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## ARSENIC IN DRINKING WATER – CASE STUDY IN GEVGELIJA

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**Abstract.** This paper analyses the situation of arsenic concentration in drinking water from the city water supply system in Gevgelija for the period 2011–2019. Under the slopes of Kozuv, the ground-water carries with them to the source of water supply, inorganic arsenic originating from the rocks themselves. A series of sampling of drinking water was performed and it was observed that in the drinking water samples from the drilled well area “Moin” the concentrations of arsenic were increased above the MAC by 10 µg/l, while samples from the Vardar well were within the permissible limits. The Institute of Public Health of the Republic of North Macedonia submitted to the Directorate of Food at the Ministry of Health an opinion on urgent measures to prevent the possible risk of arsenic through drinking water for the health of the population of the city of Gevgelija and the surrounding areas (about 20 000 inhabitants). The aim of paper was to assess the risk through analyses and evaluation of level of arsenic in drinking water, which is important for prevention of arsenic-related disease by providing safe drinking water from new wells from other side of the river Vardar.

*Keywords:* arsenic, drinking water, public health.

### AIMS AND BACKGROUND

Arsenic (As) is a heavy metal and according to the Priority List of Dangerous Substances it ranks first in human toxicity. Arsenic occurs in the water by dissolving minerals and ores. Serious health effects have been reported in populations that consume drinking water with high level of arsenic in countries around the world. Long-term exposure to inorganic arsenic, mainly through drinking water and food, can lead to chronic arsenic poisoning. Arsenic appears in water by dissolving minerals and ores. Serious health effects have been reported in people who drank arsenic-rich water for a long time in countries around the world. Acute effect: Immediate symptoms of acute arsenic poisoning include vomiting, abdominal pain, and diarrhea. These are accompanied by numbness and tingling in the extremities, muscle cramps and death in extreme cases. Long-term effects: The first symptoms of long-term exposure to high levels of inorganic arsenic (for

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example, through drinking water and food) are usually seen in the skin, and include changes in pigmentation, skin lesions, and hard patches on the forearms and feet (hyperkeratosis)<sup>1</sup>. Natural contamination of water bodies with heavy metals may be apparent because of the highly rich mineral deposits and the high rainfall available to facilitate weathering, dissolution, transmission and sedimentation of heavy metals in the sediment. Natural and anthropogenic factors may play a role in pollution processes but evidence indicates that natural factors may play an equally important role<sup>2</sup>. Its transition into the drinking water is possible when the rain water passes through the minerals containing the element. In the environment several arsenic compounds exist, formed either by its three- or five-valence ion<sup>3</sup>. Having in mind profound deleterious effects of arsenic on human health, different social, economic and technological measures are required in order to reduce arsenic concentration to acceptable limits<sup>4</sup>. The average concentration of arsenic in drinking water from the city water supply networks in the Rulebook on water safety, Official Gazette of the Republic of Macedonia No 46/08 when an MAC (Maximum allowed concentration) of 0.01 µg/l arsenic in drinking water has been established<sup>5,6</sup>.

The aim of paper was to assess the risk through analyses and evaluation of level of arsenic in drinking water, which is important for prevention of arsenic-related disease by providing safe drinking water from new wells from other side of the river Vardar.

## EXPERIMENTAL

With the preparation of the Republic computer program (1996), for issuing laboratory findings (RCP), from performed analyses of drinking water samples (physicochemical and bacteriological), according to the methodology for application of identification numbers in the computer system at the Institute of Public Health is created a basis for complete health records of drinking water supply facilities that are under health supervision – for the territory of the entire Republic. The RCP covers samples in scope for the following types of analyses: basic, periodic and study-research works. The RCP is supplemented with a section that covers surface waters that are of health interest – drinking water, bathing water, sports and recreation water, as well as water that can be used in food production and trade.

Case study was performed to assess level of arsenic in drinking water taken from the following measuring points Well No 1-Moin; Well No 2-Moin; Well No 3-Moin; Well No 4-Moin; Restaurant Seljakot-Moin way-Gevgelija; Administration JPKD Komunalec-Gevgelija; Rail way buffet “Feroturist” Gevgelija; Well “Vardar” – Gevgelija; Well “MOIN”; Primary School “Krste Misirkov”; Kinder garden “Sončogledi”, drinking water tank; m.p. Moin pump 1; m.p. Moin pump 2; m.p. Moin pump 3; m.p. Moin pump 4; v. Novo konjsko; Water supply network

(drinking water tank) “Stopanstvo”; Primary School Vlado Kantardžiev; Primary School “Josif Josifovski”, in the period from 2011 to 2019. Arsenic in drinking water was analysed by using (FAAS) – Perkin Elmer Atomic Absorption Spectrometer 3110.

