Consolidated vs new advanced treatment methods for the removal of contaminants of emerging concern from urban wastewater.

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Abstract

Urban wastewater treatment plants (WWTPs) are among the main anthropogenic sources for the release of contaminants of emerging concern (CECs) into the environment, which can result in toxic and adverse effects on aquatic organisms and consequently on humans. Unfortunately, WWTPs are not designed to remove CECs and secondary (e.g., conventional activated sludge process, CAS) and tertiary (such as filtration and disinfection) treatments are not effective in the removal of most CECs entering WWTP. Accordingly, several advanced treatment methods have been investigated for the removal of CECs from wastewater, including consolidated (namely, activated carbon (AC) adsorption, ozonation and membranes) and new (such as advanced oxidation processes (AOPs)) processes/technologies. This review paper gathers the efforts of a group of international experts, members of the NEREUS COST Action ES1403 who for three years have been constructively discussing the state of the art and the best available technologies for the advanced treatment of urban wastewater. In particular, this work critically reviews the papers available in scientific literature on consolidated (ozonation, AC and membranes) and new advanced treatment methods (mainly AOPs) to analyse: (i) their efficiency in the removal of CECs from wastewater, (ii) advantages and drawbacks, (iii) possible obstacles to the application of AOPs, (iv) technological limitations and mid to long-term perspectives for the application of heterogeneous processes, and (v) a technical and economic comparison among the different processes/technologies.

Keywords: activated carbon, advanced oxidation processes, oxidation by-products, ozonation, photocatalysis, urban wastewater

Contents

1. Introduction

Pharmaceutically active compounds (PhACs), personal care products, pesticides, synthetic and natural hormones, and industrial chemicals (such as flame retardants, plasticizers, food additives, among others) are continuously discharged into the environment through different anthropogenic sources, which can result in toxic and adverse effects on ecosystems and consequently on humans (Daughton and Ternes, 1999; Malajet al, 2014). This group of chemicals, typically detected in aquatic ecosystems and wastewater at low concentrations (ng/L - µg/L), is also referred to as contaminants of emerging concern (CECs). Conventional secondary (e.g., activated sludge process) and tertiary (such as filtration and disinfection) treatments in urban wastewater treatment plants (WWTPs) are not effective in the removal of most CECs entering WWTPs (Li and Zhang, 2011; Rizzo et al., 2015; Krzeminski et al., 2019) and as consequence, effluents from WWTPs are among the main anthropogenic sources for the release of CECs into the environment (Petrie et al., 2014). An additional concern for human health and the environment is related to the release of CECs to soils as well as their uptake by crops during wastewater reuse practices (Paz et al., 2016). The release of CECs from WWTPs into the environment has not yet been regulated (except in Switzerland) nor their occurrence in wastewater for agricultural reuse. Although regulations on wastewater reuse exist in some countries (Paranychianakiset al., 2015), a regulation shared by all European Countries is still under discussion, CECs monitoring in WWTP effluents for agricultural reuse being one of the main debated issues among scientists, policy makers and stakeholders (Rizzo et al., 2018a).

A wide range of advanced treatment methods has been investigated for the removal of CECs from wastewater, including consolidated (namely, activated carbon (AC) adsorption, ozonation and membrane filtration) and not intensively implemented (for brevity subsequently referred to as “new”) treatments, such as advanced oxidation processes (AOPs). Ozonation, AC adsorption and NF/RO membranes can effectively remove CECs. Ozonation and AC processes are increasingly implemented at full scale, especially in Switzerland (due to the implementation of a newWater Protection Act in 2016, which regulates the removal of CECs from urban wastewater (Eggen et al., 2014; FOEN, 2015) and Germany, where the implementation is carried out on voluntary basis (particularly in the two federal states of Baden-Württemberg and North Rhine-Westphalia).

Ozonation may result in the formation of oxidation/disinfection by-products (e.g., N-nitrosodimethylamine (NDMA) and bromate) and a polishing post-treatment step with a biological active sand filter is recommended (Hollender et al., 2009; von Gunten, 2018). Unlike ozonation, AC treatment is not effective in the inactivation of bacteria. Consequently, when stringent limits for reuse are requested, an additional disinfection step is needed. Membrane technology filtration with dense membranes, such as nanofiltration or reverse osmosis, has a high reported energy demand and results in potential challenges in relation to concentrate disposal, but also provides additional water quality benefits such as salt removal. Other options for advanced treatment of urban wastewater have been proposed in the last years and AOPs are among the most investigated ones. However, although they have been found effective in the degradation of CECs, in particular homogeneous photo driven AOPs (e.g., UV/H_2O_2 and photo-Fenton) (Klamerth et al., 2010; Ferro et al., 2015) and heterogeneous photocatalytic processes (e.g., UV/TiO_2) (Byrne, in press), they do not yet find application at full scale.

This review paper gathers part of the work done by a group of international experts, members of the NEREUS COST Action ES1403 “New and emerging challenges and opportunities in wastewater reuse” (Fatta-Kassinos et al., 2015), who for three years have been constructively discussing the state of the art and the best available technologies for the advanced treatment of urban wastewater. The publications available in scientific literature on consolidated (ozonation, AC and membranes) and new advanced treatment methods (mainly AOPs) are critically reviewed to analyse (i) their efficiency in the removal of CECs from wastewater, (ii) advantages and drawbacks, (iii) possible obstacles to the application of homogeneous AOPs, (iv) technological limitations and mid to long terms perspectives for the application of heterogeneous processes, and (v) a technical and economic comparison among the different processes and technologies. Finally, the main gaps are discussed in order to enable identifying the most suitable solutions for advanced treatment of urban wastewater.

1.1 Contaminants of emerging concern in urban wastewater: classification, environmental sources and legislation

A significant amount of contaminants of emerging concern (CECs) is discharged into the sewer system and further transported to WWTPs. The occurrence of CECs in WWTP secondary effluents has been investigated, highlighting the most prevalent substances and those with higher concentrations. A summary by Luo et al., (2014) covering WWTP effluents of the US, Europe (including the Western Balkan Region), and Asia (Korea,China), revealed that the concentration of major CECs ranged from 0.001 to 10 µg/L; whereby e.g. PhACs were detected in concentrations often higher than 1 µg/L (Luo et al., 2014).

The limits for CECs in wastewater discharge are still not regulated (Barbosa et al., 2016). Directive 2008/105/EC has established a list of 33 Priority Substances (PS) for surface water and their associated Environmental Quality Standards (EQS), but no PhACs were included. The Global Water Research Coalition (GWRC) developed an International Priority List of PhACs relevant for the water cycle, based on the compounds that present a potential risk in water supply (Global Water Research Coalition, 2008). According to GWRC, 44 compounds are classified in three main groups: Class I (10), Class II (18) and Class III (16), based on the following criteria: human toxicity, ecotoxicity, degradability, resistance to treatment and occurence in the environment. Switzerland (the Swiss Centre for Applied Ecotoxicology Eawag-EPFL) has also proposed EQS for CECs (including PhACs, steroidal estrogens, pesticides, industrial chemicals and complexing agents) that show a risk for aquatic organisms when the annual average concentrations in surface water exceed the chronic EQS (Robert et al., 2011). The newer European Union Directive 2013/39/EU recommended monitoring and treatment solutions for a group of 45 PS, meeting the requirements of environmental protection. The first Watch List of substances for European Union-wide monitoring was reported in the Decision 2015/495/EU of 20 March 2015 and updated in Decision 2018/840/EU of 5 June 2018. This list refers to different CECs: antibiotics (azithromycin, clarithromycin and erythromycin), synthetic (17-alpha-ethinylestradiol (EE2)) and natural hormones (17-beta-estradiol (E2) and estrone (E1)), the pharmaceutical diclofenac, pesticides (methiocarb, oxadiazon, imidacloprid, thiacloprid, thiamethoxam, clothianidin, acetamiprid and triallate), a UV filter (2-ethylhexyl-4-methoxycinnamate) and an antioxidant (2,6-di-tert-butyl-4-methylphenol) commonly used as food additive.

The wide and frequent occurrence of CECs in the environment and the inefficiency of conventional WWTPs for their removal put the attention on these substances. The limits for CECs discharge should be regulated by the European Commission and supported by national country authorities. Furthermore, there is persisting need for scientific research in this field and recommendations for advanced wastewater treatment steps or even new treatment scenarios (Bui et al., 2016; Ahmed et al., 2017). The classification, source and legislation of relevant CECs occurring in secondary treated effluents, that were included in the present review paper, are presented in Table SI1 (in supplementary information (SI)). Based on recommendations of the NEREUS COST Action, 25 CECs were selected according to criteria relevant for wastewater reuse such as (i) relevance to crop uptake, (ii) concern for human and environmental health, (iii) recalcitrance and (iv) frequency of detection. The criteria are described in detail by Krzeminski et al., (2019), where the fate of CECs in biological treatment is reviewed.

2. Effect of consolidated advanced treatment processes on CECs removal from urban wastewater

Starting with the first publications on the occurrence of wastewater-relevant CECs in the aquatic environment in the last century as reviewed by Halling-Sørensen (Halling-Sørensen, 1998), numerous studies on the fate of CECs during both biological and advanced treatment were conducted in lab-, pilot- and full scale. Ozonation and AC treatment proved to be promising and economically feasible for WWTP upgrade. Currently, both technologies are increasingly implemented at a full scale, especially in Switzerland which is the only country regulating CECs removal from urban wastewater up to date (Eggen et al., 2014; FOEN, 2015). They are also widely implemented at WWTPs that generate reclaimed water for different scenarios such as supply to homes with dual reticulation (Reungoat et al., 2012). Among the membrane technologies the most commonly implemented ones at full scale are the so-called pressure driven processes, which englobe nanofiltration (NF) and reverse osmosis (RO). In addition to dissolved organics, total dissolved solids (TDS) and inorganic ions are removed, with a higher rejection percentage in the case of RO. NF and RO processes also physically remove many pathogenic microorganisms providing a disinfection barrier, making them especially attractive for water reclamation processes.. Furthermore, prior application of membrane filtration synergistically increases the efficiency and effectiveness of posterior disinfection processes based on chemicals (e.g., ozone, chlorine) as well as germicidal light in the ultraviolet range. Specifically, treatment trains including high pressure membranes have been implemented in numerous potable reuse schemes in the southwest of the United States of America, Australia, Israel, Windhoek in Namibia, the Netherlands and Singapore. Therefore, in the present review ozonation, AC treatment and pressure driven processes were classified as consolidated processes for advanced treatment in WWTPs.

2.1 Ozonation

The first drinking water treatment plant to use ozone for disinfection was built in 1893 at Oudshoorn, Holland. Learning from this one, another one was built in 1906 at Nice, France (Shammas et al., 2005). Later ozonation was adopted also for the abatement of CECs (Rice, 2002). The knowledge from drinking water treatment can be used for the application in wastewater treatment, however, the differences in the matrix have to be taken into account.

The application of ozone generally involves two reaction mechanisms, the direct reaction by ozone and the indirect reaction of OH radicals (HO•) that are formed during ozone reactions. Ozone reacts selectively with compounds containing electron-rich moieties such as olefins, deprotonated amines or activated aromatics, exhibiting reaction rate constants k_{O3} over several orders of magnitude in the range of 1 to 10^7 M^{-1} s^{-1} (von Sonntag and von Gunten, 2012). The major source for HO• generation is the effluent organic matter (EfOM). HO• are generated from a side reaction of ozone with specific groups of EfOM such as phenols or amines. Due to the high HO• generation potential of EfOM, effluent ozonation can be considered an AOP (Buffle et al., 2006).

In contrast to ozone, HO• are characterised by low selectivity and a fast reaction with a wide range of organic and inorganic compounds, which makes the indirect reaction mechanism beneficial to the abatement of CECs refractory to ozone. The rate constants for most ozone refractory CECs vary only over two orders of magnitude ($k_{\text{HO}\bullet} = 10^8$-$10^9$ M^{-1} s^{-1}) (Table SI2). CECs degradation during ozonation depends on the reaction rate constants of the respective CECs with ozone and HO• and the oxidant exposure (Lee et al., 2013), according to equation 1.

$$ln\frac{[\text{CEC}]}{[\text{CEC}]_0} = -k_{O3}\int[O_3]\,dt - k_{\bullet OH}\int[\ {}^{\bullet}OH]\,dt \qquad (1)$$

Reaction rates can be determined with defined lab scale experiments (von Sonntag and von Gunten, 2012) and are available for many substances. Depending on the compound and its pKa, they can vary significantly with speciation and hence with pH. Thus, protonation can result in a reduction of the reaction rate constant with ozone by several orders of magnitude. Therefore, not species-specific but apparent second-order rate constants at the pH of interest need to be considered. Since the pH range of WWTP effluents vary mostly in the range between pH 7 and 8, the apparent second-order rate constant may vary for substances with pKa values close to this range, e.g. benzotriazole, methylbenzotriazole or metoprolol (Lee et al., 2014). Ozone and HO• exposure are site-specific and have to be determined for each wastewater (Schindler Wildhaber et al., 2015; Zucker et al., 2016).

2.1.1 Parameters impacting CECs degradation during ozonation

EfOM contains numerous ozone reactive moieties, resulting in a considerable oxidant consumption. For this reason EfOM is considered one of the most important parameters for ozonation when compared to other influencing factors such as pH, temperature, or bicarbonate as radical scavenger and inorganic compounds. Thus, the wastewater matrix is dominant for ozone and HO• consumption, rather than the presence of CECs (Nöthe et al., 2009). Usually, the so-called specific ozone dose (D_{spec} in g O_3/g DOC) is applied, where ozone is dosed as a function of the dissolved organic carbon (DOC) content in the wastewater. DOC typically varies between 4 and 15 mg DOC/L for municipal wastewater during dry weather conditions (Stapf et al., 2016), but deviations of these values can be found in literature. Applying the same flow proportional ozone dose (e.g., 5 mg O_3/L) would result in a variation of the specific ozone doses and subsequently CECs degradation efficiencies. Therefore, only literature indicating the applied or consumed specific ozone dose was taken into account for the present review. The data were derived from full-scale, pilot-scale and (sometimes spiked) lab-scale tests conducted with real wastewater.

Nitrite (another decisive parameter) reacts fast with ozone in a molar ratio of 1:1, consuming 3.43 g O_3 per g NO_2-N. The fast reaction results in a competition with CECs abatement. Thus, in addition to the DOC-normalisation, a nitrite compensation needs to be considered when applying the specific ozone dose as principle for feed-forward process control (Stapf et al., 2016).

UV absorption at 254 nm (UV_{254}) is a simple sum parameter that in contrast to DOC, significantly decreases during ozonation. The relative UV_{254} (ΔUV_{254}) was found to correlate with the specific ozone dose (nitrite compensated) and the CECs degradation (Bahr et al., 2007; Nöthe et al., 2009)). Consequently, ΔUV_{254} is considered as a surrogate parameter to evaluate the treatment efficacy of ozone for CEC abatement, but even a parameter suitable for feed-back process control (Chon et al., 2015; Park et al., 2017;Stapf et al., 2016; Wittmer et al., 2015). The advantage of this feed-back process control concept is the automatic consideration of nitrite since the ozone consumed by nitrite is not available for CEC oxidation and does not result in a UV decrease. Chon et al., (2015) suggest the combination of ΔUV_{254} and the change in electron donating capacity, which better represents the reactivity of EfOM and thus CEC abatement. The authors postulate that ΔUV_{254} is more suitable for assessing the abatement of highly ozone reactive CECs while the abatement of less reactive CECs is better reflected by the change of the electron donating capacity. However, the latter cannot be applied as a real-time control parameter.

