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Research on suitability of anaerobically treated green waste and sewage sludge mixtures with meat waste for soil fertilization

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To evaluate the suitability of biodegraded organic waste for soil fertilisation, an experiment aimed at determining the amounts of total nitrogen, total phosphorus and heavy metals contained in this waste and determining its pH was carried out. Organic waste used for research consisted of mixes of meat waste (MW) with green waste (GW) and meat waste (MW) with sewage sludge (SS) in proportion 3 : 1, 1 : 1 and 1 : 3. MW and SS (1 : 3) mixture was found to be the most suitable organic fertiliser to remediate exhausted soils and used-up areas. This mix was determined to contain the biggest contents of nitrogen and phosphorus, 4.89 mg/kg and 227.0 mg/kg respectively. Heavy metal concentrations were also found the highest among the researched cases, however they did not exceed the maximum permissible concentration (MPC) in soil when fertilized with sludge. The concentration of chromium amounts to 1.05 mg/kg, that of zinc – 15.73 mg/kg, lead – 7.74 mg/kg, copper – 29.88 mg/kg and nickel – 11.05 mg/kg. MW and SS (1 : 3) mixture is mostly suitable for the fertilisation of acid soils, because its pH value was 6.8.

Key words: fertilisation, soil, organic waste, total nitrogen, total phosphorus, heavy metals (Cr, Zn, Pb, Cu, Ni), pH

INTRODUCTION

Nowadays increasing amounts of generated waste are one of the most important environmental problems. Biodegradable waste accounts for more than 50% of municipal waste (Brazas, 2008). EU Directive 1999 / 31 / EC lays down the requirement to reduce volumes of waste disposed of in landfills. Anaerobic digestion of waste in bioreactors is one of the most promising techniques to convert biowaste into alternative energy and still obtain a fertiliser (Gunaseelan, 1997; Results..., 2004). It also helps to a certain extent deal with the problem of unpleasant odours. Unpleasant smelling hydrogen sulphide evolved during biodegradation is removed from the gas before it is supplied to the users (Stankevičiūtė et al., 2007) and therefore its content in treated organic waste is already low.

Treated organic waste contains biodegraded organic waste that could be used for soil fertilisation but it has to comply with the requirements for nitrogen, phosphorus, potassium and heavy metal contents in it, as well as the level of pH.

Nitrogen is vital for plant growth. Plants draw nitrogen almost only via their roots from soil and its main sources are ammonium and nitric acid salts (Tognetti et al., 2007). Consequently, plants faster assimilate nitrate nitrogen, whereas ammonia nitrogen becomes more accessible to plants only after the process of nitrification is completed, i. e. when nitrogen is converted into nitrate nitrogen (Piaulokaitė-Motuzienė, Končius, 2007). However, this form of nitrogen is rapidly leached out and therefore plants have to be fertilised (Liang, 1994).

Even though nitrogen is a fertiliser but upon accessing ground waters it can pollute them (Keeney, 1989) and therefore its content is soil is limited when fertilising it with sludge. The content of nitrogen in soil is regulated by LAND 20-2005 "Requirements for Sewage Sludge Usage for Fertilisation and Remediation". According to the LAND requirement, the maximum rate of fertilisation with sludge should ensure that soil is accessed by no more than 170 kg/ha of nitrogen per year.

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Phosphorus is also very important for plant growth. Since 98–99% of phosphorus that accesses soil is not leached out, even upon applying bigger amounts of the phosphoric fertiliser, plants can take its excess later. The amount of phosphorus in soil is regulated by LAND 20-2005 "Requirements for Sewage Sludge Usage for Fertilisation and Remediation". It stipulates that the maximum rate of fertilisation with sludge should ensure that soil is accessed by no more than 40 kg/ha of phosphorus per year.

Especially relevant is the concentration of heavy metals in soil, which is regulated by HN 60 : 2004 "The Maximum Permissible Concentrations of Dangerous Chemicals in Soil" as well as LAND 20-2005 "Requirements for Sewage Sludge Usage for Fertilisation and Remediation". Heavy metals have a long-term negative complex effect on flora and fauna as well as human beings. Various methods such as remediation are used to decrease concentration of heavy metals in soil (Jankaitė, Vasarevičius 2005; Ginneken et al., 2007).

Soil acidity also has a big effect on the quantity of microorganisms (Piaulokaitė-Motuzienė et al., 2004). Soils with the pH indicator lower than 5 nearly do not contain any bacteria introducing atmospheric nitrogen (except for lupinus tuberous bacteria). Acid soils also contain smaller quantities of other bacteria and slower degradation of plant residues, which results in worse nutrition of plants (Piaulokaitė-Motuzienė et al., 2005). If plants do not receive necessary nutrients, they are weak, less resistant to diseases and pests (Aplinkosaugos …, 2008).

Pursuant to LAND 20-2005 "Requirements for Sewage Sludge Usage for Fertilisation and Remediation", the use of all types and categories of sludge in agriculture is prohibited where the soil pH is < 5.5.

