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Agroecological assessment of sludge from wastewater treatment plants in the aim of their use in the practice

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Abstract

Sludge is an environmental problem, as it accumulates in the area of treatment plants, hindering their functioning and pollute the environment. At the same time, due to the shortage of organic sources in our country, sludge is a rich organic reserve due to the organic matter, macro and microelements contained in them. The aim of the study is to make an agroecological assessment of sludge, including agrochemical, chemical and microbiological characteristics of 5 WWTP's sludge. Verified international standardized methods as well as validated internal laboratory methods are applied.

The conducted studies of sludge from the surveyed five wastewater treatment plants (WWTP) show that in terms of chemical and agrochemical characteristics they are biomass, rich in organic matter and nutrients. The values of heavy metals in the studied batches of sludge, compared to the maximum permissible concentrations (MPC) in the normative documents, are lower and do not limit the use of sludge in agricultural practice. Sanitary and microbiological tests have shown that in some of the sludge self-cleaning and decontamination has occurred, and in others the indicator microorganisms are in quantities higher than required. This is an indication that more residence time or appropriate treatment is needed so that they can be used in practice without posing a health risk to humans.

Summarized, with proper management of WWTP sludge, after conducting chemical, agrochemical and biological studies in accordance with the requirements of the legislation, they can be used as an appropriate soil improver without environmental pollution in agriculture and forestry, for reclamation of disturbed terrains, etc. In each individual case it is necessary to develop a specific technological project.

Key words: WWTP's sludge, characteristics, practical use

Introduction

Biological treatment of wastewater from the settlements is carried out in the urban treatment plants. During this process, in addition to the liquid fraction, large amounts of sludge are obtained. On average, they represent about 0.5-1.5% of the total volume of wastewater, and in some cases can reach up to 10%. Sludge from biological wastewater treatment is an environmental problem, as it accumulates in the area of the stations, hinders their functioning and can pollute the natural environment. About 2.5 million tons of dry sludge are produced annually. In accordance with the European and Bulgarian legislation, the sludge from the wastewater treatment plants (WWTP) can be utilized in the agricultural practice under certain conditions (Directive 86/278/EEC, 1986; Ordinance on the procedure and manner of utilization of sludges from wastewater treatment through their use in agriculture, 2016). They are biomass, rich in organic matter, macro and microelements and can be used as an organic reserve, in connection with the shortage of organic fertilizers in our country and the disturbed balance of organic matter in Bulgarian soils (Marinova, 2008; Marinova et al., 2014a; Marinova et al., 2014b; Pallavi et al., 2015).

In addition to organic matter and nutrients, the sludge can also contain a number of contaminants - heavy metals, organic pollutants and pathogens. The composition of the sludge is determined by the inlet wastewater and by the processes related to the wastewater treatment. Primary, secondary and tertiary sludges are generated in the treatment plants at the different stages of treatment (Marinova et al., 2006; Colón et al., 2017; Pöykiö et al., 2019; Wang et al., 2005).

Sludge production increases every year as a result of industrial development and production intensification. According to a European Commission report published in 2010, 39% of sludge is recycled in agriculture (Lamastra et al., 2018). The literature states that in 2020 the total sludge production in the countries of the European Union (EU) will reach 12 997 000 tons of dry matter (Tytła, 2019).

Safe disposal of sludge is one of the main environmental problems worldwide. The main document indicating the order and manner of use of WWTP sludge in Bulgaria is the Ordinance on the order and manner of utilization of sewage sludge through their use in agriculture (Council of Ministers, 2016), which transposes the provisions of "Directive 86/278/EEC on the protection of the environment, and in particular the soil, when sewage sludge is used in agriculture". The Ordinance specifies the indicators that must be monitored when assessing sludge and the maximum permissible concentrations (MPC) with which the analyzes must be compared. It sets out the requirements that sludge must meet in order to ensure that it does not have an adverse effect on human health and the environment. According to this document, sludge producers are obliged to treat them by methods that provide conditions for the completion of the fermentation process. The purpose is to limit the release of unpleasant odors, as well as to prevent the spread of pathogenic organisms (Directive 86/278 / EEC, 1986; Ordinance on the procedure and manner of utilization of sludges from wastewater treatment through their use in agriculture, 2016; National strategic plan of the Republic of Bulgaria 2014-2020).