2.1.2 Abatement of CECs by ozonation

For a more applied comparison of CECs, a grouping in three categories based on the abatement at commonly applied specific ozone doses in the range of 0.4-0.6 g O_3/g DOC is suggested (Table 1). Several publications (amongst others, Bourgin et al., 2018; Gerrity et al., 2012; Lee et al., 2013, 2014) grouped the investigated CECs according to the reactivity with ozone and in some cases also with HO•. In the present review the categorization based on the ozone reactivity according to Bourgin et al., (2018) was used. The abatement refers only to the reduction during the advanced treatment by ozonation, since the degradation over the entire treatment plant can be higher if a compound is also biodegradable.

Table 1

In Figure 1 the abatement (%) of all reviewed CECs allocated to group A, B and C (group A: azithromycin, bisphenol-A, carbamazepine, ciprofloxacin, clarithromycin, diclofenac, erythromycin, metoprolol, sulfamethoxazole, and the hormones 17-alpha-ethinylestradiol and 17-beta-estradiol; group B: benzotriazole, bezafibrate, mecoprop and methylenbenzotriazole and group C: acesulfame, iopromide and primidone) is depicted for specific ozone doses from 0.1-1.6 g O_3/g DOC. If the authors reported a abatement below the LOQ, it was not considered in the figures. However, all reported data are shown in Table SI3 of the Supporting Information (SI) and the second-order rate constants of the reviewed CECs with ozone and hydroxyl radicals are given in Table SI2. Depending on the publication, either single measurements or mean calculated abatement with standard deviation were reported, and only results with a maximum of 10% standard deviation were taken into account for the present review. The percentage degradation of one representative of each group is shown in Figure 2.

Figure 1

Figure 2

CECs of group A comprise substances that predominantly react with ozone; they are characterised by electron-rich moieties and a fast reaction with ozone ($k_{O3} > 10^3$ M^{-1} s^{-1}).

Even a specific ozone dose D_{spec} as low as ≥ 0.25 g O_3/g DOC is high enough to abate more than 80% of CECs with high ozone reactivity in most of the ten effluents investigated by Lee et al., (2013). For D_{spec} above 0.5 g O_3/g DOC and compounds with $k_{O3} \geq 10^5$ M^{-1} s^{-1}, no detrimental impact of the wastewater quality on the abatement efficiencies was observed any more. A similar dependency is depicted in Figure 1, where the highest differences regarding CECs degradation as a function of the specific ozone dose were observed below D_{spec} of 0.4 g O_3/g DOC. Metoprolol shows the lowest abatement among the compounds in group A, in agreement with its lowest reactivity with ozone ($k_{O3,\ pH7}$ of 2 x 10^3 M^{-1} s^{-1}). Nevertheless, also for metoprolol an average abatement above 80% was achieved for D_{spec} of 0.4-0.6 g O_3/g DOC according to Table 1, while values higher than 94% were reported for diclofenac (Figure 2). Lower abatement occurred only for effluents with elevated nitrite concentrations and EfOM with a higher content of electron-rich moieties, respectively, as reported also by other authors (El-taliawy et al., 2017; Margot et al., 2013). The lower metoprolol abatement reported e.g. by Kreuzinger et al., (2015) was influenced by the effluent quality, but was not induced by nitrite. This emphasizes that the effluent quality, which influences the ozone exposure, plays a decisive role for the efficiency of ozonation, beside ozone reactivity, as given in Equation 1.

Endocrine disrupting compounds are currently in the focus of the European Union, and the hormones EE2, E2 and E1 are included in the Watch List (EU 2015/495 and 2018/840). The industrial chemical bisphenol-A was reviewed as a representative of estrogenic compounds. All these CECs react fast with ozone due to the phenolic moiety (Deborde et al., 2005). Hence, ozonation efficiently abates these compounds and their estrogenic effect (Deborde et al., 2005, 2008; Huber et al., 2004). Overall, ozonation was found to reduce estrogenic activity by >90% at D_{spec} >0.4 g O_3/g DOC (Altmann et al., 2012; Escher et al., 2009; Reungoat et al., 2012).

Regarding CEC belonging to group B ($k_{O3} = 10^2\text{-}10^3$ M^{-1} s^{-1}), they were removed to a lower extent than those from group A, as shown in Figure 1 and Figure 2. For this group of compounds, the ozone reactivity is still decisive for CECs abatement, and not the reactivity related to HO• (Lee et al., 2013). The abatement correlates with the specific ozone dose (Figure 2) and the apparent second-order rate constants. The variability of the observed abatement is reflected by the measured variability of the ozone exposure over a factor of approximately four at D_{spec} of 1.0 and 1.5 g O_3/g DOC (Lee et al., 2013). Higher variations may occur for compounds with p*K*a values close to the pH of the wastewater (e.g., for benzotriazole with a p*K*a of 8.6), which is affecting the apparent second-order rate constant. Lower degradation of benzotriazole reported by Kreuzinger et al., (2015) may be caused by this, beside the influence of EfOM quality. Overall, most of the reported abatement data between 0.4 and 0.6 g O_3/g DOC-were between 50 and 80% (Figure 1).

CEC of group C ($k_{O3} < 10^2$ M^{-1} s^{-1}) can be considered ozone resistant and their reactivity is influenced by their reaction with HO•. The HO• exposure correlates with the specific ozone dose (Lee et al., 2013) since they are formed during ozone reaction with matrix components. Hence the abatement correlated with D_{spec} and differences amongst representatives of group C can be attributed to $k_{HO•}$. In general, the compounds in this group exhibit an abatement smaller than 50% at D_{spec} of 0.4-0.6 g O_3/g DOC (see Figure 1 and Figure 2). Some compounds exhibited higher abatement in specific WWTPs (e.g., for primidone in Stapf et al., (2017)), what may be due to high $k_{HO•}$ values or due to EfOM effects. A good representative of group C is iopromide (Figure 2).

Data was difficult to find for metformin degradation at various specific ozone doses (Figure 1); but considering the rate constant with HO• ($k_{HO•} \sim 10^7$ M^{-1} s^{-1}, Figure 2), which is approximately two orders of magnitude below iopromide, low abatement can be expected for this compound. Perfluorooctanesulfonic acid (PFOS) and perfluorooctanonic acid (PFOA) exhibit an even lower reactivity with HO• (von Sonntag and von Gunten 2012) and no degradation was observed at an Australian water reclamation plant employing ozonation (Thompson et al., 2011). Therefore, its abatement by ozonation can be assumed to be negligible.

2.1.3 By-product formation

The formation of transformation products (deriving from CECs) and oxidation by-products (deriving from the wastewater matrix) is an important issue for ozonation since the applied doses do not result in mineralisation. Therefore, Schindler Wildhaber et al., (2015) developed a test system for evaluating the treatability of wastewater with ozone. Two important oxidation by-products that should be analysed according to the authors are bromate and NDMA. The WHO guideline value for NDMA in drinking water is 100 ng/L. NDMA can already be present in the WWTP influent (Bourgin et al., 2018). Additionally, it can be formed during ozonation from precursors like hydrazines and sulfonamides (yield >50%) (von Gunten et al., 2010; Kosaka et al., 2009; Krasner et al., 2013; Schmidt et al., 2008). Bromate is formed from bromide in the influent of the WWTP. The main bromide sources are the wastewaters discharged from waste incinerators, waste and chemical industries and to a lower extent from precipitation and geogenic sources (Soltermann et al., 2017). Seawater ingress into the sewer system in coastal areas may be another source of bromide. Since bromate formation is a slow process, it can be controlled by adjusting the ozone dose. At specific ozone doses of ≤0.4 g O_3/g DOC, little bromate is formed in ozonation, however, bromate yields are almost linearly correlated to the specific ozone dose for higher ozone doses (Chon et al., 2015; Soltermann et al., 2016). For typical specific ozone doses in wastewater treatment (0.4−0.6 g O_3/g DOC) molar bromate yields are ≤3%. Usually the bromide concentrations in municipal wastewater are in the range of ≤100 µg/L, which yields bromate concentrations in the WWTP effluent below the WHO drinking water standard of 10 µg/L. For higher bromide concentrations than 100 µg Br^-/L, bromate generation needs to be evaluated and ozonation may not be appropriate.

To evaluate wastewater quality after ozonation, the biological activity should be assessed with several bioassays targeting various modes of action that proved to be suitable. Thus, Schindler Wildhaber et al., (2015) suggest the application of five different bioassays: the Ames test, the Yeast Estrogen Screen (YES), and the combined algae assay (performed with solid phase extracted (SPE) samples to be sensitive enough), as well as the *Ceriodaphnia dubia* reproduction assay and the fish embryo toxicity test with *Danio rerio* (performed with native wastewater effluent samples to include the effect of very polar compounds not well extracted by SPE). Some ecotoxicological investigations have shown a temporary increase of toxicity after ozonation in certain tests due to the formation of labile, toxic organic reaction products (Stalter et al., 2010a, 2010b). The toxicity can be reduced again by a subsequent treatment step with biological activity, such as a sand filter or a biologically activated carbon filter (BAC) (Hübner et al., 2015; Knopp et al., 2016; Lee et al., 2016). Based on these investigations, a biological treatment after ozonation is recommended to reduce biodegradable organic reaction products with potential toxicity.

2.2 Activated carbon (AC)

2.2.1 Adsorption process

AC is generally known for its adsorption ability towards a broad spectrum of CECs (Rossner et al., 2009, Snyder et al., 2007), due to its high porosity, large surface area and high degree of surface interactions. According to the International Union of Pure and Applied Chemistry (IUPAC), the AC is classified according to the pore diameter: macroporous (≥ 50 nm), mesoporous (2-50 nm), secondary microporous (0.8-2 nm) and primary microporous (≤ 0.8 nm). Mesoporous AC was found to be the most suitable for CECs adsorption due to the reduced impact of organic matter competition on the surface sites (Budimirović et al., 2017). AC is commonly applied as a powdered slurry feed (i.e. powdered AC, PAC) into a contact reactor, or in a granular form (i.e. granular AC, GAC) in a packed bed filter (Snyder et al., 2007). The adsorption characteristics are varying depending on the nature of the material and the activation process during manufacturing of the AC (Zietzschmann et al., 2014a).

The basic principle of the adsorption process is to transfer CECs from the liquid phase to the solid one. During the adsorption competition occurs: (1) direct in between of small organic molecules for the same activated surface sites, e.g. the high energy adsorption sites in the micropores of the AC and (2) pore blocking by the large organic molecules that hinder the entrance of CEC into the appropriate adsorption sites. The competitive adsorption depends on a variety of factors related to the adsorbent surface characteristics (Ruhl et al., 2014), such as: AC surface area, particle size, pore size distribution and surface chemistry (e.g., surface charge - acidity or basicity). Moreover, the efficacy of the adsorption process is affected by the nature of the adsorbed compounds (e.g. hydrophobicity, chemical structure, and charge). The specific physicochemical properties of the target pollutants in the water matrix determine its adsorption potential or the resistance against the adsorption competition on the AC. The pH and temperature of wastewater are external factors that affect the removal of CECs in wastewater effluents (Luo et al., 2014).

The abatement capacity of CECs also depends on the wastewater quality and the operational conditions of the used WWTP technology (Mailler et al., 2015). CECs adsorption onto AC is limited by the content of the background EfOM which is present in urban wastewater. The adsorption processes for CECs removal onto AC are more efficient in water with low concentrations of competing organic content (low DOC) compared to the waters with high DOC loading (Boehler et al., 2012; Pramanik et al., 2015). AC efficacy is greatly reduced by the presence of EfOM which competes with the activated carbon structure for binding sites and can block pores (Snyder et al., 2007). However, the amount of DOC is not always sufficient to explain adsorption competitions in wastewater, and the quality of the organic matter should be considered too. The dissolved effluent organic matter presents a heterogeneous mixture of refractory organic compounds, with diverse structure and varying origin (Michael-Kardatou et al., 2015). In addition, the WWTP configuration has a substantial impact on the effluent quality also due to seasonal variations. The properties of EfOM such as size, hydrophobicity, and acidity/basicity are of paramount importance. Small size EfOM compounds are more competitive in adsorption process and low molecular-weight components of the DOC have a detrimental influence on adsorption capacity (Zietzschmann et al., 2014b). Low or medium molecular weight EfOM and molecules with low specific UV absorbance at 254 nm were found to be removed by 65-70% by PAC (Filloux et al., 2012).

The adsorption processes are controlled by the contact time (PAC) and the empty bed contact time EBCT (GAC). For PAC application a relatively short hydraulic contact time from 18-30 min (Karelid et al., 2017; Ruhl et al., 2014) to 0.7-3 h (Margot et al., 2013) is sufficient. However, PAC is kept in the reactor for a longer time by returning it to the contact tank (Siegrist et al., 2018). In this way PAC residence times of minimal 12 hours (Boehler et al. 2012) up to several days (Margot et al., 2013; Löwenberg et al., 2014) can be achieved.

For GAC the adsorption process was found to depend heavily on the empty bed contact time (EBCT) in the packed reactor (see below). In GAC adsorption, a loading with organic matter happens in the beginning up to around 5’000-15’000 bed volumes (BV), when an equilibrium is reached for DOC removal in the range of 15-20% (Altmann et al., 2016b; Bourgin et al., 2018; Reungoat et al., 2012; Zietzschmann et al., 2016). However, the adsorption of CECs is continuing with decreasing efficiency over time and BVs.

2.2.2 Activated carbon application

PAC can be dosed directly into the existing biological treatment process (Streicher et al., 2016) or dosed on top of the tertiary filter with low backflush intervals of 1-3 days (Altmann et al., 2014). Most commonly, however, a stand-alone contact reactor with a fluidized PAC bed, followed by a clarifier to retain PAC, is employed as a post-treatment after biological treatment (Mailler et al., 2015). Good removal of CECs can be achieve in all these technologies. For direct dosing into the biology, slightly higher PAC doses are needed for the same performance, however, capital cost is lower (Siegrist et al., 2018). The return of the filter backwash or of the used PAC from the clarifier back into the secondary biological treatment of the WWTP improved the overall CECs removal further by 10 to 50% compared with effluent PAC application alone (Boehler et al., 2012). However, if the sewage sludge is used for agricultural purposes, direct dosing or recycling of PAC into the biological treatment is not practicable. Different dosing locations (Streicher et al., 2016) and dosing approaches – continuous (Altmann et al., 2015a; Hu et al., 2016) or single-pulse PAC dosing (Karelid et al., 2017; Mailler et al., 2015) - have been investigated. The continuous dosing resulted in decreasing CECs effluent concentrations with increasing reactor runtime due to adsorption onto accumulating PAC in the reactor bed (Altmann et al., 2015a). For the separation of PAC from the wastewater, a sand filtration (Altmann et al., 2014; Karelid et al., 2017), or a ultrafiltration (UF) unit (Margot et al., 2013) can be used. Both pressurized (with in/out driven membranes) and submerged (with out/in driven membranes) PAC/UF systems have been tested (Löwenberg et al., 2014). The addition of a coagulant (4–15 mg $FeCl_3$/L or 0.1-0.4 gFe/gPAC) improves the subsequent separation of the PAC by UF or sand filtration. The influence of different PAC dosing procedures in in/out driven PAC/UF process has recently been studied by Ivančev-Tumbas et al., (2017). Slightly different removal efficiencies were observed in pressurized vs submerged configurations of PAC/UF processes due to different tank concentration and retention time of PAC even at the same PAC dose (Löwenberg et al., 2014). Specifically, a lower removal of sulfamethoxazole at peak loads in PAC/UF systems was achieved, presumably due to desorption processes from the membrane material. A review of such sorptive interactions for estrogen compounds on membrane surfaces has been published by Schäfer et al., (2011) as well as relevant findings related to impact of solute-solute interactions on UF filtration (Neale and Schäfer, 2012), Sheng et al., (2016) also reported interactions of CECs with an UF membrane and differences between removal by PAC applied alone and within an in-line PAC/UF hybrid process. However, the implications of the findings related to such interactions on PAC efficiency in the hybrid PAC/UF process and long-term performance remain unclear.

