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Dedicated to Professor Emil Cordoș on the occasion of his 80th anniversary

THE EVALUATION OF THE METAL CONTAMINATION OF DRINKING WATER SOURCES FROM MEDIAS TOWN, ROMANIA USING THE METAL POLLUTION INDICES

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ABSTRACT. Drinking water represents a direct source for the toxic and persistent metals entrance in the organisms. Their bioaccumulation makes them harmful for living organisms, including humans. Assessing the quality of the drinking water sources represents a significant prevention and protection measure. Quality assessment methods such as heavy metal pollution index, degree of contamination and heavy metal evaluation index are applied worldwide. The aim of current study was to evaluate the metal contamination of drinking water sources from Medias town using the pollution and quality assessment indices. The concentrations of As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn were assessed in 21 drinking water sources (19 well waters and 2 public springs). Results indicate Mn and Cd concentrations exceeding the threshold limits of the drinking water quality guidelines, only in summer season, in three samples. In the spring season the regulatory limits were not exceeded. The quality assessment indices show low contamination degree, all values being lower than the critical ones. Positive correlations were observed between pollution indices, As, Cd, Mn, Zn, Ni and Cu concentrations.

***Keywords**: drinking water sources, contamination index (C_d), heavy metal evaluation index (HEI), heavy metal pollution index (HPI)*

INTRODUCTION

Today, throughout the world, the quality of drinking water sources is declining especially due to the population and economic growth. Countless and diverse wastes, hazardous contaminants and emissions are released

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directly and indirectly into the environment [1-3]. Metals are among the most frequent contaminants that pose a hazard for the environment due to their persistence, toxicity, bioaccumulation and non-biodegradability [4, 5]. High concentrations of metals in aqueous environments led to serious problems concerning the human, animal and plant health [6, 7]. Accumulated metals in the human body may cause damage of internal organs, neurological disorders, inhibition of embryo and children development, and even death [8, 9]. Bioaccumulation of metals occurs in plants also, therewith negative effects and affection of the photosynthesis, absorption and exchange of gases [10, 11]. Anthropogenic sources (metallurgical industry, smelting, mining, galvanic processes, traffic) and geological background are reflected by the metals spatial variability [12-14]. As a worldwide concern, water quality studies are carried out to monitor and to prevent the chemical pollution [15, 16].

Metal pollution indices are significant tools for drinking water quality assessment and have been successfully applied all over the world [17-20]. Assessment methods, such as the quality index are effective in gathering a composite influence of indicators on the overall contamination [21-23]. The C_d (degree of contamination) represents an approach to evaluate the areas characterized by unsafe concentrations of defined metals [24]. The applied method considers the combined effects of the studied elements (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) with harmful effects of a consumed contaminated drinking water [25]. The *HPI* (heavy metal pollution index) is based on the weighted arithmetic quality mean method generated in two basic steps: selection of a rating scale (an arbitrary value between 0 and 1) for all quality characteristics, giving weightage to the selected elements followed by the selection of the pollution indicators, which represents the essential part of the index count [17]. For drinking water, the critical *HPI* value of 100 is considered [17].

Study results indicate exceedances of the metal maximum admissible concentrations (*MACs*) for the As, Fe, Mn, Pb and Cr concentrations, and high values for the metal pollution indices. Results present values higher as the critical ones, which are 3 for the C_d (degree of contamination), 100 for the *HPI* (heavy metal pollution index) and 400 for the *HEI* (heavy metal evaluation index) [19].

The aim of the current research was to assess the quality of the groundwater used as drinking water source, from Medias town, starting from the study carried out in 2014, in which the chemical status and inorganic contamination of well waters from urban area (Medias town) and mining areas (Tarna Mare, Trut and Turulung) were investigated and compared [26]. Considering the high values of metals (Cd and Mn exceeding the maximum admissible limits) present in the well waters from Medias town, studying the pollution status from a different view, using different methods represents a necessity due to the use of well waters as drinking water source. Hereby, in the present research metal pollution indices methods (*C_d, HPI* and *HEI*) were used as water pollution evaluation tools, summarizing the combined effects of metals concentrations (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn). High concentrations of metals are considered toxic and harmful to the water quality and ecosystems and unassailably to animals and human health [24].

