ARSENIC CONTAMINATION IN ENVIRONMENT IN THE REGION OF VOJVODINA

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SUMMARY

Ground waters in the region of Autonomous Province of Vojvodina, Republic of Serbia are endangered by arsenic (up to 0.750 mg/l). Total arsenic concentration was determined In samples of untreated and treated water from some local and central water-supply systems. Results are compared to actual regulations in the country. This analysis encompassed 324 drinking water samples of various sources, analyzed in the Institute of Public Health Novi Sad, during 2005. Determined concentration of total arsenic in drinking water varies from 0.005 to 0.450 mg/l. Arsenic concentration in the river Danube water was within recommended value for I-II class. Maximum arsenic daily intake through food and nutrition was 60.9± 22.3 µg/day in 2000. To understand importance of the problem of arsenic environment contamination in Vojvodina region, our own results, as well as the results of other authors are presented and analyzed (drinking water: 173 samples, moss deposition and daily intake through nutrition).

Key words: arsenic, environment, daily intake, human health

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INTRODUCTION

Arsenic (As) belongs to the Vth group of periodical elements and it is broadly distributed worldwide, most often as an arsenic sulfide or as a metal arsenates and arsenides. Some soil microorganisms could transform arsen into volatile organic arsine, which could be found in the air. In water environment, it attaches to sediment and some species of fishes and Crustacea class cumulate it in their tissue. Arsenicals are used commercially and industrially, every so often as an herbicide compound which use is increasing (1, 2).

Arsenic is introduced into drinking water sources primarily through dissolution of naturally occurring minerals. The most important route of human exposure is through oral intake of food, drinking water and beverages. There are a number of regions worldwide where arsenic may be present in drinking water sources, particularly in groundwater, at elevated concentration (3–9).

Province of Vojvodina (Fig. 1) represents Pannonian lowland where serious big arsenic contamination through the water and geological source can be expected. In this province live 2.031,992 inhabitants: 1.152,295 in urban and 879,697 in rural areas (10).

Most of arsenic-contaminated areas of Vojvodina are in the region of alluvial formation along the banks of the rivers Danube and Tisa (with confluent rivers from Romania: Zlatica, Begej, Tamis, Nera), Palic lake and groundwater from the rest of the province areas (depth of tube well from 60 to ≥100 meters): sub/artesian well. In groundwater in Vojvodina region arsenic concentrations are reaching levels above 0.010 mg/l. Arsenic concentration in the river Danube was within recommended value of 50 µg/l for I–II class during 1986/2003 (Official bulletin of Yugoslavia No 8/1978) (in its whole course through the Republic Serbia (11–13).

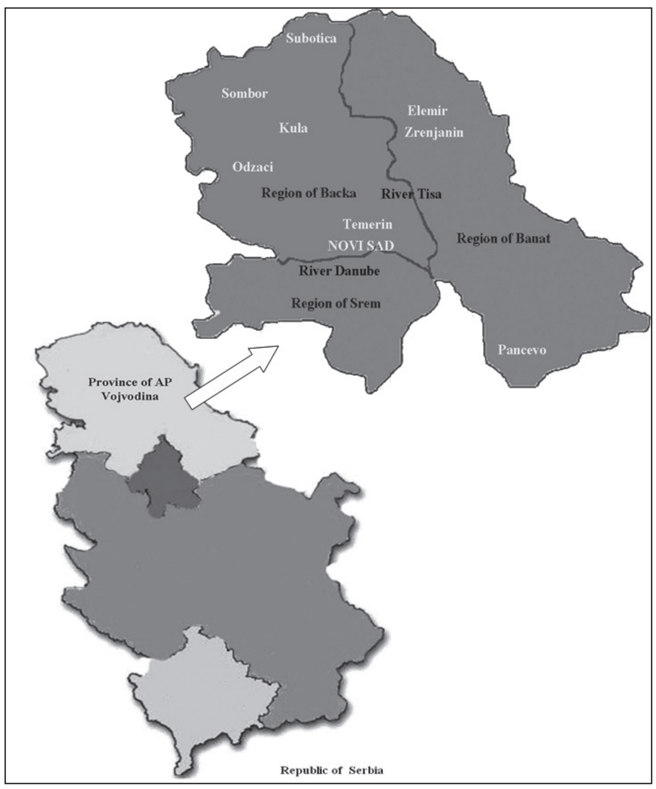

Fig. 1. Province of Vojvodina, north part of Serbia.

