Emerging pollutants removal through advanced drinking water treatment: A review on processes and environmental performances assessment

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Abstract

The presence of emerging pollutants in the aquatic environment in relatively small concentrations and the fact that they cannot be removed by conventional water/wastewater treatment processes bring new challenges in terms of adequate selection of technologies from the technical, economical and environmental points of view. Generally, literature discusses emerging pollutants' removal at significant concentrations (such as those in wastewater), while few studies consider their low concentrations occurring in raw water. This study presents a comprehensive review of the research efforts related to the occurrence, fate, health effects and impacts of emerging pollutants on advanced drinking water treatment and the environmental performance evaluation of different technological options, with a focus on pilot and full-scale installations. All presented case studies consider pollutants removed, process conditions and removal efficiencies, thus making possible comparisons between membrane processes, advanced oxidation processes and adsorption on activated carbon and other materials. The study is completed by an analysis of the environmental assessment instruments (life cycle assessment, carbon, water footprints, other type of assessments) that may be used for selecting sustainable advanced drinking water treatment processes able to remove emerging pollutants. This paper critically reviews the main research topics concerning emerging pollutants: classification, legislative framework, up-to-date removal processes and their environmental performances assessment, to offer a comprehensive analysis of the strategic issues that may constitute future research directions for sustainable water supply.

Keywords: emerging, priority, pollutants, drinking water treatment, environmental, assessments

1. Introduction

Drinking water treatment (DWT) plants face great challenges in optimizing technologies to avoid human health problems and to ensure environmental sustainability, in direct correlation with population growth, water sources lower availability, deterioration due to land use and climate changes, hydrology and water quality changes. These water related problems are better understood and controlled through the improved detection and increased knowledge of the environmental, toxicological and biological effects of an ever increasing list of compounds currently known as: *Emerging Pollutants* (EPs) or *Contaminants of Emerging Concern* (CECs) or *Micropollutants* (MPs) or Priority Pollutants (PPs) or *Persistent Toxic Substances* (PTS) or *Substances of Very High Concern* (SVHC) (Li et al., 2011; Sauvé and Desrosiers, 2014). They are characterised by environmental persistence and threats to human health (Miniero et al., 2014). Pesticides, pharmaceuticals, personal care products, detergents, disinfection by-products, drugs, flame-retardants are a few examples of EPs. Through their presence, eco-toxicological and human health effects, bio-accumulative and degradation characteristics they may influence aquatic biota and also the performance and costs of DWT technologies.

Although a vast scientific literature is available on multiple aspects of concern regarding EPs' monitoring and analysis, we consider that it is important to review in a holistic way these research efforts especially in relation to the technological developments of water supplies and their sustainability assessment. EPs' research topics usually refer to: identification/classification/regulation of new compounds; identification/characterization of toxicological effects, environmental pathways and fate and human health risks (Stiborova et al., 2017); development of analytical methods for detection and advanced drinking water treatment processes (ADWT) for their removal from water (Deblonde and Cossu-Leguille, 2011); development of tools for their environmental impact assessment (Ternes et al., 2015).

To our best knowledge, there is not yet a review paper that structures all the research efforts related to the fate and effects of EPs on DWT and the environmental performance evaluation of different technological options. As a consequence, this study aims to perform a comprehensive literature assessment regarding the presence of EPs in water sources, their occurrence and impacts on the treatment processes and to identify research hot-spots that need further investigation. In detail, this review paper approaches the following research questions (RQs):

- *RQ1) Identification, classification and regulations of EPs.* Numerous concepts, definitions and criteria, were developed to classify and characterize these compounds, leading to a certain level of controversy in the scientific community. Over 1000 substances are classified as EPs (Norman Network, 2016), however only some tens of them are recognized by international regulations and organizations, with a significant lack of consistency;

- *RQ2) Presentation of EPs' characteristics that are relevant for the choice of technology for their removal within DWT.* In detail: raw water characteristics, EPs' toxicological and chemical characteristics, fate and impacts on treatment efficiency, environment and human health;
- *RQ3) Discussion of DWT processes specifically developed for EPs' removal in pilot and full-scale installations.* Scientific literature mostly refers to waterborne EPs and research efforts mainly focus on their fate and effects in natural water sources, or on removal processes from wastewater;
- *RQ4) Investigation of the assessment instruments* that are used to evaluate the environmental performances of DWT processes dedicated to EPs' removal, as a consistent support for more sustainable water supply technologies.

2. Review methodology

The selection of scientific literature was made considering partially the integrative literature review and the following screening criteria:

a) *Relevant international databases and information sources*. Bibliometric sources such as: Web of Science, Science Direct, Scopus were used to retrieve articles, book-chapters and international proceedings articles. International databases of the European Environmental Agency and of the European Commission were used for the selection of Directives or Reports related to EPs' regulation or classification; the relevant scientific content was sourced for this review from scientific articles (119 documents – 88.2%), technical reports (9 documents – 6.6 %) and legal documents (7 documents – 5.2%);

b) *Chronological order*. The majority of the references (*81.4%*) in this review are from 2010 to 2018 (110 references from the total of 135 references), the rest of the references belonging to 2001-2009. Other 45 references (out of which 35 references from 2010-2018) are presented in Annex 1 as information to support the research concerning EPs sources and health effects;

c) *Relevant keywords for the topics of interest* (EPs' definitions, classifications, regulations, occurrence, fate and impacts, methods of analysis, ADWT processes and their pilot or full-scale application, environmental assessment of ADWT processes). The following keywords have been used in different combinations: *EPs/Priority pollutants/Toxic pollutants/Persistent pollutants/Regulation/ Methods of analysis/Drinking water treatment/Advanced drinking water treatment/Membrane processes/Advanced oxidation processes/Adsorption/Pilot/Full-scale applications/Life cycle assessment/Water Footprint/Carbon footprint/Multi-criteria assessment;*

d) *Selection of the references cited in this review based on content analysis.* After eliminating the articles that concern removal of EPs from wastewater (a search on Science Direct after the keywords: *advanced drinking water treatment* and *emerging pollutants* and *pilot scale* provided 900 results, with over 54% strictly related to wastewater treatment processes), the remaining articles/book chapters were analyzed thoroughly. Abstracts of all references left after the first screening process (by using keywords, publication years, elimination of wastewater applications), were analyzed and out of more than 680 identified literature sources, only 135 relevant references were analyzed as full content and finally included in this review, while in Annex 1 were included 45 supplementary references;

e) *Analysis of the overall process of data collection and selection*. The selected scientific literature mirrors the evolution of the studied field in terms of instrumental analysis methods for EPs' detection, development and innovation of ADWT processes and their environmental assessment. However, it was observed that many studies present inconsistencies in terms of terminology used (EPs or PPs, CECs or MPs, PTS or SVHC), many processes are limited only to laboratory-scale and environmental assessments are not always used to support pilot or full-scale applications.

