

Biological Reclamation Using a Sewage Sludge-Based Substrate – A Way to Ensure Sustainable Development of Urban Areas

Ivan Tymchuk^{1*}, Myroslav Malovanyy¹, Oleh Bota², Oksana Shkvirko¹, Olena Popovych¹

¹ Viacheslav Chornovil Institute of Sustainable Development, Lviv Polytechnic National University, S. Bandera Str. 12, Lviv, 79013, Ukraine

² Center LTD Consulting 1a/12, Lazarenka Str., Lviv, 79026, Ukraine

* Corresponding author's e-mail: i.s.tymchuk@gmail.com

ABSTRACT

The monitoring studies were carried out on the territory of the Lviv wastewater treatment plants. The volumes of accumulated stocks of the sludge site were determined, its qualitative and quantitative indicators were identified, and the presence of limiting factors was established. On the basis of the obtained data, the 2-D models characterizing the vertical and horizontal accumulation of elements were created. The mioindication studies were conducted to determine the feasibility of using sewage sludge as a growth substrate. Modeling was performed to determine the potential area of reclaimed land.

Keywords: sewage sludge, reclamation, Lviv wastewater treatment plants, substrate.

INTRODUCTION

The rapid growth of cities and the ever-growing needs of society for minerals, energy resources, building materials, as well as the irrational use of natural resources have led to significant changes in the landscape, especially natural vegetation. Large areas of land are directly affected by industrial development, landfills, as a result of which the relief of the Earth's surface changes, vegetation and soil cover is destroyed.

The main purpose of reclamation is to use biogeocenosis to transform unsuitable areas into productive areas for urban construction, agriculture, forestry and recreation. The choice of the direction of reclamation of disturbed lands is based on the rational and efficient use of land resources, the creation of landscapes that would meet the economic, environmental and sanitary needs of society. To date, most attention has been paid to the agricultural direction of land reclamation. However, such reclamation works require significant financial and natural resources.

At the same time, there are many problems associated with increasing consumption of natural resources. As a result of man-made human activities, a large amount of hazardous substances enters the environment and a significant amount of waste is generated. In most cases, waste is not reusable and therefore pollutes the environment, but there are those that can be used as secondary raw materials. For example, a lot of attention is drawn to the wastes that contain an organic component, namely: calcium-containing sludge from thermal power plants, wastes from biogas production (spent biomass), algae, wastes from coal mining, sewage sludge (SS), etc.

In recent years, in many countries around the world used the waste generated in the process of biological wastewater treatment, namely sludge as a secondary raw material. These wastes are rich in nutrients such as nitrogen, phosphorus, potassium, so there is a possibility of their use in agriculture for the manufacturing of organo-mineral fertilizers. This method of using sewage sludge is popular in the United Kingdom, Spain and Portugal (Kacprzak et al., 2017, Mininni et al., 2014, Inglezakis et al.,

2014, Fijalkowski et al., 2017). There are also other ways to dispose of sewage sludge. For example, in the United States, the anaerobic conversion of sewage sludge produces electrical and mechanical energy, and the digestion of wet sludge produces biological oil that can be fractionated into other fuels. In Japan, using various technologies (PRISA, Seaborne, AirPrex, etc.), phosphorus and nitrogen are extracted from sludge, which is then used to make nitrogen and phosphorus fertilizers, and sewage sludge is used to make bricks, cement and other building materials (Blöcher et al., 2020, Kalogo, Y., & Monteith, 2012, Zhou et al., 2020).

In Ukraine, unlike other countries, the situation with sewage sludge is quite threatening, as most sludge is stored in silt fields and pollutes the environment. Currently, only 3% of sewage sludge is used in Ukraine and mainly in agriculture. Therefore, it is important to find optimal and economically sound ways to dispose of sewage sludge in Ukraine.

Accumulation of a large amount of sewage sludge on the open areas of the Lviv wastewater treatment plants creates a significant burden on the environment, which is manifested in the coverage of large areas of fertile land, air pollution with toxic gases, the possibility of leachate into adjacent watercourses and groundwater. Insufficiently treated wastewater enters the surface water and causes eutrophication (Nykyforov et al., 2016).

