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ARSENIC-BEARING ARTESIAN WATERS OF HUNGARY

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The anomalous arsenic content of some communal drinking waters produced from artesian aquifers of the Great Hungarian Plain was recognized in 1981/82 by public health authorities in Hungary. Arsenic in excess of the sanitary threshold contcentration could be detected in the artesian waters of extensive areas providing the water supply of nearly half a million of consumers. These increased levels caused by geological and geochemical processes. Short term solutions for the most urgent problems were found by investing many thousand of millions of Hungarian forints, but finding long term solutions remained a task for the future.

The hydrogeochemical study started in 1993 was aimed at the establishment of the knowledge still missing up to that time but being indispensable for the further advance. This paper gives the summary of the first results obtained during 1993.

1. By carrying out a new evaluation of the data base built up by data on 6500 wells and using GIS we plotted a map of arsenic bearing waters. These areas turned out to be considerably bigger than it has been known hitherto.

2. On the ground of the two-peaked lognormal frequency distribution of the concentration values of arsenic we identified its largescale enrichment as the result of well definiable and characteristic geochemical processes.

3. It was found that the arsenic-bearing waters were formed in the geological-geochemical environment of the Quaternary. The process went on during several phases of the early diagenesis transforming the fluvial; floodplain and paludal sediments. According to the evidence the enrichment of arsenic is related to adsorption processes taking place on the surface of colloid oxi-hydroxides of Fe, while its remobilization was the result of reducting bacterial processes following the burial of the sediment.

4. On the basis of the new results the areas of the possible occurrence of arsenic-bearing waters could be contoured, thus pointing to new targets for further study.

Preliminaries

The poisonous effect of arsenic and its soluble compounds is well known since the ancient times. Later on it has been recognized by medical investigations that by receiving small doses of arsenic over a long period, the living organism gradually accumulates it; this enrichment may lead to inadmissible consequences, such as increased probability of the development of malignant tumours, diseases of the vascular system and — first of all — of serious damages to the skin. In accordance with the recommendations of the World Health Organization the Hungarian standard (MSz 450/1-78) puts the upper limit of the acceptable arsenic content in drinking waters to the value of 0.05 mg/l, i.e. to 50 µg/l.

Since the 1981/82 survey carried out by the Hungarian public health authorities we know that in certain areas of the country the arsenic content of subsurface waters is considerably higher than the limit acceptable for the public health. Some half a million consumers had been affected by this chemical feature of the drinking water and as a

consequence of this situation the construction of a new water supply system having 150. thousand cubic metre/day capacity was needed.

During the decade which elapsed since then the dificulties caused by the arsenic bearing waters for the communal water supply have been considerably reduced. The governmental programme of improving the water quality, which was established in 1983 and is in force even to-day resulted in the reduction of the arsenic content below the permissible threshold concentration in the case of most of the communal water works concerned. The investment costs ran to several thousands of millions of Hungarian forints. Certain areas however, like the Sárrét and Nagykunság regions and the towns of Gyula and Orosháza are difficult to supply with healthy drinking water throughout the year. The problem presents itself mainly during the summer season. (Verbal information given by Z. KÁRPÁTI)

In its most recent recommendations of the World Health Organization (1993) urges the considerable lowering of the limit for the arsenic content in drinking water, to a level as low as 10 µg/l eventually. This means that we should not overestimate the results achieved in Hungary so far, but consider them as the most urgent first steps taken. The proposed lowering of the limit puts into question not only the efficiency of the measures taken for the reduction of arsenic content (modification of the operative processes in water works, dilution, extraction of As, development of new water reserves etc.) in their present form, but the results achieved in the improvement of the water quality on the whole as well. What was done so far can be considered as partial measures at best and further action is called for.

It is certain however that the solution in the long run may be reached by the complete re-investigation of the entire scope of the problem. In the search for new solutions the hydrogeological and hydrogeochemical investigations have also a role. Realising this gave the impetus for launching the geochemical investigation of arsenic bearing waters by a survey of the entire national territory.

Results of the research work done prior to 1993

The recognition of the abnormally high arsenic content in the artesian and ground waters of Hungary is the achievement of the professional staff of the public health service, and being also the result of a process lasting some fifty years.

