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Original Research

Arsenic in Tap Water of Serbia's South Pannonian Basin and Arsenic Risk Assessment

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Abstract

The population of Serbia's South Pannonian Basin obtains its drinking water supply from groundwater. High alkalinity; the presence of iron, manganese, and the ammonium ion; low hardness; and high concentrations of organic substances and arsenic are general characteristics of these groundwaters. One of the major drinking water supply issues in the South Pannonia Basin is the high concentration of arsenic, which in some cases exceeds several times the maximum allowable concentration. A health risk assessment model was applied to calculate cumulative exposure to As as well as toxic and carcinogenic risks resulting from drinking raw groundwater contaminated by As in the South Pannonian Basin.

Keywords: groundwater quality, arsenic, risk assessment, South Pannonian Basin

Introduction

Arsenic (As) occur naturally in the environment in a number of different forms. Some are toxic, while others are not. Pure arsenic, a gray-colored metal, is rarely found in the environment. Instead, it is usually combined with one or more other elements such as oxygen, chlorine, and sulfur. This combination is referred to as inorganic arsenic. Arsenic combined with carbon and hydrogen is referred to as organic arsenic. The organic forms are usually non-toxic or less toxic than the inorganic forms. The two most common non-toxic forms of organic arsenic are arsenobetain and arsenocholine. Organic arsenic is completely excreted within 1 to 2 days after ingestion and there are no residual toxic metabolites. In natural waters, arsenic is mainly present in its inorganic forms, As (V) and As (III). Compounds of As (III) are generally considered to be more toxic than As (V). It is therefore important to determine the presence of both types of As for the qualitative analysis of groundwater.

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Groundwater is a major source of drinking water in many parts of the world, especially in Southeast Asia (Bangladesh, Taiwan, India, Vietnam, PR China), where arsenic is from a natural source [1]. In Bangladesh shows very high cancer risk assessment because concentrations of arsenic in drinking water exceed several times the MPC for As, and values in some places are about 500 mg/l [2]. In Taiwan, populations exposed to high concentrations of arsenic in their drinking-water, containing an average of 800 mg/L of arsenic, had estimates showing high cancer risk [3]. Arsenic in groundwater of Vietnam poses a serious health threat to millions of people, in the Hanoi area there are high levels of dissolved arsenic (average 121 μg/L) [4]. Arsenic concentrations in Shany, China, the worst affected of the regions in Shanxi Province, are as high as 4.4 mg/L [5]. The problem with groundwater quality and arsenic concentrations above MPC are seen in countries that share the Pannonian Basin (Serbia, Hungary, Romania, Croatia).

Earlier research of arsenic in Serbian groundwater showed As levels from 5 to 349 μg/L [6]. In several regions of the Great Hungarian Plain the As content of groundwater is higher than the acceptable limit for drinking water

(10 μg/L). About 500,000 consumers are affected by drinking water containing As above this limit [7]. According to previous research, in Romania about 50,000 people are affected by high concentrations of arsenic in drinking water (0-176 μg/L) [8]. Croatia is also known to contain elevated naturally occurring As, and the regions Osijek-Baranja and Vukovar-Srijem are suspected to be more affected than other parts of Croatia, with total As concentrations up to 491 μg/L [9].

The Pannonian Basin is a continental lowland surrounded by mountains. It extends over a length of some 600 km in an west-east direction, from the eastern edges of the Alps in Austria and Slovenia to the Carpathian Mountains in Romania. In the south, the basin is bordered by the North Dinarides in Croatia, Bosnia and Herzegovina, and Serbia, and in the north by the Tatras in Slovakia. The Danube River is also the western boundary toward Croatia, while to the north and northeast the South Pannonian Basin (Vojvodina) is bordered by Serbia's state frontiers with Hungary and Romania. The surface area of Vojvodina is 21,506 km^2 and its population is about two million. The test area, where arsenic concentrations in groundwaters were analyzed, is divided into three districts: Banat, Bačka, and Srem (Fig. 1).

Arsenic Occurrence in Groundwater

The removal of arsenic from shallow aquifer groundwater is a worldwide challenge, especially since the EPA, WHO, and other international standards reduced the maximum permissible concentration (MPC) of arsenic from 50 to 10 μg/L [10, 11]. A large number of adverse impacts of arsenic on human health have been identified, such as skin changes, effects on the cardiovascular and respiratory systems, neurological impacts, and skin and other cancers [12]. First of all, toxic arsenic compounds act on the nervous system and blood vessels, causing paralysis of the capillaries. As a result of the interruption of blood flow and neural processes occurring, pathological changes in heart, liver, intestines, kidneys, skin, and nails occur. Arsenic is a protoplasmic poison with a pronounced effect on oxidative processes in the cell. Inorganic As(III) and As(V) are the main forms of arsenic present in natural waters, followed by their methylated forms [13]. It is especially important to determine the source and mobility of arsenic in groundwater. Its mobility depends on the highly-complex geochemical processes that take place in the aquifer. An anoxic state of groundwater increases arsenic mobility [14].

Fig. 1. Geographical location of the test area.

