Treatment of sewage sludge with the use of solarization and sanitizing products for agricultural purposes

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Recycling of sewage sludge for agricultural purposes is recommended as one of the most adequate forms of final disposal of this waste. This study evaluated the effectiveness of solarization combined with chemical treatments by acid and alkali during different periods of cleaning. The experiment was conducted at the Experimental Farm of the Federal University of Uberlândia (UFU) in Uberlândia-MG. The experimental design used randomized blocks in a 5x3+1 factorial arrangement with four replications. The factor plots consisted of sanitizing products (260 mg L\(^{-1}\) peracetic acid, 2400 mg L\(^{-1}\) quaternary ammonium compounds, hydrated lime equivalent to 30% of the dry mass of the sewage sludge, 2500 mg L\(^{-1}\) sodium hypochlorite, and pure sludge) for different times: T1 = 7 days, T2 = 14 days, and T3 = 21 days. Data were also collected from the pure mud at time zero. The concentration of fecal coliforms, pH, N (Nitrogen), Na (Sodium), Al (Aluminium), Ca (Calcium), Mg (Magnesium), K (Potassium), OM (Organic Matter), C (Carbon), Cr (Chromium), Ni (Nickel), Cd (Cadmium), Pb (Lead), Cu (Copper) and Zn (Zinc) were all evaluated. Lime increased the concentration of Ca and Mg in the biosolids, reduced the level of fecal coliforms below the limits specified by environmental standards from seven days and decreased the levels of available N, Al, OM, C, Na, Cr, Ni, Cd, Cu and Zn in the biosolids.

Key words: Environment, heavy metal, mineral nutrient, micro-organism.

INTRODUCTION

Sewage, processed in treatment plants (STPs), undergoes chemical, physical, and biological processes in order to achieve the standards determined by the Brazilian environmental legislation. After treatment, the liquid is usually released into local bodies of water. The semisolid material is called sewage sludge.

Agricultural recycling of sewage sludge is one of the most adequate methods of disposing of this waste in terms of technical, economic and environmental requirements (Barbosa et al., 2007). For Camargo et al. (2013), the use of sewage sludge in agriculture, as organic fertilizer, is the most promising alternative for the final disposal.
of this waste. De Maria et al. (2007) found that the application of sewage sludge during two successive years resulted in an increase in the organic matter content and aggregate stability of the oxisol in the 0-10 cm layer.

Studies conducted by Franco et al. (2010) showed that raw sewage, when supplemented by mineral fertilizer, increased the productivity of sugarcane as compared to conventional fertilization. The sewage did not change the technical quality of the plants. Melo et al. (2007) observed that the use of sewage sludge as fertilizer on corn increased productivity as compared to the application of mineral fertilizer. Santos et al. (2014) found that sewage sludge increased fertility for seedling production, mainly in terms of nitrogen, calcium and phosphorus.

However, the principal factor limiting the use of sewage sludge in agriculture is the presence of high levels of heavy metals, various organic pollutants and pathogenic micro-organisms (Barros et al., 2011; Thomas-Soccol et al., 2000). Sewage sludge can present various pathogens capable of causing disease in humans and animals (Thomaz-Soccol et al., 2000). Among the pathogens, the following groups may be present in sewage sludge: helminths, protozoa, fungi, viruses and bacteria (Sidhu and Toze, 2009). The quantity of pathogens in sewage sludge depends on its origin, the time of year and the treatment process (Thomas-Soccol et al., 2000). With regard to the bacterial content, the CONAMA (National Council of the Environment) Resolution no 375/2006 of the Brazilian Environmental requires the analysis of fecal coliforms and the bacteria being used as a bacterial indicator for assessing the quality of the sewage sludge for agricultural use (Brazil, 2006).

The processing methods most commonly used include aerobic and anaerobic digestion, chemical treatment with an alkaline medium, composting, solar radiation and thermal drying (Andreoli et al., 2001).

Solarization is the process by which sewage is subjected to the heat and rays of the sun in containers covered by film. This is an alternative for sanitizing sewage at a low cost.

The chemical cleaning of sewage sludge using an alkaline product, normally lime, capable of raising the pH of the sludge deactivates most of the pathogenic microorganisms (Dores-Silva et al., 2011). There are also other possibilities for processing sludge, including the use of acetic acid, peracetic acid, sodium hypochlorite and quaternary ammonium salts (Barros et al., 2011; Barros et al., 2006; Daschner, 1997).