Medical and sanitary investigations concerning the occurrence of arsenic

The first signs of the anomalous arsenic content in some domestic drinking waters appeared as cases of chronic poisoning in the lowland areas between the Danube and Tisza rivers during the forties and fifties. It was supposed at first that these cases might be attributed to the pollution of the groundwaters from the surface - thus being caused by human negligence- and were regarded as local problems only. (HORVÁTH et al. 1980.)

The essential change in appreciating the problem and its sanitary importance was brought about by the results of the national survey concerning the arsenic content of the communal drinking waters. The surveys carried out in 1981/1982 under the direction of the National Institute of Public Health (Országos Közegészségügyi Intézet - OKI) have found (CSANÁDY et al. 1985) that:

- In the drinking waters of 148 settlements scattered over the area of six counties (Bács-Kiskun, Békés, Csongrád, Hajdú-Bihar, Heves and Szolnok) the amount of the arsenic exceeded the 50 µg/l sanitary limit. Almost half of the settlements concerned was found in Békés County.
- Within the "arsenic-bearing areas" the arsenic content of the communal drinking waters in 50% of the cases exceeded twice the amount of the sanitary limit (by being over 100 µg/l). The peak value observed reached 560 pg/l.
- The size of the population affected (400,000) by the problem was higher than in any similar cases known else where in the world documented up to that time (Taiwan, USA, Canada, Argentine, Chile).
- The extraordinary high arsenic content of the artesian aquifers tapped for communal water supply is a phenomenon which in spite of earlier opinions of the medical circles was not caused by external pollution, but being the result of natural — i.e. geological — processes.

Geological-geochemical investigation of the arsenicbearing artesian waters

The efforts of a decade aimed at the survey of the arsenic content of artesian drinking waters and concentrated on the urgent reduction of it resulted in a significant improvement of the water supply, but at the same time the full-scale investigation of the problem including first of all its geological-geochemical aspects was left aside. Thus:

- The anomalies of the occurrence of arsenic were not identified in geological sense: i. e. by taking into consideration the genetical conditions of the phenomenon. The potentially endangered aquifers and their extent were not yet determined either.
- The effect of the exploitation of aquifers on the arsenic content of the water produced was not investigated.
- The role of the trace elements associated with arsenic in influencing the toxicity was not taken into consideration.

The initiative to carry out these investigations was taken by two research institutes simultaneously.

The Hydrogeological Department of the Geological Institute of Hungary (MÁFI) began the hydrogeological mapping of Hungary in 1983 under the leading GY. TÓTH. Within the scope of the general reconnaissance concerning the distribution of trace elements the amount of arsenic had been determined in almost one thousand samples of drinking water, but the evaluation of the results was not completed.

The pilot project for the geological and geochemical study of arsenic-bearing waters was steered by the "ad hoc" academic committee organized in Szeged for the execution of assignments given by the Council of Békés County and later on also by the MÁFI. This committee was acting under the leadership of T. SZEDERKÉNYI, with Mrs. VARSÁNYI, B. MOLNÁR and M. ERDÉLYI collaborating in the research work. The results obtained were published in 1990 (SZEDERKÉNYI et al. 1990). At the outset the survey had covered the area of Békés County only, but later it was extended to cover the entire southern part of the Great Plain. These geochemical, genetical (VARSÁNYI 1990, SZEDERKÉNYI 1990), facies (MOLNÁR 1990) and hydrogeological (ERDÉLYI 1990, 1991) surveys were of fundamental importance for all further research activity.

Results of the hydrogeochemical investigation of the arsenic-bearing artesian waters carried out in 1993

The countrywide geological–geochemical survey of the arsenic-bearing waters was started by the Geological Institute of Hungary at the end of 1992. This activity formed part of a hydrogeochemical project. In 1993 the results of the investigations carried out by the public health authorities and of those of geological-geochemical character done up to that time were re-evaluated according to geochemical concepts. In collaboration with the National Institute of Public Health (OKI) a national data base of the arsenic-bearing waters was set up and processed in part by GIS in that year. The data base and GIS have made it possible to determine the spatial extension of the arsenic-bearing artesian waters, their geological and geochemical environment, and the geochemical processes responsible for their formation.