With proper sludge management, if they meet the requirements of the legislation, they can be used for fertilization of agricultural crops, improvement of soils with low natural fertility, land reclamation, in forestry, park construction and maintenance of lawns, composting, in floriculture, etc. (Banov et al., 2016; Marinova, 2002; Marinova et al., 2014; Marinova & Chuldiyan, 1998; Markov & Marinova, 2006).

The specific purpose of the study is to analyze sludge and on the basis of the results of agrochemical, chemical and microbiological analyzes, to make an agroecological assessment of sludge.

Material and Methods

Three times, during a certain time interval (months: March, May and September), from 5 WWTPs average samples of dewatered sludge were taken after stay. Sampling was performed in accordance with the Ordinance on Sludge and Guidance on Sampling of Sludge from Wastewater Treatment Plants, Water Treatment Plants and Production Processes, defined by BDS EN ISO 5667-13: 2004. Sampling and analysis cover the spring (March - May) and autumn (September) seasons, as it is recommended that the sludge be introduced into the soil before spring and before deep autumn plowing. The sampling was performed from predetermined batches of sludge from the different WWTPs and a residence time of 5-7-10 months. The sampling points from the designated batches of sludge are random.

The performed chemical analyzes are in accordance with international EN and ISO standards, which for the most part are approved BDS methods.

During the microbiological tests, a validated intra-laboratory test method was applied, which is a combination of approaches described in CEN / TR standards, intended for sludge testing and a methodology developed and applied by the NCPHA team. The helminths were tested by the ILM / P / 19/1995 method.

The methods by which the chemical, agrochemical and microbiological analyzes were performed are listed in Table 1.

Results and Discussion

The sediments vary greatly in their characteristics. The various indicators range quite widely and depend on many factors. The composition of the wastewater at the inlet, the chemical and biological conditions under which the treatment processes take place have the greatest influence (Marinova et al., 2016).

The results of the chemical and agrochemical characteristics of the investigated WWTP sludges are given in Table 2.

The data show that the measured dry matter value allows sludge to be transported and spread by mobile means such as manure.

Nitrogen content is of great importance for the use of sludge in agriculture. The effectiveness of nitrogen depends largely on its distribution in organic and inorganic form. It was found that the content of ammonia nitrogen is higher in sludges that have a lower content of absolute dry matter. As the amount of dry matter increases, ammonia nitrogen decreases. This is explained by losses of ammonia nitrogen during the period of storage of the sludge and scattering in the field. In order to use the organic nitrogen in the sludge, it must be converted to an ammonia form, which is further nitrified to nitrates. This depends to a large extent on the characteristics of the sludge. The mineralization of organic matter takes place faster under aerobic conditions than under anaerobic ones.

From the analyses of the sludge (Table 2) it was established that the content of total nitrogen is sufficient to satisfy the needs of the plants for nitrogen. Not only the total nitrogen content but also the amount of ammonia nitrogen is taken into account. In order to limit nitrogen losses, it is appropriate to carry out plowing, disking or cultivation immediately after spreading the sludge on the soil surface, so that the imported nitrogen can come into contact with the soil absorption complex, whereby losses are reduced to a minimum.

In biological wastewater treatment, sludge is enriched with phosphorus. It plays an important role in plant development. From this point of view, sludge is of great importance for regulating the plant diet.

The potassium content is low. This is due to the increased solubility of potassium salts, which remain in the liquid fraction during sludge formation.

The content of calcium and magnesium have different effects on the properties of the soil. Calcium affects the plants and the structure of the soil, leads to a change in the reaction of the environment and to the dynamics in the assimilation of the various elements.

Magnesium also affects plants with its physiological role. The cations of this element are easily assimilated by plants.