To evaluate the suitability of biodegraded organic waste use for soil fertilisation, an experiment aimed at determining the contents of total nitrogen, total phosphorus and heavy metals (Cr, Zn, Pb, Cu and Ni) in this waste and measuring the pH indicator was carried out.

RESEARCH METHODS

The research determines the level of nitrogen and phosphorus, the key elements ameliorating the properties of infertile soil, that of heavy metals in a biologically treated substrate as having negative effect on soil and plants, as well as the pH indicator of the treated substrate. The experiment was carried out under laboratory conditions.

Organic waste used for the experiment consisted of green waste (GW) and sewage sludge (SS) mixed with meat waste (MW) at the ratios of 3 : 1, 1 : 1 and 1 : 3 in volume. Prior to the experiment, a substrate, containing 10% of substrate mass of its total mass, was prepared. Organic waste was digested under psychrophilic conditions (22 ± 2 °C) in anaerobic bioreactors. Organic wastes were mixed twice a day. Afterwards the waste was kept under anaerobic conditions in a bioreactor for 60 days plus one day was allowed for it to settle. The mixture that remained after pouring off the filtrate was mixed up and 1 litre (5 mixed samples 0.2 l each) of the organic waste was taken for the analysis.

The total nitrogen was determined according to LAND 59: 2003 (LST EN ISO 11905-1: 2000) Water Quality. Determination of Nitrogen. Part 1: Method using oxidative digestion with peroxodisulfate (ISO 11905-1: 1997).

The total phosphorus was determined according to LAND 58: 2003 (LST EN ISO 1189-1: 2000) Water Quality. Determination of Phosphorus. Spectrometric method using ammonium molibdate.

The concentrations of the most frequent basic heavy metals whose amounts are regulated by certain documents were determined. The concentrations of heavy metals (Cr, Zn, Pb, Cu and Ni) were determined according to LST ISO 11047 : 2004 (based on the measurements of the element's concentration in an extract of the sample treated with royal vodka using the flame atomic absorption spectrometry).

pH values of the treated organic waste were measured with a pH-meter.

The results of all the experiments were presented as an average value derived from three established values of the measured parameter. Values that differed from the other two values by more than 30% were excluded.

The results were evaluated statistically with the Excel program. Error of measured total nitrogen is ± 0.12 mg/kg, total phosphorus ± 0.1 mg/kg, heavy metals ± 5% and pH ± 0.1. Error of each average value is shown as a range in the Figures.

RESULTS AND DISCUSSION

Nitrogen is one of the main biogenic elements contained in soil necessary for plant growth. Fig. 1 shows the total nitrogen concentration and error of the measured values in the anaerobically digested substrates. The highest content of nitrogen was recorded in biodegraded MW and SS mix (1 : 3), i. e. 4.89 ± 0.12 mg/kg, and the lowest in MW and GW mix (1 : 3), i. e. 3.80 ± 0.12 mg/kg.

Research carried out in Lithuania and other countries shows the dependence of agricultural plant yield and nitrogen fertiliser efficiency on the content of mineral nitrogen in soil (Mattsson, 1990; Lazauskas et al., 1996). Considering this, more nitrogen fertilisers are necessary for plants in soils with a lower nitric content, whereas with the nitric content in soil increasing, the quantity of the fertiliser has to be reduced (Mattsson, 1990; Lazauskas et al., 1996).

It is most expedient to apply biodegraded MW and SS (1 : 3) on infertile soils or land areas under remediation where the biggest amount of nitrogen is required because the highest concentrations of total nitrogen were found in biodegraded MW and SS (1 : 3) mix (Fig. 1).

In areas filled with perennial graminaceous plants there is about 8 mg/kg of mineral nitrogen, in areas with winter wheat and arables – more than 5 mg/kg (Šileika, 2003). It