RESULTS AND DISCUSSION

The descriptive statistics of As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn in the studied drinking water samples are given in table 1. The studied metals, except Mn and Cd, are below the regulatory limits of the drinking water standards. Water samples W9 and W10 have Mn concentrations (66 µg/l and 82 µg/l) higher than *MAC* in the summer season with 33 % and 41 %, respectively [26]. In the spring season, Mn level is lower as 50 µg/l [26]. Still, average values of Mn concentrations in both seasons for W9 and W10 are 49 µg/l and 50 µg/l, second value reaching the regulatory limit [26]. In the summer season, 29 % of the samples present higher Mn concentrations as in the spring season [26]. In the spring season the maximum concentration was 33 µg/l, while in the summer season was 82 µg/l, and the lowest level was 0.12 µg/l, respectively 0.20 µg/l [26]. According to Zaporozec (1981), the high concentrations of Mn have naturally occurrence in the groundwater system from the soil matrix [27].

The Cd threshold limit (5 µg/l) is exceeded for sample W12 almost two times (average of 9.6 µg/l); 71 % of the samples present higher Cd levels in summer than in spring while 67 % of the samples have concentrations, below the quantification limit (0.10 µg/l). Possible source for the high Cd concentrations is a former non-ferrous metallurgical plant, localized at a relatively short distance (almost 15 km).

Table 1. Descriptive statistics for the studied elements [26, 28]

Element (µg/l)	Minimum	Maximum	Mean	Median	Standard Deviation	MAC*
As	0.08	4.3	0.77	0.33	1.0	10
Cd	<0.10**	9.6	0.55	0.04	2.1	5
Cr	0.47	10	2.8	2.2	2.0	50
Cu	0.84	8	3.9	3.7	2.2	100
Fe	32	99	58	52	19	200
Mn	0.30	50	9.7	3.3	15	50
Ni	1.0	5.2	2.2	1.8	1.1	20
Pb	0.10	6.2	1.02	0.40	1.61	10
Zn	10	970	132	48	228	5000

*According to Romanian legislation regarding the drinking water quality [28]
**Below the quantification limit (0.10 µg/l)

The calculated pollution indices for the studied water samples are given in table 3. According to the C_d, the lowest degree of contamination was obtained for water sample W2 (-8.7), while the highest degree of contamination was determined for water samples W12 (-5.2). The studied region was found to have low degree of contamination, as the C_d average value (-8.0) indicates. Similar results were obtained by Backman et al. (1997), using the C_d for groundwater samples from Slovakia and Finland, but also values of $C_d = 1$-3 and C_d >3 for studied elements (Al, As, B, Ba, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Rb, Sb, Se and Zn) which indicates groundwater contamination, having as contamination sources: mining activities, natural occurrence and geogenic processes of metals [24]. In a different study conducted in Bangladesh, on drinking water sources, the C_d was used; values between 3.45 and 85 were obtained, results suggesting a high contamination status of the water samples [20].

Table 2 presents the indices used for the calculation of *HPI* count for water samples. The mean concentrations of the two data sets obtained in both sampling campaigns were used. The Romanian threshold limits (Law 311/2004 on drinking water quality were used as the standard permissible values [28].

According to the *HPI* results, the highest obtained value is 100.3 for sample W12, while spring sample S2 is characterized by the lowest metal pollution (1.8). Thought the *HPI* mean value indicates low metal pollution (10). The highest concentrations for Pb, Cd and As were measured in sample W12, which implies the highest value for *HPI*. Higher *HPI* values than the critical value of 100, for studied metals (As, B, Cd, Co, Cr, Fe, Mn, Pb, Se, Sr and Zn) were obtained by Ehya and Marbouti (2016) for groundwater samples collected from Iran, due to the leaking of industrial and urban sewages into the aquifers, as well as fossil fuels combustion [29]. Lower *HPI* values than 100, for studied metals (Cd, Pb, and Zn) were obtained in Tirupati, Indian groundwater samples used as drinking water source [17].

