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ENHANCEMENT OF WATER TREATMENT BY COMBINED FILTRATION-ICPS: INTEGRATED EVALUATION BASED ON EEMS, DOC, UV260 AND REMOVAL OF PATHOGENIC BACTERIA

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ABSTRACT

The impact of natural organic matter (NOM) and pathogenic bacteria in water are of great concern in water treatment due to their effect on human health. In this study, the ability to combine filtration with an inductively coupled plasma system (ICPS) to remove natural organic matter and pathogenic bacteria from water was investigated. UV260 adsorption, dissolved organic carbon (DOC) and excitation-emission matrices (EEMs) were used to evaluate the removal of NOM by the filtration-ICPS combination. The system was operated at two different flow rates, 100 and 150 ml/minute. Decreasing trend revealed for UV260 and DOC with the higher flow rate. Tryptophan-like substances generally found in water were taken as a measure of microbial activity. This preliminary research shows that the average removal efficiencies for fecal coliforms, total coliforms and salmonella were highest at a flow rate of 100 ml/minute. It can be concluded that the combined filtration-ICPS method is capable of removing NOM and pathogenic bacteria from water.

Keywords: Filtration; ICPS; Natural organic matter; Pathogenic bacteria; Water treatment

1. INTRODUCTION

Naturally occurring natural organic matter (NOM) and dissolved organic matter (DOM) refer to complex and diversified mixtures of organic compounds that arise from the microbial degradation of biomolecules (Bougdah et al., 2017). DOM occurs as a consequence of the contact between water and its precursors, such as dead and living plants, animals, microorganisms and their decomposition products that are present in the hydrological cycle (Bridgeman et al., 2011). A third to over half of the DOM in natural surface waters comprises humic substances; these are organic compounds that are microbially (by bacteria and algae) and terrestrially derived from soil humus.

DOM from wastewater effluent and/or sewage treatment plants is a source of dissolved organic matter in water environments. Humic substances are dominated by hydrocarbon bonds, causing

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lipophilic interactions. This is a significant concern in drinking water treatment due to aesthetic, operational and economic factors. The presence of humic acids will discolor the water, making it brown or black (Ahmad et al., 2017). They also unfavorably decrease the effectiveness of oxidants and disinfectants, and produce troublesome disinfection byproducts during oxidation (Juretic et al., 2015). They can contribute to biogrowth in the water distribution system and affect the performance of drinking water treatment systems which use membrane filtration and granular activated carbon (Özdemur, 2014).

The emergence of pathogenic bacteria in drinking water is a common cause of outbreaks of disease. In 2010, WHO revealed that over 2.6 billion people lacked access to clean water, contributing to around 2.2 million deaths annually (Pandey et al., 2014). In many countries, chlorine compounds are generally used as disinfectants to kill bacteria in water treatment plants. Nevertheless, potential harm to humans can still arise due to the presence of DOM in water, which encourages the production of disinfection byproducts (DBPs) when reacting with chlorine in drinking water. DBPs include haloacetic acids (HAAs), trihalomethanes (THMs), haloacetonitriles (HANs), iodo-THMs, haloketones (HKs) and nitroamines (Wu et al., 2013). The primary source of THMs is natural organic matter. DBPs that form as a result of the chlorination of NOM are probable carcinogens to humans and aquatic life forms (Hanigan et al., 2017) and can have other serious and chronic effects on human health. Thus, the removal of pathogenic bacteria and NOM is an important aspect in the production of safe drinking water.

Recently, the inductively coupled plasma system (ICPS) has been revealed to be a functional, low-energy and environmentally efficient tool for pathogenic bacteria disinfection, as well as for natural organic matter degradation in water. As a source of energetic charged particles, high electric fields, UV light and even shockwaves, plasma can trigger the production of OH^- in water (Foster, 2017). This generates radical species (H^+, O^{-2}, and OH^-) and radical molecules, such as ozone (O_3) and peroxide (H_2O_2), with high oxidation potential that can be used to remove microorganisms.

