

Mathematical Models Describing the Relations between Surface Waters Parameters and Iron Concentration

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Abstract: Iron found in surface waters in small concentrations ($\text{ppm} = \text{mg L}^{-1}$) is considered as a nutrient, but above these concentrations it increases the eutrophication phenomenon by algal mass development, generating unpleasant odours and taste changing in drinking waters. For water treatment plants is essential to know the concentration of iron in water. Existence of some mathematical models to express statistical connection between iron concentration and water parameters can prevent the occurrence of unwanted effects. The purpose of this work was to develop a series of mathematical models in order to describe the relation between waters pH, turbidity, hardness, electrical conductivity and concentrations of iron presents in surface waters. For this purpose the multiple non-linear correlation method was used. The required database was created by daily monitoring, during the year 2011, of iron concentration and physico-chemical parameters of Danube River, in the upstream of Drobeta Turnu Severin town. This area is used for town water supply. Correlation coefficients of mathematical models that describe the relation between system variables have been shown that between total iron concentration and turbidity there is a determinant connexion.

Keywords: iron, Danube, turbidity, multiple correlation.

1. Introduction

Iron is an essential element for human and aquatic organisms. Iron found in surface waters at the ppb to ppm concentrations is considered as a nutrient [1], but above these concentrations it increases the eutrophication phenomenon by algal mass development. Excessive growing of algal mass has negative effect on water used for drinking.

The acceptable level of iron in drinking water is 0.2 mg L^{-1} [2]. Over this limit, iron can significantly change the taste and colour of water and in higher doses may even be poisonous.

Given the fact that Danube River, the second largest and important in Europe, is used on some sections as source of water supply, it is necessary to know the variation tendency of iron concentration in water when the physico-chemical parameters of water are changing [3].

In the present work, the correlation of total iron concentration from Danube River (Romanian sector – upstream Drobeta Turnu Severin town) and waters' pH, turbidity (NTU), hardness (H) and electrical conductivity (EC) was studied.

For this purpose the multiple non-linear correlation method was used.

Given the fact that on a lower pH of water there is a growth of the mobility of heavy metal deposited in watercourse sediments [4, 5] and the heavy metals concentrations increase in water [6] a mathematical functions have been proposed, in which iron concentration

was considered as dependent variable and the others parameters as independent variables.

Development of a mathematical model based on statistical processing of experimental data allows the determination of mutual dependence on the variables [7].

Given the dynamic and exchange regime of Danube River and the fact that even one relation between the considered variables is nonlinear, nonlinear multiple correlation method was used [8].

The empirical nonlinear models used for describing the complex processes give better results than the linear [7].

To determine how close is the relation between iron concentration and water parameters, mathematical function of third-degree with 3 parameters and second degree with 4 and 5 parameters were proposed

The general form of equations used was [8, 9]:

$$Y(x_i) = a_0 + \sum_{i=1}^n a_{1i} x_i + \sum_{i=1}^n a_{2i} x_i^2 + \sum_{j=1}^{n-1} \sum_{i=1}^{n-j} b_{ji} x_j x_{i+j} \quad (1)$$

were: Y – computed total iron concentration in water; x_i – physico-chemical parameters of water.

The coefficients of the regression function (a_0 , a_{1i} , a_{2i} , b_{ji}) were calculated by least squares method, so that the deviations dispersion of experimental values (y) towards the calculated values (Y) by means of regression equation (1) to be minimal:

$$M(f) = M[y - Y(x_i)]^2 = \min \quad (2)$$

$$f(a_0, a_{1i}, a_{2i}, b_{ji}) = [y - a_0 - \sum_{i=1}^n a_{1i} x_i - \sum_{i=1}^n a_{2i} x_i^2 - \sum_{j=1}^{n-m-j} \sum_{i=1}^n b_{ji} x_i x_{i+j}]^2 \quad (3)$$

were: y - dependent parameter: total iron concentration in water; x - independent parameters: $x_1 = \text{pH} / \text{pH units}$, $x_2 = \text{NTU} / \text{mg SiO}_2 \text{ l}^{-1}$, $x_3 = \text{H} / ^\circ\text{Ge}$, $x_4 = \text{EC} / \mu\text{S cm}^{-1}$.

