Urban wastewater treatment plants as hotspots for the release of antibiotics in the environment: A review

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Abstract

Urban wastewater treatment plants (UWTPs) are among the main sources of antibiotics' release into various compartments of the environment worldwide. The aim of the present paper is to critically review the fate and removal of various antibiotics in wastewater treatment, focusing on different processes (*i.e.* biological processes, advanced treatment technologies and disinfection) in view of the current concerns related to the induction of toxic effects in aquatic and terrestrial organisms, and the occurrence of antibiotics that may promote the selection of antibiotic resistance genes and bacteria, as reported in the literature. Where available, estimations of the removal of antibiotics are provided along with the main treatment steps. The removal efficiency during wastewater treatment processes varies and is mainly dependent on a combination of antibiotics' physicochemical properties and the operating conditions of the treatment systems. As a result, the application of alternative techniques including membrane processes, activated carbon adsorption, advanced oxidation processes (AOPs), and combinations of them, which may lead to higher removals, may be necessary before the final disposal of the effluents or their reuse for irrigation or groundwater recharge.

Keywords: advanced wastewater treatment; activated sludge; antibiotics; disinfection; pharmaceuticals; wastewater.

48 **CONTENT TABLE**

49

1. Introduction

During the last years, it is recognized that antibiotics constitute a new class of water contaminants of emerging concern with adverse effects on the aquatic life (Kolpin et al., 2002; Kümmerer, 2009; Fatta-Kassinos et al., 2011a). The generic term "antibiotic" is used herein to denote any class of organic molecule that inhibits or kills microbes by specific interactions with bacterial targets, without any consideration of the source of the particular compound or class (Davies and Davies, 2010). Investigations for the occurrence of various antibiotics in wastewater effluents have been conducted in several European countries (Miao et al., 2004; Batt et al., 2007; Gulkowska et al., 2008; Jones et al., 2001; Heberer, 2002; Kümmerer, 2009; Fatta-Kassinos et al., 2011a). Because of the intensive use of antibiotics for human (domestic and hospital use), veterinary and agriculture purposes, these compounds are continuously released into the environment from anthropogenic sources, such as urban wastewater treatment plants (UWTPs), which are considered as one of the main 'hotspots' of potential evolution and spreading of antibiotic resistance into the environment (Hirsch et al., 1999; Díaz-Cruz et al., 2003; Brown et al., 2006; Kümmerer 2009; Le Corre et al., 2012; Czekalski 2012). The presence of antibiotics in environmentally relevant concentration levels has been associated to chronic toxicity and the prevalence of resistance to antibiotics in bacterial species (Schwartz et al., 2006; Kümmerer, 2009).

The number of studies focusing on the chronic toxicological assessment of antibiotics in the environment is constantly increasing with the aim to bridge the various knowledge gaps (*i.e.* relevant endpoints to be considered in chronic bioassays) associated with these issues. Boxall et al. (2004) and Kümmerer (2009) represent two comprehensive review articles regarding the ecotoxicity of antibiotics. Thomulka and McGee (1993) determined for example the toxicity of a number of antibiotics (*e.g.* novobiocin, tetracycline, chloramphenicol, nalidixic acid, ampicillin, streptomycin) on *Vibrio harveyi* in two bioassay methods. Almost no toxic effects were found after short incubation times when luminescence was used as an endpoint. However, in a long-term assay

using reproduction as the endpoint, a toxic effect in environmentally relevant concentrations was detected for almost all the examined antibiotics. These results are in accordance with the observations of Froehner et al. (2000) concerning chloramphenicol, nalidixic acid and streptomycin. The chronic toxicity of several groups of antibiotics towards *Vibrio fischeri* is also presented in a study by Backhaus and Grimme (1999). The chronic bioluminescence inhibition assay was shown to be sensitive against many of the high volume antibiotics used for veterinary purposes and in aquaculture. Furthermore, exposure to antibiotics may have adverse effects on the reproductive system in the early life stages of different organisms like the freshwater flea *Daphnia magna* and the crustacean *Artemia salina* (Macrí et al., 1988; Wollenberger et al., 2000). In the study by Kim et al. (2007), sulfonamides (*i.e.* sulfamethoxazole, sulfachloropyridazine, sulfathiazole, sulfamethazine, sulfadimethoxine), and trimethoprim, were examined for their acute aquatic toxicity by employing a marine bacterium (*Vibrio fischeri*), a freshwater flea (*Daphnia magna*) and the Japanese medaka fish (*Oryzias latipes*). In this study, *Daphnia magna* was in general the most susceptible in terms of effective/lethal concentrations-E/LC_{50}, among the test organisms.

Moreover, the extensive use of antibiotics has contributed to the development of antibiotic resistance genes and bacteria, reducing the therapeutic potential against human and animal pathogens (Kemper, 2008). The consequences are particularly worrying as bacteria in the aquatic environment can be continually exposed to antibiotic residues (Rosal et al., 2010). The biological treatment process creates an environment potentially suitable for resistance development and spreading, because bacteria are continuously exposed to environmentally relevant levels of antibiotics. However, it remains unclear where most of the resistant bacteria have been selected, and in particular if the low antibiotic concentrations that are present in natural environments or in human/animal body compartments during therapeutic use, are important for the selection and enrichment of resistant mutants (Gullberg et al., 2012). The extent to which human activities contribute to the development of resistant bacterial strains is still poorly understood (Auerbach et al,

2007). The number of studies, focusing exclusively on wastewater treatment systems regarding the removal of antibiotic resistance, is still however limited.

Gao et al. (2012) investigated the relationship between concentrations of tetracyclines and sulfonamides and the number of antibiotic resistance genes and antibiotic resistant bacteria in a conventional UWTP located in Michigan. Significant reductions (2-3 logs) of antibiotic resistance genes and antibiotic resistant bacteria were observed between raw influent and final effluent whereas no apparent decrease was observed in the concentrations of tetracycline resistance genes (*tetO* and *tetW*) and sulfonamide resistance gene (*sulI*) by chlorine disinfection. Moreover, Dodd (2012) provide a comprehensive overview on the significance of antibiotic resistant genes and bacteria occurrence in environmental systems, and a discussion on the role that commonly used water and wastewater disinfection processes may play in minimizing ARG transport and dissemination.

Zhang et al. (2009) reported the impact of the wastewater treatment process on the prevalence of antibiotic resistance in *Acinetobacter spp.* in the wastewater and the possible spread of antibiotic resistance to receiving water bodies. It was found that the prevalence of antibiotic resistance was significantly higher in the downstream samples than in the upstream samples, with the higher values occurred for trimethoprim (97%), followed by rifampin (74%). Other studies have reported that the prevalence of resistant bacteria in sewage may significantly vary, depending on the plant (initial quality characteristics of sewage, type of treatment, plant operation, etc.), the target bacterial population, and the antimicrobial agent under study, as well as on the methods and the breakpoint values used to determine antimicrobial resistance (Guardabassi et al., 2002).

Another issue related to the use of reclaimed wastewater for irrigation is the plant uptake of antibiotics. The accumulation may or may not affect the growth and development of plants; however, the uptake into plants may represent an important exposure pathway of these compounds to humans and other biota (European Medicines Agency-EMEA). Migliore et al. (2003) determined

the phytotoxicity of enrofloxacin on crop plants *Cucumis sativus*, *Lactuca sativa*, *Phaseolus vulgaris* and *Raphanus sativus* in a laboratory model. Enrofloxacin at concentrations between 50 and 5000 μg L^{-1} induced both toxic effect and hormesis in plants, by significantly modifying both length of primary root, hypocotyl, cotyledons and the number/length of leaves. There are also new concerns that antibiotics decrease the biodegradation of leaf and other plant materials, which serves as the primary food source for aquatic life in rivers and streams (Richardson and Ternes, 2011).

The aim of the present paper is to introduce a critical review on the removal efficiency of various antibiotics in wastewater treatment during the application of different processes, namely biological processes, advanced treatment technologies and disinfection. An effort to include as many studies as possible was made in order to highlight important findings and present the knowledge currently available on the removal efficiency of antibiotics from wastewater through a variety of treatment processes.

2. Fate of antibiotics in UWTPs

The conventional wastewater treatment generally consists of a primary, secondary and sometimes a tertiary stage, with different biological and physicochemical processes available for each stage of the treatment. Primary treatment intends to reduce the solid content of the wastewater (oils and fats, grease, sand, grit and settleable solids). This step is performed entirely mechanically by means of filtration and sedimentation and is common at all UWTPs. However, the secondary treatment, which typically relies on a biological process to remove organic matter and/or nutrients with aerobic or anaerobic systems, can differ substantially. Several biological treatments are being used in modern municipal UWTPs, but the most common method is conventional activated sludge (CAS). Membrane bioreactors (MBR), moving bed biofilm reactor (MBBR), or fixed bed bioreactors (FBR) are less common. Activated sludge plants use dissolved oxygen to promote the growth of a biological floc that substantially removes the organic material and nitrogen at given conditions. In

150 the final step, tertiary wastewater treatment processes can be applied to remove phosphorus by
151 precipitation and particles on a filter (Batt et al., 2007). In some UWTPs the effluent is also
152 disinfected before it is released into the environment, typically by chlorination or ultraviolet
153 irradiation.

154 The effect of biological treatments, membrane filtration, activated carbon adsorption, advanced
155 oxidation processes (AOPs), and disinfection on different classes of antibiotics has been widely
156 investigated in the last years; several of these studies are presented in the subsequent paragraphs.
157

158 2.1 Effect of biological treatment on antibiotics' removal

159 Elimination and transformation of antibiotics during the biological treatment is the result of
160 different processes. These processes can be biotic (biodegradation, mainly by bacteria and fungi)
161 and non-biotic or abiotic (e.g. sorption, hydrolysis, photolysis).

