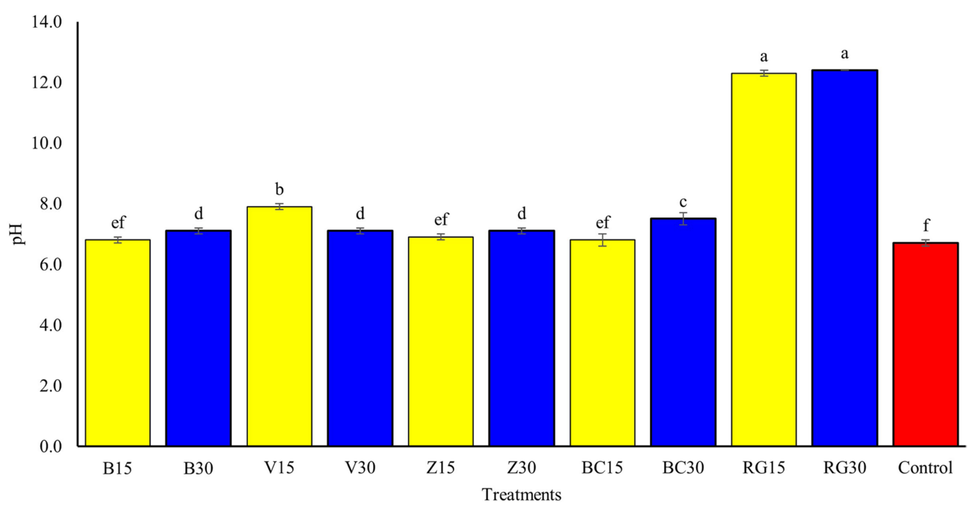

Fig. 2 pH of the treatments. B15, V15, Z15, BC15 and RG15: dewatered sewage sludge treated with 15% addition of bentonite, vermiculite, zeolite, biochar and $Ca(OH)_2$, respectively; B30, V30, Z30, BC30 and RG30: dewatered sewage sludge treated with 30% addition of bentonite, vermiculite, zeolite, biochar and $Ca(OH)_2$, respectively; Control: untreated, just air-dried dewatered sewage sludge. Values are presented as means ± standard error. Different letters indicate significant differences among means, using the protected LSD test, at $p \leq 0.05$. As far as ANOVA it concerns, p F test < 0.001

amounts of humidity in flocks and protecting them from drying (Capodaglio 2023). As it is obvious from the above-mentioned results, clay minerals and biochar contributed to accelerating sludge's drying since these materials sorb water. However, this effect on the reduction in microbial load was not the only one in the case of bentonite, vermiculite and biochar addition to sludge. It seems that also other mechanisms contributed to the observed reduction in sludge's pathogens.

Except for the limed sludge treatments, the rest of the treatments had nearly neutral to slightly alkaline pH (lower than 8.0) (Fig. 2) at both periods. As far as the C/N ratios of the sludge treated with bentonite, vermiculite or zeolite are concerned, it seems that they were not high enough to promote organic matter's mineralization because of the materials' inorganic nature and the initial low C/N ratio of the dewatered sludge (Barthod et al. 2018). The C/N ratios ranged from 6.4 to 7.9 at all treatments (including control), except for the treatments with biochar (32 and 27 C/N ratios for the BC15 and BC30 treatments, respectively). Low C/N ratios are typical of sewage sludge and facilitate N loss (Dume et al. 2023). However, in the case of bentonite, vermiculite or biochar addition to sludge, a considerable amount of N could be retained after its mineralization, thus increasing treated sludge's fertilizing capacity.