## RESULTS

Under the slopes of Kozuv, the groundwater carry with them all the way to the source of water supply, inorganic arsenic that originates from the rocks themselves. One of the most important steps in the affected communities, such as Gevgelija, is to prevent further exposure. In order to monitor the state of the arsenic content in the drinking water from the city water supply system, a series of drinking water sampling was performed.

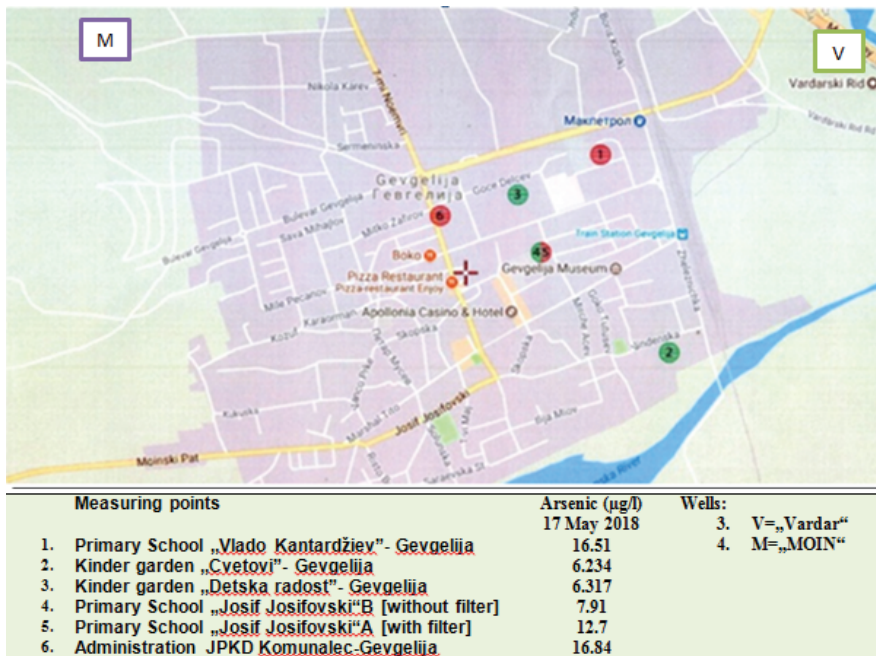


Fig. 1. Map of the water supply system with drinking water of Gevgelija with a result of arsenic content with several measuring points

It was noticed that in the samples of drinking water from the well area “Moin” the concentrations of arsenic were increased above the MPC of 10 µg/l, while the samples from the well area “Vardar” were within the allowed limits in the period from 2009 to 2019.

The water from the wells near Moin in Gevgelija has been banned for drinking and cooking since 2009, precisely because of the increased level of arsenic in the

water. The Institute of Public Health submitted an opinion to the Food Directorate of the Ministry of Health on taking urgent measures to prevent the possible risk of arsenic through drinking water for the health of the population of the city of Gevgelija and surrounding areas (about 20 000 inhabitants).

In Table 1 it can be seen that arsenic in drinking water from the wells “Moin” started to rise-up since 2011 with the highest value in water from the wells 3 and 4 (22.40 µg/l) that are closest to the mountain Kozuf, and lowest (average 10.85 µg/l) from the well No 1. The arsenic level has been increasing almost each year from the period of investigation 2011–2019. Almost all results of drinking water from the wells “Moin” were above the MAC (Maximum admissible concentration).

