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Agricultural Use of Urban Sewage Sludge from the Wastewater Station in the Municipality of Alexandria in Romania

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Abstract: Considering the increase in the amount of sewage sludge as a result of the development of urbanization, and the pollution of the environment through the storage of this sludge, the objective of this paper is to analyze the effects of the action of different doses of urban sewage sludge as a fertilizer for agricultural soil. Starting from the legislative recommendations regarding the use of urban sewage sludge in agriculture, we analyzed the sludge resulting from the sewage treatment plant in the Municipality of Alexandria, Romania. This sewage treatment plant, with two technological lines of water and sludge treatment, produces urban sewage sludge without pathogenic bacteria. We highlighted that by applying sewage sludge doses of 15 t/ha and 25 t/ha to agricultural soils, on which we planted soybean and wheat crops. We obtained high yields, and the concentration of heavy metals in the roots, stems and grains of soybean and wheat crops did not exceed the maximum standard limits allowed. Having a high content of organic matter and nutrients important for plants such as nitrogen, ammonium, potassium and zinc, sewage sludge has improved the fertility, physical, chemical and biological properties of the soil, and is able to be used as a fertilizer for degraded soils.

Keywords: soil; fertilizer; heavy metals; treatment plant; recovery; treatment; agriculture

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1. Introduction

In this paper, we analyze the possibility that, in accordance with the legislation of the European Economic Community (EEC) and of Romania, the sludge from municipal wastewater can be used effectively in agriculture as an organic fertilizer for acidic soils, without having a negative impact on the food chain and with beneficial results for human health and the environment. Through the use of sludge we aim to avoid the pollution of groundwater and surface water through the infiltration of pollutants that would be produced by sludge storage.

Water pollution affects human health, economic development and ecosystem functions. We must protect fresh water, which is an exhaustible resource essential for sustaining life, development and the environment. Currently, the development of urbanization has determined the increase in the amount of sludge from treatment plants [1]. Researchers are making efforts to be able to eliminate this sludge, without affecting the environment. One method of management is its use in agriculture as fertilizer, due to its content of the nutrients nitrogen, potassium, phosphorus, calcium, sulfur and magnesium [2–5]. The urban sewage sludge resulting from the wastewater treatment process contains essential nutrients [6–12] for plant growth [13,14]. This sludge, when applied to agricultural soil, preserves the reserves of phosphorous minerals on which the preparation of phosphate fertilizers is based, and improves the following soil characteristics: cationic exchange capacity, physico-chemical and biological properties, aeration, water retention capacity. It is therefore considered a true "biological sewage treatment plant" [15–17]. The use of urban sewage sludge in agriculture can be an alternative to the growing lack of chemical fertilizers [8,18,19] and to the continuous increase in their price against the background of an energy crisis. It can also be a complementary solution to the decrease in manure as a result of the decrease in the number of animals in the county, especially in conditions where the soil requires increased amounts of fertilizer to obtain high, stable and efficient yields [20]. Applying chemical fertilizers to the soil can degrade the soil, air and water. According to Marzougui et al. [18], the lack of chemical fertilizers has led to farmers using sewage sludge as organic fertilizer [21]. Agriculture practiced on acidic, heavy soils, with a chemical composition unfavorable to plants, reduces the profitability of farmers. The quality of the soil has been the attention of many researchers. Dexter [22] studied the physical quality of the soil, and determined that it should have good workability, good water infiltration, aeration and good rooting. On the other hand, the management of the sludge resulting from the processing of urban wastewater constitutes one of the priority problems of the water regime in large human communities, due to the large quantities that are processed.

In this context, in this paper we present the effect of using sludge from municipal wastewater treatment in Alexandria Municipality, Teleorman County, as a fertilizer on the soil-vegetation system.

Starting from the legislative recommendations regarding the use of sewage sludge in agriculture, in this paper we analyze the sludge resulting from the sewage treatment plant in the Municipality of Alexandria, Romania. The treatment plant has two treatment lines: the technological water treatment line and the sludge treatment line. It produces urban sewage sludge cleaned of pathogenic bacteria. We carried out experiments on the influence of certain doses of sludge on the content of heavy metals in the roots, stems and grains of soybean and wheat crops and found that the metal content does not exceed the maximum limits allowed by the legislation. We studied the impact of the development of these crops on the soils on which the sludge was applied and found that the total yield for two years in the variants fertilized with sludge increased compared to the non-fertilized variant. The yield of these crops increased, thanks to the micronutrients contained in the sludge. This sludge can be used as an amendment to degraded soils if it is applied in certain doses, to certain crops, in certain periods. Moreover, by using sewage sludge in agriculture, we have solved the problem of the increasing amount of sludge in sewage treatment plants due to urbanization and reduced the pressure on the environment, i.e., the negative impact on soil and water resulting from storage. Considering the aforementioned relationship, the energy crisis and long-term drought caused by climate change, we are forced to properly dispose of sewage sludge and apply urban water treatment technologies to be able to reuse these resources.

We analyzed the effect of the content of organic matter, including the forms of phosphorus and potassium, in the soil under the influence of the application of urban sewage sludge resulting from wastewater treatment.

Having a high content of organic matter and nutrients important for plants, such as nitrogen, ammonium, potassium and zinc, improving soil fertility, urban sewage sludge contributed to improving some physical, chemical and biological properties of the soil, including soil fertility, and is able to be used to condition agricultural soils. Over time, researchers have studied the effects of soil pollution with heavy metals [23–32]. In the case of the accidental pollution of agricultural land with heavy metals, the most convenient and effective cleaning method is phytoremediation. Phytoremediation means using plants to clean soil, water or air.

2. Legislative Recommendations Regarding the Use of Sewage Sludge in Agriculture

The use of sludge from urban sewage treatment plants in agriculture is regulated by Directive 86/278/EEC [33], transposed into national legislation by MAPPM Order no. 344/2004 [34] regarding the protection of the environment, especially of the soil, when sewage sludge is used in agriculture [35].

In the assortment of crops, plants that accumulate a large amount of heavy metals in consumable organs (for example, leaves, as in lettuce, spinach, cabbage, etc.) should be avoided.

The purpose of the regulations is to develop the agro-chemical potential of the sludge while preventing unwanted effects on the soil, water, crops, animals and people.

The EU Directive [33] requires that all treated sludge be subject to restrictions on use, which include periods of prohibition of grazing after application on pastures, and restrictions on sowing and harvesting of certain crops in certain periods of time after application. The national legislation through Order MAPPM no. 344/2004 [34] prohibits the use of sludge on pastures, fruit tree plantations and vegetable crops.

Romanian legislation requires that:

- The sludge should be mixed (incorporated) with the soil immediately after application. Through the incorporation process, any discomfort related to possible odors will be avoided. Properly treated sludge should not present an unpleasant odor [35];
- The application rate should not exceed the nutrient requirements of the crops. This represents an important environmental protection measure to avoid the excess of nutrients, especially nitrogen, which can infiltrate in the form of nitrates into the groundwater or surface waters;
- The pH of the soil must be maintained above the value of 6.5 [34]. The reason for this requirement is to restrict the possible assimilation of heavy metals by crops (the bioavailability of Zn, Ni and Cd increases in the conditions of an acid soil). The main concern of the Romanian regulations is the monitoring and control of contaminants, especially with heavy metals. The regulations establish the number of sludge samples that must be taken for analysis depending on the amount of sludge used in agriculture. The most frequently determined parameters are: pH, N, P, K, Ca and heavy metals. The speed with which the maximum limit values in the soil are reached is also important in the control of heavy metals.

