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Analysis of the Influence of Climatic Factors on the Concentration of Iron and Manganese in Raw Water Intended for a Water Treatment System

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ABSTRACT

In surface water body sources intended for domestic system, iron and manganese concentrations has often been presented an increase in its values, it is recurrent during the warm seasons. As the chemical dynamic of water bodies is dependent on the environmental conditions, climatic aspects which affects temperature and dissolved oxygen in the water, will also affect the concentration and structure of these ions. Therefore, the present study aims to evaluate the influence of temperature and rainfall rates on the iron and manganese concentrations of the water source of the Water Treatment System – Terras Baixas, by using statistical methods, in a series of five years of data (2013 to 2017). The results of iron and manganese concentrations shows an increase during the hot seasons, and, through the statistical methods used, it was possible to conclude that, regarding temperature and rain factors, only temperature had a significant influence on these variables.

Keywords: WTS Terras Baixas, Water quality, statistical analysis, correlation, significance

Análise da Influência de Fatores Climáticos na Concentração de Ferro e Manganês em Água Bruta Destinada a um Sistema de Tratamento de Água

RESUMO

Nos mananciais superficiais destinados ao sistema doméstico, as concentrações de ferro e manganês têm apresentado, muitas vezes, um aumento em seus valores, sendo recorrente durante as estações quentes. Como a dinâmica química dos corpos d'água é dependente das condições ambientais, aspectos climáticos que afetam a temperatura e o oxigênio dissolvido na água, também afetarão a concentração e a estrutura desses íons. Portanto, o presente estudo tem como objetivo avaliar a influência da temperatura e dos índices pluviométricos nas concentrações de ferro e manganês do manancial do Sistema de Tratamento de Água – Terras Baixas, por meio de métodos estatísticos, em uma série de cinco anos de dados (2013 a 2017). Os resultados das concentrações de ferro e manganês mostram um aumento durante as estações quentes, e, através dos métodos estatísticos utilizados, foi possível concluir que, em relação aos fatores temperatura e chuva, apenas a temperatura teve influência significativa nessas variáveis.

Palavras-chave: ETA Terras Baixas, qualidade da água, análise estatística, correlação, significância

Introduction

Water is a natural resource of extreme importance for all forms of life. Despite that, water has undergone changes that compromise its

quality. The growing of urban areas, discharge of untreated effluents, increase of pesticides uses are among the main reasons of water quality degradation. Otherwise, there are many other

activities that cause direct or indirect damage to water bodies has affecting water properties. In addition, surface water bodies have a several processes influencing its quality (Santos et al. 2020; De Oliveira et al., 2014; Kumar 2015; Leite et al. 2019; Silva et al. 2017).

The presence of chemical substances in the water system can be of natural or anthropic origin. In past years, the increase of the anthropic activities has led to the intensification of chemical compounds production. When disposed in an incorrectly way, it causes environmental contamination (Antenor et al., 2020). Among the inorganic chemical substances which contaminate surface and groundwater reservoirs are the heavy metals. In Brazil, Consolidation Ordinance 5 from 2017, provides water potability standard, and establishes, among other parameters, maximum permissible values for the concentrations of iron and manganese (Brazil, 2017).

Iron and manganese are naturally found in water in a dissolved form, respectively Fe^{+2} and Mn^{+2} , in normal pH ranges. In the presence of a oxidant, these ions are oxidized and produces insoluble products, namely, iron hydroxide ($\text{Fe}(\text{OH})_3$) and manganese dioxide (MnO_2). If the oxygen concentration in the water is sufficient, the manganese will remain with an Mn^{+4} valence and insoluble. On the other hand, when the concentration of dissolved oxygen in water declines, bringing problems for the aerobic organisms present, and disturbing their existence, and triggering harmful process in their life cycle, the manganese is reduced to form Mn^{+2} , increasing its solubility in the water, becoming liable to be carried to the Water Treatment Plants. A similar process occurs with iron, which changes from its iron form to the ferrous ion Fe^{+2} , becoming soluble in water, thus changing the characteristics of raw water (Nascimento, Monte, Correa, 2021; De Almeida et al., 2019; Moruzzi and Reali, 2012; Wallhroos, Ferreira, 2008).

