- 1 The approaches and prospects for natural organic matter-derived disinfection
- 2 byproducts control by carbon-based materials in water disinfection progresses
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#### 9 ABSTRACT

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The formation of unintended disinfection by-products (DBPs) mainly results from the reaction between the widespread natural organic matter (NOM) and disinfectants in source water and wastewater disinfection progresses. Although waterborne diseases are controlled by disinfection, the generated DBPs show a high risk to human health accompanying disinfection, and receive considerable attention. Carbon materials are widely used in different stages of water treatment progress. They show good adsorption and catalytic performance, and the low price makes them competitive in engineering application. Moreover, carbon materials show high ability in decreasing the concentration of NOM, intermediates of NOM-derived DBPs and DBPs themselves during the different stages of water treatment. This review aims to highlight the removal of NOM and NOM-derived DBPs through the application of carbon materials, and to explain the conversion and potential risks of NOM in water disinfection. The operating forms of different carbon materials including adsorbents, filling materials of membrane and catalysts of advance oxidation progresses (AOPs) are summarized systematically. Moreover, new DBP precursors introduced by the application of carbon materials are discussed. Lastly, perspectives on the challenges for DBPs control in emerging conditions such as stormwater reuse, microplastics pollution, or water treatment of public health emergency, are proposed for further development.

- **Keywords:** Disinfection by-products; Natural organic matter; Carbon materials;
- 29 Disinfection

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#### 1. Introduction

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Before the end of the twentieth century, it is widely believed that environmental disasters like endemic waterborne disease are more dangerous than chemical disasters (Li and Mitch 2018, von Gunten 2018). Chlorination disinfection is regarded as one of the greatest inventions in the world since it is helpful for water resource recovery and pathogenic bacteria extermination. After ensuring the basic sanitation and safety of water, more attention has been paid to the health risks of water accompanying disinfection like disinfection by-products (DBPs) (Fu et al. 2017). Rook first detected trihalomethanes (THMs), a representative class of DBPs, in chlorinated disinfection water in 1974, and the united states environmental protection agency (USEPA) investigated the prevalence of chloroform in chlorinated water bodies in 1976 (Liu et al. 2020a, Rook 1974). The types and contents of DBPs generated by different disinfection processes are highly uncertain. It was found that some DBPs are cytotoxic, genotoxic, mutagenic, carcinogenic, and developmentally toxic even at very low concentrations (Yang et al. 2016). Therefore, DBPs indeed pose a risk to the ecological system and human health. And under public health emergency of international concern (PHEIC) like Corona Virus Disease 2019 (COVID-19), there is a drastic increase in disinfectant which result in high DBPs formation. DBPs are mainly generated by the interaction between disinfectants and organic matter through oxidation, addition and substitution reactions in water treatment (Liu et al. 2020a). In natural and social water cycles, organic carbon precursors can contact

with disinfectants under different circumstances. Anthropogenic micropollutants in water are contributed to the production of DBPs precursors and may form DBPs in the disinfection progress. In most cases, natural organic matter (NOM), or dissolved organic matter (DOM) caused by NOM, is widely recognized as the dominating source of DBPs (Ike et al. 2019b, Jiang et al. 2020, Li and Mitch 2018, von Gunten 2018). The presence of NOM in source water and wastewater generates many problems in water treatment progresses such as taste and odor problems which have a negative impact on water quality (Brezinski and Gorczyca 2019, Gusain et al. 2020). Besides, the existence of NOM in influent solution may promote biological growth in the distribution system and increase the adsorption of other hazardous substances like heavy metal (Bhatnagar and Sillanpaa 2017). More importantly, low NOM removal capacity in the previous progress before disinfecting reactor such as grilling and sedimentation increases the dose requirement of disinfectant or other chemical reagents like coagulant, which results in increasing sludge production and formation potential of harmful DBPs (Cheng et al. 2018, Cruz-Silva et al. 2019, von Gunten 2018). In general, the ultimate purpose of NOM removal in water treatment is alleviating the formation of DBPs, thereby achieving high water quality and ensuring water safety. For drinking water or landscape water, the removal of NOM contributes to decreasing colority and turbidity additionally. To sum up, in this article, the control and removal of NOM and NOMderived DBPs will be well discussed together.

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Combined with the current processes and technologies for controlling DBPs in

water treatment, the main methods of DBPs control and removal can be concluded into the following aspects. The first method is reducing DBPs precursors especially the concentration of NOM in the source water to control DBPs formation. The second method is updating new and green disinfectants or new disinfection techniques, such as ultraviolet disinfection or ultrasonic disinfection, in the disinfection process to replace the existing chlorine disinfection techniques and then decrease the formation potential of DBPs. And the third one is adding subsequent progress such as adsorption technology using carbon materials which can reduce the amount of DBPs that have already been produced. In fact, the revolution of disinfection methods cannot eliminate DBPs formation (Ding et al. 2019, Erdem et al. 2020, Jiang et al. 2017). As for the application and popularization of new disinfection technologies in water treatment, not only holistically economic or technical assessments are required, but also relevant facilities and equipment need to be adjusted or improved. Therefore, it is more feasible to remove and control NOM and NOM-derived DBPs through other methods or to enhance efficiency of existing progress, such as adsorption, filtration and advanced oxidation progresses (AOPs). Carbon materials are widely used in pollution abatement and water resource recovery, and the diversity of carbon materials shows the possibility for different

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Carbon materials are widely used in pollution abatement and water resource recovery, and the diversity of carbon materials shows the possibility for different circumstances and applications (Chen et al. 2020, Ye et al. 2019, Yu et al. 2020). With these advantages, such as abundant pore structures, high specific surface areas, multiple functional groups or even graphitized structure and heteroatom doping favorable for

electron conduction, carbon materials are effective for NOM and NOM-derived DBPs removal in different methods as mentioned above. In the previous studies, carbon materials showed the inhibition of DBPs formation during disinfection (Erdem et al. 2020, Jiang et al. 2018, Zhang et al. 2019a). Therefore, it is necessary to give a timely review of the progress of different kinds of carbon materials used for NOM and NOM-derived DBPs removal and control. In this review, the chief objective is to highlight recent advances of carbon materials used for controlling NOM and NOM-derived DBPs, and the following content will be included: (1) different approaches of carbon materials for NOM and NOM-derived DBPs control; (2) special attention for carbon materials as new sources of DBPs precursor in relative progress; (3) DBPs control under different water requirements; and (4) promoting carbon materials for NOM and NOM-derived DBPs control in practical engineering applications.

### 2. The conversion and potential risk of NOM in water disinfection

## 2.1. The potential of NOM as DBPs precursors

NOM, the most important and widely accepted DBPs precursor, is a complex mixture with both amphipathic and amphoteric properties and structures (Fig. 1a and 1b). The different components of NOM show the potential as DBPs under different classifications. DBPs in water are usually classified in several ways: (1) N-DBPs and C-DBPs based on the presence of nitrogen or not; (2) non-halogenated DBPs and halogenated DBPs (X-DBPs, X=Cl, Br, I); and (3) aliphatic DBPs and aromatic DBPs

(Liu et al. 2020a). Phenolic structures with functional groups such as hydroxyl and carboxyl group in humic acids (HA) and fulvic acids (FA) are proved to be the main precursors of THMs (Ding et al. 2019). The THMs and haloacetic acids (HAAs) formation potential of hydrophilic and low molecule weight (MW) in NOM are higher than that of hydrophobic and high MW during disinfection (Hua and Reckhow 2007). Different results were obtained from Li et al. proposed that hydrophobic fraction of NOM including nitrogenous organic matter like protein-like materials showed the formation potential of N-DBPs and heterocyclic DBPs during disinfection (Li et al. 2020). Besides, the potential formation of halogen-containing DBPs from hydrophobic fraction was much higher than that of hydrophilic fraction during disinfection (Li et al. 2017). Different types of DBPs might convert into each other in the disinfection process (Fig. 1c). Recently, aromatic DBPs receive more attention because their higher toxicity than commonly known aliphatic DBPs and the possible precursor sources of other DBPs. High MW fraction of NOM was treated as the precursors of aromatic DBPs and could be converted into aromatic DBPs via direct or indirect pathways (Jiang et al. 2020). For example, ring opening and side chain cleavage are the decomposed mechanisms of transforming aromatic DBPs into regulated aliphatic DBPs (Liu et al. 2020a). Moreover, Hua et al. established the connection between UV absorbance and the yields of different DBPs. Higher UV absorbance resulted from C-rich NOM showed high production of C-DBPs, while low levels of C-DBPs and N-DBPs were produced from NOM with O-rich function groups which exhibited low UV absorbance (Hua et

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## 2.2. The influence of co-existing ions in NOM-derived DBPs formation

The formation of NOM-derived DBPs is affected by co-existing ions in complex water disinfection (Fig. 1d). In general, Br<sup>-</sup>, as a common and trace ion in natural water, is usually considered as co-existing ion in the disinfection byproducts formation potential (DBPFP). According to the species and molecular composition of DBPs, Br may affect the potential and toxicity of generated DBPs (Gao et al. 2019, Qian et al. 2020). Hua et al. evaluated the bromine substitution factors of DBPs during chlorination disinfection, and Br-DBPs showed higher stability to pH than Cl-DBPs which means that Br-DBPs may be generated preferably in the presence of Br (Hua and Reckhow 2012). But additional condition needs to be considered when the treated water showed more distinct characteristics. The landfill leachate contains refractory organic matters (e.g., HA, FA) as well as various hazardous matters such as, inorganic ions (e.g., Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, Cl<sup>-</sup>, Br<sup>-</sup>), and heavy metals. Considering the high concentration of NOM and halogen ions in landfill leachate, it is reported that the formation of THMs was more than 100 times higher than that of in general wastewater during disinfection treatment. It may be due to the incomplete oxidation of DBPs resulted in their accumulation (Iskander et al. 2020). In addition, during the co-treatment of landfill leachate and general wastewater, organic carbon and nitrogen of high concentration in untreated water may enhance DBPs formation after disinfection (Li and Mitch 2018). Iskander et al. investigated that DBP formation occurred in landfill leachate during Fenton oxidation treatment. It is found that in situ generated chlorine and bromine species is the key factor for DBPs formation (<u>Iskander et al. 2020</u>).

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Kim reviewed DBPs formation during seawater desalination (Kim et al. 2015). It was observed that the enhancement of Br-DBPs and I-DBPs species in bromide/iodide rich water and a shift formation from Cl-DBPs to Br-DBPs was proved with the approximately constant concentration of Cl-DBPs (Ged and Boyer 2014). And Ged et al. investigated DBPFP during disinfection in the case of seawater intrusion in which condition a certain concentration of halogen ions such as Cl-, Br- and I- were introduced. The formed THMs concentration was doubled with the 2% seawater addition and above the regulatory levels (Ged and Boyer 2014). Hua et al. proved that increasing concentration of Br exacerbated the formation of the monitored DBPs (Hua et al. 2006). And the cytotoxicity, genotoxicity and developmental toxicity of Br-DBPs were higher than the chlorinated species. Moreover, chemical disinfectants are commonly used in pretreatment technology for inhibiting bacterial growth and improving subsequent processing performance in seawater desalination. When desalinated water with high concentration of halogen ions was mixed with other source waters with high NOM concentration, the formation potential of Br-DBPs or I-DBPs would significantly increase in distribution systems (Kim et al. 2015).