Large volumes of sewage sludge generated and accumulated over decades at Ukrainian WWTPs pose a serious threat to the environment. Starting from 2016, the storage of sewage sludge has been prohibited in the EU countries, sludge sites can no longer be used for dewatering of sewage sludge (Bień & Bień, 2015). The authors of this paper propose combining the problem of reclamation of man-made disturbed lands with the utilization of organ-containing waste. In this way, several problems can be solved at once.

Thus, the conducted research was aimed at determining the accumulated reserves of sewage sludge on sludge sites, the study of their qualitative and quantitative indicators, as well as the possibility of their environmentally safe disposal.

MATERIALS AND METHODS

The object of conducted study was the silt fields of the Lviv municipal enterprise “Lvivvodokanal”. The sludge fields of the treatment

plants at the time of sampling were finely compacted, in some places significantly water-saturated sediments.

In order to obtain topographic plans, an electronic total station SOKKIA was used, which allows registering numerical and textual information and perform coding of terrain objects in the field, as well as graph plotters, which after appropriate processing of materials using software-technological complexes such as “Topograd” and “DIGITALS”, enable to automatically obtain topographic plans in digital and graphical form. The distances measured at the station to the picket points were registered in the electronic total station data terminal. At the same time, the outline was crossed out during each survey. The outlines were decorated with symbols (with explanatory inscriptions), approximately following the scale of the survey, on separate sheets for each station, oriented along the course and showing the direction of orientation. The outlines showed the situation of the area and the boundaries of the land plot. Ashtech Solution was used to calculate the vectors between the points. The evaluation of the quality of measurements was performed according to the absolute root mean square errors of location determination, the accuracy of which is in the range of 0.05–0.20 m. In-house processing of field data was performed on a personal computer using the “DIGITALS”, “INVENTGRAD” and “MAPINFO” software. The site is located in the north-eastern suburbs of Lviv, 1200 m south of the village Murovane. The river Poltva flows 200 m north of the site. The work area is located within the northern, northeastern industrial zone. The northeastern industrial area is adjacent to the northern industrial zone and the northern industrial hub. Urban wastewater treatment plants, bases, warehouses, and car companies are located here (Fig. 1).

Sampling of sludge was carried out using a metal cylindrical tube with a bottom valve. The diameter of the sampler was 128 mm with length of 1 m. A pipe was screwed to the top of the sampler, the length of which was increased by means of couplings. The thickness of the sludge was 3 meters or more. The samples were taken in the range of depths 0–0.2 m, 1.4–1.6 m, 2.8–3 m. Such intervals were chosen to study the ecological, biogeochemical and parasitological characteristics of silt in the near-surface, medium-depth and bottom conditions of their stay. The volume of the sample was 2.5 dm³. The selected mule samples were packed in double



Figure 1. Location of the investigated silt site

plastic bags. Each sample was assigned a serial number. The sample numbers were recorded in the sampling log. The coordinates of the sampling points were measured and identified on the ground using geodetic instruments.

As the State Standards of Ukraine on the methodology for determining the composition of sludge from wastewater treatment plants have not yet been developed, the research was conducted in accordance with the current regulations on soil analysis.

The organic matter content was determined gravimetrically after dry combustion of the sample in accordance with DSTU 4289: 2004.

The gross Cu content was determined by the atomic absorption method after acid decomposition of the samples in the presence of hydrogen peroxide according to MVV № 081/12-0002-01.

Determination of the mobile forms of Cu was performed with ammonium acetate buffer solution with a pH of 4.8 at a soil to solution ratio of 1: 5, followed by analysis of the obtained extracts by atomic absorption method in accordance with DSTU 4770.1:2007 – 4770.9:2007.

The bacteriological and parasitological studies of sludge were also performed, in the Lviv Regional Laboratory Center of the Ministry of Health of Ukraine by using the titration and microscopic methods, respectively.

