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THE IMPACT OF SLUDGE STORAGE ON THE SOIL. CASE STUDY: TOMESTI DEPOSIT IN IASI COUNTY, ROMANIA

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Abstract

The study was conducted in the sludge storage area in Tomesti, Iasi County. The sludge from the wastewater treatment process was stored in the Tomesti lagoon, which increased the pollutant concentrations in the soil. The goal of the present paper was to study the impact of sludge storage on the Tomesti soil by analyzing the contents of salts, heavy metals and microbiological loads (total coliform bacteria, enterococci, *Escherichia coli, Clostridium perfringens, Salmonella spp*). The total concentration of heavy metals in soil is a relevant indicator on the risk to human health and environment. Growing plants in polluted areas can be a remedial solution and can serve as an alternative tool in sustainable agriculture. Plants can influence the accumulation of heavy metals in soils, either by decreasing or increasing their accumulation. Following the carried analyses, we can say that some physical and chemical properties of the sludge were propagated to the soil on which the sludge is stored. Thus, the moisture of the soil located below the sludge layer increased by up to 20%, compared to the humidity of the neighboring soil, that is not influenced by the presence of sludge. It was also an adsorption medium for numerous cations and anions. From the group of tested heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn), the only element present at polluting level is zinc. In the first soil horizon on which the sludge is stored, the zinc contents exceed 10.8 times the maximum permissible limit value, and for the neighboring soil this is 1.6 times. From a bacteriological point of view, the soil beneath the sludge lagoon is loaded with *Salmonella* genus bacterial colonies.

Keywords: bacteria; heavy metals, nutrients, *Salmonella spp*

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1. Introduction

Due to the city's urbanization and anthropic activity, the quantity of industrial and domestic waste water increases, fact that requires a treatment process in order to reintroduce that water into the natural water circuit. As a result of wastewater treatment, two fractions of substance result, a liquid one (water) and solid-wet one (sludge), known as a wastewater treatment plant by-product or waste (Agnello et al., 2015). The EC proposals for the reduction of waste by 2050 by 50% also include the sludge from wastewater treatment plants being classified in category 19 of the waste code, according to the Decision no. 856 of August 16, 2002 (Dellisanti et al., 2016). According to EU studies conducted on sludge it results that for 15 EU countries the final sludge disposal, its reuse in agriculture and composting is applied for 53% of produced amounts, and incineration is applied for 20% of the produced amount. Regarding those 12 EU countries, which acceded after 2003, these remained the classic sludge disposal methods (Farahat et al., 2015). Due to obligations established by Directives 91/271/EC and 86/278/EEC which regulate the correct use of sludge, the most widely used methods of sludge recovery in all 27 EU countries are the agricultural

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reuse and final disposal incineration (Mahar et al., 2016).

The wastewater treatment plant sewage sludge is rich in organic and inorganic nutrients; hence, it can be applied on agricultural land as an alternative to mineral fertilizers. Land application is an increasingly popular mean of the reuse of sewage sludge as it allows for recycling of valuable components such as organic matter, N, P and other nutrients (Chunhua et al., 2020). Understanding the impact of sewage sludge treatments is essential in the correct management of agricultural land. Indeed, sewage sludge amendment to the soil modifies the soil's physico-chemical and biological properties, such as plant-available macro-/micronutrient contents and organic matter content (Lan et al., 2020)

The sludge contains organic matter and micro and macronutrients which are potentially useful for agricultural usage. However, it can be harmful when containing undesirable amounts of organic pollutants, heavy metals, or pathogens (Farsang et al., 2020).

Most scientific papers show the sludge positive effects of sludge on plant yield improvement due to the sludge macro-nutrient content (Chrysargyris and Tzortzakis, 2015; Özyazici, 2013), but also due to the better physico-chemical properties of soils on which they are used, by altering the soil bulk density, and aeration and stabilization of eroded soils (Gu et al., 2013; Mihalache et al., 2014). There are also articles that show some negative effects, concerning the increasing in heavy metals in plants and soils, pathogens and esthetical alteration of environment through unpleasant smell (Collivignarelli et al., 2015; Mazen et al., 2010; Vaitkute et al., 2010). The soils are influenced by the hydrological regime, and, as well, as by the abiotic stress factors.