162 The removal of antibiotics mainly depends on their sorption on the sewage sludge and their
163 degradation or transformation during the treatment. Hydrolysis can play a role for some
164 compounds, while photolysis is not very likely to occur due to the low exposure of the substances
165 to light during the wastewater treatment.

166 Hydrophobic (or non-polar) antibiotic residues are expected to occur at higher concentration in
167 primary and secondary sludge than hydrophilic ones because they have a greater affinity to solids
168 and hence, concentrate in the organic-rich sewage sludge (Le-Minh et al., 2010). Antibiotics can
169 also be removed from aqueous solutions onto solid particulates by ion exchange, complex
170 formation with metal ions and polar hydrophilic interactions (Diaz-Cruz et al., 2003). Antibiotics
171 that are sorbed to flocs, suspended solids and activated sludge, are removed from the aqueous phase
172 by sedimentation and subsequent disposal of excess sludge. The affinity of antibiotics sorbed to
173 sludge is most often represented by sludge sorption constants K_d (L kg^{-1}). The higher K_d values the
174 higher sorption of the compounds to sludge. A review on K_d values of several antibiotics are

175 provided in Kovalova et al. (2012). It is important to note that the sludge is often used as fertilizer
176 on agriculture fields, but in several European countries this is forbidden and the sludge is
177 incinerated. Using sludge as fertilizer can therefore be considered as another input pathway for
178 various antibiotics into the environment.

179 The tendency to accumulate in sludge solids can be assessed using the octanol-water partition
180 coefficient (K_{OW}). Rogers (1996) proposed the following guide to assess the sorption potential of
181 organic contaminants: $\log K_{OW} < 2.5$: low sorption potential (*e.g.* tetracyclines, sulfonamides,
182 aminoglycosides); $2.5 < \log K_{OW} < 4.0$ (*e.g.* β-lactams, macrolides): medium sorption potential and
183 $\log K_{OW} > 4.0$ (*e.g.* glycopeptides): high sorption potential. However, it should be emphasized that
184 the prediction of the antibiotics sorption onto solids or sludge is mainly possible for non-polar
185 compounds, while the prediction of the behaviour of polar or charged compounds is often not
186 correct. In some cases, the use of $\log K_{OW}$ values lead to an underestimation of the sorption of *e.g.*
187 fluoroquinolones (Golet et al., 2003) or tetracyclines (Kim et al., 2005) to sludge. For instance,
188 ciprofloxacin (fluoroquinolone) has a K_{OW} value of 1.8, but nevertheless sorbs onto sludge by 80%,
189 indicating that sorption is the main elimination process.

190 However, antibiotics are mostly hydrophilic and were designed to be biologically resistant; they are
191 therefore expected to mainly remain in the aqueous phase of the wastewater.

192 The main operational factors that can influence the biological removal of antibiotic residues in
193 wastewater treatment are biochemical oxygen demand (BOD_5), existence and size of anoxic and
194 anaerobic compartments, suspended solids (SS) loading, hydraulic retention time (HRT), sludge
195 retention time (SRT), food-microorganism ratio (F/M ratio), mixed liquor-suspended solids
196 (MLSS), pH and temperature (Drewes, 2008; Kovalova et al., 2012).

197 The SRT is related to the growth rate of microorganisms. High SRTs allow the enrichment of
198 slowly growing bacteria and therefore, provide greater diversity of enzymes, some of which are
199 capable of degrading the antibiotic compounds (Jones et al., 2007; Le-Minh et al., 2010). High SRT

200 can be reached with a membrane bioreactor (MBR), where the suspended activated sludge is
201 retained in the reactor by utilizing a membrane for solid/liquid separation instead of a settling tank
202 as used in CAS. Commonly, micro- or ultrafiltration membranes are used in MBRs, which do not
203 retain the antibiotics on the filter. Some studies have been performed to investigate if higher SRTs
204 enhance the elimination of antibiotics, which will be discussed in detail below (Joss et al., 2005;
205 Göbel et al., 2007; Radjenovic et al., 2009b; Tadkaew et al., 2011, Kovalova et al., 2012).

206 The performance (expressed as % removal) of some UWTPs applying biological treatment for
207 removing antibiotics as reported in the literature is summarized in Table 1. The removal is highly
208 variable for many substances (from nearly complete to very little). Frequently, however, operational
209 details are not provided in the studies available in the literature on the fate and transport of
210 antibiotic residues during wastewater treatment or have not been systematically investigated. This
211 poses a major challenge for the comparison and discussion of results. Moreover, differences in
212 reported efficiencies may, in some cases, be attributed to limitations of employed mass balance
213 techniques (Le-Minh et al., 2010). For example, short-term variations of pharmaceuticals loads in
214 influent can be significant (Göbel et al., 2005; Khan and Ongerth, 2005), thus consideration must be
215 taken when comparing influent and effluent concentrations.

216 Antibiotics can be grouped by either their chemical structure or mechanism of action. The main
217 groups of antibiotics and their potential removal during conventional wastewater treatment are
218 discussed in the following sections.

219

220 *2.1.1 β-Lactams*

221 β-lactams are not very stable due to hydrolysis of the beta-lactam ring (Hirsch et al., 1999; Längin
222 et al. 2009). β-lactams have been reported to be significantly reduced during biological treatment
223 with removals higher than 90% (Watkinson et al., 2007; Watkinson et al., 2009). According to Li et
224 al. (2009) the observed removals at an UWTP in Hong Kong were between 30.4-100%. β-lactams

were also eliminated significantly at both Shatin and Stanley UWTPs as described in the work of Li and Zhang (2011). Cha et al. (2006) investigated the fate of four β-lactams (ampicillin, cloxacillin, cephapirin, oxacillin) and the estimated removals were between 17-43%. Ampicillin was removed by 82% in an activated sludge process (Li and Zhang 2011). High removal of ampicillin (>94%) was also achieved in MBR treatment (SRT 3-60 days, Xia et al 2012). A significant removal (96%) of cephalexin from 2000 ng L^{-1} to 78.2 ng L^{-1} has been reported to occur through conventional UWTP processes in Australia (Costanzo et al., 2005). Analysis of amoxicillin conducted by Zuccato et al. (2010) in UWTPs in Italy and Switzerland showed that it is efficiently removed by CAS (100%). Similarly, Watkinson et al. (2009) showed that amoxicillin is quite susceptible to microbial degradation with removal higher than 99% and therefore it is not likely to remain in significant concentration after biological treatment systems. Cephalexin was removed by 53% at the Shatin UWTP, while it was removed by 91% at the Stanley UWTP (Li and Zhang, 2011). Cephalexin was also removed by 36-99.8% in four Taiwanese UWTPs combining biological treatment and disinfection process (UV or chlorination) (Lin et al., 2009) and by 99.6% in an Australian UWTP using CAS (Watkinson et al., 2009). Therefore, cephalexin is relatively easily eliminated in UWTPs with biological processes, whereas cefotaxime, which was only detected in Shatin UWTP, was removed by only 43% (Li and Zang, 2011).

2.1.2 Macrolides

Li and Zhang (2011) reported that roxithromycin was degraded by 40-46% during CAS. Slightly lower removal (33%) was reported for one German UWTP (Ternes et al., 2007). In the studies of Göbel et al. (2007) and Joss et al. (2005), roxithromycin was removed at two UWTPs in Switzerland by 38% during secondary treatment and by 38-57% during MBR treatment (SRT=16, 33, 60-80 days). Moreover, roxithromycin removal was reported to be higher than 53% for four UWTPs in south China (Xu et al., 2007). Clara et al. (2005) reported a removal range for roxithromycin of 52-100% during MBR treatment (SRT 10-55 days).

Erythromycin is frequently detected as its main human metabolite, the dehydrated product with an apparent loss of one molecule of water, erythromycin-H_2O. Erythromycin-H_2O was degraded by 15% and 26% in activated sludge processes at Shatin and Stanley UWTP, respectively (Li and Zhang, 2011), and up to 10% in two Swiss UWTPs (Göbel et al., 2007). Higher removals were reported in other studies, that is, 56% in four Taiwanese UWTPs (Lin et al., 2009) and 43.8-100% in an UWTP in USA (Karthikeyan and Meyer, 2006) by secondary wastewater treatment processes both employing activated sludge.

For clarithromycin highly variable elimination rates are reported, from ≤20% (Göbel et al. 2007, Spongberg et al. 2008) up to 80% (Dolar et al. 2012, Lin et al. 2009). For clarithromycin and erythromycin-H_2O an influence of sludge age was observed with enhanced eliminations at higher SRTs (26-40% at SRT= 33 days, 90% at SRT 60-80 days in Göbel et al., 2007). Reif et al. (2008) also found high removals of roxithromycin and erythromycin (77% and 91%, respectively) in an MBR with SRT of 44-72 days.

Macrolides may be sorbed to biomass via cation exchange processes due to the fact that under typical wastewater conditions (pH=7-8), many are positively charged through the protonation of the basic dimethylamino group (pK_a=7.1-9.2) while the surface of activated sludge is predominantly negatively charged (Le-Minh et al., 2010). Analysis of sludge, however, showed that sorption of macrolides is of minor importance for the elimination in conventional UWTPs with K_d of below 400 L Kg^{-1} (Göbel et al., 2005, Kovalova et al. 2012). Abegglen et al. (2009) observed a slightly higher affinity of MBR sludge to macrolides than conventional activated sludge (K_d =1400 L Kg^{-1} for azithromycin).

