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NUTRIENT AND TRACE ELEMENT DISTRIBUTION IN LOOSE FLUVIAL SEDIMENTS, KÖMLŐ AREA, HUNGARY

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Abstract. The Kömlő model area is a 250×250 m sized agricultural field in the central part of the Great Hungarian Plain. In this paper, the geological background, the hydrogeology of the area and the distribution of nutrients and trace elements of different soil levels were studied. The soil, which was formed on the clayey-silty floodplain and the sandy riverbed sediments of the old River Tisa was sampled in 25 boreholes (4 genetic levels, 108 samples) and the samples were analyzed for 23 main and trace elements. Among them, tree groups of elements were recognized: the main, rock forming elements, the trace elements of which concentration up to 1 ppm and some elements with concentrations below 1 ppm, close to the detection limit. The concentration of elements and the relation among them depend on the sediment type, on the soil level and on the presence of the groundwater table, while the areal distribution of some trace elements in the ploughed and in the root zones indicates the stream direction of the sediments during the past floods of the river Tisa.

Keywords: Great Hungarian Plain, Holocene sediments, Nutrient elements, Trace elements, Ploughed layer, Root zone, Statistical analysis, Linear correlation.

1. INTRODUCTION

Beginning 2002, in Kömlő model area, a complex mineralogical, petrographical, sedimentological, pedological and agrochemical research program was executed. The aims of these works were to establish the geological background of outcropping hydromorphous soils, emphasizing the negative factors of plant nutrition and as well, to establish the source of the seasonal Mg-salt accumulation. These studies, including the analysis of nutrient and trace element content of the sediments was part of the project NKFP No. 4/037/2001 „Precision Plant Production”

2. LOCATION

The rectangular model area, measuring 250×250 m is situated on outskirts of Kömlő village, Heves county, in the central part of Great Hungarian Plain. The land owner is TISZA FARM PRODUCE Ltd., which has supported a part of our research program.

The Heves Plain is a near perfectly, horizontal alluvial plain of Tisa river and its left affluent rivulets. 25 boreholes of 9-10m in depth were executed in right side of the small Görbe-chanel. Here, a NE-SW oriented hillock - probably, a buried sand bar - of 92.3 m altitude dominates the plain of 90.0–90.5m ASL altitude (Plate I, up).

The Heves Plain is one of the most fertile agricultural zone of Hungary (Vatai et al., 1998). Here large fields of wheat (Plate 1, photo 1), barley, corn and sunflower occur The model area covers arable land, with alternative crops of alfalfa, but they fertility is very heterogenous, considering the salt affected areas.

The Heves plain is characterized by hydromorphic soils in which the morphological and chemical characters are defined by the vertical movement of ground water. The model area is covered with Solonetz soils of different properties.

3. THE GEOLOGICAL BACKGROUND

The deep basement of the Heves Plain are represented by Maramures-Szolnok Paleogene Flysch deposits, where are covered by more, that