The results for the agrochemical parameters of the studied sludges from WWTP (Table 2) prove that they are biomass, rich in organic matter and nutrients, which characterizes them as potentially rich energy sources. Their composition resembles

Table 1. Analytical methods used for analysis of WWTP sludge

pH/H_20/	BDS EN 15933
Dry substance, %	BDS EN 12880
Organic carbon,%	BDS EN 15935
Total quantities on a dry matter basis:N, %	BDS EN 13342
P; K, %	BDS EN 16170
N-NH_4^+; N-NO_3^-, %	SD CEN/TS 16177
Metabolite rates on a dry matter basis	
Mg; Ca; P; K %	BDS EN 16170
Sulphur (water soluble, such asSO_4^{2-}), %	BDS EN ISO 10304-1
Heavy metals and arsene	
As; Hg	BDS EN 16170
Cd; Cr; Ni; Cu; Zn; Pb	ISO 11047
Microbiological indicators	LMT:ICZ 02.01:2016-08-05
Helminths	ILM/P/19/1995

Table 2. Agrochemical parameters of the investigated WWTP sludges

Indicators	WWTP № 1			WWTP № 2			WWTP № 3			WWTP № 4			WWTP № 5		
pH / H_20/	7.27	7.93	8.05	7.15	6.30	6.85	9.14	8.62	7.10	8.51	7.26	6.5	8.43	7.93	7.30
dry substance, %	85.27	69.07	79.07	85.27	95.2	96.00	95.4	97.3	95.3	28.70	29.49	34.78	34.78	29.07	48.50
organic C_2, %	27.77	19.76	20.57	17.77	19.9	18.19	35.36	23.89	29.9	37.87	27.81	28.9	27.77	32.76	26.50
Total amounts based on absolute dry matter, %															
N	6.23	6.12	7.25	3.23	2.49	3.05	2.00	3.75	1.71	5.16	8.05	3.94	3.43	4.12	2.85
P	2.18	2.24	3.02	1.15	1.21	1.11	0.71	1.44	0.72	1.27	2.71	1.15	3.18	2.21	1.98
K	0.25	0.37	0.18	0.19	0.15	0.18	0.28	0.28	0.20	0.26	0.35	0.29	0.22	0.37	0.29
N - NH_4^+,%	0.41	0.38	0.21	0.075	0.069	0.022	0.03	0.14	0.01	0.15	0.11	0.13	0.41	0.033	0.001
N-$N0_3^-$, %	0.0015	0.0018	0.0024	0.16	0.16	0.12	0.001	0.22	0.006	0.029	0.034	0.072	0.0013	0.0010	0.0010
Metabolite quantities, %															
Ca	0.64	0.88	0.70	0.92	0.93	0.97	3.99	0.71	2.18	2.28	2.97	1.13	0.64	1.15	0.42
Mg	0.11	0.10	0.072	0.11	0.13	0.13	0.21	0.055	0.16	0.41	0.26	0.165	0.11	0.16	0.15
P	17.9	15.7	38.0	0.080	0.08	0.063	0.20	0.0052	0.24	0.42	0.28	0.373	0.058	0.025	0.018
K	0.045	0.004	0.071	0.058	0.058	0.060	0.11	0.11	0.15	0.18	0.15	0.13	0.12	0.11	0.011
S/water soluble as SO_4^{2-}/,%	0.11	0.22	0.31	0.33	0.22	0.48	0.12	0.22	0.48	0.28	0.24	0.29	0.11	0.22	0.22

nitrogen-phosphorus fertilizer with the possibility of use in agriculture. Most of the nutrients are in organic form and mineralize graduallyThe content of nutrients - nitrogen (N) and phosphorus (P) are sufficient to meet the needs of plants not for one but for several years. In potassium-loving crops, potassium (K) must be added through mineral fertilizers. The sludge has reached equilibrium of humus systems and could be used as organic fertilizer in agricultural practice (Stankova et al., 2021).

When using sludge in agricultural practice, the presence of heavy metals and arsenic must be taken into account, the content of which above the maximum permissible concentrations is a limiting factor for the optimal fertilizer rate of sludge. Many of these elements in minimal amounts are necessary for the development of living organisms, but in high concentrations are toxic to plants, soil, groundwater, animals and humans.