Primary Objective

To understand the significance of the arsenic contamination problem of environment in Vojvodina, we present our results of arsenic analysis (Institute of Public Health Novi Sad, Vojvodina) in drinking waters (drinking water – treatment and non-treatment), waters of the river Danube, as well as results of other authors (drinking water – treatment and non- treatment, food and As deposition in moss as bio-indicator).

Critical Analysis of the Arsenic Contamination Problem in Environment

There are sources of arsenic ores in the soil. It can be found in minerals, coal and petroleum; it is released during exploitation and use; in nature it is mostly connected with copper and iron minerals. Humans also influence changed arsenic concentration (1, 2, 9, 11, 13, 14). Concerning mineral-geological structure, soil of the Province of Vojvodina has arsenic concentration from 0.1 to 40 mg/ per kg^{-1}, which is in accordance with results of other authors (15, 16).

Water is the most important vehicle of As in nature. Arsenic concentration in waters varies from 1–2 μg/l (9, 13). In existing literature there are very few data concerning As concentration in water resources of former Yugoslavia.

In good oxygenated surface water five-valent arsenic is the most frequent form. In the sediment of deep lakes or ground water predominant form is trivalent arsenic (9, 14). Increased pH can be the result of increased arsenic solution in water (17). Some thermal sources in New Zealand and Russia contain extremely high quantities, which is in relation to mineral structure of deep soil layers (18). A recent survey from Bangladesh has indicated that 11% of tube-well water contains arsenic in the range from 0.01 mg/l to 0.05 mg/l and 29% are above the WHO maximum permissible limit of 0.05 mg/l. None of the water samples from tube-wells of less than 18 meters depth showed arsenic level above 0.05 mg/l (19). The latest statistics indicates that 80 % of Bangladesh and an estimated number of 40 million people are at risk of arsenic poisoning-related diseases because the ground water in these wells is contaminated with arsenic (3).

Besides already explained transmission ways in ground waters, natural erosive processes are also mentioned. Industrial waste containing As and other potentially dangerous matters represent risk of fatal pollutions and many cause extensive damages (residues in pharmaceutical industry, color industry, pesticide industry etc.) (6, 8, 9, 14, 20).

Arsenic concentrations in sediment of water ecosystems in some countries are also cited: the lowest values are registered in Canada (6 mg/kg), in the Netherlands 29 mg/kg and basic concentration for Danube (period 1950–1970) was 10 mg/kg (21).

Published results of various studies on As concentration in water and their variety (comparative samples) are most probably result of different methods, techniques, apparatus applied, as well as of their identification and expression in the form of total arsenic from organic and inorganic compounds; trivalent, five-valent arsenic etc. (22, 23). High arsenic concentrations in drinking water are registered in the some parts of Vojvodina (region of Backa, Middle and North Banat and plain parts of Srem). Colleagues from Zrenjanin-Banat (24) reported that in the period 1987–2001 in all water samples from different locations in water-supply system concentration of inorganic arsenic was above recommended value.

Analysis of foodstuff samples from certain area (region Vojvodina) should on time indicate risks in nutrition chain, due to contamination, pollution of air, soil, surface and ground. Arsenic concentration in food is usually under 1 mg/kg of weight. According to WHO (8, 25) arsenic quantity in plants is around 0.4 μg/g; in fishes 1–10 μg/g (mainly in organic form) and in the group “other”: 0.25 μg/g. Results of arsenic analysis in seven-day whole-day nutrition in selected population group in our country (26) have pointed out a significantly higher intake values than those recommended by World Health Organization (8, 14, 25). Agency for registration of poisons and diseases in USA, in association with toxicological profile of arsenic cites the value of 0.3 μg/kg/day of inorganic arsenic as the limit under which there are not any undesirable effects in case of oral intake lasting longer than one year (27).