The following factors should be taken into account when choosing suitable land for the management of urban sewage sludge from the urban wastewater treatment process:

- Weather conditions. It is recommended to spread in cloudy and windy weather and to avoiding scattering this type of fertilizer in rain, snow and strong sunlight, or if the ground is heavily frozen;
- The application period should be as early as possible in the crop growth period. It is recommended to spread the sludge in the spring and autumn, during the land preparation work for summer/autumn crops;
- Sludge is applied to agricultural soils with uniform topography. Consideration shall be given to the amount of eroded soil and the potential for entrainment of sludge or its decomposed compounds by surface water and groundwater. Land slope affects the rate and amount of surface runoff; for slurry applications, a slope of less than 5% is acceptable and a slope of less than 2% is considered adequate;
- Soils that are too hard or too loose, and soils that are deeply cracked or that have undergone subsoil work in the last 12 months should be avoided;
- Soil permeability influences the distribution of water on the soil profile; in its circuit, the water entrains the sewage sludge particles as well as the compounds resulting from its decomposition. A very high or a very low permeability are not suitable for soils intended for sewage sludge recycling;
- Soil drainage directly affects all physical, chemical and biological processes that occur in the soil. The mobility of various elements is affected by the effect on the redox potential. In poorly drained soils, residues undergo anaerobic decomposition, producing unoxidized primary compounds and intermediates, many of which are toxic to plants. In soils poorly permeable to water and air, and implicitly poorly drained, the rate of decomposition of organic matter is lower. Very poorly drained and over-drained lands should be excluded from the application of sewage sludge;

- It is not recommended to apply sewage sludge on agricultural soils affected by erosion, runoff processes, on floodplains [35];
- The usable water capacity at a depth of 0–100 cm or up to the limiting layer must be greater than 1400 m^3/ha. It is not recommended to apply sewage sludge on lands with a low water capacity;
- The protection of ground water (especially that used as a source of drinking water) against pollution with pathogenic agents, mineral elements (especially N-NO_3) resulting from the decomposition of sewage sludge, and heavy metals is one of the most restrictive factors in the application of sewage sludge. Lands where the groundwater depth is low are excluded from the application of sludge [34];
- Soil pH greatly affects the flux of heavy metals, increasing or decreasing their uptake by plants. Soils with a pH below 5.5 should be excluded from the application of uncomposted or non-calcareous sewage sludge, while those with a pH between 5.5–6.5 will be affected by increasing the pH above 6.5;
- Cation exchange capacity affects the mobility of heavy metals. It is forbidden to apply sewage sludge on soils with very low or very high cation exchange capacity [34–36];

When applying sewage sludge regarding the protection of water supply sources of localities, the following must be taken into account:

- At least 1500 m in front of water collection points for localities;
- More than 500 m from wells and localities that are supplied with water from the shallow water table;
- At least 500 m from localities;
- At least 100 m in front of rivers, lakes and ponds and protective measures will be taken against side leaks;
- At least 1000 m from tourist and leisure areas;

- Plants that accumulate a large amount of heavy metals in consumable organs (for example, leaves in the case of lettuce, spinach, cabbage, etc.) should be avoided in the assortment of crops.

The use of sewage sludge in agriculture can only be allowed under the provision of OM 344/2004, if the sewage sludge is properly treated so that it meets the specific quality standards, and only if the sewage sludge is applied to the land according to best practices, used for minimal-risk crops [34].

3. Materials and Methods

In this paper we analyzed the pre-dehydrated (aerobic, anaerobic and mesophilic stabilization) and dehydrated sewage sludge resulting from the treatment of urban wastewater from the city of Alexandria, Teleorman County, Romania (Figure 1).

Figure 1. Alexandria wastewater treatment plant.

Depending on the stage of wastewater treatment, several types of sewage sludge are generated, as follows: According to Tyagi and Lo [37], primary sewage sludge is produced from primary sewage treatment (flotation, precipitation and sedimentation), by separating solid from weights and fats from oils. Typically, primary sewage sludge contains 2% to 9% solids, of which approximately 60% to 80% are organic in nature; the remaining 90%, sometimes even more (99.5%), is water. This type of sewage sludge is difficult to dewater without treatment, being digested by aerobic or anaerobic bacteria. According to Devi and Saroha [38], secondary sewage sludge (activated sludge or biological sludge) is produced during biological treatment when microorganisms break down biodegradable organic content in wastewater.

The total solid concentration can vary between 0.8 and 3.3%, depending on the type of biological treatment process used [39], with the remainder being water. The solid part of the sludge consists of microorganisms that contain the organic fraction (between 59% and 88%) and inert materials. The organic fraction in secondary sludge contains: 50–55% carbon, 25–30% oxygen, 10–15% nitrogen, 6–10% organic hydrogen, 1–3% phosphorus and 0.5–1.5% sulfur [37].

Treatment is an important issue for municipal wastewater treatment plants due to its characteristics, high energy demand and treatment cost. Of the total operating costs of the sewage treatment plant, sludge removal processes account for 50–60% [40,41]. Due to these aspects, over the years, a large number of techniques (physical, chemical and biological) have been developed to treat or minimize sludge production [40]. Thickening is usually the first step in the sewage sludge treatment process.

In Figure 2, sewage sludge fermentation tanks and gasometer (methane tanks) are shown.

Figure 2. Sewage sludge fermentation tanks (methane tanks) and gasometer. tivated sewage sludge basin (sequential biological reactors).

Due to the possibility of co-digestion with organic waste, anaerobic digestion is considered an extremely profitable technology [42–44]. These bring economic benefits; revenues from sludge thickening are important because they affect the reliability and performance of the entire sludge treatment system [41]. After thickening, the sludge is stabilized. Among all stabilization methods, anaerobic digestion is the most commonly used because it involves eliminating pathogens, converting volatile substances into biogas (and then energy), as well as obtaining stable biosolids for mechanical dehydration [42,43]. It should be noted that organic waste sent for co-digestion produces volatile substances that are later transformed into biogas (and then energy), and also brings social benefits to the environment (diversion of waste from the landfill and the ability to recover materials) [45]. Aerobic digestion is another method of sludge stabilization which takes place in a fully aerated reactor (Figure 3) and is influenced by system temperature and retention time [46]. Figure 3 shows the biological reactors RB1, RB2 and RB3. In biological reactors, biological treatment takes place, involving sequential biological modification/purification reactions, which remove excess activated sludge [46].

Figure 3. Activated sewage sludge basin (sequential biological reactors).

Composting is also included in the category of sewage sludge stabilization methods. The composting processes involves the treatment and transformation of sewage sludge into a stabilized product, which can be used as an organic fertilizer or value-added product [42].

Many of the parasites in the sewage sludge survive aerobic and anaerobic stabilization procedures.

The content of pathogens resulting from the dehydration process depends very much on the choice of the most suitable type of polymer (in our case, the polymer is ferric chloride) for sewage sludge flocculation, on ensuring the optimal dissolution conditions and on optimizing the dosage. In this phase, organic matter is stabilized, and the amount of pathogenic microbes is significantly reduced.

The fundamental reduction/elimination of pathogens is influenced by:

- Ammonia and detergents that strongly contribute to the inactivity of viruses;
- The dehydration of the sewage sludge, its thickening, and the decrease in its humidity greatly influences the inactivity of the parasites, as with viruses;
- Sewage sludge irradiation with an electron accelerator, or with a ray source. This method is particularly effective for eliminating pathogenic organisms;
- Using the prismatic composting system, in which a temperature of 60–70 °C is ensured for approximately 50 days, leads to total inactivity of viruses.

