Mobility of Some Micropollutants in a Brown Forest Soil

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Introduction

The natural cycle of elements on Earth is limited and life has adapted itself to this condition. The more mobile, partly harmful fractions have disappeared from the soil, the concentration of undesired elements in the soil solution and in natural waters are low. The situation may change drastically when the available harmful element content of the soil is increased by some orders of magnitude by means of the application of sewage sludges with high metal content, traffic, industry or settlements in some parts of the country (CSATHÓ, 1994; KÁDÁR, 1993; SZABÓ, 1996; SZABÓ-WILLIN, 1995, 1996).

To get a reliable picture on the accumulation of elements, a statistically sufficient number of samples has to be analyzed. Such analyses of a hundred of soil, plant and fertilizer samples were carried out earlier, using ICP technique (FODOR, 1995; KÁDÁR, 1995; SZABÓ & FODOR, 1996; SZABÓ & KÁDÁR, 1994).

The present paper aims to supply data on soil contamination caused by soluble salts of micropollutants added to the soil in a field experiment. Plant uptake of microelements in the experiment is presented elsewhere (FODOR, 1998).

Material and Methods

Long-term field trials were set up at experimental stations representing three of the main soil types of Hungary: calcareous loamy chernozem, calcareous sandy soil and acidic brown forest soil. Different levels of the selected metal salts were added to plots once at initiation and were mixed into the ploughed layer (0-20 cm). Treatments were arranged in a split-plot design with replications. These experimental plots are cultivated with commonly used agrotechnics, chemical fertilizers are added yearly to ensure sufficient macronutrient supply for the different crops grown each year.

The field trial on acidic brown forest soil with clay illuviation – discussed in the present paper – includes 8 toxic elements (Al, As, Cd, Cr, Cu, Hg, Pb, Zn) and 3 levels (30, 90, 270 kg/ha), i. e. 24 treatments all in triplicate with 72 plots altogether. Each plot has a size of 35 m². The experimental plants were: wheat in 1995, maize in 1996 and sunflower in 1997. The main characteristics of the soil of the experimental site (Tas-puszta Experimental Station) (ploughed layer) were: pH_{KCl} 6.2, upper limit of plasticity (K_A) 43, humus content 3%, clay content 30-35%. Treatments are shown in Table 1.

Table 1

Treatments of the field trial on acidic brown forest soil with clay illuviation (Tas-puszta Experimental Station, 1994)

Element	Loading levels, kg/ha			Form of salt applied	% of element in salt	kg salt/plot		
	1.	2.	3.			1.	2.	3.
Al	0	90	270	$Al(No_3)_3 \cdot 9H_2O$	7.14	0.00	4.41	13.23
As	30	90	270	$NaAsO_2$	57.60	0.18	0.54	1.64
Cd	30	90	270	$3CdSO_4 \cdot 8H_2O$	43.83	0.24	0.72	2.16
Cr	30	90	270	K_2CrO_4	26.77	0.39	1.18	3.53
Cu	30	90	270	$CuSO_4 \cdot 5H_2O$	25.45	0.41	1.24	3.71
Hg	30	90	270	$HgCl_2$	73.88	0.14	0.43	1.28
Pb	30	90	270	$Pb(NO_3)_2$	62.56	0.17	0.50	1.51
Zn	30	90	270	$ZnSO_4 \cdot 7H_2O$	22.73	0.46	1.38	4.16

Composite soil samples consisting of 20 subsamples are collected yearly from the ploughed layer of each plot. To check the vertical movement of the pollutants, samples are also taken from the subsoil of plots with the highest level of added salts in some years. Choosing 20-40 plants or plant parts per plot randomly, plant samples are taken annually during the vegetation period and at harvest. The total amount of elements in homogenized soil and plant samples is measured after microwave digestion cc. HNO_3 + H_2O_2. In the soil samples, the so-called „mobile" fraction is determined by NH_4 acetate+EDTA extraction according to LAKANEN & ERVIÖ (1971).

Results and Discussion

The effect of treatments on the available element content of the ploughed layer of the brown forest soil with clay illuviation is presented in Table 2.

Considering that 3 kg/ha applied element equals 1 mg/kg in the ploughed layer, it can be stated that after the first year of the experiment nearly the total amount of the applied elements could be detected in the soil in „mobile" form

Table 2

Effect of treatments on the available element content in the ploughed layer of the acidic brown forest soil with clay illuviation (NH_4 acetate + EDTA extraction, ppm)

Element	Rate of application in autumn 1994, mg/kg				$LSD_{5\%}$
	0	30	90	270	
	Determined in 1995				
Al	127	119	156	188	24
Cu	7	22	32	98	24
Pb	6	18	35	96	32
Zn	7	24	58	75	25
Cd	0.2	13	37	117	35
As	0.0	14	38	98	29
Cr	0.0	2	5	12	4
Hg	0.0	3	4	17	4
	Determined in 1997				
Al	110	113	110	126	14
Cu	7	9	25	47	10
Pb	6	18	23	36	8
Zn	7	16	20	37	6
Cd	0.3	8	22	43	12
As	0.0	0.2	7	14	7
Cr	0.0	0.1	0.3	0.7	0.2
Hg	0.0	0.1	0.3	0.6	0.2

in the case of Cu, Pb, Cd, Zn and As, while 70-80% of the applied Cr and Hg „disappeared" in the soil in other fractions.

