Use of clay minerals for reducing sewage sludge's microbial load and nitrogen losses

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Abstract

Purpose

Preserving sewage sludge's N is important for its agronomic use, although N losses are not considered by the most common sludge's stabilization processes. This could possibly be achieved by treating sludge with certain clay minerals, which could retain N as NH_4^+ into their structure.

Methods

Nine clay minerals, i.e. low and high grade bentonite, low and high grade bentonite treated with Na_2CO_3, saponite, attapulgite, mixed clay of saponite and attapulgite, thermally modified attapulgite and zeolite (i.e. clinoptilolite), and $Ca(OH)_2$ were added to dewatered sewage sludge at rates of 0, 10, 20 and 30% (wet weight basis) (treatments). The mixtures were equilibrated for 70 days and analyzed for certain properties.

Results

The microbial indicators (load) of sludge treated with both untreated bentonite, attapulgite, mixed clay of saponite and attapulgite and zeolite, added at the rate of 30%, decreased compared to the control, drastically in certain cases (100%), and as it was expected, were not detectable in all $Ca(OH)_2$ treatments. All the aforementioned minerals' treatments were performed also well regarding sludge's retention capacity of inorganic N, with the attapulgite and zeolite treatments containing the significantly highest amounts of NO_3-N and NH_4-N, respectively. On the other hand, the treatments with both bentonites treated with Na_2CO_3 had the lowest inorganic N concentration, which was similar to that of the $Ca(OH)_2$ treatment.

Conclusions

Certain clay minerals, such as natural bentonite, attapulgite, mixed clay of saponite and attapulgite and zeolite, seem promising materials for the stabilization of sewage sludge and preservation of its N content.

Keywords

Clay minerals, dewatered sewage sludge, nitrogen retention, pathogens.

Introduction

Sewage sludge represents the insoluble residue produced during municipal wastewater treatment and subsequent stabilization procedures of the dewatered sewage sludge. Land application of sewage sludge improves soil fertility and at the same time enriches the global goal of sustainability, whereas organic material is successfully recycled back to arable land, and reduces the dependence on chemical fertilizers [1-2]. The fertilizing benefits of sewage sludge come from its content in N, P, organic matter and micronutrients [1-6]. However, sludge also contains heavy metals and pathogens which can be harmful when entering the food chain [5, 7].

The Sewage Sludge Directive 86/278/EEC regulates the use of sewage sludge in such a way that any potential harmful effect on soil, vegetation, animals and human beings is prevented. According to the Greek legislation, which is in accordance to the aforementioned European directive, sewage sludge must be stabilized before being used in agriculture. As it is mentioned in both legislations, treated sludge, defines the sewage sludge which "has undergone biological, chemical or heat treatment, long-term storage or any other appropriate process so as significantly to reduce its ferment ability and the health hazards resulting from its use" [5]. The physical and chemical properties as well as the biological parameters of the treated sludge that is produced depend on its origin and treatment process [1, 5, 7-8].

Although stabilization of dewatered sewage sludge has been widely investigated, certain aspects of the sludge's stabilization processes, which are related to the agronomic use of sludge, have not been extensively investigated. Specifically, N losses from the sludge, mainly in the form of NH_3, during the stabilization period are not considered by the most common methods and especially by those of heating and alkaline treatment. During lime-stabilization and under strong alkaline conditions release of NH_3 from the sludge was observed [1, 9-10]. Similar results have been reported for thermal and aerobic treatments because of heating and microorganism digestion, respectively [5, 11-13].

Preserving sewage sludge's N is of importance for its agronomic use as N source and this could possibly be achieved by using new stabilization processes, like treatment of the sludge with certain clay minerals. Due to their structure, strong colloidal properties and their volume increase when coming into contact with water, certain clay aluminosilicate minerals could be used to retain NH_4-N [14-16]. Montmorillonites (i.e. bentonite) and zeolites (i.e. clinoptilolite) have been used for the removal of heavy metals from sewage sludge compost and aqueous solutions [12-13, 17-22]. Palygoskites and sepiolites have been widely used in many industries to adsorb a wide range of substances like oil, toxins, bacteria and viruses. In addition, they have been used to remove NH_3 in farm environments and pollutants from wastewaters. Moreover sepiolite can also be used as a carrier for methanogenic bacteria in the production of methane from wastewater [23].

