Distribution of Natural Organic Matter (NOM) and Trihalomethane Compounds (THMs) in Drinking Water Supplies of India - A Case Study



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ENVIS Monograph No. 20, 2021

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IIT(ISM) ENVIS Centre on Mining Environment Dept. of Environmental Science & Engineering Indian Institute of Technology (ISM) Dhanbad – 826 004 (INDIA) Published by Prof. Anshumali ENVIS Co-ordinator IIT (ISM) ENVIS Centre Dept. of Environmental Science & Engineering Indian Institute of Technology (Indian School of Mines) Dhanbad – 826 004 (INDIA)

Sponsored by Ministry of Environment, Forest & Climate Change Government of India, New Delhi – 110 001

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Printed in India

This monograph is dedicated to my Mother Smt. Tara Devi and Father Sri Gora Chandra Mahato For their endless love, support and encouragement

Preface

The lack of safe drinking water is an alarming concern in developing nations, where nearly half of the population suffers from the health issue associated with it. More than 1600 deaths across the world cause daily due to water-borne diseases. India is one of the fastest developing countries, bare only 4% of the world's water resource, supporting around 17% of the total human population on the earth. At the same time, about 21% of infectious diseases in India arise due to unsafe drinking water. India also ranked in the 2nd last position in the safe drinking water index out of 123 countries worldwide. During the year 2010-13, 12,901 deaths were reported due to the unsafe drinking water, with the maximum number in Uttar Pradesh (3382) followed by West Bengal (1778), Andhra Pradesh (1359), and Odisha (730). The sources of drinking water in India either form surface and groundwater reservoirs, making it even more challenging to provide potable water to the public due to microbiological contamination and Natural Organic Matter (NOM). Disinfection using chlorine is a predominantly used practice worldwide for many decades to protect the consumer against pathogens. However, the issue associated with the chlorination process cannot be ignored, as it reacts with the Natural Organic Matter (NOM) and results in the formation of disinfection by-products (DPBs). especially the cancer-causing Trihalomethanes (THMs) compounds.

This monograph enlightens the occurrence of NOM and THMs compounds in the drinking water supplies of India. It is also based on recent reviews, ideas, effects, and possible control technology adopted to minimize the formation of THMs in water with special reference to the Indian scenario. The book has structured into three different chapters with continuity to get an in-depth understating of the magnitude and gravity of the problems. For controlling the THMs, removal of NOM before the chlorination process is considered the best strategy. The monograph proved to be very helpful for the water management authority and public health departments to regulate THMs in drinking water and ensure communal health safety.

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List of abbreviations used

ADWG	Australia Drinking Water Guideline
AOC	Amounts of Assimilable Organic Carbon
AOP	Advanced Oxidation Process
BDCM	Bromodichloromethane
BDL	Below Detectable Limit
BDOC	Biodegradable Dissolved Organic Carbon
BF	Bromoform
BIS	Bureau of Indian Standard
BOM	Biodegradable Organic matter
BWTP	Water Treatment Plant, Palasuni, Bhubaneshwar, Orissa
CDBPs	Chlorination Disinfection by-products
CDCP	Centers for Disease Control and Prevention
CF	Chloroform
DBCM	Dibromochloromethane
DBPs	Disinfection by-products
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
DWTP	Water Treatment Plant, Belatand, Dhanbad, Jharkhand
EU	European Union
FA	Fulvic Acid
GAC	Granular Activated Carbon
HA	Humic Acid
HAAs	Haloacetic Acids
IGWTP	Indira Gandhi Water Treatment Plant, Kolkata, West Bengal
MCL	Maximum Contaminant Level
MGD	Million Gallons Per Day
MIEX	Magnetic Ion Exchange
MLD	Million Litter Per Day

NOM	Natural Organic Matter
POC	Particulate Organic Carbon
RC	Residual Chlorine
RW	Raw Water
RWTP	Water Treatment Plant, Ravanbhata, Raipur, Chhattisgarh
SUVA	Specific Ultraviolet Absorbance
TCU	Total Color Units
TDS	Total Dissolved Solids
THM	Trihalomethane
THMFP	Trihalomethane Formation Potential
TKN	Total Kjeldahl Nitrogen
TOC	Total Organic Carbon
TTHM	Total Trihalomethane
TW	Treated Water
USEPA	United States Environmental Protection Agency
UV	Ultraviolet
VWTP	Water Treatment Plant, Bhelupur, Varanasi, Uttar Pradesh
WHO	World Health Organisation
WTPs	Water Treatment Plants

Acknowledgements

The inspiration for preparing this monograph originated from my Ph.D supervisor Dr. Sunil Kumar Gupta, Professor, Department of Environmental Science and Engineering (ESE), Indian Institute of Technology (ISM), Dhanbad. I express my profound gratitude to Dr. Sunil Kumar Gupta, author of this monograph, for his continuous insistence to complete the monograph in time. His valuable guidance and constant encouragement had been a great source of inspiration throughout the preparation of this monograph.

I am also grateful to Dr. Anshumali, ENVIS Coordinator and Prof & Head, ESE and Dr. Sheeja Jagadevan, Co-coordinator of IIT(ISM) ENVIS Centre on Mining Environment, Department of Environmental Science and Engineering, Indian Institute of Technology (ISM), Dhanbad, for his valuable suggestion and constant encouragement.

I am thankful to all the faculty members of Department of Environmental Science and Engineering, Indian Institute of Technology (ISM), Dhanbad, for their kind guidance.

I express my deep sense of appreciation to my colleagues Dr. Ashvani Kumar, Programme Officer, Shri Bishwajit Das, Information Officer, and Shri Durga Das Chandra, IT Officer at IIT(ISM) ENVIS centre for their ready and willing co-operation in editing the monograph.

I would like to thanks Ministry of Environment, Forest and Climate Change (MoEF&CC), Government of India, New Delhi, for providing financial support.

I am deeply indebted to my parents and sisters for their endless love, support, and encouragement. Last but not the list I thanks my best friend, Bhavna Singh for always being there to complete this work successfully.

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Dhanbad: July 31, 2021

1. Natural Organic Matter and Trihalomethane: An Introduction

'Natural abilities are like natural plants that need pruning by study'.

- Francis Bacon

1.0 General

Population growth, urbanization, and industrialization exert diverse pressures on the quality and quantity of water resources and the access to safe drinking water. Still, potable water remains inaccessible to about 1100 million people globally, and more than 90 % of drinking water demands are compensated by the groundwater. The rate of groundwater withdrawal is different in all countries, where India raked top (251 km³ per vear) followed by China (112 km³ per vear), USA (112 km³ per year), Pakistan (64 km³ per year), Iran (60 km³ per year), Bangladesh (35 km³ per year), Mexico (29 km³ per year), Saudi Arabia (23 km³per year), Indonesia (14 km³ per year) and Italy (14 km³ per year) (Alsalme et al., 2021). India is the country of rivers; hence the share of surface water for drinking purposes is remarkable. Disinfection of drinking water before its distribution into water supply systems is necessary and used since the early 1900s. Besides chlorine, the first and the most widespread disinfection reagent, there are other disinfectants, such as chloramines, chlorine dioxide, ozone, and UV radiation. One of the major problems of using surface water sources in India is the content of NOM. It is responsible for giving the water a distinct vellow-brown color and result in the formation of THMs compounds during the chlorination process (Ibrahim & Aziz, 2014). All the major drinking water treatment plants (WTPs) use raw surface water as raw water in their supply systems. Hence, it is desirable to minimize the NOM concentration during treatment practices.

1.1 NOM in water

It is a non-homogenous mixture of complex organic compounds primarily comprised of humic acid (HA) and fulvic acid (FA) (Samios et al., 2017; Pasandideh et al., 2016). NOM finds its way into the water bodies by decomposing organic matter and algal metabolic activity (Mahato and Gupta, 2020; Mouelhi et al., 2016). The interaction between the hydrologic cycle and biosphere also results in the formation of NOM in surface water reservoirs (Bhatnagar and Sillanpaa, 2017). The humic substance is commonly present in the soils, sewage, surface water, compost heaps, marine and lake sediments. These are the high molecular weight compound contributing color (brown to black) to the water by secondary synthesis reactions. The HA, FA, and humin are the major components of humic substance which solubility depends upon the pH of the solution (Pettit, 2006).

HA is the mixture of weak aliphatic and aromatic carbon chains organic acids, which are only soluble in alkaline pH. It is the major component of humic substance, with molecular weight ranging from 10,000 to 100 000. Approximately

35% molecules of HA molecules are aromatic, while the remaining are aliphatic (Pettit, 2006; Booi, 2013).

FA also comprises weak aliphatic and aromatic carbon chains organic acids but soluble in all the pH conditions. The oxygen content of FA is twice that of HA and has many carboxyl (-COOH) and hydroxyl (-OH) groups. It is reported to be much more chemically reactive than HA. The molecular weight of FA range from approximately 1000 to 10000 (Pettit, 2006; Booi, 2013).

Moreover, the fraction of humic substances that are not soluble to any pH range is called humin. The molecular weight of humin ranges from approximately 100000 to 10000000 and is considered a micro-organic (very large) substance (Pettit, 2006; Booi, 2013). The schematic of humic and fulvic acid model structure is illustrated in Fig 1-2, respectively (Zularisam et al., 2006).

NOM can be present in the water as particulate and dissolved (filtration through 0.45μ m filter) forms (Mahato and Gupta, 2020). The characteristical properties of NOM may depend upon its source of origin and biodegradability of dissolved organic carbon (Ibrahim and Aziz, 2014). Moreover, seasonal change and other climatic factors like rainfall, snowmelt, and runoff also affect the concentration level of organic compounds in NOM (Thacker et al. 1996). HA is the major constituent of NOM, includes carboxylic, carbonyl, methoxyl, hydroxyl, and phenolic functional groups. It is a compound with wide ranges of molecular weight and sizes (Mouelhi et al., 2016).



Fig 1. The schematic structure of HA (Zularisam et al., 2006).



Fig 2. The schematic structure of FA (Zularisam et al., 2006).

1.1.1 Origin, composition, and characteristics

Aquatic NOM consists of dissolved and suspended organic matter and can be classified as autochthonous and allochthonous NOM (Ibrahim & Aziz, 2014). The autochthonous NOM originated from an internal source like decayed algae, bacteria, and macrophytes living in the water (Nikolaou & Lekkas, 2001). Meanwhile, NOM enters the streams from external sources such as the natural cycle (e.g., soil leaching and snow melting), and human activities (e.g., Effluent from wastewater treatment plant) are referred to as allochthonous (Hwang et al., 2001).

A various concentration range of NOM, from high-MW hydrophobic to low-MW hydrophilic compounds, are exhibited in the natural water bodies (Rodriguez & Nunez, 2011; Edzwald, 1993). The hydrophobic portion of NOM, making up 50% of the total organic carbon (TOC) in water, incredibly consists of HA, FA, and humin (insoluble in water at any pH) (Sillanpää and Matilainen, 2014). The hydrophobic fraction of NOM is enriched with aromatic carbon, phenolic structure, and a conjugated double bond (Samios et al., 2017; Pasandideh et al., 2016). In contrast, the hydrophilic portion contains more aliphatic carbon and nitrogenous compounds, like carbohydrates, amino acids, and sugar (Winterdahl, 2013). The chemical group fraction of NOM is shown in Table 1 and Fig 3. Besides, the percentage sharing of various NOM fractions was illustrated in Fig. 4.

Fraction	Chemical Groups
Hydrophobic	
Acid	
Strong	Humic and fulvic acids, high molecular weight (MW) alkyl monocarboxylic and dicarboxylic acids, aromatic acids
Weak	Phenols, tannins, intermediate MW alkyl monocarboxylic, and dicarboxylic acids
Bases	Proteins, aromatic amines, high MW alkyl amines
Neutrals	Hydrocarbons, aldehydes, high MW methyl ketones, and alkyl alcohols, ethers, furans, pyrrole
Hydrophilic	
Acid	Hydroxy acids, sugars, sulfonic, low MW alkyl monocarboxylic, and dicarboxylic acids
Bases	Amino acids, purines, pyrimidines, low MW alkyl amines
Neutrals	Polysaccharides; low MW alkyl alcohols, aldehydes, and ketones

Table 1. Chemical Group and fractionation of NOM (Edzwald, 1993)

The NOM in the water can be quantified by the parameters TOC, Dissolve organic carbon (DOC), and UV absorbance at 254 (UV₂₅₄) (Bhatnagar and Sillanpaa, 2017). Besides these, the fourth parameter of interest for NOM is the specific UV absorbance (SUVA) index. It characterized the hydrophobic and hydrophilic nature of NOM and can be expressed as the ratio between UV₂₅₄ absorbance (cm⁻¹) and the DOC (mg/l) (Rodriguez & Nunez, 2011). SUVA value interprets and measures the extent distribution of humic substances as HA and FA (Table.2) (Edzwald & Tobiason, 1999).

$$S U V A = \frac{U V_{254}}{D O C} X 1 0 0 L / m g^{-m}$$

.

Table 2. Interpretation of NOM according	g to SUVA	(Edzwald &	Tobiason, 1999)

SUVA (L/mg ^{-m})	Composition
< 2	Mostly non-humic, low hydrophobic, and low MW
2-4	Mixture of hydrophobic and hydrophilic NOM
> 4	Mostly humic, high hydrophobic, and high MW



Fig 3. The characterization of NOM (Levchuk et al., 2018)

1.1.2 Effects of NOM

Although there is no direct and immediate health impact of NOM, however, it critically affects the water treatment process and can contribute to indirect health issues. (Park et al., 2019). NOM deteriorates the quality of drinking water in many ways, like acting as a carrier for metals and hydrophobic organic, bringing undesirable changes in color, order, taste, and aesthetic value (Rodriguez & Nunez, 2011). It also exerts a chemical oxidant demand (i.e., chlorine, chlorine dioxide, ozone) that must be overcome before pathogen log inactivation requirements can be met. Besides, the level of NOM affects the performance of various treatment processing units and the biological stability of water (Park et al., 2019; Bhatnagar and Sillanpaa, 2017).As a result, disinfection is typically applied after treatment processes that remove NOM. The higher the concentration of NOM in water increases the coagulant and chlorine demand necessary for water disinfection. In the membranes, filtration of water raises the possibility of fouling.

It contributes to corrosion in the pipeline and is a source of microorganism's nutrient, promoting the growth of bacterial in the water distribution system (Park et al., 2019; Bhatnagar and Sillanpaa, 2017). NOM affects the UV transmittance at a wavelength of 254 nm that inhibits UV light penetration through the water. In general, every 10% decrease in UV transmittance results in a 50% reduction in the UV dose (Cantwell et al., 2008).

Biological stability is the concept of maintaining the microbiological quality of water from production to the point of consumption (Prest et al., 2016). In water, most of the heterotrophic microorganisms draw their energy for growth, multiplication, and biofilm production from the degradation of organic carbon compounds (Prest et al., 2016). The encouragement of bacterial growth and biofilm development in the water distribution system is a serious threat to community healths.

The composition and concentration of NOM are also greatly affected by the various treatment process. For example, when ozone or chlorine reacts with NOM, it produces biodegradable products (Reckhow et al., 2007). The ozone transforms the NOM into biodegradable organic matter (BOM), whereas the chlorine with NOM increases the amounts of assimilable organic carbon (AOC) and biodegradable DOC (BDOC). Ultimately, these can exacerbate the problem of biofilm growth in the water supply system (Reckhow et al., 2007). NOM plays a critical role in the water treatment process for many reasons:

The indirect health effects of NOM includes:

- A deterioration of pathogen log removal capability due to increased coagulant demand;
- A deterioration of pathogen log inactivation capability due to chemical disinfectant demand;
- Interference in ultraviolet (UV) disinfection;
- The development of biofilms in the distribution systems that can harbour pathogens;
- Increase corrosion in the pipelines and introduces metals toxicity;

The effects of NOM in the operation of Water treatments plants include:

- Increased chemical demands (coagulants and disinfectant dose);
- Poor floc formation or settling;
- Shorter filter run times;
- Required more frequent backwashes;
- Reduced hydraulic capacity;
- Membrane fouling, and higher transmembrane pressure and energy consumption;
- Reduced effectiveness of adsorption and ion exchange processes.

Moreover, after many years of research, it is generally accepted that the NOM is the major contributor of potentially hazardous disinfection by-products (DBPs), especially Trihalomethane (THMs) (Pasandideh et al., 2016; Park et al., 2019). Water utilities must understand the source-specific reactivity of NOM when selecting a disinfectant to mitigate the formation of potentially harmful THMs compounds.



Fig 4. Percentage fraction of NOM in surface water (Zularisam et al., 2006)

1.2 THMs in water

THMs are the highly volatile halogenated single-carbon compounds with the general formula CHX₃, where X denotes the halogen (fluorine, chlorine, bromine, or iodine) (Chawla et al., 1983). The formation of THMs in chlorinated drinking water was first acknowledged by Rook (1974) and Bellar et al. (1974). Later in early 1996, the evidence of its occurrence was also investigated in Indian drinking water by many researchers (Thacker et al. 1996; Satyanarayana and Chandrasekhar 1996). These THMs compounds includes chloroform (CHCl₃) (CF). bromodichloromethane (CHCl₂Br) (BDCM), dibromochloromethane (CHClBr₂) (DBCM) and bromoform (CHBr₃) (BF) (Clark et al. 1986), considered liquids at room temperature. These are slightly soluble in water, with solubilities less than 1 mg/ml at 25 °C. The total THMs (TTHMs) refers to the sum of these four substances (Rodriguez et al., 2004). The chemical structure of these compounds is shown in Fig.5.



Fig.5 Chemical structure (a) Chloroform (b) Bromodichloromethane (c) Diromochloromethane (d) Bromoform. (Mazhar et al. 2020).

