

## Article

# Effect of Radio-Frequency Treatment on the Changes of Dissolved Organic Matter in Rainwater

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**Abstract:** Rainwater is a potential source of drinking water, but has various components of dissolved organic matter (DOM). DOM is a reservoir of potential hazards in drinking water. Therefore, a new method is required to purify rainwater as a drinking water source in terms of DOM aspects. A radio-frequency (RF) treatment system is introduced here to purify source water with a small possibility of contamination. RF is generated by applying a frequency of 1.5 MHz through a glass reactor with a diameter of 2 mm which is wrapped by a 2 mm copper wire. The results demonstrate that UV<sub>260</sub> value and dissolved organic carbon (DOC) are reduced during RF treatment. DOC was reduced by a lower amount compared to UV<sub>260</sub>, suggesting the partial transformation of bio-refractory DOM. A fluorescence excitation-emission matrix showed that humic-like substances in rainwater were reduced faster than protein-like ones, indicating that humic-like substances are susceptible to reduction by RF treatment. The results offer information on the use of RF treatment in a rainwater purification process for the production of drinking water.

**Keywords:** dissolved organic matter; radio-frequency treatment; rainwater; humic-like substances



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## 1. Introduction

Due to the economics of water scarcity, rainwater has become popular as an alternative drinking water source in areas with a high rainfall rate, deficient centralized systems, or where pressure on water resources surpasses the recharge capacity [1]. Rainwater can be transformed into drinking water when the supply capacity is drastically reduced due to climate change [2,3]. The field of water resource management in large urban areas has emerged to support the development and sustainability of the water supply, as well as the treatment of discharges of rainwater. Rainwater harvesting is one way to effectively reduce excess surface runoff and flooding risk, while producing an alternative drinking water source, and an emergency water supply, thus reducing water bills and operational costs [4].

With the recent concerns for sustainability as well as development of safe drinking water, rainwater has gained increasing interest as a safe water supply option [5]. Urban rainwater tank systems have undergone a surge to promote water conservation in both developed and developing countries [6]. In African and Asian developing countries, including Indonesia, the utilization of rainwater as a water supply is being brought to urban areas, with the support of the government. In Australia, rainwater is used for non-drinking purposes, such as for gardening or toilet flushing; only 2% of the Australian population uses rainwater as drinking water [6]. Countries in Europe, such as Germany and Sweden, have developed rainwater harvesting systems for urban use, toilet flushing, fire protection and flood control [7,8]. However, the practice of transforming rainwater to drinking water is still rare, and only a few projects have been reported to date [9]. In Indonesia and Vietnam, rainwater has been traditionally utilized for drinking water through the boiling method [5,10]. Nevertheless, there are no standard regulations on

the use of rainwater as a source of drinking water. Substances such as dissolved organic matter (DOM) and microorganisms in rainwater may not be effectively removed through the simple disinfection method of boiling [10]. Thus, the health issues inflicted from the unsafe utilization of rainwater have been a major concern in the use and maintenance of rainwater for drinking systems.

Studies have demonstrated that rainwater contains DOM in various concentrations and of varied composition [11,12]. DOM is a part of natural organic matter (NOM) along with particulate organic matter (POM) that consists mainly of microorganisms [13]. However, the proportion of DOM is higher than POM (>65% of total NOM) [14], and thus the study on DOM in rainwater and other water sources is gaining more interest. DOM in rainwater originates from various sources, such as gaseous dissolution, soil dust re-suspension, marine organic matter evaporation, fossil fuel combustion, and biomass burning [15–17]. The presence of DOM in rainwater plays a crucial role in the changes in the atmospheric environment, cloud albedo, inputs to the land ecosystem, biogeochemical cycles, and global climate change [4,18,19]. Rainwater as an alternative source of drinking water requires a purification process to achieve water quality standards for drinking purposes [20]. Conventional treatments, such as coagulation, flocculation, sedimentation, and sand filtration have been widely used to remove large types of DOM from the water source. Adding chlorine to rainwater is an obligatory operation for reducing DOM, but simultaneously increases the chemical risks [21]. However, conventional treatments for rainwater are insufficient to completely remove pollutants with a low molecular size of DOM.