Taking the derivative of equation (3) with respect to each regression coefficients and setting them to zero it is possible to determine the values of regression coefficients.

a. Third degree correlations functions with 3 parameters [8, 10]:

$$Y(x_1, x_2) = a_0 + a_1 x_1 + a_2 x_2 + a_3 x_1^2 + a_4 x_2^2 + a_5 x_1 x_2 + a_6 x_1^3 + a_7 x_2^3 + a_8 x_1^2 x_2 + a_9 x_1 x_2^2 \quad (4)$$

b. Second degree correlations functions with 4 parameters:

$$Y(x_1, x_2, x_3) = a_0 + a_1 x_1 + a_2 x_2 + a_3 x_3 + a_4 x_1^2 + a_5 x_2^2 + a_6 x_3^2 + a_7 x_1 x_2 + a_8 x_2 x_3 + a_9 x_3 x_1 \quad (5)$$

c. Second degree correlations functions with 5 parameters:

$$Y(x_1, x_2, x_3, x_4) = a_0 + a_1 x_1 + a_2 x_2 + a_3 x_3 + a_4 x_4 + a_5 x_1^2 + a_6 x_2^2 + a_7 x_3^2 + a_8 x_4^2 + a_9 x_1 x_2 + a_{10} x_2 x_3 + a_{11} x_3 x_4 + a_{12} x_4 x_1 + a_{13} x_1 x_3 + a_{14} x_2 x_4 \quad (6)$$

After processing the experimental data, the following indicators of model adequacy were calculated [9]:

- standard deviation of measured values y from the Y values calculated using the regression equation, which can be used to calculate the confidence interval for each individual value y :

$$\sigma^2 = \sqrt{\frac{\sum_{i=1}^n (y_i - Y_i)^2}{n}} \quad (7)$$

- multiple correlation coefficient:

$$R = \sqrt{1 - \frac{\sum_{i=1}^n (y_i - Y_i)^2}{\sum_{i=1}^n (y_i - \bar{y})^2}} \quad (8)$$

were: y_i - experimental values; Y_i - calculated values

using the regression equation; n - number of data sets; \bar{y} - arithmetic mean of the experimental values.

2. Experimental

The experimental database was drawn by daily monitoring, during year 2011, of iron concentration in the Danube River waters and physico-chemical parameters of waters respectively.

Physico-chemical analyzes were performed in laboratories of water distribution operator from Drobeta Turnu Severin.

Total concentration of iron in water was measured as follow: 50 ml of analyzing sample was acidified with concentrated H_2SO_4 (97% concentration) up to $\text{pH} = 1$ and then subjected to successive rounds of boiling (in the presence of potassium persulfate) and cooling. In the end a solution of 1,10-phenanthroline was added. The absorbance of the red complex was measured with DR 2800 spectrophotometer, at a wavelength of 510 nm [11].

The pH was measured using a portable pHmeter type pH 340i according to SR ISO 10523/2009. An INOLAB 720 conductometer was used for electrical conductivity measurements according to SR EN 27888/1997.

Turbidity of the samples was established using a 2100N IS Turbidimeter. Turbidity units were expressed in $\text{mg SiO}_2 \text{ l}^{-1}$ [12].

Water hardness was measured by EDTA titrimetric method using SR ISO 6059/2008.

3. Results and Discussion

Laboratory results consist of 265 sets of values for all five water parameters monitored during the year 2011. These values were used to obtain the mathematical models presented in this work

The mean monthly values of these parameters are presented in Table 1.

