

# Analysis of Natural Organic Matter as Precursors of Trihalomethanes and Its Removal in Water Treatment Plant

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## Abstract

Disinfection is an important step in water treatment, but unpredictable compounds are produced as disinfection by-products (DBPs). DBPs are harmful products to human health so their formation must be prevented during treatment, especially for drinking water treatment. An organic compound such as natural organic matter (NOM) is the main precursor of DBPs due to its presence in river water. The Water Treatment Plant (WTP) in Indonesia, mostly uses river water as raw water. Analysis of the organic matter is very important as this research aims to determine the ability of the WTP units to reduce precursors and the formation of DBPs. The sample was filtered by a 0.45 $\mu$ m microfilter. The presence of NOM was evaluated by the dissolved organic carbon (DOC) parameters, ultraviolet at 254 nm absorbance (UV<sub>254</sub>), and specific UV absorbance (SUVA). Then measure the Trihalomethanes (THMs) as DBPs after the disinfection. The results of analysis from five WTPs, the DOC concentration is between 3.30 – 8.20 mg/L; UV<sub>254</sub> is 0.70 – 0.10 cm<sup>-1</sup>; SUVA is 0.90 – 3.13 L/mg.m. The ability of conventional WTP units such as coagulation-sedimentation-filtration in removing DOC is around 7% – 42%; UV<sub>254</sub> is 17% – 35%; SUVA is 6% – 69%. For the THMs, the concentration after the disinfection is 4.50 – 127  $\mu$ g/L. It can be concluded that conventional WTP units are not effective in reducing NOM and the potential of DBPs formation is still high.

**Keywords:** Disinfection by-products; Water treatment; Natural organic matter; Trihalomethanes

## 1. Introduction

Since it found that the disinfection process forms the by-product in the 1970s, disinfection by-products (DBPs) presence in drinking water treatment has become very important to pay attention. Until now, for more than 50 years, hundreds of types of DBPs compounds have been found in water treatment, both in drinking water treatment, wastewater treatment, and even in water use that uses chlorine such as swimming pools. Based on previous research, it has been shown that there are more than 600 types of DBPs and these compounds have the potential to cause health

problems in humans (Richardson *et al.*, 2011; Verdugo *et al.*, 2020). DBPs are classified as a harmful matter to human health and their presence in water should be of concern and effort especially for the water companies and stakeholders (Gilca *et al.*, 2020).

Among the many types of DBPs compounds, the ones that have been studied the most and have standard limit regulation of concentrations are DBPs compounds from the Carbonaceous DBPs group, namely Trihalomethanes (THMs) and Haloacetic acids (HAAs) (Chu *et al.*, 2011). The formation of

THMs (such as chloroform) and other THMs compounds in high concentrations in raw water processing was first discovered as DBPs, it was formed from the reaction of chlorine with natural organic compounds (NOM) (Rook, 1974). Another DBPs group that is often found in Nitrogenous DBPs are haloacetonitriles (HANs), halonitromethanes (HNMs), and haloacetamides (HAcAms).

Several DBPs have been regulated for the standard limits concentration by the U.S Environmental Protection Agency (EPA), such as 11 DBPs: four trihalomethanes (THM4), five haloacetic acids (HAA5), bromate, and chlorite (USEPA, 2019). The maximum concentration for THM (THM4: chloroform, bromo dichloromethane, dibromochloromethane, bromoform) is 80 µg/L and 60 µg/L for (HAA5: chloroacetic acid, bromoacetic acid, dichloroacetic acid, dibromoacetic acid, trichloroacetic acid) based on United States regulations (USEPA) and 10 ng/L permitted levels for three N-nitrosamines (N-nitrosodimethylamine (NDMA), N-nitrosodiethylamine, and N-nitrosodipropylamine) in California Department of Public Health (Furst et al., 2018).

The formation of DBPs is mostly determined by the precursor which is the main trigger when going through the disinfection process. There are many DBPs precursors including natural organic matter (NOM), algae organic matter (AOM), anthropogenic pollutants (such as pesticides, pharmaceuticals, detergents, etc.), bromine, and iodine compounds. Wastewater discharges and operational parameters in drinking water treatment such as type of disinfection agents, dose, pH, contact time, and temperature also contribute to the formation of DBPs (Alexandrou et al., 2018; Gilca et al., 2020). The concentration of DBPs depends on the presence and diversity of the number of organic compounds in the water, both natural (Natural Organic Matter, NOM) and dissolved (Dissolved Organic Matter, DOM).