3. Emerging pollutants occurrence, classification and impacts

3.1. Definitions, classifications and regulations

International organizations and regulations dedicated considerable efforts in defining and characterizing EPs (see Figure 1). "*Emerging*" refers to either new pollutants identified in aquatic media and organisms or to new characteristics and impacts of compounds that are already present in the environment. The Norman Network (2016) defined EPs as *substances detected into environment but currently not included in routine environmental monitoring programmes and which may be candidate for future legislation due to its adverse effects and/or persistency*. More than 1000 substances, gathered in 16 classes (algal toxins, antifoaming and complexing agents, antioxidants, detergents, disinfection by-products, plasticizers, flame retardants, fragrances, gasoline additives, nanoparticles, perfluoroalkylated substances, personal care products, pharmaceuticals, pesticides, anticorrosives), are classified as EPs addressing their environmental and health effects and some of their sources (see Annex 1, Table I). The European Environmental Agency considers that EPs (refered to also as "*hazardous substances and chemicals*") should be closely monitored as concentrations and effects, since they are increasingly being found in water bodies across the EU (EEA, 2012).

If EPs are relatively new and not very well regulated, this is not the case of **priority pollutants** (PPs), which are mostly part of EPs but are regulated at the international and national levels due to their high risk towards the aquatic biota and human health, hence their "*priority*" status (Richardson et al., 2007). The Water Framework Directive (WFD) 2000/60/EC (EC, 2000) and Decision 2455/2001/EC (EC, 2001) identified **33 PPs** based on their significant risks ***to*** or ***via*** the aquatic environment. Through Directive 2008/105/EC (EC, 2008) and Technical Report 2009-025 (EC, 2009), Environmental Quality Standards and strict monitoring rules regarding sampling points and analytical methods were issued. PPs' list was completed with other **13** substances and the distinction between **priority** and **priority hazardous substances** was made (for the last ones, Member States should implement necessary measures with the aim of ceasing or phasing out emissions, discharges and losses).

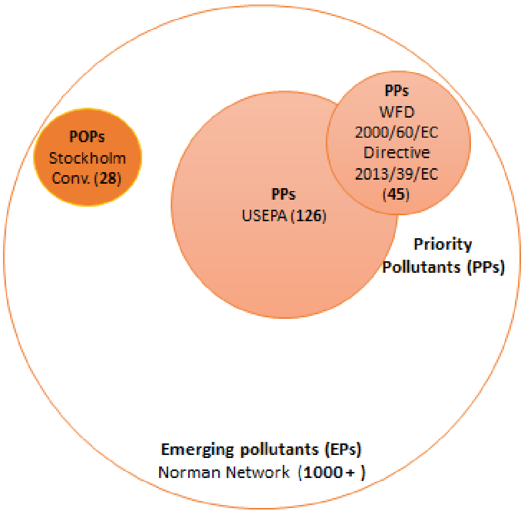

Fig. 1. Emerging, persistent and priority pollutants and specific regulations

Directive 2013/39/EU (EC, 2013) amended WFD 2000/60/EC (EC, 2000) and Directive 2008/105/EC (EC, 2008), new priority pollutants being identified and standards were targeted for implementation (by 2021 for existent PPs and by 2027 for the newly identified ones). Although this directive is based on the preventive actions and the-*polluter-pays principles*, it focuses on PPs monitoring and assessment and it does not specifically approach issues related to DWT. Directive 2013/39/EU defines a list of **45**-priority pollutants grouped as single or classes of substances, which contains pesticides, industrial additives and by-products, pharmaceuticals, personal care products, steroid hormones, drugs of abuse, food additives, flame/fire retards, surfactants and others, from which an initial 10 compounds form a *watch list*. The first PPs included in the watch list were diclofenac, 17-beta-estradiol (E2) and 17-alfaetiniestradiol (EE2); afterwards, measures to avoid the risks involved by their release into aquatic environment should be established (Barbosa et al., 2016; Geissen et al., 2015). However, since PPs are currently not included in the routine monitoring programmes at EU level, but may pose a significant eco-toxicological risk, a recent study (Brack et al., 2017) proposes some specific solutions for the forthcoming WFD review in 2019 based on the developments of EU collaborative projects and Norman Networks contributions. Thus, ten recommendations to improve monitoring and strengthen comprehensive prioritization of pollutants, to foster consistent assessment and support solution-oriented management of surface waters were developed. The United States Environmental Protection Agency regulates **126** priority pollutants, including heavy metals and organic chemicals and their specific analytical test methods (US EPA, 2014).

A smaller group of EPs includes the so called **persistent organic pollutants** (POPs) which are defined as *chemical substances that persist in the environment, bio-accumulate through the food chain and pose a risk of causing adverse effects to human health and the environment* (Stockholm Convention, 2010). Starting with **12** substances, new POPs have regularly been added into the Stockholm Convention annexes. Presently, **28** POPs are listed, grouped in 3 categories as pesticides, industrial chemicals and unintentional chemical by-products (Stockholm Convention, 2017). POPs may enter the aquatic environment from point sources (as wastewater discharges or spills), non-point sources (as agricultural runoff) or indirectly via atmospheric transport and ocean currents.

