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AQUATIC ORGANIC MATTER CHARACTERISTICS AND THMFP OCCURRENCE IN A TROPICAL RIVER

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ABSTRACT: Dissolved Organic Matter (DOM) in water sources is reactive to chlorine, forming Trihalomethanes (THMs). However, information on DOM characteristics and Trihalomethanes Forming Potential (THMFP) in tropical surface raw water sources is still limited. This study aimed to determine the characteristics of Total Organic Matter (TOM) and DOM, the correlations between the pair, as well as the correlations between DOM and THMFP during dry and rainy seasons in a polluted tropical river in Indonesia. Samples were collected from Cikapundung River, at the intake of drinking water treatment plant during both seasons. TOM and DOM were measured as COD, DOC, Chromophoric organic (A_{254}, A_{355}, A3/4) and Specific Ultra Violet Absorbance (SUVA). Total THMFP (TTHMFP) was determined from $CHCl_3$, $CHBrCl_2$, $CHClBr_2$, and $CHBr_3$ measurements. Compared to the rainy seasons, the values of all organic parameters at the intake during the dry season were higher. DOM in the dry season was more aromatic, hydrophobic and humic, consistent with higher concentrations of TTHMFP in the dry season ($\overline{X}$=636.79µg/L) than that in the rainy season ($\overline{X}$=430.13µg/L). TOM and DOM correlations for A_{254}, A_{355}, and COD in the dry season were stronger than in the rainy season. Each THMFP species was poorly correlated with any DOM parameters during the dry season, specifically at SUVA level ≥5L/mg/m. However, there were significant correlations between A_{254} and $CHBrCl_2$-FP; A_{355} and $CHCl_3$-FP; A_{355} and TTHMFP. These conditions were observed only during the rainy seasons at SUVA of 2-4L/mg/m. Thus, A_{355} can be used as a proxy of TTHMFP during the rainy season in Cikapundung river.

Keywords: Correlation, DOM characteristics, Seasonal variations, THMFP

1. INTRODUCTION

Organic matter is one of the main components in the natural water system and is ubiquitous in water sources as a result of both natural degradation of some organic substances and from human activities [1, 2]. One kind of organic matter in natural waters is Dissolved Organic Matter (DOM), which is defined as the fraction of organic matter that passes membrane filters of 0.1 – 0.7µm [3]. DOM existing in raw water for water treatment plant reacts with chlorine, generating Disinfection by Products (DBPs) such as Trihalomethanes (THMs) in drinking water that can pose significant health effects including cancer [4, 5].

Because of the heterogeneity of DOM properties, various surrogate parameters have been used to estimate its reactivity toward DBPs formation [6]. Common parameters to measure DOM in water body include Dissolved Organic Carbon (DOC), and Chromophoric DOM (CDOM). DOC is often synonymous with DOM since organic contaminants in natural systems generally represent a significant fraction of DOC [7], while CDOM is the fraction of DOM that absorbs Ultraviolet (UV)-Visible light, and is recognized as a controlling factor for the optical properties of surface waters [8]. Previous works also confirmed Specific UV Absorbents (SUVA) or UV absorbance normalized as DOC concentrations is a surrogate parameter of THMs due to its strong correlation with dissolved aromatic of the humic compound and reactive to chlorine [4, 9].

The majority of studies on seasonal changes of NOM were conducted in non-tropical regions. Few studies in tropical regions suggest that the intensity of precipitation may influence the characteristics of NOM in surface water [10]. In a pristine tropical river such as Epulu river (Republic Congo) and Maji ya Chai River (Tanzania), the concentrations of DOC measured during the rainy season were higher than those observed during the dry season [11, 12]. In these studies, the SUVA value and allochthonous humic compounds were high during the rainy season with high precipitation intensity [11, 12]. DOM was also found to be highly correlated with the potential for THMs formation (THMFP) in Bangpakong River, a polluted tropical river in

Thailand [13].