#### 3. The roles of carbon materials for NOM and NOM-derived DBPs control

#### 3.1. Adsorbent

Adsorption is the most basic and commonly used method employed in water purification (shown in Fig. 2) (Gusain et al. 2020). The relationship between the pore-size distribution or pore volume of carbon materials and the molecular weight (MW) of adsorbed target substances such as organic matter determines the adsorption effectiveness for NOM and NOM-derived DBPs. Besides, functional groups on the surface of carbon materials are also important factors affecting adsorption (Lucaci et al. 2019). Carbon materials with the advantages of strong adsorption capacity and excellent recycling ability are economically feasible and environmentally friendly which are necessary for the efficient adsorbent. In this section, the roles of carbon materials as adsorbents for NOM and NOM-derived DBPs control are discussed through several basic properties of carbon materials (Fig. 3a and Table 1).

### 3.1.1. Pore sizes and distribution

The pore structure and pore sizes distribution characteristics of carbon materials are important for NOM and NOM-derived DBPs removal. Carbon materials prepared by pyrolysis, such as activated carbon (AC) and biochar (BC), present more abundant pore distribution (Wang et al. 2019a, Zhang et al. 2019c). For example, pores of AC are divided into micropores (<2 nm, in diameter), mesopores (2–50 nm), and macropores (>50 nm). The average size of NOM ranges from 1 to 5 nm (Zhang et al. 2019a). Actually, the adsorption of NOM primarily occurs in mesopores and large micropores

(Lu et al. 2020). Tailored mesoporous biochar, produced from pinecone biomass and modified with NaOH, showed enhanced removal capacity for NOM compared to the commercial powder activated carbon (PAC), in a wide range of pH (Yazdani et al. 2019). Erdem et al. also found that the major contribution of adsorption for halogenated DBPs precursors was mesoporous region in carbon materials (Erdem et al. 2020). And the strong relationship between DBPs removal and GAC with a 5-10 nm pore range were revealed by Golea et al. (Golea et al. 2020). Zhang et al. also revealed that mesopores in AC was benefit for NOM irreversible adsorption through  $\pi$ - $\pi$  bonding and hydrophobic effects (Zhang et al. 2019a). Moreover, the existence of both micro- and mesopores of granular active carbon (GAC) applied to biological activated carbon (BAC) filters, not only high adsorption capacity is guaranteed sufficiently but also bacterial adhesion are allowed by macropores (Lu et al. 2020). The surface areas and the volume of micropores and mesopores of GAC are much higher when compared to biochar pyrolyzed at 300 °C. The measured DBPs concentrations treated with GAC are well below that of biochar. Biochar produced at high temperature showed increased surface areas, pore volumes and change in pore size distribution. The volume of micropores increased from 0.003 to 0.167 cm<sup>3</sup>/g but the diameters of different biochar from 2 nm to 300 nm were just slightly changed. The formation of trichloromethane (TCM), one of THMs, was decreased from 77.4 to 8 µg/L with the increasing production temperature from 300 to 700 °C (Zhang et al. 2019a). Therefore, the pore size and its distribution of carbon materials can influence the NOM-derived DBPs

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formation through the adsorption of NOM. In general, the abundant pore sizes of carbon materials which are similar in diameters or slightly larger than the components of NOM are positive for NOM adsorption. And the function of residual pores can be neglected. Therefore, carbon materials with abundant and tailored mesoporous pores showed superiority as adsorbent in NOM and NOM-derived DBPs removal.

## 3.1.2. Different functional groups

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Considering the similar properties between NOM and carbon materials in functional groups and aromatic rings, carbon materials possibly show directly influence on DBPs formation. In the study of Zhang et al., the formation of DBPs was significantly inhibited with the existence of AC. They concluded that the interaction between the oxygen-containing functional groups (like -OH and -COOH) on AC and NOM affects the DBPs formation via hydrogen bonding which can increase the efficiency of NOM removal (Zhang et al. 2019a). NaOH modified biochar with increasing aromatic surface area and alkali-generated hydroxyl groups result in higher adsorption affinity of NOM with low molecular when compared with pure carbon materials. More than 80% of total NOM from lake water were removed by modified biochar (Yazdani et al. 2019). Truong et al. explored that biochar with higher specific surface areas and ample functional groups particularly enhanced NOM removal via adsorption. Surface modification of biochar can change the surface charge and pore size distribution even the type of functional groups of adsorbents. In this study, the maximum capacity of biochar for NOM is about  $80.65 \pm 4.16$  mg C/g, and electrostatic

attraction, hydrogen bonding, and  $\pi$ - $\pi$  interaction are the operative mechanisms of adsorption (Truong et al. 2020).

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Compared to amorphous carbon materials such as AC and BC, CNTs are typical single-dimensional nanomaterial with specific sp<sup>2</sup> hybridized surface, defects and low oxygen level (Song et al. 2020). CNTs with more functional groups can be obtained through modification, and related research showed that the variety of functional groups and surface characteristics of the CNTs are important for adsorption (Kumar et al. 2016). There is a negative correlation between adsorption capacity of carbon material and the number of carboxylic acidity of the NOM (Ateia et al. 2017). Zhang et al. used functionalized multiwall carbon nanotubes (MWCNTs) produced by hydroxylation and carboxylation to change the numbers and types of functional groups of MWCNTs, and tried to evaluate the independent influences of the different functional groups of MWCNTs on DBP formation. The adsorption via hydrogen bonding between Ocontaining function groups (such as -OH or -COOH) may weaken the reactivity of C atom in the middle of the -OH groups, and then suppressed DBPs formation. Besides, modified MWCNTs with numerous O-containing functional groups would have a remarkable effect on decreasing the concentrations of HA which has many Ocontaining functional groups (Zhang et al. 2019a). Wang et al. also found that -OH modification showed no significant effect on MWCNTs for inhibiting TCM formation. The possible reason of no enhancing adsorption ability of -OH modification might be the smaller pore structures of MWCNTs, so the majority of NOM can only be adsorbed

on the outer surface of the OH-MWCNTs (Wang et al. 2020a).

Besides, Jamil et al. showed that GAC was proved to remove organic matter and micro-organic pollutants at the same time from reverse osmosis both in batch and column experiments. The results suggested the application potential of GAC in complicated water treatment (<u>Jamil et al. 2019</u>). In conclusion, the addition of carbon materials is positive for NOM and DOM-derived DBPs control during disinfection in most research (<u>Zhang et al. 2019a</u>).

### 3.1.3. The selectivity of carbon materials towards NOM

NOM is a complex mixture of organic compounds. It mainly includes humic substances such as HA, FA and humins, as well as non-humic substances such as carboxylic acids, carbohydrates, sugars and amino acids (proteins). Humic substances are comprised of hydrophilic fraction of NOM and mainly composed of high molecular weight (MW) components, which contributed to more than 50% of total organic carbon (TOC) in raw water (Korotta-Gamage and Sathasivan 2017). The characteristics of hydrophilic organic matter in NOM are relatively composed of low MW fractions and show low charge density.

The MW of NOM is the most important influence factor for its removal. Lee et al. observed that biochar can remove NOM with all molecular weight and adsorb preferably NOM with high molecular weight. The prepared biochar can removal 90% of NOM at 200 mg-C/L solution with an estimated capacity of 0.0064 mg-DOC/mg-C. And the key impact on adsorption progress is the interaction between active sites of

biochar and NOM (Lee et al. 2018a). Ateia et al. proposed that HA with molecular weight range from 0.5 to 2 kDa prefer to be adsorbed on CNTs. And FA in the molecular weight range of 1–3 kDa was preferentially adsorbed on CNTs. However, the smallest featured fractions (MW < 0.4 kDa) in all samples were not adsorbed in the study. The result indicated that the main interaction forces between CNTs and NOM are hydrophobic interactions and  $\pi$ - $\pi$  attractions. NOM of different derivations and decomposition degree can also affect the adsorption capacity of CNTs (Ateia et al. 2017). Shimizu et al. investigated that CNTs showed the potential for adsorbing high MW fractions of NOM selectively, but the smallest fractions of NOM were not removed (Shimizu et al. 2018). Cai et al. presented that graphene powder with the high surface area can rapidly remove NOM from water and prefer to adsorption aromatic, high molecular weight fractions of NOM (Cai et al. 2015). The source of NOM is another factor needed to be concerned. Fluorescence index (FI) and freshness index (BIX) based on excitation-emission matrix (EEM) showed significant difference between terrestrial derived and microbial derived NOM. The study of Ateia et al. indicated that terrestrial-original and decomposed NOM is preferentially adsorbed on CNTs compared to microbial-derived and freshly produced NOM (Ateia et al. 2017), and Shimizu et al. proposed that the decomposition of NOM is more easily to occur on the surface CNTs. They also explained that AC tends to adsorb higher molecular weight and freshly produced fraction of NOM. And microbialderived NOM was preferential adsorption on AC. There were obvious differences in

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the preferential adsorption of different carbon material for NOM with different properties and sources. It is reported that terrestrial derived NOM are rich in aromatic compounds and high in MW fractions than microbial derived NOM (Shimizu et al. 2018).

Jiang et al. investigated that the control of intermediate aromatic halogenated DBPs can well decrease the formation of commonly known DBPs. They explained that GAC can control halogenated DBPs formation with aromatic intermediates adsorption. The removal ability of aromatic halogenated DBPs was significantly higher than aliphatic halogenated DBPs in GAC application. It might because the difference of pKa values of aromatic halogenated DBPs and aliphatic halogenated DBPs, which are in the ranges of 1.96–6.59 and 0.09–2.65, respectively. So, the aromatic halogenated DBPs showed low degree of ionization in conventional water treatment, and the neutral forms are easier to adsorbed onto GAC (Jiang et al. 2018). The removal of total organic halogen (TOX), THMs, and HAAs were highly enhanced when adsorbing NOM by pretreatment. Furthermore, GAC was found to adsorb more than 50% of the formed nonhalogenated aromatic DBPs (Jiang et al. 2020).

In summary, adsorption is the most common way to remove DBPs precursors before DBPs formation in water treatment processes in the current application, but it is difficult for the specific carbon materials to efficiently remove the hydrophilic and hydrophobic components of NOM simultaneously in a single progress or technologies. Therefore, regulating the functional groups of carbon materials, using sustainable

carbon materials, or combining with other technologies (such as flocculation, sedimentation and biological treatment technology) to realize the efficient adsorption of all components of DBPs precursor is the focus of current research.

#### 3.2. Materials in membrane

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Membrane filtration is regarded as the most promising and extensively used technology for water purification in advance treatment (shown in Fig. 2) (Ihsanullah 2019). Membrane fouling is the major barrier of membrane technology application, and it may lead to highly energy consumption and shorten lifespan of membranes. Carbon materials, which can be used as not only direct filters but also fillers, are promising in developing novel and advanced membranes (Fig. 3b). Besides, the decreasing dispersibility of carbon materials can be obtained through the membrane preparation (Ghaffar et al. 2018). Some properties of traditional membranes such as pore size distribution, porosity, antifouling behaviors, water permeability and stability can be improved by the additional of different carbon materials (Wu et al. 2020). NOM is regarded as one of the major membranes fouling, and it can cause both severe reversible and irreversible membrane fouling (Tian et al. 2018, Wang et al. 2019b). According to the structure of membrane, carbon materials can be used as filling materials or/and modification materials and discussed in the following section (detailed in Table 2).

