

A Review on techniques used for removal of Natural Organic Matter (NOM) from the water.

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Abstract - The natural organic matter in the water continuously affects the treatment units due to their transformations. The NOM characteristics are complex, and it is challenging to clarify and remove the NOM in water. Due to the modifications of NOM during the disinfections, which results in the formation of by-products, their removal is problematic and causes adverse effects on human health. Several techniques are adopted to remove NOM from the water; one standard method involves coagulation and similar processes. The current review provides insights about the methods available for removing the natural organic matter and highlights the coagulation techniques that are gaining prominence in the current scenarios to remove the NOM from the various sources.

substances are highly soluble at the higher pH and show less solubility under acidic conditions, and even form precipitate at $\text{pH} \leq 4$. FA shows low molar masses, oxidizes faster, is polar, and is soluble at all pH [5].

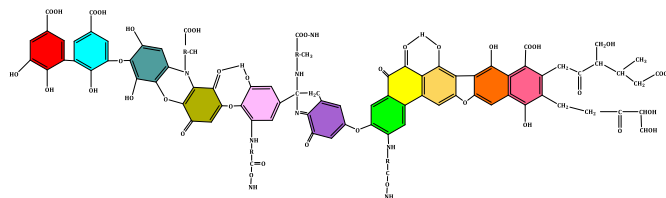


Figure 1: Humic acid structure from the literature [7, 8].

Key Words: Coagulants, Coagulation, Natural organic matter, coagulating aids.

1.INTRODUCTION

The primary sources for drinking water supply are surface water and aquifers. Most water sources consist of NOM (natural organic matter), suspended substances, and pathogenic biological substances (algae, fungi, and protozoa). NOM is a complex compound consisting of different organic materials in most natural waters [1]; NOM is a significant factor affecting water quality worldwide [2, 3]. These compounds have different chemical compositions, functionalized groups, solubility, molecular sizes, and polarity [4]. The essential NOM components are humic acid and fulvic acids in water treatment. Breakdown of the animal and plant matter results in the formation of HA. These are complex structures with molecular weights ranging from 500-5000 Daltons and highly aromatic (Figure 1). The colour of NOM is brown to yellow as these substances have various functionalized groups, generally influenced by the moieties of the phenolic and carboxylic group [5, 6]. Based on the solubility in acids/bases, the humic substances are classified into two types, i.e., HA and FA. HA

The HA and FA molecule's solubilities and charges increase with increasing pH. The functionalized compounds become more acidic and ionized; the HA and FA molecules become more negatively charged and polar [9]. The negatively charged molecules help in the complexation of mineral surfaces with cations. At the same time, macromolecules' hydrophobic regions interact with various hydrophobic complexes like pesticides, chemicals from industries, pharmaceutical wastes, and herbicides [10]. In addition to the HA and FA substances, lower molar masses of carboxylic groups, amines, pyrroles, alcohols, tannins, and polysaccharides are found in the NOM complexes [11, 12, 13].

The HA in the drinking water itself doesn't cause any effects on human health directly [19, 20], even though the HA add colour to the water and produce pungent odours and taste [21, 22, 23]. Furthermore, if water consisting of HA is treated by disinfecting agents like chlorine or chloramines results in the formation of halogenated compounds (toxic), commonly called "Disinfection By-products" (DBPS). These compounds in the drinking water cause several diseases like endocrine disorders and cancers due to their toxicity [24]. Thus, to minimize the

formation of DBPS, concentrations of NOM and HA should be reduced before the process of disinfection [26-27].

2. TECHNIQUES USED IN NOM REMOVAL

The most widely used techniques for removing NOM from the drinking water include activated carbon adsorption, membrane filtration, oxidation, and advanced oxidation process, Ballasted Flocculation Separation, Magnetic Ion Exchange (MIEX), and coagulation [28-30].

a. Activated carbon adsorption (AC)