More efforts are still required to investigate the variation in DOM characteristics across seasons and its major role in THMs formation in other tropical regions. In Indonesia, the world's 4th most populated country with a huge demand for drinking water, information of concentrations and characteristics of DOM in its rivers are still limited. This includes, in Cikapundung River, a drinking water source for nearly two million inhabitants in Bandung City as the fifth biggest city in Indonesia. The number of organic matters in Cikapundung River was only determined by measuring the unfiltered water which potentially under-reported the organic matter concentrations [14]. In addition, the raw water quality standard for DOM concentration was not available. Therefore, it is important to characterize DOM fractions and to determine its correlation with Total Organic Matter (TOM). Furthermore, chlorination has been the main disinfection method in drinking water treatment in Indonesia for many years, suggesting the potential of organic matter to generate THMs in source water becomes critical.

The main objectives of this paper were to determine: (i) the characteristics of DOM and TOM ; (ii) the relationships between DOM and TOM; and (iii) the relationships between DOM and THMFP across two different seasons. Considering THMFP measurement is time-consuming and costly, it is expected that the observed relationships between DOM and each THMFP parameter can be used to determine surrogate parameters of THMFP in a polluted tropical river such as Cikapundung River.

To the best of our knowledge, this is the first study in Indonesia that investigates seasonal variations of DOM characteristics and its effect on THMFP conducted in a polluted river which is a representative of drinking water sources for cities in Java Island. Thereby, this study provides science-based information for controlling DOM in surface water and to minimize the formation of THMs in the drinking water.

2. MATERIALS AND METHODS

2.1 Location of Study

Cikapundung River is a tributary of Citarum River that is known as the longest river in Java Island. With a length of 28 km, the river divides the city of Bandung from the north to the south. This river is a typical urban river in a tropical and developing country of Indonesia. The upper stream flow of Cikapundung River ranges between 500–600L/s of water is extracted for the raw water supply of the city Water Treatment Plant (WTP). The concentrations of organic matters in the Cikapundung river were measured the highest among the other water sources to fulfill drinking water demand for the city of Bandung [15].

The annual average precipitation in this area was 191.54mm, with the highest intensity of rainfall occurs primarily between October to June. The maximum river flow usually occurs in the rainy seasons of 7.59m^3/s while the lowest occur in the dry season of 0.54m^3/s. Land use in the upper basin mainly comprises mixed anthropogenic area (80%), including farming, agriculture, wetlands, commercials, and residential areas. The remaining areas are occupied by forest area of 19% of the total catchment area (6,933.3Ha) (Fig.1), and the intake was located in this natural area (National Park of Djuanda). Among other rivers used as raw water sources for the city WTP, Cikapundung river has the worst water quality due to its high concentrations of organic matters originated from anthropogenic activities [15].

The main sampling site (point-1) was located at the Bantar Awi intake of drinking WTP (S $6^0$51'33,1'', E $107^0$38,57'57,1''). In addition, one reference site was located at the upstream of Cikapundung River of about 7 km from the intake at the upper stream part ((S 6^o50'19.12", E 107^o43'35.52") (point-2)). This reference site was surrounded by natural forestry and was used to identify potential contaminations in the area prior to the location of point-2. The study area and the selected sampling sites are illustrated in Fig.1.

2.2 Sampling and Analytical Methods

All samples were collected through grab sampling during both rainy and dry seasons. The study consist of two sub-studies: substudy-1 focused on the TOM and DOM characterization and TOM and DOM relationships, while the substudy-2 was conducted to determine DOM and THMFP relationships. In substudy-1, DOM and TOM concentrations and characteristics were measured from the samples collected in the dry season (14 – 23 August 2017, period 1), and two periods in the rainy season: period 1 (27 Jan – 13 Feb 2017) and period 2 (9 – 17 November 2017). Following, the DOM and THMFP samples were collected during the dry season (14 – 23 August 2017) and two periods in the rainy season: period 2 (9 – 17 November 2017 and period 3 (8 – 14 Feb 2018).

