

Recent advances in the characterization and the treatment methods of effluent organic matter

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Abstract. There are many previous review articles available to summarize either the characterization methods of effluent organic matter (EfOM) or the individual control treatment options. However, there has been no attempt made to compare in parallel the physicochemical treatment options that target the removal of EfOM from biological treatments. This review deals with the recent progress on the characterization of EfOM and the novel technologies developed for EfOM treatment. Based on the publications after 2010, the advantages and the limitations of several popularly used analytical tools are discussed for EfOM characterization, which include UV-visible and fluorescence spectroscopy, Fourier transform infrared spectroscopy (FTIR), size exclusion chromatography (SEC), and Fourier transform-ion cyclotron resonance-mass spectrometry (FT-ICR-MS). It is a recent trend to combine an SEC system with various types of detectors, because it can successfully track the chemical/functional composition of EfOM, which varies across a continuum of different molecular sizes. FT-ICR-MS is the most powerful tool to detect EfOM at molecular levels. However, it is noted that this method has rarely been utilized to understand the changes of EfOM in pre-treatment or post-treatment systems. Although membrane filtration is still the preferred method to treat EfOM before its discharge due to its high separation selectivity, the minimum requirements for additional chemicals, the ease of scaling up, and the continuous operation, recent advances in ion exchange and advanced oxidation processes are greatly noteworthy. Recent progress in the non-membrane technologies, which are based on novel materials, are expected to enhance the removal efficiency of EfOM and even make it feasible to selectively remove undesirable fractions/compounds from bulk EfOM.

Keywords: organic matter; wastewater; oxidation; membrane; removal

1. Introduction

Effluent organic matter (EfOM) generally represent the refractory organic fractions in the effluent of biological treatment plants, and it is a heterogeneous mixture of microbial enzymes. For example, terrestrial and marine natural organic matters (NOM), synthetic organic pollutants and by products generated during disinfection (DBP) (Zhao *et al.* 2010). Among which, the microbial enzymes generally comprise of soluble microbial products (SMP) and extracellular polymeric substance (EPS) secreted from metabolic pathways i.e. catabolic and anabolic reactions in microbial enzymes (Sun *et al.* 2011). With the growing population and industrialization, the need for urban

wastewater treatment is highly increasing (Gude 2015a, 2015b, Sun *et al.* 2016). Biological treatments thus far have been considered as conventional, low-cost, and the preferred option to treat urban wastewater (Li *et al.* 2019). However, the intensive use of emerging pollutants, such as cosmetics, pharmaceuticals, detergents, and other refractory organics hampers the effective biological treatment, which results in the effluents with a high proportion of refractory organic matter (Fraia *et al.* 2018). EfOM plays detrimental roles in aquatic ecosystems and has the potential to produce carcinogenic disinfection byproducts (DBPs) during the chlorination of receiving water. In addition, EfOM has a tendency to make complexes with organic micropollutants and metals, and it affects their transport in an aquatic environment (Krasner *et al.* 2009, Lamelas *et al.* 2005, Meinelt *et al.* 2007, Shon *et al.* 2006). To minimize such risks, the discharge limits of EOM have become more restricted in recent years (Mathews and Tan 2016, Qu and Fan 2010).

A lot of effort has been made to understand the variations of the chemical composition of EfOM and to develop the post treatment methods to meet the restricted discharge standards for wastewater. To characterize EOM, the samples are separated into different fractions based on the aromaticity, the functional group, the molecular structure, size based fractions and the fluorescence behavior

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(Michael-Kordatou *et al.* 2015). The characterization of EfOM withholds its importance because the presence of organics in effluent wastewater may define its reactivity and stability in the receiving water body. EOM contains microbial and terrestrial origins of biodegradable and refractory organic fractions, which behave differently to the environment (Derrien *et al.* 2019, Li *et al.* 2020, Michael-Kordatou *et al.* 2015).

The successful characterization of EfOM in biological treatment processes has revealed that various operating conditions have significant effects on the nature of EfOM. For example, Maqbool *et al.* (2017) observed that lowering the organic loading rate (OLR) in a membrane bioreactor (MBR) significantly increased the retention of tryptophan-like fluorescence fraction in EfOM. Ly *et al.* (2018) reported that the activated sludge subjected to high chemical oxygen demand to nitrogen (COD/N) of influent wastewater led to the EfOM enriched with humic-like, fulvic-like, and large sized biopolymer (BP) fractions. It was found that the EfOM of an SBR system was enriched with the organics that were abundant with humic-like and fulvic-like fractions rather than tryptophan-like fraction and marine organic matter (Yu *et al.* 2015).

With the advancement in characterization tools, one can unfold the multifaceted complexion behavior of EfOM with other inorganic compounds, which are mostly found in the biological effluents. For example, characterizing the molecular composition of EfOM via Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR-MS) revealed that the EfOM was enriched with the CHOS formula as compared to the influent wastewater, which was enriched with CHO. The CHOS were related to the complexation of influent organics with surfactants (Wang *et al.* 2018). Yoo *et al.* (2016) discovered that microbial derived that the non-humic hydrophilic fractions in EfOM were responsible for the complexation with heavy metals (Ag and Cu). In another study that employed the fluorescence quenching method (Wei *et al.* 2018), it was disclosed that the fulvic-like fraction of EfOM originating from municipal wastewater was the most susceptible fraction to copper binding.

With the chronic EfOM quality being discharged into the environment, an intensive amount of effort has been concentrated on optimizing and developing the optimum treatment methods. Interestingly, the advancement was greatly assisted by the characterization of EfOM. For example, powdered activated carbon (PAC) is well known to eliminate low molecular weight organic fractions (LMW) from an MBR. Ma *et al.* (2014) purposely modified the PAC by coating the iron oxide particles to its surface in order to adsorb high molecular weight fractions (HMW) to a greater extent (Ittisupornrat *et al.* 2019). In a previous study, the foulants of EfOM present in an ultrafiltration membrane were identified by the characterizations, and the fractions with microbial origin and aromatic proteins were selectively removed by the pretreatments (Chung *et al.* 2019). Another study found that the coagulation process integrated with ozonation effectively removed the HMW fractions in an EOM, which acted like membrane fouling, and they were not readily removed by the conventional

ozonation process (Jeong *et al.* 2014). In a comparison of the advanced oxidation processes, it was revealed that ozone and per-sulphate oxidants were more effective for the removal of humic-like fractions in EfOM, and hydroxyl radicals were more likely to degrade the protein-like fractions (Chen *et al.* 2017). All these studies reflected a strong connection between the advancement in treatments and the characterizations of EfOM for the effective removal of EfOM. Even though many review articles are available to compare the physicochemical treatments for EOM removal and for various EfOM characterization tools, none of them do not particularly aim to disclose the strong benefits of characterizing EfOM for advancements in the physicochemical treatment processes. Therefore, this mini review aimed to highlight the overlooked linkage by selecting some recent literature, which was published after 2010, and present a comparison of the characterizations of EfOM and its removal via the post-treatment processes in parallel.