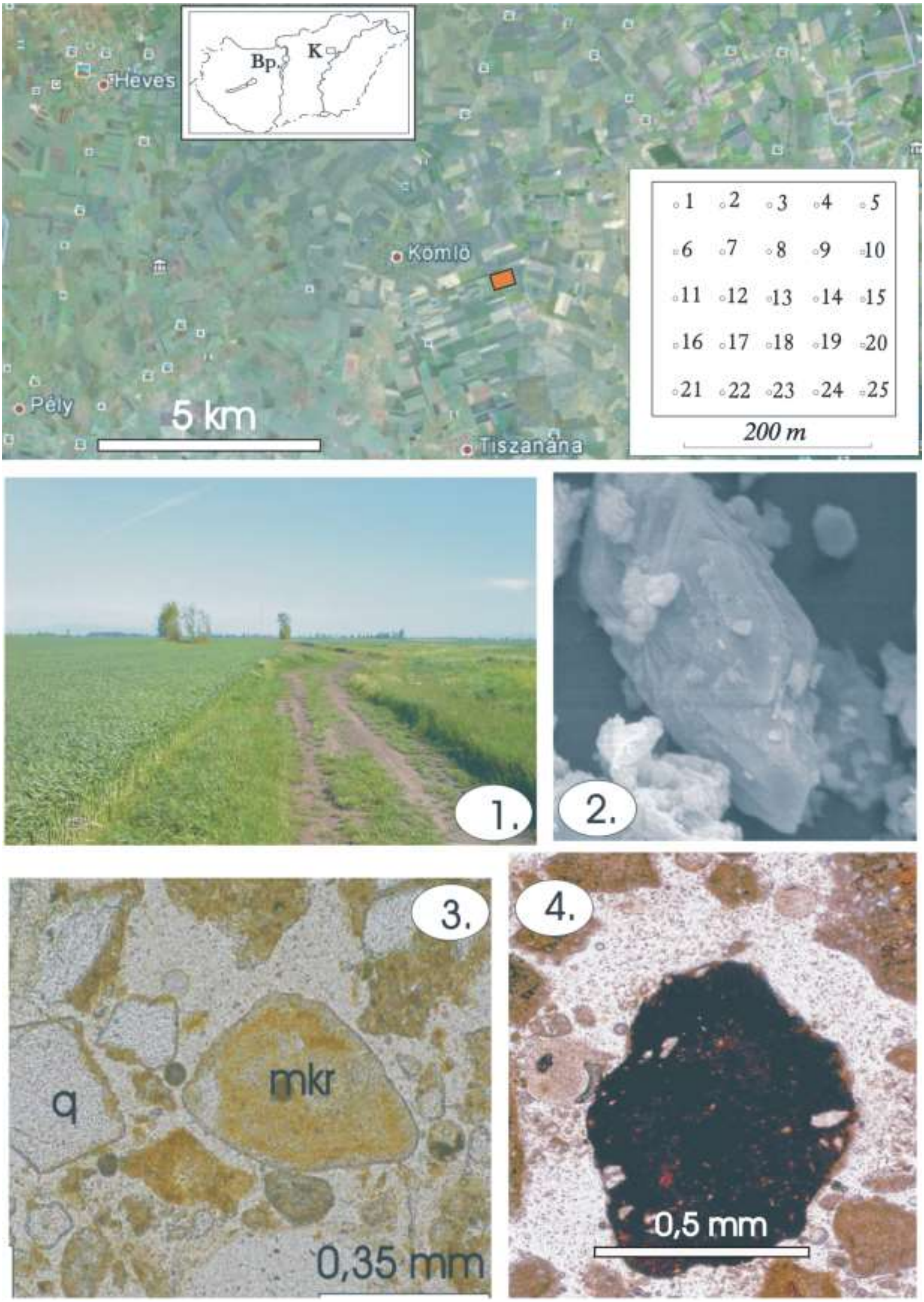

Plate I. Up: Location of Kömlő model area (K, red squadron); in frame the shallow borehole network. Photos: 1. The Heves Plain, farm path and wheat field, close to the Kömlő model area; 2. .Euhedral epsomite crystal and small halite cubes from white salt powder of soil lumps. SEM photo, 1000×; .3. Micritic limestone fragment (mkr) and quartz grain (q) in sand. Thin section, II nichols; 4. Dark brown limonite concretion in clayey sand. Thin section, II Nichols.

1000 thick Pannonian clayey-sandy deposits (Rónai et al., 1975) The upper, sandy level of Pannonian s.l. and the Pleistocene sandy deposits represent the main aquifers of this area, in which the artesian wells were deepened (Rónai, 1955).

The Heves Plain is covered by fluvial and fluvio-eluvial sandy–silty and silty–clayey deposits. Based on Mollusca, Pollen and Archaeological analyses (Sümegi, 2005), these 10-30 m thick deposits represent the Pleistocene-Holocene transition, the Lower Holocene and a part of Upper Holocene.

The correlation of our 25 shallow boreholes from 2002 and other two ones from 2006, following the recommendations of Friend (1984) indicates four sand-silt-clay sedimentary sequences, from which the last near surface sequence is incomplete (Fig. 1).

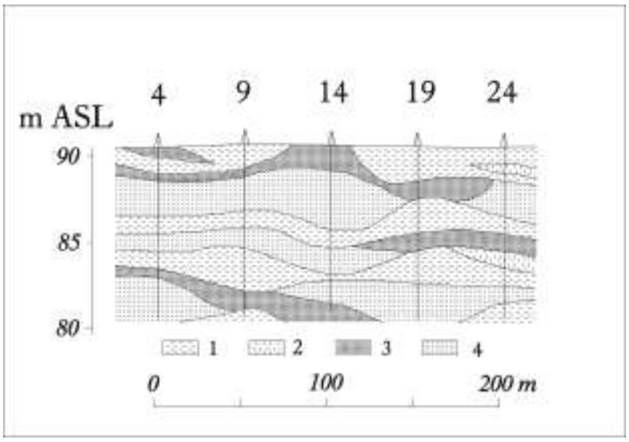

Figure 1. Geological section in Kömlő Model Area. 1. Clay; 2. Silt; 3. Silty sand; 4. Sand.

The clayey-silty sediments in which the main and trace element concentrations were measured, consists of clay and non-clay minerals. The main clay mineral is the Cheto-type, Ca-montmorillonite with normal thermical behavior, and with disordered (‘pedogenetical’) lattice. It constitutes 12-23% of the sediment. The illite, which appears in 9-12% is partly disordered, partly ordered, showing transition to hydromuscovite, as “herited” detrital mineral. Small quantities of montmorillonite-illite mixed layer mineral, of chlorite and of kaolinite are present, too.

The non clay minerals are represented by detrital (volcanic and metamorphic) quartz, by feldspars (microcline, orthoclase, albite, intermediar plagioclase), small quantities of muscovite, biotite, chlorite, amphibole, heavy minerals, lithic grains (rhyolite , pumice, andesite, sericite schists, sericite-chlorite schist, limestone, shale and carbonate concretion fragments, see plate I., photo 3). Small limonite and carbonate concretion, Ca- and Mg-sulphates as syn- and diagenetic minerals were identified, too (plate I., photo 4).

Our studies emphasize, that the original (parent) sediment of soil is the Holocene fluvial complex, in which are represented the weathering products and rock debris of all of geological formations from the Northern and North-Eastern Carpathians.

4. HYDROGEOLOGY

The groundwater table of the Kömlő model area lies at 2.5-3m depth, with max. ±1m seasonal variation. During the spring snow break or the hard rainfalls, on the Heves floodplain (including our sampling area) large inland water ponds may be formed. The direct connection between the surface water and the groundwater table is not certified yet by chemical analyses. During the summer, a part of inland water dry and in some zones white salt powder covers the soil furrows i.e. the epsomite-glauberite-halite-gypsum salt association (Plate I., photo 2.). The rest of this rainwater accumulations percolate leaching the sediments and transporting certain elements in deeper zones.

The composition of groundwater (Table 1) is characterized by dominance of Mg^{+2} and of SO$_4^{+2}$ ions; while the Na$^+$ Ca^{+2}, respectively the Cl$^-$ ions play a secondary role. Therefore, the origin of Mg-salts seems to be the capillary groundwater “pump” toward the surface.

