

## Regional patterns of the accumulation of toxic and nutrient elements in the superficial formations of Hungary

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South of the Sudetes and Tatra Mountains young sediments contain more of almost all micro- and mesoelements than those to the north of the Carpathian Mountains, which were covered by ice during the Late Pleistocene. As detailed investigations show this larger unit is a collection of many individual geochemical regions, among which there are four on the area of Hungary: the region of soil calcification in the central part of Hungary, the slope debris of the foothills of the Eastern Alps, the floodplains of the rivers of Eastern Hungary that are contaminated with the waste material of heavy industry, and the rest of the country. No general geochemical background can be given to the whole; the background value intervals for each element shall be given for each geochemical region separately. In Eastern Hungary, where those rivers flow that originate from the heavy industrial centers of Transylvania and Slovakia, the toxic elements can locally exceed the limit value. In the central part of the country, however, the micro-elements are frequently found below the minimum concentration required, since a carbonate accumulation zone develops above the groundwater level and the carbonate minerals contain low amounts of nutrient elements.

Key words: trace elements, geochemical maps, background level, regional anomalies, heterogeneity, stream sediment, floodplain sediment, soil calcification

### ***Soil-forming sediments and their origin in Hungary***

Geographers divide the loose plains and hilly regions of Hungary into five separate units (Pécsi 1967): Tisza Plain, Danube Plain, Little Plain, West Hungarian Borderline and Transdanubian Range. The near-surface and superficial formations of these re-

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gions are usually young, loose sediments, from the uppermost part of which soil forms. These regions are the main areas of agricultural production of Hungary.

The three plains are accumulation regions where rivers coming from the surrounding mountains deposit their alluvium. In regions where the groundwater level is located near to the surface the sediments could have undergone salinization.

According to the same type of division, there are two mountainous areas in Hungary: the Transdanubian Mountains and the North Hungarian Range. The mountains are predominantly built up of older, Mesozoic rocks, while the intermountain basins are filled with younger sediments. On the hard rocks mostly younger sediments are deposited with variable thicknesses.

With regard to agriculture, forestry and environmental geology, the loose sediments are of importance in the mountainous areas as well.

#### *1. Danube Plain*

The largest area of blown sand of the country is situated above the ridge of the Danube–Tisza Interfluvium where the thickness of the blown sand often exceeds 10 m, though usually it forms 2–3 m thick beds that alternate with loess beds of similar thickness. Holocene lacustrine sediments occur between the loess ridges and sand dunes.

In the valley of the Danube superficial clay is deposited on the coarse stream sediments (sand or gravel). In many places fluvial or fluvio-eolian sand can also be found on the surface.

#### *2. Tisza Plain*

The bulk (70%) of the Tisza Plain is covered with fluvial sediments. The surface is covered with clay of varying thickness, under which a succession of diverse fluvial sediments is to be found, represented by silt, sand and gravel (Horváth and Ódor 1977).

Blown sand is the characteristic superficial formation in the northeastern part of the Tisza Plain, called Nyírség. The loess is present on the surface only in smaller patches in the northern marginal parts. The loess (as well as the blown sand) has low carbonate content, in contrast with the loess found in other parts of the country.

#### *3. Little Plain*

Sediments are diverse in the Little Plain; clay, silt, sand and gravel can occur, depending on how the Danube and its tributaries have moved, and what kind of sediments have been deposited. This diversity can be traced with depth. Everywhere, however, the lowermost member of the loose sediment succession is gravel.

#### *4. West Hungarian Borderline*

Plain surfaces are covered with fluvial sediments. The dominant land type in the western parts is ‘clay-covered’, while in the eastern parts are ‘sand-covered’.

In the Sopron Mountains mainly Ordovician–Lower Carboniferous metamorphic rocks occur. In the Lower Miocene coarse clastic sediments with brown coal seams

(Brennberg) and (due to a transgression event) open-marine basinal clay, clay-marl and Lajta limestone beds were deposited. Above them sandy-pebbly sediments and biogenic limestone are to be found.

The Kőszeg Mountains and the Vashegy are made up of metamorphic rocks (quartz phyllite, conglomerate, calcareous phyllite, greenschist, and ophiolite) that had been previously assigned to the Paleozoic era. According to recent research, they are of Mesozoic age. In the foreland of the mountains Upper Pannonian sediments (clay, sand, gravel, and lignite) are known to occur.

##### *5. Transdanubian Hills*

The surface of the eastern part of the loose sedimentary lands of the Transdanubian Hills is covered with silt and loess, while the western ones are covered with blown sand. With the exception of smaller peripheral areas, finer sediments are only characteristic in the valleys of the loessic and sandy hills.

From the areas covered with sedimentary rocks the Mecsek and the Villány Mountains rise, primarily composed of Late Paleozoic clastic and Mesozoic carbonate rocks.

##### *6. Transdanubian Range*

The oldest known formations of the mountains are Late Paleozoic epimetamorphic sedimentary rocks that contain volcanic interbeds. The Devonian succession is comprised of limestone and clay slate. As a result of the Carboniferous orogeny, uplift and denudation, continental-fluvial conglomerate was formed. During the Permian red fluvial sandstone was deposited on older formations, while in lagoons evaporite and dolomite were formed. The granite mass of the Velence Mountains was intruded in the beginning of the Permian.

During the Mesozoic mainly carbonate rocks were formed (Less 2007).