3.2. Drinking water sources, occurrence, fate and impacts of EPs

Freshwater represents the main raw water source for human consumption, industry, agriculture and energy production. Seawater is considered only in cases of water scarcity due to the high energy and chemicals requirements for its treatment. Nonetheless, EPs are often detected in freshwater and ADWT technologies are required to achieve the necessary quality needed for human consumption (Ternes et al., 2015). This review focus on DWT of raw surface water, usually affected by organic and inorganic micro-pollutants that are of concern for human health and the environment. Inadequate wastewater treatment, excessive use of pesticides or hospital wastewater discharges are important causes of surface water pollution by EPs (Emmanuel et al., 2009). River freshwater is the most exposed to contamination from industrial, agricultural and animal farming discharges (*WHO*, 2011). EPs are found in surface water due to different factors (see Figure 2), then they undergo transport phenomena in natural waters and soil by runoff, erosion or leaching (Fàbrega et al., 2014). EPs' concentration can vary from wastewater discharge point to the water abstraction point because of:

biotransformation, volatilisation, photolysis, sorption, dispersion or different water sources combination, which can attenuate initial concentrations or transform pollutants.

The environmental impacts and health risks are not so well identified and characterized for all EPs (Bui et al., 2016), but there is a growing interest in the scientific community to deepen this field of knowledge. EPs' transformation through DWT processes can lead to compounds which may be more toxic, persistent and less biodegradable than their predecessors (Farré et al., 2008). The most important environmental effects of EPs refer to: bioaccumulation and biomagnification, persistency, toxicity, endocrine disruption potential, carcinogenic effects, mutagenic and teratogenic effects (Guillén et al., 2012). Some EPs can be harmful for both humans and aquatic organisms, with endocrine disturbing effects, estrogenic or hormone disruption, foetal malformation, or even DNA damages (Fawell and Ong, 2012). Human exposure pathways include: ingestion, inhalation and dermal contact through water and food (Pease and Gentry, 2016).

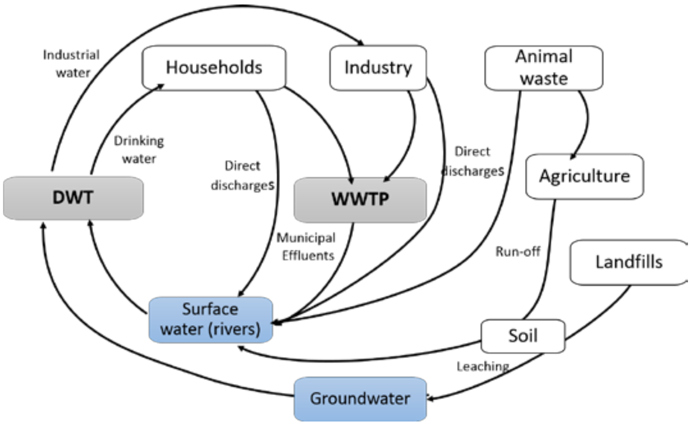

Fig. 2. EPs' pathways and related impacts related to the water uses (DWT: drinking water treatment; WWTP: wastewater treatment plant)

3.3. Methods of analysis

As most EPs are organic compounds and their concentrations are very low, their analysis is challenging and it entails continuous updates (about new compounds to be detected) and improvements (about required sensitivity). Analysis techniques include gas chromatography (GC) coupled with mass spectrometry (MS) and liquid chromatography (LC) coupled with MS (Guillén et al., 2012; Ribeiro et al., 2015; Richardson and Ternes, 2014). EPs analysis procedures are continuously adapted considering the pollutants concentration, specific media and level of precision. Some analysis methods use nuclear magnetic resonance spectroscopy

(Wu et al., 2010), solid-phase extraction coupled to LC-MS, hydrophilic interaction liquid chromatography (HILIC) (Postigo et al., 2008), liquid chromatography–tandem mass spectrometry (LC–MS/MS) analysis method for flame retardants, pesticides, personal care products (Pal et al., 2014; Rodil et al., 2012). EPs or their transformation products may be also qualitatively confirmed by nuclear magnetic resonance (NMR) spectroscopy (Richardson and Ternes, 2018).

4. Advanced drinking water treatment (ADWT) processes for EPs' removal

Conventional DWT processes are dedicated to the removal of solids of various sizes, organic matter and microorganisms/pathogens. They refer to: bar-screening, grit removal, pre-oxidation, coagulation-flocculation, sedimentation, rapid/slow sand and/or granular active carbon filtration, disinfection. The widespread use of chemicals, pharmaceuticals and personal care products (PPCPs), pesticides and solvents made necessary the adoption of advanced technologies, because conventional DWT were not designed to remove EPs (Rodriguez-Narvaez et al., 2017). This situation is exacerbated by the increasing pressures on water resources due to population growth, deterioration of natural water sources, knowledge of new EPs and therefore new guidelines and regulations involving more restrictive concentration limits.

The removal of specific EPs with low concentration in raw water by ADWT is associated with energy and chemicals consumption, which in turn leads to higher treatment costs and additional environmental impacts. From a technological point of view, ADWT may be applied (with the due modifications) for either water of wastewater treatment. To our knowledge (see Section 2), the scientific literature is far more abundant on case studies (mostly at lab-scale and to a lesser extent at pilot/full-scale) on advanced technologies for EPs' removal from wastewater, while studies specifically dedicated to drinking water production are far fewer. Due to their hydrophilic character, most EPs are difficult to remove during wastewater treatment processes, thus they reach surface water and their persistence afterwards affects DWT (Rodil et al., 2012).

Bui et al. (2016) discussed EPs regulated by the Swiss Government and the cases of advanced water or wastewater treatment for their removal (having as final end-use: water supply, wastewater recycling and reuse) that should consider at least the following criteria: (i) range of treated pollutants, treatment efficiency and removal mechanisms, (ii) environmental friendliness, (iii) simplicity of operation and maintenance, (iv) cost-effectiveness and (v) social acceptance.