Soluble metals, such as iron and manganese, commonly occurs during the warm season, presenting increased concentrations because it might be related to oxygen concentration depletion in the water body (De Almeida et al., 2019; Gantzer et al., 2009). Precipitation in turn can also exert an influence on the water body quality. In the surface waters, the levels of iron and manganese increase in the rainy seasons due to the soil carry-over and the occurrence of bank erosion process, as highlighted in the study by Marcelino et al. (2017), which emphasizes that iron and

manganese can result from water and soil interaction.

However, other studies states that during the dry period, higher concentrations of metal ions can be verified due to the reduction in the volume of water, because the balance of their concentrations is greater by volume of water (Pivelli, 2012). Corroborating this idea, Baharim et al. (2011) states that heavy rains can cause an impact in the decrease in the total concentrations of iron and manganese as they can play a significant role in the natural de-stratification of the reservoir, in which the concentration of dissolved oxygen in the bottom water increases again, resulting in a decrease in the concentration of iron and manganese in the lower portions of the water body. Furthermore, manganese can be found in different states of oxidation, but Mn^{+2} is more commonly soluble in natural waters (Ghazi and Qomi, 2015). Yet, iron is generally found under two states insoluble form as Fe^{3+} , and soluble form as Fe^{2+} (Marsidi et al., 2018).

In WTS the variations of the iron and manganese concentrations at water abstraction points are a challenge, because when these ions in the reduced form are conducted to the treatment stages, they usually pass for oxidation stage as treatment step, changing the state of oxidation of these compounds. When oxidized, iron and manganese form precipitates, which, if are not removed, they will be noticeable above the maximum permissible values, which are 0,3 and 0,1 mg/L, for iron and manganese, respectively (Brazil, 2017), providing color, odor, and taste to water (Narciso et al., 2014; Libânio, 2005; Parron et al., 2011). Besides, it causes stains on clothes and floor, which probably might be rejected by the consumers. Furthermore, it also affects operationally the WTS, by reducing the efficiency of the filters and providing problems of incrustation.

Considering the context described, evaluation of the water sources characteristics is necessary to support the decision-making process regarding treatment of water systems. In this context, one of the problems faced in monitoring water quality is the complexity in analyzing and interpreting a large number of variables (Isçen et al., 2008). According to Valentini et al. (2021a), an important tool that has been widely used to circumvent this complexity is statistical analysis. Statistical methods have the advantage of providing a simple interpretation for a complex and otherwise difficult to interpret data set, as well as greater accuracy and facilitate of data

manipulation, making the application of advanced statistical methods of great use to extract significant information without losing its accuracy (Hair, 2009; Zhao et al., 2012; Zhao et al., 2011).

The statistical methods which may be applied for analysis and monitoring of water quality are very diverse and cover different types of approaches, as reported by the authors Drose et al. (2020), Fraga et al. (2021), Valentini et al. (2020), Valentini et al. (2021b), Varol (2020), Santos et al. (2020), Santos et al. (2021), Silveira et al. (2021), among others. Correlation methods, for instance, provides a good idea of how the parameters used for assessment of water quality are correlated with each other or to assess how other factors can influence in chemical, physical, and microbiological variables used to assess water quality. Variance analysis is another statistical method widely used in the mentioned studies. This analysis is used to assess whether the analyzed parameters vary significantly, or whether the variation of a given factor, such as temperature or rainfall, for example, causes significant variation in the data studied (BILGIN, 2015). Valentini et al. (2021c), for example, used this analysis to assess whether the variation in rainfall intensity or the variation in the tide provides a significant variation on the bathing conditions of the beaches in Itapoá-SC city.

Furthermore, regression analysis is widely used to analyze data set related to water quality.