In the ingression marshes of the Eocene launching the Cenozoic, and in the boggy-marshy shores, coal measures and (in the inner part of the basin) a series of marl and limestone were deposited (Hartai 2005). In the Velence Mountains andesite intruded into the granite. During the Oligocene, in the foreland basins continental sediments were deposited with brown coal seams followed by the deposition of the diverse formations of the strongly broken shorelines of the Miocene – in places with coal seams, in other parts with reef limestone bodies, and in the basins, with clay and schlier. In the Miocene, three levels of large explosive volcanism can be distinguished. The tuff has been preserved on the limestone plateaus as well. Clay and sand of the Pannonian stage is characteristic everywhere in the peripheral areas. Basalt volcanoes were active and in the crater lakes alginite was formed. Typical formations of the Pleistocene are loess, blown sand, fluvio-eolian sand, travertine, calcareous tufa and along the rivers, gravel terraces.

### *7. North Hungarian Mountains*

The mountain range east of the Danube comprises mountains and intramontane basins. Proterozoic rocks occur near to northeastern part of the border.

In the Late Paleozoic limestone, marl, and terrigenous sediments were deposited, locally with evaporite.

In the Triassic and the Jurassic varying marine sediments were deposited, while in the Cretaceous the entire area was uplifted.

In the Cenozoic the area was flooded several times by the sea. Andesite volcanism occurred twice, first during the Eocene and later during the Miocene. Volcanic activity continued until the Pannonian, resulting in the emplacement of andesitic-rhyolitic rocks (Hartai 2005). Since the last regression event of the Middle Miocene no sediments of normal salinity have been formed in Hungary (Less 2007).

During the Pannonian lacustrine-marshy facies were formed. Locally basalt volcanoes were active at the end of the Pannonian.

### ***The process of soil formation***

The soil is the uppermost sphere of the Earth's crust that supports plant life (Stefanovits 1975). An important feature of soil is fertility. Among other things, fertility depends on the geologic media it was formed in.

Soil formation is determined by the soil-forming sediment. The material, mineral composition, physical and chemical properties of the soil-forming sediment decisively influence the type and quality of soil formed from it under the influence of other factors (vegetation, climate, topography).

Soil-forming sediment is defined on plains and in hilly regions as the loose sediment on the surface, which is the upper layer of the superficial and near-surface rock facies, and in mountainous areas as the upper layer of the sediment succession that is situated over the bedrock undisturbed by surface processes. This implies that different processes and effects are related to surface and near-surface formations of plains and hilly lands than those related to the mountainous areas. Under the influence of different soil-forming factors and processes, the sediment becomes soil. Considering its origin, these sediments can be autochthonous or allochthonous (Kuti et al. 2007b).

In plain areas the near-surface formations must be studied down to the saturated zone, but at least to a depth of 10 m (Bartha et al. 1987). In areas of mountains and foothills, the thickness, the composition and genetics of the young loose sediments situated above the older solid rocks are also of vital importance (Kuti 2009).

### ***Formation and redeposition of stream sediments, element accumulation and washout***

As a result of surface weathering, fragments of rocks enter watercourses to form fluvial sediment. The fragmenting rock may release ions into the water due to bio-

chemical processes. Later, under favorable conditions, the released ions can be adsorbed onto the surface of different fractions of the sediment, mainly onto those of the clay minerals whose quantity increases over time with weathering. Especially favorable for this process are those periods when floods disperse the sediment over the floodplain (temporarily). There, the sediment can undergo processes that result in the forming of soils. Later, with the development of the river, the sediment returns to the river to be transported further. Scientists studying the fluvial sediments of the Great Hungarian Plain (Horváth and Ódor 1977) found that given the current precipitation conditions, the Tisza River totally re-accumulates its load approximately every 1,500 years. This corresponds to the cycle length of soil formation, and indicates well that the amount of certain ions in the stream sediments depends not only on the geology of the drainage basins but also on the manner of re-accumulation.

Croatian scientists studying the Žumberak Region (Peh and Miko 2001) compared the composition of sediment samples with two regression models in order to determine with which to better correlate the concentration of the elements in stream sediments: with their areal distribution (areal model) or with the length of their occurrence beneath the river bed (linear model). They found that the linear model is more appropriate to measure the concentration of those elements (the majority of them) that accumulate mainly in the solid phase. The length of the river bed section of a rock type characterized by higher concentration of elements correlates with the quantity of these elements in the stream sediment. Those elements that are redeposited in solutions are better described by areal regression (which shows the regional spread of those rocks on the drainage basin that contain these elements in a higher concentration).

The Geochemical Atlas of Europe was based on the investigation of drainage basins (Salminen et al. 1998). Stream sediment samples were collected from the outflow points of the small drainage basins (<100 km<sup>2</sup>) that are situated in a larger drainage basin (>1000 km<sup>2</sup>) described by floodplain sediments. The median data of these two different types of samples were compared (Table 1). The results show that the differences between the two types of sediments are minimal. Significant difference occurs only in the case of sulfur; the sulfur content decreases along flow direction, since clay minerals do not adsorb the dissolved anions.

A similar pattern has been found in Hungary. During the first phase of the geochemical mapping of the mountainous areas (Hartikainen et al. 1992) the microelement content of the four sampling agents (heavy mineral concentrates, stream sediments, composite soil and composite rock fragment samples) were compared. Results showed that the expected values are similar over the drainage basins while the variability increases from the heavy mineral concentrate to the rock fragment.

### ***Purpose and methods of geochemical mapping***

Geochemistry does not exclusively deal with the Earth, but also with the history and present of the whole Universe. As a consequence, its methods are excessively diverse,

encompassing methods from isotope geochemistry to electron microscopy. On the other hand, regional geochemical mappings always serve practical purposes.