Urban sewage sludge is made by artificially dewatering the sludge through dynamic processes (centrifugation in the presented case). The main advantages of the artificial dewatering processes are: the short duration of the process, the small area required for machinery, and the lack of influence of inhibitors on the dewatering processes. After dewatering, the sludge is transported to the final storage place or directly to agricultural utilization. There are two main periods during the year when sewage sludge can be applied to the land: spring and autumn, to correlate with the processes of sowing and harvesting crops, respectively. It is necessary to store the sewage sludge during the period when incorporation into the soil is not recommended. Figure 4 shows the technological dehydration by centrifugation. Sludge processing by centrifugation has a much better yield than pre-dehydration technology. In this sludge treatment stage, the dehydrated sludge can have a moisture content between 70-80%, depending on the needs (e.g., season, stationary time in the warehouse).

The amount of sludge generated in the wastewater treatment plant in the Alexandria municipality, Teleorman County is approximately 21.8 m^3/day [45]. Research carried out in the period 2014–2021 recommends the use of urban sewage sludge in agriculture due to the lack of chemical fertilizers and the continuous increase in their price against the background of the energy crisis, considering that the soil requires a large amount of fertilizer to achieve high, stable and efficient production. There are two main periods during the year when sewage sludge can be applied to the land: spring and autumn, to correlate with the processes of sowing and harvesting crops, respectively. Therefore, it is necessary to store the sludge during the period when it is not recommended to incorporated into the soil.

Figure 4. Sewage sludge thickening room (mechanical concentration) and space for the storage of dehydrated sewage sludge.

4. Results

4.1. Economic Companies That Pollute Water

Next, we present some of the existing economic agents in Alexandria taking into account the possibility that their waste water enters the urban installations for deep biological purification. Only Koyo România S.A., Iaica S.A., Germino S.A., Electrotel S.A. and Time International Trading SRL are industrial economic agents in the true sense of the word, if we refer to the volume of discharged wastewater and consider that the minimum criterion is 10,000 cubic meters/year. SC Koyo Romania SA is part of the machinery and equipment manufacturing industry, producing bearings, gears and mechanical transmission parts, known as the bearing factory. At present, it is the only potentially significant industrial polluter in Alexandria in the event of a pollution incident.

SC Koyo Romania S.A. is equipped with an efficient emulsion treatment station, and for the resulting deposits it has a contract with a specialized company which takes over and neutralizes deposits.

The Oxygen Factory remains in operation. It does not cause pollution problems except sometimes for a single parameter, namely pH. Monitoring of the Oxygen Factory monitoring is done quarterly.

Electrotel S.A. manufactures electrical equipment (heaters, stoves, etc.); the waste water loads of Electrotel S.A are below the limits allowed by the discharge notice, with the limits for some parameters being more drastic than those in NTPA 002 [46]. The company does not have any pre-purification facility.

Time International Trading SRL is a clothing unit, with a volume of used water over 50,000 cubic meters (information from the Questionnaire). It exceeds the limits of the discharge notice for CCO-Cr, CBO5 and ammoniacal nitrogen.

In the following are presented some of the commercial companies that, although they discharge volumes of water below 10,000 m^3, have presented systematic excesses in some of the monitored indicators.

FSB Prestibe SRL is a production unit that Teflon-coats vessels and that periodically has non-compliances regarding dissolved oxygen and exceedances regarding CCO-Cr and ammoniacal nitrogen.

FSB Prestibe SRL is equipped with a settling basin and has a new phosphorus removal line. The loads of waste water discharged by the other economic agents monitored by APA SERV are generally within the limit values set by the contract and do not raise pollution problems for SEAU Alexandria. These are generally units in the field of services, mainly laundromats and car services.

4.2. Utilization of Urban Sewage Sludge in Agriculture

In order to test the effect that urban sewage sludge has on plant growth and changes in soil characteristics, experiments were carried out according to the pedological and agrochemical study of the Teleorman Pedological and Agrochemical Studies Bureau in accordance with standard legislation. Urban sewage sludge represents the agrochemical measure that positively influences the humus regime in the soil [2]. Together with the vegetable remains left in the soil from the previous crops, it is both the source of raw material for the nutritious humus, and for the synthesis of the stable humus. Both of them contribute, along with other links to plant culture technologies, to maintaining the humus content of the cultivated soils. Under the ratio of the fertilizing effect, urban sewage sludge is valuable. It brings to the soil important quantities of all the essential elements of plant nutrition, in balanced ratios compared to their requirements. The ameliorative effect on the soil is due to the appreciable supply of organic matter [2]. At the same time, urban sewage sludge increases the buffering capacity of the soil, reduces the harmful action of soil acids, and reduces acidity by forming NH and by increasing the amount of exchangeable calcium. It improves the soil's physical properties, structure, permeability, water retention capacity, and determines important changes to it.

We experimented with two doses of sludge incorporation calculated from the chemical composition of sewage sludge according to the analytical data in Table 1 with a C:N Ratio of 15:1. The urban sewage sludge was applied with the machine manure spreader (MIG) (Figure 5) in the fall before plowing.

Figure 5. Loading the Manure spreading machine (MIG) with urban sewage sludge and spreading it. tivated sewage sludge basin (sequential biological reactors).

The soybean and winter wheat crops benefited from the direct effect of the sludge application in the second year as well when the C:N ratio was 25:1, a decrease in yield was observed after the second year in which the crops were also inverted. The analytical data obtained and reproduced in Table 1 highlight the following aspects:

- The sludge resulting from the purification of urban sewage waste water from the city of Alexandria, Teleorman County can be considered an organic fertilizer due to the content of mineral nitrogen (nitrogen and ammonium ions) directly accessible to plants, the content of potassium and zinc, and the range of variation in organic carbon content;

- The content of bioaccessible heavy metals present in the activated sludge, resulting from the purification of urban sewage wastewater from the Municipality of Alexandria, Teleorman, is below the permitted limits (Table 1).

The sludge resulting from urban sewage wastewater treatment can be used in agriculture as an improver of physico-chemical properties (reaction properties and fertility, etc.) and as an organic fertilizer for acidic soils, being an ecological alternative to chemical fertilizers [47].

Table 1. The physico-chemical composition of the sludge, resulting from the purification of urban sewage waste water from the Municipality of Alexandria, Teleorman and of the soil from the perimeter studied.

Parameter	Soil	Sewage Sludge
Sand (%)	58.8	-
Silt (%)	14.2	-
Clay (%)	6.2	-
Absorbable halogenated organic compounds (AOX)	30	283
Total phosphorus as P	0.20	0.974
Moisture	0.10	40.9
Losses by calcination at 55 °C	-	34
pH (H_2O)	7.02	6.72
Total nitrogen (%)	0.08	728
Total organic carbon(%)	0.40	16.6
Cadmium	0.47	1.15
Cobalt	0.0330	<8.00
Chromium	2.70	26.9
Copper	19.8	238
Nickel	2.65	24.7
Potassium	0.16	3290
Zinc	1.77	485
Arsenic	0.667	3.19
Cadmium	1.15	1.15
Mercury	0.001	0.588

To determine the values presented in Table 1, we performed the following test in the laboratory:

A total of 12–15 individual soil samples were collected, of which only five were analyzed. In the laboratory, they were analyzed physico-chemically as follows:

- Soil reaction (pH) was determined in aqueous suspension;
- To measure (pH) and electrical conductivity (EC) we prepared a suspension of soil or sewage sludge sample in distilled water in a ratio of 1:5 (*w/v*). We measured pH and electrical conductivity (EC) with a pH meter and EC meter.
- The assimilated phosphorus content was measured in the ammonium acetate–lactate extract determined using Kjeldhal method [48];
- The assimilated potassium content was measured using photometry in the ammonium acetate–lactate extract using a the method of Jackson [48];

- The humus content was determined by the Walkley–Black method; the principle is based on determining the organic carbon in the soil by oxidizing the carbon with acid dichromate [49];
- The total organic carbon (TOC) was estimated using the potassium dichromate oxidation titrimetric method [50];
- For heavy metal quantification, soil and sewage sludge samples were digested using a mixture of HCl and HNO_3 (4:1 *v/v*). Heavy metal concentration was determined using the method of McGrath and Cunliffe [51], using an atomic absorption spectrophotometer (GBC 932 plus). Bio-available forms of different heavy metals were quantified using extraction with DTPA as suggested by Lindsay and Norvell [52].