Our growing data base now includes 9600 analysis results of the arsenic content found in 6499 water wells of 1466 settlements of the country. Hydrogeological and hydrochemical and well completion information is also included. Seventy percent of the analytical results on arsenic obtained by the highly sensitive hybrid-forming AAS procedure were given to us by public health services; of this, nearly 20% was received from the OKI. The contribution of the MÁFI is about 10%, while the remaining 20% of the data were obtained from water works.

New concepts on the geological–geochemical environment and genetical conditions of the arsenic-bearing artesian waters

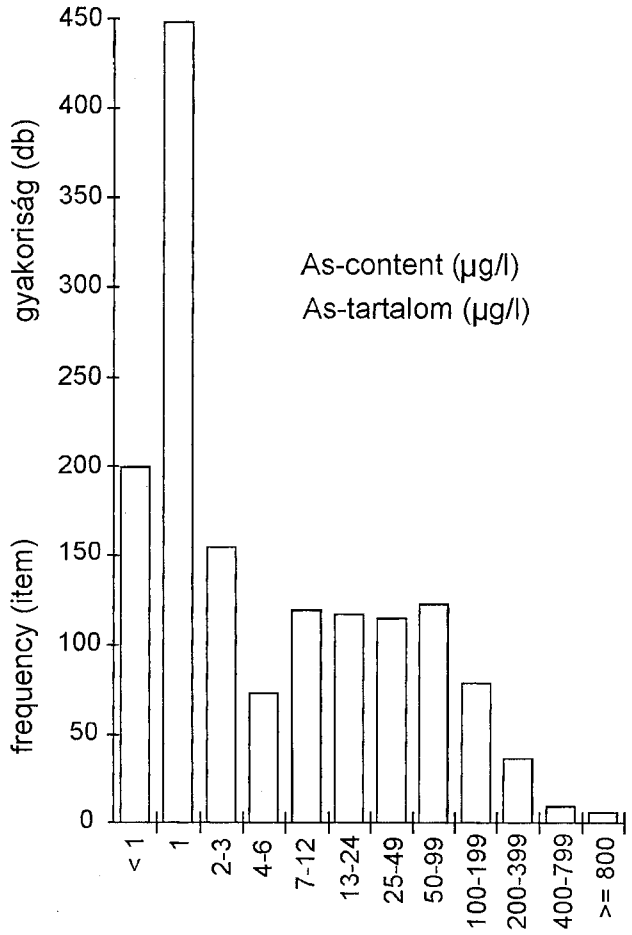

Fig. 1. Frequency of the maximum As-contents tested in the artesian waters of the settlements

1. ábra. A települések rétegvizeiben mért legnagyobb As-értékek gyakorisága

Due to the very uneven areal distribution of the samples, we took into consideration for each of the settlements the data of only one water well, the one that contained the greatest amount of arsenic.

We plotted the frequency of values on plots with logarithmic scaled axes. The smoothed curve of arsenic distribution in the chosen water wells is characteristically bimodal (Fig. 1).

The distribution pattern indicates that:

- Thc arscnic content of the analysed artesian waters lies in the 1-10 µg/1 interval - thus being in accordance with published data in scientific publications.

- The appearance of the second peak of the concentration-frequency can be interpreted as a peculiar process of the enrichment of arsenic which causes an anomaly both in geochemical and sanitary sense. The geochemical anomaly limit is likely to be in the 12-25 µg/litre concentration interval.

- According to the new data base the arsenic content of the drinking water exceeds the sanitary limit in 358 settlements. By taking into consideration the greatest values of the arsenic content found at separate settlements, our ideas on the geographical extent of the arsenic-bearing artesian waters have to be modified (Fig. 2).

- Besides the areas mentioned hitherto with emphasis (Békés and Csongrád Counties moreover the southern part of the Danube-Tisza Interfluve) arsenic-bearing waters occur also in the whole area of the Danube-Tisza Interfluve, in the Nagykunság, Jászság and southern Heves areas, in Szabolcs-Szatmár County, in the Bihar and Hajdúság regions and in extensive marginal zones of the Nyírség.

- Scattered geochemical anomalies of arsenic can he found between the Sajó and Hernád rivers, along the Dráva river, on the flood plain of the Kapos and Zala rivers and, according to recent information, in the Szigetköz area as well.