Table 1. Groundwater analyses from several boreholes of Kömlő model area

Sample	Ko-1	Ko-4	Ko-13	Ko-18	Ko-25
Na$^+$	171.00	117.00	281.57	182.95	106.00
K$^+$	0.33	0.67	0.41	0.60	1.00
Ca^{+2}	220.07	127.00	167.00	145.00	363.12
Mg^{+2}	118.00	122.00	290.68	362.51	134.00
Fe^{+2}	0.03	0.06	0.10	0.26	0.04
NH$_4^*$	0.12	0.14	0.14	0.10	0.16
Mn^{+2}	0.05	0.01	0.01	<0.01	0.01
Cl$^-$	227.67	271.50	114.00	263.59	81.00
HCO$_3^-$	538.63	402.60	351.37	338.55	327.57
SO$_4^{-2}$	396.00	489.00	384.00	417.00	484.00
NO$_3^-$	34.85	162.00	150.00	114.00	181.00
NO$_2^-$	<0.10	<0.10	0.10	<0.10	<0,10
PO$_4^-$	0.12	0.63	0.09	0.06	0.20
Total	1706.87	1692.61	1739.47	1824.62	1678.1

5. GEOCHEMICAL SAMPLING: METHODS AND AIMS

All of 25 boreholes were sampled for grain size, carbonate and pH, resulting 9-12 samples for each borehole. Samples for mineralogical studies, including X-ray and DTA were taken, too. The sediment column was sampled for geochemical analyses (i) sediments from the arable level (0-30 cm); (ii) from the root zone (max. until 1,6 m); (iii) from the oscillation zone of groundwater table (1.5-2.0 m) and (iv) from the permanently, groundwater covered zone (Kuti, 2009).

Table 2. Analytical data of main and trace elements

Elements	Clay+silt, ppm		Sand, ppm	
	Average	Dispersoin	Average	Dispersion
Al	18,523.48	4747.88	12,846.29	4860.09
As	5.42	1,72	5.09	10.86
Ba	165.92	113.20	79,18	25.81
Ca	20,210.87	28,479.92	20,563.55	17,483.74
Cd	0.18	0.06	0.27	0.28
Co	10.46	5.67	7.57	2.99
Cr	75.44	91.79	128.34	185.87
Cu	74.64	5.72	13.62	24.59
Fe	27,743.48	5553.20	23,970.16	18,185.69
K	3620.98	1344.08	3015.66	830.23
Li	16,09	3.95	11.51	4.24
Mg	9059.78	6128.95	9630.92	5092.17
Mn	540.33	237.33	330.13	193.00
Mo	1.05	1.00	1.85	2.24
Na	684.02	404.00	831.45	406.05
Ni	35.95	25.57	30.65	34.73
P	556.60	184.45	454.86	206.52
Pb	18.88	4.92	12.53	5.45
S	229.30	306.82	117.54	49.65
Sr	52.64	37.37	44.70	22.53
Ti	63.77	55.46	52.05	43.59
V	33.21	8.05	22.13	8.73
Zn	65.31	14.25	48.90	18.10

5.1. Analytical results

For our geochemical studies, the analytical results of 23 elements of 108 samples were used. For the research area, the distribution of these elements (i) related to sediment types, (ii) to the soil levels and (iii) to their horizontal distribution pattern on the 250×250 m area will be examined.

Showing the magnitude order of the concentration of analyzed elements, we present the minimal, the maximal and the average concentration, as well the value of their dispersion.

In table 2 may be observed three groups of elements: (i) the main, rock forming elements with concentration from thousand to hundred fifty thousand (0.1-15%) ppm, as Al, Ca, Fe, K and Mg; (ii) trace elements of which concentration varies between a few ppm and thousand ppm, as As, Ba, Co, Cr, Cu, Li, Ni, P, Pb, S, Sr, Ti, V and Zn, and (iii) some elements with concentrations mainly below 1 ppm, as Cd and Mo, close to the detection limit, for which the analytical error may be greater, that their measured concentration in sediments.

Statistically, Al, Ca, Fe and a part of trace elements (as As. Ba, Cu, Mn, Ni, P, Sr V and Zn) show normal distribution; a few of its come close to the lognormal distribution, while the rest of elements as Co, K, Li, S, Pb and Ti (Fig. 2) present irregular or hyperbolic distribution.

5.2. Trace elements and the sediment type.

From geochemical point of view, two main sediment types were recognized: the clay rich and the clay poor (mainly sandy) sediments.

Comparing the individual samples, the average content and the dispersion for analyzed elements, the difference between them seems to be evident: the clayey-silty (foodplain) sediments contain between 10-50% more trace and nutrient elements, comparing with the sandy (riverbed) sediments.

There are exceptions: among the main elements, Ca, Mg and Na; among the trace elements: Cr, Cd and Mo.

If the <<1 ppm differences of Cd and Mo contents are close to the analytical error domain, the increased Ca, Mg and Na contents of sands are coming from the presence of alkaline feldspars (mainly albite and sanidine), of acid glass or pumice and of carbonate rock (mainly Eocene limestone) as lithic fragments (Plate I., photo 3). In our mineralogical studies in all of sand samples these clasts were recognized. Chrome may be related to the rare (spinel?) grains mentioned in our analyses as “euhedral dark minerals”.

