

RESIDUAL ALUMINIUM IN DRINKING WATER SUPPLIES IN RELATION TO PEATLAND CATCHMENT AREAS IN MUKAH, SARAWAK

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Abstract: This study focused on the relationship between peatland catchment areas and aluminium (Al). For some major development areas in Sarawak, the only available water sources are peat waters. Thus, the objectives of this study were to quantify the levels of Al and other trace metals in raw and treated water from peat and non-peat sources and also to quantify the levels of organic matter and nutrients in raw water from peat and non-peat sources. The study was conducted at four selected catchment areas in Mukah, Sarawak. In particular, this study shows that the treated and raw water sources from both peat and non-peat sources have high levels of Al, ranging from 0.089 – 3.458 mg/L for raw water and 0.235 – 4.574 mg/L for treated water. In addition, results show that treated water from two of the peat water sources were significantly higher in mean levels of Al as compared to before treatment in both sampling trips. Treated water from non-peat source also was significantly lower in mean levels of Al than the treated water from peat sources. Low pH, a typical peat swamp water characteristic was shown to have a big impact on the levels of Al in treated water, mainly due to the higher solubility of Al in acidic condition, with the use of Al-based coagulants in its water treatment processes. Thus, considerations have to be made on the use of Al-based coagulants.

Keywords: Raw water, trace metals, treated water, water quality

Introduction

Challenges in health and aesthetic parameters in public watersupply are major concerns in drinking water treatment. What have always been feared since the practice of using chemical treatment in water are the residues and by-products it produces. For example, trihalomethane (THM) is a widely known carcinogenic by-product produced by the application of chlorine in water disinfection process (Richardson & Postigo, 2012). In order to provide potable water supply, surface water has always been treated with aluminium (Al)-based chemicals like Al sulphate ($Al_2SO_4)_3$, polyaluminium chloride (PAC) or Al chlorohydrate (ACH). Addition of these chemicals are favorable due to their effectiveness in the removal of particulates and colloidal and dissolved substances by the formation of visible flocs through coagulation and flocculation that can then be eliminated through other series of physical treatment processes such as sedimentation and filtration before water can be disinfected using chlorine

(Srinivasan *et al.*, 1999; Binnie & Kimber, 2009). Often, this treatment will produce a certain level of residues; thus, all water purveyors in Malaysia are to comply with the National Drinking Water Quality Standards (NDWQS) set by the Ministry of Health (MOH). For residual Al, the maximum permissible limit is set at 0.2 mg/L (NDWQS, 2004) for treated water. Although acute exposures to high doses of Al are well tolerated, some studies had shown possible relationship with neurodegenerative diseases such as Alzheimer's (AD) and Parkinson's Dementia (PD) (Gidding, 1998; Flaten, 2001; Virginie *et al.*, 2009) regardless that the risk factor for these diseases has not been resolved conclusively. Dosing of Al-based chemicals in water treatment is suggested to be the major source of residual Al in drinking water; however, it is not the only cause, as Al also occur naturally in soil and water bodies (ATSDR, 2008).

There are two naturally abundant types of soil available in Sarawak, namely Histosols and Ulvisols, and these largely contribute to the

acidity and organic content of surface water sources (Hashim, 2003). This study focused on the relationship between peatland catchment areas and Al occurrences with the aim of providing a better understanding of how Al occurrences in natural ecosystem had impacted treated water supply. Histosols or peats are soils with a surface layer containing more than 30% organic matter in 40 cm of the upper 80 cm of the profile (FAO-UNESCO, 1990). Water draining from peat land is black owing to its high levels of humic and fulvic acids, polyphenols and other products of organic matter decomposition (Wosten *et al.*, 2008). With the development of modern industry, acid precipitation is becoming more and more common. This has led to a large amount of Al dissolving from soil to natural waters which may increase the concentration of residual Al in drinking waters (Wang *et al.*, 2010; Gerhard *et al.*, 2002). For some major development areas in Sarawak, the only available water sources are peat waters. The peatland areas of the state's coastal zones are often characterized by high sulphide content and iron (Fe) Al complexes. Peat soils also have high content of humic substances, thus as suggested by Lovely *et al.* (1996) might be dynamically involved in carbon and electron flow in anaerobic environments, indicating their potential role in pollutant transformation. Due to the presence of humic substances that contributes to acidity problem, it may also affect water treatment processes, producing treated water that have odour, colour and taste (Sim & Murtedza, 2007). Several studies have been conducted on water quality of non-peat river water (Ling *et al.*, 2017a, b; Sim *et al.*, 2016; Ling *et al.*, 2014) but for peat river water, little has been studied. Rosli *et al.* (2010) studied

water quality at peat swamp forest converted into oil palm plantation in Sarawak. Thus, the objectives of this study are to quantify the levels of Al and other trace metals in raw water and treated water from peat and non-peat sources, and to quantify the levels of organic matter and nutrients in raw water from peat and non-peat sources.

Materials and Methods

Catchment areas in Daro District, Mukah, were chosen as the study site. Figure 1 shows the location of the sampling stations. Two types of water were collected for the purpose of this study: raw and treated water. For raw water, there were five stations where samples were collected from peat (R1-R3, R5) and non-peat (R4) sources (Table 1). Peat waters in this study were river waters draining from peat land whereas non-peat waters were from a river with no surrounding peat soils. For treated water, samples were collected from the tap water outlet at each water treatment plant (WTP) which draws water from the raw water source. The stations at WTP were Matu (T1), Tian (T2), Daro (T3) and Basong (T4). Sampling trips made to complete the data collection were 1st November and 13th December 2013. Both sampling trips started at 10 am and ended at 4 pm with no changes in the weather (sunny) during the whole trip. Triplicate grab samples of water were collected using 2L polyethylene bottles. All samples of raw water were collected at the surface water layer, not more than 5 cm from the surface, avoiding the top 1 – 2 cm to avoid collecting floating debris. Prior to transport to the laboratory, samples were kept in a cooler box filled with ice ($\leq 4^{\circ}\text{C}$).

