

Assessing the Aquatic Environment Quality Contaminated with Heavy Metals as a Result of Polymetallic Mining in the North-West Region of Romania Using Pollution Indices

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Abstract—The main objective of this paper is the assessment of surface water quality polluted with various heavy metals (HM) using the quality indices and the information submitted may be useful for the competent authorities and the public. Cadmium (Cd), iron (Fe), manganese (Mn) and zinc (Zn) were determined from water samples of Socea Valley using the Atomic Absorption Spectrometry technique. The calculated water quality indices indicate that the monitored surface water between 2011-2014 years was deteriorating due to the former mining activities from this area. Most of the HM levels were highest compared to the acceptable standard limit. The level of Heavy Metal Evaluation Index (HEI) shows that the water quality falls within the low quality class and the values of Degree of Contamination (CdI) shows a high contamination. Also, the correlation matrix was performed between the analyzed heavy metals. The results obtained for the Pearson's coefficient showed positive values for all heavy metals and between Fe and Mn the value of r being 0.933. All data revealed that the water from Socea Valley is not safe especially for drinking and irrigation purposes.

Index Terms—Heavy metals, mining, polluted aquatic environment, water quality indices.

I. INTRODUCTION

Because the water is very important natural resources on the earth, its quality becomes a global concern [1], [2]. For this reason, the monitoring and the assessment of the water pollution are a critical area of study and are the primary aims of the Water Framework Directive 2000/60/EC, approved by the European Commission [3], [4]. The pollution of aquatic environments with heavy metals (HM) is a serious ecological issue due to their stability in the environment [5]-[7]. Some of HM like Fe and Zn are detrimental to the living organisms at

higher concentrations [3], [4], [8].

The exploitation of natural resources (as mining) are the basic activities associated with the economic growth that in time produce changes in the environment such as HM entering into the natural cycles (water, soil and air cycles) [8]. At international level, the extractive industry legislation has undergone significant changes to ensure a high degree of environmental protection [9]. In Romania, from 1990 to 2007 due to the oversized of the mining sector in the previous period, were halted an impressive number of mining sites, mining companies and societies (462 mining sites between 1998 and 2006, 13 companies in 2006, 85 mining sites in 2007 and 5 in 2010) [10].

To be mentioned is the ecological disaster that occurred in the mining region of Baia Mare in January 2000. Because of a breach that appeared in the walls of the decantation dam, about 100.000 cubic meters of tailing waters, which contained cyanide (50 to 100 tons) and the heavy metals were released into surroundings. The released substances were traveled beyond the Romania borders, into the Tisa River and then into Danube [11].

The surface water taken into consideration in this paper is located in the North West of Romania (Satu Mare County), a traditional area in the field of mining [10]. This research focuses on Cd, Fe, Mn and Zn concentration levels found in the surface water from this interest rural area.

II. MATERIALS AND METHODS

In order to assess the surface water near the closed mining activities located the North West of Romania in terms of pollution, four HM (Cd, Fe, Mn and Zn) were chosen. In the area of Socea Valley (the studied surface water) a number of 160 water samples were taken during 2011-2014.

A. Sampling

The water samples collection was performed under the provisions of SR EN ISO 5667.1-2007 and SR EN ISO 5667.3-2013 [12], [13]. Thus, the sampling points along the Socea Valley were established and named from A1 to A10. From ten different points (hereafter referred to as A1 - A10) the water samples were collected at depth between 0.10 ÷ 0.50 cm. The samples were stored at low temperature (4 °C) during transportation in metal-free 2L polypropylene recipients prior to be prepared and analyzed [4].

B. Analytical Procedure

The preparation stage of the water samples was receiving a special attention in order to achieve real data. The

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polyethylene recipients of 100 mL capacity were used to the prepared water samples. According to the standardized methods specifications in the SR ISO 8288-2001 [14], SR 13315-1996 [15] and SR 8662.2-1996 [16] the samples were analyzed. All the standardized methods applied use the atomic absorption spectrometry technique and the operating instrument was VARIAN type [4]. For the determination of the Cd, Fe, Mn and Zn, the solutions were prepared using double deionized water and the reagents used were of analytical grade (e.g. nitric acid 65% - Merck).

C. Legislative Reference

The Order no. 161/2006 and the Law no. 458/2002 (modified) are considered for the research results discussion [17], [18]. The Order 161/2006 establishes five classes of environmental status for the aquatic ecosystems of lakes and rivers type: (I) very good, (II) good, (III) moderate, (IV) poor and (V) bad [17]. Table I presents the limits of each

environmental status (classes) and of drinking water for the determined HM discussed in this paper. The knowing of the ecological status of surface waters is important for establishing a monitoring program at regional level, especially if the water will fall in class V of water quality.