**Table 1.** Water supply system with drinking water of Gevgelija with a result of arsenic content (µg/l) from several measuring points (2011–2019)

| Measuring points                               | 2011  | 2012  | 2013  | 2014 | 2015  | 2016 | 2017 | 2018  | 2019 | Average |
|--|-------|-------|-------|------|-------|------|------|-------|------|---------|
| 1  | 2     | 3     | 4     | 5    | 6     | 7    | 8    | 9     | 10   | 11      |
| Well No 1 – Moin                               | 9.00  | 12.70 |       |      |       |      |      |       |      | 10.85   |
| Well No 2 – Moin                               | 20.30 | 11.90 |       |      |       |      |      |       |      | 16.10   |
| Well No 3 – Moin                               | 22.40 | 12.60 |       |      |       |      |      |       |      | 17.50   |
| Well No 4 – Moin                               | 17.80 | 14.83 |       |      |       |      |      |       |      | 16.32   |
| Restaurant Seljakot-Moin way-Gevgelija         | 20.40 | 15.70 |       |      |       |      |      |       |      | 18.05   |
| Administration JPKD Komunalec-Gevgelija        | 18.00 | 15.30 |       |      |       |      |      | 16.87 |      | 16.72   |
| Rail way Station-buffet “Feroturist” Gevgelija | 20.40 |       |       |      |       |      |      |       |      | 20.40   |
| Well “Vardar”-Gevgelija                        | 2.77  |       |       |      |       |      |      |       |      | 2.77    |
| Well “MOIN”                                    | 13.21 | 13.40 |       |      |       |      |      |       |      | 13.31   |
| Primary School “Krstе Misirkov”                |       | 11.70 |       |      | 18.45 |      |      |       |      | 15.08   |
| Kinder garden “Sončogledi”                     |       | 12.33 | 17.40 |      | 17.30 |      |      |       |      | 15.68   |

to be continued

Continuation of Table 1

| 1   | 2 | 3 | 4     | 5     | 6     | 7     | 8     | 9     | 10 | 11    |
|---|---|---|-------|-------|-------|-------|-------|-------|----|-------|
| Drinking water tank                                     |   |   | 20.20 | 20.31 |       | 22.00 | 22.00 |       |    | 21.13 |
| m.p. Moin pump 1  |   |   | 11.73 |       |       |       |       |       |    | 11.73 |
| m.p. Moin pump 2  |   |   | 15.00 |       |       |       |       |       |    | 15.00 |
| m.p. Moin pump 3  |   |   | 14.70 |       |       |       |       |       |    | 14.70 |
| m.p. Moin pump 4  |   |   | 14.49 |       |       |       |       |       |    | 14.49 |
| v. Novo konjsko   |   |   |       |       | 11.34 |       |       |       |    | 11.34 |
| Water supply network (drinking water tank) "Stopanstvo" |   |   |       |       |       | 21.45 |       | 20.14 |    | 20.80 |
| Primary school "Vlado Kantardžiev"                      |   |   |       |       |       |       | 10.22 | 16.51 |    | 13.37 |
| Primary school "Josif Josifovski"                       |   |   |       |       |       |       |       | 12.70 |    | 12.70 |
| Average   |   |   |       |       |       |       |       |       |    | 14.90 |
| MAC = 10 µg/l   |   |   |       |       |       |       |       |       |    |       |

MAC – Maximum admissible concentration.

From Table 1 we can see that the average value in the period from 2011–2019 exceeds (14.90 µg/l) the maximum allowed value of 10 µg/l. Provisional tolerable weekly intake (mg/kg body weight) for arsenic through contaminated food/water is 0.015.

In Table 2 are shown rates of malignant neoplasm of the skin, bladder and lungs. It can be seen that the rate per 100 000 inhabitants are increasing in Gevgelija and Republic of North Macedonia from 2011–2019. The same is happening with the rate of malignant neoplasm of the bladder, with exception in Gevgelija and a little bit in North Macedonia. But for the rate of malignant neoplasm of lungs is quite opposite. Regarding the fact that water from the wells "Moin" has been prohibited for drinking since 2009, rates of malignant neoplasm of the skin, bladder and lungs can not be related to the provisional daily intake of arsenic from drinking water. Gevgelija's inhabitants have been drinking bottled water since 2009.