2. EfOM and its characteristics

Effluent organic matter (EfOM) is considered as complex mixture of organic matters existing in treated outflow of wastewater treatment plants (WWTPs). EfOM can be classified into dissolved EfOM (dEfOM), which is depicted as filtered EfOM fraction by a 0.45 μm membrane filter and particulate EfOM (pEfOM) for the remaining fraction on the filter. The pEfOM includes high molecular weight cellulose fibres, single-celled eukaryotes, bacteria flocs from activated sludge process, phytoplankton with size range from 0.45 to 1000 μm (Shon *et al.* 2006). For dEfOM, there is no established methods to date to fully determine the structures of all constituting compounds, which are merely categorized into the groups of common chemical characteristics. The three major compositions of dEfOM are dissolved natural organic matter (dNOM), soluble microbial products (SMPs) and microcontaminants (Michael-Kordatou *et al.* 2015).

The dNOM is a bulk material of dissolved organic matters containing humic substances (fulvic acid, humic acid and humin) and algogenic substances such as carbohydrates and proteins. The dNOM constituents and features can reflect the origin of the water sources (Murphy *et al.* 2014). The allochthonous dNOM, originating from decaying vegetation or soils, is composed of terrestrially derived organic materials with highly polycondensed and aromatic compounds. In contrast, the autochthonous dNOM with the origin of photosynthesis in aquatic ecosystems comprises recently biologically produced organic matters such as marine humic-like and plankton-derived components (Li *et al.* 2014). Profound research on dNOM requires further dNOM fractionation based on different hydrophobicity and functional groups. As the highest portions of dNOM in water resources, hydrophobic acidic fraction is mostly represented by humic substances, whereas hydrophilic fraction possesses a high content of aliphatic hydrocarbons and organic nitrogen compounds. (Yan *et al.* 2018).

Soluble microbial products (SMPs) occur at the secondary treatment stage of wastewater where bacteria are grown and consume biodegradable soluble organic matters and thus, the produced SMPs from biomass metabolism and decomposition (Barker *et al.* 1999). The significant components of SMPs are saccharides, amino acids, and humic substances. The generation and properties of SMPs are influenced by many operating parameters including the concentrations of substrate and biological mass, the types of bioreactor, the contents of biodegradable components present in the influent, and hydraulic residence time, etc (Liang *et al.* 2007). Esparza *et al.* found that use of sequential batch reactors can reduce SMPs production due to the increase of solid retention time from 3 to 30 days (Esparza-Soto *et al.* 2011). Barker *et al.* in the study of treating municipal wastewater by anaerobic reactor concluded that the production of SMPs was reduced with decreasing temperature and that SMPs at different ranges of molecular weight were formed at different chambers of the reactors (Barker *et al.* 2000). To be specific, lipopolysaccharides (>100 kDa) appeared at the 2nd chamber and carboxylates (<1 kDa) were discovered at the 1st chamber. In general, the number of articles on characterizing SMPs are still limited and the origin of SMPs formation is still a subject of controversy. More studies are required to focus on optimizing the conditions of biotreatment of wastewater concurrently with the reduction of SPSs production.

Detected micro-organic contaminants in effluents encompasses polynuclear aromatic hydrocarbons, polychlorinated biphenyls, endocrine disruptors and many other emerging contaminants such as detergents, organo-fluorine compounds, plasticizers, etc (Khetan *et al.* 2007, Michael-Kordatou *et al.* 2015). With the development of analytical tools and techniques, the microcontaminants can be found in effluent at the concentration lower than ppb level. This suggests that the common treating methods are incapable of totally removing the substances, which can be discharged and raise the concerns relating to environment impacts and human health.

Disinfection by-products (DBPs) generated from disinfection process are also listed as microcontaminants. dNOM serves as the main precursor of disinfection by-products (DBPs) when chemical disinfectants (chlorine, hypo chlorite, bromine chloride, etc) are applied (Krasner *et al.* 2009). More than 600 DBPs have been identified (Richardson, 2003), in which two most common groups of DBPs are trihalomethanes and haloacetic acids. Another group of DBPs commonly detected in effluents are nitroamine compounds due to the interaction of nitrogen-containing organic compounds like SMPs with chloramine. Other halogenated DBPs including brominated and iodinated DBPs have withdrawn more attention when they present strong carcinogenic, cytotoxic or teratogenic effects to health compared to chlorinated analogues (Dong *et al.* 2019). Although the production of DBPs from wastewater disinfection is less noticed than that from drinking water process, alternative methods of using chemical disinfectants have been taken into account to suppress the production of DBPs such as ultraviolet and light-emitting diode disinfection process (Nguyen *et al.* 2019).

Highly interactive capability and complex formation with metals is a remarkable aspect of dEfOM in wastewater effluents. dNOM diminishes the potential bioavailability of the micropollutants to aquatic organisms. On the other hand, it can promote the toxicity by its absorbability on cell membranes. Pernet-Coudrier *et al.* reported that hydrophilic compounds in dNOM including proteins or carbohydrates might play a more notable role in copper complexation compared to the hydrophobic fractions and thus lessen the copper toxicity toward planktonic crustacean *Daphnia magna* (Pernet-Coudrier *et al.* 2008). Worms *et al.* proved that cadmium complexation by dEfOM was greater than that of lead and their intracellular contents in *Chlorella kesslerii* species were reduced due to the complexation. In contrast to the positive role above, dEfOM also exhibited the adverse impacts on marine organisms (Worms *et al.* 2010). Several articles reported regarding the sorption of dNOM on algal membrane and fish gill surfaces, which then modified the membrane structures, permeability of algal membranes, and caused the oxidative stress and physiological disorders of fish gills (Lamelas *et al.* 2009, Cui *et al.* 2014).

3. Characterization tools for EfOM

3.1 UV-visible spectroscopy

It is one of the most commonly used techniques for quantitative determination of organics as well as inorganic compounds in solution. In this tool, the spectrum is created by the absorption of ultraviolet (UV) or visible light by chemical compounds. Its principle follows the Beer-Lambert law. UV-visible spectroscopy has been widely applied in EfOM characterization due to several advantages, such as easy monitoring, rapid responses, simple sample treatments, and low costs. This tool has been used in EfOM investigation for 70 years (Li and Hur 2017). The spectroscopy provides a wealth of information about the aromaticity, the origin, and the reactivities of EfOM (Li *et al.* 2014). Even though it can only detect the light-absorbing compounds in EfOM, several indices derived from the spectra provide a lot of information. For example, the absorption coefficient, the absorption ratios, and the spectral slope have been extensively used to estimate the aromatic content, the humification degree, and the molecular weight (MW) of EfOM. In general, the EOM contains mostly the human and the microbial cell-derived products, which reveal a higher E2 (absorbance at 254 nm)/E3 (absorbance at 365 nm) ratio and a lower UV absorbance at 254 nm per mg carbon (SUVA₂₅₄) (X. Zhang *et al.* 2019). The information of the UV absorption parameters for an EfOM investigation are depicted in Table 1. The high SUVA values suggest the existence of mostly hydrophobic and high MW organic compounds in EfOM (Michael-Kordatou *et al.* 2015). However, this tool does not provide a clear understanding of the whole structural characteristics in EfOM (Michael-Kordatou *et al.* 2015). In particular, this tool cannot detect non-UV absorbing compounds, such as amino acids (Xie *et al.* 2017, Mori *et al.* 2006).

