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Effects of Soil-Like Materials Mix from Drill Cuttings, Sewage Sludge and Sawdust on the Growth of *Trifolium pratense* L. and Transfer of Heavy Metals

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ABSTRACT

Approximately 80000 tons of drilling waste is produced in Poland annually. This type of waste is relocated and deposited in waste neutralization plants or landfills. Eventually, it must be managed. Reuse of solid wastes might constitute a method of their utilization. The article investigates the use of drill cuttings, sewage sludge and sawdust for production of soil-like mixtures. The studies on evaluation of the possibility of economic application of soil-like materials were carried out. The assessment was conducted on the basis of metal content in soil-like materials and plants cultivated on substrates produced of soil-like materials. In accordance with the valid legal regulations, the concentration of metals in soil-like materials enables their application on class II soils, i.e. arable land. The concentration of heavy metals in the plants cultivated on the substrates produced with soil-like materials is low and does not exceed the concentration of metals determined in animal feeding standards.

Keywords: drill cuttings, wastes, landfarming, recultivation

INTRODUCTION

The total production of wastes in Europe approximates 3 billion tons. Only about 40% of this amount was recycled, and the rest was deposited or combusted [Woodard and Curran 2006]. Reuse is the key element of waste management. The most natural direction for municipal waste and sewage sludge management is composting. The produced compost is used as a fertilizer, improving the soil properties [Boruszko 2011]. Low nutritional value of compost along with low mineralization may result in increased requirement of compost in order to satisfy their nitrogen and phosphorus uptake [Cambardella 2003]. There are studies in which mineral wastes are added in order to increase the amount of mineral compounds in compost [Belyaeva 2010, Zmora-Nahum et al. 2007]. The resultant soil-like materials can be utilized in land application.

In recent years the amount of drilling wastes in Poland has increased due to the exploration of new deposits of conventional and non-conventional hydrocarbons [Macuda 2017]. Part of the mining wastes is stabilized, e.g. by means of cement or calcium hydroxide and finds application in road construction. However, over 70% is deposited in waste neutralization plants [Leonard et al. 2010]. Drilling wastes – cuttings, i.e. the excavated material with spent drilling fluid produced in the process of drilling. They are characterized by a high content of such elements as: calcium, magnesium, potassium, sodium. The pH of drilling wastes exceeds 10, which indicates the possibility of neutralizing acidic soils [Abbe 2009]. Drilling wastes could be introduced to degraded soils. There are studies [Zha et al., Zvomuya et al. 2011, Chaineau 1996] indicating that the addition of drilling wastes improves the sorption properties of soil and the composition of soil complex. However, the Polish legal system, in contrast to the American, prohibits the direct introduction of drilling wastes to soil [Regulation of the Minister of Environment of 20th January 2015 on the recovery process of R10 [Journal of Laws 2015, item 132].

Creation of a soil-like material from drill cuttings, compost and sawdust was proposed in the paper. The physical and chemical properties of these components indicate that the resulting mixture may improve the soil properties. There are concerns that the high content of heavy metals both in compost as well as drilling wastes may infiltrate to ground waters, soils, and plants. The bioavailability of metals for plants is the main method for the assessment of heavy metals toxicity.

The article contains an assessment pertaining to the feasibility of economic application of the soil-like material. The assessment was conducted on the basis of the metal content in the produced soil-like materials and plants cultivated on the substrates.

MATERIALS AND METHODS

The following components were used for the production of soil-like materials:

- compost from the organic municipal waste fraction, produced in a composting plant located on the premises of Wilcze Doły landfill in Kraśnik,
- drill cuttings collected from the layer of waste deposited in a reclaimed object in Luchów Górny, Tarnogród commune,
- sawdust from a sawmill located in Luchów Górny, Tarnogród commune,

The above-mentioned materials were mixed in various weight ratios, yielding four mixture in the proportions presented in Table 1.

Determination of physicochemical properties:

- Electrical conductivity – in the slurry (soil weight : volume 1:2.5) with conductometric titration (by means of a VERSA STAR meter manufactured by ORION);
- pH value by means of potentiometric method in water solution and in 1 M solution of potassium chloride (PN-ISO 10390: 1997P soil quality. Determination of pH),
- dry matter content by means of loss on drying method in the temperature of 105˚C (PN EN ISO 11465:1999P soil quality. Determination of dry matter content of soil and water in soil per dry mass of soil. Gravimetric method),
- total exchangeable bases (S) by Pallmann method in 0.5 mol/dm3 NH4Cl at pH 8.2,
- carbonate content was determined with Scheibler's volumetric method,
- total organic carbon by means of TOC-5050A analyzer Schimadzu (PN-ISO 4335 standard),
- total nitrogen content by Kjeldhal method using 1002 Kjeltech distillation unit
- total heavy metals were determined by ICP-OES Ultrace 238 (Jobin Yvon-Horriba France) using direct calibration method after microwave digestion (Multiwave 3000, Anton Paar). The samples of homogenized soil (1 g) were digested in acid mixture of HNO3: HCl (5:2) and water samples (15 g) were digested in HNO3 (3 mL). The digestion process lasted 45 min at 180˚C and at the pressure of 18 bars. Metal concentrations were determined at different wavelengths (213–395 nm). Detection limits for particular metals did not exceed 10 ppb.

