Modeling and Prediction of the Total Manganese Concentration Levels In The Finished Water of the Three Water Treatment Plants in the Linggi River Basin of Negri Semblian, Malaysia

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Abstract**—**Water quality parameters of the raw and finished water of the three water treatment plants in the Linggi river basin of Negri Semblian, Malaysia , including pH, dissolved oxygen (DO), conductivity (cond.), turbidity (Tur.), temperature (T), chlorine (Cl_2) and potassium permanganate ($KMnO_4$) dosages were measured to determine their effects on the concentration level of total Mn in their treated water. Sub-sets modelling procedure was applied to develop the final model. It was found that a multiple linear model is efficient enough to explain the variability among the total Mn concentration levels in the finished water of the three plants. The majority of the variables resulted in significant impact. However, the conductivity and turbidity were found not to have correlations with the total Mn concentration levels in the finished water.

Keywords**—**Malaysia, Manganese, Modeling and Water quality.

I. Introduction

THE Linggi River Basin is one of the major polluted river basins in Malaysia. It is located at South-western part of the State of Negri Semblian, Peninsular Malaysia. Seremban, the capital of Negeri Sembilan, is located on the Linggi River. The Linggi River drainage basin is the major source of the drinking water for the townships of Semblian (60 %) and Port Dickson (100 %) in Negri Semblian [1]. The quality of this resource is threatened by rapid development that taking place in this basin. Several studies have shown that the Linggi River is in a state of serious pollution with trace metals such as manganese (Mn) and iron (Fe) and requiring more extensive and effective treatments [2-5], [1], [6-8] and [9]. However, due to resource constraints, the Linggi River is continuously required to be used as the major source of the drinking water for the state of Negeri Semblian [10].

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Modelling procedures that predict the total Mn concentration levels in the finished water of the three water treatment plants namely Pantai Water Treatment Plant (PWTP), Terip River Water Treatment Plant (TWTP) and Linggi River Water Treatment Plant (LWTP) in the Linggi river basin, Malaysia were developed. Linear regression analysis was used to investigate and model the relationship between the total Mn in the finished water of the three water treatment plants and some predictors. These predictors were concentration levels of the total Mn in raw water source, concentration level of the total Mn after sedimentation process, concentration level of the total Mn after filtration process, chlorine and potassium permanegenat dosages, and some raw water quality parameters (pH, DO,cond., Tur., T, Cl_2 and $KMnO_4$ dosages). The results of the correlation of these predictors with the measured concentration level of the total Mn in the finished water was considered helpful in finding out to what extent these predictors influence the concentration level of the total Mn in the finished water of the subjects. For the purpose of the model development, the best subsets model-selection technique was used to screen out predictors does not associate with the concentration levels of the total Mn in the finished water.

II. Methodology

Data used for generating these models were obtained from the previously work which done by Sarmani and Bobaker [9]. The raw and finished water samples of the three WTPs were collected weekly for a period of one year from January to December 2003. Total Mn analysis were done by neutron activation analysis (NAA) as recommended by Sarmani et al. [8]. Five physical parameters were measured *in-situ* using the relevant apparatus. Temperature, pH, conductivity and DO were measured using a Water Quality Test Kit (Ciba Corning Diagnostics, Essex, England), turbidity measured using a turbidity meter (HACH 16800 model). Chlorine (Cl_2) and potassium permanganate ($KMnO_4$) dosages were obtained from the plants authorities.

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III. Results and Discussion

A. Best subsets regression

Best subsets regression procedure identifies the best fitting regression models that can be constructed with the predictor variables that were specified. It was being used to select a group of likely models for further analysis of this study. The general method is to select the smallest subset that fulfils certain statistical criteria. The reason that it should be used as a subset of variables rather than a full set is that the subset model may actually estimate the regression coefficients and predict future responses with smaller variance than the full model using all predictors. It is an efficient way to identify models that achieve the goal of this study with as few predictors as possible [11].

