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Fates of Metal in a Water Reticulation System: A Case Study of Negeri Sembilan / Malaysia

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ABSTRACT

The availability of drinking water is one of the most stringent challenges facing the world. This study was conducted in Negeri Sembilan/Malaysia by taking water samples from raw water intake point along the distribution system. Chemical analysis is conducted to determine the level of contamination of 10 heavy metals by ICP-OES. pH, temperature, salinity, conductivity, and dissolved oxygen were measured on site, while Free chlorine, BOD, COD, TOC, TSS and NH_3-N were measured in the laboratory. The results for physical-chemical parameters showed similar parametric temperature for water. Ammonical nitrogen (NH_3-N) and dissolved oxygen in the tap water vary monthly, exceeding WHO's standard. The levels of Ca, As, Se, and K also exceeded standards set by WHO in certain months throughout the study period. Statistical analysis was applied to determine the relationship between the concentrations of heavy metals and other parameters.

KEY WORDS: Contamination, Heavy metals, Water quality parameters, WHO standards.

1. INTRODUCTION

Water contamination is one of most crucial environmental problems in the recent decade (Muhammad, 2011). Many countries, especially in Africa and Asia, lack access to adequate potable water. Although 70% of the globe's surface is water, it is mostly made up of salt water, with only 3% fresh water, represented by rivers and lakes, which can be safely consumed (Mebrahtu & Zerabruk, 2011). The source of water in Malaysia is rivers and streams, with rainfall rate from 2000-2500 mm annually (Rahmanian, 2015). Surface water represents 99% of water source, amounting to 580 km^3 annually (Hafiza, 2015). The quality of the drinking water affects the human health. The presence of heavy metals in drinking water is a problem that needs to be addressed to ensure access to clean and potable water. There are many sources of heavy metals, such as industrial discharges, sewage effluents, and the earth's crust (Mohod & Dhote, 2013). Pollution in Linggi River (Malaysia) can be attributed to industrial and agricultural activities, and human settlements. Palm plantations, rubber trees, and fishing also contribute to pollution. The estate development and industry that began in the 1970s led to many environmental problems due to reduced areas of forestation and agriculture on the rivers (Ngah & Othman 2011). There are two types of Linggi River water; the first is polluted and needs treatment, and the second is water that require special treatment process due to its high levels of pollution (Lonercan & Vansickle, 1991). For more on Linggi River water (Ibrahim & Mustafa, 2010; Kasim, 2015; Khalik & Abdullah, 2015; Khan, 1992; Khan & Begham, 2012; Mokhtar, 2002.

There are four stages of treating water in plants; the first stage (11A) represents raw water, the addition of $Al_2(NO_3)$ to the water is the 11B stage, filtered water is in the 11C stage, and the final stage (11D) distribute water to the customers. Azrina (2011) and Khoo (2011), determined the quality of tap water in Negeri Sembilan by measuring the concentration of metals in the tap water in all of Malaysia via two locations from every state. The first author used spectroscopy methods to measure the level of metals in the water, while the second author focused only on five metals. Their results showed that most of the metals' concentrations are below standard levels, meaning that the water is safe to drink. Khalid & Abdullah (2013) investigated water quality at Sri Menanti, Negeri Sembilan via three locations during the wet season. He used ex-situ analysis to determine the concentrations of seven parameters, and the results showed that the TSS concentration exceeded that of the standard level.

Rahman (2012) and Bobaker (2014), compared three stages of heavy metals concentrations in Linggi River; raw, portrait and fresh. Rahman (2012), used (ICP-MS) to determine the concentrations of seven metals, then compared the concentrations at the three aforementioned stages. His results showed that most metal concentrations decreased after treatment. Bobaker (2014), studied the effect of the parameters, such as pH and DO on the total concentration of Mn of the finished water in three plants on the river. This study was conducted over a period of four months, from June - September 2016, by taking 25 samples every month from Linggi River, treatment plant, and tap water. We intend to assess drinking water quality in Negeri Sembilan over time, followed by the water treatment process. Next, the concentrations of heavy metals and parameters in the water through three stages were compared: river, plant, and tap water. We will then clarify the relationship between heavy metals and other parameters.