It can be seen according to the data in Table 2 that, at a higher concentration of sludge applied to the soil of 25 t/ha compared to 15 t/ha, the content of heavy metals increases slightly, but does not exceed the standard limit.

Table 2. The influence of different doses of sludge on the content of heavy metals in the stems, roots and grains of soybean plants.

Sludge Dose	Pb	Cd	Cu	Ni	Zn
Roots					
Simple soil test	1.9	0.10	12.4	11.2	22.2
Urban sewage sludge (15 t/ha)	2.2	0.12	13.9	12.3	23.1
Urban sewage sludge (25 t/ha)	2.44	0.13	14.2	13.7	24
Strains					
Simple soil test	2.101	0.09	6.4	5.2	16.2
Urban sewage sludge (15 t/ha)	2.308	0.11	6.7	5.8	16.8
Urban sewage sludge (25 t/ha)	2.502	0.12	6.9	6.1	17.1
Grains					
Simple soil test	0.95	0.08	6.3	5.1	16.8
Urban sewage sludge (15 t/ha)	1.1	0.10	6.5	5.4	17
Urban sewage sludge (25 t/ha)	1.3	0.11	6.8	5.6	17.2
CMA	3–15	10	15–20	30	200

Note: CMA (Concentration maximum admissible).

The chemical analyses performed on soybean plants (stems, roots) and grains (Table 2) highlighted the following aspects:

- In all experimental variants, soybean stems and roots in the variants fertilized with sludge had a slightly higher lead content than in the control variant but below phytotoxic levels, and soybeans had a lead content within normal limits in all variants;
- The cadmium content in soybean stems recorded values below normal limits. In soybean roots they were higher than in stems, but below the limits of the toxicity threshold. In soybeans the lower level of cadmium was 0.11 mg/kg s.u. in the variant treated with 15 t/ha, and 0.12 mg/kg in the variant treated with 25 t/ha of sludge;
- The concentration of Ni, Cu and Zn in stems, roots and grains was far below the toxicity level in all organs of the soybean plant regardless of the dose applied. However, it

is necessary to mention the fact that with the increase in the doses of fertilizer, there was a slight tendency to accumulate heavy metals in all the organs of the plants, which requires a rigorous control of the amounts of sludge applied.

The field tests of dewatered urban sewage sludge showed that in the first year of application of the sludge, the soybean crop increased significantly by 450 kg after the dose of 15 t/ha, and 750 kg /ha after the dose of 25 t/ha (Table 3) [39].

Table 3. The influence of the sludge from the wastewater treatment process in the Municipality of Alexandria, Teleorman on the harvest of soybeans and autumn wheat kg/ha.

Variant of Experience	Variant of Experience			The Second Year of Action at the Culture of Autumn Wheat			Total Harvest for 2 Years in Grain Units	The Increase in the Total Harvest Over 2 Years in Grain Units	
	Harvest	The Crop Growth		Harvest	The Crop Growth		Harvest	The Crop Growth	
		kg/ha	%		kg/ha	%		kg/ha	%
Simple soil test	2600	-	-	4100	-	-	6700	-	-
Urban sewage sludge (15 t/ha)	3050	450	15	5200	1100	21	8250	1550	19
Urban sewage sludge (25 t/ha)	3350	750	22	5000	720	26.25	8706	2115	24

In the second year of the experiment, the autumn wheat harvest increased considerably. At the dose of 15 t/ha, grain production was 21% higher than in the unfertilized control variant. On the plots where 25 t/ha of sludge was applied, the yield increase was 26.25%, i.e., 1.25 times more than in the variant treated with 15 t/ha. (An important factor was also the supply of water from the soil).

Table 3 shows the influence of sludge from the wastewater treatment process in the Municipality of Alexandria, Teleorman on the harvest of soybeans and winter wheat in kg/ha.

The productivity of soybean and wheat agricultural crops to which urban sewage sludge was applied reflects the state of the nutrient regime of the soil in relation to the water intake from the soil (Table 4).

Table 4. The influence of the amount of precipitation in the first and in the second year for soybean and autumn wheat agricultural crops.

The Year	Sep—Mar		April		May		June		July		August		Agricultural Year	
	mm	%	mm	%	mm	%	mm	%	mm	%	mm	%	mm	%
First year	324	126	39	93	33	42	37	61	10	19	15	25	458	84
Second year	153	60	38	90	114	215	48	61	59	97	22	36	434	80

Because of its relatively high content of organic matter (Table 5) and nutrients, urban sewage sludge contributed to the improvement of some physical, chemical and biological properties of the soil. We find that the total harvest over two years in the variants fertilized with sludge was 8250–8350 kg/ha compared to 6700 t/ha in the non-fertilized variant. The specific yield increase from 1 ton of sludge was 103.3 kg grain units at the dose of 15 t/ha and 84.6 kg/t at the 25 t/ha. Small doses of 12–18 t/ha of sludge will be more efficient both agronomically and economically, as smaller amounts of sludge will be used per unit of land.

Table 5. The effect of the content of organic matter, the forms of phosphorus and potassium from the soil under the influence of the application of urban sewage sludge resulting from the treatment of wastewater from the Municipality of Alexandria, Teleorman.

Variant of Experiment	Organic Matter, %		P_2O_5 ppm *		K_2O ppm *	
Entry 1	The Content	The Difference	The Content	The Difference	The Content	The Difference
Simple soil test	3.1	-	22.5	-	190	-
Urban sewage sludge (15 t/ha)	3.25	0.15	32.1	9.6	260	70
Urban sewage sludge (25 t/ha)	3.35	0.25	38.2	15.7	280	90

Note: * ppm—mg/kgsol.

The effect of using urban sewage sludge in agriculture contributes to the improvement of some physical, chemical and biological properties of soils as follows:

It maintains the balance between humification and mineralization, between organic and mineral fertilizers [1].

It increases the water capacity of the soil, increases the buffering capacity of the soil, decreases the harmful action of soil acids and reduces acidity through the formation of ammonia, and increases the amount of exchangeable calcium [2];

It improves the physical properties, namely: the structure of coarse-textured soils, the structure of fine-textured soils, the water holding capacity in coarse-textured soils [14];

It intensifies the activity of microorganisms and stimulates plant growth, produces heat through the decomposition of organic substances, releases large amounts of carbon dioxide—the main factor responsible for soil reaction—and increases the amount of humus;

The process of mineralization of soil organic matter, which brings nitrogen into the soil in mineral form, sometimes occurs at low rates [53]. During this time there are considerable losses of nitrogen in gaseous form. In the transformed organic matter, 90% of the nitrogen is bound in the form of organic compounds, a fact that represents a great advantage, constituting an important source of nitrogen [14]. Within the limits of the established legalities, the process of transformation of organic matter incorporated with urban sewage sludge, was manifested. A significant increase, by 0.15–0.25% of the soil mass, was observed in the content of organic matter in the arable layer in the fertilized variants. The content of mobile phosphorus increased, compared to the unfertilized version, by 9.6–15.7 ppm, and that of exchangeable potassium by 70–90 ppm [54]. These increases in plant-accessible phosphorus and potassium are due not only to the amount of sludge applied, but also to the solubilizing influence of the sludge on soil minerals containing phosphorus and potassium [6].