TABLE 1. Mean monthly values of water parameters monitored during year 2011 [13]

Month	pH_{mv}^*	NTU_{mv} , $\text{mg SiO}_2 \text{ L}^{-1}$	H_{mv} , $^\circ\text{Ge}$	EC_{mv} , $\mu\text{S cm}^{-1}$	$(\text{C}_{\text{Fe}})_{\text{mv}}$, mg L^{-1}
January	7.95	32.6	11.6	462.4	1.002
February	8.01	10.4	12.83	524.8	0.334
March	8.08	8.57	12.62	531.8	0.247
April	8.02	7.93	12.41	484.3	0.220
May	7.86	4.75	11.93	457.2	0.133
June	7.79	4.84	11.01	403	0.140
July	7.86	4.07	10.33	393.4	0.113
August	7.93	4.61	9.984	381.3	0.127
September	8	3.09	9.942	394.1	0.081
October	7.92	2.86	11.01	425.6	0.078
November	7.96	1.9	10.19	432.9	0.056
December	7.99	1.43	11.65	498.6	0.039

*mv - mean value

Two softwares were used for experimental data processing: Matlab 7.0 for correlations functions with 3 parameters and MathCad for correlation functions with 4 and 5 parameters.

In order to describe the relationship between the values of iron concentration in water and two physico-chemical parameters of water, multiple nonlinear correlations functions were generated. These functions were materialized by polynomial equations of second degree (b) and third degree (c). To make a comparison with steady state flow regime, multiple linear correlation functions (a) have been proposed also [14]. All these functions were presented in Table 2.

TABLE 2. Correlation functions with 3 parameters

Type of variation	Mathematical correlation functions	σ^2	R
$C_{Fe} = f(H, NTU)$	(a) $C_{Fe} = -4.6989 - 0.1582 \cdot H + 30.3611 \cdot NTU$	56.6214	0.9792
	(b) $C_{Fe} = -45.3805 - 0.0990 \cdot H + 48.8607 \cdot NTU - 1.1658 \cdot H \cdot NTU + 0.1606 \cdot H^2 - 0.1310 \cdot NTU^2$	54.4780	0.9808
	(c) $C_{Fe} = -1.1052e+003 + 305.9018 \cdot H + 14.4081 \cdot NTU + 3.0872 \cdot H \cdot NTU - 28.4531 \cdot H^2 + 0.5145 \cdot NTU^2 + 0.8905 \cdot H^2 - 0.0088 \cdot NTU^3 - 0.1874 \cdot H^2 \cdot NTU - 0.0024 \cdot H \cdot NTU^2$	52.3327	0.9823
$C_{Fe} = f(pH, NTU)$	(a) $C_{Fe} = -348.2983 + 43.0721 \cdot pH + 30.3263 \cdot NTU$	56.4650	0.9793
	(b) $C_{Fe} = -8.3303e+003 + 2.1946e+003 \cdot pH - 156.6682 \cdot NTU + 23.9887 \cdot pH \cdot NTU - 144.5603 \cdot pH^2 - 0.0981 \cdot NTU^2$	54.0546	0.9811
	(c) $C_{Fe} = -7.5135e+005 + 2.6755e+005 \cdot pH + 2.5488e+004 \cdot NTU - 6.3786e+003 \cdot pH \cdot NTU - 3.1651e+004 \cdot pH^2 - 7.7593 \cdot NTU^2 + 1.2435e+003 \cdot pH^3 - 0.0054 \cdot NTU^3 + 399.4719 \cdot pH^2 \cdot NTU + 1.0142 \cdot pH \cdot NTU^2$	49.8273	0.9840
$C_{Fe} = f(EC, NTU)$	(a) $C_{Fe} = -40.5192 + 0.0772 \cdot EC + 30.2905 \cdot NTU$	56.4742	0.9793
	(b) $C_{Fe} = -170.7945 + 0.4449 \cdot EC + 50.0606 \cdot NTU - 0.0328 \cdot EC \cdot NTU - 2.5986e-004 \cdot EC^2 - 0.1225 \cdot NTU^2$	55.0369	0.9804
	(c) $C_{Fe} = 2.6693e+003 - 15.3939 \cdot EC - 126.9107 \cdot NTU + 0.5535 \cdot EC \cdot NTU + 0.0295 \cdot EC^2 + 2.5142 \cdot NTU^2 - 1.8787e-005 \cdot EC^3 - 0.0099 \cdot NTU^3 - 4.8793e-004 \cdot EC^2 \cdot NTU - 0.0043 \cdot EC \cdot NTU^2$	51.8466	0.9826

In figures 1 to 6 are presented the experimental data and the surfaces generated by the functions from Table 2.