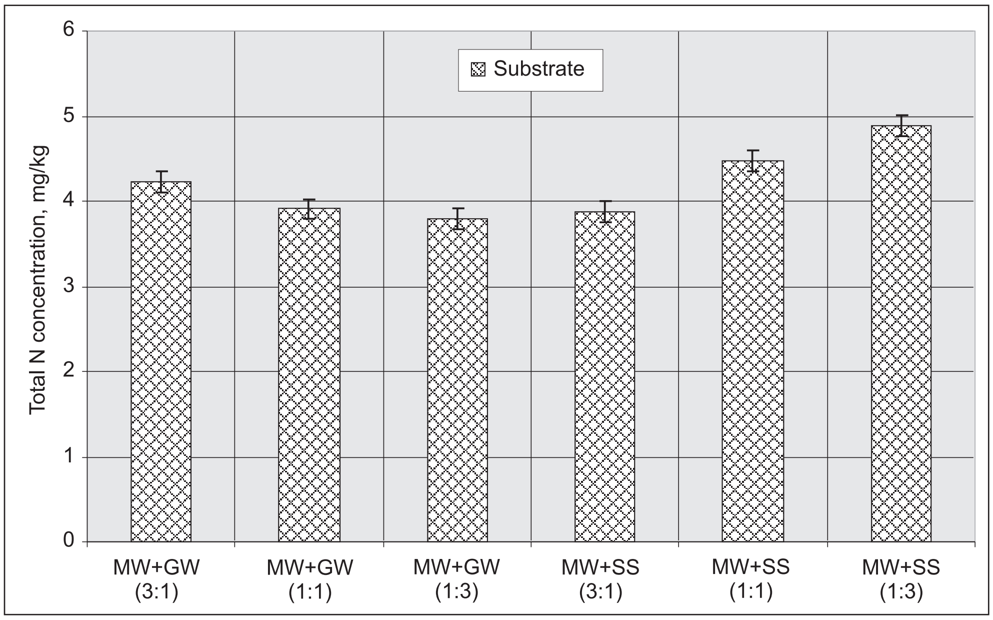

Fig. 1. Total nitrogen concentration in anaerobically digested substrates

is obvious that the researched substrates are not rich in nitrogen, so they could be used exceptionally for arables fertilization where most of mineral materials are migrated or leached. Bučelienė et al. (2003) found that concentrations of total nitrogen varied from 1.52 mg/kg to 2.27 mg/kg in Graisupis watershed.

Upon expressing the total nitrogen concentration in anaerobically digested substrates in percent and equating the maximum concentration in MW and SS (1 : 3) with 100%, the total nitrogen concentration in other substrates was found to be equal to: 91.4% in MW and SS (1 : 1), 79.3% in MW and SS (3 : 1), 86.5% in MW and GW (3 : 1), 80.0% in MW and GW (1 : 1), 77.7% in MW and GW (1 : 3).

Phosphorus is another important biogenic element necessary for plants. The total phosphorus concentration and error of the measured values are shown in Fig. 2. The highest content of phosphorus is present in biodegraded MW and SS (1 : 3) mix, i. e. 227.0 ± 0.1 mg/kg, and the lowest, 61.7 ± 0.1 mg/kg, in MW and GW (1 : 3) mix. Soils with a rich content of phosphorus contain around 200 mg/kg of phosphorus, whereas those with a low content of phosphorus – up to 100 mg/kg (Mažvila et al., 2003). Bučelienė et al. (2003) found that concentrations of phosphorus were rather high and varied from 113 mg/kg to 135 mg/kg in Graisupis watershed. Consequently, it is most expedient to apply biodegraded MW and SS (1 : 3) mix on infertile soils or land areas under remediation where the biggest amount of phosphorus is required (Fig. 2).

A biodegraded MW and GW (1 : 3) substrate with a lower content of phosphorus can be applied on soils where the deficiency of phosphorus is less.

Upon expressing the total phosphorus concentration in anaerobically digested substrates in percent and equating the

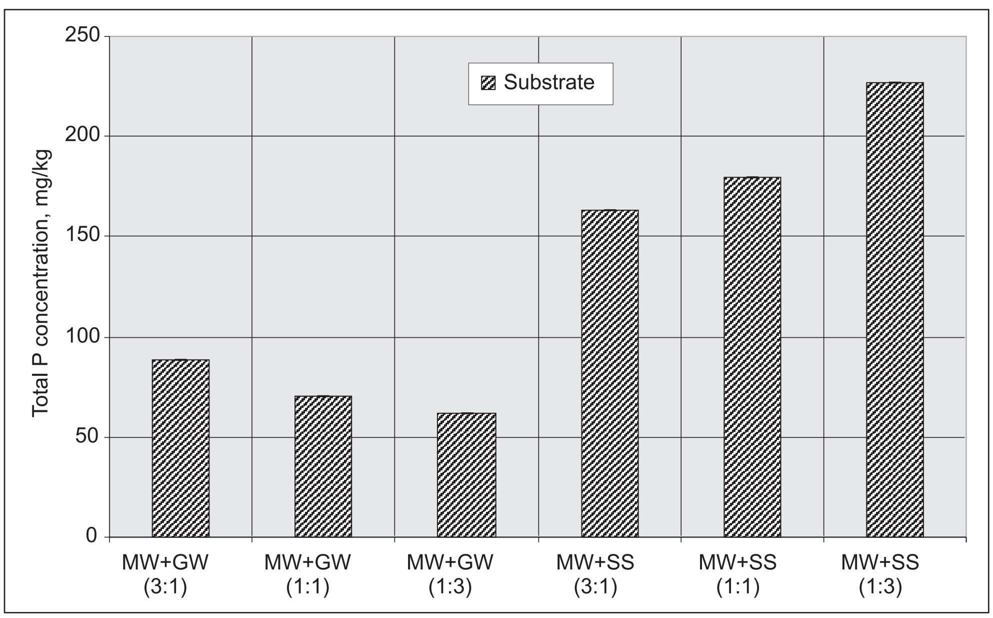

Fig. 2. Total phosphorus concentration in anaerobically digested substrates

maximum concentration in MW and SS (1 : 3) with 100%, the total nitrogen concentration in other substrates is equal to: 79.0% in MW and SS (1 : 1), 71.9% in MW and SS (3 : 1), 38.9% in MW and GW (3 : 1), 31.1% in MW and GW (1 : 1), 27.2% in MW and GW (1 : 3).

