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Original scientific paper

Concentration of heavy metals in spring waters of Kochani and their impact on human health

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Abstract

The concentration of lead (Pb), cadmium (Cd), aluminium (Al), manganese (Mn), and iron (Fe) in spring waters in the vicinity of Kochani was investigated to ascertain the health risk exposure. Heavy metal water contamination is possible mostly due to wrongful human activities either industrially or residentially and exposure to toxic doses of heavy metals can cause acute and chronic diseases.

To examine heavy metal contamination of spring waters as well as possible seasonal variations, water samples in two seasons, autumn 2021 and spring 2022 were collected. For this aim, ten locations within the area of Kochani were selected. The spring water samples were collected directly from the source or from the catchment that was made. The samples were collected in appropriate bottles, previously washed with detergent and rinsed with distilled water. The examination of heavy metals was carried out immediately after sampling using UV spectrophotometric method, with an absorption spectrum of 190-1100 nm. The results have revealed no major deviations from the maximum concentrations allowed for Pb, Cd, Al, Mn, and Fe defined within the Official Guidelines in the Republic of North Macedonia, except moderately increased levels of lead in two samples during autumn season and manganese in four samples in the spring season 2022.

Key words: heavy metal levels, spring water, UV spectrophotometry, human health

Introduction

Drinking water is important resource which influences human life and the source of good quality portable water is considered as a priority for public health. The most common sources of drinking water are natural surface water (lakes, ponds, rivers, streams etc.) and ground water (bore holes and well water). Besides the vital role in biochemical processes in the human body, water is important for domestic and industrial supply, as well as for irrigation. Population growth, rapid industrialization and urbanization are recognized as major source for water contamination. The contamination of ground water is not easy to restore. Hence, it is necessary to protect quality of ground water (Tadiboyinaa and Ptsrkb, 2016).

Heavy metals are elements with high density or an actual volume of more than 6 g/m^3 (Tavakoli-Hosseinabady et al., 2018) or specific density of about 4-5 times as much as that of water (Duruibe et al., 2007). They are found naturally in the earth crust, but due to different human activities their geochemical and biochemical balance has been drastically changed and they can easily reach the drinking water supply. Many spring waters are passing near the areas of agriculture; mining and industrial smelting activities as well as fossil fuel burning plants, municipality waste and sewage, all considered being important sources of metal pollution. Heavy metals in

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water can exist in colloidal, particulate and dissolved phases with their occurrence in water bodies being either of natural origin (e.g. eroded minerals within sediments, leaching of ore deposits and volcanism extruded products) or of anthropogenic origin (i.e. solid waste disposal, industrial or domestic effluents). Heavy metals cannot be easily degraded or decomposed and they have ability of bioaccumulation once get into the ecosystem via drinkable water impairing the quality of water and subsequently human health (Baby et al., 2010; Meng et al., 2021). There are some biologically essential heavy metals which human body requires in trace amounts such as iron, cobalt, copper, zinc, molybdenum, and manganese. These entities are needed at low levels as catalyst for enzyme activities (Udeagbara et al., 2019), but when present in excessive amount, are detrimental to human health. Other heavy metals such as lead, mercury, arsenic, and cadmium can be found in drinking water too, but have no beneficial effects for the human body. They are of great concern as they can harm different organs even at low exposure levels and their accumulation into the body can cause severe health problems. Heavy metals in water can be found as ions in the form of easily diluted compounds able to adhere to enzymes and inhibit their function with subsequent physiological consequences. Exactly, how risky a metal can depend on several factors, including the dose and means of exposure, as well as the temperature and pH value affecting the release of the metal ions from the medium (Marković et al., 2020).