# 3.2.1. Filling materials of membrane

Graphene, graphene oxide (GO) and functionalized GO have been widely used for the preparation of membranes because they can improve surface areas, high chemical and mechanical stabilities of membranes, and no additional recycling process is required. Cohen-Tanugi and Grossman et al. firstly proposed graphene-based membranes applied to water treatment progresses and desalination plants (Cohen-Tanugi and Grossman 2012). The superiority of asymmetric GO membrane assembling a dense GO barrier layer on the back side by layer assemblage was explained by Hu et al. The affinity of GO membrane to organic foulants is about 4 to 5 times higher than the PA membrane, but the presented water flux was less affected by foulant adsorption, because the water molecular mainly entered the oxidized edges of GO membrane. And organic foulants are adsorbed on the basal plane of GO nanosheets rather than trapped inside the porous structure of membrane which is main reason of irreversible membrane fouling (Hu et al. 2016). You et al. investigated that GO membrane can reject more than 99.9% NOM, and membrane fouling occurred in 6.0 h with the constant water flux of 65 L m<sup>-2</sup>/(h·bar) (You et al. 2018). Lee et al. revealed that aromatic molecules preferentially adsorbed to the GO and the progress is well affected by pH and available sites of GO (Lee et al. 2015). Maqbool et al. concluded that hydrophobic interactions and  $\pi$ - $\pi$  interactions were the main mechanism for fluorescent NOM removal during non-oxidative interactions (Magbool et al. 2018). However, by introducing functional groups at different locations, the main mechanism for NOM removal may change. In the study of Qin et al., they proposed that it is easier to anchor HA via functional groups of the GO surface than stacked onto graphene surfaces via  $\pi$ - $\pi$  interaction. Modification can change the hydrophilic of the membrane surface, and then influence adsorption

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kinetics and the proportion of reversible adsorption of HA (Qin et al. 2020). Besides, Xia et al. prepared ultrathin GO membrane cross-linked by diamine monomers to remove NOM for the first time. C–N covalent bonds formed in the membrane can effectively suppress the interlayer spacing and stretching (Xia et al. 2015a).

#### 3.2.2. Modified materials of membrane

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In the study of Hwang et al., AC is used for PPSU/PEI/PEG composite membranes modification. The filtration flux and permeability of membranes is improved with the increased AC addition. The removal efficiency of HA is about 80% (Hwang et al. 2013). Ghaffar et al. synthesized biochar-PVdF composites membranes through thermal phase inversion method. The presence of biochar contributed to the enhanced antifouling of membrane. And the active sites on aromatic structure of biochar are responsible for the enhancement of NOM adsorption. The adsorption and sieving capabilities of the biochar composite membranes can be regenerated after simple physical treatment (Ghaffar et al. 2018). However, in most cases, adsorption is not the main mechanism of carbon material composite membrane for NOM removal. Modification by carbon materials can reduce the directly interaction of NOM and surface of membrane, and then decrease membrane fouling. The addition of carbon materials might change the surface charge, roughness, hydrophilicity or pore size distribution of membrane to reduce membrane fouling (Bai et al. 2015, Qin et al. 2020). Bai et al. used functionalized MWCNTs to prepare membranes. Modified membrane showed stronger resistance to NOM fouling with only

a slight sacrifice in membrane flux (Bai et al. 2015). And the combination of CNTs and polyamide (PA) membranes change the roughness and mobility of NOM at molecule scale on the surface of membranes. So the irreversible adsorption of NOM is decreased (Cruz-Silva et al. 2019). The antifouling properties of graphene-based membranes are important indicators of evaluation. Different modification methods and composite technology make the graphene-based membrane more competitive. Lv et al. indicated that massive oxygencontaining groups in GO-CNC/PVDF composite on account of low irreversible fouling of GO membrane (Fig. 4b) (Lv et al. 2018). Zhao et al. reported that 2D GO sheets membranes modified by 0D GO quantum dots (GO QDs) showed 3-4 times higher water permeability than pure GO membrane. And the rejection of GO or GO QDs/GO membranes for HA was more than 99% (Fig. 4a) (Zhao et al. 2019). Liu et al. found that GO membranes modified by novel cation (Al<sup>3+</sup> or Fe<sup>3+</sup>) showed enhanced stability. Moreover, the removal capacity of trivalent metal cation cross-linked graphene oxide membranes for NOM is 99.6% in HA solution and 70-80% in actual water (Liu et al. 2017). Graphene-based materials used for other membranes modification also showed good performance. Gu et al. found that permeability and anti-organic fouling properties of the GO QDs chemically grafted onto ceramic microfiltration membranes were improved. And the irreversible fouling of membrane is decreased (Fig. 4c) (Gu et al. 2020). Algamdi et al. fabricated graphene oxide incorporated polyethersulfone hybrid

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ultrafiltration membranes. The result revealed that the recyclability and antifouling capabilities of the membrane were enhanced by GO addition. With the increasing content of GO (from 1-5%), the irreversible fouling ratio values of membrane decreased obviously from 22.3 to 2.4% (Algamdi et al. 2019). More importantly, rejection ability of membranes for NOM was improved prominently with the existence of GO (Fig. 4d) (Algamdi et al. 2019, Chu et al. 2017). And Xia et al. prepared GO modified polyamide thin-film composite membranes used in water treatment. With the increasing GO loadings (from 0 to 0.012 wt%), the removal rates of NOM became higher. The addition of GO can change hydrophilicity, surface roughness and surface charge of membranes (Xia et al. 2015b), and all the transformation of membranes after modification is positive for NOM rejection. Moreover, Xia and Ni proposed that the addition of GO improved antifouling properties of PVDF membranes, which increased the pore density of the top-layer and macrovoids in the sub-layer and smoother the membrane surface. The systemized PVDF/GO membrane showed better removal for NOM of low MW fractions than unmodified membrane (Xia and Ni 2015). In summary, carbon materials with regular microstructure, such as graphene, graphene oxide or carbon nanotube, have been widely used for the preparation of carbon-based membranes than amorphous carbon. The chosen carbon materials for membrane preparation generally used to change the smoothness, hydrophilicity, and pore size of membrane surface, which are positive for membrane application in water treatment. Membranes made by graphene-based materials can be used for NOM

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management in conventional water treatment plants (You et al. 2018), and almost complete removal for NOM is achieved by graphene-based membrane in synthetic water. But in real water treatment with complex conditions, the removal capacity and efficiency of graphene-based materials for NOM may decrease because of membrane fouling by competitive adsorption of other substances (Gu et al. 2020). However, graphene-based materials still show the potential for NOM removal and DBPs control as well. Besides, carbon materials produced from cheap and sustainable raw materials with regular morphology and abundant functional groups are also deserve to explore.

## 3.3. Catalyst in AOPs

Chemical oxidants have been used in water treatment for more than a century, not only as disinfectants, but also to reduce various pollutants (von Gunten 2018). Free radical of high standard reduction potential is the key substance in AOPs. The advantage of AOPs is to achieve complete mineralization for most organic pollutants or improve the biodegradability of the decomposition component. Moreover, AOPs eliminate the diversion of NOM and DBPs from one phase to another which is regarded as more environmentally friendly than adsorption. In previous researches, carbon materials, as the widely studied efficient catalysts in AOPs showed great potential for pollutant removal and environmental remediation (Ye et al. 2020b, Yu et al. 2020). Besides, a growing number of researches are published about the control and removal of NOM and DBPs which are newly emerging and target contaminants especially in drinking water treatment (Bhatnagar and Sillanpaa 2017). And carbon material based-

AOPs were regarded as efficient methods to remove NOM from water supply system and then alleviate the formation of DBPs (Fig. 2 and Table 3). HA was used as a typical NOM to simulate the situation of actual water, which usually regarded as a scavenging effect to free radical. The interaction between DOM and three free radical species was studied by Varanasi et al. (Varanasi et al. 2018). Aliphatic components, aromatic and olefinic moieties of DOM were influenced and transferred by free radical. And free radicals can lead to the conversion of NOM to low molecular weight and result in the improvement of membrane flux. Generally, AOPs, as membrane pretreatment, showed alleviation of membrane fouling because of the reducing accumulation of high molecule weight organic matter on membrane surface (Fig. 3c) (Cheng et al. 2018). Moreover, lke et al. observed that unactivated oxidant rather than free radical which is commonly held is principally contributed the generation of DBPs during AOPs (Ike et al. 2019a). Therefore, the dosages of oxidant and catalyst should be considered cautiously.

### 3.3.1. Photocatalytic degradation

Photocatalytic oxidation is a highly efficient and eco-friendly technology that has got increasing attentions (Fig. 5a) (Truong et al. 2019). The advantages of photocatalysis are making good use of renewable resources and only introducing relatively simple or clean substance. Reactive oxygen species (ROS) scavenging and competitive adsorption by NOM is the critical inhibition mechanism in photocatalysis. The higher molecular weight fraction of NOM prone to be adsorbed on the catalyst results in decreasing photocatalysis efficiency. Besides, NOM has been considered as a

prominent absorber of light and sensitizer that can produce reactive intermediates. Kohantorabi et al. found that NOM can produce <sup>3</sup>NOM\* and <sup>1</sup>O<sub>2</sub> under solar irradiation which can be used as important oxidants for bacteria inactivation (Kohantorabi et al. 2019). Drosos et al. found that low NOM concentration enhanced contaminant photocatalysis through creating attraction sites on the catalyst (Drosos et al. 2015). The transformation of NOM indicated the degradation potential of NOM in water treatment. High-energy photocatalysis is not feasible and economical although it controls the DBPs production. Therefore, synthesis high-efficiency and low-cost carbon-based material for photocatalysis catalysts is of importance (Liang et al. 2021, Yang et al. 2020d). Truong et al. synthesized a novel catalyst based on a hybrid composite of zincbismuth oxides and g-C<sub>3</sub>N<sub>4</sub> (ZBO-CN), which showed high NOM removal capacity even in low-intensity visible light irradiation. The result pointed out that the dominant mechanism of NOM removal is direct oxidation on the catalyst. High molecular weight or more condensed structured NOM because of the greater adsorption towards catalyst's surface is vulnerably attacked during catalysis oxidation progresses (Truong et al. 2019). g-C<sub>3</sub>N<sub>4</sub> was used by to investigate the photocatalytic degradation of two typical DBPs, THMs and haloacetonitriles (HANs), under visible light condition. The results showed that holes generated by materials played a dominant role for THMs and HANs degradation. The main degradation pathway of THMs achieved by hydrogen abstraction or/and dehalogenation, and the degradation of HANs mainly due to the breakage of C-C bonds (Chang et al. 2019). There are certain differences in

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photosensitivity between different DBPs, so more research is needed on the status of diverse DBPs in actual water. However, Mayer et al proposed that the limitation of photocatalysis for DBPs removal (Mayer et al. 2014). Incomplete mineralization of DBPs precursors because of low-energy and limited photocatalysis may exacerbate the DBPs formation.

