

Mercury content in the superficial geological formations of Hungary

Ubul Fügedi, József Vatai, László Kuti

Geological Institute of Hungary, Budapest

Among the periodic system's elements, mercury (Hg) is most liable to dispersion and, simultaneously, most liable to secondary enrichment. Consequently, mercury enrichments can occur as a result of a number of geologic as well as anthropogenic processes. If the geologic processes cease, quite extended dispersion halos can form around a former accumulation center.

Hydrothermal mineralization is a typical process giving rise to mercury concentration. As a result, regional mercury impacts can occur in the floodplains of rivers flowing from the mining and heavy industrial regions of Transylvania and Slovakia. Elsewhere, mercury anomalies detectable at the intermediate scale (1:50,000) can be found in the Zemplén and Mátra Mountains and, subordinately, in the Börzsöny Mountains. Typically mercury anomalies develop above major structural lineaments as well, unless they are buried under thick young sediments. A remarkable example is the deep fault separating the Pilis and Visegrád Mountains. Another group of Hg anomalies is caused by well-known mercury contamination sources (Kazincbarcika, Balatonfűzfő), which are truly local: they cannot be detected at the scale of the given study.

Key words: mercury, geochemical anomalies, background, Hungary, regional anomalies, Transylvania, mineral exploration, contamination

Introduction: Mercury in the Earth's crust

Mercury is typically a microelement: its Clark value in the Earth's crust is 56 $\mu\text{g/kg}$ (Wedepohl 1995). Newer sources quote higher crustal Hg concentration (80 $\mu\text{g/kg}$ – Risher 2003; 67 $\mu\text{g/kg}$ – WebElements, 2010). It is characterized by a dual geochemical behavior: it is liable to extreme concentration and to dispersion,

Addresses: U. Fügedi, L. Kuti, J. Vatai: H-1143 Budapest, Stefánia út 14, Hungary
e-mail: fugedi@mafi.hu

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the latter resulting in an approximately entirely even concentration. Its ability for concentration can clearly be characterized by the ratio of the threshold of its economic ability to be mined (in large occurrences) to its crustal average value, which is presented for comparison with some other elements in Table 1.

Its ability for dispersion is clearly manifested by the fact that mercury is the second most equally dispersed element (of lowest variance) after oxygen in the

Table 1
Global lithosphere baselines (b) and approximate workabke concentrations (wc) of some elements

Element	b*	wc**	wc/b
Iron (%)	6,3	30	4,8
Nickel (mg/kg)	90	6000	67
Copper (mg/kg)	68	5000	74
Zink (mg/kg)	79	20 000	250
Arsenic (mg/kg)	2,1	50 000	24 000
Mercury (µg/kg)	67	5 000 000	75 000

* WebElements, 2010
** Magakyan, 1974

the mercury in the Earth's crust occurs in the elementary state and a substantial majority thereof is in capillary water (Fursov 1977). The main reason for mercury's geologic mobility is that its tension largely exceeds that of the other metals.

If the geologic processes inducing its enrichment cease, the combination of its concentration and dispersion results in the dispersion of the concentrated mercury, giving rise to extended geochemical anomalies around its accumulations. Given that mercury is enriched by an extremely wide variety of geologic processes, from the formation of hydrocarbon to hydro-thermal mineral occurrences, it can be regarded as a universal geochemical indicator of young geologic effects (Saukov 1946); its dispersion halos are more extensive than those of any other element (Sergeev 1957).

In its industrial applications mercury is predominantly used in the

Earth's crust in Cretaceous and pre-Cretaceous sequences, irrespective of their age and lithology (Fursov 1983). Its expected concentration in rocks of different age and composition (Table 2) shows hardly any variation – commonly with slightly lower values in carbonate than in silicate rocks.