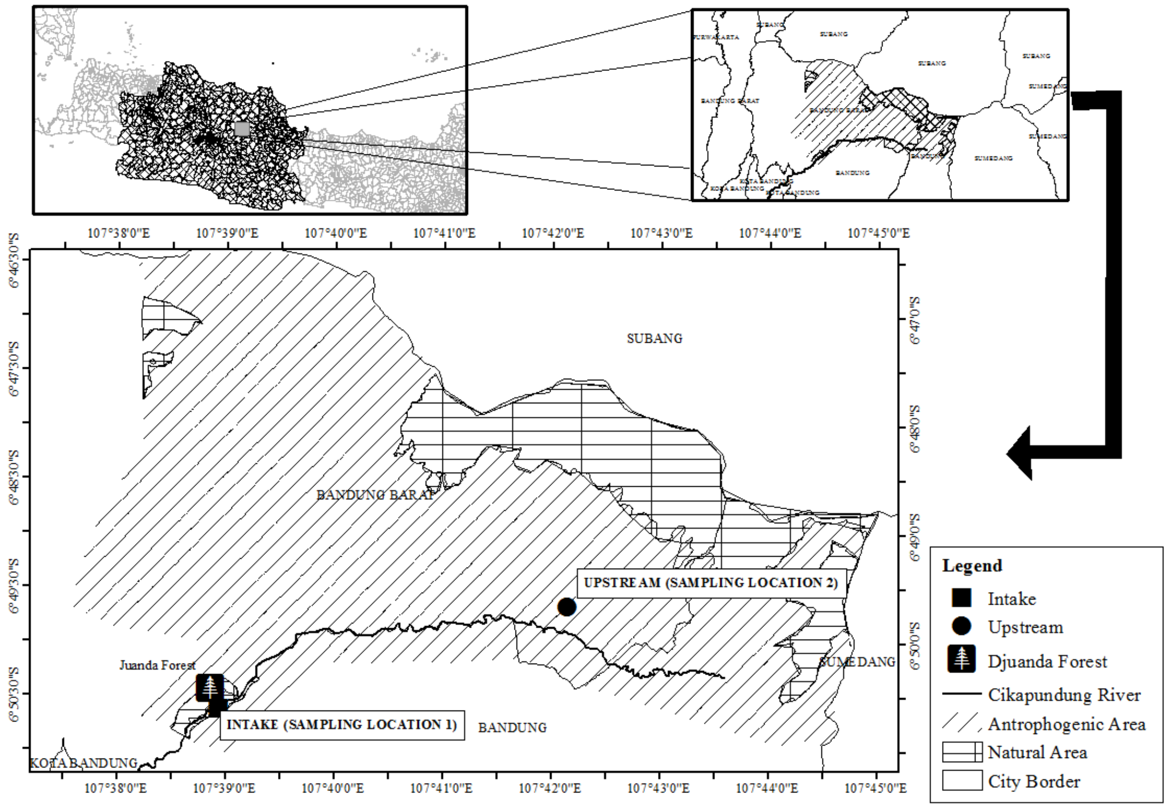


Fig.1 Study area and distribution of water sampling sites in Cikapundung River. The circle represents the sampling site at the upstream (point 2). The square represents the sampling site at the intake (point 1).

Samples at the upstream (point-2) were only collected during the rainy season period-1 due to limited availability of sampling equipment. Samples were placed in a 5L-polyethylene bottle, shipped in a cooled container and stored in a refrigerator at 4°C prior to analysis. Samples were prepared and analyzed in the Environmental Engineering Laboratory of Institut Teknologi Bandung and Integrated Laboratory of Politeknik Kesehatan Bandung. Measurement of pH and water temperature was carried out on-site during the sampling.

The characteristics of DOM were identified as Chemical Oxygen Demand (COD), DOC, and CDOM which were measured as UV absorbance at wavelength of 254 nm (A_{254}), UV absorbance at wavelength of 355 nm (A_{355}), ratio between A_{300} and A_{400} (A3/4), and ratio of absorbance at 254 to the concentrations of DOC (SUVA). A_{254} indicates humic and aromatic compounds in water, while A_{355} describes CDOM derived from terrestrial[16], and a ratio between A_{300} and A_{400} (A3/4) was measured to show the proportion of humic and fulvic acid [17]. DOC, A_{254}, and SUVA are surrogate parameters for THMFP in the alternative disinfectants and oxidants guidance manual [4]. While COD is listed in Indonesia standard for raw water quality (Government Regulation 82/2001), thus it is the most common parameter of NOM in raw water, measured by the water company in Indonesia. Other considerations that these parameters are easy to measure, relatively do not time-consuming, and cheaper than THMFP measurements.