Table 1. Compost, drill cuttings and sawdust treatments used in the experiments.

Soil-like mixtures	Compost	Drill cuttings	Sawdust
	[% weight]		
A12	72.5	2.5	25
A13	45	5	50
A14	40	10	50
A15	60	15	25

Greenhouse experiment: The studies were conducted using clover seeds (*Trifolium pratense L.*). Twelve pots (350 ml each) were prepared and divided into 4 series (each in triplicate):

12 seeds of wheat, originating from the same source, were sown. The plants were watered with distilled water until they reached 75% of total water capacity. The cultivation was carried out in accordance with PN-EN ISO 11269–2:2013–06 standard.

The collected plants were separated into roots and stalks. The biomass of plant roots and shoots obtained in each series was weighed. The cleaned plant material from each pot was dried in 70°C and ground in laboratory ball mill. The content of Ba, Cd, Cr, Cu, Ni, Pb, Zn was determined in the plant material, following its mineralization in nitrogen and hydrochloric acid (5:2) performed by means of Microwave 3000 by Anton Paar. The elements were indicated using mass spectrometry method with inductively coupled plasma by means of ICP-OES JY 238 Ultrace (Jobin Yvon-Horriba, France). On the basis of the obtained metal contents in plant biomass, bioaccumulation factor was calculated using the formula:

$$BCF = C_P/C_G \quad (1)$$

where: C_P – concentration of metals in the surface/underground parts of the plant, mg/kg;
C_G – concentration of metal in soil at the beginning of the process, mg/kg.

Mean values with standard deviation were calculated for the results of this study. Then, the statistical analyses based on the one-way analysis of variance (ANOVA) and multiple T-Tukey tests with the significance level α = 0.05 were carried out. T-Tukey's multiple comparative tests provided a detailed comparative analysis of mean values by isolating statistically homogeneous groups (homogeneous groups). The matrix of correlation coefficients between the variables was determined.

RESULTS AND DISCUSSION

There are studies which confirm the positive influence of drilling wastes on lightweight soils [Zha et al. 2017]. The research conducted by Kominko et al. on short- and mid-term results of various organic fertilizers such as: sewage sludge, municipal wastes and manure on the physical properties of soil showed that composting of solid wastes and introducing sewage sludge to soil improves its stability [Kominko et al. 2017].

Table 2 presents the selected physicochemical properties of soil-like mixtures. The mixtures are characterized by neutral pH. The highest value of pH – over 6.9 – was obtained by A12 and A15 mixtures, which is a statistically significant difference from the pH of A13 and A14 mixtures. This pH is optimal for the growth of majority of arable crops.

The content of carbonates increases along with the content of drill cuttings in mixtures. Drill cuttings are characterized by high carbonate content. The studies carried out by Gonet et al. also indicated that the addition of drilling wastes greatly increased the content of carbonates in the soil with drilling waste addition [Gonet et al., 2006] The contents of carbonates in mixtures are statistically significantly different.

A similar situation occurs with the value of total exchangeable bases. Total exchangeable bases in soil-like materials are statistically significantly different. The research conducted by Gonet indicates that the addition of drilling wastes greatly changes the total exchangeable bases with the addition of drilling wastes [Gonet et al. 2006].

The value of nitrogen in soil-like materials amounted to about 1%. The addition of drilling fluid reduced the nitrogen content in the produced mixtures. The content of nitrogen in the compost approximates 1.5% [Belayaeva 2011]. Drill cuttings contain a very small amount of nitrogen [Zvomuya et al. 2011]. The contents of nitrogen in A12 and A13 mixtures are statistically significantly different from A14 and A15. The concentration of nitrogen in Polish soils ranges from 0.1 to 0.3% [Kabata-Pendias et al. 2015]. The produced materials are characterized by the content of about 1%.

The mixtures contain roughly 20% of carbon. The content of carbon in mixtures is not statistically significantly different.

The C:N ratio in materials reaches about 20. The optimal value of C:N for plant growth is assumed as 23 (Ozimek A. et al. 2012). The C:N ratio greater than 30 indicates low activity of biological processes and accumulation of undecomposed organic substance (Mroczkowski et al. 2016).

