

EXPERIMENTAL STUDY OF Cd, Hg, Mo, Pb AND Se MOVEMENT IN THE SOIL-PLANT-ANIMAL SYSTEM

POKUSNO ISTRAŽIVANJE KRETANJA Cd, Hg, Mo, Pb i Se U SUSTAVU TLO - BILJKA - ŽIVOTINJA

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SUMMARY

The purpose of this work was to evaluate the movement of some microelements in soil-plant-animal food chain. The long-term field trial with polluting microelements was set up on a calcareous loamy chernozem soil in 1991. The experimental site contained in its plow layer 25% of clay, 5% of CaCO₃, 3% of humus. The 13 elements/chemicals were added on 4 levels each as soluble salts to plots at initiation and mixed into the soil. The 52 treatments were arranged in a split-plot design with 2 replications. Elements applied were Al, As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Se, Sr, Zn. Loading levels were: 0, 90, 270, 810 kg/ha. Plant material derived from the field experiment was partly eaten by animals in feeding trials conducted by the Animal Nutrition Department of the Veterinary University. Main conclusions drawn from the field experiments and the 1992-year carrot feeding trial with rabbits:

From the applied elements/chemicals proved to be phytotoxic at harvest time for corn in 1991 were Cr, Mo, Pb; for carrot in 1992 and potato in 1993 Cr, Hg and Se. Mo was not mobile in soil, but accumulated extremely in plant tissues. Rabbits' organs also absorbed it weakly, maximum concentration showed kidney with 3.5 mg/kg D.M. It was excreted by both faeces and urine. Cd and Pb were mobile in soil, but accumulated in plant tissues. Rabbits' organs also absorbed these elements moderately while kidney showed maximum concentration with 3-5 mg/kg D.M. Excretion followed by faeces. Hg was not mobile in soil, but accumulated in carrot rot. It was absorbed extremely only by kidney with over 50 mg/kg D.M concentration. The residue excreted by faeces. Se was mobile both in soil, crop and animal organs. The residue was excreted by both faeces and urine. From the elements studied Se has the highest capability of moving through the soil-crop-animal chain unhindered.

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INTRODUCTION

The purpose of this work was to evaluate the movement of some important contaminants in soil-plant-animal food chain. The general research program based on experimental studies had in view the following goals for investigation:

1. Behaviour of these elements in soil: fixation, availability, leaching, volatilization, transformation.

2. Effect of these elements on soil life: soil biological activity, recording of macro- and micro organisms in soil, etc.

3. Absorption of these elements by plant roots and their transport within the plants: their accumulation in the shoots, leaves, stems and grains.

4. Effect of these elements on the animals. The plant material derived from the field experiments is was given to animals in feeding experiments led by the Department of Animal Nutrition, University of Veterinary Sciences.

MATERIAL AND METHODS

The soil of the Experimental Station is a calcareous chernozem with about 20% of clay, developed on loess. In its ploughed layer it contains humus in about 3% and CaCO_3 in about 5%. To ensure a sufficient macronutrient supply in the whole experiment, 100-100 kg/ha N, P_2O_5 and K_2O are given yearly. The 13 selected micro elements were applied in spring 1991 on 4-4 levels. The $13 \times 4 = 52$ treatments was arranged in a split-plot design with 2 replications. Each plot had an area of 21 m^2 . Maize was grown in the first, carrot in the second, potato in the third experimental year with commonly used agrotechnique. The treatments and the chemical used are given in Table 1.

Soil samples were taken each year. Each composite sample consisted of 20 subsamples drawn from the plow layer of each plot. Plant samples were taken during the vegetation period at least twice, using 20-40 plants or plant parts per plot randomly. Plant material and animal organs were dried, milled and digested in teflon

bombs using cc. $\text{HNO}_3 + \text{H}_2\text{O}_2$ and their total element content was determined, while soil samples were extracted by ammonium-acetate + EDTA (LAKANEN and ERVIO 1971) and their available element content was measured, using in all cases the ICP technique. The procedures, material and methods and the main conclusion of the feeding experiments are described elsewhere (FEKETE et al. 1994).

Table 1. Treatments in the field experiment. Calcareous chernozem soil - Nagyhorcsok Research Station, 1991.

Tablica 1. Tretiranja u poljskom pokusu. Tlo vapnenački černoze - Istraživačka stanica Nagyhorcsok, 1991.