- Due to lack of adequate information the danger to public health can not be established in the southern parts of Pest, Heves and Borsod Counties.

Current data suggest that these areas, along with in the deeper levels of the water base of the Kisalföld region, belong to the unfavourable zone.

Fig. 3 shows the distribution of the arsenic-bearing aquifers by depth in Békés County almost all of which is a contiguous anomaly. Arsenic-bearing aquifers are most frequent there in the 300–400 m depth interval.

- Extraordinary enrichments of arsenic can be found in thermal waters ascending from considerable depths as

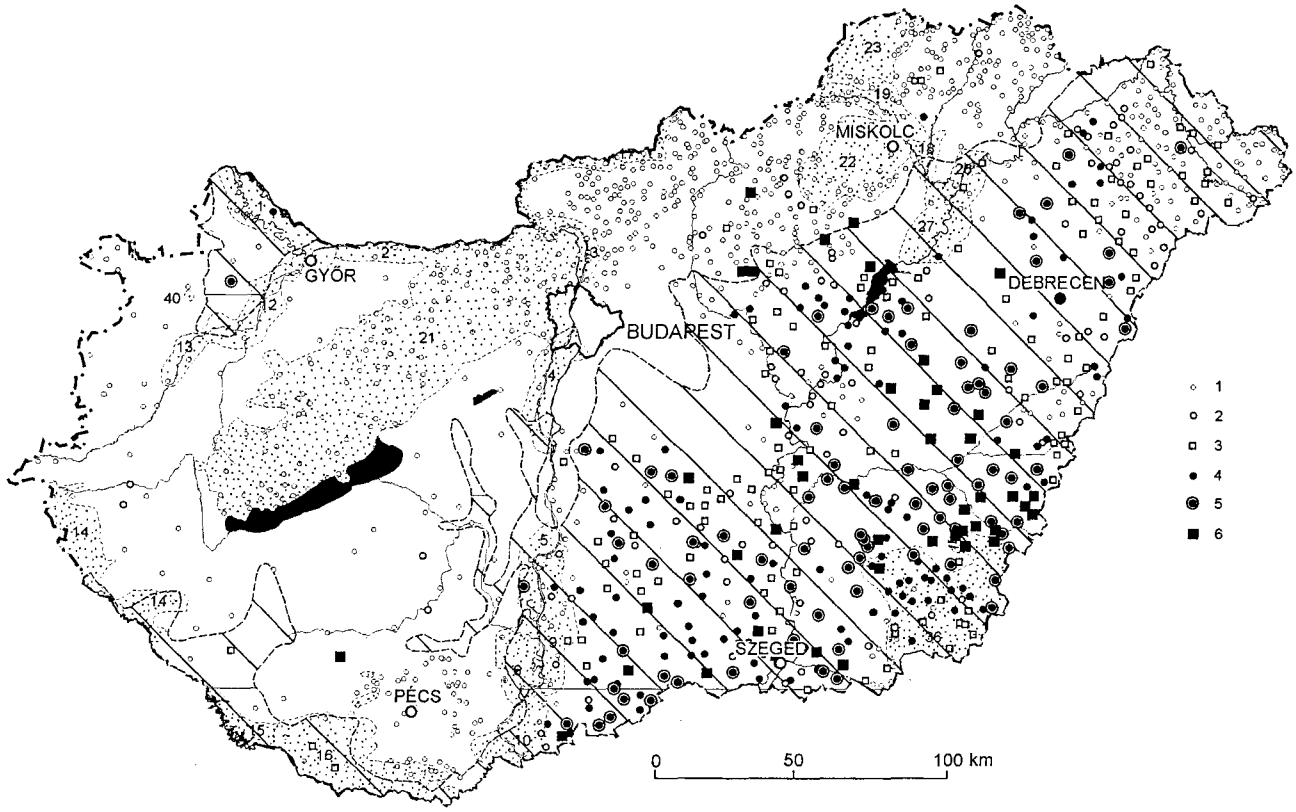

Fig. 2. The maximum As-contents tested in the settlements investigated; (the sanitary threshold contentration is 50 µg/l in Hungary)
1. <12.5, 2. 12.5–25, 3. 25–50, 4. 50–100, 5. 100–200, 6. >200 µg/l, 7. Territories of Quaternary sedimentary rocks and sediments with thickness more than 50 m, 8. Protected water-bearing areas (after F. FRANYÓ)