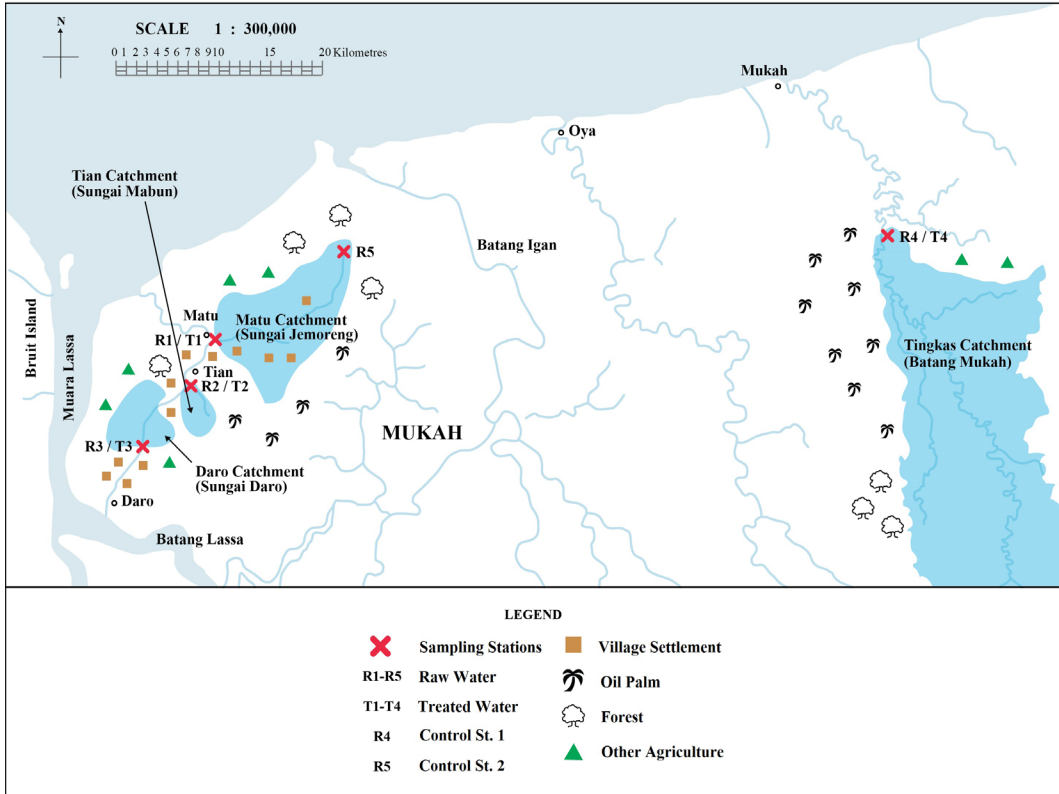


Figure 1: Sampling stations of raw water and treated water and the land use at the study site

Table 1: Sampling stations and site information of the peat and non-peat raw water sources

Station	Name	Type	Description	Coordinates
R1	Sg. Jamoreng	Peat water	Raw water source for Matu WTP, flowing	N 5295925.08 m E 2208665.49 m
R2	Sg. Tian	Peat water	Raw water source for Tian WTP, slightly stagnant	N 5291017.45 m E 2203173.84 m
R3	Sg. Daro	Peat water	Raw water source for Daro WTP, slightly stagnant	N 5284240.24 m E 2197455.39 m
R4	Control St 1	Non-Peat Water	Raw water source for Basong WTP, flowing	N 5306172.84 m E 2278462.27 m
R5	Control St 2	Peat Water	Upstream of R1 – R3, flowing	N 5296396.04 m E 2209190.33 m

Field parameters of pH, water temperature (Tm), turbidity (Tb), conductivity (Cn) and dissolved oxygen (DO) were measured using a pH meter (CLEAN PH30/Temp Tester), turbidimeter (HACH 2100P), Cn meter (CLEAN CON30 Tester) and DO meter (Milwaukee SM600). Analyses of total metals

in water were conducted according to Method 3111 B and 3111 D (APHA, 2005). Water samples in 100 mL polyethylene bottles were acidified using concentrated nitric acid. Then, the water samples were analyzed for metals (Al, Pb, Cu, Zn, Cd, Cr, Fe, Mn and Ni) using Flame Atomic Absorption Spectrophotometer (Thermo

Scientific iCE3XXX). A blank sample was also carried through the complete preparation and analytical procedure. In order to calculate the concentration of total metals in the samples, a standard curve was prepared before the analysis of samples began by taking a series of working standards in the detectable range for each metal.

Chemical oxygen demand (COD) (0 – 150 mg/L) was measured using the Standard Methods (Reactor Digestion Method) (APHA, 2005; HACH, 2003). Water samples were homogenized before transferring into the COD digestion reagent vial and heated in a COD reactor for two hours. After heating and cooling to room temperature, the COD concentration was measured using a spectrophotometer (Model HACH DR2800). Total Reactive Phosphorus ($\text{PO}_4^{3-}\text{-P}$) was measured using the Ascorbic Acid Method (4500-P E.) and HACH PhosVer3 Method (APHA, 2005). All samples were mixed with activated carbon and filtered prior to analysis. A powdered reagent packet was added to 10 mL of sample. After at least 10 minutes, sample was placed inside a vial of the spectrophotometer (Model HACH DR2800). The reading in mg/L was measured at 880 nm. A blank added with the powdered reagent was carried throughout the experiment as the reference solution, and measurement for the blank treated with activated carbon was also conducted. An individual calibration curve from a series of standards within the phosphate ranges was prepared. Distilled water blank was used with the combined reagent to make photometric readings for the calibration curve. Absorbance vs. phosphate concentration was plotted to give a straight line passing through the origin. At least one phosphate standard was tested with each set of samples.