In nature, NOM is formed from biological activities such as microbes, vegetation, water, and other wild animals. NOM decomposition in the environment is affected by chemical compounds and microbial activity

(Huang et al., 2020). NOM is usually found in the concentration range of 2 - 10 ppm in raw water from drinking water treatment plants (Levchuk et al., 2018). Both NOM and DOM when reacting with disinfection agents form DBPs such as trihalomethanes (THMs), haloacetic acids (HAAs), haloacetonitriles (HANs), and others which are harmful to human health (Sharma et al., 2021; Badaro et al., 2021).

The river is one of the main sources of raw water in WTP Indonesia, its currently experiencing significant changes both in terms of quantity and quality. There are socio-cultural dynamics in the surrounding area such as high population growth, infrastructure development, excessive natural resource exploitation, and land transformation (deforestation, agriculture, livestock, and other activities) which have influenced the decline of river water quality as raw water (Benitez et al., 2021). The high load of pollutants into the river further worsens the water quality and can threaten the health and safety of humans who consume it (Benitez et al., 2021; Bagatin et al., 2014). Decreasing raw water quality can affect the drinking water treatment process which has been designed for its ability to remove contaminants, but the high pollutant can reduce the performance efficiency of the processing unit. This can affect the presence of NOM and DOM precursors which are increasingly high and difficult to remove with conventional water treatment and the potential for the formation of DBPs is even greater.

The removal of DBPs precursors is very important as an effort to prevent the formation of DBPs (Benitez et al., 2021; Ersan et al., 2019; Li et al., 2017). The removal of DBPs precursors is one of the appropriate strategies to reduce the potential of DBPs formation in the water treatment process. Therefore, knowing the presence of precursor in water treatment greatly determines their removal targets. Analysis of precursors starts from the raw water source that will be used, then the analysis of each treatment unit of WTP to determine the unit's ability to reduce the precursors. Coagulation and filtration units can remove the organic matter precursor for 50 – 60% depending on the molecular weight

of precursors (Zhang *et al.*, 2020; MacKeown *et al.*, 2020). But generally, NOM still escapes after treat by the treatment unit of the Water Treatment Plant (WTP) and then reacts with chlorine in the disinfection process.

In Indonesia, research about the DBPs formation and the presence of NOM as the precursor of DBPs in WTP is still lacking. Moreover, there are no specific regulations regarding quality standards for NOM and DBPs for drinking water. Therefore, this paper is important to analyze the presence of NOM in WTP as the main precursor of DBPs by the measurement of UV absorbance ( $UV_{254}$ ), dissolved organic carbon (DOC), and specific UV absorbance (SUVA) from raw water to the effluent of the filtration unit. Knowing the presence of precursors NOM in water treatment can be a good consideration in analyzing the formation potential of DBPs and removal strategy.

## 2. Methodology

### 2.1 Water Samples

The selection of WTP is determined based on the location of the source of raw water: (1) WTP at Bogor because Bogor has Cisadane River (WTP Bogor 1) and Ciliwung River (WTP Bogor 2) as upstream rivers, then (2) Depok in the middle city flowed by several rivers such as Ciliwung River (WTP Depok 1) and Kali Angke River (WTP Depok 2), and last (3) Tangerang has Cisadane river as downstream river (WTP Tangerang). The specific location of WTP can be seen in

Figure 1. There are 5 different sampling point locations in each WTP, namely: (1) raw water (river), (2) intake channel, and treated water (3) effluent sedimentation unit, (4) effluent of filtration unit, and (5) reservoirs.

One-liter sample was taken using the grab sampling method. During the sampling, some in-situ parameters such as temperature, pH, dissolved oxygen, and turbidity were measured directly. The samples are stored in jerry cans and then put in a cooled box during transport to the laboratory. After that sample must be immediately taken to be prepared for organic compound analysis (no more than 2 hours).