2. ábra. A vizsgált településeken mért legnagyobb arzéntartalom; µg/l (az egészségügyi határérték 50 µg/l)
1. <12.5, 2. 12.5–25, 3. 25–50, 4. 50–100, 5. 100–200, 6. >200 µg/l, 7. Az 50 m-nél vastagabb negyedidőszaki üledékek elterjedése (FRANYÓ F. után), 8. A védett vízbázisok területe

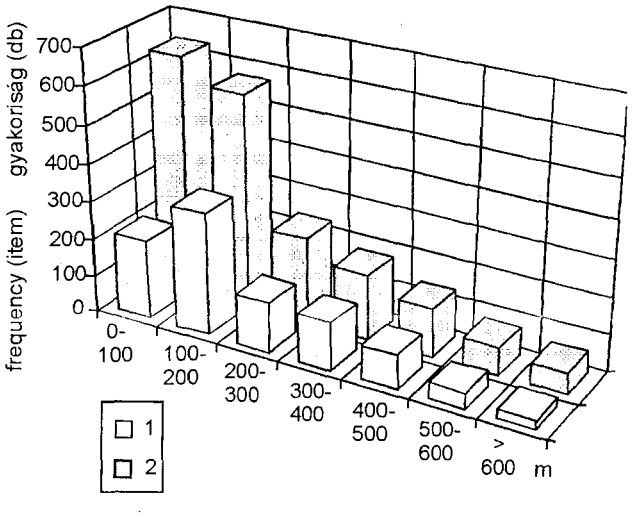

Fig. 3. The As-anomalies vs depth in Békés County
1. Anomalous samples, 2. The total of the samples

3. ábra. A Békés megyei arzénanomáliák a mélység függvényében
1. Anomális (As>50 µg/l) minták 2. Összes minta

well. It is proven however that neither areal nor, consequently, genetical connection exists between the arsenic-bearing waters of the shallow and deep aquifers.

Fig. 4 shows the percentage of shallow depth As anomalies as a fimction of their depth in Békés County. Fig. 4 indicates two important facts:

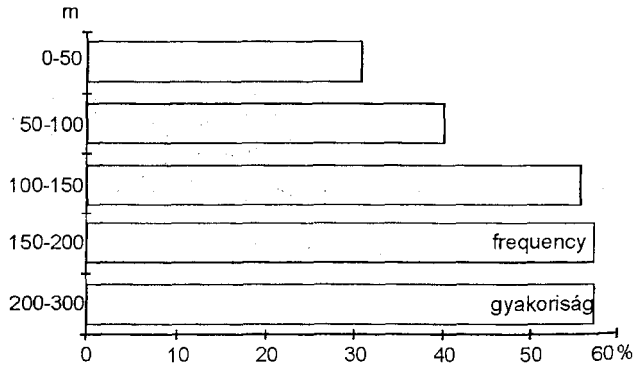

Fig. 4. Percentage of the anomalous samples to the total vs depth inside the 0–300 m interval in Békés County

4. ábra. A Békés megyei sekély mélységű arzénanomáliák az összes mintához viszonyított %-os arányának változása a mélység függvényében

- The frequency of the shallow-depth As anomalies decreases towards the present surface, thus it can be assumed that their origin is not connected to recent or very young supergene processes.