Table 1  
Medians (mg/kg) of the element composition of floodplain and stream sediments in the Geochemical Atlas of Europe. Analysed following aqua regia digestion (Salminen et al. 2005a)

Element	Stream sediment	Floodplain sediment
As	6	6
Ba	86	82
Co	8	8
Cr	63	59
Cu	14	16
Mn	452	446
Ni	16	18
Pb	14	16
S	510	287
V	29	29
Zn	60	56

The differences of the purposes of geochemical investigation result in the differences of data structures. In the case of theoretical research, usually the precisely measured absolute concentration (or concentration ratio) of several elements or those of the isotopes of one or two elements is required. However, practical investigations require the examination of many samples (possibly for as many elements as possible), necessarily with less precision, but with relatively inexpensive analyses. During the second third of the 20<sup>th</sup> century two major schools of geochemical mapping had been formed: the ‘Soviet’ school, that processed the semi-quantitative analyses of many soil samples, and the ‘Western’ school, that dealt with the more precise analyses of far fewer, but more representative samples.

For a long time, the main purpose of regional geochemical research was to locate ore (mainly rare-earth element, non-ferrous metal and noble metal) deposits. This still applies to today’s geologically less well-known regions. However, in the industrialized countries of the world, the geologic knowledge concerning the upper part of the Earth’s crust has continuously increased (ore deposits that could be found by geochemical methods have been explored), and the need of environmental protection has been highlighted due to the increasing pollution. Therefore the role of regional geochemical research in the more developed regions has shifted increasingly from mineral ore exploration to the assessment of environmental status. Simultaneously, instead of ‘whole’ analyses, it became more important to study the element content able to be mobilized by various methods: in the field of environmental protection mainly aqua regia extraction is used (since it can be used to determine element content that can be mobilized over a longer period), while in agrochemical research the Lakanen–Erviö solution is preferred. The integrated geochemical atlas of Hungary was based on the analytical results of aqua regia extraction.

The floodplain sediments that represent large areas are suitable to represent both the original sedimentary environment and the current – occasionally contaminated – environment. For such investigations two samples must be collected from every sampling site. Samples from the lower zones represent the ‘original’, near-natural status of the sedimentary environment, and the denudation processes at the time of the deposition of the sediment. The effects of human activity (such as regional/local contaminations originating from agriculture and industry) are reflected by the upper 10 m part of the section. Obviously precise analytical methods are needed in order to detect low concentrations – especially in case of elements that show pollution with high sensitivity (like Cd, Hg, As).

The type of the media to be sampled primarily depends on the purpose and scale of the examination, since the areal representation capacity of the sample types is highly variable. The composition of soil samples – depending on several factors (like vegetation, micro-morphology etc.) – shows rapid spatial change. The results obtained through the analyses of soil samples taken on the Great Hungarian Plain can be extended around the points over a distance of 500–1000 m (Kuti 2009). This length is at least one magnitude shorter in mountainous regions. Moreover, the area to be described is thus prolonged downhill (Aristov 1984). Stream sediments and floodplain sediments may be representative even for an area of several thousands of km<sup>2</sup>.

### ***Regional geochemical mapping in Hungary***

After World War II the main aim of geochemical exploration in Hungary was to support mining operations. Therefore, geochemical mapping was focused on areas (as well as their immediate surroundings) considered to be prospective for a particular mineral (Gedeon 1964; Csongrádi 1984, and many unpublished reports).

The first geochemical exploration over a larger area was launched by MÁFI’s Department of North Hungary (Magyar Állami Földtani Intézet, Geological Institute of Hungary) in 1975. 800 × 800 m cells had been designated in order to collect samples from the A horizon of soils. The Bükk Range and the Darnó Hill were the designated sample sites in the first year (Böjtösné Varrók 1977); later exploration was continued in the Aggtelek–Rudabánya Mountains (Böjtösné Varrók, unpublished report). The powdered samples were analyzed by semi-quantitative optical emission spectrometry. Though the goal was still mainly ore exploration, it was also important to determine the concentrations of all micro elements above the detection limit. The exploration was unsuccessful due to several reasons not gone into here; the predicted Zn-anomaly proved to be a methodological mistake (Böjtösné Varrók 1974).

The research program (Böjtösné Varrók et al. 1970, and others) aiming to explore for rare earth elements was also inefficient, partly because there are no rare earth metal deposits in Hungary of economic viability; furthermore, it was ultimately revealed by this study that optical emission spectrometry is at best applicable to comprehensive testing, and even then only combined with other analytic methods. The last successful



example of this was provided by the geologic mapping (1:50,000) of the Central and Western Mátra (Nagy 1983); in this program many new ore dykes (non-ferrous metal) were identified by metallometry.

The “swan song” of optical spectrum analysis was the geochemical mapping of the Zemplén Mountains. This research served as the basis for those national mapping operations, the results of which could later be compared to those of other European countries. Apart from the search for ore resources, the main aim of this program was to survey the environmental status of the area. Thereafter the main purpose of geochemical research became the evaluation of environmental problems – solely by quantitative and satisfyingly accurate tests.

For the geochemical mapping of Hungary (Ódor et al. 1997) the floodplain sediments of 300–400 km<sup>2</sup>-large drainage basins were examined with the ICP-OES method, following an aqua regia digestion. Based on the principal components, the main geochemical regions of the country could be identified and the expected values of those micro elements that are significant with regard to environmental geology, could be detected.

For the geochemical mapping of the mountainous areas of Hungary (Fügedi et al. 2007) stream sediment samples from drainage basins of approximately 4 km<sup>2</sup> area were analyzed. We found that the expected values of the results of the two methods are very close to each other, but a greater variability was found in the stream sediment samples of smaller drainage basins (Tables 2 and 3). This combination of the two methods (large drainage basins by floodplain sediments, smaller drainage basins by stream sediment samples) was accepted by the work group responsible for defining the methodology of the Geochemical Atlas of Europe (Bølviken et al. 1990) as well.