In the next sections, the most used and promising (in terms of removal efficiencies) ADWT technologies for EPs' specific removal at pilot and/or full-scale will be presented.

4.1. Membrane processes

Membrane processes effectively remove a wide variety of organic, inorganic and solid particles from surface and seawater or wastewater, through semipermeable materials that allow the separation of water (permeate) and of the concentrate (retained at the membrane surface). Membranes are produced from different materials, which provide specific characteristics (pore size, surface charge and hydrophobicity) that determine what type of contaminants can be retained (Gupta and Ali, 2013a). A wide variety of chemically and thermally stable polymers or polymer blends, but also other materials, such as ceramics, metals, glasses or mixed matrix membranes may be used (Arribas et al., 2015).

For drinking water production, several membrane processes may be used, which can be classified considering the force that drives the separation (Arribas et al., 2015):

a) *Pressure driven processes*: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO),

b) *Electric potential gradient*: electrodialysis (ED),

c) *Concentration gradient*: forward osmosis (FO).

DWT requirements, the influent quality, the necessity to remove specific target pollutants and their concentrations dictate the process type and membrane material. The major operational parameters refer to: driving force characteristics (pressure, electric potential), mode of operation (cross flow, dead-end), water flow and initial solute concentrations, pH, membrane material, pore size and retention capacity (Rodriguez-Narvaez et al., 2017). The risk of fouling, clogging or concentration-polarisation depends on the water matrix (concentrations and type of contaminants), membrane material and process control, and therefore different cleaning methods (flushing, backward flushing, air flushing or chemical cleaning) have been developed (Sperlich et al., 2010).

A disadvantage of membrane processes (when comparing them to advanced oxidation processes) is that the pollutants are transferred into the concentrate streams and not destroyed, therefore the concentrate requires further treatment and disposal (Rodriguez-Mozaz et al., 2015). This is compensated by advantages as: ease of operation and adaption to existent treatment facilities, modular design, very small chemical requirements, low energy consumption. The most suitable membrane processes for EPs removal, due to their small pore sizes are: UF, NF and RO (Arribas et al., 2015). Among these processes, RO is the most effective in removing pesticides, PPCPs, toxic metals and cyanides (Malaeb and Ayoub, 2011). Over 99% of total organic and inorganic species were removed from surface waters and groundwater through RO (Gupta and Ali, 2013b; Schoonenberg Kegel et al., 2010). *Ion-*

exchange membranes (cation/anion exchange and bipolar membranes) are used in DWT, especially for sea water desalination by ED or reverse electrodialysis (Alzahrani and Mohammad, 2014; Singh, 2014). The most effective membrane processes that may be used for EPs removal at pilot and full-scale level are presented in Table 1, either as stand-alone or combined treatment.

4.2. Advanced oxidation processes (AOPs)

AOPs are based on the production of hydroxyl radicals (HO•), one of the strongest oxidants (oxidation potential 2.8 V) that may be used for the removal/destruction of EPs for several purposes: raw water pre-treatment, DWT, wastewater treatment (Antonopoulou et al., 2014). The production of hydroxyl radical can be achieved by many pathways, which allow to choose the adequate AOP according to the specific characteristics of the raw water/wastewater and the necessary treatment targets (Catrinescu et al., 2011).

AOPs may be classified by considering the method to generate hydroxyl radicals into chemical, electro-chemical, sono-chemical and photochemical processes, combinations of AOPs with other processes being also frequently used (Molinari et al., 2017; D. Wang et al., 2015). Depending on how reactants get in contact, AOPs may be homogeneous or heterogeneous processes. In the heterogeneous processes, a catalyst such as metal supported catalysts, clays, carbon materials or semiconductors such as TiO_2, ZnO, WO_3, Cu_2O or composite materials are also used (Enesca et al., 2016; Orha et al., 2017). EPs' removal efficiencies in homogeneous processes is dependent on the interactions between the chemical reagents and target compounds; while in heterogeneous processes, the adsorption of reactants and desorption of products that occur at the active sites of the catalyst surface are also very important (Klavarioti et al., 2009).

The most common radical generators/catalyst systems for AOPs include (X. Wang et al., 2015): Fenton's reagent involving iron species (Fe^{2+} or Fe^{3+}) and Hydrogen peroxide (H_2O_2), Ozone (O_3), UV light (UV), H_2O_2/UV, H_2O_2/O_3, H_2O_2/O_3/UV, Photocatalysts/UV or solar radiation, Fenton combined processes (ultrasound/electro/photo Fenton with addition of H_2O_2 and Fe^{2+}). Hybrid processes that combine membrane separation and heterogeneous photocatalysis such as the photocatalytic membrane reactors are also interesting because each process complements the advantages and overcomes the challenges of the other (Molinari et al., 2017).

Although there are many lab-scale applications of AOPs for EPs removal from water/wastewater, there are only few studies performed at pilot-scale (Antonopoulou et al., 2014; Bui et al., 2016). It is worth mentioning that the major challenges referring to the use of

AOPs for EPs' removal from water are related to: a) the small concentrations of these compounds in water, b) the generation of reaction intermediates (when complete mineralisation of target compounds is not achieved), c) relatively high cost of processes when large scale installations are considered. The most effective AOPs that may be used for EPs' removal at pilot and full-scale level are presented in Table 1.

4.3. Adsorption on activated carbon and other materials

Adsorption is an important ADWT process that is efficient and cost-effective if the adsorbent has high porosity and specific surface area, is easy to operate and regenerate (thermally or chemically) and is available in sufficient quantities. The adsorbents' removal capacity depends on densities (material, particle and bulk), porosities (particle and bulk), external surface area, internal surface area, pore-size distribution (among macropores, mesopores and micropores), surface chemistry and operational parameters (temperature, pH, contact time) (Zaitseva et al., 2013).