2.1.3 Sulfonamides

The concentrations of these antibiotics in UWTP influents and effluents vary significantly, depending on consumption patterns and the types of wastewater treatment processes employed. For example, sulfamethoxazole has been reported at concentrations as high as 5450-7910 ng L^{-1} in sewage influent in China and was completely removed during the treatment (Peng et al., 2006). In a Taiwanese UWTP, sulfamethoxazole was detected in influent at concentration range of 500-10000 ng L^{-1} and the removal was 65-96% after the biological treatment (Yu et al., 2009). Sulfamethoxazole has been reported to be removed up to 81% (initial concentration 1090 ng L^{-1}) (Yang et al., 2005), 69-75% (initial concentration in the range 13-155 ng L^{-1}) (Pailler et al., 2009), 68.2-95.7% (initial concentration in the range 146-355 ng L^{-1}) (Li et al., 2009) and 93% (initial concentration in the range 3000 ng L^{-1}) (Watkinson et al., 2009). However, in other studies lower removal rates of 20-24% were reported (Brown et al., 2006; Ternes et al., 2007).

At this point it is worth mentioning that, there is only little knowledge on the environmental fate of humans' metabolites of antibiotics, which are excreted from the human body, often in considerable amounts and can be found predominantly in the environment (Hollender et al., 2008). Humans' metabolites are often omitted when analyzing antibiotics; a notable exception is the sulfamethoxazole's acetylated metabolite. N_4-acetylsulfamethoxazole usually accounts for more than 50% of an administered dose in human excretion and can occur in UWTP influents at concentrations of 2.5-3.5 times higher than concentrations of the parent compound (Göbel et al., 2007). Significant removal efficiencies (81-96% and 68-92%, respectively) of N_4-acetylsulfamethoxazole during secondary treatment were reported by Göbel et al. (2007) and Joss et al. (2005). N_4-acetylsulfamethoxazole can also de-conjugate into sulfamethoxazole during wastewater treatment (Göbel et al., 2007), leading to an underestimation of removal efficiency for sulfamethoxazole if this metabolite is not considered. This might be a reason for the highly varying observed elimination rates.

299 Higher removal rates were observed for sulfadiazine during activated sludge process at Shatin
300 (72.8%, 100%) and Stanley (87%) UWTPs (Li et al., 2009; Lin and Zhang, 2011). However, the
301 removal rate for sulfadiazine was only 50% in a Chinese UWTP (Xu et al., 2007).

302 Sulfamethazine was removed to concentrations below detection in the study of Lin and Zhang
303 (2011), Karthikeyan and Meyer (2006), Choi et al. (2007) and Yang et al. (2005), achieving
304 removal rates higher than 80%. Yu et al. (2009) reported a removal of 32-85% in a UWTP in
305 Colorado. Many other sulfonamides were eliminated during conventional processes with removal
306 efficiencies varying from <0 to 100%, but sorption to sludge was found to be negligible for
307 sulfonamides (Yang et al., 2005; Choi et al., 2007; Göbel et al., 2007; Watkinson et al., 2007;
308 Spongberg et al., 2008; Abegglen et al., 2009; Pailler et al., 2009; Watkinson et al., 2009; Tambosi
309 et al., 2010).

310 The variation of sulfonamides removal may possibly be explained by the deconjugation of
311 metabolites, but also by the differences in UWTP operating conditions such as HRT and the
312 presence of an anaerobic compartment. Higher SRT, though, was not found to increase the
313 elimination of sulfamethoxazole and sulfapyridine (Göbel et al., 2007, Radjenovic 2009b).

314

315 *2.1.4 Trimethoprim*

316 The presence of trimethoprim can generally be correlated to that of sulfamethoxazole since the two
317 drugs are often administered in combination (Göbel et al., 2005). The removal of trimethoprim has
318 been reported as 13% and 42% by Lin and Zhang (2011). The removal of this compound was found
319 to fluctuate within the same levels in various UWTPs in USA (50-100%), in Germany (69%) and in
320 Taiwan (74%) (Brown et al., 2006; Karthikeyan and Meyer, 2006; Ternes et al., 2007; Yu et al.,
321 2009). Higher removals were obtained in five UWTPs in Australia yielding 94% (Watkinson et al.,
322 2009) and 93.3% (Li et al., 2009). In contrast, the removal of trimethoprim was negligible as
323 reported in the studies of Lindberg et al. (2005) and Roberts and Thomas. (2006).

Some studies have indicated that nitrifying microorganisms appear to be capable of degrading trimethoprim. This suggests an important role for aerobic conditions for the biotransformation of trimethoprim (Perez et al., 2005; Batt et al., 2006). Moreover, trimethoprim elimination was found to be increased at higher SRTs (Göbel et al., 2007; Radjenovic 2009b; Tambosi 2010; Kovalova et al., 2012).

2.1.5 Quinolones

Removal efficiencies of quinolones during wastewater treatment in Sweden were reported to be 87% for norfloxacin and ciprofloxacin and 86% for ofloxacin (Lindberg et al., 2005). A later study reported the removal of ciprofloxacin (>90%), ofloxacin (56%), and norfloxacin (>70%) during activated sludge treatment followed by chemical coagulation/flocculation (Zorita et al., 2009). Sorption to sewage sludge has been suggested by Golet et al. (2003) as the primary removal mechanism for fluoroquinolones (ciprofloxacin and norfloxacin) during secondary wastewater treatment, resulting in the removal of 78-84% of the aforementioned fluoroquinolones from the aqueous phase. High removals of ofloxacin were achieved in UWTPs in Cyprus (>83%) (Fatta et al., 2010) and in China (100%) (Peng et al., 2006). Removal of ciprofloxacin in an MBR treating hospital wastewater (SRT=30-50 days) was only 51% (Kovalova et al. 2012). This relatively low removal might have been caused by the lower sludge production in MBR than in conventional activated sludge, leading to lower sorption.

2.1.6 Tetracyclines

Tetracycline is one of the most frequently detected antibiotics in wastewater (Watkinson et al., 2007). According to the study of Yang et al. (2005) tetracycline was removed by 85% in an UWTP in Colorado. Lin and Zhang (2011) reported removals of 24-36% at two plants while higher removals (67.9-100%) were reported by Karthikeyan and Meyer (2006) and four Taiwanese UWTPs (66-90%) by Lin et al. (2009)

The removal rates for chlortetracycline as reported by Lin and Zhang (2011) were in the range of 82% and 85%. Furthermore, for chlortetracycline and doxycycline, after secondary treatment and chlorination, the removal efficiencies were reported to be 78% and 67%, respectively (Yang et al., 2005). Choi et al. (2007) reported even higher removal values for minocycline and democlocycline (92 and 89%, respectively). High removal was also achieved for tetracyclines in MBR treatment (SRT=3-60 days, Xia et al 2012).

Tetracyclines have complexing properties and can easily bind to calcium and similar ions, thus forming stable complexes, which can bind to suspended matter or sewage sludge (Drewee, 2008). Kim et al. (2005) found no evidence of tetracycline biodegradation during the biodegradability test, but sorption was found to be the principal removal mechanism in activated sludge. These properties might explain why tetracyclines are detected in many cases in low concentration levels (ng L^{-1}) in treated secondary effluents.

2.1.7 Other antibiotic groups

Several studies reported the occurrence of lincosamides antibiotics such as lincomycin and clindamycin in wastewater influents and effluents with maximum removal efficiencies of 67% (Zuccato et al., 2010; Kovalova et al., 2012). Clindamycin may be transformed back from the main human metabolite clindamycin sulfoxide in the denitrification process, resulting in increased concentration (Kovalova et al., 2012). A study by Watkinson et al. (2009) showed that removals of polyether ionophores (monensin and salinomycin) in wastewater were up to 95%. Metronidazole, an imidazole antibiotic, was removed up to 23% during CAS (Kasprzyk-Hordern et al., 2009; Jelic et al., 2011) and 45% in an MBR treating hospital wastewater (SRT=30-50 days, Kovalova et al., 2012). Metronidazole is rapidly transformed into 1-(2-hydroxyethyl)-2-hydroxymethyl-5-nitroimidazole (Mahugo-Santana et al., 2010). Limited information on the behaviour of polyether ionophores through UWTP processes is available, due to the less likely occurrence of these

antibiotics in urban wastewater except where there is runoff from agricultural lands into sewers. Glycopeptides such as vancomycin was analysed by Lin and Zhang (2011) and the removal after the activated sludge process was found to be as high as 52%. The aminoglycoside gentamicin was found in hospital wastewater, although is a compound that is adsorbed very strongly (Löffler and Ternes, 2003).

In summary, biological treatment cannot completely remove antibiotics in wastewater treatment. Accordingly, alternative treatment processes are considered as necessary in order to provide further elimination of these compounds from wastewater effluents and to better manage environmental and human exposure to these contaminants.

In the following sections, other techniques including membrane filtration, activated carbon adsorption and advanced oxidation processes (AOPs) are discussed. The removal of antibiotics by these processes is depicted in Table 2 along with other relevant and important information. The upgrading of UWTPs and the application of such technologies is regarded as a possible optimization of the biological treatment with regard to antibiotics' removal.