Element used Primijenjen element	Applied in spring 1991 Primijenjeno u proljeće 1991. kg/ha				Chemical used Primijenjena kemikalija
	0	1	2	3	
Al	0	90	270	810	AlCl_3
As	30	90	270	810	As_2O_3
Ba	0	90	270	810	BaCl_2
Cd	30	90	270	810	CdSO_4
Cr	0	90	270	810	K_2CrO_4
Cu	0	90	270	810	CuSO_4
Hg	30	90	270	810	HgCl_2
Mo	0	90	270	810	$(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}$
Ni	0	90	270	810	NiSO_4
Pb	0	90	270	810	$\text{Pb}(\text{NO}_3)_2$
Se	30	90	270	810	Na_2SeO_3
Sr	0	90	270	810	SrSO_4
Zn	0	90	270	810	ZnSO_4

For feeding experiments those carrot root samples were selected which showed considerable microelement concentrations (Mo 39, Se 36, Hg 30, Pb 4, Cd 2-3 mg/kg D.M.) The experiment was carried out in the animal house of the Department of Animal Nutrition, University of Veterinary Sciences, Budapest. A total of 30 New

Zealand White rabbits of 2.3 kg average body weight were divided into 6 groups (n=5) according to the fed diet. The control group continued to receive the basal diet *ad libitum* while all 5 animals in each experimental groups were pre-fed over a period of 10 days. During that period the daily intake of basal diet was restricted to 50 g and a carrot diet containing Mo, Cd, Pb, Hg and Se was offered *ad libitum*.

This was followed by a 20-day feeding period when a "feed mixture" containing basal diet and carrot was fed. At the end of the experiment blood samples were taken from each animal and different blood parameters were determined. The rabbits were weighed, killed painlessly and subjected to pathological examination. The different animal organs were weighed and analysed. During both experimental periods the total daily faeces and urine volume was collected from each animal.

RESULTS AND CONCLUSIONS

Immediately before slaughter the body weight of experimental rabbits was significantly lower than that of the control animals. This can probably be attributed to the fact that the animals dry matter uptake from the carrot diet was rendered difficult by physical factors rather than by the heavy metal content of the carrot. This is also supported by the findings that the body weight of the experimental groups did not differ. Histological examination revealed that the rate of spermatogenesis in the testis was reduced in the treated animals. A large number of syncytial giant cells and degenerated cells indicating abnormal meiotic divisions were found among the spermatogenic cells.

With regard to the changes in blood parameters, it should be noted that Cd intake resulted in a considerable decrease in GGT and an increase in ALP activity. Both Cd and Hg had a similar effect on GGT and ALP activity. All experimental treatments decreased the activity of cholinesterase ALT, AST, CK and creatinine levels did not show appreciable changes. The carrot diet high in Mo, Cd and Se increased GSH-Px activity by 22-50%, that rich in Pb lowered it by 18%, while Hg caused no change in the activity of that enzyme.

Concerning the soil-plant-animal movement of elements the following conclusions could be drawn:

1. From the applied elements or chemicals proved to be phytotoxic at the time of harvest for corn 1991: Cr, Mo, Pb and Se. Carrot root in 1992 and potato tuber yield in 1993 decreased significantly in the Cr, Hg and Se treatments. Other elements did not effect the yield substantially (Table 2.)

Table 2. Phytotoxicity of some soil pollutant Calcareous chernozem, Nagyhorcsok, 1991-1993. t/ha*

Tablica 2. Fitotoksičnost nekih biljnih zagađivača vapnenačkog černozema

Sign of element Oznaka elementa	Applied element, kg/ha, spring 1991 - Primjenjeni elementi, proljeće 1991.				LSD _{5%}	Mean Prosjeak
	0	90	270	810		
Corn grain, 1991 - Zrno kukuruza, 1991.						
Cr	8.1	5.2	1.9	1.6		4.2
Mo	8.5	8.4	7.4	4.7	1.5	7.2
Pb	8.9	8.4	7.8	6.4		7.9
Se	8.5	7.6	5.7	4.3		6.5
Carrot root, 1992 - Mrkva						
Cr	13.0	7.1	-	-		5.0
Hg	15.6	15.3	13.8	10.8	4.8	13.9
Se	15.3	14.4	7.2	-		9.2
Potato tuber, 1993 - Krumpir						
Cr	12.0	11.3	7.9	4.9		9.0
Hg	11.6	9.3	8.0	7.9	3.5	9.2
Se	11.6	10.5	3.8	1.5		6.8

* Other treatments did not effect the yield significantly

* Ostali tretmani nemaju učinka značajne količine

- No harvestable yield

- Neusklađene količine

2. If we take in calculation that 1 mg/kg equals kg/ha in the 0-20 cm plow layer, we can state that 100% of Pb, 84% of Cd, 30% of Se, 23% of Hg and 16% of maximum applied Mo could be detected in the second experimental year in available ammoniacal + EDTA soluble form in soil. (Table 3.)