In the clayey-silty sediments, with the increased iron content the trace element content increased with 5-25%. In sample No. 2305, described as “grey, plastic clayey silt with reddish brown films and patches”, the ~4% iron is present as finely dispersed hydroxide, while in sample No. 704: “yellowish grey, fine sand with 1-2 cm lens like and irregular hard, dark brown limonite concretions” on the contrary, the 15.5% Fe content make to decreases

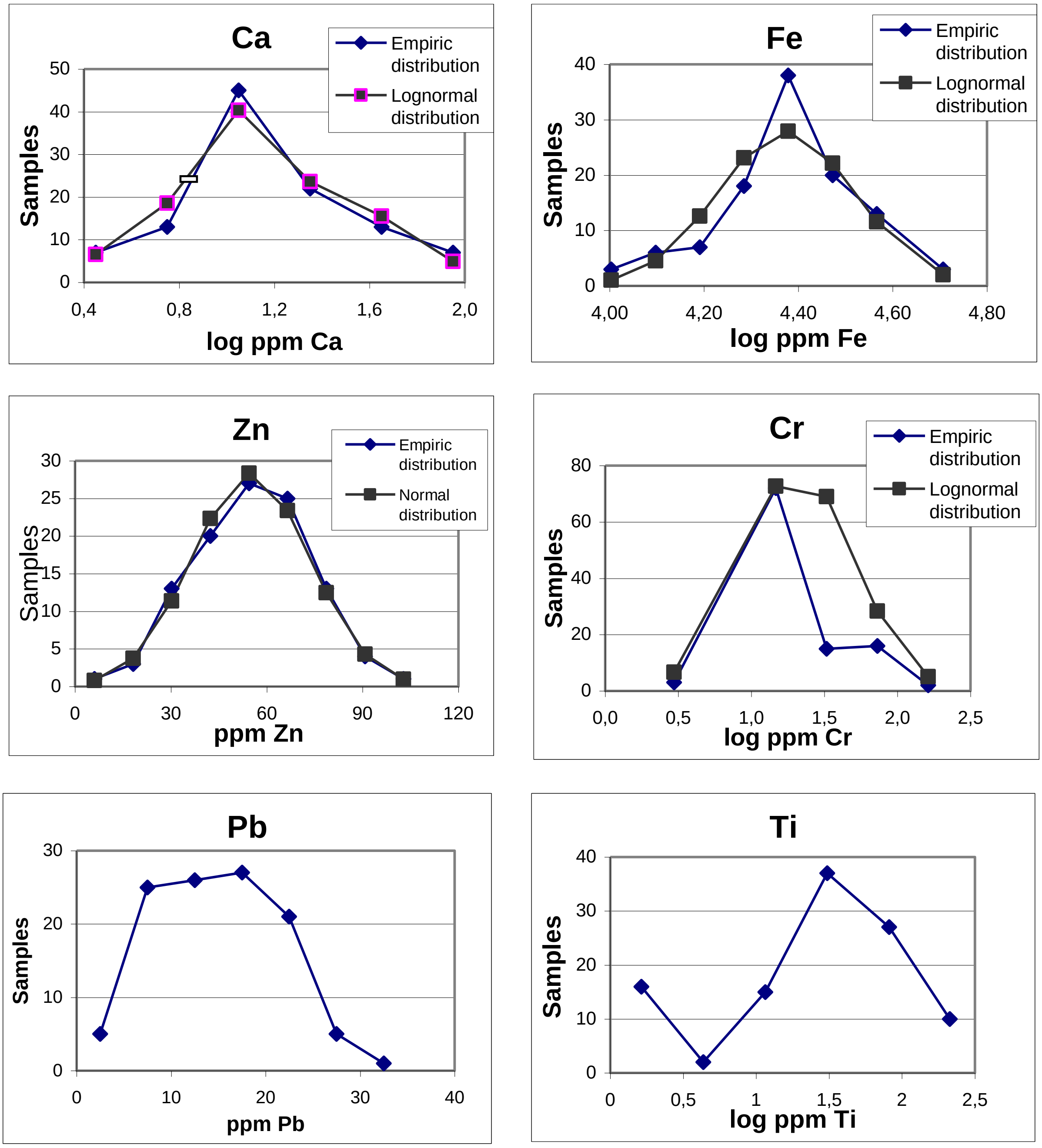

Figure 2. Distribution diagrams of some elements. lognormal distribution (Ca, Fe); normal distribution (Zn); close to lognormal distribution (Cr), irregular (Pb) and bimodal (Ti) distribution.

with 5-15% the concentration of trace elements, including some siderophyle ones, as As, Co, Cr, Ni and V. Concluding, the “host” mineral phases for trace elements is, beside of clay minerals, the dispersed iron hydroxides It is known, that the well crystallized iron hydroxide minerals as goethite or hydrohematite bring forth slight quantities of these elements (Schwertmann & Cornel, 2000). In photo 4 from the plate I., a concretion body with zonar structure was represented, with hard crust having fibrous structure.

5.3. The elements and the soil levels

5.3.1. The main (nutrient) elements

The concentration of the main and trace elements varies in vertical sense, diminishing from the surface (plough level) to the root zone and to the ground water covered zone (Fig. 3). From agricultural viewpoint, the variation of the nutrient elements as K, P, S and as well, of Mg and Ca is important to known.