Models are evaluated based on R^2, adjusted R^2 (R^2 adj.), Mallows' C-p statistic (C-p), and standard error of regression (s). The adjusted R^2 and C-p were used to compare models with different numbers of predictors. In this case, choosing the model with the highest adjusted R^2 is equivalent to choosing the model with the smallest mean square error (MSE). The C-p statistic is expressed as the summation of the mean-squared errors of the fitted response values divided by the variance of the error term. A small value of C-p indicates that the model is relatively precise (has small variance) in estimating the true regression coefficients and predicting future responses.

Results of the best subsets regression between the concentration levels of the total Mn in the finished water of the three water treatment plants and the predictor variables are presented in the Tables 1, 2 and 3. In all Tables, each line of the output represents a different model. The number of variables or predictors in the model is listed in the first column. The statistics R^2, adjusted R^2, C-p, and s are displayed next. R^2 and adjusted R^2 are converted to percentages. An X indicates predictors that are present in the model.

TABLE I
RESULTS OF THE BEST SUBSETS REGRESSION PROCEDURE OF THE PANTAI WATER TREATMENT PLANT (PWTP)

No. of Variable	R^2	R^2 adj.	C-P	S	Mn Raw	Mn Sedimentation	pH	DO	Cond.	Tur.	T	Cl_2
1	80.7	80.3	95.3	4.0897	X							
1	23.9	22.3	517.9	8.1274		X						
2	89.0	88.6	35.5	3.1163	X	X						
2	83.1	82.4	79.3	3.8635	X		X					
3	91.9	91.4	16.4	2.7106	X	X	X					
3	91.3	90.8	20.7	2.8038	X	X		X				
4	94.0	93.5	2.8	2.3595	X	X	X					X
4	92.6	91.9	13.3	2.6215	X	X	X	X				
5	94.2	93.5	3.3	2.3449	X	X	X		X			X
5	94.0	93.4	4.3	2.3712	X	X	X	X				X
6	94.2	93.4	5.1	2.3633	X	X	X	X	X			X
6	94.2	93.4	5.2	2.3677	X	X	X		X		X	X
7	94.2	93.3	7.0	2.3883	X	X	X	X	X		X	X
7	94.2	93.3	7.1	2.3899	X	X	X	X	X	X		X
8	94.2	93.1	9.0	2.4159	X	X	X	X	X	X	X	X

TABLE II
RESULTS OF THE BEST SUBSETS REGRESSION PROCEDURE OF THE TERIP WATER TREATMENT PLANT (TWTP)

No. of Variable	R^2	R^2 adj.	C-P	S	Mn Raw	Mn Sedimentation	Mn Filtration	pH	DO	Cond.	Tur.	T	Cl_2
1	72.5	72.0	190.5	8.6946	X								
1	32.7	31.3	536.8	13.614							X		
2	81.6	80.8	114.1	7.1947	X	X							
2	76.7	75.8	156.1	8.0841	X		X						
3	84.1	83.1	94.0	6.7491	X	X	X						
3	83.2	82.2	101.7	6.9353	X	X							X
4	87.8	86.8	64.1	5.9804	X	X	X					X	
4	86.0	84.8	80.0	6.4122	X	X	X	X					
5	89.9	88.8	47.5	5.4913	X	X	X	X				X	
5	89.3	88.1	53.1	5.6628	X	X	X	X	X				
6	93.2	92.3	21.4	4.5725	X	X	X	X	X			X	
6	91.7	90.6	34.4	5.0494	X	X	X	X				X	X
7	95.0	94.2	7.5	3.9582	X	X	X	X	X			X	X
7	93.4	92.4	21.2	4.5392	X	X	X	X	X	X		X	
8	95.1	94.2	8.2	3.9417	X	X	X	X	X	X		X	X
8	95.0	94.1	9.0	3.9828	X	X	X	X	X		X	X	X
9	95.2	94.1	10.0	3.9807	X	X	X	X	X	X	X	X	X

TABLE III
RESULTS OF THE BEST SUBSETS REGRESSION PROCEDURE OF THE LINGGI RIVER WATER TREATMENT PLANT (LWTP)

