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Dissolved organic matter released from paper trash in water during ultraviolet irradiation: impacts on trihalomethane formation

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ABSTRACT

When exposed to ultraviolet (UV) light, paper trash could release dissolved organic matter (DOM) as a disinfection byproduct (DBP) precursor in water. This study was undertaken to assess the regulated DBPs, trihalomethanes-4 (THM4) generation potential, and estimated cytotoxicity of paper trash soaked in water during UV radiation. The UV irradiation was carried out at 1, 6, 12, 24, and 48 hours. The released organic material was determined using ultraviolet-visible (UV-Vis) adsorption at wavelengths ranging from 200 to 700 nm. Chlorination was carried out for 24 hours to determine THM4 formation potentials. According to the research findings, the longer the radiation period, the more THM4 species are formed in plain paper samples. On the other hand, the production of trichloromethane (TCM) on paper samples containing writing decreased as the UV radiation period increased. THM4 levels reached 103.12 and 150.57 µg/L for plain and writing paper samples, respectively. UV absorbance at 272 nm (A272) shows a 92% association with THM4 production, making it suitable as an initial characterization approach. Paper trash pollution in water could form carcinogenic DBPs, THM4, if used as a water source in a water treatment plant using chlorine disinfection.

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1. INTRODUCTION

Paper is an irreplaceable resource even as time passes. Paper is utilized in office buildings, schools, and shopping malls. This is also reinforced by the COVID-19 epidemic, in which it is utilized as envelopes and package wrappers in the buying and selling process, which is frequently done online. The growing demand for paper has contributed considerably to a rise in residential waste in Indonesia and around the world [1]. In Indonesia, garbage disposal is not limited to the final disposal site [1], [2]. People's unhealthy habits result in vast garbage being dumped into water bodies such as rivers and lakes, which are the primary water supplies for drinking water treatment plants (WTP) and the ocean [3]. Paper waste is known to release organic compounds in water bodies [4]–[6], lowering the dissolved oxygen and causing water pollution [7]. Waste disposed of in landfills can leach, causing dissolved organic matter (DOM) to contaminate ground and surface water, particularly when exposed to ultraviolet (UV) rays from the sun [4]. Directly tossing rubbish, on the other hand, is more likely to release organic material and pollute the water [5].

Water disinfection is crucial for protecting human health from aquatic diseases such as cholera, dysentery, and leptospirosis [8]. The use of chlorine as a final disinfectant has the advantage of allowing residual chlorine to combat bacteria during water storage and distribution [9], [10]. The presence of DOM in water bodies may cause disinfection byproducts (DBPs) such as trihalomethanes (THMs), haloacetic acids (HAA), haloacetonitriles, halonitromethanes, and other DBPs formation during water disinfection with chlorine [11], [12]. In general, the concentrations of THMs were substantially greater than those of other by-products [13].

THMs are halogenated organic molecules in which three of the four hydrogen atoms in methane (CH4) are replaced with halogens. THMs are also environmental contaminants, and several are considered carcinogenic [14], [15]. THM4 (trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform (TBM)) is a group B carcinogen (it has been shown to cause cancer in experimental animals). The United States Environmental Protection Agency (USEPA) established a maximum contamination threshold of 80 µg/L for total THM4 [15]. The WHO's guidelines for drinking water quality (GDWQ) set limits for total THMs of 300 µg/L for TCM, 100 µg/L for TBM, 60 µg/L for BDCM, and 100 µg/L for DBCM [16].

Ultraviolet-visible (UV-Vis) absorbance has been used to identify DOM in water and is linked to the development of DBPs [17]–[19]. The advantage of UV-Vis absorbance is that it can identify DOM's chromophore characteristics [20]. On the other hand, chromophore DOM has been shown to have high DBP precursor properties [21]. Several investigations have employed UV-Vis absorbance to detect DOM and the possible generation of DBPs such as THMs [22], [23], HAAs [24], and phenolic DBPs [25]. Previous studies mostly only relate UV-Vis spectra to the presence of natural organic matter (NOM) while the relationship with anthropogenic organic matter is still very little known [22]–[25]. To the best of our knowledge, publication on the detection of organic matter in water from paper trash associated with THMs formation is still lacking.

Given the increasing production of paper trash, mitigation must be implemented to determine the impact of this waste on the presence of organic compounds in polluted water. Using UV-Vis's absorbance spectra, this work has developed a novel method to investigate the characteristics of organic compounds created by paper waste in water under UV irradiation. The possibility for THM4 formation and the resulting cytotoxic risk assessments were investigated to estimate the impact of employing water polluted with paper waste as a water source in the water treatment process using chlorine disinfection.