The K extracted by *aqua regalis* leaching represents the adsorbed and mineralogical fixed

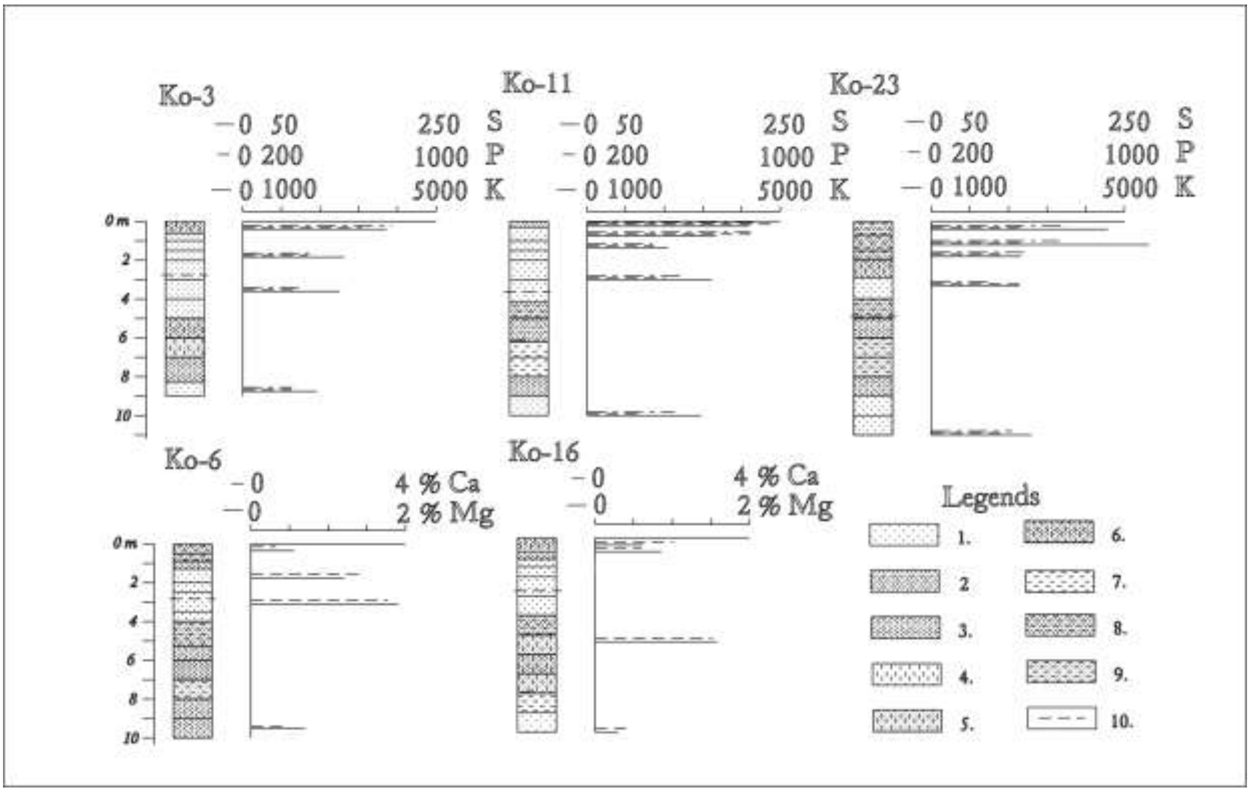

Figure 3. Nutrient elements in sediments in some boreholes (Ko3…Ko23). Legends: 1. Sand; 2. Silty sand; 3. Clayey sand; 4. Silt; 5. Sandy silt; 6. Clayey silt; 7. Clay; 8. Silty clay; 9. Sandy clay; 10. Hydrostatic level.

potassium, excepting a part of K content of feldspar grains. The contribution of plant rests in this concentration is unknown yet. The concentration decreases in depth, often till half of the content of ploughed layer and seems to stabilize around 3000 ppm below groundwater table.

The phosphorus represents almost all of organic and inorganic phosphate content of sediment. A negligible part of P tied in some heavy minerals is missing here. The relatively high contents of surface samples decrease more, that two times in the root zone and below the groundwater table.

Despite of presence of some sulphatic minerals at the surface, the S content of arable soil layer is low, under 0.1%. The iron sulphides, evidenced in X-ray analyses in the grey clayey sediments seem to be not enough for compensate the decrease of S contents in the depth.

The acid soluble Mg and Ca come from carbonates, sulphates and from adsorbed ions of clay minerals, as well Mg from phyllosilicates as biotite and chlorite. Both elements show an accumulation zone in the pH-changing level (Merin, 1976) i.e. at 1-3 m depth, and low contents, below 1% on the surface and in groundwater covered sediments.

5.3.2. The trace elements

It is known, that the trace elements having low concentrations cannot form individual mineral phases. Excepting some Ti oxides and probably, the barite (clastic) grains, they are hidden in rock forming minerals: iron oxy-hydroxides, carbonates, sulphates and silicates, and as well, in living and/or decomposed organic matter. It is evident, that in sediments, as well as in lithified rocks there are affinities of certain trace elements to one or other main “host” element.

Because in our samples, both the main and trace elements show normal (lognormal) distribution or close to these ones, it is possible to prove the liaison of two elements considering the linear correlation between them. In case of positive correlation coefficient *r*, which satisfy simultaneously two (e.g. *t* and *Romanowski* S_r) significance criteria, the element pair may be considered as forming a geochemically dependent system (Fig. 4). The classic example is the system Ca–Sr (r=0.915; S_r=9.349) or Fe–V (r=0.959; S_r=16.209).

The “attachment” of trace elements to the “host” ones is influenced mainly by pH and redox potential values and as well, by mobility of the host element-trace element system. The mobility of these systems is tied to the capacity of some elements to form mineral or organo-mineral complexes.