### 2.2 Analytical Natural Organic Matter (NOM)

The DOC parameter is a quantitative description of organic compounds in water. Meanwhile,  $UV_{254}$  shows the aromatic nature of a compound in water, where aromatic compounds will tend to absorb UV at the 254 nm wave and show the main part as a DBPS precursor. SUVA is a hydrophobicity value that correlates with the potential for DBPs formation (Tak *et al.*, 2018). SUVA is a widely parameter to describe the presence of organic compounds in the water as DOC and the reactivity of organic compounds with chlorine in forming DBP (Hua *et al.*, 2015; Tak *et al.*, 2018).

The water sample is filtered first using a 0.45  $\mu\text{m}$  syringe filter, then the dissolved organic carbon (DOC) parameter is measured using a TOC-L 00466 Shimadzu Total Organic Carbon Analyzer, and the absorbance

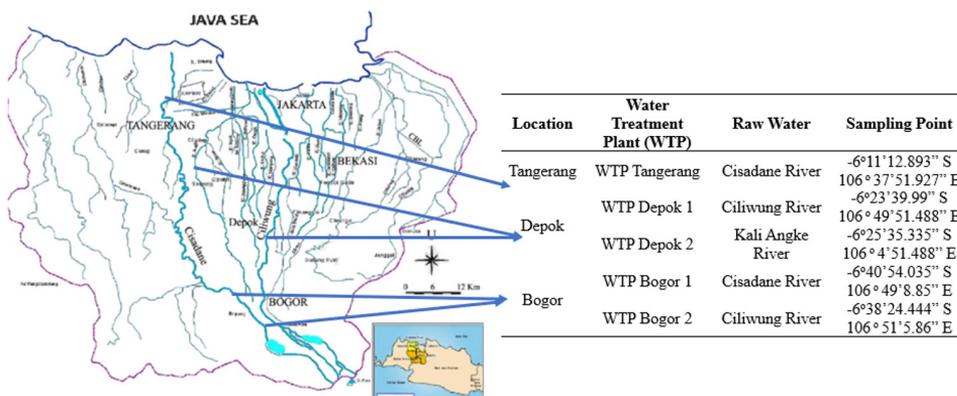


Figure 1. Sampling location of WTP

at 254 nm is measured with a UV-1800 Shimadzu Spectrophotometer ( $UV_{254}$ ) for NOM analysis. Then, calculated SUVA value as an indicator of hydrophobicity and the correlation of DBPs formation (Hua *et al.*, 2015). The value of the SUVA was calculated by comparing the value of  $UV_{254}$  to DOC as Equation 1.

$$SUVA \text{ (L/mg.m)} = \frac{UV_{254}}{DOC} \quad (\text{Eq.1})$$

After measuring the presence of organic matter, the capability of each treatment unit of the WTP is determined by calculating the NOM removal efficiency according to Equation 2.

$$\% C_{removal} = \frac{C_{initial} - C_{final}}{C_{initial}} \times 100\% \quad (\text{Eq.2})$$

### 2.3 Analysis of Trihalomethanes

Four forms of Trihalomethanes (THMs) were investigated including trichloromethane, bromodichloromethane, chlorodibromomethane, and tribromomethane. THMs identification was carried out using USEPA Method 555.1. A total of 50 mL of sample was extracted with 3 mL of methyl-tert-butyl-ether (MTBE) and 10 g of NaCl then shaking by hands for 4 minutes. Then let it sit for about 2 minutes until the water and MTBE solution separate. Take 2 mL of the MTBE solution to be transferred to the autosampler and analyzed using a Gas Chromatograph (GC) Agilent Technologies 7890B equipped with a micro-electron capture detector (1-ECD).