The purpose of the current paper is to present, with the help of laboratory analysis, the effects of sludge disposal from wastewater on the soil and plants. The research results can be used as a scientific basis in the development of an adequate soil management system in the study area.

2. Material and methods

The study was carried out in the area of the Tomesti sludge storage, in Iasi County. The sludge that originates from the wastewater treatment process was stored on the Tomesti lagoon, this resulting in increased concentrations of pollutants in soil. At the same time, the infiltration water that passes through the sludge layer and the underlying soil determines the fact that groundwater gets loaded with pollutants and chemical compounds.

The Tomesti sludge lagoon (Fig. 1) was built in 1994 by removing the soil layer from an area of 9.1 hectares of land and by constructing surrounding and dividing dams. Apart from compacting the underlying clay layers, no other protective measures have been taken. Currently the lagoon has a total active area of 15 ha, out of the total area of 18.9 ha made available by the Tomesti local council. The lagoon was divided into 11 compartments, of different surfaces. Between the compartments there are water and mud circulation gaps. The lagoon's total volume is reaching a value of 225.000 m^3. The lagoon's average elevation is 34 - 35m, and in the surrounding areas the elevation reaches values of up to 200 m.

Fig.1. Satellite image of the sludge lagoon Tomesti, Iasi County

The research on sewage sludge stored in the Tomesti lagoon, started fifteen years ago, in 2005. Over the year's sludge changed from a high-water content raw sludge, to a lower humidity sludge, due to drainage but also due to a moisture consumption performed by the grown vegetation. The area is characterized by a dynamic activity during time with significant degradation due to surface washing, erosion and landslides (Murariu et al., 2007).

The total concentration of heavy metals in soil is a relevant indicator of risk posed to human health and environment (David and Janez, 2018). Plant cultivation in polluted areas can be a remedial solution and can serve as an alternative tool in sustainable agriculture.Plants can influence the accumulation of heavy metals in soils, either by decreasing or increasing their accumulation (Boglárka et al., 2018). The objective of the present paper was to study the impact of the sludge storage on the alluvial soil of Tomesti.

Experimental research was conducted in order to highlight the evolution of heavy metals content in sludge and, as well, in the subsoil and in the soils from the lagoon vicinity. The sludge resulting from processes that take place in the Iasi wastewater treatment plant, one of the largest in the country, was stored for about 10 years (between 1995 and 2006), in lagoons created on a land area of 18.920 m², concessioner from the Tomesti commune council. The sludge was conveyed to the lagoon via a pumping station and a discharge pipe. In 2006 the lagoon was decommissioned for rehabilitation. A year before that, the first research was carried out in order to assess the effect of the sludge on the alluvial soil on which it stagnates and on the soils from boundary areas.

In order to study the effect of sludge storage from the Tomesti platform on the soil, two drillings were performed: the first drilling (F1) was performed at a depth of 300 cm on a control soil located near the lagoon and the second drilling (F2) was performed in the lagoon's compartment 8 at the same depth of 300 cm. The surface sample (0-20 cm) from the F2 drilling represents the sludge itself and down beyond a depth of 100 cm it's the soil which the sludge was stored.

Drilling F2 was located in compartment 8 of the lagoon because the infiltration water flows from compartment 1 to compartment 8. Thus, the samples from this compartment fully characterize the sludge and its influence on the soil.

The sampling of the stored material (the sludge) was carried out in increments of 20 cm to 20 cm by using boreholes (F1) down to a depth of 300 cm (S1, S2,..., S5). There was also a blank drill F2, in the vicinity of the lagoon, at the same depth (SE1, SE2, ..., SE5). From the analysis of sampled material from the lagoon (S1,… S5) it was observed that at depth of 0-20 cm and below this depth the material is relatively well structured, with smaller structural aggregates, the lack of groundwater causing an increase in consistency, plasticity and reduction of interspacing between aggregates.

