TEMPORAL VARIABILITY OF SELECTED HEAVY METALS´ LEVELS IN WELL WATER SAMPLES IN NORTHWEST SLOVAKIA

ČASOVÁ VARIABILITA HLADÍN VYBRANÝCH ŤAŽKÝCH KOVOV VO VZORKÁCH STUDNIČNEJ VODY NA SEVEROZÁPADNOM SLOVENSKU

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ABSTRACT

Introduction: Although most of the Slovak population employs public water supplies, there are rural areas with dominant use of individual wells. Heavy metals are quite common contaminants of well water and can potentially affect human health.
Aim: he study analyses levels of manganese (Mn), copper (Cu) and lead (Pb) and studies possible temporal variability of these elements in well water one year apart.
Sample and methods: The study was realized in Korňa village and included 15 wells. Water samples were collected in a period of two subsequent years in May 2018 and May 2019. The levels of Mn, Cu and Pb were determined by atomic absorption spectroscopy with graphite furmance GF AAS. The measured results were complemented by self-administered questionnaire data on well maintenance, water use and its quality monitoring.
Results: Median Mn level in 2018 was 8.9 μg/l. Parametric value for manganese was exceeded in 3 samples. In 2019 median Mn level was 11.0 μg/l. Parametric value (PV) was exceeded in 2 samples. Median copper level in 2018 was 9.6 μg/l and in 2019 13.3 μg/l. In 2019 PV for Cu was exceeded in one sample. Median Pb level in 2018 was 4.8 μg/l and one sample exceeded PV. In 2019, median Pb level was 4.1 μg/l. Effect of weather patterns on heavy metals' levels was insignificant.
Conclusions: Heavy metal contamination of well water presents a potential thread of human health. Proper and regular well stewardship play an important role in prevention of potential health impact.

Key words: Heavy metals. Water contamination. Well water.

ABSTRAKT

Úvod: Hoci väčšina slovenskej populácie využíva verejné zdroje vody, existujú vidiecke oblasti, kde sa väčšinovo používajú individuálne studne. Ťažké kovy sú častými kontaminantmi studničnej vody a predstavujú potenciálne ohrozenie zdravia ľudí.
Cieľ: Štúdia analyzuje hladiny mangánu (Mn), medi (Cu) a olova (Pb) a sleduje ich možnú časovú variabilitu v studničnej vode v priebehu jedného roka.
Vzorky a metódy: Štúdia sa realizovala v obci Korňa a zahŕňala 15 studní. Vzorky vody sa zhromažďovali v období dvoch nasledujúcich rokov v máji 2018 a máji 2019. Hladiny Mn, Cu a Pb sa zisťovali atómovou absorpčnou spektroskopiou s grafitovou pieckou GF AAS. Namerané výsledky sa doplnili dotazníkovými údajmi od majiteľov o údržbe studní, spôsobu použitia vody a monitorovania jej kvality.
Výsledky: Medián hladiny Mn v roku 2018 bol 8,9 μg/l. Parametrickú hodnoty prekračovali tri vzorky. V roku 2019 bol medián hladiny Mn 11,0 μg/l. Parametrickú hodnoty prekračovali dve vzorky. Medián hladiny Cu v roku 2018 bol 9,6 μg/l v roku 2019 13,3 μg/l. V roku 2019 parametrickú hodnotu pre Cu prekračovali dve vzorky. medián hladiny Pb v roku 2018 bol 4,8 μg/l a jedna vzorka prekračovala parametrickú hodnotu. V roku 2019 bol medián hladiny Pb 4,1 μg/l. Vplyv počasia na hladiny ťažkých kovov sa nepreukázal ako signifikantný.
Závery: Kontaminácia studničnej vody ťažkými kovmi predstavuje potenciálne ohrozenie zdravia ľudí. Dôkladná a pravidelná údržba hrá významnú úlohu v prevencií možných dopadov na zdravie.

Kľúčové slová: Ťažké kovy. Kontaminácia vody. Studničná voda.

INTRODUCTION

Although more than 90 % of Slovak population is supplied by public water supplies, individual sources (wells) are quite common, similarly, as in other European countries [1]. It applies namely for rural areas, where hilly terrain represents a problem with further construction of public water supply´s infrastructure. However, the water quality monitoring is left on their owners [2] and experience has shown that most of them do not analyze well water quality regularly and there are also problems with an appropriate stewardship [3].