Biochar increased the C/N ratio at the respective treatments and probably for this reason, among sludge treatments with clay minerals or biochar, the highest reduction in microbial load was observed at the BC15 treatment. Nafez et al. (2015) reported a six-logarithmic units reduction in total *coliforms* and absence of *Salmonella spp.* during sewage sludge's composting with green plant waste and a mixture of dry leaves and pruning waste, with all composts having a C/N ratio 28–49 and pH 7.5–7.9. However biochar's condensed recalcitrant poly-aromatic structure, which was developed after pine tree residues pyrolysis (Kalderis et al. 2020), showed great stability against organic matter's decomposition and did not help mineralization which could cause temperature to increase (Singh et al. 2012). On the other hand, the literature reported that biochar enhanced organic matter's decomposition (Yu et al. 2019) and caused temperature increase during sewage sludge's composting (Malinska et al. 2014). Pathogen reduction could have also been supported by biochar's large surface area and pores, which facilitate entrapment of huge amounts of organic matter that supports the reproduction and metabolism of pathogens (Muoghalu et al. 2023).

Consequently, the bactericidal properties of bentonite, vermiculite and biochar as well as air-drying had a considerable effect on microbial load reduction in sewage sludge. However, the materials could not support a higher reduction in pathogens at the end of the 60 days period. As is reported in literature, pathogens inactivation at sludge treatment is a critical issue which is not fully satisfied even at lime treatment, where although pathogens become undetectable, bacterial spores and helminth eggs with strong resistance remain (Li et al. 2022). In addition, disrupted bacterial cells upon treatment processes can act as a nutrient substrate for pathogens to grow (Fane et al. 2021).

Effect of the clay minerals and biochar on the chemical properties of sewage sludge

At the end of air-drying period, pH remained almost neutral or increased to the slightly alkaline range in all cases, except for the RG treatments, which had strongly alkaline reaction (pH around 12) due to lime addition (Fig. 2). The EC was high at all treatments, while the RG treatments along with the untreated sludge (control) had the significantly highest values (two to five times higher than those of the rest of the treatments) (Fig. 3). It is worth noting that the increased water-soluble Ca concentrations of both limed sludge treatments were responsible for the low SAR values obtained (Fig. 4). The high EC values were attributed to the increased concentrations of basic cations (especially that of Na) of sewage sludge (Table 3), which predominated at all treatments. Thus, in the perspective of using sludge treated with the particular clay minerals or biochar as a soil amendment, a possible salinization risk should be taken under consideration. However, this risk seems to be more pronounced in the case of applying limed sludge to soil, along with possible alkalinity risk due to its high pH (Brady and Weil 2008). Balidakis et al. (2022) reported that application of limed