## 3. Results and Discussions

### 3.1 Analysis of Natural Organic Matter as Precursor of DBPs

**Table 1.** Quality of Raw Water

Raw Water Sample	pH	DO (mg/L)	Turbidity (NTU)	$UV_{254}$ ( $cm^{-1}$ )	DOC (mg/L)	SUVA (L/mg.m)
WTP Bogor 1	6.54	8.03	23.5	0.0800	8.217	Th
WTP Bogor 2	6.87	7.92	28.9	0.0720	4.886	1,474
WTP Depok 1	6.17	5.13	23.3	0.1040	3.319	3,133
WTP Depok 2	5.41	6.29	19.2	0.1120	3.579	3,129
WTP Tangerang	7.73	4.54	14.2	0.0870	5.921	1,469

The quality of raw water is affected by the pollutants entering the river water from the disposal of several industries, and also the poor lifestyle of the community (Qadafi *et al.*, 2023). Pollutants come from different sources such as industrial waste, agricultural runoff, and nutrient abundance which causes algae blooms, domestic wastewater, rainwater runoff, forest fires, and increased bromine and iodine matter which can increase levels of organic compounds as precursors on the formation of DBPs (Ding *et al.*, 2019). The high concentration of organic compounds also provides a high opportunity for the formation of DBPs in drinking water treatment.

From the analysis, data on water quality and organic matter are obtained as in Table 1. The highest  $UV_{254}$  (0.1120  $cm^{-1}$  and 0.1040  $cm^{-1}$ ) and SUVA (3.129 L/mg.m and 3.133 L/mg.m) values are shown by raw water from WTP in Depok City which comes from the Angke Rivers and Ciliwung Rivers, while the highest DOC concentration (8.217 mg/L) is actually in raw water from WTP in Bogor City which comes from the Cisadane River and then the next highest DOC concentration (5.921 mg/L) from the raw water WTP Tangerang that the raw water source comes from the Cisadane River. The high SUVA indicates that the organic matter is high in the raw water. The potential for pollution of raw river water increases due to domestic and home industry activities along the river. The SUVA value between 2 - 4 L/mg m indicated that organic matter is a mix of aquatic and other NOM, typical of hydrophobic and also hydrophilic and it has a good correlation with THMs formation (Tak *et al.*, 2018). The low SUVA value (less than 2 L/mg m) means that the organic matter in raw water mostly contains non-humic substances and has low hydrophobicity (Wang *et al.*, 2016).

Analysis of the organic compounds NOM in raw drinking water was also carried out by comparing the results of previous studies as shown in Table 2. The SUVA value shows almost the same range in several raw water sources in the range of 2.00 - 4.21 L/mg.m. The SUVA value shows that raw water contains organic compounds in the form of NOM and DOM. Organic compounds in water indicate that water has received various forms of contaminants, both naturally and or as a result of activities around water bodies. The SUVA value of more than 4 means that organic matter is mostly aquatic humic, and hydrophobic and it tends to produce high THMs (Hua *et al.*, 2015).

However, not many people realize that the presence of NOM, is difficult to remove by conventional treatment even though the concentration is relatively small, the effects from the DBPs formed are toxic. Before the formation of DBPs, prevention efforts can be made by removing the precursors in the raw water. In Indonesia, the existence of organic compounds NOM and DOM has not been specifically regulated. Meanwhile, the presence of NOM indicated by the value of DOC, SUVA, E4/E6 ratio, and color content in the water has a correlation with THM formation (Zevi *et al.*, 2022)

### 3.2 Analysis of Capability of WTP Treatment Unit

Conventional technologies in WTP such as coagulation, sedimentation, sand filtration, and ultrafiltration units can actually control the formation of DBPs by removing precursors in the water (Xu *et al.*, 2019). During the stage of treatment of drinking water treatment, NOM can also decrease because of the pollutant elimination mechanism.

Figure 2 shows the removal of NOM, the SUVA removal efficiency obtained is around 4% - 63% from the five WTPs observed. As for the DOC parameter, the removal efficiency is smaller, around 7% - 42%. This shows that the ability of conventional water treatment units is not effective enough in eliminating NOM. Coagulation and sand filtration of WTP are unable to remove organic matter with low molecule weight, it's only effective in removing high molecule weight organic matter such as humic acid (Qadafi *et al.*, 2023).