2.2 Membrane processes

Removal of antibiotics in membrane processes can occur through multiple mechanisms. First, removal can be governed by adsorption where antibiotics that are hydrophobic or have strong hydrogen-bonding characteristics, readily adsorb to membranes at the initial stages of filtration. In many cases though, removal can occur through steady-state rejection due to either steric effects for uncharged solutes or combined steric and electrostatic effects for charged solutes. These mechanisms are dependent on the physicochemical properties of the compound (molecular weight cutoff (MWCO), pK_a, hydrophobicity/hydrophilicity), the solution (pH, ionic strength), and the membrane characteristics (material, surface morphology, pore size) (Le-Minh et al., 2010).

While the pores in micro- and ultrafiltration are too large to reject micropollutants, the lower membrane pore size used in nanofiltration (NF, pore size range: 0.001 μm) and reverse osmosis (RO, pore size range < 0.001 μm) have been shown in recent years to effectively remove low-molecular-weight pharmaceutical compounds, including antibiotics, during wastewater treatment. Various studies showed up to 90% removal of several antibiotics including quinolones, sulfonamides, tetracyclines and trimethoprim (Kimura et al., 2004; Morse and Jackson, 2004). A study undertaken by Kosutic et al. (2007) on the treatment of model wastewater of a manufacturing plant producing pharmaceuticals for veterinary use showed that sulfonamides were effectively removed by NF and RO. Zhang et al. (2006) reported a high removal efficiency (98.5-99.7%) for amoxicillin from wastewater, which contains high level of TOC using RO. In a study of Li et al. (2004) oxytetracycline at very high concentration (1000 mg L^{-1}) in wastewater from pharmaceutical manufacturing was reduced to 80 mg L^{-1} (<92% removal).

Given the complementary treatment capacity of MBR and NF/RO membrane filtration, there is significant scope for the coupling of these two treatment processes to achieve an overall enhanced performance (Alturki et al., 2010; Dolar et al., 2012). Excellent overall removal of target antibiotics with removal rates above 99% were achieved with MBR/RO (Dolar et al. 2012).

Some investigations reveal that the fouling of membranes can also lead to improved rejection of many solutes (Schafer et al., 1994; Drewes et al., 2006; Xu et al., 2006). This interesting observation is believed to be due to increased negative surface charge leading to increased electrostatic rejection of ionic species; along with simultaneously increased adsorptive capacity for non-ionic solutes (Xu et al., 2006).

2.3 Activated carbon adsorption treatment

Adsorptive treatment with activated carbon can be used for removing many hydrophobic and also some charged pharmaceuticals from water (Le-Minh et al., 2010). The adsorption mainly involves

424 the following steps: (i) solute transport in the bulk-adsorbate movement by the stagnant liquid film
425 surrounding the adsorbent, (ii) film diffusion-adsorbate transport along the film, (iii) pores
426 diffusion-adsorbate diffusion through the porous structure to the active sites (molecular diffusion in
427 the pore and/or in the adsorbent surface), (iv) adsorption-interaction between adsorbate and porous
428 structure (Homem and Santos, 2011).

429 The removal effectiveness of the activated carbon adsorptive treatment system depends on the
430 properties of the adsorbent (*e.g.* specific surface area, porosity, surface polarity and physical shape
431 of the material), and the characteristics of the compound (*e.g.* shape, size, charge and
432 hydrophobicity). Moreover, the sorption efficiencies of antibiotics to activated carbon may be
433 significantly altered by the initial concentrations of the target compounds, the pH, the temperature
434 and the presence of other species in the solution (Aksu and Tunc, 2005). Non-specific dispersive
435 interactions (*e.g.* van der Waals interactions) are the dominant mechanism of removal for organic
436 compounds, including antibiotics, in activated carbon adsorption systems, removing most non-polar
437 antibiotics with $\log K_{OW} > 2$. However, electrostatic interactions between ionic antibiotics and the
438 charged groups on the surface of activated carbon can result in removal of polar antibiotics (Snyder
439 et al., 2003). The removal of antibiotics by activated carbon has been reported during wastewater
440 treatment in some studies (Adams et al., 2002; Westerhoff et al., 2005; Putra et al., 2009; Rivera-
441 Utrilla et al., 2009; McArdell et al. 2011; Boehler et al. 2012). A post-treatment with powdered
442 activated carbon (PAC) after biological treatment has been mostly investigated. The concentrations
443 of several antibiotics in wastewater with PAC dosages between 10 mg L^{-1} and 20 mg L^{-1} have been
444 reduced by 49-99% after 4 h contact time (Adams et al., 2002; Westerhoff et al., 2005). In a study
445 on hospital wastewater treatment, macrolides, fluoroquinolones, trimethoprim and clindamycin
446 were removed well at PAC dosages of 20-40 mg L^{-1}, while sulfamethoxazole and metronidazole
447 showed poor removals (McArdell et al., 2011). PAC can also be added directly into the biological
448 reactor, where higher concentrations of carbon are required. Serrano et al. (2011) added 1 g L^{-1} into

449 a sequential membrane bioreactor and found elimination of 42-64% for erythromycin, 71-97% for
450 roxitromycin whereas no significant removal was obtained for TMP. Putra et al. (2009) compared
451 the adsorption capacity of activated carbon and bentonite and reported that 94.67% of amoxicillin
452 was removed from wastewater using activated carbon at a dose as high as 30 g L^{-1}.

453 It should be noted that in the case of the application of the activated carbon adsorption process in
454 wastewater effluents, the natural dissolved organic matter (DOM) in wastewater matrix competes
455 for adsorption sites and decreases the activated carbon capacity for antibiotics and other
456 micropollutants (Snyder et al., 2003).

457

458 2.4 Advanced Oxidation Processes (AOPs)

459 Advanced Oxidation Processes (AOPs) are quite efficient novel methods for water and wastewater
460 treatment (Legrini et al., 1993; Klavarioti et al., 2009; Malato et al., 2009). These processes involve
461 the use and generation of powerful transitory species, principally the hydroxyl radical ($HO^{\bullet}$)
462 (Goslich et al., 1997; Andreozzi et al., 1999). $HO^{\bullet}$ are powerful oxidizing agents leading to
463 oxidation and mineralization of organic matter (Litter, 2005), while this species is characterized by
464 lack of selectivity of attack. This property is of great importance in wastewater treatment because
465 radicals attack the oxidisable part of organic molecules with rates usually in the order of 10^6-10^9 M^{-1}
466 s^{-1} (Andreozzi et al., 1999). Several studies have reported the effective AOPs treatment for
467 removal of antibiotics in wastewater effluents (Adams et al., 2002; Arslan-Alaton et al., 2004;
468 Saritha et al., 2007; Naddeo et al., 2009; Elmolla and Chaudhuri, 2011). It is worth noting the fact
469 that most studies do not include information on the by-products formed during the application of
470 oxidation or any information related to the antibiotic activity of the by-products. Therefore, AOPs
471 should be carefully monitored and ecotoxicological investigations should be accompanied to
472 investigate the formation of potentially toxic transformation products (Hollender et al., 2009; Rizzo,
473 2011). The effectiveness of oxidative processes for degrading antibiotics will be largely determined

by the specific water matrix. However, the effects of water matrix quality on antibiotics removal are much less well understood than for other technologies. For example, the presence of natural dissolved organic matter (DOM) can result in the formation of oxidation by-products that may cause water quality to deteriorate beyond its initial state of contamination. Similarly, the presence of nitrates, carbonates and DOM, can interfere with the destruction of the target antibiotic(s) and ultimately reduce the effectiveness of the selected AOP.

The versatility of the AOPs is enhanced by the fact there are different ways of producing hydroxyl radicals, facilitating compliance with the specific treatment requirements. The most common AOPs that have been used and evaluated (mainly at a bench scale but many of the processes are being developed at a pilot scale as well) are: photolysis under ultraviolet (UV) irradiation; combinations of hydrogen peroxide (H_2O_2), ozone (O_3) and UV irradiation; homogeneous photocatalysis with Fenton reagent, heterogeneous photocatalysis with semiconductor materials (*e.g.* TiO_2) and sonolysis under ultrasound irradiation.

2.4.1 Ozonation

Ozone is a powerful oxidant and has been increasingly used for the treatment of wastewater whereas it has been traditionally employed in drinking water treatment (Litter, 2005). Huber et al. (2005) and Hollender et al. (2009) observed that using ozone at a dose of 2 mg L^{-1} (0.3-0.4 g g^{-1} DOC) more than 80% of sulfonamides, trimethoprim and macrolides were removed in the effluent of secondary wastewater treatment. Similar results between different wastewater treatment plants are achieved if the dose of ozone per amount of dissolved organic carbon (DOC) is compared. The study by Adams et al. (2002) showed that ozonation removed more than 95% of several sulfonamides and trimethoprim from river water within 1.3 min contact time at ozone dose of 7.1 mg L^{-1}. Clindamycin was already removed by 95% with an ozone dose of 2 mg L^{-1} (0.40 g g^{-1} DOC) (Hollender et al., 2009) and tetracycline by 100% with an ozone dose of 1.5 mg L^{-1} (Huber et

al., 2005). Balcioglu and Otker (2003) found that up to 80% of β-lactams removal from wastewater was observed during ozonation treatment after 60 min and ozone dose 2.96 g L^{-1} h^{-1}. In a study of Arslan-Alaton et al. (2004) the COD of an antibiotic formulation effluent containing penicillin (COD=830 mg L^{-1}) was removed by 10-56% during ozonation process while the addition of small amounts of hydrogen peroxide increased the removal efficiency (83%). In another study of Arslan-Alaton and Dogruel (2004) the COD and TOC of the formulation effluent containing penicillin was removed by 49% and 52% respectively under alkaline conditions (pH=11), whereas the removal efficiency was much lower under acidic conditions (pH=3) (COD removal max=15%; TOC removal max=2%). Many authors (Balcioglu and Ötker, 2003; Arslan-Alaton et al., 2004; Andreozzi et al., 2005) suggested that pH is a critical parameter in the ozonation process and a decrease of pH usually affects the reaction rate and also the absorption rates of ozone. During wastewater ozonation, many antibiotics, including β-lactams, sulfonamides, macrolides, quinolones, trimethoprim and tetracyclines, have been shown to be transformed predominantly via direct oxidation by O_3 whereas penicillin G, cephalexin and N_4-acetylsulfamethoxazole were transformed to a large extent by hydroxyl radicals (Dodd et al., 2006).