Activated carbon adsorption methods are widely accepted and used in drinking water treatment. These methods are proven to be efficient in removing micro-pollutants, chemicals, pesticides, toxins, taste, and odours. "Granular-activated-carbon" (GAC) or "powdered-activated-carbon" (PAC) materials broadly utilized for the NOM removal; however, due to competition for adsorption sites, these materials' removal efficiencies decrease and results in less removal of other pollutants (Figure 2). Due to the NOM heterogeneity, understanding these compounds' adsorption behaviours is complex [31]; Newcombe et al., 2002, [32] revealed that the activated carbon adsorption process could be controlled NOM molecular size and AC pore size distribution. Thus, utilization of GAC for NOM adsorption is not generally recommended as HA molecules' presence saturates the media's capacities causes blocking of pores; to avoid the blocking of filters, pre-coagulation techniques are used before the GAC filtration. Pre-coagulation processes remove some NOM moieties, reduce the filter's load, and enhance the filter's life [33]. The AC post-filtration method showed their efficiencies by protecting the treated water and avoiding DBP formation. AC filters can be integrated with the heterotrophic biomass to form BAC (Biological Activated carbon adsorption process), removing biodegradable compounds. PAC's addition reduces the coagulation dose and further enhances the coagulation efficiency [34]. The thermal reactivation might improve the GAC filters life span; however, this technique can increase the pore's size (to macropores) due to the burning of filters; increasing pore size increases the removal of NOM moieties

with higher molar mass and reduces the disposal of NOM with low molar masses [35].

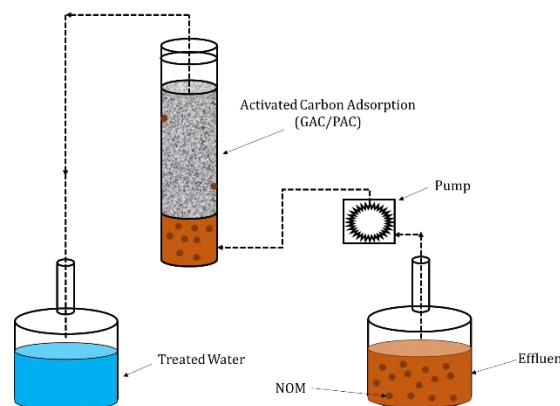


Figure 2: Schematic representation of activated carbon adsorption process for the NOM removal and water treatment.

Guirguis et al., 1978 reported that the pre- ozonation techniques could improve GAC filters' performance by making some organic substances absorbable/biodegradable; at a dosage of 3 mg/L and 14 mg/L during pre-ozonation, it was observed that effluent quality, AC life span, and removal efficiencies were enhanced. GAC adsorption can be improved by integrating lower doses of ClO₂ oxidation or by ozonation. This process enhances the molar masses of the NOM moieties and increases the NOM molecular weight; GAC adsorption sites attain more significant affinities in adsorbing the NOM [37]. However, the addition of higher dosages of ClO₂ might break down the NOM larger molecules into smaller molecules [38]. Furthermore, interactions among these compounds might lead to DBPS which are toxic. Recently, GAC materials have been substituted by the new sustainable materials obtained from recycling waste. Biochar is such material obtained from the pyrolysis of organic waste proven efficient [39, 40].

b. Membrane filtration

Over the past few decades, the increase in membrane technologies used for water treatment, including nanofiltration, ultrafiltration, and reverse osmosis, has increased [41, 42]. These filtration techniques are very effective but require high energy and create complexity for the water plants. The pore size of the ultrafiltration membranes ranges from 0.1 µm to 0.002 µm and requires a pressure of 2×10² to 7×10² kPa [43]. Ultrafiltration membranes are proven to be efficient in removing the microbial species [44] and NOM with higher

molar mass; however, this technique cannot remove the NOM with lower molar masses and results in pore blockage [45]. Gibert et al., 2015 [45] stated that using the ultrafiltration (UF) technique reduced the dissolved organic carbon from river water from 3.1 mg/L to less than 0.05 mg/L. Xu et al., 2019 [46] reported similar results and removed the DOC from 3.55 to 3 mg/L using the UF technique. Nanofiltration (NF) outperformed UF as their pore sizes are less than 0.003 μm with a feed pressure of 6×10^2 to 103 kPa higher than the UF [43]. Recently Ogutverici et al., 2016 [47] reported that NF techniques removed the triclosan from the feed from $1.862 \times 10^3 \mu\text{g/kg}$ to $1.160 \times 10^3 \mu\text{g/kg}$ [48]. Further, Shen and Schäfer, 2015 [48] reported that at the feed rate of 270 mg carbon/L, NOM removal by NF was found to be eighty per cent.