These parameters were analyzed in both filtered (DOM) and unfiltered (TOM) samples. To obtain filtered samples, samples were passed through 0,45µm membrane prior to analysis. COD was analyzed based on the Standard Method protocol 5220C (close reflux method) [18]. Before reuse, glassware was cleaned first and heated at 450°C in the muffle furnace for 4 hours. The concentrations of DOC were measured using TOC Analyzer SIEVER Innovox 0545 with a high temperature of combustion, following the Standard methods 3510 B[18]. CDOM parameters were measured using a spectrophotometer (Shimadzu-1700 UV/Vis with a 1-cm quartz cell).

The potential of THMs formation in the filtered samples was measured according to the Standard Method 5710B [18]. The method included the addition of NaOCl and phosphate buffer for adjusting the pH of samples to be at 7, chlorinating samples with excess free chlorine, and incubating samples for 7 days at 25°C. The extraction of THMs was conducted based on USEPA 551.1 standard protocol (Liquid-liquid extraction by n-pentane). The extract was then analyzed by gas chromatography (GC 7890 A Agilent) and mass spectrophotometry (MS 5975 C). Standard Method 6232B was used for measurement using GC MS with splitless injection. THMs mixed Standard (Supelco /Sigma–Aldrich Co., USA) comprised of chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$), and dibromochloromethane ($CHClBr_2$), bromoform ($CHBr_3$) was used in this study.

3. RESULTS AND DISCUSSION

3.1 NOM Characteristics

The characteristics of water samples which include COD, A_{254}, A_{355}, A3/4, DOC, and SUVA were taken at point-1 and point-2 and the results are presented in Table 1 and 2, respectively. During each sampling period, the measured temperature of water samples ranged between 23 and 25^{0}C, and pH values were in a neutral range (6.7-7.1).

The measured average of DOM-COD and DOC levels at the point-2 (DOM-COD= 3.87mg/L; DOC=3.54mg/L) were lower than those measured at the point-1 (DOM-COD= 19.94mg/L; DOC=6.56mg/L) during the rainy period 1 (Table 1). The national standard for the quality of raw water sources in Indonesia does not regulate organic parameters except for COD. It was found that COD average concentration at the point-1 in dry and rainy seasons was above the national standard (10mg/L). One of the possible reasons was because the land use around the point-2 was dominated by natural forest, while the intake was surrounded by anthropogenic activities such as households, agricultural, and cow husbandry (within 7km) (see Fig. 1).

Higher concentrations of DOM-COD at the point-1 were potentially attributable to the direct discharges of cow manures biogas residues into the stream and domestic activities around the catchment. This comparison showed a logical fact that the catchments with more forests and less anthropogenic activities have better water quality in term of oxidizable pollutants and dissolved carbon.

Table 1 The characteristics of water samples from the point-1 in dry and rainy seasons

Parameters	Season					
	Dry (N = 9)		Rainy periode-1 (N = 9)		Rainy periode-2 (N = 9)	
	range	$\bar{X}$	range	$\bar{X}$	range	$\bar{X}$
TOM-COD(mg/L)	19.20-60.8	45.16	20.60-38.40	28.96	19.20-30.00	26.18
DOM-COD(mg/L)	13.60-38.4	25.18	13.60-27.70	19.94	11.20-30.00	19.12
TOM-A_{254}(cm^{-1})	0.35-0.58	0.43	0.25-0.38	0.31	0.35-0.58	0.43
DOM-A_{254}(cm^{-1})	0.21-0.48	0.35	0.15-0.22	0.19	0.06-0.26	0.19
TOM-A_{355}(cm^{-1})	0.11-0.39	0.24	0.11-0.26	0.18	0.08-0.22	0.13
DOM-A_{355}(cm^{-1})	0.08-0.35	0.18	0.03-0.06	0.05	0.05-0.13	0.08
TOM-A3/4	1.20-2.21	1.66	1.18-2.56	1.77	0.71-1.79	1.27
DOM-A3/4	1.70-2.80	2.23	2.60-4.30	3.47	1.15-2.94	2.28
DOC(mg/L)	4.26-8.66	6.98	4.20-9.35	6.56	3.02-5.50	4.36
SUVA(L/mg/m)	2.70-9.84	5.7	1.67-4.07	2.86	1.81-5.80	4.32