Determination of the quality of the growth substrate was performed with the generally accepted method (DSTU ISO 11269-1: 2004, DSTU ISO 11269-2:2002, 2004). In this method, plants are preferred to establish the possibility of using a growth substrate because they characterize the state of the environment in which they grow, multiply

rapidly, react differently to harmful factors and thus allow choosing the most appropriate response for a particular study. The use of such a method makes it possible to determine the compatible biological activity of the influence of physicochemical factors on the natural environment. The method is suitable for all types of soils, soil-forming materials, precipitated waste or chemicals that may be introduced into the soil. According to the method, the growth substrates were studied mixture and control soil, which is known to be of good quality.

Two types of plants belonging to Category 1 (according to DSTU ISO 11269-2: 2002) were selected for research, i.e. monocotyledonous plants: rye, ryegrass, rice, oats, wheat, barley, sorghum, corn. Before using the seeds, each culture was analyzed to determine their germination and germination energy. During the experiment, 10 identical seeds of the selected species were planted in each of the vessels. For each replicate in each embodiment, the percentage of seed germination relative to the average germination in the control vessels was calculated. The length of the longest roots of each plant was measured and the average length of the longest root for each investigated growth substrate was determined. Statistical analysis was used to determine the smallest significant discrepancies between controls and test concentrations.

For the study, in accordance with this method, different types of substrate based on soil, sewage sludge and natural sorbents were used and compared according to the control soil in order to determine the quality of the substrate from the above-mentioned plants used: barley (*Hordeum vulgare*) and ryegrass (*Lolium perenne*).

RESULTS AND DISCUSSION

The topographic and geodetic works allowed building a map of the silt field with a clear reference to local coordinates. However, these studies allowed calculating quite accurately the area of the silt field, which is 12.9006 ha, i.e. 129 006 m². On the basis of the results of topographic and geodetic surveying, a map of the silt field surface morphology and its three-dimensional model were constructed (Tymchuk et al., 2020), which clearly show a decrease in the absolute marks of the silt surface in the eastern, north-eastern directions from almost 247 m to a little more than 244 m. The elevation between the highest and lowest points reaches about 3 m.

On the basis of the results of the obtained data, the volume of sludge sediments was calculated. The calculation was carried out by multiplying the area of the silt field by the sludge power calculated from the power isolines. Power was calculated on a grid with a step of 25 m, the number of points was 173:

$$V = S \cdot \left(\frac{h_1 + \dots + h_{173}}{173} \right) \quad (1)$$

The calculated average power is 3.16 m. The total area of the silt field, according to topographic and geodetic mapping is 129 006 m². The total volume of silt sediments of the entire silt field is 407 659 m³.

A three-dimensional model of the silt field bottom morphology was built based on the synthesis of the map of the silt field surface morphology and sludge capacities in the wells (Tymchuk et al., 2020). The difference between the absolute highest and lowest points is slightly greater than 1.5 m. The general morphology of the bottom decreases in the eastern direction.

After calculating the available sludge reserves, a qualitative analysis was performed. In particular, the content of organic and mineral components was determined. The content of organic matter in the sewage sludge is in the range of 28–44 wt.%. In most wells there is a clear pattern of reducing the amount of organic matter with the depth of selection, this may be due to the activity of reducers.

The total mineral content in the sewage sludge of the studied sludge field is 46–60 wt.% (Fig. 2). In most wells, the number of mineral increases with depth. However, here such a clear correlation, as in organic matter, with depth is not observed.

In the near-surface depth range (0.0–0.2 m) the mineral content is 46–55 wt.%. Its minimum

values are characteristic of the central part of the silt field, decreasing isomorphically to the peripheral parts (Fig. 2a).

In the range of 1.4–1.6 m the amount of mineral matter is in the range of 49–58 wt.%. Moreover, the maximum values are characteristic of its central part (Fig. 2b).