In figure 5, we present these interrelations in four situation: up to groundwater table: (i) on the surface (ploughed layer), (ii) in deep (root) zone, and below groundwater table: in (iii) sandy sediments and in (iv) silty-clayey sediments. The big circles represent the main elements, which carry with them the trace elements marked by small circles.

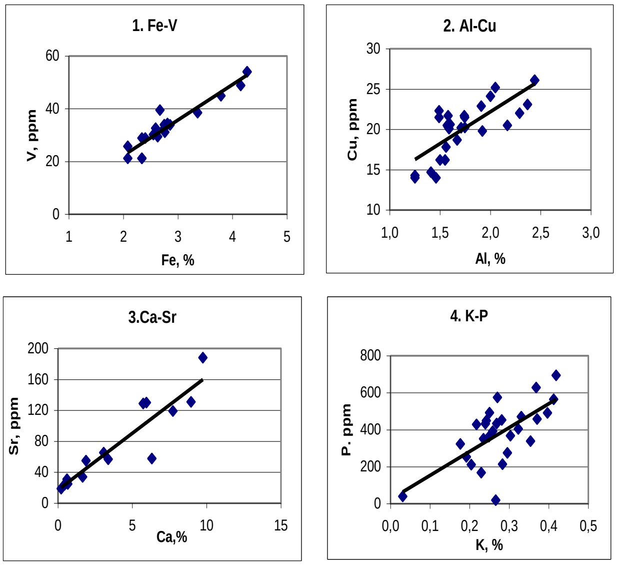

Figure 4. Correlation diagrams. 1. Between Fe and V in plough zone; 2. Between Al and Cu in root zone; 3. Between Ca and Sr in clayley-silty sediments below the grounwater table; 4. Between K and P in sand, below the grounwater table.

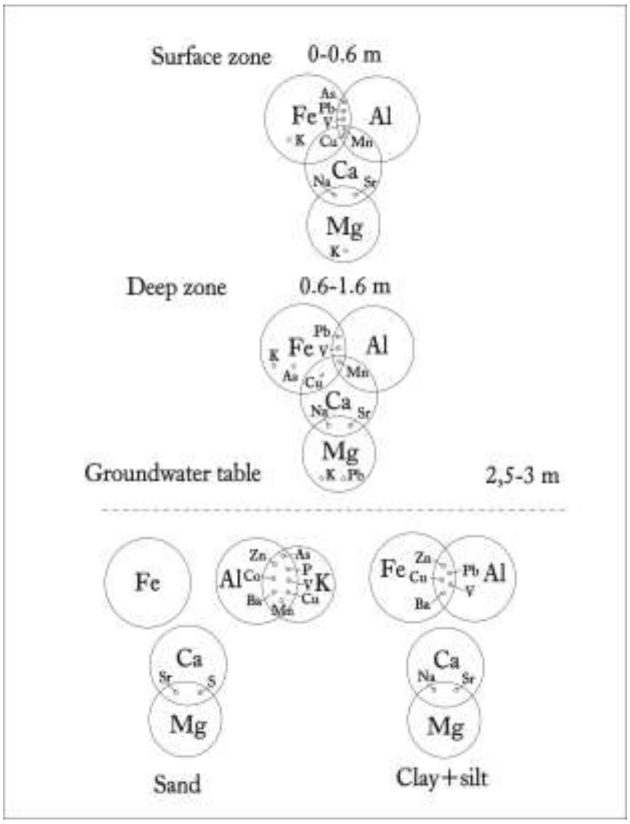

Figure 5. The trace elements (small circles), which correlate with main elements (big circles) in different levels of the lithologic column.

Up to groundwater table, in case of the sandy and clayey sediments, the differences for majority of elements are inscribed in analytical error domains. Furthermore, the separation of sandy and clayey-silty sediment types in any cases is not sure, because the randomly mixed, disturbed sediments.

In surface zone, including the ploughed layer and the soil level traversed by the roots the Fe-Al-Ca triad keep together almost all of trace elements and potassium, while Ca and Mg correlate only with Na and Sr. In this zone, upon the natural geochemical background, the results of biological and of agricultural activity were superposed.

In deeper zone, which corresponds to D soil level ("parent sediment"), the iron and the aluminum "share" these common trace elements: As and Cu pass to Fe, while Mg gains Pb. It is probable, that this zone conserves the initial (both sedimentary and geochemical) situation, before the beginning of the agricultural activity.

Below the groundwater table, a part of sediments has clayey-silty character, and the other part is represented by fine or coarse sand. The last one belongs to well distinguish riverbed facies of Old Tisa river system.

Therefore, in water saturated sand, the iron is free to correlable trace elements. As we has discussed above, during crystallization, the iron minerals eliminated these ones. They were hosted mainly in Al-rich mineral phase: phyllosilicates, with or without K. In this level, Ca and Mg correlate with Sr and S and K with P (muscovite with apatite inclusions?). In this situation, is important to know, the main character of groundwater is magnesian and sulphatic.

On the other hand, in clayey-silty sediments, the Fe and Al correlate and both carry with them Ba, Cu, Pb, V and Zn, while the other trace elements are dispersed randomly in different mineral phases, including the pore- and hygroscopic water. Ca and respectively Mg keep Na and Sr without correlation with Fe and Al. Here, because the presence of water in sediment pores the "united behavior" of main elements was brook up.

5.3.3. Horizontal variation of some trace elements

Because the Kömlő model area is situated far to industrial and communal pollution sources, and the use of the chemical fertilizers was ceased almost twenty years ago, the trace element contents of sediments depend only to the geological-pedological factors and may be considered as (local) geochemical background.

