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Effect of aerial deposition on an experimental station near to Budapest, Hungary

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Abstract

The composition of precipitation and element loads originating from rainwater were examined at an experimental station (Őrbottyán) in Hungary, in order to identify the aerial input of elements and define their effects on agriculture and environment. Twenty-five characteristics were analysed: pH, EC, NO_3-N, NH_4-N, and concentrations of the main macro- and microelements. The observation represents the dry and wet depositions together.

Generally, the lower amount of monthly precipitations resulted in higher EC, pH, NH_4-N, Ca, Na, K concentrations. The highest element yields however, were typical for the wet months. Acidic precipitation under pH 5 was rich in nitric acid forming NO_3-N, but poor in NH_4-N at the Experimental Station. Emission of the nearby cement plant in February and March 2006 caused an increase of Ca, Mg, Na, Sr elements with an order of magnitude compared to the other months and the concentrations of NH_4-N, S, Zn, As, Cr, Pb also lifted considerably in the precipitation. The pH reached 7.0 at this site.

Aerial input has considerable agronomical and environmental significance. According to this study the aerial deposition could substitute for 10% K, 15% Mg, 20% P, 30% Ca and N, 40% S element demand of an average 5 t/ha cereal grain yield with its 5 t/ha straw. The deposition of Zn, Mn, Fe, Cu, B elements in this site were similar to previous Hungarian and Austrian data, however Pb, Ni, Cd, Co depositions were lower with an order of magnitude, which demonstrates the result of the successful heavy metal pollution control in Europe since 1990.

Key words: aerial deposition, macro- and microelements, agricultural importance, environmental consequences, nutrient balances

Introduction

In Hungary, Kazay (1904) analysed the ammonium and nitrate content of the rainfall at Ó-Gallya Station between 1902 and 1904. Concentration of NH_4-N was 12 kg/ha and NO_3-N was 5 kg/ha in 1902, so he found a 17 kg/ha/year N deposition. Data from the literature also supports that content of NH_4-N can be 2-3-fold compared to NO_3-N. NH_4-N can be enriched in lower strata clouds, since it is mostly emitted by the soil surface. Maximum values are typical for January, minimum values for July, because water can absorb less gas at higher temperature. The first raindrops and the hail in summer are also rich in ammonia. N deposition of an abundant rainfall with available nitrate and ammonia could be equal to 30 kg/ha N fertilization, according to Kazay (1904).

Based on data from the 1980's Nriagu and Pacyna (1988) and Nriagu (1989) estimated, that human activity is responsible for 96% of Pb, 85% Cd, 75% V, 66% Zn, 65% Ni, 61% As, 59% Hg and Sb, 56% Cu, 52% Mo, 42% Se, 41% Cr of the total aerial amount of these elements on a global scale. It was also established, that toxic elements accumulate fast in air, waters, soils and in the whole food chain, which is an unknown risk for the future generations.

Sager (2009) in Austria calculated the atmospheric deposition in 1999-2000 from own and literature data as follows: Zn 308, Cu 110, Ni 30, Pb 24, Cr 12, Cd 2 g/ha/year. The Cr and V pollution was made mainly by

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fertilizers, while Pb and Zn pollution came mainly from aerial deposition. In the case of As, Cd, Ni elements the two mentioned sources had similar effects.

Ammonia emission was reduced by 60% in Hungary between 1980 and 2000. About 94-98% of this emission originates from agriculture. The main sources are manuring, N-fertilization, animal farms, communal sewage and garbage production (CSOH 2003). Since 1990, the decreasing trend in N-fertilization, animal husbandries and manuring are responsible for most of the emission reduction in Hungary. In Northwestern Europe efforts were also successful for the reduction of air pollution (SO_2, NOx, NH_3) since 1990 (Boxman et al. 2008).

Table 1 presents an estimated microelement-balance in the 1980's in Hungary. According to our analytical data, phosphorous fertilizers had the highest content of microelement pollutants. Fertilization caused about 30 g As, 8 g Zn and Cu, 4-5 g Pb, 1-2 g Se, 0.8 g Cd and 0.4 g Ni per hectare load yearly as an estimation in the 1980's due to the practice of large-scale fertilizer application. Fertilization made only 5-10% of the total pollution, so generally its role was almost negligible, but this source was responsible for the 2/3 of the total As load. Manure application is a considerable source of Zn, Pb, Cu, Ni, As elements. However, greatest pollution of Zn, Pb, Cd and partly Ni came from atmospheric precipitation.