Valentini et al. (2021d), for example, used multiple linear regression analysis to model a new equation to calculate a new water quality index for Lagoa Mirim, taking into account only the variables that most correlated with the original water quality index of this water resource. The present study investigates the concentration of manganese and iron of raw water that supplies Terras Baixas water treatment system (WTS), water treatment supply mentioned in studies such as Franz et al. (2020), by using same techniques described in the study by Valentini et al. (2021d)

Therefore, this study aims to evaluate the influence of temperature and rainfall rates on the concentrations of iron and manganese in the raw water of the Terras Baixas Water Treatment System, by using of statistical methods that aim to establish a possible relationship between the interfering climatic agents and the variation of the water chemical parameters analyzed.

Methods

Study Area

The study area took place in the municipality of Capão do Leão (RS), more specifically, at the Terras Baixas Water Treatment System (TB-WTS), which is used in the treatment of raw fresh water from the Arroio Padre Doutor (Figure 1).

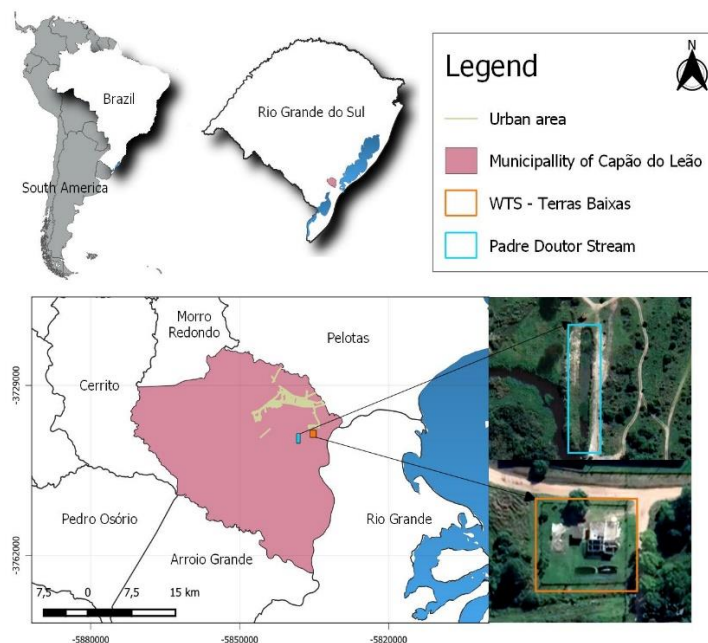


Figure 1: Location of the study area. Coordinate system: SIRGAS 2000. Projection system: UTM 22S. Sources: Google Satellite and IBGE (2010).

The TB-WTS in question is administered by the Federal University of Pelotas (UFPEL) together with the company Embrapa Clima Temperado. It is system with drinking water, through conventional treatment by complete cycle for the Capão do Leão campus of UFPEL, the Embrapa-Estação Terras Baixas, and a community village around (houses, a church, and elementary school), the actual flow rate of treatment is 23m³/h (Franz et al., 2020).

The Padre Doutor water body is localized in the Mirim – São Gonçalo catchment, and according to annual environmental quality report of the municipality of Pelotas there is no resolutions which approves the classification of water related to Mirim – São Gonçalo catchment yet. Thereby, according to article 42 of resolution of CONAMA 357/2005, it is established that while the respective classifications of water body have not been approved, freshwater must be considered as class 2 (Brazil, 2005).

It is also worth to mention that Padre Doutor water body flows through urban, industrial, and rural zones, and has a great importance in the economic and social aspects of the region. This water body is used for system, recreation, industrial activity, irrigation, and dilution of domestic and industrial sewage dumps (Churiara, 2007; Churiar et al., 2012).

Chemical and climatic analysis

For the present study, monitoring data for the following parameters were considered, shown in table 1, which characterize and interfere in the source that feeds the WTS - Terras Baixas.

Table 1: WTS monitoring parameters – Terras Baixas, used.

Parameter	Unity
Iron - Fe	mg/l
Manganese - Mn	mg/l
Temperature	°C
Rainfall	mm

The results of the monitored chemical parameters were made available by the technical responsible of Terras Baixas - WTS, therefore it is secondary data, and correspond to collections carried out weekly for 5 years corresponding to the period from 2013 to 2017, sampling that generated large detailed of data in overtime, which might be useful for analyze the iron and manganese variability in monitoring water quality for water resources management (Fraga et al., 2020).