Based on the average *HEI* value (1.0) and the metal indices calculation, the studied water samples are assessed as having low metal level. Sample W2 has the lowest *HEI* value (0.34), while sample W12 has the highest *HEI* value (3.8), indicating low metal pollution level. Bhuyan et al. (2010) used as well the *HEI* for drinking water samples and obtained results between 10.26 to 367 [20].

Correlations between the measured metal concentrations and metal pollution indices were calculated using the correlation matrix.

Table 2. *HPI* calculation for drinking water sources of Medias town, $\sum W_i$=0.51 $\sum W_i \times Q_i$=5.2; *HPI*=10

Metals	Mean concentration -M_i-(μg/l)	Standard permissible value -S_i-(μg/l)	Unit weightage -W_i-	Sub index -Q_i-	$W_i \times Q_i$
As	0.77	10	0.10	7.7	0.77
Cd	0.55	5	0.20	11	2.2
Cr	2.8	50	0.02	5.5	0.11
Cu	3.9	100	0.01	3.9	0.04
Fe	58	200	0.01	29	0.14
Mn	9.7	50	0.02	19	0.39
Ni	2.2	20	0.05	10.9	0.54
Pb	1.0	10	0.10	10.2	1.0
Zn	132	5000	0.0002	2.6	0.001

Table 3. Calculation of the metal pollution evaluation indices

Sample no.	C_d	Mean deviation	*HPI*	Mean deviation	*HEI*	Mean deviation
W1	-7.9	-0.07	8.6	1.7	1.1	-0.07
W2	-8.7	0.66	2.1	8.2	0.34	0.66
W3	-8.5	0.47	3.5	6.8	0.53	0.47
W4	-8.2	0.20	7.8	2.9	0.80	0.20
W5	-8.3	0.26	4.1	6.2	0.74	0.26
W6	-8.5	0.45	3.5	6.8	0.55	0.45
W7	-8.5	0.53	2.8	7.5	0.47	0.53
W8	-7.8	-0.21	5.2	5.1	1.2	-0.21
W9	-7.3	-0.68	6.6	3.7	1.7	-0.68
W10	-7.5	-0.45	6.3	4.0	1.5	-0.45
W11	-8.3	0.26	3.8	6.5	0.75	0.26
W12	-5.2	-2.8	**100.3**	-90	3.8	-2.7
W13	-8.2	0.22	7.2	3.1	0.78	0.22
W14	-7.3	-0.67	19	-8.9	1.7	-0.67
W15	-8.2	0.21	4.8	5.5	0.79	0.21
W16	-8.4	0.38	3.5	6.8	0.62	0.38
W17	-8.4	0.41	3.6	6.7	0.59	0.41
W18	-7.9	-0.05	5.9	4.4	1.1	-0.05
W19	-8.1	0.13	7.3	3.0	0.87	0.13
S1	-8.1	0.15	8.6	1.6	0.86	0.15
S2	-8.6	0.56	1.8	8.5	0.44	0.56
Minimum	-8.7		1.8		0.34	
Maximum	-5.2		100.3		3.8	
Mean	-8.0		10		1.0	

Thus, positive relationships between metal concentrations can be observed, such as Zn and Mn and Zn and Cu. Also, there is a positive and direct correlation between As, Cd and Pb. The level of Cd is as well directly related to the Ni concentration (Tabel 4).

Table 4. Correlation matrix for the obtained metal concentrations and metal pollution indices values (n=21)

	As	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Cd	HPI	HEI
As	**1**											
Cd	**0,797***	**1**										
Cr	0,081	0,176	**1**									
Cu	0,184	0,190	0,339	**1**								
Fe	-0,038	-0,116	0,421	0,125	**1**							
Mn	-0,089	-0,091	-0,040	0,332	-0,198	**1**						
Ni	0,425	**0,534**	0,012	-0,046	0,136	-0,218	**1**					
Pb	**0,838**	**0,720**	0,233	0,047	-0,067	0,018	0,282	**1**				
Zn	0,161	0,352	0,093	**0,580**	-0,210	**0,579**	0,017	0,147	**1**			
Cd	**0,785**	**0,861**	0,272	0,375	-0,015	0,347	0,426	**0,788**	**0,547**	**1**		
HPI	**0,628**	0,285	0,306	0,307	0,002	0,275	0,154	**0,744**	0,194	**0,573**	**1**	
HEI	**0,785**	**0,861**	0,272	0,375	-0,015	0,347	0,426	**0,788**	**0,547**	**1,000**	**0,573**	**1**