1.2.1 Mechanism of THMs Formation

Chlorination disinfection by-products (CDBPs) are chemical compounds that form when water containing NOM (the decay products of living things such as leaves, human and animal wastes, etc.) is chlorinated (Kumari et al., 2015, Bellar et al., 1974). Chlorine disinfection of water can lead to the formation of several chlorination by-products, of which THMs are only one subgroup (Singer, 1994). In the literature, only limited information is available to reveal the mystery of THMs formation during the chlorination process. According to the Chlorine residual testing fact sheet of the Centers for Disease Control and Prevention (CDCP, 2012), various transformations occurred when the chlorine was added to the water (Fig. 6). Initially, the addition of chlorine results in the formation of hypochlorous acid (Eq 1). Since the hypochlorous acid is a weak acid that again dissociates partially in water and gives hypochlorite ion (Eq 2).

$$Cl_2+H_2O = HOCl + H^+ + Cl^-$$
(1)
HOCl = H^+ + OCl⁻ (2)

Ultimately, the free chlorine (hypochlorous acid and hypochlorite ion) in water reacts with the NOM precursor such as humic and fulvic acid in raw water and results in the formation of THMs (Mishra and Dixit, 2013). During this formation process, multi-step reactions occur; in the first steps, organochlorine intermediates are produced, and then in the second stage, it is converted into THMs (Kumari and Gupta, 2018). The formation of THMs is generalized by the following equation (3):

$$Precursor + HO_{X} \longrightarrow CHX_{3}$$
(3)

Where X may be chlorine or bromide, and CHX₃ may be regarded as a general formula for THMs.

Eq. (3) clearly illustrated that THMs are the class of chemical compounds mainly derived from methane (CH₄), where three of the four hydrogen atoms have been replaced by halogens. The chloroform was found to be the principal compound in chlorinated drinking water (Milot et al., 2000). However, in the water containing bromides, the concentrations of CHCl₃ decrease with the formation of brominated THMs (Bellar et al., 1974). Bromide, an inorganic ion, does not react with NOM directly. However, inorganic bromide can be oxidized by chlorine or ozone to hypobromous acid or hypobromite, depending on the pH. By analogy with hypochlorous acid and hypochlorite, both hypobromous acid and hypobromite react with NOM to form brominated DBPs.

Bromine is more reactive with NOM than chlorine. In water-containing bromide, brominated DBPs are formed upon chlorination and ozonation. Since bromine uses the chlorine substitution sites, the formation of chlorinated species is reduced. In addition, bromide is rapidly oxidized by the free chlorine into hypobromous acid (HOBr), which react with NOM precursors and results in the formation of brominated and mixed chlorobromo by-products (CHB_{r3}, CHBrCl₂, and CHClBr₂) (Thokchom et al., 2020; Bellar et al., 1974) (Eq 4, 5 and 6). Since bromine (atomic weight 80) is much heavier than chlorine (atomic weight 35.5), the concentration of the correlated bromoform will be twice that of the chloroform. Therefore, under given chlorination conditions, an increase in bromide could significantly increase the concentration of the four THMs regulated by USEPA. An increase in bromide level also increases the formation of brominated HAAs and reduces chlorinated HAAs. This shows a higher affinity of hypobromous acid (HOBr) with organic precursors than hypochlorous acids resulting in a higher yield of brominated THMs than the chlorinated THMs.

The basic concept of the THMs formation pathway is illustrated in Fig. 7. The THMs formation is also greatly influenced by some water quality parameters like temperature, pH, the concentration of NOM, and contact time (Kumari et al., 2015; Singer, 1994). Moreover, It is generally accepted that the reaction between chlorine and humic substances, a major component of NOM, is responsible for producing organochlorine compounds during drinking-water treatment. Humic and fulvic acids show a high reactivity towards chlorine and constitute 50–90% of the total DOC in river and lake waters. Other fractions of the DOC comprise the hydrophilic acids (up to 30%), carbohydrates (10%), simple carboxylic acids (5%), and proteins/amino acids (5%). Hydrophilic acids such as citric acid and amino acids will react with chlorine to produce chloroform and other products and contribute to total organochlorine production.

$$HOCl + Br \longrightarrow HOBr + Cl$$
 (4)

$$HOCl + HOBr \longrightarrow NOM + DBPs$$
 (5)

$$HOCI + Br - + NOM \sim Brominated THMs$$
 (6)



Fig.6 Chlorine addition flowchart (CDCP, 2012)



Fig. 7: Basic concept of THMs formation pathway

1.2.2 Factors influencing the formation of THMs

THMs in drinking water are primarily formed due to the chlorination of organic matter present in raw water supplies. The rate and degree of its formation depend upon many factors and quality parameters, including TOC, DOC, UV₂₅₄, water temperature, residual chlorine, and pH (Li and Mitch, 2018; Padhi et al., 2019). The extent of THMs formation varies with the water quality characteristics and different

treatment processes (Thokchom et al., 2020). The reaction chemistry of chlorine and organic molecules is complex and, although extensively studied, but poorly understood yet. However, the essential factors which dramatically influence the formation of THMs in drinking water are discussed below:

1.2.2.1 Effects of NOM

Natural organic matter exists in surface and groundwater at concentrations between 2-10 mg/l (Bolto et al. 2002), although much higher levels are sometimes found depending mainly on the watershed state. Groundwater generally has a lower concentration of NOM than surface water. TOC, DOC, and UV₂₅₄ are the essential surrogate measures of NOM, act as a key precursor for THMs formation (Sung et al., 2000). The concentration of level of these parameters was found significantly correlated with the formation of THMs in water by many researchers (Padhi et al., 2019, Chang et al, 2001). The THMs formation rate is equal to that consumption of TOC, thus increasing in organic content of water, upswing the formation of THMs (Chang et al., 2001; Hassani et al., 2010; Arora et al., 1997). It was also reported previously that a water sample with high TOC could produce more THMs if enough RC is available (Babcock and Singer, 1979).

DOC constitutes approximately 83-98% of TOC in water and generally more representative of the soluble organic carbon than TOC (Owen, D. M., Amy, G. L., & Chowdhury, 1993). There was a strong and significant correlation between TOC and DOC. Thus, concerning THMs formation, DOC is also equally responsible for its formation as TOC (Westerhoff et al., 2000; Muller, 1998).

UV₂₅₄ is another important key surrogate of NOM after TOC and DOC, provides an insight into the nature of organic content, and liable to form the THMs in the essence of NOM (Edzwald et al., 1985). Because of easy measurement, UV₂₅₄ offers potentially simple and reliable methods to quantify the contribution of organic carbon in water to the formation of DBPs during chlorination. The correlation coefficients of TOC with THMs were slightly higher than the DOC and UV_{254} , indicating TOC as more influential parameters. Moreover, it was also noticed that a slow reaction between chlorine and NOM results formation of THMs under second-order reaction to TOC, especially for the long-term (Draper and Smith, 1981). Thus it is a multistage process that operates through an initial reaction of TOC with residual chlorine followed by many possible pathways to produce THMs. The second step is found to be rate determining through which the reactive chlorinated intermediates are formed in the initial step (Trussell and Umphres, 1978). With respect to NOM, DOC and UV₂₅₄ were found second and third most influential parameters after TOC responsible for THMs formation, respectively (Hua et al., 2015). The formation of THMs can be affected by the concentration and characteristics of the NOM in two ways. First, an increase in NOM concentration raises the level of THMs precursors. Second, an increase in the NOM concentration increases the chlorine demand of the water. A high chlorine dosage will be necessary to maintain proper chlorine residual in the distribution system.

1.2.2.2 Effects of pH and alkalinity

pH and alkalinity are the other essential parameters that significantly affect the THMs formation. pH showed a positive correlation with THMs; in other words, increasing in pH formation of THMs also increases (Roccaro et al., 2014; Hong et al., 2013; Kim et al., 2003). The oxidation process of chlorine is more prevalent in alkaline pH required more chlorine may support the greater THMs formation. In contrast, acidic pH lowered the reactivity of the chlorine pathway and strongly disfavored the THMs formation (Navalon et al., 2008). Besides, during the chlorination process, when chlorine comes in contact with water leads to the formation of hypochlorous acid (HOC1) and a hypochlorite ion (OC1⁻). The formation of these two species is pH-dependent, as in acidic conditions, HOC1 is found to be dominated, whereas in alkaline pH OC1 (Uyak et al., 2005). Many researchers also widely accepted that base-catalyzed reactions play a major role in THM formation (Reckhow et al., 1990; Peters et al., 1980). Zang et al., (2010) and Stevens et al., (1976) observed that when the pH was decreased to 7.0, the THMs concentration decreased by 50%. These results indicated that maintaining a low pH during disinfection could reduce THM problems, and the pH can be raised once free chlorine residual is no longer present. Depending on the source of organics and chlorination conditions, 30% to 50% increase in THMs formation was noted when the pH was increased from 7 to 11 (Oliver and Lawrence, 1979; Iriarte et al., 2003) showed that a slight pH reduction in the water treatment plant influent stream could significantly lead to trihalomethane reduction in the water leaving the plant when the pH varied between 7.43-7.84 units. In this regard, pH, and alkalinity seems to be an important operational parameter in controlling the THMs formation.

1.2.2.3 Effects of water temperature

THMs formation is proportional to the temperature; the higher the temperature greater the formation (Hua and Reckhow, 2008). It was observed that every 10°C increase in the temperature doubles the rate, enhancing the activation energy of the reaction between organic matter and residual disinfectant (Engerholm and Amy, 1983; Chowdhury and Champagne, 2008). Krasner, (1999)also reported that the formation of THMs was higher during summer when there was high temp. Stevens et al. (1976) and Singer (1999) found the same tendency. The lower temperature in the winter suggests less reactivity and a lower production rate of end products. Stevens et al. (1976) performed experiments at three different temperatures (3°C, 25°C and 40°C), constant pH of 7 and chlorine dose 10 mg/L using Ohio River water from the Cincinnati water treatment plant. The formation of THMs was found to be 1.5 - 2 times higher at each stage of temperature change. The increase in THMs formation per 10oC increase in temperature has been estimated to range between 25-50% (Engerholm and Amy, 1983).

1.2.2.4 Effects of residual chlorine (RC) in the water

The high range of RC present in treated water consequently increased chlorinated THMs (Chowdhury and Champagne, 2008). However, the availability of organics beyond the chlorination breakpoint is so less that the THMs were not found to increase significantly after that point (Sung et al., 2000; Chowdhury and

Champagne, 2008). Many research reported that RC has positively correlated with THMs (Chowdhury and Champagne, 2008; Krasner, 1999; Uyak et al., 2005). Hence, the THMs yield attains higher value in water due to the greater availability of RC (El-Dib and Ali, 1995).

1.2.2.5 Effects of Bromide ion

The presence of bromide ions during water disinfection can lead to the formation of brominated by-products (Barrett *et al.*, 2000). When the ratio of chlorine dosage to bromide ion increases, the formation of brominated THMs is favored (Nokes et al., 1999). During chlorination, the bromide ions are oxidized to hypobromous acid (HOBr), which reacts more readily with organic precursors than chlorine, forming brominated THMs (Stevens et al., 1976; Singer and Chang, 1989). The combined action of chlorine and hypobromous acid leads to the formation of mixed chloro-/bromo-THMs and other halogenated by-products (Singer and Chang, 1989). This may be attributed to the fact that most surface waters, other than coastal regions, do not have significant bromide ions (Black et al., 1996). The formation of brominated THMs with bromide ions is very well known (Symons et al., 1993).

1.2.3 Toxicological /health effect of THMs

Out of approx. Seven hundred well-documented disinfection by-products (DBPs), THMs are among the most prevalent compounds that are also considered key indicators of DBP exposure in epidemiological studies (Stalter et al., 2016; Kumari et al., 2015). The occurrence of THMs in chlorinated waters in India has been reported earlier (Satyanarayana et al., 1996; Thacker et al., 1996, 1997). These compounds in drinking water are considered potential to carcinogenic by the United States Environmental Protection Agency (USEPA, 1999). The various epidemiological studies also confirmed that THMs compounds had been associated with multiple adverse health outcomes like bladder cancer, low birth weight, reproductive, stomach, and rectum problems, etc. (Wang et al., 2019; Hrudey et al., 2015). High doses of THMs compounds showed many carcinogenic, mutagenic, and teratogenic effects.

DBPs are increasingly being recognized as an issue of high risk to human health as they give rise to renal, bowel, and other cancers and reproductive disorders (Arora et al. 1997). These THMs not only cause central nervous system depression but may also cause hepatoxicity, nephrotoxicity, teratogenicity, and carcinogenicity. Association between the ingestion of chlorinated drinking water in excess with risk of bladder and rectal cancer followed by mortality have been reported (USEPA, 1999). Since water contact involves daily activities like drinking, cooking, washing, bathing, showering, etc. There are many possible ways and routes of THMs exposure (oral ingestion, dermal absorption, and inhalation exposure) to the human body (Wang et al., 2019).

1.2.3.1 Adsorption and exposure of THMs in the human body

The CF is the most predominant THM species and present in chlorinated drinking water, found in the highest concentration (Stalter et al., 2016). It may be absorbed into the human body through ingestion, inhalation, skin and showed various

toxicological effects. (USEPA, 1986). In India, the largest source of THMs exposure in humans is from the consumption of chlorinated drinking water (Kumari et al., 2015). Besides the consumption, other water uses in various activities like breathing in CF vaporized into the air and passing through the skin during bathing may contribute significantly to the total exposure of CF (Wang et al., 2019). The chlorinates swimming pools will also contribute to the total exposure of CF. The CF in humans and animals may accumulate in various body organs like adipose tissue, brain, liver, kidneys, adrenals, blood cells, and ultimately damaged it (USEPA, 1986). In addition, exposure to CF also caused progressive central nervous system and depression (USEPA, 1986).

The effects of brominated THMs (DBCM, BDCM) also showed several negative health issues like cytotoxic, genotoxic, and mutagenic effects, but the concentration level of these compounds in Indian drinking water was found nominal (Kumari et al., 2015).

1.2.3.2 Acute or Short-Term Effects of THMs

Evidence for the acute effects of CF was noticed primarily during its use as an inhalation anesthetic. The CF as anesthesia was associated with cardiac arrhythmias and abnormalities of the liver and kidneys. The high and secondarily level inhalation exposure of CF are toxic to the liver and kidneys, respectively (Nazir and Khan, 2006). Moreover, the contact with skin may cause only burning sensation, redness, and blistering (Hrudey et al., 2015).

1.2.3.3 Long-Term or Chronic Effects of THMs

The long-term or chronic effects of CF on the human body at higher doses adversary affect the heart, kidneys, liver, and central nervous system (USEPA, 1999). At the same time, studies in animals showed decreased body weights in rats and mice and an increased incidence of respiratory disease at higher doses (Wang et al., 2019; Hrudey et al., 2015). Besides, liver and kidney toxicity was also observed during the chronic exposure of THMs.

1.2.3.4 Carcinogenic or cancer-causing Effects of THMs

The consumption of chloroform predominant drinking water results in small increases in the rectal incidence and colon and bladder cancer, which was observed consistently (Nazir and Khan, 2006). However, the CF was not identified as the sole carcinogenic agent due to the probability of other possible carcinogenic agents in water (USEPA, 1999). The weight of evidence for the genotoxicity of chloroform is considered negative. Based on sufficient animal and inadequate human evidence of carcinogenicity, the United State Environmental Protection Agency (USEPA) classified CF as group B2 or "probable human carcinogen" (USEPA, 1999). The most universally observed toxic effect of chloroform is damage to the centrilobular region of the liver. Moreover, cancer from chloroform exposure can only occur due to cell toxicity if that threshold is exceeded.

Human exposure to BDCM may increase tumors of the kidney, liver, and large intestine and has been classified as probably carcinogenic to humans, with

sufficient evidence in animals and inadequate evidence in humans (IARC 1991). BDCM gave both positive and negative results in a variety of in vitro and in vivo genotoxicity assays. Exposure to BDCM has also been linked to a possible increase in reproductive effects (increased risk for spontaneous abortion or stillbirth).

Moreover, the IARC has classified both BF and DBCM in Group 3 (not classifiable as to its carcinogenicity to humans). However, the presence of BF in drinking water increased tumors of the large intestine.

1.2.3.5 Developmental or Reproductive Effects of THMs

Previous literature on animal studies confirmed that CF could cause miscarriages, birth defects, and delays in fetal development. Results have generally been inconclusive regarding THMs exposure and adverse reproductive or developmental effects in humans. However, recent research findings suggest an increased risk of early-term miscarriage from high levels of THMs in tap water, mainly due to the presence of BDCM. Table 3 illustrated the summary of various health effects of THMs.

Table 3 Various health effects of THMs.

1.	Acute Effect •	Acute effects of THMs are not well documented in the literature but are expected to be similar to chloroform.					
	•	Cardiac arrhythmias and abnormalities of the liver and kidneys.					
	•	Undiluted exposure to chloroform may cause a burning sensation, redness, and blistering.					
2.	Chronic Effect •	Chronic oral exposure of humans to chloroform at high doses results in adverse effects on the					
	•	The central nervous system, liver, kidneys, and heart.					
	•	Liver toxicity and decreasing were also observed during studies in animals.					
3.	Carcinogenic •	Liver tumors					
	Effects	Bladder cancer.					
4.	Developmental/ • Reproductive Effects	Cause birth defects, miscarriages, and delays in fetal development.					

[Source-NH Department of Environmental Services 2006].

1.2.4 Regulatory guideline of THMs

Disinfection of drinking water by using chlorine was first practiced in Chicago (United States) in the year 1908, which spread quickly worldwide (Mazhar et al., 2020). Later, in 1974 the discovery of THMs alarmed the world to concern about its guideline value in order to minimize the possible adverse health effect.

1.2.4.1 Guideline of THMs in India

Indian interest in THMs had increased in early 1996-1997 when Thacker et al. (1996) confirmed THMs in chlorinated drinking water. However, the guideline value of THMs in India was promulgated late in the year 2004 by BIS. In this year, Indian authorities set the individual permissible for all THMs compounds similar to the guideline value of the 3^{rd} edition of WHO (1998-2004). During the second revision of the draft Indian standard drinking water specification (IS 10500) in 2009, BIS established a single guideline value of 100 µg/L for all four THMs compounds (IS 10500 2009). Further in 2012, in the second revision of the final Indian standard drinking water specification, these guideline values were again revised and suggested to remain the same as per IS 10500 (2004) (Table. 4).

