TOXIC ELEMENTS IN THE SEWAGE SLUDGE – SOIL – PLANT CHAIN

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According to statistical data the amount of sewage sludge does not increase as it was prognosed some years ago. Although this is a positive effect, we have to solve the problem of recycling of the sewage sludge. The appropriate chemical composition of municipal sewage sludge compost is suitable for nutrient supply in the agriculture. The small-plot experiment with sewage sludge compost was started in the spring of 2003. Three doses of compost (9, 18, 27 t ha^{-1}) and a control treatment were used in the experiment. The small-plot experiment was re-treated in the fall of 2006 and 2009. In the composting process bentonite, rhyolite and wheat straw were used as additives for improving the quality of the compost. Our purpose was to stabilize or increase the fertility of sandy soil and contribute to the reduction of environmental pollution. In the plot experiment there were three test plants: triticale, maize and pea. These three plants were sawn in crop rotation. We examined the concentration of nickel (Ni) and zinc (Zn) in two soil layers, 0-30 and 30-60 cm in 2008-2009 years. We could not observe toxic effects on the test plants. The aim of the experiment was to investigate whether the application of sewage sludge compost cause hazardous accumulation of toxic elements in the soil and plants.

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Introduction

Sewage sludge is an organic waste which usually contains high levels of nitrogen and phosphorous as well as significant concentrations of micronutrients [1,2]. The use of sewage sludge in soil has very beneficial effects on the quantity and availability of nutrients, on the restoration of structural stability of the soil and on resistance against soil erosion [3,4].

Composting offers an effective method for treating sewage sludge by stabilizing organic matter, reducing odour, and killing human pathogens and weed seeds by the heat generated [5,6]. The products can be used as soil conditioner to improve soil physical properties by increasing soil organic matter content. In this way the water holding capacity and aggregate stability are increased, but soil bulk density is reduced[7]. Sewage sludge compost also represents a source of nitrogen (N), phosphorus (P), potassium (K) and trace elements such as zinc (Zn), copper (Cu) and molybdenum (Mn) for plant growth[8].

However, the presence of toxic elements in composts is the main cause of adverse effects on animal and human health, transmitted through the food chain from soil, groundwater and plants[9].

The sewage sludge contains toxic elements within an organic matrix. This may lead to the increased solubility of some of the metals (due to the formation of soluble organic complexes) or to their immobilization and subsequent reduced possibility of being assimilated by the plant[10]. Consequently, exact analyzing the contents of toxic elements in composts is very important for the routine monitoring and risk assessment and regulation of environmental protection.

In the present study, we aimed to measure toxic element, such us nickel (Ni) and potential toxic element, like zinc (Zn). During the experiment we would like to follow the movement of toxic elements in the food chain.

Experiments

Experimental design

The small-plot experiment was established in spring, 2003 at the Research Institute of CAAES of University of Debrecen, at the town of Nyíregyháza, located in the NE part of Hungary. It was re-treated in 2006, when the field experiment was re-arranged and the size of plots was enlarged.

The size of small-plots was 19 x 36 meters. The experiment was done in five replications on sandy soil. Our compost fills the requirements of the Agricultural Ministry Order of 36/2006 (V.18.). The compost includes 40% sewage sludge, 25% straw, 30% rhyolite and 5% bentonite. These materials have good effects on light textured sandy soils especially when they are mixed with organic material (e.g. sewage sludge). The compost was applied at 9, 18, 27 t ha^{-1} doses, ploughed into the 0-30 cm soil layer before sowing. The effect of the applied compost on triticale (*x Triticosecale Wittmack*), maize (*Zea mays*) and pea. (*Pisum sativum L.*) was studied.

Toxic elements of plant samples were measured after harvesting. In the case of triticale and maize the toxic elements content of grain was measured. The toxic element content of pea plants were measured in its three parts (root, straw + pods and grain). Composite soil samples were mixed from 5 subsamples in each plot from 0-30 and 30-60 cm soil layers after harvesting of all test plants.