The chosen of such parameter (Fe and Mn) for this study was since floodplain soils of Rio Grande do Sul (state which WTS – Terras Baixas is located) are originated from different materials (basalt, sandstone, siltstone, and granite), which contribute to variation in the total content of Fe and Mn oxides (Gonçalves et al. 2011). Moreover, according to Davis and Masten (2016) the presence of these substances in water intended for domestic supply are very common in Brazil, both in soluble and ionic forms.

In the statistical analyzes were used the monthly average of the studied parameters. Whereas the results of the meteorological variables, temperature, and rainfall rates, were obtained through the data available on the portal Weather Research and Climate Studies Center from Brazilian National Institute of Space Research for the Capão do Leão weather station corresponding to the period from 2013 to 2017.

Statistical analysis

Normality test

In order to analyze the sample distribution of the data in this study, we proceeded with the Kolmogorov-Smirnov (K-S) and Shapiro-Wilk (S-W) tests at a significance level of 0.05. For these tests, the null hypothesis (H0) considers the distribution to be normal and for p-value <0.05 this hypothesis is rejected.

Analysis of variance

Certain statistical tests can infer if the variation in one parameter causes significant variation in another or if it does not cause a statistically noticeable effect. Thus, with the objective of verifying if meteorological variables (temperature and rainfall) cause significant variation in the chemical variables (iron and manganese) studied, the significant test was carried out. For these analyzes, for p-value <0.05 the null hypothesis will be rejected, resulting in a significant variation (Pinto et al., 2020).

Different tests do this type of analysis, but the standard mechanism is the same, the significant test of null hypothesis. According to Ellenberg (2015), this method has been used to evaluate results of scientific research for decades. In this type of test, a null hypothesis (H0) is evaluated, in order to retain or reject it. One of the most commonly limits used by researchers for this purpose is 0.05. Thus, null hypothesis must be rejected if there is less than a 5% chance of finding a result which consider H0 true. To this limit, name

the level of significance and it represents the upper limit of the probability of observing an event if the null hypothesis is true (Wheelan, 2016).

Correlation analysis

Further, in order to assess the correlation between chemical variables with temperature and rainfall variables, a correlation matrix with its coefficients and significance p-value was used, with the coefficient to be used depending on the data distribution. For data that follows a normal distribution, for example, Pearson's coefficient usually is the most used, whereas the Spearman Rhô coefficient, being the coefficient used in non-parametric correlation analyzes, is more suitable for data that do not have sample normality (Guimarães, 2017).

As for the intensity of the correlation, the threshold defined by Helena (2000) will be used, which defines as intense the correlations with coefficients equal to or greater than 0.5 in module. Furthermore, the correlations that obtain a p-value <0.05 , regardless of whether they are intense or not, are considered significant.

Regression analysis

Regression analysis allows to assess the relationship that specific variables have with a result of interest. This analysis typically uses a methodology called ordinary least squares (OLS). Through the OLS method, the linear regression analysis traces a line with a more accurate fit in the center of the data distribution, minimizing its quadratic residuals (Wheelan, 2016).

Regression models for the variables iron and manganese will be evaluated, according to the correlations obtained in the correlation matrix. In these models, iron and manganese will be the dependent variables and temperature and rainfall will be the predictors, also called independent variables. Subsequently, the model equation that presents the best adjustments will be extracted.

Results

Normality test

The chemical parameters, as well as the temperature and the pluviometry, were evaluated regarding their sampling distribution. The results of these tests can be seen in Table 2. According to the K-S and S-W tests, only the temperature and rain variables follow a normal distribution, while iron and manganese obtained p-value <0.05 demonstrating that the sampling of these parameters does not follow a normal distribution.