Sl	Edition		References				
No		TTHMs	CHCl ₃	CHCl ₂ Br	CHClBr ₂	CHBr ₃	
1	IS 10500, (2004)		200	60	100	100	IS 10500 (2004)
2	IS 10500, (2009)		100	100	100	100	IS 10500 (2009)
3	BIS, 2012		200	60	100	100	BIS 2012

Table 4 THMs guideline history of BIS

1.2.4.2 Guideline of THMs in other Countries

Canada became the first country to set the guideline value of total THMs (TTHMs) (350 μ g/L) in 1978 (Hrudey and Charrois 2012). This guideline was revised in 1996 and reduced the value to 100 μ g/L. Again in the year 2006, the revision of the Guideline for Canadian Drinking-water Quality re-affirmed the value of TTHMs at 100 μ g/L. Also, it proposed an individual guideline value for CHCl₂Br (16 μ g/L) (Hrudey and Charrois, 2012). In the second revision of the guideline for Drinking Water Quality 1985, China proposed its standards for CHCl₃ (60 μ g/L) (Wang et al., 2015). Further, the standards were revised in 2006 and set different guideline values for all THMs compounds (Wang et al., 2015).

For the first time in 1995, the European Union (EU) recommended the permissible limit for two THMs compounds viz. $CHCl_3$ (40 µg/L) and $CHCl_2Br$ (15µg/L) (Premazzi et al. 1997). Later in 1998, the EU established a single guideline value only for TTHMs, which was brought forward to 2007 and in 2014

(EU 2014). The standard for THMs in Australia was first drawn in 1996 under the Australia Drinking Water Guideline (ADWG). A guideline value of 250 μ g/L was proposed for TTHMs (Hrudey and Charrois 2012). The ADWG 1996 was revised in 2004 and then in 2011, but no changes were made in the guideline of THMs (Table.5).

Many countries and regions across the world follow the THMs guideline value of either WHO or USEPA. However, countries like the UK, Taiwan, New Zealand, South Africa, Japan, Italy, and Korea promulgated their THMs standards for drinking water to ensure public health safety (Table.5).

Sl	Country/	Guideline value (µg/L)					References
No	Organization	TTHMs	CHCl ₃	CHCl ₂ Br	CHClBr ₂	CHBr ₃	
1	CANADA (2012)	100	16				Hrudey and Charrois (2012)
2	CHINA (2014)		60	60	100	100	Wang et al., (2015)
3	EU (2014)	100					EU (2014)
4	AUSTRALIA (2013)	250					Hrudey and Charrois, (2012)
5	JAPAN (2009- 10)	100	60	30	100	90	HASANI et al., (2010); Sharma et al. (2009)
6	UK (2010)	100					HASANI et al., (2010)
7	Korea (2010)	100					HASANI et al., (2010)
8	Tiwan (2010)	100					HASANI et al., (2010)
9	New Zealand (2009)		400	60	150	100	Sharma et al., (2009)
10	Italy (2005- 2017)	30					Villanueva et al., (2017)
11	South Africa (2015)		300	60	100	100	SANS, (2015)

Table 5 THMs guideline value in various countries

1.2.4.3 WHO Guideline of THMs

In the first edition of *Guidelines for Drinking-water Quality* by *WHO* in 1984, no guideline values for THMs other than CHCl₃ were proposed (Hrudey and Charrois 2012). Though the chloroform was the most commonly encountered member of the THMs group, a health-based guideline value of 300μ g/L was recommended (WHO 2006). Later, in the second edition published in 1993, WHO lowered the permissible limit of CHCl₃ (200μ g/L) and established separate guidelines for all THMs compounds (Table. 6). The same guideline value of all four THMs compounds was brought forward to the third edition (1998-2004). In the latest edition, released in 2011, WHO recommended following the guideline of the first edition only for CHCl₃, whereas the guideline value remained the same for other compounds as of the third edition. The complied guideline value of all the editions proposed by WHO is illustrated in Table.6.

Sl	Edition	Guideline value (µg/L)					References
No		TTHMs	$CHCl_3$	$CHCl_2Br$	CHClBr ₂	CHBr ₃	-
1	1st edition (1984)		300				WHO (2006)
2	2nd edition (1993)		200	60	100	100	WHO (2006)
3	3rd edition (1998- 2004)		200	60	100	100	WHO (2006)
4	Latest edition (2011)		300	60	100	100	WHO (2011)

Table 6 THMs guideline history of W.H.O

1.2.4.4 USEPA Guideline of THMs

In the five years after THMs discovery (1979), USEPA established the permissible limit for total THMs (TTHMs) ($100\mu g/L$) under the safe drinking water act (Simpson and Hayes 1998). Later, in 1998 under the Stage 1 DBPs Rule, USEPA lowered its permissible limit to 80 $\mu g/L$ (Tak et al. 2020). Although, the implementation of the Stage 2 DBPs rule in March 2006 maintained the same guideline value as of Stage 1. (USEPA 2006). According to 2012, Edition of the Drinking Water Standards and Health Advisories of USEPA, a maximum contaminant level (MCL) of 80 $\mu g/L$ was set for all the individual THMs compounds, and no change was made to its latest edition of 2018 (USEPA 2012; USEPA 2018). Table. 7 depicted the THMs guideline development history of USEPA.

Sl	THMs rule Maximum Contamination Level (MCL) µg/L					References	
No		TTHMs	CHCl ₃	$CHCl_2Br$	CHClBr ₂	CHBr ₃	
1	THMs Rule (1979)	100					Simpson and Hayes (1998)
2	Stage 1DBPs rule (1998)	80					Tak et al. (2020)
3	Stage 2DBPs rule (1998)	80					USEPA (2006)
4	Drinking- Water Standards and Health Advisories (USEPA, 2012-18)		80	80	80	80	USEPA (2012-18)

Table 7 THMs guideline History of USEPA

2. Distribution of NOM and THMs species in drinking water- An experimental approach

All philosophy in two words – sustain and abstain

- Epictetus

2.0 Background

The global drinking water scenario presents a dismal picture, with most developing countries fighting the scarcity and contamination of drinking water. NOM is ubiquitously distributed over the earth's surface (soils, sediments, and natural water resources) (Sillanpaa et al., 2018). The interaction between the hydrologic cycle and the biosphere results in the occurrence of NOM in the drinking water sources (Bhatnagar and Sillanpaa, 2017). Usually, the organic matter in water is an undesired contaminant and being a nutritional supply for microbiological activity. It may also produce various toxic organic substances during the oxidizing steps in the water treatment process. Over the past 20 years, an increase in the level of NOM in surface water reservoirs has occurred. The impact of such a change in the quality of drinking water was almost unknown. The consequences of such change result in the i) change of color, ii) an increase in biodegradable organic matter and bacterial growth in distribution systems, iii) higher demand for a coagulation agent, iv) an increase in disinfectant demand and disinfection by-products formation (Sillanpaa et al., 2018; Bhatnagar and Sillanpaa, 2017). Especially for countries where drinking water is distributed without or with low chlorine residual, changes in the raw water quality could have a relevant impact on the change of the bacterial drinking water quality in the distribution system. Disinfection using chlorine is the most widely used method for this purpose.

To date, more than 600 DBPs were reported in the literature, of which THMs were found to be the most investigated chlorinated DBPs in drinking water (Hrudey and Charrois 2012). The concentration level of these compounds has increased globally in the last 30 years. The high level of THMs in water has raised a lot of attention in the water treatment process due to its potent to carcinogenicity and other negative health impacts (Wang et al., 2019; Hrudey et al., 2015).

2.1 Distribution of NOM in water

NOM is the major contributor to the brownish-yellow color in water. The first observation of changing the surface watercolor (NOM) was reported in Swedish in the late 1980s. The color contribution of the various composition of NOM was illustrated in Table. 8 (Zularisam et al., 2006). Later, in the last few decades, increasing the concentration range of NOM on the surface was found a worldwide phenomenon. The variation in the global climatic factors like temperature,

precipitation, and decline in acid deposition are reasonable explanations for the increasing concentration level of NOM in surface water bodies.

The occurrence of NOM varies significantly from one source to another due to the unique features of each water body. NOM in the raw surface water is the net effect of hydrological cycles and runoff processes, whereas, in the case of groundwater aquifer, it mainly because of the biogeochemical and leaching processes (Eckhardt and Moore, 1990). NOM concentration in groundwater resources is typically lower than surface water bodies because organic matter is subjected to adsorption and microbial degradation processes as transported through the soil (Aitkenhead-Peterson et al., 2003).

Generally, the NOM in the waster can be determined by the parameters TOC, DOC, adsorption of UV light at 254 nm (UV₂₅₄), and specific UV absorbance (SUVA). It contributes brownish-yellow color in water indicate NOM contents (Edzwald & Tobiason, 1999). The determination of all these parameters is fast and does not require sophisticated sample preparation and analytical instruments.

2.1.1 TOC and DOC in water

TOC represents the amount of particulate and DOC when the existing inorganic carbon is removed by acidification. At the same time, the DOC is a fraction of organic carbon in water that filtrates through the 0.45μ m filter paper (Mahato and Gupta, 2020). TOC and DOC are both the most convenient parameters that genuinely represent the amount of NOM in water. The technique for the monitoring of these parameters involves oxidizing agents burning and radiation. A nondispersive infrared radiation detector measures the resulting CO₂ as NOM present in water.

2.1.2 Specific UV absorbance (SUVA) in water

SUVA is the ratio of UV absorbance at 254 and the concentration of DOC. The value of SUVA indicates the hydrophobic and hydrophilic nature of NOM. The water containing SUVA value > 4 $\text{Lmg}^{-1} \text{ m}^{-1}$ signified the humic nature (hydrophobicity) with high molecular weight and value < 2 $\text{Lmg}^{-1} \text{ m}^{-1}$ indicate non-humic nature (hydrophilic) with low molecular weight. In contrast, a range value between 2 to 4 $\text{Lmg}^{-1} \text{ m}^{-1}$ suggests a mixture of hydrophobic and hydrophilic NOM fractions (Owen et al., 1995).

Table 8	The color	r contributio	n of the	various	composition	of NOM

(Zularisam et al., 2006)

	FA		НА		
Light	Yellow	Dark	Grey	Black	
yellow	brown	brown	black		
2.2 Global occurrence of NOM in water

The NOM in various surface and groundwater reservoirs can be variable due to numerous biogeochemical and hydrological processes (Zularisam et al., 2006). Its concentration range in water may also vary as per the geographical location of the region. Since carbon is the principal constituent of NOM, particulate organic carbon (POC), DOC, and TOC had historically been considered quantitative measures of particulate, dissolved, and total organic matter, respectively (Mazhar et al., 2020). DOC is the major contributor of NOM, accounts for > 90% TOC of water (Mahato and Gupta, 2020). It represents colloidal and dissolved fractions of organic molecules and tends to be the most problematic NOM component for the water treatment process. Earlier, the strength of watercolor was also used to measure the humic and fulvic. A diverse concentration range of NOM in the surface water of various countries was noticed globally viz.USA (3.9 to 5.7 mg/L) (Marhaba & Lippincott, 2000), Malaysia (2.4 to 2.6 mg/L) (Sillanpaa et al., 2018), Canada (4.0 to 7.9 mg/L) (Hua & Reckhow, 2007), China (2.0 to 5.6 mg/L) (Chen et al., 2008), Iran (4.72 to 10.37 mg/L) (Bazrafshan et al., 2012), as well as in Nordic countries (Finland, Sweden and Norway) (3.6 to 54 mg/L) (Vogt et al., 2001). The increased level of NOM brings various changes in water quality which create severe challenges to the drinking water industry (Matilainen et al., 2011). The occurrence of NOM found in the various regions of the world describe below:

2.2.1 Occurrence of NOM in Canadian water

Globally, monitoring data of previous literature illustrates that the NOM concentration and color content of surface water bodies tend to be higher than groundwater (Sillanpää, 2014). The distribution of NOM in surface and groundwater resources are the net effects of hydrological and biogeochemical processes, respectively (Bolto et al., 2002). Table.9 depicted the variation in NOM concentration in surface and groundwater resources of Canada during the years 2009 and 2010 (Environment Canada, 2017). The local environmental condition also plays a significant role in establishing the concentration and character of NOM (Sillanpaa et al., 2018).

Jurisdiction	ТОС	(mg/L)	Colour (TCU)			
	Groundwater Surface Water		Groundwater	Surface Water		
Newfoundland	2.0	7.0	14.3	53.5		
Nova Scotia	2.3	5.8	11.7	43.5		
New Brunswick	2.1	4.8	16.0	40.5		
Quebec	3.1	6.2	NA	53.2		
Manitoba	4.0	11.6	14.0	31.5		

 Table 9 Total organic carbon and color data for raw water from select jurisdictions in Canada

TCU - total color units, NA- Data not available

2.2.2 Occurance of NOM in Europe and North American water

During the past 10 - 20 years, a significant increase in the NOM content was observed in the surface water resources of Europe and North America. Color and UV absorption appeared to increase more than TOC, implying an increase in SUVA levels. Typically, the range value of DOC and SUVA varied from 2.4 to 5 mg/L and 3.8 to 4.8 L/mg^{-m}, respectively. The increased NOM level for this region seems related to global warming or changes in precipitation patterns. There is a strong covariance between NOM concentration and precipitation intensity discharge from the forested sites. The increase in the runoff intensities upswing discharge of water from the upper parts of the soil profile rich with soil organic matter. Moreover, changing the temperature further accelerates soil organic matter degradation, thus generating more NOM in water, at least for the short term. (Eikebrokk et., 2004).

2.2.3 Occurance of NOM in Australian water

Surface water in Australia serves as the primary source of drinking water, contains a high level of NOM throughout the year. The concentration range of NOM in the raw (RW) and treated water (TW) of various regions here are shown in Table.10. The RW seems to have the highest level of UV₂₅₄, color, DOC, and SUVA in all these areas. However, NOM in the treated water was significantly declined. The Australian waters were also found with a low humic content; hence the increased amount hydrophilic fraction of NOM was contributed to the water by autochthonous sources (algae and bacteria). Moreover, the vegetation grew in the vicinity catchment area of the surface water reservoir is one possible reason for the high level of NOM. The Australian waters authorities following conventional coagulation processes, which reduced color by 89%, followed by UV₂₅₄ 82%, DOC 59%, and SUVA 57% (Fabris et al., 2008).

			U								
Regions	NOM parameters										
Regions	Colour (TCU)		DOC	DOC (mg/L)		4 ^{cm-1}	SUVA (L/mg ^{-m)}				
	RW	TW	RW	TW	RW	TW	RW	TW			
Myponga	72	9	12.8	5.4	0.46	0.11	3.6	2.0			
Happy Valley	86	9	10.1	4.7	0.37	0.09	3.9	1.9			
Jandakot	93	10	12.2	4.9	0.51	0.09	4.2	1.8			
Moondarra	26	3	12.9	4.9	0.13	0.10	2.5	1.7			

 Table 10 Concentration range of NOM in the surface water of various regions in Australia

2.2.4 Occurrence of NOM in African water

The high variability in the concentration range of NOM in South African surface water sources raises a significant challenge to the water treatment industries. The

concentration level of NOM in the raw water of various water treatment plants (WTPs) in this region is shown in Table 11. It can be observed from this table a high concentration range of DOC (4.64 to 21.44 mg/L) was monitored in African water as compared to the other part of the world reported above. These WTPs showed good potential for DOC removal by 35.2% to 88.1%. Owing to allochthonous (terrestrial organic carbon input) or autochthonous (phytoplankton and macrophyte activity within a water source) origin, natural organic matter (NOM) abundantly occurs in this region (Nkambule et al., 2012).

WTPs	NOM parameters					
	DOC (mg/L)	SUVA (L/mg ^{-m})				
Johannesburg	9.91	7.61				
Plettenberg Bay	21.44	5.98				
Rietvlei	7.94	1.85				
Stilfontein	7.60	1.86				
Wiggins	4.64	0.30				
Umzoniana	8.35	1.11				
Lourie	7.61	2.46				

Table 11 Concentration range of NOM in the raw water of various WTPs in South Africa

2.2.5 Occurrence of NOM in surface water of Southern Quebec

The greatest absolute variability of NOM concentration was reported in the surface water of southern Quebec. Table 12 depicted the DOC concentration of various streams draining eight small catchments in southern Quebec. Such an elevated concentration range of DOC (3.5 to 40 mg/L) was measured in this region, which a grave concern with respect to drinking water quality. DOC was introduced into the catchment by precipitation, decomposition of vegetation, leaching, and soil organic horizons. The mean annual rainfall in southern Quebec is about 1 008 mm, of which 500-600 mm is runs off. During runs off, the DOC-rich water percolating into the subsoil and passes through soil horizons change in NOM concentration. However, much of the DOC produced by the vegetation and soil can reach the stream channel (Eckhardt and Moore, 1990).

Table12 Concentration range of NOM in various catchment area of southern Quebec

of southern Quebec									
Catchment	1	2	3	4	5	6	7	8	
DOC (mg/L)	3.9	3.5	7.2	3.8	14.5	32.2	21.9	40.0	

2.3 Occurance of NOM Indian water

India is a country of the river where the surface reservoir is the major source of raw water for WTPs to produce safe drinking water to the communities. The surface water resources in India contain a versatile range value of NOM (TOC 2.1 – 44.1mg/L) (Mahato and Gupta, 2020; Selvam et al., 2018). However, very few studies have been carried out in Indian to evaluate the distribution of NOM in surface and groundwater resources in the last decade (Table.13). The concentration and variability of NOM in Indian water depend upon many factors.