At the base of the last survey sampling segment, the soil material is still under the influence of water. The research is in its 13th year of investigation, soil samples were taken every year in the first part of July. The hydromorphism influences the degree of the aggregation and the particles structure, which is moderate, even low, and the plasticity is reduced, while the soil material on the upper horizon of the soil is still gleyed.

The analysis of soil properties (pH, humus content, NO_3, N_{total}, P, K) were performed at T National Research and Development Institute for Soil Science, Agrochemistry and Environment – ICPA Bucharest.

- The humidity of the sludge was determined gravimetrically, leaving the samples in the oven at a temperature of 105ºC.

- The reaction of the sludge and the soil, highlighted by the pH index, was potentially determined, in aqueous suspension, using a combined glass-calomel electrode.

- The estimation of the organic carbon content was performed according to the Walkley-Black method, in the donut modification.

- The mobile form of nitrogen ($N\text{-}NO_3$) was determined potentiometrically with an electrode selective for $N\text{-}NO_3$. The determination of the total nitrogen content was performed by the Kjeldahl method.

- The content of mobile phosphorus (PAL) and mobile potassium (KAL) were determined in activated solution with ammonium lactate at pH = 3.7 (after Egnér-Riehm-Domingo) and dosing by spectrophotometry and flame photometry, respectively.

- The total content of soluble salts was determined by the conductometric method, and the soluble anions were determined volumetrically; Flammphotometry (Ca_2^+, K^+, Na^+) and atomic absorption spectrometry ($Mg_2^{+)}$ were used to determine the cations.

The testing of heavy metals content in soil (Cd, Co, Cr, Cu Fe, Mn, Ni, Pb and Zn) was performed by means of atomic absorption spectrometry (Sudip et al., 2018), Zeenit 700 P Analytik Jenna, from the ENERED laboratory of the Department of Hydrotechnical Engineering, Geodesy and Environmental Engineering from the "Gheorghe Asachi" Technical University in Iasi.

Microbiological analyses were performed on samples of muds and soils from deep horizons. Different profiles of the following bacteriological indicators were studied: total coliform bacteria, enterococci, *Escherichia coli, Clostridium perfringens, Salmonella sp*. The soil sample was taken on 07.08.2019, and the period of analysis of the samples was: 13.08.2019 - 21.08.2019.

3. Results and discussion

Following the analyses carried out, the reaction of the fermented sludge is weakly alkaline with pH values between 7.58 and 8.99. Its humidity is slightly higher at the depth of 30-50 cm, compared to the surface horizon, as expected (Table 1). Organic carbon content varies between 16.3 and 21.7% in the first case and between 11.1 and 11.5% in the second case. Compared to primary, active and fermented sludge, the content in mobile nitrogen ($N-NO_3$ and $N-NH_4$) is up to 17 times higher in the case of nitric nitrogen and up to about 4 times in the case of ammoniacal nitrogen. The C/N ratio values are in the range 10-15 for the upper horizons and between 6 and 9 for the lower horizons. These values are close to those of normal, agricultural soils. The alluvial soil of the Bahlui Plains in which the sludge lagoon is located has a high load of soluble salts reaching up an amount of 1.829 mg/100 g soil in the horizon from 60 to 80 cm. This value shows a very strong salinization which is still present at a depth of 120 cm, from 40 to 160 cm. The upper horizons from 0 to 40 cm show a low salinization and the lower ones from 160 to 300 cm show a moderate or weak salinization.

Table 1. Moisture and main chemical properties of soil samples collected from control drilling (F1) and drilling in the sludge deposit (F2)