Although the purpose of stabilization of dewatered sewage sludge is the reduction of health hazards, the 86/278/EEC directive in its current form aims mainly at controlling the accumulation of heavy metals in soils after sludge application. In addition, sludge treatment standards with regard to pathogens′ reduction are not clearly defined [8]. Some countries in the E.U. in their national legislation have limit values for pathogens, Greece is not one of them [1, 24]. Furthermore, the U.S. Environmental Protection Agency classifies stabilized sewage sludge in either an end product containing pathogens below detection limits (class A) or sludge whereby the pathogen numbers are reduced but still detectable (class B) [1].

In view of the above, nine clay minerals were applied to dewatered sewage sludge at various rates and the objective of the study was to investigate the ability of the minerals in: (a) reducing sludge′s microbial indicators and (b) retaining its N during the stabilization process, in comparison to untreated and limed sludge.

Materials and Methods

Four samples of bentonite were taken from Milos island, i.e. low (B1) and high (B2) grade bentonite, low (B3) and high (B4) grade bentonite treated with Na_2CO_3. In addition, four samples of saponite or attapulgite, i.e. saponite (S), attapulgite (A), mixed clay of saponite-attapulgite (SA) and thermally modified attapulgite (ThA), and one sample of zeolite (Z) (i.e. clinoptilolite) were taken from areas of N. Greece. All samples were ground to pass a 2 mm sieve and analyzed for pH at a 1:10 w/v suspension with water and cation exchange capacity (C.E.C.) [25] (Table 1).

Dewatered sewage sludge was taken from the urban sewage treatment plant of Thessaloniki city (N. Greece). The sludge was tested for total coliform, Escherichia coli (*E. coli*), enterococcus using the Most Probable Number (MPN) methods [26-27]. In addition, salmonella spp. was also detected [28]. In addition, pH was measured at a 1:5 w/v suspension with water, dry matter was determined after drying at 105°C, organic matter was estimated by loss on ignition (LOI) [29] and KCl extractable NO_3-N and NH_4-N were determined [30] (Table 2).

At the end of September 2017, the nine minerals and a tenth material, i.e. reagent grade $Ca(OH)_2$ (RG), were mixed with the dewatered sewage sludge at rates equal to 0 (control), 10, 20 and 30 % (wet weight basis), in three replications. The mixtures (treatments) were placed in plastic pots and left for equilibration in a greenhouse for more than 70 days, with periodic mixing. The experimental design was the completely randomized (CRD) and randomization was repeated every 15 days.

During the equilibration period, sub-samples of the mixtures were collected 35 and 70 days after the initiation of the experiment and analyzed for pathogens, pH, dry matter, organic matter and available NO_3-N and NH_4-N, using the aforementioned methods. In addition, total C and N were determined by elemental microanalysis and the C/N ratio was calculated.

For each parameter determined, within each sampling, ANOVA (10 materials x 4 rates) was conducted and the LSD test, at $p \leq 0.05$, was used for mean comparisons, which were performed among rates within the same material or among materials within the same rate.

Results and discussion

After 35 days of equilibration, a reducing trend of the sludge's microbial indicators was observed upon the addition of certain minerals in comparison to the control and that trend was confirmed by the results of the second sampling (Table 3). The most efficient means of microbe's removal were the two untreated bentonites, attapulgite, mixed clay of saponite-attapulgite and zeolite at the rate of 30% and they reduced the microbial indicators by one to almost three logarithmic units (Table 3). Reduction of the microbial indicators of sludge by at least two logarithmic units, after its treatment by conventional methods, is common. On the other hand, a four logarithmic units′ reduction of sludge′s microbial indicators, achieved by advanced treatment processes, is evidence of hygienised sludge, i.e. sludge virtually free of pathogens [31].

Although the reduction observed in the present study could be considered low in terms of logarithmic units, it represented 80 to 100% reduction of fecal indicators relative to the control treatments, because of the relatively low initial microbial load of the sludge that was used (see Table 2). Moreover, although salmonella spp. was identified in the control treatments, it was not observed in the aforementioned mineral treatments (Table 3). As it was expected, the fecal indicators were not detectable in all treatments of $Ca(OH)_2$, meaning that the reduction was at 4 logarithmic units (see Table 2), and this was attributed to the pH increase in the strongly alkaline range (12.2-12.6). According to Carrington et al. [32], raising the pH of sludge to 12 by addition of slacked lime can reduce many pathogens to insignificant numbers.