Heavy metal concentrations in drinking water exceeding the permissible limits set by national and international organizations are related to acute and chronic diseases ranging from nonfatal, such as muscle and physical weakness or teeth decay, to fatal, such as neurogical diseases, adverse reproductive outcomes, cardiovascular diseases and even cancer (Saghali et al., 2014; Singh et al., 2022). In addition, heavy metal water pollution is a global problem due to the nature of heavy metals being non-biodegradable with long biological half-lives resulting in accumulation inside the human body. Thus, for the protection of human health and environmental water quality, constant monitoring and control of heavy metal levels is of prime importance. Considering the essential role of water to be an indispensable part of the human life and environment, safe and good quality drinking water is the basis for good health.

In this study, five heavy metals (lead, cadmium, iron, aluminium and manganese) in water samples were analyzed. The examination was carried out in two seasons, autumn and spring, in order to determine the content of the selected heavy metals in samples of natural spring waters taken at 10 different measuring sites, as well as to determine potential variations in the concentration of heavy metals with the change of weather conditions or other influencing factors.

Material and methods

Study area and sampling

A total of ten water samples from different locations within the area of Kochani (Table 1), in autumn 2021 and then in spring 2022 were collected. This city is located in the centre of the east region in the Republic of North Macedonia and it is the major large-scale manufacturer of rice in the country. Besides agricultural processing, other manufacturing industries such as textile, construction industry, paper processing plant, are active and in addition mining activities are located at 40 km northeast of Kochani. The sampling plan was to determine whether different factors in some way contribute to the rise of lead, iron, aluminium, manganese, and cadmium concentration in water. Seasonal sampling lasted for over half a year starting from October 2021. The spring water samples were collected directly from the source or from the catchment that was made after flushing water for 10-15 min in order to remove the stagnant water. For sample collection appropriate tight-capped high-quality polyethylene dark colored glass bottles, previously washed with detergent and rinsed with distilled water were used. Each sample was properly marked at the time of the collection. The samples were transported to the laboratory under cold conditions and all analyses were performed without delay.

Chemicals and measurement

Lead, cadmium, iron, aluminium, and manganese concentration was determined in the ten water samples examined, following the manufacturer’s protocol. All test kits used were supplied from the manufacturer Merck KgaA, Germany. According to the specification of the test kits for lead and manganese analysis, lowest level of 0.010 mg/L can be measured for both metals to the levels of 5.000 mg/L lead and 10.000 mg/L manganese. According to the specification of aluminium test kit, levels in a range of 0.020 to 1.200 mg/L can be determined. The cadmium test kit has a measurement range of 0.002-0.500 mg/L, while the iron test kit specifies the concentration range of 0.0025 to 5.000 mg/L. Stock standard solution at a concentration of 10 mg/L for the investigated heavy metal was supplied from Merck KgaA, Germany, separately of the respective kit intended for analysis of lead, cadmium, iron, aluminium and manganese.

Ultraviolet-visible spectrophotometer, a Spectroquant Prove 300 (Merck KgaA, Germany), with an absorption spectrum of 190-1100 nm was used to analyze the presence and concentration of selected heavy metals in this work. Appropriate cuvettes with a size of 20 mm and 50 mm were used for the measurements. Apart from the Specrtoquant Prove 300, a pH Meter pH 1100L (VWR Phenomenal, Germany) was also used as an additional equipment, since the pH of the sample needed to be in the range of 3-6 for the lead test. The pH was adjusted by adding nitric acid.

Table 1. Measuring sites of samples collected in the region of Kochani.

Sample	*Measuring point*
1	*Usova`s fountain*
2	*Udarnichka`s fountain*
3	*Fountain – belski dol*
4	*Fountain village Leshki*
5	*Vojnichko kladenche*
6	*Marenska fountain*
7	*Gorna fountain*
8	*Remiro fountain*
9	*Fountain near the church*
10	*Fountain near the mosque*

All measurements were done in triplicate and the average value was calculated. The absorbance was measured at a wavelength according to the test instructions against a corresponding reagent blank. The heavy metal concentration in a tested water sample was determined using concurrently prepared calibration graph. The obtained values for the concentration of each heavy metal per sample were expressed in mg/L. Least significant difference test was applied for means comparisons with the level of significance at $p<0.05$.

