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EFFECT OF MUNICIPAL SEWAGE SLUDGE ON WINTER RAPE AND SOYBEAN PRODUCTION AND HEAVY METAL CONTAMINATION OF SOIL

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ABSTRACT – Investigations conducted during 2004-2008 at the Podu-Iloaiei Agricultural Research Station have studied the influence of different rates of sewage sludge on yield and soil agrochemical characteristics. Sewage sludge was applied annually at rates of 20, 40 and 60 t/ha, together with mineral fertilizers, differentiated according to the growing plant. Applying rates of 20 t/ha DM sewage sludge resulted in the accumulation of mobile phosphate stock in soil of 58 ppm; the microelement content (mobile forms from soil) was of 19.0 ppm at Cu, 0.51 ppm at B, 149 ppm at Zn and 397 ppm at manganese. The obtained results have shown that by applying a rate of 30 t/ha of raw sewage sludge (18.5 l t/ha dry matter), the allowable maximum limits stipulated by Norm 86/278/EEC, were not exceeded. By applying rates of 60 t/ha raw sewage sludge (37 t/ha dry matter), the limits established by Norm 86/278/EEC and Order no. 49 from January 14, 2004 have been exceeded only at zinc. The application of the rates of 40 t/ha sewage sludge has resulted in increasing the degree of plant supply by mineral elements (especially, nitrogen and phosphorus). It has also resulted in a mean annual supply of 3.8 t/ha highly humificated organic matter; this explains the increase in soil humus content (after 4 years) from 2.82 to 3.79%. The use of fermented sewage sludge, at a rate of 40 t/ha, has resulted in getting mean yield increases of 65 % (1075 kg/ha), in winter rape, and of 63 % (937 kg/ha), in soybean. For slope lands degraded by erosion, sewage sludge with other organic resources may contribute to the improvement in soil characteristics. The application of rates of 20 t/ha sewage sludge DM has resulted in the accumulation of mobile phosphates in soil of 58 ppm, and the content of microelements, mobile forms from soil, was of 19.0 ppm at Cu , 0.51 ppm at B, 149 ppm at Zn and 397 ppm at manganese.

Key words: sewage sludge, heavy metals, winter rape, soybean, soil chemical characteristics

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REZUMAT – Efectul nămolului orăşănesc asupra producţiei de rapiţă şi soia şi contaminarea solului cu metale grele. Cercetările efectuate în perioada 2004-2008, la Staţiunea de Cercetări Agricole Podu-Iloaiei, au studiat influenţa diferitelor doze de nămol orăşenesc asupra producţiei şi a caracteristicilor agrochimice ale solului. Nămolurile oraşeneşti au fost aplicate în doze de 20, 40 şi 60 t/ha, împreună cu îngrăşăminte minerale, în funcţie de planta cultivată. Aplicarea unor doze de 20 t/ha SU nămol orăşănesc a dus la acumularea unei rezerve de fosfaţi mobili în sol de 58 ppm, iar conţinutul de microelemente (forme mobile în sol) a fost de 19.0 ppm Cu, 0.51 ppm B, 149 ppm Zn şi 397 ppm mangan. Din rezultatele obţinute se constată că, prin aplicarea unei doze de 30 t/ha nămol brut (18.51 t/ha materie uscată), nu s-au depăşit limitele maxime admisibile prevăzute în Directiva 86/278/EEC. Prin aplicarea unei doze de 60 t/ha nămol brut (37.0 t/ha materie uscată), doar la zinc s-au depăşit limitele stabilite prin Directiva 86/278/EEC şi prin Ordinul nr. 49 din 01/14/2004. Aplicarea unor doze de 40 t/ha nămol a determinat creşterea gradului de asigurare a plantelor cu elemente minerale, în special cu azot şi fosfor, şi s-a asigurat un aport mediu anual de 3.8 t/ha materie organică puternic humificată, fapt care explică creşterea conţinutului de humus din sol, doar după 4 ani, de la 2.82 la 3.79%. Folosirea nămolului fermentat, în doză de 40 t/ha, a determinat obţinerea unor sporuri medii de producţie de 65 % (1075 kg/ha) la rapiţa de toamnă si 63% (937 kg/ha) la soia. Pentru terenurile în pantă degradate prin eroziune, nămolurile, împreună cu alte resurse organice, pot contribui la ameliorarea însuşirilor solului. Aplicarea unei doze de 20 t/ha nămol materie uscată a determinat acumularea unei rezerve de fosfaţi mobili în sol de 58 ppm, iar conţinutul de microelemente, forme mobile din sol, a fost de 19.0 ppm la Cu, 0.51 ppm la B, 149 ppm la Zn şi 397 ppm la mangan.