Excessive amounts of trace elements do harm to all live microorganisms and therefore the reduction of heavy metal concentrations is very important. The discussion of the obtained experimental findings concerning heavy metal concentrations in an anaerobically treated substrate covers their comparison with the MPC regulated by LAND 20-2005.

The analysis of chromium concentration in the mentioned digested substrates shows that the treated MW and SS (1 : 3) mixture contains the highest concentration of chromium, i. e. 1.05 ± 5% mg/kg of a dry mass, whereas the lowest chromium concentrations are MW and GW mixes, i. e. 0.07 ± 5% mg/kg (3 : 1), 0.08 ± 5% mg/kg (1 : 1) and 0.11 ± 5% mg/kg (1 : 3) of a dry mass, respectively. Background concentration of chromium in Lithuanian sandy and loamy soil is 7 mg/kg and 10 mg/kg respectively (LAND 20-2005) (Fig. 3).

The comparison of the MPC of this metal and the concentrations in substrates produces the following values: 0.14% (sand / sandy loam) and 0.09% (silt loam / loam) in MW and GW (3 : 1), 0.16% (sand / sandy loam) and 0.10% (silt loam / loam) in MW and GW (1 : 1), 0.22% (sand / sandy loam) and 0.14% (silt loam / loam) in MW and GW (1 : 3), 0.84% (sand / sandy loam) and 0.53% (silt loam / loam) in MW and SS (3 : 1), 1.48% (sand / sandy loam) and 0.93% (silt loam / loam) in MW and SS (1 : 1) and 2.10% (sand / sandy loam) and 1.31% (silt loam / loam) in MW and SS (1 : 3).

A higher value of chromium in treated MW and SS mixes is directly dependent on the composition of sewage and gets higher with increasing amount of sewage sludge.

Experiments carried out by Eitminavičiūtė et al. (2007) showed the content of chromium of 106.0 mg/kg in soil that formed after landfills had been remediated with sewage sludge for 9 years.

This is a very high content compared with the results of the experiment covered by this paper, as the difference reaches even 100 times. This might be the result of the improved clean-up quality of sewage in Vilnius in the recent decade. Also concentrations of heavy metals in digested substrates could decrease with leachate of substrate liquid phase. In sewage sludge, concentrations of heavy metals may vary even several times. Oleszczuk (2008) measured concentration of heavy metals in sewage sludge. Concentration of Cr varied from 28.8 mg/kg to 80.0 mg/kg.

The analysis of zinc concentration in the mentioned digested substrates shows that the treated MW and SS (1 : 3) mix contains the highest concentration of zinc, i. e. 15.73 ± 5% mg/kg of a dry mass, whereas the lowest zinc concentrations are found in MW and GW mixtures, i. e. 2.68 ± 5% mg/kg (3 : 1), 3.02 ± 5% mg/kg (1 : 1) and 4.11 ± 5% mg/kg (1 : 3) of a dry mass, respectively. Background concentration of zinc in Lithuanian sandy and loamy soils is 20 mg/kg and 30 mg/kg respectively (LAND 20-2005) (Fig. 4).

Thus, the situation is similar to the previous analysis on chromium concentration – a larger content of zinc was determined in treated sewage sludge.

The comparison of the MPC of zinc and the concentrations in substrates produces the following values: 1.68% (sand / sandy loam) and 1.03% (silt loam / loam) in MW and GW (3 : 1), 1.89% (sand / sandy loam) and 1.16% (silt loam / loam) in MW and GW (1 : 1), 2.57% (sand / sandy loam) and 1.58% (silt loam / loam) in MW and GW (1 : 3), 3.48% (sand / sandy loam) and 2.14% (silt loam / loam) in MW and SS (3 : 1), 6.04% (sand / sandy loam) and 3.72% (silt loam / loam) in MW and SS (1 : 1) and 9.83% (sand / sandy loam) and 6.05% (silt loam / loam) in MW and SS (1 : 3). The highest zinc concentration in sewage sludge is likely to depend on sewage composition.

Previously mentioned experiments carried out by Eitminavičiūtė et al. (2007) also showed a 306.5 mg/kg content of zinc in the forming soil of landfills remediated with sewage sludge for 9 years. Again, the obtained results show