*Values in bold are different from 0 with a significance level alpha=0.05

The Pearson correlation matrix revealed that the metal pollution indices (C_d, *HPI* and *HEI*) are directly affected by As, Cd, Pb and Zn concentrations. Additionally, the comparison between the used assessment methods indicates a significant correlation, especially between C_d and *HEI*, positively correlated (table 4). For example, the highest As, Pb and Cd concentrations in sample W12, is directly correlated with the highest *HPI*.

CONCLUSIONS

The studied drinking water sources from Medias town present high concentrations of Cd and Mn, exceeding the *MAC*s, while As, Fe, Cr, Cu, Zn, Pb, Ni do not exceed the threshold limits. The Mn level exceeds the *MAC* in the summer season with 33 % (W9) and 41 % (W10), but in the spring season does not exceed the corresponding *MAC*. In addition, in summer season Cd *MAC* is surpassed in 71 % of the studied samples. There is a positive correlation between Pb, Cd and As, similarly between Zn and Cu and Zn and Mn concentrations. Metal pollution indices (C_d, *HPI* and *HEI*) showed that water samples are characterized with a low contamination degree ($C_d < 0$) and the calculated *HPI* was below the critical value of 100, except sample W12, which exceeds the threshold with 0.003 %. Sample W12 presents the highest As, Cd and Pb concentrations for which the highest *HPI* was obtained, which indicates a positive correlation between the metal concentrations and the HPI results. The *HEI* classifies the water samples as low contamination. Methods C_d and *HEI* present a significant correlation and positive correlation between C_d and *HPI*, *HEI* and C_d.

EXPERIMENTAL SECTION

General description of the study area

Study area is located in Medias, Sibiu County, Transylvania, Romania (figure 1). At 15 km South-West from Medias, in Copsa Mica town, one of the biggest Romanian non-ferrous metallurgical industry unities was localized. Copsa Mica area is known as a metal pollution *hot spot* in Romania and in Europe [30, 31]. The main activity of the company was the production of Pb, Sn, Cu and Zn, until 2005 [30, 31]. The area was the subject of numerous studies regarding the environment pollution, indicating high level of metals, which exceed the *MAC* and deteriorate the health condition after the exposure. As a result, mortality and morbidity among the inhabitants of Copsa Mica area increased [32].

Medias area is characterized by hill formations localized on the Tarnava Mare terraces [33]. Smooth cliffs and horizontal fragmentation characterize Medias town surroundings [34]. The climate is continental temperate and it is governed by the average annual temperature of 8 ºC, 600-800 mm for the mean annual precipitations and average annual humidity of 87 % [34].

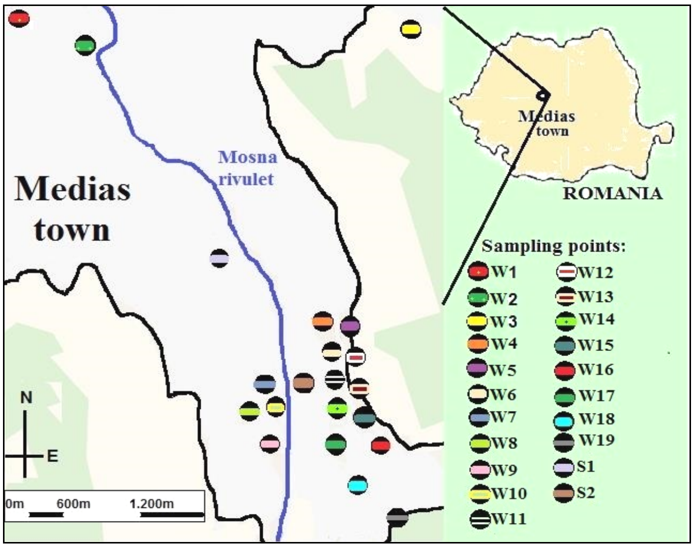

Figure 1. Study area including sample location map

Town is crossed from Vest to East by Tarnava Mare River and from South to North by its right side tributary, Mosna Rivulet [35]. Tarnava Mare's water is used as drinking water source as well as the natural springs. The groundwater is directly used from private well waters and public natural springs by the inhabitants.