The data on the content of heavy metals and arsenic in the investigated WWTP sludges in the present study are given in Table 3. They are compared with the requirements for these elements given in the Ordinance on the use of sludge in agriculture and with the European norms.

The obtained results for arsenic and mercury content in the analyzed samples are below the limits of determination of the analytical procedures (LOQ 10 mg / kg and LOQ 1 mg / kg, respectively) for these elements. The established values for Cd, Cr, Ni, Cu, Zn and Pb are below the maximum permissible concentrations for heavy metals in sludge, according to the cited documents and do not limit their use in practice (Stankova et al., 2021). A similar conclusion is reached by the authors of another Bulgarian study (Sidjimov et al., 2013).

Sanitary-microbiological tests (Table 4) found that in sludge №1 and №2 self-cleaning and decontamination occurred, and in №3, №4 and №5 in some of the batches the presence of E.coli and Clostridium perfringens was observed in values higher than required. These sludges need more residence time or appropriate treatment to be used without posing a health risk. In the studied sludge there are no pathogenic microorganisms from genus Salmonella, and no viable helminth eggs are observed. Bacteria of the genus Salmonella are characterized by the difficulty of surviving in this environment due to the presence of a large number of other microorganisms with which they compete for nutrients. Other studies conducted in Bulgaria received similar negative results regarding the absence of pathogenic bionts in sludge from WWTPs (Georgieva et al., 2017).

In addition to the microbiological parameters laid down in the normative documents - *E. coli, Clostridium perfingens, Salmonella spp.* - the samples are analysed for enterococci and coliforms, which are also of sanitary importance. They provide information on the condition of the sludge in terms of their load of organic matter of anthropogenic nature and the degree of self-purification (Sidjimov et al., 2016). In most of the analysed samples these microorganisms are in large quantities.

Although all members of coliforms are associated with faecal contamination, the ratio between the individual genera and species included in this group varies considerably under certain conditions. Fresh faecal contamination is dominated by typical *E. coli*. As self-cleansing progresses, the microbial ratio shifts to other representatives of coliforms - the genera *Enterobacter, Citrobacter* and *Klebsiella*. No less important are enterococci, which are more often used as indicators of fresh faecal contamination and are more resistant to environmental conditions. The presence of sulfite-reducing anaerobic clostridia - including *Clostridium perfingens*, in the absence of coliforms, is an indicator of old fecal contamination. Thus, each of the used sanitary-microbiological indicators reflects a certain moment related to the pollution and self-cleaning of the sludge. In the case of pre-treatment of fresh sludge with coagulants or flocculants and mechanical dehydration with belt filter presses, self-cleaning occurs after drying fields, and their complete decontamination ends after 12-15 months (Marinova, 2008).

There are complex relationships and interactions between soils and sludge, which has been the subject of many studies and publications. Imported into the soil, sludge becomes a source

Table 3. Content of heavy metals in WWTP sediments (in mg/kg)

WWTP	As	Cd	Cr	Ni	Cu	Zn	Pb	Hg
№1	<10	2.9	109	75.0	405.4	1090	70.4	<1
	<10	2.1	54.2	45.8	316.5	1054	47.9	<1
	<10	2.5	77.7	36.3	310.0	1030	67.5	<1
№2	<10	2.9	47.5	25.0	205.9	1090	70.4	<1
	<10	2.2	50.9	27.4	215.4	1000	64.2	<1
	<10	2.0	49.2	25.5	214.1	980.0	72.5	<1
№3	<10	< 0.25	44.7	26.7	136.0	483.3	41.8	<1
	<10	< 0.25	32.5	16.3	91.21	732.3	23.4	<1
	<10	< 0.25	30.4	18.8	125.9	540.4	51.2	<1
№4	<10	2.9	109	75.2	405.2	1090	2.71	<1
	<10	0.98	61.5	27.2	204.2	1340	21.6	<1
	<10	< 0.25	59.4	43.5	132.9	410.5	36.4	<1
№5	<10	2.9	109	75.0	405.4	1090	70.4	<1
	<10	1.2	57.0	68.0	210.0	894.0	82.3	<1
	<10	2.8	23.8	44.3	179.0	769.0	78.0	<1
MPC * Ordinance	25	30	500	350	1600	3000	800	16
MPC ** Directive	-	40	-	400	1750	4000	1200	25