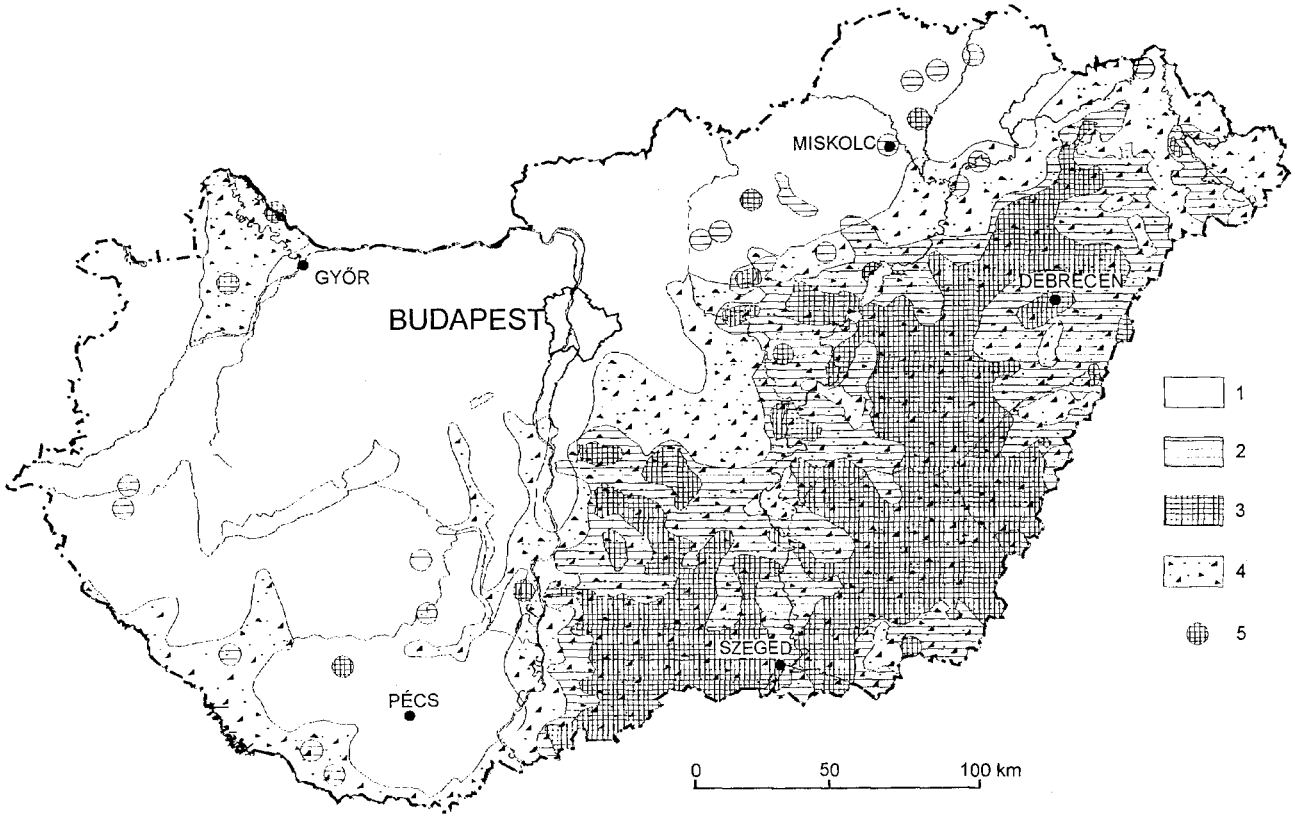

Fig. 5. Areas of the known (1–3) and possible (4) occurrences of As-bearing artesian waters based the maximum As-contents tested in the settlements

1–3. The As-content of the artesian waters (µg/l): 1. <12.5, 2. 12.5–50.0, 3. >50.0, 4. Areas of the possible occurences of the As-bearing artesian waters, 5. Single tests

5. ábra. Az arzénes rétegvizek ismert (1–3) és lehetséges (4) elterjedése a településeken mért legnagyobb arzéntartalom alapján

1–3. A rétegvizek arzéntartalma (µg/l): 1. <12.5, 2. 12.5–50.0, 3. >50.0, 4. Az arzénes vizek lehetséges megjelenési területei, 5. Egyedi arzénadatok

- In the phreatic zone (taken in the broad sense) the frequency of the anomalies reaches 30%. This zone is only inadequately investigated all over the country.

The stratigraphical and facies conditions determining the occurrence of the arsenic-bearing artesian waters are shown by Fig. 5. Contours are drawn according the manuscript map of F. FRANYÓ (1993). These show an overlap with the more than 50 m thick Quaternary sediments. Consequently this map verifies former views regarding to the geological background of the extent of As anomalies (CSÁKI et al, 1983).

The stratigraphic dating and facies analysis of the aquifers storing arsenic-contaminated waters is mostly still outstanding. Available data suggests that these waters are connected mainly to sediments of fluvial, flood plain and paludal origin. These formations are described in the monograph of A. RÓNAI (1985). It is remarkable that there is no enrichment of arsenic in the loess areas of the same Quaternary age.