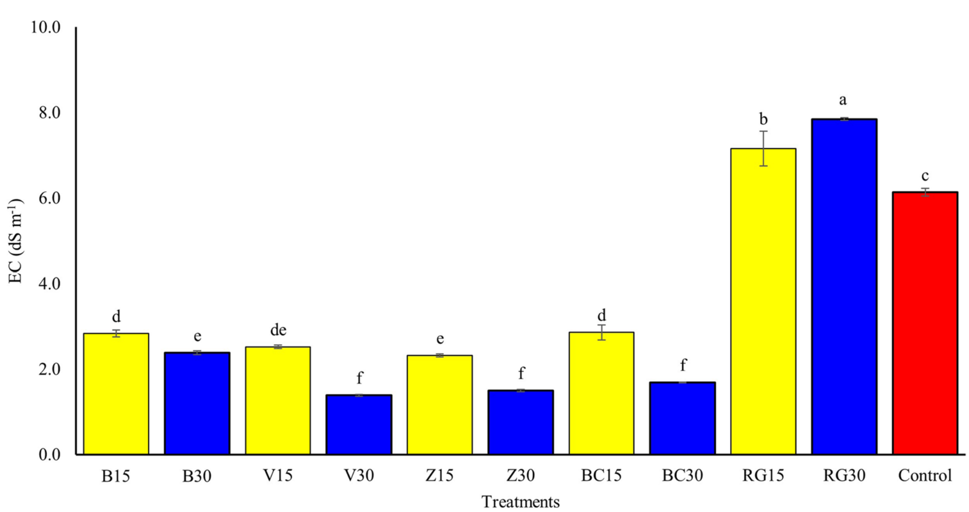

Fig. 3 Electrical conductivity (EC) of the treatments. B15, V15, Z15 and BC15: dewatered sewage sludge treated with 15% addition of bentonite, vermiculite, zeolite and biochar, respectively; B30, V30, Z30 and BC30: dewatered sewage sludge treated with 30% addition of bentonite, vermiculite, zeolite and biochar, respectively; Control: untreated, just air-dried dewatered sewage sludge. Values are presented as means ± standard error. Different letters indicate significant differences among means, using the protected LSD test, at $p \leq 0.05$. As far as ANOVA it concerns, p F test < 0.001

sludge to an acid and an alkaline soil resulted in a negative effect on the growth of white clover.

The water-soluble concentrations of NO_3-N and NH_4-N significantly decreased in almost all cases of the treated sludge in comparison with control, except for the RG15 treatment where NO_3-N concentration was the highest and the RG30 treatment where NO_3-N concentration followed that of the untreated sludge (Table S1). This was attributed to the strongly alkaline reaction of both limed treatments, which favored N nitrification (Rigby et al. 2016). As far as the NH_4-N concentrations are concerned, both limed sludge treatments had the lowest concentrations (Table S1), probably due to their alkaline reaction which led to hydrolysis of urea to NH_3 (Arthurson 2008). Based on the above, a considerable amount of bioavailable N of limed sludge could be lost through NH_3 volatilization or NO_3^- leaching either during stabilization period or upon soil application, respectively. On the other hand the B15, V15 and BC15 treatments had the significantly highest NH_4-N concentrations after the untreated sludge (Table S1), which indicates that both clay minerals, i.e., bentonite and vermiculite, as well as biochar retained a considerable amount of sludge's NH_4-N. Balidakis et al. (2023) reported lower available NH_4-N concentration when limed sludge was applied to an acid soil and lower available NO_3-N concentrations when limed sludge was applied to an alkaline soil, compared to the corresponding treatments which received treated sludge with bentonite, vermiculite or biochar.

In general, N loss in the form of NH_3 from dewatered sewage sludge is inevitable due to the decomposition of its organic matter; however, it can be minimized by the sludge's stabilization method which is applied. Malinska et al. (2014) reported a reduction of NH_3 loss up to 69% from sewage sludge compost with wood chips and biochar. They also reported that biochar increased temperature, enhanced sludge's organic matter decomposition and sorbed NH_4-N, which probably remained bioavailable (Taghizadeh-Toosi et al. 2012). As far as the clay minerals are concerned, Bourliva et al. (2010), after treating sewage sludge with bentonite, reported high sorption efficiency of N species, and Awasthi et al. (2018) mentioned that Ca-bentonite increased the bioavailable nutrient concentrations during sewage sludge's composting process. In the current study, both bentonite and vermiculite had high CEC values (Table 4), which can justify the substantial retention of NH_4-N in exchangeable, and thus bioavailable form. In addition, the fixation of NH_4-N in the interlayer space of vermiculite cannot be excluded (Nommik and Vahtras 1982).

The concentrations of water-soluble P and K significantly decreased at all sludge treatments with the materials compared to control (Table S1). However, the BC15, BC30 and B15 treatments had the highest P and K content, after the untreated sludge. Probably due to their strongly alkaline reaction, P was undetectable at both limed sludge treatments (Table S1). It is well known that P precipitates in the form of insoluble substances at the strongly alkaline and strongly acid pH ranges (Brady and Weil 2008). Overall, in almost all cases, sewage sludge treated with 15% addition of all materials had significantly higher N-P-K concentrations in comparison with 30% treatments because of increased sludge's amount in the former case (Table S1). From the water-soluble microelements, only B was detectable at all treatments,