Based on a review of different literature sources, regions featuring high arsenic concentrations are generally associated with groundwater in geothermal areas or with low-temperature groundwater systems. Given that the subject matter of this research was groundwater used for drinking water supply, the focus is on the theoretical basis of low-temperature systems. Conditions that favor arsenic migration are associated with a reducing medium in which desorption and dissolution take place. Low Eh levels (<50 mV), the absence of dissolved oxygen, high concentrations of Fe, Mn, and NH_4^+, elevated alkalinity, the presence of organic substances, and low sulfate concentrations (<5 mg/L) are general hydrochemical characteristics of groundwater featuring elevated arsenic concentrations [15].

Some authors believe that the origin of arsenic in groundwater in the Pannonian Basin is anthropogenic. But, in hydrogeological terms, Vojvodina is comprised of thick Quaternary sediments with numerous water-bearing media of different structural porosities and clay interbeds that prevent pollutants from permeating from the ground surface. It should be noted, however, that water supply systems are comprised of relatively deep (70-250 m) drilled wells which tap Pliocene-Pleistocene sediments, while no elevated arsenic concentrations were recorded in the "first" (shallow) aquifer tapped by village wells (up to 10 m) [9], such that the origin of arsenic in groundwater within the Pannonian Basin is undoubtedly geological. Many authors have researched the origin of arsenic and associated it with clay minerals. Goldberg, S. (1988) believe that arsenic can be absorbed on the surface layer of mineral clay and that in sediments with clay interbeds there is generally more arsenic than in sands and limestones [16, 17]. In most of the affected areas, the sediment fraction is covered by a clay layer of varying thickness, which effectively prevents the ingress of oxygen into the aquifer. This, along with large amounts of organic substances deposited in the sediments, has resulted in the creation of a reducing medium where arsenic mobilization is enhanced [18].

Of the total amount of abstracted groundwater in the South Pannonian Basin, 5810 L/s (or more than 85%) is used to provide public water supply to towns and municipal centers. Based on estimates, only about 1000 L/s, or 15% of the total, is used for drinking water supply in rural settlements. More than 600,000 inhabitants of Banat and Bačka (or about 40% of the population of Vojvodina) are exposed to drinking water whose arsenic and ammonia concentrations exceed drinking water standards. Arsenic concentrations vary from 50 to 100 μg/L, with some municipalities recording up to 250 μg/L [19].

The aim of our study was to report current distribution of arsenic in drinking water from the public water supply system in the South Pannonian Basin and to show the effects of drinking arsenic-rich water.

Experimental Procedures

Sampling, Field Measurements and Laboratory Analyses

Groundwater samples from water supply wells within the territory of Vojvodina were used to perform arsenic concentration analyses as part of studies conducted by the University of Belgrade, Faculty of Mining and Geology [20], "Geozavod" from Belgrade [21], as well as the Faculty of Natural Sciences of the University of Novi Sad [22]. Total arsenic concentrations were measured in 470 water samples from public water supply systems from 2004 to 2009. Sampling and analyses of drinking water were performed at the following laboratories: at the Institute of Public Health of Vojvodina (in Novi Sad), the Institute of Public Health in Subotica, and at the Institute of Public Health in Belgrade. Current Serbian regulations limit arsenic levels at 10 μg/L [23]. The laboratory of Public Health in Subotica was accredited in 2006 as well as the laboratory in Belgrade. Sampling was undertaken in accordance with the Drinking Water Sampling and Laboratory Analysis Rulebook [24]. The laboratory for chemical testing of the environment at the Faculty of Natural Sciences of the University of Novi Sad used advanced and specialized tools for the precise required analysis. Arsenic concentrations were determined by atomic absorption spectrophotometer (Perkin Elmer Analyst 700), applying the graphite technique. The practical quantitation limit (PQL) of the method was 0.50 μg/l [22]. The metals (Fe, Mn, Na, K, Ca, Mg) are determined by atomic absorption spectroscopy after acidification with HNO_3 in accordance with EPA methods.

Based on the results of the measurement of As concentrations in the raw groundwater, cumulative exposure to arsenic for most endangered cities of Vojvodina and average daily dose were calculated.

Chemical analyses (470 samples of groundwater) were statistically processed to assess and interpret hydrochemical data and to generate a prognostic hydrochemical map. The data were statistically processed and graphically interpreted by water supply source and by populated area using statistical software IBM SPSS 15.0. The hydrochemical map showing zones of arsenic concentrations in groundwater in Vojvodina was generated using ESRI ArcGIS 9.3 and a scale of 1:650.000.