**Table 2.** Rate of newly registered cases of malignant neoplasm of the skin (C44), bladder (C67) and lungs (C34) in the Republic of North Macedonia, period 2011–2019

| Years | Malignant neoplasm of the skin (C44)     |                                    | Malignant neoplasm of the bladder (C67)  |                                    | Malignant neoplasm of the lungs (C34)    |                                    |
|-------|--|------------------------------------|--|------------------------------------|--|------------------------------------|
|       | rate per 100 000 inhabitants – Gevgelija | rate per 100 000 inhabitants – RNM | rate per 100 000 inhabitants – Gevgelija | rate per 100 000 inhabitants – RNM | rate per 100 000 inhabitants – Gevgelija | rate per 100 000 inhabitants – RNM |
| 2011  | 2.9                                      | 12.7                               | 8.6                                      | 11.9                               | 72.0                                     | 637.0                              |
| 2012  | 2.8                                      | 10.9                               | 8.7                                      | 7.2                                | 55.0                                     | 43.0                               |
| 2013  | 20.2                                     | 22.7                               | 5.8                                      | 9.7                                | 34.7                                     | 37.1                               |
| 2014  | 20.3                                     | 27.4                               | 14.5                                     | 16.0                               | 58.1                                     | 46.1                               |
| 2015  | 14.6                                     | 15.4                               | 23.3                                     | 16.8                               | 78.6                                     | 44.2                               |
| 2016  | 14.6                                     | 24.6                               | 11.7                                     | 16.0                               | 55.4                                     | 38.3                               |
| 2017  | 35.0                                     | 35.4                               | 17.5                                     | 14.1                               | 81.7                                     | 44.2                               |
| 2018  | 26.4                                     | 35.3                               | 17.6                                     | 14.8                               | 64.6                                     | 27.2                               |
| 2019  | 50.1                                     | 45.8                               | 8.8                                      | 13.3                               | 41.2                                     | 38.5                               |

Source: Institute of Public Health of the Republic of North Macedonia.

## NEW WELLS

In order to provide the required quantities of water of 200 l/s with safe drinking water (with no arsenic in the water) based on the performed research works and the results obtained from the testing of IEB-1, as well as data processing and mathematical modelling, it was recommended to perform a total of four wells, three more exploitation wells, with the IEB-1 well being used as an exploitation well in the future. New wells (EB-2, EB-3 and EB-4) to be constructed to a depth of 35 m. The locations of the wells are arranged linearly and parallel to the river Vardar, at a mutual distance of  $L = 60$  and 100 m.

The three wells (IEB-1, EB-2 and EB-3) are located within the plot determined by geodetic experts of the municipality of Gevgelija, as land of the Republic of North Macedonia, and the fourth well (EB-4) is determined in a private plot, 100 m north of the planned well EB-3. The mutual influence, radius of influence, which the wells will have during joint work, as well as their work in conditions of hydrological minimum, low water level of the river Vardar, has been taken into account.

Expected yield of each constructed well, when working together in a system of four exploitation wells, will be 50 l/s, in total for the entire well system the expected-projected yield will be 200 l/s, that is quite suitable for water supplying of Gevgelija.

The hydraulic calculation given below determines the technical and hydraulic characteristics of submersible pumps (Figs 2a, b). The pumps are located at a depth of 20.00 m from the field in accordance with the recommendations of the performed

hydrogeological investigations. The pumps are adopted for a projected flow of  $Q = 50 \text{ l/s}$ , and a calculated manometer height of  $H = 110 \text{ m}$ . A submersible well pump with a motor power of  $75 \text{ kW}$  has been selected for these characteristics.



Fig. 2a. Location of new wells



Fig. 3b. Location of new wells

### WELL HOUSE

Above each well it is planned to build a well house with dimensions of basically  $4.00 \times 5.50 \text{ m}$ , in which it is planned to place all the hydraulic equipment as well as the cabinets of the pumps. The floor of the building is planned to be placed at an altitude of  $50.00 \text{ m}$ , due to the protection of the building from high waters of



the river Vardar, which for this area are at an altitude of 50 m. The architectural-construction project is completely processed.

In the building from the wellhead, which is made of steel pipe F600 mm, with flange and welded FF piece F200/600, the pressure pipeline starts, with diameter F200 mm. On the pressure pipeline in the building are placed air valve F100 mm, non-return valve F200 mm, electromagnetic flow meter F200 mm, shock absorber F100 mm, for proper operation of the system. The equipment in the house is designed to be at a working pressure of 20 bar.