Expected daily arsenic intake in adults ranges from 16.7 to 129 μg and in children from 1.26 to 15.5 μg in this region. It is assumed that 25% of arsenic intake is of inorganic source and 75% of organic source (9, 25). Participation of As (III) in total arsenic ranges from 40–46% (9). Meacher et al. (28) in investigation that included over 100,000 persons have determined that the highest quantity of arsenic intake is through food and then through water while intake through air is insignificant. In this study, daily inorganic arsenic intake from food, water, soil and from airborne particle inhalation in US adults was estimated. To account for variations in exposure across the USA, a Monte Carlo approach was taken using simulations for 100,000 individuals representing the age, gender profile and country of residence of the US population based on census data. The exposure is the best represented by the ranges of inorganic arsenic intake (at the 10th and 90th percentiles) which were 1.8 to 11.4 μg/day for males and 1.3 to 9.4 μg/day for females.

Inorganic arsenic in dietary staples (i.e. yams and rice; in the 1995) may have substantially contributed to exposure and adverse health effects observed in an endemic Taiwanese population historically exposed to arsenic in drinking water (29). These data support a likely mean dietary intake of 50 μg/day with a range of 15 to 211 μg/day. Consideration of dietary intake may result in a downward revision of the assumed potency of ingested arsenic as reflected in EPA toxicity values.

METHODS AND PROCEDURES

Institutes of Public Health in Novi Sad, Vojvodina, in all seven regions of the Vojvodina collect random water samples from own territories, according to law regulations (30) and WHO recommendations (8, 9). Institute of Public Health Novi Sad has used VDM 016 method (Standard Methods for the Examination of Water and Wastewater, 20th edition, Baltimore Maryland, 1998) for identification of total arsenic with atomic absorption spectrophotometer (AAS) – hydride technique at apparatus Perkin Elmer Analyst 300 with addition for hydride technique MHS 15 (method is accredited) (31). Chemicals of p.a. purification were used. The analysis included sample concentration by nitrogen acid (HNO_3 65%; 1.40 g/l). This As was reduced in arsine with natriumborhydride. Arsine was eliminated from solution by aeration with nitrogen in acetylene flame, where it was determined by atomic absorption at 193.7 nm.

Quality control within analytic procedure included validation of method and determination of allowed inaccuracy. Results were expressed as total arsenic, which was in agreement with existing law regulations in the Republic Serbia (30) as well as World Health Organization recommendations (9) and Council Directive 98/83/EC (32): 0.010 mg/l (independently from the arsenic form).

__Table 1.__ Arsenic concentration in purified chlorinated drinking water during 2005 – Southern Backa region

Regional water supply system Novi Sad /cities No.	No. of samples	Detected value /mg/l	Recommended value /mg/l*
12	29	<0.005	0.01

**Limit method detection /AAS/: <0.005 mg/l*

__Table 2.__ Arsenic concentrations in non-purified chlorinated drinking water during 2005 – Region of Southern Backa

Local water supply system /cities No.	No. of samples	Minimum detection range /mg/l	Maximum detection range /mg/l	Recommended value /mg/l
21	55	<0.005–0.084	<0.005–0.152	0.01

__Table 3.__ Arsenic concentrations in non-purified drinking water during 2005 – Region of Srem

Local water supply system /cities No.	No. of samples	Minimum detection range /mg/l	Maximum detection range /mg/l	Recommended value /mg/l
4	13	< 0.005–0.010	0.006–0.022	0.01

__Table 4.__ Arsenic concentrations in purified chlorinated drinking water during 2005 – from different cities in Vojvodina (regions of Backa, Srem and Banat)

Regional water supply system /cities No.	No. of samples	Minimum detection range /mg/l	Maximum detection range /mg/l	Recommended value /mg/l
9	43	< 0.005–0.048	< 0.005–0.180	0.01

__Table 5.__ Arsenic concentrations in non-purified chlorinated drinking water during 2005 – from different cities in Vojvodina (regions Backa, Srem and Banat)