Table 1 Different UV absorption parameters for the EOM analysis

UV absorption parameters	Information	References
A ₂₀₀₋₂₂₆ (UV absorption spectrum)	Amount of N, NO ₃ ⁻ , NO ₂ ⁻ , and other inorganic ions	(Li <i>et al.</i> 2014)
A ₂₂₆₋₄₀₀ (UV absorption spectrum)	Content of Benzene ring containing compounds	(Wang <i>et al.</i> 2009, Huo <i>et al.</i> 2008)
SUVA ₂₈₀ (UV absorption at 254 nm)	Molecular size, aromaticity, and degree of humification	(Chin <i>et al.</i> 1994)
E ₂₅₃ /E ₂₀₃ (ratio of absorbance at 253 nm and 203 nm)	ET band, BZ band, types/substitution in aromatic ring, oxidation of aliphatic chain of aromatic rings into smaller fragments	(Peuravuori and Pihlaja, 1997, Korshin <i>et al.</i> 1997)
E ₂₅₀ /E ₃₆₅ (ratio of absorbance at 250 nm and 365 nm)	Degree of humification and molecular weight of compost OM	(Peuravuori and Pihlaja, 1997, WANG <i>et al.</i> 2009)
E ₄₆₅ /E ₆₆₅ (ratio of absorbance at 465 nm and 665 nm)	Humic substances, condensation degree, molecular structure, extent of polymerization, and aromaticity in compost OM	(Fuentes <i>et al.</i> 2006, Kang <i>et al.</i> 2002, Fialho <i>et al.</i> 2010)
S ₂₇₅₋₂₉₅ (spectral slopes from 275nm to 295 nm)	Content of aromatic carbon, molecular weight, composition of chromophoric OM, and origin	(Fuentes <i>et al.</i> 2006, Fialho <i>et al.</i> 2010, Westerhoff and Anning, 2000, John <i>et al.</i> 2009, Hur <i>et al.</i> 2009)

3.2 Fluorescence spectroscopy

Fluorescence is one of the types of luminescence that shows the emission from the excited single states. It also yields a ground state singlet. Fluorescence is the emission of photons when fluorophores were irradiated with a high-intensity light source. Fluorescence spectroscopy is a favored technique to detect EfOM due to the abundance of fluorescing compounds in EfOM and the high correlations with the bulk organic matters and other indicators (Li *et al.* 2020). Fluorescence spectroscopy has been successfully applied to monitor discharged wastewater into natural systems and to optimize wastewater treatment processes (Goldman *et al.* 2012, Guo *et al.* 2018, Ignatev *et al.* 2019). In two recent decades, fluorescence spectroscopy has undergone tremendous development. The most common display of the fluorescence spectrum is the excitation-emission matrix (Yang *et al.* 2015). The generation of three-dimensional plots enable one to visualize the distribution of multiple fluorophores, but it requires another interpretation method to analyze the immense array of data and to unfold

the meanings behind the fluorescence features. The earliest method, which is called the peak-picking method, is based on the experiences and the recorded data of the distinctive positions of certain components on the EEM map. However, the overlapped components with different physicochemical properties make it difficult to identify the independent components and to compare the different samples in a quantitative way (Goldman *et al.* 2012). To resolve this problem, fluorescence region integration (FRI) was proposed, which the EEM spectrum is divided into individual regions that represent different components/origins. However, the FRI method revealed a strong drawback of assigning a multi-peak component to different components, which is due to the regional integration without considering the peak intensity and the gradient (Murphy *et al.* 2014).

The parallel factor analysis (PARAFAC) is now the most popular chemometric technique to decompose the EEM data into hidden components (Murphy *et al.* 2011, Murphy *et al.* 2014, Yang *et al.* 2015). PARAFAC has been widely employed as a post-processing method to analyze the EOM (Cawley *et al.* 2012, Mostofa *et al.* 2010, Guo *et al.* 2018, Xiong *et al.* 2018). The unavoidable disadvantages can be the multiple-step data processing, such as the reconciliation of dataset, the correction of the spectral bias, the removal of the inner filter effects, and normalization, which make PARAFAC inapplicable for a continuous monitoring (Carstea *et al.* 2016).

The combination of fluorescence spectroscopy with other effective methods, such as chromatography can expand the analytical window into other properties of fluorophores and the associated changing mechanisms. As reported from recent publications (Ignatev *et al.* 2019, Li *et al.* 2014, Xiao *et al.* 2016), using size exclusion chromatography with a fluorescence detector (FLD) permits tracking the changing behavior of similar fluorescent moieties that have different physicochemical features, such as hydrophilicity-hydrophobicity or molecular weight.

Among many analytical tools applied to characterize EfOM, fluorescence spectroscopy can be superior as a monitoring tool due to its benefits that include rapid preparation, reagentless preparation, high sensitiveness, and cost-effectiveness (Murphy *et al.* 2014, Yang *et al.* 2015). With strong correlations between fluorescent intensity and other wastewater quality parameters (BOD, COD and TOC) (Carstea *et al.* 2016) and the convenience in combination with other analytical methods, the prospect of fluorescence-based monitoring tools is expected to focus on developing in-field fluorimeters and enhancing the capacity of EEM data interpretation.

3.3 Fourier transform infrared spectroscopy (FTIR)

Fourier transform infrared spectroscopy (FTIR) is arguably one of the most commonly used spectroscopic instruments after fluorescence spectroscopy. One of the greatest advantages of this technique is that almost any sample in any state can be analyzed (El Fels *et al.* 2015). This spectroscopy exploits the fact that the molecules absorb frequencies that are the characteristics of their structure, which is the vibrational characteristics of

Table 2 Recent progress in SEC for EfOM analysis

SEC techniques	Applications	Results	Reference
SEC-ESI/APCI-MS/MS	Analysis of bound residues <i>via</i> interaction of fungicides, DOM and humic acid	Release of small quantity of dihydroxyanilazine in the low molecular range (originated from adsorbed dihydroxyanilazine)	(Klaus <i>et al.</i> 2000)
HPSEC/UV-vis diode array detectors	Investigation of stormwater quality, parameter correlations, and influences	Elimination of interferences in inorganic components during MW determination/physicochemical characterization (MW and size distribution)	(Huang <i>et al.</i> 2016)
HPSEC-URI	Identification and characterization of biopolymers in water	1.59 for a humic acid, 1.88 for fulvic acid, and 13.5 for BSA as well as Evaluation of functional group properties	(Her <i>et al.</i> 2008)
HPSEC-multiwavelength	Photocatalytic oxidation of OM in surface water	Formation of low molecular weight as well as low aromaticity in oxidation by-products	(Liu <i>et al.</i> 2010)
SEC/HPLC/UV/NDIR	Chemical and physical properties of OM in lake	Reduction of analytical time, enhancement of sensitivity and efficient evolution of molecular size distribution (less than 4000 Da)	(Kawasaki <i>et al.</i> 2011)
HPLC/HPSEC-FLD	Characterization and behavior of OM in wastewater	Evaluation of Physicochemical properties, polarity, and AMW distribution (25 kDa-4 kDa)	(Li <i>et al.</i> 2013)
SEC/LC-OCD-OND/UV	Separation of OM into different fractions based on sizes and chemical functions in surface water	Separation of fractions (Biopolymers, Humic Substances, Building Blocks, Low Molecular-weight Acids, Low Molecular-weight neutrals, and Hydrophobic Organic Carbon) with recoveries of high molecular weight compound (Pullulane: 95.7 to 125.2 %, Dextran: 103 %, and Dextran: 79.3 to 106.3 %)	(Huber <i>et al.</i> 2011)
HPLC-SEC/multiwave absorbance	AMW distribution of OM in river water	Detection of humic substances (16 kD), fulvic acids (11 kD), low AMW acids (5 kD), low AMW molecules (proteins and their amino acid building blocks: 3 kD), and humic substances (6–10 kD)	(Yan <i>et al.</i> 2012)
HPSEC/UV	Determination of MW into OM	Effective way of MW measurement by changing columns unique and demonstration of correlation between SUVA ₂₈₀ and M _n	(McAdams <i>et al.</i> 2018)
HPSEC/UV-DAD/ESI-MS	Relationship between the apparent size distribution (ASW) and molecular complexity in OM (stream water)	Accurate understanding of OM and its influences in natural environments	(Hawkes <i>et al.</i> 2019)