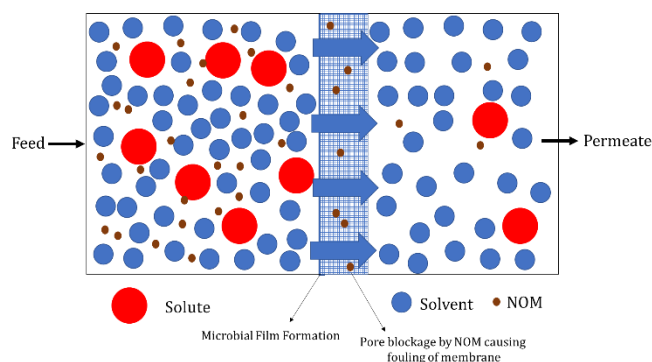


Figure 3: Schematic representation of membrane filtration for the NOM removal and water treatment.

Due to their efficiency, many desalination plants adapted reverse osmosis (RO) techniques to remove NOM and other contaminants like inorganic salts, microbes, and other pollutants [49]. The RO pore sizes are around 1nm and require a pressure of 3×10^3 kPa for the operation, higher than NF. Kimura et al., 2016 [50] reported that solute rejection was high in RO compared with NF; thus, it is evident that RO rejects NOM with both higher and lower molar masses in the influent.

The foulants in the water and the NOM deposit on the filter in membrane filtration and form a cake layer. NOM is the primary food source for micro-organisms; the deposition of NOM on membrane filters enhances microbes' growth, resulting in microbial films that block the membrane's pores,

leading to membrane fouling which reduces the flux of the membrane filtration (Figure 3) [51, 52, 53]. Regular physical and chemical cleaning remove the organic fouling in the membrane filters [54]. Furthermore, membrane techniques' operation cost is very high compared to the other NOM removal methods [55, 56]. Overall, membrane filtration is an effective treatment method for water purification. The high energy and pressure requirements integrated expensive material used for an operation, making them more impractical and cost-prohibitive in several situations where influents' purification is required.

c. Advanced oxidation process (AOPS)

AOPS is considered an alternative for minimizing DBPS and NOM removal. This process involves the generation of radicals with high reactivity (hydroxyl groups) at ambient temperatures.; these compounds are highly reactive and can oxidize the moieties of NOM in the water. Combined catalytic oxidants methods like O₃-UV (ozone-ultraviolet) and H₂O₂-UV (ozone- ultraviolet) are generally used for the generation of hydroxyl groups (OH) (Figure 4). Even though AOPS produces the hydroxyl radicals, the ozone-ultraviolet process produces the hydroxyls (OH) with maximum efficiency [57]. Murray & Parsons, 2004 [58] reported that the Fentons and Photo-Fenton's process removed the NOM from the drinking water more efficiently than AC and UF technologies. Matilainen & Sillanpää, 2010, [59] in a review, stated that the AOP process effectively removes NOM from the drinking water. Nevertheless, these methods involve the operation of susceptible and accurate instruments, enhancing the treatment cost. Thus, these factors make the AOP process less suitable for removing NOM in many cases as these technologies can't be operated in rural, small communities and developing countries.

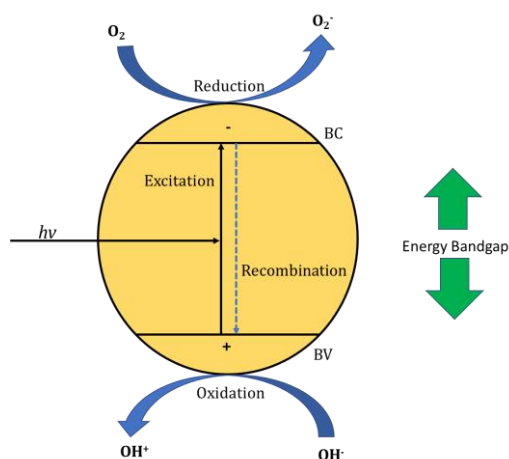


Figure 4: Illustrates the Advanced oxidation process for the removal of NOM.

d. Ballasted Flocculation Separation (BFS)