The ammoniacal nitrogen analysis was conducted using the titration method according to APHA (2005). Prior to the analysis, 250 ml of each water sample was distilled to avoid excessive colour and turbidity which potentially interfere in colorimetric analysis. At least 200 mL distillate were collected in boric acid solution and titrated with 0.02 N H_2SO_4 . A

blank was carried out through all the steps. The amount of ammonia nitrogen was calculated using the equation [1].

$$\text{Ammonia Nitrogen} = [(A - B) / \text{ml sample}] \text{ mg/L} \quad [1]$$

where A is the volume of 0.02 N H_2SO_4 titrated for the sample (mL) and B is the volume of 0.02 N H_2SO_4 titrated for the blank (mL). Sulphide analysis was done according to the Methylene Blue Method (HACH, 2013). Sulphide 1 (0.5 mL) and Sulphide 2 (0.5 mL) reagents were added to a 10 ml sample. Hydrogen sulfide and acid-soluble metal sulfides react with N, N-dimethyl-p-phenylenediamine sulfate to form methylene blue. The intensity of the blue color is proportional to the sulfide concentration. After the five minutes reaction period, samples were measured in mg/L at 665 nm using a spectrophotometer.

The one-way ANOVA with Tukey test was performed to explore if there was any significant difference between water quality parameters in each station and different raw water sources. Bivariate correlations were performed at 5% level of significance to test whether there were correlations between the water quality parameters. One sample *t*-test was used to analyze the significant difference as compared to a standard value and previous studies results. To compare the means of peat water and non-peat water, independent *t*-test was conducted to see the difference between the two types of water. Comparison of raw water and treated water of the same type was analyzed using the paired *t*-test to study the relationship before and after treatment.

Results

Two water quality standards from the Malaysian Ministry of Health and the Department of Environment were used to evaluate the results. Parameters which are not stated in the recommended limit of National Drinking Water Quality Standards (NDWQS) (MOH, 2004),

were classified based on the National Water Quality Standard (NWQS) (DOE, 2009).

Results of *in situ* parameters (Cn, DO, pH, Tm and Tb) for raw water are shown in Table 2a. All of the peat raw water stations violated the recommended range for pH and were significantly different from ($P<0.05$) the non-peat raw water station R4 with low pH. Dissolved oxygen level for all the peat raw water stations falls under Class III and IV and were also significantly different from ($P<0.05$) the non-peat raw water station R4 DO of non-peat raw water at R4 falls in the range reported at Serin River and Baram River (Ling *et al.*, 2014; 2017b). Results for *in situ* parameters for treated water are shown in Table 2b. All of the treated water stations showed significant difference ($P<0.05$) among each other in all of the *in situ* parameters tested for both sampling trips, except for Tm during the second trip. More than half of the stations violated the recommended level of pH 6.5–9.0 for treated water (NDQWS). Likewise, the Cn levels for all treated water stations were significantly different between each other ($P<0.05$). All stations of treated water were significantly different ($P<0.05$) between each other in Tb level, with the highest mean value of 4.50 ± 0.00 NTU at station T3 during the

first trip and 5.17 ± 0.06 NTU in the second trip. Table 3 shows the mean values of $\text{NH}_3\text{-N}$, COD, TS and TP at raw water stations. All samples exceeded the recommended value for COD level (10 mg/L). Means for both TS and TP from peat raw water stations were all significantly different from non-peat raw water station R4 though they were all below or within Class IIA.

Trace metals were tested in both raw water and treated water in this study. Table 4a shows mean values of trace metals for raw water. During both trips, for raw water samples, station R2 showed alarming levels of Al (Class V), ranging from 2.133 ± 0.122 to 4.784 ± 0.565 mg/L, and they were significantly different from the rest of the stations ($P<0.05$). Table 4b shows the mean values of trace metals for treated water sampled from four outlets of WTPs drawing water from the sampled stations of raw water. Samples at Station T1 to T3 were pipe water treated from peat water sources, whilst station T4 from non-peat source. There was a significant difference in Al level between all of the stations ($P<0.05$). Comparing treated water with its raw water, all of the stations showed a significant increase in the level of Al in treated water ($P<0.05$), except for station 4 on the first trip and station 2, on both trips.

Table 2a: Mean values of Cn, DO, pH, Tm and Tb of raw water at the five sampling stations

Parameter	Station	Trip 1	Trip 2	Mean \pm SD
Cn (\square S/cm)	R1	171.33 \pm 0.06 ^a	172.40 \pm 0.00 ^a	171.87 \pm 0.59 ^a
	R2	110.87 \pm 0.06 ^b	112.10 \pm 0.00 ^b	111.48 \pm 0.68 ^b
	R3	206.67 \pm 1.53 ^c	208.67 \pm 0.58 ^c	207.67 \pm 1.51 ^c
	R4	102.00 \pm 0.00 ^d	100.33 \pm 0.58 ^d	101.17 \pm 0.98 ^d
	R5	105.43 \pm 0.06 ^c	106.33 \pm 0.06 ^c	105.83 \pm 0.50 ^c
DO (mg/L)	R1	†1.27 \pm 0.06 ^a	†1.80 \pm 0.10 ^a	†1.53 \pm 0.30 ^a
	R2	‡3.27 \pm 0.55 ^b	‡3.53 \pm 0.31 ^b	‡3.40 \pm 0.42 ^b
	R3	‡3.30 \pm 0.10 ^b	‡3.43 \pm 0.06 ^b	‡3.37 \pm 0.10 ^b
	R4	#7.53 \pm 0.06 ^c	#7.23 \pm 0.06 ^c	#7.38 \pm 0.17 ^c
	R5	†1.27 \pm 0.06 ^a	†1.70 \pm 0.10 ^a	†1.48 \pm 0.25 ^a
pH	R1	*3.48 \pm 0.01 ^a	*3.99 \pm 0.01 ^a	*3.74 \pm 0.28 ^a
	R2	*4.01 \pm 0.02 ^b	*4.05 \pm 0.00 ^b	*4.03 \pm 0.02 ^b
	R3	*4.02 \pm 0.01 ^b	*4.05 \pm 0.01 ^b	*4.04 \pm 0.02 ^b
	R4	7.18 \pm 0.03 ^c	7.10 \pm 0.00 ^c	7.14 \pm 0.05 ^c
	R5	*3.83 \pm 0.01 ^a	*4.06 \pm 0.04 ^b	*3.95 \pm 0.13 ^b
Tm(\square C)	R1	30.47 \pm 0.06 ^a	29.70 \pm 0.00 ^a	30.08 \pm 0.42 ^a
	R2	29.87 \pm 0.06 ^b	29.80 \pm 0.00 ^b	29.83 \pm 0.05 ^a
	R3	32.67 \pm 0.15 ^c	30.60 \pm 0.00 ^c	31.63 \pm 1.14 ^b
	R4	32.50 \pm 0.00 ^c	31.20 \pm 0.00 ^d	31.85 \pm 0.71 ^b
	R5	30.10 \pm 0.00 ^d	29.80 \pm 0.00 ^b	29.95 \pm 0.16 ^a
Tb (NTU)	R1	1.74 \pm 0.05 ^a	1.67 \pm 0.01 ^a	1.70 \pm 0.05 ^a
	R2	7.54 \pm 0.09 ^b	8.19 \pm 0.01 ^b	7.87 \pm 0.36 ^a
	R3	35.00 \pm 0.00 ^c	34.00 \pm 2.00 ^a	34.50 \pm 1.38 ^b
	R4	115.00 \pm 0.00 ^d	137.33 \pm 3.06 ^d	126.17 \pm 12.38 ^c
	R5	1.70 \pm 0.03 ^a	1.51 \pm 0.01 ^a	1.60 \pm 0.11 ^a