Geochemical relationships of the origin of arsenic-bearing artesian waters

The geochemistry of the arsenic, the conditions of its enrichment and mobilization moreover the effects of oxidation-reduction and adsorption processes of the sedimentation on the distribution of the arsenic are well known in general (BELZILE et al. 1990, FERGUSON et al. 1972, HEM 1977, KANAMORI 1962, MATISOFF et al. 1982, ONISHI et al. 1955, SZÁDECKY-KARDOSS 1955). As usuallly the practical difficulties arise from the fact that theoretical knowledge can be applied to complex and variable natural systems with certain restrictions and simplifications only.

The geological-geochemical investigations aimed at the origin of the arsenic-bearing waters, and their results published in the scientific literature were restricted both in space and time. As the amount of analysis data was limited, in spite of valuable results some contradictions occurred as well.

The key-question of the origin of these waters remained unsolved; why did the biggest anomaly of arsenic content ever described in scientific publications from drinking water develop just in Hungary, and precisely in the Alföld region of the national territorry?

Because of the many unanswered questions about the genetic conditions we deemed it necessary to elaborate a new genetic model. We had to reconsider the existing patchwork of data on arsenic-bearing artesian waters and apply also the evidence in foreign publications. This model aimed to create an action plan for further research. The new conceptual model was based on the following considerations:

1. The former assumption concerning the source area of enriched arsenic content in the artesian waters (CSÁKI et al. 1983, SZEDERKÉNYI et al. 1990, ERDÉLYI 1990, 1991) must be modified. The Carpathian and Transsylvanian belt of ore-mineralized magmatites can not be regarded as the exclusive source of the arsenic in our domestic arsenic-bearing waters. The following facts must be taken into consideration:

- Arsenic-bearing artesian waters occur in sediments arriving not only from the Carpathian-Transsylvanian territory but also from source regions of entirely different character, i.e. from the catchment areas of the Danube, of the Dráva and Zagyva rivers.

- The average of the arsenic content differs only insignificantly in the various magmatites and sedimentary rocks. These slight differences can not be regarded as explanation for the differences of concentration in the secondary enrichments exceeding primary ones by orders of magnitude.

- Due to the diluting and averaging character of the processes of wheathering the presence of arsenic-rich hydrothennal formations in the catchment areas can not be regarded as determining factor.

2. The genetical model based on the supposed origin of the arsenic-bearing waters from great depth (SZEDERKÉNYI 1990) is in contradiction to the chemical composition of these waters especially with regard to the distribution of the anions (verbal information of I. HORVÁTH). The composition does not point to abyssal origin. Also, the distribution of the arsenic-bearing waters by depth does not support this genetic model.

From these considerations it can be deduced that the arsenic-bearing waters do not originate from great depths - i.e. from the basement, moreover their occurences can not be connected to well defined erosional areas. They may also be found in such regions of the country which are far from the Carpathian-Transsylvanian areas.

Regarding the mode of transportation of the arsenic two possibilitites are to be considered. Arsenic may be transported either in dissolved state or included in the the crystal lattice of the (mostly) silicate minerals occurring in the fine grained detrital material of source rocks. This form of the arsenic is not affected by surface processes; it may exceed the dissolved amount by 2-3 orders of magnitude. It is important to note that arsenic-bearing minerals which might be considered as sources of this element by their weathering and sedimentation were not found by micromineralogical investigations in Hungarian or Romanian (Transsylvanian) fluvial deposits of the Quaternary.

We have no evidence relative to the formation of arsenic-bearing waters by the interaction of rock and interstitial water. Even the character of the occurrences as it is determined by geographical setting, time of formation and facies gives rise to contradictions in every model based on purely mineralogical and geochemical ideas.

It is very probable that the arsenic accumulated in artesian waters originates from an external source and enters into the sedimentary basin in dissolved state. A very convincing explanation for this assumption is furnished by the process of mobilization of the soluble arsenic content of sulphide minerals in the form of arsenate anions freed by oxidative surface weathering in the source area.