GAC treatment has the benefit that it can be filled into existing deep bed reactors (sand filters). GAC is usually applied either as a monomedia adsorbent (Altmann et al., 2016b; Grover et al., 2011; Zietzschmann et al., 2016), or as a replacement for the upper layer of a tertiary dual media filter (Altmann et al., 2016b). Earlier studies had shown that the AC usage is higher for GAC application compared to the PAC form, for the same DOC value and similar removal of CECs (Karelid et al., 2017). In a setup using internal recirculation the PAC system achieved a 95% removal applying a fresh dose of 15-20 mg/L, while variations of GAC dosage were much broader and ranged up to 230 mg/L, depending on the carbon product (Karelid et al., 2017). Boehler et al., (2012) demonstrated that more carbon (about 3-5 times more) is needed with GAC than with PAC for the same elimination of CECs when using an adsorption reactor with EBCT of around 10-15 minutes. However, an EBCT of 25 minutes turned out to be sufficient for a good performance for a 1.18 - 2.36 mm granulation at low DOC (5-6 mg/L) with similar carbon usage as in PAC treatment (Wunderlin et al., 2017). At EBCT higher than 30 minutes, no influence of EBCT on the performance was found anymore (Reungoat et al., 2011).

UV_{254} absorbance measurements present a reliable parameter for monitoring and controlling the removal of CECs in AC treatment. CECs removal was found to correlate with the decrease of UV light absorbance. UV_{254} measurements may also predict CECs removals, but are not accurate for biodegradable compounds (Altmann et al., 2016a; Anumol et al., 2015; Ziska et al., 2016).

2.2.3 Influence of CECs physico-chemical properties of the compounds on their removals

The adsorption process is affected by interactions between the carbon surface and the adsorbate. The AC surface is predominantly hydrophobic but may also contain heteroatoms (oxygen, hydrogen, chlorine, nitrogen and sulfur), which determine the acid/base character of the surface and specific interactions with adsorbed compounds. When the AC is in contact with an aqueous solution, an electric charge is a result either of dissociation of the surface functional groups or the adsorption of ions from the solution (Dias et al., 2007). The adsorption of the EfOM, which is generally negatively charged in wastewater, alters the initial AC surface (neutral or positive charged) or increase (in case of initially negative surface sites) the total charge of the AC surface (Mailler et al., 2015). The physico-chemical characteristics of the CECs such as polarity, molecular weight and the presence of different functional groups and charges (and their interaction with the AC surface sites altered by EfOM) are crucial parameters that determine their removal (Jekel et al., 2015, Kovalova et al., 2013; Sotelo et al., 2014; Yang et al., 2011). The removal efficiency of ionisable compounds is strongly pH dependent (Verlicchi et al., 2012). High molecular weight organic compounds are more sensitive to direct competition for the adsorption surface sites (Delgado et al., 2012). The parameters for selected CECs relevant for wastewater effluents are presented in Table SI4.

Hydophobicity is often characterized by the log of the octanol-water partition coefficient, K_{ow}. Higher logK_{ow} values lead to better adsorption of nonpolar (hydrophobic) compounds controlled by nonspecific dispersive interactions with AC (Altmann et al., 2014). For charged compounds, the acid-base speciation needs to be taken into account at a certain pH for the octanol−water distribution, given by D_{ow}. However, a simple estimation from log D_{ow} values for charged polar compounds can lead to an under-estimation of elimination efficiencies for many compounds (Kovalova et al., 2013). Additional electrostatic interaction with the functional groups of the AC need to be accounted for. Such ionic interaction cannot be predicted simply from physical-chemical characteristics. As matter of fact, Altmann et al., (2015a, 2016b) found that highly polar negatively charged CECs such as sulfamethoxazole, primidone and iopromide, are only partially adsorbed similar to the highly polar sweetener acesulfame (Mailler et al., 2015), despite a predominantly positively charged AC surface. On the other side, the zwitterionic compounds ciprofloxacin and atenolol acid (Kovalova et al., 2013; Yang et al., 2011), and the positively charged metoprolol with low D_{ow} (Margot et al., 2013a) show very high removal rates.

2.2.4 Removal of CEC by powdered AC and granular AC

The removal efficiencies of selected CECs by advanced wastewater treatments with PAC and GAC, summarized from different studies, are presented in Table SI5 and SI6, respectively. The literature data on CEC removal are selected from studies in bench, pilot and, preferably, full-scale applications. Despite the frequent detection of CECs in urban wastewater (influents and effluents), their removal by advanced treatment at full scale is quite limited (Boehler et al., 2012; Grover et al., 2011; Yang et al., 2011). Because of the high influence of the matrix and DOC content, the data summarized in Table SI5 and Table SI6 are restricted to real urban wastewater effluents and the use of AC as advanced treatment in WWTPs.

CEC removal is highly related to the applied AC dosage. Average removal of 80% was reported for a PAC dose of 7-20 mg/L, depending on the DOC of the wastewater ranging normally from 5-10 mg/L (Boehler et al., 2012; Karelid et al., 2017; Löwenberg et al., 2014;

Margot et al., 2013). As a rule of thumb, about 1.5 g PAC/g DOC needs to be applied in municipal wastewater with PAC recycling to the activated sludge treatment, or 2-3 g PAC/g DOC for direct application to activated sludge treatment (Siegrist et al., 2018). Strongly adsorbing compounds like carbamazepine or clarithromycin can be eliminated by more than 90% also at low PAC doses of 5-10 mg/L (Boehler et al., 2012; Mailler et al., 2015). To remove weakly adsorbing CECs (e.g., primidone, sulfamethoxazole, gabapentin) by 80%, higher amounts than 2 g PAC/g DOC are necessary (Altmann et al., 2015b).

GAC is packed into a deep bed reactor, and breakthrough curves are observed. Well-adsorbing CECs (e.g. carbamazepine) were removed more than 80 % up to 8,000-10,000 BVs, whereas weakly to moderately adsorbing compounds (e.g., primidone and sulfamethoxazole) showed removal less than 80% at <5,000 BVs for EBCTs of 14 minutes (Altmann et al., 2016b; Bourgin et al., 2018). It is worth noting that in addition to adsorption, biological degradation can occur. This was observed for diclofenac and benzotriazole in a GAC reactor with high bed volumes in the range of 30,000- 50,000 BV (Bourgin et al., 2018). This is in line with a previous publication by Reungoat et al., (2012) which reported that BAC (a fixed bed GAC supporting the growth of bacteria attached to its surface) has a good potential for the removal of the investigated CECs hydrochlorothiazide, tramadol, venlafaxine, and metoprolol (>90% at >68,000 BV) as well as many other compounds investigated in that study. However, CEC abatement may be attributed to sorption and/or biodegradation processes, which were indistinguishable in these studies .

2.3 Powdered AC vs. ozonation

As treatment with AC and ozone are established technologies in full-scale treatment to abate CECs, a more detailed comparison is reasonable. Compared to AOPs, the adsorption onto AC offers the advantage of a lower energy consumption at ther WWTP and no by-product formation (Knopp et al., 2016; Mousel et al., 2017). However, the production of AC comprises a high primary energy demand. Moreover, the exhausted adsorbents with adsorbed CECs may be considered as hazardous waste and demand adequate disposal. GAC can be reactivated and reused, therefore having a smaller CO_2 footprint compared to PAC. The sustainability of GAC filters is greatly affected by the frequency of adsorbent replacement and/or regeneration. Despite the advantage of potential reuse of exhausted GAC (Hu et al., 2015), its regeneration is associated with high energy demand for desorption of high-molecular-weight compounds (Bui et al., 2016). In addition, the hot stream with desorbed pollutants derived from GAC regeneration should be managed as hazardous waste. An extended PAC life time is obtained by its recirculation to the aerobic activated sludge tank, which increases CECs removal, but also sludge volume (Margot et al., 2013). Unlike GAC, PAC cannot be regenerated and must be separated from the wastewater and finally incinerated (Bui et al., 2016). PAC that was recycled into the biological treatment can be incinerated together with the excess sludge, as practiced in certain countries like Switzerland (Boehler et al., 2012).

For a comparison, removal rates from wastewater effluents of selected CECs by PAC (reported in Table SI5) and ozonation process (with specific ozonation dose, expressed in g O_3/g DOC) are summarized in Table 2. Margot et al (2013), Kovalova et al., (2013), and Jekel et al., (2015) concluded that PAC gave a higher average removal for some CECs (e.g., fluconazole, valsartan, benzotriazole), while ozone performed better for others (e.g., gabapentine, sulfamethoxazole and diclofenac). Negatively charged iodinated contrast media were not removed with high efficiency regardless of the process applied, but neutral contrast media like iopromide are slightly better removed with AC (Knopp et al., 2016). Acesulfame, PFOA and PFOS are not well removed by either PAC treatment or ozonation (Altmann et al., 2015a, 2016a; Mailler et al., 2015; Margot et al., 2013; Thompson et al., 2011). The removals of steroid hormones (e.g., EE2 and E2) are high with ozone (Sun et al., 2017) as well as with PAC (Margot et al., 2013).

Overall, it cannot be concluded if ozonation or treatment with AC is more beneficial. More recently, the combination of ozonation at low specific ozone doses with PAC or GAC was tested for their performance and economic evaluation (Bourgin et al., 2018; Knopp et al., 2016; Yang et al., 2011).

Table 2

2.4 Membrane filtration processes

2.4.1 Some engineering aspects of membrane filtration

The two primary objectives of the application of low pressure membranes (microfiltration (MF) and ultrafiltration (UF)) in the advanced treatment of urban wastewater tend to be the removal of total suspended solids (TSS) and microorganisms through the provision of a physical barrier. Common nominal pore sizes vary from 0.1-1 μm for MF to 0.01-0.04 μm in UF (Crittenden et al., 2012). A common characteristic of the employed polymer chemistries are their hydrophilicity and their chemical resistance over a wide pH range and oxidizing conditions. These material properties together with the engineering approach make these membranes quite robust, allowing the use of hydraulic backwashing, air scouring, and soaking in a variety of chemicals including hypochlorite solutions to maintain their functionality and control fouling reversibly. The water recovery of low pressure membrane processes in wastewater tends to be between 96 and 98%.

NF and RO membranes on the contrary usually operate on already pre-filtered water with a very low TSS concentration. In water reclamation, these processes are usually employed to reduce the content in total dissolved solids (TDS), whereby the treatment objectives can encompass the removal of major inorganic solutes to reduce electrical conductivity, specifically hardness, or trace metal contamination, as well as organic contaminants including bulk EfOM and CECs. While in water reclamation most of the named water quality benefits are at least desired for applications such as potable reuse or other high human exposure scenarios, in reality NF and specifically RO tend to be implemented, when at least partial desalination is required.

From an engineering point-of-view the by far dominant industry standard is cross-flow filtration, whereby leafs of membranes are packaged in a geometry denominated as spiral-wound membrane modules, whereby the modules are installed sequentially in pressure vessels. The resulting designs are highly modular as pressure vessels are installed in parallel flow conveniently addressing flow requirements flexibly. As water crosses the membrane from feed to permeate, the resulting retentate stream diminishes in volume and hence also in cross-flow velocity as the feed flow progresses through the pressure vessel. In practice this means that after filtering approximately 50% of the feed flow in a pressure vessel the remaining retentate will be combined with the retentate of another pressure vessel to feed another pressure vessel in a subsequent stage to maintain cross-flow velocity in the acceptable range. A variety of staged designs with two or three stages are common in water reclamation to achieve overall water recoveries of 70 to 85%.

From a membrane material point-of-view, the typical commercial NF or RO membrane is a so-called thin film composite with a loose polyester layer providing structural support, a second layer similar to an UF membrane in structure consisting of polyethersulfone and an ultrathin top-layer (10-100nm) of cross-linked polyamide, which is the part of the membrane that retains TDS. This polyamide layer is less chemically resistant compared to the materials employed in MF and UF membranes. Hence, it is vulnerable to strong chemical oxidants such as hypochlorous acid or ozone. This limits the application of cleaning agents on the membrane as well as the in-situ control of biofouling via germicidal chemical agents. Due to the dense structure of the polyamide layer, it is not feasible to conduct hydraulic backwashing as the polyamide layer may peel off the support layer, if the pressure gradient is reversed in the course of backwashing.

2.4.2 Mechanisms of removal of CECs by high pressure membranes

The molecular weight of CECs lies typically in the range of 100-400 Dalton with some exceptions such as macrolidic antibiotics that can be substantially larger. These values translate to molecular radii that are typically less than 1nm, i.e. they are not retained by clean MF and UF membranes, with the exception of minor removal due to adsorption on membrane surfaces or fouling layers. Hence, this section focuses entirely on NF and RO membranes.

For the rejection of organic compounds by NF and RO membranes three rejection mechanisms have been identified in literature: size exclusion, Donnan exclusion and adsorption (Van der Bruggen et al., 1999), which are conceptually visualized in Figure 3 (adapted from Verliefde, 2008). Those three rejection mechanisms are not only governed by the solute properties and membrane properties but also by the operational conditions, module and system design, and the feed water quality. Membrane fouling has also been found to influence the solute rejection due to altering the membrane surface and its inherent properties (Zularisam et al., 2006).

Size exclusion occurs due to the solutes being larger than the effective pore size of the polyamide layer of the membrane. Size exclusion is assumed to be the dominant rejection mechanism for ‘large’ molecules with a molecular weight >200 g/mol. NF is generally capable of obtaining considerable removal of organics with a molecular weight larger than 200 g/mol, whereby this may vary strongly depending on the NF membrane employed. On the other hand, RO achieves good removal for solutes with a mass of 100 to 150 g/mol (Bellona et al., 2004).

The surface of NF and RO membranes has been designed in such a way that it bears negative charges at the surface leading to a negative zeta potential and the formation of Helmholtz electric double layers that lead to the formation of a so-called Donnan potential. The Donnan potential influences the incoming ions increasing the overall ion rejection of the membrane (Ong et al., 2002). This is important for salt removal of the membrane and also impacts upon charged organic solutes. Consequently, specifically high rejections have been reported for negatively charged organic contaminants, whereby the opposite can be the case for positively charged compounds (Bellona et al., 2004; Yangali-Quintanilla, 2010).

Dissolved organic compounds can also adsorb to the membrane. This may lead to an enrichment on the membrane and an increased chemical potential that promotes the transport through the membrane towards the permeate stream and hence a lower than the expected rejection. This phenomenon has been observed by a number of authors for fairly hydrophobic CECs like estradiol and related compounds (Kimura et al., 2003; Nghiem et al., 2004a).