Recycling of sewage sludge for use in agriculture thus requires cleaning according to the standards established by the requirements of CONAMA, Resolution n° 375/2006, to ensure health and environmental safety (Brazil, 2006).

The objective of this study was to evaluate the efficiency of solarization combined with chemical treatment using acid and alkaline during different cleaning times.

MATERIALS AND METHODS

The experiment was conducted at the Glória Experimental Farm of the Federal University of Uberlândia (UFU) during 21 days of August, 2013. The sewage sludge was collected from the Uberabinha Sewage Treatment Plant of DMAE - Department of Water and Waste water, located in Uberlândia, MG. The sewage sludge was extracted from an anaerobic reactor UASB type (Upflow Anaerobic Sewage Sludge Blanket) from domestic sources after passing through a dewatering process by adding cationic polymers (FeCl₃) and centrifuging to a moisture level of 71.21 and 28.79% dry matter. The material was collected at the exit of the reactor.

The experimental design was a randomized block in a factorial 5×3+1, with four replications of factor plots consisting of sanitizing products (260 mg L⁻¹ peracetic acid, 2400 mg L⁻¹ quaternary ammonium compounds, hydrated lime equivalent to 30% of the dry mass of the sewage sludge, 2500 mg L⁻¹ sodium hypochlorite, and pure mud) for different times: T₁ = 7 days, T₂ = 14 days, and T₃ = 21 days. Data were also collected from the pure sewage sludge at time zero. The total experiment thus consisted of 64 subplots (Table 1).

The plots consisted of metal boxes of 0.30 × 0.23 × 1.0 meters on pedestals in order to eliminate as much interference (humidity and temperature) from the ground, as possible. Each drug treatment with peracetic acid, sodium hypochlorite, quaternary ammonium compounds and hydrated lime, with the addition of pure sludge, was blended in a mixer for three minutes. In each metal box, corresponding to one experimental unit (plot), 30 kg of the mixture, containing sewage sludge and sanitizing material, was placed for processing to pure mud. Instruments to measure temperature were implanted in each box at a depth of five cm in the sludge mass and 40 cm from the end of the receptacle. Data were stored in a datalogger, model CR 1000 (Campbell Scientific®), calibrated to record daily temperatures at 30 min intervals throughout the experimental period.

Each unit, composed of five boxes, was covered with transparent glass of a thickness of 5.0 mm, in order to form a greenhouse and prevent the entrance of moisture, such as rainfall, from the external environment. A ribbon was placed between the glass and the enclosures to prevent the entrance of air and moisture. A spatula that had been autoclaved was used for the collection of each sample. Within each plot, four sub-samples were collected at different depths of the sludge, extending to the bottom of the box. The analysis of thermo tolerant fecal coliforms was carried out in the Environmental Microbiology Laboratory of the Federal University of Uberlândia using the technique of multiple pipes, recommended by the United States Environmental Protection Agency for sludge analysis (USEPA, 2006) (Table 2).

Sewage sludge samples were submitted to nitropercloric digestion. The total contents of P, K, Ca, Mg, Cu, Zn, In, Ni, Cd, Pb, Al and Mg were determined in the extract by means of atomic absorption spectrophotometry with an acetylene flame and via spectrometer examination of the plasma, respectively, analysis ICP/OES, simultaneously, according to the methodology proposed by EMBRAPA (2009) (Table 2). In order to determine the total nitrogen, sulfuric acid digestion was performed (Kjeldahl method). To determine the pH and humidity at 105°C, the methodology of EMBRAPA (2009) was used. The method for determining the organic carbon was based on the oxidation of the organic matter.
Table 1. Treatments used in the sewage sludge cleaning process and their concentrations at 7, 14 and 21 days.

<table>
<thead>
<tr>
<th>Treatments</th>
<th>Chemical concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sludge + peracetic acid</td>
<td>260 mg L⁻¹</td>
</tr>
<tr>
<td>Sludge + Quaternary compounds of ammonium¹</td>
<td>2400 mg L⁻¹</td>
</tr>
<tr>
<td>Sludge + Hydrated lime</td>
<td>30% of the sludge dry matter</td>
</tr>
<tr>
<td>Sludge + Sodium hypochlorite</td>
<td>2500 mg L⁻¹</td>
</tr>
<tr>
<td>Pure sludge without chemical</td>
<td>-</td>
</tr>
</tbody>
</table>

¹ Dicetyl ammonium chloride, alkyl amido propyl chloride, dimethylbenzyl ammonium, alcohol and water.