Results and discussion

Significant part of the population in the area of Kochani uses water from existing spring waters for drinking and other purposes. The adopted waste management strategies to keep pace with the industrial growth and urbanization often fail, especially in low income countries. Unsuitable handling of industrial waste as well as use of fertilizers and pesticides can be considered as potential source of heavy metal contamination to the environment. In fact, although most of the heavy metals present in different types of water samples are derived from natural weathering of the earth's surface, effluents from urban and industrial sewage and wastewater used for irrigation could migrate and primarily get at groundwater impairing the water quality (Assubaie, 2015).

The determination of heavy metal content (iron, lead, aluminium, cadmium, and manganese) in water samples can be performed by various sophisticated techniques such as AAS, ICP-AES, and ICP-MS. Despite their sensitivity, they require expensive instrumentation and may represent financial burden for laboratories. Therefore, the UV spectrophotometric method for determination of heavy metals can be suitable alternative. The low cost of the instrument, easy handling, and speed of the analyses has proven UV spectrophotometry a technique of choice, particularly in laboratories of developing countries with limited budgets (Ullah and Haque, 2010). The sensitivity of determination is as low as few µg/L and relatively small standard deviation (Fig. 1 and Fig. 2) indicated that the applied method is reliable to measure the concentration of the studied metals in drinking water samples. In addition, this method is routinely applicable providing measurement of any of the five heavy metals at a level that is significantly lower than the maximum permitted level given in the Official Guidelines for drinking water of 2018 (Official Gazette of Republic of Macedonia № 183, 2018). The results obtained from the measurements in both seasons (autumn 2021 and spring 2022) are presented in Fig. 1 and Fig. 2, respectively. The investigated heavy metals (iron, lead, aluminium, cadmium, and manganese) were detected in all water samples analyzed in both seasons. The determined concentrations were generally lower than the maximum allowed concentrations according to the Official Guidelines for drinking water of 2018 (Official Gazette of Republic of Macedonia № 183, 2018). In all samples, heavy metal concentrations during spring were found higher compared to the fall season, however no significant deviations when compared to the maximum allowed concentrations defined with the national legal framework. These changes are most likely due to the change in weather conditions.

Lead

Lead is a toxic metal which occurs naturally in the environment, but its concentration is increased due to human activities. Lead easily enters into environment through the exhaust of vehicles, and it reaches drinking water through leaching from lead containing pipes, facets