Knowing the regional and local variability of background values in Hungarian main geological regions (Fügedi et al., 2012), we tested the homogeneity for eight elements for 0.0-0.6 m and 0.6-1.6 m, drawing isoline maps for each element (Figs. 6 and 7).

The first observation is that the range of concentrations, i.e. the difference between the positive and negative anomalies (excepting As, Mn and Ni) do not pass up 60%). In 5–20 ppm concentration values, these anomalies is the same magnitude, that the analytical error and/or the geological variability of the sediments.

It is evident, that the concentration of elements decreases in depth with 20-50% both in minimal zones and in the maximal ones. The single exception is the manganese in northern corner of the model area.

An other observation is the different pattern of iso-lines for surface and deep zone (Figs. 6 and 7), passing from slightly similitude (Ba, P) to complete difference (As, Ni). Nevertheless, the main positive and a part of negative anomalies appear in same site; indeed, they appear in same two or three sites for different elements (e.g. on the northern corner,in same place As, Mn., Ni and Zn positive anomalies appear).

The statistical measurement of iso-lines shows two preferential directions: the main N-S direction and the secondary, NW-SE direction. We note, that the buried sand bar which culminates in the 92.3 m high hillock has approx. N-S orientation, similarly to the old Tisa branches, evidenced by grain size analysis of sand levels (Kuti et al., 2007). In this case, the main direction indicates the mean current line, while the secondary direction is due to water (and mud) movement during the withdraw of muddy water in riverbed during final phase of the floods (see Walker & Douglas, 1984).

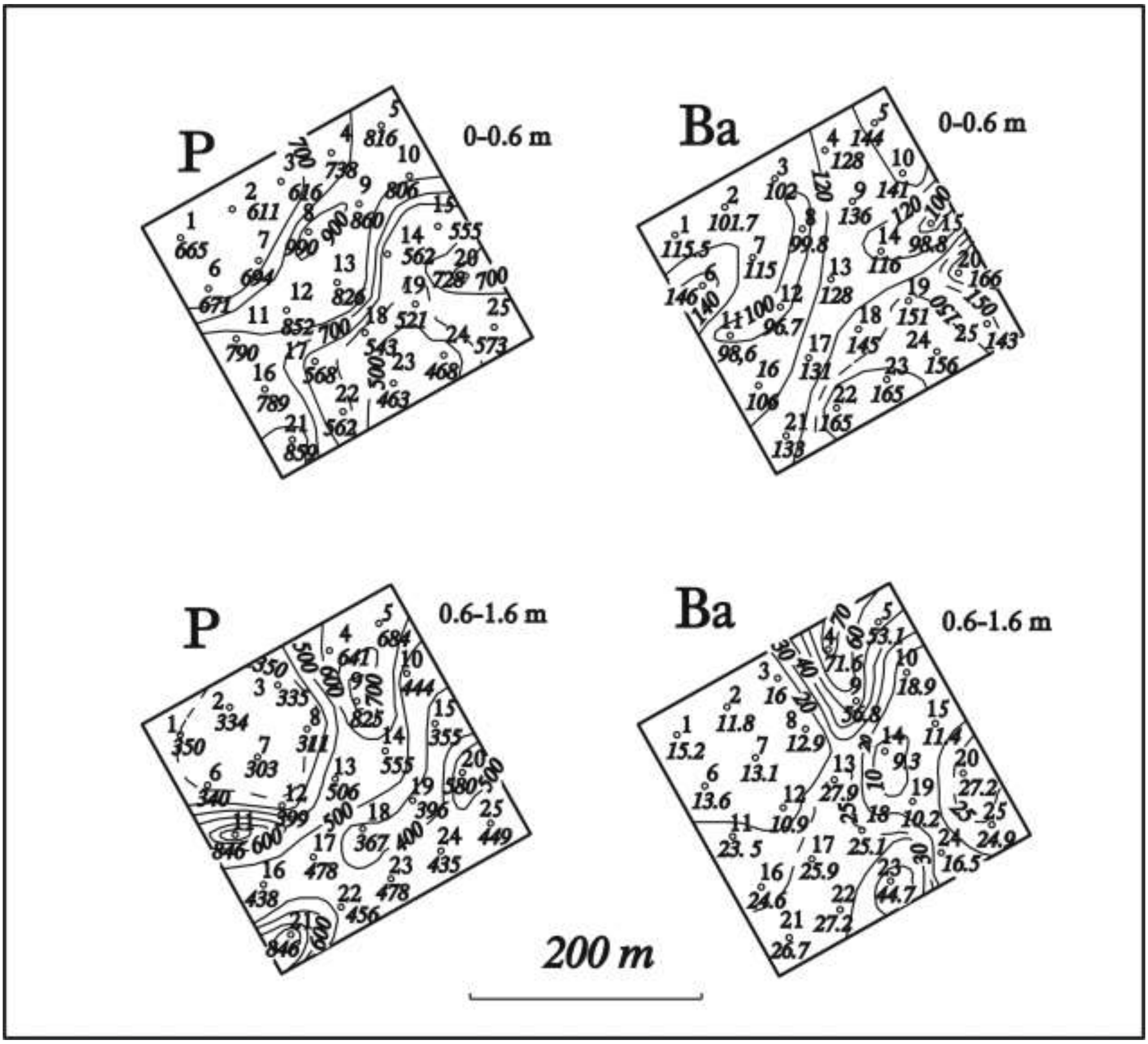

Figure 6. The areal distribution of Ba and P in sediments, between 0.0-0.6 m and 0.6-1.6 m depth