To achieve the reduction of DOM in rainwater through a chemical process, numerous purification processes have been used in terms of the oxidation process that promotes a faster reduction of DOM with low molecular size in water. Radio-frequency (RF) is an electromagnetic field system in the frequency range of 20 kHz to 300 GHz [22–24]. RF technology has been used for diverse purposes in many different areas. RF treatment is among the advanced treatments used to purify the water source with a lower possibility of contamination. RF generates electrons, free radicals, ions, and neutral ionized gas that can enable plasma substances to reach thermal equilibrium and decompose organic compounds in water [25,26]. RF currents in conductors can radiate into space as radio waves and induce heat to provide physical and chemical effects in solution that are capable of degrading high molecular weight organic pollutants [27,28]. RF can generate oxidant species such as hydroxyl radicals ( $\bullet\text{OH}$ ), hydrogen peroxide ( $\text{H}_2\text{O}_2$ ), and ozone ( $\text{O}_3$ ), and these oxidizing species have a high potential to decompose organic compounds in water [29–31].

Numerous studies have focused on a single oxidant species generated by an advanced oxidation process on DOM transformation. Voronstov [32] and Ya et al. [33] demonstrated that  $\bullet\text{OH}$  could oxidize most organic matter, and even completely mineralize it, ultimately generating carbon dioxide and water. DOM was significantly degraded by  $\text{O}_3$  with a specific ozone dose of 0.5–1.5  $\text{mgO}_3/\text{mgC}$  and reached sufficient degradation in 30 min [34]. This oxidation treatment can promote a high removal efficiency for removing organic compounds, yet the demerit of this oxidation treatment is that corrosion may occur on the electrode surface due to direct contact with water in the reactor. Therefore, the utilization of RF can promote DOM reduction efficiency without causing corrosion because there is no direct contact with the water. In the current study, the purpose was to validate the potential utilization of RF treatment to reduce the DOM composition and concentration in rainwater. The validation study was assessed by the reduction of DOM during treatment and to help further the development of an optimization strategy for utilization rainwater as a drinking water source.

## 2. Materials and Methods

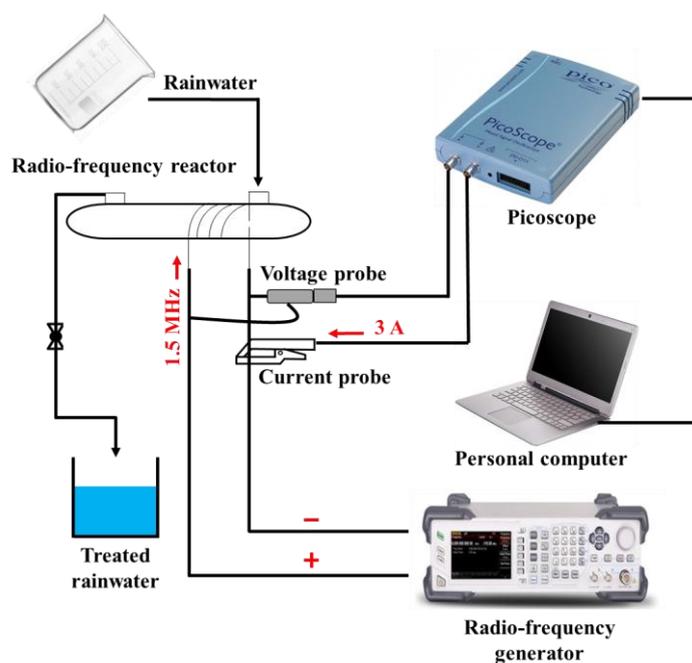
### 2.1. Rainwater

Rainwater was collected from domestic residence roof catchments located in Padang, West Sumatra, Indonesia, in daily rainfall events for 8 days (25 June–8 July 2019). Rainwater was collected using 500 mL pre-chlorinated glass bottles and 8 samples were collected

during the investigation. The nearest meteorological station to the sampling location is Maritim Teluk Bayur (8 km). The available data of average rainfall for the site was 214 mm, originating over a mountainous area [35]. A sterile glass bottle was used to collect rainwater manually.

## 2.2. RF Treatment

Figure 1 depicts the experimental set-up used in this study. The system consists of a MI007 passive oscilloscope probe, a current probe (P6021A Tektronix), an RF generator and a reactor. The RF reactor has a length of 30 cm and a diameter of 7.62 cm, coupled by a copper wire ( $\varnothing$  2 mm), with a frequency of 1.5 MHz and an electric current of 3 A. The collected rainwater was transferred to the water tank, pumped with a flow rate of 200 mL/min through a cartridge filter and allowed to flow through the reactor. The treated rainwater was analyzed for dissolved organic carbon (DOC), ultra-violet absorbance at 260 nm ( $UV_{260}$ ), fluorescence excitation-emission matrix (EEM), and molecular weight (MW) distribution.