Figure 3

The impact of fouling layers on the membrane on rejection arises from a combination of a couple of different factors. First, adsorption effects can promote an enrichment of the organic solute in the vicinity of the membrane, if the solute in question has a chemical affinity to the fouling layer, either through hydrophobic or charge interactions. Secondly, fouling layers will enhance so-called concentration polarization as solvent (i.e. water) is convectively transported to the membrane and permeates the membrane, whereas the rejected solutes accumulate in the vicinity of the membrane surface as diffusive and convective transport back into the bulk of the solution is reduced by the fouling layer compared to a clean membrane surface. This phenomenon is often called cake-enhanced concentration polarization (Hoek and Elimelech, 2003; Kimura et al., 2009; Zularisam et al., 2006). In addition to organic fouling layers this effect has also been observed for biofouling (Botton et al., 2012).

Due to their importance, mathematical modelling of mass transport in membrane filtration processes has been attempted from early onwards. One of the first proposed models was the Spiegler-Kedem model (Spiegler and Kedem, 1966), published in the inaugural issue of the Desalination journal decades before the processes became truly commercial. Remarkably, this simple model is still often used nowadays as a straightforward solution to practical problems. Another simple and frequently applied model is the solution-diffusion model (Wijmans and Baker, 1995; Williams et al., 1999). Later, authors have attempted to modify those models by introducing additional terms (Verliefde et al., 2009). Alternative approaches have modelled rejection rather based on molecular properties for a particular system establishing quantitative structure activity relationships that describe the rejection behavior. Such an approach has for instance been reported for CEC (Yangali-Quintanilla et al., 2010) and disinfection byproducts (Doederer et al., 2014).

2.4.3 Removal of CECs by membrane processes

Within the scope of this review, it does not appear pertinent to provide vast details on the removal of many individual CECs as a large diversity of membranes have been tested and several reviews and PhD theses exist already providing good overviews and collections of experimental data (e.g., Bellona et al., 2004; Ge et al., 2017; Le-Minh et al., 2012; Nghiem, 2005; Plakas and Karabelas, 2012; Siegrist and Joss, 2012; Verliefde, 2008; Yangali-Quintanilla, 2010).

In this regard, the following information is focused on a set of contaminants that are relatively well known and have been mentioned in the past frequently as critical compounds either due to environmental, health or social perception issues in the context of water reuse. Specifically, here we reviewed diclofenac, a negatively charged pharmaceutical at pH values typically prevalent in water reuse processes; carbamazepine, another pharmaceutical but without charge; E2 a natural steroid hormone that is fairly hydrophobic (logK_{OW} 4.01); and finally NDMA, a potential carcinogen, that can be generated as an undesired by-product of oxidation and disinfection processes, specifically chloramination and ozonation. This set of CECs appears suitable to provide the reader with an overview on potential variation in treatment performance, while at the same time illustrating the above mentioned mechanisms involved in contaminant rejection and impacting factors.

As summarized in Table SI7**,** carbamazepine and diclofenac are generally well removed with the rejections for the reported NF membranes ranging from around 60 to 90% for most reported studies. However, a carbamazepine rejection of only 32-40% was reported for a NF membrane (Vergili, 2013). On the other hand, the only study carried out at large scale (Radjenovic et al., 2008) reported very high rejection percentages of carbamazepine (> 97%). The same study reports rejection percentages of above 99% in the case of RO membranes. Thus, first, as pointed out above, retention percentages are generally high for both NF and RO membranes when dealing with typical pharmaceutical compounds, apparently increased rejection being obtained when RO membranes are employed. The molecular weights of carbamazepine and diclofenac are respectively 236.3 and 296.1 g/mol, as a rough indication of molecular size, without going into further detail of geometric molecular descriptors such as different hydrodynamic radii or projection areas. Second, diclofenac is consistently better rejected than carbamazepine. This may be related to a slightly higher molecular size but also, as mentioned previously, to the Donnan exclusion generated due to its negative charge at ambient pH. The latter is likely the most important explanation for this behavior. For comparison, the rejection behavior of ibuprofen (molecular weight 206.0 g/mol, negatively charged) closely resembles the rejection observed for diclofenac (Vergili, 2013). Thirdly, one has to be cautious when extrapolating laboratory results typically obtained on small-scale flat sheet apparatus to full-scale rejections, as evidenced by the diverging results obtained, when going to the industrial-scale engineered process (Radjenovic et al., 2008). Such observed differences may be due to an inadequate reproduction of the hydraulic conditions on lab-scale, influencing concentration polarization and transport phenomena more generally speaking. Also, the small water recovery in lab-scale processes needs to be considered. Finally, small membrane samples used on lab-scale may not always be representative of the average performance of large membrane surfaces industrially manufactured.

The second example chosen is the rejection of E2, an uncharged steroid hormone with relatively high logP (E2 log P = 4.01 vs. carbamazepine log P = 2.77) and a molecular weight of 272.4 g/mol (carbamazepine, 236.3 g/mol). Since E2 is more hydrophobic than carbamazepine, despite being a larger molecule, its rejection in NF is lower (63-67% versus 77-79%), when analyzed under equal conditions (Ge et al., 2017). This is due to the higher affinity of the compound to the membrane material and hence a resulting enrichment. It should be noted that log P is only one molecular descriptor that relates to the solute-membrane affinity and many more molecular descriptors can be used to approximate this interaction. For instance, Kimura et al., (2004) used the dipole moment as another molecular descriptor relevant and Doederer et al., (2014) employed the polar surface area of the molecule besides the dipole moment, when describing contaminant transport. Other studies (Nghiem et al., 2004b; Semiao and Schäfer, 2013) also evidenced the considerable difference that can be observed, when comparing the filtration results obtained by NF and RO membranes. Even comparing NF membranes operated under identical conditions, the differences for rejection can be as large as 35-55% with one NF membrane and 80-85% with a second NF membrane tested (Semiao and Schäfer, 2013).

The final example is the NDMA molecule, which is very small (74.1 g/mol), neutral, and hydrophilic ($\log K_{ow} = -0.57$). In water reclamation plants, it may be formed as an undesired byproduct of chloramination employed to control membrane biofouling as secondary effluent is rich in NDMA precursors (Farré et al., 2011). Due to its small size and hydrophilicity it is not well rejected, even by RO membranes. Different full-scale studies show large variations in the rejection of NDMA by RO membranes ranging from almost no retention to up to 86% rejection (Fujioka et al., 2012; Fujioka et al., 2013a). Most of these full-scale plants use very similar commercial membranes that are dominating the water reclamation market. Another study of the same authors (Fujioka et al., 2013b) shows very well in a laboratory scale study, how the NDMA rejection increases by employing different membranes ranging from a tight NF membrane (Dow NF90) to a low pressure RO membrane (Hydranautics ESPA2) very frequently applied in water reclamation processes and finally to a high rejection seawater RO membrane (Hydranautics SWC5). The respective rejection percentages reported are 8%, 32-42%, and 79-85%.

In summary, it is clear that NF and RO membranes can provide high rejection percentages for many contaminants. However, hydrophobic contaminants can adsorb on the membrane, which will decrease its rejection compared to hydrophilic or charged contaminants. Sufficiently small molecules in turn may also be badly rejected as size exclusion becomes inefficient. Fouling layers can as well increase or reduce contaminant passage through the membrane. Finally, it should be reiterated that membrane processes are separation processes, i.e. the contaminant load in the feedwater is not really removed. It is rather enriched towards a concentrate stream that may require further treatment depending on local discharge opportunities (Joo and Tansel, 2015; Xu et al., 2013).

3. Homogeneous advanced oxidation processes with short-term perspectives

There has been a rise in the number of developed homogeneous AOPs during the last decade and the areas of potential application of these have increased dramatically (Klavarioti et al., 2009). Various efforts have been made by many research teams to critically review the findings of the relevant studies investigating the potential of homogeneous AOPs to degrade various CECs (Klavarioti et al., 2009; Malato et al., 2009; Rizzo et al., 2013; Oturan and Aaron 2014; Ribeiro et al., 2015; Barbosa et al., 2016; Formisano et al., 2016). However, to the best of our knowledge, no full-scale application and operation of these processes has been reported so far. Full-scale operation of these processes bears various inherent restrictions that slow their development and application at full scale. Similar to the conventional oxidation processes, these are: (i) the absence of explicit regulations for the elimination of CECs from wastewater and (ii) the wide presence of diverse scavengers in wastewater, including both organic (e.g., humic and/or fulvic acids, amino acids, proteins and carbohydrates) and inorganic species (e.g., sulphide, carbonate, bicarbonate, bromide and nitrate) that hinder the degradation of CECs by quickly engaging HO•. In order to address this last constraint, it is thus suggested that bench- and pilot-scale research encompassing homogeneous treatment processes be performed using realistic matrices, namely urban WWTP effluents in order to gather information as close as possible to the real-case circumstances.

The experimental application of treatment processes is also limited by the variability of the effluents, a factor that cannot be excluded during the application of wastewater treatment processes (e.g., location, point and non-point sources of pollution, type of treatments applied, production of transformation products in the WWTP, etc.) (Pera-Titus et al., 2004; Song et al., 2009). A literature survey is thus herein conducted on the studies reporting the application of homogeneous AOPs not established at full scale to remove the selected CECs from real urban wastewater effluents. Only publications dealing with real urban wastewater were considered (including spiked, real wastewater).

The UV/H_2O_2 oxidation involves the photolysis of H_2O_2 by UV radiation which is absorbed at <300 nm wavelengths, producing a homolytic scission of the O-O bond of the molecule and leading to formation of HO• radicals, which in turn contribute to H_2O_2 decomposition by secondary reactions (Liao and Gurol, 1995). UV radiation can also be employed to enhance the ozone decomposition by producing highly reactive HO• radicals. The fact that COD is reduced while DOC only changed slightly during the ozonation process indicated that ozone treatment transformed the structure of organic matter and formed transformation products mainly via direct oxidation (Pešoutová et al., 2014). UV photolysis of O_3 in water yields H_2O_2, which in turn reacts with UV radiation or O_3 to form HO•. The degradation of less reactive compounds can be enhanced by HO• radicals. As a consequence, the UV/O_3 treatment achieved a much better DOC reduction (Pešoutová et al., 2014). The role of pH is important when conducting homogeneous AOPs, as different AOPs operate optimally at different pH values. Possible examples include UV/H_2O_2, UV/peracetic acid (PAA) and photo-Fenton processes. AS matter of fact, the reaction rate of UV/H_2O_2 photolysis is higher in alkaline media, which may be attributed to the fact that the HO_2^- anion resulting from the ionization of H_2O_2 can strongly absorb UV radiation and produce HO•, the superoxide radical anion $HO^{\bullet}_2$ and singlet oxygen O^*_2 (López-Peñalver et al., 2010). UV/PAA has been recently investigated in the abatement of CECs from wastewater (Cai et al., 2017; Rizzo et al., 2018b). In particular, it was shown to be highly efficient at near-neutral pH for the degradation of pharmaceuticals since the pKa value of PAA (i.e. 8.2) falls within the inherent pH of the wastewater (Cai et al., 2017). The synergistic effect of combined UV and PAA has been also attributed to the formation of HO• and 'active oxygen' by the photolysis of PAA.

Photo-Fenton treatmentinvolves the catalytic breakdown of H_2O_2 in reaction with ferrous iron in an acidic medium (optimum pH = 2.8) to form active transitory species such as HO•, in the presence of artificial UV-Vis or sunlight. The photo-Fenton oxidation has been widely studied for wastewater treatment, due to its high effectiveness for the elimination of recalcitrant CECs present in complex aqueous matrices. The increase in the reaction rate observed (compared with classical Fenton) when an irradiation source is added, is due to the reduction of Fe^{3+} to Fe^{2+} ions, a reaction which produces HO• and regenerates Fe^{2+} ions that can further react with H_2O_2 molecules (Will et al., 2004). This increases the amount of Fe^{2+} and the Fenton reaction is accelerated (Tamimi et al., 2008). In addition, operating the Fenton process with solar irradiation has resulted in added advantages to this type of photochemical AOP, as the process is simple and more efficient than solely chemical AOPs. However, the main shortcomings of this process (e.g., the need for pH adjustment of the water matrix, sludge treatment and disposal as well as high cost due to H_2O_2 and catalyst consumption) still limit its broader full-scale application (Pliego et al., 2015).

Many Fenton-based processes have risen in the last years, suggesting the future intensification of the use of the classical Fenton process coupled to radiation or electrochemistry, and/or involving heterogeneous catalysts. Photo-Fenton represents a promising AOP for the abatement of a wide variety of CECs present in urban wastewater due to its environmentally friendly application and the prospect of operating under natural solar irradiation hence, lowering the operation cost considerably. The efficiency of the photo-Fenton system in degrading CECs is driven by several operating parameters, among others the dose of the Fenton reagent (i.e. H_2O_2 and iron concentrations), pH, and organic/inorganic content of the wastewater matrix. Malato et al., (2009) and recently Wang et al., (2016) presented in their reviews the main process parameters that affect the Fenton/photo-Fenton efficiency with respect to the abatement of various CECs dissolved in water or wastewater. The optimization of the catalyst and oxidant doses make the process capable of treating complex water matrices such as urban wastewater effluents, with many cases resulting in rapid and complete abatement of CECs. It has been clearly demonstrated in the scientific literature that the increase of H_2O_2 concentration results in higher generation of HO•, which in turn leads to the increase of the degradation rate. Nevertheless, the use of excessive oxidant concentration is not encouraged either, since massive amount of H_2O_2 can induce antagonistic reactions (i.e. reaction of the oxidant with the produced hydroxyl radicals) and thus the formation of radicals that are less reactive than the hydroxyl radicals. Some researchers reported that the stepwise addition of H_2O_2 is a good way to improve the treatment efficiency (Klamerth et al., 2010) due to a moderate concentration of the oxidant in the reaction system. Despite the limitations of the process, the high efficiency of the photo-Fenton technology for the treatment of various CECs present in urban wastewater has prompted its investigation at pilot scale through the development and application of solar concentrating parabolic collectors (CPCs). Then, natural solar light can be exploited, which dramatically lowers the operational cost of the process and, thus, provide a major step towards full-scale application. The results obtained from the pilot-scale applications, are quite satisfactory regarding the complete abatement of a plethora of PhACs, among others antibiotics (Michael et al., 2012; Karaolia et al., 2014), nonsteroidal anti-inflammatory drugs (Radjenović et al., 2009), analgesic drugs (Klamerth et al., 2009; Radjenović et al., 2009), hormones (Klamerth et al., 2009) and x-ray contrast media (De la Cruz et al., 2012). The final separation of soluble iron species from the treated wastewater, in order to comply with the regional regulatory limits for effluent discharge, is generally not necessary if the concentration of Fe is kept below 5 mg/L, a typically effective catalyst concentration.

Another important factor that strongly influences homogeneous processes performance with regard to the abatement of CECs, is the complex chemical composition of dissolved effluent organic matter (dEfOM) present in wastewater. dEfOM components react readily with hydroxyl radicals (10^8-10^{10} M^{-1} s^{-1}), thus reducing the radical concentration and the direct attack of HO• towards the target CECs. Under the inherent wastewater pH, ferric iron (Fe^{3+}) can be complexed by dEfOM resulting in the formation of stable and soluble complexes (Fe^{3+}-dEfOM) that can participate in further reactions. This approach has removed the burden of the economic limitation of the process associated with the chemical cost for pH rectification. However, CECs degradation during photo-Fenton tends to be slower at neutral pH than at the optimum pH value (De la Cruz et al., 2012). It has also been demonstrated that by adding iron at different steps (i.e. sequential iron dosage), it is possible to operate photo-Fenton at initial neutral pH without substantially decreasing the reaction rate compared to photo-Fenton at pH 2.8 (Carra et al., 2013). It was also confirmed by some studies that the occurrence of inorganic anions (i.e. carbonate, chlorides, sulfates) in wastewater, can influence the degradation rate of CECs during the photo-Fenton treatment by consuming hydroxyl radicals. The inorganic anions scavenge the HO• to generate inorganic radicals which, in turn, react with the organic contaminants, albeit at a slower rate (Klamerth et al., 2009; Michael et al., 2012). Phosphate is a specific case as it can precipitate the iron catalyst in a wide range of pH.