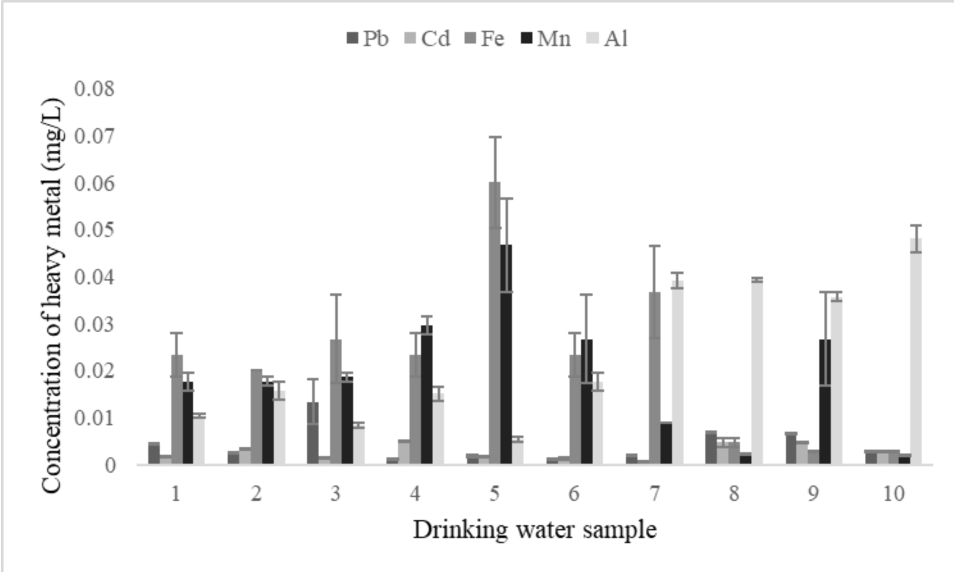

Fig. 1. Mean concentration (mg/L) of five heavy metals (lead, cadmium, iron, manganese, and aluminium) in ten locations of Kochani region during autumn 2021 season.

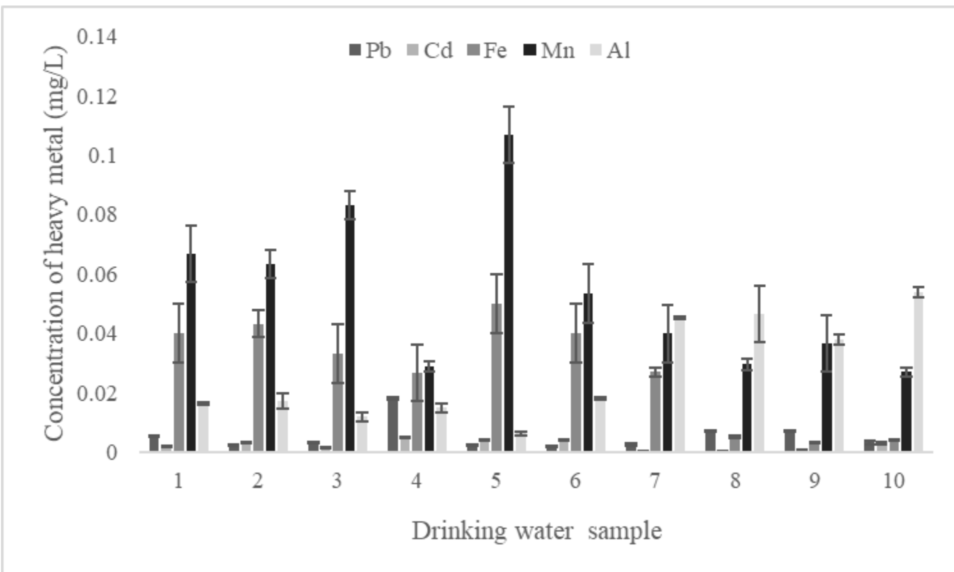

Fig. 2. Mean concentration (mg/L) of five heavy metals (lead, cadmium, iron, manganese, and aluminium) in ten locations of Kochani region during spring 2022 season.

or when plumbing materials that contain lead corrode. Small amounts of lead in water were reported to show adverse effects on the functioning of the nervous system as well as hypertension, impaired thyroid function and preterm birth associated with various neurodevelopmental problems (Caussy et al., 2003; Payne, 2008). Children below six-year-old are at the highest risk (Tadiboyina and Ptsrkb, 2016).