For the values presented above, the determinations were made in the laboratory:

We collected 10–12 individual soil samples under the influence of sewage sludge application, of which we analyzed only five soil samples.

In the laboratory, they were analyzed as follows:

- We measured the assimilated mobile phosphorus content in the ammonium acetate–lactate extract determined by the Kjeldhal method [48];
- The assimilated mobile potassium content was measured photometrically in the ammonium acetate–lactate extract using the Jackson method [48];
- The humus content was determined by the Walkley–Black method.

Heavy metals in the soil (total forms) underwent few changes [55]. Compared to the non-fertilized variant, a slight increase in the content of Cd, Cu, Cr was observed (Table 6), but their concentration does not exceed the maximum limits allowed in the soil [31].

Table 6. The variation of the content of heavy metals (total forms) in the soil fertilized with urban sewage sludge from the wastewater treatment process in the Municipality of Alexandria, Teleorman [34,35].

Type of Sewage Sludge	Cd	Cu	Ni	Pb	Cr	Zn
Sample sludge	0	10	12	18	21	28
Urban sewage sludge (15 t/ha)	0.5	12	13	20	23	29
Urban sewage sludge (25 t/ha)	0.6	13	14	19	26	30
[34]	1–3	50–140	30–75	50–300	-	150–300
[35]	3	50	30	50	100	150

Note: Zn, Ni and Cd increases in the conditions of an acid soil.

Figure 6 shows the pH values at which each element is bioavailable. A high acidity of the soil reduces the absorption of Mo, and blockages in the absorption of Mn, Zn or B can occur with an inadequate application of calcium amendments (to correct the pH of a soil). The diagram also demonstrates that the assimilation of elements is most optimal in the case of a pH between 6 and 7 (in our case the pH of the sludge is 6.72, as can be seen in Table 1) [56]. As can be seen from the diagram, the toxicity of aluminum is controlled by a pH level of 5.5.

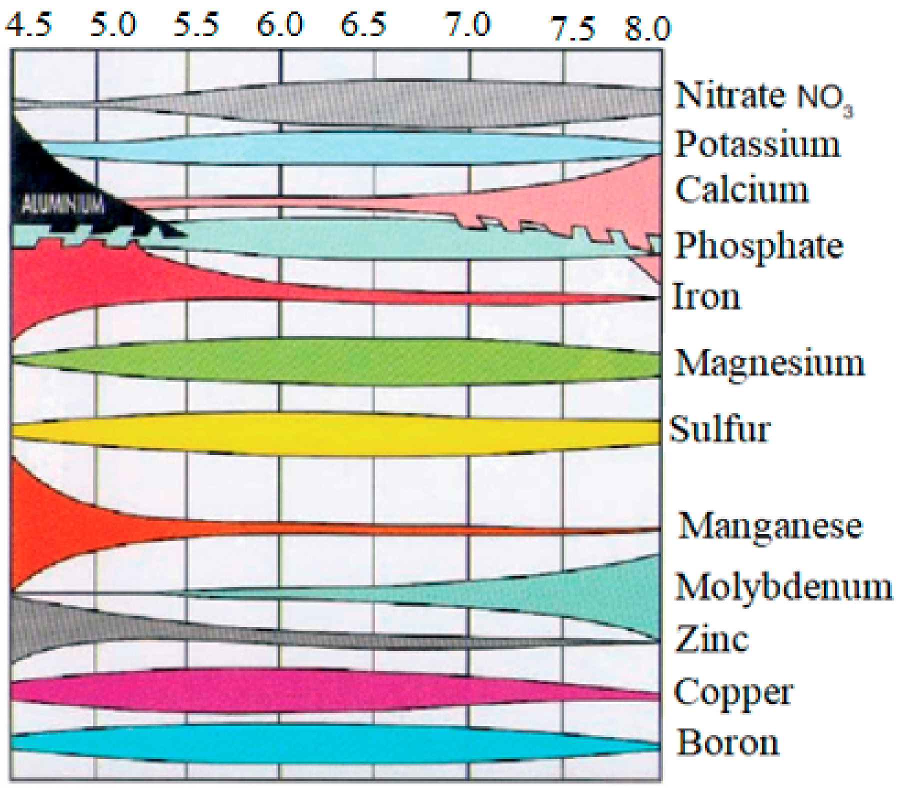

Figure 6. Assimilation of nutrients by plants depending on soil pH (adapted from [57]).

In the case of acidic soils that maintain heavy metals in a form accessible to plants as a result of high solubility at pH < 6, it is recommended to limit the individual contamination indices for the Pb—Cd—Ni system, with an impact on heavy acidic soils, to a total value of maximum 1, [56,58]:

$$Pb/75 + Cd/2 + Ni/75 \leq 1 \quad (1)$$

Cereals and technical plants can be grown on soils improved with sludge; the analytical data obtained on their chemical composition indicated that the maximum allowed values are not exceeded.

To evaluate the level of phytotoxic contamination/pollution with heavy metals of plants grown on soils improved with biosolids, we propose the use of a synthetic index, rendered mathematically in the form [56,58]:

$$I_p = 2\ (Cu/30) + Ni/50 + Zn/140 \quad (2)$$

$$Ip = (Cu/30)2 + (Ni/50) + (Zn/140) \quad (3)$$

4.3. The Biological Properties of the Soil on Which the Sewage Sludge Was Applied

The use of urban sewage sludge for the fertilization of agricultural land positively influences the humus regime, brings to the soil important quantities of essential elements for plant nutrition, has an ameliorating effect on the soil, increases the buffering capacity of the soil, reduces the harmful action of soil acids, improves the physical properties of the soil, its structure, permeability, water retention capacity, and causes important changes in the aerohydric regime.

According to the data presented in the tables below, the physicochemical properties of all treatments were routinely quantified on days 0–90 using standard protocols. Microbial biomass (C) and microbial biomass nitrogen (N) content of samples were estimated using chloroform fumigation extraction [59]. The difference between extractable C and N fumigated and non-fumigated extracts were converted to microbial biomass (C) and microbial biomass (N) using a K_c factor of 0.45 and a K_n factor of 0.54 [57]. Dehydrogenase (DHA) activity in different treatments was determined by the reduction of 2,3,5-triphenyltetrazolium chloride to triphenylformazan according to the method of Casida et al. [57]. Urease activity (UA) was measured as $NH_4{}^+$–N released per gram of soil per hour by incubating 5.0 g of wet soil samples with 0.2 M urea substrate at 37 °C [60].

Alkaline phosphatase (APA) was determined by measuring the rate of p-nitrophenol (PNP) formation during incubation of the soil sample with disodium p-nitrophenyl phosphate solution for 1 h at 37 °C [61].

Carbon mineralization was determined by measuring CO_2 evolution during the 4 weeks of incubation according to the method of Pramer and Schmidt [62] with slight modifications. CO_2 was trapped in a NaOH solution (0.5 N) and titrated against 0.5 HCl after the addition of freshly prepared saturated $BaCl_2$. Potentially mineralizable nitrogen was determined by extracting 10 g of a soil sample using 100 mL of 2M, KCl before and after the 90-day incubation period at 30^0 C with 60% water holding capacity. Inorganic nitrogen (NO^--N and $NH_4{}^+$-N) was determined using steam distillation using MgO and the net mineralization of nitrogen and alloy was calculated by subtracting the initial inorganic N values from that were obtained after 90 days [6,63,64].