Cuvinte cheie: nămol orăşenesc, metale grele, rapiţa de toamnă, soia, caracteristici chimice ale solului

INTRODUCTION

Sewage sludge is the most commonly studied residue. Most of investigations have followed its effects on environment.

The investigations carried out under different climate and soil conditions showed that applying sewage sludge has determined the increase of pH and organic carbon content from soil and improved plant nutrition with macro and microelements (Tsadilas, 2005; Madrid, 2007; Hargreaves, 2008). Rape is used for eatable oil, which is recommended for a healthy diet and for the production of ecological fuel (biodiesel) (Guş, 2003).

According to Directive 30/EC/2003, the member countries of EU will have to replace until 31 December 2010 at least 5.75% of the consumption of classical fuels used in transports by biofuels.

The obtained results on the cambic chernozems from the National Agricultural Research and Development Institute of Fundulea have shown that in rape growing, 80-160 kg N/ha, 50-80 kg P_2O_5/ha and 80 kg K_2O/ha must be applied (Sin, 2005).

In Germany (Rheinland-Pfalz), where the farmers using sewage sludge are stimulated, of the total used nutrients 311.5 kg/ha NPK (139.2 N; 57.1 P_2O_5; 115.2 K_2O), 152 kg are provided by mineral fertilization, 4.4 kg/ha by biocompost, 5.9 kg/ha by sewage sludge and 149.2 kg/ha by manure. Regulations of the German Government foresee the limitation of total nitrogen to 210 kg/ha in perennial grasses and 170 kg/ha on arable lands. In sewage sludge, maximum allowable amounts are 5 t/ha DM once in three years, or 10 t/ha well-treated sewage sludge (Jeebe, 1999). On arable lands, sewage sludge is applied within rotation, in crops, which accumulate low amounts of heavy metals.

The successive application for seven years of a rate of 100 Mg ha^{-1} municipal sewage sludge in wheat and maize crops, placed on a soil with silt clay loam texture determined the decrease of soil bulk density from 1.37 to 1.03 Mg m^{-3}, compared to unfertilized control, and the increase with a percent of organic carbon from soil (Aghilinategh, 2009). In soils from arid and semi-arid areas of Iran, applying rates of 250 kg urea ha^{-1} and 250 kg ammonium phosphate ha^{-1} for seven years, in wheat-maize crop rotation did not result in significant increases of the content of organic carbon from soil (1.73%), compared to unfertilized control (1.53%) (Aghilinategh, 2009).

MATERIALS AND METHODS

Investigations conducted during 2004-2008 at the Podu-Iloaiei Agricultural Research Station have followed the influence of different sewage sludge rates on yield and soil agrochemical characteristics. Experiments were set up in a five-year crop rotation (soybean-wheat-maize-sunflower-wheat). Sewage sludge was applied annually at rates of 20, 40 and 60 t/ha, with different mineral fertilizer rates, differentiated according to crop.

The Cambic Chernozem used for experiments has a clayey-loam texture (415 g clay, 305 g loam and 280 g sand), a weakly acid reaction and a mean supply with mobile phosphorus and a very good one with mobile potassium.

The soil on which physical and chemical analyses were done was sampled at the end of plant growing period. Soil response was determined in water suspension by potentiometrical means with glass electrode. The content of organic carbon was determined by the Walkley-Black method and the content of mobile phosphorus from soil was determined by the Egner-Riechm Domingo method, in solution of ammonium acetate-lactate (AL), and potassium was measured in the same extract of acetate-lactate (AL) at flame photometer. Determinations of cadmium, chromium, cobalt, copper, lead, manganese, nickel, and zinc in extracts of soil were measured by flame and electro-thermal atomic absorption spectrometric method, with the International Standard ISO 11047:1998. ANOVA was used to compare treatment effects.

RESULTS AND DISCUSSION

Fermented sewage sludge from the Iasi Municipal Treatment Station had a neutral reaction, an organic matter content of 44.3% and an organic carbon content of 29-30%.

The climatic conditions recorded during 2004-2008 have resulted in a good valorisation of mineral fertilizers and sewage sludge in main crops.