Desmiarti et al. (2015) investigated the removal of pathogenic bacteria from different water sources (river, groundwater and spring water) by ICPS, using three different reactor sizes (diameters of 1, 2 and 3 inches) and three different applied frequencies (3, 3.3 and 3.7 MHz) in batch experiments. It was found that increasing the applied frequency in addition to decreasing the plasma reactor diameter led to a reduction in the death yield of pathogenic bacteria. It was also reported that the removal efficiency of total coliforms was 86, 91 and 100% for river water, groundwater and spring water respectively, after 20 minutes of treatment using an applied frequency of 3.7 MHz and a 2-inch plasma reactor.

Another pathogenic bacterium in water is Salmonella typhi (S. typhi). As explained by Levatensi et al. (2012), S. typhi can be categorized as a food-borne bacterium that spreads through drinking or tap water and can cause intestinal illnesses such as paratyphoid fever and typhoid. These are known to be cause of human deaths in Indonesia, with more than 100 cases per population of 100,000 reported (Amicizia et al., 2017). The removal of S. typhi by an ICPS system has been studied using continuous flow experiments (Desmiarti et al., 2017). It was found that its removal strongly depends on the flow rate. On the other hand, the disinfection efficiency of S. typhi and the death rate constant decreased from 75% to 48% and from 0.57/h to 0.33/h respectively, with an increase in the initial number of S. typhi bacteria. Compared to the flow rate, the initial number of S. typhi bacteria had only a slight effect on energy consumption and death yield; energy consumption fell from 0.28 to 0.07 KWh/l, while the death yield increased from 784 to 1889 CFU/KWh, with an increase in flow rate from 5 to 20 ml/minute. It was concluded that flow rate is the key parameter in predicting the disinfection rate of pathogenic bacteria in drinking water treatment.

Desmiarti et al. (2018a, 2018b) have also investigated the influence of flow rate on removal efficiency (RE), death yield and the death rate of bacteria when using an inductively coupled plasma system. Energy consumption was also calculated in their studies. Water was taken from Kuranji River in Padang, West Sumatra, Indonesia and the applied radio frequency was 4.6 MHz. After 30 minutes of treatment, the disinfection efficiency of total coliforms, other coliforms and fecal coliforms increased to 44, 54 and 73%, respectively. Disinfection efficiency increased to 74, 89 and 100% with respect to total coliforms, other coliforms and fecal coliforms respectively, after treatment was run for 60 minutes. Compared to flow rate, the frequency applied in the ICPS had a less significant effect on the efficiency of the disinfection of pathogenic bacteria.

Regarding the degradation of organic matter by means of ICPS, little has been reported. Therefore, this research aims to investigate the application of a combination of microfiltration and ICPS to remove DOM and pathogenic bacteria from river water as a drinking water resource. The most significant chromophoric fraction is proteinaceous material related to extracellular microbial products and plant litter degradation products, such as fluorescent amino acids, tryptophan and tyrosine. These amino acid-like fluorophores form at lower emission wavelength ranges than typically observed for humic fluorophores. More chemical information can be obtained from the fluorescent classes of natural organic substances present in DOM by knowing the wavelength ranges at which the molecules both absorb and emit light than by absorbance information. For this reason, the observation of ubiquitous fluorescent organic substances in water samples through fluorescence spectroscopy has been proven to be a powerful technique for investigating the source and chemical composition of DOM. Hence, the characteristics of the DOM were measured based on three-dimensional excitation emission matrix fluorescence spectroscopy (3D-EEMs). Total DOM as dissolved organic carbon (DOC) and the ultraviolet absorbance with wavelength 260 nm (UV260) were investigated to analyze the content of the natural organic compounds in the treated water. The disinfection efficiencies of salmonella, total coliforms and fecal coliforms were also revealed.

2. METHODS

2.1. Water Source

River water from the largest river in Padang city, Indonesia was utilized in the experiment. The water properties are shown in Table 1.