In our study, in nine of total ten examined water samples in both seasons, lead was found to be below the maximum allowed concentration in drinking water according to the international and national legislation. Permissible level for lead in drinking water of 10 µg/L was revised in 2011 by the European Commision (SCHER, 2011), but remained at the same value such as this pollutant is regulated in our legislation (Official Gazette of Republic of Macedonia № 183, 2018). In the autumn season, Sample 3 has shown slight increase over a borderline concentration of lead (13.33 µg/L, Fig. 1), while in the spring season significantly lower concentration for more than three times was observed (3.27 μg/L, Fig. 2). Such a difference between the seasons may due to some transient pollution. However, this water was not suitable for consumption considering that low levels of lead are toxic. In this case, people should be warned. An increase in lead content in the spring season was registered in Sample 4 only (18.33 μg/L, Fig. 2, while the lead level in this sample was within the acceptable concentration range in the last autumn (1.13 μg/L, Fig. 1). Considering that US Environmental Protection Agency (US EPA, 2022) has set the maximum allowable concentration of lead in public drinking water at 15 μg/L (although the maximum contaminant level goal is zero), it can be concluded that the waters at the locations 3 and 4 (Table 1) have acceptable safety. On the other hand, according to the European legislation, maximum level of lead in drinking water should be ultimately reduced to a 10 μg/L as recommended by the World Health Organization (WHO Guidelines for Drinking Water Quality, 2017). Lead concentrations can vary according to the period in which the water has been in contact with the lead-containing materials, and hence special attention in monitoring of lead concentrations in Samples 3 and 4 is required.

Cadmium

Cadmium is commonly a byproduct in the process of refining other metals. This metal can enter the groundwater through the effluent of zinc, carbonate and sulfide ore, urban sewage (mainly phosphate detergents that contain cadmium as an impurity) or in the form of phosphate fertilizers that are usually used in certain areas, battery, galvanizing and other industrial effluents (Assubaie, 2015; Drozdova et al., 2018; Tadiboyina and Ptsrkb, 2016). Due to its high accumulation capacity, even low exposure level can cause learning disabilities and hyperactivity in children (Hunt, 2003).

Cadmium levels determined in all samples from both seasons were within the maximum permissible limits defined by the Official national guidelines of 5.0 μg/L (Official Gazette of Republic of Macedonia № 183, 2018). According to the World Health Organization, the maximum permissible level of cadmium is 3.0 μg/L (WHO Guidelines for Drinking Water Quality, 2017). According to the WHO Guidelines, cadmium in drinking water is usually found at concentration levels less than 1.0 μg/L. Herein, cadmium levels less than 1.0 μg/L were determined in only one of the total ten water samples analyzed in autumn (Sample 7, 0.47 μg/L) and in three samples analyzed in spring (0.53 μg/L in Sample 7 and 0.50 μg/L in both Sample 8 and 9). Slight variations were observed in Samples 5 and 6 that showed increased cadmium level in spring 2022 compared to autumn 2021. On the contrary, the level measured in Sample 8 was significantly higher in autumn than in spring the following year. Such a variation is rather due to the occurrence of cadmium as a contaminant of the soil than naturally present in the environment. It was already reported that it is possible storm water to drain and runoff from nearby roads and agricultural farms entering sewage plants thus increases the levels of Cd in wet seasons (Agoro et al., 2020).

Iron

Iron is naturally occurring metal in the form of magnetite, hematite etc. that enter into water through a series of extraction processes of “iron ore”. Another important source of contamination is aluminium waste products which contain iron and are often discharged into water. Although essential for the normal function of hemoglobin, high concentrations can cause tissue damage (Bahiru, 2020). The presence of excess iron in drinking water imparts a strong metallic taste and can cause staining (Ramesh and Damodhram, 2016). However, no guideline value for iron in drinking water was proposed by the WHO claiming that iron is not of health concern at levels found in drinking water (WHO Guidelines for Drinking Water Quality, 2017). In our study, variable iron levels in autumn 2021 and spring 2022 comprising the respective samples were observed, but the measured concentrations were much less than the maximum permissible limits according to the national guidelines (0.2 mg/L) (Official Gazette of Republic of Macedonia № 183, 2018). The results showed that the Fe concentrations in all examined samples varied from 0.0027 to 0.0600 mg/L in autumn 2021, and almost the same concentration range was evidenced in spring 2022 (0.0033-0.0500 mg/L). Lowest iron levels were found in Samples 8, 9 and 10 comprising both measuring seasons (Fig. 1 and 2).