2. MATERIALS AND METHOD

Study area description: The area of study is Negeri Sembilan, which is one of the states in Malaysia (Figure.1). It lies south of Kuala Lumpur and borders Selangor, Pahang and Malacca from north, east, and south, respectively. The population of the state is one million, covering an area of 6686 km^2. Sampling was performed on the Linggi River by taking 11 samples from different areas of the river facing the streets, houses, trees, and shops on the riversides. A total of 14 samples was collected from the treatment plant; 11 stations of tap water and 3 from other sources.

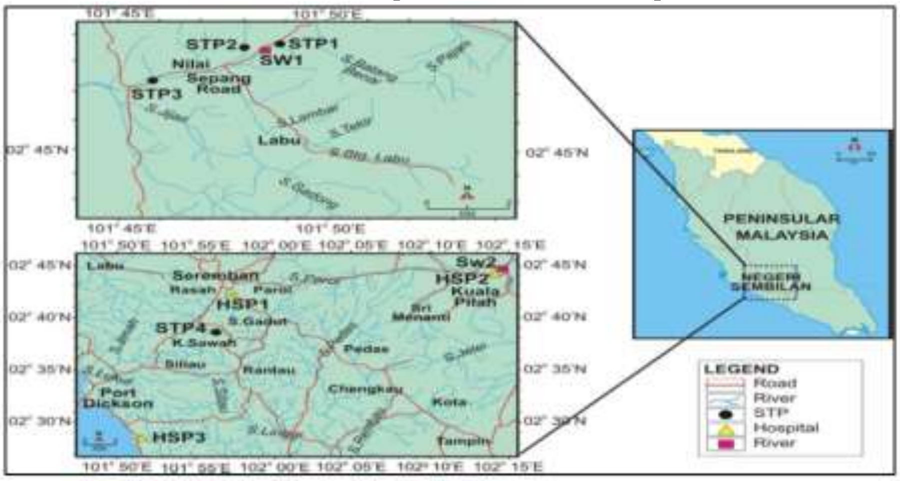

Figure.1. Negeri Sembilan

Sampling: A total of 4 samples was collected from various drinking water sources in Negeri Sembilan from Jun-Sep 2016. This included 11 samples from the rivers; Sg. Batang penar (before sand mining), Sg. Batang Penar (after sand mining), Intake water Treatment plant (WTP) Sg. Terip, Sg. Batang penar-after sand mining 2, Sg. paroi, Sg.Temiang, Sg. Senawang, sg. Kepayang, Sg. Mantau, Sg. Kayu Ara, and WTP Sg. Linggi-intake. Three samples were collected from the treatment plant; WTP-sedimentation Tank, WTP-Filter, and WTP-clear water tank. 11 samples were collected from tap water (residence); Bandar sunggala, Si Rusa RES 2 Outlet, Si Rusa RES 3 Outlet, Si Rusa RES 4 Outlet, Si Rusa RES 5 Outlet, Jalan lama (A:038), Taman Ria (A:034), Tanah Merah Site C, Tanah Merah Site A, Pekan Chuah (A:025), and Bandar Baru Springhill. The water samples were collected in a clean and dry polyethylene bottle pre-washed with 65% nitric acid (HNO3) and double-distilled water. Some of the parameters were analyzed on site, such as temperature, Dissolved Oxygen (DO), pH, and conductivity using an instrument (YSI 560 Multiparameter probe). Other parameters such as (Biochemical Oxygen Demand (BOD), Total organic carbon (TOC), total suspended solids (TSS), Chemical Oxygen Demand (COD) and ammonical nitrogen (NH_3-N) were measured in the laboratory. The water samples were filtered, and a few drops of HNO_3 were added before the samples were transported to the laboratory and filtered using Vacuum filtration. Also, the samples' heavy metal content was determined using (ICP-OES), and the samples were then refrigerated at 4°C.