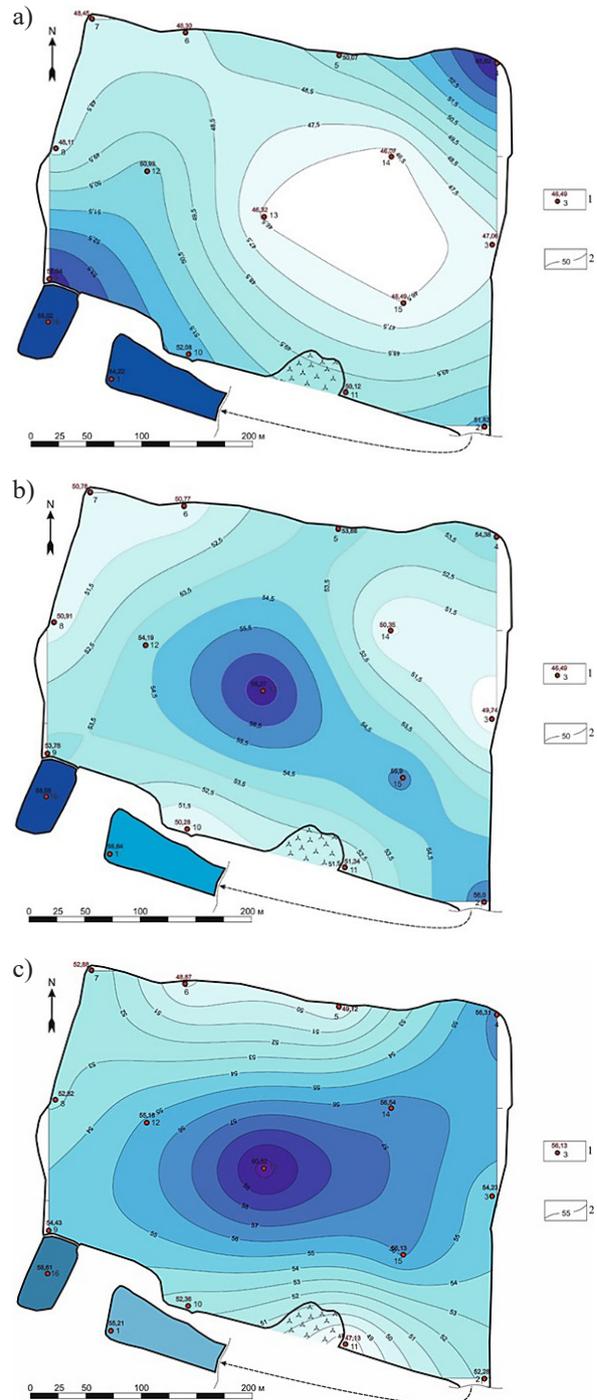


Figure 2. Map of mineral component content in the depth range a) 0.0–0.2, b) 1.4–1.6, c) 2.8–3.0 m: 1 – well number and mineral component content, mg/kg; 2 – isolines of the same content of mineral component

In the bottom range (2.8–3.0 g) the mineral content is 47–60 wt.%. Laterally, the morphology of the distribution of mineral matter (Fig. 2c) is similar to the middle interval of the silt field.

The amount of mineral matter increases along with the depth of the sewage sludge. The reason for this is the gravitational differentiation of the silt substrate, which is manifested in the deposition of heavier mineral components. An additional factor is their leaching from the silt by meteorological waters, followed by movement down the section of the silt field.

In order to determine the disposal methods, one of the mandatory evaluation parameters is to establish the content of heavy metals in the sludge on the sludge site. However, it should be noted that the storage of sludge in the silt field did not take place in chronological order, so it is extremely difficult to trace the dependence of changes in heavy metals on their historical features (over time in Lviv decreased the number of enterprises with hazardous wastewater discharges. Heavy metals in the soil can be in various forms of solubility and mobility, namely: insoluble, which are part of soil minerals; exchange, which are in dynamic equilibrium with the ions

of this metal in the soil solution; movable and soluble forms. There is not only a close relationship between them, but also the possible transformation of some forms into others.

The results of analytical studies for the determination of copper (Cu) in the depth range 0 – 0.2; 1.4 – 1.6; 2.8 – 3.0 m on the silt site are presented in Table 1.

Mobile forms of metals can accumulate in the soil to high concentrations, which cause their toxicity to both soil biota (Sehin et al., 2020) and plants. It is believed that mobile forms of metals in soils are the most susceptible to accumulation by plants. On the basis of these features, analytical determinations of the metal content in sewage sludge in both exchange and gross forms were made.