The initial high moisture content of the dewatered sewage sludge (84%) was significantly and drastically reduced in all treatments and ranged from 7 to 14 % for both samplings in all mineral treatments and even the control. In the $Ca(OH)_2$ mixtures, the moisture content was significantly lower than all the other treatments and ranged from 5 to 8 % (data not shown). It is noteworthy that in all cases of sludge [with the minerals or the $Ca(OH)_2$ or just airdried], its moisture content was lower than that of sludge treated by energy costly thermal processes reported in literature. Fytili and Zabaniotou [5] mentione moisture content of ≈ 15% of sludge after pyrolysis or combustion.

As far as the pH of treatments is concerned, the results obtained in both samplings were similar and those of the second sampling are presented in Table 4. The pH of the control treatments decreased to acidic levels (Table 4) in comparison to the alkaline pH of the initial samples of the dewatered sewage sludge (Table 2). The addition of all materials to the sludge increased significantly the pH compared to control (Table 4) due to their alkaline reaction (Table 1). However, except for the two treated with Na_2CO_3 bentonites and the $Ca(OH)_2$, the addition of the rest materials at all rates increased a little the pH into the slightly acidic to slightly alkaline range. On the contrary, the addition of the two treated bentonites to the sludge increased pH into the alkaline to strongly alkaline range depended on the addition rate, probably due to the Na_2CO_3 treatment which increased the pH of the particular bentonites above 10.0 (Table 1). As it was expected all treatments containing $Ca(OH)_2$ had a strongly alkaline reaction (Table 4).

The reduction of the initial pH of the dewatered sewage sludge (Table 2) during the stabilization period (Table 4, see control) maybe ascribed to the production of humic acids, as organic matter biodegrades [33]. Shaheen et al. [20] and Murtaza et al. [34] recognized the acidifying effects of the decomposable products of sewage sludge in soil pH. The increase of sludge′s pH following the minerals′ addition, compared to the control, could be attributed to their ability to bind cations and thus H^+. Filippides et al. [17] observed an increase in the pH of an acidic solution with the addition of zeolite, which was attributed to the binding of H^+ to basic sites and to lesser extend to the H^+ retention through cation exchange reactions. On the other hand, no clear relationship between pH and zeolite's concentration was observed during composting process of sludge [12, 22].

Organic matter of the dewatered sewage sludge decreased significantly with the addition of all materials and the results of the 35-days equilibration period were almost similar to those of the 70-days, which are presented in Table 5. The decrease was not proportional to the increase of the addition rate of the materials to the sludge and depended on the material. Over the two samplings and at the 30% addition rate of the minerals, the organic matter content of the respective treatments ranged from 29 to 44 % of that of the control. Moreover, for the particular rate the highest organic matter content was obtained for saponite, followed by attapulgite and at all addition rates the lowest values were observed for $Ca(OH)_2$ (Table 5).

Although organic matter content is of outmost importance for sewage sludge′s agronomic use, this issue is totally ignored by the most common stabilization processes. As is reported in the literature, thermal treatment removes the organic part of the sludge, leaving only the ash component for the final disposal [5[. Moreover, zeolite was used in composting process of sewage sludge in order to enhance the degradability of the sludge's organic matter [12, 22].

As far as the available NO_3-N and NH_4-N of the treatments are concerned, almost similar results were obtained for both samplings and the results of the second sampling are presented in Tables 6 and 7, respectively. The NO_3-N and NH_4-N concentrations depended on the material and the addition rate. Within the same material, in almost all cases the significantly lowest NO_3-N concentrations were observed at the 30% rate. At the particular rate, the significantly highest NO_3-N concentrations were obtained for the mixtures of sludge with the two untreated bentonites, attapulgite and zeolite in the first sampling and with the same minerals and additionally the thermally modified attapulgite and the mixed clay saponite-attapulgite in the second sampling (Table 6). It is noteworthy, that within each addition rate, the significantly lowest NO_3-N concentrations were obtained for the treatments of the two bentonites treated with Na_2CO_3, followed by the $Ca(OH)_2$ treatment (Table 6).

At both samplings and for all addition rates, the highest concentrations of NH_4-N were observed in the zeolite treatments and the lowest in the $Ca(OH)_2$ treatments. At the rate of 30%, the mixtures with the rest of the minerals had similar NH_4-N content (see Table 7 for the second sampling). In most of the treatments with minerals, NH_4-N retention capacity was higher or equal to those reported for most common stabilization processes, where N losses can reach ≈ 60 % [5, 10-11].