Chemical analysis: All acidified water samples were analyzed for its content of heavy metals (Al, Ba, Fe, K, Mg, Na, As, Pb, Se, Co, Cr, Cu, Mn, Ni, Pb, and Zn) using Inductively coupled plasma optical emission spectrometry ICP-OES. It offers multi-element analysis and absorption spectrometer, but this technique is not extensively used in developing countries due to its difficulty in implementation and maintenance costs. The temperature, DO, pH, and conductivity were being measured on site using (YSI 560 Multiparameter probe), while the concentration of Biochemical Oxygen Demand (BOD), Total organic carbon (TOC), total suspended solids (TSS), Chemical Oxygen Demand (COD), and ammonical nitrogen (NH_3-N), was measured in the ALIR laboratory of UKM. Total Suspended Solid (TSS) were tested by using APHA - Standard methods for examination of water and wastewater (21st Edition, 2005) (Majd & Othman 2015). The Aurora TOC analyzer was used to measure the total Organic Carbon (TOC), while Chemical Oxygen Demand (COD) and Ammonical Nitrogen (NH_3-N) were tested using the HACH standard method (DR 3900) (Zakaria, 2015).

Statistical analysis: Statistical analysis was performed to determine the relationship between heavy metals and the aforementioned parameters. Then, ANOVA was used to determine the effect of each heavy metal on other metals and parameters by testing the following hypothesis:

$$H_0: B_0 = B_1 = B_2 = 0 \dots.$$

$$H_1: B_0 \neq B_1 \neq B_2 \neq 0 \dots.$$

Where, H_0 : null hypothesis (no effect)

H_1 : alternative hypothesis (effect)

3. RESULTS AND DISCUSSION

The values of the most critical parameters detected during the water treatment plant and water distribution network in several locations in Negeri Sembilan were analyzed to determine the concentration of (Ba, Fe, K, Mg, As, Pb, Se, Mn, Pb, Ca and Zn), and physical-chemical parameters, such as pH, conductivity, and DO. Concentrations of Al, Ba, Fe, Mn, Pb, Mg and Zn in tap water is below the standards set by WHO, while the concentration of As in tap water exceeded that of the standards set by WHO in August and September (Figure.12), and Se exceeded that level in July and August (Figure.14). The concentration of Ca was out of control for all of the measured months (Figure.6), while the level of K was under control only in June (Figure.9). The type of pipe used in the treatment and distribution network is asbestos cement. Some metals, such as Ca and Pb, were detected in tap water due to erosion-corrosion of the piping system (Azrina, 2011; Colter & Mahler, 2006). The presence of heavy metals can be attributed almost entirely to leaching caused by corrosion of the distribution pipes. Studies have shown that corrosion products attached to pipe surfaces or accumulated as sediments in the distribution system can protect microorganisms from disinfectants (Agatemor & Okolo 2008). The total concentrations of the heavy metals increased in stations 18-22, as well as station 12.

This could be due to the distance between the treatment plant and the stations, and pipe aging (Figure.4). The concentrations of pH, Cond., Residual Cl_2, TSS, COD, and BOD in the tap water was under control, as per the standards set by WHO. The concentration of temp in the tap water exceeded WHO in June, July, and August, while NH_3-N exceeded that level in July. The concentration of Do in tap water was out of control in all months (Table.1). The level of DO depends on the chemical, physical, and biological activities in the water (Khalid, 2013). The water temperature in the pipe can change as per the weather, which affects the source of water, and also pipe corrosion, as it differs from one season to another depending on the ambient temperature. The temperature also affects the DO, which is one of the parameters that increases corrosion (Jung, 2009; Mcneill & Edwards 2002). There are no controls on water treatment over time in the case of Ba, Ca, Mg, K, Mn, Zn, As, and Se. A pipe affects the concentrations of Ca, As and Pb in the plant and tap (Figures.6, 12, and 13).