No. of Variable	R^2	R^2 adj.	C-P	S	Mn Raw	Mn Sedimentation	Mn Filtration	pH	DO	Cond.	Tur.	T	Cl_2	$KMnO_4$
1	73.0	72.5	397.5	26.117	X									
1	36.6	35.3	999.2	40.041		X								
2	83.1	82.4	233.3	20.889	X	X								
2	79.1	78.2	299.5	23.232	X									X
3	89.1	88.4	135.6	16.927	X	X								X
3	85.9	85.1	188.3	19.249	X	X			X					
4	90.9	90.1	108.5	15.656	X	X			X					X
4	90.5	89.7	114.4	15.959	X	X							X	X
5	93.8	93.1	62.8	13.082	X	X		X					X	X
5	93.2	92.5	71.8	13.638	X	X		X	X					X
6	96.1	95.5	27.2	10.533	X	X	X	X					X	X
6	95.3	94.7	39.8	11.505	X	X	X	X	X					X
7	97.4	97.0	7.5	8.6952	X	X	X	X	X				X	X
7	96.5	96.0	21.5	10.002	X	X	X	X				X	X	X
8	97.5	97.0	7.8	8.6288	X	X	X	X	X			X	X	X
8	97.4	96.9	9.1	8.7574	X	X	X	X	X		X		X	X
9	97.5	97.0	9.2	8.6700	X	X	X	X	X		X	X	X	X
9	97.5	96.9	9.7	8.7202	X	X	X	X	X	X		X	X	X
10	97.5	96.9	11.0	8.7496	X	X	X	X	X	X	X	X	X	X

As shown in Table I, the best one-predictor model uses Mn in raw water stage (R^2 adj = 80.3) and the second best one-predictor model uses Mn sedimentation stage (R^2 adj = 22.3). Moving from the best one-predictor model to the best four and five-predictor models increased the adjusted R^2 from 80.3 to 93.5. R^2 usually increases slightly as more predictors are added even when the new predictors do not improve the model. The best four-predictor model might be considered as the minimum C-p (2.8) and maximum adjusted R^2 (93.5). Then, based on these preferred measures the best four-predictor model (Mn raw, Mn sedimentation, pH and Cl_2 dosage) was selected to explain the variation of Mn concentration levels in the finished water of the PWTP.

Similarly, the best seven-predictor model was selected to explain the variation of the total Mn concentration levels in the finished water of the other two plants. For TWTP the predictor variables were concentration level of the total Mn in the raw source, concentration level of the total Mn after sedimentation process, concentration level of the total Mn after filtration process, pH, DO, Cl_2 dosage and T (Tables II). The LWTP predictor variables were concentration level of the total Mn in the raw water source, concentration level of the total Mn after sedimentation, concentration level of the total Mn after filtration, pH, DO, Cl_2 and $KMnO_4$ dosages (Tables III).

The pearson method of correlation was used to examine the correlation between the total Mn concentration level in the finished water of the three water treatment plants and the above selected predictor variables. The correlation between the total Mn concentration level in the finished water and the selected predictor variables was classified accordance to Guilford's rule of thumb. The results are shown in Table 4.

TABLE IV
CORRELATION BETWEEN CONCENTRATION LEVELS OF TOTAL MN IN THE FINISHED WATER OF PWTP, TWTP AND LWTP AND SELECTED PREDICTOR VARIABLES

Predictor Variable	PWTP		TWTP		LWTP	
	Pearson r	P	Pearson r	P	Pearson r	P
Mn Raw	0.898**	0.0001	0.852**	0.0001	0.841**	0.001
Mn Sedimentation	0.488**	0.0001	0.475**	0.0001	0.527**	0.001
Mn Filtration	-	-	0.400**	0.003	0.408**	0.006
pH	-0.353**	0.001	-0.355**	0.010	-0.399**	0.003
T	-	-	0.277*	0.047	-	-
Cl_2 Dosage	-0.314*	0.023	-0.321*	0.020	-0.361**	0.009
$KMnO_4$ Dosage	-	-	-	-	-0.352*	0.011
DO	-	-	-0.379**	0.006	-0.400**	0.003