**Figure 1.** Experimental set-up of an RF treatment.

## 2.3. DOM Analysis

The DOC was quantified with a TOC analyser (TOC-V<sub>wet</sub>, Shimadzu, Japan). Pure water was measured prior to the standard solution and samples to confirm the sensitivity of the measurement. Five different concentrations (0.0; 0.5; 1.0; 2.0 and 5.0 mg/L) of standard solution containing 1,000 mg-C/L of potassium hydrogen phthalate was used for DOC measurement. The linear regression obtained from the measurement of the standard solution was 0.9998. The  $UV_{260}$  was measured by a UV-vis spectrophotometer (UV-2600, Shimadzu, Tokyo, Japan). A 1-cm cuvette was used to measure pure water prior to the samples to confirm the sensitivity of the analysis. The scanning wavelength of 220–500 nm was used with medium scanning speed (400 nm/min) and the data pitch of 1 nm. pH, electric conductivity (EC), and total dissolved solids (TDS) were measured with the procedures described in the APHA standard methods. Dissolved ions: nitrite ( $NO_2^-$ ), nitrate ( $NO_3^-$ ), ammonium ( $NH_4^+$ ), sulfate ( $SO_4^{2-}$ ), calcium ( $Ca^{2+}$ ), magnesium ( $Mg^{2+}$ ) and potassium ( $K^+$ ) were measured using an ion chromatography system (LC-20AD SP, Shimadzu, Japan).

The fluorescence EEM spectra of the rainwater were measured by a spectrofluorometer (RF-5300, Shimadzu, Japan). A 1-cm cuvette and high-sensitivity cell holder were used for the measurement. The excitation and emission scans showed a wavelength in the range of 200 nm and 550 nm, at 5 nm increments. The obtained fluorescence intensity of rainwater was normalized using the quinone sulphate unit (QSU) by dividing the fluorescence intensity values of rainwater by that of 10 µg/L quinone sulphate (in a 0.05 M sulfuric acid solution), at the designated excitation wavelength (Ex) of 350 nm and the emission wavelength (Em) of 450 nm. High pressure size exclusion chromatography (HPSEC) was used to evaluate the MW characteristics of the DOM at a wavelength of 260 nm. HPSEC consisted of a silica chromatographic column (GL-W250-X, 10.7 × 450 nm, Hitachi, Tokyo, Japan) and a UV detector (LC-10AV, Shimadzu). Pure water containing 0.02 M of Na<sub>2</sub>HPO<sub>4</sub> and 0.02 M of KH<sub>2</sub>PO<sub>4</sub> was used as the eluent, and was introduced to the column at a constant flow rate of 0.5 mL/min.