2. RESEARCH METHOD

2.1. Samples and chemicals

The paper samples used are plain paper and written paper from campus garbage. Deionized water is used to avoid organic contaminants that could disrupt the research process. Sigma Aldrich (USA) provided the THM4 standard, while Merck (Germany) supplied methyl tertiary butyl ether (MTBE) and potassium bromide. Local (Indonesia) sodium sulfate and sodium hypochlorite were purchased from Bratachem and Pudak Scientific, respectively.

2.2. UV irradiation

Figure 1 shows a schematic diagram for the UV irradiation process. Each piece of paper was weighed at 5 grams and soaked in 500 mL of deionized water in a glass beaker. During immersion, UV light (254 nm) was exposed for 1, 6, 12, 24, and 48 hours to simulate solar radiation during that time. After UV exposure, the sample was filtered through a 0.45 µm filter to remove suspended particles before being prepared for UV absorbance measurement and chlorination.

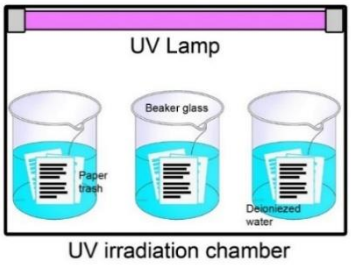

Figure 1. Schematic diagram of the UV irradiation process

2.3. Chlorination and THM4 extraction

The chlorination process was carried out for 24 hours on 96 mL of sample water by adding 1 mL of 10% sodium hypochlorite solution, 1 mL of potassium bromide, and 1 mL of pH 7 phosphate buffer. The

samples were maintained at room temperature and in the dark for 24 hours. After 24 hours, 1 mL of saturated sodium sulfite solution was added to terminate the chlorination process, and the sample was ready for THM4 extraction.

The THM4 extraction technique followed the USEPA's [26] liquid extraction (LLE) guidelines. A total of 50 mL of sample was deposited in a 60 mL EPA vial, then 20 grams of sodium sulfate and 3 mL of MTBE were added. The vial was then closed and shaken thoroughly for four minutes. After 4 minutes, the sample was allowed to split into two phases for 5 minutes. After the phases were separated, 1 mL of the MTBE phase was collected with a micropipette and transferred to a 1.5 mL autosampler vial for THM4 measurement.

2.4. Analytical methods

A UV-Vis spectrophotometer (Shimadzu UV-1700) was used to examine the chemical substances produced by paper samples. The sample is examined in the 200-700 nm wavelength range to identify the absorbance peak. Agilent 7890A gas chromatography combined with Agilent 5975C mass selective detector (MSD) (GC-MS) was utilized for THM4 analysis under conditions based on previous research [9]. Based on THM4 formation data, estimated cytotoxicity was calculated by dividing the molar concentration of each reported THM4 (MTHM4) by the provided cytotoxicity (LC50) values (MLC50) when exposed to Chinese hamster ovarium (CHO) cells (1) [27].

$$Est.cytotoxicity = \frac{M_{THM4}}{M_{LC50}} \quad (1)$$

3. RESULTS AND ANALYSIS

3.1. UV-Vis absorbances

Figure 2 depicts the UV-Vis absorbance of paper samples that were soaked in water and exposed to UV light for 1, 6, 12, 24, and 48 hours. Plain paper samples exhibit two absorbance peaks at wavelengths of 272 and 350 nm (A272 and A350), as shown in Figure 2(a). The A272 value rises as the UV light exposure time increases. However, the fluctuations in A350 values follow a random pattern. Based on these UV absorbance measurements, peak A272 can be attributed to the breakdown of DOM in paper. The A350 value, on the other hand, is a derivative or a change in the form of DOM from paper that is further degraded by UV light; hence, the variations are categorized as random [28]. The decreasing trend of UV absorbance is in line with previous research, which states that the UV-Vis spectrum of water containing DOM has a decreasing trend as the wavelength increases [24]. The peak at 272 nm is in line with previous studies that reported the presence of NOM in drinking water [29]. A272 indicates the presence of dissolved phenolic compounds in water [29]. On the other hand, A350 indicates the existence of simpler organic molecules and has a lower molecular weight [30]. Similar to the plain paper sample, written papers show two prominent peaks appearing at 272 and 350 nm (Figure 2(b)). After being subjected to UV light radiation for more than 24 hours, written paper samples also show a peak at 630 nm (A630), which distinguishes them from plain paper samples. Even while A272 has a higher value in the first hour of written paper samples than plain paper samples, the increase in UV absorbance of written paper samples occurs faster. This leads to A272 after 48 hours being significantly higher than plain paper. This is possible because UV light dissolves not only organic molecules from paper but also pigments found in writing ink [31]. Despite its irregularity, the A350 values increase after 1 and 48 hours of radiation exposure. The increase was caused by the creation of byproducts from the breakdown of dissolved organic compounds in paper and ink under UV light radiation [32].