The capability of conventional treatment is highly dependent on the size of the precursor molecular weight (MW) to be removed. MW with size > 5kDa can be significantly removed in conventional treatment (Zhang *et al.*, 2020). The size of MW can also change due to adsorption and flocculation processes in water treatment so its removal becomes more difficult for those with low MW (Han *et al.*, 2015). Each stage of WTP units contributes to the pollutants reduction of water including DBPs precursor. Based on the literature in, DOC can be reduced by WTP (consisting of conventional treatment and biological ammonia oxidation unit) up to 70% DOC removal (MacKeown *et al.*, 2020). From the 5 WTPs, the largest DOC removal was only 42% after passing through the coagulation-sedimentation-filtration processing unit.

In the literature, precursors that are dominant in the reactive hydrophobic fraction will be difficult to remove by coagulation and filtration (MacKeown *et al.*, 2020). In coagulation of drinking water treatment, the main objective is to remove turbidity. The phenomenon of floc destabilization in the coagulation process and agglomeration process involves a role in reducing NOM in them, although it is

**Table 2.** Analysis of Organic Matter in Several Sources of Raw Water

Parameters	WTP in Indonesia (This Study)	WTP in India*	WTP in The United States**	WTP in China***
UV <sub>254</sub>	0.09 ± 0.02	0.20 ± 0.12	0.18 ± 0.21	0.06 ± 0.03
DOC	5.18 ± 1.99	4.01 ± 0.51	5.08 ± 4.11	3.14 ± 1.47
SUVA	2.04 ± 1.02	4.21 ± 2.08	2.79 ± 1.27	2.04 ± 0.59

\*Kumari *et al.*, 2022 and Tak *et al.*, 2020; \*\* Uzun *et al.*, 2017 and Gustafson, 2012; \*\*\* Wang *et al.*, 2016 and Liu *et al.*, 2011

not yet clear which is dominantly reduced, whether the hydrophobic or hydrophilic fraction (Tak *et al.*, 2018). However, during the process, NOM is also eliminated in the coagulation process. Coagulation can remove hydrophobic organic matter such as

humic acid and also able to lower the UV absorbance of organic matter). Coagulation is not effective in removing the hydrophilic fraction, and some adsorption processes can remove them (Notodarmojo *et al.*, 2023).