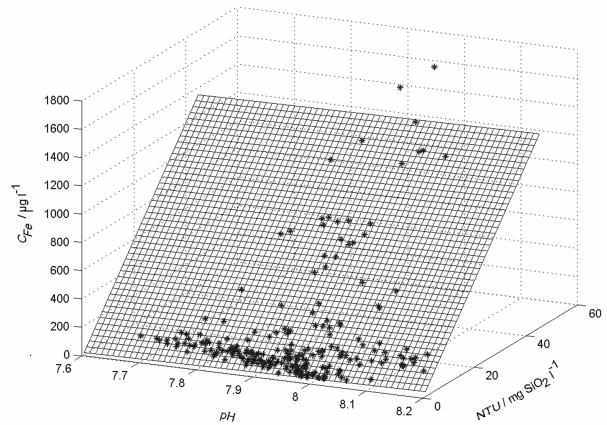


Figure 1. Variation of iron concentration with water pH and turbidity – linear equation

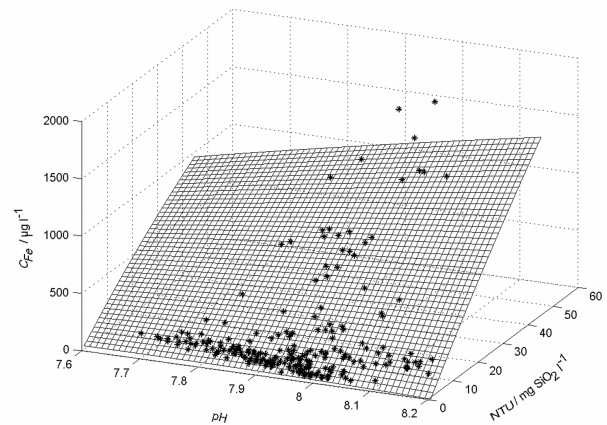


Figure 2. Variation of iron concentration with water pH and turbidity – second degree polynomial equation

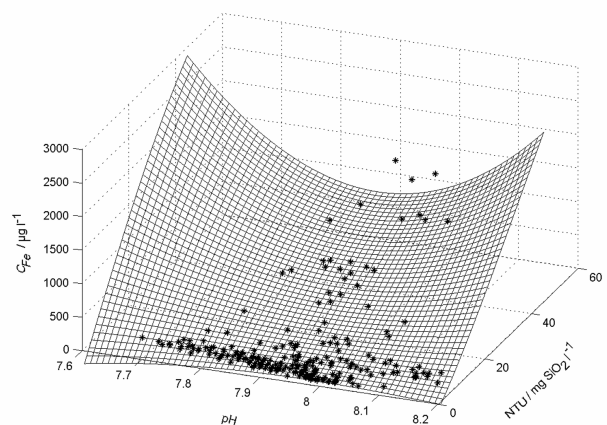


Figure 3. Variation of iron concentration with water pH and turbidity – third degree polynomial equation

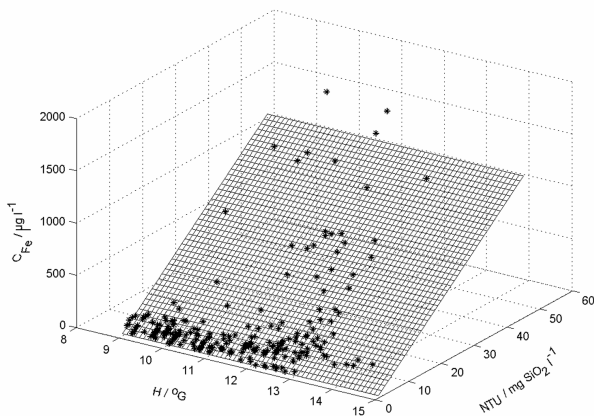


Figure 4. Variation of iron concentration with water hardness and turbidity – linear equation