The treatment processes, increased the concentration of Ca, Mg, and Se relative to their respective concentrations in the river and the plant (Figures.6, 8, &12). The treatment processes were ineffective in the case of K (Figure.9). The additional chlorination in the treatment process led to increased levels of As (McNeill & Edwards, 2002).

Correlation and ANOVA were used to clarify the relationship between the concentrations of the heavy metals and physical- chemical parameters. F value is a significant in the case of Ba (Eq. 1) (Lyman Ott & Longnecker, 2010). From P-value, we can deduce that Fe, Mn and NH_3-N are significant (less than 0.05). Based on the Coefficients value, there is a positive relation between Fe, Mn and NH_3-N from side and Ba from another side. 49% from variations in Ba explained by Ca, Fe, Mn and NH_3-N, according to the value of the adjusted R Square. By using the same manner, we found:

- 78% from variations in Ca explained by K, Mg and pH (positive relation), and Mn (negative relation) (Eq.2).
- 64% from variations in Fe explained by Mn and As (positive relation) (Eq.3).
- 45% from variations in K explained by Mg and As (positive relation) (Eq.4).

Heavy metals concentration: Figures (2a & 2b) show the heavy metals concentrations in tap water for four months.

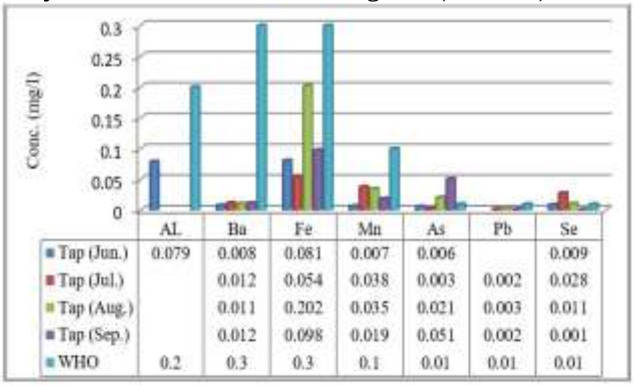

Figure.2.a. Concentrations of heavy metals in the tap water, according to the four months

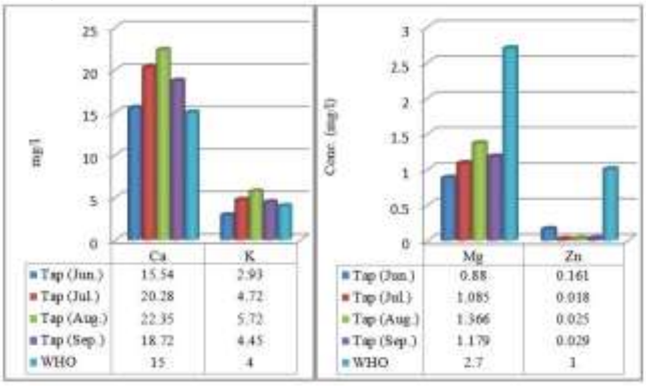

Figure.2.b. Concentrations of heavy metals in the tap water, according to the four months

Figure.3, shows the changing in heavy metals concentrations in the tap water, according to the stations.