No. crt	*Identification*	*Depth cm*	*Humidity 1 %*	*Humidity 2 %*	*pH*	*$CaCO_3$*	*Co_{rganic} %*	*N_{total}*	*C/N*	*$N-NH_4$**	*$N-NO_3$* ppm*	*Pal***	*Pal*	*Kal*
1	F1-sample	0-20	17	15	7.85	6.4	4.4	0.369	14	traces	4.9	32.4	56.2	424
2		20-40	26	21	8.19	5.9	1.9	0.214	11	8.8	4.9	19.2	46	350
3		40-60	37	27	8.28	3.6	1.3	0.142	11	8	6.4	10.5	28.3	295
4		60-80	44	31	8.4	1.9	1	0.146	8	5.9	5	6.1	20.1	323
5		80-100	42	30	8.35	1.9	1.1	0.13	10	8.3	4.3	6	18.1	332
6		100-120	41	29	8.35	1.9	0.8	0.113	8	14.8	4.2	10.4	31.4	282
7		120-140	37	27	8.28	3.2	0.7	0.058	15	8.8	4	7.9	21.4	259
8		140-160	38	33	8.17	5.7	0.5	0.097	6	5.6	4	6.4	14.9	268
9		160-180	39	28	8.44	2.1	0.6	0.099	7	5.7	4	6	21.2	277
10		180-200	41	29	8.71	1.5	0.6	0.095	7	9.9	3.8	1.4	11.5	263
11		200-220	29	23	8.85	5.2	0.6	0.082	8	9	3.5	0.4	13.8	231
12		220-240	28	22	8.93	11.7	0.3	0.068	6	4.5	3.2	traces	7.6	186
13		240-260	28	22	8.99	12.8	0.2	0.019	9	6.7	3.2	traces	6.7	172
14		280-300	41	29	8.85	11.1	0.5	0.07	8	11.5	3.5	0.4	13.8	295
15	F2	0-20	575	85	7.65	8.5	10.4	0.986	12	2866	73	236	358	758
16		100-120	117	54	7.58	9.6	8.3	0.767	13	1048	6.5	230	336	460
17		120-140	58	37	7.87	8.1	2.2	0.289	9	192	19.4	84.7	149	378
18		140-160	51	34	8.12	8.5	1.5	0.198	9	160	21.7	49.9	111	330
19		180-200	44	30	8.17	5.3	1	0.15	8	30.2	17.3	20.7	48.6	305
20		240-260	43	30	8.21	5.3	0.7	0.118	7	14.2	7.7	13.1	32.3	373
21		280-300	41	29	8.63	2.1	0.6	0.115	6	9.9	19.4	4.2	24.6	277

*1- moisture relative to the mass of the wet material 2- moisture relative to the mass of the dry material at 105 °C * - recalculated values for dry soil at 105 °C** - recalculated values depending on the soil reaction*

Table 2. Total content of soluble salts and the anionic and cationic composition of soil samples collected from the control well (F1) and from the drilling carried out in the sludge deposit (F2)

Localization	***Depth***	***HCO_3"***	***SO_4^{2-}***	***Cl^-***	***Ca^{2+}***	***Mg^{2+}***	***Na+***	***K+***	***Rez.cond.***	***Rez. min.****
		mg/100 g /soil							***mg/100 g /soil***	
F1-sample	0 -20	46	1	14	5	19	12	4	76	92
	20-40	47	70	27	7	35	41	3	186	205
	40-60	37	790	69	53	55	270	5	1149	1248
	60-80	34	1479	77	196	111	333	7	1829	1717
	80-100	34	1357	77	165	96	338	7	1707	1660
	100-120	35	932	63	85	55	297	4	1272	1330
	120-140	38	1330	50	211	80	288	5	1516	1437
	140-160	37	1044	35	145	61	252	5	1180	1235
	160-180	66	322	40	14	15	158	3	449	618
	180-200	76	231	42	15	33	86	2	267	485
	200-220	81	46	20	6	8	46	1	160	210
	220-240	73	55	24	5	12	44	1	173	219
	240-260	70	49	30	5	10	46	1	176	216
	280-300	76	28	25	5	4	47	2	183	190
F2	0-20	53	732	49	126	55	144	18	918	916
	100-120	46	595	28	137	44	71	13	775	627
	110-140	43	310	24	76	25	41	8	481	424
	140-160	41	280	21	48	27	53	5	510	476
	180-200	44	1116	34	172	78	225	7	1246	1248
	240-260	44	840	30	104	45	230	5	996	1101
	280-300	43	449	32	36	25	162	3	566	750

The total content of soluble salts and the anionic and cationic composition of soil samples collected from the control well (F1) and from the drilling carried out in the sludge lagoon (F2) are presented in Table 2. In terms of heavy metals contents in fermented sludge there was a slight concentration of these chemical elements, without major differences between the values recorded in the sludge stored on the platform inside the treatment plant and in the sludge from the Tomesti lagoon. If we compare these contents with the maximum permissible values of heavy metals in sludge, in order for the sludge to be accepted as soil fertilizer (Table 3), we find that the analytical data of the heavy metal contents in sludge are lower than the values of the maximum permissible limits, except for zinc. In the case of this last chemical element, its load exceeds up to 6.4 times the admissible value, the average of the exceeding being of about 3 times.