Ozone and/or hydroxyl radicals deactivate bactericidal properties of antibiotics by attacking or modulating their pharmaceutically active functional groups, such as N-etheroxime and dimethylamino groups of macrolides (Lange et al., 2006; Dodd et al., 2009), aniline moieties of sulfonamides (Huber et al., 2005), thioether groups of penicillins, unsaturated bonds of cephalosporin and the phenol ring of trimethoprim (Dodd et al., 2009). The high removals (>90%) by ozonation were achieved for those compounds with electron-rich aromatic systems, such as hydroxyl, amino (*e.g.* sulfamethoxazoles), acylamino, alkoxy and alkyl aromatic compounds, as well as those compounds with deprotonated amine (*e.g.* erythromycin, ofloxacin and trimethoprim) and non-aromatic alkene groups since these key structural moieties are highly amendable to oxidative attack (Dickenson et al., 2009).

524 Research conducted so far demonstrates that ozonation is a promising approach to degrade
525 antibiotics. According to Table 2, ozonation was found to be an effective process for removing β-
526 lactams, macrolides, sulfonamides and trimethoprim, quinolones, tetracyclines and lincosamides
527 The energy consumption for upgrading a Swiss municipal wastewater treatment plant with
528 ozonation was evaluated by Hollender et al. 2009. For an ozone dose of 0.6 g O_3 g^{-1} DOC (effluent
529 DOC ~5 g m^{-3}), 0.035 kWh m^{-3} wastewater was consumed, which is 12% of the total energy
530 consumption of a typical nutrient removal plant (0.3 kWh m^{-3} wastewater). Additionally, 0.01-0.015
531 kWh m^{-3} was needed for pure oxygen production. Ozone treatment performance may be enhanced if
532 ozone is combined with UV irradiation, hydrogen peroxide or catalysts (usually iron or copper
533 complexes) (Klavarioti et al., 2009). However, optimal process and operating conditions have yet to
534 be determined for the various water and wastewater types as well as for the different types of
535 antibiotics (Yargeau and Leclair, 2008).
536
537 *2.4.2 Fenton oxidation*
538 Fenton's oxidation is a homogeneous oxidation process and is considered to be a metal-catalyzed
539 oxidation reaction, in which iron acts as the catalyst (Tekin et al., 2006; Saritha et al., 2007). The
540 main disadvantage of the process is the low pH value required in order to avoid iron precipitation
541 that takes place at higher pH (Melero et al., 2007; Santos et al., 2007).
542 Trovó et al. (2008) observed that amoxicillin degradation was not influenced by the source of the
543 irradiation during the photo-Fenton process and the removals of the antibiotic obtained were 89 and
544 85% under black light and solar irradiation, respectively. A similar study by Bautitz and Nogueira
545 (2007) showed that tetracycline was removed by 80% during the photo-Fenton treatment using two
546 types of iron and irradiation. Moreover, in a study by Arslan-Alaton and Dogruel (2004) adequate
547 COD and TOC removal rates were achieved during the photo-Fenton and photo-Fenton-like
548 treatment of a formulation effluent containing penicillin. Trimethoprim was completely removed

549 during solar-Fenton process in the study of Michael et al. (2012a) and it was found that the
550 presence of organic carbon and higher salt content in the simulated wastewater and real secondary
551 effluent, led to lower mineralization though per dose of hydrogen peroxide compared to ultrapure
552 water. It is important to highlight that a new approach aimed at performing photo-Fenton treatment
553 at neutral pH has been proposed by Klamerth et al. (2010) and De la Cruz et al. (2012). The
554 efficiency of the modified photo-Fenton system is based on the reaction of dissolved organic matter
555 (DOM) present in wastewaters with Fe^{2+} leading to the formation of soluble iron-complexes.
556 However, contaminants degradation and mineralization tend to be slower at neutral pH than at pH
557 3.0.

558 Michael et al., (2012b) investigated the application of a solar photo-Fenton system for the
559 degradation of antibiotics at low concentration level (μg L^{-1}) in secondary treated domestic
560 effluents at a pilot-scale. The examined antibiotics were ofloxacin and trimethoprim and the pilot
561 treatment plant consisted of a compound parabolic collector reactor. The results demonstrated the
562 efficiency of the process in removing enterococci, resistant to these two antibiotics, while the
563 compounds themselves were completely eliminated. The total cost of a full-scale unit for the
564 treatment of 150 m^3 day^{-1} of secondary wastewater effluent was estimated to be 0.85 € m^{-3}. This
565 value was found to be in agreement with a previous study of the photo-Fenton process in a pilot
566 scale set-up (Jordá et al., 2011).

567 Another approach was taken by Lee et al. (2009) who used ferrate (Fe(VI)) to oxidize
568 micropollutants and remove phosphate by formation of ferric phosphates in wastewater. They
569 showed that Fe(VI) doses higher than 5 mg Fe L^{-1} were capable of eliminating sulfamethoxazole
570 and ciprofloxacin by more than 85%. In comparison to ozone, Fe(VI) was as effective or slightly
571 less effective in terms of micropollutants oxidation, with Fe(VI) having the benefit of phosphate
572 removal. In general, Fenton process has been extensively used with success for the oxidation of
573 many classes of antibiotics including β-lactams, quinolones, trimethoprim and tetracyclines.

574 *2.4.3 Heterogeneous photocatalysis with TiO_2*

575 Heterogeneous photocatalysis by TiO_2 semiconductor is achieved usually by the illumination of a
576 suspension of TiO_2 in aqueous solution with light energy greater than its bandgap energy. This
577 leads to the formation of high energy electron-hole pairs (e^-/h^+) which can migrate on the surface of
578 the catalyst and can either recombine producing thermal energy, or participate in redox reactions
579 with the compounds that are adsorbed on the catalyst's surface (Herrmann et al., 1993; Schiavello
580 et al., 1993; Robertson et al., 1996). The valence holes are strong oxidants and are able to oxidize
581 various contaminants, as well as water, resulting in the formation of $HO^{\bullet}$ while the conduction band
582 electrons are good reductants reducing the dissolved oxygen to $O_2^{\bullet-}$ (Munter et al., 2001).

583 The study of Elmolla and Chaudhuri (2011) examined the feasibility of using combined TiO_2
584 photocatalysis ($UV/TiO_2/H_2O_2$) and sequencing batch biological reactor (SBR) process for the
585 treatment of an antibiotic wastewater containing amoxicillin and cloxacillin. The complete removal
586 of these compounds was observed at TiO_2 and H_2O_2 doses of 1000 and 250 mg L^{-1}, respectively.
587 Amoxicillin was also completely removed from urban wastewater treatment plant effluent using
588 $[TiO_2]$=0.8 g L^{-1} after 120 min of treatment as reported by Rizzo et al. (2009). Ofloxacin in
589 wastewater samples was removed by 60% using $[TiO_2]$=3 g L^{-1} (Michael et al., 2010) while
590 Hapeshi et al. (2010) reported that the DOC of a solution contained ofloxacin at 10 mg L^{-1} was
591 reduced by 79% after 120 min of photocatalytic treatment using $[TiO_2]$=250 mg L^{-1} and
592 $[H_2O_2]$=0.07 mmol L^{-1}.

593 Besides some drawbacks of the heterogeneous photocatalysis (*e.g.* the rather small quantum yield
594 of the process; the relatively narrow light-response range of TiO_2; the need of post-separation and
595 recovery of the catalyst particles from the reaction mixture in aqueous slurry systems), TiO_2 seems
596 to possess some interesting features, such as high chemical stability in a wide pH range, strong
597 resistance to chemical breakdown and photocorrosion, commercial availability and good
598 performance. The catalyst is also cheap and can be reused (Andreozzi et al., 1999; Malato et al.,

599 2009). The properties of antibiotics to be treated such as pK_a and molecular structure will determine
600 not only the efficiency of their photocatalytic degradation but also the mechanisms of the oxidation
601 products formation (*i.e.* contribution of $HO^{\bullet}$ radical and valence band holes oxidation pathway).
602
603 *2.4.4 Sonolysis*
604 Ultrasound irradiation or sonolysis is a relatively new process in water and wastewater treatment
605 and therefore, has unsurprisingly received less attention than other AOPs. This is also reflected by
606 the small number of publications concerning the treatment of pharmaceutical compounds.
607 Ultrasound enhances chemical and physical changes in a liquid medium through the generation and
608 subsequent destruction of cavitation bubbles. These bubbles grow over a period of a few cycles to
609 an equilibrium size for the particular frequency applied. It is the fate of these bubbles when they
610 collapse in succeeding compression cycles that generates the energy for chemical and mechanical
611 effects (Parsons, 2004). The sonochemical degradation in aqueous phase involves several reaction
612 pathways and zones such as pyrolysis inside the bubble and/or at the bubble-liquid interface and
613 hydroxyl radical-mediated reactions at the bubble-liquid interface and/or in the liquid bulk.
614 Pyrolytic reactions inside or near the bubble as well as solution radical chemistry are the two major
615 pathways of sonochemical degradation (Emery et al., 2005).