Two years later, in 1997, the NH_4 acetate + EDTA soluble mobile fraction of the elements Cu, Pb, Zn and Cd also decreased drastically. About two-thirds of the applied amount cannot be followed in soil by this method. At the same time, Cr and Hg almost completely „disappeared". There is also a transformation or fixation of these elements in less mobile form.

After harvesting the second crop in 1996, the „total" and „mobile" fractions were both measured in the 0-30, 30-60 and 60-90 cm soil layers of the plots with the highest (270 kg/ha) load. Data of the analysis are presented in Table 3.

The conclusions concerning the vertical movement of the elements can be summarized as follows:

As the elements are basically left in the upper 0-30 cm layer, deeper layers have not been contaminated significantly yet. Cd mobility cannot be proven either, deeper layers did not show any increase in total or in mobile concentration. Cr, applied as Cr(VI) was washed down to the 60-90 cm layer with precipitation, which might have been caused by the rainy and wet weather of 1996.

Table 3

The effect of 270 kg/ha As and heavy metal load on the total and mobile element content (ppm) of the acidic brown forest soil with clay illuviation (Tas-puszta Experimental Station, 1996)

Depth of sampling	„Total" content				„Mobile" content			
	I	II	III	Average	I	II	III	Average
				As				
0-30	54	61	110	75	12.6	13.6	33.6	19.9
30-60	12	11	13	12	0.3	0.0	0.5	0.3
60-90	11	11	11	11	0.1	0.0	0.1	0.1
				Cd				
0-30	99.6	108	61	90	83.6	88.1	58.2	76.6
30-60	0.8	0.6	0.5	0.6	0.2	0.3	0.2	0.2
60-90	0.4	0.4	0.5	0.4	0.1	0.1	0.2	0.1
				Cr				
0-30	96	128	80	101	2.0	2.3	2.2	2.2
30-60	37	35	35	36	0.4	0.3	0.4	0.4
60-90	38	30	34	34	0.2	0.2	0.3	0.2
				Cu				
0-30	59	88	100	82	39.0	62.2	49.5	49.9
30-60	22	23	21	22	5.4	4.5	5.5	5.1
60-90	16	18	20	18	3.5	3.6	3.6	3.6
				Hg				
0-30	19	17	80	39	0.8	0.5	8.7	3.3
30-60	0	0	0	0	0.0	0.0	0.0	0.0
60-90	0	0	0	0	0.0	0.0	0.0	0.0
				Pb				
0-30	61	29	49	46	4,.9	15.3	32.1	29.4
30-60	33	16	15	21	17.2	5.4	5.5	9.4
60-90	14	14	12	14	3.8	4.2	3.4	3.8
				Zn				
0-30	136	114	200	150	35.4	19.5	92.6	49.2
30-60	89	88	68	82	7.9	9.1	3.7	6.9
60-90	74	74	65	71	4.7	2.9	2.0	3.2

In the control soil:
„Total" ppm: Hg 0, Cd 0.6, As 9, Pb 20, Cu 25, Cr 30, Zn 85
„Mobile" ppm: Hg 0, Cd 0.3, As 0, Pb 7, Cu 7, Cr 0, Zn 10

Cu, Hg and Zn were obviously fixed in the upper layer, the soil profile showed no contamination with these elements. Pb, like As moved downwards somewhat, the mobile fraction in the 30-60 cm layer increased as compared to the control. Of course, the leaching of elements with time cannot be excluded in the long run.

When the mobile fractions were taken and expressed in the percentage of the total amount, the following mobility order was reached: Cd (85%) > Pb (65%) > Cu (61%) > Zn (33%) > As (26%) > Hg (8%) > Cr (2%).

Summary

In a field trial on acidic brown forest soil the effect of 8 micropollutants was studied on the soil's total and mobile element content. The As, Cd, Cr, Cu, Hg, Pb and Zn salts were applied in soluble form with the rate of 30, 90 and 270 kg/ha in 1995. The soil of the experimental site was characterized by pH_{KCl} of 6.2, upper limit of plasticity (K_A) 43, 3% humus content, 30-35% clay content. The total element content was measured after microwave digestion with cc. HNO_3+H_2O_2, while the mobile amount with NH_4 acetate + EDTA extraction (LAKANEN & ERVIÖ, 1971).

After the first year of the experiment, nearly the total amount of applied Cu, Pb, Cd, Zn and As could be detected in the ploughed layer in mobile form, while the Cr and Hg basically „disappeared" from this fraction. Two years later only about one-third of the applied Cu, Pb, Cd, Zn and As was traced in available forms, while Cr and Hg nearly completely disappeared. With time there is a fixation of these elements in less mobile/soluble forms. Cd, Cu, Hg and Zn showed no downward movement, while As, Cr(VI) and Pb mobile fractions reflected some accumulation in the 30-60 and 60-90 cm layers following the harvest of the second crops.

These observations need further investigation in the long run.

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