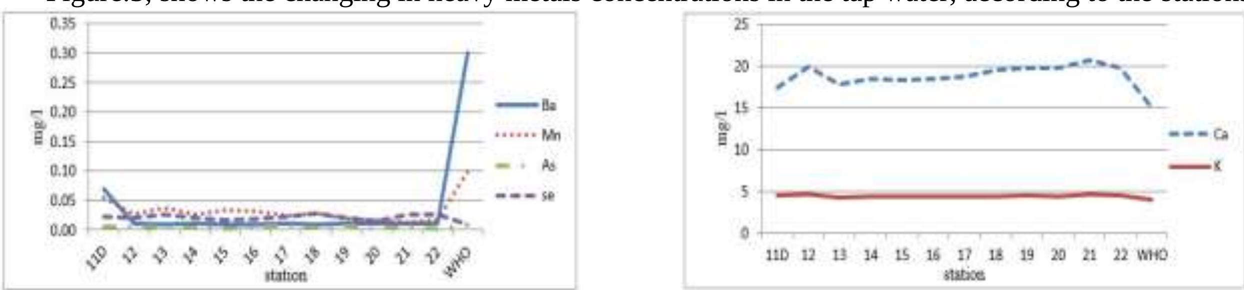

Figure.3. Concentrations of heavy metals in the tap water, according to the stations

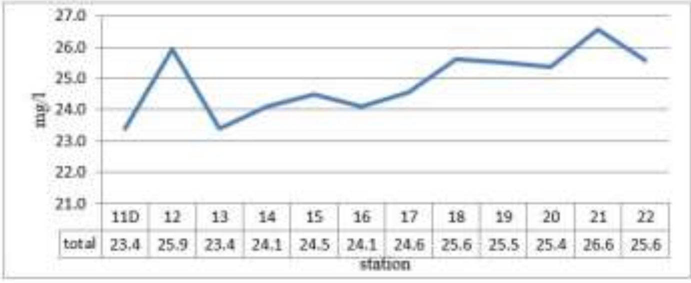

Figure.4. The total concentrations of heavy metals in the tap water, according to the stations

Parameters: Table.1, shows the concentrations of parameter from source to consumption over time.

Table.1. value of Parameters in the Tap Water

Month		June	July	August	September	WHO
PH (unit)	River	6.72	7.54	6.71	6.97	6.5-8.5
	Plant	6.860	7.090	8.940	8.350	
	Tap	6.900	7.197	6.962	7.135	
Temp (c0)	River	27.71	30.03	30.5	28.83	30
	Plant	28.590	29.770	30.400	28.770	
	Tap	30.625	30.928	30.482	29.918	
Cond (µs/cm)	River	66	146	229.7	153	1000
	Plant	92.000	168.000	244.200	151.000	
	Tap	125.636	157.451	179.182	201.627	
Salinity (ppt)	River	0.03	0.07	0.04	0.07	-
	Plant	0.040	0.080	0.050	0.040	
	Tap	0.058	0.069	0.078	0.070	
DO (ppt)	River	3.73	4.5	2.42	3.65	4
	Plant	5.010	3.160	6.320	4.890	
	Tap	4.650	5.966	5.986	5.970	
Residual Cl_2 (mg/l)	River	0.53	0.27	0.17	0.14	5
	Plant	0.100	0.400	1.960	0.240	
	Tap	0.083	0.812	0.358	0.942	
Tss (mg/l)	River	1198	204	106	0.206	500
	Plant	8.000	18.000	4.000	0.022	
	Tap	6.000	11.091	9.309	0.009	
NH_4 (mg/l)	River	1.12	3	3	3	0.5
	Plant	0.010	3.000	0.880	1.000	
	Tap	0.015	1.052	0.330	0.325	
COD (mg/l)	River	4	24	26	12	100
	Plant	2.000	5.000	8.000	4.000	
	Tap	7.545	3.545	15.273	6.455	
BOD (mg/l)	River	2.2	2.282	3.5	2.1	10
	Plant	4.500	1.7	1.800	4.900	
	Tap	3.136	2.082	1.827	3.082	
TOC (ppm)	River	3.267	5.964	7.101	1.395	-
	Plant	1.971	4.355	7.096	0.790	
	Tap	2.05	3.883	3.646	0.18	

A control on the tap water treatment: Figures.4-13, illustrate the concentrations of the heavy metals from source to consumption Over time.