We have been participating in the joint mapping operations of the Eurogeosurveys (formerly WEGS and FOREGS) from the very beginning. For the database and maps later published as the Geochemical Atlas of Europe, the continental area of Europe was divided into cells (Salminen et al. 2005b), corresponding approximately to a survey at 1:5,000,000 scale). The sampling grid was based on GTN grid cells developed for the purpose of Global Geochemical Baseline mapping (Darnley et al. 1995). This grid divides the entire land surface of the Earth into 160 km × 160 km cells. Where it was possible we took one sample from every cell from all the following sample types: humus, topsoil, subsoil, stream sediment, stream water, and floodplain sediment.

Similarities between the four solid sample media (subsoil, topsoil, stream sediment, floodplain sediment) were investigated by factor analysis (Batista et al. 2006). From Hungary we had to collect the samples of 15 cells and we managed to do it with satisfying efficiency (Table 4). The acid-soluble concentrations of the floodplain and stream sediments were integrated into the Geochemical Atlas of Hungary.

The most remarkable feature of the maps (Fig. 1) is the zone that separates the northern part of the British Isles, the northern part of the former GDR and Poland from the rest of Europe (the position of the line differs slightly for different elements and



Table 2

The expected value of the background (median, g/t) in floodplain sediments and in stream sediments on the Central Hungary geochemical region (after Ódor et al. 1997; Fügedi et al. 2007)

Element	Floodplain sediment (196)	Pilis-Buda Hills, stream sediment (50)	Gerecse, stream sediment (63)	Vértes, stream sediment (74)	Bakony, stream sediment (329)
As	6.3	7.4	4.7	4.7	4.8
Ba	68	78	56	58	62
Co	5.7	10.5	6.6	5.7	6.4
Cr	14.5	19	16	16	17
Cu	15	9	5.6	4.4	5.7
Hg	0.08	0.05	0.03	0.05	0.03
Ni	16	24	17	14	16
Pb	13	12.5	7	11	6
Zn	46	56	41	45	41

Table 3

The background maximum (g/t) in floodplain sediments and in stream sediments on the Central Hungary geochemical region (after Ódor et al. 1997; Fügedi et al. 2007)

Element	Floodplain sediment	Pilis-Buda Hills, stream sediment	Gerecse, stream sediment	Vértes, stream sediment	Bakony, stream sediment
As	13	20	15	10	15
Ba	158	200	200	>106	>230
Co	10	>17	20	>9	20
Cr	32	60	40	>32	60
Cu	33	25	>14	>12	30
Hg	0.37	0.3	>0.11	0.27	0.3
Ni	30	50	50	>27	50
Pb	23	40	>18	>37	40
Zn	180	200	>90	>80	200

Table 4

The number of different sample types collected in Hungary in the frame of FOREGS

Floodplain sediment	Humus	Subsoil	Topsoil	Stream sediment	Stream water
14	3	14	14	14	10

sample types). The younger soils of northern Europe are commonly depleted in many elements.

On the maps of the GEMAS (Geochemical Mapping of Agricultural and Grazing Land Soil; Reimann et al. 2009) (Figs 2 and 3) it can be clearly seen that this line continues east to Poland and into North Ukraine. This break in concentration coincides with the maximum extent of the last glaciations (Reimann et al. 2012).

No aqua regia digestion was used on the GEMAS samples; only the total concentrations were analyzed by X-ray fluorescence spectroscopy. Therefore these measurements could not be integrated into our atlas. On one hand the calculated principal com-

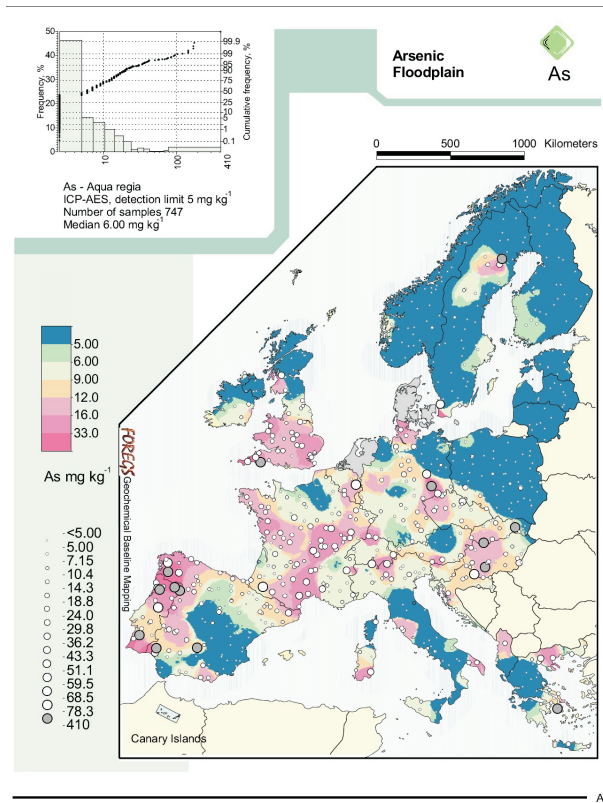


Fig. 1  
Arsenic (mg/kg) in the  
floodplain sediments of Europe  
(Salminen et al. 2005a)

ponents correlated very well with those distinguished earlier. On the other hand a new factor appeared unequivocally, indicating the accumulation of heavy minerals (placer mineral deposits) (Fügedi et al. in press).

The European surveys showed, as external independent examinations, that our conclusions on the geochemical regions of Hungary and on their features were correct. Moreover, they provide information on the continental background, on which those regional patterns show up that were derived from our more (one or two orders of magnitude higher) detailed surveys.

Apart from the geochemical mapping program, we also participated in the research program ‘Geochemistry of European Bottled Water’ (Fügedi et al. 2010; Reimann and Birke 2010).

### Data processing

The main task of geochemical research (be it ore indication or detecting the source of pollution) is to separate background and anomalies.