III. AQUATIC ENVIRONMENT ASSESSMENT METHODOLOGY

Generally, the parameters assessment presents a particular importance because it shows the existence of natural conditions, minor alterations, or extent of anthropic impact and respectively, the situation of water bodies quality in a given period of time. In this paper, the pollution indices used for the assessment of surface water quality regarding HM are described in Table II. In order to calculate the unit weightage (W_i) of the Heavy Metal Pollution Index the constant of proportionality (k) value is considered to be 1 [22].

TABLE I: QUALITY STANDARDS FOR ESTABLISHING THE ECOLOGICAL STATUS AND POTABILITY [17], [18]

Specific toxic pollutants of natural origin	Unit	Quality class					Potability limits
		I	II	III	IV	V	
Cadmium (Cd^{2+})	$\mu\text{g/L}$	0.5	1	2	5	>5	5
Iron ($\text{Fe}^{2+} + \text{Fe}^{3+}$)	$\mu\text{g/L}$	300	500	1000	2000	>2000	200
Manganese ($\text{Mn}^{2+} + \text{Mn}^{7+}$)	$\mu\text{g/L}$	50	100	300	1000	>1000	50
Zinc (Zn^{2+})	$\mu\text{g/L}$	100	200	500	1000	>1000	5000

TABLE II: POLLUTION INDICES DESCRIPTION [1], [8], [19]-[21]

Pollution indices	Acronym	Formulas	Variables	Interpretation
Degree of Contamination – pollution index	CdI	$C_f^i = \frac{C_{\text{sample}}}{C_{\text{background}}} \quad (1)$ $\text{CdI} = \sum_{i=1}^n C_f^i \quad (2)$	<ul style="list-style-type: none"> - C_f^i is the contamination factor - C_{sample} and $C_{\text{background}}$ refer to the mean concentration of i^{th} element - n is the number of contamination factors 	<ul style="list-style-type: none"> $\text{CdI} < 8$ low contamination (LC) $8 \leq \text{CdI} < 16$ moderate contamination (MC) $16 \leq \text{Cd} < 32$ considerable contamination (CC) $\text{Cd} \geq 32$ high contamination (HC)
Heavy Metal Pollution Index	HPI	$W_i = \frac{k}{S_i} \quad (3)$ $Q_i = \frac{100V_i}{S_i} \quad (4)$ $\text{HPI} = \frac{\sum_{i=1}^n Q_i W_i}{\sum_{i=1}^n W_i} \quad (5)$	<ul style="list-style-type: none"> - W_i is the unit weightage for i^{th} element - S_i is the recommended standard for i^{th} element - k is the constant of proportionality - Q_i is the sub-index of the i^{th} element - V_i is the monitored value of the i^{th} element - 100 represents the critical pollution index value - n is the number of HM evaluated 	<ul style="list-style-type: none"> $\text{HPI} < 100$ low HM pollution (LP) $\text{HPI} = 100$ HM pollution on the threshold risk (PTR) $\text{HPI} > 100$ high HM pollution (HP)
Heavy Metal Evaluation Index	HEI	$\text{HEI} = \sum_{i=1}^n \frac{H_i}{H_{\text{mac}}} \quad (6)$	<ul style="list-style-type: none"> - H_i is the concentration value for the i^{th} element - H_{mac} is the standard allowed value for i^{th} element 	<ul style="list-style-type: none"> $\text{HEI} < 400$ low water quality (LWQ) $400 < \text{HEI} < 800$ moderate water quality (MWQ) $\text{HEI} > 800$ high water quality (HWQ)
Metal Index	MI	$\text{MI} = \sum_{i=1}^n \frac{C_i}{\text{MAC}_i} \quad (7)$	<ul style="list-style-type: none"> - C_i is the concentration value for the i^{th} element - MAC_i is the standard allowed value for i^{th} element 	<ul style="list-style-type: none"> $\text{MI} < 1$ suitable for drinking, irrigation and aquatic life water utilizations $\text{MI} = 1$ threshold of danger $\text{MI} > 1$ threshold of warning

IV. RESULTS AND DISCUSSIONS

A. Descriptive Statistics

The quality indicators are considered continuous variables

in statistical terms and a large volume of values are necessary [23]. Therefore, the basic statistical method discussed in this paper was taken into consideration.

Table III and IV presents the basic features of the HM data found in water samples and the Pearson correlation matrix based on the same concentration levels. The r is the Pearson's correlation coefficient and p is the value that indicates a significant correlation between the elements. The data given in the Table V were used to the interpretation of the obtained r coefficient values. All the values of HM found were in $\mu\text{g/L}$.