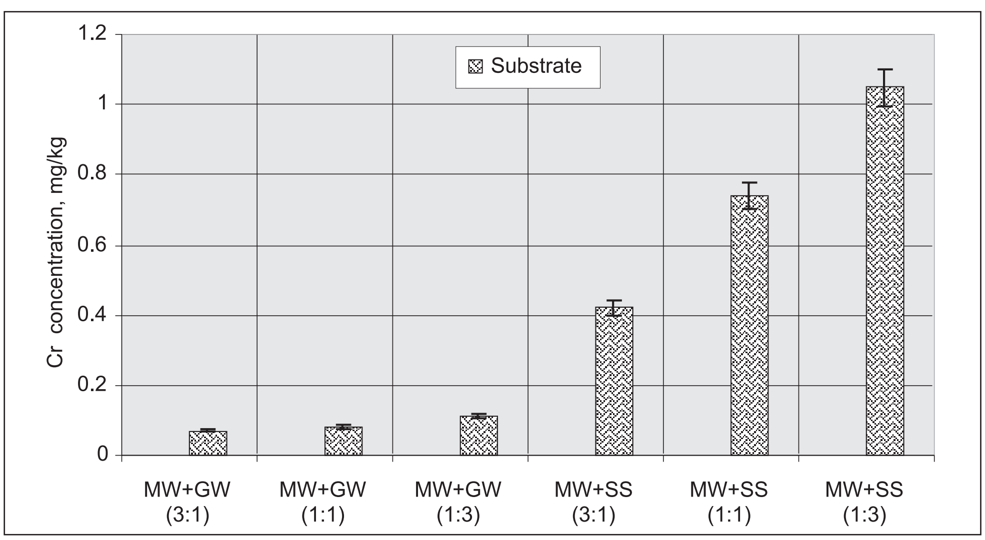

Fig. 3. Chromium concentration in anaerobically digested substrates

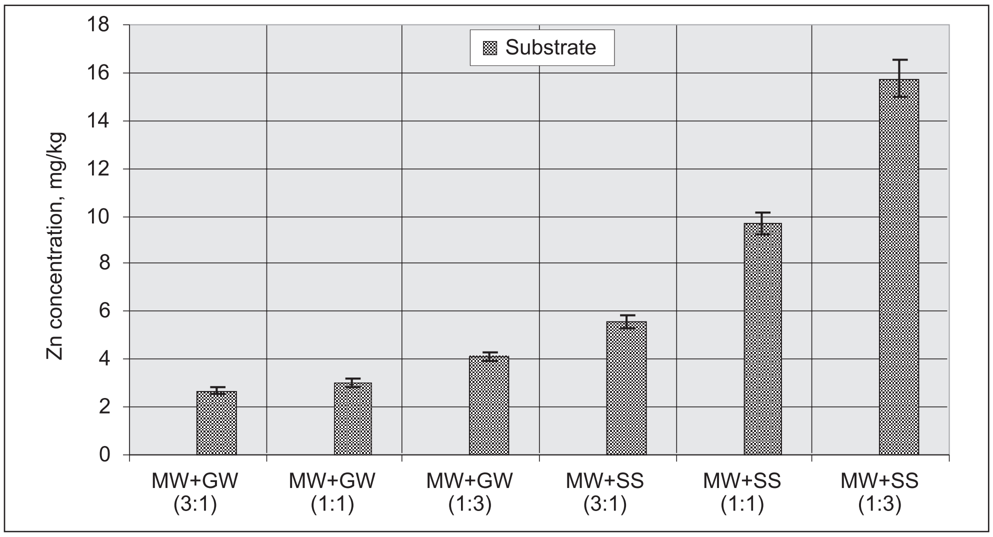

Fig. 4. Zinc concentration in anaerobically digested substrates

a difference of more than 19 times, which most probably was influenced by the changed sewage clean-up quality and leaching of zinc with substrate liquid phase. Oleszczuk (2008) measured concentration of heavy metals in sewage sludge. Concentration of Zn varied from 0 mg/kg to 1680 mg/kg.

Lead is another crucial heavy metal whose content in soil has to be controlled.

The analyses of lead concentration in the mentioned digested substrates as in above described cases shows that the treated MW and SS (1 : 3) mix contains the highest concentration of lead, i. e. 7.74 ± 5% mg/kg of a dry mass, whereas the lowest lead concentrations were present in MW and GW mixtures, i. e. 0.68 ± 5% mg/kg (3 : 1) of dry mass. The difference is 11 times. Such a concentration of lead in mixtures with sewage sludge might have been determined by sewage from industrial facilities. Oleszczuk (2008) measured concentration of heavy metals in sewage sludge. Concentration of Pb varied from 10.6 mg/kg to 28.2 mg/kg. Anyway, this is not a big concentration since a normal lead concentration in soil is around 10 mg/kg (Pichtel et al., 2000). Background concentration of lead in Lithuanian soils is 15 mg/kg (LAND 20-2005) (Fig. 5).

The comparison of the MPC of lead and the concentrations in substrates produces the following values: 1.36% (sand / sandy loam) and 0.85% (silt loam / loam) in MW and GW (3 : 1), 2.18% (sand / sandy loam) and 1.36% (silt loam / loam) in MW and GW (1 : 1), 4.44% (sand / sandy loam) and 2.78% (silt loam / loam) in MW and GW (1 : 3), 5.20% (sand / sandy loam) and 3.25% (silt loam / loam) in MW and SS (3 : 1), 9.98% (sand / sandy loam) and 6.24% (silt loam / loam) in MW and SS (1 : 1) and 15.48% (sand / sandy loam) and 9.68% (silt loam / loam) in MW and SS (1 : 3).