The organic matter content exceeds 30%; this value is 5 times greater than the maximum value of organic matter found in Polish soils [Report OSCR 2012]. On the basis of organic matter content (Table 2), in line with PN-EN ISO 14688–2 standard, the mixtures may be considered as highly organic soils (>30%).

Organic matter plays an important role in maintaining the soil quality. It improves the structure of soil, may increase the infiltration rate and reduce the soil erosion [Rattan 2016]. Such high

Table 2. Physicochemical properties of artificial soil

Artificial soil	pH	OM [%]	$CaCO_3$ [%]	N [%]	C [%]	S [mmol]
A12	6.94[a]	33.97[a]	1.16[a]	1.13[a]	19.75[a]	30[a]
A13	6.38[b]	35.69[a]	1.63[b]	1.08[a]	20.75[a]	32.03[b]
A14	6.46[c]	34.04[a]	1.78[c]	0.77[b]	19.79[a]	33.37[c]
A15	6.98[a]	33.2[a]	2.21[d]	0.77[b]	19.30[a]	34.67[d]

The superscript [a] next to the average content values in the Table (Tukey Homogeneous Groups) indicates statistically homogeneous groups. The presence of the indicator designates the lack of statistically significant difference between them.

Table 3. The concentration of heavy metals in artificial soils (mg/kg).

Artificial soil [mg/kg]	Ba	Cd	Cr	Cu	Ni	Pb	Zn
A12	304.20[c]	0.12[a]	3.29[a]	0.59[a]	0,59[a]	7,17[a]	34,05[b]
A13	325.39[a]	0.11[a]	4.39[b]	1.44[b]	1,44[b]	7,55[a]	47,99[a]
A14	340.36[a]	0.23[b]	6.73[d]	3.72[c]	2,48[d]	7,51[a]	89,98[d]
A15	382.81[b]	0.28[b]	7.53[c]	7.43[d]	3,72[c]	8,56[b]	99,75[c]
	400	*2*	*200*	*200*	*150*	*200*	*500*

The superscript [a] next to the average content values in the Table (Tukey Homogeneous Groups) indicates statistically homogeneous groups. The presence of the indicator designates the lack of statistically significant difference between them.

organic matter content may limit the availability of heavy metals in the produced soil-like materials.

The concentration of heavy metals in soil-like materials was presented in Table 4. Along with the increase in the percentage content of drill cuttings, the concentration of heavy metals increases as well. However, the concentrations of heavy metals in all mixtures are lower than the ones stated in the Regulation of the Minister of Environment of 1st September 2016 on the conduct of the assessment of contamination of the surface of the earth (Journal of Laws, 2016, item 1395). Therefore, the threat resulting from the application of soil-like materials on the environmental pollution may be controlled by creating waste mixtures in appropriate ratios. The content of metals enables the application of waste components for class II soils, i.e. agricultural soils.

The concentration of barium in the created soil-like materials is approximately 3 times greater than the maximum concentration of barium in Polish soils, reaching approximately 100 mg/kg [Report OSCR 2012]. The highest concentration of barium, reaching 382.81 mgBa/kg was obtained for A15 mixture, which is statistically significantly different from the concentration of barium in other soil-like materials. Despite such high concentration of barium, its value does not exceed the permissible levels stated in the Polish law; moreover, barium is hardly soluble in water, does not infiltrate to ground waters and is not taken up by plants [Kabata-Pendias et al. 2001]. The created mixtures are characterized by lower concentration of cadmium than the average cadmium concentration in Polish soils, amounting to 0.59 mgCd/kg [Czech T. 2012]. In relation to A12 and A13, the content of cadmium in A14 and A15 mixtures is twice as high, which is statistically significant.

The highest concentration of lead, i.e. 8.56 mgPb/kg was achieved with A15 soil-like material. This is statistically significantly different from the concentration of lead in other soil-like materials, but lower than the average content of lead in Polish soils, amounting to about 23.3 mgPb/kg [Czech T. 2012].

The average concentrations of chromium in Polish soils approximate 11.3 mgCr/kg, copper 9.9mgCu/kg, nickel 9.4 mgNi/kg, zinc 78.8 mgZn/kg [Report OSCR 2012]. The concentrations of these elements in the produced mixtures are lower. The concentrations of chromium, copper, nickel, nickel, zinc in particular soil-like materials are statistically significantly different.

The biomass of clover cultivated on soil-like mixtures is presented in Figure 1. Along with the increase of drill cuttings content in soil-like materials, the biomass is reduced. The same results were obtained by Gonet [Gonet et al. 2006]. The decrease in biomass results from the increased

Table 4. The concentration of heavy metals in *Trifolium pratense L.* (mg/kg).