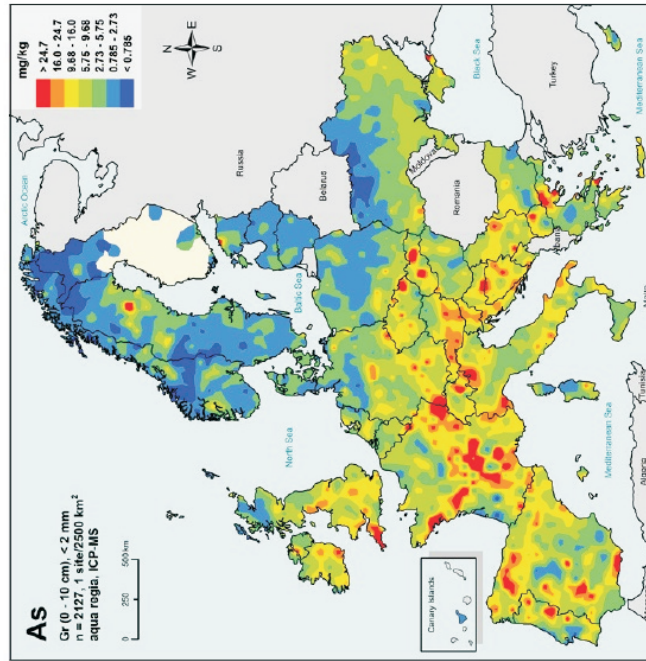


Fig. 3  
 Arsenic content of meadows and grazing lands in Europe (mg/kg)

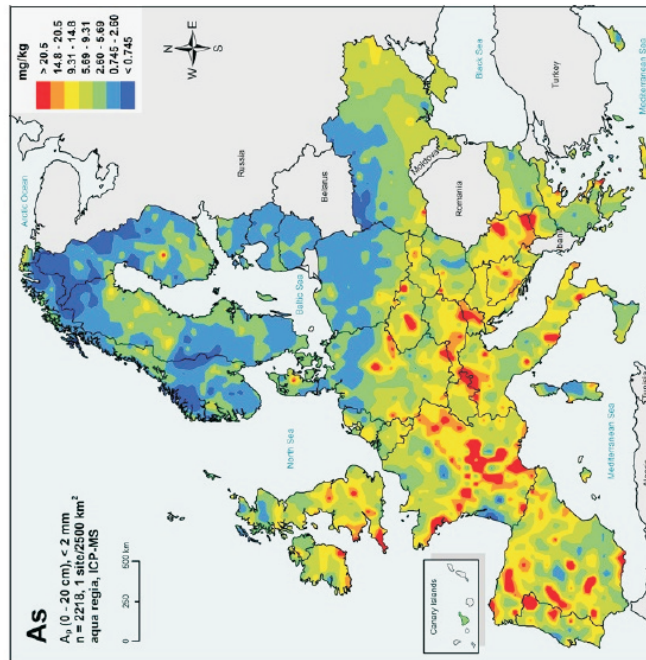


Fig. 2  
 Arsenic content of arable lands in Europe (mg/kg)

In our conception (Fügedi et al. 2012) the background and the anomalies can be interpreted only together, by focusing on the relation of one to the other. They can only be defined on the basis of statistical distribution patterns, always with regard to the possible heterogeneity of the background. The background can never be a distinct value, but rather ranges of concentrations. The heterogeneity of the background is always relative and depends strongly on the scale. Those areas that seem to be homogeneous at first sight, might fall apart during detailed research into smaller zones of anomalies. In cases (like in Hungary) where the distinguished groups of samples result from the same geologic processes on related surfaces or surfaces of similar genetics, they are not considered to be anomalies, but we can talk of background heterogeneity. In order to decide which one applies, one has to clarify the causes of the differences shown on the maps.

Though single-element maps depict the variability of concentrations, they provide only little help for data interpretation. In cases where the concentration of several elements changes mutually and significantly along a borderline (as seen on the maps of the Geochemical Atlas of Europe), given an in-depth knowledge of other fields of geology and environmental sciences, the interpretation of these data is reliable. However, in cases where there are only minor changes and at the given scale of research the changes are continuous, it is necessary to turn to multidimensional statistical tools.

In the case of a sufficient amount of data, factor analysis gives a result that is closer to reality than the principal components, since non-independent variables are present in the geologic processes and the concentration of certain element groups can vary significantly due to several geochemical processes. However, in the case of an insufficient amount of samples, the factors best matching the data do not really indicate certain geologic processes. Deleting or adding just one element in the input matrix can drastically change the results of factor analysis (Reimann et al. 2002).

### ***The geochemical regions of Hungary: specification and formation***

The four geochemical regions of Hungary (Fig. 4) were first identified in the Geochemical Atlas of Hungary, using principle component analysis (Ódor et al. 1997). Geologic processes were linked to the first four principal components (one for each) and on the basis of this the drainage basins were assigned to the geochemical regions (Table 5).

The mosaic-map depiction is misleading in case of Region 4, since here only the floodplain area of the main watercourse of the drainage area differs significantly from the geologic image of the other regions of the country. In addition, it should be taken into consideration, in the case of all regions and samples, that beside the dominant process, other geologic processes might be taking place simultaneously. For example the first principal component in every sample, and the second and the third one in the samples of the border zone of the central and the western regions.