616 According to the authors' best knowledge, only one paper is available up to now in the literature on
617 the applicability of sonolysis to remove antibiotics from wastewater effluents. Naddeo et al. (2009)
618 evaluated the ultrasonic process on the degradation of amoxicillin spiked in urban wastewater
619 effluent. It was found that the amoxicillin conversion was enhanced at increased applied power
620 densities, acidic conditions and in the presence of dissolved air and the maximum removal observed
621 was 40%.

622 It is important to note that there is limited literature (Hernández-Sancho et al., 2010; Mahamuni and
623 Adewuyi, 2010; Jordá et al., 2011; Hollender et al. 2009; Michael et al., 2012b) dealing with
624 advanced wastewater treatment process economics although this aspect is a very important issue.
625
626 2.5 Effect of disinfection on antibiotics removal
627 *2.5.1 Chlorination*
628 Limited studies have focused on the removal of antibiotics during wastewater treatment with
629 chlorine. Chlorination is by far the most common method of wastewater disinfection and is used
630 worldwide for the disinfection of pathogens before discharge into receiving streams, rivers or
631 oceans. From the chlorinated species, hypochlorite (ClO^-) has the highest standard oxidation
632 potential (E_0=1.48 V), followed by chlorine gas (E_0=1.36 V) and chlorine dioxide (E_0=0.95 V)
633 (Homem and Santos, 2011). The two major disadvantages of using chlorine based disinfectants are
634 (i) the safety hazards associated with storage, transportation and handling of chlorine, and (ii) the
635 potential formation of disinfection by-products.
636 The effective removal of antibiotics by chlorination from wastewater requires sufficient free
637 chlorine concentration and contact time. For example, cephalexin which was removed by 91% in
638 activated sludge treatment at the Stanley WWTP was further removed in the following disinfection
639 process by 99%, resulting in a total removal of 100% in the Stanley WWTP whole treatment
640 process (Li and Zhang, 2011). Li and Zhang (2011) also reported that during chlorine disinfection
641 process roxithromycin was eliminated by a further 18% (total removal 53%), erythromycin-H_2O by
642 24% (total removal 43%), sulfamethoxazole by 27% (total removal 73%) and trimethoprim by 40%
643 (total removal 65%).
644
645
646

2.5.2 Ultraviolet irradiation

Ultraviolet (UV) disinfection is increasingly finding applications in UWTPs. Photolytic degradation can be either direct or indirect. In direct photolysis, the target contaminant (in this case the antibiotic compound) absorbs a solar photon, which leads to a break-up of the molecule. In an indirect photolysis mechanism, naturally occurring molecules in the system such as dissolved organic matter (DOM) act as sensitizing species which generates strong reactive agents *e.g.* singlet oxygen ($^1O_2$), hydroxyl radicals ($HO^\bullet$) or alkyl peroxyl radicals ($^\bullet OOR$) and hydrate electrons under solar radiation (Arnold and McNeill, 2007; Fatta-Kassinos et al., 2011b). Generally, the degradation of a compound by UV irradiation is affected by the UV energy absorption and the quantum yield of the compound. UV energy absorption is expressed as molar extinction coefficient, which is a measure of how strongly a chemical species absorbs light at a given wavelength that can be used for its degradation (Kim et al., 2009).

Ultraviolet irradiation has been widely used for the treatment of waters and wastewaters worldwide. Several studies have reported the effective treatment of UV irradiation for removal of antibiotics in wastewater effluents (Adams et al., 2002; Ryan et al., 2011; Yuan et al., 2011). It has been recently reported that at high UV doses of nearly 11000-30000 mJ cm^2, an almost complete removal of tetracyclines and ciprofloxacin was achieved (Yuan et al., 2011). Kim et al. (2009) reported that sulfonamides (sulfamethoxazole and sulfadimethoxine) and quinolones (norfloxacin and nalidixic acid) showed high removal efficiency in the range of 86-100% during the UV process. In contrast to this, macrolides (clarithromycin, erythromycin and azithromycin) were removed by 24-34%. Among tetracyclines, chlorotetracycline concentration decreased to less than limit of detection during the UV process while only 15% removal efficiency was achieved for tetracycline. This can be explained by the low molar extinction coefficient of tetracycline (4108 M^{-1} cm^{-1}) comparing to that of chlorotetracycline (18868 M^{-1} cm^{-1}).

671 Another study of photolysis was conducted by Arslan-Alaton and Dogruel (2004) in which
672 penicillin in the form of formulation effluent with total COD=1555 mg L^{-1} was treated under UV
673 irradiation or UV combined with H_2O_2. In this study, the removal efficiency was very low
674 compared to the others described above (COD removal max=22% and TOC removal max=10%
675 with 30 and 40 mM of peroxide respectively) and this may be attributed to the complexity of the
676 formulation effluent (high COD and TOC values). Zuccato et al. (2010) also reported complete
677 elimination of amoxicillin in Varese WWTP with UV-light treatment. The addition of H_2O_2 to UV
678 has proven to be more efficient in removing antibiotics than UV alone, and lower fluence doses
679 need to be applied for the same removal (Kim et al. 2009, Yuan et al. 2011, Rosario-Ortiz et al.
680 2010).

681 Many of the antibiotics have aromatic rings, structural moieties (such as phenol and nitro groups)
682 heteroatoms, and other functional chromophore groups that can either absorb solar radiation or react
683 with photogenerated transient species in natural waters (*e.g.* photo excited natural organic matter-
684 NOM) (Fatta-Kassinos et al., 2011b). The organic matter (DOC, COD), UV dose, contact time and
685 the chemical structure of the compound are important factors governing the removal efficiency of
686 antibiotics during direct photolysis. This technology is only applicable to wastewater containing
687 photosensitive compounds and waters with low COD concentrations (*e.g.* river, drinking waters)
688 (Homem and Santos, 2011). Furthermore, wastewater effluents have different organic compounds
689 that may either inhibit or enhance the process by scavenging or generating oxidant species (humic
690 and inorganic substances like dissolved metals) (Jiao et al., 2008). Generally, photolysis has proved
691 to be less effective in degrading antibiotics in wastewater effluents and more energy demanding
692 (Katsoyiannis et al., 2011) than e.g. ozonation.
693
694
695

3. Conclusive remarks and future trends

The conventional sewage treatment facilities were never designed to deal with pharmaceutical compounds. Due to their highly variable physicochemical properties (chemical structure, solubility, octanol/water partition coefficient) as well as the operational conditions of the biological process, the efficiencies by which pharmaceuticals are removed vary substantially. Unfortunately, the lack of data concerning the biological treatment processes does not allow comparison among the various studies conducted, and there are only few studies, which comprehensively and systematically investigated operating conditions of the biological treatment. In general, MBR systems have been reported to be equal to or slightly more effective in removing some antibiotics compared to CAS treatment systems (Le-Minch et al., 2010); MBR is more expensive, but provides a more hygienic effluent due to the filtration. As a consequence of the inability of the most commonly applied biological treatments to sufficiently remove antibiotics, the latter are regarded as pseudo-persistent contaminants due to their continual introduction into the environment and permanent presence.

Advanced treatment, downstream of conventional biological process, can significantly improve antibiotics removal before effluent disposal. Although capital and operational costs of an advanced treatment increase the costs of conventional process, further improvement of micropollutants and other antibiotics removal, in line with possible stringent regulations might be difficult to achieve without advanced treatment. The installation of treatment techniques to remove antibiotics in wastewaters should also be flexible and allow their implementation not only in UWTPs, but also at important source points such as hospitals and the pharmaceutical industry.

More comprehensive studies are required to thoroughly understand the behaviour of antibiotics under both conventional sewage treatment and advanced treatment processes and to gain more knowledge on the elimination processes within the UWTPs including sorption onto sewage sludge.

720 Furthermore, studies should provide all basic treatment plant operational parameters since these are
721 essential for later comparison or assessments.

722 It is important to underline also the fact that only little information is currently available with regard
723 to transformation products formed in the environment or UWTPs and during oxidative treatment.
724 Future research should include a dedicated focus on the potential formation of pharmacologically
725 active or more toxic products during treatment processes. Additionaly, it is necessary to conduct
726 research on the occurrence, fate and removal of humans' metabolites in UWTPs. Most antibiotics
727 and their metabolites are excreted by humans after administration and therefore discharged to the
728 municipal sewage; however, only little is known about their biodegradability in the aquatic
729 environments.

730 From a practical point of view, it is necessary to study process integration to maximize the
731 treatment performance in removing antibiotics and for disinfection including those that can use
732 renewable energy resources to power the processes. Moreover, both environmental and economic
733 assessments are considered necessary in the framework of industrial scale applications for the
734 removal of antibiotic residues from wastewater.

735 Finally, evaluation of the negative impacts (i.e. antibiotic bacteria and resistance genes evolution,
736 toxicity on organisms and plants) caused by the presence of antibiotics in the environment are
737 considered as a necessity in order to reduce the risk for humans.

738

739 **Acknowledgements**

740 Nireas-International Water Research Center is a project co-funded by the European Regional
741 Development Fund and the Republic of Cyprus through the Research Promotion Foundation
742 (Strategic Infrastructure Project ΝΕΑΥΠΟΔΟΜΗ/ΣΤΡΑΤΗ/0308/09), and the University of
743 Cyprus. The authors would like to thank EU for the support provided through Cost Action
744 "TD0803: Detecting evolutionary hot spots of antibiotic resistances in Europe (DARE)". Luigi

745 Rizzo wishes to thank University of Salerno for funding the project “Effect of solar photolysis on
746 antibiotic degradation, antibiotic resistant bacteria inactivation as well as on their capacity to
747 develop antibiotic resistance in surface water”, Ex 60%, anno 2011.