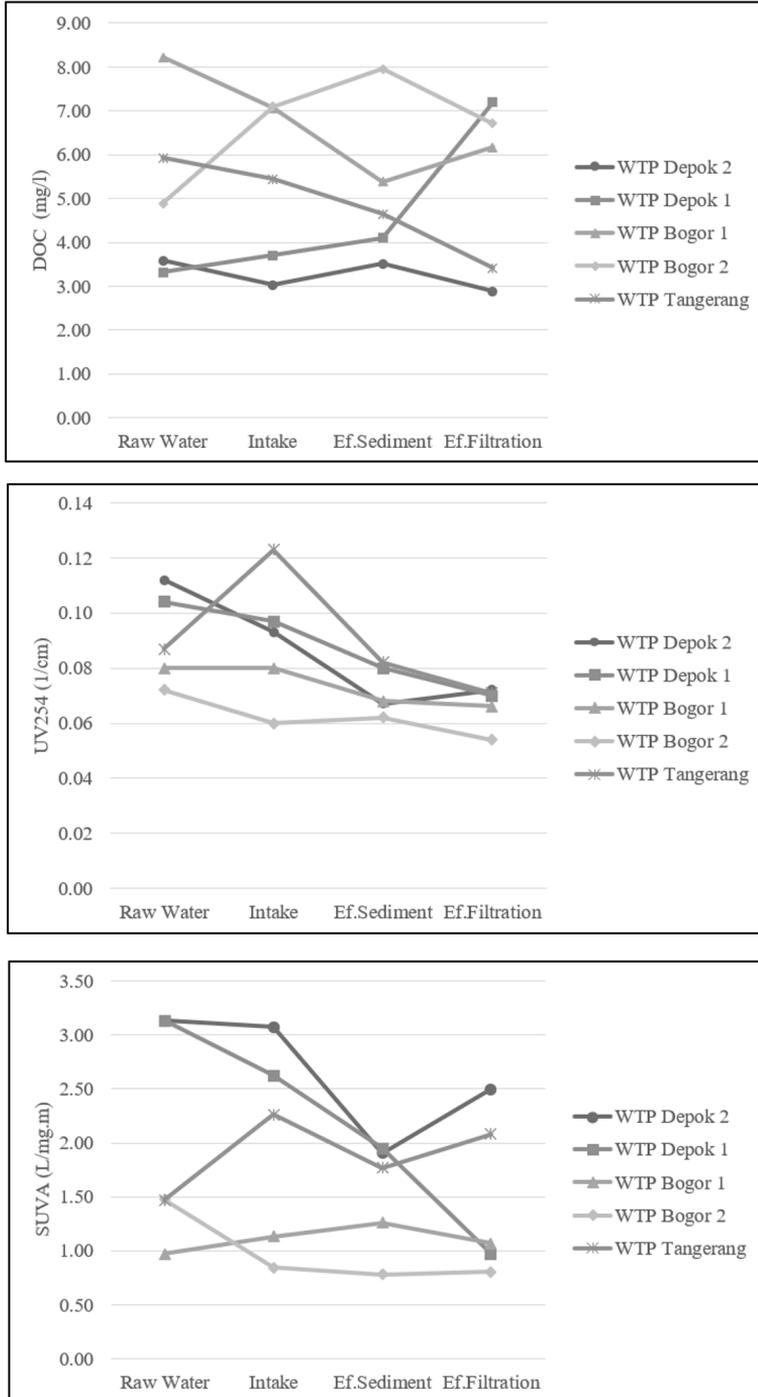


Figure 2. Organic Matter Removal of WTP Treatment Units

There is no specific treatment for removing DOC or NOM, several technologies in water treatment also play a role in reducing levels of organic compounds even though they do not reach optimal conditions. Conventional water treatment such as coagulation, sedimentation, filtration, and even adsorption is not efficient enough to remove NOM (Tak *et al.*, 2018).

### 3.3 Analysis of Trihalomethanes Formation

Trihalomethanes (THMs) formation analysis was carried out as a reference for the potential formation of DBPs compounds in water treatment. THMs are the most common type of DBPs found in water treatment and NOM is the main precursor for forming THMs. The water sample is the intake from the Angke River at the WTP Depok 2. THM has 4 groups of compounds that most often appear in the formation of DBPs when the organic matter reacts with chlorine: Chloroform, Bromodichloromethane, Chlorodibromomethane, and Bromoform, these four compounds are often referred to as THM4. THMs concentration is not only affected by the presence of organic precursors but pH, chlorine dose, reaction time, and halogen concentration can also increase THMs formation (Badaro *et al.*, 2021; Wagner and Plewa, 2017). The formation of THM4 in water after the disinfection are shown in Table 3.

Table 3 shows that the most dominant of THMs are Chlorodibromomethane and Bromoform. The high levels of halide compounds (bromine and iodine) indicate the presence of organic contamination from wastewater that enters the raw water (Richardson and Postigo, 2015). The high levels of bromide in THMs indicate that

a decrease in DOC results in less chlorine acting so chlorine will tend to react with bromide to form reactive bromine species (MacKeown *et al.*, 2020). Ion bromide in water reacted with chlorine and formed hypobromous acid (HOBr) then reacted with organic matter to form the brominated THM4. A high concentration of bromide ions can be an indication that water has been polluted by industrial wastewater (Qadafi *et al.*, 2023).

## 4. Conclusion

The raw water in drinking water treatment plants contains NOM as organic matter. The 5 WTPs show that NOM in the DOC concentration range is 3.30–8.20 mg/L; UV<sub>254</sub> is 0.70 – 0.10 cm<sup>-1</sup>; SUVA is 0.90 – 3.13 L/mg.m. Although the treatment units in the WTP such as coagulation, sedimentation, and filtration can reduce NOM, the efficiency is only about 63% of the reduction in SUVA values and 42% of DOC removal. At WTP Depok 2, the concentration of THM4 is between 4.57–127.02 µg/L, and chlorodibromomethane is the largest concentration of THMs followed by bromoform. Removing precursor levels in raw water can reduce the potential formation of DBP, and advanced treatment may be needed to remove DBPs in water treatment.

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**Table 3.** Concentration of THM4

Trihalomethanes (THMs)	Concentration (µg/L)
Chloroform	4.57
Bromodichloromethane	8.91
Chlorodibromomethane	127.02
Bromoform	59.93

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