Indirect data indicate that subsequent to mobilization the arsenic enters into the area of sedimentation in very dilute solutions or by adsorption on the surface of humic colloids or colloidal Fe-hydroxides.

By taking into consideration the very low arsenic content (ranging 1-10 μg/l) of the surface waters it is clear that the formation of artesian waters having more than 50 μg/l arsenic content has to be the result of a geochemical process, by which the considerable accumulation takes place.

In the given geological environment, considering also the possibilities of accumulation known from published geochemical references the enrichment of arsenic by adsorption on the surface of colloidal oxi-hydroxides of iron (HEM 1977) can be singled out as the most likely genetical process responsible for this phenomenon.

The formation of iron oxi-hydroxides is well known in Quaternary fluvial flood plain and paludal formations (like the bog-iron ores and variegated clays) of Hungary. The possibility of the arsenic-enrichment bound to crystalline and colloidal ferric oxi-hydroxides is supported by our analytical results which indicate that the arsenic (As) content of the domestic bog-iron ores exceeds 1000 ppm regardless of the site of occurrence. The measure of enrichment by adsorption is characterized by the fact, that the arsenic content of the investigated bog-iron ores exceeds three times the respective clark value of rocks and five-six times the average value given for waters by the scientific literature.

The enrichment of arsenic on colloids of Fe hydroxides can occur in the sediments of any flood-plain, irrespective of the geological environment and of the arsenic content in the surface waters of the given area.

It follows from the geochemical character of the arsenic, that following burial the colloid ferric oxi-hydroxide minerals are beginning to be decomposed by reducing diagenic processes. The soluble arsenic freed from the adsorptive bond enters into the interstitial water. The redox limits of the remobilization of the arsenic, i.e. the limits of the

fonnation of arsenic-bearing pore waters are well known. The upper limit coincides with the ferric/ferroan - boundary: it is indicated by the formation of arsenite anion and minerals of Fe^2 (ankerite, siderite). The lower limit is marked by the appearance of sulphide anions and sulphide minerals due to the increased intensity of the reduction.

According to these characteristics the remobilization process of the arsenic i.e. the formation of arsenic-bearing artesian waters takes place during early diagenesis in a narrow redox domain of transitional and moderately reductive character. Bacterial activity and the organic matter content of the system are the most important factors in this process. Proof for this is the correlation between arsenic, methane, iron, manganese and ammonia content of the waters. The genetic model outlined here answers also the following questions:

- Why are the arsenic-bearing waters bound only to the most recent Quaternary sediments which are in early stage of diagenesis, and why is arsenic missing from older formations which went through a complete diagenesis?

- Why do we find in Hungary the greatest occun-ence of the arsenic-bearing artesian water known in the world? Has this to do with having very thick Quaternary sequences or other peculiar geological conditions?

So far we have completed only the first steps in the study of arsenic-bearing artesian waters; still many problems are left to be solved. It seems that the most important one of them is whether the arsenic-bearing waters are autochtonous. This problem has not been investigated yet. The ^{14}C isotope dating carried out by J. DEÁK (in RÓNAI 1985) indicates an age of approximately 30 thousand years. From these data the following conclusions can be drawn:

- The age of arsenic-bearing artesian waters differs from that of aquifer; thus the waters were not formed in the area of their present occurrence.

- The areal extent of this type of artesian waters is strongly influenced by the systems of underground flow.

Further isotope geochemical investigations are needed to bring the question of the provenance to the waters to a well founded conclusion and to indicate the best direction for further work. Answering these questions allows setting the priorities of studies of rock-and-water interaction and the regional ground water flow systems.

Apparently there are still considerable deficiences of our present knowledge. We know already that arsenic-bearing artesian waters can occur also in regions of the country which had been deemed free of this phenomenon up to now - but only scattered investigations were done in these areas. We find important targets for further research in the valleys of the rivers filled by Quaternary formations. Within these units the protected bases of the communal water supply (like the Dráva-valley, the alluvial fan of the Maros river and the Szigetköz-Hanság region) are of primary importance. The regions of extant and drained bogs, swamps and protected flood plains are fields for further investigation as well.

At present we have little data about the arsenic contained in domestic groundwaters and in fluvial, floodplain and paludal formations which are older than the Quaternary.

Acknowledgement

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