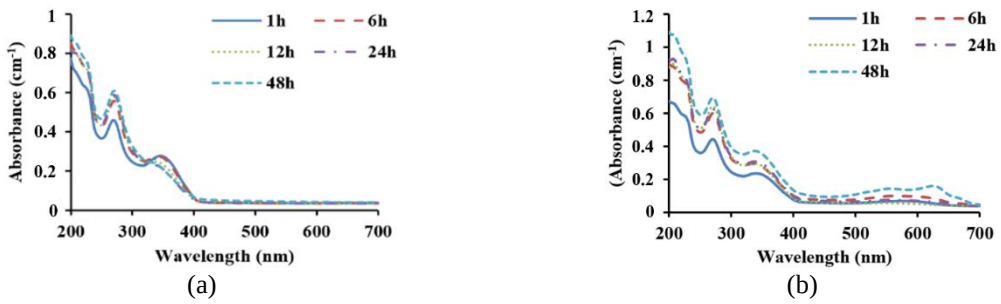

Figure 2. UV-Vis's absorbance of deionized water immersed by (a) plain paper samples and (b) written paper samples during the UV irradiation process

3.2. THM4 formations

Figure 3 depicts the influence of UV irradiation time on THM4 formation in plain and written paper samples that had been soaked in water after 24 hours of chlorination. In plain papers, all THM4 species increased as the irradiation time increased (Figure 3(a)). After one hour of UV irradiation, TCM, BDCM, CDBM, and TBM reached 6.14, 4.98, 6.75, and 6.63 µg/L, respectively. After 48 hours of irradiation, TCM concentration increased by more than twice, whereas CDBM concentration increased by one and a half times. The creation of BDCM and TBM grew significantly more than TCM and CDBM. The formation of TBM and BDCM increased by 722.34 and 424.58%, respectively. The presence of hydrophilic organic molecules generated during the UV radiation process can cause a considerable rise in TBM [24]. UV light can also generate radicals and convert the DOM in paper to simpler, hydrophilic organic matter [33]. The rise in TCM is not significant because organics leached from paper are not the primary precursor of TCM.

As the radiation period rose in written paper samples, the number of THM4 species that bind to bromine (DBCM, CDBM, and TBM increased (Figure 3(b)). TCM formation, on the other hand, had the reverse outcomes, with formation decreasing as UV radiation time increased. THM4 production in written paper samples is higher than in plain paper, particularly for non-combined THM4 (TCM and TBM). After one hour of UV exposure, TCM, BDCM, CDBM, and TBM formation rates were 79.15, 2.36, 0.47, and 30.82 µg/L, respectively. After 48 hours of irradiation, TCM formation was reduced by more than three times its prior concentration. The creation of CDBM and TBM rose significantly compared to the prior concentration, while CDBM formation elevated, although not dramatically. The formation of BDCM and TBM increased far more than TCM and CDBM. The formation of BDCM and TBM rose by 1258.05 and 280.66%, respectively. TBM levels can rise significantly when hydrophilic chemical compounds from paper and ink are produced during the UV radiation process [34]. This is also caused by the release of bromine during the UV radiation process due to the presence of bromide in the water [35]. Additionally, DOM in paper can be broken down by UV radiation into simpler, hydrophilic organic materials and radicals [36]. TCM formation may decrease as a result of the breakdown of phenolic chemicals leached from paper and ink during the UV radiation process. The THM4 formation for DOM released by paper trash has a higher formation compared to that released by microplastics [37]. Paper is created from wood, which contains organic components such as lignin and cellulose, which are precursors of DBPs [38], whereas ink may contain synthetic organic dyes, which are also precursors of DBPs [39]. The total THM4 formation is shown in Figure 3(c).