According to Table 1 the balances of the observed microelements were positive. Inputs of Pb and As were 5-fold, Cd 4-fold, Se 2.5-fold, Zn and Ni 1.5-fold compared to outputs of these elements. The situation has changed by nowadays. After the 1990's Pb load decreased to 1/5 with the introduction of unleaded fuel, and As decrement was similar due to the reduction of superphosphate application. Income of Pb, Cd and Zn from the Silesian "Black triangle" also declined as the former industrial area has been collapsed.

Table 1. Estimated microelement-balance in agricultural cultivated soils in Hungary in the 1980's, g/ha (In: Kádár et al. 2009)

Balance items	Zn	Pb	Cu	Ni	As	Cd	Se
Input							
Mineral fertilizers	8	5	8	<1	30	0.8	1.5
Organic fertilizers	180	30	60	15	15	1.5	1.5
Sewage sludge	50	17	17	3	2	0.3	1.7
Liming materials	2	<1	1	<1	<1	<0.1	<0.1
By-products	80	8	60	7	<1	0.8	0.5
Precipitation	200	70	24	15	1	5.0	*1.0
Total	520	130	170	40	47	8.4	6.2
Output							
Plant uptake	200	10	100	10	1	1.0	1.0
Leaching	20	10	5	5	4	1.0	0.5
Volatilisation	-	5	-	-	5	-	1.0
Total	220	25	105	15	10	2.0	2.5
Balance	+300	+105	+65	+25	+38	+6.4	+3.7
Input% compared to output	236	520	162	267	480	420	248

*Verbal information from Ágnes Molnár, Atmospheric chemistry group, University of Veszprém

Earlier, a comprehensive research was done at two Experimental Stations of the Research Institute for Soil Sciences and Agricultural Chemistry in order to estimate the amount of the aerial element input and its effects on agriculture and environment (Kádár et al. 2009, Kádár and Ragályi 2010).

Material and methods

Samplings were conducted at one of the Experimental Sites of the Research Institute for Soil Sciences and Agricultural Chemistry in Őrbottyán city (Danube-Tisza mid region), where amount of monthly precipitations were measured for almost 50 years. The measuring equipment was installed 1 m high above the ground and emptied every day at 7 a.m. according to the general meteorological practice. The accuracy of the measurement is 0.1 mm. The solid snow, sleet, freezing rain and hail are measured after melting. The dew, frost and hoar are not considered to be precipitation.

The precipitation samples were stored in glass bottles at 7°C and analysed monthly for 26 attributes. Measuring of mineral elements were conducted by ICP technique. Electrical conductivity, pH as well as As, Ba, Ca, Cd, Co, Cr, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, P, Pb, Se, Sr, Zn, S, B, NH_4-N and NO_3-N contents were determined. Element yield or atmospheric deposition was calculated by multiplying the rainfall amount with element concentrations. The measuring equipments collected not only rain but also aerosol particles of dry fall-out so wet and dry inputs were sampled together. However according literature data the proportion of dry deposition is diverse depending on elements, but makes only 5-10% of the total aerial input. The incidental pollution from the measuring equipment was additionally observed. Measurements of macro- and microelements were made by ICP-OES device excluding nitrogen.

Results and discussion

The monthly data of amount, conductivity, pH as well as element concentrations and yields of rainfall are shown in *Table 2* between January and July 2007.

Table 2. Characteristics of the monthly precipitation as well as element yield in 2007 (Danube-Tisza mid region, calcareous sandy soil, Őrbottyán)

Months	Precipitation	Conductivity	pH	Ca	NH_4-N	NO_3-N	S	Na	K
in 2007	mm	µS/cm		Concentration, mg/l					
I.	31	62	4.5	3.6	0.7	4.0	n.d.	1.1	1.1
II.	46	54	4.8	3.4	1.1	3.6	n.d.	1.1	0.9
III.	39	78	4.2	4.4	0.8	4.0	n.d.	0.7	0.7
IV.	4	140	7.2	4.7	11.6	0.0	n.d.	1.2	3.1
V.	58	71	6.9	1.9	5.5	1.7	n.d.	1.2	1.2
VI.	61	86	4.5	7.7	0.7	5.8	n.d.	0.9	2.0
Months	Precipitation	Conductivity	pH	Ca	NH_4-N	NO_3-N	S	Na	K
in 2007	mm	µS/cm		Element yield, kg/ha					
I.	31	62	4.5	1.1	0.2	1.2	n.d.	0.3	0.3
II.	46	54	4.8	1.5	0.5	1.6	n.d.	0.5	0.4
III.	39	78	4.2	1.7	0.3	1.6	n.d.	0.3	0.3
IV.	4	140	7.2	0.2	0.5	0.0	n.d.	0.0	0.1
V.	58	71	6.9	1.1	3.2	1.0	n.d.	0.7	0.7
VI.	61	86	4.5	4.7	0.4	3.6	n.d.	0.5	1.2
Total:	239	-	-	10.3	5.2	9.0	-	2.4	3.1