Composting of municipal sewage sludge used in agriculture depends on the production of good quality compost, which is mature and low in metals and salt content. The best method of reducing metal content and improving the quality of municipal sewage sludge is early source of separation (Hargreaves et al., 2008). Low increases of total soil Zn concentrations were observed where municipal sewage sludge was applied at a rate of 15 Mg ha^{-1} on an alkaline sandy soil, but no increases were seen when the compost was applied on an acidic sandy soil or a neutral sandy loam (Sebastiao et al., 2000).

The sewage sludge from the Iasi Station contained more zinc and copper (*Table 1*). These elements could be used in maize and beans crops, where, on slope lands, zinc deficit was frequent. Out of the nine studied heavy metals (Co, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn), only zinc concentration had high values (*Table 2*). The concentration of other heavy metals from fermented sewage sludge was similar to the normal content from soils.

Table 1- Content of heavy metals in sewage sludge from the Iasi Station, used for experiments (mg/kg DM)

Heavy metals	Heavy metals content in the following years:					Mean	LSD 5%
	2004	2005	2006	2007	2008		
Zn	2475	3260.0	2461	2254	2542	2598	186
Cu	42.9	63.9	61.3	72	69	62	4.5
Fe	20668	21629.0	24559	29088	32028	25594	650
Mn	315	323.0	325	370	432	353	16
Pb	187	174.0	164	135	152	162	10
Ni	38	57.0	64	59	52	54	8.3
Cr	87	86.0	74	61	76	77	5.2
Co	37	35.0	33	20	26	30	3.2
Cd	3.6	3.5	3.3	2.2	2.7	3.1	0.18
Hg	2.02	1.9	1.82	1.8	1.5	1.8	0.12

Table 2 - Maximum allowable concentrations for farming sewage sludge, according to Directive 86/278/EEC (further amended by Council Regulation 1882/2003/EC) and Order no. 49 (mg/kg DM)

Heavy metals	Heavy metal content from Iasi Municipal Treatment Station (mg/kg DM)	Directive 86/278/EEC, Council Regulation 1882/2003/EC) (mg/kg DM)	Maximum allowable concentrations for farming sewage sludge, according to Order no. 49 (mg/kg DM)
Zn	2598	2500-4000	2000
Cu	62	1000-1750	500
Fe	25594	-	-
Mn	353	-	-
Pb	162	750-1200	300
Ni	54	300-400	100
Cr	77	-	500
Co	30	-	50
Cd	3.1	20 – 40	10
Hg	1.8	16 – 25	

Organo-mineral resources as sewage sludge, correctly applied, could be a substitute for a great part of expensive technological consumptions (mineral nutrients) and could contribute to the improvement of organic matter content from soil. The results have shown that the application of a rate of 30 t/ha sewage sludge determined the increase in plant supply degree with mineral elements (especially, N and P). It has also provided a mean annual supply of 4 t/ha organic matter, highly humificated, which explained the increase in soil humus content from 2.82 to 3.79% (*Table 3*). By using sewage sludge, plant nutrition was improved in secondary nutrients (calcium, manganese and sulphur) and in micronutrients (boron, iron, manganese and zinc).

Table 3 - The evolution of chemical characteristics under the influence of sewage sludge applied on the cambic chernozem at the Podu-Iloaiei Agricultural Research Station

Agrochemical indices	Applied rates				LSD 5%
	Unfertilized control	20 t/ha sewage sludge, DM	30 t/ha sewage sludge, DM	$N_{100}P_{80}$ + 30 t/ha sewage sludge, DM	
pH, H_2O	6.6	6.7	7.1	6.7	0.11
Nt, %	0.142	0.187	0.196	0.199	0.01
P-AL, ppm	29	58	62	84	4.0
K-AL, ppm	235	275	289	291	12.0
Humus, %	2.82	3.38	3.79	3.76	0.02

Utilization of sewage sludge in agriculture has a major interest, due to nitrogen, phosphorus and microelement supply (Zn, Cu). For slope lands degraded by erosion, sewage sludge together with other organic resources can contribute to the improvement in soil characteristics (Tsadilas., 2005; Ailincăi, 2007, 2009; Jităreanu, 2007, 2009).

The application of rates of 24 t ha^{-1} sewage sludge has increased the contents of organic C from 13.3 to 15.0 g kg^{-1} soil and total N from 1.55 to 1.65 g kg^{-1} soil. There were significant increases in dehydrogenase (9.6%), β-glucosidase (13.5%), urease (15.4%), nitrate reductase (21.4%) and phosphatase (9.7%) activities (Crecchio, 2004).