Table 2 The characteristics of water samples from the point-2 in the rainy season period 1 (N=9)

Parameters	Range	$\bar{X}$
DOM-COD(mg/L)	3.80-4.00	3.87
DOM-A_{254}(cm^{-1})	0.13-0.19	0.15
DOM-A_{355}(cm^{-1})	0.09-0.14	0.11
DOM-A3/4	1.14-1.25	1.18
DOC(mg/L)	2.81-4.00	3.54
SUVA(L/mg/m)	3.45-5.16	4.18

The average level of A_{355} at point-2 (0.11cm^{-1}) during the rainy season was larger than that measured for point-1 (0.05cm^{-1}), indicating DOM in the upstream area may be predominantly contributed by the natural decomposition of plants such as lignin [19]. Lignin is a very complex organic compound and mostly consists of aromatic compounds [20]. Although DOM at the point-1 was richer in oxidizable pollutants and dissolved carbon, the changes in A_{254} levels in both locations were not significant. The results indicated the anthropogenic activities during the rainy season along the water body between the two points did not contribute significantly to chromophoric organic. Compared to other studies of tropical rivers such as in Maji ya Chai river, Tanzania [11] and Central Kalimantan peat water [21], the measured A_{254} values taken from the intake were considered low. The higher level of A_{254} in Maji ya Chai river and peat water in central Kalimantan was due to the contribution of humic substances originated from natural sources of NOM. In addition, higher SUVA (>4 L/mg/m) and the lower level of measured aromatization ratio (A3/4) (<5) values at the point-2 than those at the point-1 indicated DOM in the upstream was dominated by humic acids and hydrophobic substances. Compared to other studies, DOM-A_{355} values at the point-1 were within the range of those reported in Xiaoqing River (0.04–0.17 cm^{-1}) [22], but lower than those reported in the estuary (average of 0.06cm^{-1}) [23] and lake water body (0.004–0.013cm^{-1}) which have a higher load of organic matter originated from natural sources [24].

DOM characteristics and concentrations in natural water can vary seasonally. In this study, seasonal variations in the measured values of COD, A_{254}, A_{355}, A3/4, DOC, and SUVA were observed in both TOM and DOM samples, with the highest average values occurred in the dry season (Table 1). The lower values of these parameters during the rainy season were not expected, considering greater entry of allochthonous/ terrestrial materials through surface runoff may occur in the rainy season [11, 12]. Seasonal changes of river flow might explain the concentrations and characteristics differences over seasons. According to the Meteorology and Geophysics Agency, the average monthly rainfall intensity during the dry season (August 2017) was only 48.4 mm, with a total of six rainy-days observed only during the month. The rainfall intensity during rainy season period 1 range between 65.3mm-199.3mm, reached 442.2mm during period 2 with total rainy-days at least 23 days/month of during the time course (January-February 2017; and November 2017). Therefore, there was a potentially less dilution effect from the rainwater in the river during the dry season. On the other hand, organic matters from nearby anthropogenic sources were consistently discharged into the stream throughout the year. This continuous entry of organic matters from anthropogenic activities throughout the year coincidence with the minimum flow of Cikapundung river during the dry season may result in higher DOM level in this season.