Note: Means within a column followed by the same letters are not significantly different at 5% level. *Violation of the standard values according to the NDWQS raw water quality criteria. Parameters which are not stated in NDWQS are classified based on DOE NWQS water quality classification and are indicated with symbols: #Class I, ‡Class III, †Class IV.

Table 2b: Mean values of Cn, DO, pH, Tm and Tb of treated water at the four sampling stations

Parameter	Station	Trip 1	Trip 2	Mean \pm SD
Cn (\square S/cm)	T1	200.10 \pm 0.00 ^a	190.30 \pm 0.00 ^a	195.20 \pm 5.37 ^a
	T2	130.40 \pm 0.00 ^b	135.20 \pm 0.00 ^b	132.80 \pm 2.63 ^b
	T3	242.10 \pm 0.00 ^c	238.30 \pm 0.00 ^c	240.20 \pm 2.08 ^c
	T4	135.00 \pm 0.00 ^d	139.60 \pm 0.00 ^d	137.30 \pm 2.52 ^b
DO (mg/L)	T1	†5.17 \pm 0.06 ^a	†5.23 \pm 0.06 ^a	†5.20 \pm 0.06 ^a
	T2	†6.40 \pm 0.00 ^b	†6.43 \pm 0.06 ^b	†6.41 \pm 0.04 ^b
	T3	†6.77 \pm 0.12 ^c	†6.83 \pm 0.06 ^c	†6.80 \pm 0.09 ^c
	T4	‡12.57 \pm 0.06 ^d	‡12.57 \pm 0.06 ^d	‡12.57 \pm 0.05 ^d
pH	T1	*6.19 \pm 0.02 ^a	*6.00 \pm 0.00 ^a	*6.10 \pm 0.10 ^a
	T2	6.90 \pm 0.00 ^b	*6.27 \pm 0.06 ^b	6.58 \pm 0.35 ^a
	T3	*6.00 \pm 0.00 ^c	6.57 \pm 0.06 ^c	*6.28 \pm 0.31 ^a
	T4	7.20 \pm 0.00 ^d	*5.59 \pm 0.02 ^d	*6.39 \pm 0.88 ^a
Tm (\square C)	T1	29.00 \pm 0.00 ^a	28.07 \pm 0.06 ^a	28.53 \pm 0.51 ^a
	T2	28.00 \pm 0.00 ^b	28.00 \pm 0.00 ^a	28.00 \pm 0.00 ^a
	T3	30.00 \pm 0.00 ^c	31.13 \pm 0.06 ^b	30.57 \pm 0.62 ^b
	T4	28.20 \pm 0.00 ^d	29.00 \pm 0.00 ^c	28.60 \pm 0.44 ^a
Tb (NTU)	T1	2.33 \pm 0.01 ^a	4.10 \pm 0.00 ^a	3.22 \pm 0.97 ^a
	T2	2.50 \pm 0.00 ^b	4.79 \pm 0.01 ^b	3.65 \pm 1.26 ^a
	T3	4.50 \pm 0.00 ^c	3.47 \pm 0.06 ^c	3.98 \pm 0.57 ^a
	T4	1.00 \pm 0.00 ^d	*5.17 \pm 0.06 ^d	3.08 \pm 2.28 ^a

Note: Means within a column followed by the same letters are not significantly different at 5% level. *Violation of the standard values according to the NDWQS treated water quality criteria. Parameters which are not stated in NDWQS are classified based on DOE NWQS water quality classification and are indicated with symbols: ‡Class I, †Class IIA.

Table 3: Mean values of NH3-N, COD, TS, and TP of raw water at the five sampling stations

Parameter	Station	Trip 1	Trip 2	Mean±SD
NH3-N (mg/L)	R1	‡0.11±0.00 ^a	‡0.22±0.00 ^{ab}	‡0.17±0.06 ^a
	R2	‡0.30±0.06 ^b	‡0.30±0.06 ^b	‡0.30±0.06 ^b
	R3	‡0.22±0.00 ^{bc}	‡0.17±0.06 ^a	‡0.20±0.05 ^a
	R4	‡0.15±0.06 ^{ac}	‡0.15±0.06 ^a	‡0.15±0.06 ^a
	R5	‡0.11±0.00 ^a	‡0.11±0.00 ^a	‡0.11±0.00 ^a
COD (mg/L)	R1	*87.7±1.5 ^a	*125.0±3.0 ^{ab}	*106.3±20.6 ^a
	R2	*153.3±3.1 ^b	*130.3±3.2 ^a	*141.8±12.9 ^b
	R3	*160.0±1.0 ^c	*122.3±3.2 ^b	*141.2±20.7 ^b
	R4	*15.3±1.2 ^d	*10.7±1.2 ^c	*13.0±2.8 ^c
	R5	*114.0±2.0 ^c	*76.7±1.5 ^d	*95.3±20.5 ^a
TS (□g/L)	R1	13.7±0.6 ^a	13.0±0.0 ^a	13.3±0.5 ^a
	R2	11.7±0.6 ^b	11.0±0.0 ^a	11.3±0.5 ^a
	R3	12.0±0.0 ^b	12.0±0.0 ^a	12.0±0.0 ^a
	R4	1.3±0.6 ^c	1.0±0.0 ^b	1.2±0.4 ^b
	R5	14.7±0.6 ^a	18.0±3.6 ^c	16.3±2.9 ^c
TP (□g/L)	R1	‡226.7±5.8 ^a	‡213.3±5.8 ^a	‡220.0±8.9 ^a
	R2	60.0±0.0 ^b	56.7±5.8 ^b	58.3±4.1 ^b
	R3	193.3±23.1 ^c	223.3±5.8 ^a	‡208.3±22.3 ^a
	R4	20.0±0.0 ^d	20.0±0.0 ^c	20.0±0.0 ^c
	R5	66.7±5.8 ^b	60.0±0.0 ^b	63.3±5.2 ^b