Therefore, for the analysis of variation and the correlation matrix, non-parametric tests will be used. Several studies, such as Cardoso et al. (2021), Valentini et al. (2021a), Valentini et al. (2021e), Valentini et al. (2021f) and Santos et al. (2020), have also used normality tests to assess the fit of the data set for correlation and variance analysis which were used in their studies, corroborating the importance of sample distribution assessment before the application in a certain statistical analyzes.

Table 2: Evaluation of the sample distribution - Kolmogorov-Smirnov (K-S) and Shapiro-Wilk (S-W) tests

	p-value	
	K-S	S-W
Iron	0,000	0,000
Manganese	0,005	0,000
Temperature	0,200	0,129
Rainfall	0,200	0,092

Analysis of variance

Regarding the variation of chemical variables in relation to the meteorological variables, their significance was assessed by the Kruskal-Wallis non-parametric test, and its results can be seen in Table 3. As shown, the temperature variable obtained p-value <0.05 for the variables Fe and Mn. In other words, the temperature variation causes a significant variability in the concentrations of these chemical elements in the water. Whereas the rainfall variable did not have a statistically distinguishable effect, with a significance level of 0.05, in relation to chemical variables. The significant variation of Fe and Mn as a function of temperature variation might be explained by the fact that, according to Almeida et al. (2019), there is a relationship between these metals presence in a soluble form and dissolved oxygen concentration in the water.

It is worth noting that the temperature analyzed in the present study was the ambient temperature, which has higher amplitude than the water of surface water bodies, which are regulated by latitude, altitude, season, time of day, depth, and flow (CETESB, 2018). As for the Padre Doutor stream, the water body source addressed in this study, the localization, the sampling collection period, and the rainy season favored low thermal amplitude.

Further, the fact that rainfall does not cause significative influence on the metals concentrations reported in this study, but temperature may be due to localization of water abstraction point suffers

less influence of carrying of these substances through runoff caused by rainfall. Otherwise, the abstraction point was sidetracked from the original watercourse Padre Doutor stream, where the water is conducted by gravity until the abstraction point. At this point, water accumulates on the surface in a shallow water table, which vary according to weather conditions. Thus, this water accumulation provides a stationary characteristic, when comparing to original source, becoming more susceptible to thermal stratification effects which may significantly influence reservoirs water quality as reported by Noori et al. (2018). Therefore, the significant variation in Fe and Mn content as function of environmental temperature may be related to thermal stratification. Furthermore, Fe and Mn in neutral oxide

conditions typical in waters, are commonly present in sediments as insoluble forms (hydroxide and oxide minerals). However, under the reduction conditions, that commonly develop in thermally stratified reservoirs, the reductive dissolution of Mn and Fe hydroxides can release soluble metals in the water column (Munger et al., 2017).

It is known that, according to Mondal et al., (2015), the stratification of the water column due to temperature variation is a seasonal phenomenon in lakes and reservoirs. The study by Grande et al., (2015), on stratification of Metallic and Sulfate Loads, carried out in Dams in the Iberian Pyritic Belt in southwestern Spain, showed that Fe and Mn concentrations increase in the deeper reservoir layer.

Table 3: Analysis of variance - Kruskal test - Wallis

	p-value	
	Temperature	Rainfall
Iron	0,000	0,627
Manganese	0,001	0,814

Although according to Huang et al., (2014), Oliveira and Cunha (2014), Girarde et al., (2016), Ling et al., (2017) and Rostami et al., (2018), rainfall events influence water bodies, by changing their quality and hydrological conditions, in this study it did not have as much influence as temperature did. Still the study by Conceição et al. (2020) shows that, in surface waters, the iron concentration increases during the rainy seasons due to the carry-over of soils and the occurrence of erosion processes of the banks. On the other hand, studies like the one by Fraga et al. (2012), shows that precipitation was not the factor of greatest influence on the water quality of the source addressed in their study. Yet, it should be noted that the water blade at the point of abstraction of water

in the present study is shallow and, therefore, quickly sedimentation might occur, corroborating the non-significant variation of chemical variable in relation to pluviometry in this study.