This process is physicochemical separation involving high-density additives for promoting the heavier flow than usual, which helps settle faster. For instance, a BFS model named Actiflo-turbo was filed tested in France and reported by Aliverti et al., 2011 [60]. This model consists of a micro-sand, which acted as a ballasting agent and enhanced the velocities of the floc for settling and flow rate by reducing the footprints compared to the traditional coagulation techniques and strengthening the BFS flocculation efficiency. Based on the influent characteristics, different types of coagulants, flocculants, and micro sand are much needed to purify and maintain water quality. It is to be noted that these systems on demand can be stepped up and can accommodate the large flows of the influent without any performance losses [61]. The schematic representation of the BFS for potable water treatment is shown in Figure 5. This process effectively eliminated the colour, turbidity, bacteria attached to the suspended particle, and natural organic matter. The dosage might vary based on the application and the influent characteristics. Not consistently, it is evident that a more significant amount of dosage additives might enhance the efficiency of the removal and require to be optimized following influent load and the quality of the water. Thus, the chemical dosage is needed to determine while performing the jar test methods. The significant limitations of the BFS model are a higher requirement of chemical dosing than the conventional treatment process pump problems owing to

ballistic materials and new technology without having a prolonged performance history.

Further development in these technologies is needed by using magnetic powder as ballistic materials instead of micro sand. Magnetic materials generated more compact forms larger flocs than micro sand. Magnetically ballasted flocs are nearly three folds larger than micro sand and six folds more significant than the conventional coagulation techniques. Thus, the suspended particulate matter can be removed and enclosed by the flocs [62].

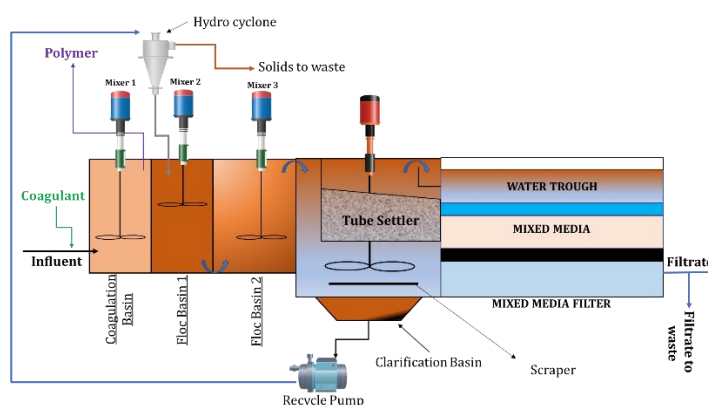


Figure 5: Schematic representation of BFS model for potable water treatment.

e. Magnetic Ion Exchange (MIEX)

This method has strong anion resin for exchange with polyacrylic macroporous structures with larger surface areas than conventional ion exchange for removing contaminants under the magnetic field [64]. MIEX adsorbs the anionic charged NOM by the ion exchange and regenerates quickly from the saline aqueous medium (Figure 6); further, it can remove high amounts of NOM even after several cyclic performances. Additionally, MIEX can be combined with several other potable treatment processes and, combined with the MIEX, found improved NOM removal efficiencies. MIEX techniques can be used as a pretreatment for the conventional coagulation process to enhance efficiency, reducing the coagulation dose and sludge formation. The MIEX combined treatment with coagulation improved the removal efficiencies of NOM to 45% [65]. Jarvis et al., 2008 [66] reported that using MIEX combined coagulation reduced the disinfection byproducts formation and turbidity load during the treatment and enhanced the removal of NOM by forming larger flocs

compared to the traditional coagulation technique. Fearing et al., 2008, reported that combining MIEX-coagulation reduced the dosages of alum and acted as an effective treatment for the solutions consisting of higher organic loads. The experimental results revealed that the molar masses of the dissolved organic carbons are reduced as MIEX models removed the organic carbon and ultraviolet adsorptive materials from the aqueous medium and further enhanced the removal efficacy on various aquaphobic NOM elements.

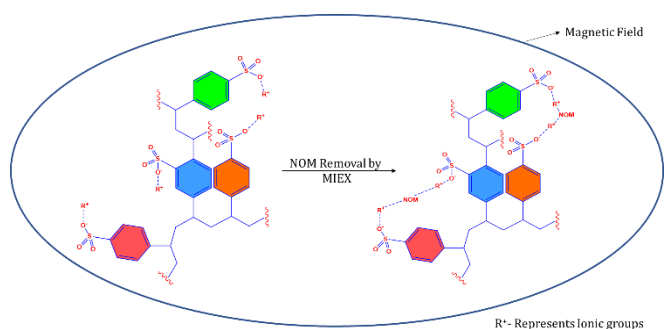


Figure 6: Diagrammatic representation of the MIEX process.