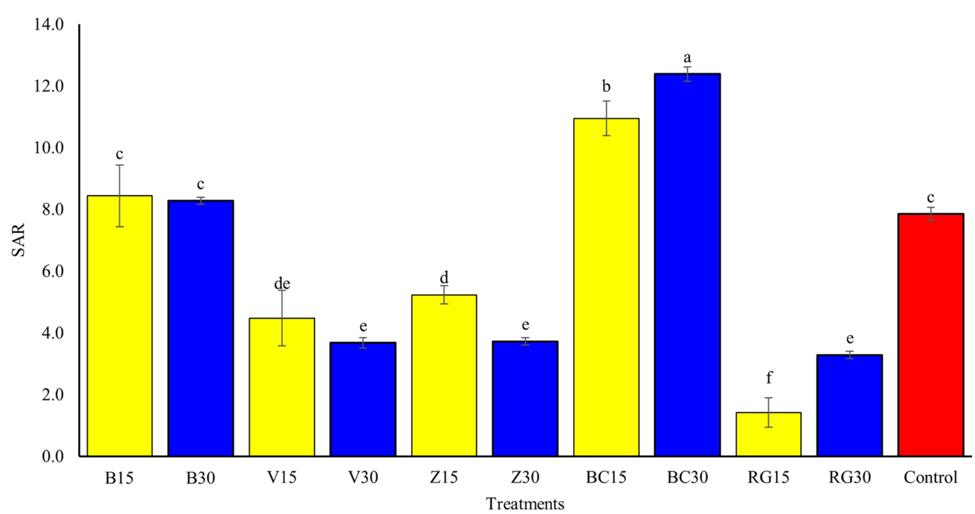

Fig. 4 Sodium adsorption ratio (SAR) of the treatments. B15, V15, Z15 and BC15: dewatered sewage sludge treated with 15% addition of bentonite, vermiculite, zeolite and biochar, respectively; B30, V30, Z30 and BC30: dewatered sewage sludge treated with 30% addition of bentonite, vermiculite, zeolite and biochar, respectively; Control: untreated, just air-dried dewatered sewage sludge. Values are presented as means ± standard error. Different letters indicate significant differences among means, using the protected LSD test, at $p \leq 0.05$. As far as ANOVA it concerns, p F test < 0.001

including control (Table S1), and this was attributed to the initial high B concentration of the dewatered sewage sludge (Table 3). The significantly highest B concentrations were obtained at the B15, V15 treatments and control, whereas the lowest concentrations were observed at the RG15 and RG30 treatments, again probably because of pH in the latter case. It is well-known that B sorption is proportionally pH-dependent and maximizes at the strongly alkaline pH range (Goldberg et al. 1993). Based on the aforementioned and for reasons previously mentioned, consideration should be given in respect to possible B phytotoxicity risk, after soil application of sewage sludge treated with the studied clay minerals or biochar.

Due to dilution of the sludge's organic matter at the treatments with materials, LOI was significantly lower than that of control in all cases, except for biochar treatments (BC15 and BC30) (Fig. 5). The same was evidenced for organic C content, whereas total N content of the untreated sludge was significantly the highest (Table S2). In general, total concentrations of the rest macro- and microelements of the treated sludge (Table S2) ranged at levels of similar magnitude to those of the untreated sludge (Table 3). However, in certain treatments macro- and microelements were increased because of the materials' total elemental composition (Table 4). For example, Mg was higher at the V15 and V30 treatments because of vermiculite, K was higher at the Z15 and Z30 treatments due to zeolite, B was higher at the BC15 and BC30 treatments because of biochar and Ca was higher at the RG treatments because of $Ca(OH)_2$. Total concentrations of all heavy metals, except for Cd and Hg which were undetectable, were far below the permissible levels regarding the agronomic use of sewage sludge, according to legislation. As for Cr, its total concentrations were far below the critical limits proposed by the 3rd Draft of "Working Document on Sludge" (European Commission 2000) (Table S2 and compare with Table 1).