As most AOP investigations up to the present focus on the assessment of operational parameters and kinetic investigations of specific compounds, a more comprehensive approach as to the abatement of a wide array of CECs in real situations is needed. As shown in Table SI8, experiments with AOPs in more realistic conditions, such as under real sunlight rather than under simulated solar irradiation, as well as their execution at pilot scale is needed. Moreover, the more extensive use of real WWTP secondary CAS-treated effluent during the assessment of CECs abatement by AOPs rather than ultrapure/deionised water and simulated wastewater effluents is crucial to attain substantial conclusions regarding the production of treated effluents that are safe for disposal into the environment, or reuse for other applications e.g. agricultural irrigation.

4. Perspective methods

UV/TiO_2, heterogeneous photo-Fenton, photocatalytic ozonation, photocatalytic membrane processes, electrochemical oxidation and hybrid processes, among others, are some processes/technologies that have been investigated in the removal of CECs but, due to technological limitations and costs, have not yet been applied at full scale as advanced treatment of urban wastewater. They can be considered as long-term perspective methods that have received less attention than other AOPs in urban wastewater treatment so far (Klavarioti et al., 2009; Malato et al., 2009; Rizzo et al., 2013; Oturan and Aaron 2014; Ribeiro et al., 2015; Barbosa et al., 2016). Considering the extensiveness of the subject, this section intends to provide the state-of-the-art surrounding the application of the abovementioned perspective processes, presenting mostly valuable perspectives on the fundamental variables and design parameters that affect the processes' efficiency with regard to the removal of CECs. Figure SI1 shows the results of the search based on the Scopus database using as keywords the name of each perspective process and "*wastewater*" (i.e. this figure is the only one including results with unrealistic matrices, such as distilled water), while Figure SI2 shows the results obtained from the search to prepare Tables SI8 and SI9 (i.e. solely publications dealing with simulated and real urban wastewater (spiked or not), describing the abatement of CECs). Also here, only a few studies have been conducted with realistic matrices (Figure SI2), mainly with spiked real wastewater, heterogeneous photocatalysis being the most investigated process.

4.1. Heterogeneous photocatalysis (UV/TiO_2) and photocatalytic ozonation

. The elimination of various CECs through heterogeneous photocatalysis (particularly that based on titanium dioxide, TiO_2) has been explored by many researchers (Figure SI1). CECs can be degraded by radicals formed from photoexcited electrons or photoinduced holes, or through direct oxidation by holes on the surface of the catalyst (Malato et al., 2009). This process has been employed at both bench- and pilot-scale, mainly using TiO_2-based materials as catalysts. Aeroxide® TiO_2 P25 has been revealed to be frequently the most active photocatalyst, among the numerous semiconductors so far investigated, due to its specific features such as crystalline phases, particle size, among others.

Two core configurations have been established in a TiO_2/UV reaction system: the catalyst can be either suspended (i.e., slurry design) or held on a carrying material (i.e. immobilized design). The efficiency of a heterogeneous photocatalytic slurry system is mostly dependent on the irradiation, catalyst load, initial concentration of target contaminants and wastewater physical-chemical characteristics (such as pH and dEfOM), matrix effects being limiting aspects for application of photocatalytic-based systems in urban WWTPs (Ribeiro et al., 2015). Normally, a low catalyst amount might end in a surface site controlled reaction and therefore in lacking generation of reactive radicals, whereas a high catalyst dose (above the optimum load) can decrease the transmittance of the UV radiation due to the augmented turbidity. TiO_2/UV systems experience considerable interferences by the constituents of dEfOM in wastewater, which can prevent the degradation of CECs. The optimum TiO_2 load and reaction time required for the degradation of a particular CEC are therefore dependent on the water characteristics, and are often established through bench-scale studies using the wastewater matrix of interest. Furthermore, there are some organic compounds able to improve the efficiency of the process due to their photosensitizing properties when exposed to sunlight. In fact, photosensitizing components occurring in a water matrix can promote photo-assisted processes by producing reactive species, as already reviewed (Tsydenova et al., 2015), but the process hindering is most frequently observed (Malato et al., 2009). The main causes of diminished abatement rates of CECs through matrix effects are: (i) scavenging of hydroxyl radicals by anions (e.g., bicarbonate, chloride, sulphate), producing radicals with lower oxidation potentials; (ii) screening effect, when matrix components and the catalysts have light absorption at same wavelengths; (iii) turbidity that might avoid light transmission through the bulk of the solution; and (iv) adsorption onto catalyst surface of some organic and inorganic species (e.g., phosphate and carbonate). The removal of CECs by heterogeneous photocatalysis is also pH-dependent since the charge of both the catalyst particles and the CECs relies on the medium pH value, interfering as a result with the adsorption and degradation of the contaminants. The performance of the process can be assisted by adding a strong oxidant such as H_2O_2, which can accept an electron from the conduction band, reducing the electron-hole recombination (Wang et al., 2016).

In particular, solar-driven photocatalysis is a topic that gained a huge attention over the recent years and several reviews on this subject were published (Malato et al., 2002, 2009; Oller et al., 2011; Spasiano et al., 2015). Regardless of the successful demonstration that solar-driven TiO_2 photocatalytic processes are effective in eliminating a multiplicity of CECs from wastewater, there are some major technical obstacles limiting their performance. The main weakness of this process is the restricted sunlight application due to the narrow overlap (small fraction of the UV range) between the absorption spectrum of the reference commercially available photocatalyst TiO_2 and that of the solar light (Ribeiro et al., 2015). Different approaches have been developed to broaden the absorption of TiO_2 toward the visible region to improve the photocatalytic treatment efficiency. These approaches also include TiO_2 doping by non-metallic species such as N (Rizzo et al., 2014; Ata et al., 2017). Moreover, from an engineering perspective, the available active surface is much higher if a suspension of the catalyst is used; however, the catalyst particles have to be separated from the treated water. Fixed bed photocatalytic reactors have been applied to circumvent this problem (Malato et al., 2009; Vaiano et al., 2016; Sacco et al., 2018). Another possibility to avoid the constraints related to the recovery of the catalyst is the use of photocatalytic membrane reactors (PMRs), where the catalyst is confined in the reaction solution through a membrane, being already tested for the removal of various CECs (Mozia et al., 2010). Other examples of hybrid membrane filtration-AOPs systems have been described in the literature (Ganiyu et al., 2015), including physically separated photocatalytic and membrane units or photocatalytic membranes (TiO_2 coated membranes). For instance, TiO_2 modified ceramic membranes and graphene oxide-based ultrafiltration membranes (Athanasekou et al., 2015) were recently described for the degradation of various CECs; however their application in urban wastewater effluents has not been investigated yet.

The integration or combination of AOPs is also a common methodology. As predictable, when two or more approaches are associated, a better performance is achieved in comparison to the single treatments, with additive effects or even synergistic effects where the efficiency of the whole treatment is superior to the sum of that of each individual process (Agustina et al., 2005). For instance, photocatalytic ozonation combines the best features of photocatalysis and ozonation processes and some reports were already published in this domain (Figure SI2), photocatalytic ozonation being described as more cost-effective in the elimination of some CECs (Xiao et al., 2015). Photocatalytic ozonation suffers from neither the poor mineralization often attained by ozonation nor the low oxidation degree of photocatalysis when treating realistic matrices. The strong oxidizing power of ozonation joined with photocatalysis promotes a fast degradation of recalcitrant CECs with an improved TOC reduction (Agustina et al., 2005). Photocatalytic ozonation was recently reviewed by some authors (Mehrjouei et al., 2015; Xiao et al., 2015) who stressed the catalysts typically applied, the kinetics and mechanisms of reaction, the cost-effective aspects, as well as the effect of operational parameters, such as the effect of pollutant concentration, pH, temperature, light intensity, ozone dosage and catalyst properties and dosage. These authors emphasized the challenging need of developing catalysts highly active towards visible light, immobilising the photocatalyst particles and designing novel reactors to overcome their mass transfer limitation. The cost related to the conventional UV lamps can be overcome by the application of more efficient and long-lasting UV sources (Xiao et al., 2015), such as light emitting diodes (LEDs) (Moreira et al., 2016). Employing reflective materials in UV reactors is another efficient way to reduce the energy cost when using renewable sunlight. The photoreactors can be classified in three categories: parabolic trough collectors (PTCs), non-concentrating collectors (NCCs) and compound parabolic collectors (CPCs) (Spasiano et al., 2015).

4.2. Electrochemical oxidation

A limited number of bench-scale studies have been conducted for the assessment of the capability of electrochemical oxidation processes to remove CECs from urban wastewater, as most of the available studies have been focused on the removal of organic content, in terms of COD and DOC. Only few works reported the electrochemical oxidation of some CECs spiked in wastewater effluents, such as antibiotics, caffeine and BPA (Fabianska et al., 2014; Martin de Vidales et al., 2015; Rodrigo et al., 2010; Chen et al., 2014; Zaviska et al., 2012). Electrolytic cells offer multiple technical benefits, including mild operation conditions (lack of chemicals), amenability to automation, compact and modular reactor design, and an ability to adjust to variable organic loads. However, one critical challenge to wider adoption of electrochemical oxidation for wastewater treatment is the relatively high cost of electrodes and concerns related to the generation of toxic organic chlorine- and bromide-containing transformation products in the treated water. To gain insight into the use of electrochemical treatment processes under conditions likely to be encountered in wastewater applications, recent research on these processes was reviewed focusing on the effect of various operational parameters on process performance (Sirés et al., 2014; Radjenovic 2015). The efficiency of electrochemical treatment processes highly depends on the electrode material. Boron-doped diamond (BDD) electrodes have been studied extensively in recent years. The distinct features of BDD electrodes (“non active” anodes), such as high overpotential for oxygen evolution, make them better suited for the direct oxidation of contaminants than metal oxide anodes. The electrochemical oxidation is strongly pH dependent. Even though there are many scientific reports on the influence of pH, the results are controversial due to the different organic structures and electrode materials that have been examined. Usually, the oxidation potential of an electrochemical system in acidic medium is higher than that in alkaline medium. The performance of the process in removing CECs is also affected by the presence of inorganic anions and dEfOM intrinsically present in wastewater, which can react with the electro-generated hydroxyl radicals and other reactive oxygen species. A comprehensive review on the application of different electrochemical processes for the abatement of pharmaceutical residues from both synthetic and real wastewater effluents was already published (Sirés and Brillas, 2012), with antibiotics and non-steroidal anti-inflammatory drugs being the most studied. Electrochemical membranes (EMs) are hybrid systems, in which degradation can occur at the EMs surface, but the mode of pollutants' rejection is not well understood. The need for research on this topic was already emphasized, addressing several challenges (Ganiyu et al., 2015).

4.3 Sonolysis and hydrodynamic cavitation

Among different AOPs, sonolysis is a process which has not been widely examined in the currently available scientific literature, with even fewer studies existing on sonolytic degradation of CECs in urban wastewater effluents. For instance, the abatement of various PhACs including diclofenac, amoxicillin and carbamazepine in real urban wastewater effluents was investigated (Naddeo et al., 2009, 2013), and the findings revealed that the conversion of the examined compounds is enhanced at increased applied electrical power densities, in acidic conditions and in the presence of dissolved air, indicating the high operational financial costs required for the optimum operation of sonolysis in real-world scenarios. Other studies have not shown significant benefits of sonolysis for wastewater treatment in comparison to other AOPs, as regards DOC removal and energy consumption (Dialynas et al., 2008).

Cavitation is a physical phenomenon, where the formation, growth and subsequent collapse of small bubbles (cavities) in a liquid, release high amounts of energy that can drive chemical and mechanical effects. Generally, there are two kinds of cavitation, hydrodynamic and acoustic. In hydrodynamic cavitation (HC), bubble inception and collapse are the result of an increase in fluid velocity and accompanied decrease in static pressure. This phenomenon can occur, when the fluid passes through a constriction (e.g., valves), or gets a rotational impulse, as in the case of hydraulic machines. HC is usually generated either by high-velocity passage of the liquid through a constriction such as an orifice plate or Venturi pipe, the use of high-speed homogenizers, devices based on the rotor-stator principle or by a rotating propeller blade. In the literature, there are many reports investigating the potential application of the HC phenomenon, where HC was used as a tool for disinfection (Heath et al., 2013), cell disruptions (Jyoti et al., 2003), preparation of nanoparticles (Save et al., 1997), and lately also for removal of organic compounds in wastewater treatment (Joshi et al., 2012; Patil et al., 2012; Wang et al., 2009).

In the case of acoustic cavitation (ACa), formation, growth and subsequent collapse of bubbles is a result of high frequency acoustic irradiation (normally in the range of 20 to 1000 kHz) of liquids (Klavarioti et al., 2009; Sangave et al., 2004). The extreme conditions occurring during ACa, trigger production of HO• by decomposition of water molecules. To improve the efficiency of the process, e.g. to increase the amount of HO• formed, ACa can be employed in combination with, for example, ozone, H_2O_2 and Fenton’s reagent. The improved efficiency can be exploited for treatment of more complex matrices (i.e., wastewater).

From a literature survey investigating the efficiency of ACa and HC for abatement of organic contaminants, it is evident that most studies on the abatement of CECs deal with matrices less complex than wastewater, namely deionized water and groundwater. Recent studies investigating the efficiency of ACa focus mostly on:

(i) employing ultrasound alone (Campbell et al., 2015; Lin et al., 2015; Kim et al., 2016; Sutar et al., 2009), with focus on the effects of power density, frequency, solution pH, temperature and compound concentration;

(ii) investigating ultrasound in combination with different chemicals to increase efficiency (i.e. O_3, H_2O_2, Fenton's reagent, persulfate oxidant, surfactants, zero valent iron) (Lim et al., 2014; Hao et al., 2014; Lin et al., 2016; Prado et al., 2017);

(iii) employing ultrasound combined with different catalysts to increase efficiency (i.e.TiO_2, SiO_2, SnO_2, and titanosilicate) (Hou et al., 2013; Hassani et al., 2017);

(iv) studying sonoelectrolysis and sono-photoelectrolysis (Finkbeiner et al., 2015; Martin de Vidales et al., 2015);

(v) applying a combination of microwaves and ultrasound (Horikoshi et al., 2011) or UV and ultrasound (Torres et al., 2007).