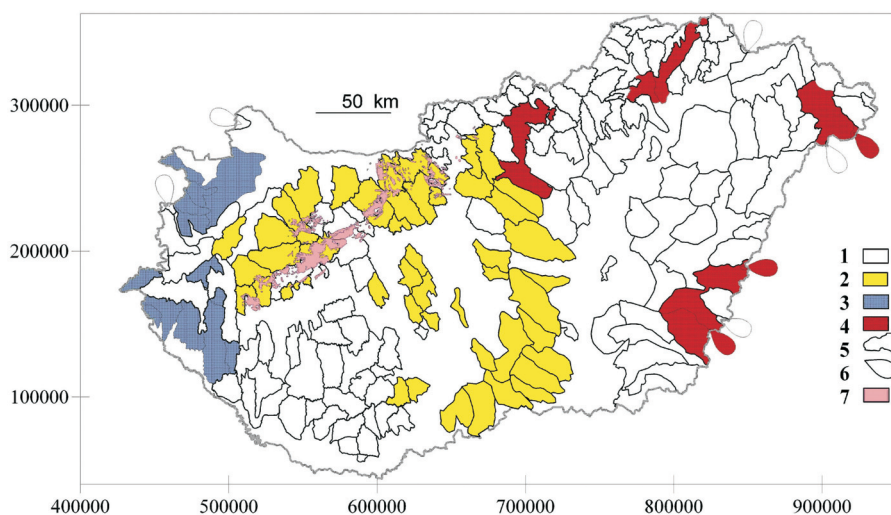


Fig. 4  
Geochemical regions of Hungary

Legends: 1 – Region 1; 2 – Region 2 (Central); 3 – Region 3 (Western); 4 – Region 4 (Eastern); 5 – Boundary of drainage basins; 6 – Drainage areas beyond the country's border; 7 – Distribution of friable carbonate sediments on the surface

Table 5  
Principal components with regard to the floodplain sediments in the Geochemical Atlas of Hungary (Ódor et al. 1997)

Principal component	1	2	3	4
Elements	Al, Co, Cr, Fe, K, Li, Ni (Cu, Pb, Zn)	Ca, Mg, Sr (PO <sub>4</sub> , SO <sub>4</sub> )	Co, Cr, Ni, Fe	Ag, Au, Cd, Hg, Pb (Cu, Zn)

The **first principle component** can be detected throughout the country, because it does not indicate any kind of specification, but a kind of (primarily) biogenic accumulation of elements due to pedological processes. In this principle component most of the examined elements show positive correlation. Not unexpectedly, the only exception is silica, since the two main element-accumulating phases of floodplain sediment successions are those of organic materials and clay minerals. These two usually correlate positively with one another; the humus content of cohesive soils is higher than that of loose, coarse-grained sands. Consequently, the sandier the floodplain sediment is (the more silica it has), the less total micro-element content it can be characterized with (Fig. 5). This applies to the stream sediments as well. Above the groundwater level, in the 3-phase-zone the conditions are often oxidative. In such cases limonite precipitates

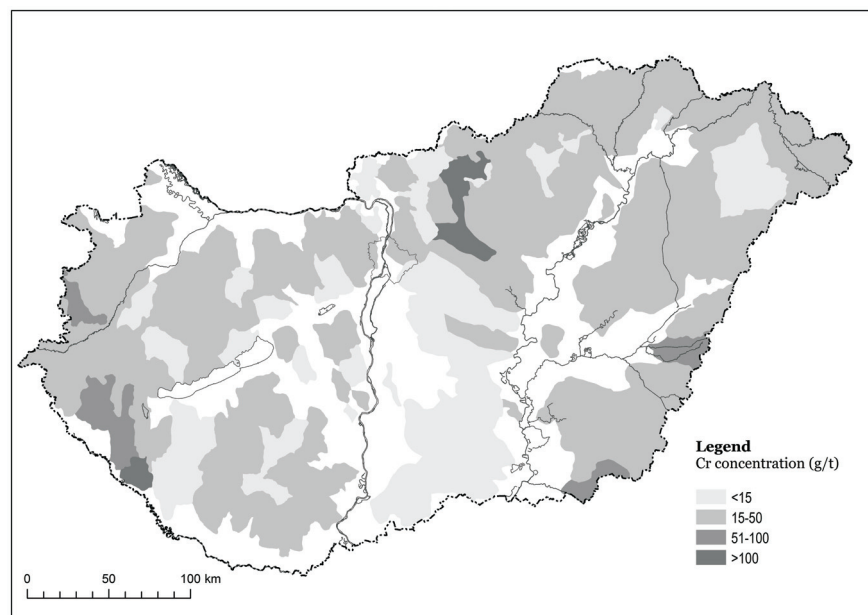


Fig. 5  
Chromium content (g/t) of the floodplain sediments of Hungary

in the open pores of the sand. In addition to iron, several other black metals (Mn, Co) (with variable valences) also enter the crystal lattice of oxy-hydroxides. The limonite, which has a large specific surface, adsorbs the arsenic from the temporary rising groundwater or the arsenic of precipitation.

Table 6  
Principal components of the Hungarian GEMAS samples (Fügedi et al., in press)

Principal component	1 (64.7% of total variance)	2 (11.3%)	3 (5.9%)
Elements	Si, Ti, Al, Fe, Mn, Mg, K, P, F, As, Ba, Ce, Co, Cr, Cs, Cu, Ga, La, Nb, Ni, Pb, Rb, Sc, Th, V, Y, Zn, Zr	Mg, Ca, Sr (SO <sub>4</sub> )	Hf, Zr

Both the clay minerals and the organic matter are characterized by the concentration of dissolved elements; those accumulating in solid form (minerals of placer deposits) can only be detected by analysis of total concentration values (Table 6).

The elements of the **second principal component** are the characteristic components of carbonate minerals and they are in negative correlation with all the other elements. This is the most important factor determining the concentration of the elements: communality is very high, with values above 0.6 both in the upper (0–10 cm) and the lower (50–60 cm) sampling level. The anomalies of this association outline a large



area in the middle part of the country reflecting the presence of calcareous soils. This geochemical region mostly covers the Transdanubian Range, the middle and eastern parts of the Transdanubian Hills, as well as the Danube Plain.