Still, it should be noted that the analysis of significance presented here measures only if the variation was significant or not, it does not measure the intensity of that variation. The intensities of the variations given as significant (temperature-Fe and temperature-Mn) can be seen in Figure 2. As can be seen in this figure, there is indeed an increase in the concentration of the elements iron and manganese as the temperature increases, and this variation is intensified in the higher temperature ranges.

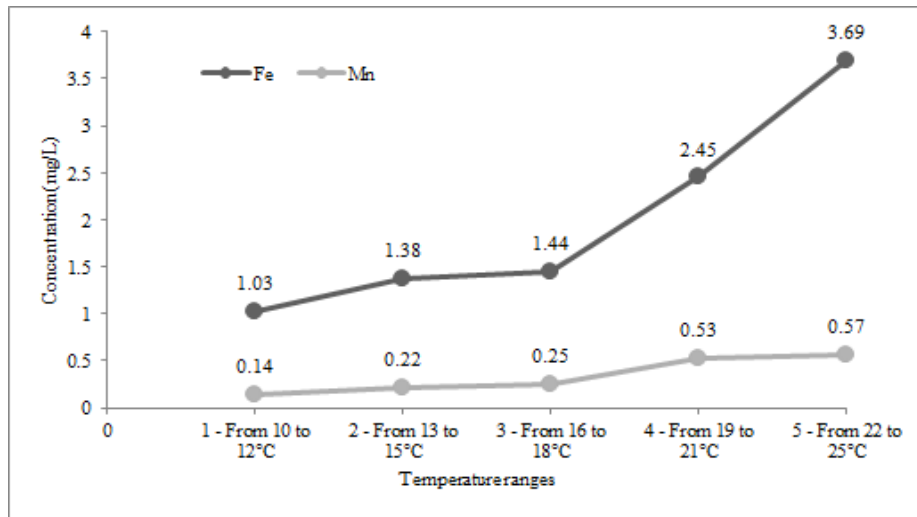


Figure 2: Variation of Fe and Mn concentrations in relation to temperature ranges.

Correlation matrix

Once the sampling distribution of the data has been evaluated, it follows with the correlation matrix with Spearman coefficients. This matrix, with its coefficients and p-values of significance, can be seen in Table 4. The focus is given on the correlations of iron and manganese with the variables temperature and rainfall. As defined, correlations with a Spearman coefficient greater than or equal to 0.5 in module will be considered strong. Nevertheless, a correlation can have an intensity coefficient of less than 0.5 and still be significant, that is, have a p-value <0.05.

As can be seen in Table 4, the temperature variable has a strong (0.819) and significant correlation with Fe, and a strong (0.605) and significant correlation with Mn. The rainfall variable did not show a distinguishable correlation with any of the studied parameters.

Table 4: Parameter - Temperature and Parameter correlation coefficients - Pluviometry and their respective significances

		Fe	Mn
Temperature	C.	0,819	0,605
	Sig.	0,000	0,000
Rainfall	C.	0,194	0,023
	Sig.	0,208	0,880

The results of the positive correlation between temperature and the Fe and Mn parameters presented in Table 4 highlight what was shown in Figure 2, i.e., they show that as higher temperature is higher will be Fe and Mn concentrations in the water. Different from the

study in question, CETESB (2017) and De Almeida et al. (2018) relate the increase of Fe and Mn parameters in water due to the increase of precipitation, which ends up carrying such elements through leaching from the soil to the water body. However, as far as this study is concerned, there was no strong correlation between rainfall and metal concentration, which, as previously discussed, may be related to the fact that the metals carried by rain to abstraction point of water analyzed here can be quickly precipitated due to the shallow depth of the water sheet.

The importance of statistical analyses such as correlation analysis for water quality monitoring is also worth mentioning. Several studies in Brazil and around the world have used these analyses in order to infer the processes that influence the water quality, helping to identify sources of pollution and to take measures to maintain the water quality (Bilgin, 2015; Kumar et al., 2016; Oliveira et al., 2019; Santos et al., 2020; Silveira et al., 2021; Valentini et al., 2021a; Valentini et al., 2021b; Wu et al., 2018).