3. Mo was not mobile in soil, but accumulated extremely in plant tissue. Rabbits' organs absorbed it, however, weakly. It was excreted mainly by faeces and partly through urine. Maximum concentration shows kidney with 3,5 mg/kg D.W. (Tables 3, 4.)

4. Cd was very mobile in soil, but it accumulated moderately in plant tissues. Rabbits' organs also absorbed it poorly, maximum concentration found in kidney with 2.6 mg/kg D.W. It is excreted basically through faeces (Tables 3, 4.)

Table 3. Effect of Mo, Cd, Pb, Hg and Se treatment in the available element content of soil in plow layer and the total element content of carrot crop. Field experiment on calcareous chernozem soil. Nagyhorcsok, 1992. (mg/kg D.M.)

Tablica 3. Djelovanje Mo, Cd, Pb, Hg i Se tretiranja u sadržaju raspoloživog elementa tla u oraničnom sloju i ukupni sadržaj elemenata mrkve. Poljski pokus na vapnenačkom černozeu. Nagyhorcsok, 1992. (mg/kg S.T.)

Soil and carrot crop Tlo i urod mrkve	Applied in spring 1991, kg/ha Primjenjeno u proljeće 1991,				LSD _{5%}
	0	90	270	810	
Molybdenium (Mo) - Molibden					
In soil - U tlu	0.0	12	22	43	16
In carrot - U mrkvi					
Canopy - Nadzemni dio	0.1	117	270	434	33
Root - Korijen	0.0	21	54	99	12
Cadmium (Cd)					
In soil - U tlu	0.2	18	62	228	14
In carrot - U mrkvi					
Canopy - Nadzemni dio	0.1	2.9	6.6	11.2	4.2
Root - korijen	0.0	3.1	5.4	5.8	1.8
Lead (Pb) - Olovo					
In soil - U tlu	8	65	131	280	13
In carrot - U mrkvi					
Canopy - Nadzemni dio	0.2	3.1	5.3	7.8	2.0
Root - Korijen	0.3	3.6	4.1	4.1	1.2
Mercury (Hg) - Živa					
In soil - U tlu	0.0	1	13	61	4
In carrot - U mrkvi					
Canopy - Nadzemni dio	0.0	1.2	9.3	16.9	7.2
Root - Korijen	0.0	0.5	13.4	23.8	7.4
Selenium (Se) - Selen					
In soil - U tlu	0.0	7	66	81	13
In carrot - U mrkvi					
Canopy - Nadzemni dio	0.0	38	64	-	15
Root - Korijen	1.0	33	63	-	7

- No yield harvested because of phytotoxic effect – Nisu spremljene količine zbog fitotoksičnog učinka

Soil data for ammonacetate + EDTA soluble content by LAKANEN and ERVIO (1971). Plant data for total content using cc. HNO₃ + cc. H₂O₂ digestion.

Podaci tla za ammonacetat + EDTA topivi sadržaj po LAKANEN i ERVIO (1971). Podaci biljaka za ukupni sadržaj upotrebom HNO₃ + cc. H₂O₂ probavljičnost

Table 4. Effect of fodder carrot enriched with Mo, Cd, Pb, Hg and Se on the mineral composition of rabbit's organ, hard excrement and urine. Feeding experiment at the Institute for Animal Feeding. Analysis at the Research Institute for Soil Science and Agricultural Chemistry, 1992.

Tablica 4. Djelovanje mrkve za krmivo obogaćene s Mo, Cd, Pb, Hg i Se na mineralni sastav organa kunića, feces i urin. Hranidbeni pokus u Institutu za hranidbu životinja. Analiza u istraživačkom institutu za znanost o tlima i poljoprivrednu kemiju, 1992.