		NO	DM		
Location	TOC	DOC	UV 254	SUVA	References
	(mg/L)	(mg/L)	(cm^{-1})	$(L.mg^{-1}.m^{-1})$	
Bhelatand, Dhanbad	3.112	3.101	0.077	2.49	Mahato and
Source - Barakar River					Gupta, (2020)
Kolkata, West Bengal	4.43	3.5	0.157	4.4	
Source - Hubli River					
Durgapur, West Bengal	2.1	1.9	0.11	5.47	
Source - Damodar river					
Jamadoba, Dhanbad	4.3	3.6	0.1	3.3	Minashree and S G
Source - Damodar river					(2014)
Maithon, Dhanbad	4.4	4.3	0.2	4.5	
Source - Barakar River					
Swarnrekha, Ranchi	2.42	2.20	0.105	4.77	
Source – Swarnrekha River					
Tiruchirappalli, Tamil Nadu	43.4	NA	NA	NA	
Source – Cauvery River					Selvam et
Srirangam, Tamil Nadu	44.1	NA	NA	NA	al., (2018)
Source – Cauvery River					
Kanpur, Uttar Pradesh	18.8	NA	NA	3.1	Mishra and
Source – Ground Water					Dixit, (2013)
Haridwar, Uttrakhand	15	NA	NA	NA	Dash et al.,
Source – Ganga River					(2010)

Table13 Distribution of NOM in surface and ground of India

NA- Data not available

2.4 International considerations of NOM

Increasing the level of NOM in water resources is an alarming concern across the world. It has a fundamental impact on water quality and drinking water treatment processes aimed to protect the community's health. Hence, in order to minimize the effect of NOM, some jurisdictions have established their regulatory requirements for drinking water quality.

2.4.1 USEPA regulatory

Since the year 1998, USEPA mandates the treatment of NOM in their drinking water supplies system to minimize the DBPs formation. All the water industries were instructed to follow the conventional water treatment process using lime if the TOC level in the source water was found > 2 mg/L. The sets some criteria for selecting the treatment technique based on alkalinity and the concentration level of TOC. The water source containing higher NOM that the coagulation process can not remove is permitted to conduct jar test to determine alternative performance criteria and avoid excessive alum use. In order to maintain the water quality, it was strictly instructed to develop and implement the monthly sampling plans for TOC, DOC, UV₂₅₄, SUVA, and THMs in the raw and treated water. However, the monitoring of TOC was made mandatory quarterly (USEPA, 1998).

2.4.2 WHO regulatory

The World Health Organization suggests optimized NOM removal to depreciate biofilm growth in the water distribution system. They also considered organic carbon as an operational parameter in water safety plans to monitor control measures and prescribed the drinking water standard for it as 5 mg/L (WHO, 2011).

2.4.3 European Union (EU) regulatory

The drinking water regulations of the EU consider TOC as a general water quality parameter for their supplies system. A parametric guideline value of 5 mg/L is specified with "no abnormal change" in it. Similarly, the French regulatory body established the guideline limit of TOC for human consumption as 2 mg/L (EU, 2014).

2.4.4 Australian regulatory

Australian Drinking Water Guidelines Framework also developed some guidelines to help water utilities understand and controlling the effects of NOM. (Cooperative Research Centre for Water Quality and Treatment, 2005).

2.5 An experimental approach to know the distribution of NOM in various major WTPs of India

As reported earlier, very few studies have been conducted on the occurrence status of NOM in Indian drinking water. However, no work performed yet to know the distribution pattern of NOM in their water supply system. Previous literature confirmed that this country's surface and groundwater resources contain a high level of NOM (TOC 2.1 – 44.1mg/L) (Mahato and Gupta, 2020; Selvam et al., 2018). Hence, in the present approach, five major drinking water treatment plants (WTPs) from five different states of India, i.e., Jharkhand, Utter Pradesh, Chattishgarh, West Bengal, and Odisha, have been selected to monitor the distribution patterns of NOM in drinking water supply system. The sample WTPs were selected based on the locations, accessibility, geographical locations, source of intake water & other geo-climatic conditions, keeping in mind to cover at least five nearby states in India. The water samples were collected from four well-defined sampling points of each WTPs as follow:

- I. Raw water from the inlet (RW).
- II. Before chlorination (BC).
- III. After chlorination (AC).
- IV. At the consumer end (CE).

2.5.1 Water Treatment Plant, Bhelupur, Varanasi (UP) (VWTP)

Varanasi is an ancient religious city on the bank of the holy river Ganga, with a total population of 3,676,841 (Census 2011). Surface water and groundwater sources contributing almost equally to the drinking water supply to this city. Varanasi Jal Sansthan looks after the water supply system here. The Bhelupur WTP was established in 1892 with a designed capacity of 311 MLD. River Ganga is the source of raw water for this plant. The conventional treatment process was used to treat the raw water of this WTP. The process flow chart of the treatment system was given in Fig. 8. The physicochemical water quality and distribution pattern of NOM in the various treatment stage, from the raw water to the final treated water, is shown in Table 14.

The Physico-chemical water quality characteristics of VWTP good complied with the guideline value of the Bureau of Indian Standards (BIS) for drinking purposes (IS 10500:2012). However, the turbidity in the raw water was monitored slightly higher (7.26 NTU). The concentration range of all the potential surrogates of NOM decreased as it passed through the treatment system. The value of SUVA indicating that the NOM in the water is hydrophilic in nature with low molecular weight. Poly aluminum chloride (PAC) and alum are the most widely used coagulants in VWTPs to treat raw water. Besides, disinfection of the final treated water was done using chlorine gas. Overall, VWTPs producing good quality of drinking water for the consumers concerning these parameters.

Sl No	Parameters	<u> </u>	Samp	ole Id		Drinking-Water Std. (IS 10500:2012)		
		RW	BC	AC	CE	Acceptable Limit	Permissible Limit	
On-	Site Parameter							
1.	pН	7.62	7.63	7.54	7.69	6.5-8.5		
2.	Temperature (°C)	27.8	27.5	27.8	25.7			
3.	DO (mg/l)	7.2	6.8	7.2	7.4			
4.	Residual Chlorine (mg/l)	Nil	Nil	0.2	Nil	0.2	1	
Off	Site Parameter							
1.	Turbidity (NTU)	7.26	2.72	4.29	3.74	1	5	
2.	TDS (mg/l)	285	270	267	280	500	2000	
3.	Conductivity (µScm ⁻¹)	231.3	233.2	231.9	260.2			
4.	Total Alkalinity (mg CaCO ₃ /L)	180	165	161	185	200	600	
5.	Total Hardness (mg CaCO ₃ /L)	162	166	164	176	200	600	
6.	Ca Hardness (mg CaCO ₃ /L)	104	84	80	106			
7.	TKN (mg/l)	0.28	Nil	Nil	Nil			
8.	Ammonical Nitrogen (mg/l)	Nil	Nil	Nil	Nil			
9.	Nitrate (mg/l)	1.621	1.409	1.099	1.095	45	No relaxation	
10.	Br ⁻ (mg/l)	BDL	BDL	BDL	DBL			
11.	Total Coliform (MPN/100ml)	≥1600	≥1600	<1.1	<1.1	Shall not be any 100 i	detectable in nl sample	
12	Faecal Coliform (MPN/100ml)	≥1600	≥1600	<1.1	<1.1	Shall not be any 100 i	detectable in nl sample	

Table14 Water quality status and distribution of NOM in VWTP

Sl	Parameters		Sam	ole Id		Drinking-Water Std.				
No			-			(IS 1050)0:2012)			
		RW	BC	AC	CE	Accentable	Permissible			
		IC W	DC	ne	CL	Limit	Limit			
						LIIIII	Limit			
NOM										
1	TOC (mg/l)	3 838	3 613	3 083	2 999					
	100(119,1)	2.020	0.010	0.000						
2	DOC (mg/l)	3 715	3 409	3 040	2 636					
2.	DOC (IIIg/I)	5.715	5.407	5.040	2.050					
2	$\mathbf{U}\mathbf{V}$ (and \mathbf{I})	0.0522	0.0424	0.0206	0.0121					
3.	UV_{254} (cm ⁻¹)	0.0525	0.0424	0.0296	0.0131					
		1 400	1	0.074	0.405					
4.	SUVA (L.mg	1.408	1.244	0.974	0.497					
	¹ .m ⁻¹)									



Fig. 8: Treatment process flow chart of Bhelupur WTP, Varanasi

2.5.2 Water Treatment Plant, Belatand, Dhanbad, Jharkhand (DWTP)

Dhanbad is situated in Jharkhand state, better known as the coal capital of India, with a total population of 26 84,487 (Census 2011). The Drinking-Water and Sanitation Department of Dhanbad manages and looks after the drinking water supply in the city. In order to fulfill the need for water demands, Belatand WTPs was established in the year 2007, with a capacity of 77 MLD. The Barakar river, which is a tributary of Damodar, is the raw water source for this plant. The

schematic layout of the treatment processes at DWTP is shown in Fig. 9. The physicochemical water quality and distribution pattern of NOM in the various treatment stage, from the raw water to the final treated water, is shown in Table 15.

As per the guideline value of BIS, the quality of water in DWTP seems suitable for drinking purposes. In fact, the concentration level of dissolved oxygen (DO) at all the treatment process stages is higher, indicating the less load of pollutants. However, residual chlorine (RC) value exceeds the permissible limit of BIS, just after chlorination points. In context to the NOM, the TOC, DOC, and UV_{254} levels were found relatively good, and they also showed a decreasing trend as it passes through the treatment process. Moreover, the SUVA value indicating the hydrophobic nature of NOM for this WTP with high molecular weight. DWTP uses alum and lime for coagulation as well as the chlorine gas for the disinfection process

Sl No	Parameters		Samj	ple Id		Drinking- (IS 1050	Water Std.
110		RW	BC	AC	CE	Acceptable Limit	Permissible Limit
On-S	Site Parameter						
1.	pН	7.30	7.30	7.35	7.45	6.5-8.5	
2.	Temperature (°C)	20.6	19.4	20.7	19.5		
3.	DO (mg/L)	8.5	8.2	8.0	7.3		
4.	Residual Chlorine (mg/L)	Nil	Nil	1.59	Nil	0.2	1
Off	Site Parameter						
1.	Turbidity (NTU)	1.36	1.12	0.81	1.0	1	5
2.	TDS (mg/L)	140	126	149	130	500	2000
3.	Conductivity (µScm ⁻¹)	90.85	87.89	136.9	151.4		
4.	Total Alkalinity (mg CaCO ₃ /L)	82	74	96	76	200	600
5.	Total Hardness (mg CaCO ₃ /L)	84	72	88	76	200	600
6.	Ca Hardness (mg CaCO ₃ /L)	52	44	44	40		
7.	TKN (mg/L)	Nil	Nil	Nil	Nil		

Table15 Water quality status and distribution of NOM in DWTP

Cha	pter	2
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Sl No	Parameters		Samj	ple Id		Drinking-	Water Std. 0.2012
100		RW	BC	AC	CE	Accentable	Permissible
		10.00	Be	ne	CL	Limit	Limit
8.	Ammonical Nitrogen (mg/L)	Nil	Nil	Nil	Nil		
9.	Nitrate (mg/L)	0.220	0.165	0.177	0.233	45	No relaxation
10.	Br ⁻ (mg/L)	BDL	BDL	BDL	BDL		
11.	Total Coliform (MPN/100ml)	$\geq 160 \\ 0$	≥1600	<1.1	1.1	Shall not be detectable in any 100 ml sample	
12.	Faecal Coliform (MPN/100ml)	$\geq 160 \\ 0$	≥1600	<1.1	< 1.1	Shall not be any 100 r	detectable in nl sample
NON	4						
1.	TOC (mg/L)	1.669	1.591	1.633	1.726		
2.	DOC (mg/L)	1.484	1.469	1.513	1.484		
3.	UV 254 (cm ⁻¹)	0.042 6	0.0319	0.0436	0.032		
4.	SUVA (L.mg ⁻¹ .m ⁻¹)	2.870	2.1715	2.8816	2.169		



Fig. 9: Treatment process flow chart of DWTP

2.5.3 Water Treatment Plant, Ravanbhata, Raipur, Chhattisgarh (RWTP)

Raipur is the capital city of Chhattisgarh, having a population of 10 10,433 (Census, 2011). The Municipal Corporation of Raipur is responsible for managing water supply to the town from both surface and groundwater sources. The RWTP is in Ravanbhata, located in the southern part of Raipur. The Kharoon river, a tributary Mahanadi, is the primary source of raw water for this plant.

RWTP consists of three units with different capacities of 150 MLD, 80 MLD, and 47 MLD, which serve approx 4.6 lakhs consumers of the city. The schematic layout of the treatment processes at RWTP is shown in Fig. 10. The quality of water and distribution NOM in this plants is illustrated in Table 16.

All the Physico-chemical quality of water was found suitable for drinking and other domestics activities, except RC, just after the chlorination point (Table 15). Its concentration exceeded the permissible limit of BIS (1 mg/L) due to inappropriate dosing of chlorine during the disinfection process. The distribution of NOM in all the treatment stages bared appropriate for drinking and showed decreasing trend during the treatment. PAC/Alum and chlorine gas are the main chemicals used for producing safe drinking water in this WTP. Moreover, when the chlorine gas not available, a prepared slurry of bleaching powder was used for chlorination.

Sl No	Parameters		Sample Id			Drinking-Water Std. (IS 10500:2012)		
		RW	BC	AC	CE	Acceptable Limit	Permissible Limit	
On	Site Parameter							
1.	рН	7.66	7.71	7.73	7.61	6.5-8.5		
2.	Temperature (°C)	23	22.5	22.8	22.1			
3.	DO (mg/L)	7.5	6.5	7.7	7.1			
4.	Residual Chlorine (mg/L)	Nil	Nil	2.5	Nil	0.2	1	
Off	Site Parameter							
1.	Turbidity (NTU)	3.65	0.59	0.75	1.2	1	5	
2.	TDS (mg/L)	95.67	93.17	92.73	85.82	500	2000	

Table16 Water quality status and distribution of NOM in RWTP

Sl No	Parameters		Sampl	e Id		Drinking-Water Std. (IS 10500:2012)	
		RW	BC	AC	CE	Acceptable Limit	Permissible Limit
3.	Conductivity (µScm ⁻¹)	155.31	186.2	185.4	172.8		
4.	Alkalinity (mg CaCO ₃ /L)	78	78	76	74	200	600
5.	Total Hardness (mg CaCO ₃ /L)	60	65	79	74	200	600
6.	Ca Hardness (mg CaCO ₃ /L)	47.25	55.65	68.25	55.65		
7.	TKN (mg/L)	Nil	Nil	Nil	Nil		
8.	Ammonical Nitrogen (mg/L)	Nil	Nil	Nil	Nil		
9.	Nitrate (mg/L)	1.160	1.179	1.091	0.982	45	No relaxation
10	Br ⁻ (mg/L)	BDL	BDL	BDL	BDL		
11	Total Coliform (MPN/100ml)	≥1600	≥1600	<1.1	1.1	Shall not be any 100 r	detectable in nl sample
12	Faecal Coliform (MPN/100ml)	≥1600	≥1600	<1.1	< 1.1	Shall not be any 100 i	detectable in nl sample
NO	М						
1.	TOC (mg/L)	2.491	1.990	2.019	2.146		
2.	DOC (mg/L)	2.008	1.812	1.962	1.971		
3.	UV 254 (cm ⁻¹)	0.0177	0.0118	0.007 8	0.003 8		
4.	SUVA (L.mg ⁻ ¹ . m ⁻¹)	0.881	0.651	0.398	0.193		



Fig. 10: Treatment process flow chart of RWTP

2.5.4 Water Treatment Plant, Palasuni, Bhubaneshwar, Orissa (BWTP)

Bhubaneshwar is the capital city of Orissa, having approx. 7 Lakhs of the population (Census, 2001). In order to supply safe drinking water to the community, BWTP was established in 1983 by the Public Health Engineering Department of Orissa. The Kuakhai river, which is about 10 KM far away from the city, is the raw water source. There are three units in this WTP with the capacities of 13.63 MLD, 27.26 MLD, and 41 MLD. The schematic layout of the treatment processes is shown in Fig.11. The distribution of NOM and physicochemical quality of water is depicted in Table 17.

The Physico-chemical water quality characteristics of BWTP good complied with the guideline value of the Bureau of Indian Standards (BIS) for drinking purposes (IS 10500:2012).

However, the RC just after the chlorination point was monitored higher (2.57). The concentration range of all the potential surrogates of NOM, i.e., TOC, DOC, and UV_{254} decreased as it passed through the treatment system. The concentration level of these NOM parameters was found lower than all above WTPs. The value of SUVA indicating that the NOM in the water is hydrophilic in nature with low molecular weight. Treatment of water is done conventionally by rapid sand filters using lime alum and PAC as well as the chlorine gas.

Sl No	Parameters		Sam	ple Id		Drinking- (IS 105	Water Std. 00:2012)
		RW	BC	AC	CE	Acceptable Limit	Permissible Limit
On-	Site Parameter						
1	nH	7 37	7 56	7 43	631	6 5-8 5	
2.	Temperature (°C)	20.6	20.6	20.6	20.8		
3.	DO (mg/L)	7.8	8.1	8.2	7.8		
4.	Residual Chlorine (mg/L)	Nil	Nil	2.57	Nil	0.2	1
Off	Site Parameter						
1.	Turbidity (NTU)	0.94	0.37	0.27	0.91	1	5
2.	TDS (mg/L)	121	130.4	101.6	151.6	500	2000
3.	Conductivity (µScm ⁻¹)	204.6	184.1	194.2	230.1		
4.	Alkalinity (mg CaCO ₃ /L)	78	74.5	70.5	78.66	200	600
5.	Total Hardness (mg CaCO ₃ /L)	49.5	47.5	45.5	52	200	600
6.	Ca Hardness (mg CaCO ₃ /L)	24	26	30	38		
7.	TKN (mg/L)	Nil	Nil	Nil	Nil		
8.	Ammonical Nitrogen (mg/L)	Nil	Nil	Nil	Nil		
9.	Nitrate (mg/L)	0.235	0.243	0.202	0.240	45	No relaxation
10.	Br ⁻ (mg/L)	BDL	BDL	BDL	BDL		

Table17 Water quality status and distribution of NOM in BWTP

Sl No	Parameters	Sample Id				Drinking-Water Std. (IS 10500:2012)		
		RW	BC	AC	CE	Acceptable Limit	Permissible Limit	
11.	Total Coliform (MPN/100ml)	≥1600	≥1600	<1.1	1.1	Shall not be any 100	detectable in ml sample	
12.	Faecal Coliform (MPN/100ml)	≥1600	≥1600	<1.1	< 1.1	Shall not be any 100	detectable in ml sample	
NO	М							
1.	TOC (mg/L)	1.902	1.613	1.464	1.303			
2.	DOC (mg/L)	1.663	1.573	1.420	1.121			
3.	UV 254 (cm ⁻¹)	0.0332	0.028 7	0.027	0.0201			
4.	SUVA (L.mg ⁻¹ . m ⁻¹)	1.996	1.825	1.944	1.793			



Fig. 11: Treatment process flow chart of BWTP

2.5.5 Indira Gandhi Water Treatment Plant, Barrackpore, Kolkata, West Bengal (IGWTP)

Palta Waterworks, which is now rechristened as the Indira Gandhi Water Treatment plant, is located at Barrackpore, Kolkata. It is unique of its kind, one the largest in the Asian Sub-Continent, spreading over a sprawling stretch of 480 acres, was constructed during 1864-70 with an initial capacity of 6 MGD. In the year 2006, the capacity of this WTP was upgraded to 260 MGD. Hooghly River is the primary source of raw water for this plant which served more than 70% population of Kolkata. The schematic layout of the treatment processes at IGWTP is shown in Fig. 12. The quality of raw and treated water and the distribution of NOM in this plants are illustrated in Table 18.