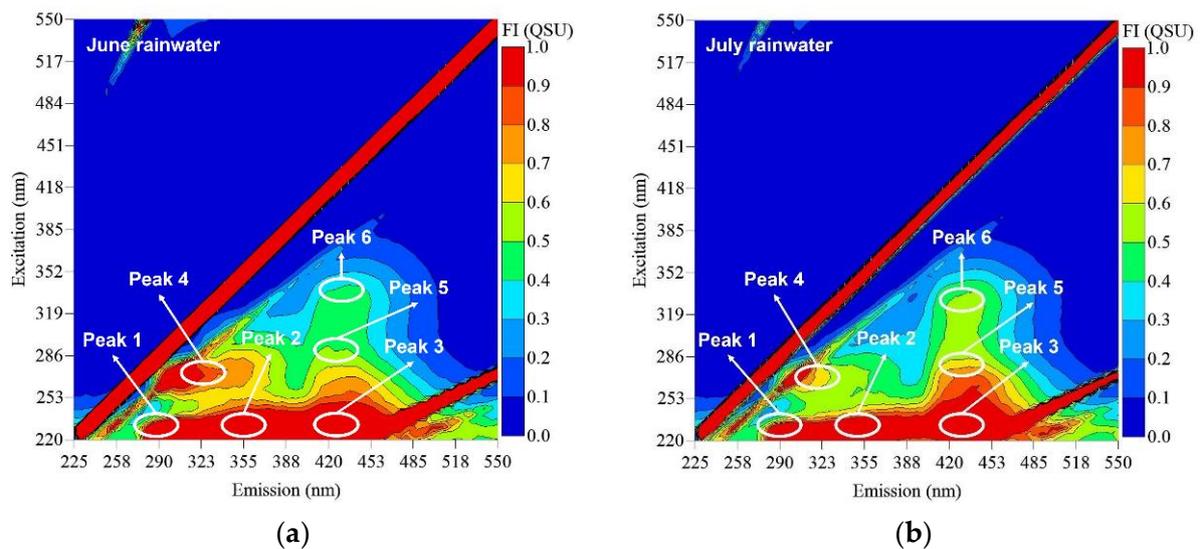
### 3. Results and Discussion

#### 3.1. Characteristics of Rainwater

A RF treatment for the degradation of DOM was investigated to improve rainwater quality as an alternative source for drinking water. Several prominent water quality parameters have been analyzed, including pH, EC, TDS, dissolved ions, and DOC and DOM compositions evaluated by fluorescence EEM analysis. The initial pH, EC, and TDS values were  $6.9 \pm 0.17$ ,  $19.5 \pm 0.98$  mS/m and  $9.17 \pm 0.38$  mg/L, respectively ( $n = 8$ ). The UV<sub>260</sub> value of rainwater was in the range of 1.31–21.8 m<sup>-1</sup> during rainfall events. The mean UV<sub>260</sub> was  $2.97 \pm 0.31$  m<sup>-1</sup> in June ( $n = 5$ ), and  $7.29 \pm 0.44$  m<sup>-1</sup> in July ( $n = 3$ ). The significant difference in UV<sub>260</sub> suggests that rainwater carried different UV-absorbing DOM from the atmosphere across the sampling periods. The lower absorption in June suggests that the DOM in rainwater was scavenged from the atmosphere below the clouds, as the rainwater amount increased [36]. The DOC concentration was low in June (1.65 mg/L), suggesting that the contribution of organic carbon input is low in terms of DOC in rainwater in Padang.

Six fluorescent components in rainwater were identified by the fluorescence EEM peak-peaking method (Figure 2). The DOM that is identified by fluorescence EEM in rainwater was assigned to protein-like tyrosine (Peak 1: Ex/Em: 230/302 and Peak 4: Ex/Em: 275/302) which derived from autochthonous origin [37,38]. Peak 2 was described as protein-like tryptophan, located at Ex/Em: 240/338, and was also found in rainwater. Peak 3 (Ex/Em: 305/414) was like the terrestrial humic-like fluorophores [39]. Peak 5 (Ex/Em: 320/390) was described as marine humic-like and was often observed in the ocean [37]. Peak 6 (humic-like) was attributed to the UV-absorbing humic fluorescence, which was in Ex/Em: 230/386 and was observed in other rainwater studies [40,41] and considered as microbial humic fluorescence [39]. The intensities of Peaks 1, 2 and 3 were higher than other peaks. There was no significant time variation in DOM composition. The average relative contributions of all peaks to the total value were 32%, 27%, 21%, 10%, 5% and 5%, respectively.

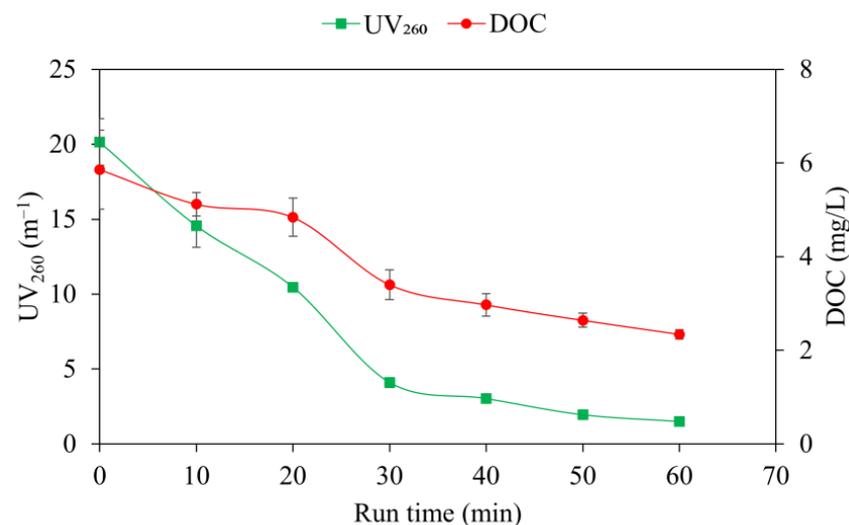
The average concentrations of NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> were  $1.61 \pm 0.17$ ,  $3.87 \pm 0.11$ , and  $5.74 \pm 0.04$  mg/L, respectively ( $n = 8$ ). NH<sub>4</sub><sup>+</sup> was generally considered to originate from fertilizer, soil, livestock manure and plants. Padang is covered by farmland and the local emissions may influence the changes in ammonium ions in rainwater. NO<sub>2</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup> mainly originate from vehicle emissions and chemical fuel combustion. Motor vehicles lead to an increase and deposition of NO<sub>x</sub> in rainwater [42]. SO<sub>4</sub><sup>2+</sup> may likely be from a nearby cement industry, which resulted in the presence of SO<sub>4</sub><sup>2+</sup> in rainwater. Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup> in rainwater mainly originate from dust particles and dissolve in rainwater.