#### 3.3.2. Fenton-like oxidation

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H<sub>2</sub>O<sub>2</sub> is a common oxidant for AOPs. However, when compared to the acidic and alkaline conditions, the oxidation capability of H<sub>2</sub>O<sub>2</sub> at neutral pH is lower. Considering actual water treatment, it is necessary for synthesizing high-efficiency catalysts used in the natural environment. Yang et al. synthesized carbon-doped Fe<sub>3</sub>O<sub>4</sub> peroxidase-like nanozyme (CFPN) and used it for NOM control (Fig. 5b) (Yang et al. 2020b). An acidic microenvironment can be generated by CFPN because of oxygen-containing groups on the carbon structure, which allowed hydroxyl radical produced by H<sub>2</sub>O<sub>2</sub> in neutral conditions. But NOM in CFPN/ H<sub>2</sub>O<sub>2</sub> system prefers to be hydroxylated and carboxylated rather than degraded by hydroxyl radical. In general, THM formation potential of hydrophobic fraction exhibited about two times larger than that of hydrophilic fraction hydrophilicity of NOM. After CFPN/ H<sub>2</sub>O<sub>2</sub> treatment, the increasing hydrophilicity of NOM is positive for following disinfection (Korotta-Gamage and Sathasivan 2017). There are few pieces of research on carbon materials-based Fenton-like oxidation

for NOM and NOM-derived DBPs control, since it related to the transformation

between Fe<sup>2+</sup> and Fe<sup>3+</sup> or the involvement of other transition metal ion. If Fenton like oxidation used for NOM and NOM-derived DBPs removal, the problem of metal leaching from catalyst deserved more attention especially in drinking water treatment.

The main free radical of sulfate radical-based AOPs (SR-AOPs) with high redox

## 3.3.3. Sulfate radical-based AOPs

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potential  $(E_0 (SO_4^-) = 2.5 \text{ eV} - 3.1 \text{ eV})$  helps decompose high molecule NOM into low molecule NOM. Besides, Ike et al. proposed that  $SO_4^{\bullet-}$  with higher selectivity may achieve enhanced degradation of hydrophobic fractions of NOM and the increased mineralization of overall NOM, and both effects may directly reduce DBP formation potential (Ike et al. 2019b). Catalytic performance of carbon materials in SR-AOPs was well discussed before (Yu et al. 2020), and this technology showed efficient removal and control potential of NOM and DBPs since the persulfate radical of high standard reduction potential played an important role. Cheng et al. found that PMS activated by activated carbon can mitigate membrane fouling mainly originate from NOM (Cheng et al. 2019). Considering the problem of toxic metal leaching, in the study of Magbool et al, nitrogen-doped reduced GO (NrGO) was used as a catalyst of PS for NOM removal with about 86.9%, 59.9%, and 60.3% removal efficiency for actual NOM, HA, FA, respectively. Moreover, the humic-like component was more easily to be removed compared with the fulvic-like component (Fig. 5c) (Magbool et al. 2018). Cheng et al. reported that both reversible and irreversible fouling resulted from different NOM

fractions with the increasing effluent of organic content could be reduced with PAC/PMS pretreatment (Cheng et al. 2019).

However, Guan et al. found that PDS/CNT/I<sup>-</sup> system resulted in high formation potential of I-DBPs in the presence of NOM. There are absorbable organic iodine compounds (AOI) as well as specific iodoform and iodoacetic acid detected in treated water (Guan et al. 2017). Before the novel oxidation process practical application in water treatment, there are still more conditions need to be considered. Regardless of DOM type, the formation potential of DBPs can be decreased with the high radical dose and the DBPs precursors presented extensive mineralization. But an excessively residual dose of externally added PS bring to an increasing generation of inorganic oxidation byproducts (Ike et al. 2019b). Therefore, from the standpoint of the overall progress and application, the dose of oxidant should be paid attention.

#### 3.3.4. Catalytic oxidation or ozonation

Aeration is convenient and easy to operative oxidation in engineering applications. Because of the low efficiency of separate aeration for oxidation, high-efficiency catalysts deserve to be studied. Fe–N–C oxidase-like nanozyme (FeNZ) was firstly used by Yang et al. to catalytic oxidation of NOM. And the removal degree of NOM was increased more than 6 times with a low dose of FeNZ (10 mg/L) than that without catalyst. The mechanism of NOM removal by FeNZ included adsorption, activation and oxidation. FeNZ/O<sub>2</sub> not only broken high MW NOM into low MW fractions, but also mineralized small-molecule components (Fig. 5d) (Yang et al. 2020a).

It is knowledgeable that O<sub>3</sub> can react with numerous contaminants. But the selectivity of O<sub>3</sub> for pollutant oxidation will lead to low mineralization efficiency of pollutants, or even bring in toxic intermediate products such as DBPs generation (Wang and Chen 2020). Besides, the low solubility of O<sub>3</sub> in water results in low service efficiency and high operating cost. In some areas, catalytic ozonation is directly used as a disinfection progress. Besides, in those main areas where the chlorination disinfection process is the predominate technology, catalytic ozonation can be used as technology to control the DOMs and ammonia before disinfection which can significantly reduce the precursors of DBPs (Chu et al. 2012, Chuang and Mitch 2017). O<sub>3</sub>/BAC systems have been broadly applied in municipal wastewater treatment. Because O<sub>3</sub> requires field preparation and the price of O<sub>3</sub> is relatively high, O<sub>3</sub> is under restrictions in widely application and it always used for pretreatment. And the combination of O<sub>3</sub> and BAC showed a considerable synergetic effect (Sun et al. 2018). High MW NOM is destroyed with strong oxidation ability of O<sub>3</sub>, and small fraction is ready to be adsorbed by GAC. DOC levels and aromaticity of untreated water, which are two important indicators of DBPF/P, can be effectively reduced by O<sub>3</sub>/BAC filtration. And the combination of O<sub>3</sub> and BAC was available in decreasing the new phenolic DBPs concentrations more than 80%. Huang et al. showed that GAC filtration immediately after ozonation can dramatically reduce the content of new phenolic DBPs by 97.3% (Huang et al. 2018, Jiang et al. 2020).

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Although ozonation can limitedly remove dissolved organic carbon (DOC), besides

the decomposition of the molecular weight of NOM, it can change NOM polarity, decrease the formation potential of DBP/mg DOC, and enhance the efficiency of the BAC obviously. Yan et al. found that the formed yield of THMs decreased from 201.3 mg to 140.3 mg after the biological granular activated carbon (BAC) in pilot-scale tests (Yan et al. 2010). Chu et al. proposed that O<sub>3</sub>/BAC system was effective at controlling the formation of the TOX and representative halogenated nitrogenous DBPs (N-DBPs) during chlorination (Chu et al. 2012). Chen et al. considered that O<sub>3</sub>/BAC decreased the sum of DBPFP by 49.8 % to 57.5% at 0.7 mg of O<sub>3</sub>/mg of DOC (Chen et al. 2019). The service life of BAC is limited, because of the reducing of microbial community diversity and decreasing of metabolism strength of the attached biofilm with the increasing amount of treated water. The DBPs precursor removal of old BAC filters is much less than fresh BAC filters. The study of Zhang et al. showed that the decreasing in abiotic adsorption capacity and microbial metabolism properties result in low removal capacity of N-DBP precursors by the old BAC filter (Zheng et al. 2018) And O<sub>3</sub> increased toxic potencies partly due to the formation of brominated DBPs (Chen et al. 2019). Besides, intermittently pre-ozonation before BAC filtration resulted in higher concentration DBPs formation (Chu et al. 2015a). As mentioned above, it is difficult to achieve complete degradation and mineralization of DBPs and its precursors if only AOPs were used with general oxidant agent dose merely. Common radicals roughly showed a similar impact on DBPs formation. Moreover, the molecule structure of NOM is unspecific and the DBPs is

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trace and difficult to be attacked by oxidant. In this condition, it is challenging to study the control of NOM and DBPs by carbon material combined with AOPs. To some extent, NOM and DBPs can be regarded as a representative class of pollutants, which are ubiquitous and unmanageable. But under high radical doses or with the ubiquity of halide ions, the advantage of AOPs may only be temporary or of limited impact in practical conditions (Luo et al. 2020). Meanwhile, high light intensity or high concentration oxidants will require a reconsideration of technical feasibility, economic rationality, and environmental risk (Ike et al. 2019b). Therefore, the combination of different technologies is more necessary. The application of carbon materials and oxidizing agent are a good choice because of the combined effect of adsorption and oxidation process.

#### 4. The potential environmental influence of carbon materials

4.1. New sources of DBPs precursor result from the application of carbon

## 640 materials

NOM is widely believed to be the most dominating precursor of DBPs. However, NOM with certain concentrations in untreated water is not adequately transformed into known and experimentally measured DBPs in finished waters in some cases. More attention has been paid to other non NOM-derived DBPs precursors which can transfer into more known or unknown DBPs species (<u>Du et al. 2016</u>). Recent studies have highlighted that unintended chemical agents existed in water treatment influence DBP

formation, especially as a DBPs precursor (Ding et al. 2019). In the previous section, various carbon materials can be used as adsorbents, filling materials of filter system, and catalysts, which show the potential to revolutionize the traditional water treatment processes. The adoption of carbon materials at full-scale water treatment facilities have been raised, many problems including technical challenges, public acceptance, and uncertain environmental risks of carbon materials need to be concerned (Adeleye et al. 2016, Yang et al. 2020c). Zhang et al. found that even mg/L residual of carbon materials can be detected in downstream after 6 to 12 months because of the corrosion and disruption of carbon materials (Zhang et al. 2019a). For carbon materials with a large proportion of micropores, NOM may only adsorb on the surface of carbon materials and it can be oxidized by following disinfection progress and then probable formed DBPs (Shimizu et al. 2018). Considering the artificial addition of carbon material used for NOM and DBPs control, and carbon materials themselves can be the precursors of DBPs during water treatment. Therefore, in following section, carbon material-based DBPs precursors will be discussed. nC<sub>60</sub> is usually referred to as aqueous colloidal aggregate which is a stable existing form of C<sub>60</sub>. The nC<sub>60</sub> in nature water might be released from relative industrial wastewater. Wang et al. proposed that nC<sub>60</sub> in an aqueous suspension can be transformed into products with various O- and Cl-containing functional groups after chlorine exposure (Fig. 6a) both under the darkness and light condition (Wang et al. 2012). Double bonds rather than the simple C–C bonds of  $nC_{60}$  are preferentially to be

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attacked during chlorination disinfection. It is proved that carbon materials can be attacked by disinfectants.