SUVA values correspond to the nature or characteristics of organic matters, providing a quantitative measure of aromatic and hydrophobic fractions of the DOM in water [25]. Higher average values of SUVA (>5mg/L) as well as TOM-A_{254} (0.43cm^{-1}) and DOM-A_{254} (0.35cm^{-1}) during the dry season, suggesting domination of aromatic and hydrophobic compounds to DOM during the dry season than those during the rainy season. The majority of hydrophobic compounds was acidic compounds such as fulvic and humic acids originated from both natural decompositions of lignin and anthropogenic activities [25]. A further measurement such as through Fluorescence Excitation-Emission Matrix is needed to confirm the origin of DOM [26]. Along with the results for SUVA and A_{254}, A3/4 ratio values across dry and rainy seasons were observed <5. Thus it enhances the finding that humic acid compounds were more dominant than fulvic acid compounds, as reported by Artinger [2000] that A3/4 <5 indicates higher concentrations of aromatic and humic acid compounds in the water.

3.2 The relationships between TOM and DOM

Fig. 2 A-B shows a relationship between TOM- and DOM-A_{254} (R=0.78;p=0.01), as well as -A_{355} (R=0.75;p=0.02) during the dry season. TOM-COD was well correlated with DOM-COD in this season (R=0.72; p=0.03), as seen in Fig.2 C.

For the rainy season, the relationships between TOM and DOM for all parameters were weaker (Fig. 2). This was because TOM of the rainy season was possibly dominated by terrestrial particulate organic matter carried by the runoff. Yet, TOM of the dry season was composed of DOM predominantly rather than particulate organic matter. As seen in Table 1, the proportions of CDOM in Chromophoric-TOM (CTOM) in the rainy season were smaller than that in the dry season (CDOM/CTOM = 49-66% in the rainy; 73-82% in the dry season). It can be suggested that for the dry season, the level of DOM can be predicted solely based on the measurement of TOM. However, both TOM and DOM should be measured during the rainy season as they were not well correlated.

3.3 DOM and THMFP relationships

The measured total forming potential and forming potential of each THM compound across three different sampling periods are summarized in Table 3. The table shows, the formation of $CHCl_3$-FP was the highest, followed by $CHBrCl_2$-FP. However, the $CHClBr_2$-FP and $CHBr_3$-FP were detected below the Detection Limit (DL). Both $CHCl_3$ and $CHBrCl_2$ were the most common THMs species detected in tropical water [13, 27].