Cavitation efficiency in the removal of a model compound, sulfamethoxazole, was investigated in different matrices (deionized water, synthetic wastewater and "real" wastewater) (Table SI9). Even though the studies cannot be directly compared due to different experimental conditions, the difference in removal efficiency of the parent compound is evident. Accordingly, the highest removal of the parent compound was obtained in the simplest matrix. No removal was achieved by single sonolysis in synthetic wastewater, whereas a removal efficiency of 68% was observed by combining sonolysis and ozonation, being the abatement by ozone alone lower. This synergistic effect of sonolysis and ozonation is in agreement with a study reporting that sonolysis can improve the cleavage of S-N bond, so that sulfamethoxazole might be more easily attacked by ozone (Prado et al., 2017). The same effect was also observed employing HC with the addition of H_2O_2, to study the removal of diclofenac and carbamazepine (Zupanc et al., 2013, 2014). Two types of HC experimental set-ups were tested: pulsating HC (PHC) using a symmetrical Venturi constriction and shear-induced HC (SHC). In both cases, removal efficiency was tested in wastewater effluents. Both types of cavitation were optimized in the terms of H_2O_2 addition, temperature and time of cavitation. The SHC reactor was more efficient to remove the two recalcitrant PhACs carbamazepine (62%) and diclofenac (79%).

Literature reveals that not many compounds have been studied applying cavitation and using synthetic or “real” wastewater matrices (Table SI9). Results show that the highest removals are achieved when ACa or HC are combined with other treatments (e.g., O_3) or by addition of different chemicals (e.g., H_2O_2) (Table SI9).

Unfortunately only a few studies available in scientific literature addressed the use of cavitation to remove CECs from wastewaters (Table SI9). However, some results are promising, e.g., the recalcitrant antiepileptic drug carbamazepine was removed at a high extent (> 96%), when HC was coupled to ACa at optimized conditions (Braeutigam et al., 2012). In addition, the maximum extent of removal of the biologically resistant non-steroidal anti-inflammatory drug diclofenac (66%) was obtained using combined HC/UV process, whereas removal rates of 27% and 43% were obtained in single HC and UV processes, respectively (Bagal et al., 2014). More rare are studies involving HC in real wastewater. When HC was applied as pre-treatment step to biological treatment, all tested compounds (e.g., clofibric acid, ibuprofen, ketoprofen, naproxen, diclofenac, carbamazepine that are not well degradable) were removed to below the limits of detection (Zupanc et al., 2013, 2014). When compared to ACa, HC as an AOP has several advantages including lower investment costs and easier scale-up (Braeutigam et al., 2012; Gogate 2002, 2005). This makes HC worthy of investigation, but a cost benefit analysis is needed before any firm conclusions can be drawn.

4.4 Catalytic wet (air or peroxide) oxidation

In catalytic wet air oxidation (CWAO) oxygen is dissolved in the liquid-phase at high temperatures and pressures, whereas catalytic wet peroxide oxidation (CWPO) employs H_2O_2 and a suitable catalysts not limited to iron-based ones (as in the case of the Fenton process). These AOPs have not been widely investigated for the treatment of realistic urban wastewater, some examples being the degradation of industrial compounds by CWPO (Rueda-Marquez et al., 2015) and PhACs from urban effluents by CWAO (Benitez et al., 2011; Ribeiro et al., 2016).

5. Consolidated vs new processes: process comparison, advantages and drawbacks

In this paragraph an attempt to compare consolidated and new processes was made, summarizing some of the comparison studies available in scientific literature on real wastewater in full or pilot scale as reviewed in the previous paragraphs. Adsorption by PAC and GAC, and ozonation were selected as examples of consolidated processes, photo-Fenton and UV/H_2O_2 as example of homogeneous AOPs with short-term perspectives and UV/TiO_2 was selected as example of a mid to long term perspective process. To make the comparison as close as possible to real scenarios only studies on real wastewater at full and pilot scale were summarized in Table 3. Economic and energetic process performances are discussed in the subsequent section 6.

When different processes are investigated in the same work, the comparison is more reliable because the respective tests are typically performed under comparable operating conditions. For example, oxidation by ozone (followed by sand filtration) and PAC adsorption (followed by either ultrafiltration (UF) or sand filtration) processes were investigated at pilot scale in parallel, over more than one year, at the municipal WWTP of

Lausanne, Switzerland (Margot et al., 2013). 70 CECs were removed on average over 80% compared with raw wastewater, with an average ozone dose of 0.78 g O_3/g DOC or a PAC dose between 10 and 20 mg/L. The authors considered PAC-UF treatment to be the most suitable option at this site, because the strongest decrease in toxicity and better disinfection was observed with this treatment.

Photo-Fenton (UV-C/Fe/H_2O_2) process was compared to UV-C/ H_2O_2 process and high abatement of the target CECs was observed at pilot scale when the optimal operating condition (Fe/ H_2O_2 ratio) was established (De la Cruz et al., 2013). Interestingly, good results were also observed with photo-Fenton at neutral pH (6-7), but it is worthy to mention that UV-C/ H_2O_2 resulted in higher efficiencies compared to photo-Fenton process. Anyway, when (solar) photo-Fenton process was operated at neutral pH through the “support” of chelating agents (namely ethylenediamine-N,N′-disuccinic acid, EDDS), high abatements (>95%) were observed for all the target CECs (Klamerth et al., 2013). Although solar photo-Fenton at pH3 resulted in a shorter treatment time, the need for acidification and subsequent neutralization increases effluent salinity as well as treatment costs, making this option not attractive. Solar photo-Fenton was also compared to sunlight/TiO_2 and ozonation processes in the abatement of 66 micropollutants from urban wastewater showing that sunlight/TiO_2 was the slower process (Prieto-Rodrıguez et al., 2013a).

Advantages and drawbacks of advanced technologies discussed in the previous paragraphs are summarized in Table 4. Additionally, where relevant, recommendations are provided. It is not possible to provide a “best” technology to minimize the release of CECs into the environment. The decision on best technology needs to be made for each location depending on the local conditions (e.g., available space and solar energy, cost of electricity), the water quality derived from the biological treatment and on the required effluent quality (reuse requirements, disinfection needs).

Table 3

Table 4

6 Economic feasibility and cost evaluation

Advanced treatment of urban wastewater has been in operation for direct or indirect reuse of wastewater for drinking water purposes to overcome water scarcity, mostly as a managed aquifer recharge system to obtain good groundwater quality. However, full-scale evidence of advanced wastewater treatment for protecting the aquatic ecosystem from adverse effects caused by CECs being discharged from WWTPs can be mainly found in Europe and are further discussed here.

Past investigations of the last decade on pilot- and full-scale compared different processes of advanced wastewater treatment methods for their technical and economic feasibility (Hollender et al., 2009; Abegglen and Siegrist 2012; Margot et al., 2013; Prieto-Rodrıguez et al., 2013; De la Cruz et al., 2013). The filtration with tight membranes as used in nanofiltration or reverse osmosis were generally found to be more cost-intensive. In geographical areas with high yearly average solar irradiation (between latitude 40ºN and 40ºS), solar driven AOPs, after further technology development, may well be competitive with other advanced treatment technologies for CECs abatement from urban wastewater.

In all treatment processes except the treatment with AC or solar energy processes (solar photo-Fenton), main costs arise from electricity consumption. In Table 5, rough estimations on the required electrical energy are summarized.

Table 5

Advanced urban wastewater treatment with a target of CECs abatement has been implemented cost-efficiently by adsorption (with both PAC and GAC) and by ozonation so far. Current implementations are based on this state of the art. Currently Switzerland is the only country to have a legislation for advanced treatment of urban wastewater to protect the environment. The new Swiss water protection act entered into force on January 2016 and requires an upgrade of selected WWTPs by 2040 (www.bafu.admin.ch). According to that CECs need to be removed by 80% relative to the raw wastewater (Eggen et al., 2014). The treatment target is defined by the abatement of a selection of CECs from a list of twelve defined compounds (Bourgin et al., 2018). So far several plants are in full-scale operation in Switzerland, either with ozonation or PAC treatment. Germany currently has the highest number of plants designed to remove CECs with around 20 installations, although the actual legislative situation does not explicitely require the construction of advanced treatment units. The issue is still being discussed controversially in different states of the country. However, two federal states North Rhine-Westphalia (NRW) and Baden-Württemberg (BW) have long decided to take an action. Several municipal WWTPs have been upgraded with ozonation, PAC or GAC units. Three competence centres were founded in the two German states NRW and BW, as well as in Switzerland to ensure knowledge exchange on the application of advanced wastewater treatment (www.kompetenzzentrum-mikroschadstoffe.de; www.koms-bw.de; www.micropoll.ch). In

Austria, pilot-scale experiments with ozonation and subsequent BAC are running with foreseen full-scale application in the near future for specific situations such as missing receiving surface water and subsequently infiltration ponds, resulting in ground water recharge (Kreuzinger et al., 2015; Haslinger et al., 2017). Also in other countries like France and the Netherlands full-scale application with ozonation or AC treatment are in operation.

Moreover, the state NRW has also been funding feasibility studies for the upgrading of municipal WWTPs with a CECs removal step (Antakyali 2016). The studies evaluate the suitable process configurations depending on the individual features of the treatment plants. In most cases ozonation, PAC treatment and GAC reactors are assessed comparatively for constituting the state of the art in CECs removal. Besides the technical feasibility of the processes, monetary costs play an important role and eventually the best option can be highlighted after the assessment. Since 2013 the number of the studies in NRW increased remarkably, creating a database on the monetary costs of the above mentioned techniques. Detailed cost calculations for advanced treatment for Germany and Switzerland are summarized in the following section.

6.1 Cost evaluation: methodology and assessment

A cost evaluation was conducted for ozonation, PAC and GAC adsorption processes according to the German guidelines for comparative cost calculations (KVR-Guidelines). Investments basically consist of the construction costs (civil work), process control (electrical work), machinery and incidentals. Reinvestment costs are considered according to the given life cycle of each group, which are 30 years for civil works, 15 for machinery and 10 for electrical works. Regarding the operation of the plants, the costs are divided as energy, personal, material, maintenance and in case of PAC also disposal costs. Investment and operation costs are adjusted to a selected base year after being calculated separately as functions of life cycle and assumed interest rates, to allow comparability of the studies conducted in different years. The cost evaluation given in Figure SI3, Figure SI4 and Figure 4 is based on data originating from the 42 readily completed feasibility studies conducted in the NRW state of Germany in the years 2009-2016. Swiss data are compiled from the cited references.

Figure SI3 presents the specific capitalised investment costs, which consider the initial investments and the required re-investments per treated m³ wastewater. Ozonation exceeds its alternatives at machinery costs, due to the relatively high costs of ozone generation and dosing systems. The implementation of PAC system require higher costs for civil works, when separate contact and/or settling tanks are constructed. GAC plants are basically preferred only when a filtration plant is readily available in the wastewater treatment plant, which is reflected in the moderately lower investment costs. If a new GAC plant is to be constructed, investment costs are expected to be significantly higher. According to these figures, the investment-related total costs vary between 0.035 and 0.05 € per treated m³ wastewater.

Operation costs are presented in Figure SI4. Studies which constitute a basis for this evaluation mainly consider a specific ozone dose of 0.6-0.8 gO_3/g DOC and a PAC doses of 10-20 mg PAC/L. Main costs for ozonation arise from the electrical energy needed for the production of ozone from air or liquid oxygen on site. For AC treatment, material costs of the carbon are controlling the costs. The variation of cost estimations in different plants is remarkably small, with the exception of the material costs for the GAC process. Relatively high variation in this cost element resulted from the insufficient full-scale experience to predict a realistic bed volume until the breakthrough of CECs. Yet when the total operation costs are compared, no remarkable difference is seen among different processes, as all yield to a median value of 0.04 €/m³.

The addition of capitalised investment-related and operation cost yield to the annual costs for the given life cycle. Specific annual costs for CECs abatement through ozonation, PAC and GAC processes are given in Figure 4, which enable a rough cost estimation varying with the treatment plant size. For the extension of small WWTPs with a CECs removal unit, costs may vary in a wide-range. From mid-scale plants (~50.000 PE) the costs rather drop to a range of 0.10 to 0.15 €/m³, decreasing further with the increasing plant size.

The presented results are based on plant designs assuming a certain ozone and PAC dose as well as a bed volume for GAC filters. Recent studies show that process optimisation may help increase the efficiency, e.g. by increasing the contact time of the GAC with pollutants to be removed, which in turn may be reflected in further reduction of the costs.

Figure 4

In Switzerland, costs were estimated within the project Micropoll, where full-scale installations of ozonation and treatment with powdered activated carbon were evaluated (Abegglen and Siegrist, 2012; Hollender et al., 2009; Margot et al., 2013). Ozone was generated on-site from liquid oxygen. Primary energy was calculated for oxygen and PAC (production and disposal via incineration with activated sludge). No robust data for PAC production was available. Results are shown in Table SI10 and are in a similar range as the cost evaluations for Germany.

Solar photo-Fenton was compared with ozonation in the abatement of 66 CECs from urban wastewater at pilot scale and respective treatment costs were evaluated in terms of reagent consumption, labour, electricity, and investment costs based on a design flow of 5000 m^3/day and CECs abatements as high as 90% and 98%, respectively (Prieto-Rodrıguez et al., 2013a). The main costs in solar treatment arise from the investment for solar collectors. Treatment costs were estimated as 0.188 €/m^3 and 0.358 €/m^3 for 90% and 98% CECs abatement by solar photo-Fenton treatment and 0.450 €/m^3 and 0.560 €/m^3 for ozonation, respectively, using a dose close to 9.5 gO_3/m^3. However, nowadays ozonation for this application at full scale presents costs substantially lower and close to 0.25 €/m^3 for 9.5 g O_3/m^3. The overall conclusion is that solar photo-Fenton could be competitive with ozonation under certain boundary conditions and after more technological development, and therefore a choice to explore at least in sunny countries.

UV-C/H_2O_2 process was investigated at pilot scale with a reactor placed at the end of the treatment process of a WWTP (Vidy, Lausanne) and operated in continuous mode (De la Cruz et al., 2013). Operating costs were evaluated for 4 different flow rates (in the range 48-336 m^3/d), CECs (5 selected among the 22 investigated) abatements higher than 80% and different operating conditions (residence time in the range 10-67 s; H_2O_2 dose in the range 20-50 mg/L). Per m^3 cost decreased as the flow rate was increased from 0.202 CHF/m^3 (0.18 €/m^3) for 48 m^3/d to 0.142 CHF/m^3 (0.12 €/m^3) for 336 m^3/d.

7. Concluding remarks and gaps

Consolidated advanced urban wastewater treatment methods, namely AC adsorption (with both PAC and GAC), ozonation and filtration by nanofiltration or reverse osmosis membranes, can effectively remove CECs. Several plants employing AC adsorption and ozonation have recently been implemented cost-efficiently at full scale in Germany and

Switzerland. Filtration with tight membranes as used in nanofiltration or reverse osmosis was found to be more cost-intensive. Nevertheless, reverse osmosis membranes have been implemented on full scale in potable reuse schemes in the United States of America, Singapore and Australia because of the additional benefit provided regarding salinity and metal reduction. For membrane filtration processes alternatives for the treatment of the concentrated waste stream should though be further evaluated.

In geographical areas with high yearly average solar irradiation (between latitude 40ºN and 40ºS), solar driven AOPs appear competitive with other advanced treatment technologies for CECs abatement from urban wastewater, though they are currently developed to a lower Technology Readiness Level, which makes comparison difficult. The same situation exists for many innovative processes and novel combinations of existing processes, which often have been studied only at small-scale or under non-realistic source water conditions so far.

The abatement of CECs from wastewater by AOPs depends on the operating parameters, the matrix composition and the abatement mechanisms occurring during the employment of each treatment technology. When applying ozonation or AOPs the generation of oxidation transformation products with potential biological effects, derived from either the CECs or the dEfOM itself, generates the need to perform ecotoxicological studies to evaluate the effect of these new products. A post-treatment with sand filters or BAC has been proven a suitable strategy to cope with this issue following ozonation, but increases treatment costs.