Molnár (1980) established that in the Danube–Tisza Interfluve there is considerable amount of carbonates (often 20–30%) in the sand and in the loess, respectively. Carbonate mud is also precipitated at the bottom of saline ponds in the hilly parts of the Danube–Tisza Interfluve. According to Kuti et al. (2002) the calcium and magnesium (necessary for the carbonate accumulation) accompanying the salinization in the groundwater fluctuation zone, are dissolved from clastic calcite and dolomite grains. Both the calcite and dolomite are crystalline; the grains corrode under subsurface conditions (Kuti et al. 1999). In the Trans-Tisza area, where no clastic carbonate grains are found, salinization is not accompanied by carbonate accumulation (Kuti et al. 2002).

The geologic causes of the carbonate accumulation in the soils in the middle part of the country were clarified by Fügedi et al. (2008). The blown sand forming the Danube–Tisza Interfluve Ridge is derived from the Danube alluvium and the carbonate minerals from the friable dolomite and limestone of the Transdanubian Mountains. The loess was carried to its present location by the wind in the periglacial area during the Ice Age. Three processes were responsible for the accumulation of most of the elements in the consolidated sediments: soil formation, development of small intermittent

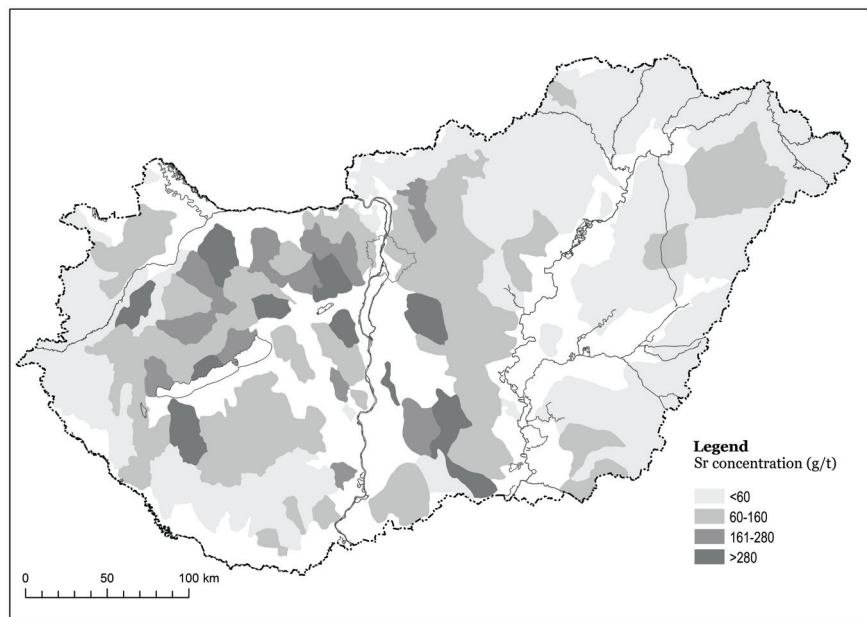


Fig. 6 Strontium content (g/t) of the floodplain sediments of Hungary



lakes with no outlet, and capillary water movement. These processes depend on the composition of the sediment, the precipitation conditions and the groundwater level.

Corroded carbonate crystals have a relatively higher Sr content than those originally deposited ( $\text{Ca/Sr} = 410$ ), while those of the precipitates from evaporated capillary water are lower ( $\text{Ca/Sr} = 730$ ), as calcium is more readily dissolved out of wind-blown carbonate crystals than strontium (Fig. 6). The amount of calcium in the carbonate mud of lakes and in the carbonate accumulation horizon below the paleosoil horizon is also relatively low, since it is more intensely absorbed by plants than magnesium or strontium in the course of syn- and anadiagenetic processes.

The **third principal component** typically assembles elements characteristic of basic magmatites. These are present in the western borderline of Hungary (West Hungarian Borderline and the western part of the Transdanubian Hills) in a concentration higher than what is typical for other regions (Fig. 7). The West Hungarian Borderline is the eastern margin of the Pleistocene alluvial fan of the Alps. The material of the alluvium has been settled by creeks and smaller streams regularly changing the position of their beds (Kuti et al. 2007a). In the stream sediments of the watercourses dissecting the basic and ultrabasic magmatites of the Eastern Alps, the concentration of black metals is higher than what is typical for the other parts of the country.