Assessment of the Risk Incurred through Drinking Water

The main source of arsenic exposure for the general population is the ingestion of drinking water with high levels of arsenic [25, 26]. Inorganic arsenic, which is extensively metabolized in humans into even more toxic methylated arsenicals, is a potent carcinogen. Consumption of drinking water highly contaminated by arsenic causes serious health problems in some countries in southeastern Asia, and arsenic poses problems for drinking-water safety world-wide [27]. The chronic exposure of humans through consumption of high levels of inorganic arsenic-contaminated drinking water is associated with skin lesions, peripheral vascular disease, hypertension, and cancers [28]. Inorganic arsenic has been determined to be a human carcinogen based on epidemiological evidence [29, 30]. It also induces a number of non-cancer effects: arsenic-specific skin lesions, conjunctivitis, peripheral neuropathy, and respiratory diseases [31]. The non-cancerogenic and the carcinogenic effects on individuals who consume As-rich groundwater for drinking were computed using the health risk assessment model derived from the US Environmental Protection Agency, Intergrated Risk Information System [32-34]. Based on the results of the measurement of As concentrations in the raw groundwater and the assumption that average 70-kg adults with an average lifespan of 73.9 years in Serbia would intake 1.5 L/day of untreated groundwater rich in As for 30 years, 365 days per year, cumulative As ingestion (CAI) of South Pannonian Basin (Serbia) is calculated. The cumulative As exposure index (CAI) was estimated using four parameters: the concentration of As in the groundwater . The cumulative As exposure index (CAI) was estimated using four parameters: the concentration of As in the groundwater (C_{water}), the age of the well (exposure duration, ED), the annual ingestion rate of groundwater (frequency exposure, EF) and daily water consumption (exposure rate, IR):

$$CAI=C_{water} \cdot IR_{water} \cdot EF \cdot ED$$

The average daily dose (ADD in mg/kg·day) was calculated using the same assumptions used to calculated CAI. Toxic risk was defined for non-carcinogenic exposure. This is calculated in terms of a Hazard Quotient (HQ). Toxic risk is considered to exist if HQ>1.00. The following equations were used:

$$HQ=ADD/RfD$$
$$ADD= (C_{water} \cdot IR_{water} \cdot EF \cdot Ed)/(AT \cdot BW)$$

RfD = 0.000304 mg/kg·day, the oral reference dose for non-cancer health effects of arsenic [33].

Carcinogenic risk is the probability of incidence of cancer from chemical exposure. It is calculated using the following formula:

$$R=1\text{-}exp(-(SF \cdot ADD)$$

...where: SF is the oral carcinogenic slope factor and for As, it is equal to 1.5 mg/kg·day.

Results and Discussion

Quality of Groundwater

The quality of groundwater used for drinking water supply in the South Pannonia Basin is considered to be conditionally poor. Groundwater suitable for public water supply is found only up to a depth of 300 m, possibly 450 m in

Table 1. Comparison of Regulations of Republic of Serbia with the EU Directives with MPC.

	EU Directive 1998/83/EC drinking water	Regulation of Hygienic Correctness of Drinking Water (Official Gazette of the SFRY No. 42/98 and 44/99)
As [μg/L]	10	10
Fe [mg/L]	0.2	0.3
Mn [mg/L]	0.05	0.05
NH_4^+ [mg/L]	0.5	0.5
pH	6.8-9.5	6.8-8.5
Na^+ [mg/L]	150	150
Ca^{2+} [mg/L]	200	200
Mg^{2+} [mg/L]	50	50
HCO_3^- [mg/L]	-	-
SO_4^{2-} [mg/L]	-	250
Cl^- [mg/L]	250	200
F^- [mg/L]	1.5	1.5

some cases. Below this depth, the physicochemical properties of the groundwater are extremely unfavorable and would necessitate costly and complex technological treatments. Iron, ammonium ion, organic substances, and arsenic are the major parameters that do not meet requirements, MPC [23, 24] (Table 1).

In general, the groundwaters in Vojvodina have stable chemical composition, because of low recharge from atmospheric precipitation, water exchange is slow enough that the groundwater is in constant interaction with the sediments. For purposes of this paper, 22 water supply systems comprised of 470 wells were assessed; groundwaters for supply are HCO_3-Na to Ca-Mg type (Fig. 2).

Arsenic Occurrence in Groundwater of Banat District

In Banat, out of the 244 analyzed samples of groundwater used for drinking water supply, arsenic concentrations were found to exceed MPC in 69 wells, meaning that 72% of the samples were chemically compliant with regard to arsenic in drinking water (Fig. 3).

Groundwater in Banat is alkaline in regards to pH (pH: 7.4-8.9). The color is generally yellow, caused by high concentrations of naturally-occurring organic substances, and the presence of humic and fulvic acids and their salts. This is a result of their inability to break down into inorganic end products due to a lack of oxygen and bacteria in the groundwater, such that the organic substances are virtually preserved. Based on its chemical composition, the groundwater is Na-HCO_3 type (Central Banat). It is dominated by sodium, in concentrations of 200-300 mg/L, while the concentrations of calcium (<50 mg/L) and magnesium (<30 mg/L) are low, which is rare in deep aquifers. The dominant anions are hydrocarbonates, while the concentrations of sulfates (3-90 mg/L), chlorides (5-50 mg/L), and fluorides (0.1-1 mg/L) are low compared to the usual levels found in groundwater [35].