Note: Means within a column followed by the same letters are not significantly different at 5% level. *Violation of the standard values according to the NDWQS raw water quality criteria. Parameters which are not stated in NDWQS are classified based on DOE NWQS water quality classification and are indicated with symbol: ‡Class IIA.

Table 4a: Mean values of trace metals (mg/L) of raw water at the five sampling stations

	St	Trip 1	Trip 2	Mean±SD		St	Trip 1	Trip 2	Mean±SD
Al	R1	0.025±0.006 ^a	†0.340±0.038 ^{ab}	†0.182±0.174 ^a	Fe	R1	*1.277±0.015 ^a	0.703±0.008 ^a	0.990±0.315 ^a
	R2	*2.133±0.112 ^b	*4.784±0.565 ^c	*3.458±0.150 ^b		R2	*2.292±0.038 ^b	0.294±0.023 ^a	*1.293±1.095 ^{ab}
	R3	0.025±0.003 ^a	*0.898±0.003 ^b	†0.462±0.480 ^a		R3	*4.594±0.028 ^c	*1.509±0.014 ^b	*3.051±1.690 ^b
	R4	0.028±0.000 ^a	†0.150±0.042 ^a	0.089±0.072 ^a		R4	*1.527±0.016 ^d	*4.172±0.088 ^c	*2.850±1.450 ^{ab}
	R5	†0.465±0.131 ^c	*0.519±0.007 ^{ab}	†0.492±0.088 ^a		R5	*1.526±0.015 ^d	*1.373±0.437 ^b	*1.450±0.290 ^{ab}
Cd	R1	0.003±0.000 ^a	0.003±0.001 ^a	0.003±0.000 ^a	Pb	R1	0.038±0.013 ^a	0.008±0.001 ^a	0.023±0.018 ^a
	R2	0.003±0.000 ^a	*0.007±0.001 ^b	*0.005±0.002 ^{ac}		R2	0.046±0.009 ^a	0.006±0.001 ^a	0.026±0.022 ^a
	R3	0.003±0.000 ^a	*0.011±0.001 ^c	*0.007±0.004 ^{bc}		R3	0.047±0.009 ^a	0.007±0.001 ^a	0.027±0.023 ^{ab}
	R4	0.003±0.000 ^a	*0.009±0.001 ^d	*0.006±0.003 ^{bc}		R4	*0.108±0.003 ^b	0.003±0.001 ^b	*0.056±0.058 ^b
	R5	0.003±0.000 ^a	*0.005±0.000 ^c	*0.004±0.001 ^{ab}		R5	0.034±0.001 ^a	0.006±0.001 ^{ab}	0.020±0.016 ^a
Cr	R1	*0.069±0.003 ^a	*0.065±0.003 ^a	*0.067±0.003 ^a	Mn	R1	0.044±0.004 ^a	0.023±0.004 ^a	0.034±0.012 ^a
	R2	0.005±0.000 ^b	0.005±0.000 ^b	0.005±0.000 ^b		R2	0.061±0.001 ^b	0.015±0.003 ^a	0.038±0.025 ^a
	R3	0.008±0.001 ^b	0.012±0.001 ^c	0.010±0.002 ^c		R3	0.158±0.000 ^c	0.044±0.004 ^b	0.101±0.063 ^b
	R4	0.005±0.000 ^b	0.005±0.000 ^b	0.005±0.000 ^b		R4	0.079±0.000 ^d	0.071±0.000 ^c	0.075±0.005 ^{bc}
	R5	0.026±0.002 ^c	0.027±0.001 ^d	0.027±0.001 ^c		R5	0.061±0.002 ^b	0.021±0.006 ^a	0.041±0.022 ^{cc}
Cu	R1	0.002±0.000 ^a	0.005±0.000 ^a	0.003±0.002 ^a	Zn	R1	0.059±0.016 ^a	0.010±0.001 ^a	0.035±0.029 ^a
	R2	0.004±0.001 ^b	0.005±0.000 ^a	0.004±0.001 ^a		R2	0.058±0.001 ^a	0.003±0.000 ^b	0.031±0.030 ^a
	R3	0.021±0.001 ^c	0.019±0.005 ^b	0.020±0.003 ^b		R3	0.105±0.002 ^b	0.043±0.002 ^c	0.074±0.034 ^{ab}
	R4	0.004±0.001 ^b	0.008±0.004 ^a	0.006±0.003 ^a		R4	0.026±0.004 ^c	0.048±0.001 ^c	0.037±0.012 ^a
	R5	0.003±0.000 ^{ab}	0.004±0.000 ^a	0.004±0.000 ^a		R5	0.167±0.006 ^d	0.018±0.003 ^d	0.092±0.082 ^b

Note: Means within a column followed by the same letters are not significantly different at 5% level. *Violation of the standard values according to the NDWQS raw water quality criteria. Parameters which are not stated in NDWQS are classified based on DOE NWQS water quality classification and are indicated with symbol: †Class IV, ‡ Class IIA, □Class V. No Al standard limit was set for raw water in NDWQS.