Investigations conducted on the influence of sewage sludge treatment on heavy metals content from soil have shown that the continuous application for four years of sewage sludge on the same field resulted in the accumulation of heavy metals in soil.

By comparing the concentration of heavy metals from soil with maximum allowable limits established by EU countries, we found that sewage sludge from Iasi Municipal Treatment Station was within these limits and by applying rates of 20 t/ha sewage sludge dry matter, the limits foreseen by the Directive 86/278/EEC were not exceeded. At the rate of 60 t/ha raw sewage sludge (30 DM), the limit foreseen by the Directive 86/278/EEC was exceeded only at zinc (358 mg/kg soil DM) (*Table 4*).

Table 4 - Content of heavy metals from soil (mg/kg DM) at different rates of chemical fertilizers and sewage sludge, applied on the cambic chernozem of the Podu-Iloaiei Agricultural Research Station

Heavy metals from soil (0-20 cm)	Applied rates			LSD 5%	Limits foreseen by Directive 86/278/EEC
	Unfertilized control	20 t/ha sewage sludge, DM	30 t/ha sewage sludge, DM		
Zn	61.2	149	358	4.8	150-300
Cu	9	19	42	1.5	50-140
Mn	312	397	768	14.0	
Pb	14	32	59	6.2	50-300
Ni	31	41	49	2.1	30-75
Cr	29	41	52	1.1	
Co	10	17	25	0.9	
Cd	0.12	1.02	1.08	0.05	1-3

Cd and Ni concentration from soil, achieved by applying for 4 years rates of 30 t/ha sewage sludge, was lower compared to the mean allowable limit, established by Directive 86/278/EEC

The obtained results have shown that by applying a rate of 30 t/ha raw sewage sludge (19.51 t/ha), maximum

allowable limits were not exceeded. By applying a rate of 30 t/ha sewage sludge dry matter, the limits established by Norm 86/278/EEC and Order no. 49 of 14 January 2004 were exceeded only at zinc. Heavy metal content from fermented sewage sludge at the Iasi Municipal Treatment Station was higher at zinc (2598 ppm) and copper (62 ppm), requiring the limitation of sewage sludge rate to 20-30 t/ha. The application of rates of 20 t/ha sewage sludge DM has resulted in the accumulation of mobile phosphates in soil of 58 ppm and the content of microelements, mobile forms from soil, was of 19 ppm at Cu ,0.52 ppm at B, 149 ppm at Zn and 397 ppm at manganese (*Tables 3 and 4*).

The mean yield obtained in winter rape was of 1660 kg/ha in unfertilized control and 3380 kg/ha at the rate of 40 t/ha sewage sludge + $N_{80}P_{60}$ (*Table 5*). The mean yield increases obtained in the last three years in winter rape, by applying rates of 40 t/ha sewage sludge, were of 1075 kg/ha (65%), compared to untreated control. Mean yield increases obtained in winter rape, for each tone of sewage sludge, were of 17.9 kg/t. Nitrogen and phosphorus fertilizers resulted in mean yield increases of 1130 kg/ha (68 %) , while sewage sludge applied at rates of 40 and 60 t/ha resulted in yield increases of 1075 (65 %) and, respectively, 1455 kg/ha (88 %).

Table 5 - Influence of sewage sludge and mineral element fertilization on winter rape yield (Bravour Variety)

Treatment	Mean yield		Difference, kg/ha	Signif
	kg/ha	%		
Unfertilized control	1660	100		
$N_{80}P_{60}$	2790	168	1130	xxx
20 t/ha sewage sludge	2450	148	790	xxx
20 t/ha sewage sludge+ $N_{80}P_{60}$	3150	190	1490	xxx
40 t/ha sewage sludge	2735	165	1075	xxx
40 t/ha sewage sludge+ $N_{80}P_{60}$	3380	204	1720	xxx
60 t/ha sewage sludge	3115	188	1455	xxx
60 t/ha sewage sludge+ $N_{80}P_{60}$	3545	214	1885	xxx
Yield (Y) = 1954.03 + 5.19 NP + 17.86 sewage sludge (t/ha), R^2=0.926, obs=8				
LSD 5%= 250 kg/ha, LSD 1%= 340 kg/ha, DL 0.1% = 460 kg/ha				

The use of a rate of $N_{100}P_{80}$+30 t/ha sewage sludge in winter rape has resulted in getting yield increases of 10-15% (257-395 kg/ha) in Karibik, Bravour and Fair varieties, compared to the mean of trial.