The quality of water at IGWTP seems slightly alkaline, and the temperature was also observed higher than other WTP discussed above. The turbidity in raw water at the inlet point exceeded the permissible limit of BIS; however, after treatment, its level has gone down. The distribution of NOM at various stages of the treatment process followed a similar trend as other WTPs. The characteristic of NOM was observed hydrophilic with low molecular weight as the value of SUVA < 2 L.mg⁻¹. The plant consists of different types of filters bed (Slow sand filter, rapid gravity sand filter). The coagulation and disinfection process was done by using Alum and chlorine gas, respectively.

Sl No	Parameters		Sample Id			Drinking-Water Std. (IS 10500:2012)		
		RW	BC	AC	CE	Acceptable Limit	Permissible Limit	
	On-Site Parameter							
1.	pH	8.6	8.4	8.2	8.4	6.5-8.5		
2.	Temperature (°C)	29.7	30	30.1	30			
3.	DO (mg/L)	6	5.2	4.7	4.2			
4.	Residual Chlorine (mg/L)	Nil	Nil	0.8	0.4	0.2	1	
Off-	Site Parameter							
1.	Turbidity (NTU)	5.6	0.8	0.8	0.7	1	5	
2.	TDS (mg/L)	216	255	220	174	500	2000	
3.	Conductivity (µScm ⁻¹)	344	328	314	330			
4.	Alkalinity (mg CaCO ₃ /L)	165	158	141	150	200	600	
5.	Total Hardness (mg	155	147	130	131	200	600	

Table18 Water quality status and distribution of NOM in IGWTP

Sl No	Parameters		Samp	le Id		Drinking-Water Std. (IS 10500:2012)	
		RW	BC	AC	CE	Acceptable Limit	Permissible Limit
	CaCO ₃ /L)	-					
6.	Ca Hardness (mg CaCO ₃ /L)	92	77	74	80		
7.	TKN (mg/L)	0.33	0.336	0.224	0.112		
8.	Ammonical Nitrogen (mg/L)	Nil	Nil	Nil	Nil		
9.	Nitrate (mg/L)	0.016	0.009	0.08	0.059	45	No relaxation
10.	Br ⁻ (mg/L)	BDL	BDL	BDL	BDL		
11.	Total Coliform MPN/100ml)	≥1600	≥1600	<1.1	< 1.1	Shall not be any 100 i	detectable in ml sample
12.	Faecal Coliform (MPN/100ml)	≥1600	≥1600	<1.1	< 1.1	Shall not be any 1	detectable in 00 ml
N	OM						
1.	TOC (mg/L)	2.712	2.411	2.294	2.388		
2.	DOC (mg/L)	2.676	2.366	2.233	2.37		
3.	UV 254 (cm ⁻¹)	0.038	0.033	0.018	0.018		
4.	SUVA (L.mg ⁻¹ . m ⁻¹)	1.431	1.42	0.824	0.772		



Fig. 12: Treatment process flow chart of IGWTP

2.6 Overall distribution of NOM species in all selected WTPs

The occurrence of NOM in various treatment processes of water utilities greatly influences its efficiency for the production of safe drinking water (Mahato and Gupta, 2020). Fig. 13a–d depicted the distribution of NOM species (TOC, DOC, UV₂₅₄, and SUVA) at the defined sampling point of all five selected WTPs. The concentration of TOC and DOC and were monitored higher in VWTP followed by IGWTP > RWTP > BWTP and > DWTP (Fig. 13a–b). The biodegradability of these two parameters played an essential role in changing the physicochemical properties of water and causes many problems during the coagulation processes (Menya et al., 2018). UV absorbance at 254 nm (UV₂₅₄) is a well-defined surrogate to represent the NOM content in natural water (Chen et al., 2008). The trends of UV₂₅₄ in the study area were found in the order of VWTP> DWTP> IGWTP > BWTP and >RWTP (Fig. 13c). The range value of SUVA (0 to 2.8 L/mg^{-m}) indicating the mixed nature (hydrophobic and hydrophilic) of NOM for all the WTPs (Mahato and Gupta, 2020) (Fig. 13d).

The concentration range of these NOM species was observed to decrease as it passes through the various treatment system slightly. However, no change was noticed for DOC in the case of DWTP (Fig. 13a–d). It was also detected that the water quality characteristics of VWTP and IGWTP were found identical, maybe due to the fact that of having the same sources of raw from the river the Ganga.



Fig. 13(a). Distribution of NOM species TOC



Fig. 13(b). Distribution of NOM species DOC



Fig. 13(c). Distribution of NOM species UV₂₅₄



Fig. 13(d). Distribution of NOM species SUVA

2.7 Efficacy evaluation of existing treatment system for NOM removal

The purpose of this study is to evaluate the efficiency of existing treatment technology of selected WTPs with respect to the percentage removal of turbidity and NOM species. Since all the WTPs having various capacities and specifications, they follow traditional treatment processes comprised of aeration followed by coagulation (alum, PACl, and lime), flocculation, sand filtration, and finally, chlorination (bleaching powder or chlorine gas). The efficacy of WTPs was evaluated by using the following formula:

Efficacy (%) = (Raw water quality – Treated Water quality)/ Treated water quality x 100

Fig. 14 depicted the percentage efficiency of the conventional treatment process for the removal of turbidity and NOM species. Efficacy of existing technology in these WTP seems very good for turbidity (up to 82 %), whereas in terms of NOM it showed marginal efficiency viz. SUVA (up to 48 %) followed by UV_{254} (up to 47 %), DOC (up to 37 %), and TOC (up to 34 %). Literature survey also revealed that the conventional treatment system like coagulation, sedimentation, and rapid sand filtration can remove only part of NOM i.e. up to 30% from the drinking water (Randtke and Stephen, 1988). According to the USEPA Stage I (1999) the implementation of enhanced coagulation or softening is necessary when the concentration TOC is higher than 2 mg/L in raw water. Hence, conventional

treatment is not enough for the eradication of NOM, as the Indian raw drinking water supplies content NOM higher than 2 mg/l.



Fig.14 Efficiency of various WTPs for % removal of NOM species and Turbidity

3.0 Global occurrence of THMs in drinking water

The occurrence of THMs in chlorinated drinking water was first reported by Rook, (1974) and Bellar et al., (1974). After this discovery, in 1975, the national organic reconnaissance survey was conducted for the water supplies of 27 large cities in the United States by Symons et al., (1975). This study confirmed the four THMs (CF, BDCM. DBCM, and BF) are widespread in chlorinated drinking waters at trace concentrations. This discovery alarmed the world's drinking water supply authorities to concern about THMs due to their detrimental to human health and set the trend of regular monitoring in drinking water networks. Hence, in the last 3 to 4 decades, research on THMs received massive attention due to their unacceptable concentration in drinking water and having a potential risk of reproductive disorders and many cancers (Milot et al. 2000; Mahato and Gupta, 2020; Thokchom et al., 2020). The previous research demonstrated that the major factors affecting the formation of THMs viz. level of residual chlorine, concentration and type of NOM, contact time, pH, and water temperature (Thacker et al. 1996; Satyanarayana and Chandrasekhar 1996). The concentration range of THMs in drinking water may vary from country to country due to their geographical location, climatic factor, and many other possible reasons (Thokchom et al., 2020). A diverse range value of these compounds was reported in different countries as follow (Table.19):

Sl.	Country/Location	The	concentrat	s (µg/l)	Reference		
No		CF	BDCM	DBCM	BF	TTHM	-
			Asian	Countries			
1	Islamabad, Pakistan	105	9.15	6.55	BDL	120.7	
2	Rawalpindi, Pakistan	172	10.65	7.32	BDL	189.7	Abbas et al., (2015)
3	Street 10E-7 Islamabad, Pakistan	417.7	10.11	1.11	BDL	428.8	
4	Askari 3 Rawalpindi,	415.1	14.09	2.11	BDL	431.2	
	Pakistan						
5	E-8, Islamabad, Pakistan	575.9	15.16	1.59	3.11	595.8	
6	Karachi, Pakistan					173.2	Siddique et al., (2003)
7	Japan					378	Imo et al., (2007)
8	Kagithane Celebi, Turkey					102	Uyak et al., (2005)
9	Jiangsu Province, China					39.0	Wang et al., (2019)
10	Greater Kandy, Sri Lanka					16	Amarasooriya et al., (2018)
11	Kandy South, Sri Lanka					54	Amarasooriya et al., (2018)
12	Tampin, Malaysia					0.1204	
13	Sabak Bernam, Malaysia					0.1365	Abdullan et al., (2003)
		N	orth Ame	rican coun	tries		
1	Canada					137.8	Milot et al., (2000)

Table19 Concentration range of THMs compounds across the world

S1.	Country/Location	The	concentrati	s (µg/l)	Reference					
No		CF	BDCM	DBCM	BF	TTHM				
2	USA					237	Weinberg et al., (2002)			
3	Toluca, Mexico					13.02	Garrido and Fonseca, (2010)			
4	New Jersey, US					64	Kolb et al., (2017)			
South American countries										
1	Messejana, Brazil	61.4	43.1	18.7	BDL	123.2				
2	Downtown, Brazil	72.1	72.1	72.1	BDL	72.1				
3	Aldeota, Brazil	66.2	44.1	13.3	BDL	123.5	Viana et al.,			
4	Mucuripe, Brazil	72.5	37.0	37.0	BDL	122.9	(2009)			
5	Floresta, Brazil	62.8	62.8	62.8	BDL	105.5				
6	Barra Ceará, Brazil	58.2	38.7	16.1	BDL	113.0				
7	Montese, Brazil	67.9	35.1	16.0	BDL	119.0				
8	Cocorote, Brazil	60.8	60.8	13.6	BDL	120.5				
9	Caucaia, Brazil	61.7	41.5	14.3	BDL	117.5				
10	Maracanaú, Brazil	55.6	35.1	14.6	BDL	105.3				
11	Cali, colombia					40	Montoya et al., (2018)			
12	Huancayo, Peru					39.6	Ore et al., (2019)			
13	Talcahuano, Chile	111.6	25.5	1.0			Loyola et al., (2009)			

S1.	Country/Location	The	concentrat	s (µg/l)	Reference		
No		CF	BDCM	DBCM	BF	TTHM	-
		E	uropean U	nion Cou	ntries		
1	Austria					1.1	
2	Belgium					85.1	
3	Denmark					2.2	
4	Estonia					127	Evlampidou et al., (2020)
5	Finland					93.0	
6	Hungary					771.0	
7	Ireland					255.0	
8	Italy					129.5	
9	Poland					146	
10	Portugal					301	
11	Spain					349	
12	Sweden					100	
13	United Kingdom					100.5	
			African	Countrie	s		
1	Cape Town, South Africa					32.82	Booi, (2013)
2	Hossana Town, Ethiopia					135.7	Zelelew et al., (2018)

Chapter 2

Sl.	Country/Location	The	concentrat	ion range o	of THM	s (µg/l)	Reference
No		CF	BDCM	DBCM	BF	TTHM	-
3	Lagos, Nigeria					997.83	
4	Ogun, Nigeria					825.04	Benson et al., (2017)
			Au	stralia			
1	Melbourne, Australia					153.2	Alexandrou et al., (2017)
2	Sydney, Australia					66.1	
3	Hunter, Australia					66.4	Rahman et al., (2014)

3.1 Occurrence and status of THMs in drinking water

The Indian interest in THMs was received in early 1996, when Thacker et al., (1996); and Satyanarayana and Chandrasekhar, (1996) reported the first occurrence of THMs compounds in chlorinated drinking water of various major cities, i.e., Delhi (66.2 µg/l), Kolkata (51.35 µg/l), Mumbai (62.95µg/l), Chennai (52.73µg/l), Agra (85.12), Goa (100µg/l), Nagpur (88.3µg/l), and Guna (51.46 µg/l). Further, many research has been carried in this domain, like seasonal variation and trihalomethane formation potential (THMFP) of the drinking water in Mumbai, Delhi, and Kanpur city by Thacker et al., 2002; Hasan et al., 2010; and Mishra et al., 2012, respectively. The presence of THMs in drinking water of India was taken into a health concern when the linearity range concentration of CF (45 - 880 μ g/L), BDCM (20 - 900 µg/L), DBCM (65 -900 µg/L) were monitored higher than the prescribed guideline value of WHO (300 µg/L) (Hasan et al., 2010). Moreover, the concentration of THMs (CF- 231.26 µg/l, BDCM - 87.36 µg/l, DBCM -50.20 µg/l, and BF - 46.78 µg/l) in municipal drinking water of Mumbai also exceed WHO standards (Thacker et al., 2002). Later, by the year, the elevated concentration range of CF was observed in the chlorinated drinking water of cities like Kolkata (466 µg/l), Dhanbad (503 µg/l) (Minashree Kumari, 2014), Bokaro (594) (Mishra et al., 2014), Varanasi (380.9 µg/l), Raipur (324.3 µg/l), and Bhubaneswar (319.7 µg/l) (Mahato and Gupta, 2020). In these studies, the CF was reported to be the most dominant THMs compound in chlorinated drinking water. In another study by Basu et al., (2011), an unexpected range value of CHBr₃ (9.78–1854.90 µg/l) was monitored in the water sample of the various thermal power station (Bokaro, Chandrapura, and Durgapur). The variation in the concentration range of these THMs compounds is greatly influenced by the fluctuation of operational parameters (pH, residual Chlorine, temperature) and the seasonal and geographical distribution of the location (Minashree Kumari, 2014; Hasan et al., 2010). Later, multi-exposure cancer and non-cancer risk assessment associated with THMs

compounds are also investigated by many researchers (Mishra et al., 2014; Kumari et al., 2015; Priya and Mishra, 2017; Basu et al., 2011). The concentration range of THMs in Indian drinking, reported in previous literature, is illustrated in Table 20.

	Table 20 The concentration range of TTHVIS in Indian drinking										
Sl.	City/Location	The con	centration ra	ange of THM	ls (µg/l)	Reference					
No		CHCl ₃	CHCl ₂ Br	CHClBr ₂	CHBr ₃						
1	Agra, Ahmedabad, Bombay, Calcutta, Delhi, Goa, Guna, Kanpur Madras, and Nagpur	Presence	Presence	Presence	Presence	Thacker et al., (1996)					
2	Hyderabad	0.0 to 86.5				Srikanth, (1997)					
3.	Mumbai	29.1 to	87.36	0.63 to	46.78	Thacker et al.,					
		231.26		50.2		(2002)					
4	Gantok	36.50	8.70	7.70	5.10	Sharma and					
						Goel (2007)					
5	Delhi	311 to 377	113.3			Hasan et al., (2010)					
6	Bokaro (Thermal Power Station)	232.00	41.70	41.70	868.00						
7	Chandrapura (Thermal Power Station, Bokaro)	203.00	32.10	105.00	428.00	Basu et al., (2011)					
8	Durgapur (Thermal Power Station)	222.00	97.70	19.20	1850.00						
9	Lucknow	13.84 to 74.12	4.71 to 62.69	1.10 to 8.46	1.06 to 5.23	Singh et al., (2012)					
10	Kalpakkam	98	185	201		Rajamohan et al., (2012)					
11	Gwalior	6.03	16.16	2.04	0.13	Nisha et al., (2013)					
12	Kanpur	77.6 to 259.64				Mishra and Dixit, (2013)					
13	Kolkata	466	12	2							
14	Durgapur	255	8	11		Minashree					
15	Ranchi	236	14	31		Kumari,					

Table 20 The concentration range of THMs in Indian drinking

S1.	City/Location	The con	centration ra	ange of THN	ls (µg/l)	Reference
No		CHCl ₃	CHCl ₂ Br	CHClBr ₂	CHBr ₃	-
16	Dhanbad	503	4	2		(2014)
17	Bokaro Steel City	594				Mishra et al., (2014)
18	Dhanbad, Raniganj,	231 to				Kumari and
Barrackpore, and Ranchi	484				Gupta, (2015)	
19	Varanasi	380.9	18.3	15.5		
20	Dhanbad	360.2	16.9	12.3		
21	Raipur	324.3	21.7	14.2		Mahato and
22	Bhubaneswar	319.7	20.3	8.5		Gupta (2020)
23	Barrackpore (Kolkata)	353.1	18.8	12.1		

3.1.1 An experimental approach to know the distribution of THMs in various major WTPs of India

Previous research showed a significant variation in the concentration level of THMs in Indian drinking water. Hence, in order to establish the concentration range and observed their distribution pattern in the water supplies system, five major drinking WTPs from five different states of India, i.e., Jharkhand (DWTP), Utter Pradesh (VWTP), Chattishgarh (RWTP), West Bengal (IGWTP), and Odisha (BWTP), have been selected as earlier.