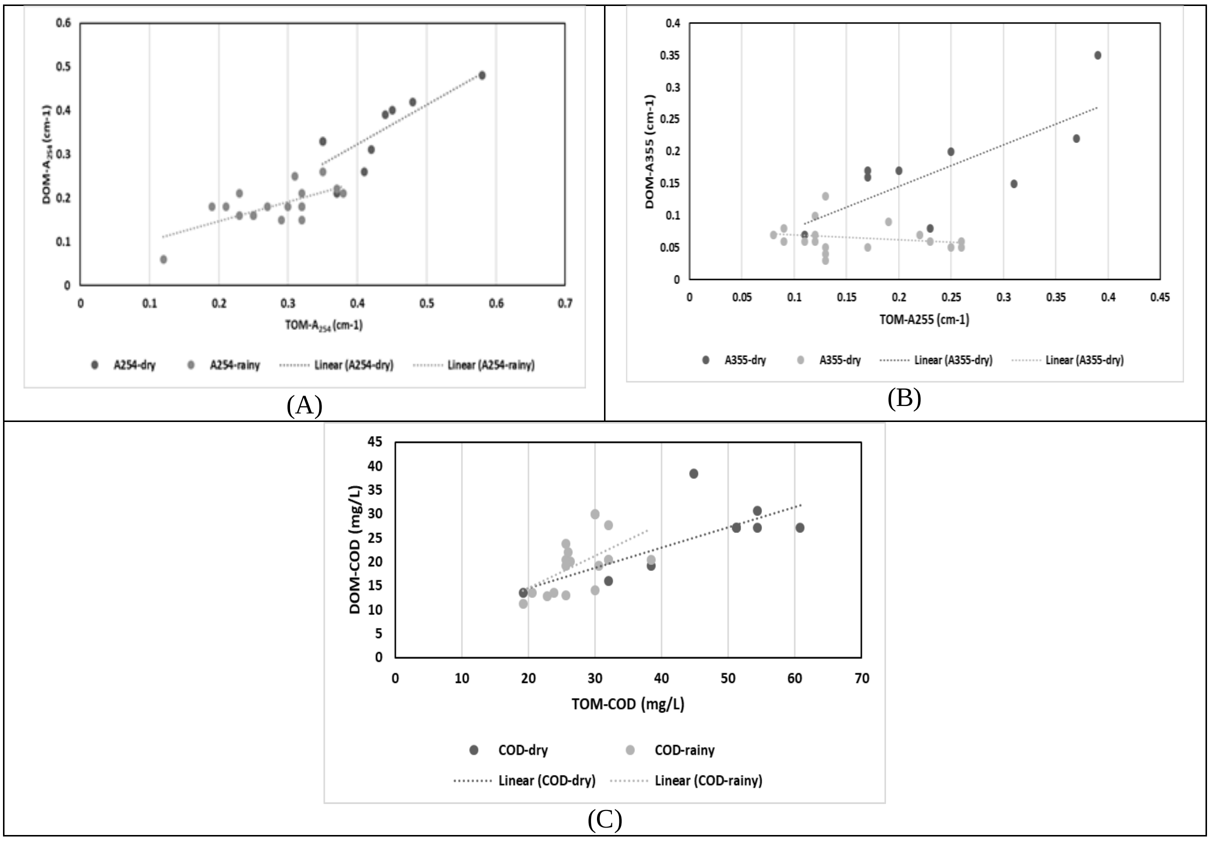


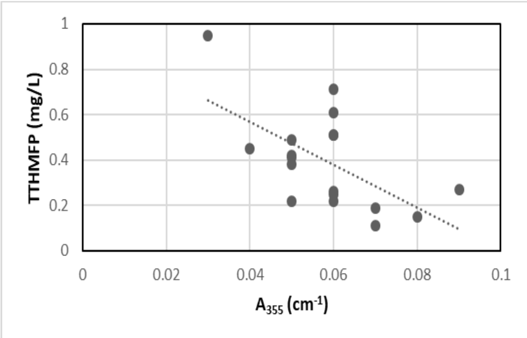
Fig.2 The relationships between TOM and DOM during dry and rainy seasons: (A) TOM-A_{254} and BOM-$A_{254;}$ (B) TOM-A_{355} and BOM-A_{355}; (C) TOM-COD and DOM-COD

Table 3. THMFP concentrations **(µg/L)** during dry rainy and rainy seasons period 2-3 at point-1

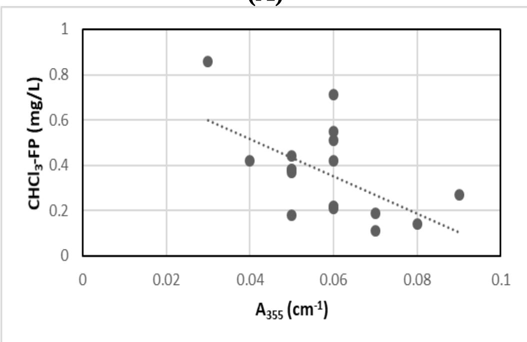
Parameters	Ranges	$\overline{X}$
$CHBrCl_2$ -FP		
Dry	14.43-97.93	66.87
Rainy	<DL-120.00	33.57
$CHCl_3$ -FP		
Dry	140.00-937.39	568.63
Rainy	110.00-859.98	405.69
TTHMFP		
Dry	154.43-1021.88	636.79
Rainy	100.00-867.26	430.13

Furthermore, the results indicated that $CHCl_3$-FP was the predominant species of THM in the water samples, consistent with those reported in both tropical and sub-tropical rivers [9, 13, 28-32]. Seasonal variations were also observed for the occurrence of THMFP. The average concentrations of total THMFP, $CHCl_3$-FP, and $CHBrCl_2$-FP during the dry season were higher than those during the rainy season (Table 3).