f. Biological treatment

Microbial metabolism was used for the NOM and ammoniacal nitrogen removal from the aqueous mediums in this system. The widely used technique for biological treatment is biofiltration, which is utilized for NOM fraction degradability. Thus, it is essential for controlling the formation of the disinfection by-products, microbial growth and trihalomethane formation potential formation [67]. These filter efficiencies are affected by several factors like water quality, temperature, backwash system and filter design parameters. The optimization process is required to improve the knowledge of this method to know about these factors' relative importance and influences on biofilter operation. Few researchers researched the performances of biofilters efficiencies under various conditions, i.e. changing the medium of the filter, concentrations of the filter bed, contact time and doses of the pre-ozonation. The natural organic matter, total organic carbon, and biological dissolved organic constituents react with the disinfectants products to form oxidized products used to quantify these filter performances [69].

Numerous experiments showed that the filter design that supports the higher biomass enhanced the removal of natural organic matter from the aqueous medium [70, 71]. The growth of the biomass and its activity varies with the temperatures resulting in the removal of biological organic matter. Also, it acts as a limiting factor for the development of microbes in the filters of the effluents and distribution channels [72]. Biofiltration is followed chiefly after flocculation/sedimentation in the filtration units helps remove the particulate matter. An adequately designed biofiltration process enhances the reduction of natural organic matter and prevents the disinfection by-product formation. This process is time-consuming and might release hazardous bacteria on the suspended water after the treatment [73]. The majority of the potable water sources experiences a seasonal change in temperature ranging from 20-30 °C. Variability in the water temperatures might impact the biofilters' biological organic matter and particulate matter. The microbial structures in the biofilters are also affected by these temperature variations. The research assessing the temperature impacts of biofiltration is limited due to the unavailability of long-term full-scale studies; however, filters were usually operated at the ambient temperatures, and seasonal changes were monitored (Figure 7). Sereďyńska-Sobecka et al., 2005 [74] state that total organic carbon and chlorine demand elimination efficiencies are reduced during the winter months; further reported that removal efficiencies of ozonation byproducts like methylglyoxal and glyoxal in the biofiltration process were reduced during the winter compared to summer; this might be attributed to the efficiency of this filter in eliminating the bio-refractory substances and requires pre-oxidation. The most commonly used oxidant for this purpose is ozone, as this process enhances the biodegradability of the organic matter and enhances the effectiveness of the filtration process. The primary advantage of these biofilters is they remove biodegradable components, i.e. undesirable fractions of organic matter [73]. Nevertheless, excess biomass generation might clog the filter's pores, causing heavy head losses. On the contrary, the weak biofilms will not effectively remove the total organic carbon from the aqueous medium.

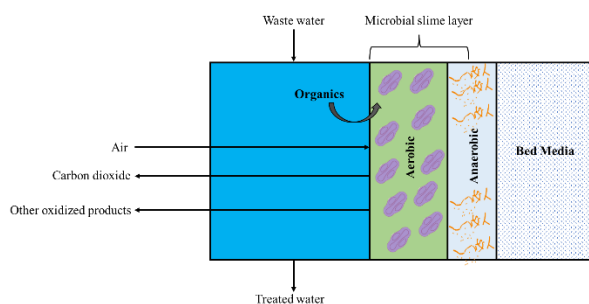


Figure 7: Illustrates the biological treatment process.

Comprehensively according to the available literature, it is evident that membrane technologies are very efficient in removing the pollutants from the aqueous mediums. However, the higher operating pressures, expensive materials involved in the treatment and complex plant structures hindered the usage of these treatment technologies in many situations where water treatment is required. The strengths of conventional coagulation methods are simple, require less energy, low-cost operation, and are easy to establish in rural and small communities residing in remote locations compared to the other treatment technologies. The below sections are considered by us and found essential for applying these technologies for the water treatment.

g. Conventional coagulation process

Coagulation involves repulsive forces of the colloids, with the double layer having an electrical charge which can be reduced by adding the counterions (Figure 8) [59]. In general, NOM in an aqueous medium consists of the negative charge due to the ionization of the functional groups like phenols and carboxyls. The NOM with positive charges reduces the potential, the solubility of the molecules and is aquaphobic, resulting in the aggregation of these compounds [76]. Flocculation enhances the size of the particle from submicroscopic to micro flocs or suspended particulate matter [77]. These formed micro flocs contact the other flocs by mixing; collisions between the flocs result in attaching and forming larger pin flocs. These flocs continuously increase their size by further collisions, and the process can be enhanced by adding organic polymers [78]. The addition of polymers with high molar masses might help reinforce the flocs improve the flocs' weight. The size of the flocs and thickness of the formed sludge during the

coagulation-flocculation process is significant for designing and controlling the parameters [80].