*significant at 5 %
** significant at 1 %

As noted in Table IV, the concentration levels of the total Mn in the finished water of the PWTP, TWTP and LWTP showed positive and highly significant correlations with the concentration levels of the total Mn in the raw water sources of the three water treatment plants (r = 0.898, r = 0.852, and r = 0.841, respectively). However, positive and moderate correlation with substantial relationship were found between the concentration level of the total Mn in the finished water and that after sedimentation process of the three water treatment plants (r = 0.488, r=0.475 and r= 0.527, respectively). Likewise, positive and moderate correlation with substantial relationship was found between the concentration of the total Mn level in the finished water of the TWTP and LWTP and those after filtration process (r=0.400, and r = 0.408, respectively). However, this effect was also noted by other investigators [12], who reported increase of the concentration level of the total Mn in finished water as a result of the high concentration level of the total Mn in the raw water source.

Negative and low definite correlation with substantial relationship were obtained between the level of the total Mn in finished water of the PWTP, TWTP and LWTP and pH values of the raw water sources of the three water treatment plants (r = -0.353 for PWTP, r = -0.355 for TWTP and r = -0.399 for LWTP). This negative relationship indicated that decreased pH resulted in high total Mn and similarly increases pH results in low total Mn in water column. This due to the fact that the acidic pH water will convert insoluble Mn form in the sediment to the soluble Mn form that will result to increase the total Mn concentration levels in the water column [13], [14], [15]. These findings are consistent with Bratby [16], Kassium [6], Marble et al. [17] and Preda & Cox [18], who reported decreased Mn level as a result of higher pH. However, this parameter seems to be very important factor to control the concentration levels of total Mn in the finished water. Several studies confirmed that the pH is a very important factor in controlling total Mn in the finished water [13], [19], [20], [14].

The temperature was found to have a positive definite correlation with low relationship with the total Mn in finished water for the TWTP (r=0.277). This could be attributed to the fact that the raw water of TWTP is stored in impounding reservoirs where natural purification processes take place prior to treatment. Warm weather and light winds however, may causes reservoirs to become thermally stratified. One of the problems of the thermal stratification is the creation of anoxic conditions in the bottom layer, which leads to dissolution of trace Mn from sediment into the water column [13], [21], [22], [23], [24]. So, this effect may be the cause of correlation between the total Mn in the finished water and temperature.

Chlorine dosages were found to have negative and low but definite with substantial relationship with the level of the total Mn in finished water of the PWTP and TWTP (r = -0.314, r = -0.321 and r = -0.361, respectively). This negative relationship indicated that increases chlorine dosages will result in decreasing the concentration levels of the total Mn in the finished water. This was attributed to the reaction of chlorine with the soluble Mn to form insoluble Mn which can be then removed by the filtration process. The reaction is as follows:

$$Mn^{2+}_{soluble} + Cl_2 + 2H_2O \rightleftharpoons MnO_{2\,insoluble} + 2Cl^- + 4H^+$$

This finding accords with, Graveland and Heertjes [25], Knocke et al. [21] and Wong [15] who reported a decrease in the concentration levels of the total Mn in the finished water as the chlorine dosage increased.

Likewise, potassium permanganate ($KMnO_4$) was found to be negatively and significantly contributing to the Mn level in finished water of the LWTP (r = - 0.352). This was attributed to the reaction of $KMnO_4$ with the soluble Mn to form insoluble Mn which can be then removal by the filtration process. The reaction is as following:

$$3Mn^{2+}_{soluble} + 2KMnO_4 + 2H_2O \rightleftharpoons 5MnO_{2\,insoluble} + 2K^+ + 4H^+$$

This finding accords with Knocke et al. [21], who reported a decrease in the Mn level as the potassium permanagate dosage increased.

The correlation between the DO concentration level and the total Mn level in finished water of TWTP and LWTP were found to be negative and significant (r = -0.379 and r = -0.400, respectively). This negative relationship indicated that increasing the DO concentration levels will result in decreasing levels of the total Mn in the finished water. This is because oxygen gas oxidizes the soluble Mn to insoluble Mn form which can be then removal by the filtration process. Oxidation of Mn^{2+} by molecular oxygen may be described by the following chemical reaction:

$$2Mn^{2+}_{soluble} + O_{2(g)} + 2H_2O \rightleftharpoons 2MnO_{2\,insoluble} + 4H^+$$

This result is agrees well with that obtained by other investigators, who reported that the concentration levels of the total Mn decrease with an increase of DO [26], [6], [21], [17], [27].