The analysis of copper concentration in the mentioned digested substrates shows that the treated MW and SS (1 : 3) mix contains the highest concentration of copper, i. e. 29.88 ± 5% mg/kg of a dry mass, and the lowest copper concentrations are found in MW and GW mixes, i. e. 5.26 ± 5% mg/kg (3 : 1) of a dry mass, respectively. Background concentration of copper in Lithuanian sandy and loamy soils is 8.1 mg/kg and 11 mg/kg, respectively (LAND 20-2005) (Fig. 6).

The comparison of the MPC of copper and the concentrations in substrates produces the following values: 10.52% (sand / sandy loam) and 6.58% (silt loam / loam) in MW

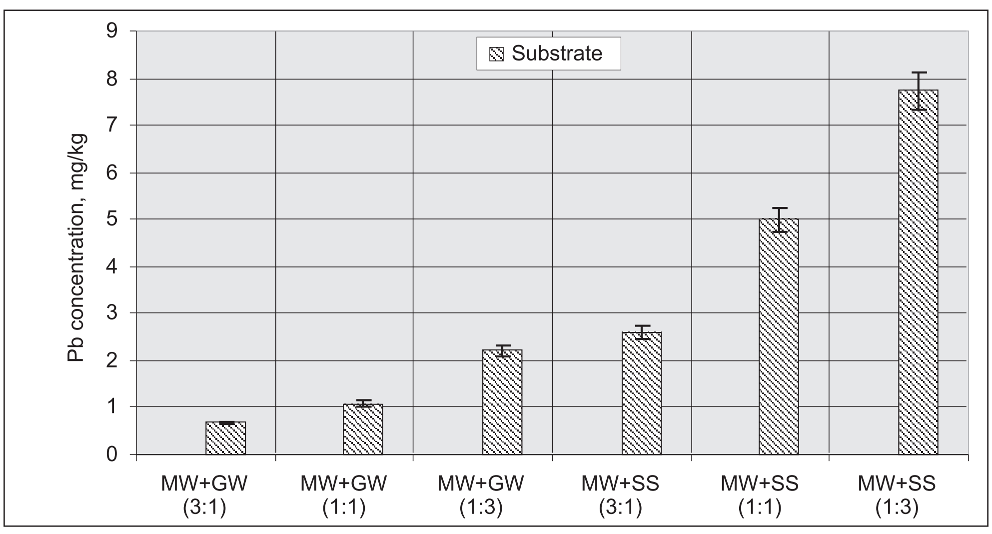

Fig. 5. Lead concentration in anaerobically digested substrates

and GW (3 : 1), 11.96% (sand / sandy loam) and 7.48% (silt loam / loam) in MW and GW (1 : 1), 12.70% (sand / sandy loam) and 7.94% (silt loam / loam) in MW and GW (1 : 3), 20.1% (sand / sandy loam) and 12.56% (silt loam / loam) in MW and SS (3 : 1), 35.48% (sand / sandy loam) and 22.18% (silt loam / loam) in MW and SS (1 : 1) and 59.76% (sand / sandy loam) and 37.35% (silt loam / loam) in MW and SS (1 : 3).

As in previous cases, when amount of sewage sludge is increased, concentration of copper becomes higher proportionally. Higher concentration of the heavy metal mentioned depends on sewage composition. Oleszczuk (2008) measured concentration of heavy metals in sewage sludge. Concentration of Cu varied from 88.2 mg/kg to 161.0 mg/kg.

The analysis of nickel concentration in the mentioned digested substrates shows that the treated MW and SS (1 : 3) mix contains the highest concentration of nickel, i. e. 11.05 ± 5% mg/kg of a dry mass, the lowest nickel concentrations are present in MW and GW mixes, i. e. 2.77 ± 5% mg/kg (3 : 1) of a dry mass, respectively. Background concentration of nickel in Lithuanian sandy and loamy soils is 8.1 mg/kg and 11 mg/kg respectively (LAND 20-2005) (Fig. 7).

The concentration of nickel in soil, like those of other heavy metals, is regulated. The comparison of the MPC of nickel and the concentrations in substrates produces the following values: 5.54% (sand / sandy loam) and 4.62% (silt loam / loam) in MW and GW (3 : 1), 6.06% (sand / sandy loam) and 5.05% (silt loam / loam) in MW and GW (1 : 1), 6.56% (sand / sandy loam) and 5.47% (silt loam / loam) in MW and GW (1 : 3), 6.68% (sand / sandy loam) and 5.57% (silt loam / loam) in MW and SS (3 : 1), 14.32% (sand / sandy loam) and 11.93% (silt loam / loam) in MW and SS (1 : 1) and 22.10% (sand / sandy loam) and 18.42% (silt loam / loam) in MW and SS (1 : 3).

As seen in Fig. 7, when increasing the amount of sewage sludge, concentration of nickel becomes higher proportionally. Oleszczuk (2008) measured concentration of heavy metals in sewage sludge. Concentration of Ni varied from 15.7 mg/kg to 55.5 mg/kg.