The strong dispersion ability of mercury is due to the reduction of its compounds to metal mercury, which is a rather slow process discernable only on a geologic scale. More than 80% of

Table 2
Median mercury concentrations (µg/kg) in some rock types (Fursov 1983)

Granite, granodiorite	67
Diorite	67
Gabbro	75
Ultrabasic rocks	65
Rhyolite	54
Andesite	51
Basalt	51
Marble	42
Quartzite	94
Clay	91
Siltstone	66
Sand, sandstone	74
Gravel and conglomerate	70
Limestone	52
Dolomite	37

elementary state. For this reason dispersion halos in the vicinity of industrial mercury contamination sources are very limited, much more so than around any geogenic source (Smith and Smith 1972; Yanin 1997; Ping et al. 2008; etc.). The dispersion halos formed in the gaseous phase within the soil feature the highest contrasts (Fursov 1983). Unfortunately, appropriate equipment for their study is lacking in Hungary. However, during the last 20 years it was possible to obtain enough quality analyses from near-surface sediment samples to appraise the mercury content of these geologic formations and to detect mercury accumulation and contamination at different scales. Based on these data we can attempt to characterize the main distribution patterns and natural variation of mercury in the surficial geologic formations of Hungary. In the present paper we summarize the results of our geochemical investigations completed in this period.

Analyses

Until 1997 the cold-vapor AAS method (Bertalan and Bartha 1999) was used for the analysis of mercury after leaching with hot aqua regia in Teflon bombs. Since 1998 the determination of total mercury has been carried out using a cold vapor atomic absorption technique, involving pre-concentration on a gold amalgam before detection with an "Advanced Mercury Analyser" (AMA-254, ALTEC) instrument (Sandström et al. 2005). The analysis was performed directly on solid samples without any sample preparation. Mercury is liberated from the sample during a programmed temperature elevation to 850 °C and amalgamated with gold. Mercury vapor is then released from the amalgam by heating and detected using atomic absorption spectrometry. The detection limit of the atomic absorption method is 30 µg/kg and the detection limit with the AMA-254 is 0.1 µg/kg.

Background and anomalies

Neither the terms background nor anomaly are well defined. As Reimann and Garrett (2005) have shown more than 10 quite different definitions are in use in the scientific literature. The Hungarian rule of law (10/2000., 219/2004.) and most of the EU documents (for example EU, 2008) in various forms define the background of some component as its "natural" concentration(s) without (any) anthropogenic influence.

In directly opposition to this conception, in our opinion (Fügedi et al. 2006) there is no reason to estimate where geochemical cycles of individual atoms have been influenced by any human activities, and where activities considered as "natural" end and the "anthropogenic" influences begin. Our (purely technical, i.e. formal) conception is that background and anomalies need to be viewed together, and can only be defined on the basis of statistical distribution patterns,

always taking, however, the possible heterogeneity of the background (geologic-geographical setting) into account.

There exist a number of different techniques for the statistical calculation of the background (see Reimann et al. 2005). In our experience the most effective method is the separation of the so-called "uncorrelated background" from the well-correlated anomalies. In presenting the results of some technical problems in this paper we followed another method: the background of mercury always falls into the same range (between two frequency minimums of the polymodal distribution), in which the majority of the values occur. The positive and negative anomalies are placed outside of this range. If the distribution is unimodal, all samples may be considered as background.

It is important to point out that the background is, in most cases, heterogenic: the concentration may be quite different in different rock types. The variability (the range of the background concentrations) is dependent on the location of the sample type: e.g. overbank sediments represent a larger catchment basin (with lower variability) than stream sediments, while stream sediment samples represent larger territories than a soil sample.

Previous investigations

1. Continental scale. In the FOREGS Geochemical Baseline Mapping Programme (Salminen et al. 2005) 14 overbank sediment and 14 stream sediment samples were collected from Hungary. All the solid samples of this program were analyzed for mercury in the laboratory of MÁFI using a AMA 254 instrument.