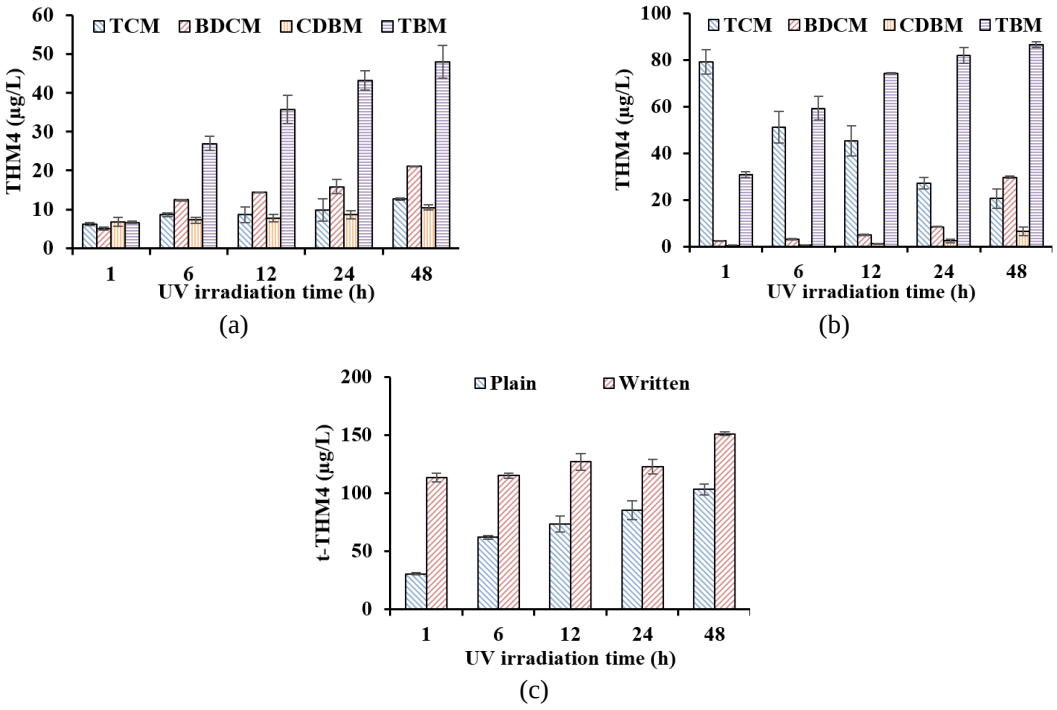

Figure 3. Effect of UV irradiation time on THM4 formation during chlorination of water immersed by plain and written paper of (a) plain paper, (b) written paper, and (c) t-THM4 formation (24 hours chlorination, Cl2 10 mg/L, and Br- 1 mg/L)

Because the formation of all THM4 species increased during the UV irradiation of plain and written paper samples in water following chlorination, total THM4 (tTHM4) increased significantly. The concentration of t-THM4 in plain paper samples increased from 30.48 µg/L after 3 hours of UV light to 103.12 µg/L after 48 hours, exceeding the USEPA standard. Based on this, the presence of paper in water leads to the formation of THM4 if the water receives exposure to UV light because it can release organic chemicals, which are THM4 precursors. The concentration of t-THM4 in the written paper samples increased from 113.51 µg/L after 1 hours of UV light to 150.97 µg/L after 48 hours, exceeding the USEPA standard. Based on this, the presence of paper and ink in water adds to the creation of THM4 when exposed to UV radiation, as it can release chemical molecules that are THM4 precursors.

3.3. Estimated cytotoxicity

Figure 4 shows the effect of UV radiation time on the cytotoxicity calculation of the formed THM4. The changing trend of cytotoxicity based on THM4 formation is in line with the formation of THM4 itself. However, the difference in cytotoxicity of each THM4 species makes the cytotoxicity trend slightly different with the concentration of THM4 formation (Figure 4(a)). Since the LC50 concentration value of TCM is smaller than that of BDCM, the cytotoxicity of TCM is greater than that of BDCM [27]. This causes the cytotoxicity value of TCM to be greater and equivalent to BDCM, even though the concentration of TCM formation is smaller. TBM still has a large cytotoxicity compared to other THM4 species due to the large formation concentration and small LC50 concentration value. The presence of ink in written paper not only increased the formation of higher THM4, but also increased its cytotoxicity (Figure 4(b)). The decrease in TCM formation as UV radiation time increased led to an increase in THM4 cytotoxicity in samples with writing that was not as significant as in plain paper samples. The cytotoxicity of THM4 formed from DOM released by paper waste has a higher value than organic matter influenced by textile dye [22] reported by a previous study. The total THM4 estimated cytotoxicity is shown in Figure 4(c).