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CNTs, especially the functionalized CNTs, are revealed to serve as the potential precursor of halogenated DBPs such as THMs, HAAs and HNMs. The structures of CNTs are more regular and compact when compared to NOM, but the structures on the end of CNTs are similar to that of NOM, especially the aromatic rings with different functional groups (shown in Fig. 6c). Then, the mechanism of DBPs formation caused by CNTs and NOM might be comparable. The -NH<sub>2</sub>, -OH and -COOH functionalized MWCNTs were investigated by Du et al. The results showed that relatively higher THMs production was observed in the presence of OH-MWCNTs with rich Ocontaining functional group. Moreover, Du et al. compared the DBPFP of GO and HA with similar amount of -OH functional groups on their surfaces. The detected THMs concentration after chlorination in GO sample was lower than that in several HA samples even with higher TOC concentration, and the result was further proved by THMs formation kinetics. However, they reported that CNTs are not the major precursor of DBPs in comparison of NOM (Du et al. 2016), and Verdugo et al. reported only 1 µg DBPs/ mg-CNT generated after chlorination of 4 h (Fig. 6d) (Verdugo et al. 2016).

Biochar itself can be a precursor substance of DBPs. The ratios of C/O and O-containing functional groups influenced by the pyrolysis temperature of biochar affect the formation of different DBPs. Zhang et al. found that the lowest concentration of

TCM formation during the chlorination of biochar produced in high temperature. But the concentration of trichloroacetic acid (TCAA) showed a converse tendency with the produced temperatures of biochar. There are negligible differences of DBPs formation among various biochar pyrolyzed by different raw biomass at the same temperature. It might attribute to the similar structures and O-containing functional groups of biochar under same pyrolysis temperature. In general, the aromatic ring structures and various O-containing functional groups of the biochar are the main precursor for DBPs formation (Zhang et al. 2019b). And DOM leached from biochar should take into consideration. In the study of Lee et al. 2018, the potential of DOM leached from biochar as precursors of DBPs has been studied. The result demonstrated that the sources of biochar are the main influence factor of DOM characteristics rather than the contact time for extraction. Moreover, the precursors for THMs were more easily leached from biochar than those of HAAs or DOC, and condensed aromatic structures could be suggested as a predictor of THMs formation. As shown in Fig. 6b, the initial leaching of DOM derived from biochar is important and severe in intermittent contact between water and biochar (Lee et al. 2018b). It is proposed that many factors such as production conditions, pyrolysis temperature and biomass sources, can influence the type and amount of DOM released from biochar (Quan et al. 2020). The optical properties of DOM are highly correlated with different biomass feedstocks and more than 50% DOM was extracted from total cumulative carbon during the first day of the extraction (Lee et al. 2018b). More importantly, the initial extraction from biochar

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showed greater formation potential for THMs compared to HAAs. The result showed a strong relation between terrestrial humic-like or fulvic-like components and THMs formation potential. And condensed aromatic structures in BC-derived DOM can be regarded as the detectable precursors which contributed for THMs formation (Huang et al. 2019, Lee et al. 2018b). Besides, Chu et al reported that BAC in the filter can be the source of N-DBP precursors. In the BAC treatment process, soluble microbial products (SMPs) produced by microorganisms, which including many DON compounds such as polysaccharides, proteins and exocellular enzymes, can be monitored in the BAC effluent and became the N-DBP precursors (Chu et al. 2015a). However, the existence of SMPs played an important role for contamination removal (Korotta-Gamage and Sathasivan 2017). The application of BAC beyond service life without replacement or regeneration might result in the increasing release of SMPs (Sun et al. 2018). Moreover, Yan et al. proposed that the SMPs leaked from the BAC reactor show a high formation potential of THMs in the following continued chlorination process (Yan et al. 2010). In conclusion, carbon materials used for water treatment and purification show great potential as precursors for DBPs formation. On the one hand, more experiments and theoretical researches are needed for the DBPFP of different carbon material-derived components. On the other hand, considering the environmental concentration of carbon materials and the low generation potential of DBPs compared with that of NOM, environmental and health problems resulted from carbon materials themselves such as particle size and impurities should be paid more attention (Wang et al. 2020a).

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#### 4.2. Sustainability of carbon materials

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As mentioned above, carbon materials can play different roles in water treatment progresses. Considering the potential environmental risks of carbon materials during production and application in water treatment, especially the sustainability, more relative issues of different carbon materials should be discussed. The negative effects of carbon materials in water treatment can mainly divided into several parts including: harmful gas emissions during the production process, ecotoxicity resulted from micro/nano dimension, harmful components and adverse properties or structure of carbon materials, in addition to the potential of carbon materials as the precursor of DBPs we mentioned above. Those potential environmental influence unintentionally accompanied with production or subsequent aging. From the production methods and the characteristic of carbon material itself, the nanostructures that already exist may be the source of negative influence. For example, ball milled biochar and its composite nanomaterials can induce the ROS formation which further inactivated cell (Liu et al. 2019, Liu et al. 2020b). And carbon materials undergo subsequently inevitable aging progress during application, for example, broken, oxidation, collapse and so on, are another reason of potential negative effects. Aged carbon materials may have different characteristics from the original or ideal (Wang et al. 2020c). The negative effects of the production and use of carbon materials are the reasons that affect their sustainability.

Until now, the sustainability of carbon materials have been well discussed especially

focus on environmental catalysis, energy storage, soil recovery and so on Correa et al.
2019, Kumar et al. 2021, Shen et al. 2020a, Wang et al. 2021). There is still obvious
tradeoff among different carbon materials used for water treatment because of cost,
efficiency, ecological risk, and sustainability. Therefore, more recent researches paid
attention to carbon materials derived from sustainable biomass resources and green
preparation methods (such as mechanochemical), it can increase their sustainability and
decrease environmental influence during preparation and application (Nasrollahzadeh
et al. 2021). Biomass resources, which regarded as one of the most promising renewable
feedstocks nowadays, are more likely to be used in actual water treatment progresses.
Meanwhile, considering carbon neutralization, the application of renewable resources,
which have little impact on the natural environment, is encouraged (Shen et al. 2020a).
However, biomass-derived carbon materials prepared by simple thermal transformation
may not satisfied with complicated environment problem. Starting from sustainable
resources, by adjusting some parameters in production, targeted regulation of the
production of materials that meet the requirements is a more popular research in the
field of environment (Nasrollahzadeh et al. 2021).
Moreover, it is necessary to do sustainability evaluation of carbon materials in water
treatment based on life cycle assessment (LCA), as a method which is chiefly used for
evaluating a certain material or technology related environmental factors and potential
impact Polative analysis and research are call for further avaluation

#### 5. DBPs control in wastewater disinfection progress under different condition

and the potential treatment methods by carbon materials

## 5.1. Selection of carbon materials in different progresses and methods in

## municipal wastewater treatment

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In water treatment, NOM and DBPs can be removed through multiple progresses (Zhang et al. 2017). During water transportation, treatment and distribution, carbon materials can be used in different stages and assisted with different technologies. Carbon material could be used as filling material of fixed beds or prepared into the membrane to reduce recovery process and dissipation loss of powder. Considering the impact of disinfection on subsequent treatment, important issues include not only the selection of carbon materials but also the application sequence of carbon materials and chlorinated disinfectants. For example, pre-chlorination is recently introduced as a promising method to reduce the formation of DBPs. Pre-chlorination adjusted the sequence of NOM contact with disinfectants and carbon materials in water treatment to reduce DBPs formation. Erdem et al. investigated that dosing chlorine before GAC contactors is effective for DBPs control. With GAC treatment after pre-chlorination, about 58% decreasing in the formation of DBPs has been found. And higher removal efficiency was observed by NOM with higher aromaticity (Fig. 7a) (Erdem et al. 2020). Jiang et al. also proposed that the adsorption of GAC targeting for intermediate aromatic halogenated DBPs instead of NOM can effectively decrease the formation of THMs, HAAs, and total organic halogen (TOX) in disinfected water. Besides, treated water

showed considerably less developmental toxicity than the traditional approach (Jiang et al. 2018, Jiang et al. 2017). Therefore, by adjusting methods and water disinfection progress can control DBPs formation in water. Disinfection is not only necessary for drinking water treatment, but also important in tap water production and supplement, swimming pool water purification or even medical wastewater treatment. Due to the different sources and demands of water, the specific process of water treatment needs to be designed, and coordinated with the local development situation.

#### 5.2. Problems caused by emerging contaminations like microplastics

In the water bodies worldwide, MPs have been detected as a new emerging micropollutant in the natural system, and they become a hot topic in the recent study. Increasing research reported migration, transformation and the negative effect of MPs and relatively deuterogenic substances on creatures such as fragmentation and additives release (Fu et al. 2020, Ye et al. 2020a). MPs are widely found in surface water from discharge of industrial wastewater and domestic wastewater, due to the abrasions and decomposition of environmental plastic wastes. Besides, atmospheric deposition is also a potential source of MPs for the water environment (Ding et al. 2019). In the wastewater treatment system, the source of primary MPs mainly results from daily personal care products (including facial cleanser, scrubbing cream, toothpaste and various cosmetics) and laundry wastewater (Shen et al. 2019). During water transportation and distribution, undesirable DBPs precursors are emerging such as fragmented plastic and aging supplement pipes (Ding et al. 2019). Besides, overdosing

a certain degree of chlorine disinfectant is necessary to effectively control the number of pathogenic microorganisms in the urban pipe network system, which a relatively closed and stable environment that provides an ideal living environment for some harmful pathogenic microorganisms. Internal attach biofilms will formation and organic matter may adsorb on the inner tube of pipes in the long term., overdosing a certain degree of chlorine disinfectant in the pipe network system is necessary (Ding et al. 2019). In addition, the layer of organic and inorganic substances adhered on MPs are formed during natural-ageing progress gradually. The organic matters will remarkably change the surface properties of MPs as well as become the potential precursor of DBPs. Besides, sunlight illumination and water flow can accelerate structural damage of MPs and DOM leaching from MPs. Lee et al. investigated the DOM leaching capacity from MPs for DBPs formation potential. The main fraction of MP-derived DOM is low in molecular weight, which is highly correlated with DBPs formation in treated water according to previous studies. And MPs-derived DOM showed comparable DBPs formation potential compared to that of aquatic NOM (Lee et al. 2020). Considering the variety of MPs, Kelkar investigated that various sources of MPs exposed to different disinfection conditions representative of dosages used in the disinfection of drinking water, wastewater, and heavily contaminated surfaces. There was a few chemicals and no physical changes of MPs detected with normal chlorination disinfectant doses in wastewater treatment (the concentration-time values is about 150 mg min/L), but

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obvious differences between several types of MPs were detected in higher disinfectant doses and temperatures and presented chemical and physical changes. Polystyrene (PS) showed degradation at low chlorine doses (75 mg min/L), but polyethylene (HDPE) and Polypropylene (PP) were resistant even in high chlorine doses (150 mg min/L) that is equivalent to the characteristic of wastewater disinfection (Kelkar et al. 2019).

In recent research, the removal capacity of MPs by carbon materials was discussed. Tong et al. 2019 found that biochar or Fe<sub>3</sub>O<sub>4</sub>-biochar can retention the transportation of MPs, and about 74% MPs can be recovered from biochar. Extreme situations such as a sudden rainstorm was stimulated through a high flow rate. The result showed that the release of MPs decreased from 16.5% to 7.5% with Fe<sub>3</sub>O<sub>4</sub>-biochar existence (Tong et al. 2019). Therefore, carbon materials can be used for potentially DBPs precursors like microplastic removal and then decreasing DBPs formation.