Table 1 Initial water properties

Parameter	Unit	Value
pH	-	10–10.8
Total dissolved solids (TDS)	mg/l	49–65
Temperature	°C	30
Dissolved organic carbon (DOC)	mg/l	28.75
Fecal coliforms	CFU/100 ml	3,800
Total coliforms	CFU/100 ml	16,400
Salmonella typhi	CFU/100 ml	2,800

2.2. Experiment

The experimental set-up for the combined microfiltration-ICPS system has been described by Desmiarti et al. (2018a) and is shown in Figure 1. The equipment consists of microfiltration filters with sieves of 5 μm and 1 μm pore size, a plasma reactor, and a plasma generator. The volume of the plasma reactor is 100 mL and the glass reactor is wrapped with 2 mm copper wire. A current probe P6022 (Tektronix Inc., USA), a high-voltage P6015A (Tektronix Inc., USA), and a Picoscope 4424 (Picotech, United Kingdom) were used to check the applied voltage and discharged currents. The applied frequencies were established at 1.8−1.9 MHz. The experiments were conducted at flow rates of 100 and 150 ml/minute to investigate the performance of the filtration-ICPS system, with a peristaltic pump used to control the flow rate.

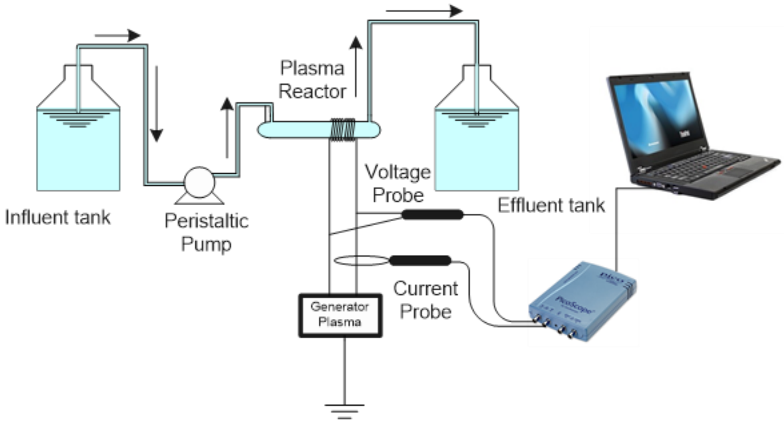

Figure 1 Experimental set-up of the combined filtration-ICPS system

2.3. Analysis

To examine the number of pathogenic bacteria, the plate count method is commonly used (Desmiarti et al. 2015, 2018a). Five ml of melted nutrient agar (brilliant green lactose bile 2% broth made by Oxoid, UK) was poured onto a petri dish (diameter 5 cm) and then 1 ml of water influent or effluent was added. The petri dish was stored in an electrical incubator at 37 ºC for 24 hours. The total number of bacteria was measured in a colony-forming unit (CFU/100 ml) by counting purple colonies as total coliforms and pink colonies as fecal coliforms. Additionally, the number of salmonella bacteria was measured according to the method explained in Desmiarti et al. (2017). The water samples were filtrated using a 0.2 μm cellulose acetate membrane filter (produced by ADVANTEC Corporation). The UV-absorbance of the filtrated water was analyzed at 260 nm (by a U-3210, manufactured by Hitachi Co., Ltd). The total dissolved organic carbon (DOC) was analyzed using a TOC-V, manufactured by Shimadzu. 3D-EEM spectroscopy (RF-5300 PC spectrofluorometer, Shimadzu) was used to distinguish between the different types and sources of natural organic matter in the water samples. By determining the emission wavelength, fluorescence intensity and excitation wavelength, it is possible to obtain an accurate fingerprint of the water samples, since specific excitation-emission (E_x/E_m) wavelengths can be correlated with certain molecular structures.

3. RESULTS AND DISCUSSION

3.1. UV260 and DOC

The behavior of natural organic matter with variation in flow rate is shown in Figure 2. DOC concentration fell after over 180 minutes of the experiment, and DOC concentration removal efficiency at flow rates of 100 and 150 mL/minute was 46.61% and 33.04%, respectively. Decreasing the flow rate resulted in an increase in removal efficiency, as the contact time increased. However, the optimum flow rate has yet to be decided, as the experiment was preliminary research to investigate the performance of the combined filtration-ICPS system.

Degradation of natural organic substances using an ICPS system may occur in many chemical reactions. NOM can be degraded into low molecular substances, forming CO_2 and water, thus decreasing DOC concentration. The removal efficiency of UV_{260} absorbance was 47.35% and 39% at flow rates of 100 and 150 mL/minute, respectively. During the experimental runs for the two flow rates, the trends of the UV_{260} absorbance fluctuated, so it was difficult to determine clear behavior. It is thus reasonable to assume that the effluent consisted of effectively degraded UV_{260} constituents. Meanwhile, the other constituents not detected by UV_{260} were less favorably removed.