Regional water supply system /cities No.	No. of samples	Minimum detection range /mg/l	Maximum detection range /mg/l	Recommended value /mg/l
20	37	< 0.005–0.107	< 0.005–0.108	0.01

__Table 6.__ Arsenic concentrations in non-purified chlorinated drinking water during 2005 – from different cities in Vojvodina (regions Backa, Srem and Banat)

Regional water supply system /cities No.	No. of samples	Minimum detection range /mg/l	Maximum detection range /mg/l	Recommended value /mg/l
32	147	<0.005–0.088	<0.005–0.450	0.01

MAIN RESULTS AND DISCUSSION

Survey of Identified Arsenic Concentrations in Drinking Waters of Vojvodina According to Regions

Southern-Backa region. Institute of Public Health Novi Sad analyzed in water quality surveillance this region. During 2005 out of 324 water sources investigated, in 93 (28.70%) samples arsenic concentration was above the World Health Organization Recommended Value (RV) (2004) of 0.01 mg/l and 231 (71.26%) samples were within the RV. Among these 321 samples, 27 (8.33%) samples had arsenic concentrations above the WHO Maximum Permissible Limit (MLP) of 0.05 mg/l. Single maximum concentration of 0.450 mg/l was detected in the city of Temerin (25 km far from Novi Sad, capital of the Province of Vojvodina).

In purified, chlorinated drinking waters As concentrations above recommended values have not been identified (Table 1). These levels are exceeded in non-purified groundwater systems from wells and from various depths: 60–200 m (Tables 2 and 3). Regional water supply system Novi Sad takes water from alluvial parts of the river Danube by Reni wells from 40–60 m depth. Purification system is as follows: raw water – aeration, coagulation, sand filter, activated carbon, chlorination (gas chlorine).

Drinking water consumers from various regions of Vojvodina were concerned for their health, so they took samples of their own to the Institute of Public Health Novi Sad, where analyses were carried out. The method of sampling was explained in details to each person (Tables 4–6).

After identification of increased As concentrations in drinking water, it was recommended to consumers not use it for drinking. Municipality authorities had provided good condition drinking water in tankers (cars) in all problematic cities. Some settlements still use drinking water from cisterns.

West region of Backa. This region is in charge of the Institute of Public Health Sombor (22). Problem of arsenic in drinking water of this town and surroundings appeared in recent days. In central water-supply system in Odzaci, in all seven actual existing sources – wells, as well as from samples collected from the system, values above recommended value were identified (Table 7).

Table 7. *Arsenic concentration in drinking water in municipality of Odzaci – West region of Backa during 2001–2002 (mg/l)*

Place of laboratory where analyses were done	As conc. in distribute system	Well - 1	Well - 2	Well - 5	Well - 6	Well - 7	Well - 8	Well - 9
		146 m	240 m	240 m	138 m	138 m	90 m	81 m
Institute of Public Health, Novi Sad	0.3	-	-	<0.01	0.27	0.16	0.38	-
Institute of Public Health, Belgrade	0.38	0.7	0.022	0.016	0.64	0.72	0.52	0.018
Institute of Public Health, Subotica	0.125	0.609	0.049	0.046	0.456	0.582	0.605	0.05

Middle part of the region Backa. Bijelic et al. (33) have analyzed arsenic concentration in drinking water of 7 settlements in Kula municipality. Population use water from 25 wells that is drawn from depth of 60–200 m. All analyses were performed in “Bio-ecological Center” in Zrenjanin, using analytic procedure AAS (31, 34). Arsenic concentration in drinking water in four settlements was within of RV (Crvenka, Nova Crvenka, Lipar, Kula), while in other settlements (Sivac, Ruski Krstur, Kruscic) it was 1.02–5.59 times above value defined in the Regulations (30). Authors (33) have noticed connection of arsenic concentration values and depth of the well, applying Pierson’s correlation coefficient of 0.5. Obtained correlation coefficient was characterized as modestly strong, which somehow indicated that increased depth of the well was related to increased arsenic concentration and vice versa.