Carrot fodder, animal organs - Mrkva kao krmivo, organi životinje	Mo ppm		Cd ppm		Pb ppm		Hg ppm		Se ppm	
	Control Kontrola	Treated Pokus	Control Kontrola	Treated Pokus	Control Kontrola	Treated Pokus	Control Kontrola	Treated Pokus	Control Kontrola	Treated Pokus
Fodder with carrot Krmivo s mrkvom	0.53	39.00	0.14	2.30	1.58	4.01	0.00	30.00	0.00	36.20
1. Heart - Srce	0.06	1.23	0.00	0.00	0.39	0.00	0.00	0.00	0.58	19.4
2. Lung - Pluća	0.03	1.21	0.01	0.03	0.64	0.51	0.00	0.00	0.73	14.7
3. Liver - Jetra	1.26	1.88	0.12	0.72	1.72	1.85	0.00	3.53	1.74	65.0
4. Kidney - Bubrež	0.75	3.46	1.12	2.59	0.04	4.66	0.00	50.48	4.10	38.6
5. Milt - Slezena	0.00	1.08	0.01	0.00	0.76	0.15	0.00	0.08	1.99	15.4
6. Testis	0.24	0.73	0.00	0.02	0.21	0.00	0.00	0.00	1.00	22.4
7. Fat tissue Masno tkivo	0.00	0.06	0.00	0.00	0.14	0.06	0.00	0.00	0.00	0.6
8. Muscle - Mišić	0.00	0.37	0.00	0.00	0.00	0.13	0.00	0.13	1.33	13.5
9. Bone - Kost	0.00	1.20	0.00	0.00	0.00	0.65	0.00	0.00	0.00	3.2
10. Hair - Dlaka	0.00	0.41	0.00	0.00	0.36	0.0	0.00	0.00	1.37	2.7
11. Faeces	0.42	25.34	0.46	9.07	3.02	9.38	0.00	32.06	0.00	11.7
12. Urine	0.42	6.60	0.00	0.01	0.04	0.05	0.00	0.02	0.06	3.2
LSD _{5%}	1.41		0.38		1.14		11.04		2.21	

5. Pb was very mobile in soil, but it accumulated weakly in plant tissues. Rabbits' organs absorbed it also weakly, its maximum concentration found in kidney was 4.7 mg/kg D.W. It was excreted through faeces. (Table 3, 4.)

6. Hg was not mobile in this calcareous soil and accumulated moderately in plant tissues. It was absorbed extremely in rabbits' kidney with over 50 mg/kg D.W. concentration. The residue excreted by the faeces. (Table 3, 4.)

7. Se was relatively mobile in soil and it accumulated intensively in the canopy and roots. Its maximum concentration was found in liver and kidney, however, Se was absorbed practically by every organs by an order of magnitude. The residue excreted through faeces and urine. From the elements studied, Se has the highest capability of moving through the soil-plant-animal chain unhindered.

LITERATURE

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SAŽETAK

Svrha ovog članka bila je procijeniti kretanje nekih mikro-elemenata u hranidbenom lancu tlo-biljka-životinja. Dugotrajni poljski pokusi s mikroelementima zagađivačima postavljeni su na vapnenačkom tlu 1991. godine. Mjesto pokusa sadrži u svom oraničkom sloju 25% gline, 5% CaCO₃ i 3% humusa. Na parcele je pri započinjanju dodano 13 elemenata/kemikalija na 4 razine u obliku topivih soli i umiješano u tlo. Pedesetdva tretmana postavljena su u obliku razdvojene parcele u dva ponavljanja. Primijenjeni elementi bili su Al, As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Se, Sr, Zn. Razine opterećenja bile su: 0,90, 270, 810 kg/ha. Biljnim materijalom dobivenim u pokusu djelomično su hranjene životinje u pokusima hranjenja, kojeg je provodio Odjel za hranidbu životinja Veterinarskog fakulteta. Glavni zaključci dobiveni u pokusima na terenu i u pokusima hranidbe kunića mrkvom 1992. godine:

Od primijenjenih elemenata/kemikalija dokazanih da su fitotoksični za žitarice u 1991. bili Cr, Mo, Pb; za mrkvu 1992. i krumpir 1993. Cr, Hg i Se. Mo nije bio pokretan u tlu ali se u velikoj mjeri akumulirao u tkivima biljaka. Organi kunića slabo su ga apsorbirali, maksimalna koncentracija nađena u bubregu bila je 3,5 mg/kg S.T. Izlučena je fecesom i urinom. Cd i Pb bili su pokretni u tlu ali su se slabo akumulirali u tkivima biljaka. Organi kunića također su umjereno apsorbirali ove elemente, maksimalna koncentracija nađena u bubregu bila je 3 do 5 mg/kg S.T. izlučena je fecesom. Hg nije bio pokretan u tlu ali se nakupljao u truleži mrkve. Apsorbirao ga je u velikoj mjeri samo bubreg u koncentraciji preko 50 mg/kg S.T. Talog je izlučen fecesom. Se je bio pokretan u tlu, usjevima i organima životinja. Talog je izlučen urinom i fecesom. Prema istraživanim elementima Se ima najveću sposobnos nesmetanog kretanja kroz lanac tlo-usjev-životinja.

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