The content of mobile forms of Cu ranges from 1.36 to 5.40 mg/kg of air-dry sample of sludge. As currently no limiting values of mobile forms of heavy metals for sewage sludge for use as fertilizers have been established, a comparative analysis of the content of metals in the sewage sludge from Lviv wastewater treatment plants was conducted relative to the maximum allowable concentrations (MPC) of mobile forms of heavy metals in agricultural

Table 1. The content of mobile and gross forms of Cu in sludge (for air-dry sample)

Sample number	Cu mobile form, mg/kg			Cu gross content, mg/kg		
	0–0.2 m	1.4–1.6 m	2.8–3.0 m	0–0.2 m	1.4–1.6 m	2.8–3.0 m
Well №1	3.14	2.78	2.05	220.6	231.0	225.4
Well №2.	2.10	4.10	1.37	236.7	233.3	226.7
Well №3.	2.35	3.10	2.65	270.0	220.0	220.0
Well №4.	2.64	4.30	4.50	220.0	235.7	225.3
Well №5.	2.15	1.60	1.95	260.0	240.5	252.5
Well №6.	2.30	1.60	2.90	240.0	286.7	243.3
Well №7.	5.40	2.80	4.20	273.3	260.0	263.3
Well №8.	2.20	2.70	5.00	262.5	270.0	265.0
Well №9.	1.95	2.60	3.40	220.0	232.3	222.6
Well №10.	2.45	2.80	4.00	250.0	260.0	265.0
Well №11.	2.20	1.64	1.70	250.0	243.3	220.0
Well №12.	3.22	3.58	1.36	265.3	254.6	240.5
Well №13.	4.59	3.64	2.03	262.4	251.5	208.4
Well №14.	2.57	2.6	1.55	255.8	238.2	219.5
Well №15.	4.68	3.55	3.21	286.2	245.8	216.8
Well №16.	3.65	2.88	2.39	214.1	230.5	215.5
MPC for soil (mobile form of metal) (Yatsuk & Baliuk, 2013)	3.0	3.0	3.0	–	–	–
Limited value (Council Directive 86 / 278 / EEC)	–	–	–	1000–1750	1000–1750	1000–1750
DSTU ISO 7359:2013. 2014	–	–	–	100–300	100–300	100–300

soils (Yatsuk & Baliuk, 2013) and MPC was – 3.0 mg/kg. The average data showed from different horizons showed that at a depth of 0–0.2 m the average concentration is 2.97 mg/kg, 1.4–1.6 m – 2.89 mg/kg, 2.8–3.0 m – 2.77 mg/kg, so in the vertical distribution there is a decrease in the concentration of mobile forms of Cu with depth.