Heavy metals include over 50 elements. Some of them such as copper and chromium are essential for a human metabolism. On the other hand, there are ones being toxic even in low levels and their chronic exposure can be associated with health disorders [4-6].

Individuals are most likely exposed to heavy metals through water consumption. Water can be polluted either by naturally occurring metals in earth crust or by anthropogenic pollution [7, 8]. A secondary water pollution in the water supply system can present another problem due to adverse hydraulic conditions of the network, combined with pipelines ageing or chemical instability of water. Such events can lead to formation of sediments providing a reservoir of accumulation of toxic substances, including also heavy metals. [8].

GOALS

The aim of the study was to analyze levels of manganese (Mn), copper (Cu) and lead (Pb) in well water in northwest Slovakia. We also analyzed interannual variability of these heavy metals.

METHODS

Water samples from the private wells were collected in a period of two subsequent years in the village of Korňa, which is unique by its settlement and frequent use of private wells. The first sampling was realized in May 2018 and the second one in May 2019.

The village of Korňa (18 ° 32'10 "E, 48 ° 24'42" N) is situated in northwest Slovakia (Figure 1) having a population of 2057 inhabitants and is typical for its 36 traditional dispersed settlements called "kopanice" or "osady". They are situated mostly in higher elevations of valleys or mountains, not close to a community. The public water supply has been established in 2012. However, especially individuals living in remoted dispersed settlements has only limited access to this and are depended on their private wells. In 2018, only 78.3 % of inhabitants were connected to the public water supply.

Geotechnical profile of the studied area is built by flysch layers, made up of sandstones, claystones and clayshales. The previous geochemical studies in the area confirmed higher levels of manganese in ground water with relatively high variability. The village is known by its open natural oil-gas seepage (protected natural heritage of Slovakia).

The prevailing wind in the Korňa has northwest direction. There are no significant industrial pollution sources close to the area. On the other hand, possible air pollution from nearby Moravian-Silesian Region cannot be omitted.

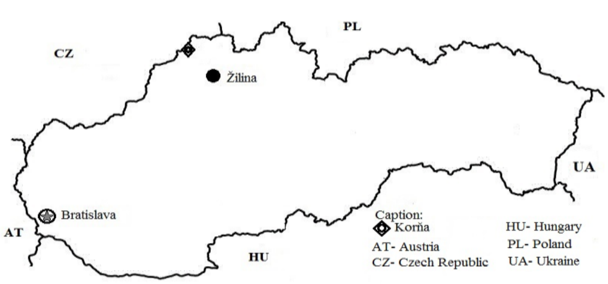

Figure 1. Location of the village of Korňa

Meteorological characteristics:

The village of Korňa belongs to a slightly warm, very wet to cold climate zone [9]

1st data collection (May 2018): The temperatures in a winter and spring were highly about the average. In April, dry season started. The rain started one days after the water sampling.

2nd data collection (May 2019): The first winter months to the first part of January got a lot of snowfall; the temperature was in normal. From the January significant dry season started. The rain season started one week after the water sampling.

Samples

Data collection started in May 2018. To guarantee the owners ‘privacy, the samples were taken by the owners themselves with the assistance and supervision of the activists trained beforehand. The well owners received from the activists information leaflet including all information about the project together with respective contacts, sterile polyethylene bottles and the complementary questionnaire. Both, the sample container, and the questionnaire were labelled with the same identification code.

The samples were collected in 250 ml sterile polyethylene bottles. Immediately before sample taking, water was run for 1 to 5 minutes and then the bottles were rinsed three times with the water. If the water was sampled directly from the well, the string was attached to the bottle neck. Subsequently, the bottle was filled up to 2/3 of the maximum volume, shaken and poured them out. Then, the bottle was fully filled with the water up to the edge. All the samples were transported to the laboratory within 24 hours.

In the laboratory, 67 – 69 % of trace metal grade HNO_3 was added into the samples (500 µl of HNO_3 in 100 ml of well water) and afterwards each sample was filtered by cellulose filtration paper. The levels of the Mn, Cu and Pb were determined by atomic absorption spectroscopy with graphite furnace GF AAS (AAS GBC XplorAA 5000 with GBC GF 5000) equipped with hollow cathode lamps. Argon was used for the inert gas flow and deuterium lamp was employed for the background correction. The chemicals for the experiments were in a high purity grade. For the Pb analysis we used 1 % Pd modifier. We used ultrapure water Type 1 (UPV H_2O) with resistance 18.2 MΩ.cm for the blank, standards preparations and water dilutions. All the containers used for the laboratory analysis were washed up