Maximum arsenic concentrations in groundwater used for drinking water supply were recorded in Central Banat (217 μg/L). Arsenic concentrations in groundwater did not significantly correlate with depth, but a stratum tapped at a depth of 130 m, which serves as a source of drinking water supply for Central Banat, featured the highest concentrations of arsenic. The Piper diagram in (Fig. 2) shows the groundwaters of Central Banat which are characterized by high arsenic concentrations (the average was 50 μg/L); these groundwaters are of the Na-HCO_3 type. In South Banat arsenic is not a main cause of physicochemical non-compliance of groundwater, except for the fact that the groundwater is loaded with iron and manganese. Additionally, at many locations the ammonium ion exceeds regulated levels. Only

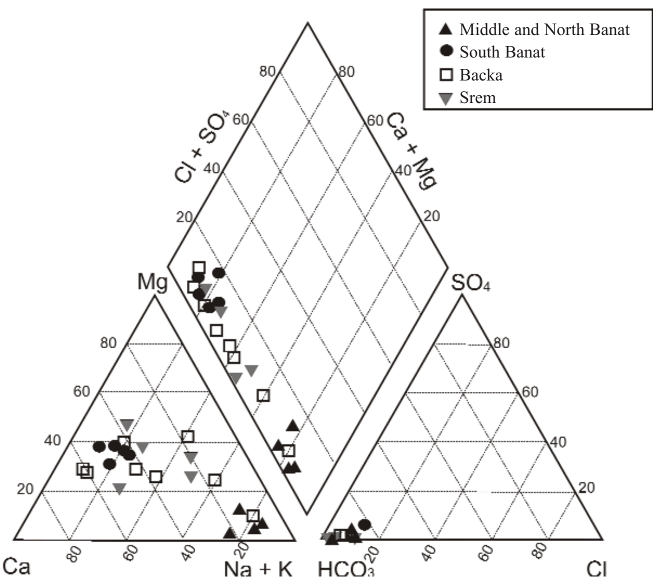

Fig. 2. Piper diagram of the composition of groundwater used for drinking water supply (22 water supply systems).

certain samples of drinking water collected in several towns within the riparian zone of the Danube measured elevated arsenic concentrations. In South Banat the ammonium ion concentrations are 0.72-2.75 mg/L, while the MPC for this ion in drinking water is 0.5 mg/L [24].

Arsenic Occurrence in Groundwater of Bačka District

Bačka generally obtains its drinking water supply from centralized public water supply systems. Rural settlements rely on local groundwater sources. With regard to the cation composition, sodium dominates all locations and all water-bearing media, except the Quaternary sediments in the extended area of Subotica, where the cation composition was primarily Ca-Mg [21]. The average arsenic concentration in Bačka groundwaters was 48 μg/L (Fig. 4).

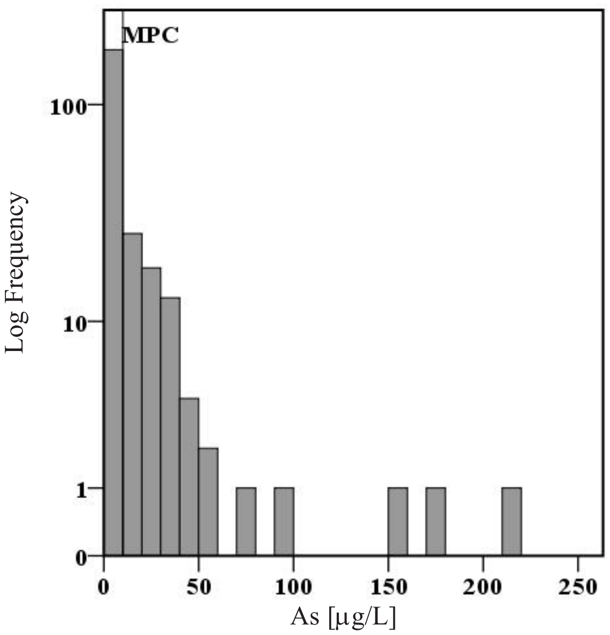

Fig. 3. Histogram of arsenic in groundwater used for drinking water supply in Banat.

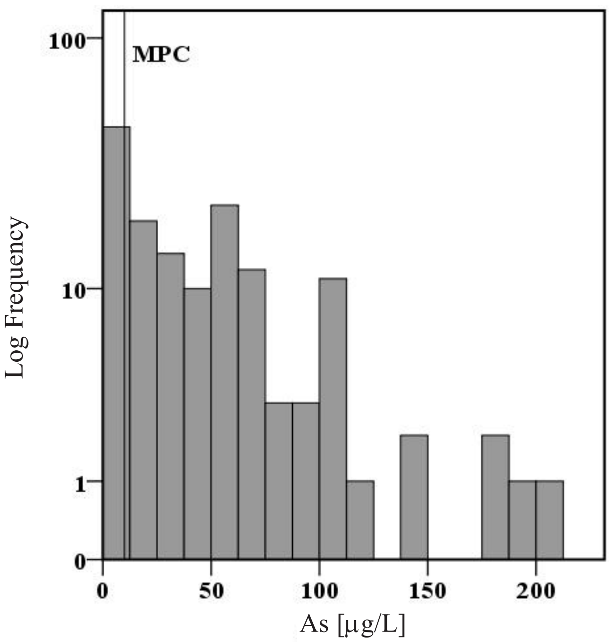

Fig. 4. Histogram of arsenic concentrations in groundwater used for drinking water supply in Bačka.