**Figure 2.** Fluorescence spectra of rainwater from different sampling periods (a) June (b) July. Peak 1: protein-like tyrosine; Peak 2 and 4: protein-like tryptophan; Peak 3: fulvic acid-like; Peak 5: marine humic-like; and Peak 6: humic-like from fluorescence EEM.

### 3.2. Impact of RF on DOC and $UV_{260}$

Figure 3 shows the reduction in  $UV_{260}$  absorbance and DOC observed during RF treatment. The reduction of  $UV_{260}$  may be attributed to the loss of aromatic and conjugated double bond structures of DOM, due to the oxidation process.  $UV_{260}$  was observed to be reduced by 28% in 10 min, and 93% in 60 min, while DOC was reduced by 13% in 10 min, and 60% in 60 min. DOC reduction was lower because of the partial oxidation of DOM to other parameters, like  $UV_{260}$ . The high reduction of  $UV_{260}$  with a lower reduction of DOC suggests that RF can remove the conjugated double bonds with minimal mineralization [43]. The rapid decrease of  $UV_{260}$  in 30 min is probably due to the extent of aromatic DOM destruction by the oxidant species generated from the RF reactor. Previous studies reported that organic matter could be highly oxidized in 10 min through specific reactions [44,45]; thus, rapid degradation of DOM in term of  $UV_{260}$  in this study is explained by the high reactivity of its carbon double bond. A very slight decrease of  $UV_{260}$  value after 30 min indicates that the remaining UV-absorbing constituents contain moderately activated aromatic rings and were probably attacked by  $\bullet OH$  and  $O_3$  [45].



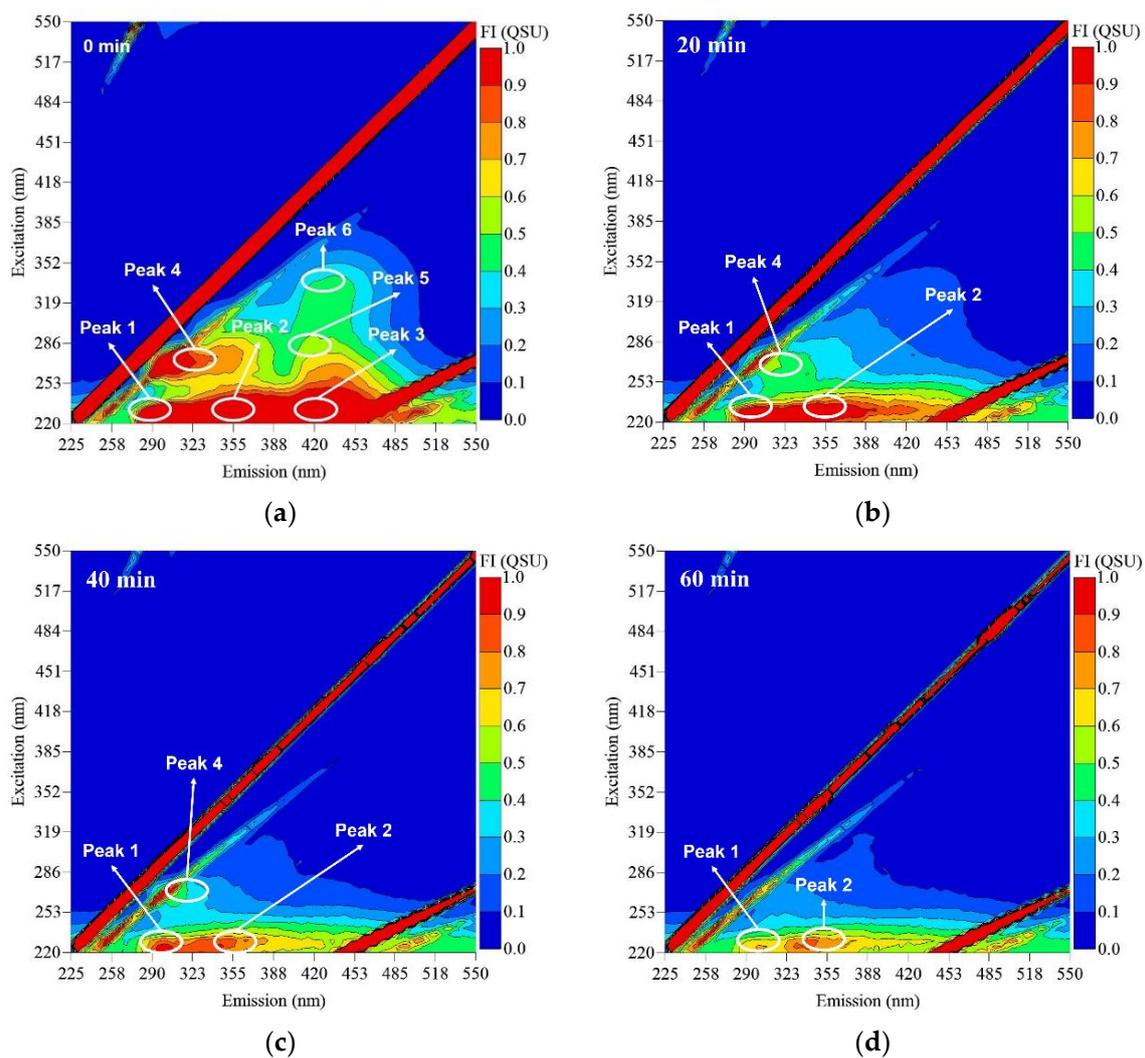
**Figure 3.** The reduction of  $UV_{260}$  and DOC during RF treatment ( $n = 8$ ).