With reference to a similar method, the microwave technique has been found to be effective in degrading larger molecules into smaller compounds (Usman et al., 2018). A low-power microwave apparatus was used to drive the bond breaking of chitosan into nanoparticles with different sizes (3–17 nm). Moreover, the use of biosorbent with trivalent metals (e.g. La, Nd, Al, Mg and Ti) was found to have potential for the removal of fluoride ions from water (Kusrini et al., 2015). Both of these techniques can be taken into consideration to increase the removal efficiency of DOC through this experiment.

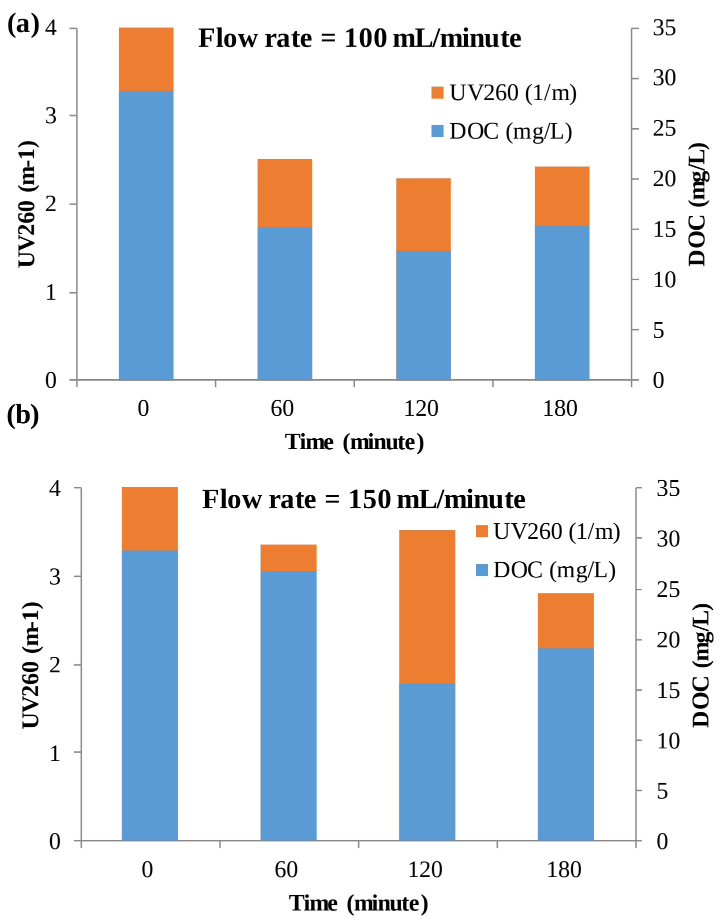

Figure 2 Profile of organic matter by UV260 absorbance and DOC concentration: (a) flowrate = 100 mL/minute; and (b) flowrate = 150 mL/minute

This result is in accordance with Tubic et al. (2011), who conducted advanced oxidation processes (AOP) in a pilot drinking water treatment system plant with a capacity of 2 m^3/h to remove natural organic matter (NOM) from groundwater. The pilot plant was set up to investigate a variation of method scheme, consisting of a series of processes such as sand/anthracite filtration, granular activated carbon (GAC) adsorption, ozone (O_3), intermediate ozonation, combined O_3/H_2O_2, and coagulation/sedimentation. The results showed that 10–14% of UV_{254} absorbing material was removed in the pre-oxidation step and that the reduction increased to 71–78% after intermediate ozonation, suggesting that degradation of natural organic substances by ozone can remove NOM from water.

3.2. Profile of Pathogenic Bacteria Disinfection Efficiency

The profile of pathogenic bacteria disinfection efficiency is shown in Figure 3.