SUVA is generally known as the major precursors to the formation of disinfection by-products [4]. Hence a higher SUVA should increase THMFP level [33, 34]. In this study, during dry season however, poor correlations were observed between DOM enriched with aromatic and hydrophobic than hydrophilic compounds (SUVA>5L/mg/m) and TTHMFP (with a Pearson's correlation (R) of -0.37 and p-value of >0.05), $CHCl_3$-FP (R of -0.37 and p-value of 0.05), and $CHBrCl_2$-FP (R of -0.28 and p-value of >0.05). This indicated the presence of non-reactive aromatic compounds which were originated from anthropogenic activities near the intake area of Cikapundung River. Previous studies demonstrated that for water samples with high SUVA level, not all aromatic fraction reacted with chlorine to form THMs [35]. Similar results were also found in a shrimp farm contaminated by fertilizers with SUVA value above 5L/mg/m [13]. Accordingly, SUVA >5L/mg/m tends to be a poor indicator for THMFP and measured DOM parameters relationship in a polluted water body with a high level of aromatic and hydrophobic compounds as also found in Cikapundung River.



(A)



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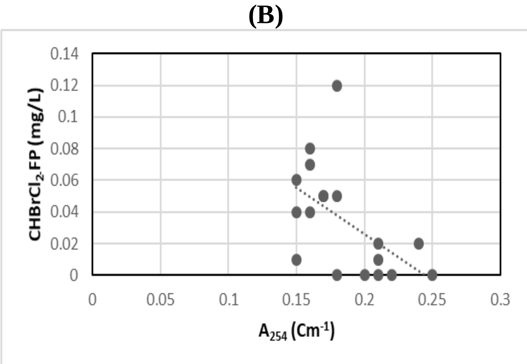

(C)

Fig. 3 The relationships between DOM and THMFP during rainy seasons: (A) A_{355} and TTHMFP; (B) A_{355} and CHCl3-FP; (C) A_{254} and CHBrCl2-FP

The value of DOM during the rainy season indicated balance proportion of aromatic and non-aromatic as well as balance proportion of hydrophobic and hydrophilic compounds, which was consistent with SUVA values in the range of 2–4L/mg/m. Significant correlations ($p<0.05$) were found between A_{355} and $CHCl_3$-FP and TTHMFP, A_{254}, and $CHBrCl_2$-FP during the rainy season (Fig.3). This suggests the aromatic compounds reactive to chlorine and bromine in water samples when SUVA values ranged from 2 to 4L/mg/m [33]. Significant correlations between CDOM and THMFP during the rainy season were also reported in a river surrounded by anthropogenic activities [36] and in a river with high flow rates [35], similar with those observed in Cikapundung River during the rainy season. The results may confirm terrestrial DOM was an important precursor of THMs in that season. Therefore, A_{355} can be used as a surrogate parameter during the rainy season in the Cikapundung river and other tropical polluted rivers with similar environmental conditions. The findings are of great importance and concern since THMFP measurement is costly and timely.

4. CONCLUSIONS

This study showed that the highest levels of all organic parameters in the Cikapundung river occurred during the dry season. In this season, good relationships between TOM and DOM indicated the level of DOM can be predicted based on the measurement of TOM. DOM of dry season comprised a larger proportion of aromatic and hydrophobic compounds at SUVA levels >5L/mg/m). However, DOM parameters were poorly correlated with THMFP species which possibly because not all aromatic fraction reacted with chlorine to form THMs. Thus SUVA at levels above 5L/mg/m can be used as an indicator of poor relationships between DOM and THMFP.

DOM during the rainy season comprised balance proportion of aromatic and non-aromatic as well as hydrophobic and hydrophilic compounds (SUVA of 2-4L/mg/m). In this condition, a significant correlation between A_{355} and TTHMFP; A_{355} and $CHCl_3$-FP; and A_{254} and $CHBrCl_2$-FP were observed. Therefore, terrestrial DOM (A_{355}) was concluded as an important precursor of THMs and A_{355} can be used as a surrogate parameter to control THMFP during the rainy season. This may help to replace THMFP measurement with A_{355} measurement to avoid the high cost and time-consuming measurement.

5. ACKNOWLEDGMENTS

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