Moreover, zeolite's good performance in retaining NH_4-N could be attributed to its high C.E.C. Although the external C.E.C. was lower than the total C.E.C. of the other minerals (Table 1), the total C.E.C. of the clinoptilolite used is expected to be ≈ 200 $cmol_c$ kg^{-1} [14]. According to literature, zeolite has been used during composting of sludge and increased NH_4^+ retention capacity due to reactions of cation exchange and adsorption [12-13]. However, Zorpas et al. [22] report that during sludge′s composting process the concentration of NH_4^+ in the final product decreased when the amount of zeolite increased at the addition rates of 25 and 30 %.

The C/N ratio of the treatments was significantly affected by the material and the application rate and an increasing trend was evidenced with the increase of the application rate of the materials, except for zeolite (Table 8). In all cases of the treatments, even the control, the C/N ratio ranged at levels which are considered low for organic materials. However, such values ($C/N < 20$) are permissible for the potential agronomic use of treated sludge as soil amendment [35].

Conclusions

The results of this study showed that certain clay minerals, such as natural bentonite, attapulgite, mixed clay of saponite-attapulgite and zeolite, seem promising materials to be used (alone or in combination with other substances) for sewage sludge's sanitation. Furthermore, they can preserve more sludge's N than that of other conventional treatments. In addition, they are economic materials found in Greece and they could be used from the local communities to treat sludge and apply it in agricultural land. In other words, it is possible that the use of certain clay minerals could be an alternative way to treat sludge for its agronomic use.

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Table 1. The pH and cation exchange capacity of the nine minerals

Property	Minerals								
	B1	B2	B3	B4	S	A	SA	ThA	Z
pH (1:10 H_2O)	7.9[a] ± 0.1[b]	9.5 ± 0.0	10.5 ± 0.2	10.1 ± 0.1	8.6 ± 0.6	8.3 ± 0.1	8.5 ± 0.1	7.6 ± 0.2	8.9 ± 0.1
C.E.C. ($cmol_c\ kg^{-1}$)	66.9 ± 0.2	80.9 ± 0.8	81.1 ± 1.6	78.1 ± 0.9	55.2 ± 0.5	30.0 ± 0.2	34.9 ± 0.3	38.2 ± 0.0	12.7[c] ± 1.6

[a] Mean

[b] Standard deviation

[c] External C.E.C.

Table 2. Certain properties of the dewatered sewage sludge[a]

Total coliform (MPN g^{-1})	*E. coli* (MPN g^{-1})	Enterococcus (MPN g^{-1})	Salmonella spp. (per 25 g)
3.5 10^4	2.9 10^4	3.4 10^4	Detectable

pH (1:5 H_2O)	Dry matter (g kg^{-1})	LOI (g kg^{-1})	NO_3-N (mg kg^{-1})	NH_4-N (mg kg^{-1})
8.1[b] ± 0.0[c]	162 ± 2	656 ± 2	66.5 ± 4.3	715 ± 39

[a] All concentrations are expressed on wet weight basis.

[b] Mean

[c] Standard deviation

Table 3. Reduction of the fecal indicators (in logarithmic scale) and salmonella spp. of the 30% treatments in respect to the dewatered sewage sludge, after 70 days of equilibration[a]

Microbial load	Material								
	B1	B2	B3	B4	S	A	SA	ThA	Z
T. colifrom	1.8	1.1	0.4	0.0	0.8	2.4	2.8	2.2	2.5
E. coli	2.0	1.0	0.3	1.2	1.9	2.1	1.7	1.7	1.4
Enterococcus	1.4	1.3	1.3	0.3	0.0	0.7	2.1	0.0	1.9
Salmonella spp.	ND[b]	ND	D[c]	ND	D	ND	ND	ND	ND

[a] In all cases of the RG treated sewage sludge the microbial load was non-detectable.

[b] Non-detectable

[c] Detectable

Table 4. The pH of all treatments, after 70 days of equilibration

Rate	Material									
	B1	B2	B3	B4	S	A	SA	ThA	Z	RG
0	6.6lm*	6.5mn	6.5mn	6.6lm	6.5mn	6.4no	6.5mn	6.5mn	6.5mn	6.3o
10	6.9ij	6.8jk	7.8f	7.5g	6.8jk	6.7kl	6.7kl	6.7kl	6.7kl	12.2b
20	6.7kl	6.9ij	8.4d	7.9f	7.0i	7.0i	7.0i	6.8jk	6.7kl	12.6a
30	6.8jk	7.2h	8.8c	8.2e	7.0i	7.2h	7.2h	7.0i	6.8jk	12.6a
p F-test										
Material $p < 0.001$										
Rate $p < 0.001$										
Interaction $p < 0.001$										

* Means followed by different letters are statistically different using the LSD test, at $p \leq 0.05$. Mean comparisons were conducted among rates within the same material (column) or among materials within the same rate (row).