Table 7 shows the chemical properties of the modified soil with different sewage sludge concentrations.

It can be observed that by amending the soil with amounts of urban sewage sludge of approx. 15–25 t/ha, the chemical properties of the soil change.

Table 8 shows the ratio between the microbial biomass and the microbial coefficient of the soil modified with different concentrations of sewage sludge.

It can be seen that the microbial coefficient of the soil changes depending on the sewage sludge concentration (Table 9).

Table 7. Chemical properties of soil modified with different concentrations of sewage sludge.

Treatments	pH *		Organic C ** (%)		Nitrogen *** (%)	
Days	0	90	0	90	0	90
Sample sludge—C	7.02	6.9	0.40	0.38	0.08	0.09
Urban sewage sludge (15 t/ha)	7.15	7.09	0.70	0.63	0.101	0.105
Urban sewage sludge (25 t/ha)	6.80	6.68	0.91	0.84	0.121	0.126

Note: Critical Difference (CD) for treatments (A) = 0.108 *. 0.02 **, CD for days (B) = 0.062 *. 0.09 **, CD for factor A × B = 0.153 *. 0.03 *** [6]. * The pH of the soil to which the sludge was added; ** The concentration of soil organic carbon; *** Nitrogen concentration.

Table 8. Ratio of microbial biomass C (C_{mic}) to organic C (C_{org}) or microbial quotient of soil amended with different concentration of sewage sludge.

Treatments	C (C_{mic})/C (C_{org}) (%)	
Days	0	90
Sample sludge—C	3.90	3.78
Urban sewage sludge (15 t/ha)	4.20	4.11
Urban sewage sludge (25 t/ha)	4.63	4.45

Note: Critical Difference (CD) for treatments (A) = 0.201, CD for days (B) = 0.17. CD for factor A × E = 0. 153 [6].

Table 9. Basal respiration in soil determined as gross flux of CO_2, amended with different concentration of sewage sludge during different time intervals.

Treatments	CO_2-C (mg 100g^{-1} Soil Incubation Period)						Total Carbon Mineralization mg (mg kg^{-1} Soil)
Days	0	0–5	5–15	15–30	30–60	60–90	
Sample sludge	4.1	9.1	8.5	8.9	8.6	8.1	47.3
Urban sewage sludge (15 t/ha)	11.2	18.9	17.3	17.1	15.8	15.9	96.2
Urban sewage sludge (25 t/ha)	12.2	2.1.7	19.3	19.1	18.4	17.3	108

Note: Critical Difference (CD) for treatments (A) = 0.025, CD for days (B) = 0.025, CD for factor A × B 0.092, $CO_2 - C$= mass of CO_2-carbon generated (mg) [6].

It can be seen that using different sewage sludge concentrations in different time intervals changes the base respiration parameters in the soil determined as CO_2 flow.

Soil microbial biomass represents the amount of organic matter in the soil; in this case the control sample (C) and different doses of sludge (N) of 15–25 t/ha over a certain period of time (days) were studied as the transfer factor. It should be emphasized that sewage sludge stimulates microbial populations and their activities, as most microbial populations are heterotrophic and respond quickly to new changes. In particular, the high concentration of organic matter in sewage sludge provides a potential source of energy for various heterotrophic microorganisms, thus leading to higher values of microbial biomass in the control samples during the first days of culture, but over time the readily available carbon was consumed, which resulted in lower microbial biomass in the control samples, as shown in Table 8. As shown in Table 6, sewage sludge with low concentrations of heavy metals will improve the quality of organic matter and therefore will not have a negative impact on microbial biomass [63]. Soil hydrolytic enzymes, namely alkaline phosphatase, dehydrogenase, urease and glucosidase, enhance the degradation and stabilization of various soil organic matrices and affect the availability of essential nutrients for plants and microorganisms [65]. Soil enzyme activities indicate the impact of environmental factors on various microbial functions [65]. Substantial changes in various enzyme activities were observed at different incubation periods with increasing sewage sludge application rates. The addition of sewage sludge increases soil organic matter and serves as a source of macronutrients and available organic carbon that contributes to higher enzyme activities [65]. Dehydrogenase activity is an indicator of soil biological activity. Dehydrogenase is part of microbial oxidoreductase metabolism and its activity can be used to represent the potential of soil to support various biochemical reactions that are crucial for maintaining soil health and fertility [65].

5. Conclusions

In this paper, we have researched the use of urban sewage sludge as a fertilizer for degraded soils over two years. We added doses of sludge of 15–25 t/ha, equivalent to a nitrogen content of 62 and 103 kg/ha, respectively, to the agricultural soil on which we planted soybean and wheat crops. The concentration of heavy metals did not exceed the maximum permissible limits in the soil, roots, stems and grains, and the yield of these crops increased. Sewage sludge applied to agricultural soil has improved the physical, chemical and biological properties of the soil and can be used as a fertilizer for degraded soils.

This indicates that urban sewage sludge in doses greater than 25 t/ha can be used on compact soils with sufficient moisture. For average soils, lower doses (15–20 t/ha) are indicated, because nitrification is more intense, and part of the nitrogen is lost.

At the same time, soil improvement with sewage sludge does not pollute the soil or plants with heavy metals, nitrites or nitrates, if the following conditions are met: its content in heavy metals and nitrogen is according to standard legislation, the optimal dose, the period of application, and the quality of the sludge is high in organic matter and important nutrients for plants such as: nitrogen, ammonium, potassium and zinc.

Moreover, by using sewage sludge in agriculture, we have solved the problem of increasing the amount of sludge in sewage treatment plants due to urbanization and reduced the negative impact on soil and water resulting from storage. The energy crisis and prolonged drought caused by climate change force us to properly dispose of municipal sludge and to apply urban water purification technologies to be able to reuse these resources.

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References

1. Rusănescu, C.O.; Rusănescu, M.; Constantin, G.A. Wastewater Management in Agriculture. *Water* **2022**, *14*, 3351. [CrossRef]
2. Rusănescu, C.O.; Rusănescu, M.; Voicu, G.; Paraschiv, G.; Biriș, S.Ș.; Popescu, I.N. The Recovery of Vermicompost Sewage Sludge in Agriculture. *Agronomy* **2022**, *12*, 2653. [CrossRef]
3. Usman, K.; Khan, S.; Ghulam, S.; Khan, M.U.; Khan, N.; Khan, M.A.; Khalil, S.K. Sewage sludge: An important biological resource for sustainable agriculture and its environmental implications. *Am. J. Plant Sci.* **2012**, *3*, 1708–1721. [CrossRef]
4. Angin, I.; Aslantas, R.; Gunes, A.; Kose, M.; Ozkan, G. Effects of sewage sludge amendment on some soil properties, growth, yield and nutrient content of raspberry (*Rubus idaeus* L.). *Erwerbs-Obstbau* **2017**, *59*, 93–99. [CrossRef]
5. Angin, I.; Yaganoglu, A.V. Effects of sewage sludge application on yield, yield parameters and heavy metal content of barley grown under arid climatic conditions. *Int. J. Agric. Biol.* **2012**, *14*, 811–815.