2. Regional scale (Darnley et al. 1995) geochemical mapping (1: 500 000) in Hungary was completed in 1991–1995 (Ódor et al. 1997, 1998). In regions with well-developed drainage systems, 196 catchment basins of approx. 400 km² were delineated and flood-plain (overbank) deposits sampled at their outlets. Two samples were collected at each site, one from a depth of 0 to 10 cm and one from 50 to 60 cm. The most important result of these investigations was that in Hungary there is no single background because the territory is divided into four geochemical regions (Fig. 1) with different characteristics. The larger part of the country (the "main" Region 1) does not show a characteristic association of elements, i.e. the dominant part of the variations resulted from processes of accumulation and leaching. In Central Hungary (the "limy" Region 2) the association of Ca-Mg-Sr-CO₃²⁻-PO₄³⁻-SO₄²⁻ reflects the presence of limy soils. Near the western border line (the "ferrous" Region 3) the iron alloy metals Co-Cr-Ni-Fe originate from basic and ultrabasic rocks of the Alpine belt. In the flood-plain deposits of rivers discharging from Transylvanian mining areas and from some heavy industrial centers ("eastern" Region 4) we can find characteristic Ag-Au-Cd-Pb-(Cu-Zn) anomalies in the overbank sediments of lower courses, and also Hg originates from the processing of ore from low- to medium-temperature hydrothermal ore deposits.

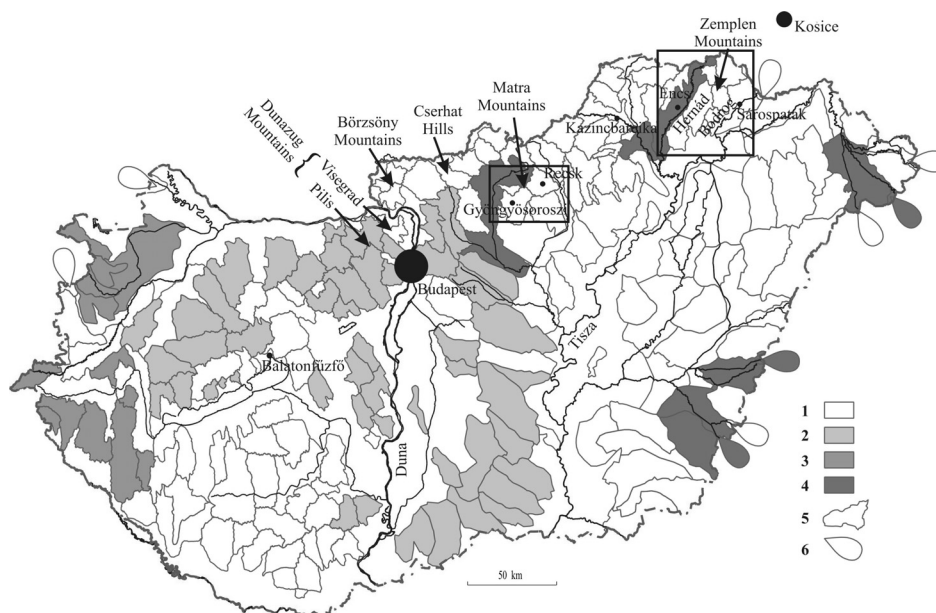


Fig. 1
Geochemical regions of Hungary (after Ódor et al. 1997, corrected). 1. Region 1 ("main"), 2. Region 2 ("limy"), 3. Region 3 ("ferrous"), 4. Region 4 ("eastern"), 5. catchment area in Hungary, 6. catchment area outside the national boundaries

On the mercury distribution map (Fig. 2) yet another tendency is visible. More variable Hg concentrations occur in the mountainous areas than in the plains. The reason for this is that during the weathering process mercury vaporizes and precipitates continuously, and that during transportation the concentrations become more and more equalized.