Northern Backa region of Vojvodina (Institute of Public Health, Subotica). According to investigations conducted at more than 80 locations (total number of samples: 80) in surroundings of Subotica and banks of the river Tisa arsenic concentrations were above 0.01 mg/l level (in 63% of cases from 0.023 to 0.123 mg/l) (23). Data from Jovanovic & Stanic (15) concerning North part of Backa region (raw water and water from water-supply system) indicated that out of 30 samples, in 6 were the arsenic concentrations within recommended value (from 0.001 to 0.006 mg/l) while in 24 samples concentrations ranging from 0.024 to 0.168 mg/l were identified. The subject of analysis was comparison of raw and purified waters from the same sources, with regards to concentrations of total arsenic and iron. It was noticed that after water chlorination, iron decreased, as well as arsenic concentration, which was explained by adsorption of arsenic particles on iron, and then, by their mutual elimination.

Middle-east region of Banat (Institute of Public Health, Zrenjanin). Existing system of water supply of Zrenjanin is based on drawing ground water from source from the northern part of the town. Ground source is capped by the system of 30 deep wells (70–130 m), settled in two lines with mutual distance of around 1200 m. Distance between wells in the same line is around 300 m. Through high pressure pumps is water from wells directly injected into water supply system and treated by gas chlorine; in such a way water reaches its consumers without any technological processing. Depending on the needs of the town, different number of wells is used (24). According to results of the Institute of Public Health in Zrenjanin, in the period 1987–2001 concentration of arsenic in five of seven analyzed wells from the source of the central water supply system in Zrenjanin was 5–10 times above RV while in two samples it was 2–3 times above this value (Table 8) (24). Dissimilar to the main water source, in two of five public wells in Zrenjanin which are located far a way from the area of active agricultural production and earth gas source, concentration of arsenic was in accordance with requirements of Regulation on drinking water safety in Serbia (1998).

In the Table 9 there are presented arsenic concentrations in public wells in Elemir (village near Zrenjanin where are located the greatest oil and earth gas wells in the country) and in Zrenjanin (24).

Aiming to overcome the arsenic problem in drinking water, pilot plant for purification of drinking water located in Elemir was analyzed. This plant was constructed according to the project of an expert from the Faculty of Physical Chemistry Belgrade, and function on the principle of combined filters, transforming trivalent in five-valent arsenic compounds and afterward eliminating arsenic residues as well as other matters by various adsorbents – ferrous hydroxide and magnesium oxide (35). Here we have not obtained desirable results.

In order to improve drinking water quality (35) a new Pilot plant in cooperation with Department of Chemistry – Science faculty in Novi Sad has been put in operation in the city of Zrenjanin in 2005. Investigations are still going on.

Table 8. *Arsenic concentration in wells of the water source in Zrenjanin (mg/l) (N = 16)*

Year	Well - 6	Well - 9	Well - 25	Well - 26	Well - 28	Well - 29	Well - 30
1987	0.080	0.07	0.09	0.07	0.05	0.100	
1990	0.015			0.18			
1991	0.006				0.11		
1995		0.076			0.05		
2000	0.021	0.065				0.034	0.053

Table 9. Arsenic concentration in drinking water of public wells (mg/l) (N =7)

City/year	Well - 1	Well - 2	Well - 3	Well - 4
Elemir/1990	0.31	0.14	0.048	
Zrenjanin/2001	0.100	0.051	0.011	0.003

Table 10. Daily arsenic intake by nutrition in Serbia

	Year				
Element	1997	1998	1999	2000	2001
As μg/day	9.6±2.3	14.6±2.2	16.7±7.0	60.9±22.3	42.7±14.7

Arsenic in the Air and Soil

Airborne concentration of arsenic ranges from 1 ng/m^3 to 10 ng/m^3 in rural areas and from a few nano-grams per cubic meter to about 30 ng/m^3 in non-contaminated urban areas. It could be found as the result of atmospheric deposition in suspended form, with other particles (20, 36).