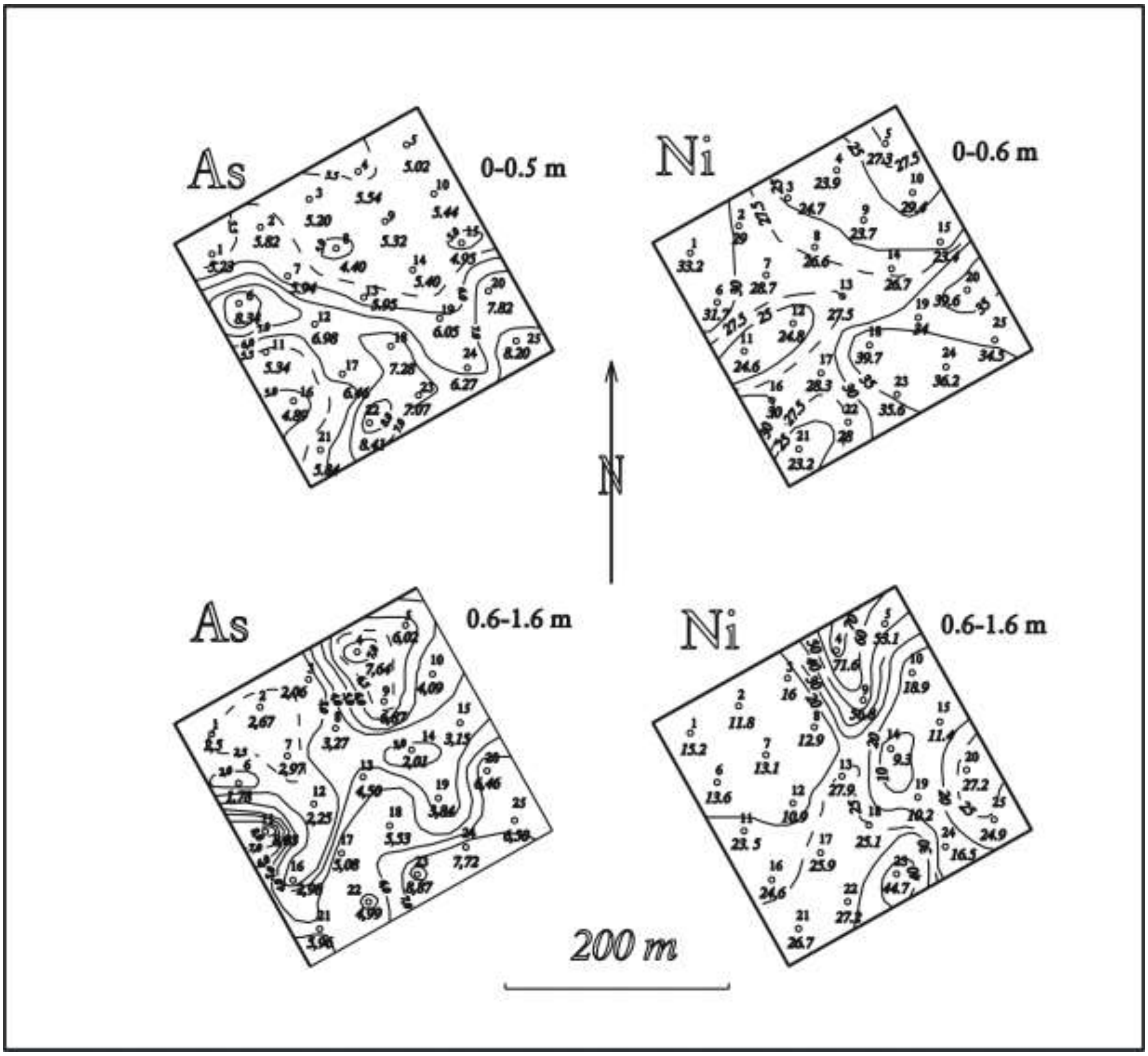

Figure 7. The areal distribution of As and Ni in sediments, between 0.0-0.6 m and 0.6-1.6 m depth

6. CONCLUSIONS

In Kömlő model area (Heves plain, Hungary), the distribution of the *aqua regalis* soluble 23 main and trace elements and their mineralogical, sedimentological and statistical relationship were examined. The concentration of these elements depends mainly to the lithology of the sediments, i.e. their grain size: the sandy sediments show lower concentration, that the clayey-silty ones, excepting Ca, Cr, Mg and Na.

The concentrations decrease in depth, excepting Ca and Mg, which form an accumulation zone up to the groundwater table, i.e. in the oscillation zone of the hydrostatic level. The minimal concentrations of elements are observed in water saturated zone, both in sandy and clayey-silty sediments.

From agricultural point of view, it is important to observe the concentration of the nutrient elements (excepting Ca and S) close to the surface, both in ploughed layer and in zone of roots. After the published data of Loch & Nosticzius (2004) for Hungarian soils, in this area the Ca, K, Mg and P contents satisfy the necessity of plants. Only the S most is completed by fertilizers.

Examining the correlation between the main and trace elements, we concluded, that at surface, by the biological processes, an increased concentration of elements were resulted. Therefore, in deeper zones, the concentrations represent the natural, geological background values, close to Salminen's (2005) values for floodplain sediments.

Under groundwater table, a diminishing of element contents is observed, caused, mainly by leaching and by weak content of host mineral phases: phyllosilicates and well crystallized iron hydroxides.

The horizontal distribution of some trace elements both at the surface and in depth show small amplitude variation between the maximal and minimal values. They depend on the internal structure of these sediments, resulting from stream direction of water and mud during floods of Tisa River in the past.

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