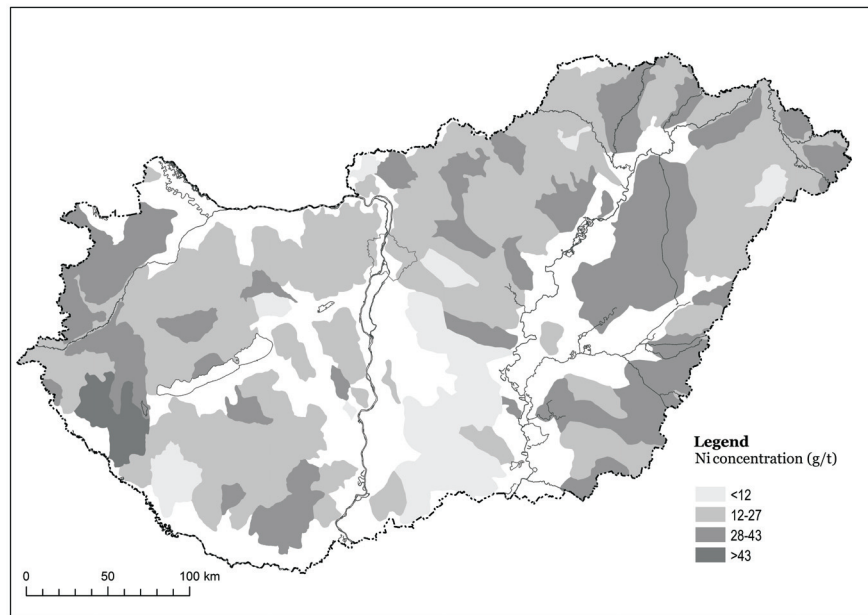


Fig. 7  
Nickel content (g/t) of the floodplain sediments of Hungary

As it has been indicated in the beginning of this chapter, the Eastern geochemical region marked by the **fourth principal component** (unlike the Central and the Northern regions) is not homogeneous; it extends only to the floodplains of certain rivers. Its characteristic elements are non-ferrous (Fig. 8) and noble metals, as well as accompanying elements (probably mercury and, it is suspected, certain half-metals). The formation of this geochemical region is the result of environmental pollution, since this element group is present where heavy industry, especially mining and metallurgy, is significant in the area of the drainage basin. To give an example, rivers originating from Transylvania have been continuously spreading the byproducts (mining waste, slag) of mining and metallurgy onto the floodplain; the Hernád River, on the other hand, has been transporting the pollution from Košice. Pollution sources in Hungary are the heavy industry of Kazincbarcika (polluting the floodplain area of the Sajó), the manufacturing plants of Salgótarján and the one-time ore mine of Gyöngyösoroszi (both polluting the stream sediments of the Zagyva).

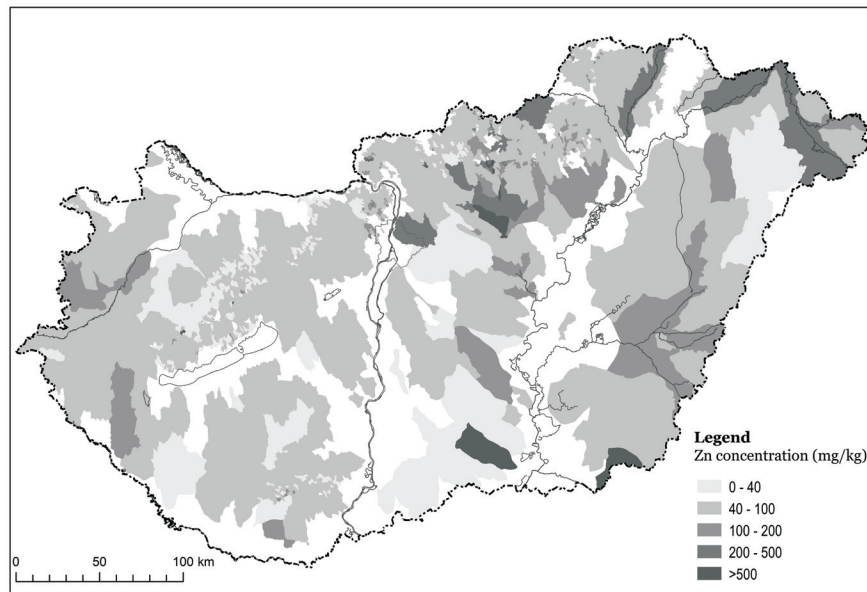


Fig. 8  
Zinc content (mg/kg) of the stream and floodplain sediments of Hungary

That the pollution is not only present at surface, but also in the ‘lower’ samples collected from a depth of 50–60 cm, applies to all the floodplain areas of the region. On the one hand it shows that heavy industry has been polluting these regions for a longer period of time; on the other it indicates that the trace element content of floodplain

Table 7  
Background concentrations (mg/kg) of some environmentally important elements in the upper level of floodplain sediments (Ódor et al. 1997)

Element	Region 1		Region 2 (Central)		Region 3 (Western)		Region 4 (Eastern)	
	Background range	Median	Background range	Median	Background range	Median	Background range	Median
As	<2.5–19	7.3	<2.5–57	6.3	5.8–13	8	5.4–22	12
Ba	53–158	95	22–158	68	87–190	135	88–160	113
Cd	<0.5–1.5	<0.5	<0.5–3.4	<0.5	<0.5	<0.5	<0.5–10.4	1.0
Co	4.9–13	9	1.7–10	5.7	10–15	12.8	9.4–14	11.1
Cr	8–39	21	4–32	14.5	25–39	36	27–92	36
Cu	8.5–42	19	5.5–33	15	18–32	24	21–103	40
Hg	0.04–0.2	0.08	0.03–0.37	0.08	0.06–0.12	0.09	0.08–0.75	0.14
Ni	11–36	22	7–30	16	25–37	32	25–41	29
Pb	10–34	17	5.3–23	13	16–26	18	32–90	46
Zn	32–150	65	14–180	46	69–96	82	100–600	132

soil profiles is quite even due to the flushing and washing occurring several times a year. In contrast with regions that are not flooded with water regularly, no prominent vertical zoning has been developed.

### **Conclusions**

The expected values of the characteristic elements on the four geochemical regions differ significantly (Table 7). Therefore it is not possible to characterize the area of Hungary with values of a homogeneous geochemical background, which should be taken into consideration in decision-making (e.g. legislation).

Regionally, the concentration of only two elements (lead and cadmium) exceeds the environment limit for soils as regulated by Joint Order No. 10/2000. Corresponding to the above, both elements occur on the floodplains of the Eastern geochemical region in higher concentrations and over larger areas. As the lead uptake of plants is excessively low, only a small amount of lead enters the food chain. Therefore, it does not cause significant environmental and health problems. Cadmium is, however, similarly to zinc, bioactive and highly toxic. As a consequence, the cadmium concentration of the cereals grown on polluted floodplains often exceeds the limit.

Consequently, the most important environmental problem of Hungary is not environmental pollution but the lack of essential elements. Soils (especially sandy soils) are depleted in several essential elements (Cu, Zn and some other elements not indicated in this table, including B, Se, Mo – Kádár 2001; Terbe 2013) in most parts of the Central geochemical region (2). Other pollutions are of local nature.

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