Table 4a: Mean values of trace metals (mg/L) of raw water at the five sampling stations

	St	Trip 1	Trip 2	Mean□SD		St	Trip 1	Trip 2	Mean±SD
Al	T1	*1.577±0.160 ^a	*0.950±0.047 ^a	*1.264±0.360 ^a	Fe	T1	*0.875±0.037 ^a	0.004±0.000 ^a	*0.440±0.478 ^a
	T2	0.028±0.000 ^b	*1.668±0.095 ^b	*0.848±0.900 ^a		T2	*0.548±0.010 ^b	0.004±0.000 ^a	*0.276±0.298 ^b
	T3	*8.008±0.563 ^c	*1.141±0.094 ^a	*4.574±3.778 ^b		T3	*0.623±0.026 ^b	0.004±0.000 ^a	*0.314±0.340 ^{ab}
	T4	0.028±0.000 ^b	*0.441±0.045 ^c	*0.235±0.228 ^a		T4	*0.439±0.038 ^c	0.004±0.000 ^a	0.221±0.240 ^{ab}
Cd	T1	0.003±0.000 ^a	*0.008±0.001 ^a	*0.005±0.003 ^a	Pb	T1	*0.047±0.002 ^a	*0.013±0.000 ^a	*0.030±0.018 ^a
	T2	0.003±0.000 ^a	*0.005±0.000 ^b	*0.004±0.001 ^b		T2	*0.048±0.006 ^{ab}	*0.013±0.000 ^a	*0.031±0.020 ^a
	T3	0.003±0.000 ^a	*0.008±0.000 ^a	*0.005±0.003 ^a		T3	*0.059±0.004 ^b	0.004±0.002 ^b	*0.032±0.030 ^a
	T4	0.003±0.000 ^a	*0.009±0.000 ^a	*0.006±0.003 ^a		T4	*0.046±0.003 ^a	0.006±0.003 ^b	*0.024±0.022 ^a
Cr	T1	0.005±0.000 ^a	0.005±0.000 ^a	0.005±0.000 ^a	Mn	T1	0.062±0.001 ^a	0.012±0.001 ^a	0.037±0.028 ^a
	T2	0.005±0.000 ^a	0.023±0.007 ^a	*0.014±0.011 ^a		T2	0.064±0.000 ^a	0.021±0.001 ^b	0.042±0.024 ^a
	T3	0.005±0.000 ^a	*0.180±0.068 ^b	*0.093±0.105 ^b		T3	*0.126±0.001 ^b	0.023±0.001 ^b	0.074±0.057 ^b
	T4	0.005±0.000 ^a	0.018±0.006 ^a	0.012±0.008 ^a		T4	0.044±0.002 ^c	0.002±0.000 ^c	0.023±0.023 ^a
Cu	T1	0.004±0.001 ^a	0.005±0.000 ^a	0.004±0.001 ^a	Zn	T1	0.076±0.000 ^a	0.001±0.000 ^a	0.039±0.041 ^{ab}
	T2	0.005±0.000 ^a	0.005±0.000 ^a	0.005±0.000 ^a		T2	0.097±0.012 ^b	0.003±0.000 ^b	0.050±0.052 ^a
	T3	0.005±0.000 ^a	0.005±0.000 ^a	0.005±0.000 ^a		T3	0.080±0.000 ^{ab}	0.003±0.000 ^b	0.042±0.042 ^{ab}
	T4	0.005±0.000 ^a	0.005±0.000 ^a	0.005±0.000 ^a		T4	0.046±0.002 ^c	0.003±0.001 ^b	0.025±0.024 ^{ab}

Note: Means within a column followed by the same letters are not significantly different at 5% level. *Violation of the standard values according to the NDWQS treated water quality criteria.

Discussion

Overall Status

The overall status of peat water investigated in this study compared to other peat swamp waters studied in Malaysia is depicted in Table 5. The values of Tb, pH, DO, Cn, Tm, NH₄-N, as compared to other peat swamp waters in Malaysia, are similar. Based on the results obtained, the most notable parameters that violated the recommended raw water quality of NDWQS are pH and DO. Both parameters are below the recommended value. As depicted in Table 5, low pH and thus acidic water condition

are typical characteristics of peat swamp water. In water treatment, pH is an important factor in determining the efficiency of chemical treatment, as most coagulant used has an optimum pH working range of above 6.9. Oxygen level is also important as higher oxygen level in water helps to eliminate odor. Further relation of peat water characteristics to treated water quality are discussed in other sections of this discussion.

Comparing Al of non-peat raw water (R4) with the baseline value of 0.34 mg/L at Baleh River (Sim *et al.*, 2016), both trips in the present study showed much lower values.

Table 5: Selected water quality values for peat swamp waters in Malaysia

Study	Tb	pH	DO	Cn	Tm	NH ₃ -N
Beamish <i>et al.</i> (2003)	1.0 – 53.4	3.4 – 5.5	2.2 – 6.1	0.022 – 0.168	26 – 28	0 – 0.29
Yule & Gomez (2009)	N.D.	2.6 – 3.8	1.8 - 16	N.D.	25 – 32	N.D.
Rahim <i>et al.</i> (2009)	N.D.	4.55	1.15	0.021	26.6	0.81
Gasim <i>et al.</i> (2007)	1.5 – 17.2	3.53 – 4.55	0.5 – 1.76	0.053 – 0.062	26.2 – 28.9	N.D.
Rosli <i>et al.</i> (2010)	1.27 – 5.33	3.74 – 3.80	3.02 – 4.34	0.063 – 0.088	27.69 – 30.07	0.18 – 0.32
Irvine <i>et al.</i> (2013)	1.2	3.63	0.31	0.083	27.3	<0.5
The present study	1.60 – 7.87	3.74 – 4.04	1.48 – 3.40	0.105 – 0.207	29.83 – 31.63	0.11 – 0.30

(Modified from Irvine *et al.*, 2013); N.D.–Not determined.