6. Dhanker, R.; Chaudhary, S.; Goyal, S.; Garg, V.K. Influence of urban sewage sludge amendment on agricultural soil parameters. *Environ. Technol. Innov.* **2021**, *23*, 101642. [CrossRef]
7. Hamdi, H.; Hechmi, S.; Khelil, M.N.; Zoghlamia, I.R.; Benzartic, S.; Mokni-Tlilia, S.; Hassen, A.; Jedidi, N. Repetitive land application of urban sewage sludge: Effect of amendment rates and soil texture on fertility and degradation parameters. *Catena* **2019**, *172*, 11–20. [CrossRef]
8. Macdonald, C.A.; Clark, I.M.; Zhao, F.J.; Hirsch, P.R.; Singh, B.K.; McGrath, S.P. Longterm impacts of zinc and copper enriched sewage sludge additions on bacterial, archaeal and fungal communities in arable and grassland soils. *Soil Biol. Biochem.* **2011**, *43*, 932–941. [CrossRef]
9. Werle, S.; Wilk, R. A review of methods for the thermal utilization of sewage sludge: The Polish perspective. *Renew. Energy* **2010**, *35*, 1914–1919. [CrossRef]
10. Kumar, V.; Chopra, A.K.; Kumar, A. A review on sewage sludge (Biosolids) a resource for sustainable agriculture. *Arch. Agric. Environ. Sci.* **2017**, *2*, 340–347. [CrossRef]
11. Kehrein, P.; van Loosdrecht, M.; Osseweijer, P.; Garfí, M.; Dewulf, J.; Posada, J. A critical review of resource recovery from municipal wastewater treatment plants—Market supply potentials, technologies and bottlenecks. *Environ. Sci. Water Res. Technol.* **2020**, *6*, 877–910. [CrossRef]
12. Shah, M.; Hashmi, H.N.; Ali, A.; Ghumman, A.R. Performance assessment of aquatic macrophytes for treatment of municipal wastewater. *J. Environ. Health Sci. Eng.* **2014**, *12*, 106. [CrossRef]
13. Chen, H.; Yan, S.H.; Ye, Z.L.; Meng, H.J.; Zhu, Y.G. Utilization of urban sewage sludge: Chinese perspectives. *Env. Sci Pollut Res* **2012**, *19*, 1454–1463. [CrossRef]
14. Rusănescu, C.O.; Voicu, G.; Paraschiv, G.; Begea, M.; Purdea, L.; Petre, I.C.; Stoian, E.V. Recovery of Sewage Sludge in the Cement Industry. *Energies* **2022**, *15*, 2664. [CrossRef]
15. Purdea, L.; Rusănescu, C.O.; Tucureanu, M.C. Alternative for the Use of Sewage Sludge in Romania. *Rev. Chim.* **2019**, *70*, 1967–1970. [CrossRef]
16. Rusănescu, C.O.; Rusănescu, M.; Jinescu, C.; Durbacă, I. Recovery of Treated Sludge. *Rev. Chim.* **2019**, *70*, 3477–3481. [CrossRef]
17. Zhang, Q.H.; Yang, W.N.; Ngo, H.H.; Guo, W.S.; Jin, P.K.; Dzakpasu, M.; Yang, S.J.; Wang, Q.; Wang, X.C.; Ao, D. Current status of urban wastewater treatment plants in China. *Environ. Int.* **2016**, *92–93*, 11–22. [CrossRef] [PubMed]
18. Marzougui, N.; Ounalli, N.; Sabbahi, S.; Fezzani, T.; Abidi, F.; Jebari, S.; Melki, S.; Berndtsson, R.; Oueslati, W. How Can Sewage Sludge Use in Sustainable Tunisian Agriculture Be Increased? *Sustainability* **2022**, *14*, 13722. [CrossRef]
19. Savci, S. An Agricultural Pollutant: Chemical Fertilizer. *Int. J. Environ. Sci. Dev.* **2012**, *3*, 77–80. [CrossRef]
20. Sharma, B.; Vaish, B.; Singh, U.K.; Singh, P.; Singh, R.P. Recycling of Organic Wastes in Agriculture: An Environmental Perspective. *Int. J. Environ. Res.* **2019**, *13*, 409–429. [CrossRef]
21. Diacono, M.; Montemurro, F. Long-Term Effects of Organic Amendments on Soil Fertility. *Sustain. Agric.* **2011**, *2*, 761–786.
22. Dexter, A.R. Soil physical quality: Part I. Theory, effects of soil texture, density, and organic matter, and effects on root growth. *Geoderma* **2004**, *120*, 3–4, 201–214. [CrossRef]
23. Golia, E.E.; Angelaki, A.; Giannoulis, K.D.; Skoufogianni, E.; Bartzialis, D.; Cavalaris, C.; Vleioras, S. Evaluation of soil properties, irrigation and solid waste application levels on Cu and Zn uptake by industrial hemp. *Agron. Res.* **2021**, *19*, 1–92.
24. Angelaki, A.; Dionysidis, A.; Sihag, P.; Golia, E. Assessment of Contamination Management Caused by Copper and Zinc Cations Leaching and Their Impact on the Hydraulic Properties of a Sandy and a Loamy Clay Soil. *Land* **2022**, *11*, 290. [CrossRef]
25. Bourioug, M.; Alaoui-Sossé, L.; Laffray, X.; Raouf, N.; Benbrahim, M.; Badot, P.M.; Alaoui-Sossé, B. Evaluation of sewage sludge effects on soil properties, plant growth, mineral nutrition state, and heavy metal distribution in European larch seedlings (*Larix decid*). *Arab. J. Sci. Eng.* **2014**, *39*, 5325–5335. [CrossRef]
26. Delibacak, S.; Voronina, L.; Morachevskay, E.; Onguna, A.R. Use of sewage sludge in agricultural soils: Useful or harmful. *Eurasian J. Soil Sci.* **2020**, *9*, 126–139. [CrossRef]
27. Delibacak, S.; Ongun, A.R. Influence of treated sewage sludge applications on total and available heavy metal concentration of sandy clay soil. *Desalination Water Treat.* **2018**, *112*, 112–118. [CrossRef]
28. Lee, L.H.; Wu, T.Y.; Shak, K.P.Y.; Lim, S.L.; Ng, K.Y.; Nguyen, M.N.; Teoh, W.H. Sustainable approach to biotransform industrial sludge into organic fertilizer via vermicomposting: A mini-review. *J. Chem. Technol. Biotechnol.* **2017**, *93*, 925–935. [CrossRef]
29. De Ona, J.; Osorio, F. Application of sludge from urban wastewater treatment plants in road's embankments. *J. Hazard Mater.* **2006**, *B131*, 37–45. [CrossRef]
30. Sherene, T. Mobility and transport of heavy metals in polluted soil environment. *Biol. Forum Int. J.* **2010**, *2*, 112–121.
31. Fytili, D.; Zabaniotou, A. Utilization of sewage sludge in EU application of old and new methods—A review. *Renew. Sustain. Energy Rev.* **2008**, *12*, 116–140. [CrossRef]
32. Nicholson, F.A.; Smith, S.R.; Alloway, B.J.; Carlton-Smith, C.; Chambers, B.J. An inventory of heavy metals inputs to agricultural soils in England and Wales. *Sci. Total Environ.* **2003**, *311*, 205–219. [CrossRef]
33. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:31986L0278&from=RO (accessed on 12 December 2022).
34. Order No. 344/708/2004 for the Approval of the Technical Norms Regarding the Protection of the Environment and Especially of Soils, When Sewage Sludge Is Used in Agriculture. Available online: https://www.adideseuribn.ro/legislatie/Ordin%20344_2004.pdf (accessed on 29 October 2022).