The huge local diversity of CECs and of the water matrix make optimisation essential for each application (adsorbent and/or flocculants choice, selection of membranes, dosing procedures, systems configurations, mixing conditions, etc). This indicates the need for knowledge systematisation and development of tools for prediction of CECs behaviour in wastewater treatment.

The lack of comparative investigations between consolidated (AC adsorption and ozonation) and new processes (namely novel AOPs) hamper the conclusive evaluation of the most suitable and cost effective solution/s for advanced treatment of urban wastewater. In any case, site-specific limitations (e.g., availability of space and solar energy, cost of electricity) may lead to different conclusions for two different sites. Most importantly, these comparative investigations should be designed and performed by taking into account different relevant end-points for a safe effluent discharge or reuse, such as CECs abatement, effluent toxicity, bacteria inactivation, by-products minimization or abatement, antibiotic resistance control and treatment cost.

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Figure captions

Figure 1. Abatement of 18 reviewed CEC as a function of the specific ozone dose. CEC are grouped according to their ozone reactivity as shown in Table 1.

Figure 1. Abatement of representatives for group A (diclofenac), group B (benzotriazole) and group C (iopromide) including the group-specific boundaries for the average abatement at 0.4-0.6 g O_3/g DOC according to Table 1. Nonlinear regression fit with exponential rise to maximum (f = a*(1-exp(-b*x))).

Figure 3. Visualisation of the three removal mechanisms by high pressure membranes. Adapted from Verliefde, 2008..

Figure 4. Specific capital annual costs of CECs abatement (selected process options only) (Antakyali 2017).

Table 1. Categorization of CEC according to their abatement during ozonation of biologically treated wastewater (0.4-0.6 g O_3/g DOC) and their reactivity with ozone

Group	**Abatement**	**Reactivity with ozone**	**Reaction rate k_{O3} (M^{-1} s^{-1})**
A	> 80	High	$> 10^3$
B	50-80%	Intermediate	10^2-10^3
C	<50%	Low	$< 10^2$

Table 2. Categorization of CECs according to their abatement by PAC and ozonation

CEC	**PAC**				**Ozonation**		
	DOC , mg/L	**PAC dose, mg/L**	**Abatement[a]**	**Reference**	**Specific ozone dose, g O_3/g DOC**	**Abatement[a]**	**Reference**
Sulfamethoxazole	5-10	15	Intermediate	(Boehler et al. 2012)	0.54 ± 0.05	High	(Bourgin et al. 2018)
Erythromycin	5.6 ± 0.9[b]	5-10	Intermediate	(Mailler et al. 2015)	0.56	High	(Schaar et al. 2010)
Clarithromycin	5-10	15	High	(Boehler et al. 2012)	0.54 ± 0.05	High	(Bourgin et al. 2018)
Azithromycin	7.3 ± 1.9[b]	10-20(12)[c]	High	(Margot et al. 2013)	0.67 ± 0.03	High	(Bourgin et al. 2018)
Ciproflaxacin	5.6 ± 0.9[b]	5-10	High	(Mailler et al. 2015)	0.64 ± 0.01	High	(Kovalova et al. 2013)
Diclofenac	5.0	3 (+50)[d]	High	(Altmann et al. 2015a)	0.54 ± 0.05	High	(Bourgin et al. 2018)
Carbamazepine	5.6 ± 0.9[b]	5-10	High	(Mailler et al. 2015)	0.50	High	(Altmann et al. 2014b)
Metformin	7.3 ± 1.9[b]	10-20 (12)[c]	Intermediate	(Margot et al. 2013)	0.78	Low	(Margot et al. 2013)
Metoprolol	5.0	3 (+50)[d]	High	(Altmann et al. 2015a)	0.54 ± 0.05	High	(Bourgin et al. 2018)
Bezafibrate	5.6 ± 0.9[b]	5-10	High	(Mailler et al. 2015)	0.50	Intermediate	(Altmann et al. 2014b)
Primidone	7.3 ± 1.9[b]	10-20(12)[c]	Intermediate	(Margot et al. 2013)	0.78	Low	(Margot et al. 2013)
Iopromide	5-10	15	Intermediate	(Altmann et al. 2015a)	0.54 ± 0.05	Low	(Bourgin et al. 2018)
17-Alpha - ethinylestradiol	-	20	Intermediate	(Sun et al. 2017)	0.12	High	(Sun et al. 2017)
17-Beta estradiol	7.3 ± 1.9[b]	10-20 (12)[c]	Intermediate	(Margot et al. 2013)	0.44	High	(Nakada et al. 2007)
Mecoprop	5-10	15	Intermediate	(Boehler et al. 2012)	0.54 ± 0.05	Intermediate	(Bourgin et al. 2018)
Bisphenol A	7.3 ± 1.9[b]	10-20 (12)[c]	High	(Margot et al. 2013)	0.56	High	(Schaar et al. 2010)
Benzotriazole	12	5-100	Intermediate	(Zietzschmann et al. 2014a)	0.54 ± 0.05	Intermediate	(Bourgin et al. 2018)
Methylbenzotriazole	7.3 ± 1.9[b]	10-20 (12)[c]	Intermediate	(Margot et al. 2013)	0.78	Intermediate	(Margot et al. 2013)
Acesulfame	11.4	20	Low	(Altmann et al. 2016b)	0.54 ± 0.05	Intermediate	(Bourgin et al. 2018)
Perfluorooctanic acid	-	-	-	-	5[g]	Low	(Thompson et al. 2011)
Perfluorooctanesulfonic acid	5.6 ± 0.9[b]	5-10	Low	(Mailler et al. 2015)	5[g]	Low	(Thompson et al. 2011)

[a] Abatement (High: >80%, Intermediate: 50-80%, Low: <50%)

[b] The average DOC content of the wastewater (± standard deviation).

[c] Median PAC dosage (mg/L).

[d] Continuous PAC dosing (initial dosage of 3mg/L, plus 50 mg/L).

[e] The average EBCT (± standard deviation).

[f] The average DOC content from four collected samples (seasonal variation).

[g] Applied Ozone dose (mgO_3/L).

Table 3. Studies on real wastewater based on consolidated processes (PAC, GAC, ozonation), homogeneous AOPs with short-term perspectives (photo-Fenton, UV/H_2O_2) and mid to long term perspective process (UV/TiO_2). Only studies dealing with actual wastewater at full- and/or pilot-scale are presented.

CEC	**Process**	**Scale of study**	**Water matrix[1]**	**Organic matter (mg/L)**	**CEC initial concentration**	**Comments**	**CEC abatement (%)**	**Reference[2]**
Sulfamethoxazole	PAC	Pilot/full	RMW	5-10 (DOC)	171 ng/L (data only from 1 paper)	10-20 mg PAC/L. 0.3-1h contact time. Data from 2 papers.	58-64	Boehler et al. 2012; Margot et al. 2013
	GAC	Pilot	RMW	5.8 (DOC)	145 ng/L	7400 bed volumes treated. 14 min EBCT. Data from 1 paper.	59	Bourgin et al. 2018
	O_3	Pilot/full	RMW	3.5-8.6 (DOC)	-	0.61±0.04 gO_3/gDOC. Data from 3 papers.	94-97	**Table SI2**
	Solar photo-Fenton (CPC rector)	Pilot	RMW/SRMW	10.2-42.7 (DOC)	5.5 ng/L – 1879 µg/L	Fe: 5 – 10 mg/L; H2O2: 20 – 100 mg/L; pH: 2.8 or neutral (chelating agent used). Data from 5 papers.	56-100	**Table SI8**
	Solar photo-Fenton (Raceway pond)	Pilot	RMW	40 (DOC)	282 ± 36.7 ng/L	Continuous mode.Two liquid depths (5, 15 cm) and three HRTs (80, 40, 20 min); Fe: 5.5 mg/L; H_2O_2: 30 mg/L. pH 2.8. Data from 1 paper.	81-100	Arzate et al., 2017
	Photo Fenton	Pilot	RMW	5-7.5 (TOC)	355 ng/L	20-50 mg H_2O_2/L; 2-4 mg Fe/L. pH 6-7 (no chelating agents added). 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	3-82	De la Cruz et al., 2013
	UV-C/H_2O_2	Pilot	RMW	5-7.5 (TOC)	355 ng/L	20-50 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	38-99	De la Cruz et al., 2013

	sunlight/TiO_2 (CPC reactor)	Pilot	SRMW	13 (DOC)	100 µg/L	Data from 1 paper. TiO2 immobilized on glass spheres.	100	Miranda-García et al. 2011
Erythromycin	PAC	Pilot	RMW	5.6±0.9 (DOC)	50±38 ng/L	5-10 mg PAC/L; 0.4-0.8h contact time; data from 1 paper.	70	Mailler et al. 2015
	GAC	Pilot	RMW	4.2±0.1 (DOC)	300±200 ng/L	25000 bed volumes treated. Data from 1 paper.	>99	Knopp et al. 2016
	O_3	Pilot	RMW	8.6	-	0.64±0.01 gO_3/gDOC. Data from 1 paper.	>93	Kovalova et al., 2013
	Solar photo-Fenton (Raceway pond)	Pilot	RMW	40 (DOC)	119 ± 15.5 ng/L	Data from 1 paper. Continuous mode.	100	Arzate et al., 2017
	Sunlight/TiO_2 (CPC)	Pilot	RMW	23.2 (DOC)	41-78 ng/L	Data from 1 paper. 0.02 g TiO_2 powder/L.	>88	Prieto-Rodríguez et al 2013a
Clarithromycin	PAC	Pilot/full	RMW	5-10 (DOC)	54-440 ng/L	10-20 mg PAC/L. 0.3-1h contact time. Data from 3 papers.	88-95	**Table SI5**
	GAC	Pilot	RMW	4.4 (DOC)	155 ng/L	23400 bed volumes treated. 14 min EBCT. Data from 1 paper.	54	Bourgin et al. 2018
	O_3	Pilot/full	RMW	4.2-8.6 (DOC)	-	0.64 gO_3/gDOC. Data from 2 papers.	99-100	Hollender et al. 2009; Kovalova et al. 2013
	Solar photo Fenton (CPC reactor)	Pilot	RMW/SRMW	5-42 (DOC)	100 ng/L – 100 µg/L	Data from 3 papers. Fe: 5 – 10 mg/L; H_2O_2: 20 – 100 mg/L; pH: 2.8 or neutral	77-84	**Table SI8**
	Photo Fenton	Pilot	RMW	5-7.5 (TOC)	209-487 ng/L	20-50 mg H_2O_2/L; 2-4 mg Fe/L. pH 6-7 (no chelating agents added). 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	79-82	De la Cruz et al., 2013

	UV-C/H_2O_2	Pilot	RMW	5-7.5 (TOC)	209-487 ng/L	20-50 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	81-89	De la Cruz et al., 2013
	sunlight/TiO_2 (CPC reactor)	Pilot	RMW	15-50 (DOC)	24-54 ng/L	Data from 2 papers. 0.02-0.2 g TiO_2 powder/L.	30-88	Bernabeu et al. 2011; Prieto-Rodríguez et al 2013b
Diclofenac	PAC	Pilot	RMW	7.3(±1.9) (DOC)	1187 ng/L	10-20 mg PAC/L; 0.3-0.7h contact time; data from 1 paper.	69	Margot et al. 2013
	GAC	Pilot	RMW	4.4 (DOC)	1008 ng/L	23400 bed volumes treated. 14 min EBCT. Data from 1 paper.	72	Bourgin et al. 2018
	O_3	Pilot/full	RMW	3.5-8.6 (DOC)	-	0.61(±0.04) gO_3/gDOC. Data from 4 papers.	98-100	**Table SI2**
	Photo-Fenton	Pilot	RMW	5-7.5 (TOC)	925 ng/L	20-50 mg H_2O_2/L; 2-4 mg Fe/L. pH 6-7. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	93-100	De la Cruz et al., 2013
	Solar photo-Fenton (CPC rector)	Pilot	RMW/SRMW	10.2-36 (DOC)	1 – 5100 µg/L	Data from 4 papers. Fe: 5 – 10 mg/L; H_2O_2: 20 – 60 mg/L; pH: 2.8 or neutral (chelating agent used).	80 - 100	**Table SI8**
	UV-C/H_2O_2	Pilot	RMW	5-7.5 (TOC)	925 ng/L	20-50 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	99-100	De la Cruz et al., 2013
	sunlight/TiO_2 (CPC reactor)	Pilot	RMW/SRMW	13-23 (DOC)	414 ng/L-100 µg/L	Data from 4 papers. 20 mg/L TiO_2 and supported TiO_2, neutral pH.	85-100	**Table SI9**
Carbamazepine	PAC	Pilot/full	RMW	5-10 (DOC)	221-461 ng/L	10-20 mg PAC/L; 0.3-1h	90-92	**Table SI5**

						contact time; data from 3 papers.		
	GAC	Pilot	RMW	4.4 (DOC)	110 ng/L	23400 bed volumes treated. 14 min EBCT. Data from 1 paper.	72	Bourgin et al. 2018
	O_3	Pilot/full	RMW	3.5-7.6 (DOC)	-	0.61±0.04 gO_3/gDOC. Data from 3 papers.	97-100	**Table SI2**
	Solar photo-Fenton (CPC rector)	Pilot	RMW/SRMW	10-36 (DOC)	70 ng/L- 100 µg/L	Data from 4 papers. Fe: 5 mg/L; H_2O_2: 50 – 60 mg/L; pH: 2.8 or neutral (chelating agent used).	24 - 100	**Table SI8**
	Solar photo-Fenton (Raceway pond)	Pilot	RMW	40 (DOC)	422 ± 54.9 ng/L	Data from 1 paper. Two liquid depths (5, 15 cm) and three HRTs (80, 40, 20 min); Fe: 5.5 mg/L; H_2O_2: 30 mg/L. pH 2.8	86-96	Arzate et al., 2017
	Photo-Fenton	Pilot	RMW	5-7.5 (TOC)	333 ng/L	20-50 mg H_2O_2/L; 2-4 mg Fe/L. pH 6-7. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	66-94	De la Cruz et al., 2013
	UV-C/H_2O_2	Pilot	RMW	5-7.5 (TOC)	333 ng/L	20-50 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	82-99	De la Cruz et al., 2013
	sunlight/TiO_2 (CPC reactor)	Pilot	SRMW	13 (DOC)	100 µg/L	Data from 1 paper. TiO2 immobilized on glass spheres.	50-80	Miranda-García et al. 2011
	sunlight/TiO_2 (CPC reactor)	Pilot	RMW	15-50 (DOC)	56 ng/L	Data from 1 paper. 0.2 g TiO2 powder/L.	65-80	Bernabeu et al. 2011
Metoprolol	PAC	Pilot	RMW	5.8-7.3 (DOC)	653-1203 ng/L	10-20 mg PAC/L; 0.3-1h contact time; data from 2 papers.	95-100	Karelid et al. 2017; Margot et al. 2013
	GAC	Pilot	RMW	4.4 (DOC)	191 ng/L	23400 bed volumes treated. 14 min EBCT. Data from 1	85	Bourgin et al. 2018

					paper.		
O_3	Pilot/full	RMW	3.5-8.6 (DOC)	-	0.61±0.04 gO_3/gDOC. Data from 3 papers.	80-98	**Table SI2**
UV-C/H_2O_2	Pilot	RMW	5-7.5 (TOC)	255 ng/L	20-50 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	80-97	De la Cruz et al., 2013.
Photo-Fenton	Pilot	RMW	5-7.5 (TOC)	255 ng/L	20-50 mg H_2O_2/L; 2-4 mg Fe/L. pH 6-7. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2. Data from 1 paper.	68-90	De la Cruz et al., 2013.
sunlight/TiO_2 (CPC reactor)	Pilot	RMW/SRMW	20-23 (DOC)	21 ng/L, 200 µg/L	Data from 2 papers. 0.02-0.2 g TiO_2 powder/L.	85-100	Prieto-Rodríguez et al 2013a; Quiñones et al., 2015.