MPC * - Ordinance on the procedure and manner of utilization of sludges from wastewater treatment through their use in agriculture
MPC **- Council Directive 86/278 / EEC on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture

for the formation of humus and available to plants nutrients - nitrogen, phosphorus, potassium, sulphur and microelements - iron, manganese, zinc, copper and others. Only a small part of this nitrogen is in an easily digestible form for plants. In the process of mineralization, which depends on many factors (humidity, soil temperature, pH, humus supply, etc.), the amount of nitrogen forms available to plants increases (Banov et al., 2016; Marinova, 2008; Marinova et al., 1994; Toncheva et al., 2014).

Technical, grain, fodder and vegetable crops, as well as perennials, can be fertilized with sludge. The fertilization rate depends on the type of crops grown, the composition of the sludge and the condition of the cultivated soils (Marinova, 2008; Marinova et al., 1994; Pallavi et al., 2015).

Under the influence of sludge fertilization, the yield of winter wheat increases significantly at a sludge rate of 1-2 t / da with 30% humidity (Marinova et al., 2012b).

Studies have shown that the optimal sludge rate for potatoes at 50% humidity is 3t/da, and for cereals - 6t/da. Further increase of the norm is ineffective (Marinova et al. 1999).

As a result of field experiments with alternation of wheat - cabbage - tomato, it is proved that the yield increases significantly when sewage sludge is applied. As an appropriate norm, 20 t/ha of sludge is recommended, which is also in accordance with the climatic conditions in the area where the experiments were performed (Özyazici, 2013).

In the coming years, stabilized sludge is expected

Table 4. Microbiological and parasitological characteristics of WWTP sludge

WWTP	Salmonella spp.	Coliforms	E. coli	Enterococci	Clostr. perfingens	Viable helminth eggs
№1	Absence in 20g	<0.30	<0.30	<0.30	<0.30	Not detected
	Absence in 20g	<0.30	<0.30	0.36	24	Not detected
	Absence in 20g	2.9	2.9	<0.30	<0.30	Not detected
№2	Absence in 20g	>1.1x10^{4} (>11 000)	<0.30	1.1x10^{2} (>110)	>1.1x10^{4} (>11 000)	Not detected
	Absence in 20g	>1.1x10^{2} (>110)	<0.30	>1.1x10^{2} (>110)	2,3	Not detected
	Absence in 20g	>1.1x10^{2} (>110)	<0.30	>1.1x10^{2} (>110)	24	Not detected
№3	Absence in 20g	>1.1x10^{3} (>1 100)	<0.30	>1.1x10^{3} (>1 100)	>1.1x10^{3} (>1 100)	Not detected
	Absence in 20g	>1.1x10^{5} (>110 000)	<0.30	>1.1x10^{3} (>1 100)	>1.1x10^{4} (>11 000)	Not detected
	Absence in 20g	>1.1x10^{4} (>11 000)	>1.1x10^{4} (>11 000)	>1.1x10^{3} (>1 100)	2.3	Not detected
№4	Absence in 20g	>1.1x10^{4} (>11 000)	<0.30	>1.1x10^{2} (>110)	>1.1x10^{4} (>11 000)	Not detected
	Absence in 20g	>30	2.4x10 (24)	2.4x10 (24)	>1.1x10^{2} (>110)	Not detected
	Absence in 20g	>1.1x10^{3} (>1 100)	<0.30	>1.1x10^{4} (>11 000)	>1.1x10^{4} (>11 000)	Not detected
№5	Absence in 20g	2.4.10^{6} (2 400 000)	2.4x10^{6} (2 400 000)	>1.1x10^{4} (>11 000)	1.5x10^{2} (150)	Not detected
	Absence in 20g	2.4x10^{4} (24 000)	<0.30	>1.1x10^{2} (>110)	<0.3	Not detected
	Absence in 20g	2.4x10 (24)	>1.1x10^{6} (>1 100 000)	>1.1x10^{2} (>110)	>1.1x10^{5} (>110000)	Not detected
Requirements	Not allowed in 20 g	It is not regulated	Up to 100/g (up to 1x10^{2}/g)	It is not regulated	Up to 300/g (up to 3x10^{2}/g)	1 per 1 kg of dry matter