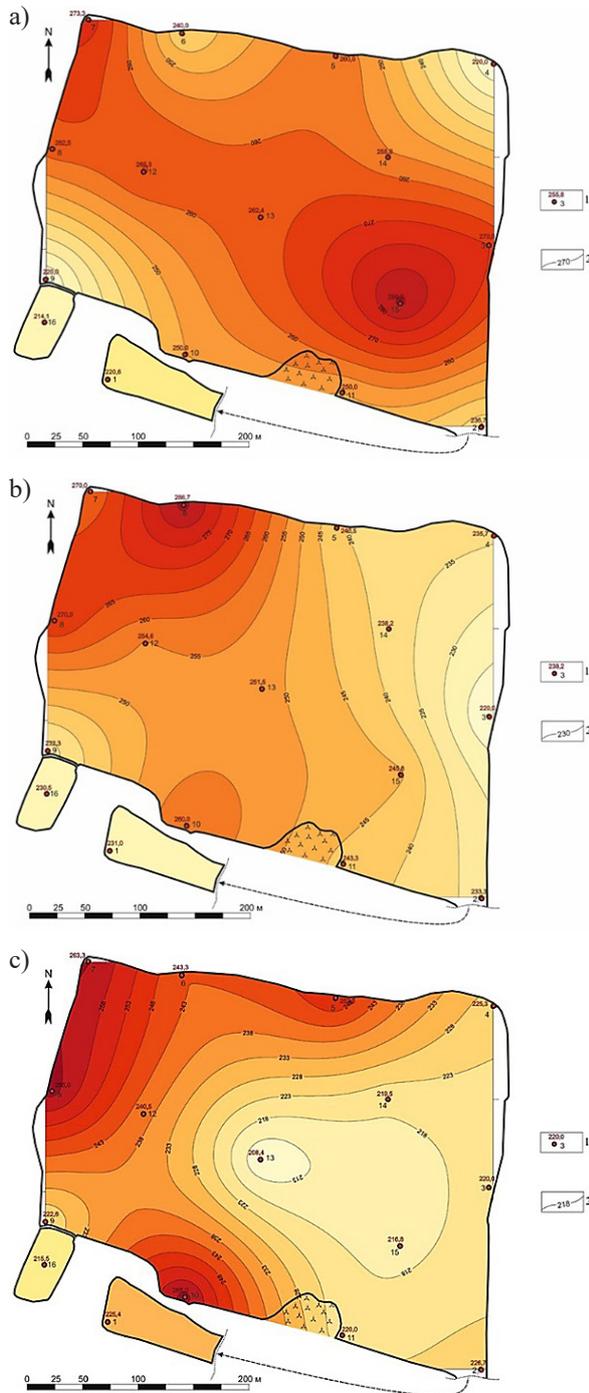


Figure 3. Schematic map of Cu content in the depth range a) 0–0.2, b) 1.4–1.6, c) 2.8–3.0 m: 1 – well number and Cu content, mg/kg; 2 – isolines of the same content of Cu

Determination of the gross Cu concentration in sludge is 208.4–286.7 mg/kg of air-dry sample of sludge (Fig. 3). The comparison of heavy metal content was carried out in relation to the limits approved by the EU Directive for sewage sludge for use in agriculture (Council Directive 86 / 278 / EEC) is 1000–1750 mg/kg, and the permissible heavy metal content in sewage sludge according to group I (Use or manufacture compost in doses adequate to standard fertilizers) (DSTU ISO 7359:2013, 2014) – 100–300 mg/kg.

The average data from different horizons showed that at a depth of 0–0.2 m the average concentration is 249.18 mg/kg, 1.4–1.6 m – 245.84 mg/kg, 2.8–3.0 m – 233.11 mg/kg. Therefore, in the vertical section of the silt field, the maximum contents of this metal are characteristic of the surface layer, and the regularity of the decrease in the concentration of the gross Cu content with depth is traced. Laterally, there is a certain spatial correlation of the Cu content with the content of organic matter in the sludge: the maximum metal concentrations in the near-surface and middle intervals are concentrated in the central part of the silt field, in the bottom interval – in the peripheral parts (Fig. 3).

A series of studies consisting of 4 gradual stages were conducted and described in detail in previous publications (Vankovyň et al., 2021, Tymchuk et al., 2021a, Tymchuk et al., 2021b) [17–19]. These studies confirmed the possibility of using sludge as a component for growth substrate, the proportion of sludge that does not have an inhibitory effect is ≈ 20%, but the addition to the substrate of a small proportion of natural sorbents 5–10% doubles the use of sludge and allows using ≈ 40% of sewage sludge without negative consequences for growth and plant development (Fig. 4.).

The results obtained show that it is possible to use sewage sludge to create a substrate that can be used for the biological reclamation of the land disturbed by man. For example, the largest such facility that can be considered for implementation is the Hrybovytsia landfill (Savchyn et al., 2020). The land freed from the deposited sediments can be used for the production of green energy (Shapoval et al., 2017, Fedoryshyn et al., 2008).

Thus, all available sewage sludge from the sludge site is used – 407659 m³ – and a growth substrate is created from them, a significant area can be provided for the cultivation of man-made disturbed lands, namely 1,019,147 m² or 101.9 ha (using the upper layer thickness in 1 m.).