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[1] RMW= real municipal wastewater; SRMW= spiked real municipal wastewater; [2] when data refer to more than two papers the reader is addressed to the corresponding table in the supplementary material.

2210 **Table 4**. Advantages, drawbacks and recommendations for each advanced treatment

Advanced Treatment	Advantages	Drawbacks	Recommendations
UV/H_2O_2	• Moderate-good CEC removal at lab/pilot scale • Effective as disinfection process too	• Formation of oxidation transformation products • No full-scale evidences on CEC removal • Higher energy consumption compared to ozonation, specifically when high organic matter concentration acts as inner filter for UV radiation.	• Toxicity tests recommended
Photo-Fenton	• High CEC removal • Use of solar irradiation • Effective as disinfection process too	• Formation of oxidation transformation products • No full-scale evidences on CEC removal • At neutral pH 7 addition of chelating agents necessary. • Large space requirements for solar collectors	• Toxicity tests recommended
UV/TiO_2	• High CEC removal • Use of solar irradiation • Effective as disinfection process too	• Low kinetics • Formation of oxidation transformation products • Catalyst removal • Large space requirements for solar collectors	• Not possible to apply until more efficient photocatalysts (at least one order of magnitude) will be developed
Ozonation	• High CEC removal • Full scale evidence on practicability • Partial disinfection • Lower energy demand compared to UV/H2O2 and membranes	• Formation of by-products (NDMA, bromate) and other unknown oxidation transformation products • Need for a subsequent biological treatment (e.g., slow sand filtration) to remove organic by-products	• Toxicity tests recommended • NDMA and bromate should be monitored
Powdered activated carbon	• high CEC removal • full scale evidence on practicability	• PAC must be disposed • Post-treatment required (membrane, textile or sand filter) to prevent discharge of PAC	• Test with different products/process configurations

(PAC)	• additional DOC removal • no formation of by-products • Partial disinfection possible by the combination with membrane filtration (UF)	• production of PAC needs high energy • adsorption capacity may fluctuate with each batch	recommended
Granular activated carbon (GAC)	• high CEC removal • full scale evidence on practicability • additional DOC removal • no formation of by-products • An existing sand filtration can relative easily be replaced by GAC • GAC can be regenerated	• production of GAC needs high energy • Still under investigation if more activated carbon is needed compared to PAC • Less flexible in operation than PAC and ozonation to react to changes in wastewater composition • Adsorption capacity may fluctuate with each batch	• Test with different products recommended
NF and RO	• high CEC removal • RO can reduce salinity • effective disinfection • full rejection of particles and particle-bound substances	• High energy requirements • High investment and re-investment costs • Disposal of concentrated waste stream • Need for pre-treatment to remove solids	

Table 5. Estimations on the required electrical energy in different treatment methods to reduce CECs by around 80% in wastewater treatment.

Method	**Required amount**	**Energy**	**Ref.**
ozonation	5 mg/L ozone	0.33 kWh/m^3 (1)	Abegglen and Siegrist (2012)
UV / H_2O_2	10^{-1} cm path length	0.7-2.28 kWh/m^3	Katsoyiannis et al. (2011)
Nanofiltration & reverse osmosis	6-15 bar pressure**(2)**	0.6-0.9 kWh/m^3	Crittenden et al (2012)

(1) For on-site production of ozone from liquid oxygen (0.06 kWh/m^3 for O_3 from liquid O_2 and 0.27 kWh/m^3 primary energy for liquid O_2 production)

(2) Typical values that depend strongly on feedwater salinity; more energy and pressure is needed as feedwater salinity increases, e.g. around 65 bars and 3 kWh/m^3 for seawater desalination. Values for NF are slightly lower than for RO

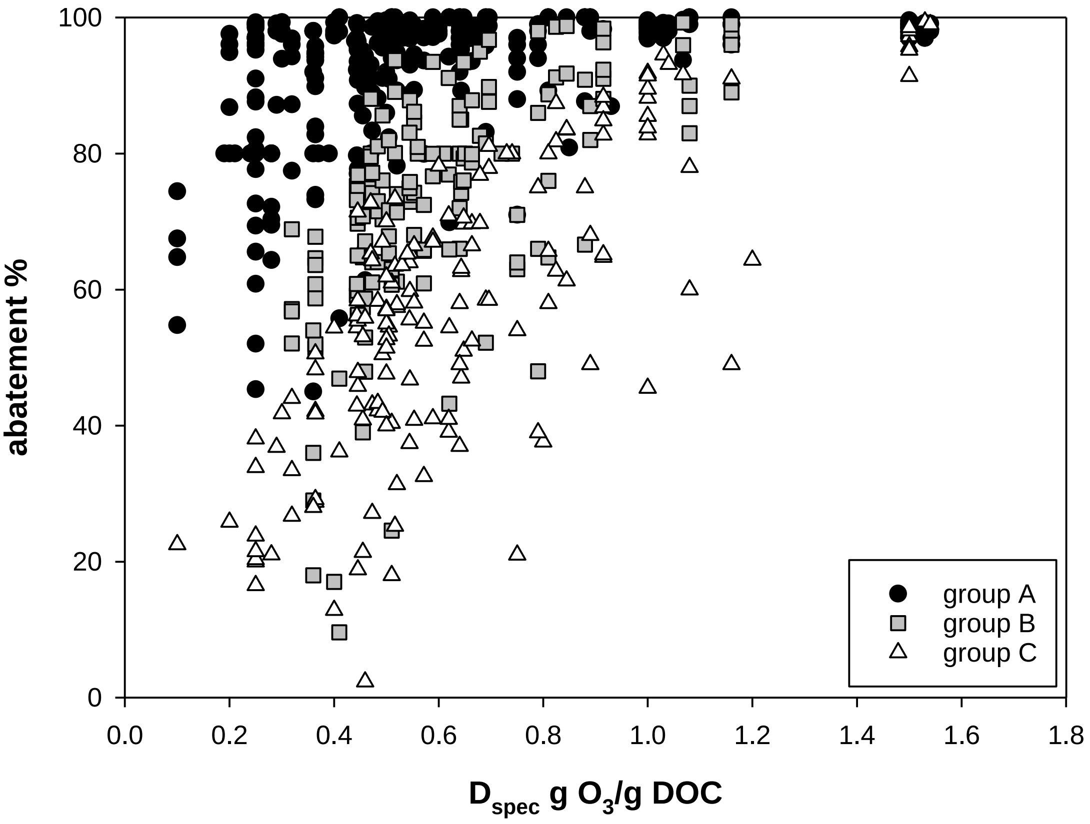

Figure 1. Abatement of 18 reviewed CEC as a function of the specific ozone dose. CEC are grouped according to their ozone reactivity as shown in Table 1.

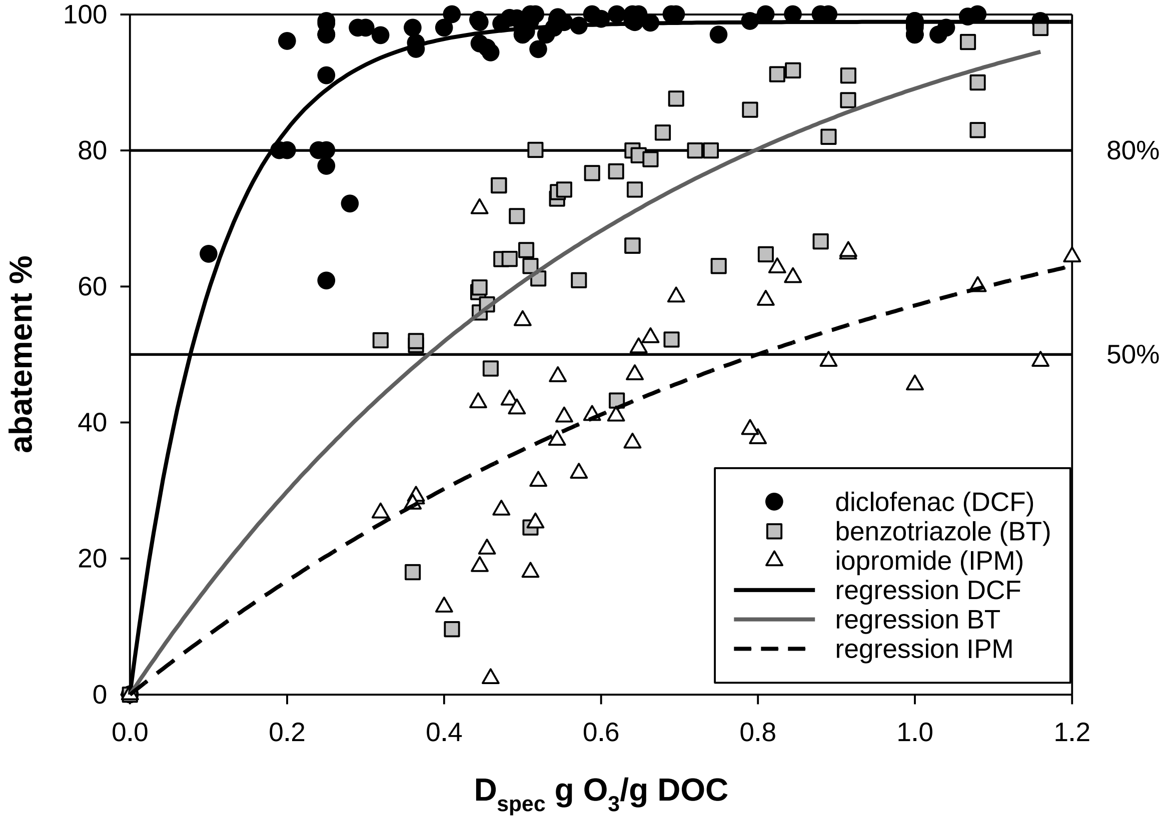

Figure 2. Abatement of representatives for group A (diclofenac), group B (benzotriazole) and group C (iopromide) including the group-specific boundaries for the average abatement at 0.4-0.6 g O_3/g DOC according to Table 1. Nonlinear regression fit with exponential rise to maximum (f = a*(1-exp(-b*x))).

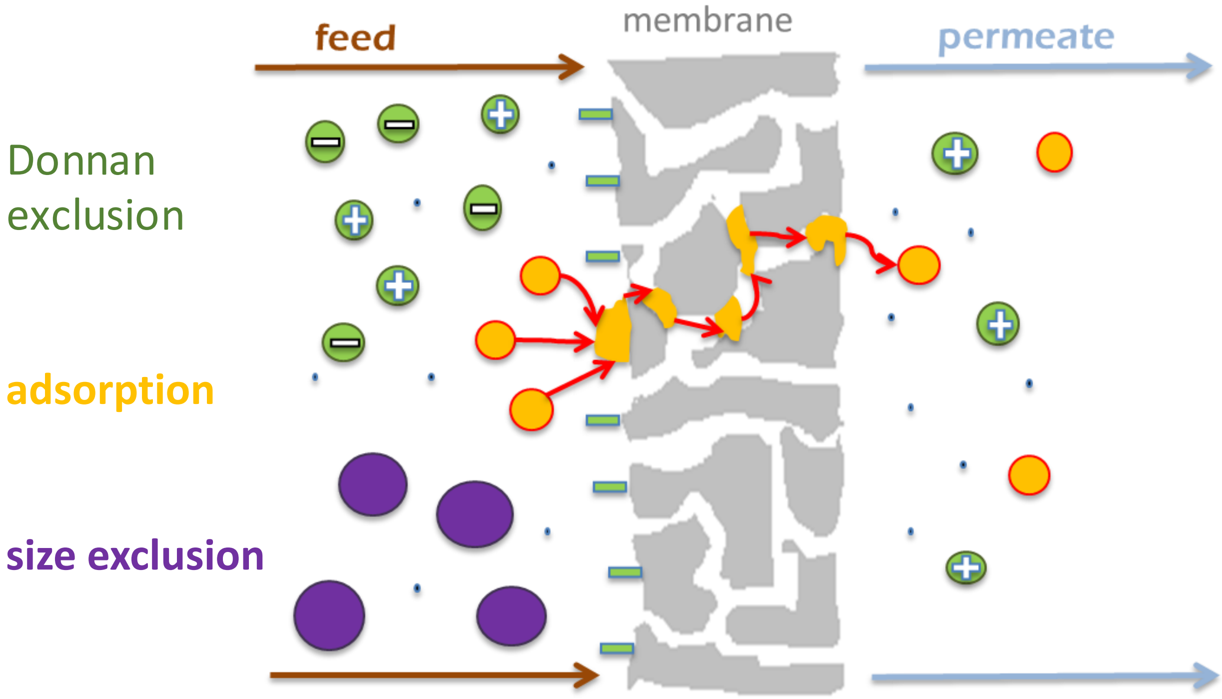

Figure 3. Visualisation of the three removal mechanisms by high pressure membranes. Adapted from Verliefde, 2008.

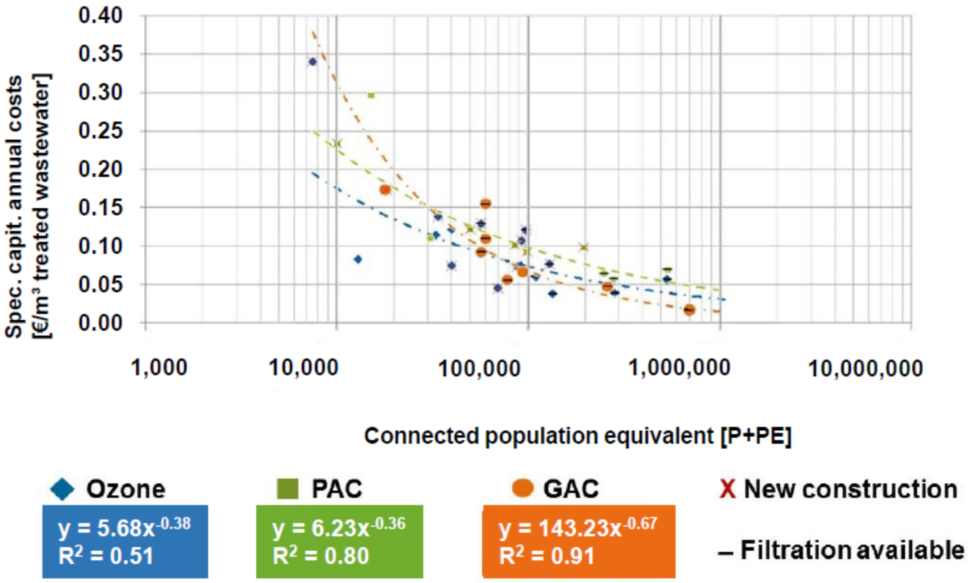

Figure 4. Specific capital annual costs of CECs abatement (selected process options only) (Antakyali 2017)