Artificial soil [mg/kg]	Ba	Cd	Cr	Cu	Ni	Pb	Zn
A 12 shoot	48.54[a]	0.15[c]	1.42[a]	8.63[a]	2.23[a]	0.82[a]	403.02[a]
A13 shoot	49.76[a]	0.14[c]	2.07[a]	8.87[a]	2.77[a,c]	1.01[a]	405.44[a]
A 14 shoot	46.73[a]	0.21[a]	2.95[b]	8.47[a]	2.59[c]	1.85[a]	445.39[a]
A 15 shoot	48.33[a]	0.34[a]	5.11[c]	8.28[a]	3.64[d]	1.93[a]	489.54[a]
		0.5	20	30	10	10	100

The superscript [a] next to the average content values in the Table (Tukey Homogeneous Groups) indicates statistically homogeneous groups. The presence of the indicator designates the lack of statistically significant difference between them.

content of mineral salts. The highest biomass was found in the plants cultivated on A12 mixture. The biomass values obtained on particular substrates do not differ in a statistically significant way.

The content of heavy metals accumulated in clover was investigated, because it has a major influence on the possibility of assessing the feasibility of utilizing the produced soil-like materials for agricultural purposes. There are works in which a significant increase of metal content was observed in the plants cultivated on compost [Belyaeva et al., 2009, Zmora-Nahum et al. 2007] or drill cuttings with the addition of soil [Gonet 2006, Zvomuya 2011]. The content of metals in the plants cultivated on soil-like materials is low and does not exceed the permissible concentrations of heavy metals in feed, determined by Kabata-Pendias et al. and the Regulation of the Minister of Agriculture and Rural Development of 23rd January 2007 on the permissible concentrations of undesirable substances in feed [Kabata-Pendias et al. 2001].

The reference values of metals in plants equal: 0.05 mgCd·kg^{-1}, 1.5 mgCr·kg^{-1}, 10 mgCu·kg^{-1}, 1.5 mgNi·kg^{-1}, 1 mgPb·kg^{-1}, 50 mgZn·kg^{-1}, respectively [Market 1992]. The concentrations of all the considered metals in the plants cultivated on soil-like materials are higher than the reference values.

The concentration of chromium in the plants cultivated on A12 soil-like material is approximately three times greater than in the plants cultivated on A12 soil-like material, which is statistically significant. The concentration of cadmium in the plants cultivated on A15 substrate is twice as high as the one of the plants frown on A12 substrate, which too is significant. The concentrations of barium, copper, lead and zinc in the plants cultivated on particular substrates are not statistically significantly different.

Metals from substrates may be transported to plants. The heavy metal accumulation capacity of plants can be expressed by means of the bioconcentration factor (BCF) which was presented in Table 5. The BCF values were below 1, except for Ni, Cu, and Zn. The BFC for Ni, Cu, and Zn in the surface part, in all mixtures, exceeded 1, which indicates that clover easily accumulates these compounds.

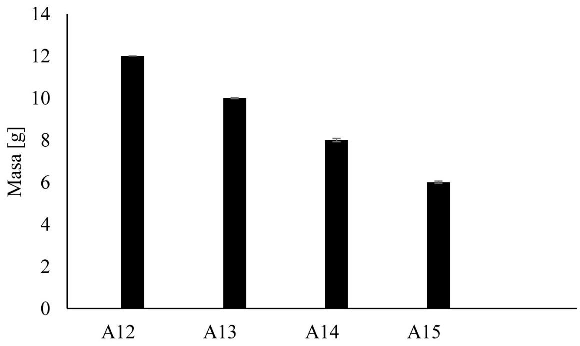

Fig 1. Biomass of *Trifolium pratense L.* cultivated on soil-like materials.

The results presented in table 5 depict this discrepancy in the BFC values and heavy metal absorption in plants, which can be related to the changes in the application doses of drill cuttings and composts.

CONCLUSIONS

1. The proposal of producing soil-like mixtures from such materials as compost, sawdust, and drill cuttings may become an alternative method of their management.
2. The soil-like mixtures are highly organic materials. pH and ratio C/N the soil-like mixtures are optimal for the growth of majority of arable crops.
3. The presented results indicate that the content of heavy metals in the produced soil-like materials is characterized by limited bio-availability for the environment. However, the long-term impact of soil-like mixtures was not studied.
4. The biomass of clover cultivated on soil-like mixtures is reduced along with the increase of drill cuttings content in soil-like materials. The plants cultivated on soil-like materials can be used as feed. The clover easily accumulates Cu, Ni, Zn.

Table 5 . BCFs of heavy metals in *Trifolium pratense L.*

Mixture	BCF						
	Ba	Cd	Cr	Cu	Ni	Pb	Zn
A12	0.11	0.15	0.48	1.21	1.27	0.29	1.10
A13	0.10	0.13	0.41	1.61	1.81	0.59	1.11
A14	0.15	0.21	0.39	1.44	1.43	0.27	1.20
A15	0.15	0.34	0.75	1.89	1.98	0.40	1.20

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