The applied compost

In the Table 1 can be seen the important parameters of the applied compost

Table 1. Main characteristics of the applied compost

Parameter	Value
pH (H_2O 1:10)	7.2
Dry matter content [m/m% raw matter]	54
Organic matter content [m/m % dry matter]	26
Total N-content [m/m% dry matter]	1.68
Total P_2O_5-content [m/m% dry matter]	0.7
Total K_2O-content [m/m% dry matter]	0.4
As (mg kg^{-1})	5.26
Cd (mg kg^{-1})	0
Co (mg kg^{-1})	0
Cr (mg kg^{-1})	21.96
Cu (mg kg^{-1})	149.7
Hg (mg kg^{-1})	1.42
Ni (mg kg^{-1})]	12.09
Pb (mg kg^{-1})	23.33
Se (mg kg^{-1})	2.51

In the composted sewage sludge there were nitrogen (N), phosphorus (P) and potassium (K), which are useful for enhancing the available nutrients content of soil. The toxic elements content of compost was low therefore it is good for agricultural utilization. In the applied compost amount of nickel was under the limit value determined by the Agricultural Ministry Order of 36/2006 (V.18.).

Chemical and statistical analysis

The Zn and Ni content of soil and plant samples were measured according to the MSZ –08-0012/16-87. Briefly, the samples were digested with cc. $HNO_3 : H_2O_2$ and after filtering, the heavy metal contents of solutions were measured by using an ICP-OES emission spectrometer.

Statistical significance ($P<0.05$) of the differences was determined by one-way ANOVA and Tukey's multiple range test. Statistical analysis was performed with SPSS 13.0., with five replications.

Results and Discussion

Toxic elements in the soil

The utilization of sewage sludge in agriculture may cause to get a certain amount of toxic elements into the soil. This could result in the contamination of soil and plants by toxic elements. In our data reveal the fate of Zn and Ni in the sewage sludge-soil-plant chain.

The heavy metal content in soils comes from two sources: first, from the natural heavy metal content of soil and on the other hand, as a result of soil pollution. The measurable concentration of the micronutrients in the soil usually increases when the amount of heavy metals entering into the soil is higher than their leached or utilized quantity[11].

In the Table 2 the Ni and Zn concentrations of soil of the three test plants measured in 2008 are shown. The Ni and Zn content of the soil were below the 40 and 200 mg kg^{-1} limit in all treatments in this year. Concentration of Zn was increased after the treatments in the upper 0-30 cm soil layer, but this increase was not statistically significant. The measured Zn concentrations were around 30 mg kg^{-1}. In Canada[12] various crop responses were measured to a mixed municipal solid waste compost and the fate of certain metals associated with compost were examined. Plant and soil samples were collected after the compost application to analyse for content of Arsenic (As), Cupper (Cu), Zn, Mercury (Hg) and Lead (Pb). The research results showed that the compost slightly increased the heavy metal concentrations of the soil. The amount of Ni did not increase in the top soil under triticale.

Similar results were found when sea weeds compost was applied (leaves of beached *Posidonia oceanica* were used), which had high concentrations of heavy metals. Except of Ni, they measured higher heavy metal concentrations in soil samples amended with compost than in the control ones. The low concentration of Ni in soluble and exchangeable form explains its low mobility and biological activity [13].

Toxic elements content of soil samples measured in 2009 can be seen in Table 3. Ni concentrations of soil samples were similar in both soil layers (7.35 – 10.09 mg kg^{-1}) in 2009 and neither was above the 40 mg kg^{-1} threshold limit. The compost treatment did not increase its concentration. Low increase of Zn concentration in all treatments was found in the top soil samples collected from green pea's plots. On the contrary a researcher[14] observed significantly higher levels of Zn, manganese (Mn) and Cu in soil amended with compost at a rate of 19 t ha^{-1} but their applied compost had high heavy metal content (Zn: 566 mg kg^{-1}, Mn: 176 mg kg $^{-1}$ and Cu: 226 mg kg $^{-1}$).