In Toccata and Formula varieties, the obtained yields were lower by 342-638 kg/ha (13-24%), compared to the mean of trial (*Table 6*).

The mean yield obtained in soybean crop, grown in 5 year rotation, during 2004-2008 was of 1476 kg/ha in unfertilized control and 3115 kg/ha at the rate of 40 t/ha sewage sludge + $N_{60}P_{60}$ (*Table 7*). The mean yield increases obtained in the last 5 years in soybean crop, by applying rates of 40 t/ha sewage sludge, were of 937 kg/ha (63%), compared to untreated control. The mean yield increase obtained in the last 5 years in soybean crop, for each tone of sewage sludge, was of 18.3 kg/t.

Table 6 – Mean yields obtained in different winter rape varieties fertilized with mineral fertilizers and sewage sludge

Variety	N_0P_0	$N_{100}P_{80}$	$N_{100}P_{80}$+30 t/ha sewage sludge	Mean	%	Difference, kg/ha
Astoria	1743	2974	3340	2686	101	21
Formula	1635	1890	2555	2027	76	-638
Karibik	2115	3130	3520	2922	110	257
Petrol	1895	2605	3195	2565	96	-100
Royal	2045	2890	3145	2693	101	28
Toccata	1723	2510	2735	2323	87	-342
Fair	2300	3285	3595	3060	115	395
Nemax	2305	2635	3065	2668	100	3
Bravour	2405	3075	3500	2993	112	328
Recital	2010	2850	3285	2715	102	50
Mean	**2018**	**2784**	**3194**	**2665**	**100**	
%	100	138	158			
Difference	0	766	1176			
LSD 5%						150
LSD 1%						240
LSD 0,1%						315

Table 7 - Influence of sewage sludge and mineral element fertilization on soybean yield

Treatment	Mean yield		Difference, kg/ha	Signif
	kg/ha	%		
Unfertilized control	1476	100		
$N_{60}P_{60}$	2475	168	999	xxx
20 t/ha sewage sludge	2033	138	557	xxx
20 t/ha sewage sludge+ $N_{60}P_{60}$	2795	189	1319	xxx
40 t/ha sewage sludge	2413	163	937	xxx
40 t/ha sewage sludge+ $N_{60}P_{60}$	3115	211	1639	xxx
60 t/ha sewage sludge	2761	187	1285	xxx
60 t/ha sewage sludge+ $N_{60}P_{60}$	3392	230	1916	xxx
Yield (Y) = 1622.8 + 6.45NP + 18.27 sewage sludge (t/ha), R^2=0.982, obs=8				
LSD 5%= 220 kg/ha, LSD 1%= 310 kg/ha, LSD 0.1% = 430 kg/ha				

CONCLUSIONS

The fermented sewage sludge from the Iasi Station, by its chemical and biological characteristics can be used as fertilizer on most of soils, except the salted soils, due to their high content in soluble salts (1.0 g/100 g dry sewage sludge) and amendment measures, necessary in these cases.

The heavy metal content from fermented sewage sludge at the Iasi Municipal Treatment Station is higher at zinc (2598 ppm) and copper (62 ppm), requiring the limitation of sewage sludge rate to 20-30 t/ha.

The use of fermented sewage sludge, at a rate of 40 t/ha, resulted in getting mean yield increases of 65 % (1075 kg/ha) in winter rape, and of 63 % (937 kg/ha), in soybean.

The highest sewage sludge rate, which may be applied on fields, without exceeding the maximum charge rates of 30 kg/ha/year for zinc and 12 kg/ha/year for copper, foreseen by Directive 86/278/EEC and Order of the Romanian Government no. 49 from 14 January 2004, is of 30 t/ha.

The four-year application of 30 t/ha fermented sewage sludge resulted in the increase of mobile phosphorus content from soil (compared to untreated control) from 29 to 62 ppm and in the increase of humus content from soil from 2.82 to 3.79%.

The content of heavy metals from soil at the treated variant for four years with 20 t/ha sewage sludge has increased (compared to control) by 88 ppm at zinc, 10 ppm at copper, 10-12 ppm at Cr and Ni, 18 ppm at Pb and by 0.9 ppm at Cd.

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