#### 5.3. Guideline of disinfection for the public health emergency

In the past 20 years, Severe Acute Respiratory Syndrome Coronavirus 2 (SARS-CoV-2) becomes the third emerging coronavirus and results in high alert to global public health institutions (Carducci et al. 2020, Wang et al. 2020b). Medical wastewater is a potential source of health risks, especially during the epidemic. The treatment of hospital wastewater has attracted special worldwide attention (Wang et al. 2020b). However, in the routine examination and relevant regulation, the concentration and variety of DBPs are not monitored. And the wastewater treatment plants seldom equipped with real-time monitoring. In particular, under the COVID-19 pandemic

context all over the world, it is of great significance to reduce the health risks to the public and society. In addition to possible viruses, hospital wastewater may contain high concentrations of drugs, such as cardiovascular drugs, painkillers, antibiotics, cell inhibitors, and X-ray contrast agents. Considering the multiple contaminants and pathogenic bacteria of high concentration, medical wastewater undergone certain treatments are permitted to discharge into municipal networks (Carducci et al. 2020). Therefore, hospitals are required to equip with the relevant medical wastewater treatment system, and appropriately disinfection for hospital wastewater is necessary before transported or discharged into municipal sewage networks (Wang et al. 2020b). Under the public health emergency, the dosage of disinfectant for medical water treatment per unit is rapidly increased and the total quantity of disinfectant is tensive supply. Effluent from the water treatment plant is requested to maintain high concentrations of residual chlorine under the public health emergency compared to that of in general conditions. Yang et al. found that residual Cl/Br and DBPs formation showed positively association with disinfectant dosage rather than dosing methods (Yang et al. 2016). Therefore, DBPs formation potential will prominently increase during the epidemic viruses with increasing and accumulating disinfectants dose. In the previous study, different carbon materials show the possibility of bacterial retention as well as DBPs and its precursor removal. But further investigations are needed for the removal potential and efficiency of carbon materials for specific pathogenic bacteria or virus. Under important public health paroxysmal incident,

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carbon materials can be an additional and feasible technology in water treatment for treating the pollution of pathogenic bacteria or viruses as well as reducing the potential of DBPs formation.

## **5.4.** Challenges of DBPs control in stormwater

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Nowadays, the utilization of stormwater is widely considered as a crucial element for achieving regional and sustainable water management and circulation, especially in urban areas severing water shortage (Furlong et al. 2017, Wijesiri et al. 2020). Unfortunately, it is difficult for traditional stormwater recycling system to reach the pathogens removal standard on the basis of relevant policies and regulations, so additional disinfection technologies are required (Feng et al. 2018, Shen et al. 2020b). Stormwater collection and centralized management can reduce the impact of heavy rainfall on wastewater treatment plants and ensure treatment efficiency. Stormwater, flowed through the ground, was collected into drainage systems. Organic matter in the atmosphere can become a kind of DOM source of the surface water through wet and dry precipitation. Particulate matters dispersed on atmosphere or at road surface can be gathered by rainfall, and increase DOM concentration in the stormwater stream (Hou et al. 2018, Zhang et al. 2020). Rain washing in highly urbanized areas will collected a portion of deposited particulates with the source of automobile exhaust and industrial dust. You et al. found that NOM in raw water will increase around 40% after some heavy rain event in some areas (You et al. 2018). Warner and Saros et al. found that the sudden accumulation in quantity and quality of DOC resulted from precipitation events

in a short time (Warner and Saros 2019). Considering the high NOM concentration in stormwater requiring further treatment progress for its reapplication, there is a contact possibility of organic matter and disinfector with the chance of DBPs formation. He et al. deduced that DOM resulted from atmospheric wet deposition is an important source DBPs precursor, and the relatively high contribution of DOM from dry deposition particles are hydrophilic and low MW fractions (He et al. 2020). Furthermore, Hou et al. proposed that aromatic proteins and soluble microbial product-like compounds showed highly formation potential capacity of DBPs. Although according to the statistic information, more than 80% of the total DOM contribution in stormwater originated from particulate matter, the contribution of atmospheric organic matter could not be neglected (Hou et al. 2018). The application of carbon materials as the replaceable filling materials or substrates

amendment in LIDs construction, which are representative and important construction for stormwater management, can increasing the resiliency of LID (Mohanty et al. 2018). Carbon materials shows the potential of removing different contamination in stormwater runoff and some of them are regarded as the possible DBPs precursors. Furthermore, the widespread application of carbon materials such as biochar can even lower the cost of the whole construction as well as increase the maintainability and efficiency of LID (Yang et al. 2020c, Zhou et al. 2019), and it can decrease potential DBPs precursors and control DBPs in the following disinfection progress.

#### 6. Perspectives

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921 Although some researches and achievements have been made, there are still many 922 challenges to utilizing carbon materials for DBPs and their precursors' control in water 923 treatment. Some points are listed as follows: 924 (1) The control of DBPs during water treatment should focus on reducing the interaction 925 between potential DBPs precursors and disinfectant. The additional carbon materials-926 based progress such as pre-chlorination and adsorption progress before disinfection can 927 be considered in application. Selecting the type and amount or the methods of carbon 928 materials in DBPs control treatment and optimizing the sequence of existing disinfection process, and then achieving the purpose of DBPs control by carbon 929 930 materials. 931 (2) The multiple effects of carbon materials in water disinfection progress should take 932 into more consideration and make the utmost use of the advantage in application. From 933 the above discussion, for example, when carbon materials used in AOPs for DBPs 934 control, the function of catalyst and adsorbent are exhibited at the same time. Various 935 function of carbon materials can be the advantages for its further and widely application integrated DBPs control. 936 937 (3) It is necessary to establish the connection between DBPfP and other influence 938 factors such as contact time, disinfectant dosage, residuals or even carbon materials. 939 According to the complex actual water treatment situation, computationally adjustable 940 and optimizable of relevant parameter in disinfection is important. DBPs formation potential should be an indicator of the selection and evaluation of disinfection methods. Different disinfection methods show different influence and results of DBPs formation. And the emerging and unregulated DBPs accompanying with new disinfection progresses should be taken into consideration. Therefore, a comprehensive assessment of the disinfection methods should be carried out. (4) The oxidation mechanism of DBPs precursors and the toxicity of reaction intermediates should be deeply investigated. A wide variety of disinfection byproducts have been found, and it is important to do more researches on model design and development. And the possibility of emerging DBPs control by carbon materials should

#### 7. Conclusion

be examined.

In conclusion, this review summarized the removal of DBPs and their precursors by different carbon materials. The removal performance and capacity of carbon materials for DBPs and their precursors are divided into methods including adsorption, membrane filtration and AOPs on different stages and scales. In general, it is generally accepted that removing DBPs precursors such as NOM before disinfection are more effective than decreasing DBPs that are already formed. The possible and new source of DBPs precursor resulted from the application of carbon materials also be overviewed. Moreover, combined with the actual situation, some suggestions deal with the emerging problems such as MPs pollution and stormwater treatment on the application of carbon

materials used for DBPs control have been proposed.

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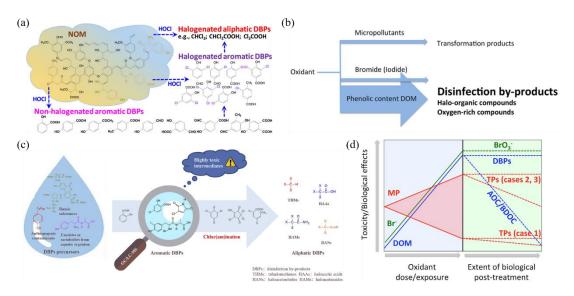
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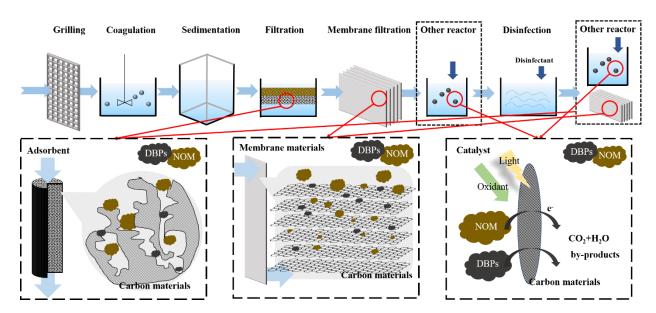
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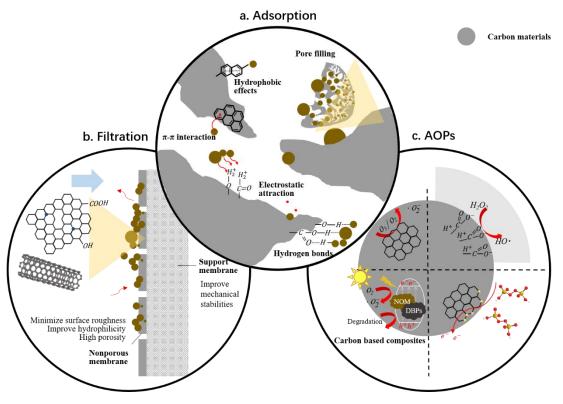
# Figure captions: 1342 1343 1344 Fig. 1. A summary to the conversion and potential risk of NOM and DBPs. 1345 Fig. 2. The methods of carbon materials for NOM and NOM-derived DBPs control in 1346 different progresses of water treatment. Fig. 3. The mechanism of carbon materials for NOM and NOM-derived DBPs control 1347 in the three methods including adsorption, filtration and AOPs. 1348 1349 Fig. 4. Membrane made by graphite-based materials used for NOM and NOM-derived 1350 DBPs control. 1351 Fig. 5. Membrane made by graphite-based materials used for NOM and NOM-derived 1352 DBPs control. 1353 Fig. 6. New sources of DBPs precursor result from the application of carbon materials. Fig. 7. DBPs control in wastewater treatment under different conditions. 1354 1355



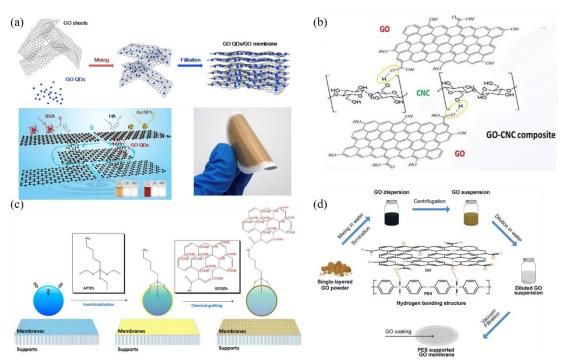
**Fig. 1.** A summary to the conversion and potential risk of NOM and DBPs. (a) NOM is regarded as the most widely concerned DBP precursors (Jiang et al. 2020); (b) DOM mainly contributed from NOM is the major consumer of oxidants and showed high DBPs formation potential (von Gunten 2018); (c) DBPs derived from diverse sources of NOM from show different characteristics (Liu et al. 2020a). (d) Oxidation progress in disinfection of water brings in increasing toxicity to human health with DBPs formation (von Gunten 2018).