Soil quality depends not only on the biogenic elements (nitrogen and phosphorus) analysed in this paper and heavy

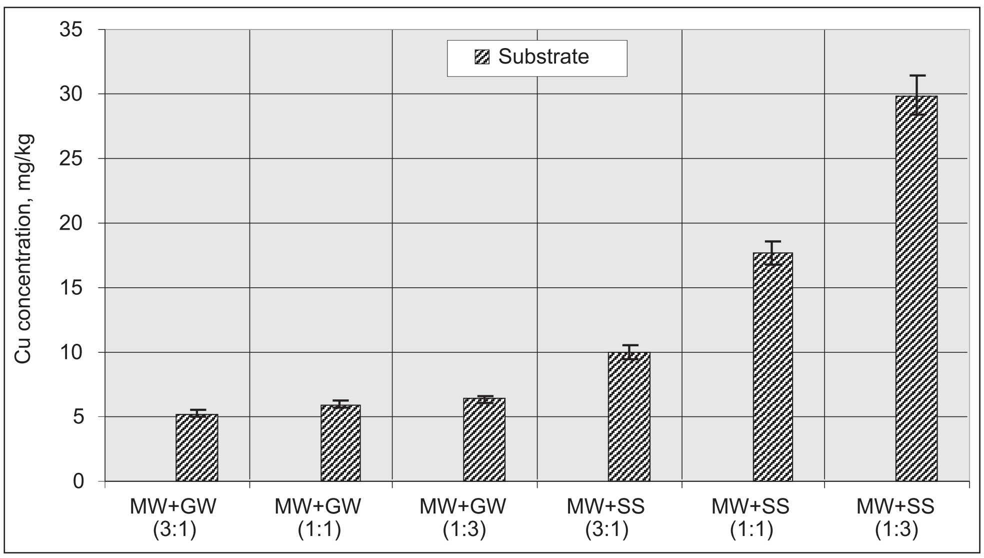

Fig. 6. Copper concentration in anaerobically digested substrates

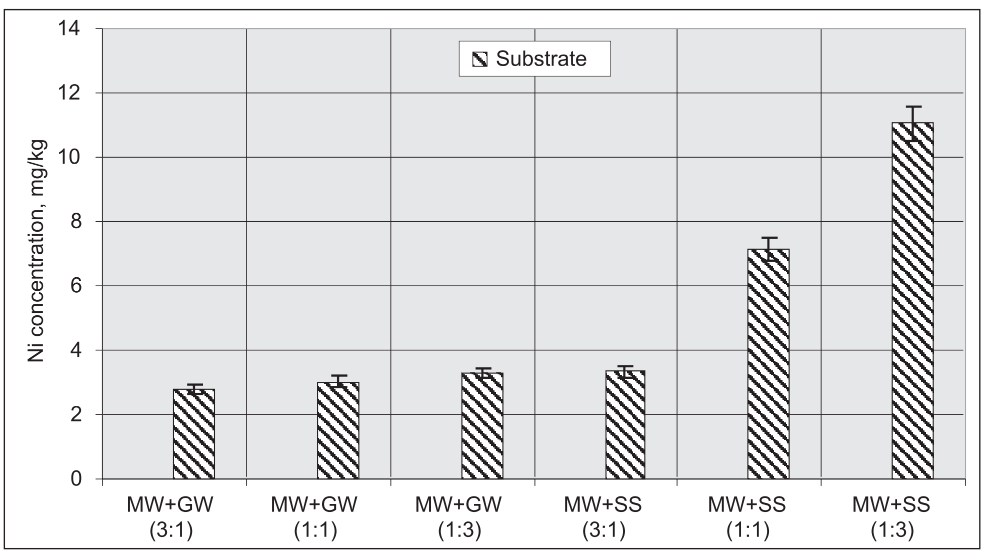

Fig. 7. Nickel concentration in anaerobically digested substrates

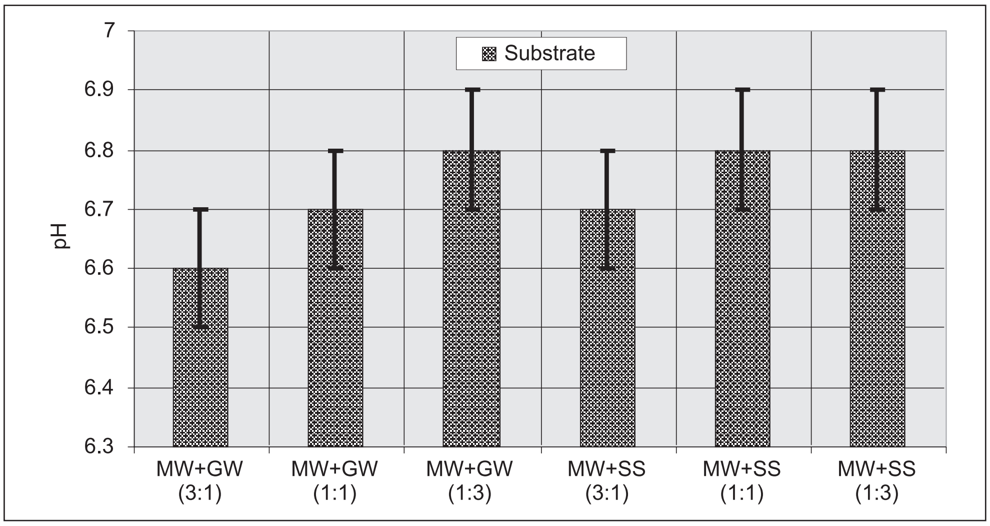

Fig. 8. pH of anaerobically digested substrates

metals concentrations in soil but also on soil acidity. Species of plants depend on the medium of pH, and where pH is low (<5), the process of growth is very complicated and difficult. Soils with pH < 5 must be limed. Mineral fertilizers and manure are more effective in limed soil (Daugėlienė et al., 2005). Acid soils also contain smaller quantities of bacteria introducing atmospheric nitrogen, other bacteria and slower degrade plant residues, which results in worse nutrition of plants (Piaulokaitė-Motuzienė et al., 2005).

The research results show that anaerobically treated MW and GW (1 : 3), MW and SS (1 : 1), MW and SS (1 : 3) have the highest indicator of pH, and MW and GW (3 : 1) – the lowest, i. e. 6.8 ± 0.1 and 6.6 ± 0.1, respectively (Fig. 8).

Regarding pH value, all organic waste researched in this article could be used for acid soil fertilisation. The average pH value of all anaerobically treated organic waste was 6.7. Marschner et al. (2005) found that growth of wheat was the best in neutral soils, lower in the acidic, and poorest in the alkaline soils. Other data showed that increase in soil pH from 4.6 to 6.6 significantly decreased plant dry weight and shoot nutrient content of noninoculated and inoculated seedlings (Aggangan et al., 1996).

CONCLUSIONS

1. The highest contents of nitrogen and phosphorus were determined in anaerobically treated MW and SS (1 : 3) and therefore it is most suitable as an organic fertiliser to remediate exhausted soils and used-up areas. It has been determined that this mix contains 4.89 mg/kg of nitrogen and 227.0 mg/kg of phosphorus.

2. The lowest contents of biogenic elements, nitrogen and phosphorus were determined in anaerobically treated MW and GW (1 : 3) mix, where nitrogen amounts to 3.80 mg/kg and phosphorus to 61.7 mg/kg. Considering the concentrations of nitrogen and phosphorus, this mix is not useful for fertilization.