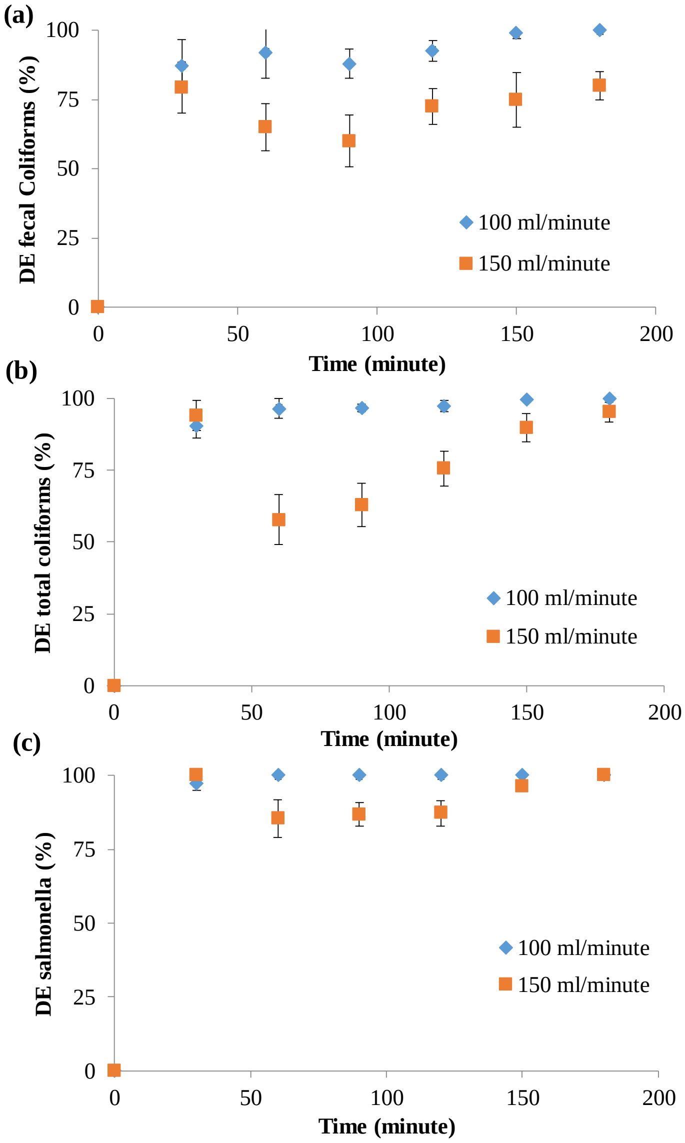

Figure 3 Profile of pathogenic bacteria disinfection efficiency:
(a) fecal coliforms; (b) total coliforms; (c) salmonella

In all the runs, the disinfection efficiency of fecal coliforms, total coliforms and salmonella decreased after the first 60 minutes. After 90 minutes, stable increasing trends were observed until 180 minutes of the experiment. This suggests that the disinfection of the pathogenic bacteria increased during the running process. However, an increase in flow rate prompted a decrease in pathogenic bacteria disinfection efficiency. Average disinfection efficiency was 87.0–100%, 90–100% and 90–100% for fecal coliforms, total coliforms and salmonella respectively, at a flowrate of 100 mL/minute. Increasing the flow rate to 150 ml/minute was found to reduce average disinfection efficiency to 60–79.9%, 58–95.3% and 85–100%, also for fecal coliforms, total coliforms and salmonella respectively. These results suggest that the disinfection efficiency of pathogenic bacteria decreases with an increase in the flow rate. Further research is needed in which the frequency applied is increased to observe if all the pathogenic bacteria in the water can be removed.

3.3. Fluorescence Properties

The chemical composition of the DOM contained in the water samples was investigated through its 3D-EEM properties (Hudson et al., 2007). Fluorescence in different spectral regions represents different functional groups, based on their excitation and emission wavelengths (Rajapaksha et al., 2019). Figures 4 and 5 show the 3D-EEM fractions of the DOM from the river water and treated water at flow rates of 100 and 150 ml/minute.