Heavy metal load (Sommers et al. 1976) and speciation (Merrington et al. 2003) differ among sewage sludges and stabilization methods. For example, Shrivastava and Banerjee (2003) reported that sludge's concentrations of Cu, Zn, Pb, Ni, Cr and Cd found in the exchangeable fraction were the lowest, implying the low bioavailability of the metals, and Ignatowicz (2017) reported a similar trend. Composting sludge with organic bulking agents might yield greater reduction in microbial load (Amir et al. 2005); however, organic matter's breakdown effect on heavy metal mobility is dubious (Nomeda et al. 2008). Breakdown of sludge's organic matter to stable humic species tends to immobilize heavy metals; nevertheless, increased Mn and Zn mobility is also reported (Nomeda et al. 2008). Liming favors the immobilization of heavy metals because of alkaline pH (Arthurson 2008). In the present study, the bioavailable forms (water-soluble) of heavy metals were not detectable at all treatments (including the untreated sludge), and organic matter breakdown was not extensive (low C/N ratios even for the biochar treatments) to promote distribution of heavy metals to more mobile fractions. In any other case, clay minerals and biochar could control the mobility of sludge-born heavy metals since such a trend was observed by Qiao and Ho (1997), when red mud was added at 10% and 20% to sludge before composting.

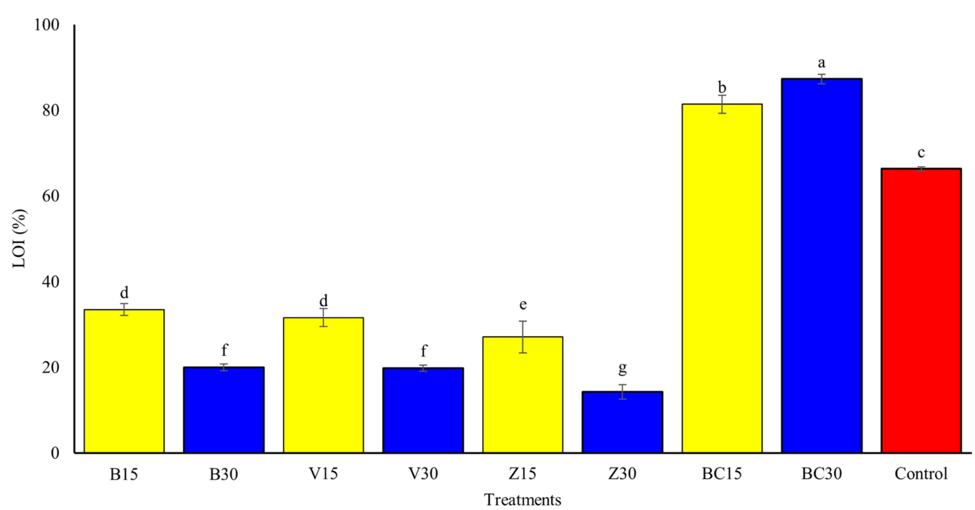

Fig. 5 Loss on ignition (LOI) of the treatments. B15, V15, Z15 and BC15: dewatered sewage sludge treated with 15% addition of bentonite, vermiculite, zeolite and biochar, respectively; B30, V30, Z30 and BC30: dewatered sewage sludge treated with 30% addition of bentonite, vermiculite, zeolite and biochar, respectively; Control: untreated, just air-dried dewatered sewage sludge. Values are presented as means ± standard error. Different letters indicate significant differences among means, using the protected LSD test, at $p \leq 0.05$. As far as ANOVA it concerns, p F test < 0.001