Linear regression analysis

Having defined that only temperature has a strong and significant correlation with iron and manganese, regression models will be built with the temperature variable as an independent variable and Fe and Mn as dependent variables. That is, a Temperature-Mn regression model and another Temperature-Fe regression model, in order to assess what influence this variable actually has on the concentrations of these two metals in the water resource studied here. The summary of these models can be seen in Table 5.

Table 5: Regression analysis - summary of the models

Model	Dependent variable: manganese	Dependent variable: iron
1	Adjusted R square 0,279	Adjusted square 0,658
	Sig. 0,000	R 0,000
Predictors: (constant), Temperature		

As can be seen in Table 5, for both manganese and iron, there was significance in the models using temperature and a constant as predictors (independent variable). It is also observed that the Temperature-Mn regression model obtained an adjusted R^2 of 0.279 and the Temperature-Fe model, in turn, 0.658. According to Wheelan (2016), the R^2 , used here adjusted R^2 , is a measure of the total size of the variation that is explained by the regression equation of this model. That is, according to the models used, the temperature variable explains 27.9% of the Mn

concentration and 65.8% of the Fe concentration for the studied site.

To assess the fit of the models, it was proceeded with the analysis of the residual statistics of each model. The values of this analysis can be seen in Table 6. As observed in this table, the residuals and the predicted standard values are within a range of three standard deviations more or less for the Temperature-Fe model, corroborating the adequacy of the model. As for the Temperature-Mn model, there were residues more distant than the established one, indicating that the regression of this model was not satisfactory.

Table 6: Residual Statistics

	Model Temperature-Mn				Model Temperature-Fe			
	Minimum	Maximum	Average	Standard deviation	Minimum	Maximum	Average	Standard deviation
Predicted value	0,0778	0,6626	0,371	0,157	0,401	3,823	2,114	0,920
Residue	-0,470	1,046	0,000	0,243	-1,193	1,910	0,000	0,652
Default Predicted Value	-1,862	1,857	0,000	1,000	-1,862	1,857	0,000	1,000
Standard Residue	-1,913	4,258	0,000	0,988	-1,808	2,894	0,000	0,988

The histogram graphs of the standard residuals and the graphs of cumulative expected and observed probability are shown in the figures 3 and 4, which demonstrated the fit of the models. As can be seen in figures 3 and 4, the residuals of the model Temperature-Mn, in fact, depart to a

greater extent from the regression line. Whereas, for the Temperature-Fe model, on the other hand, it presented a satisfactory fit, corroborating the adequacy of this regression model.

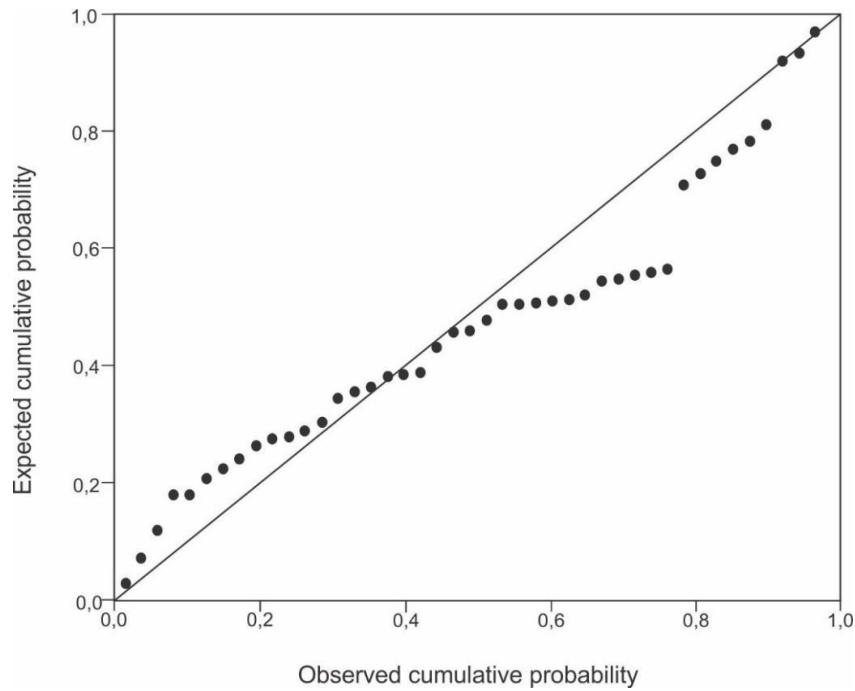


Figure 3: Residual distribution of temperature-Mn model.