to be used in large quantities in agriculture in many EU Member States, such as France, Germany, Italy, Spain, as well as in the United Kingdom (Colón et al., 2017). In the Mediterranean regions, e.g. in France, about 30% of the sludge produced is used for agricultural purposes (Marinova et al., 2012a; 2014; 2015).

Apart from being a fertilizer in agriculture, sludge can be used for reclamation, which allows to accelerate the creation of vegetation on the disturbed lands. The soils in such lands have unfavorable physicochemical properties for plant development. The use of heavy machinery in the process of agricultural cultivation also leads to compaction, which negatively affects the development of plants and creates a risk of erosion and water pollution (Banov et al., 2016; Marinova, 2002; Marinova & Tsolova, 2005). In Western Europe, extensive experience has been gained in the use of sludge for the reclamation of disturbed terrains from the extraction of ore and non-ore minerals. High rates for sludge application apply. For example, in the UK, about 10% of the total volume of sludge produced by wastewater treatment plants is used for land reclamation (Mozafa, 1994).

The use of sludge in forestry has a positive effect on the growth of forest trees. Higher rates are used for fertilizing forest soils. Sludge reclamation on disturbed forest terrains is effective. It is recommended to apply solid or composted sludge (Trifonova et al., 2013).

Sludge can also be used in urban landscaping. The use of sludge in park construction and maintenance of lawns has a positive impact on the areas of cities and their surroundings. In Norway, compost is produced by German methods for accelerated decomposition of waste in a bioreactor or by the method of field composting using forced aeration. The application of compost maintains the high water holding capacity of the soil in the green areas. The water-physical properties are improved and the danger of erosion is reduced. All this has a positive impact on the development of tree and shrub species. To reduce the risk of contamination of groundwater with nitrates, special studies are conducted. It is necessary to determine the time, place and grass mixtures for the restoration of lawns (Marinova, 2000).

The use of sludge in floriculture requires composting. The composted sludge is loose, with good structure. The additional treatment of the sludge with lime leads to the improvement of the organoleptic properties necessary in floriculture. In these cases, the recommended norms usually exceed the norms for agricultural soils (Marinova, 2008).

The main principle that we must adhere to is that the soil should not serve as a final receiver of pollutants. In each case it is necessary to develop a specific technological project, including assessment of sediments, soil, terrain and agricultural conditions, mode of transportation, fertilization rates, type of crops, the economic effect of the activities and more (Marinova, 2008; Marinova et al., 1994; Pallavi et al., 2015).

Conclusions

The results of the studies of sludge from the surveyed five wastewater treatment plants (WWTP) can be summarized in the following conclusions:

1. The chemical and agrochemical characteristics of the studied sludges show that they are biomass, rich in organic matter and nutrients.

2. The values of heavy metals in the studied batches of sludge, compared to the maximum permissible concentrations (MPC) in the normative documents, are lower and do not limit the use of sludge in agricultural practice.

3. Sanitary-microbiological tests have shown that in some of the sludges self-cleaning and decontamination has occurred, and in others the indicator microorganisms are in quantities higher than required. This is an indication that more residence time or appropriate treatment is needed so that they can be used in practice without posing a health risk to humans.

4. Pathogenic microorganisms of the genus Salmonella, as well as viable helminth eggs, are absent in the studied sludge.

Summarized, with proper management of WWTP sludge, after conducting chemical, agrochemical and biological research in accordance with the requirements of the legislation, they can be used as a suitable soil improver without environmental pollution in both agriculture and forestry, for reclamation of disturbed terrains, etc. In each case it is necessary to develop a specific technological project.

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