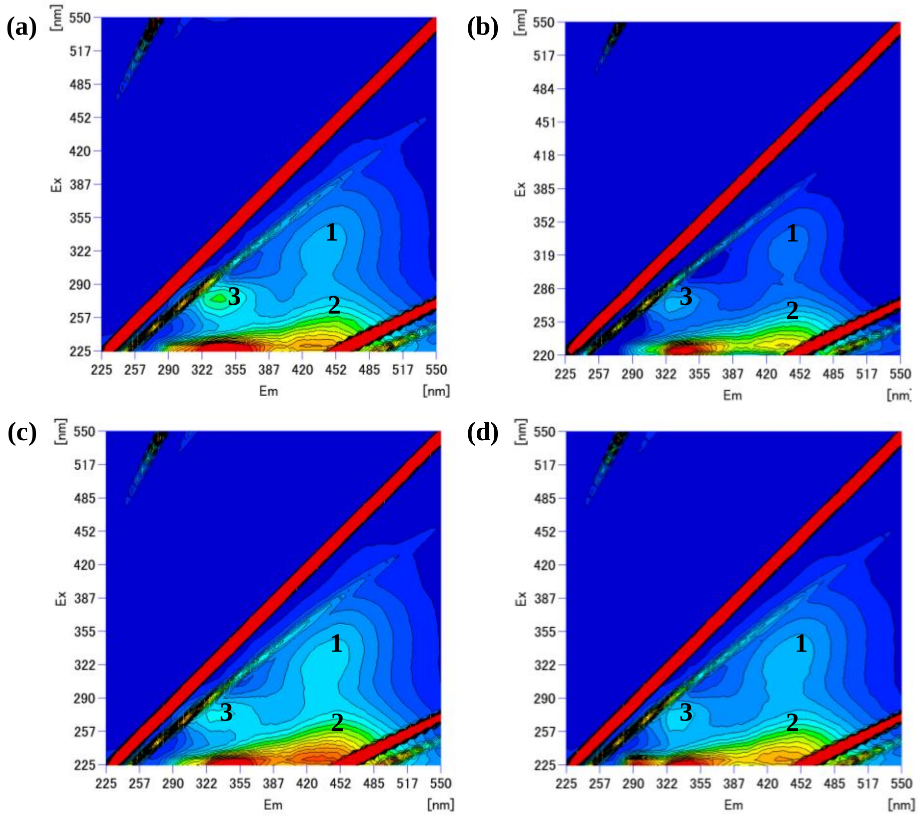

Figure 4 3D-EEMs at a flow rate of 100 ml/minute: (a) raw water; and after (b) 60 minutes; (c) 120 minutes; and (d) 180 minutes of treatment. The horizontal and vertical lines dividing the EEMs into three peaks represent the DOM fractions; i.e. **peak 1** (E_x/E_m with E_x: 320–350 nm and E_m: 435–450 nm), and **peak 2** (E_x/E_m with E_x: 250–260 nm and E_m: 430–460 nm), which both represent humic-like substances, and **peak 3** (E_x/E_m with E_x: 270–280 nm and E_m: 320–350 nm), which represents tryptophan-like substances

The fluorophores exhibiting fluorescence at excitation/emission wavelengths suggested by **peak 1** (E_x/E_m with E_x: 320–350 nm and E_m: 435–450 nm) and **peak 2** (E_x/E_m with E_x: 250–260 nm and E_m: 430–460 nm), as shown in Figures 3 and 4, represent humic-like substances derived from the breakdown of microbes and plants in water. Similar to humic-like material, tryptophan-like material, as free molecules or bound in proteins and amino acids, also indicate fluorescence at distinctive wavelengths in water, as represented by **peak 3** (E_x/E_m with E_x: 270–280 nm and E_m: 320–350 nm).

DOM is made up of decaying plants and microbes. In water environmental systems it is always accompanied by an active microbial community, which consumes oxygen, leading to high levels of biological oxygen demand (BOD), causing drops in oxygen levels that can adversely affect aquatic ecosystems. Protein found in the cell walls of these microorganisms has been shown to fluoresce in the same region as one of the amino acids, tryptophan. Thus, tryptophan-like fluorescence (TLF) is also likely to identify the input of polluting organic matter and is often associated with organic carbon originating from sewage or farm waste; it can be used as a measure of microbial activity within a water body (Hudson et al., 2008).