For the study, the grab samples in triplicates were collected from each WTPs in dry/clean 30-mL glass vials with a polypropylene cap and PTFE-faced rubber septa (Fig.15). The sodium sulphite (0.010 g) was added as a dechlorination agent to eliminate any further THM formation immediately after the sample collection. Furthur the THMs was analyzed in the Department of Environmental Science and Engineering Laboratory, IIT (ISM) Dhanbad, by Gas chromatograph (CHEMITO CERES-800 PLUS). The instrument is equipped with the Ni⁶³ electron capture detector (ECD). Fused silica capillary column DB5, 30M x 0.5mm (id) was used for the chromatographic separation of individual THMs. Injector and detector temperatures were kept at 200°C and 250°C, respectively. The oven temperature was programmed to remain constant at 40°C for 3 min and rise to 150°C at a ramp rate of 8°C/min. Nitrogen was used as a carrier gas at a flow rate of 60 ml/min.



Fig.15 Photograph taken during the sample collection

3.1.2 Distribution of THMs species

The seasonal distribution of THMs compounds in the water supplies system of various selected WTPs is separately summarized in Table 21. The concentration range of TTHMs observed relatively higher in pre-monsoon (348 to 414 μ g/L) than post-monsoon (319 to 356 μ g/L) exceeded the prescribed guideline value of Bureau of Indian Standard (IS 10500: 2012) (200 μ g/l) and USEPA (80 μ g/l) and WHO (300 μ g/l) in all five WTPs. The rates of chlorine decay are high at elevated temperatures; hence, it required higher doses of chlorine for treatment in pre-monsoon, which ultimately reacts with available NOM, thus providing more THMs in processed water (Uyak et al. 2005). Besides, high organic content in water will also require a higher chlorine dose (Rodriguez et al., 2001). Temperature and NOM content in water during post-monsoon were observed slightly lower, resulting in lesser chlorine demand (Rodriguez et al., 2004). Thus comparatively lower THMs formed in this season. Fig.16a-d illustrated the gas Chromatogram of these compounds.

The percentage distribution of all four THMs species shown in Fig. 17a1-5, b1-5 indicated that the CF shared the maximum concentration (84-92%). Next to the CF, BDCM exhibits high concentration followed by DBCM; the occurrence of these compounds often does not exceed the permissible limit of Bureau of Indian Standard, i.e., $60\mu g/l$ and $100\mu g/l$, respectively. According to a report by Chowdhury et al., (2010), more than 90% of the THMs in the chlorinated drinking water supplies typically consist of CF, while BDCM and DBCM contribute up to 2.1–14%. This observation in the present work is suitable in line with the finding of Zhang et al., (2011), where they reported that the contribution of CF was up to 94% to that of other THMs compound in 13 WTPs of China. The concentration level of BF in all five WTPs were not detected, which may be due to the absence of Bromide ion in the treated water. Source water without bromide ions forms more chlorinated THMs than the brominated one (Diehl et al., 2000;Nikolaou et al., 2001), which can also be seen in this study. It was also noticed that chlorinated THMs were predominated over brominated THMs in all the WTPs. The variation in TTHM concentration among the selected WTPs are VWTP> DWTP>IGWTP> RWTP> BWTP and VWTP> IGWTP> BWTP> RWTP> DWTP for pre-monsoon and post-monsoon season respectively (Fig.18a-b). The difference in TTHMs level may be attributed to fluctuation in the composition of NOM (TOC, DOC, UV₂₅₄, and SUVA), availability of free RC, and other operational parameters (Padhi et al., 2019). It may also greatly affected by the geographical distribution and climatic condition of the location.

The level of organic content and the temperaturewere also appeared to be higher during the pre-monsoon, which may favor the greater THMs formation (Nikolaou et al., 2001).

Level of THMs Species (µg/l)										
Pre-Monsoon										
Water utility	CF	BDCM	DBCM	BF	TTHMs	-				
VWTP	3.81E+02	1.83E+01	1.55E+01	ND	4.15E+02					
DWTP	3.60E+02	1.69E+01	1.23E+01	ND	3.89E+02					
RWTP	3.24E+02	2.17E+01	1.42E+01	ND	3.60E+02					
BWTP	3.20E+02	2.03E+01	8.45E+00	ND	3.49E+02					
IGWTP	3.53E+02	1.88E+01	1.21E+01	ND	3.84E+02					
		Post-Monso	oon							
VWTP	3.26E+02	1.69E+01	1.31E+01	ND	3.56E+02					
DWTP	2.88E+02	1.84E+01	1.38E+01	ND	3.20E+02					
RWTP	2.96E+02	2.01E+01	1.39E+01	ND	3.30E+02					
BWTP	3.08E+02	2.15E+01	1.17E+01	ND	3.41E+02					
IGWTP	3.14E+02	2.05E+01	1.38E+01	ND	3.48E+02					
	Water utility VWTP DWTP RWTP BWTP IGWTP IGWTP DWTP RWTP BWTP IGWTP	Level Water utility CF VWTP 3.81E+02 DWTP 3.60E+02 RWTP 3.24E+02 BWTP 3.20E+02 IGWTP 3.53E+02 VWTP 3.26E+02 DWTP 2.88E+02 RWTP 2.96E+02 BWTP 3.08E+02 IGWTP 3.14E+02	Level of THMs Spectromer Spectr	Level of THMs Species (μg/l) Pre-Monsoon Water utility CF BDCM DBCM VWTP 3.81E+02 1.83E+01 1.55E+01 DWTP 3.60E+02 1.69E+01 1.23E+01 RWTP 3.24E+02 2.17E+01 1.42E+01 BWTP 3.20E+02 2.03E+01 8.45E+00 IGWTP 3.53E+02 1.88E+01 1.21E+01 Post-Monsoon VWTP 3.26E+02 1.69E+01 1.31E+01 DWTP 2.88E+02 1.84E+01 1.31E+01 DWTP 2.96E+02 2.01E+01 1.39E+01 BWTP 3.08E+02 2.15E+01 1.17E+01 IGWTP 3.14E+02 2.05E+01 1.38E+01	Level of THMs Species (μg/l) Pre-Monsoon Water utility CF BDCM DBCM BF VWTP 3.81E+02 1.83E+01 1.55E+01 ND DWTP 3.60E+02 1.69E+01 1.23E+01 ND RWTP 3.24E+02 2.17E+01 1.42E+01 ND BWTP 3.20E+02 2.03E+01 8.45E+00 ND IGWTP 3.53E+02 1.88E+01 1.21E+01 ND VWTP 3.26E+02 1.69E+01 1.31E+01 ND WTP 3.26E+02 1.69E+01 1.31E+01 ND DWTP 2.88E+02 1.84E+01 1.38E+01 ND RWTP 2.96E+02 2.01E+01 1.39E+01 ND BWTP 3.08E+02 2.15E+01 1.17E+01 ND IGWTP 3.14E+02 2.05E+01 1.38E+01 ND	Level of THMs Species (μg/l) Pre-Monsoon Water utility CF BDCM DBCM BF TTHMs VWTP 3.81E+02 1.83E+01 1.55E+01 ND 4.15E+02 DWTP 3.60E+02 1.69E+01 1.23E+01 ND 3.89E+02 RWTP 3.24E+02 2.17E+01 1.42E+01 ND 3.60E+02 BWTP 3.20E+02 2.03E+01 8.45E+00 ND 3.49E+02 IGWTP 3.53E+02 1.88E+01 1.21E+01 ND 3.84E+02 VWTP 3.26E+02 1.69E+01 1.31E+01 ND 3.56E+02 DWTP 3.26E+02 1.69E+01 1.31E+01 ND 3.20E+02 DWTP 2.88E+02 1.84E+01 1.38E+01 ND 3.20E+02 RWTP 2.96E+02 2.01E+01 1.39E+01 ND 3.30E+02 BWTP 3.08E+02 2.15E+01 1.17E+01 ND 3.41E+02 IGWTP 3.14E+02 2.05E+01 1.				

Table 21 Distribution of THMs species in various WTPs



Fig.16(a)Gas Chromatogram of THMs compounds



Fig.16(b) Gas Chromatogram of THMs compounds



Fig.16(c) Gas Chromatogram of THMs compounds



Fig.16(d) Gas Chromatogram of THMs compounds



Fig.17a(2) The percentage distribution of all four THMs species in DWTP for pre-monsoon season



Fig.17a(3) The percentage distribution of all four THMs species in RWTP for pre-monsoon season



Fig.17a(4) The percentage distribution of all four THMs species in BWTP for pre-monsoon season



Fig.17a(5) The percentage distribution of all four THMs species in IGWTP for pre-monsoon season



Fig.17b(1) The percentage distribution of all four THMs species in VWTP for the post-monsoon season



Fig.17b(2) The percentage distribution of all four THMs species in DWTP for the post-monsoon season



Fig.17b(3) The percentage distribution of all four THMs species in RWTP for the post-monsoon season

BWTP



Fig.17b(4) The percentage distribution of all four THMs species in DWTP for the post-monsoon season



Fig.17b(5) The percentage distribution of all four THMs species in IGWTP for the post-monsoon season


Fig.18(a) The TTHMs in various WTPs in pre-monsoon



Fig.18(b) The TTHMs in various WTPs in post-monsoon

3.1.3 Correlation study of THMs with various parameters

The Pearson correlation matrix method was used to observe the correlation among the THMs formation, NOM (TOC, DOC, and UV_{254}), and other water quality parameters (pH, water temperature, and RC). The established co-relation matrix is illustrated in Table 22a-b for pre and post-monsoon season, respectively.

3.1.3.1 Correlation with NOM

Wide varieties and range concentrations of NOM are found in natural waters and are monitored as TOC, DOC, and UV₂₅₄. The Pearson correlation matrix showed a strong relationship (pre-monsoon r = 0.956, post-monsoon r = 0.886) between the TTHM formation and TOC for both the season (Table 22a-b). An increase in TOC concentration generally leads to the greater formation of THMs in chlorinated water (Arora et al., 1997; Chang et al., 2001). Statistical data of the present study showed similar trends as the variation in TTHM concentration among the selected WTPs varies according to the concentration of their TOC level present in the water. Babcock et al., (1979) also reported that water samples having a higher concentration of TOC, more THMs will be formed. Moreover, it was also investigated in the research; consumption of TOC is equivalent to the rate of THMs formation (Adin et al., 1991).

Besides TOC, DOC is another key surrogate of NOM liable to form the THMs as much as of the TOC. The DOC also had a strong correlation with TTHMs (pre-monsoon r = 0.876, post-monsoon r = 0.864) (Table 22a-b), which further signifying that rise in the concentration of DOC attributed to formed more THMs in water (Westerhoff et al., 2000). A similar correlation of DOC with THMs formation was also observed by Ye et al., (2009); Reckhow et al., (1990); Sung et al., (2000).

The concentration of organic matter could also be measured as UV₂₅₄, which can provide an insight into the nature of the organics molecules present in water and their potential for THMs formation. UV₂₅₄ showed significant correlations with TTHM (pre-monsoon r= 0.867, post-monsoon r= 0.822,) and furthermore it strongly correlated with TOC (pre-monsoon r= 0.965, post-monsoon r= 0.974) as well as with DOC (pre-monsoon r= 0.973, post-monsoon r= 987) (Table 22a-b) for each season. Overall, TOC was found to be major precursors for THMs, followed by DOC and UV₂₅₄.

3.1.3.2 Correlation with pH

The data of the Pearson correlation matrix present in Table 22a-b depicted that the pH and TTHMs were positively correlated (pre-monsoon r = 0.699, post-monsoon r = 0.667), which signifies that pH value significantly affects the formation of THMs. Many researchers also reported that the concentration of TTHMs increases with an increase in pH (Oliver and Lawrence, 1979; Kim et al., 2002). Under alkaline conditions, base-catalyzed hydrolysis prevails, yielding more THMs, whereas, in an acidic environment, trihaloacetic acids will be formed. Besides, it was also noticed that the chlorine consumption is higher in alkaline pH give rise to

greater THMs formation. In contrast, acidic pH strongly disfavors all the chlorine reactivity pathways, including THMs formation (Navalon et al., 2008).

3.1.3.3 Correlation with Temprature

In general, water temperature appears to be the most influential parameter for DBPs formation, as the concentration of THMs increases with an increase in water temperature (Milot et al., 2000). In the present investigation Pearson correlation matrix of analyze data showed significant correlation of temperature with TTHMs (pre-monsoon r = 0.634, post-monsoon r = 0.886). Temperature provides the activation energy required for the reaction of organic matter, with the residual disinfectant, which results in the greater formation of THMs (Kavanaugh et al., 1980).

3.1.3.4 Correlation with Residual chlorine (RC)

The high range of RC present in treated water consequently increased the formation of chlorinated THMs (Chowdhury and Champagne, 2008). Pearson correlation test in this study indicated that RC has moderately correlated (pre-monsoon r = 0.463, post-monsoon r = 0.610) with TTHMs. When chlorine comes in contact with water leads to the formation of hypochlorous acid (HOC1) and a hypochlorite ion (OC1⁻). The formation of these two species is pH-dependent, as in acidic condition HOC1 is found to be dominated, whereas in alkaline pH OC1⁻ (Uyak et al., 2005). In the present study, pH was noticed in the range between 7.40 to 7.91. Hence, HOCI is the more prevalent chlorine species responsible for the formation of THMs. In this context, when the concentration of THMs increases, the level of HOCI decreases. This result appeared to be inconsistent with the finding of Uyak et al., (2005); and Wei et al., (2010).

Pearson Correlation Matrix									
Parameters	рН	Temp. (°C)	Resudual Chlorine (mg/l)	TOC (mg/l)	DOC (mg/l)	UV 254 (cm ⁻¹)	TTHMs (µg/L)		
pН	1	0.535	-0.171	0.469	0.284	0.325	0.699		
Temp. (°C)		1	-0.169	0.633	0.493	0.649	0.634		
Resudual Chlorine	(mg/l)		1	0.582	0.688	0.515	0.463		
TOC (mg/l)				1	.972**	.965**	.956*		
DOC (mg/l)					1	.973**	0.876		
UV 254 (cm ⁻¹)						1	0.867		
TTHMs (µg/L)							1		

Table 22a The Pearson correlation matrix of variables in pre-monsoon season

*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).

			Pearso	Pearson Correlation Matrix						
Parameters	pН	Temp.	R. Chorine	TOC	DOC	UV ²⁵⁴	TTHMs			
		(°C)	(mg/l)	(mg/l)	(mg/l)	(cm ⁻¹)	(µg/L)			
рН	1	.400	530	.603	.614	.643	.667			
Temp. (°C)		1	895*	.943*	.920*	.851	.886*			
R. Chorine (mg/l)			1	833	801	701	.610			
TOC (mg/l)				1	.998**	.974**	.886*			
DOC (mg/l)					1	.987**	.864			
UV 254 (cm-1)						1	.822			
TTHMs (µg/L)							1			

Table 22b The Pearson correlation matrix of variables for post-monsoon

*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).

3. Global Control of Technology of NOM and THMs with special reference to India

Progress is impossible without change; and those who cannot change their minds cannot change anything

- Francis Bacon

3.0 Background

The global scenario of potable water presents a dismal picture, where most of the developing nations are fighting with the scarcity and contamination of drinking water. Disinfection of water prior to its distribution is necessary and has been used since the early 1900s. Chlorine, the first and the most widely used disinfectant, effectively destroys the pathogen in the water supply system and can reduce the risk of water-borne diseases up to 90% (E.V.Ohanian et al., 1989). However, the issue associated with the chlorination process cannot be ignored during the water treatment process due to the presence of NOM in raw water. Chlorine reacts with the NOM and results in the formation of potentially hazardous DBPs in water (Satyanarayana et al., 1996; Thacker et al., 1996, 1997). Although more than 600 DBPs have been reported in the literature, only a small number has been assessed in quantitative and health effects studies (Chowdhury et al., 2010; Navalon et al., 2008; Kavanaugh et al., 1980). Among the many THMs are most often present in the greatest concentration in the drinking water and used as indicators of total DBPs formation (Bolto et al., 2002). In the last 30 years, identification of these THMs compounds and concern over their possible adverse health impacts has promoted considerable research interest worldwide in order to minimize risks of cancers other possible diseases such as depression of the central nervous system, hepatoxicity, nephrotoxicity, and teratogenicity

Since the discovery, United States Environmental Protection Agency (USEPA), World Health Organization (WHO), and European Union (EU) have already set up regulations for THMs in drinking water. Besides, most developed nations have also promulgated guidelines to control DBPs to minimize consumers exposure to hazardous compounds while, at the same time, maintaining adequate disinfection and control of targeted pathogens (Stalter et al., 2016; Kumari et al., 2015; Satyanarayana et al., 1996; Thacker et al., 1996, 1997).

Controlling THMs in drinking water supplies has become an important issue across the world. Though considerable information on the control and other aspects related to THMs to date is available, it is randomly scattered in the literature. Therefore, the authors feel that a concise review of this significant area is highly appropriate and timely to catalyze further advancements with particular reference to India. The various methods of THMs control commonly adopted worldwise are shown in Fig. 19, as reported by Levchuk et al., (2018).



Fig.19 The various methods of THMs control commonly adopted worldwise (Levchuk et al., 2018)

3.1 Methods for controlling THMs in drinking water

Public water systems could have difficulties when attempting to meet both THMs limits and disinfection regulations. The regulations developed for disinfection and THMs control are of equal importance, and both regulations must be met simultaneously. The formation of THMs in treated water can be controlled through the following methods.

1) Source control of organic precursor at the point of disinfection.

- 2) Use of alternative disinfectant and dose optimization
- 3) Removal of organic precursor.

3.1.1 Source Control

Source control strategies manage the source water to lower the concentration of NOM and bromide in the water. It has been analyzed (Oliver and Shindler; 1980) that the algal growth leads to the production of THMs precursor (NOM). One approach to controlling THMs formation is to prevent nutrient input to waters that

are used as drinking water sources to limit the water's algal growth potential. The algal control strategy is the control of the nutrient cycle in reservoirs and impoundment. Another control measure aimed at controlling bromide level is preventing saltwater or brine intrusion into the water source. By drawing raw water from the water source during seasons when the quality of the raw water is best, storing the water after treatment in controlled storage aquifers, and then recovering the stored water for distribution to consumers, THMs formed during treatment can be eliminated. Additionally, the precursor concentrations can be lowered so that lower levels of THMs are produced from subsequent chlorination.