chemical bonds. The resulting absorption is a unique imprint of compounds, which allows the identification of functional groups. Although this technique has the advantage of being applicable to both dissolved and particulate fractions of organic matters, it is currently rarely used alone and preferably combined to other methods, which include NMR and fluorescence, to confirm the interpretation of the results associated with the changes during the biochemical mechanisms and/or to control the quality and the effectiveness of the water remediation processes (Derrien *et al.* 2019). The resulting FTIR spectral pattern is analyzed and compared with the known signatures of the identified materials. An IR spectrum represents a fingerprint of a sample with the absorption peaks that correspond to the vibrational frequencies between the bonds of the atoms that make up the material. Because each individual material is a unique combination of the atoms, no two compounds produce the same infrared spectrum. More information about the functionality of humic substances

(HS) can be acquired from the infrared (IR) spectra. There are several studies that used the IR to understand the changes in the structural characteristics of the HS during the oxidation processes (Maqbool *et al.* 2018, Ly *et al.* 2019). Even though the IR can be used to determine the functional groups, the characterization is mainly qualitative and only some specific bands can be generally clearly identified (Baghoth 2012).

3.4 Size exclusion chromatography (SEC)

SEC is method for separation of different compounds based on their size. It has been widely used to understand the molecular size distribution of EfOM. High-performance size exclusion chromatography (HPSEC) has been extensively applied for this purpose, because it can eliminate the interferences of inorganic components during the MW determination (Huang *et al.* 2016). It is mainly used for the separation of dissolved organic matter (DOM)

according to the apparent molecular weight (AMW). It has a strong point to exclude the inorganic matters (MW<0.25 kDa) from EfOM during the analysis. To get more information about the structural characteristics of EfOM, the SEC has been frequently coupled with different types of detectors.

Her *et al.* suggested the coupling of UV detectors (210 nm and 250 nm) with HPSEC with a UV absorbance ratio (URI) to evaluate the different functional groups of the DOM (Her *et al.* 2008). Such a multi-wavelength SEC system was also utilized by Liu *et al.* to probe the low molecular weight by-products as well as the aromatic structures of wastewater (Liu *et al.* 2010). Huang *et al.* coupled SEC with a UV-vis diode array detector to evaluate the changes in the storm water quality (Huang *et al.* 2016). Meanwhile, McAdams *et al.* suggested the improvement of an accurate MW measurement by changing the standard materials and the column packing (McAdams *et al.* 2018). However, the above-mentioned SEC technologies cannot describe the relationship between the apparent size distribution (ASW) and the molecular complexity of the OM in a natural environment. Moreover, there is a big limitation, because it cannot detect the non-UV absorbing components, which are abundant with functional groups. To overcome the limitations, the SEC can be coupled with a non-dispersive infrared (NDIR) detector, which allows the determination of the chemical and the physical properties of EfOM (Kawasaki *et al.* 2011) despite the low sensitivity. Huber *et al.* reported the successful combining of the SEC with an organic carbon detector (OCD) and an organic nitrogen detector (OND). The SEC-OCD/OND is applied to obtain the isolation of EfOM into different fractions on the basis of the sizes and the chemical functional groups (Huber *et al.* 2011). More recently, an SEC equipped with a UV-DAD and an ESI-MS was proposed to obtain a more detailed chemical composition across different size fractions of the DOM (Hawkes *et al.* 2019). The recent progress in SEC for an EfOM investigation is listed in Table 2.

3.5 Fourier transform-ion cyclotron resonance-mass spectrometry (FT-ICR-MS)

FT-ICR-MS has become a reliable tool for in-depth molecular characterization of complex mixtures, such as EfOM. Over the past decade, FT-ICR-MS had a dominant position with the chemical characterization of EfOM at the molecular level. In combination with electrospray ionization (ESI), this method provides the necessary resolution to determine with high accuracy from hundreds of ions to several thousands of ions with an *m/z* range that is typically from 200 to 1000 Da. Furthermore, due to the high mass resolution and accuracy (less than 0.5 ppm), elemental formulas are assigned with a high level of reliability (Ly and Hur, 2018, Derrien *et al.* 2019). Since an ESI-FT-ICR-MS analysis generates a large amount of data and empirical formulas, the data is commonly classified into 6–8 main classes of compounds, which include lipids, proteins, carbohydrates, unsaturated hydrocarbons, lignin's and/or carboxyl-rich alicyclic molecules (CRAM), tannins,

and condensed aromatic compounds, in accordance with their H/C and O/C ratios. An example of this is the van Krevelen diagram. Some indices related to the degree of unsaturation (DBE: double bond equivalent index) and/or the aromaticity of the formulae (AI or AI_{mod}: modified aromaticity index), which have also been developed. A comparison of the samples on this molecular specificity scale made it possible to identify the specific classes of the compounds in accordance with their sources or production methods (Koch *et al.* 2005, Koch and Dittmar, 2006) as well as to highlight the extreme isomeric complexity in the DOM across aquatic environments (Hawkes *et al.* 2018). Currently, this method of organic characterization is considered as the most powerful tool that can be used to characterize the structures and the molecular properties of EfOM.

4. Major treatment processes for EfOM

The removal of EfOM during wastewater treatment mainly depends on the characteristics of the present organic matter, its concentration, and the removal methods applied. HMW EfOM fraction is more amenable to removal than low molecular weight (LMW) fraction, predominantly with MW of 500 Dalton (Da). Many treatment methods have been used to remove EfOM during wastewater treatment with a fluctuating degree of success.

4.1 Enhanced coagulation

Organic matter removal in a conventional water treatment process may be achieved through the addition of a chemical coagulant. Coagulation with aluminum and iron salts is effective in the removal of EfOM, which is measured by the total organic carbon, and the removal efficiencies in the range of 25% to 70% (Owen *et al.* 1993). Coagulation removes the hydrophobic fraction and the high molecular weight EfOM in preference to the hydrophilic fraction and the low molecular weight compounds (Owen *et al.* 1993). The former is composed of primarily HS (fulvic and humic acids), which are rich in aromatic carbon and phenolic structures, but the latter is mainly composed of aliphatic and nitrogenous organic carbon, such as carboxylic acids, carbohydrates, and proteins. Enhanced coagulation for the removal of EfOM requires elevated coagulant doses, which is 5–100 mgL⁻¹ for Al and Fe salts. However, the increased coagulant dose leads to excess sludge production and an increased cost of pretreatment particularly for low alkalinity waters. Enhanced coagulation is recommended for waters with hydrophobic and relatively high molecular weight OM, which is indicated by moderate to high specific ultraviolet absorbance values (SUVA) (Bagthoth 2012).