Concentrations of measured elements were under the limit value (Ni: 40 and Zn: 200 mg kg^{-1}).

Toxic elements in the test plants

Pea (Pisum sativum L.)

In Table 4 presents the ratio of toxic elements content in different parts of pea in the sewage sludge compost treatment. The amount of Ni measured in the pea grain yield was slightly higher than the plants growing in unpolluted sites but among the treatments the differences are not significant. The amount of Zn was higher in the grain yield compared to other two parts of the plants in the year 2008 and 2009. Due to the 18 and the 27 t ha^{-1} treatment the Zn concentration increased in different parts of the plants. In 2009 the amount of Zn increased only in the 9 and 27 t ha^{-1} treatments compared to the control plots, while the Zn concentrations measured in the 18 t ha^{-1} treatments were similar to the control ones.

Table 2. Nickel and zinc content in two layers of soil under test plants in the sewage sludge compost experiment in 2008

Plant	Dose of compost (t ha $^{-1}$)	Ni (mg kg^{-1})				Zn (mg kg^{-1})			
		0-30 cm		30-60 cm		0-30 cm		30-60 cm	
		Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
Triticale	control	4.724	6.364	9.116	1.648	33.84	2.64	29.22	1.13
	9	4.230	5.649	9.274	1.494	35.26	1.79	28.40	1.54
	18	4.280	5.588	8.295	0.246	36.76	4.29	28.45	1.39
	27	4.058	5.065	10.768	3.353	35.48	4.97	29.36	1.81
Maize	control	0.065	0.055	9.860	1.120	32.53	2.29	28.46	1.11
	9	3.878	5.236	9.380	1.176	33.04	2.17	29.17	0.32
	18	4.118	5.110	9.393	0.405	34.98	2.33	29.42	2.51
	27	3.908	4.917	9.394	0.973	35.96	1.35	29.24	1.81
Green pea	control	0.123	0.037	10.478	1.100	32.15	7.48	26.88	1.28
	9	3.828	4.808	10.010	1.174	36.46	10.37	31.64	6.85
	18	3.746	4.974	9.862	2.059	33.94	3.57	27.84	2.85
	27	4.894	6.488	9.584	1.741	33.40	1.78	27.80	1.98
approved limit[1] (mg kg^{-1})		40				200			

[1] (50/2001. Government regulation). There was not significant difference among treatments, according to the Tukey's test ($p>0.05$)

Table 3. Nickel and zinc content in two layers of soil under test plants in the sewage sludge compost experiment in 2009

Plant	Dose of compost (t ha $^{-1}$)	Ni (mg kg^{-1})				Zn (mg kg^{-1})			
		0-30 cm		30-60 cm		0-30 cm		30-60 cm	
		Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
Triticale	control	8.256	1.177	8.294	0.275	21.857	2.547	21.971	2.703
	9	7.924	0.734	8.992	0.816	22.201	2.172	21.989	1.370
	18	7.761	1.130	8.616	1.576	22.137	1.726	19.755	4.213
	27	8.058	0.377	7.994	1.623	22.580	2.325	20.258	5.766
Maize	control	8.510	1.528	10.099	1.228	20.082	1.423	20.270	5.449
	9	9.625	3.265	9.411	2.593	23.016	4.989	22.221	9.451
	18	8.322	1.680	8.381	2.760	20.941	1.057	17.681	3.592
	27	7.703	1.617	8.390	1.729	20.673	0.989	17.249	3.363
Green pea	control	8.203	1.297	8.346	2.680	22.677	2.779	20.359	3.167
	9	8.503	1.454	8.509	0.426	23.665	3.686	22.375	1.463
	18	8.034	1.308	7.356	2.572	24.137	2.708	21.104	4.664
	27	8.161	1.511	7.845	3.304	27.806	3.471	19.502	4.702
approved limit[1] (mg kg^{-1})		40				200			