Table 1 Specific parameters for the analysis of selected heavy metals

Element	Hollow cathode lamp (wavelength, current)	Std. level[1]	2018		2019	
			LOD[2]	LOQ[3]	LOD	LOQ
Mn	279.5 nm, 5 mA	6 µl/l	0.1 µg/l	0.4 µg/l	0.4 µg/l	1.2 µg/l
Cu	175.0 nm, 4 mA	24 µg/l	0.4 µg/l	1.0 µg/l	1,3 µg/l	3,9 µg/l
Pb	217.0 nm, 5 mA	26 µl/l	1.2 µg/l	4.0 µg/l	1.0 µg/l	3.35 µg/l

Legend: [1]Std. concentration standard concentration, [2]LOD limit of detection, [3]LOQ limit of quantitation

with ultrapure water, subsequently sank in a trace metal grade HNO_3 for a day and then rinsed with UPV H_2O. The calibration solutions standards were acidified by trace metal grade HNO_3 to the same levels of concentration as the samples. Table 1 shows specific parameters of the heavy metals´ analysis

The measured levels were complemented by questionnaire data on numbers of potentially exposed persons in household, well design and its construction characteristics, well maintenance frequency, well water use and the water quality monitoring. Levels of heavy metals were compared with parametric values (PV) given in in the Resolution of the Ministry of Health of Slovak Republic No. 247/2017 Coll. corresponding to Annex I to Directive 98/83/EC on the quality of water intended for human consumption. In addition, levels of manganese were also compared with PV 200 µg/l given in the Resolution of the Ministry of Health of Slovak Republic No. 247/2017 Coll. This PV applies for the areas with high natural occurrence of manganese in the earth crust.

The measured levels were graphically expressed as decadic logarithms.

RESULTS

We analyzed 15 well water samples and questionnaires. As much as 62 individuals used the well water, including 9 children. Most of them (80 %) used the water for drinking. Median usage of the wells was 27 years. Majority of them (73.3 %) were dug and only 4 owners used a filtration system. Only around half of them realized inspection of water well system annually; three owners never inspect their system. More than a quarter (26.7 %) realized at least one water quality testing since the start of the well´s operation (Table 2).

Manganese

We quantified manganese in 15 samples. Median level in 2018 was 8.9 µg/l, maximum was 805.0 µg/l. Parametric value (PV) in drinking water 50 µg/l was exceeded in 3 samples. Three samples exceeded the Slovak PV for manganese (200 µg/l), 2 of them were used for drinking. In 2019, the median was 11.0 µg/l and maximum 295.3 µg/l (Table 3). PV for manganese in drinking water 50 µg/l exceeded 2 samples and only one sample exceeded Slovak PV 200 µg/l. To summarize, from 3 initial samples exceeding PV 50 µg/l, all of them were below the PV after the second analysis. On the other hand, from three initial levels exceeding PV 200 µg/l, only one sample did not meet after the subsequent analysis the PV. We registered declined levels in 11 samples. The deepest decrease was by 801.8 µg/l. In 4 samples we noticed an increase. All these samples were in 2018 below manganese PV (50 µg/l), but two of them reached levels over PV in 2019 (Fig. 2).

Copper

We quantified copper in all 15 samples. Median level in 2018 was 9.6 µg/l and maximum was 641.6 µg/l. None of the samples exceeded PV for cooper in drinking water (Table 3). On the other hand, median level in 2019 raised to 13.3 µg/l. and maximum was 3442.0 µg/l. This maximum level was the only one event, when PV for cooper was exceeded. An increase of concentration in this sample was by 3435.0 µg/l resp. 491.7 times higher in comparison with initial level. Water from this well was used for drinking and has been operating for 34 years. In general, levels raised in 11 samples (73.3 %) and only 2 of them increased in more than 100 % (7.0 µg/l to 3442.0 µg/l; 9.7 µg/l to 145.2 µg/l). We noticed in one sample decrease from 641.6 µg/l to 13.3 µg/l.