4.2 Adsorption processes

Adsorption is another effective and easy method for EfOM removal, which can be set up for different flow rates and a wide range of EfOM concentrations (Bhatnagar *et al.*

Table 3 Summary of recent applications of adsorption in EfOM treatment

Methods	Effluents	Effluent removal	Results	Reference
<i>Carbon based materials</i>				
GAC	Pulp and paper industry effluent	46.72 mg HA /g GAC	- Reducing time and operated energy for electrocoagulation. - Optimum condition: pH 4 and 150 mg/L Na ₂ SO ₄ .	(Barhoum <i>et al.</i> 2019)
Coconut PAC	Cork effluent	455 mg/g PAC (COD)	- Optimum condition: pH 3.5, 1g AC for 0.2 L of effluent, adsorption time of 10 min - Proposal of microwave-assisted regeneration	(Ge <i>et al.</i> 2018)
Magnetic PAC	Membrane bioreactor effluent	12.5 mgC/g PAC	- 4 g/L dosage quickly adsorbed EOM in 5 min of contact	Ittisupornrat <i>et al.</i> 2019)
GAC	Wastewater treatment plant effluent		- Smaller-size fractions of EOM performed a strongly competitive impact onto micro-pollutant adsorption. - Ozone degraded EOM, reduced their molecular weight and aromaticity and thus, diminished their adsorbability on AC - The removal of EOM hydrophilic fractions depended on degree of functionality.	(Zietzschmann <i>et al.</i> 2015)
CNTs, functionalized CNTs and functionalized biochar	Biological treatment plant effluent	$q_{\max}(\text{BC}) = 7.81 \mu\text{g/g}$, $q_{\max}(\text{CNT}) = 39.29 \mu\text{g/g}$ $q_{\max}(\text{CNT-COOH}) = 36.23 \mu\text{g/g}$ $q_{\max}(\text{CNT-OH}) = 40.68 \mu\text{g/g}$	- Adsorbability of hydrophobic fractions was highest for original CNTs. - Hydrogen Bonding, Electron Donor - Acceptor and electrostatic attraction were primary adsorption mechanism.	(Almed <i>et al.</i> 2015)
Magnetic biochar	Municipal effluent	$q_{\max}(\text{MBC}) = 56.14 \text{ mgC/g}$	- EOM removal order: fulvic, humic-like > protein-like component. - EOM (MW<500 Da) were removed most efficiently.	(Wei <i>et al.</i> 2016)
Nanosized SWCNTs and MWCNTs, expand graphite	Sewage treatment plant effluent	$q_{\max}(\text{SWCNT}) = 16.22 \text{ mgC/g}$, $q_{\max}(\text{MWCNT}) = 19.27 \text{ mgC/g}$, $q_{\max}(\text{EG}) = 19.27 \text{ mgC/g}$,	- Nanosized MWCNTs was the highest EOM adsorption capacity. - Microbial products, humic and fulvic components were removed most by nanosized SWCNTs. - Both materials were preferred to adsorb large size EOM portions	(Jeong <i>et al.</i> 2017)
<i>Mineral and polymeric adsorbents</i>				
Fly ash	Secondary wastewater	0.2 mgC/g Fly ash	- At the optimum dose, 15 g/L fly ash can adsorb 25% of DOC (12.9 mg/L) and preferentially removed hydrophilic EOM fractions.	(Wei <i>et al.</i> 2011)
Nanosized TiO ₂	Olive effluent	450 mg/g n-TiO ₂ (COD)	- The effect of adding salt, temperature, pH, adsorbent dose was investigated - No experiment described the reusability of the adsorbent	(Bsoul <i>et al.</i> 2019)
Modified polystyrene HPA)	Coking effluent (A-after biological treatment)	52.2 mgC/g A-HPA	Adsorption mechanism based on synergetic effects of π - π interactions, acid-base interactions and micropore filling data of 3-years in situ monitoring indicated the effective performance of the recycled adsorbent without remarkable capacity loss	(Yang <i>et al.</i> 2017)
Purolite A502PSM and GAC	Municipal effluent	$Q_{\max}(\text{GAC}) = 13.4 \text{ mgC/g}$	- GAC was more effective than A502PS in fluidized bed. Application of plug flow model in predicting the influence of operating conditions.	(Shanmuganathan <i>et al.</i> 2014)

2017). The recent applications of adsorption in an EOM treatment are summarized in Table 3. The carbon-based materials are the most widespread adsorbents applied in treating EfOM. The materials include activated carbon, carbon nanotube, graphene, and biochar and their derivatives, which have a large surface area, a compatible sp² hybridization structure with EOM, an abundance of porosity, and the capacity of being activated and structurally modified by physical and chemical processes to alter their functions. Activated carbon (AC) commonly signifies carbon-rich material possessing sp² hybridized planar layers (Gamal *et al.* 2018). From the physical aspect, AC was classified to powdered AC (PAC), granulated AC (GAC), spherical AC, metal-impregnated AC, polymer coated carbon based on their particle size, and

characteristics of specific surface area (Gamal *et al.* 2018). Utilization of the AC as an adsorbent for EfOM removal has been widely conducted for many years in the past. Recently, researchers focused mainly on treating EfOM by combining adsorption on the AC with other methods. Afef Barhoumi *et al.* studied the removal from wastewater that contained a high amount of humic acid from the pulp and paper industry by combining electrocoagulation using Al electrodes with granulated AC adsorption. This combination showed an economic improvement to treat certain wastewater via reducing time and operated energy with optimum conditions (Barhoumi *et al.* 2019). Ge *et al.* introduced the microwave-assisted regeneration of coconut powder AC in the purification process of cork effluent after a coagulation treating step (Ge *et al.* 2018). Fast AC-based

adsorption performance was reported using magnetic powdered AC in the study of Suda *et al.* for the pretreatment of a membrane bioreactor effluent (Ittisupornrat *et al.* 2019). Zietzschmann *et al.* in their studies investigated the competitive adsorption of EfOM and the micro-pollutants on granular AC. They suggested adding ozonation to increase the efficiency of the organic micropollutants removal by the AC (Zietzschmann *et al.* 2014, Zietzschmann *et al.* 2015). The results are promising, and they explained that ozone degraded HMW EfOM compounds and thus diminished their adsorbability on the AC. Besides the AC, carbon nanotubes (CNTs) and biochar are carbonaceous materials to be applied as the adsorbents for EfOM removal. Biochar is a solid by-product from the thermochemical process of the biomass (Lehmann 2011). The adsorptive removal behaviors of EfOM from the biological treatments were investigated using CNTs, functionalized CNTs, and functionalized biochar (Almed al., 2018). Dong Wei *et al.* synthesized the magnetic biochar to examine the capacity of adsorptive removal towards different EOM fluorescing components (Wei *et al.* 2016). The EfOM removal occurred following the order of fulvic and humic-like components > protein-like component. Jeong *et al.* used nanosized SWCNTs and MWCNTs to compare their adsorbability with EfOM from a sewage treatment plan (Jeong *et al.* 2017). Nanosized MWCNTs was the most effective in the study.

Besides carbonaceous materials, other synthesized materials or natural materials have been applied to treat EfOM. In the study of Wei *et al.*, fly ash was utilized to adsorb secondary wastewater (Wei *et al.* 2011). At the optimum dose of 15 g/L, the adsorbent removed 25% of EfOM in terms of the dissolved organic carbon (DOC). Abeer *et al.* used nanosized TiO₂ as an EfOM adsorbent to control an olive mill effluent. The effect of adding salt, temperature, pH, and an adsorbent dose were investigated to find out the optimal operation condition (Bsoul *et al.* 2019). It can be concluded from the recent studies that although different types of adsorbents, which encompassed from organic to inorganic materials, were introduced for EfOM removal, and some of the novel materials showed low adsorption efficiencies compared to the common adsorbents, such as activated carbon or nanoparticle Fe₃O₄.