[1] (50/2001. Government regulation) There was not significant difference among treatments, according to the Tukey's test ($p>0.05$)

Table 4. Nickel and zinc concentrations in different parts of pea in the sewage sludge compost experiment

	Dose of compost (t ha $^{-1}$)	**Ni, mg kg^{-1}**				**Zn, mg kg^{-1}**			
		2008		**2009**		**2008**		**2009**	
		Mean	**S.D.**	**Mean**	**S.D.**	**Mean**	**S.D.**	**Mean**	**S.D.**
Root	control	33.58	23.25	15.08	5.25	20.52	8.24	26.33	6.23
	9	32.87	23.47	14.25	7.64	15.26	11.77	29.16	6.72
	18	36.39	30.23	5.77	2.69	22.24	12.18	22.87	9.07
	27	52.40	51.23	3.64	1.47	23.88	8.10	32.59	5.66
Straw + Pods	control	1.75	1.03	12.48	7.33	13.04	5.14	25.29	3.66
	9	2.59	2.75	20.88	15.83	15.04	2.70	21.45	3.81
	18	1.61	1.24	33.30	30.31	13,60	2.57	24.27	4.99
	27	1.71	0.71	25.52	18.45	19.10	8.11	24.78	6.93
Grain	control	12.36	8.87	7.92	3.48	48.27	11.13	46.91	5.27
	9	7.61	5.34	5.55	2.52	43.99	20.87	44.72	7.73
	18	8.69	3.22	2.38	0.94	51.08	4.20	45.15	4.11
	27	9.02	2.95	2.39	1.40	51.82	5.95	49.13	2.80
in unpolluted plants, mg kg^{-1}		0.1 - 5.0				25 - 150			

There was not significant difference among treatments, according to the Tukey's test ($p>0.05$)

Maize (Zea mays)

In Figure 1 presents Zn content in maize in the sewage sludge compost experiment. Zn concentration was under 30 mg kg^{-1} in the test plant. The quantity of Zn measured in maize was higher in the 9 t ha^{-1} dose of compost treated plots in 2008.

In 2008 we did not find significant differences between the three treatments, as it is showed in Figure 2. Other researchers found significantly increased Ni concentration in the roots of the lettuce plants after compost application. The Ni concentration was particularly higher in the leaves of the control sample[13]. Quantity of Ni in the maize crop was lower than detection limit in 2009.

Triticale (x Triticosecale Wittmack)

Figure 3 shows Zn content of triticale crop in the sewage sludge compost experiment in 2008 and 2009. We can see that amount of Zn was increased in the treatments in 2008. Measurements by Chaney[15] demonstrated that in plant tissue, the concentration of Zn was the highest in the compost treatment which reflect the high Zn concentrations in the compost material.

In the year 2009 the amount of Zn increased only in the 9 t ha^{-1} treatment in comparison with the control plot, while the measured Zn concentrations of the other two treatments were similar to the concentrations of control samples. Figure 4 presents the Ni concentration in triticale in the sewage sludge compost treatment. Effect of compost treatment did not increase the amount of Ni in 2008.