Table 2. Characteristics of anaerobic sewage sludge from the treatment plant STP - Uberabinha, Brazil.

<table>
<thead>
<tr>
<th>Determinations</th>
<th>Units</th>
<th>Analytical Results¹</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermo tolerant coliforms</td>
<td>MPN g⁻¹ of ST</td>
<td>2.87 × 10⁷</td>
<td>USEPA, Method (1681)</td>
</tr>
<tr>
<td>pH CaCl₂ 0.01 mol L⁻¹</td>
<td>-</td>
<td>8.62</td>
<td>EMBRAPA² (2009)</td>
</tr>
<tr>
<td>Nitrogen – N</td>
<td>g kg⁻¹</td>
<td>30.01</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Sodium – Na</td>
<td>g kg⁻¹</td>
<td>0.75</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Aluminum – Al</td>
<td>g kg⁻¹</td>
<td>39.34</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Phosphorus – P</td>
<td>g kg⁻¹</td>
<td>9.60</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Calcium – Ca</td>
<td>g kg⁻¹</td>
<td>17.30</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Magnesium – Mg</td>
<td>g kg⁻¹</td>
<td>2.60</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Potassium – K</td>
<td>g kg⁻¹</td>
<td>1.0</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Organic matter - OM</td>
<td>g kg⁻¹</td>
<td>573.91</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Organic carbon – C</td>
<td>g kg⁻¹</td>
<td>332.89</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Chromium – Cr</td>
<td>mg kg⁻¹</td>
<td>166.72</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Nickel – Ni</td>
<td>mg kg⁻¹</td>
<td>31.93</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Cadmium – Cd</td>
<td>mg kg⁻¹</td>
<td>0.94</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Lead – Pb</td>
<td>mg kg⁻¹</td>
<td>ND</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Copper – Cu</td>
<td>mg kg⁻¹</td>
<td>211.00</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Zinc – Zn</td>
<td>mg kg⁻¹</td>
<td>1500.00</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>Humidity to 105°C</td>
<td>-</td>
<td>71.21</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>MS at 105° C</td>
<td>-</td>
<td>28.79</td>
<td>EMBRAPA (2009)</td>
</tr>
<tr>
<td>C/N – Total</td>
<td>-</td>
<td>9.91</td>
<td>EMBRAPA (2009)</td>
</tr>
</tbody>
</table>

¹ = value on a dry matter basis; MPN: Most Probable Number; MS - Dry Mass; ND-Not Detected. ²/ EMBRAPA (Brazilian Agricultural Research Corporation).

RESULTS AND DISCUSSION

The average concentration of thermo tolerant coliforms in the sewage sludge before cleaning was 2.87 × 10⁷ g MPN ST⁻¹, an amount within the range established by Sidhu and Toze (2009) (Table 3).

The hydrated lime, with an increase in pH (12.65) and production of NH₃, reduced the concentration of fecal coliforms to acceptable values according to the CONAMA Resolution nº 375/2006, established for 10³ MPN g⁻¹ at seven days after mixing with the sewage sludge (Table 3).
Fia et al. (2005) reported that the liming process, which raises the pH of the sludge to slightly more than 12 through the addition of hydrated lime (Ca (OH)$_2$), eliminated most of the pathogens present in the residue.

There was no significant difference in the reduction of fecal coliforms in the treatment with hydrated lime among the seven, 14 and 21 days. The concentration of coliforms in the limed sludge after 14 and 21 days was unchanged, indicating that even the coliforms still present in the residue showed no regrowth. In the external environment microorganisms do not, typically, multiply, requiring an intermediate host (Thomaz-Soccol et al., 2000). Quaternary ammonium salts produced no efficient reduction in coliforms. The measured values did not reach the standard set by the Brazilian Environmental Standard for fecal coliforms as elaborated in CONAMA Resolution nº 375/2006. Similar results have been reported by Daschner (1997) and Miyagi et al. (2000), noting that the quaternary ammonium compounds do not exhibit effectiveness in reducing microorganisms in conditions of a high level of organic matter or media with high salt contents. It was noted that these compounds have low inhibitory effects on gram-negative bacteria such as those belonging to the group of fecal coliforms. The sludge, without the addition of sanitizing products, presented decreased levels of fecal coliforms on days 7, 14 and 21, but did not reach the limits established in CONAMA Resolution nº 375/2006 (Table 3) for agricultural use. Regardless of the method of sanitation, a decrease was observed in the concentration of fecal coliforms at 21 days, influenced mainly by the reduction of moisture in the residue.