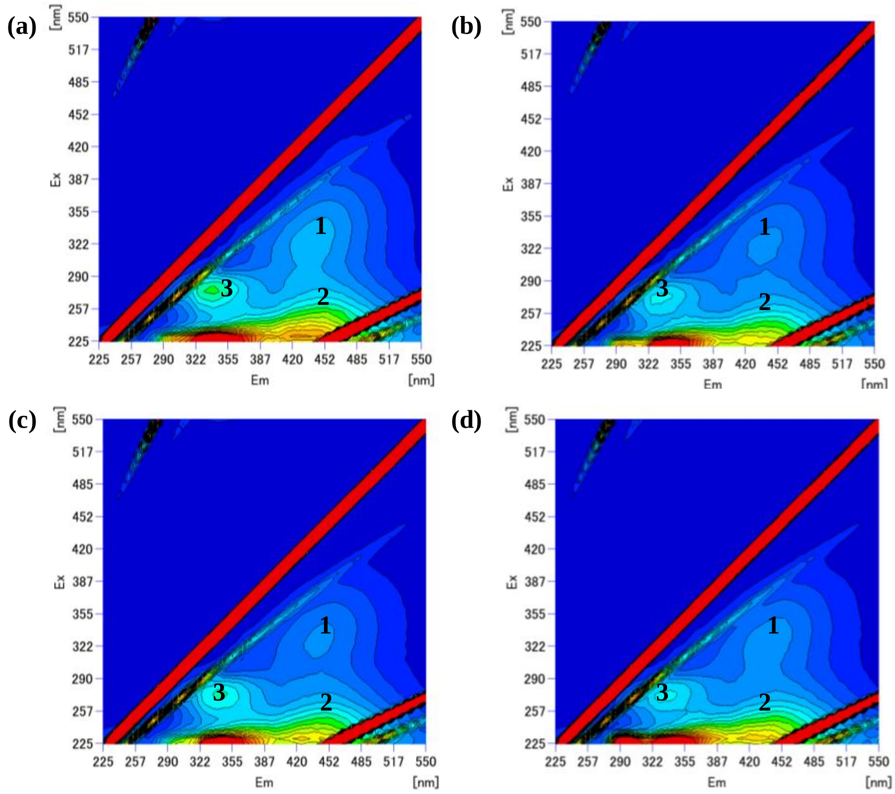

Figure 5 3D-EEMs at a flow rate of 150 ml/minute: (a) raw water; and after (b) 60 minutes; (c) 120 minutes; and (d) 180 minutes of treatment. The horizontal and vertical lines dividing the EEMs into three peaks represent the DOM fractions; i.e. **peak 1** (E_x/E_m with E_x: 320–350 nm and E_m: 435–450 nm) and **peak 2** (E_x/E_m with E_x: 250–260 nm and E_m: 430–460 nm), which both represent humic-like substances, and **peak 3** (E_x/E_m with E_x: 270–280 nm and E_m: 320–350 nm), which represents tryptophan-like substances

The TLF peak is generally associated with excitation at 270–280 nm and emission at 320–350 nm. From Figures 4 and 5, it can be observed that after 180 minutes of the experiment, only the peak intensity of the tryptophan-like substances (peak 3) had been reduced at both flow rates, suggesting the removal of pathogenic bacteria. The ICPS has several properties that are detrimental to bacteria. Reactive species and UV radiation will break one or both of the long strands of microorganism DNA, resulting in cell death.

The active species will also attack the nearest stable molecule and steal its electron. When the attacked molecule loses its electron, it becomes a free radical, starting a chain reaction. Once the process has been started, it can cascade and ultimately lead to the disruption of living cells (Kashmiri & Mankar, 2014). As can be seen from peaks 1 and 2, the ratio of the maximum Ex/Em wavelengths of the humic-like components was the same at both flow rates.

4. CONCLUSION

A combined filtration-ICPS was studied to remove pathogenic bacteria, as well as natural organic matter from river water. The disinfection efficiencies of fecal coliforms, total coliforms and *Salmonella* typhi fell with an increase in flow rate, suggesting that this is an important parameter. The combined filtration-inductively coupled plasma system was also found to be sufficiently proficient to remove organic matter constituents, as suggested by the UV260 absorbance and DOC measurements. Furthermore, the 3D-EEMs results suggest that removal of tryptophan-like substances from raw water occurred, indicating the removal of pathogenic bacteria. Thus, the combined filtration-inductively coupled plasma system was found to be a prospective advanced technology in drinking water treatment, capable of simultaneous organic and microbial removal by filtration and degradation mechanisms.

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