Lead

We quantified lead only in 7 water samples in 2018. Median level was 4.8 µg/l and maximum was 20.0 µg/l. One sample exceed PV 10.0 µg/l of lead in drinking water (Table 3). The respective well was used for 40 years for garden watering. After a year we quantified 11 samples with levels higher than LOQ. Median level was 4.1 µg/l. Maximum in 2019 was 8.9 µg/l, none sample exceeded Pb PV. The

Table 2 Selected characteristics of private well water supply

Characteristics	Categories	Korňa (N=15)
Type of well	drilled	4
	dug	11
Well water components	filtration system	4
Inspection of water well system	annually	6
	every few years	6
	never	3
Water testing	at least once	4
Well water use	drinking	12
	well water only	*3*
	bottled and well water	*9*
	watering	7
	personal hygiene	10

Table 3 Measured manganese, copper and lead levels in well water samples in 2018 and 2019

Parameter	Mn		Cu		Pb	
	2018	2019	2018	2019	2018	2019
>LOQ (No)[1]	15	15	15	15	7	11
average (µg/l)	151.5	46.8	54.1	250.9	7.0	5.0
geo. mean[2] (µg/l)	25.8	15.9	13.9	22.3	5.9	4.7
min.[3] (µg/l)	2.9	3.2	5.4	8.4	4.4	3.4
25. percentile	5.8	4.3	11.5	4.7	4.7	3.8
median (µg/l)	8.9	11.0	9.6	13.3	4.8	4.1
75. percentile	114.7	38.8	18.8	19.8	5.2	5.6
max.[4] (µg/l)	805.0	295.3	641.6	3442.0	20.0	8.9
>PV (No) [5]	6	3	0	1	1	0

Legend: [1]Number of samples above limit of quantitation; [2]Geometric mean (µg/l); [3]Minimum. (µg/l); [4]Maximum (µg/l);[5] Number of samples exceeding parametric value given in in the Resolution of the Ministry of Health of Slovak Republic No. 247/2017

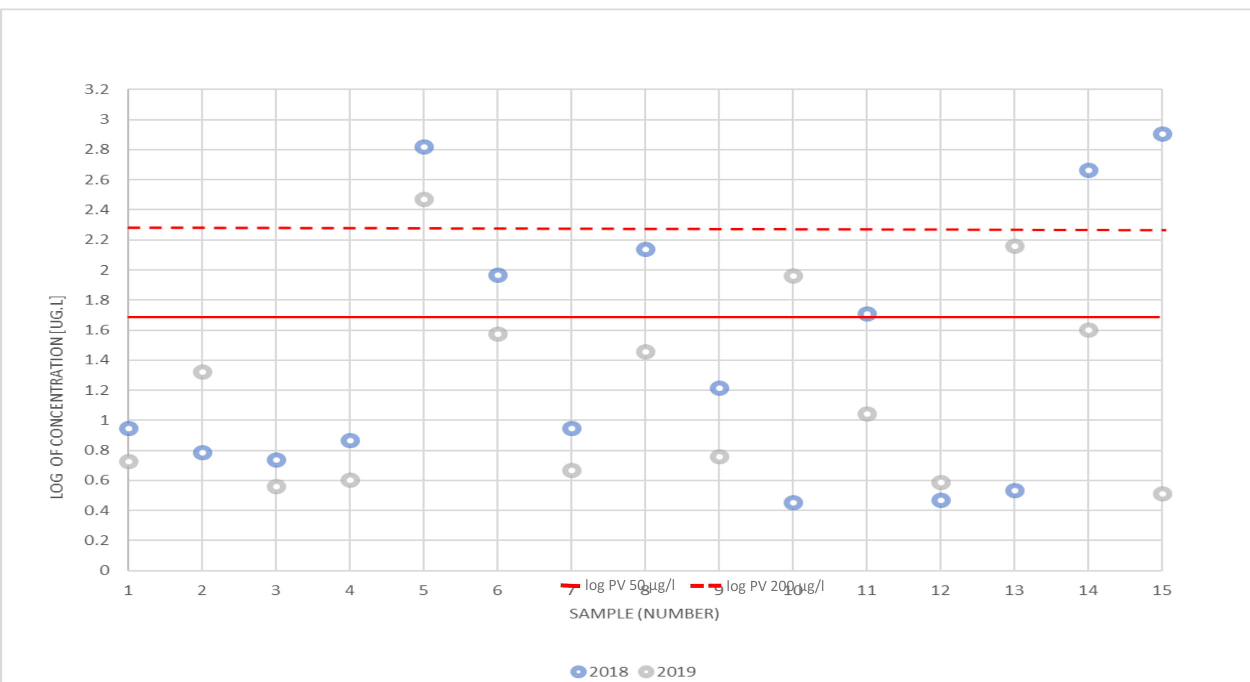

Figure 2 Manganese levels in Korňa village in 2018 and 2019

sample reaching 20.0 µg/l in 2018 decreased to the unquantified level in 2019.