3. Intermediate scale (1: 50 000): In addition to the surveys mentioned above, a large part of the hilly and mountainous regions of Hungary was subjected to more detailed geochemical mapping, based on stream sediment sampling of small, approx. 4 km² catchment basins (Hartikainen et al. 1992; Ódor et al. 1999; etc.) between 1989 and 1998. The most important result of these investigations was that no significant difference between the composition of stream and overbank sediments eroded from carbonatic, volcanic and siliciclastic rocks were found (Fügedi et al. 2007). The concentrations of elements are mainly determined by the young (Miocene–Holocene), easily eroded, soil-forming sediments (tuffs, loess, etc.). Against this background only the influence of rare, extreme geologic formations becomes visible: some ore deposits, alkaline ultrabasic rocks, etc. (Fügedi et al. 2006). Therefore, in 2000–2010, most of the detected anomalies were resampled.

The results of these three mapping programs were integrated in 2008–2010. Now the uniform database contains 1,863 records with 1,629 mercury analyses.

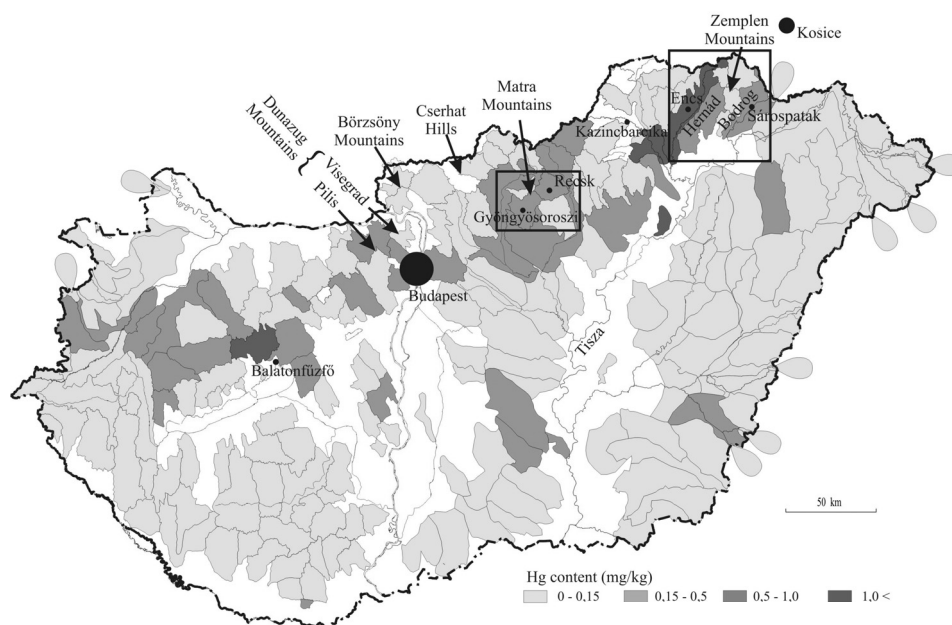


Fig. 2
Mercury background concentrations in Hungary based on overbank sediments

Some parts of the integrated geochemical atlas of Hungary (Figs 3, 4) are published for the first time in this paper.

The most important (recent) mercury contamination sources in Hungary were described in the NATO ASI Programme "Regional and Global Mercury Cycles: Sources, Fluxes and Mass Balances", in 1995–1996. A large contamination (about 600 t Hg) was located at Kazincbarcika resulting from the production of hydrochloric acid. A second is located at Balatonfűzfő close to the Nitrochemistry Works (Fügedi et al. 1995, 1996). Unusually high values of Hg with a limited geographical extent were detected in the Budapest agglomeration area. Such very local Hg contamination sites remain difficult to detect with traditional geochemical mapping methods.

The regular content of mercury in Hungary

The background levels of mercury (Table 3) in the geochemical regions of Hungary have been determined within the framework of completing the 1:500,000 geochemical atlas of Hungary.

In full compliance with the above presented features of mercury its expected values are almost equal in the geochemical regions, notwithstanding substantial differences in geology. The exception is the eastern Region 4 with 1.5–4 times higher Hg-values than elsewhere in the country.