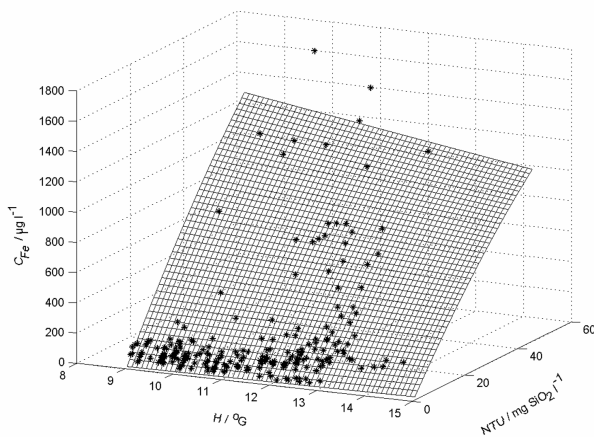


Figure 5. Variation of iron concentration with water hardness and turbidity – second degree polynomial equation

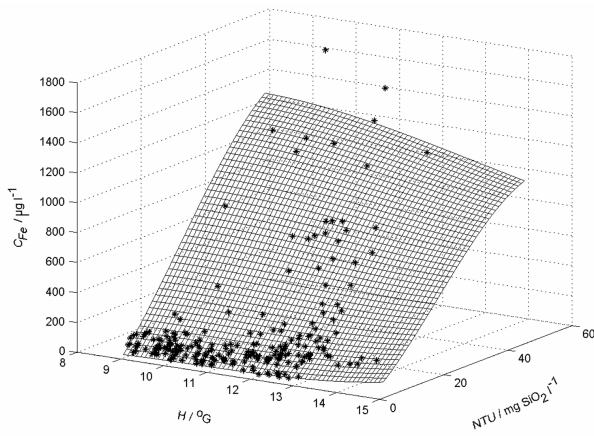


Figure 6. Variation of iron concentration with water hardness and turbidity – third degree polynomial equation

The second degree polynomial functions that describe the relationship between the values of iron concentration in water and three physico-chemical parameters of water are presented in Table 3.

TABLE 3. Correlation functions with 4 parameters

Type of variation	Mathematical correlation functions	σ^2	R
$C_{Fe} = f(\text{pH}, \text{NTU}, \text{H})$	$C_{Fe} = -5.096 \cdot 10^3 + 1.782 \cdot 10^3 \cdot \text{pH} - 277.928 \cdot \text{NTU} - 209.343 \cdot \text{H} - 147.277 \cdot \text{pH}^2 - 0.396 \cdot \text{NTU}^2 + 2.337 \cdot 10^3 \cdot \text{H}^2 + 45.005 \cdot \text{pH} \cdot \text{NTU} - 3.656 \cdot \text{NTU} \cdot \text{H} + 28.496 \cdot \text{pH} \cdot \text{H}$	25.21	0.964
$C_{Fe} = f(\text{pH}, \text{NTU}, \text{EC})$	$C_{Fe} = -1.145 \cdot 10^3 + 637.318 \cdot \text{pH} - 207.721 \cdot \text{NTU} - 3.328 \cdot \text{EC} - 59.898 \cdot \text{pH}^2 - 0.448 \cdot \text{NTU}^2 + 1.154 \cdot 10^3 \cdot \text{EC}^2 + 32.665 \cdot \text{pH} \cdot \text{NTU} - 0.032 \cdot \text{NTU} \cdot \text{EC} + 0.303 \cdot \text{pH} \cdot \text{EC}$	26.89	0.959
$C_{Fe} = f(\text{NTU}, \text{H}, \text{EC})$	$C_{Fe} = 133.525 + 43.615 \cdot \text{NTU} + 9.562 \cdot \text{H} - 1.029 \cdot \text{EC} - 0.695 \cdot \text{NTU} - 1.972 \cdot 10^3 \cdot \text{H}^2 + 6.096 \cdot 10^4 \cdot \text{EC}^2 - 2.811 \cdot \text{NTU} \cdot \text{H} + 8.98 \cdot 10^3 \cdot \text{H} \cdot \text{EC} + 0.067 \cdot \text{NTU} \cdot \text{EC}$	25.82	0.962

In order to describe the relationship between iron concentration and measured physico-chemical parameters of water (pH, NTU, H and EC) a second degree polynomial function was generated. It is presented in Table 4.