Microbiological tests performed on sludge and soil samples from horizons from different profile depths were focused on bacteriological indicators (total coliform bacteria, enterococci, *Escherichia coli, Clostridium perfringens, Salmonella sp*,). The calculation and expression of the results for the Enterococci bacteriological indicator was performed according to Standard SR EN ISO 8199/2008 - "Water quality: Guidelines for counting microorganisms in the culture environment".

For the analysis of bacteriological indicator in F1, the soil seeded plaques from the Tomesti lagoon did not show the presence of Salmonella genus bacterial typical colonies, at the dispersion and isolation stage (Table 4). For the analysis of the F2 bacteriological indicator, the soil-seeded plaques from Tomesti lagoon presented colonies typical for the *Salmonella* genus, at dispersion and isolation stage. Following the automatic identification analysis carried with the BIOLOG-OMNILOG system, the presence of the *Salmonella enterica* species was detected (Table 5). For the sewage sludge ecological rehabilitation, we propose the technological solution based on the planting of some rapidly growing trees from the *Populus, Salix* or *Paulownia* species to accelerate the crystallising process, drainage, installation of a normal plant growth regime and to begin a normal use regime of the muddy surface.

This will also solve the problem of excess humidity, which still exists at depths greater than 60 cm or more, and at the same time the technique shall decrease the sludge zinc content, that will be absorbed by plants absorbing from the sludge and will be removed from the system at the trees cutting stage (trees are to be used for other purposes).

Another way to recycle the sewage sludge would be to use it as a fertilizer on soils in the Central Moldavian Plateau, which, as a rule, have lower zinc contents and where frequently corn is deficient. In order to achieve this goal, experiments should be carried out in vegetation vessels or on field in order to establish the sludge doses that are to be spread on soils.

Table 3. Heavy metal content (total forms) of soil samples collected from the control well (F1) and from the drilling carried out in the sludge deposit (F2) compared to the limit values maximum allowed (LMA *) and with the values of the average soil content (CN **)

No.	*Identification*	*Depth*	*Zn*	*Cu*	*Fe*	*Mn*	*Pb*	*Ni*	*Cr*	*Co*	*Cd*
		cm	*ppm*								
1	F1-sample	0-20	127	10.2	22697	577	15	43.2	44.2	15.8	1.55
2		20-40	92	10.9	25977	591	22.5	35.5	53.1	29.2	0.2
3		40-60	84	13	26563	559	22.5	50.9	53.1	32.5	0.1
4		60-80	85	13 5	26680	356	15	54.8	62	32.5	0.25
5		80-100	103	14.5	30429	430	29.9	58.6	62	29.2	0.25
6		100-120	107	12.5	33358	462	22.5	62.5	48.6	22.5	3.75
7		120-140	100	10.9	31366	399	44.8	43.2	66.5	29.2	0.1
8		140-160	84	12	34412	624	29.9	77.9	39.7	29.2	0.95
9		160-180	126	16	37926	982	22.5	81.8	70.9	35.8	2.2
10		180-200	92	14.5	30663	845	15	77.9	44.2	29.2	0.4
11		200-220	74	13.2	26797	607	22.5	81.8	39.7	25.8	0.05
12		220-240	51	9.4	23283	1043	22.5	70.2	35.2	32.5	0.1
13		240-260	49	8.9	23048	1180	29.9	62.5	44.2	29.2	1.15
14		280-300	93	13	33592	913	37.3	77.9	48.6	52.5	2
15	F2	0-20	2534	28.7	27500	652	52.2	62.5	39.7	39.2	2.6
10		100-120	3236	31.2	30077	683	96.8	77.9	70.9	35.8	3.05
17		120-140	492	15.5	30429	779	44.8	66.4	44.2	39.2	0.75
18		140-160	187	9.7	27149	773	15	66.4	48.6	39.2	2.05
19		180-200	116	13	27851	611	15	58.6	35.2	42.5	0.45
20		240-260	113	12.7	30194	623	22.5	74.1	44.2	45.8	1.35
21		280-300	78	11.2	20588	844	22.5	58.6	39.7	39.2	0.4