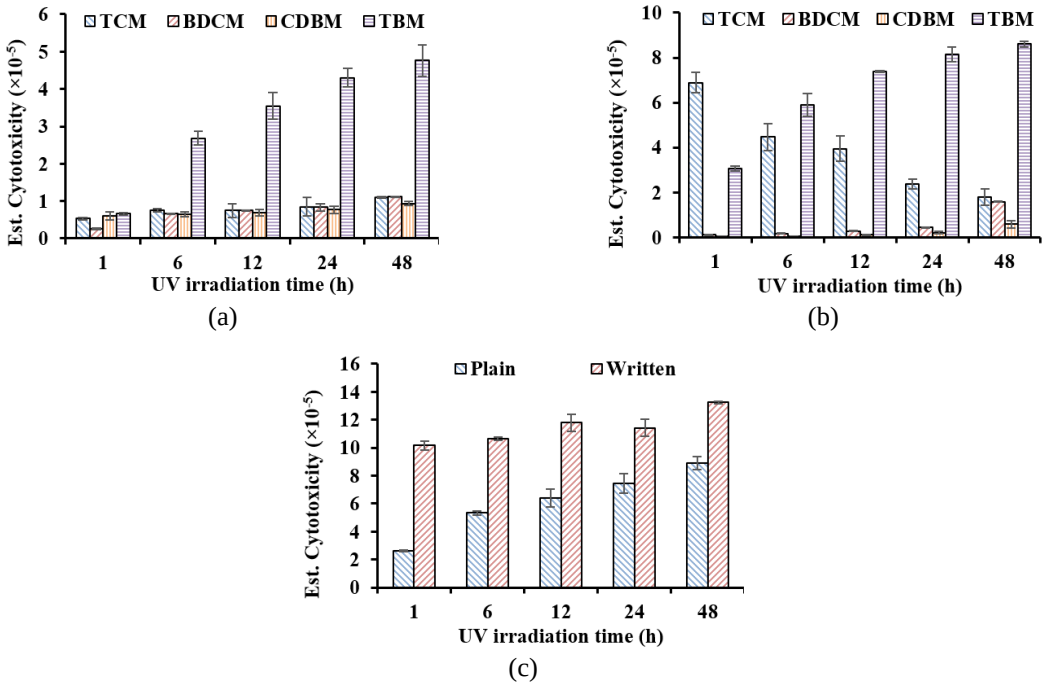

Figure 4. Effect of UV irradiation time on THM4 estimated cytotoxicities during chlorination of water immersed by plain and written paper of (a) plain paper, (b) written paper, and (c) t-THM4 (24 hours chlorination, Cl2 10 mg/L, and Br- 1 mg/L)

3.4. Correlation between UV-Vis absorbances and THM4 formations

Figures 5 and 6 show the correlation between the UV absorbance peak (A272 and A350, respectively) and the formation of THM4. In plain paper samples, the value of A272 has a very strong correlation with THM4 formation (92%) (Figure 5(a)). On the other hand, the correlation between A272 and THM4 formation in the written paper samples had a moderate correlation (54%) (Figure 5(b)). A strong negative correlation was also found between A350 and t-THM4 formation in plain paper samples (84%) (Figure 6(a)). The negative value may be due to the degradation of organic compounds released from the plain

paper sample to form smaller molecular weight compounds. An equally strong but positive correlation exists between A350 and t-THM4 formation (84%) (Figure 6(b)). The peak at 272 nm is in line with a previous study [22], where this wavelength can interpret the presence of DOM associated with THM4 formation potential.

The positive value may occur because the released compounds with a smaller molecular weight are more than the degraded compounds due to the presence of ink on the paper. UV absorbance has long been used to predict the presence of organic compounds in water related to the formation of DBPs [22], [40]. The formation of DBPs depends on the type of precursors present in water, including the type of organic compounds and halogen ions such as bromide [12]. The findings indicate that higher UV absorbance was associated with the UV-irradiation time. The proposed method may benefit from one major pollutant source that is exposed to UV radiation, such as in a natural river.

Previous research found a correlation between THM4 formation potential and UV-Vis absorbance of water affected by textile effluents [22] and NOM [41]. In this study, the impact of waste paper on THM4 formation and its correlation with UV-Vis absorbance spectra was investigated. The proposed method in this study tended to have an inordinately high proportion of correlation between the UV absorbance peak and THM4 formation potential of DOM released from paper trash in bromide-containing water. This method could be used as a preliminary study to determine THM4 formation in paper waste-polluted water during the chlorination process. Based on the UV-Vis scan results, a proper water treatment method can be considered to remove THM4 precursors from paper trash-polluted water.