DISCUSSION

Water quality depends on hydro-chemical characteristics of the area [10]. The selected area is not classified as an anthropogenic polluted one, but it is characterized by higher levels of manganese (Mn) in a geological substratum [11]. Moreover, Mn levels in groundwater may vary over time, especially when water from the unconfined aquifer mixes with water from the upper confined aquifer [12]. Therefore, PV 200 µg/l is required (the limit can be applied only when the sensory water quality parameters are not changed) while drinking water health risk assessment. We detected in May 2018 three samples exceeding Slovak PV for Mn in drinking water, two of them were used for drinking (463.9 µg/l, 660.0 µg/l). Here we can see a problem with Mn overexposure and its possible health effects. Several studies stated that chronic low-level exposure to Mn affects neurobehavioral functions, especially in children [6, 13]. Therefore, the situation deserves an attention.

Hydrogeological survey in the given area did not confirmed disturbing levels of copper (Cu) in the environment [14]. It corresponds with our results as none of the water samples exceeded the PV in 2018. The increased Cu level in 2019 can by caused by anthropogenic contamination by a fungicide agent or by a release of copper from materials used in plumbing [15]. However, median usage of wells was 27 years, so ageing of water system can play an important role, too [16].

The negative health effects of lead (Pb) can occur while drinking contaminated water [4, 5]. Similarly, individuals using well water for watering their fruit and vegetable are at risk, too [17]. Although levels of Pb were normal except one sample exceeding PV (20.0 µg/l) [18], we should pay attention to the situation considering recently adopted revised Directive EU 2020/2184. This Directive, taking into account current body of knowledge, defines borderline of abnormal Pb values much more rigorously (5 µg/l) [19]. According to this two wells would not meet PV in 2018 and four in 2019. There are several factors which can have an impact on Pb levels in well waters. Firstly, Korňa village is famous by its natural spills of crude oil [20]. Previous research on Pb isotopes confirmed, that the element occurs in parents' rocks and its higher levels can be found in upper layers of soil [21]. Secondly, Pb can release into the well water from pure lead pipes, galvanized iron pipes, lead solders or other piping materials containing lead, especially when they corrode [22]. Therefore, a regular maintenance of the well is recommended (at least once a year) [3, 17]. Unfortunately, only around half of well owners do the inspection annually. Tertiary, as the previous studies confirmed, higher levels of Mn in well water may result in a Pb release from the water system [23]. Therefore, we should make efforts to eliminate possible health risks through targeted preventive measures such as using appropriate materials for plumbing and providing proper maintenance of the system.

Several studies described seasonal variation in heavy metals levels [24-26]. Our analysis did not confirm any variations in Mn or Cu levels between 2018 and 2019. Climatic conditions were from winter 2018 to May 2019 similar and the changes in precipitation or temperature could not affect these levels. On the other hand, we noticed the significant decrease of extreme Mn levels. We suppose it as an effect of preventive measures after the first testing in May 2018. We recommended all the owners with exceeded Mn PV to get a manganese filters. Changes in water quality can also depend on frequency and intensity of water pumping [27]. Although Cu levels in 73.3% samples increased, median Cu levels between 2018 and 2019 were not significantly altered. However, the weather patterns in the studied period were similar and we did not suppose higher demand on water. Therefore, changes in physicochemical processes in the water system can explain the increase [28]. We informed the well owner after the first analysis about changes in sensory parameters as well as risk of corrosion in the system. Reverse osmosis can be considered as an effective and not expensive tool to lower Cu levels [29].

Our study has two limitations. Firstly, number of samples were relatively small. Some wells in dispersed settlements of Korňa are very old, build and used without appropriate permission so the inhabitants felt worried about possible sanctions. Secondly, we undertook well water sampling only twice – spring 2018 and spring 2019. Both sampling periods were in terms of hydrometeorological conditions similar regarding temperature and precipitation.

Eventually, our analysis pointed out that heavy metals levels in well water vary across time and repeated measurements are needed to obtain a reliable picture on the actual situation. The results further indicate that an appropriate stewardship rather than weather conditions determines this variability. Therefore, knowledge and attitudes of wells' owners plays an important role. Namely, campaigns organized in occasion of the World Water Day (22 March) would be better focused on these issues and to increase public awareness on a significance of low level concentrations of heavy metals.

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