Various studies demonstrated that granular or powder activated carbon adsorption is one of the best technologies used for EPs' removal from surface water (Zhang et al., 2016; Rodriguez-Narvaez et al., 2017) along with silica gel (Sharma and Bhattacharya, 2016), activated alumina (Kumar et al., 2014), zeolites (Lofrano, 2012) and metal oxide adsorbents (Amin et al., 2014), but the energy used to produce adsorbents is very high (Arena et al., 2016).

In order to increase EPs' removal efficiencies, organic or inorganic materials were developed to obtain nanoadsorbents or engineered nanomaterials (which possess a minimum of one external dimension ranging from 1 to 100 nm) with improved adsorption properties, being able to remove efficiently EPs with various molecular sizes, hydrophobicity and speciation behaviour (Thines et al., 2017).

Engineered nanomaterials are classified as: carbonaceous nanomaterials (C-ENMs, carbon nanotubes, carbon nanofibers, fullerenes, graphenes and carbonaceous composites), metal and metal oxides (nanoscale zero-valent iron, TiO_2, Ag and ZnO) and magnetic-core composite nano/micro particles that have cores made with magnetic elements such as iron, nickel, cobalt or their oxides and alloys with ferromagnetic or superparamagnetic properties, and shells (silica, alumina or polymers or surfactants).

4.4. Single and combined ADWT

Taking into account the wide range of contaminants that belong to EPs category, it is obvious that many processes could be adopted independently or in combination and that their overall removal efficiencies depend on the water matrix (contaminants structure and properties) and on the operational parameters (Bui et al., 2016; Verlicchi et al., 2010). These two criteria, namely the removal efficiency and the operational conditions represent the most used instruments for technical performance evaluation and ADWT comparison in the vast majority of studies.

However, ADWT such as AOPs, adsorption on activated carbon or other materials, RO and NF demonstrated their efficiency independently in EPs removal (Hofmann et al., 2011; Jin and Peldszus, 2012). The same stands for ADWT combined treatments such as: ozonation coupled to granular activated carbon (van der Aa et al., 2012), UV/H_2O_2 and UV/H_2O_2/O_3 oxidation (Lester et al., 2011; Scheideler, 2011), ion exchange combined with ceramic microfiltration (Galjaard, 2011). Specifically considering pharmaceuticals in surface water, it was demonstrated that while DWT guarantee 50% removal, ADWT are able to reach >90% efficiency (van der Hoek et al., 2014; WHO, 2012). As a general remark, literature generally discusses EPs' removal at significant concentrations, while very few studies are available considering the low concentrations occurring in surface water (Barbosa et al., 2016; Ribeiro et al., 2015).

Nanomaterials may be used in stand-alone applications (adsorption) to remove contaminants (organic or inorganic) or may be incorporated in conventional membranes, chemical degradation, photodegradation or even in chemical disinfection steps (Simate et al., 2012). Adeleye et al. (2016) compared ADWT nanomaterials based processes with the conventional ones and found out that for EPs removal from surface waters, RO or ozonation are more expensive than C-ENMs or nano/micro particles.

The most effective ADWT processes that are used for EPs removal at pilot and full-scale level are presented in Table 1.

Table 1. Overview of EPs' removal by ADWT at pilot and full scale

Treatment Processes	**EPs class target**	**Removal efficiencies**	**References**
Single processes			
Ozonisation	Pharmaceuticals	>98%	(Talib and Randhir, 2016)
UV-photolysis	Pharmaceuticals	30-70%	(Pal et al., 2010)
Nanofiltration	Pharmaceuticals	15 – 100%	(García-Vaquero et al., 2014)
GAC adsorption	Mix of 30 pharmaceuticals, pesticides	20- 50% as DOC	(Kennedy et al., 2015)
Combined processes			
Dioxychlorination , coagulation/flocculation, sand filtration, ozonation, carbon filtration and final disinfection with chlorine	Pharmaceuticals and drugs of abuse	>98%	(Boleda et al., 2011)

Treatment Processes	EPs class target	Removal efficiencies	References
Coagulation, ultrafiltration, and GAC filtration	Pharmaceuticals, hormones, antibiotics and flame retardants	99%	(Kim et al., 2007)
UV pre-disinfection, filtration, nanofiltration/reverse osmosis, remineralisation and chlorine disinfection	Pharmaceuticals	>85%	(Radjenović et al., 2008)
Chlorine, coagulation/flocculation, sedimentation, filtration,chloramine, ozonation, GAC filtration, chlorine disinfection	Drugs of abuse	89 – 100%	(Huerta-Fontela et al., 2008)
Chlorine, coagulation/flocculation, filtration, ozonation, granular activated carbon, final chlorination	Drugs of abuse	88 – 100%	(Boleda et al., 2009)
Coagulation/ flocculation, filtration, final chlorination	Drugs of abuse	0 – 18%	(Rodayan et al., 2016)
Filtration, coagulation /flocculation and final chlorination	Pharmaceuticals, flame retardants, plasticizers, biocides, pesticides, herbicides, UV filters	>60%	(Rodil et al., 2012)
Coagulation, flocculation, sedimentation, rapid sand filtration, ozonisation, two-step GAC filtration and Ultraviolet disinfection	Pharmaceuticals	51-95%	(Vieno et al., 2007)
Advanced oxidation	Pharmaceuticals	>99%	(Klavarioti et al., 2009)
Ultrafiltration and nanofiltration	Estrogens	>90%	(Sanches et al., 2012)
Photocatalysis and solar photolysis	Pharmaceuticals	66-82%	(Kanakaraju et al., 2014)
UV / H2O2 integrated into an existing full scale plant	10 pesticides, Pharmaceuticals, Microorganisms	60-98% 67-98% 100%	(Kruithof et al., 2007)
Ultraviolet -photolysis	Pharmaceuticals	80%	(Lekkerkerker-Teunissen et al., 2013)
Dioxychlorination, coagulation/ flocculation, settling, sand filtration, ultrafiltration, ultraviolet disinfection, reverse osmosis and remineralization	perfluorooctane sulfonate (PFOS) / perfluorooctanoate (PFOA)	≥99%	(Flores et al., 2013)

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5. Environmental assessments of ADWT processes

As presented in the previous sections, ADWT technologies are needed for EPs' removal implying the use of additional energy and/or chemicals, which translates into higher investment and operational costs (Bui et al., 2016). These environmental impacts increase in indirect correlation with EPs' concentrations (i.e. large water quantities to be treated usually by combined ADWT so as to remove small pollutant loads). Therefore, the selection of ADWT processes for EPs' removal should take into account complex criteria involving technical, environmental and economic aspects to ensure a sustainable technological option. These criteria may consider: environmental assessment instruments (e.g. life cycle assessment, carbon and water footprint, multi criteria assessments, etc.), technical performance indicators (removal efficiencies, specific energy consumption, reagent quantities, etc.), and economical evaluation tools (life cycle costing, cost-benefit analysis). In the next sections the most used environmental evaluation instruments for the assessment of ADWT technologies are discussed.