The metal concentrations were not significantly different between sampling points along the Socea Valley. In the Table III, the mean concentration levels for all HM are higher than the value of the last quality class presented in Table I. So, the Socea's water fall within V class of water quality. Based on the mean concentration and abundance, the heavy metals are ranked as $\text{Fe} > \text{Zn} > \text{Mn} > \text{Cd}$. The total amount of Fe found in each water sample is due to the acid mine waters (AMW) from the Socea mining area.

A very high positive correlation was observed between Fe and Mn with $r = 0.933$ and between the pairs: Cd-Fe, Mn-Zn, Cd-Mn, Cd-Zn and Fe-Zn were obtained a high positive correlation. The pair Mn-Zn has the lowest r coefficient, being 0.794.

B. Aquatic Environment Assessment by Pollution Indices

To calculate the indices CdI, HEI and HPI, the standard values of II quality class for each HM were considered (see Table I). In order to calculate the MI index, the standard values for drinking water were considered (see Table I). Measured pollution indices for the surface water samples in the Table VI are given. The indices values obtained for the each HM are the mean value of the ten water samples.

The indices values ranged as: for CdI from 9.02 to 140.03, for HPI from 1904.60 to 14003.42, for HEI from 10.02 to 91.45 and for MI from 2.00 to 182.91. The values of CdI and HPI indicate a strong pollution of the Socea Valley, excepting the CdI value obtained for cadmium that indicates a moderate contamination. According to the HEI index values for HM, the overall quality of the surface water is low in the sampling

period. Also, the water from Socea Valley is non-suitable for drinking, irrigation and aquatic life water utilizations according to MI index value obtained.

TABLE III: DESCRIPTIVE STATISTIC DISPLAYED FOR THE FOUR HM STUDIED

Element	Mean	StDev	CoefVar	Sum	Min	Median	Max
Cd	10.02	1.54	15.37	100.23	8.74	9.51	13.71
Fe	22412	9706	43.31	224118	12988	19512	48546
Mn	9145	6060	66.26	91453	6205	7472	26296
Zn	14103	3059	21.69	141034	11114	12614	21208

TABLE IV: PEARSON CORRELATION MATRIX FOR THE STUDIED HM

Coefficient	Element $i \rightarrow$	Cd	Fe	Mn
r	Fe	0.811		
p		0.004		
r	Mn	0.846	0.933	
p		0.002	0.000	
r	Zn	0.865	0.800	0.794
p		0.001	0.005	0.006

TABLE V: INTERPRETATION OF r 'S AND p 'S VALUES [24]-[26]

Value	Interpretation
r	
0.90 to 1.00 (−0.90 to −1.00)	Very high positive (negative) correlation
0.70 to 0.90 (−0.70 to −0.90)	High positive (negative) correlation
0.50 to 0.70 (−0.50 to −0.70)	Moderate positive (negative) correlation
0.30 to 0.50 (−0.30 to −0.50)	Low positive (negative) correlation
0.00 to 0.30 (0.00 to −0.30)	Negligible correlation
p	
$P = 0.05$ or $p < 0.05$	Statistical significance correlation (95% confidence level)
$P = 0.01$ or $p = (0.01 - 0.001)$	Highly significant correlation (99% confidence level)
$p = (0.001 - 0.000)$	Very high significance correlation (99% confidence level)
$P > 0.05$	Insignificant correlation

TABLE VI: POLLUTION INDICES VALUES FOR THE HM STUDIED AND THE CONTAMINATION STATUS OF SOCEA'S WATER

Element	Values of Pollution Indices				Contamination Level							
	CdI	HPI	HEI	MI	CdI		HPI		HEI		MI	
					Individual	Overall	Individual	Overall	Individual	Overall	Individual	Overall
Cd	9.02	1904.60	10.02	2.00	MC		HP		LWQ		threshold of warning	
Fe	43.82	4484.99	44.82	112.06	HC		HP		LWQ		threshold of warning	
						HC		HP		LWQ		non-suitable for drinking
Mn	90.45	9149.85	91.45	182.91	HC		HP		LWQ		threshold of warning	
Zn	140.03	14003.42	70.52	2.82	HC		HP		LWQ		threshold of warning	

V. CONCLUSION

The study shows that the water of Socea Valley exceeds the limits of the Order 161/2006 and presents a bad ecological status. The Pearson's correlation indicates that exist a

significant positive correlation between all four heavy metals studied and ranged from a high positive correlation to one very high. The CdI and HPI indices place the surface water quality in high contamination level. In addition, the HEI index shows that Socea's water quality is bad (meaning LWQ) and the MI index indicates non-suitable water for different uses

such as the household. This paper amplifies the utility of the pollution indices and the statistical analysis in the environmental quality control.

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