3.1.2 Use of alternative disinfectant

The alternative oxidants involve supplementing or replacing the use of chlorine. There are many disinfection options, and all of the agents work in a somewhat different way in destroying bacteria, viruses, and protozoa. Some of these alternatives serve only a limited function, such as an alternative primary or secondary disinfectant. However, alternative disinfectants to chlorine are to be concerned must meet several criteria like:

- easily generated,
- effective as biocides,
- easily measurable as the residual,
- less DBP forming than chlorine, and
- cost-effective, and they must still be used in conjunction with chlorine.

The oxidative potential of various alternative disinfectants is shown in Table 23. Many of these disinfecting agents effectively oxidize the organic molecules and other inorganic compounds present in the water.

Species	Oxidation Potential (25°C)
Chlorine	1.36
Hydroxyl free radical	2.80
Ozone	2.07
Hydrogen peroxide	1.77
Chlorine Dioxide	1.27
Monochloramine	1.66
Hypochlorous acid	1.49
Hypobromous acid	1.33

Table 23. Oxidative potential of various alternative disinfectants

3.1.2.1 Chloramines (NH₂Cl)

Monochloramine (NH₂Cl) is an excellent alternative to chlorine for controlling THMs, which does not produce appreciable amounts of any known DBPs. These compounds are formed by combining a specific ratio of aqueous chlorine and ammonia in water. It is useful as a disinfectant for the control of THMs formation, as it does not oxidize bromide to bromine and reduce the formation of brominated THMs. It seems to be less effective than free chlorine, but due to its persistent nature can be used as an attractive secondary disinfectant for the maintenance of a stable distribution system residual. It has the potential to destroy the coliform bacteria and biofilm growth in the water supply system. Monochloramine is most commonly practiced in countries like Finland, Great Britain, Spain, Sweden, etc. According to the drinking water regulation of USEPA, the Maximum Residual Disinfectant Level (MRDL) of monochloramine should not be more than 4.0 mg/L.

3.1.2.2 Chlorine dioxide (CIO₂)

In contrast to monochloramine, chlorine dioxide (CIO₂) is a good disinfectant (relatively low CT values) and an effective oxidant for taste and odor control and iron and manganese oxidation. It does not produce halogenated DBPs to any significant degree, except for chlorite (ClO₂). Chlorine dioxide reacts with NOM to produce oxidation by-products (aldehydes, Aldo- and ketoacids) that are most likely similar to those produced by ozone (Richardson et al., 2000). Chlorine dioxide does not chlorinate rather disinfects by oxidation. It is most commonly used in Germany and Italy; however, it is occasionally practiced in other parts of the world like Austria, Belgium, Finland, France, Great Britain, The Netherlands, Portugal, Spain, Sweden, etc. Chlorine dioxide is more effective than chlorine to treat taste and odor and potentially destroy the *Cryptosporidium sp*. The WHO and USEPA have set their guideline value of 0.7 mg/L and 0.8 mg/L, respectively. Chlorine dioxide requires a high level of technical competence to operate and monitor equipment products, increasing operation costs.

3.1.2.3 Ozone (O₃)

Ozone (O₃) is the most effective oxidant and disinfectant used in water treatment practice. It has the lowest CT values, but disinfection credit is based on residual molecular ozone. It can effectively oxidize the organic matter and can be a primary disinfectant against Cryptosporidium sp. at higher concentrations. It is highly unstable and does not produce a persistent disinfectant residual. Therefore, ozone is an excellent alternative primary disinfectant to free chlorine (Huang et al., 2008). It can also be used in conjunction with a persistent secondary disinfectant like monochloramine. The combination of ozone and monochloramine as primary and secondary disinfectants, respectively, appears to be an attractive combination for minimizing DBP formation while achieving effective disinfection (Čehovin et al., 2017). However, ozone reacts with NOM to produce various oxidation by-products like Aldehydes, Aldo- and ketoacids, Hydrogen peroxide, and acids. It most dominantly practices in the Netherlands, Austria, Belgium, Finland, France, Germany, Great Britain, Ireland, Italy, Spain, etc (Agbaba et al., 2016).

3.1.2.4 Ultraviolet (UV)

Ultraviolet (UV) radiation, generated by mercury arc lamps, is a non-chemical disinfectant. It penetrates the cell wall of an organism, damages genetic material, and prevents cell reproduction. UV radiation effectively inactivates the many pathogens (protozoa, bacteria, bacteriophage, yeast, viruses, fungi, and algae) while forming limited disinfection byproducts (Čehovin et al., 2017). However, it has a little track record in drinking water applications on a large scale, which generates hydroxyl radicals that readily reacts with a wide range of organic compounds without the formation of THMs and any other known DBPs in water (Agbaba et al., 2016). It is also effective against Cryptosporidium but does not provide oxidation, residual protection. UV treatment is most commonly practiced in countries like Austria, Belgium, Finland, Germany, Great Britain, Netherlands, etc (Huang et al., 2008).

3.1.2.5 Hydrogen Peroxide (H₂O₂)

Hydrogen peroxide showed good germicidal activity and attested to bactericidal, virucidal, sporicidal, and fungicidal properties (Agbaba et al., 2016). When added to water produces hydroxyl free radicals that destroy membrane lipids, DNA, and other essential cells of microorganism (Huang et al., 2008). It is active against a wide range of microorganisms, including bacteria, viruses, fungi, yeasts, and spores. It can be an excellent alternative to chlorine, but it does not have residual properties, which may trigger the regrowth of microorganisms in the water distribution system (Čehovin et al., 2017).

3.1.3 Removal of Organic precursors

The formation of THMs is primarily related to the characteristics and level of organic precursors before the chlorination process. It is one of the best strategies to lowering the concentration of THMs in finished water. Enhanced coagulation, adsorption by granular activated carbon (GAC), and membrane filtration are the most widely practiced technologies for removing THMs precursors. Aluminum and ferric salts are the most commonly used coagulant in water treatment industries. However, the use of GAC adsorption and membrane filtration are relatively expensive processes.

3.2 Global control technology of THMs

The control of THMs compounds is highly required in the water distribution system before it reaches the consumers. Though there are various technologies for NOM removal; however, coagulation and adsorption are among the most commonly used techniques to abolish NOM bulk during the drinking water treatment process to minimize the formation of THMs. The other methods of NOM removal including enhanced coagulation, biological treatment, magnetic ion exchange (MIEX), advanced oxidation process (AOP), and membrane filtration.

3.2.1 Coagulation

The coagulation process mainly involves four mechanisms: viz. charge neutralization, entrapment, precipitation, and adsorption (Matilainen et al., 2010). The aluminum (aluminum sulfate) and iron salts (ferric chloride) are the most common inorganic coagulants used for drinking water treatment (Sillanpää et al., 2018; Matilainen et al., 2010). The dissociation of these salts results in the formation of the Al³⁺ and Fe³⁺ ions into the water. Further, they hydrolyzed and formed soluble complexes with high positive charges, which attract the negatively charged NOM colloids in the water (Mahato and Gupta, 2020; Kumari & Gupta, 2020; Sillanpää et al., 2018). It was also observed in the previous research that the efficiency of ferric-based coagulants is higher (approx. 15%) than aluminum ones concerning NOM removal (Matilainen et al., 2010). Later, pre-polymerized coagulants like poly ferric chloride, poly ferric sulfate, and poly aluminum chloride (PACl) set a new trend for the coagulation process (Jiang, 2001;Xu et al., 2004). These coagulants showed better performance than conventional ones due to their stability toward pH and temperature (Matilainen et al., 2010). Table 24 listed the efficiency of various coagulants used for NOM removal in previous research. The efficiency of any coagulants for NOM removal is highly dependent upon its type and dosage, water temperature, and pH, as well as upon the characteristics of NOM itself (Lindqvist et al., 2002; Chaukura et al., 2020; Duan and Gregory, 2003).

This technology is most commonly practiced globally due to the more effortless operation and less capital cost. Coagulation offers distinct advantages over THMs control (Edzwald and Tobiason,1999). However, the generation of solid sludge during the coagulation/flocculation process is a concerning issue.

A luminum based ecoquilants										
Alumnum-based coagulants										
Sl	Coagulants		% NOM	I Remov	References					
No.		TOC	TOC DOC UV254 SUVA		SUVA	-				
			43.2	41.2		Zhao et al., (2014)				
			57.3			Zhao et al., (2011a)				
1.	Aluminum sulfate		65			Pivokonsky et al., (2015)				
	$[Al_2(SO_4)_3]$		54			Chow et al., (2008)				
					28	Uyguner et al., (2007)				
			45			Qin et al., (2006)				
			50-60			Chow et al., (2009)				
			63			Fabris et al., (2012)				

Table 24 Efficiency of various coagulants used for THMs precursor removal in previous research

Aluminum-based coagulants									
Sl	Coagulants		% NOM	l Remov	al	References			
No.		TOC	DOC	UV254	SUVA	-			
			32			Tubić et al., (2010)			
2	Aluminum Chloride		42.7	59.4		Wan et al., (2019)			
	(AlCl ₃)								
		Ire	on-based	l coagula	ants				
					29	Uyguner et al., (2007)			
			45			Uyak et al., (2007)			
3	Ferric chloride		57.9	47.6		Zhao et al., (2014)			
	[FeCl ₃]		59.4			Zhao et al., (2011a)			
			51		30	Tubić et al., (2013)			
			53.4	62.4		Wan et al., (2019)			
			38			Tubić et al., (2010)			
			75			Fabris et al., (2012)			
4	Ferric sulfate				26.31	Heiderscheidt et al., (2016)			
	[2(~ - 1)5]		77			Fabris et al., (2012)			
		Рс	olymeric	Coagula	ants				
5	Polyaluminum		77	91		Zhao et al., (2011b)			
	chloride (PACl)		62			Fabris et al., (2012)			
			57			Tubić et al., (2010)			
6	Polyferric sulfate		81	94		Zhao et al., (2011b)			
	(PFS)		71			Fabris et al., (2012)			
7	Polytitanium		61.1	88		Zhao et al., (2015)			
	tetrachloride (PTC)		36.1	89.6		Chekli et al., (2015)			
8	Polytitanium sulfate (PTS)		76	70		Zhao et al., (2017)			

3.2.2 Adsorption

Adsorption is the surface phenomenon where a liquid phase's constituent is transferred into the solid phase (Dąbrowski, 2001). After coagulation, it is one of the most widely accepted technology for water purification due to its convenience, easy operation, and most straightforward design (Bhatnagar and Sillanpää, 2017). It

is a mass transfer phenomenon that involves the accumulation of substances between the two-phase interface, like liquid-solid, gas-liquid, liquid-liquid, or gassolid (Dąbrowski, 2001). The sorption of a dissolved constituent from the liquid phase to the solid surface may occur by the attractive interaction, mostly due to the van der Waals and electrostatic force (Zhang and Liu, 2010). Other mechanisms involving in the adsorption process are hydrogen bonding, anion and ligand exchange, surface complexation, entropic effects, hydrophobic interaction, and cation bridging (Gu et al., 1994; Newcombe, 1999). Among these, it is very difficult to know which mechanism is dominant over another. Hence it needs to implement the adsorption isotherms, kinetics, and thermodynamics model to reveal the interaction of adsorbate and adsorbent. In addition, the process of adsorption is also greatly influenced by the physicochemical characteristics of an adsorbent, such as surface area, porosity, size, shape, pore-volume, functional group presence in the surface, and point of zero charges (PZC) (Gibert et al., 2013; Iriarte et al., 2008).

The behavior of NOM adsorption is challenging to understand because of its heterogeneous nature. However, the characteristical aspects of NOM that influence the adsorption process include molecular weight and charge distribution, hydrophobicity/hydrophilicity, and the group capable of hydrogen bonding with the surface (Newcombe, 1999). It is one of the best methods to control THMs formation before the chlorination process. The literature was available on a wide variety of adsorbents for THMs control is illustrated in Table.25.

Sl	Adsorbent		% NOM	Remov	References	
No.		TOC	DOC	UV254	SUVA	
1.	Granular activated carbon (GAC)	65				Gibert et al., (2013)
2.	GAC	43- 65				Capar and Yetis, (2001)
3.	GAC and sand filter	75				Rasheed et al., (2016)
4.	Modified GAC		68			Cheng et al., (2005)
5.	Stratified layer of sand, GAC & Pyritic fill.		40			Grace et al., (2016)
6.	Powdered activated carbon (PAC) with aluminium sulphate			40		Fabris et al., (2004)
7.	Chitosan- PAC		69			Zhang and Liu,

Table 25 Efficiency of various adsorbent used for THMs control in previous research

	composite adsorbent					(2010)
8.	Nano-scale carbon black	70				Wang et al., (2010)
9.	Carbon nanotube (CNT)	36- 66				Ajmani et al., (2014)
10.	Multiwalled CNT	NR	NR	NR	NR	Lu and Su, (2007)
11.	TiO2 Nanoparticles		76.5 4			Gora and Andrews, (2017)
12.	Magnetic Fe ₃ O ₄ nano- particles	60				Zulfikar et al., (2016)
13.	Magnetic chitosan nanoparticle	50				Dong et al., (2014)
14.	Fly Ash		23.7	25.9		Wei et al., (2011)
15.	Iron-oxide particle		17-25			Choo and Kang, (2003)
16.	Silica particle coated with an aminosiloxane SAM (NH ₂ -SAM).		70	60		Chow et al., (2009)
17.	Silicate rocks (tobermorite and zeolite)	50				Kaneco et al., (2003)
18.	Iron coated pumice and H ₂ O ₂ hybrid process.		74			Kaplan et al., (2016)
19.	Surfactant modified Zeolite	40				Niri et al., (2015)
20.	Self-assembled monolayer of Silica			60		Chow et al., (2009)

NR-% removal	not re	ported
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3.2.3 Advance oxidation process (AOPs)

The AOPs are other available alternative technology for the treatment of THMs in drinking water. It has promising potential to eradicate the NOM and various pollutants in water and wastewater (Sillanpaa et al., 2018b). The practice of AOPs includes various catalytic and photochemical methods that involve the generation of highly reactive radicals intermediate such as hydroxyl radicals (OH*) (Glaze et al., 1992). This radical has been reported to be highly efficient for the oxidation of a wide range of organic compounds (Matilainen and Sillanpää, 2010). It can also mineralize organic contaminants instead of only transferring NOM from one phase to another, as seen in the conventional drinking water treatment (Tak et al., 2020).

Mainly three mechanisms involved during the interaction of hydroxyl radicals generated by AOPs with the organic constituent of water: (i) hydrogen abstraction from aliphatic carbon atoms yielding carbon-centred radicals, (ii) electron transfer reactions, where HO* gains an electron from an organic substituent, and (ii) electrophilic addition to double bonds or aromatic rings (Parsons, 2004; Matilainen and Sillanpää, 2010). This radical's highly oxidizing capabilities made AOPs gain complete oxidation or mineralization through a process that operates near ambient temperature and pressure. The oxidation rate of contaminants depends on radical, oxygen, and its concentration level. Moreover, the formation of radicals during the AOPs is affected by factors like ions, pH, temperature, type of pollutant, and scavengers such as bicarbonate ion (Parsons, 2004). In general, for commercial drinking water treatment systems, AOPs are applied under low to moderate oxidation conditions, where the NOM is partially oxidized, and high MW fractions are transformed into low MW compounds (aldehydes and carboxylic acids) (Liu et al., 2015; Sillanpaa et al., 2018b). Overall, AOPs showed good potential to remove THMs precursors from water. The list of various oxidants used in the previous literature is shown in Table 26.

Sl	Oxidant/AOPs		% NOM	References		
No		TOC	DOC	UV254	SUVA	-
1.	O ₃	4-15		10-62	1-55	Agbaba et al., (2016)
2.	O ₃		19.2	71.6	64.8	Čehovin et al., (2017)
3.	O ₃ / hydrodynamic cavitation (HC)		11.7	76.6	73.5	Čehovin et al., (2017)
4.	UV	10		13.04		Agbaba et al., (2016)
5.	H_2O_2/UV	5.17		18.18	15.38	Agbaba et al., (2016)
6.	H ₂ O ₂ /UV		23.1	44.9	28.3	Čehovin et al., (2017)
7.	H ₂ O ₂ /UV/HC		30.8	45.3	21.0	Čehovin et al., (2017)
8.	O ₃ /UV	8.62		50	43.58	Agbaba et al., (2016)
9.	O ₃ /UV		45.7	82.2	67.9	Čehovin et al., (2017)
10.	O ₃ /UV/HC		23.5	78.0	71.2	Čehovin et al., (2017)

Table 26 Efficiency of various AOPs for THMs precursors removal

Sl	Oxidant/AOPs		% NOM	References		
No		TOC	DOC	UV254	SUVA	-
11.	O ₃ /H ₂ O ₂ /UV	15.51		68.18	56.41	Agbaba et al., (2016)
12.	H_2O_2/O_3	10-25				Peleato et al., (2017)
13.	H_2O_2/O_3		43.6	73.7	53.3	Čehovin et al., (2017)
14.	H ₂ O ₂ /O ₃ / HC		40.5	77.5	62.1	Čehovin et al., (2017)
15.	H_2O_2/O_3	78				Alsheyab et al., (2006)
16.	Degussa TiO ₂ slurry/		50	50		Gerrity et al., (2009)
	Photo-Cat Lab					
17.	TiO ₂ /UVA		80			Liu et al., (2010)
18.	Low-Pressure (LP) UV			42-46		Dobrovic et al., (2007)
19.	TiO ₂ /UV		5-32	10-33	15-47	Uyguner et al., (2007)
20.	TiO ₂ /LP UV		50	85		Huang et al., (2008)
21.	Iron oxides/Fe ⁰ /H ₂ O ₂ /		60			Nie et al., (2010)
	High Pressure (HP)- UV					
22.	UV/H ₂ O ₂	23		60		Lamsal et al., (2011)
23.	$Fe^{2+\prime}H_2O_2$		55			Molnar et al., (2015)

3.2.4 Membrane Filtration

In the water treatment industries, the membrane filtration (MF) technique has been available for several decades, but its application towards THMs precursors removal has increased dramatically in the recent era. MF offers various advantages over the conventional water treatment system like superior water quality, less energy consumption, simple maintenance, etc. (Metsamuuronen et al., 2014; Huang et al., 2009). On the basics of membrane pore size MF is classified into (i) reverse osmosis (RO), (ii) microfiltration (McF), (ii) ultrafiltration (UF), and (iv)

nanofiltration (NF) (Metsamuuronen et al., 2014; Zularisam et al., 2006). Among these, NF has proven to be the most effective technique for the complete removal of THMs precursors and hardness (Schafer et al., 2001). At the same time, UF can partially remove NOM and virus, whereas McF only treats the particulate or bacteria but no dissolved compound (Siddiqui et al., 2000;Thorsen, 1999). The MW cut-off value of NF membranes lay in the range of 100–500 Da, capable of removing the compounds from macromolecular size to multivalent ions (Metsamuuronen et al., 2014). It can be an alternative technique for controlling THMs for drinking water, but its high operation cost limits its application on a large scale.