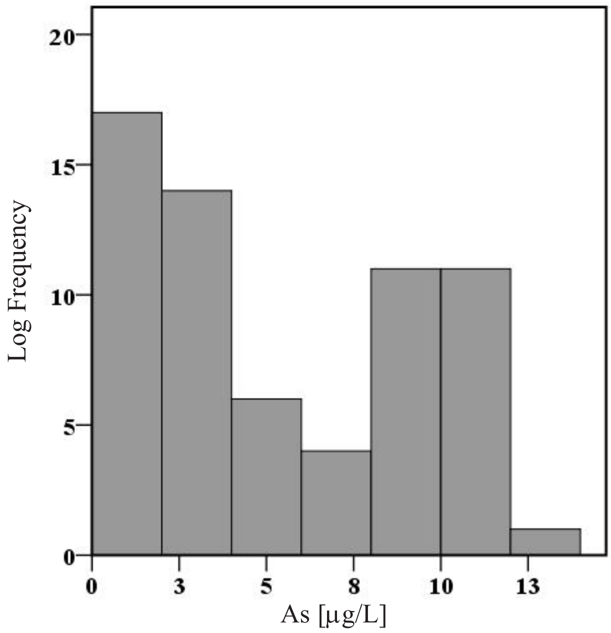

Fig. 5. Arsenic concentrations in Srem groundwaters.

The largest city in this district is Subotica (population 140,000), and this is the only city with a water treatment plant. The extended area obtains its drinking water supply from 13 groundwater sources. Based on the analyses, the average concentration of arsenic at these groundwater sources was 110 μg/L. The presence of the ammonium ion and absence of nitrates in groundwater of the test area suggests a reducing medium that creates favorable conditions for arsenic migration in groundwater. Due to the low solubility of iron oxides, hydroxides, and carbonates in groundwater, their concentrations in groundwater were found to be rather low. The iron concentration in the groundwater used for drinking water supply in Subotica measured from 0.04 to 0.96 mg/L.

Arsenic Occurrence in Groundwater of Srem District

The Srem District of Vojvodina features groundwater of good quality, with arsenic concentrations falling within the allowed range and 77% of samples having arsenic concentrations below 10 μg/L (Fig. 5). The most frequent causes of physicochemical non-compliance in Srem are concentrations of iron and manganese higher than MPC [19].

In the Srem District, the mineral content of groundwater was found to be lower than in other parts of Vojvodina. Total hardness measured from 5°dH to 17°dH. Concentrations of organic substances ($KMnO_4$ demand) were below 12 mg/L, while Fe concentrations measured about 0.3 mg/L. The public water supply is generally provided by tapping aquifers up to a depth of 250 m, which meet drinking water standards. Depths below 250 m exhibited elevated total mineralization [19].

Arsenic Risk Assessment by the Drinking Water Pathway

The end result of the assessment of groundwaters used for drinking water supply in the South Pannonian Basin should point to the importance of finding an alternative water supply for most exposed populations. This paper gives a prognostic hydrochemical map showing arsenic concentrations (Fig. 6) and assessment of the risks incurred through drinking water. According to the Provincial Secretariat of Science and Technology Development, more than 600,000 inhabitants of Banat and Bačka (or some 40% of the population of Vojvodina) obtain drinking water supply with arsenic concentrations exceeding maximum allowable concentrations in drinking water (10 μg/L) [19]. However, the actual number of the people in Vojvodina exposed to As may be lower due to the use of bottled drinking water and/or the use of private or community shallow wells, which are assumed to be As free. On the other hand, the residents of some of the villages in the study area may be exposed to drinking water with As concentrations that could lead to cancer. In comparison with studied in West Bengal, India, more than 6 million people are exposed to arsenic through their drinking water, among which approximately 300,000 people manifest signs of chronic arsenicosis [36]. It was found (in West Bengal) that it produces various systemic manifestations such as chronic lung disease, characterized by chronic bronchitis, chronic obstructive and/or restrictive pulmonary disease, and bronchiectasis; liver diseases, such as non-cirrhotic portal fibrosis; polyneuropathy; peripheral vascular disease; hypertension; nonpitting edema of feet/hands; conjunctival congestion; weakness; and anemia [37]. In Serbia the impact of arsenic on human health is insufficiently researched. The Institute of Public Health of Serbia sees association between exposure to arsenic in drinking water and the occurrence of type 2 diabetes in the Middle Banat region. The study has shown a significantly higher odds ratio and higher standardized incidence rates for the occurrence of type 2 diabetes in the population exposed to arsenic, compared to the unexposed population in Serbia [38].

The highest safe standard for cancer risk is 1 per 10,000 and the lowest standard 1 per 1,000,000 [39]. Furthermore, in the study area the cancer risk indices (R) ranged from 0.13 to 2.6 per 1,000 persons. To assess the health effects of chronic As exposure it would be necessary to accurately characterize residents' consumption patterns, namely to track changes in the sources of drinking water, duration of use, and drinking rate.

Central Banat and North Bačka featured the highest arsenic concentrations in groundwaters used for drinking water supply. At most water supply systems in South Banat arsenic concentrations remained below MPC, while levels in excess of 10 μg/L were recorded in populated areas in the riparian zone of the Danube. The Srem District generally exhibited the best quality of groundwater used for drinking water supply, with arsenic concentrations remaining below 10 μg/L.