Sodium hypochlorite, at a concentration of 2500 mg L$^{-1}$, did not reduce the fecal coliform values to the level recommended by CONAMA Resolution nº 375/2006, due mainly to the amount of solids in the sewage sludge, the high content of organic matter, low moisture content and the initial pH of the alkaline sludge (Table 3). Aisse et al. (2001) pointed out that the presence of large quantities of solids can protect micro-organisms from disinfecting action and, under conditions of elevated pH; the germicidal effect of chlorine is reduced. The presence of a high moisture content in the sewage sludge permits a longer time of contact of the disinfecting agent with microorganisms. Barros et al. (2011) observed, however, that sludge with 98% humidity, sanitized with sodium hypochlorite at 2500 mg L$^{-1}$, produced material that was free of pathogens.

Peracetic acid, also, did not reduce the level of fecal coliforms to the limits established by the CONAMA Resolution of the Brazilian Environmental Standard (Table 3). With the application of organic acids such as peracetic acid, the cleaning process of sewage sludge is normally conducted in liquid sludge with high moisture content and therefore a high potential for inactivation of micro-organisms, thus requiring only 10 minutes for a complete reduction (Barros et al., 2006). In the present study the acid was mixed with semi-pasty sludge, with 71.21% moisture and pH of 8.62, resulting in a low efficiency in reducing fecal coliforms. Peracetic acid has a higher efficiency in the inactivation of micro-organisms when the sludge has an acid pH and high humidity conditions (Barros et al., 2011).

The average temperature of the sewage sludge ranged from 19.66°C to 22.88°C from the initial to the final time of each day, with peak temperatures at 13 h and 16°C of variation between 35.24 to 39.98°C among the treatments evaluated (Figure 1).

The temperatures recorded in the experimental plots helped reduce the thermo tolerant fecal coliform level of 1.67 log but did not reach the thresholds required by CONAMA Resolution nº 375/2006 for agricultural use. Additional cleaning processes for adequate reduction of fecal coliforms were thus necessary. For Tchobanoglous et al. (1993), the lethal temperature for *Escherichia coli*, principal representative of the thermo tolerant coliforms, is 55°C for 60 min. The time of year, characterized by shorter days and lower daily temperatures, contributed to...
lower average temperatures in the mass of the mud.

The statistical model, which analyzed the split plots for inorganic zinc, copper, cadmium and nickel, yielded significant differences between the main treatments (pure mud and sludge homogenized with sanitizing products) on average, compared by the Scott-Knott Test (Table 4). Pronounced effects among secondary treatments (time differences) were only found for zinc and cadmium. There was no significant interaction between the main factor and the secondary factor for all of the analyzed inorganic substances.

The concentration of lead in the sewage sludge was below the detection limit and therefore excluded from statistical analysis. The additional (control) treatment, when compared with each of the primary and secondary treatments, did not yield significant differences, indicating that additions of sanitizing products had no effect on the sewage sludge attributes: zinc, copper, cadmium, chromium and nickel (Table 4). The times of evaluation did not demonstrate influence on the concentrations of copper, chromium or nickel present in the sanitized sewage sludge (Table 4). The sludge mixed with hydrated lime, on the other hand, significantly reduced the availability of zinc, copper, cadmium, chromium and nickel in relation to other means tested. The Ni, Cd, Zn, Cr and Cu levels were also influenced by the pH and basic conditions, which favored the passage of soluble forms over those with lower solubility. In general heavy metals are less soluble under intense alkaline conditions (pH>12.0). Hydrated lime was used to raise the pH above 12.0. This provoked precipitation of the heavy metals in the form of insoluble compounds, hydroxides, phosphates and carbonates with the organic matter of the sewage. Similar results have been reported by Akrivos et al. (2000), who observed smaller amounts of Ni, Zn, Cr, Cd and Cu in limed sludge. For Matos and Matos (2012), the limed sludge complies with environmental legislation. It had low concentrations of heavy metals readily available for plants, indicating a lower risk of environmental contamination.

The values found indicated that the sludge subjected to cleaning did not exceed the limits of metals set by environmental standards for agricultural recycling. Even the pure sludge, without the addition of chemicals, was below the required limits. According to Rocha et al. (2003), sewage sludge, of exclusively domestic origin, has low levels of heavy metals. Levels of zinc had the highest concentration at 21 days, with a value of 2,670 mg kg\(^{-1}\). For Haynes et al. (2009) and Houhou et al. (2009) domestic sewage sludge is generally rich in Zn.
The hydrated lime, in a proportion of 30% of the dry mass of the sludge, produced the highest mean pH with 12.65 (Table 5). Similar results have been reported by Fia et al. (2005) and Matos and Matos (2010).