3.2.5 Electrochemical Method

Electrochemical methods are other alternative techniques for THMs control, trending over the conventional ones (coagulation,flocculation sedimentation/flotation, and filtration). This water treatment technique and disinfection have been the subject of growing interest in recent years (Rathod et al., 2020; Sarkka et al., 2015). The methods of electrochemical techniques involve (i) electrocoagulation (EC), (iii) electrolocation (EF) and, (ii) electrochemical-oxidation (EO), have proved their efficiency not only for the removal of THMs precursors but also for coliform and algae in surface water (Chen, 2004).

3.3 Scenario of THMs control and their toxicity in India

The Indian interest in THMs was received in early 1996, and long after years, their guideline value was also established in 2004 by BIS. Although several reports are available for the monitoring and distribution of THMs on the Indian drinking water supply system. But very limited work has been carried out on their control technology and toxicity in India. However, the concentration range of THMs (231-511 μ g/l) reported in previous research is a serious concern to human health safety.

3.3.1 Studies of THMs cancer risk assessment in India

Many works have been carried out on human health risk assessment of THMs compounds through chlorinated drinking water in India. Possibly the first study was conducted by Sharma et al., (2007) to evaluate the long-term impacts of chlorinated drinking water in the northeast region viz. Sikkim and Gangtok. The monitored concentration range of individual THMs species in this region were $36.50 + 67.43 \mu g/L$ (CHCl₃), $8.70 + 1.34 \mu g/L$ (CHBrCl₂), and $7.70 + 1.06 \mu g/L$ (CHClBr₂), at the same time, the level of TTHMs was found $61.60 + 10.33 \mu g/L$. They have conducted a population-based cross-sectional study where the door-to-door survey was performed to know the adverse health outcomes of the community consuming chlorinated water. Three groups have been identified, viz. exposed group – people exposed to chlorinated water for the last 30 years; control group 1 (CG1) – people exposed for less than 30 years; and control group 2 (CG2) people with no exposure. Throughout this study, 23 cases of cancer were reported, 14 from the exposed group, 4 from CG1, and 5 from CG2.

Another study of THMs cancer risk assessment in India was carried out by Basu et al., (2011). In this investigation, multiple exposures (oral, dermal, and inhalation) lifetime cancer risk and hazard index of THMs compounds were estimated in the drinking water of 11 WTPs from two different states, i.e., West Bengal and Jharkhand. Exposure through the inhalation route contributed to a higher risk (80–90%) for THMs followed by oral ingestion (>10⁻⁶). However, the exposure risk via dermal contact was observed insignificant during this study. CF imparted the highest cancer risk through inhalation exposure compared to other THMs compounds. In addition, males were at a higher risk of cancer than females for inhalation exposure (Fig. 20a-b). The male's female's exposure to THMs via other routes viz. oral and dermal contact are shown in Fig. 21 a-b and Fig. 22a-b, respectively. The average hazard index of total THMs through the oral route was higher than unity, indicating high noncarcinogenic risk.



Fig. 20(a) Lifetime cancer risk through inhalation exposure in Male



Fig. 20(b) Lifetime cancer risk through inhalation exposure inFemale



Fig. 21(a) Lifetime cancer risk through oral exposure in Male



Fig. 21(b)Lifetime cancer risk through oral exposure in Female



Fig. 22(a) Lifetime cancer risk through dermal contact in Male



Fig. 22(b) Lifetime cancer risk through dermal contact in Female

Mishra et al., (2014) evaluated the multipath way lifetime cancer risks of THMs through ingestion, dermal absorption, and inhalation exposure based on daily average water intake for adult males (4 L/day/ per person) and females (3 L/day/ per person) as per the Indian condition. In addition, average body weight and expectancy of life were also considered as major factors while assessing cancer risk. For the THMs analysis, the water sample was collected from 8 different WTPs of two-state, i.e., Jharkhand and West Bengal. The concentration range of THMs (269 and 594 μ g/L) in these WTPs was determined high, exceeded the guideline value of BIS (200 μ g/L), WHO (300 μ g/L), and USEPA (80 μ g/L).

The estimated total cancer risk in males and females were reached 8.99E-04 and 8.92E-04, respectively (Fig. 23 a-b). The inhalation route contributed higher risk of cancer followed by ingestion and dermal contacts. It was also observed that the average risk due to chloroform was highest among the three THMs compounds. The overall study suggested that the quality of drinking water in these WTPs was unsafe in terms of cancer risk evaluation.



Fig. 23(a) Multiple pathway cancer risks of THMs in males



Fig. 23(b) Multiple pathway cancer risks of THMs in females

Another study of multi-exposure cancer and non-cancer risk assessment of THMs compounds was carried out by Kumari et al. (2015) in the drinking water supplies of the Eastern region of India. This study was based on the concentration level of THMs compounds considering the factors average body weight, exposure frequency, water intake, and duration of exposure as per the guideline of the Indian Council of Medical Research (2009). For this study, five WTPs were selected from the Eastern region of India, viz. Asansol Durgapur Development Authority WTP, Raniganj (ADDA), Swarnrekha WTP, Ranchi (SWTP), Mineral Area Development Authority WTP, Dhanbad [MADA (N)], Maithon WTP, Maithon (MWTP) and Indira Gandhi WTP, Kolkata (IGWTP II). The river Ganga and Damodar were the raw water source for these WTPs. The lifetime THMs cancer risks in supply water were found 100 times higher than prescribed by USEPA. It was observed that a higher risk comes from oral ingestion followed by inhalation (Fig. 24a-c). However, the insignificant cancer risk was determined from dermal exposure. The calculated average total cancer risk indicated that females (379 times) were at a higher risk of cancer than males (316 times).



Fig. 24(a) Lifetime cancer risk of THMs through dermal contact



Fig. 24(b) Lifetime cancer risk of THMs through oral ingestion



Fig. 24(c) Lifetime cancer risk of THMs through inhalation exposure

Lately, Anchal et al., (2020) assessed the human health risk of halogenated disinfection by-products (CF) in swmining pools water. For the THMs analysis, sampling was executed in various indoor swimming pools in and around the Dhanbad district of Jharkhand. The concentration level of THMs in water and air was monitored 197.18 \pm 16.31 µg/L and 0.033 µg m³⁻¹, respectively. USEPA methods for Swimmer Exposure Assessment Model (SWIMODEL) was used to estimate cancer risk. The exposure through an inhalation route contributed a maximum (up to 99%) risk to total cancer during the swimming activity. However, cancer risk due to accidental ingestion and dermal contact with skin was found insignificant and negligible. They suggested that regular monitoring and control of THMs in the swimming pool are highly required to minimize the potential risk. They also recommended determining the degree of bacteriological contamination to reduce the necessary dose of chlorine for disinfection to decrease the formation of THMs.

3.3.2 Studies of THMs control in India

The elevated concentration range of THMs in Indian drinking water supplies warrants in-depth attention to the regulatory agencies and management authority for controlling their levels in supply water to protect the community from probable health hazards. According to USEPA, the best way to prevent the formation of THMs in water is to eradicate or limit the concentration of NOM before the chlorination process. In India, many research has been carried out to remove NOM from drinking water since the last decade.

Priya et al., (2018) investigated the efficiency of zirconium oxychloride and aluminum sulfate as a coagulant to control the formation of THMs. They have conducted a bench-scale coagulation experiment in jar test apparatus using synthetic water prepared by HA. The optimized dose of coagulant was added to 1 litter of synthetic water was subjected to rapid mixing at 200 rpm for 2 min followed by slow mixing at 40 rpm for 15 min. After the coagulation study, the concentration of NOM was analyzed in terms of UV absorbance and adsorption slop index (ASI). Zirconium oxychloride was much more effective than aluminum sulfate for removing the slow and fast reducing agents, which correspond to the reduction rate of phenolic groups. This group eventually decreased THMs cancer risk by ~ 2.3 times. Zirconium oxychloride also showed rapid chlorine decay and can reduce the ASI and phenol by 57.98% and 49.02%, respectively, from NOM enriched water. It possessed higher charged ions and a rapid hydrolysis rate, which made it an effective coagulant for removing suspended particles and aromatic moieties of NOM. The surface characteristics of Zirconium and aluminum NOM flock and a three-dimensional plot for coagulant activity for ASI and phenol reduction are shown in Fig. 25a-b and Fig. 26a-b.



Fig. 25(a) FE-SEM image of Zirconium -NOM Flocs





Fig. 25(b) FE-SEM image of Aluminum -NOM Flocs.



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Fig. 26(a) Three-dimensional plot for coagulant activity of ASI



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Fig. 26(b) Three-dimensional plot for coagulant activity of phenol

Kumari and Gupta, (2018) developed a surfactant modified magnetic nanoadsorbents (MNPs) for treating the aromatic and hydrophobic fractions of NOM. The magnetic nanoadsorbents was synthesized by co-precipitation method using the salts of $FeCl_2$ and $FeCl_3$ in conc. HCl with the presence of ammonia. Furthur, it was coated with polyethylene glycol (PEG) for biocompatibility and chemical stability of the particles. They have conducted batch adsorption study with various dose (0.1 to 3 g/L) and contact time (5, 10, 15, 20, 25, 30, 35, 40, 50, 60, 90, and 120 min) using synthetic water having initial TOC concentration of 5 mg/L. The distilled and tap water in the ratio of 1:1 were mixed for preparation of synthetic water. The PEG-modified MNPs showed excellent potential for removing DOC (94.49%) and UV₂₅₄ (89.32%) in the netural pH (7) condition. The adsorption process routed through the multilayer chemisorption via chemical interaction between aromatic and humic compounds of NOM with MNPs. Due to the excellent adsorption and regeneration potential, MNPs can effectively be used for the treatment of NOM in water industries for Indian condition. The surface characteristics and NOM (DOC and UV₂₅₄) removal efficieny of MNPs are shown in Fig. 27a-b and Fig. 28a-c, respectively.



Fig. 27(a) Surface morphology of MNPs (a)FESEM and (b) TEM image



Fig. 27(b) TEM imageof MNPs



Fig. 28(a) Effect of MNPs on the removal DOC



Fig. 28(b) Effect of MNPs on the removal UV_{254}



Fig. 28(c) Effect of MNPs on the removal THMPF

An aluminum-based electrocoagulation (EC) method was tested by Snehi et al., (2019) for the treatment of NOM (reactive fractions) from chlorinated water. In this approaches the sample water was collected from Jamadoba WTP, Dhanbad and their characteristics has been altered by adding mine water in different ratios (1:1, 1:2, 1:3, and 1:4) to improve the NOM removal efficiency of EC method. For this study a reactor with the dimension of 160 mm \times 130 mm \times 120 mm (capacityl L) was fabricated along with an aluminium based single cathode and anod electrods. The efficiencies of EC method was monitored at different current densities (0.50, 1.50, 2.50, 3.50, and 4.50 mA/cm²), and pH (2.5, 4, 5.5, 7, and 8.5), for a contact time of 10–30 min. In an optimized condition (pH 5.35, current density 3.07 mA/cm² and contact time 25 min) it showed great efficiency for UV₂₅₄ (86%) and DOC (75%) removal (Fig. 29a-b). This methods is also found to be good effective for eradicating the turbidity and phenolic content of NOM which greatly affects the formation potential of THMs (THMFP) (Fig. 29c-d).



Fig. 29(a) Three-dimensional plot of EC for the removal UV_{254}



Fig. 29(b) Three-dimensional plot of EC for the removal of DOC



Fig. 29(c)Three-dimensional plot of EC for the removal of Turbidity



Fig. 29(d)Three-dimensional plot of EC for the removal of Phenol

An another study by Mahato and Gupta, (2020) efficiency of acid modified Bael fruit shell (AM-BFS) was investigated for the removal of NOM from the real water samples collected form Belatand water treatment plant Dhanbad (DWTP) Jharkhand. Thermo-chemical treatment was given to the raw Bael fruit shell using 88% ortho-phosphoric acid (H_3PO_4) in 2:1 (w/v) ratio. This treatment enhanced the surface properties like specific (8 times) and external surface area (13 times) of raw Bael fruit shell (VBFS) for better adsorption (Fig. 30a-d). The batch adsorption study was conducted with obtained AM-BFS by varing the operational parameters (pH 6-9, time 200-280 min and dose 3-9 mg/L) (Fig. 31a-d). It demonstrated the excellent potential for NOM adsorption which is approx. 2 and 3 times higher than the commercially available granular and powder activated carbon, respectively. The enhanced surface characterstics and positive surface charge of AM-BFS provoked higher adsorption. Owing to the easy availability of raw material and facile synthesis techniques, AM-BFS could prove to be an effective adsorbent for the removal of NOM, from drinking water and thus provide a vital corridor for obliterating the formation of THMs and ensuring public health and safety.





Fig. 30(b) SEM images of AM-BFS



Fig. 30(d) SEM enlarged view of AM-BFS after NOM adsorption



Fig. 31(a) Three-dimensional plot of effects dose of and pH for the NOM removal



Fig. 31(b) Three-dimensional plot of effects of time and pH



Fig. 31(c) Three-dimensional plot of effects of time and dose



Fig. 31(d) The plot of zeta potential for AM-BFS
Lately, Mahato and Gupta, (2021) examined the potential of cerium oxide nanoparticles (CONPs) for the adsorptive removal of various spectral different indices of NOM. These indices include DOC, UV₂₅₄, adsorption slop index (ASI), Phenolic, and Carboxylic content of NOM. They have synthesized the CONPs by two methods, viz. efficient microwave combustion (ECM) and hydroxide mediated approach (HMA) and labeled as CONP-I and CONP-II, respectively. The batch adsorption experiments were performed using 100 ml of synthetic water at an initial concentration of 50 mg/L of DOC. Both the developed nanoparticles were found identical in their physicochemical properties. However, CONP-I possessed slightly higher adsorption capacity for the simultaneous removal of DOC (94%), UV₂₅₄ (93%), ASI (95%), Phenolic (88%), and Carboxylic content (73%). In addition, the maximum adsorption capacity (238.09 mg/g) was monitored 1.2+-5 times higher than other reported nanomaterial at neutral pH. The specific physic-chemical, redox, and optical properties made the CONPs a suitable adsorbent. The instant and straightforward synthesis technique with very few lab assets made this adsorbent quickly available for effective NOM removal. The surface morphology, elemental composition and effect of operation parameters are shown in Fig. 32a-d and Fig. 33a-f, respectively.



Fig. 32(a) FESEManalysis of CONP-I



Fig. 32(b) EDX analysis of CONP-I



Fig. 32(c) FESEM analysis of CONP-II



Global Control of Technology of NOM and THMs with special reference to India

Fig. 32(d) EDX analysis of CONP-II



Fig. 33(a) Effect Time for CONP-I







Fig. 33(c) Effect pH for CONP-I



Fig. 33(d) Effect pH for CONP-II



Fig. 33(e) Effect Dose for CONP-I



Fig. 33(f) Effect Dose for CONP-II

4.0The Study revealed the following facts

NOM is liable to change the watercolor from yellow to brownish and is mainly responsible for THMs formation in chlorinated drinking water.

A diverse concentration range of NOM in the surface water of various countries was noticed globally viz.USA (3.9 to 5.7 mg/L), Malaysia (2.4 to 2.6 mg/L), (Canada (4.0 to 7.9 mg/L), China (2.0 to 5.6 mg/L), Iran (4.72 to 10.37 mg/L), as well as in Nordic countries (Finland, Sweden and Norway) (3.6 to 54 mg/L).

The concentration range of NOM in Indian surface-water varied from 2.1 - 44.1 mg/L, which significantly influences the water treatment processes of various WTPs. The efficiency of the conventional treatment process of Indian WTPs was found to be marginal for the removal of NOM species viz. SUVA (up to 48 %) followed by UV₂₅₄ (up to 47 %), DOC (up to 37 %), and TOC (up to 34 %).

The Indian interest in THMs was received in early 1996, and later by the year, the elevated concentration range of THMs was observed in the chlorinated drinking water of various major cities of India viz. Kolkata (466 μ g/l), Dhanbad (503 μ g/l) Bokaro (594), Varanasi (380.9 μ g/l), Raipur (324.3 μ g/l), and

Bhubaneswar (319.7 μ g/l), etc. which exceed guideline value of WHO (300 μ g/L), USEPA (300 μ g/L) and BIS (200 μ g/L).

The concentration range of THMs was noticed relatively higher in premonsoon (348 to 414 μ g/L) than post-monsoon (319 to 356 μ g/L), where the CF shared the maximum concentration of TTHMs followed by BDCM and DBCM.

The human health risk assessment of THMs compounds revealed inhalation route contributed higher risk of cancer followed by ingestion and dermal contacts. It was also observed that the average risk due to chloroform was highest among the three THMs compounds and the females in India were at more risk of cancer than Males.

India developed many technologies for removing NOM (MNPs, AM-BFS, CONP, aluminum-based electrocoagulation, etc.) from drinking water to minimize the formation of THMs and ensure public health safety.

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