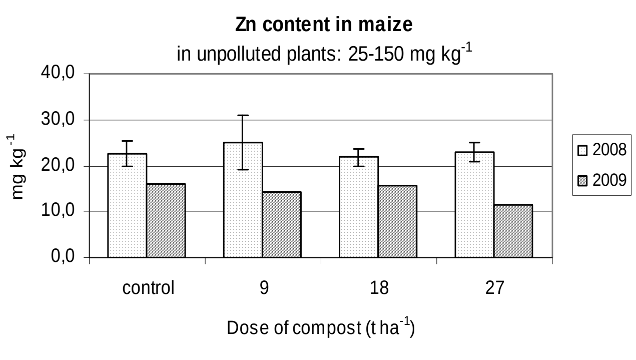

Figure 1. Zinc (Zn) content of maize in the sewage sludge compost experiment

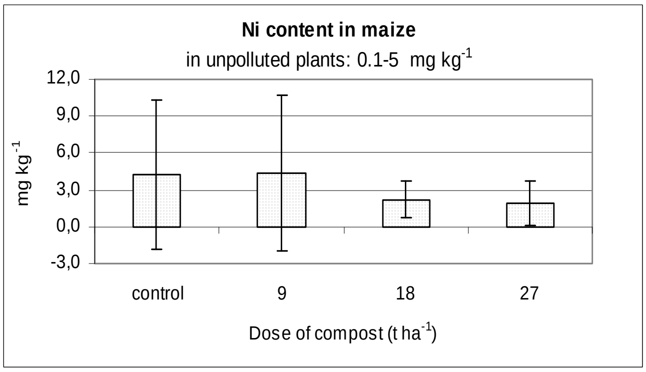

Figure 2. Nickel (Ni) content of maize in the sewage sludge compost experiment in 2008

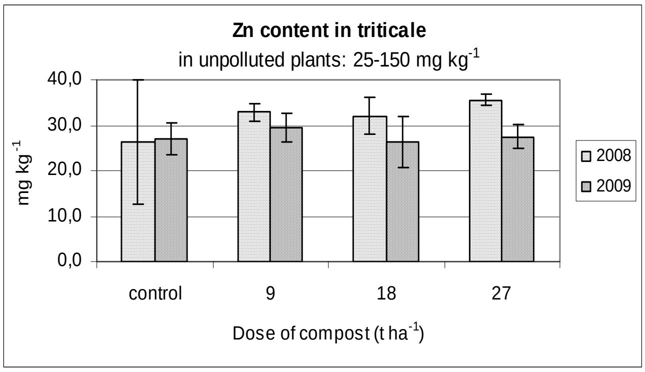

Figure 3. Zinc (Zn) content of triticale seed in the sewage sludge compost experiment

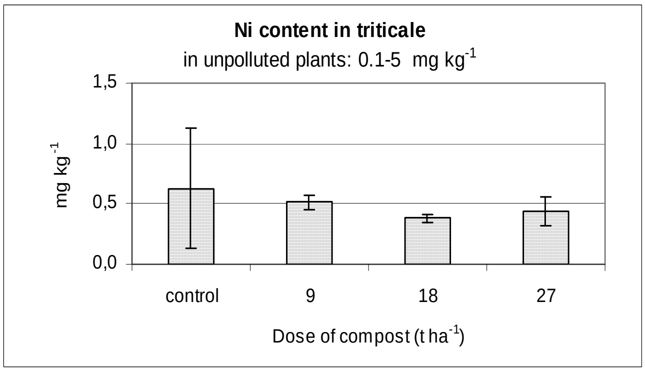

Figure 4. Nickel (Ni) content in triticale seed in the sewage sludge compost treatment in 2008

Similar to the Ni content of the corn, Ni content of triticale samples could not be measured in 2009.

Conclusion

Our results proved that the repeated use of composted sewage sludge for crop production did not cause any danger for agriculture and feed/food safety. We have not found toxic elements accumulation in the measured layers of the soil. The green pea accumulated Ni primarily in its root , which is important from food safety aspect, because it is not edible part of the plant. The other two test plants (triticale and maize) contain similar quantity of Zn in the grain while green pea concentrated this element in its grain with a higher quantity. The content of Zn and Ni were the lowest in the grain of maize.

Both measured elements were in the soil and plants, but these Zn and Ni did not accumulate hazardous in soil-plant chain. In conclusion it can be stated that the good quality and the circumspect application of the sewage sludge compost could guarantee its safe utilization.

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