The main objectives of the current research are to determine the metal content of the groundwater used as drinking water sources and to assess the water quality using the metal pollution indices.

Sampling, sample preparation and instrumentation

A number of 21 groundwater samples were collected from drinking water sources (19 well waters, W1-W19 and 2 public springs, S1-S2) localized in Medias town [26]. Samples were collected in two sampling campaigns (spring and summer 2014). Polyethylene bottles (1000 ml) were used for the collection of the water samples, which were preserved with HNO_3 (for acidification at pH 2) and until the chemical analysis kept at 4 ºC; 0.45 µm acetate cellulose filter membrane were used for sample filtration. The metal content (As, Cd, Cr, Cu, Fe, Mn, Pb and Zn) was measured by inductively coupled plasma mass spectrometry (ICP-MS) using an ELAN DRC II (Perkin-Elmer, Canada) instrument.

Evaluation methods

Three quality evaluation methods were used in the study, namely the degree of contamination (C_d), the heavy metal pollution index (*HPI*) and the heavy metal evaluation index (*HEI*).

The degree of Contamination (C_d)

The C_d is calculated using according to equation 1 and 2 [24] each water sample separately, as a sum of contamination factors of the studied metals exceeding the upper permissible values [19, 24].

$$C_d = \sum_{i=1}^{n} C_{fi} \tag{1}$$

$$C_{fi} = \frac{C_{Ai}}{C_{Ni}} - 1 \tag{2}$$

Where, C_{fi} represents the contamination factor for the *i*-th element (i = As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn), C_{Ai} is the analytical value for the *i*-th element and C_{Ni} is considered to be the upper permissible concentration of the *i*-th element; with *Ni* as the normative value regularized by the Romanian legislation [19, 24]. Studied metals maximum admissible concentrations presented by Law 311 from 2004, regarding the quality of the drinking water were used [28]. The C_d values are classified in three categories, reflecting the contamination level (*CL*): low *CL* (C_d < 1), medium *CL* (1 < C_d < 3) and high *CL* (C_d > 3) [19, 24].

Heavy metal pollution index (HPI)

The quality of water with respect to metals (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) is given by *HPI*. The *HPI* was calculated according to equations 3 and 4, [17]:

$$HPI = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i} \tag{3}$$

$$Q_i = \sum_{i=1}^{n} \frac{\{M_i(-)I_i\}}{(S_i - I_i)} \times 100 \tag{4}$$

The unit weightage of each *i*-th parameter is represented by W_i with *n* as the total number of the indicators considered [17]. The Q_i and the M_i are the sub-index and the monitored value of the *i*-th parameter, while the standard value and the desirable value of the *i*-th parameter are S_i *and* I_i, respectively [17]. The W_i was defined as inversely proportional to S_i, which was taken according to the Romanian legislation [28]. In current study the desirable value, I_i is considered 0, because in the Romanian legislation no ideal value was established.

Heavy metal evaluation index (HEI)

The *HEI* represents a quality assessment tool with respect to metals. The method is expressed using equation (5) [19]:

$$HEI = \sum_{i=1}^{n} \frac{H_c}{H_{mac}} \tag{5}$$

The technique uses H_c as the monitored value and H_{mac} as the MAC of the *i*-th parameter [19]. In the current research, the considered MAC, is used according to Romanian legislation, Law 311, from 2004 [28]. Bhuyan et al. (2010) suggested *HEI* criteria for groundwater, with a baseline established by Edet and Offiong, but for surface water (2001): low pollution (*HEI* < 40), medium (*HEI* = 40-80) and high pollution (*HEI* > 80) [19, 20].

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