



## Distribution of dissolved organic matter fractions along several stages of a drinking water treatment plant



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### ABSTRACT

The present study was conducted to track dissolved organic carbon (DOC) through the different stages of a drinking water treatment plant that includes a conventional treatment CT (coagulation/flocculation, sedimentation and sand filtration) and two parallel advanced treatments consisting of ozonation plus activated carbon filtration (AT1) and ultrafiltration (UF) and reverse osmosis (RO) (AT2), respectively. Fractionation of DOC was performed by high-performance size exclusion chromatography (HPSEC), whereby DOC fractions are separated according to their molecular weight. Results demonstrated that the dominant fraction in the raw feed water was constituted by humic substances (HS) (28–54%), followed by the fractions building blocks (BB) and low molecular weight neutrals (LMWN) (20–25%). The fraction with highest molecular weight constituted by biopolymers (BP) was found in the range 4–8%. These fractions exhibited different treatability along the treatment units. High molecular weight compounds (fractions BP and HS) were more effectively removed by CT than low molecular weight compounds (fractions BB and LMWN), which were better removed by AT1 and AT2 than CT. Differences in DOC fractions removal were also observed between AT1 and AT2. The passage through AT1 led to a decrease of BP and HS during ozonation, in opposition to BB and LMWN, which were removed to a greater extent during GAC filtration. In AT2, BP was removed by UF and RO, while HS, BB and LMWN were marginally removed by UF but largely by RO. Backwash stream water from UF membranes was also analysed to provide insight into the fouling reversibility of DOC fractions.

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### 1. Introduction

The effective removal of natural organic matter (NOM) from water is nowadays one of the main challenges faced by drinking water treatment plants (DWTPs) worldwide. NOM can (i) cause aesthetic problems to the produced water, (ii) lead to the formation of undesirable disinfection by-products (DBPs), (iii) adversely affect the performance of DWTP processes, (iv) serve as a media for the transport of pollutants, and (v) promote biofilm growth in the distribution system, threatening the safety of the distributed water [1,2].

Controlling NOM in DWTPs is crucial to minimize the above-mentioned effects and comply with the latest stricter regulations for drinking water treatment. General practice for NOM removal currently comprises the combined use of conventional treat-

ments (coagulation/flocculation, sedimentation, sand filtration) and advanced treatments (ozonation, activated carbon (bio) filtration and, more recently, membrane filtration) [1,3,4].

NOM is ubiquitous in aquatic ecosystems. It comprises a heterogeneous mixture of organic compounds with very different physicochemical properties, including humic and fulvic substances, proteins, amino acids, lipids, polysaccharides and biopolymers, among others [5–7].

NOM is characterized and quantified with parameters such as total organic carbon (TOC), dissolved organic carbon (DOC), UV-absorbance at 254 nm (UV254), color or KMnO<sub>4</sub> number [6,8]. However, there are no established relationships between parameters for quantifying NOM and, in practice, none alone can predict NOM behavior in a given system. The reason of such variability is that these parameters do not take into account the different constituents of NOM, which can differ in their removal in the different treatment stages, reactivity toward chemicals commonly added in DWTPs such as coagulants and disinfectants, potential of DBP formation, adsorbability onto sorbents used in DWTPs,

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biodegradability and potential of membrane fouling [6,9–13]. Characterizing NOM content along a DWTP is thus essential for a better understanding and optimization of the performance of each treatment unit process (e.g. coagulant and disinfectant doses, activated carbon characteristics, membrane cleaning strategy...).

A variety of methods have been developed to characterize NOM constituents. Current approaches focus on grouping organic compounds into fractions according to their physicochemical properties such as molecular weight (MW). The isolation of such fractions rely on adsorption/desorption on resins [4,11,14], membrane filtration [9,11], fluorescence excitation emission matrices [2,7] and high-performance size exclusion chromatography (HPSEC) [7,11,15], among other methods. The latter, where NOM fractions are separated according to their MW, has gained much attention as a powerful method for quantitative and qualitative characterization of NOM [15].

Previous studies have applied HPSEC for NOM fractionation at lab- and pilot- scales [3,10,16–18] or at individual treatment units at full-scale DWTPs such as activated carbon [12] and ultrafiltration [19]. However, there are to date relatively few studies that have applied HPSEC along the whole treatment train of full-scale DWTPs. Moreover, most of the DWTPs reported in these latter studies incorporate exclusively conventional treatments (i.e. coagulation/flocculation, sedimentation, sand filtration) [9] and only a small number include advanced treatments such as activated carbon [1], ozonation [20,21], a combination of both [7] and, more rarely, ultrafiltration [5] and reverse osmosis [22]. Previous studies are also found on the application of HPSEC on UF fouling characterization, in particular on its composition and reversibility by backwashing. However, most of them are limited to bench- and pilot- scale experiments [19,23–25] or to treatment scenarios that differ from DWTP (e.g. wastewater treatment plants) [26,27].

The objective of this study was applying HPSEC (1) to characterize NOM in the raw water feeding the DWTP of study in terms of DOC and its fractions, (2) to track DOC contents along the different stages of the DWTP including both conventional and advanced treatments, and (3) to determine the organic fractions retained by the UF membrane (i.e. organic fouling composition) and more readily backwashed from it. The fact that the DWTP of study comprises two parallel treatment lines that cover many distinct treatment units (including coagulation/flocculation, sedimentation, sand filtration, ozonation, activated carbon filtration, ultrafiltration, reverse osmosis) makes this DWTP of particular interest.