Manganese

Manganese, together with zinc and copper, belongs to the group of heavy metals that have important physiological functions in living tissue and regulate many biochemical processes at trace levels (Assubaie, 2015). On the other hand, manganese is toxic to a number of crops usually in acid soils and in the form of suspended particulates from industrial emissions, soil erosion, volcanic emissions and the burning human activities may be responsible for water contamination (Tadiboyina and Ptsrkb, 2016). As well as iron, elevated levels of manganese may give water an unpleasant appearance and taste.

Manganese concentrations in all ten samples in the autumn season were within the maximum permissible limit of 50 μg/L according to the national guidelines (Official Gazette of Republic of Macedonia № 183, 2018) (Fig. 1), while during the spring 2022 significantly higher concentrations of manganese than the allowed (50 μg/L) were observed in Samples 1, 2, 3 and 5 (Fig. 2). In addition, except Sample 4, all other investigated waters showed increased manganese concentrations in spring 2022 than in the autumn previous year. The levels increased during the spring probably due to the melting of the snow or rainy season rather than soil pollution since manganese is one of the most abundant metals in earth crust and easy can enter into atmosphere. In addition, WHO had not set a maximum permissible level for manganese in drinking water since at levels normally causing acceptability problems in drinking water it is not of health concern. Thus, only a health-based value of 0.4 mg/L was defined that is based on the upper range value of manganese intake of 11 mg/day, identified using dietary surveys, at which no adverse effects were observed (WHO Guidelines for Drinking Water Quality, 2017).

Aluminium

Aluminium is the third most abundant element in Earth's crust. Water turbidity from waste water effluents and solid waste of industries and human activities as well as muddy soil in nature may increase aluminium content of the environment. When aluminium accumulates acutely in tissues such as the brain, it has the potential to cause serious neurological effects (Tareen et al., 2014). However, WHO declared that orally ingested aluminium is rarely indicated to induce acute toxicity to humans despite the widespread occurrence in foods, drinking water and many antacid preparations, thus no guideline value is suggested (WHO Guidelines for Drinking Water Quality, 2017).

In our study, no exceeding levels of aluminium in all samples (significantly below than the guideline value of 200 µg/L), comprising both seasons, were detected (Official Gazette of Republic of Macedonia № 183, 2018). The concentration of aluminium was found to show only slight variations among samples from different measurement points in the same season (ranging from 0.005 to 0.048 mg/L in autumn and 0.006 to 0.054 mg/L in spring) as well as between the respective samples taken from the same location in different seasons. This finding indicated that, in the investigated samples, aluminium levels were stable.

Heavy metals and human health

It is very important to identify the relationship between the presence of heavy metals in drinking water and the prevalence of renal failure, liver cirrhosis, hair loss, and chronic anemia diseases. The prevalence of these diseases was markedly increased in the last few years due to air pollution, water pollution, and hazards over uses of pesticides in agriculture. Trace amounts of metals are common in water, and these are normally not harmful to our health. In fact, some metals are essential to sustain life. Calcium, magnesium, potassium, and sodium must be present for normal body functions. Cobalt, copper, iron, manganese, molybdenum, selenium, and zinc are needed at low levels as catalysts for enzyme activities. Drinking water containing high levels of these essential metals, or toxic metals such as aluminium, arsenic, barium, cadmium, chromium, lead, mercury, selenium, and silver, may be hazardous to human health. Metals in our water supply may occur naturally or may be the result of contamination. Naturally occurring metals are dissolved in water when it comes into contact with minerals and rocks or soil material. Other important sources of metal contamination are corrosion of pipes and leakage from waste disposal. Heavy metal concentrations in drinking water that exceed the permissible limits set by national and international organizations are related to acute and chronic diseases ranging from nonfatal, such as muscle and physical weakness or teeth decay, to fatal, such as neurogical diseases, adverse reproductive outcomes, cardiovascular diseases and even cancer (Saghali et al., 2014; Singh et al., 2022).