There are sources of arsenic ores in the soil. It can be found in minerals, coal and petroleum (particularly appears after combustion of these fossil fuels) etc. In nature it is most frequently linked to minerals of copper and iron. Well-known fact is that above 65% of arsenic come from mineral raw materials where it is produced from copper.

Applying bio-monitoring of heavy metal deposition in moss: a) Hylocomium splendes; b) Pleurozium schreberi; c) Hypnum cupressiforme, Krmar & Radnovic (37), have composed the map of arsenic space deposition in the territory of Vojvodina within the project Atmospheric Heavy Metal Deposition in Europe – estimations based on moss analysis (Fig. 2). The greatest concentrations are identified in the Eastern Serbia where the copper mine Bor is located. In Vojvodina identified arsenic concentrations in moss were under 13 ppm.

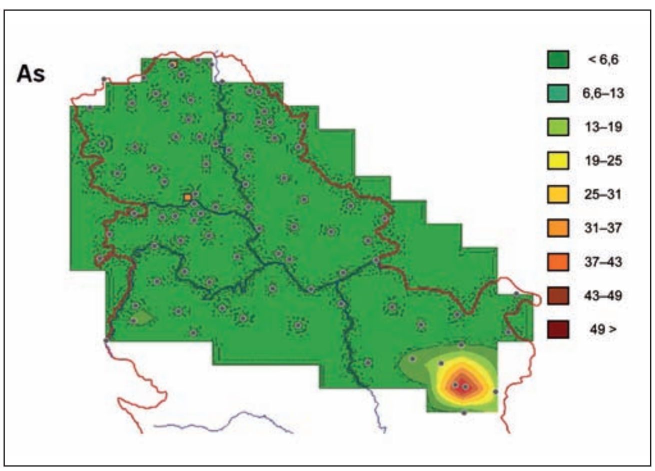

Fig. 2. Survey of space arsenic concentration in Vojvodina (in ppm).

Food and Nutrition

Concentration of some elements in soil and their concentration in food, i.e. in food chain, is in relation to many geological and anthropogenic factors. Military wars in the territory of former Yugoslavia in the last decade of previous century may serve an example.

WHO data (20) have indicated that average daily arsenic intake by nutrition is 30 μg/kg.

In Serbia the group of experts (26) has followed-up quality of nutrition with particular emphasis on toxic metal and metalloid residues. Intake of some macro and microelements was followed-up during seven-day nutrition survey in the same chosen group of persons (N=12) from different localities in Serbia, from 1997 to 2001 in the spring period, with particular emphasis on the war year in 1999. Authors have indicated that As intake increased in our country (Table 10).

In 2000, the year after bombardment of the country, arsenic intake by nutrition reached 60.9±22.3 μg/day, which was 6.34 times larger than in 1997. In 2001 arsenic intake by everyday nutrition decreased to 42.7±14.7 μg/day, which was lower by 18.2 μg/day compared to 2000.

Higher values for As intake by nutrition were identified in nearly the same seven-day nutrition survey of volunteers in Pancevo, Zrenjanin – Vojvodina, and Kragujevac (middle part of the Republic Serbia) (26).

CONCLUSIONS

The presented study indicates that there is an arsenic problem in untreated drinking water in the Province of Vojvodina pumped from deep wells (depth 60–200 m), that provide water for more than 60% population all over the province. These waters have high arsenic concentrations (0.72 mg/l – middle part of the region Backa and Temerin 0.450 mg/l – town near Novi Sad, southern part of Vojvodina).

Treated drinking waters have As concentration within recommended value. Average daily As intake through nutrition increased after war in 1999, in comparison with the earlier period: 60.9±22.3 μg/day (2000) and 42.7±14.7 μg/day (2001). These values are significantly higher than those recommended by Environmental Protection Agency and World Health Organization. Arsenic deposition in moss which was analyzed 2 years after war situation, had not categorized this problem as highly risk.

Prevention of the arsenic contamination problem of drinking water in Vojvodina can be achieved by construction of great regional systems for ground water purification, which would cover all settlements. In this way population will be in position to have safety drinking water.

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