## DISCUSSION

Consumption of arsenic greater than its permissible limit through drinking water and food is fatal for human health. Chronic exposure to arsenic can cause harm to the human cardiovascular, dermal, gastrointestinal, hepatic, neurological, pulmonary, renal and respiratory systems and reproductive system. The exposure of human to arsenic contaminated water and food can lead to some physical changes on the skin such as the appearance of small black or white marks (melanosis), then thickening of the skin on the palms and the feet (keratosis), followed by skin lesions and eventually skin cancer. The long-term ingestion or exposure (10–15 years) of arsenic can lead to a disease called “arseniasis, arsenicosis, and arsenicism”. Non-cancer endpoints include hyper-pigmentation, hypo-pigmentation, keratosis of skin, peripheral vascular disease (black foot disease), cardiovascular disease, hypertension and neurotoxicity. Most of the symptoms of above-mentioned diseases were observed in the field during the study. Arsenic exposure from contaminated drinking water of more than 50 µg/l is a significant cancer risk. The development of internal cancer in humans may take 10 years to develop and is often the result of long term exposure to arsenic. Chronic exposure to arsenic has been linked to carcinogenic effects in both humans and animals. These include cancer of the various skin and various internal organs (lung, bladder, liver and kidney) reproductive and developmental effects; cardiovascular disease; reduced intellectual function in children and mortality. There are some claims that chronic exposure of arsenic may also cause diabetes development and prostate cancer<sup>7</sup>. Exposure to arsenic is one of the major global health problems, affecting > 300 million people worldwide, but arsenic’s effects on human reproduction are uncertain<sup>8</sup>.

Ecological studies have indicated associations between in utero and/or early life exposure to arsenic at high levels and increases in mortality from cancer, cardiovascular disease and respiratory disease. Additional data from epidemiologic studies suggest intermediate effects in early life that are related to risk of these and other outcomes in adulthood. Experimental animal studies largely support studies in humans, with strong evidence of transplacental carcinogenesis, atherosclerosis and respiratory disease, as well as insight into potential underlying mechanisms of

arsenic's health effects<sup>9</sup>. The migration of water soluble forms of arsenic (and other micro elements) dominantly depends on the condition of the soil solution. There is permanent translocation of the solution along the soil profile, which depends on the soil properties and external factors such as: gravity, capillary phenomenon in soil, hydrological conditions, concentration gradient, osmotic pressure, etc.<sup>10</sup> In sample of drinking water taken from the well "Moin" Gevgelija in 2000 has been found arsenic at the level of the MAC (Ref. 11). Isolated pockets in 86 districts in ten States of India in 2003–2004 have been reported affected by groundwater arsenic content beyond maximum permissible limit of 50 ppb (Ref. 12). Though arsenic occurrence in groundwater is subject to considerable uncertainty, UNICEF and EAWAG have developed maps to assist in predicting the risk of arsenic contamination that are freely available on the Groundwater Assessment Platform website ([www.gapmaps.net](http://www.gapmaps.net)). Estimates from 2009 suggest that arsenic contamination could affect more than 140 million people globally<sup>13</sup>. Consumption of arsenic greater than its permissible limit through drinking water and foods is fatal for human health. Chronic exposure to arsenic can cause harm to the human cardiovascular, dermal, gastrointestinal, hepatic, neurological, pulmonary, renal and respiratory systems and reproductive system. Non-cancer endpoints include hyper-pigmentation, hypo-pigmentation, keratosis of skin, peripheral vascular disease (black foot disease), cardiovascular disease, hypertension and neurotoxicity<sup>14</sup>.

## CONCLUSIONS

Long-term actions are required to reduce exposure to arsenic from mining, metal smelting and refining, combustion of low-grade coal, pesticide use and timber treatment. In particular, action is needed to reduce the intake of arsenic from drinking-water and food in areas with naturally high levels in the groundwater.

Lab testing would historically yield more reliable results than field tests. Wherever arsenic removal technologies are applied, operational monitoring should be carried out regularly to confirm that systems are functioning as expected. Water-quality surveillance should then be carried out to confirm that systems are delivering water to expected quality standards.

Despite the higher level of arsenic in drinking water in Gevgelija in the period of investigation, due to banning of underground water from the wells "Moin" for drinking since 2009, rates of malignant neoplasm of the skin, bladder and lungs cannot be related to the provisional daily intake of arsenic from drinking water. Gevgelija's inhabitants have been drinking bottled water since 2009.

For the needs of the citizens, new wells were made in 2018, which in a series of series of tests at the Institute in 2018–2019 were registered results below the MAC in terms of the level of arsenic in drinking water. The most important public health measure is the prevention of further exposure by providing safe drinking water.

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