Table 4. Quantitative microbiological analyses from control well F1

No.	***Test performed***	***U.M.***	***Sample symbol / Determined values***	***Test method***
			2151B	
1	Total coliform bacteria	UFc/100 cm^3	3x104	Guidebook ICIM cap. IV.2 POL 16 Ed.l. RO
2	Enterococi (*Streptococi fecali*)	UFC/IOO cm^3	1x104	Guidebook ICIM cap. IV.4 POL 16 Ed.l. RO
3	*Escherichia coli*	UFC/IOO cm^3	1x102	EPA 1680
4	*Clostridium perfringens*	UFC/IOO cm^3	21x105	Internal method
5	*Salmonella spp.*	UFC/100 cm^3	Abs	EPA 1682 POL 16 Ed.l. RO

Table 5. Quantitative microbiological tests from the F2 control drilling

No.	***Test performed***	***U.M.***	***Sample symbol / Determined values***	***Test method***
			2149B	
1	Total coliform bacteria	UFC/IOO cm^3	1733x104	Guidebook ICIM cap. IV.2 POL 16 Ed.l, RO
2	Enterococi (*Streptococi fecali*)	UFC/IOO cm^3	157x104	Guidebook ICIM cap. IV.4 POL 16 Ed.l, RO
3	*Escherichia coli*	UFC/IOO cm^3	228x102	EPA 1680
4	*Clostridium perfringens*	UFC/100 cm^3	32x105	Internal method
5	*Salmonella spp.*	UFC/IOO cm^3	Present	EPA 1682 POL 16 Ed.l, RO

4. Conclusions

The total concentration of heavy metals and salts in soils is a relevant indicator of the risk posed to human health and environment. Knowing the total heavy metals, macro-elements and salts loads in soils is the basis for justifying the remedial measures that are to be implemented for contaminated soils.

Following the carried analyses, we can say that some physical and chemical properties of the sludge have been transmitted to the soil on which the sludge is stored. Thus, the soil moisture below the sludge layer increased by up to 20%, compared to the humidity of the neighbouring soil that is not influenced by the presence of sludge.

Colloidal clay content up to 83% together with the liquid phase of the sludge constituted a geochemical barrier to the leaching, as regards the sludge chemical elements. It was also an adsorption medium for numerous cations and anions.

The contents of organic carbon, total nitrogen, and N-NH_4 in the first two soil horizons below the sludge lagoon increased 123 times compared to the contents of the same chemical elements present in the neighbouring soil, which is not influenced by the presence of the sludge. Likewise, the contents in mobile forms of P and K from the lagoon soil, in the first soil contact horizon were 7 times, respectively 1.3 times higher than the values in the upper horizons of the neighboring soil, taken as a witness.

Therefore, the sludge contributed to a significant increasing in the organic carbon, total nitrogen, mobile phosphorus and N-NH_4 loads in the first soil horizons, for soil on which the sludge is stored.

The sludge liquid phase led to the washing of the soluble salts from the first two soil horizons and consequently to the pH decreasing by up to 0.61 pH units, but the reaction domain remained the same: weak - alkaline.

From the group of tested heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn), the only element presents at polluting level is zinc. In the first soil horizon for the soil on which sludge is stored, the zinc contents exceed 10.8 times the maximum permissible limit value, and for the other this is 1.6 times.

From a bacteriological point of view, the soil beneath the sludge lagoon, is loaded with Salmonella genus bacterial colonies.

The research results can be used as a scientific basis for developing an adequate soil management system in the studied area.

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