4.3 Ion exchange processes

In the water treatment process, the ion exchange (IE) technique can be described as a reversible transfer of ions between the resins and the solution. Compared to coagulation, IE is more efficacious with removing charged EOM components (Chen *et al.* 2018). The comparative experiment of humic acid removal by adsorption on different materials which includes AC, metal oxides, and IE resin, demonstrated that the IE process was the most effective solution (Fettig *et al.* 1999). In an effluent treatment, the magnetic ion-exchange resin (MIEX) was the most commonly used resin. The core of the resin was the magnetic ion oxide, which was covered with polyacrylic polymer in a chloride form. The resin allowed the exchange of chloride anion with anion organic matter, and the magnetic iron oxide promoted rapid aggregation and

settlement throughout adsorption. (Drikas *et al.* 2011). Nguyen *et al.* tested the MIEX ability with removing EfOM from a bio-treated effluent when conducted in batch and fluidized bed column modes (Nguyen *et al.* 2011). In a batch experimental setup, resin with a 10 mg/L dose can remove 77% of EOM (DOC) after 0.5 hours much more effectively than polystyrene resin. In a fixed bed column, 60% DOC of sewage effluent was removed after a 170-bed volume, and MIEX still performed stable EOM removal efficiency after 4 times of regeneration. Fan *et al.* compared the removal of textile EOM fractions by MIEX with AC. The results demonstrated that MIEX was more effective than AC (15% DOC higher) (Fan *et al.* 2014). With a major removal effect toward ionic organic substances, the ion exchange method was reported to show a synergetic effect with treating effluent organic matters in combination with other methods. Kim and Dempsey found that MIEX was more useful in reducing the membrane fouling in microfiltration and ultrafiltration than other resins, such as IRA-958 or DAX-8. (Kim *et al.* 2010) They concluded that MIEX removed most of the acidic EfOM, which was the main factor that caused membrane fouling. Alternate options to conduct MIEX displayed similar DOC removal but lead to different filtration resistance. Chen *et al.* investigated the impact of pH on the EfOM removal by MIEX-ozonation operation (Chen *et al.* 2019).

Besides MIEX, novel resins were synthesized and studied to treat EOM. Ahmad employed Purolite®A500PS in a fluidized bed mode to treat synthetic wastewater (Ahmad *et al.* 2012). The resin expressed steady EfOM removal effectiveness with an 80% DOC reduction of the first 900 bed volumes of wastewater. The particle dimension exerted a strong impact on EfOM removal. The resin in size range of 150-300 µm showed the highest performance and can remove 98.4% of biopolymer, 87% of humic substance, and 83% of low MW neutral fraction (Bassandeh *et al.* 2013). Other kinds of conventional anionic resins, which include Amberlite IRA-401 and Dowex-1x4 (Sun *et al.* 2015), and resins based on polymer structures, which include styrene-divinylbenzene, styrene-acrylate divinylbenzene (Abreu *et al.* 2018), Tianjin 717, and Tianjin D301R (Sun *et al.* 2018) were also studied on the EfOM removal effectiveness toward treating petroleum refinery effluents or a second effluent in a sewage treatment system.

It was noticed that the IE technique was reported to effectively treat EfOM from various types of effluent. Most of the resins seemed to only greatly absorb anionic organic components, which are required to integrate with other methods, such as coagulation or ozonation for the optimal treating the EfOM. Other common drawbacks were reported as blinds active groups when resins activity was affected by the phosphate accumulation on the porous resins (Levchuk, *et al.* 2018), biofilm growing on the working surface, and ungenerated resins due to blocking by recalcitrant organic substances (Ciputra *et al.* 2010). Since a recent study introduced nanosized ion exchange resin, which exhibited outstanding adsorption capacity and reach equilibrium in only 10 seconds, nano-resins are expected to be a novel attracted trend for scientists to conduct further studies to improve the IE techniques in removing EfOM (Johnson *et al.* 2016).

4.4 Advanced oxidation processes

Advanced oxidation processes (AOPs) have been widely applied in wastewater treatments for decades (Klavarioti *et al.* 2009). For AOPs, $\bullet\text{OH}$ radicals are one of the most powerful active species in EOM removal. This non-selective oxidant produces several reactions in photolysis, ozonation, and the heat treatment process (Matilainen and Sillanpää, 2010). From the view of mechanism related EfOM removal by AOPs, $\bullet\text{OH}$ radicals can attack the double bond that causes the abstraction of the H-atom from the carbon containing alternate sigma and the pi-bonds. In this reaction, carbon centered radicals, which include carbocations and carbanion, are generated. The formation of a positive charge or a negative charge in the carbon makes it more reactive, so it can further react with oxygen to produce peroxy radicals. These types of reactive oxygen species are responsible for the decomposition of EfOM. Moreover, different parameters, such as pH, temperature, nature of pollutants, ions, and scavengers can change the rate of oxidation via $\bullet\text{OH}$ radicals (Michael-Kordatou *et al.* 2015, Wang and Xu, 2012, Hodges *et al.* 2018, Dong *et al.* 2010).

For a few years, UV-based applications are extensively used for EfOM treatments. In this process, the absorption of emitted light (mercury vapor lamps or natural sunlight) can produce a triplet excited state of the EOM (McNeill and Canonica, 2016, Michael-Kordatou *et al.* 2015). During the photolysis process, singlet oxygen ($^1\text{O}_2$), $\bullet\text{OH}$ and peroxy radical ($\bullet\text{OOR}$) are responsible for the mineralization of EfOM. In addition, the researcher enhances the photooxidation ability of EfOM by using H_2O_2 in the light source (UV/ H_2O_2) (Umar *et al.* 2015). However, this system suffers from being unstable, overheating, less photonic efficiency, and a short lifetime period. In order to solve this problem, a LED light source is used instead of Hg lamps. However, it consists of high costs and low efficiency as compared to Hg UV lamps (Michael-Kordatou *et al.* 2015).

To enhance the oxidation tendency of EfOM, researchers use an ozone-based application, because O_3 is a strong oxidant. It can destroy various Gram-positive and Gram-negative pathogens in biological wastewater as well as organic matter in EfOM. O_3 enhances the oxidation process with the generation of $\bullet\text{OH}$ radicals. O_3 reacts with the double bond of alkene in EOM to form ozonide, which is further converted into aldehyde or ketone that contains organic compounds. The high MW hydrophobic organic compounds are changed into low MW. It significantly reduces the aromaticity (Lee and Von Gunten, 2016, Gassie and Englehardt, 2019, Wang and Xu, 2012). The oxidation efficiency can be increased by combining O_3 with UV/ H_2O_2 . This ozone-based application depends upon the dose, the concentration, the pH, and the lamps (Michael-Kordatou *et al.* 2015, Trigueros *et al.* 2019, Jung *et al.* 2017, Guzmán *et al.* 2016). The ozonation process is also increased by using a catalyst, which is catalytic ozonation. It degrades the organic matter rapidly (Manickavachagam *et al.* 2015, Hodges *et al.* 2018). However, the stability and the reuse of the catalyst limits its application. Also, the

ozone-based bromine waste water treatment process is an effective process for decomposing the high MW into low MW organic fragments (Jung *et al.* 2017, Guzmán *et al.* 2016, Michael-Kordatou *et al.* 2015). However, the formation of cariogenic compounds is the main problematic issue in this process.