Aluminium levels in raw water and treated water

Based on the results of this study, Al levels from all the stations for treated water violated the NDWQS limit of 0.2 mg/L. However, mean Al of treated water from peat source, showed significant difference from the permissible limit of 0.2 mg/L (one sample *t*-test, $P < 0.05$), with an overall increase of 63% in Al level from the raw water used. Water treated from non-peat source still violated the permissible limit, but is not significantly different (one sample *t*-test, $P = 0.089$). Also, in both trips, Peat water stations 1 and 3 both show significant increases ($P < 0.05$) in Al levels compared to before

treatment. With the use of Al based coagulants in water treatment, it is impossible not to have some low level of Al in treated water, and as a consequence of Al-based chemical treatment of surface sources, the level of Al in the treated water were often higher than those in raw water (Diaconu *et al.*, 2009). From the results, the use of Al based coagulants for peat waters may not be as efficient as its use in the treatment for non-peat waters. Comparing the present results with those of Qaiyum *et al.* (2011), there was no significant difference from the maximum level of Al in treated water drawn from non-peat source (one sample *t*-test, $P = 0.091$), but there was a significant difference from treated water drawn from peat sources (one sample *t*-test,

$P < 0.05$). Before the treatment, the level of Al in raw water of peat source was already above the permissible limit for drinking water. Although there are more sources of aluminum in food uptake for example, Al in drinking water has been postulated to be more readily available for biological uptake as compared to Al from other sources as the chemical treatment introduces a significant amount of dissolved bioavailable Al (Schintu *et al.*, 2000; Diaconu *et al.*, 2009).

Key influence of Aluminium occurrences

The significant difference of Al levels in treated water could be explained by the diverse properties of the raw water itself. There was a significant difference ($P < 0.05$) between the means of Tm in peat and non-peat raw water stations. Tm of water is an important factor in determining Al solubility (Schintu *et al.*, 2000; Diaconu *et al.*, 2009). At lower temperature (4°C), the pH of minimum solubility increases, resulting in alum coagulation and hence resulting in higher residual Al levels (Srivinasan *et al.*, 1999).

In the second sampling trip, the temperature for non-peat raw water was significantly higher than the peat raw waters. This is also when Al levels in the non-peat water station was significantly lower than the rest of the peat water station. The treated water pH, the pH of coagulation and the temperature and turbidity of water were found to have an effect on the amount of residual aluminum (Driscoll & Letterman, 1995). The amount of residual aluminum is high when raw water is cold even if the raw water KMnO_4 and poly-aluminum chloride (PAC) dosages are at low levels. When the raw water temperature is higher and raw water KMnO_4 rises, the amount of residual aluminum is relatively low. This shows that the effectiveness of the water treatment process is better when raw water is warmer which is very common in most of the WTP (Tomperi, 2012). Station 2 had the highest mean levels of Al and $\text{NH}_3\text{-N}$, and as the correlation results show, Al correlates positively with $\text{NH}_3\text{-N}$. According to experiments conducted by Lidon *et al.* (1998),

the presence of Al modulates nitrate to ammonia reduction in plants as the Al concentration in plants increases. Excess Al resulting from increased membrane permeability decreases the rate of nitrate uptake possibly through inhibition of the activity of nitrate transporters. Higher root Al concentrations are possibly associated with increased binding of Al to the membrane channel proteins or components of the induction system for net nitrate assimilation. This would increase the membrane permeability and decrease both the acidification capacity and the concurrent root nitrate concentrations (Lidon *et al.*, 1998). Thus, Al occurrences may have indirectly increased due to the use of nitrate-based fertilizers such as ammonium nitrate, in the agricultural activities nearby.

Iron can be found in natural waters at levels ranging from 0.5 to 50 mg/L and Mn usually occurs together with this element (WHO, 1993). No industrial activities were seen near sampling stations. Other anthropogenic sources in raw water include surface water runoff from discarded batteries and agricultural products (WHO, 1993). There are significant differences in Fe levels among stations of peat and non-peat raw water stations (Table 4a). All stations for peat water sources are surrounded by villages and a wide range of small scale agricultural activities such as fruit trees, paddy fields, sago, rubber trees and oil palm (except for station R3), the same goes for the station of non-peat source with an addition of larger scale sago plantation located downstream of the river intake, and oil palm plantation outside the buffer zone of catchment area. The higher levels of Fe found in peat raw water as compared to the non-peat raw water station could be due to the water flow condition of the peat swamp catchment. All peat raw water catchment area in this study are rather stagnant or have very low water flow. As for the non-peat raw water catchment, water flows are faster, thus increasing oxygen levels naturally.

Fine soil may also carry some naturally occurring trace metals. This study also observed significantly higher Tb (Table 2a) in the non-

peat station compared to the peat water stations. Erosion from upstream of the non-peat raw water catchment had also caused low water level sometimes, forcing the water authority to draw some raw water from other sources which are partially mixed with peat water. Throughout the sampling trips for this study, however, no mixture of peat water was done during the operation. In relation to residual Al in peat water, Tb was significantly lower, thus more of the trace metals found in water may not be from the particulates but more of the dissolved form. Thus, this may have caused the efficiency of the treatment to be slightly lower because in order for dissolved elements to form floc during treatment, it may require better mixing of coagulants for the reaction to be completed, while in the treatment plants for the peat water, only conventional treatment with manual and hydraulic mixing of water treatment chemicals was applied. It has also been established that the treatment with Al sulphate removes most of Al associated with particular matter (Diaconu *et al.*, 2009).

Both Fe and Mn can also occur at high concentrations in some source waters that are anaerobic (Fawell & Nieuwenhuijsen, 2003). This is supported by some significantly lower level of DO in peat water compared to the non-peat source (Table 2a). Low oxygen levels in water lowers the chance of Fe and Mn to be oxidized, and thus increases the color in raw water, leading to higher doses of coagulants needed to remove the color in water treatment process. It was also noted that all the WTPs using peat water source in this study did not provide structure for aeration in their treatment. However, in the conventional water treatment process, Mn probably was not efficiently oxidized from raw water only by aeration process.

According to Berkowitz *et al.* (2005), surface water pH plays an important role in alum treatment effectiveness and environmental impacts because the resultant distribution of Al phases and floc quality are pH-dependent.

Although the correlation analysis shows no significant relationship (Bivariate, $P > 0.05$), it is noted that violations of pH level (below pH 5.5) on all samples of raw peat water from this study could play a major role in the high levels of Al in treated water. These results correspond with a statement made from a previous study that the Al content in finished water is a function of the pH of water as it is an important factor in determining Al solubility and consequently residual aluminum in the treated water (Diaconu *et al.*, 2009). This is because inorganic coagulants will decrease the alkalinity of water, thus the pH of the chemically-dosed raw water will decrease. Al sulphate tends to work best at a dosed-water pH of 5.8-6.5. If the pH is lower or higher than the optimum range, then problems of high residual Al may occur.