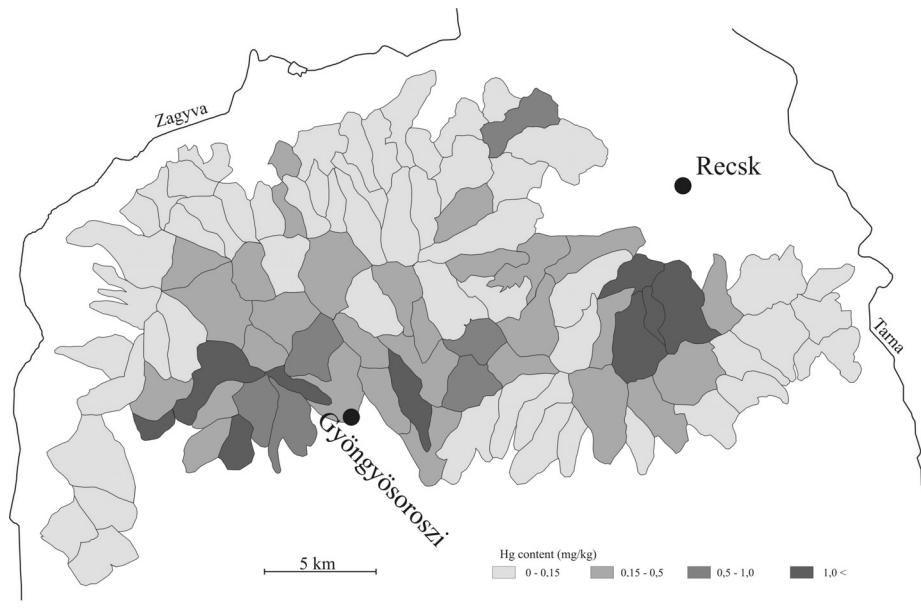


Fig. 3
Mercury concentrations in the Mátra Mountains based on stream sediment sampling

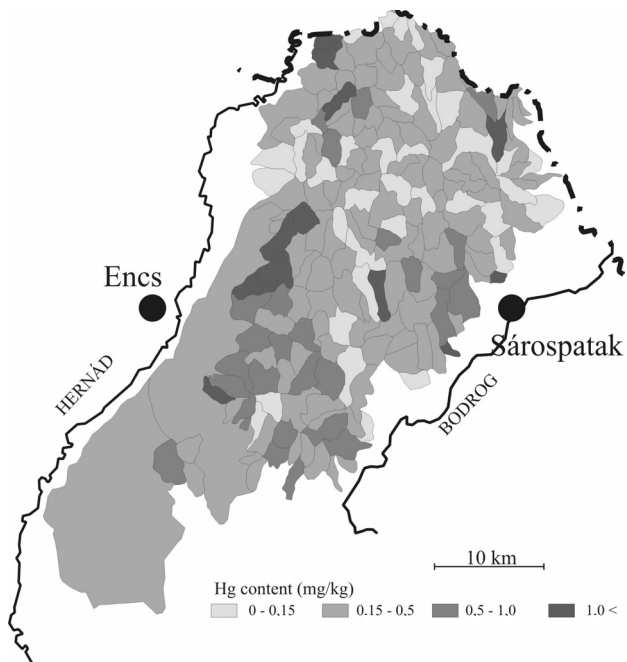


Fig. 4
Mercury concentrations in the Zemplén Mts based on stream sediment sampling

Table 3
Background concentrations ($\mu\text{g/kg}$) of mercury in the geochemical regions of Hungary (Ódor et al. 1998)

Region/background	From	To	Me
1.	40	200	80
2. (limy)	30	370	80
3. (ferrous)	60	120	90
4. (eastern)	80	750	140

Me – median

Mountains and Cserhát Hills were sampled at the beginning of the project and analyzed with the cold vapor AAS technique).

The anomaly in Mátra (Fig. 3) is due to a number of processes, including the erosion of extensive ore zones (Gedeon et al. 1959; Gedeon 1964), environmental contamination resulting from the Gyöngyösoroszi ore mine (Fügedi 2004) and the As-Sb-Hg low-temperature hydrothermal mineralization discovered in the area of the southwestern part of the mountains (Csongrádi 1984). At the same time the background values in the Börzsöny and Zemplén Mountains (Fig. 4) are not affected by mineralization, due to its substantially smaller extent: the anomalies are separate from the background. A slight but characteristic anomaly was detected between the Visegrád and Pilis Mountains.