Regarding the time periods evaluated, it was found that the seven day values had the highest average pH. At 14 and 21 days, there was a decline in mean pH values. No significant differences were found between 14 and 21 days (Table 5). The other tested treatments: peracetic acid, quaternary ammonium compounds, sodium hypochlorite and pure mud, did not differ among themselves for the pH parameter, as tested by the Scott-Knott Test at 5% significance.

The limed sludge had the lowest mean values of organic matter: 368.60 g kg⁻¹, with 213.8 g kg⁻¹ organic carbon and 20.88 g kg⁻¹ nitrogen (Table 5). According to Nascimento et al. (2014) and Pedroza et al. (2006), oxidation of the organic matter and loss of nitrogen are associated with the volatilization of ammonia, under alkaline conditions.

The times analyzed did not influence the total concentration of the OM, C or N parameters studied. According to these parameters, the additional (control) treatment had higher averages than the limed sludge values for the cleaning processes evaluated (Table 5).

Regarding aluminum (Al), calcium (Ca) magnesium (Mg) and phosphorus (P), it was found that the limed sludge, in a proportion of 30% of the sludge dry matter, had the highest average values of calcium (302.0 g kg⁻¹) and magnesium (4.2 g kg⁻¹) because hydrated lime has calcium and magnesium in its composition. It also has, however, lower total levels of aluminum (20.10 g kg⁻¹) and phosphorus (1.6 g kg⁻¹). The reduced availability of aluminum and phosphorus can also be explained by the...
formation of less soluble compounds such as calcium phosphate and aluminum hydroxide. According to Andreoli et al. (2001), limed sludge, with a pH above 12.0, causes the fixing of heavy metals and phosphorus insolubility.

Considering the secondary factor (times), there was no significant difference in the parameters of Al, Ca, Mg and P, but significant change was found in the further treatment (control) by the Dunnett test at a 5% level of significance. Thus, the limed sludge increased the calcium and magnesium, reducing the availability of phosphorus, aluminum and sodium. Regarding the total content of sodium, it was found that the sludge subjected to treatment with sodium hypochlorite at a dosage of 2500 mg L⁻¹ had increased levels, with mean values of 4.10 g kg⁻¹, different from the initial mean values of 0.75 g kg⁻¹ (Table 5). No significant differences were observed among the other means tested and there was no influence of time on the total sodium concentration. Barros et al. (2011), by applying 50 t ha⁻¹ of sewage sludge, sanitized with sodium hypochlorite (2500 mg L⁻¹), obtained a concentration of 1200 mg kg⁻¹ of sodium in the stalks of corn without any observed phytotoxicity. However, high levels of sodium in the sludge can cause problems of phytotoxicity to the crop and continued use could lead to an increase in sodium levels in the soil. Andreoli et al. (2001) reported that 1.4g kg⁻¹ of sodium in the sludge is considered high and, in the soil, can lead to problems of salinity.

The average concentration of potassium in the sewage sludge was low, with mean values of 1.0 g kg⁻¹ (Table 5). Similar results have been reported by Franco et al. (2010), who found values of 1.0 kg⁻¹ potassium in sewage sludge, indicating the need for mineral supplementation if it is to be used in agriculture. The values for potassium showed no significant effect from treatment with peracetic acid, sodium hypochlorite, hydrated lime or clean mud during the times evaluated. There was also no difference between the control treatment and the main and secondary factors. Thus, it was observed that the treatments to produce hygienic material did not influence the total potassium concentration. The times also showed no effect on the average concentrations of these sanitizing products.

Conclusion

Sewage sludge, homogenized with quaternary ammonium compounds, sodium hypochlorite and peracetic acid, subjected to heat did not reduce the concentration of fecal coliforms below 1,000 MPN g⁻¹ of total solids (TS). The heavy metal (Zn, Cu, Cd, Cr, Ni and Pb) contents, however, were below the limits established by CONAMA Resolution nº 375/2006.

The lime, applied in a quantity equal to 30% of the weight of the dry matter of the sludge, showed strong alkalinity and decreased the concentration of fecal coliforms below the limits established by the CONAMA Resolution. The increase in the pH of limed sludge resulted in lower availabilities of N, P, Na, MO, C, Al, Zn, Cu and increases in Ca and Mg.

Conflict of Interests

The authors have not declared any conflict of interests.

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