## 2. Methods

### 2.1. Plant description

The DWTP of study is located in Sant Joan Despí (Barcelona, Spain) and has a nominal capacity of 5.3 m<sup>3</sup>/s, supplying a population of over 1 million people in the metropolitan area of Barcelona. The raw water used by the DWTP comes from the Llobregat river (and occasionally its aquifer), which presents moderately high total organic carbon (3.4–4.9 mg/L), high turbidity (70–230 FNU) and high conductivity (1160–1939  $\mu$ S/cm). Groundwater exhibited lower TOC concentrations (1.1–1.4 mg/L) and turbidity (0.2–0.5 FNU), but similar conductivities (1970–2012  $\mu$ S/cm).

#### 2.1.1. Conventional treatment CT and advanced treatment AT1

The whole treatment train applied in the DWTP includes a conventional treatment (CT) comprised of pre-chlorination, coagulation/flocculation, subsequent sedimentation and sand filtration, followed by an advanced treatment (AT1) comprised of ozonation, granular activated carbon (GAC) filtration and post-chlorination.

Pre-chlorination is carried out by dispensing in-situ generated ClO<sub>2</sub> into the collected river water for the purposes of disinfection and pre-oxidation of certain metals and NOM. Coagulation is carried out by the addition of Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> to enable the separation of flocs by gravity. The dosage of coagulant (typically in the range of 60–110 mg/L) is readjusted automatically depending on the water quality and the flow to be treated. The sedimentation phase takes place in 88 static, cone-shaped decanters (of 100 m<sup>2</sup> each), at a retention time of approx. 2 h. Flocculant polydiallyldimethylammonium chloride (poly-DADMAC) was sporadically added (at a maximum concentration of 1 mg/L) at the time of the study when the water quality required it. After clarification, water passes through 20 open sand filters (of 100 m<sup>2</sup> each, with a bed thickness 0.6 m) at a filtration velocity of approx. 10 m/h to remove traces of particles and certain microorganisms such as Giardia and Cryptosporidium. These sand filters are backwashed in order to restore their initial permeability.

Once entered in AT1, water is ozonated with in-situ generated O<sub>3</sub> for its biocidal and oxidizing effects. The ozonation results in a break-down of NOM into smaller and lighter fragments. Ozone is dosed at a concentration between 3.0 and 4.8 mg/L, yielding a residual concentration of 0.2 mg/L. The ozonated water is then filtered through GAC (Chemviron Carbon F400) in 20 filters (of 100 m<sup>2</sup> and a bed height of 1.5 m each) to remove organic compounds through adsorption. The water is passed downwards at an average flow rate of 0.2 m<sup>3</sup>/s. GAC filters are backwashed every 2–3 days with air at a flow rate of 0.5 m<sup>3</sup>/s for 5 min and then with sand filtered water at a flow rate of 0.3 m<sup>3</sup>/s for 14 min to avoid long-term clogging phenomena.

#### 2.1.2. Parallel advanced treatment AT2

The need to improve water quality beyond that achieved by CT plus AT1 prompted the installation in 2010 of an additional membrane-based advanced treatment (AT2) run in parallel to AT1. AT2 treats half of the sand filtered water and consists of ultrafiltration (UF), ultraviolet (UV) irradiation, cartridge filtration, UV irradiation, reverse osmosis (RO) and remineralization (RM) units.

The diverted sand-filtered water is acidified with H<sub>2</sub>SO<sub>4</sub> and passed through 0.02  $\mu$ m-pore size submerged PVDF hollow fiber UF membranes (ZeeWeed 1000, GE Water & Process Technologies - ZENON, USA) operating under an outside in mode at a transmembrane pressure of 0.8 bar, for the elimination of bacteria and also possible remaining suspended matter. The UF stage consists of a total of 9 chambers each accommodating 9 cassettes with 57 modules each, totalling 4104 modules (with a total membrane surface area of 228,575 m<sup>2</sup>). The UF modules are periodically backwashed to remove hydraulic reversible fouling from the membrane. The permeation and backwash cycle durations are 45 min and 10 min, respectively.

Prior to RO, ultrafiltered water undergoes a pre-treatment consisting of UV radiation (to eliminate viruses that have survived the previous phases); the addition of H<sub>2</sub>SO<sub>4</sub> (to lower pH and avoid undesirable precipitation on RO membranes), bisulphite (to eliminate traces of oxidants) and a dispersant (to prevent the crystallization of the salts present in the water); cartridge filtering (with selectivity of 5  $\mu$ m) (to provide additional protection to the RO membranes in the event of leaks during the UF phase) and more UV radiation (to avoid bacterial re-growth).

Treated water is then passed through spiral-wound RO membranes (Filmtec LE 440-I, Dow Chemical, USA) that act as a total barrier to viruses and practically all organic and inorganic compounds present in the water. The RO unit contains 10 trains, each configured as a single pass with three stages containing respectively 90, 40 and 28 pressure vessels (with 7 membranes/vessel). The RO unit operates at a pressure of 10–15 bar, the conversion

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