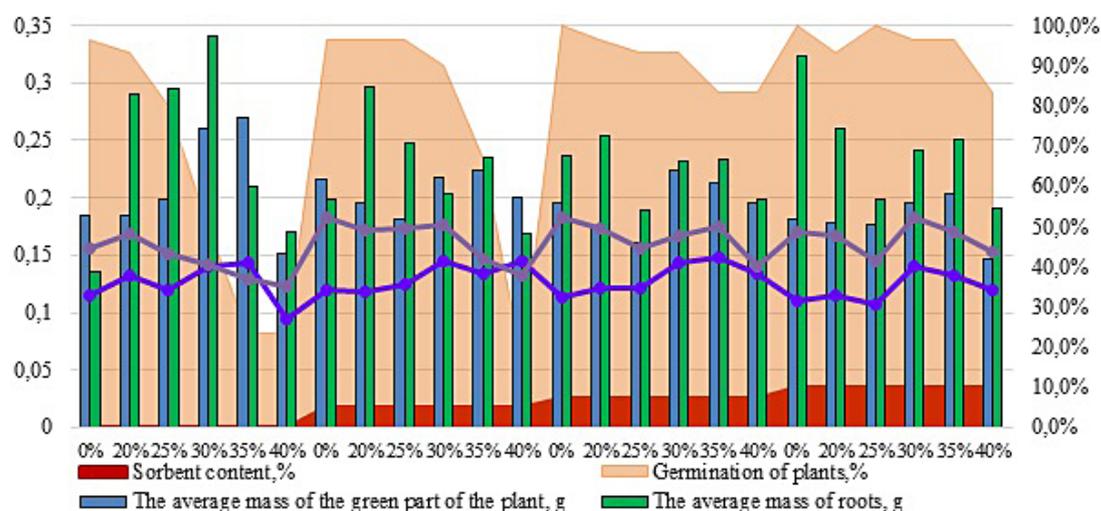


Figure 4. Development of bioindicator plants depending on the composition of the substrate

CONCLUSIONS

Monitoring studies were carried out on the territory of the sludge sites of Lviv wastewater treatment plants. The amount of sludge that can potentially be used for reclamation purposes, their main chemical and biological indicators, as well as limiting factors for the use of sludge for reclamation purposes were identified. The content of organic matter in the sludge of wastewater is in the range of 28–44 wt.%, whereas mineral content – 46–60 wt.%. The dependence on the distribution in horizontal and vertical horizons was also established.

Analytical studies showed that the content of mobile forms of Cu range from 1.36 to 5.40 mg/kg of air-dry sample of sludge. The average data from different horizons showed that at a depth of 0.0–0.2 m the average concentration is 2.97 mg/kg, 1.4–1.6 m – 2.89 mg/kg, 2.8 – 3.0 m – 2.77 mg/kg. Therefore, in the vertical distribution, there is a decrease in the concentration of mobile forms of Cu with depth. Determination of the gross concentration of Cu in the sludge is 208.4–286.7 mg/kg of air-dry sample of sludge. The average data show that at a depth of 0.0–0.2 m the average concentration is 249.18 mg/kg, 1.4–1.6 m – 245.84 mg/kg, 2.8–3.0 m – 233.11 mg/kg. Therefore, in the vertical section of the silt field, the maximum contents of this metal are characteristic of the surface layer, and the regularity of the decrease in the concentration of the gross Cu content with depth was traced. The obtained results of the qualitative composition of the sediments indicate their relatively safe chemical composition for use as a substrate.

Laboratory research conducted using the bioindication methods confirmed that sewage sludge can be effectively used as a component of the substrate for biological land reclamation. However, there are some features regarding the components of the substrate. In order to increase the share of sewage sludge from 20% to 40%, it is recommended to use natural sorbents (5–10%). It is necessary to study the effect of substrate based on sewage sludge and natural sorbents on plant growth and development.

REFERENCES

1. Bień J.D., Bień B. 2015. Zagospodarowanie komunalnych osadów ściekowych metodami termicznymi w obliczu zakazu składowania po 1 stycznia 2016. *Inżynieria Ekologiczna*, 45, 36–43.
2. Blöcher C., Niewersch C., Melin T. 2012. Phosphorus recovery from sewage sludge with a hybrid process of low pressure wet oxidation and nanofiltration. *Water Research*, 46(6), 2009–2019. DOI: 10.1016/j.watres.2012.01.022
3. Council Directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC) / Official journal of the European communities. L 181/6.– 4.7. 86.
4. Fedoryshyn R., Matiko F., Pistun Y. 2008. Prospects for improving the accuracy of natural gas accounting and for reducing gas unbalances. Paper presented at the Annals of DAAAM and Proceedings of the International DAAAM Symposium, 485–486.
5. Fijalkowski K., Rorat A., Grobelak A., Kacprzak M.J. 2017. The presence of contaminations in sewage sludge – The current situation. *Journal of Environmental Management*, 203, 1126–1136.