Sludge treatment with 15% addition of bentonite, vermiculite or biochar can be a low-cost, efficient treatment method for areas with mineral and biochar resources since it can reduce the cost and the complexity of the treatment process. When applied to soil at a 2% addition rate, the particular treated sludge can act as an organo-mineral soil amendment, which can improve soil chemical properties and supply plants with nutrients (Balidakis et al. 2022, 2023). Caution should be exercised as pathogen dissemination to the environment is concerned, since there is still insufficient information on pathogen species and concentrations, exposure pathways and corresponding dose–response effect (Li et al. 2022). Storage conditions of treated sludge should also be carefully monitored since they are a critical factor affecting pathogen survival and re-growth (Fane et al. 2021). Along with cogitation of specified environmental regulatory standards and technical costs, a life cycle assessment of sludge treated with clay minerals or biochar and its soil application should be conducted to assess the environmental impact of treated sludge and help achieve consistency in policy and environmental planning, thus sludge can turn from "waste" to "product" (Ding et al. 2021). Except for life cycle assessment, other tools such as exergy analysis could be employed for sustainability assessment. Specifically, the exergoenvironmental method can cope with the drawbacks of life cycle assessment by allocating the environmental burdens at the component level and measuring the environmental burdens of intermediate products. On the other hand, extended exergy accounting can translate all energy flows, material streams, labor fees, economic inputs and environmental remediation costs into a unified exergy scale (Aghbashlo et al. 2022). As far as our study is concerned, it would be interesting to assess whether sewage sludge treatment with bentonite, vermiculite or biochar creates less environmental burden compared to other treatment processes through exergoenvironmental analysis.

Conclusion

In the perspective of finding new treatment methods to increase sludge's merits for agriculture, addition of 15% bentonite, vermiculite or biochar to dewatered sewage sludge seems a promising simple and inexpensive method for sludge's stabilization, as far as reduction in its microbial load is concerned. In addition, load of heavy metals can be controlled also, through metals' sorption by the particular materials, especially in the case of treated sludge with the clay minerals. Regarding the potential agronomic use of the produced treated sewage sludge, the proposed method can preserve sludge's bioavailable N content, through NH_4-N sorption by the clay minerals and biochar and increase its overall fertilizing capacity (e.g., in respect to P and K), compared to the common method of sludge's liming. Moreover, biochar treated sludge seems to have a notable potential for C sequestration to soils because of biochar's recalcitrant nature. In the case of liming sewage sludge, the particular method can promote N loss, through either NH_3 volatilization or NO_3-N leaching after soil application of treated sludge in the latter case, and can decrease bioavailability of sludge's P. The use of the produced treated sewage sludge with 15% bentonite, vermiculite or biochar as soil amendments must be carefully exercised and monitored because of possible soil salinization and B phytotoxicity risks. However, the soil salinization risk is more pronounced in the case of using limed sludge as a soil amendment, in conjunction to soil alkalinity problems. In any case, these risks depend also on application rates of treated sludge to soil, soil properties and plant species.

Although most sewage sludge treatment processes can inactivate pathogens through different mechanisms, combined treatment processes are still suggested as a strategy to enhance control performance. Thus, in the near future, it would be interesting to combine this treatment process with others, like composting, vermicomposting, etc., in order to improve organic matter decomposition and pathogen load reduction. The drying effect of minerals and biochar on sludge will facilitate better conditions for subsequent composting. In addition, further insight on how clay minerals and biochar facilitate pathogen load reduction is required along with a better understanding of the life cycle of pathogens during sludge's treatment and land application.

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Author contributions AB conducted the laboratory and statistical analyses, participated in the interpretation of the results and wrote the first draft of the ms. TM had the initial idea of this research work, designed the experiment, supervised the laboratory work and the statistical analysis of data and participated in the interpretation of the results and writing of the ms. II participated in the designation of the experiment. DK produced and characterized the biochar. All authors participated in the review-editing of the ms and read and approved the final manuscript.

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Data availability All data produced during the current study are included in the article and its supplementary material.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

Ethical approval This article does not contain any studies with human participants or animals performed by any of authors.

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