The natural baseline concentrations of As in the groundwater defined during the hydrochemical study and presented in this paper were used for As risk assessment. The results obtained indicated that residents who consume raw

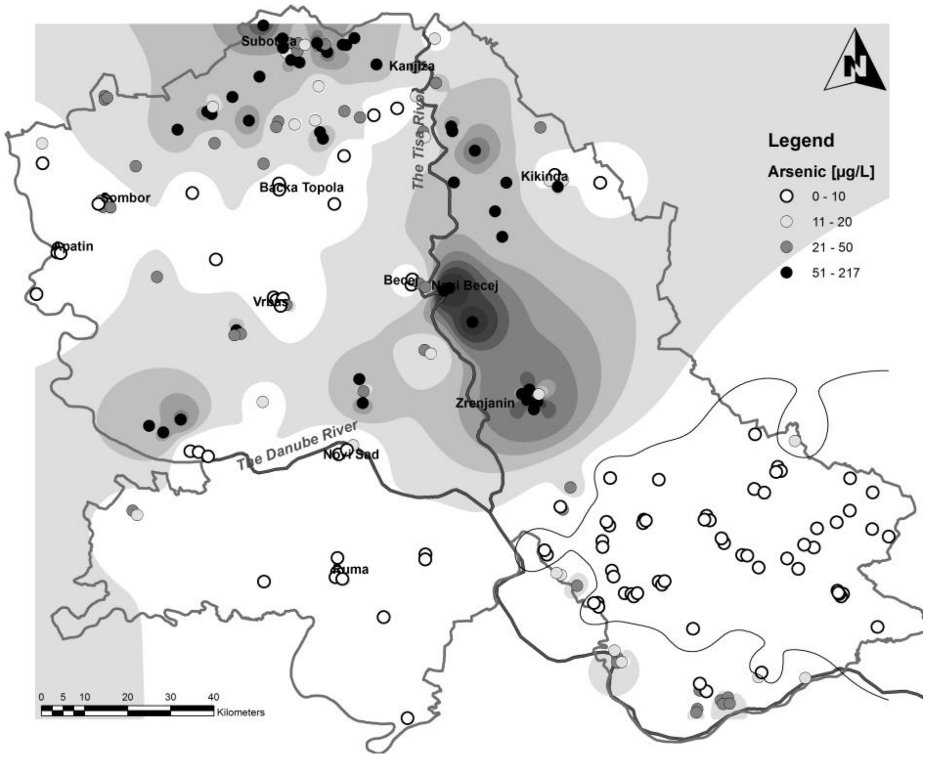

Fig. 6. Prognostic hydrochemical map with arsenic zoning of cities in Vojvodina.

Table 2. Calculated average daily dose (ADD), hazard quotient (HQ), and cancer risk (R) of residents in Vojvodina in water supplies containing arsenic concentrations higher than 10 µg/L.

As [µg/L]	ADD [mg/kg· day]	HQ	R
10	0.000087	0.29	0.000130
20	0.000174	0.57	0.000261
30	0.000261	0.86	0.000261
40	0.000348	1.14	0.000261
50	0.000435	1.43	0.000652
100	0.000867	2.86	0.001304
200	0.00174	5.72	0.002606

groundwater containing As concentrations between 10 µg/L and 200 µg/L ingest between 0.000087 and 0.00174 mg/kg day. Toxic risk calculated in terms of a hazard quotient (HQ) is considered to exist if HQ>1.00. The results of toxic and cancer risk indices calculated from the same parameters as CAI and ADD have toxic risk (HQ) ranging from 0.29 to 5.72 (Table 2). The cities with the highest As concentrations indentified in this study, and consequently with higher estimated toxic and carcinogenic risk should be connected to the new treatment for water supply. The minimum As water gave HQ and R values of 0.29 and 0.13 per 1,000 persons, respectively, and the maximum As water resulted in HQ and R of 5.72 and 2.6 per 1,000 persons. In Eastern Croatia the cancer risk is higher than in Serbia, and indices (R) ranged from 7.46 per 1,000 persons to 1.98 per 100,000 persons [40]. Interestingly, the highest maximum HQ and R values observed in Serbia are similar to those observed in Kandal Province, Cambodia. The minimum As water gave HQ and R values of 0.14 and 0.6 per 1,000 persons, and the maximum As water resulted in HQ and R of 8.8 and 3.9 per 1,000 persons [41].

Conclusions

Most of the groundwaters used for drinking water supply in the South Pannonian Basin exhibited an unsatisfactory water quality, which is a result of complex geological and hydrogeochemical characteristics. In view of the size of the test area, it was divided into three districts: Banat, Bačka, and Srem. Based on the examples presented, the variation in arsenic concentrations in groundwater differs and depends on local geological and hydrogeochemical conditions. Viewed by district, the best quality of groundwater used for drinking water supply in the South Pannonian Basin was found in Srem, characterized by the lowest percentage of arsenic concentrations exceeding MPC and relatively uniform concentrations, largely up to 10 µg/L. Conversely, the broadest range of concentrations was found in Central Banat and North Bačka. In Banat, 72% of the samples measured arsenic concentrations below 10 µg/L, with values falling within the interval from 0 to 217 µg/L. In Bačka, only 15% were below MPC, with the highest arsenic concentration being 234 µg/L, while in Srem 77% of the samples were compliant. The importance of this research of the distribution of arsenic in groundwaters used for drinking water supply, in addition to its scientific contribution, is primarily related to the impact of arsenic on human health. More than 600,000 inhabitants of Banat and Bačka (or some 40% of the population of the South Pannonian Basin) obtain drinking water supply with arsenic concentrations exceeding maximum allowable concentrations in drinking water (10µg/L). Cities with As concentrations higher than 30 µg/L have hazard quotients exceeding 1.0, which indicates the existence of toxic risk. Futhermore, in the study area the minimum As water gave HQ and R values of 0.29 and 0.13 per 1,000 persons, respectively, and the maximum As water resulted in HQ and R of 5.72 and 2.6 per 1,000 persons, and this should indicate the importance of developing a regional water supply system.