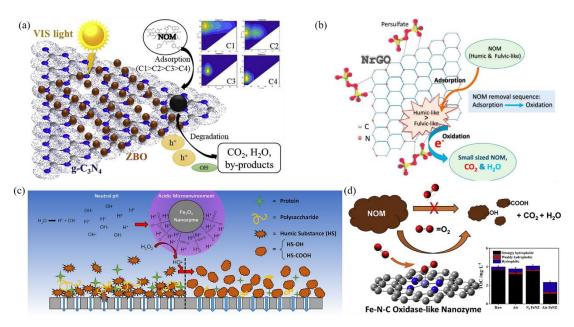
**Fig. 2.** The methods of carbon materials for NOM and NOM-derived DBPs control in different progresses of water treatment. It mainly discusses the removal mechanism and efficiency of NOM and NOM-derived DBPs by carbon materials as adsorbents, catalysts and membrane filling materials.



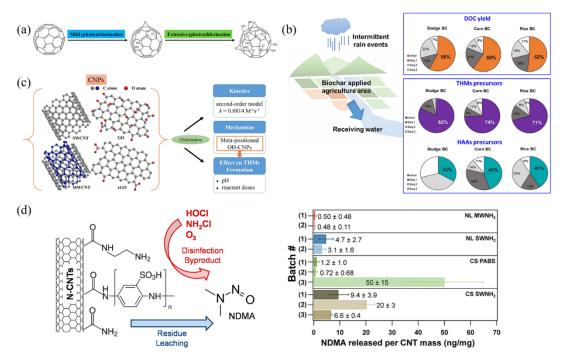
**Fig.3.** The mechanism of carbon materials for NOM and NOM-derived DBPs control in the three methods including adsorption, filtration and AOPs.



**Fig. 4.** Membrane made by graphite-based materials used for NOM and NOM-derived DBPs control. (a) GOQDs can prevent the stacking of GO membrane and showed well antifouling performance (Zhao et al. 2019); (b) GO-CNC composite membrane preformed high porosity and water permeability (Lv et al. 2018); (c) Membrane modified with GO presented improved hydrophilicity and minimized surface roughness (Gu et al. 2020); (d) the increasing hydrophilic of membranes because of coated GO showed high HA rejection trends (Chu et al. 2017).

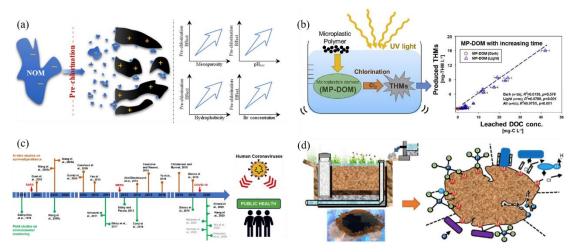


**Fig. 5.** Membrane made by graphite-based materials used for NOM and NOM-derived DBPs control. Carbon materials and specific oxidation methods used for NOM and NOM-derived DBPs control including: (a) g-C<sub>3</sub>N<sub>4</sub>-based photocatalyst under visible light (<u>Truong et al. 2019</u>); (b) carbon-doped Fe<sub>3</sub>O<sub>4</sub> peroxidase-like nanozyme catalyze Fenton reaction (<u>Yang et al. 2020b</u>); (c) N-doped reduced graphene oxide-activated persulfate (<u>Maqbool et al. 2018</u>); (d) Fe–N–C oxidase-like nanozyme for catalytic oxidation (<u>Yang et al. 2020a</u>).



**Fig. 6.** New sources of DBPs precursor result from the application of carbon materials.

(a) Chlorination transfer nC60 transformed into products containing carbon-chlorine, epoxy and hydroxyl functional groups (Wang et al. 2012); (b) DOM leached from BC showed the potential for DBPs precursors (Lee et al. 2018b); (c) DBPs were formed during chlorination of Carbon nanoparticle (Du et al. 2016); (d) N-functionalized carbon nanotubes were proved to be the source and precursor of DBPs (Verdugo et al. 2014).



**Fig. 7.** DBPs control in wastewater treatment under different conditions. (a) The progress or methods of carbon materials in wastewater treatment (Erdem et al. 2020); (b) Problems caused by emerging contaminations like microplastics (Lee et al. 2020); (c) Guideline of disinfection for public health emergency (Carducci et al. 2020); (d) Challenge of DBPs control in stormwater (Zhou et al. 2019).

# **Table captions:**

- **Table 1.** The application of carbon materials as adsorbent for NOM-derived DBPs and its precursors control.
- **Table 2.** The utilization of carbon materials in membrane for NOM-derived DBPs and its precursors control.
- **Table 3.** Different carbon materials used in AOPs for NOM and NOM-derived DBPs control and its mechanism, efficiency, advantages and disadvantages in the progress.

**Table 1.** The application of carbon materials as adsorbent for NOM-derived DBPs and its precursors control.

Carbon materials	Adsorbed	Relative	Performance	Mechanism	Advantages and	Reference
	substance	concentration			disadvantages	
Granular activated	NOM	DOC=2 mg/L	58% decrease in THM, HAA,	Halogenated DBP intermediates	high mesoporosity adsorbed	(Erdem et
carbon (with pre-		Br=200 μg/L	HAN and TOX formation	adsorbed mostly at mesoporous	more halogenated DBP	al. 2020)
chlorination)		pH=7.5		region of the adsorbent	precursors	
Granular activated	Nonhalogenated	3 mg C/L SRHA	>50% removal for	The higher the log D value of a DBP	Removal efficiency:	(Jiang et
carbon	and halogenated	solution (with 90 mg/L	nonhalogenated aromatic	showed the higher the removal	halogenated aromatic	al. 2020)
	aromatic DBPs	NaHCO <sub>3</sub> as CaCO <sub>3</sub> )	DBPs and 96% removal for	tendency	DBPs > nonhalogenated	
			halogenated aromatic DBPs		aromatic DBPs	
Granular activated	aromatic	3 mg C/L SRHA	73% TOX removals	GAC adsorption preferred neutral	aromatic DBPs are	(Jiang et
carbon	intermediates of	solution	doubled TOX removals	form rather than ionized form	preferable to be adsorbed on	al. 2018)
	halogenated		compared with the traditional		GAC over aliphatic DBPs	
	DBPs		approach			
			(i.e., using GAC to remove			
			NOM before chlorination)			
Powdered activated	NOM	4.51 mg DOC/L	the removal rates are 46%	The THMFP removal was closely	Removal sequence:	(Park et al.
carbon			DOC, 84% UV <sub>254</sub> and 70%	associated with CF removal and the	HANFP>THMFP>HAAFP	2019)
			SUVA reduction (with	precursors of MW < 1,000Da.		
			200 mg/L PAC)			
Powdered activated	carbonaceous	3.4 mg/L (raw water	60–90% for six C-DBPs and	synergistic impact of permanganate	permanganate oxidation	(Chu et al.
carbon (with	and nitrogenous	from the east of Taihu	64-93% for six N-DBPs	and PAC	chemically altered the	<u>2015b</u> )
permanganate	DBPs precursors	Lake)			molecular structures of	
oxidation)		$1\ mg\ /\ L\ KMnO_4$			nitrogenous organic	

		20 mg / L PAC			compounds and increased	
					the adsorption capacity of	
					the DBP precursors	
Powdered activated	NOM (EOM)	synthetic algal-rich			UV/Cl/PAC: decreased	(Xing et al.
carbon (before UV		feed water			reversible and irreversible	<u>2019</u> )
and UV/chlorine					fouling resistance	
oxidation)					UV/PAC: reducing	
					reversible membrane fouling	
Activated carbon	NOM	5.0-25 mg NOM/L	Qe= 1.268 or 1.266 (nm mg)	Chemical adsorption (well-fitted to	small MW (<0.4 kDa) faster	(Shimizu
		constant adsorbent		Freundlich model R <sup>2</sup> > 0.95)	than higher MW	et al. 2018)
Carbon nanotubes		dose of 250 mg/L	Qe=1.393 or 1.288 (nm mg)		High MW (>1 kDa) was	
					adsorbed; the smallest	
					fraction (<0.4 kDa) was not	
					adsorbed	
Carbon nanotubes	HA (NOM)	2,4,8 mg DOC/L(with	removal efficiency increase	the interaction between HA	the removal efficiency for	(Hui et al.
(compounded with		5 mM NaNO <sub>3</sub> and	from 41.84 to 80.56%(2, mg	molecules	DOC in acidic or alkaline	2016)
polyalumium		4mM NaHCO <sub>3</sub> )	DOC/L)		conditions	
chloride)		0.02-0.4 mmol	,			
,		Al/L(0.25, 0.50, 1.0 g,				
		1.5 g, 2.0 g CNTs/60				
		mL AlCl <sub>3</sub> )				
		3/				
Biochar	NOM (collected	1.04 mg C/L (DOC)	Remove 90% of organic	interaction of active sites with	All molecular weights can	(Lee et al.
	from the junction		matter in 20 min of contact	organic molecules controls the	be removed	<u>2018a</u> )
	of North Nanshi			adsorption process		

	River and South		And the capacity is 6.4 mg-			
	Nanshi River)		DOC/g C			
Polyethyleneimine	NOM	$5.0 \pm 0.3$ mg C/L HA	80.65 ±4.16 mg C/g	higher surface area, the electrostatic	low solubility substance of	(Truong et
modified biochar		solution	3–4 times higher adsorption	attraction, hydrogen bonding, $\pi$ - $\pi$	HA is advantageous for	<u>al. 2020</u> )
			capacity than pristine	interactions and new cation- $\pi$	adsorption	
			adsorbents at pH=3	interactions		
Activated biochar	Humic and	7.5 mg/L HA and 2.5	HA $K_F$ =1.39 (mg/g)(mg/L) <sup>1/n</sup> )	abundant adsorption sites	synergistic effect of	(Jung et al.
(with the presence of	tannic acids	mg/L TA solutions	TA $K_F$ =32.4 (mg/g)(mg/L) <sup>1/n</sup> )		combination	<u>2015</u> )
aluminum sulfate)		(over 10 mg/L)				
Biological activated	NOM	1.8 and 3.5 DOC mg/L	Stable operation for 245 days	Adsorption is important in the	Well distributed pore-size	(Lu et al.
carbon				performance of the BAC filters and	structure	<u>2020</u> )
				renewability was driven by bacterial		
				metabolic activity		
Graphene oxide	НА	10-60 mg C / L	~ 20 mg C/g (pH=4)	Preferentially adsorbed onto GO	Highly related to solution	(Lee et al.
				surface	pH and the available GO	2015)
					surface sites	

<sup>\*</sup> SRHA: Suwannee River humic acid

Table 2. The utilization of carbon materials in membrane for NOM-derived DBPs and its precursors control.