6. Inglezakis V.J., Zorpas A.A., Karagiannidis A., Sklari S. 2014. European Union legislation on sewage sludge management. *Fresenius Environmental Bulletin*, 23(2A), 635–639.
7. Kacprzak M., Neczaj E., Fijałkowski K., Grobelak A., Grosser A., Worwag M., Singh B. 2017. Sewage sludge disposal strategies for sustainable development. *Environmental Research*, 156, 39–46.
8. Kalogo Y., Monteith H. 2012. *Energy and Resource Recovery from Sludge*. IWA Publishing. 11. DOI: 10.2166/9781780404653
9. Mininni G., Blanch A.R., Lucena F., Berselli S. 2014. EU policy on sewage sludge utilization and perspectives on new approaches of sludge management. *Environmental Science and Pollution Research*, 22(10), 7361–7374.
10. Nykyforov V., Malovanyy M., Kozlovska T., Novokhatko O., Digtar S. 2016 *Eastern-European Journal of Enterprise Technologies*, 5(10), 11. DOI: 10.15587/1729-4061.2016.79789
11. Savchyn I., Lozynskiy V., Petryk Y., Marusazh K. 2020. Geodetic monitoring of the protective dam of the Lviv MSW landfill after reconstruction. Paper presented at the Geoinformatics 2020 - XIXth International Conference, Geoinformatics: Theoretical and Applied Aspects.
12. Sehin T.B., Hnatysh S.O., Maslovska O.D., Halushka A.A., Zaritska Y.H. 2020. Biochemical indicators of green photosynthetic bacteria *chlorobium limicola* response to Cu²⁺ action. *Ukrainian Biochemical Journal*, 92(1), 103–112. DOI: 10.15407/ubj92.01.103
13. Shapoval S., Shapoval P., Zhelykh V., Pona O., Spodyniuk N., Gulai B., Savchenko O., Myroniuk K. 2017. Ecological and energy aspects of using the combined solar collectors for low-energy houses. *Chemistry and Chemical Technology*, 11(4), 503–508.
14. Tymchuk I., Malovanyy M., Shkvirko O., Yatsukh K. 2021b. Sewage sludge as a component to create a substrate for biological reclamation. *Ecological Engineering and Environmental Technology*, 22(4), 101–110.
15. Tymchuk I., Malovanyy M., Shkvirko O., Chornomaz N., Popovych O., Grechanik R., Symak D. 2021a. Review of the global experience in reclamation of disturbed lands. *Ecological Engineering and Environmental Technology*, 22(1), 24–30.
16. Tymchuk I., Malovanyy M., Shkvirko O., Vankovych D., Odusha M., Bota O. 2020. Monitoring of the condition of the accumulated sludge on the territory of Lviv wastewater treatment plants. *Conference Proceedings of International Conference of Young Professionals, GeoTerrace-2020*, V. 2020, 1–5. DOI: 10.3997/2214-4609.20205714
17. Vankovyňh D., Bota O., Malovanyy M., Odusha M., Tymchuk I., Sachnyk I., Shkvirko O., Garasymchuk V. 2021. Assessment of the prospects of application of sewage sludge from Lviv wastewater treatment plants for the purpose of conducting the biological reclamation. *Journal of Ecological Engineering*, 22(2), 134–143. DOI: 10.12911/22998993/130892
18. Yatsuk I.P., Baliuk S.A. 2013. Methods of agrochemical certification of agricultural lands, 104.
19. Zhou K., Barjenbruch M., Kabbe C., Inial G., Remy C. 2017. Phosphorus recovery from municipal and fertilizer wastewater: China's potential and perspective. *Journal of Environmental Sciences*, 52, 151–159. DOI: 10.1016/j.jes.2016.04.010