TABLE 4. Correlation function with 5 parameters

Type of variation	Mathematical correlation functions	σ^2	R
$C_{Fe} = f(\text{pH}, \text{NTU}, \text{H}, \text{EC})$	$C_{Fe} = 0.083 + 3.845 \cdot 10^{-6} \cdot \text{pH} - 65.909 \cdot \text{NTU} + 61.74 \cdot \text{H} + 1.169 \cdot \text{EC} + 0.571 \cdot \text{pH}^2 - 0.712 \cdot \text{NTU}^2 - 0.436 \cdot \text{H}^2 + 1.385 \cdot 10^{-3} \cdot \text{EC}^2 + 14.186 \cdot \text{NTU} \cdot \text{pH} - 0.855 \cdot \text{H} \cdot \text{NTU} - 0.01 \cdot \text{H} \cdot \text{EC} - 0.277 \cdot \text{pH} \cdot \text{EC} - 5.571 \cdot \text{pH} \cdot \text{H} + 0.011 \cdot \text{NTU} \cdot \text{EC}$	32.7	0.954

After determination of model equations, a comparison between model predictions and experimental data is necessary to make. This validation was made for all mathematical models presented in this paper. The data set used to validate the mathematical model that describe the variation of iron concentration with water pH and turbidity (third degree polynomial equation) and the values of model adequacy indicators are presented in Table 5.

TABLE 5. Experimental data used to validate the mathematical model that describe the variation of iron concentration with water pH and turbidity and the values of model adequacy indicators

Month	pH	NTU	Exp. $C_{Fe}, \mu\text{g L}^{-1}$	Calc. $C_{Fe}, \mu\text{g L}^{-1}$	σ^2	R
January	7.91	27.40	830.0000	835.0023	4.719	0.9998
February	7.85	22.10	740.0000	732.4819		
March	8.14	5.28	150.0000	153.1431		
April	8.06	7.66	220.0000	223.7788		
May	7.82	4.79	144.0000	136.7781		
June	7.75	4.35	112.0000	109.5664		
July	7.90	3.15	88.0000	88.6918		
August	7.95	3.90	107.0000	110.8868		
September	8.04	3.04	80.0000	81.2773		
October	7.88	2.17	50.0000	56.9017		
November	8.01	1.27	40.0000	36.7594		
December	8.02	1.28	38.0000	35.5873		

The values of model adequacy indicators allow its validation, demonstrating that it accurately reflects the real phenomenon.

Analyzing the graphs from figures 1 to 6 it can be noticed that the shape of surfaces generated by the correlation equations are not so different from a linear surface. The mathematical functions generated by multiple correlations are mathematical models. For the studied situations, a mathematical model is appropriate if it describe very well the relationship between iron concentration and the considered physico-chemical parameters of water. On the other side, a mathematical model shouldn't have a high degree of complexity [15].

Correlating the values of model adequacy indicators from Tables 2, 3 and 4 with the number of independent variable considered in the mathematical model, it can be noticed that increasing the number of variables leads to lower values of adequacy indicators. This is normal because the complexity of the model increase with increasing the number of independent variables taken into consideration.

Analyzing the correlation coefficients presented in Tables 2, 3 and 4 we can see that their value exceeds 0.95, in all the cases. This means that there are strong connections between the dependent and independent variables.

Simultaneously, it should be noted that, the turbidity parameter (NTU) is present in all the considered functions.

Without the turbidity factor, the correlation coefficients indicates a weak connection between the total concentration of iron in water and the other physico-chemical parameters (the coefficients have values between 0.437 for $Fe = f(H, EC)$ and 0.53 for $Fe = f(pH, H)$ and $Fe = f(pH, EC)$). These last values can lead us to the conclusion that between water turbidity and iron concentration in water there is a determinant connection.

Highest values of multiple correlation coefficients were obtained for mathematical functions with 3 parameters, presented in Tabel II. These functions will be discussed and analyzed.

A) Fe correlation with pH and NTU

Analyzing the mathematical functions obtained is seen that the best R correlation coefficient was obtained for the third degree polynomial function (c)

The pH coefficient ($2.6755e+005$) and turbidity ($2.5488e+004$) from these function reflect the strong connection between Fe, pH and NTU.