n.d. - not detectable

The maximal electrical conductivity, pH, NH_4-N, Na and K concentrations, as well as the lack of NO_3-N connected to the lowest 4 mm amount of rainfall in April. The highest element yield resulted however in the rainiest May and June. The precipitation in March was strongly acidic with pH 4.2. Generally, when the nitric acid forming NO_3-N concentration is 3-5-fold more than the pH increasing NH_4-N concentration, the pH value goes under 5. The NO_3-N excess is also related to the high atmospheric N yields.

During the examined half year total aerial deposition was 14 kg N, 10 kg Ca, 2-3 kg Na and K per hectare. S could be detected only in traces. During the second half-year the amount and composition of the rain did not undergo such dramatic changes so it will not be presented in details.

Table 3 gives an overview about the amount and observed characteristics of measured aerial deposition between 2005 and 2008. The Ca, Mg, Na and Sr concentrations in rainwater raised an order of magnitude in February and March 2006. NH_4-N, S and Zn depositions were also high.

Table 3. Atmospheric deposition to soil at the Őrbottyán Experimental Station (Danube-Tisza mid region, 2005-2008)

Soil characteristics	Unit	2005 July-Dec.	2006 Total	2007 Total	2008 January-June
NO_3-N	kg/ha	10.7	10.0	19.9	4.3
NH_4-N	kg/ha	5.7	38.0	9.5	9.2
Total N	kg/ha	16.4	48.0	29.4	13.5
Ca	kg/ha	8.0	60.1	13.3	8.6
K	kg/ha	6.0	16.5	6.3	3.1
S	kg/ha	5.8	21.1	2.2	0.0
Na	kg/ha	3.0	13.3	4.1	0.8
Mg	kg/ha	2.8	15.8	2.4	1.3
P	kg/ha	1.2	5.6	2.5	1.5
Zn	g/ha	430	1 391	264	67
Ba	g/ha	60	79	40	15
Sr	g/ha	27	202	35	24
Cu	g/ha	33	153	21	10
Precipitation	mm	406	523	466	273
pH					
minimum		5.2	4.7	4.2	5.0
maximum		6.8	7.0	7.2	5.9
Mean		6.1	6.0	5.5	5.4
EC					
minimum	µS/cm	30	47	25	26
maximum	µS/cm	179	1 996	140	149
Mean	µS/cm	71	320	68	61

These emissions originated mainly from the cement works in city Vác, which is about 14 km far away north-westward from where usually the wind blows. The pH reached 7.0 and conductivity was near to 2000 µS/cm. Yearly element yields were outstanding: 60 kg/ha Ca; 48 kg/ha N; 21 kg/ha S; 16 kg/ha K and Mg; 13 kg Na; 5-6 kg/ha P. The yields of the notable microelements were as follows: Zn 1391 g/ha; Sr 202 g/ha; Cu 153 g/ha; Pb 7 g/ha; As 4 g/ha; Cr 3 g/ha.

Conclusions

Aerial deposition varied widely. The deposition of Zn, Mn, Fe, Cu, B elements were similar to previous Hungarian and Austrian data. However, Pb, Ni, Cd, Co depositions were lower with an order of magnitude compared to the earlier data from Mészáros et al. (1993), which demonstrates the result of the successful heavy metal pollution control in Europe since 1990.

The site was poor in N, P and K elements. The yearly 25-50 kg/ha N, 6-10 kg/ha K and 2-4 kg/ha P aerial depositions have agronomical importance. Yields are increasing year by year in PK treatments of a rye monoculture experiment since 1960 at this site, probably due to aerial N input (Kádár et al. 1984). In dry and unfavourable years the cereal yields remain below 2 t/ha, no N-effects observed. N supply could be covered by atmospheric N deposition because of the low crop yields. The Zn and Cu inputs are important among the microelements. This site is poor in Zn and Cu so the aerial deposition can totally cover the supply of these elements for the average yields. The adverse effect of the occasional acid rains can be compensated by the aerial deposition of Ca, Mg and Na.

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