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References

1. JACK C. NG, JIANPING W., AMJAD S. A global health problem caused by arsenic from natural sources, Chemosphere, **52**, (9), 1353, **2003**.
2. SMITH A. H., LINGAS E. O., RAHMAN M. Contamination of drinking-water by arsenic in Bangladesh. A public health emergency: http://www.who.int/bulletin/archives/78(9)1093.pdf)
3. CHEN C.J., CHEN C.W., WU M.M., KUO T.L. Cancer potential in liver, lung, bladder and kidney due to ingested inorganic arsenic in drinking water. Brit. J. Cancer, **66**, 888, **1992**.
4. BERG M., KIM TRANG P.T., STENGEL C., BUSCHMANN J., HUNG VIET P., VAN DAN N., GIGER W., STÜBEN D. Hydrological and sedimentary controls leading to arsenic contamination of groundwater in the Hanoi area, Vietnam: The impact of iron-arsenic ratios, peat, river bank deposits, and excessive groundwater abstraction, Chem. Geol., **249**, (1-2), 91, **2008**.
5. SUN G.F., PI J.B., LI B., GUO X.Y., YAMAUCHI H., YOSHIDA T. Progresses on researches of endemic arsenism in China: population at risk, intervention actions, and related scientific issues W.R. Chappell, C.O. Abernathy, R.L. Calderon (Eds.), Arsenic exposure and health effects IV, Elsevier, Amsterdam, pp. 79-85, **2001**.
6. JOVANOVIĆ D., JAKOVLJEVIĆ B., RAŠIĆ-MILUTINOVIĆ Z., PAUNOVIĆ K., PEKOVIĆ G., KNEZEVIĆ T. Arsenic occurrence in drinking water supply systems in ten municipalities in the Vojvodina Region, Serbia. Environ. Res. **111**, 315, **2011**.
7. VARSANYI I., KOVACS O.L. Arsenic, iron and organic matter in sediments and groundwater in the Pannonian Basin, Hungary. Appl. Geochem. **21**, 949, **2006**.