5.1. Life cycle assessment

Life Cycle -Assessment (LCA) identifies possible environmental impacts generated in all life cycle stages from "*cradle-to-grave*" (raw material, production, use and disposal, including recycling and reuse) of products (Vince et al., 2008). LCA follows a standardized procedure according to ISO 14040:2006 (Bonton et al., 2012; ISO, 2006) which includes a planning phase (goal and scope definition), a input/output analysis phase (life cycle inventory), an environmental impact assessment phase (LCIA- Life cycle impact assessment) and an interpretation phase.

LCA has received a growing attention in evaluation of the whole water use cycle (Barjoveanu et al., 2014; Loubet et al., 2014) and environmental assessment of water and wastewater treatment technologies (Corominas et al., 2013). When LCA considers the whole water cycle (Barjoveanu et al., 2014; Lemos et al., 2013; Loubet et al., 2014) the analysis focuses on identifying, describing and comparing impacts of all phases (water production, distribution, wastewater collection, wastewater treatment). Referring strictly to drinking water, LCA was extensively used on the operational phase of drinking water production, mainly to compare the environmental impacts of various treatment processes, technologies and development scenarios (Table 2), and only a few studies have considered the construction and decommissioning phases of water production facilities (Igos et al., 2014), or other relevant aspects for water production like distribution systems performance (Sanjuan-Delmás et al., 2015) and alternative water sources (Godskesen et al., 2013; Lundie et al., 2004).

In Table 2, a selection of LCA applications for ADWT is presented considering the studies objectives, system limits and most important environmental impacts.

The most used functional unit is the water production volume (usually 1 m^3) and most LCA studies focus on process or technology performance from an environmental and sometimes economic point of view (Barrios et al., 2008; Jeswani et al., 2015). With regard to the present review it is important to notice that, with very few exceptions (Amini et al., 2015; Bonton et al., 2012; Gifford et al., 2017), LCA studies do not consider aspects like raw water quality, water quality standards, treatability in the definition of the functional unit. Amini et al. (2015) discussed the importance of functional unit definition for LCA evaluation of treatment technologies in a study on ion exchange technology performance, and showed that the lower the initial concentration of the target substance, the higher the associated environmental impacts will be. Bonton et al (2012) mentioned 4 usual indicators in the definition of the functional unit (1 m^3 treated water), but did not mention EPs nor the methodology to include them in the functional unit definition. A limited number of studies compare human health risks and risk assessment with LCA; most of these focus on the assessment or comparison of processes and technologies like membrane processes (Manda et al., 2014), ion exchange (Amini et al., 2015; Choe et al., 2013), sorption (Gifford, 2016). Gifford et al. (2017) explored through LCA the trade-offs between DWT impacts and the reduction in human health but mainly considers inorganic contaminants.

Critical impacts of ADWT in LCA studies refer to: electricity consumption, and subsequent carbon emissions; chemicals production which contribute to eutrophication and eco-toxicity, but the way these are described varies greatly due to differences in analysed systems and different assessment methods. More examples for comparing DWT and ADWT alternatives by means of LCA studies are presented in Table 2.

Table 2. Overview of LCA applied for different ADWT/DWT processes evaluation (water supply: surface water)

Goals and objectives	**System boundaries (technological stages)**	**LCIA impacts and contributors**	**References**
Technology performance comparison			
Eco-design of DWT	DIS-Oz,C/F,SED,F,DIS-Oz,GAC,DIS-NaClO	Electricity consumption due to ozone generation system, GAC regeneration using fossil resources and reagents	(Ahmadi et al., 2016)
Reduction of operating cost and environmental impacts of DWT through six algorithms	DIS-OZ,C/F,SED,F,DIS-OZ,GAC,DIS	Ozonation, GAC regeneration and coagulant dose	(Capitanescu et al., 2016)
Two NF systems compared with conventional DWT	cellulose acetate membrane filtration system vs. polyethersulfine membrane filtration system (pilot-scale) vs. GAC	Electricity and chemicals for membrane production; GAC production and energy for DWT	(Manda et al., 2014)
Comparison of 2 DWT plants	a) DIS-OZ, pre-MIN, C/F, OX, MIN, SED, SF, UF, DIS-NaClO b) Pre-MIN, C/F, SED, OX, MIN, SED, post-MIN, SF, UF, DIS-NaClO	Site A: higher electricity consumption; site B: impacts due to reagent consumption and DWT process complexity.	(Igos et al., 2014)
Comparison of 2 DWT plants to assess impacts due to choice of energy sources and chemicals	1. F, NF, DIS-Cl2 2. C/F,SED, SF, GAC, DIS-Cl2	Conventional DWT's impact is higher due to electricity consumption, reagents and GAC	(Bonton et al., 2012)
Comparison of "Arvia" process vs GAC. Focus on adsorbents recovery	1. Avira process: SF / adsorption onto Nyex, SED 2. SF, GAC	Electricity consumption for GAC and Nyex regeneration.	Jeswani *et al.*, 2015)
Scenario analysis			
Comparison of 5 scenarios of drinking water sources and identification of environmental impacts caused by drinking water consumption	S1: C/F,SED,SF,GAC,DIS (Oz, Cl2) S2: (S1+RO and remineralization); S3: (S1 + domestic RO); S4: mineral water in plastic bottles; S5: mineral water in glass bottles.	S1 is the most convenient alternative compared with PET and glass bottles. Comparing S1 with S2 and S3, electricity consumption and reagents lead to higher environmental impacts	(Garfí et al., 2016)