Table 4
Median background concentrations of mercury ($\mu\text{g/kg}$) in some mountainous or hilly areas of Hungary

Zemplén Mts.	28	Börzsöny–Visegrádi Mts.	70
Mecsek–Villány	25	Cserhát	30
Bakony	35	Mátra	140
Vértes	50	Heves-Borsod Hills	40
Gerecse	30	Bükk	90
Pilis-Budai Mts.	50	Aggtelek-Rudabánya Mts.	80

The background levels of the mountain- and hilly regions (Table 4) are also quite close to each other and these are very similar to the sub-regional medians of the Geochemical Atlas of the Republic of Croatia (Halamić and Miko 2009, 35–105 $\mu\text{g/kg}$). It can be suggested that the smaller part of the relevant values below background level could have been due to analytical reasons (the Zemplén Mountains, Pilis-Buda Mountains, Börzsöny-Visegrád

Discussion

In terms of their size mercury anomalies occurring in Hungary can be assigned to three main categories as follows.

1. Regional level

In the (eastern) geochemical region (No 4), beside Hg the precious and base metals Ag, Au, Cd, Cu, Pb, and Zn, as well as their accompanying elements (As, Sb) also occur in anomalous concentrations. These anomalies do not extend throughout the entire surface of the catchment areas displayed on the map. They are restricted to the rivers' floodplains beneath heavy industrial complexes in the Transylvanian mining region and of Kazincbarcika and Košice. The contaminated sedimentary sequence is at least 60 cm thick; the length of the contaminated floodplains totals several hundred kilometers; their width, unmeasured, is probably in the order of one kilometer. The highest value of Hg detected in these sediments is 96 $\mu\text{g/kg}$ mercury. This is still just one order of magnitude above the Clarke for Hg.

As shown by the first results of our analyses performed within the framework of Romanian-Hungarian cooperation (Horvath et al. 2009) Hg contamination can apparently be detected from the mining area down to the mouth of the Tisza River.

2. Subregional level

Natural mercury anomalies occur extensively as a result of hydrothermal ore mineralization processes (Zemplén Mountains, Mátra and Börzsöny), as well as along some deep faults, like the fault zone between the Pilis and Visegrád Mountains. Their individual surface area amounts to some or some dozen km^2 ; their intensity varies between 300 and 3000 $\mu\text{g/kg}$ Hg. In some anomalies (Zemplén Mountains, Mátra and Börzsöny) mercury occurs as the accompanying element of precious and base metals. However, Hg alone occurs in the form of occasional scattered metacinnabarite elsewhere (Zemplén and Dunazug Mountains). These natural anomalies have occasionally been substantially enhanced and strengthened by mining activities (Telkibánya, Gyöngyösoroszi, Nagybörzsöny and Recsk-Lahóca).

3. Local level

The industry-induced mercury anomalies of Hungary were surveyed within the framework of NATO's ASI program (Fügedi et al. 1996). Industrial plants (Kazincbarcika, Balatonfűzfő) as well as thermal power stations have been the main emitters in the past and continue to be so today. The concentration of mercury can attain several percent in the surficial geologic sequence beneath

electrolytic plants as well. The contamination caused by factories is so small and the signal caused by power stations is so weak that these possible sources remain undetectable at the scale of our geologic survey.

Conclusions

In Hungary, natural enrichments of mercury are the result of Neogene postvolcanic activities; they are not very significant. Substantial mercury contamination was detected only in the immediate vicinity of large mines and some major industrial complexes. Since they are found mainly in Transylvania, it would be quite reasonable to study the removal of ore minerals jointly with Rumanian colleagues. The 1:50,000 scale is still not sufficiently detailed to detect the well-known domestic contamination.

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