The oxidation tendency of EfOM can be further enhanced by using Fenton and photo-Fenton. In this process, the Fe^{3+} -EOM complex (stable and soluble) compound is formed, which can involve a further reaction during the wastewater treatment process. Also, the protein like substances and the soluble microbial products are easily removed with this process. The oxidation efficiency of the OM depends on the concentration ($\text{H}_2\text{O}_2/\text{Fe}^{3+}$), the pH, and the reaction time (Asaithambi *et al.* 2017, Poblete and Pérez, 2020, De la Cruz *et al.* 2012). Nowadays, researchers pay more attention towards the semiconductor photocatalytic oxidation process for removing organic pollutants from water, because it is green technology and utilizes the natural energy from sunlight (Iboukhoulef *et al.* 2019). However, this system requires additional post-separation and reuse steps. As a result, immobilized photocatalytic membrane can solve the issues of post-separation and reuse (Michael-Kordatou *et al.* 2015).

Persulfate decontamination technologies is one of the advanced oxidation processes that is applied for the removal of organic contaminants from wastewater. It is driven by the radical or the electron transfer process. In this process, secondary oxidants (CO_3^{2-} , $\bullet\text{OH}$, $\text{O}_2^{\bullet-}$, and $^1\text{O}_2$) can affect the transformation efficiency as well as the products. However, changes in the pH and the production of hazardous contamination are the problematic issues in this process (Wacławek *et al.* 2017, Zhou *et al.* 2019, Ghauch *et al.* 2015, Wang *et al.* 2018). The electrochemical oxidation (EO) process is also used for the water treatment process (de Oliveira Marcionilio *et al.* 2019). It consists of direct (anodic: boron-doped diamond, Ti_4O_7 , transition metal/rare earth doped $\text{PbO}_2/\text{SnO}_2$, Pt decorated etc.) and indirect (electrogenerated active chlorine) oxidation. Also, photoelectron-Fenton and solar electro-Fenton processes are reported in electrochemical oxidation treatment (Martínez-Huitle and Panizza, 2018). The recent progress in AOPs for EfOM treatment is shown in Table 4.

4.5 Membrane separation processes

Organic pollutants in secondary effluent have become one of the most serious environmental problems due to their persistence, toxicity, and being bio-refractory. Traditional processes, such as adsorption, AOP, and coagulation are not very effective for the complete degradation or removal of organic contaminants. These technologies mainly required complicated equipment, high energy consumption, and high operating costs. Also, some of them require large amounts of chemicals, which leads to the production of waste and sludge (Pan *et al.* 2019). In this context, membrane technology is considered one of the most promising methods for water decontamination due to its advantages of high separation selectivity, low energy consumption, less requirements for additional chemicals, ease of scaling up,

Table 4 Recent progress in AOPs for EfOM treatment

AOPs	Results	Reference
O₃ (Ozonation)		
O ₃ (2.24 ± 0.17 mgL ⁻¹)	Removal of dissolved EOM (oxidation of HOA and HON fractions) from wastewater treatment plant	(Jin <i>et al.</i> 2016)
O ₃ (0-0.25 mg)	Removal of fulvic acid at UV ₂₅₀₋₂₆₀ from EOM	(Yu <i>et al.</i> 2019)
O₃/ H₂O₂/UV		
UV/H ₂ O ₂ (8000 mgL ⁻¹)/Fe ³⁺ (170 mgL ⁻¹)	93 % COD and 97 % color removal of OM in textile effluents (120 min)	(Trigueros <i>et al.</i> 2019)
O ₃ (15.3 mg)/H ₂ O ₂ (36 mgL ⁻¹), UV/H ₂ O ₂ or UV/O ₃ (62.4 mgL ⁻¹)	50 % humic acid removal of total EOM in WWTP	(Hofman-Caris <i>et al.</i> 2017)
O₃/Catalyst (Catalytic ozonation)		
(LaCoO ₃ : 0.25 gL ⁻¹ / O ₃ : 1 mgL ⁻¹)	Degradation of benzotriazole (100 µgL ⁻¹) into aldehyde (87. 9 µgL ⁻¹) in 90 min reaction time from secondary effluent matrix and EOM	(Zhang <i>et al.</i> 2019)
O ₃ (0-1.64 mg)/activated carbon	Fragmentation of humic acid organic micro pollutants (Acesulfame: 20 µgL ⁻¹ , benzotriazole: 19 µgL ⁻¹ , bezafibrate: 38 µgL ⁻¹ , carbama-zepine: 21 µgL ⁻¹ , diclofenac: 24 µgL ⁻¹ , 4-Formylaminoantipyrine: 20 µgL ⁻¹) into smaller components from	(Zietzschmann <i>et al.</i> 2015)
MnO ₂ /O ₃	Removal of EOM (TOC: 13.24 %, UV ₂₅₄ : 60.83 %, colority: 85.42 %,)	(Wen <i>et al.</i> 2018)
O₃/Fenton/UV/ H₂O₂/Cl₂		
O ₃ /H ₂ O ₂ /MW/PS	66.93 % (O ₃ /H ₂ O ₂) and 86.06 % (MW/PS) for elimination of refractory organics in landfill leachate	(Chen <i>et al.</i> 2019)
O ₃ (4 mgL ⁻¹)/Fe ³⁺	Removal of EOM (significant decrease in high MW fraction) in wastewater	(Jeong <i>et al.</i> 2014)
Solar photo-Fenton/ O ₃	Removal of OM (76.4 % COD, 74.9 % color, 50 % nitrate, 12.8 % ammonium, and 73.3 % humic acid) in landfill leachate	(Poblete and Pérez, 2020)
Fe ³⁺ (0.6 mM)/Fe ²⁺ (40 mgL ⁻¹)/UV/ H ₂ O ₂ (320 mgL ⁻¹)/NaOH	0.09 mgL ⁻¹ residual nickel and > 58 % TOC removal form carboxyl complexed Ni containing synthetic and authentic effluent	(Jiang <i>et al.</i> 2019)
Fenton/ozonation (0.0-10.0 g h ⁻¹)/ H ₂ O ₂ (97.2 mM)/UVQS	ΔUV ₂₅₄ : 0.016ΔCOD (ozonation) and ΔUV ₂₅₄ : 0.011ΔCOD (Fenton) in municipal landfill leachate	(Jung <i>et al.</i> 2017)
O ₃ /UV (254 nm)/H ₂ O ₂ (15,937 mgL ⁻¹) /Solar light/Fenton (510 mgL ⁻¹)	76 % COD and 53 % DOC removal in citrus wastewater (30 min reaction time)	(Guzmán <i>et al.</i> 2016)
Coupled Fenton-Denitrification (CFD) (Fe ³⁺ /H ₂ O ₂ : 50 % wt. ratio)	90 % TOC and 75 % nitrogen removal from coking plant (72 h batch bioreactor and 40 days pre-acclimated denitrifying biomass)	(Razaviarani <i>et al.</i> 2019)
Photodegradation/photocatalyst/S₂O₈		
Solar light (photodegradation)	Photodegradation of F1 and F2 fraction (C1: 81.24, C2: 86.41 and C3: 51.16%) in EOM	(Zhang <i>et al.</i> 2019)
(photodegradation)	Photodegradation of micropollutant (sulfamethoxazole, sulfadimethoxine: 75% effluent/25% river water via •OH, cimetidine: >95 % via ¹ O ₂ and caffeine: >95 %)	(Bodhipaksha <i>et al.</i> 2017)
BiFeO ₃ photocatalyst (0.6 g)/O ₃ (600 mgL ⁻¹)/S ₂ O ₈ (0.05 M)	82.9 % and 98.0 % decrease in phenolic compounds and COD, respectively for degradation of olive Mill wastewater	(Iboukhoulef <i>et al.</i> 2019)
UV/Chlorine/PAC/ultrafiltration	54 % removal (UV ₂₅₄) in algal polluted water	(Xing <i>et al.</i> 2019)
Photo-electrochemical oxidation		
Photo-electro-Fenton (PEF) (H ₂ O ₂ : 9000 mgL ⁻¹ , Fe ²⁺ : 60 mgL ⁻¹ , electrolysis time: 45 min, pH: 3.5-4.5, current: 2.3 A	89 mg Pt-CoL ⁻¹ of color, 254 nm = 0.18 a.u., 370 mgL ⁻¹ of TC, 315 mgL ⁻¹ BOD ₅ , 782 mgL ⁻¹ COD, and BOD ₅ /COD = 0.4 removal in sanitary landfill leachate	(Seibert <i>et al.</i> 2019)
Electrochemical oxidation (BDA) under hydrodynamic condition	complete mineralization (Re > 2000) of 1-butyl-3-methylimidazolium chloride solutions	(de Oliveira Marcionilio <i>et al.</i> 2019)