During RF treatment, the reduction of DOM was attributed to the chemical oxidation of the DOM present in rainwater by reactive oxidant species, such as  $\bullet\text{OH}$  and  $\text{O}_3$ . However, exponential decays of  $\text{UV}_{260}$  and DOC were observed, although the chemical degradation kinetics differ between them (data not shown). A previous study revealed that DOC still fell slightly even after five hours with  $\text{O}_3$  [46]. It is suggested that oxidant species produced in this study were inefficient in reducing DOC. The low change of DOC might also be limited by the ability of electric discharge from RF reactor to transform DOM into lower MW compounds; only a partial transformation of bio-refractory DOM into biodegradable compounds was facilitated. The preliminary results confirm that RF can effectively degrade DOM in less than 1 h reaction under the adopted experimental conditions. The findings are in agreement with a previous study on the transformation of micropollutants in wastewater using an ozone-based oxidation process [45]. However, since the RF treatment discharges various oxidant species, the specific oxidants that are responsible for the degradation mechanisms of DOM in this study require further investigation.

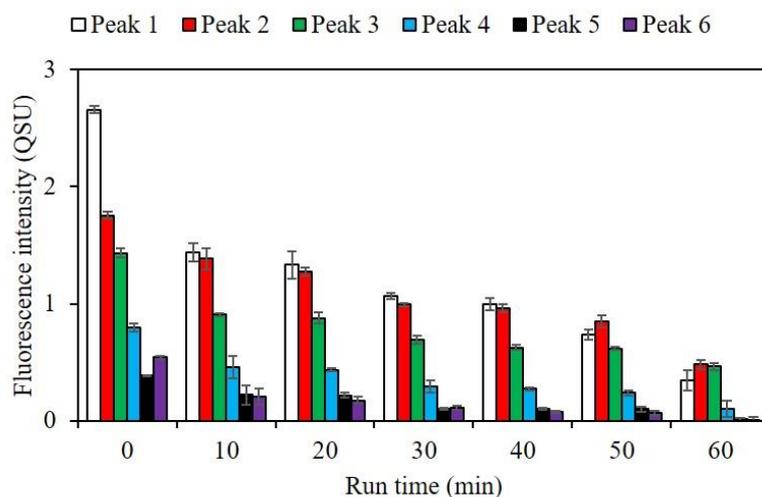
### 3.3. Impact of RF on Fluorescent DOM Composition

As is shown in Figure 4, the fluorescence intensities of Peak 1, Peak 2 and Peak 3 were much higher than others; thus, they were the main components represented in rainwater and subjected to the degradation process during plasma treatment. Peak 5 and Peak 3 may contain the carboxyl and hydroxyl groups that led to the favorable formation of  $\bullet\text{OH}$  and accelerated the degradation rate of organic compounds. The rapid degradation of Peak 5 in 10 min indicates that the humic-like component was depleted faster than the protein-like component. The protein-like component may have a large molecular weight that results in the incomplete dehydrogenation of aromatic rings [47,48]. A previous study demonstrated oxidation treatment using parallel electrodes during direct electrolysis treatment with the addition of chloride ion [49]. The results show that aromatic macromolecular organic compounds were degraded into small molecular substances, such as acetic acid and phenol. The reasons may be due to the increase of  $\text{H}^+$  that could improve the reaction efficiency between organic matter and reactive species. Simultaneously, it promoted the formation of highly reactive oxygen species, such as  $\bullet\text{OH}$ , and facilitated the oxidation and decomposition of organic compounds [49]. In this study, RF can generate several reactive species that may assist in the decomposition and degradation of DOM in rainwater during treatment.