Goals and objectives	System boundaries (technological stages)	LCIA impacts and contributors	References
Comparison of different scenarios in Paris metropolitan area	Two DWT plants with conventional treatment: C/F,SED,SF,GAC,DIS-Oz, Dis-UV, CL Membrane process: F,SED, Pre-F,UF,NF,DIS-UV, DIS-Cl)	Complex evaluation of scenarios and impacts. Water resources depletion and electricity consumption	(Loubet et al., 2016)
Environmental assessment of coagulant and PAC dose; chemicals alternative; different alternative technologies for ozone production and electricity consumption.	a) C/F,SED,GAC,SF, DIS-Oz b) C/F,SED/SF,DIS-Oz, GAC c) C/F,SED,SF,DIS-Oz, GAC	Chemicals and electricity consumption.	(Mery et al., 2014)
Environmental impacts produced by 2 DWT	Conventional: SF,DIS-Oz, pH adjustment, GAC, pH Adjustment, SF Alternative 1: DIS-Oz,GAC,pH adjustment,SF,RO Alternative 2: SF,RO, pH adjustment.	Electricity, chemicals, GAC.	(Mohapatra et al., 2002)

Notes: DIS-Disinfection (Oz–ozonation, NaClO–sodium hypochlorite, HClO–hypochlorous acid, Cl_2–chlorine gas) C/F–coagulation/flocculation, F–Filtration (SF–sand filtration), GAC–Adsorption on granular activated carbon, MIN-mineralization, OX–oxidation, DWD–Drinking water distribution, WWC–wastewater collection, WWT–wastewater treatment

5.2. Carbon Footprint and Water Footprint

The footprint family indicators was developed in the last two decades to quantitatively measure the impacts of human activities over natural resources and the environment, as well as to assess sustainability (Čuček et al., 2012; Fang et al., 2014). The most used footprints are: ecological footprint, water footprint, and carbon footprint although other types of footprints are also used for different studies such as: social, economic, energy, chemical nitrogen footprints (Fang et al., 2014).

Carbon footprint (CF) is defined as the amount of CO_2-equivalent emissions caused directly and indirectly by an activity (Wiedmann and Minx, 2008), or as the total amount of greenhouse gas (GHG) emissions over the life cycle of a process or product (BSI, 2008). CF takes into account non-CO_2 emissions (e.g. CH_4, N_2O and fluorinated gases) whose global warming potentials are much higher than that of CO_2, and the CF gives the responsibility for global warming to consumers (Strutt et al., 2008; Wiedmann and Minx, 2008). CF assessment is usually based on the GHG (that are process relevant) accounting, considering certain boundaries and it is expressed as a sum of mass units or mass flows of CO_2 equivalents (Pandey et al., 2011).

CF analysis may be used to evaluate alternatives for water treatment and technological facilities distribution and storage, in order to reduce emissions and the associated impacts and costs. Software instruments such as **CCaLC**-Carbon Footprinting Tool (2013) or **ECAM**-*Energy performance and Carbon emissions Assessment and Monitoring Tool* (2015) are available, however there are few studies (Qi and Chang, 2013; Wu et al., 2015) that assess the CF of DWT plants due to the difficulty of quantifying the emissions and estimating the associated costs.

Qi and Chang (2013) compared through CF actual DWT production capacity (surface and groundwater sources) with the expansion of 5 different water sources (groundwater, surface water, transferred water use permit, regional water and other options). The impact generated on climate change by GHGs and the costs involved in each stage of DWT plants construction, production, transportation, treatment, use (drinking water distribution and wastewater collection), wastewater treatment and discharge into rivers was evaluated. CF and cost analysis for water supply systems boost continuous improvements for the related technologies, cost-effectiveness of each stage of drinking water production and reduction of generated impacts (Qi and Chang, 2013).

In a recent study, Wu et al. (2015) have used life cycle CF accounting (LCA-CF) and life cycle costing (LCC) to optimise the placement of the water treatment works of Ningbo city (China) considering the performance of other water systems elements (raw water transport, water treatment facility and the downstream distribution network). The analysis showed that a clear accounting of LCA-CF and the inclusion of CF costs as a decision-making component affect the priority ranking of various DWT siting options, facilitating the decision for an alternative with the lowest LCC (with CF cost) when achieving the same water supply benefits.

A comprehensive mathematical model was developed in order to estimate and compare on-site and off-site CO_2 emissions, from conventional and ADWT plants in South Korea (Kyung et al., 2013). Coagulation-flocculation, sedimentation and filtration were chosen as conventional treatment scheme, while advanced treatments involved coagulation-flocculation followed by MF and ozone disinfection. A sensitivity analysis to examine the effect of operation factors and conditions on CO_2 emission and to suggest better options for sustainable operation was also carried out. This study results, apart from providing basic knowledge on CO_2 emissions from different types of DWTs, suggest possibilities to efficiently reduce CO_2 emission while enhancing the quality of treated water, and offers guidelines to properly operate each unit in the water treatment process (Kyung et al., 2013).

Water footprint (WF) concept was introduced in 2003 by A. Hoekstra and is defined as the cumulative virtual water content of all products and services consumed by individuals or communities within a given region (Hoekstra et al., 2011). WF assessment is used to evaluate the extent to which a product, process or activity may affect the environment, social or economic sustainability. As in LCA, WF can be expressed in a specific functional unit (e.g. m^3/t of production, per hectare of cropland, etc.). WF has three components: a) *Blue water footprint* refers to consumption of blue water resources (surface and groundwater); b) *Green* *water footprint* refers to consumption of green water resources (rainwater as it does not become run-off); c) *Grey water footprint* is defined as the volume of freshwater required to assimilate the pollutants loads considering the natural background concentrations and existing ambient water quality standards.