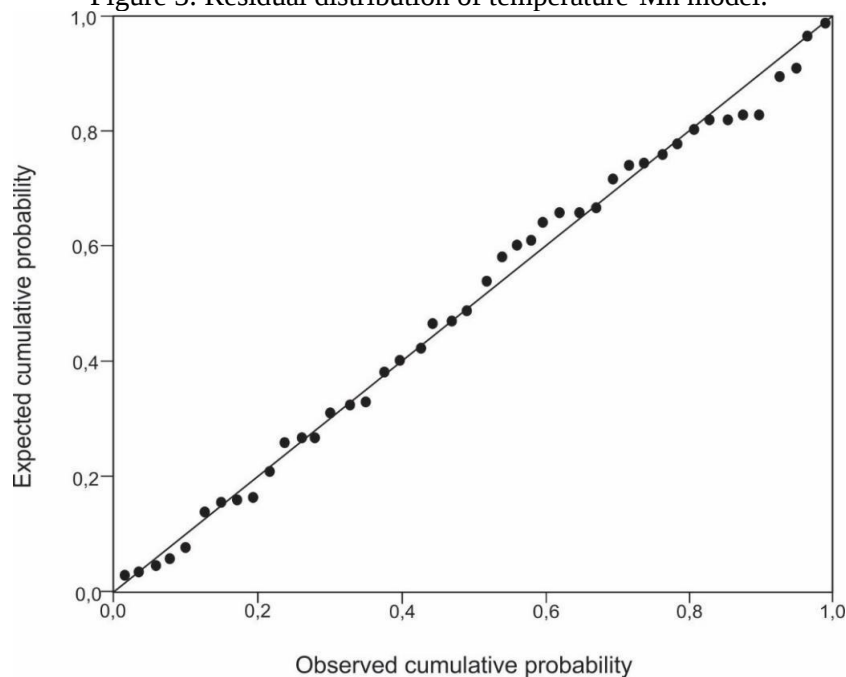


Figure 4: Residual distribution of temperature-Fe model.

Regression analysis has been widely used in several water quality and hydrological modelling studies. Valentini et al. (2021d) in their study regarding Lagoa Mirim watershed used linear regression analysis to model a new equation of WQI for this watershed, finding very satisfactory results and corroborating the use of this technique for water quality analysis. In this same approach, Santos et al. (2021), have also used linear regression analysis in their study concerning Moreira-Fragata stream water quality, emphasizing

the applicability and importance of this technique in water quality studies. Further, Cassalho et al. (2019) have made the use of linear regression analyses by using a potential model, for modelling regional equations to predict maximum flows for Rio Grande do Sul state. In worldwide, several other studies have made similar analysis mentioned in the present study, showing that statistical analysis as regression analyses (whether linear or potential, simple, or multiple) may be widely used in studies regarding water quality and hydrological

modelling, otherwise these analyzes introduces a significant improvement to the interpretation of the results analyzed by these studies (Ewaid et al., 2018; Kangabam, et al., 2017; Saleem et al., 2012).

Conclusion

Based on the results obtained by this study, it is possible to conclude that the temperature plays a significant role for the quality of the raw water of the WTS - Terras Baixas, with regard to the concentrations of iron and manganese. Still, the relevance and applicability of the statistical methods proposed for the evaluation of the effects of the parameters rain and temperature on the metals evaluated in this study are highlighted, after all, it was through the statistical methods that it can be proved that the rain does not cause significant influence about the chemical variables studied here, while the temperature has a great effect.

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