B. Linear regression analysis

Linear regression analysis was used to determine how the level of total Mn in the finished water of the three water treatment plants changes as a predictor variable change. It was also used to predict the value of the total Mn level for any value of the predictor variable, or combination of values of the predictor variables. The regression equation took the following form:

$$Y_i = \alpha + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_4 X_4 + \beta_5 X_5 + \beta_6 X_6 + \beta_7 X_7 + \beta_8 X_8 + \beta_9 X_9 + \beta_{10} X_{10} + \mu_i$$

where;

Y = denotes the concentration level of total Mn in finished water (Mn finished) measured in μgl^{-1},

X1 = refers to the concentration level of total Mn in the raw water (Mn Raw) measured in μgl^{-1},

X2 = refers to the concentration level of total Mn after sedimentation process (Mn Sedimentation) measured in μgl^{-1},

X3 = refers to the concentration level of total Mn after filtration process (Mn filtration) measured in μgl^{-1},

X4 = refers to the raw water pH,

X5 = refers to the temperature (T) of the raw water measured in °C,

X6 = refers to turbidity of the raw water (Tur.) measured in NTU,

X7 = refers to chlorine dosage (Cl_2 dosage) measured in mgl^{-1},

X8 = refers to potassium permanegenat dosage ($KMnO_4$ dosage) measured in mgl^{-1},

X9 = refers to conductivity of the raw water (Cond.) measured in μmhos,

X10 = refers to the dissolved oxygen of the raw water (DO) measured in mgl^{-1}, and

µ = an error term assumed to capture the contribution of other unknown variables, α is a constant, and β1, β2, β3, . . . β10 are the coefficients of the predictor variables that to be estimated.

i Subscript of the weekly record. Total sample size equals to 52 weeks.

Only the best predictor variables selected from the Tables I, II and III were employed in the above equation.

Results of the multiple linear regressions between the concentration levels of the total Mn in the finished water of the three water treatment plants and the particular predictor variables are presented in the Table 5. Nevertheless, the results of the diagnostic tests are all satisfactory, they included the detection of multicollinearity between the predictor variables, heteroscedasticity or unequal variance, and autocorrelation between the disturbances or partial correlation. These findings support the validation of these models and confirm that the models are sound and effective for the purpose of this study.

As seen from Table V, the three models have reasonably high explanatory power with coefficients of determination R^2 = 93.5 %, R^2 = 94.2 % and R^2 = 97.0 %, respectively. These indicate that 93.5, 94.2 and 97.0 percents of the total Mn concentration levels in the finished water of the three water treatment plants are explained by particular predictor variables. The high values of multiple R (96.9 %, 97.5 % and 98.7 % for the PWTP, TWTP and LWTP, respectively) that measure the correlation between the observed and predicted values of the total Mn concentration levels; indicate that there are obviously linear relationships between the concentration levels of the total Mn in the finished water and the contribution of the particular predictor variables.

Predicated R^2, which measures the amount of variation in the new data explained by these models; accounted for 92.5 %, 93.1% and 96.2 %, respectively. These findings indicate that less than 10 % of the total variation is not explained by the models. Hence, these models could properly be applied for future data.

Further, the F-values of 183.01, 119.27 and 232.64 indicate the overall significance of the three models at 1 percent level of significance, respectively. This means that the rejection of null hypothesis that the regression coefficients β1 = β2 = β3= …= βk = 0 and the acceptance of the alternative hypothesis that at least one predictor variable coefficient is not equal to zero. Moreover, they indicate that such association between the total Mn concentration levels and the predictor variables under considerations could not be of random origin.