The sedimentation process occurs after the flocculation, and these tanks were designed for separating the supernatant and flocs in the aqueous medium. Nevertheless, few operations don't require the sedimentation tanks as the particles were trapped during the filtration process. Sedimentation tanks store temporary water flows to separate the flocs and suspended matter from the aqueous medium. Different tank designs were used to rapidly settle the flocs [81]. Sedimentation of flocs by the gravitational pulls are the most commonly used process than centrifugal sedimentation [82]. The sedimentation process, like laminar clarifiers, plate and tube settlers, and dissolved air floatation, is employed when the particulate matter has lower specific gravity than water to enhance the settling [83].

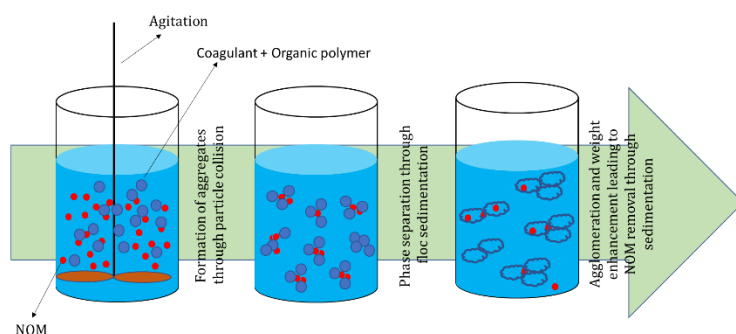


Figure 8: Illustrative representation of the conventional coagulation process.

3. THEORY OF COAGULATION

This process destabilizes and agglomerates colloidal contaminants to larger particles (form flocs) separated by the physicochemical process like rapid/slow mixing, sedimentation, and filtration. Colloidal pollutants in the water include microbes, inorganic/organic solids, dye producing substances and natural organic matter. The conventional coagulation process destabilizes the contaminants in the aqueous medium by the charge neutralization after the coagulant dosing. In contrast, they occur to form nano-sized particles in the flocculation process. Even though the initial process of the flocculation and coagulation happens to occur quickly in practical applicability, we found slight

dissimilarities. Dempsey and O'Melia, 1984 [84] reported that these mechanisms could achieve the colloidal particles in the coagulation process by the following: Compression of double-layer, charge-neutralization, sweep-coagulation and interparticle bridging. This method destabilizes the particulate matter by adding coagulant (electrolyte) to form suspended solids. Under appropriate conditions, the colloids exhibit higher stability where they cannot aggregate together due to thick double layer formation. The addition of the electrolyte alters the double layer's ionic strengths around the collides, resulting in the shape of destabilized colloid [85]. This process was found to be essential for destabilizing the colloids.

The literature shows that coagulation and integration of these techniques with the advanced treatment options are efficient in treating and removing natural organic matter from the aqueous medium. Thus, the authors propose integration techniques to fill the literature gaps for future research. Karnena et al., 2021 [86] reviewed the NOM characteristics and showed how the cations interact with the natural organic matter. Later Karnena et al., 2022 [87] removed the NOM from the wastewater using the coagulation techniques and stated that scaling up of studies are required for moving these techniques to be adopted by the water industry [88-90].

4. CONCLUSIONS

The online version of the volume will be available in LNCS Online. Members of institutes subscribing to the Lecture Notes in Computer Science series have access to all the pdfs of all the online publications. Non-subscribers can only read as far as the abstracts. If they try to go beyond this point, they are automatically asked, whether they would like to order the pdf, and are given instructions as to how to do so. NOM is a complex compound with different organic materials in most natural waters. NOM is a significant factor affecting water quality worldwide. Removing NOM from the aqueous medium is a tedious process. Even though there are many techniques available for removing the organic matter and new technologies are advancing like MOF, Carbon nanotubes etc., based on the cost-benefit analysis, the coagulation techniques using the coagulants are found to be effective in terms of economy for developing countries like India. Further, there is a

need for researching the integration of techniques with coagulation to fill the gaps in the literature.

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