and continuous operation (Pan *et al.* 2019). Basically, membrane is classified into organic (polymer: cellulose, PAN, PVDF, PP, PVA, PI, PTFE, PES, PSU etc.) inorganic (ceramic: metal oxide, metal carbide, zeolite etc., metal: porous metal/dense metal, carbon: graphene, CNTs, coal etc. and hybrid membrane). In addition, membrane filtration is classified as microfiltration (MF), ultrafiltration (UF), nano filtration (NF), forward osmosis (FO), and reverse osmosis (RO). MF and UF membrane systems have already proven their advantages in terms of economic efficiency as well as product water quality. NF and RO membranes are also used in a broad range of wastewater reclamation. However, these membrane processes often suffer from their own membrane fouling, which consequently decreases the permeate flow (Shon *et al.* 2011).

Membrane fouling has been a major challenge for the better and continuous operation of the membrane processes. Membrane fouling can occur due to many reasons, which includes 1) biological fouling that is due to the unwanted growth of biological species on the membrane surface that blocks the membrane pores and ultimately reduces the permeate flow while increasing the pressure, 2) colloidal fouling, which also results the loss of permeate flux through the membrane, 3) organic fouling due to the deposition of organic substances, and 4) scaling, which is the deposition of the mineral or inorganic substances on the membranes surface (Shon *et al.* 2011).

Moreover, "The materials accumulated on a membrane surface, which cannot be removed by cross-flow, backwashing or back pulsing, can be named as irreversible fouling, resulting in permanent permeability loss." Fouling that results from concentration polarization has been considered a loose reversible fouling layer. However, if this layer reaches a critical concentration, a condensed layer may form, which can become irreversible (Holman *et al.* 2007). Organic fouling is the initial source of membrane fouling, which is associated with the molecular size, its shape, and the chemical characteristics of the organic matter. The organic fouling can occur due to precipitation, adsorption, and the interaction with cations. Normally, irreversible fouling is caused by organics and needs careful chemical treatment (Shon *et al.* 2011). The membrane fouling mechanism is different from MF, UF, NF, and RO, which is mainly related to the size of the organic matter. Fouling in MF and UF is significantly affected by suspended solids, particulate organic and large organic matter. On the other hand, NF and RO, which are non-porous and meet smaller sizes of organic matter due to pretreatment, are caused by less than 30kDa of organic molecules.

EfOM contains additional bulk organic matter in the form of soluble microbial products (SMPs) derived from the microbiota in the biological treatment. SMPs are the mixture of organic compounds, which are highly diverse nature constituents with a broad spectrum of physicochemical properties, such as humic-like substances, proteins, polysaccharides, lipids, and DNA (Ly *et al.* 2019). Particularly, the extracellular substances produced by the microorganisms in the biological treatment of the wastewater appear to play a vital role in membrane fouling.

Evenblij *et al.* investigated the influence of substrate conditions on the production of extra-cellular polymeric substances (EPS), and they suggested that the optimization of the biological processes could result in lower EPS production, which would consequently affect the fouling (Evenblij *et al.* 2005). Rosenberger *et al.* (2006) found for two parallel membrane bioreactors that macromolecules, which included polysaccharides, proteins, and organic colloids with a molecular weight of >120 KDa, make a significant contribution to membrane fouling. At higher concentrations of these substances, higher fouling rates were observed (Laabs *et al.* 2006). Although many attempts have been made to control membrane fouling, it is still the principal limitation of a membrane's performance. In addition to proper design parameters, the fouling potential can be controlled by other selected mechanisms, which include chemical cleaning, hydraulic cleaning, and pretreatment, such as the chemical addition in the feed stream. The methods to reverse or prevent fouling are dependent on the type of fouling.

5. Conclusions and future directions

EfOM adversely affects water treatment processes involving coagulation, oxidation, adsorption, and membrane filtration. This influences the color, taste and smell in the final treated water and can serve as a precursor for the formation of DBPs. The biodegradable EfOM fraction can contribute to the growth of microorganisms in water distribution networks, predominantly in the systems which do not maintain a residual disinfectant in the distribution networks. The efficiency of water treatment is affected by both the quantity and composition of EfOM. Through systematic characterization of NOM, problematic fractions can be identified and targeted for the removal and/or transformation. Therefore, the proper characterization of EfOM or the wastewater at different individual treatment steps can be an important step stone for the selection of water treatment processes.

Among the many analytical tools applied to characterize EfOM, fluorescence spectroscopy can be superior as a monitoring tool due to the benefits of rapid preparation, reagent less preparation, a high sensitiveness, and cost-effectiveness. Recently, much effort has been made to diversify the detection objects contained in the heterogeneous EfOM by coupling advanced detectors with SEC systems to probe the variations of the chemical functionalities of EfOM across different size fractions of EfOM. Despite the advancements, there is no doubt that FT-ICR-MS is the most powerful tool to characterize the EfOM at molecular levels. Enhanced coagulation is recommended for the removal of relatively hydrophobic and high molecular weight EfOM fractions. Even though there are many reports available to introduce the alternative adsorbents based on novel materials, many of them have shown lower removal efficiencies for EfOM compared to the conventional adsorbent activated carbon. Ion exchange is found to be advantageous for the selective removal of certain EfOM components. A variety of AOP has been

suggested to enhance the oxidation efficiency towards EfOM, which include Fenton and photo-Fenton oxidation, persulfate-assisted oxidation, and photoelectron-Fenton and solar electro-Fenton processes. Despite the proven removal efficiency of membrane filtration, membrane fouling mitigation is still hampering its effective operation. The cost-effective pre-treatment method should be developed to cope with the persisting problem.

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