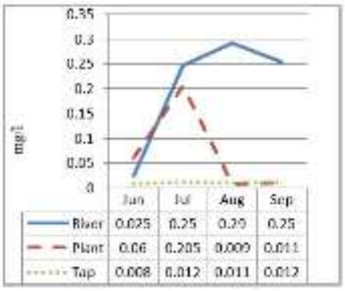

Figure.5. Concentration of Ba

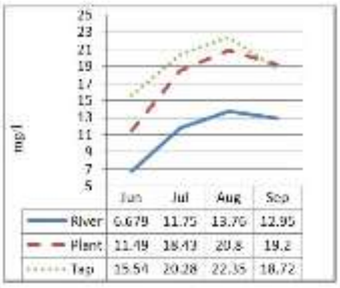

Figure.6. Concentration of Ca

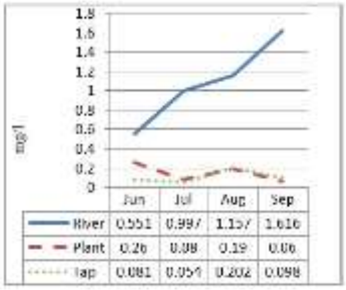

Figure.7. Concentration of Fe

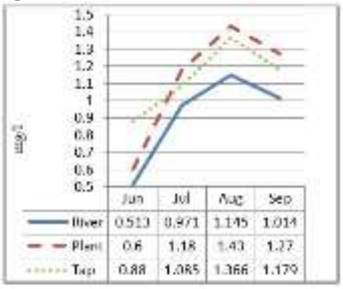

Figure.8. Concentration of Mg

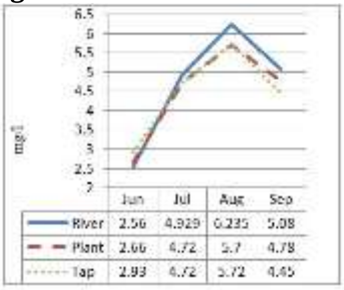

Figure.9. Concentration of K

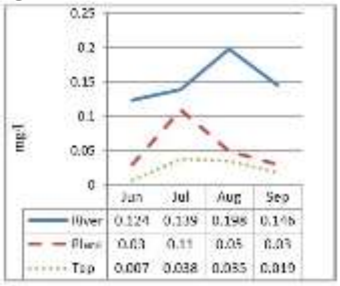

Figure.10. Conc. of Mn

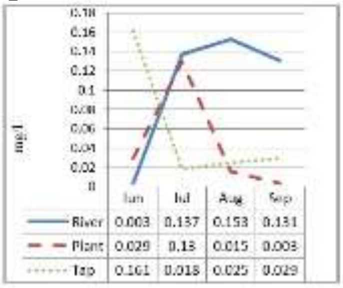

Figure.11. oncentration of Zn

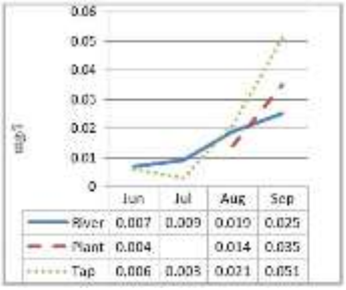

Figure.12. Concentration of As

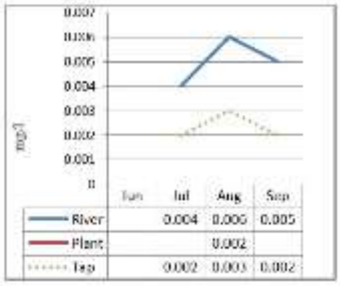

Figure.13. Concentration of Pb

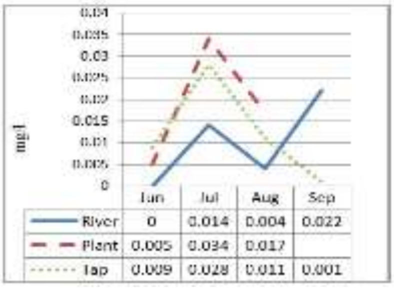

Figure.14. Concentration of Se

The Relation between heavy metals and parameters:

Correlation: The correlation matrix is as shown in Table.2.