3. The highest contents of heavy metals were determined in anaerobically treated MW mixtures with SS, where the concentration of chromium amounts to 1.05 mg/kg, that of zinc – 15.73 mg/kg, lead – 7.74 mg/kg, copper – 29.88 mg/kg and nickel – 11.05 mg/kg. Taking into account the concentrations of the determined heavy metals, all researched mixes are suitable for land fertilization.

4. The average pH value of all anaerobically treated organic waste was 6.7. Researched mixes of anaerobically treated waste are most suitable for fertilization of acid soils to make them more alkaline.

5. The evaluation of all the analysed parameters of the suitability of anaerobically treated organic waste for soil fertilisation shows that mixture of MW and SS (1 : 3) treated in a bioreactor is most suitable for soil fertilization.

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ANAEROBIŠKAI PERDIRBTŲ ŽOLINIŲ ATLIEKŲ BEI NUOTEKŲ VALYMO DUMBLO MIŠINIŲ SU MĖSOS ATLIEKOMIS TINKAMUMO DIRVOŽEMIUI TRĘŠTI TYRIMAI

Santrauka

Siekiant įvertinti anaerobiškai bioreaktoriuose suskaidytų organinių atliekų tinkamumą dirvožemiui tręšti, buvo atliktas eksperimentas, kurio tikslas – ištirti bendrojo azoto, bendrojo fosforo, sunkiųjų metalų kiekius šiose atliekose ir nustatyti jų pH. Tyrimams naudotos organinės atliekos: mėsos atliekų (MA) ir žolinių atliekų (ŽA) bei mėsos atliekų (MA) ir nuotekų valymo dumblo (NVD) mišiniai (3 : 1, 1 : 1 ir 1 : 3). Nustatyta, kad tręšimui yra tinkamiausias MA ir NVD (1 : 3) mišinys. Jame nustatyta daugiausia azoto ir fosforo – atitinkamai 4,89 mg/kg ir 227,0 mg/kg. Nors sunkiųjų metalų nustatytos taip pat didžiausios koncentracijos, tačiau dumblu patręštame dirvožemyje jos buvo ne didesnės už didžiausią leistiną koncentraciją. Chromo koncentracija siekė 1,05 mg/kg, cinko – 15,73 mg/kg, švino – 7,74 mg/kg, vario – 29,88 mg/kg ir nikelio – 11,05 mg/kg. MA ir NVD (1 : 3) mišinys tinkamiausias rūgščių dirvožemių tręšimui, nes nustatytas pH 6,8.

Raktažodžiai: tręšimas, dirvožemis, organinės atliekos, bendrasis azotas, bendrasis fosforas, sunkieji metalai (Cr, Zn, Pb, Cu, Ni), pH