Among the peat raw water stations, Station R3 showed the highest in most of the trace metals analyzed (Cd, Cr, Cu, Fe, Mn and Zn, Trip 2). This could be due to the highest Tb found in this station. Tb that arise from particulates suspended or dissolved in the water, may be the main cause of the higher concentrations of most trace metals at this station. Suspended matter work as a trap for trace metals. Tb will also increase water Tm due to the increased heat absorption by the water (Ridanović *et al.*, 2010). The results also supported this, as station 3 had the highest Tm and were significantly higher than the rest of the peat raw water stations. The increased heat was also most probably due to the turbidity levels as all peat raw water stations have similar amounts of canopy cover. In addition, at Station 4, the non-peat raw water station showed the highest mean levels of Pb. Contamination of Pb in this case could be due to discharge from agricultural activities. Station R5 had shown the highest level of Zn, and according to the results, this station also had the highest level of TS compared to the rest of the stations. Based on a study by Saarinen *et al.* (2013), the increasing oxidation of sulphidic materials can considerably lower the pH and increase metal concentrations (maximum Al, Cu and Zn concentrations of 1010, 257 and 186 µg/L, respectively) in runoff waters. Station

R5 also, based on the soil characteristics, had thicker organic layer (>150 cm) compared to soils at other sampling stations.

Based on Table 2b, the mean Cn level shows significant difference between the treated water stations. A slight increase in Cn level in treated water as compared to before treatment was because treatment chemicals used introduce other inorganic dissolved solids such as calcium (with the use of calcium hypochlorite) and Al cations (with the use of Al-based coagulants). Raw water that is nearer to the sea may have higher Cn level due to the presence of chloride such as in station R3, thus its treated water station T3 also have higher Cn.

Results of this study showed Al has negative correlation with pH in treated water. For the first sampling trip, violation of pH value at station T1 and T3 showed level of Al to be the highest during that time as compared to the other stations. High Al concentration in drinking water affects Tb (Srivinasan *et al.*, 1999; Kvech & Edwards, 2001) also because suspended Al was the major species at pH below 7.5 (Wang *et al.*, 2010). The treated water pH, the pH of coagulation and the Tm and Tb of water as mentioned by Driscoll & Letterman (1995), were found to have an effect on the amount of residual aluminum in finished water.

All Pb levels in treated water during the first trip violated the required standard regardless of the source. But, all the peat sources-treated water increased in Pb levels as compared to before treatment. For non-peat source treated water, the mean level of Pb decreases despite being still above the permissible limit. This shows that treatment of non-peat water is more efficient in reducing Pb levels compared to the

treatment of peat waters. As mentioned earlier, conventional treatment using chemicals require optimum pH range (5.8 – 6.5) in order for water treatment coagulants to work efficiently in removing turbidity and other contaminants, but for peat swamp raw water, this pH range is difficult to achieve as the removal of color in typically highly colored peat water requires lower pH range (5.0 - 5.5), and this could lead to lower efficiency in removing other contaminants such as Pb.

According to Tyagi & Mehra (2000), in most countries, public water supplies rarely contain clinically significant levels of Pb. Lead is present in tap water, to some extent as a result of its dissolution from natural sources, but primarily from household plumbing systems containing Pb in aged pipes, solder, or pipe fittings (WHO, 1993). From the increased levels of Pb in all the three treated water from peat source, it may indicate corroded system in the WTPs. No lead pipes are used in the treatment system but, Pb can also corrode from metal faucets and fixtures made from brass, an alloy of copper and zinc that often contains lead impurities, including chrome-plated brass fixtures. The amount of lead corroded from metal plumbing generally increases as water corrosivity increases. Water corrosivity is controlled primarily by the water's acidity. Low pH of peat waters may have contributed to the corroded system. In relation to Al levels with higher levels of other trace metals in treated water, it may correlate because high Al concentration leads to pipe wall deposition (Wang *et al.*, 2009). This is also supported by a significant increase of Cd at station T1 and Mn levels at station T2 during the second trip. Table 6 shows the discussion summary of the key influences of Al occurrences.

Table 6: Summary of key influences of Aluminium occurrences

Parameter		Raw water		Treated water
pH	-	Although no statistical correlation was found, violations of pH level (below pH 5.5) on all samples of raw peat water from this study could play a major role on the high levels of Al as it is an important factor in determining Al solubility	↓pH ↑Al	Lower pH increases Al solubility
Fe	↑Fe ↑Al	Peatland areas are often characterized by high Fe – Al complexes	-	
NH3-N	↑NH3-N ↑Al	Al occurrences may have indirectly increased the use of nitrate based fertilizers such as ammonium nitrate	-	-
DO	↓DO ↑Al	Fe also occur at high concentrations in some source waters that are anaerobic	-	No statistical correlation found in treated water but, low oxygen level in water lowers the chance of Fe to be oxidized and thus increases the color in raw water, leading to higher doses of Al based coagulants needed to remove color in water treatment process
Pb	-		↑Pb ↑Al	Low pH of peat waters may have contributed to the corroded system also, high Al leads to pipe wall deposition leading to higher Pb levels in treated water
Tb	-		↑Tb ↑Al	High Al in drinking water affects Tb because suspended Al was the major species at pH below 7.5; also, suspended matter work as a trap for trace metals

Conclusion

This study shows that both treated and raw water sources have high levels of Al. There is no hard evidence showing peat water sources have higher levels of Al than the non-peat raw water source. However, results show that treated water from two peat water sources were significantly higher in mean levels of Al as compared to raw water samples in both sampling trips. Treated water from non-peat source was also significantly

lower in mean levels of Al than the treated water from peat sources. Peat swamp water typical characteristics such as low pH have an impact on the levels of Al in its treated water, mainly due to the higher solubility of Al in acidic condition with the use of Al-based coagulants in its water treatment processes. Besides that, the low dissolved oxygen levels further add to the difficulty of treatment process as the existence of higher concentration of metals such as Fe and Mn reduces the efficiency of Al-based coagulants

in coagulation and flocculation processes. As such, the treatment of raw peat water may pose a challenge especially in rural treatment plants where application of technological advancement is limited. Considerations need to be made on the continued use of Al-based chemicals in water treatment for drinking purposes.

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