8. ROWLAND H.A.L., OMOREGIE E.O., ROMAIN M., JIMENEZ C., MERTENS J., BACIU C., HUG S.J., BERG M. Geochemistry and arsenic behaviour in groundwater resources of the Pannonian Basin (Hungary and Romania). Appl. Geochem. **26**, 1, **2010**.
9. UJEVIĆ M., DUIĆ Ž., CASIOT C., SIPOS L., SANTO V., DADIĆ Ž., HALAMIĆ J. Occurrence and geochemistry of arsenic in the groundwater of Eastern Croatia. Appl. Geochem. **25**, 1017, **2010**.
10. US EPA United States Environmental Protection Agency, [Online] **2006**: http://water.epa.gov
11. WORLD HEALTH ORGANIZATION [Online]: http://www.who.int
12. PETRUSEVSKI B., SHARMA S.K., SCHIPPERS J.C., SHORDT K. Arsenic in drinking water. Delft: IRC International Water and Sanitation Centre, **17**, 1, **2007**.
13. BEDNAR A.J., GARBARINO J.R., RANVILLE J.F., WILDEMAN T.R. Field and laboratory arsenic speciation methods and their application to natural-water analysis. 2001. Water Res., **38**, (2), 355, **2004**.
14. KIM Y.T., YOON H.O., YOON C., WOO N.C. Arsenic species in ecosystems affected by arsenic-rich spring water near an abandoned mine in Korea. Environ. Pollut. **157**, (12), 3495, **2009**.
15. SMEDLEY P.L., KINNINBURGH D.G. A review of the source, behaviour and distribution of arsenic in natural waters. Appl. Geochem. **17**, 517, **2002**.
16. GOLDBERG S. Chemical Modeling of Specific Anion Adsorbtion on Oxides, Clay Minerals, and Soils, ENVIROSOFT, CML Publications, Ashurst Southampton, U.K. **86**, pp. 671-688, **1986**.
17. MANNING A.B., GOLDBERG S. Modeling arsenate competitive adsorption on kaolinite, montmorillonite and illite. Clay. Clay Miner. **44**, (5), 609, **1996**.
18. ANWAR J., Argument against natural origin of arsenic in groundwater. Abstract from Arsenic Poisoning in Bangladesh, [Online] **2011**: http://www.sos-arsenic.net/english/natural_origin/1.html#sec4
19. PROVINCIAL SECRETARIAT OF SCIENCE AND TECHNOLOGY DEVELOPMENT (PSSTD). Water Supply and Water Protection Strategy for the Autonomous Province of Vojvodina. University of Novi Sad, Faculty of Science and Mathematics, **2009** [In Serbian]. http://www.eko.vojvodina.gov.rs/files/file/dokumenti/sajt%20strategija%20vodosnab devanja%20i%20zastite%20voda%20apv.pdf
20. PAPIĆ P. Groundwater quality of North Backa, utility assessment and treatment. Final report of the project: SUDEHSTRA. University of Belgrade, Faculty of Mining and Geology, Department of Hydrogeology, Belgrade, pp. 71-75, **2009**.
21. MANDIĆ M., PAPIĆ P. A Contribution to the Understanding of the Qualitative Properties of Groundwater in Vojvodina. Novi Sad: 14th Conference of Geologists of Serbia and Montenegro, pp. 385-390, **2005** [In Serbian].
22. DALMACIJA B. Monitoring of arsenic in public water supply wells in South Banat (in Serbian). Novi Sad, **2008**. http://www.eko.vojvodina.gov.rs/files/file/monitoring/voda/2008/juzni%20banat%20arsen%202008.pdf [In Serbian].
23. EU Directive 98/83/EC. Council Directive of 3 November 1998 on the quality of water intended for human consumption. Official Journal of the European, **1998**.
24. REGULATION OF HYGIENIC CORRECTNESS OF DRINKING WATER. Official Gazette of the SFRY No. 42/98 and 44/99
25. WORLD HEALTH ORGANIZATION, Environmental health criteria 18: arsenic, Switzerland, **1981**.
26. AGENCY FOR TOXIC SUBSTANCES AND DISEASE REGISTRY, Toxicological profile for arsenic Atlanta, **1993**.
27. SCHUHMACHER-WOLZ U, DIETER HH, KLEIN D, SCHNEIDER K., Oral exposure to inorganic arsenic: evaluation of its carcinogenic and non-carcinogenic effects. Crit. Rev. Toxicol., **39**, (4), 271, **2009**.
28. SAMS R., WOLF D. C., RAMASAMY S., OHANIAN E., CHEN J., LOWIT A., Workshop overview: Arsenic research and risk assessment. Toxicol. Appl. Pharm., **222**, (3), 245, **2007**.
29. USEPA, Recommendations for and documentation of biological values for use in risk assessment. EPA 600/6–78/008, NTIS PB88–179874/AS, **1988**.
30. NRC, Arsenic in Drinking Water, National Academy Press, Washington, DC, **1999**.
31. DE CHAUDHURI S., KUNDU M., BANERJEE M., DAS J.K., MAJUMDAR P., BASU S., ROYCHOUDHURY S., SINGH K.K., GIRI A.K. Arsenic-induced health effects and genetic damage in keratotic individuals: Involvement of p53 arginine variant and chromosomal aberrations in arsenic susceptibility, Mutat. Res.-Rev. Mutat., **659**, (1-2), 118, **2008**.
32. IRIS (Integrated risk information system) Arsenic, inorganic. CASRN 7440-38-2; [Online] – March **2012**. http://www.epa.gov/iris/subst/0278.
33. US EPA (United States Environmental Protection Agency). Integrated Risk Information System [Online]. - March **2012**. http://cfpub.epa.gov/ncea/iris/compare.cfm.
34. WHO [Online]. – May **2012**. http://www.euro.who.int/en/where-we-work/member-states/serbia/facts-and-figures.
35. VUKOJE M.S. Water Supply in Zrenjanin (in Serbia). Yugoslav Association of Water Supply and Sewerage, pp. **52**, 87-90, **2001** [In Serbian].
36. CHAKRABORTI D., RAHMAN M.M., PAUL K., CHOWDHURY U.K., SENGUPTA M.K., LODH D., CHANDA C.R., SAHA K.C., MUKHERJEE S.C., Arsenic calamity in the Indian subcontinent: what lessons have been learned. Talanta, **58**, 3, **2002**.
37. MAZUMDER D.G., DASGUPTA U.B., Chronic arsenic toxicity: Studies in West Bengal, India, The Kaohsiung Journal of Medical Sciences, **27**, (9), 360, **2011**.
38. JOVANOVIC D., RASIC-MILUTINOVIC Z., PAUNOVIC K., JAKOVLJEVIC B., PLAVSICA S., MILOSEVIC J., Low levels of arsenic in drinking water and type 2 diabetes in Middle Banat region, Serbia, Int. J. Hyg. Envir. Heal. Available online 10 February **2012**: http://www.sciencedirect.com/science/article/pii/S1438463912000028
39. SMITH A. H., BIGGS M. L., MOORE L., HAQUE R., STEINMAUS C., CHUNG J., HERNANDEZ A., LOPIPERO P, Cancer Risk from Arsenic in Drinking Water. Implications for Drinking Water Standards, Elsevier Science, pp. 191-199, **1999**.
40. UJEVIĆ BOŠNJAK M., CAPAK K., JAZBEC A., CASIOT C., SIPOS L., POLJAK V., DADIĆ Ž., Hydrochemical characterization of arsenic contaminated alluvial aquifers in Eastern Croatia using multivariate statistical techniques and arsenic risk assessment, Sci. Total Environ. **420**, 100, **2012**.
41. STHIANNOPKAO S, KIM KW, CHO KH, WANTALA K, SOTHAM S, SOKUNTHERA C, KIM J.H., Arsenic levelin human hair, Kandal Province, Cambodia: the influences of groundwater arsenic, consumption period, age and gender. Appl. Geochem., **25**, 81, **2010**.