One of the major symptoms of chemical toxicity seems to be a breakdown of the immune system, which opens the gateway for all kinds of diseases in the body. Also, another major symptom seems to be damage to the nervous system and increased nervousness. Toxic doses of chemicals cause either acute or chronic health effects. The levels of heavy metals in drinking water, however, are seldom high enough to cause acute health effects. They are more likely to cause chronic health effects that occur long after exposure to small amounts. The production of reactive oxygen species resulting in oxidative damage and health related adverse effects has been reported to be the main mechanism involved in heavy metal-induced toxicity (Jamshaid et al., 2018; Rehman et al., 2018; Singh et al, 2022).

Apart from the two samples with increased lead levels, all metals in the rest of the samples were considerably below the respective limits permitted by our national legislation and the drinking water standard of the World Health Organization (WHO Guidelines for Drinking Water Quality, 2017). The analyses of the samples taken in spring 2022 also revealed good quality of investigated water samples, with the exception of manganese levels have been measured above the recommended limit in four of total ten samples. This outcome suggests that the analysis of heavy metals is important aspect of environmental monitoring, including drinking water quality.

Conclusion

Water contamination has become a major threat in today's world. Heavy metals that are potentially toxic get distributed to different areas through various pathways gaining upon drinking water.

This study was done in order to determine the levels of five heavy metals (Pb, Cd, Mn, Al, Fe) in spring waters in Kochani and its surroundings, as well as to examine the impact of eventually contaminated water on human health. The analyses were performed using UV spectrophotometry. The study was carried out in two seasons, and the obtained results showed no major deviations from the maximum concentrations allowed by the Official Guidelines in our country. Concentration above the legal limit for lead in two samples analyzed in the autumn season and manganese in four samples analyzed in the spring season 2022 was detected. As a result, continuous monitoring of drinking waters using simple, sensitive, selective and effective alternative method as presented in this study is strongly recommended. In a lack of studies of spring water quality in Kochani region where continuous agricultural and industrial activities occur, the results obtained in this study may serve for evaluation of the potential health risk for people who use water from investigated natural sources. Further, these results provide the evidence for spring water quality that may be useful to map safe water sources around Kochani.

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Резиме

Концентрација на тешки метали во изворските води во околината на Кочани и нивното влијание врз здравјето на луѓето

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Клучни зборови: тешки метали, изворски води, UV-спектрофотометрија, влијание врз здравјето на луѓето

Концентрациите на олово (Pb), кадмиум (Cd), алуминиум (Al), манган (Mn) и железо (Fe) во изворските води во околината на Кочани се испитани со цел да се утврди влијанието врз здравјето на луѓето во овој регион. Нивоата на тешки метали во водата за пиење ретко се доволно високи за да предизвикаат акутни или хронични ефекти врз здравјето на луѓето. Сепак, контаминацијата на водата е можна најмногу поради погрешни хумани или индустриски активности, па доколку е човекот изложен на токсични дози на тешки метали може да дојде до појава на акутна и хронична токсичност.

За да се испита можната контаминација на изворските води со тешки метали, како и сезонските варијации во нивоата на тешки метали, сите анализи се извршени во две сезони, есен 2021 и пролет 2022 година. Избрани се десет локации на подрачјето на Кочани. Примероците од изворските води се собрани директно од изворот или од сливот што беше направен, во соодветни шишиња, претходно измиени со детергент и исплакнати со дестилирана вода. Испитувањето на тешките метали е спроведено веднаш по земањето на примероците со помош на Spectroquant Prove 300 (Merck KGaA Germany), со апсорпционен спектар од 190-1100 nm. Добиените резултатите не покажуваат големи отстапувања од максималните дозволени концентрации за Pb, Cd, Al и Fe дефинирани во Правилникот за безбедност и квалитет на водата за пиење во Република Северна Македонија, освен за олово во два примероци анализирани во есен 2021 и за манган во четири примероци анализирани во пролет 2022.