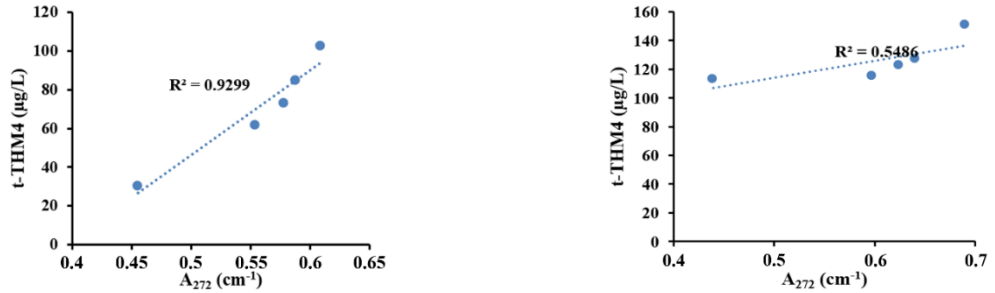

Figure 5. Relationship between A272 and t-THM4 formation in (a) plain paper and (b) written paper

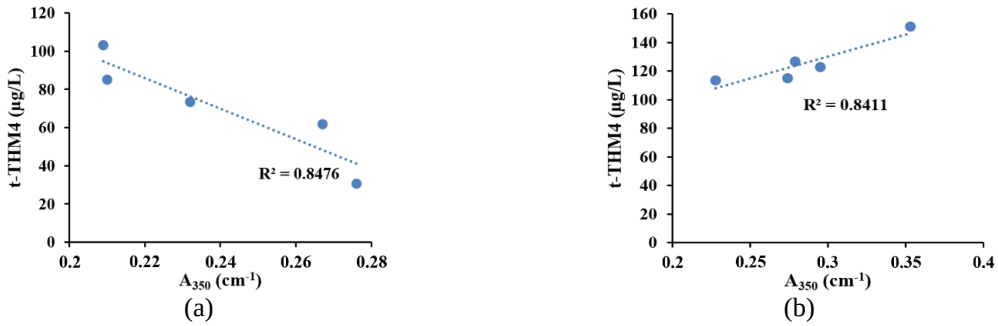

Figure 6. Relationship between A350 and t-THM4 formation in (a) plain paper and (b) written paper

4. CONCLUSION

The study was conducted to determine the effect of UV irradiation on the degradation of organic compounds on plain paper and paper with writing in water on the formation of THM4 and estimation of its cytotoxicity. The UV-Vis scanning method was also conducted to predict the formation. Based on the results, there are two peaks at wavelengths of 272 and 350 nm. The longer the UV radiation time, the greater the absorbance value of A272 and A350, which has an impact on the greater formation of THM4 and its cytotoxicity. A272 and A350 also have a strong correlation (up to 92%) with THM4 formation, so that they can be used as an initial quantification method. The findings of this study offer definitive proof that the change of UV-Vis's absorbance spectra during UV-irradiation of paper trash in water is linked to THM4 formation potential alteration, rather than being caused by increased quantities of DOM. This study helps to know how paper trash releases DOM in water during UV radiation in the environment, and its impact on toxic THMs formation during the water treatment process using chlorine. To find out which organic

compound species are cytotoxic and which other DBP species are degraded and formed when paper samples are exposed to UV light in water, more research is required.

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AUTHOR CONTRIBUTIONS STATEMENT

This journal uses the Contributor Roles Taxonomy (CRediT) to recognize individual author contributions, reduce authorship disputes, and facilitate collaboration.

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Kahar	✓		✓	✓			✓			✓	✓		✓	
Muammar Qadafi	✓	✓	✓		✓					✓	✓	✓		
Wisnu Prayogo					✓		✓			✓		✓		

C : **C**onceptualization
M : **M**ethodology
So : **So**ftware
Va : **Va**lidation
Fo : **Fo**rmal analysis
I : **I**nvestigation
R : **R**esources
D : **D**ata Curation
O : Writing - **O**riginal Draft
E : Writing - Review & **E**diting
Vi : **Vi**sualization
Su : **Su**pervision
P : **P**roject administration
Fu : **Fu**nding acquisition

CONFLICT OF INTEREST STATEMENT

The authors state no conflict of interest.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author, [NYH], upon reasonable request.

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