Table 5. Organic matter (as LOI %) of all treatments, after 70 days of equilibration

Rate	Material									
	B1	B2	B3	B4	S	A	SA	ThA	Z	RG
0	63.1a*	63.4a	62.5a	62.6a	64.6a	64.8a	64.2a	64.6a	64.3a	63.2a
10	38.6cde	37.6def	41.8bcd	40.3bcd	43.7b	43.3b	43.1b	40.5bcd	42,1bc	34.9efg
20	27.8jkl	27.3jkl	26.1jkl	27.1jkl	33.7fgh	28.1jk	32.9ghi	30.0hij	28,9ij	18.4n
30	20.9mn	20.7mn	20.6mn	20.3mn	28.7ij	27.8jkl	23.9klm	23.5lm	24,2klm	1.6o
p F-test										
Material $p < 0.001$										
Rate $p < 0.001$										
Interaction $p < 0.001$										

* Means followed by different letters are statistically different using the LSD test, at $p \leq 0.05$. Mean comparisons were conducted among rates within the same material (column) or among materials within the same rate (row).

Table 6. Available NO_3-N (mg kg^{-1}) of all treatments, after 70 days of equilibration

Rate	Material									
	B1	B2	B3	B4	S	A	SA	ThA	Z	RG
0	764no*	1071jkl	1221efghij	976klm	1125hijk	1197fghij	1616ab	1688ab	1723ab	1304defg
10	1375de	1590bc	534q	717op	1370def	1369def	1617ab	1429cd	1774ab	959klm
20	1107ijk	1237efghij	226r	244r	1162ghij	1087ijkl	1237efghij	1255defghi	1287defgh	586pq
30	925lmn	802mno	166r	145r	776no	1417cd	861mno	842mno	975klm	496q
p F-test										
Material $p < 0.001$										
Rate $p < 0.001$										
Interaction $p < 0.001$										

* Means followed by different letters are statistically different using the LSD test, at $p \leq 0.05$. Mean comparisons were conducted among rates within the same material (column) or among materials within the same rate (row).

Table 7. Available NH_4-N (mg kg^{-1}) of all treatments, after 70 days of equilibration

Rate	Material									
	B1	B2	B3	B4	S	A	SA	ThA	Z	RG
0	450nop*	633jklm	680ijklm	804hij	707hijkl	1308d	1635b	1335cd	1510bc	820ghi
10	988efg	1025ef	531lmno	376opq	1054e	1012ef	1018ef	1115e	1869a	72r
20	567klmn	514mno	358opq	384opq	870fgh	749hij	746hijk	669ijklm	1348cd	64r
30	374opq	255q	310pq	351opq	374opq	380opq	383opq	406nopq	1099e	54r
p F-test										
Material $p < 0.001$										
Rate $p < 0.001$										
Interaction $p < 0.001$										

* Means followed by different letters are statistically different using the LSD test, at $p \leq 0.05$. Mean comparisons were conducted among rates within the same material (column) or among materials within the same rate (row).

Table 8. The ratio C/N of all treatments, after 70 days of equilibration

Rate	Material									
	B1	B2	B3	B4	S	A	SA	ThA	Z	RG
0	6.9hij*	6.9hij	7.0ghi	6.9hij	6.9hij	6.8hijk	6.8hijk	6.8hijk	6.7ijkl	7.0ghi
10	6.8hijk	7.0ghi	7.4def	7.5de	6.8hijk	6.7ijkl	6.6jkl	6.5kl	6.0m	7.7cd
20	7.1fgh	7.1fgh	8.0bc	7.7cd	7.0ghi	7.1fgh	6.5kl	6.4l	5.6n	7.6de
30	7.3efg	7.3efg	8.4a	8.2ab	7.3efg	7.5de	7.4def	6.6jkl	5.3n	7.6de

p F-test

Material $p < 0.001$

Rate $p < 0.001$

Interaction $p < 0.001$

* Means followed by different letters are statistically different using the LSD test, at $p \leq 0.05$. Mean comparisons were conducted among rates within the same material (column) or among materials within the same rate (row).