Figure 5 shows the changes of fluorescence DOM during RF treatment of rainwater. Peak 1 had the highest intensity at 0 min, indicating that protein-like tyrosine accumulated in rainwater. The intensity of the fluorescent components in the rainwater gradually decreased during plasma treatment. Peaks 1, 2 and 4, which are described as protein-like compounds, were reduced by 87%, 73% and 87%, while Peaks 3, 5 and 6, which are described as humic-like compounds, were reduced by 67%, 97% and 98%, respectively, from 0 to 60 min. The results indicate that RF consists of high oxidative agents, such as  $\bullet\text{OH}$  and  $\text{O}_3$ , which could effectively degrade the humic-like compounds in rainwater. The decrease of DOM composition by plasma was in the following order: Peak 5 > Peak 3 > Peak 1  $\geq$  Peak 4 > Peak 2 > Peak 3. A fast decrease of Peak 1 and Peak 6 was observed in 10 min of treatment, because of the degradation and mineralization of the DOM with an aromatic structure that reacts quickly with  $\bullet\text{OH}$  [50,51]. Humic substances are known to be bio-refractory with high stability and aromaticity and can be transformed to become more biodegradable by oxidants generated during RF treatment [41].



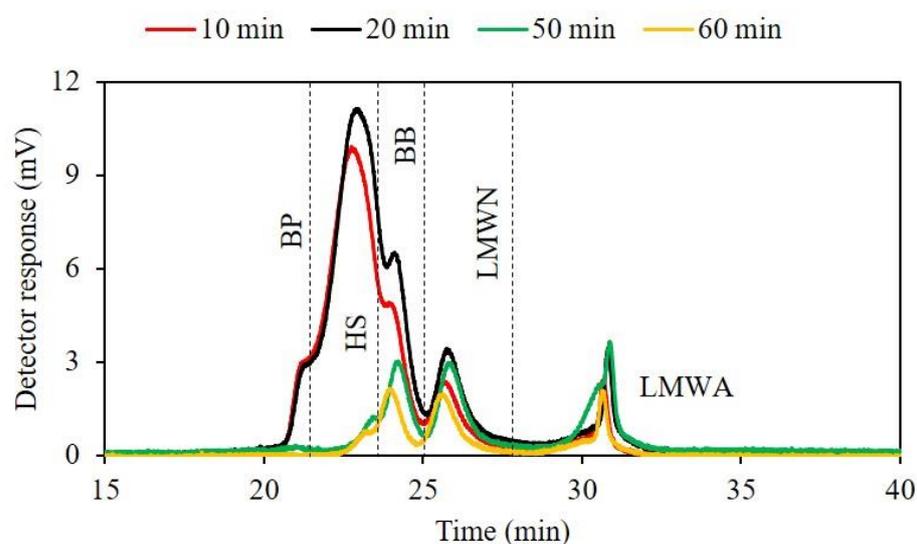
**Figure 4.** Fluorescence spectra of rainwater before and after RF treatment. (a) 0 min, (b) 20 min, (c) 40 min, and (d) 60 min. Peak 1: protein-like tyrosine; Peaks 2 and 4: protein-like tryptophan; Peak 3: fulvic acid-like; Peak 5: marine humic-like; and Peak 6: humic-like from fluorescence EEM.



**Figure 5.** The changes of fluorescence DOM composition during RF treatment ( $n = 8$ ). Peak 1: protein-like tyrosine; Peaks 2 and 4: protein-like tryptophan; Peak 3: fulvic acid-like; Peak 5: marine humic-like; Peak 6: humic-like.