Table.2. The correlation matrix

	Ba	Ca	Fe	K	Mg	Mn	Zn	As	se	PH	Temp	Residual Cl	Tss	NH_4
Ba	1.00													
Ca	-0.45	1.00												
Fe	0.58	-0.42	1.00											
K	0.23	0.48	0.07	1.00										
Mg	-0.08	0.66	-0.12	0.58	1.00									
Mn	0.64	-0.76	0.65	-0.13	-0.39	1.00								
Zn	0.30	-0.17	0.20	0.08	-0.12	0.17	1.00							
As	0.23	0.04	0.47	0.40	0.09	-0.01	0.04	1.00						
Se	-0.18	0.36	-0.24	0.19	0.32	-0.32	-0.08	-0.16	1.00					
PH	-0.20	0.40	-0.09	0.13	0.25	-0.21	-0.10	-0.02	0.09	1.00				
Temp	-0.14	0.14	-0.14	-0.06	-0.02	-0.23	0.00	-0.04	0.00	0.05	1.00			
Residual Cl	0.12	-0.19	-0.03	-0.05	-0.03	0.25	-0.05	-0.23	0.16	-0.17	-0.13	1.00		
Tss	0.12	-0.38	0.06	-0.26	-0.28	0.37	0.07	-0.04	-0.15	-0.25	-0.13	0.36	1.00	
NH_4	0.40	-0.25	0.25	0.17	0.04	0.27	0.20	0.05	-0.08	-0.06	-0.11	0.02	-0.03	1.00

Anova: From correlation matrix, we can deduce there is a relation between the independent variables $(Ca, Fe, Mn \,\&\, NH_3 - N)$ and the dependent variable (Ba). Thus the linear regression equation of *Ba* is:

$$Ba = B_0 + B_1 Ca + B_2 Fe + B_3 Mn + B_4 NH_3 - N + e \qquad (1)$$

The results with 95% significance level as shown in Table.3.

Table.3. The Regression Statistics based on equation 1

Regression Statistics					
Multiple R	0.712258				
R Square	0.507311				
Adjusted R Square	0.486567				
Standard Error	0.10007				
Observations	100				
Anova					
	df	**SS**	**MS**	**F**	**Significance F**
Regression	4	0.979566	0.244891	24.45489	6.26E-14
Residual	95	0.951331	0.010014		
Total	99	1.930896			
	Coefficients	**Standard Error**	**t Stat**	**P-value**	
Intercept	-0.04444	0.055309	-0.80344	0.423726	
Ca	0.001928	0.002751	0.700912	0.485071	
Fe	0.061056	0.024348	2.507671	0.013849	
Mn	0.661584	0.184346	3.588817	0.000528	
NH_3-N	0.00938	0.003061	3.064166	0.002841	

$$Ca = B_0 + B_1Fe + B_2K + B_3Mg + B_4Mn + B_5Se + B_6PH + B_7tss + e \quad (2)$$

The results with 95% significance level as shown in Tables 4

Table.4. The Regression Statistics based on equation 2

Regression Statistics					
Multiple R	0.89391				
R Square	0.799075				
Adjusted R Square	0.783787				
Standard Error	2.665049				
Observations	100				
Anova					
	df	**SS**	**MS**	**F**	**Significance F**
Regression	7	2598.673	371.239	52.26886	2.39E-29
Residual	92	653.429	7.102489		
Total	99	3252.102			
	Coefficients	**Standard Error**	**t Stat**	**P-value**	
Intercept	-4.12611	3.617308	-1.14066	0.256973	
Fe	0.077798	0.673019	0.115596	0.908225	
K	1.142007	0.283695	4.025474	0.000117	
Mg	4.080754	1.110635	3.674255	0.000401	
Mn	-34.0024	4.137722	-8.21765	1.27E-12	
Se	6.352091	10.36361	0.612923	0.54144	
PH	1.877464	0.493606	3.803564	0.000256	
Tss	7.83E-05	0.000153	0.511883	0.60996	

$$Fe = B_0 + B_1Mn + B_2As + e \quad (3)$$

The results with 95% significance level as shown in Table.5.