TABLE V
REGRESSION OF THE TOTAL MN CONCENTRATION LEVELS IN THE FINISHED WATER ON THE PREDICTOR VARIABLES OF THE THREE WATER PLANTS MODELS

Predictor Variable	Coefficient		
	PWTP	TWTP	LWTP
Constant	19.34*	18.90	282.97**
	(10.620)	(19.310)	(21.060)
Mn Raw	0.41**	0.29**	0.05**
	(0.021)	(0.016)	(0.003)
Mn Sedimentation	0.15**	0.10**	0.03**
	(0.019)	(0.016)	(0.004)
Mn Filtration		0.17**	0.04**
		(0.020)	(0.007)
pH	-7.48**	-12.26**	-27.69**
	(1.344)	(1.933)	(2.876)
T		1.94**	
		(0.352)	
Cl_2 Dosage	-1.79**	-3.57**	-2.23**
	(0.443)	(0.892)	(0.377)
$KMnO_4$ Dosage			-16.94**
			(1.742)
DO		-2.84**	-3.97**
		(0.525)	(0.846)
Standard Error of Regression (S)	2.360	3.958	8.695
Multiple R	96.9%	97.5%	98.7%
R^2 (adjusted)	93.5%	94.2%	97.0%
R^2 (predicted)	92.5%	93.0%	96.2%
F-value	183.01**	119.27**	232.64**
Lack of Fit	(P>0.1)	(P>0.1)	(P>0.1)

The figures in the parentheses are the estimated standard errors of the regression coefficients
*significant at 10 %
** significant at 1 %

For all models, the lack of fit P-value of more than 0.1 indicates that the linear predictor variables alone are sufficient to explain the variation in the total Mn concentration levels in the finished water [28]. On the other hand, the coefficients of the three models represent the estimated change in the mean of total Mn concentration level for each unit change in the particular predictor value. In other words, it is the change in y that occurs when x increases by one unit. The estimated linear regression models are as follow:

PWTP model:

$[Mn]_{Finished} = 19.34 + 0.41\ [Mn]_{raw} + 0.15\ [Mn]_{sedimentation} - 7.48\ pH - 1.79\ [Cl_2]_{dosage}$

TWTP model:

$$[Mn]_{Finished} = 18.90 + 0.29\,[Mn]_{raw} + 0.10\,[Mn]_{sedimentation} + 0.17\,[Mn]_{Filtration} - 12.26\,pH + 1.94\,T - 3.57\,[Cl_2]_{dosage} - 2.84\,[DO]$$

LWTP model:

$$[Mn]_{Finished} = 283.97 + 0.05\,[Mn]_{raw} + 0.03\,[Mn]_{sedimentation} + 0.04\,[Mn]_{Filtration} - 27.69\,pH - 2.23\,[Cl_2]_{dosage} - 3.97\,[DO] - 16.94\,[KMnO_4]_{dosage}$$

Moreover, the estimated results of partial correlation showed that the concentration of the total Mn in raw water to the total Mn concentration levels in the finished water of PWTP, TWTP and LWTP ranked first with 94.1 % (both PWTP and TWTP) and 93.7 %, respectively. The remaining predictor variables according to their concentration are ranked as follows:

PWTP model:

The Mn sedimentation (75.4 %), pH (63.0 %) and the Cl_2 dosage (50.8 %).

TWTP model:

The Mn filtration (77.6 %), pH of the raw water (69.1 %), Mn sedimentation (68.6 %), T (63.8 %), DO (63.2 %) and the Cl_2 dosage (57.7 %).

LWTP model:

The remaining factors according to their contribution are anked as: the $KMnO_4$ dosage (82.6 %), pH of the raw water (82.3 %), Mn sedimentation (76.5 %), the Mn filtration (69.6 %), Cl_2 dosage (66.4 %) and DO (57.7 %).

The significant results of these fitted models would leave a platform to others to formulate appropriate models for the purpose of building explicit, and theoretically interesting comparisons into the proposed models.

IV. Conclusion

Certain water quality parameters of the raw and finished waters of the three water treatment plants in the Linggi river basin, Malaysia were evaluated to investigate the correlation between the concentration levels of total Mn in the finished water of the three WTPs and these water quality parameters. The results indicated that, there is a close relationship between raw water quality and Mn levels in finished water at these three water plants. It was also found that the multiple linear model is efficient enough to explain the variability among the total Mn concentration levels in the finished water of the three water plants. So, this model can be used to control the high levels of Mn into the permissible level.

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