35. Available online: https://www.eea.europa.eu/publications/GH-10-97-106-EN-C/file (accessed on 29 October 2022).
36. Available online: https://aric.org.ro/?p=1081 (accessed on 29 October 2022).
37. Tyagi, V.K.; Lo, S.-L. Sludge: A waste or renewable source for energy and resources recovery? *Sustain. Energy Rev.* **2013**, *25*, 708–728. [CrossRef]
38. Devi, P.; Saroha, A.K. Utilization of sludge based adsorbents for the removal of various pollutants: A review. *Sci. Total Environ.* **2017**, *578*, 16–33. [CrossRef] [PubMed]
39. Suárez-Iglesias, O.; Urrea, J.L.; Oulego, P.; Collado, S.; Díaz, M. Valuable compounds from sewage sludge by thermal hydrolysis and wet oxidation. A review. *Sci. Total Environ.* **2017**, *584–585*, 921–934. [CrossRef]
40. Anjum, M.; Makishaha, N.H.A.; Barakat, M.A. Wastewater sludge stabilization using pre-treatment methods. *Process Saf. Environ. Prot.* **2016**, *102*, 615–632. [CrossRef]
41. Zhen, G.; Lu, X.; Kato, H.; Zhao, Y.; Li, Y.Y. Overview of pretreatment strategies for enhancing sewage sludge disintegration and subsequent anaerobic digestion: Current advances, fullscale application and future perspectives. *Renew. Sustain. Energy Rev.* **2017**, *69*, 559–577. [CrossRef]
42. Zhang, Q.; Hu, J.; Lee, D.J.; Chang, Y.; Lee, Y.J. Sludge treatment: Current research trends. *Bioresour. Technol.* **2017**, *243*, 1159–1172. [CrossRef] [PubMed]
43. Nghiem, L.; Koch, K.; Bolzonella, D.; Drewes, J. Full scale co-digestion of wastewater sludge and food waste: Bottlenecks and possibilities. *Renew. Sustain. Energy Rev.* **2017**, *72*, 354–362. [CrossRef]
44. Semblante, G.; Hai, F.; Bustamante, H.; Guevara, N.; Price, W.; Nghiem, L. Biosolids reduction by the oxic-settling-anoxic process: Impact of sludge interchange rate. *Bioresour. Technol.* **2016**, *210*, 167–173. [CrossRef]
45. Available online: https://www.afd.fr/en/carte-des-projets/sludge-treatment-east-alexandria-wastewater-treatment-plant (accessed on 30 October 2022).
46. Available online: https://legislatie.just.ro/Public/DetaliiDocumentAfis/98310 (accessed on 12 December 2022).
47. Kim, H.G.; Chung, T.H. Performance of the sludge thickening and reduction at various factors in a pilot-scale MBR. *Sep. Purif. Technol.* **2013**, *104*, 297–306. [CrossRef]
48. Jackson, M.L. *Soil Chemical Analysis*; Prentice Hall of India Pvt. Ltd.: New Delhi, India, 1973; p. 498.
49. Bremner, J.M. Total Nitrogen. In *Methods of Soil Analysis Part 2: Chemical and Microbiological Properties*; Agronomy Monographs; American Society of Agronomy: Madison, WI, USA, 1965; pp. 1049–1178. [CrossRef]
50. Kalembasa, S.J.; Jenkinson, D.S. A comparative study of titrimetric and gravimetric methods for the determination of organic carbon in soil. *J. Sci. Food Agric.* **1973**, *24*, 1085–1090. [CrossRef]
51. McGrath, S.P.; Cunliffe, C.H. A simplified method for extraction of the metals Fe, Zn, Cu, Ni, Cd, Pb, Cr, Co and Mn from soils and sewage sludges. *J. Sci. Food Agric.* **1985**, *36*, 794–798. [CrossRef]
52. Lindsay, W.L.; Norvell, W.A. Development of a DTPA Soil Test for Zinc, Iron, Manganese, and Copper. *Soil Sci. Soc. Am. J.* **1978**, *42*, 421–428. [CrossRef]
53. Regional Water and Wastewater Infrastructure Development Project from Teleorman County in the Period 2014–2020. Available online: https://www.anpm.ro/documents/27905/38872680/MP+Apa+Serv.pdf/28ec2c57-b4d3-49bc-821d-2333dc326e30 (accessed on 30 October 2022).
54. Nazari, L.; Yuan, Z.; Santoro, D.; Sarathy, S.; Ho, D.; Batstone, D.; Xu, C.; Ray, M. Low-temperature thermal pre-treatment of municipal wastewater sludge: Process optimization and effects on solubilization and anaerobic degradation. *Water Res.* **2017**, *113*, 111–123.
55. Genot, V.; Colinet, G.; Brahy, V.; Bock, L. L'état de fertilité des terres agricoles et forestières en région wallonne (adapté du chapitre 4—Sol 1 de « L'État de l'Environnement wallon 2006–2007 »). *Biotechnol. Agron. Soc. Environ.* **2009**, *13*, 121–138.
56. Trașcă, F.; Mihăilescu, D.; Ionescu, N.; Trașcă, G.; Mincă, G.; Ciodaru, I. Research on the possibility to use sewage sludge as organic fertilizer of heavy acid soils. *Agric. Crop Tech. An. I.N.C.D.A. Fundulea* **2011**, *LXXIX*, 2.
57. Casida, L.E.; Klein, D.A.; Santoro, T. Soil dehydrogenase activity. *Soil Sci.* **1964**, *98*, 371–376. [CrossRef]
58. Vance, E.D.; Brookes, P.C.; Jenkinson, D.S. An extraction method for measuring soil microbial biomass C. *Soil Biol. Biochem.* **1987**, *19*, 703–707. [CrossRef]
59. Kaiser, E.A.; Mueller, T.; Joergensen, R.G.; Insam, H.; Heinemeyer, O. Evaluation of methods to estimate the soil microbial and the relationship with soil texture and organic matter. *Soil Biol. Biochem.* **1992**, *24*, 675–683. [CrossRef]
60. Bremner, J.M.; Mulvaney, R.L. Urease activity in soils. In *Soil Enzymes*; Burns, R.G., Ed.; Academic Press: New York, NY, USA; London, UK, 1978.
61. Tabatabai, M.A.; Bremner, J.M. Use of p-nitrophenol phosphate for the assay of soil phosphatase activity. *Soil Biol. Biochem.* **1969**, *1*, 301–307. [CrossRef]
62. Pramer, C.; Schmidt, A. Organic matter. In *Methods of Soil Analysis Part 2*; American Society of Agronomy, Inc.: Madison, WI, USA, 1964; pp. 1396–1397.
63. Keeney, D.R.; Nelson, D.W. Nitrogen-Inorganic forms. In *Methods of Soil Analysis Part 2: Chemical and Microbiological Properties*, 2nd ed.; Page, A.L., Miller, R.H., Keeney, D.R., Eds.; American Society of Agronomy: Madison, WI, USA, 1982; pp. 643–698.

64. Hidri, Y.; Fourti, O.; Eturki, S.; Jedidi, N.; Cheref, A.; Hassen, A. Effects of 15-year application of municipal wastewater on microbial biomass, fecal pollution indicators, and heavy metals in a Tunisian calcareous soil. *J. Soils Sediments* **2014**, *14*, 155–163. [CrossRef]
65. Navnage, N.P.; Patle, N.P.; Ramteke, P.R. Dehydrogenase activity (DHA): Measure of total microbial activity and as indicator of soil quality. *Int. J. Chem. Stud.* **2018**, *6*, 456–458.