Table.5. The Regression Statistics based on equation 3

Regression Statistics					
Multiple R	0.804931				
R Square	0.647913				
Adjusted R Square	0.640654				
Standard Error	0.332263				
Observations	100				
Anova					
	df	**SS**	**MS**	**F**	**Significance F**
Regression	2	19.70616	9.85308	89.2501	1.03E-22
Residual	97	10.70866	0.110399		
Total	99	30.41482			
	Coefficients	**Standard Error**	**t Stat**	**P-value**	
Intercept	-0.10391	0.055877	-1.85954	0.065982	
Mn	3.562792	0.328083	10.85941	1.89E-18	
As	28.87801	3.660479	7.889133	4.61E-12	

$$K = B_0 + B_1 Mg + B_2 As + e \quad (4)$$

The results with 95% significance level as shown in Table.6.

Table.6. The Regression Statistics based on equation 4

Regression Statistics	
Multiple R	0.676716
R Square	0.457944
Adjusted R Square	0.446768
Standard Error	0.884122
Observations	100

45% from variations in *K* explained by *Mg* and *As*.

Anova					
	Df	**SS**	**MS**	**F**	**Significance F**
Regression	2	64.05678	32.02839	40.97418	1.26E-13
Residual	97	75.82223	0.781672		
Total	99	139.879			
	Coefficients	Standard Error	t Stat	P-value	
Intercept	2.118009	0.290178	7.299001	8.02E-11	
Mg	1.963709	0.270075	7.270991	9.18E-11	
As	46.23775	9.778038	4.728735	7.66E-06	

$$Mg = B_0 + B_1 Mn + e \quad (5)$$

The results with 95% significance level as shown in Table.7.

Table.7. The Regression Statistics based on equation 5

Regression Statistics	
Multiple R	0.390926
R Square	0.152823
Adjusted R Square	0.144179
Standard Error	0.305567
Observations	100

$$Mn = B_0 + B_1 Se + B_2 tss + e \quad (6)$$

The results with 95% significance level as shown in Table.8.

Table.8. The Regression Statistics based on equation.6

Regression Statistics	
Multiple R	0.461426
R Square	0.212914
Adjusted R Square	0.196686
Standard Error	0.091231
Observations	100

$$Re = B_0 + B_1 tss + e \quad (7)$$

The results with 95% significance level as shown in Table.9.

Table.9. The Regression Statistics based on equation.7

Regression Statistics	
Multiple R	0.357464
R Square	0.127781
Adjusted R Square	0.118881
Standard Error	0.605977
Observations	100

4. CONCLUSIONS

In this paper, we analyzed 100 samples from Linggi River water and tap water over a period of four months, from June - September 2016. The concentrations of heavy metals and parameters were determined and compared with the standard concentrations set by WHO and Malaysia.

The concentration of Ca and Do were out of control in all of the tested months in the tap water. High levels of Ca mean the occurrence of erosion corrosion in the pipes. The pipe also affects the concentrations (increase or decrease) of the heavy metals in the water. For example, Pb was detected in tap water in July and September, despite not being present in the plant water. There was no stability in the water treatment over time. Water treatment decreased the concentration of most heavy metals. As, Se, K, temperature, and NH_3-N exceeded the standards set by WHO in certain months. Statistical analysis showed that the K, Mg, Mn and PH affect the Ca by 78%; Mn and As affect Fe by 64%; Fe, Mn and NH_3-N effect on the Ba by 49%; Mg and As affect K by 45%.

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