The explanation of natural phenomena must take into account that the turbidity is due to the colloidal organic and inorganic matter from water, not settles in time [16].

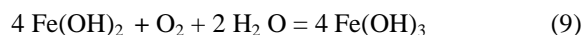
Fe in water is found both as free species Fe^{+2} and especially in the form of chelates with organic compounds.

Fe present in water as free species, according to a slightly alkaline pH such as the Danube, contribute to naturally formation of iron hydroxides and oxo-hydroxides [4].

This hydroxides and the heavy metal hydroxides from natural waters are generally in the form of gelatinous, fine

precipitates like $Zn(OH)_2$, $Mn(OH)_2$ or flocculent precipitate such as $Fe(OH)_2$ [17, 18].

In the presence of oxygen in water $Fe(OH)_2$ leads to formation of trivalent iron hydroxide $Fe(OH)_3$, a precipitate with deposit tendency [17].



But in surface waters with strong eddy currents like Danube river, precipitate formed is prevented from filing. Therefore, the precipitates of iron hydroxides remain in suspension, leading to increased water turbidity.

At the same time, in natural waters Fe is also found in the form of chelates with organic compounds, of which humic acids have significant weight. The humic acids contain predominantly COOH and OH groups, able to form complexes with Fe ions [19].

At low pH (pH = 4.5) these iron complexes could coagulate and sediment [20].

But in the slightly alkaline pH conditions encountered in natural water, coagulation and sedimentation of iron compounds does not occur. They remain in waterbody leading to increased turbidity.

Therefore, increased concentrations of iron in water at high pH, contributes to increased water turbidity, as seen in the graphic representations of Figures 1, 2 and 3.

B) Fe correlation with hardness (H) and NTU

After analyzing and comparing the adequacy indicators of the three mathematical functions identified, is seen that the third degree polynomial function (c) best describes the correlation between Fe, hardness and turbidity.

The values of hardness coefficient (+305.9018) and turbidity coefficient (+14.4081) within this function, and also the graphical representation from figure 6 shows an increase in Fe concentrations while increasing hardness and turbidity.

The explanation of natural phenomena must take into account that the hardness is due to the carbonates of Ca and Mg in water.

In the presence of iron salts in water, Ca and Mg carbonates forms ferrous carbonate $FeCO_3$, a white precipitate with amorphous structure [17].

Given the dynamic regime of water flow rate and the conditions of pH and temperature, these amorphous precipitates may lead to increased turbidity.

C) Fe correlation with conductivity (EC) and NTU

Analyzing the mathematical functions obtained for this correlation is seen that the third degree polynomial functions best describe the relationship between Fe, conductivity and turbidity.

Analyzing the values of conductivity coefficient (-15.3939) and turbidity coefficient (-126.9107) within this function, also can observe the inverse variation of Fe and the two studied parameters.

The explanation of real phenomena must take into account that under a slightly alkaline pH, electrical conductivity of water is due to an excess of OH-ions and alkali metal.

Metal ions are essential components of natural waters. Depending on the aquatic conditions (pH, oxidation-reduction potential, and presence of ligands) they enter into the composition of various inorganic and organo-metallic compounds [21].

The phenomenon of incorporating these metals in oxy-hydroxides of iron present in the water leads to the coprecipitation and accumulation in sediments [4].

Therefore, an increase in the phenomenon of sedimentation causes a decrease in turbidity.

4. Conclusions

This work was studied the relationship between physico-chemical parameters of water and concentration of Fe in surface waters, using multiple linear correlation functions.

Analyzing mathematical functions identified was observed that between Fe, pH and turbidity there is a strong relationship

Validation with experimental data of a mathematical model describing the relationship between Fe, pH and turbidity proved accurate real phenomenon.

The mathematical models developed can be used for calculate iron concentration knowing the others parameters of water.

This can be very useful in the laboratory of water treatment plants because can be compared the measured values of Fe concentrations with the calculated value, based on mathematical models.

Also, based on mathematical models obtained can be estimated Fe concentration in water before the measurement is taken in the laboratory, which would allow early intervention on the technological process of drinking water and harm reduction.

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