748

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Table 1. Removal of antibiotics from wastewater effluents through biological treatment.

Antibiotic group Antibiotic	Initial concentration (ng L^{-1})	Effluent concentration (ng L^{-1}) / (% Removal efficiency)	Reference
β-Lactams			
Amoxicillin	280	Primary / 270 (3.6%*) CAS / *nd* (100%*)	*Watkinson et al., 2007*
	18	CAS / *nd* (100%**)	*Zuccato et al., 2010*
	6940	50 (99%*)	*Watkinson et al., 2009*
Ampicillin	17	13 (23.5%*)	*Cha et al., 2006*
	nd-389.5	CAS / 126.4±6.6 (67.5%**) CAS + chlorination / *nd*	*Li et al., 2009*
	(<34.4); 77.2-383	CAS_{Shatin} (*ne*) $CAS_{Stanley}$ (82%**) Disinfection (91%**) Final (97%**)	*Li and Zhang, 2011*
	$5*10^5$	MBR (94.4, 99.6, 99.9, 99.9%**)$_{[STR=3, 10, 30, 60 days]}$	*Xia et al., 2012*
Cephalexin	2000	78.2 (96%*)	*Costanzo et al., 2005*
	5600	Primary / 3900 (30%*) CAS / *nd* (100%*)	*Watkinson et al., 2007*
	670-2900	240-1800 (~9-89%**)	*Gulkowska et al., 2008*
	1563-4367	10-994 (36-99.8%**)	*Lin et al., 2009*
	64000	250 (99.6%*)	*Watkinson et al., 2009*
	175.4-534.9	CAS / 375.6±19.7 (30.4%**) CAS + chlorination / *nd* (100%**)	*Li et al., 2009*
	658-1718; 65.7-525	CAS_{Shatin} (53%**) $CAS_{Stanley}$ (91%**) Disinfection (99%**) Final (100%**)	*Li and Zhang, 2011*
Penicillin G	29	*na* (<LOD**)	*Gulkowska et al., 2008*
	10	300 (29%*)	*Watkinson et al., 2009*
Penicillin V	160	Primary / 10 (94%*) CAS / 20 (87.5%*)	*Watkinson et al., 2007*
	13800	2000 (86%*)	*Watkinson et al., 2009*
Cloxacillin	320	Primary / *nd* (100%*) CAS / *nd* (100%*)	*Watkinson et al., 2007*
	13	9 (31%*)	*Cha et al., 2006*
	4600	700 (85%*)	*Watkinson et al., 2009*
Cefaclor	980	Primary / 800 (18%*) CAS / *nd* (100%*)	*Watkinson et al., 2007*
	6150	1800 (71%*)	*Watkinson et al., 2009*
Cefotaxime	24-1100	34 (<LOD**)	*Gulkowska et al., 2008*
	38.4-93.0; *nd*	CAS_{Shatin} (~43%**) $CAS_{Stanley}$ (*ne)* Disinfection (*ne*) Final (*ne*)	*Li and Zhang, 2011*
Cephapirin	18	15 (17%*)	*Cha et al., 2006*
Oxacillin	14	8 (43%*)	*Cha et al., 2006*
Macrolides			
Roxithromycin	18	Primary / 9 (50%*) CAS / 60 (<0%*)	*Watkinson et al., 2007*
	10-40	Primary / 10-50 (3-9%**) CAS / 10-30 (-18-38%**) MBR (38, 60, 57%*)$_{[SRT 16, 33, 60-80 days]}$ FBR (~24%*)	*Göbel et al., 2005; Göbel et al., 2007*
	26-117	CAS / 36-69 (<0*) MBR / (*nd*, 31, 42)$_{[SRT=10, 27, 55 days]}$ (100, 52, 64%*)	*Clara et al., 2005*
	500	500 (0%*)	*Watkinson et al., 2009*
	3.5-25.3	CAS / 14.2±1.1 (43.9%**) CAS + chlorination / 2.9±0.0 (17.1%**)	*Li et al., 2009*
	810±420	540±70 (33%**)	*Ternes et al., 2007*
	102±32; 164±31; 75±14; 156±29	CAS + chlorination / 36±21 (65%*) Oxidation ditch + UV / 278±46 (<0%*) CAS / 35±8 (53%*) Chemically enhanced + Chlorination / 37±11 (76%*)	*Xu et al., 2007*
	50	(40, 60, 55%*)$_{[STR=16, 33, 60-80 days]}$	*Joss et al., 2005*
	35.6-135; 4.2-141	CAS_{Shatin} (46%**) $CAS_{Stanley}$ (40%**) Disinfection (18%**) Final (53%**)	*Li and Zhang, 2011*
	Na	<(5)-31	*McArdell et al., 2003*
	600	MBR/RO MBR (89.5±7.7%**)$_{[SRT>40 days]}$ RO (99.6±0.4%**) CAS-UF/RO UF (81.4±10.1%**)	*Sahar et al., 2010*

		RO (99.9±0.1%**)	
	500-1000	MBR (>50%**)[STR>100 days]	*Abegglen et al., 2009*
	10^4	(77%**)[SRT=44-72 days]	*Reif et al., 2008*
	$5*10^4$	MBR (57%**)[SRT=15 days] MBR (81%**)[SRT=30 days]	*Tambosi et al., 2010*
Azithromycin	152	96 (37%*)	*Gros et al., 2006*
	90-380	Primary / 80-320 (10-33%**) CAS / 40-380 (-26-55%**) MBR (<0, 5, 25%*)[SRT 16, 33, 60-80 days] FBR (12.5%*)	*Göbel et al., 2005; Göbel et al., 2007*
	4.5-53	4-23 (11-57%*)	*Loganathan et al., 2009*
	$1150_{(UWTP\ I)}$; $660_{(UWTP\ II)}$; $1680_{(UWTP\ III)}$	Secondary UWTP I / 1600 (<0*) UWTP II / 300 (55%*) UWTP III / 530 (68%*) Outlet UWTP I / 180 (84%*) UWTP II / 200 (70%*) UWTP III / 30 (98%*)	*Fatta et al., 2010*
	139	MBR (21%**)[STR=30-50 days]	*Kovalova et al., 2012*
	500-1000	MBR (>50%**)[STR>100 days]	*Abegglen et al., 2009*
	110-142	MBR-RO (75%**)[STR=45 days]	*Dolar et al., 2012*
Tylosin	55	Primary / *nd* (100%*) CAS / 20 (64%*)	*Watkinson et al., 2007*
	60	3400 (<0%*)	*Watkinson et al., 2009*
	1150±70	60±4 (95%*)	*Yang et al., 2004*
Clarithromycin	59-1433	12-32 (99%**)	*Lin et al., 2009*
	330-660	Primary / 160-440 (11-14%**) CAS / 150-460 (-45-20%**) MBR (54, 40, 90%*)[SRT 16, 33, 60-80 days] FBR (~10%*)	*Göbel et al., 2005; Göbel et al., 2007*
	319	CAS / 117 (13%**)	*Zuccato et al., 2010*
	105.7-724.2	(<LOQ)-610.6 (16%*)	*Spongberg et al., 2008*
	460±100	210±40 (54%**)	*Ternes et al., 2007*
	Na	57-328	*McArdell et al., 2003*
	1500	MBR/RO MBR (91.4±5.4%**)[SRT>40 days] RO (99.2±0.8%**) CAS-UF/RO UF (93.2±5.0%**) RO (99.2±0.8%**)	*Sahar et al., 2010*
	2555	MBR (50%**)[STR=30-50 days]	*Kovalova et al., 2012*
	500-1000	MBR (>50%**)[STR>100 days]	*Abegglen et al., 2009*
	700-2720	MBR-RO (87%**)[STR=45 days]	*Dolar et al., 2012*
Erythromycin	71-141	145-290 (79%**)	*Roberts and Thomas, 2006*
	12	CAS / 52 (0%**)	*Zuccato et al., 2010*
	$380_{(UWTP\ I)}$; $280_{(UWTP\ II)}$; $700_{(UWTP\ III)}$	Secondary UWTP I / 200 (47%*) UWTP II / 250 (11%*) UWTP III / 420 (40%*) Outlet UWTP I / 30 (92%*) UWTP II / 400 (<0*) UWTP III / <LOD (100%*)	*Fatta et al., 2010*
	830±270	620±440 (25%**)	*Ternes et al., 2007*
	751±109; 1978±233; 253±22; 469±38	CAS + chlorination / 430±73 (43%*) Oxidation ditch + UV / 2054±386 (<0*) CAS / 216±34 (15%*) Chemically enhanced + chlorination / 259±20 (45%*)	*Xu et al., 2007*
	1000	MBR/RO MBR (90.4±8.2%**)[SRT>40 days] RO (99.3±0.7%**) CAS-UF/RO UF (72.2±6.8%**) RO (99.3±0.7%**)	*Sahar et al., 2010*
	32-80	MBR-RO (80%**)[STR=45 days]	*Dolar et al., 2012*
	10^4	(91%**)[SRT=44-72 days]	*Reif et al., 2008*
Erythromycin-H_2O	470-810	510-850 (-12-19%**)	*Gulkowska et al., 2008*
	226-1537	361-811 (56%**)	*Lin et al., 2009*
	(<50)-1300	(<50)-300 (43.8-100%**)	*Karthikeyan and Meyer, 2006*
	60-190	Primary / 40-190 (-8-4%**)	*Göbel et al., 2005; Göbel et al.,*