Although there are many studies related to WF of products, production processes, industries (Hoekstra, 2015; Lovarelli et al., 2016; Skouteris et al., 2018), there are only few studies related to water supply and ADWT. At global level, the WF of municipal water supply has been estimated as 3.6% of the total WF of humanity (Hoekstra, 2015). Considering that the ratio of net to gross abstraction has been estimated to be 5-15% in urban areas and 10-50% in rural areas, the blue WF of drinking water from the tap can vary in the interval 0.065-0.65 L/L, assuming that the rest of the water returns to the water system from which it was abstracted (Hoekstra, 2015). To our current knowledge, other studies that consider ADWT assessment through WF were not found in the international scientific literature.

5.3. Other type of assessments

Cost performance analysis

Below are discussed several evaluation efforts that have been targeted towards finding a trade-off between the benefits of implementing various EPs' removal technologies and the technical and environmental problems and impacts that come along with these technologies. This is also the framework into which the few ADWT cost-performance analysis studies have been realized. Adeleye et al. (2016) evaluated the applications of various engineered nanomaterials and their potential use in ADWT. This assessment has considered technical performances (targeted compounds, removal efficiencies), environmental aspects (direct impacts, secondary pollution and toxicity), as well as cost estimates (where available) with comparisons to conventional technologies for the same target EPs. The results showed a great variability of performances and costs, but the authors advocate for the benefits of these emerging technologies over the conventional ones.

The cost performance analysis performed by Igos et al (2014) show that both capital costs for retrofitting/upgrading of existing DWT to advanced processes, as well as operational costs increase with decreasing concentrations of target contaminants.

Solutions to minimize the costs and environmental impacts in the water sector should approach operational aspects and associated impacts by switching to electricity consumption from renewable sources, increasing membrane life-time and cleaning periods (Ahmadi et al., 2016; Chollom et al., 2017; Liikanen et al., 2006). Control parameters for optimizing environmental impacts and operational costs of DWT technologies improvements have been proposed: temperature, pH, turbidity, UV absorbance, dissolved organic carbon, microorganisms, inorganic compounds, micropollutants and by-products (Capitanescu et al., 2016).

Multi criteria assessment

Multi-criteria assessment (MCA) tool has been used by researchers as a support for decision making processes, evaluation, generation or comparison of alternatives, in order to better understand the aim of contextual analysis, some interests or relations between processes and stakeholders (Ahmadi and Tiruta-Barna, 2015; Paneque Salgado et al., 2009). The main goal of multi-criteria evaluation techniques is to discover connections and relations between the evaluated actions and actors involved in the decisional process, in order to have a clearer vision on how the entire assessment process takes place, not only to identify or compare different alternatives, depending on certain set criteria. MCA is supposed to identify all social, economic and governance issues, from multiple points of view, solving all interest disputes, many information from different disciplines and the involvement of social, governmental and communities representatives being needed (Martin-Ortega and Berbel, 2010; Yan et al., 2016).

Multi-criteria assessment of ADWT technologies for EPs removal was realized in a small number of studies. Mery et al. (2013), have considered in their assessment: type of treated pollutants (organic micropollutants, PPCPs and steroid hormones), operational parameters and removal efficiencies (reagents dose, pH, contact time and water characteristics) and treatment technologies (GAC filters, AOPs, membrane processes and membrane bioreactors processes. A similar complex MCA was perfomed by Sudhakaran et al (2013) to compare EPs removal of several DWT and ADWT processes: riverbank filtration, various oxidations (O_3, AOP and UV-AOP using H_2O_2), GAC adsorption, membrane processes (RO and NF). The analysis criteria were: treatability, costs, technical considerations, sustainability and residence time period.~~,~~ Other MCA focus on environmental impacts (energy consumption, reagents used, by-products, waste production and disposal) (Ahmadi et al., 2016), technological features (construction, operation, maintenance, flexibility and reliability of treatment methods) or economic aspects (DWT plant scale, treatment targets, technologies applied and electricity consumption) (Capitanescu et al., 2016) and social aspects (Bui et al., 2016; Müller et al., 2016)

6. Conclusions

In the last two decades consistent efforts have been made towards the identification, characterisation, classification, international regulation, evaluation of toxicity and fate of EPs in the aquatic environment and their impacts on human health, as well as to the identification and assessment of various treatment technologies for removing such pollutants (especially from wastewater). Emerging pollutants are classes of chemical compounds with different origins and aquatic routes that have increased risks for the human health and aquatic biota.

This paper critically reviews the main research topics related to the importance of emerging pollutants removal for drinking water production by addressing both the issues of pilot/ full scale advanced treatment options and the instruments for environmental performance assessment.

There are important issues that need to be further studied or to be addressed by scientists and water companies about the safe and sustainable supply of drinking water from surface sources:

- Consistent definitions, criteria for classification and regulation of maximum allowable concentrations of EPs need to be addressed at the level of international and national regulation bodies towards water supplies and DWT, especially considering the risks for human health;
- Removal or degradation of EPs is performed by ADWT, which usually completes (as pre-treatment or final treatment) the conventional stages for drinking water production from surface sources. Thus, membrane processes, AOPs and adsorption on activated carbon or other materials found many applications at pilot and full scale, and their selection is mainly based on technical and economic issues;
- Comparisons of ADWT options in relation to the "degradation" (AOPs) or "phase change" (membrane processes, adsorption) processes is rarely made for DWT, due to the relatively small concentrations of target pollutants in the influent, although there are many studies at laboratory scale that support these applications. However, these comparisons should be used for scaling up studies that refer to EPs removal;
- Various assessment instruments may complete the sustainability profile for the selection of ADWT options for EPs' removal, since all these alternatives involve additional energy and material consumption. There are only few studies that consider this type of approach to support full-scale installations. Although many of them pointed out that electricity consumption is one of the most significant impact generators, there are few investigations that consider the impact of using renewable energy sources. Finally, these assessment tools need to be refined to better capture the associated environmental impacts of removing EPs.

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