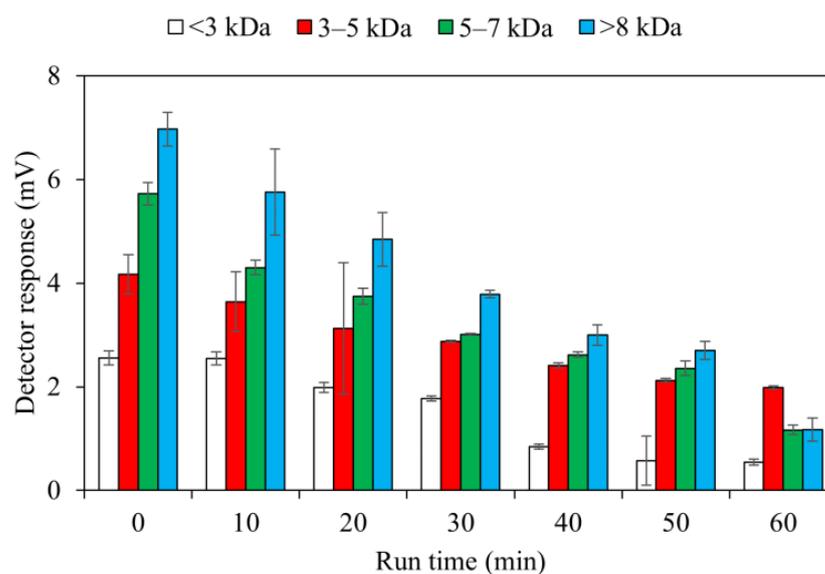
### 3.4. Impact of RF on MW of DOM

Figure 6 shows the MW distribution of the DOM in rainwater measured by HPSEC and is given in terms of detector response of UV absorbance at 260 nm. The distributions of MW fractions extracted from HPSEC in rainwater were 32%, 27%, 18%, 12% and 11% for HS, LMWN, BP, LWMA and BB, respectively. HS are not easily biodegradable. However, oxidation by RF can remove 78% of HS that can be attributed to the molecule breakdown into a low MW size. Rainwater featured a large MW, and the total response decreased after RF treatment was applied. The result suggests that RF facilitated the oxidation of aromatic or double bond organic matter into lower MW compounds. This study showed that the apparent high MW that consisted of BP and HS may contain high aromaticity humic molecules that are easy to degrade through oxidation process. The oxidants generated by the RF reactor assisted the unsaturated functional groups to break down into smaller molecules which are more biodegradable [52].



**Figure 6.** MW distribution in rainwater sampled on 27 July 2019. BP: biopolymer; HS: humic substances; BB: building blocks; LMWN: low MW neutral; LMWA: low MW acid.

RF was found to reduce the >8 kDa and 5–7 kDa MW fractions of organic matter by 83% and 79.6%, respectively, in a 60 min treatment (Figure 7). RF demonstrated that larger MW DOM was reduced more than that of lower MW DOM, indicating that the higher reaction rate is constant among oxidant species, such as  $\bullet\text{OH}$  and  $\text{O}_3$  produced from RF plasma and the larger MW compounds [53]. Higher MW compounds tend to be more aromatic, so they may have a larger number of reaction sites compared to smaller MW compounds [43]. The decreasing trend of MW size of DOM during RF treatment is consistent with the decreasing trend of  $\text{UV}_{260}$  value, where aromatics transformation leads to the formation of low MW products. RF treatment removed DOM with molecular size < 3 kDa less than DOM with molecular size > 5 kDa. This result confirms that RF treatment facilitates the faster degradation for high MW. This study is concordant with previous studies on the effects of advanced oxidation processes on DOM removal [52,53].



**Figure 7.** The changes of molecular size of DOM during RF treatment ( $n = 8$ ).

#### 4. Conclusions

This study investigated the impact of RF treatment on the levels of DOM in rainwater. DOM in rainwater was reduced during RF treatment through oxidation by reactive oxidant species produced by the RF. This study demonstrated that RF could remove 96% of UV<sub>260</sub> substances that may comprise humic molecules by oxidation through high mineralization. DOM compositions assessed by fluorescence EEM showed that humic-like compounds are more easily degraded than protein-like compounds. Humic-like compounds were reduced by 97%, indicating that oxidant species produced by RF could transform non-biodegradable aromatic humic molecules to become biodegradable. Oxidant species produced by RF can also transform high MW fractions into low ones. •OH and O<sub>3</sub> can easily attack and oxidise the reaction site in an aromatic structure. However, the dominant oxidant species produced from RF in this study remain unclear. Further work is needed to identify specific reactive oxidant species that correspond to the reduction of DOM in rainwater. The current operational study of RF also shows the need for further exploration on the DOM reduction. This work offers valuable information on understanding potential RF treatment for water source and drinking water conservation and treatment.

**Author Contributions:** Conceptualization, R.D. and M.Y.R.; methodology, R.D. and A.H.; validation, R.D., A.H. and F.L. investigation, R.D.; writing—original draft preparation, M.Y.R. and R.D.; writing—review and editing, M.Y.R., R.D., A.H. and F.L.; supervision, A.H. and F.L. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors report no conflict of interest.

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