Carbon materials	Type of membrane	Performance	Mechanism	Advantages	Reference
Graphene	Graphene-derived membranes	\	the transport of HA is retarded in the presence of -COO- on graphene surfaces	A hydrophilic GE surface increases the reversibility of HA adsorption	(Qin et al. 2020)
Graphene oxide	Graphene oxide quantum dots onto ceramic microfiltration membranes	\	hydrophilic and negatively charged surface characteristics	uniformity in pore size improved water permeability	(Gu et al. 2020)
Graphene oxide	GO-based NF membranes	>99% (20ppm HA)	Laminated GO sheets acted as molecular sieving	GO quantum dots act as nano wedges to prevent stacking	( <u>Zhao et al.</u> 2019)
Graphene oxide	GO incorporated polyethersulfone hybrid ultrafiltration membranes	The greatest HA solution flux value = 193.78 L/m <sup>2</sup> h	The enhancement of the hydrophilicity of the GO-membrane surface	Improve the membrane recyclability and antifouling capabilities	(Algamdi et al. 2019)
Carbon Nanotubes	Nanocomposite multiwalled carbon nanotubes-polyamide membranes		Decrease roughness and polymer mobility on the surfaces of the membranes at the molecular scale	Increase chlorine resistance properties, antifouling performance, and reduced scaling in the presence of calcium ions	(Cruz-Silva et al. 2019)
Graphene oxide	graphene oxide modified polyamide thin film composite membranes	reject >99.9% (5 mg/L NOM) with constant water flux of 65 L/(m² h bar) for ~6.0 h	defect-free and have pores in sub- nanometer level or of the size of just above the water molecule's size	layer-by-layer laminated structure and different thickness of GO membrane can be regulated synthesis	(You et al. 2018)
Graphene oxide	Graphene oxide-cellulose nanocrystal composite functionalized PVDF membrane	\	The formation of a hydrated layer and electrostatic repulsion	Introduced massive oxygen- containing	( <u>Lv et al.</u> 2018)
Graphene oxide	An asymmetric membrane with a dense GO barrier layer on the	Antifouling performance: GO>PA (operated in PRO	Water enters the GO membrane primarily around the oxidized edges of GO	Almost immune to irreversible fouling and suitable for engineered osmosis	( <u>Hu et al.</u> 2016)

D: 1	back (i.e., porous) side	mode) GO≈PA (operated in forward osmosis mode)		T	(Chaffar at
Biochar	Biochar-PVdF composites membranes		electrostatic repulsion and hydrophilicity improvement	Increase regeneration, stability and anti-biofouling effects	(Ghaffar et al. 2018)
Graphene oxide	graphene oxide-coated ultrafiltration membranes	the humic acid rejection of GO-coated membranes (85.3– 93.9%)	GO multilayers on the membrane surface may be held by hydrogen bonding	Increase hydrophilic and showed less flux decline and high humic acid rejection trends	(Chu et al. 2017)
Graphene oxide	Fe <sup>3+</sup> (particularly) and Al <sup>3+</sup> cross-linked GO membranes	99.6%( UV <sub>254</sub> ) or 90- 95% (TOC) for 10 mg/L HA	\	The trivalent cations greatly enhance the stability of the GO membrane	( <u>Liu et al.</u> 2017)
MWCNTs	Functionalized MWCNTs modified UF membranes	stabilities during pure- water backwashing at TMP of 60 kPa, with dislodged ratios of 1.9% and 4.54%.	The decreased direct membrane—foulant contact due to the MWCNT coating layer	Stronger resistance to NOM fouling Stability under backwashing	( <u>Bai et al.</u> 2015)
Graphene oxide	Ultrathin graphene oxide nanosheet membranes (cross-linked by diamine monomers)	DOC rejection improved to 34.3%, 24.8% and 25.3% (12.5% in contrast)	the formation of C–N covalent bonds between the GO and diamine effectively suppressed stabilized interlayer spacing	remove high molecular weight NOM	( <u>Xia et al.</u> 2015a)
Graphene oxide	GO-PVDF membranes	better removed NOM, especially relatively low molecular weight components	GO could lead to a smoother membrane surface and increased pure water permeability	GO increased pore density of the top-layer and macrovoids in the sub-layer	(Xia and Ni 2015)
Graphene oxide	GO-PVDF membranes	62.03%( UV <sub>254</sub> ) or 41.13% (TOC)	The effect of charge repulsion	membrane hydrophilicity, surface morphology and surface charge	(Xia et al. 2015b)
Activated carbon	AC/PPSU/PEI/PEG composite membranes	80% HA removal	\	filtration flux and permeability improved	( <u>Hwang et al. 2013</u> )

**Table 3.** Different carbon materials used in AOPs for NOM and NOM-derived DBPs control and its mechanism, efficiency, advantages and disadvantages in the progress.

Type of AOPs	Carbon materials	Condition	Performance	Mechanism	Advantages and disadvantages	Reference
Photocatalytic oxidation	zinc-bismuth oxides- graphitic carbon nitride(ZBO-CN)	1.5 g/L ZBO-CN	74% for DOC and 88% for UV <sub>254</sub> removal	the dominant operation of p-p and/or hydrophobic interactions	Selective removal is more pronounced for visible light than UV- activated systems	(Truong et al. 2019)
Photocatalytic oxidation	g-C <sub>3</sub> N <sub>4</sub>	5 g/L g-C <sub>3</sub> N <sub>4</sub> 0.1 THMs and HANs solution (pH=7)	The degradation percentages for CHCl <sub>3</sub> , CHBrCl <sub>2</sub> , CHBr <sub>2</sub> Cl, CHBr <sub>3</sub> , C <sub>2</sub> Cl <sub>3</sub> N, C <sub>2</sub> HCl <sub>2</sub> N, C <sub>2</sub> H <sub>2</sub> ErN, and C <sub>2</sub> HBr <sub>2</sub> N were 35.7%, 33.9%, 36.1%, 36.1%, 35.2%, 30.0%, 31.0%, 38.6%, and 41.9%, respectively.	Holes played dominant roles for THMs and HANs degradation. THMs degradation was achieved through hydrogen abstraction or/and dehalogenation. The photocatalysis removed HANs mainly through C-C bond breakage	PAC should be avoided to contact with membrane	(Chang et al. 2019)
Photocatalytic oxidation	bamboo charcoal (compounded with ZnO and TiO2)	10–70 mg/L HA solution (pH=2-10) Real wastewater (TOC=15.2 mg/L,pH=7.9)	Over 99% HA removal and 80% TOC removal in real wastewater in 180 min	Langmuir-Hinshelwood mechanism a synergistic mechanism of adsorption and photocatalysis	the competitive effects of co-existing substances in real wastewater decrease the degradation rate	(Wang et al. 2013)
Fenton like oxidation	Carbon doped Fe <sub>3</sub> O <sub>4</sub> peroxidase-like nanozyme (CFPN)	10 mg/L HA 1 mM H <sub>2</sub> O <sub>2</sub> 10 mg/L CFPN	Drop value of membrane flux deceased 20% and the flux recovered to approximately 100% after backwashing	mitigating membrane fouling by increasing the hydrophilicity of NOM	applied at neutral pH (hydroxyl radical-based treatment)	(Yang et al. 2020b)
SR-AOPs	powdered activated carbon	3.0 mg C/L (with 1 mM CaCl <sub>2</sub> , 1 mM NaHCO <sub>3</sub> , 6	73.3% DOC removal	PAC and PMS showed a significant synergistic effect in	the co-existence of PAC and HA even	( <u>Cheng et al. 2019</u> )

		mM NaCl) 1.5 mM PMS		the reduction of different NOM fractions	exacerbated fouling	
SR-AOPs	Ndoped reduced graphene oxide (NrGO)	pH=7.0 ±0.1 17 mg C / L NOM solution 0.6 mM PS 0-230 mg/L NrGO	the reduction rates of SRHA, SRFA, SRNOM are 86.9%, 59.9%, and 60.3%, respectively (with 230 mg/L NrGO)	NrGO-driven PS activation effectively removed NOM via adsorption and oxidation	May be heavily impacted by the presence of NOM in real water matrix	(Maqbool et al. 2018)
Catalysis oxidation	Fe-N-C oxidase-like nanozyme(FeNZ)	10 mg/L HA 0.4 L/min air	TOC decreased by 57.7% (at least 6 times than control	Organic mineralization and conversion of hydrophobic to	Over a broad pH range	( <u>Yang et al.</u> 2020a)
Catalysis oxidation	Biological activated carbon (with 1 year of usage time)	10 mg/L of FeNZ Raw water (3.35-7.86 mg/L DOC, 0.27-0.88 DON mg/L)	groups) Removal rate are for 39.6%, 40.2%, and 68.8% DCAN, DCAcAm, and TCNM	hydrophilic components Abiotic adsorption and microbial degradation	Aging leads to decreased performance	(Zheng et al. 2018)
Catalysis oxidation	biological granular activated carbon	Raw water 3.80 mg DOC/L Synthetic water 4.48 mg DOC/L	51% and $38%$ decrease for UV <sub>254</sub> and DOC, respectively	Synergetic effect between the ozone and the activated carbon was observed	Avoid the leakage of microbes and/or the products of metabolism	( <u>Yan et</u> al. 2010)
Pre-ozonation	Biological ctivated carbon	2.40 mg/L DOC, 0.51 mg/L DON dissolved ozone dosage was 2.0–2.5 mg/L	A stable removal of DOC (29.7–32.6%) and DON (33.0–36.7%) from the first to fifth backwash cycles	Ozonation transforms higher molecular weight compounds into smaller molecules and thereby increases the biodegradable organic matter	Terminating pre- ozonation increases N- DBP levels in BAC- filtered water upon chlorination	(Chu et al. 2015a)
Catalysis ozonization	biologically activated carbon	0-1.1 mg O <sub>3</sub> /mg DOC average influent: 4.17 mg DOC/L	NDMA decreased from 60 ng/L to 10 ng/L	Biodegradation by microbial population on spent or regenerated GAC can remove NDMA, while regenerated GAC can adsorb chlorinated THM and HAA precursors	Old biologically activated carbon needs to be regenerated in time	( <u>Sun et al. 2018</u> )
Catalysis ozonization	biological activated carbon	ozone concentration: 0.35 mg O <sub>3</sub> /mg DOC, 0.70 mg O <sub>3</sub> /mg DOC or 1.00 mg	lowered the sum of DBP FP by 49.8 %e57.5% at 0.7 mg of O <sub>3</sub> /mg of DOC and 15	1	Ozonation increased toxic potencies partly due to the formation of	( <u>Chen et al. 2019</u> )

		O <sub>3</sub> /mg DOC Treated water: 5.0 ±0.1 mg DOC/L	min empty bed contact time.		brominated DBPs.	
Catalysis ozonization	biological activated carbon	Dissolved ozone dosage was 1.5–2.0 mg/L	Improve the removal of turbidity, DOC, UV254, NH <sub>4</sub> <sup>+</sup> and DON from 98–99%, 58–72%, 31–53%, 16–93% and 35–74%, respectively	Chemically altered the molecular structures of the precursors and increased the biodegradability of N-containing organic compounds	Effective at controlling the formation of the total organic halogen	( <u>Chu et al. 2012</u> )
Catalysis ozonization	Activated carbon	3.0 mg C /L SRNOM solution (with 90 mg/L as CaCO <sub>3</sub> of NaHCO <sub>3</sub> , different concentrations (0, 0.1, 0.4, 1.0 mg/L) of Br <sup>-</sup> ) dissolved ozone dosage was 1–4 mg/L	decrease the overall level of the new DBPs by 94.6% (99.9% for halobenzaldehydes, 99.9% for halobenzoic acids, 93.2% for halosalicylic acids, and 88.9% for halophenol	High molecular weight NOM to small molecules	Effective for controlling new phenolic DBPs via reduction of their precursors	( <u>Huang</u> et al. 2018)