		CAS / 50-140 (-22-7%**) MBR (32, 26, 90%*)[SRT 16, 33, 60-80 days] FBR (~25%)	*2007*
	16.7-51.3	CAS / 96.3±6.0 (55.6%**) CAS + chlorination / 37.9±0.6 (26.1%**)	*Li et al., 2009*
	200±10	80±5 (60%*)	*Yang et al., 2004*
	258-409; 169-374	CAS_{Shatin} (15%**) $CAS_{Stanley}$ (26%**) Disinfection (24%**) Final (43%**)	*Li and Zhang, 2011*
	Na	<(20)-199	*McArdell et al., 2003*
	820	CAS (35.4±50.5%**) MBR HF-UF (25.2±108.9%**)[SRT>60 days] MBR FS-MF (43.0±51.5%**)[SRT>60 days]	*Radjenovic et al., 2009b*
	188	MBR (<60%**)[STR=30-50 days]	*Kovalova et al., 2012*
	242-6755; 144-10025	Trickling filter beds / 292-2841 (0%**) CAS / 23-2772 (50%**)	*Kasprzyk-Hordern et al., 2009*
Spiramycin	603	CAS / 454 (25%**)	*Zuccato et al., 2010*
Sulfonamides			
Sulfamethoxazole	500	Primary / 570 (<0%*) CAS / 200 (60%*)	*Watkinson et al., 2007*
	179-1760	47-964 (26-88%**)	*Lin et al., 2009*
	1090	210 (~81%**)	*Yang et al., 2005*
	450	(<30) (>93%*)	*Choi et al., 2007*
	590	390 (34%*)	*Gros et al., 2006*
	390	310 (20%**)	*Brown et al., 2006*
	nd-145	CAS / 18-50 MBR / (56, *nd*, *nd*))[SRT=10, 27, 55 days] (61, 100, , 100%*)	*Clara et al., 2005*
	$5450_{(GZ\text{-}UWTP1)}$; $7910_{(GZ\text{-}UWTP2)}$	$GZ\text{-}UWTP_1$ Primary / 9460 (<0*) Secondary / *nq* Tertiary / *nd* $GZ\text{-}UWTP_2$ Primary / *nq* Secondary / *nq* Tertiary/ *nd*	*Peng et al., 2006*
	(<80)-674	(<80)-304 (42%**)	*Lindberg et al., 2005*
	20	70 (<0**)	*Bendz et al., 2005*
	(<50)-1250	(<50)-370 (17.8-100%**)	*Karthikeyan and Meyer, 2006*
	250-640	250 (67%**)	*Carballa et al., 2004*
	230-570	Primary / 90-640 (-21-(-5)%**) Secondary / 130-840 (-138-60%**) MBR (38, 40, 37%*)[SRT 16, 33, 60-80 days] FBR (~62.5%*)	*Göbel et al., 2005; Göbel et al., 2007*
	246	CAS / 46 (81%**)	*Zuccato et al., 2010*
	3000	200 (93%*)	*Watkinson et al., 2009*
	146.5-355.5	CAS / 46.6±2.6 (68.2%**) CAS + chlorination / 15.3±0.3 (95.7%**)	*Li et al., 2009*
	500-10000	(65-96%**)	*Yu et al., 2009*
	na	UWTP I Secondary / (<60)-640 Chlorination / (<50)-70 UWTP II Secondary / 100-1600 UV / 330-2140	*Renew and Huang, 2004*
	13-155	4-39 (69-75%*)	*Pailler et al., 2009*
	Na	Amherst (Primary / 2800±300; CAS / 1200±3; Nitrification / 700±40; Tertiary / 630±60; Final / 680±30) East Aurora (Primary / 880±80 ; Secondary / 200±3; Tertiary / 190±5 ; Final / 220±20) Holland (Primary / 750±40; Secondary / 480±30; Tertiary / 450±20; Final / 500±60) Lackawana (Primary / 720±60; Secondary / 460±40; Final / 380±30)	*Batt et al., 2007*
	820±230	620±90 (24%**)	*Ternes et al., 2007*
	16±5; 118±17; 10±3; 25±7	CAS + chlorination / 16±7 (0%) Oxidation ditch + UV / 78±13 (34%*) CAS / 12±3 (<0*) Chemically enhanced + chlorination / 9±4 (64%*)	*Xu et al., 2007*
	52.0-127; 163-230	CAS_{Shatin} (90%**) $CAS_{Stanley}$ (62%**) Disinfection (27%**) Final (73%**)	*Li and Zhang, 2011*

	93	CAS (73.8±12.7%**) MBR HF-UF (78.3±13.9%**)[SRT>60 days] MBR FS-MF (80.8±12.2%**)[SRT>60 days]	*Radjenovic et al., 2009b*
	500	MBR/RO MBR (69.6±7.3%*)[SRT>40 days] RO (97.6±2.4%*) CAS-UF/RO UF (60.3±21.7%*) RO (97.6±2.4%*)	*Sahar et al., 2010*
	3476	(7%**)	*Kovalova et al., 2012*
	500-1000	MBR (75-90%**)[STR>100 days]	*Abegglen et al., 2009*
	$5*10^5$	MBR (88.5, 96.9, 99.3, 99.5%**)[STR=3, 10, 30, 60 days]	*Xia et al., 2012*
	20-268	MBR-RO (69%**)[STR=45 days]	*Dolar et al., 2012*
	10^4	MBR (52%**)[SRT=44-72 days]	*Reif et al., 2008*
	$5*10^4$	MBR (55%**)[SRT=15 days] MBR (86%**)[SRT=30 days]	*Tambosi et al., 2010*
	<3-150; 20-274	Trickling filter beds / <3-23 (0%**) CAS / 4-44 (70%**)	*Kasprzyk-Hordern et al., 2009*
N^4-Acetylsulfamethoxazole	850-1600	Primary / 570-1200 (9-21%**) CAS / <20-150 (81-96%**) MBR (90, 75, 70%*)[SRT 16, 33, 60-80 days]	*Göbel et al., 2005; Göbel et al., 2007*
	1000	(92, 75, 68%*)[STR=16, 33, 60-80 days]	*Joss et al., 2005*
	2394	MBR (81%**)[STR=30-50 days]	*Kovalova et al., 2012*
Sulfamethazine	150	(<30) (>80%*)	*Yang et al., 2005*
	4010	(<30) (>99%*)	*Choi et al., 2007*
	110-210	(<50) (100%**)	*Karthikeyan and Meyer, 2006*
	2000-10000	(32-85%**)	*Yu et al., 2009*
	(<LOQ)-26.9	<LOQ (100%*)	*Spongberg et al., 2008*
	3.2-54.7; 17.8	CAS_{Shatin} (100%**) $CAS_{Stanley}$ (100%**) Disinfection (*ne*) Final (100%**)	*Li and Zhang., 2011*
	3	MBR/RO MBR (90.2±9.8%*)[SRT>40 days] RO (93.5±6.5%*) CAS-UF/RO UF (73.5±16.2%*) RO (93.5±6.5%*)	*Sahar et al., 2010*
	500-1000	MBR (75-90%**)[STR>100 days]	*Abegglen et al., 2009*
Sulfadiazine	$5100_{(GZ\text{-}UWTP1)}$; $5150_{(GZ\text{-}UWTP2)}$	$GZ\text{-}UWTP_1$ Primary / 4180 (19%*) Secondary / *nd* Tertiary / *nd* $GZ\text{-}UWTP_2$ Primary / *nd* Secondary / *nd* Tertiary / *nd*	*Peng et al., 2006*
	nd-73.0	CAS / 16.2±0.0 (72.8%**) CAS + chlorination / *nd*	*Li et al., 2009*
	72±22	CAS + chlorination / 36±13 (50%*)	*Xu et al., 2007*
	36.0-55.4; 4.4-530	CAS_{Shatin} (100%**) $CAS_{Stanley}$ (87%**) Disinfection (4%**) Final (88%**)	*Li and Zhang, 2011*
	1896	MBR (-23%**)[STR=30-50 days]	*Kovalova et al., 2012*
	500-1000	MBR (75-90%**)[STR>100 days]	*Abegglen et al., 2009*
	$5*10^5$	MBR (93.8, 97.5, 99.6, 99.7%**)[STR=3, 10, 30, 60 days]	*Xia et al., 2012*
Sulfathiazole	40	Primary / *nd* (100%*) CAS / *nd* (100%*)	*Watkinson et al., 2007*
	10570	180 (98%*)	*Choi et al., 2007*
	300	600 (<0%*)	*Watkinson et al., 2009*
	(1.0)-2.0	(<1.0) (100%*)	*Pailler et al., 2009*
Sulfamerazine	1530	(<30) (>98%*)	*Choi et al., 2007*
Sulfachloropyridazine	1560	60 (>93%*)	*Choi et al., 2007*
Sulfadimethoxine	70	(<30) (>57%*)	*Yang et al., 2005*
	460	(<30) (>93%*)	*Choi et al., 2007*
	2000-10000	(61-96%**)	*Yu et al., 2009*
	(<(LOQ)-2.6)	(<LOQ)-1.9 (27%*)	*Spongberg et al., 2008*
	(1.0)-26	(1.0)-9.0 (65%*)	*Pailler et al., 2009*
Sulfapyridine	60-150	Primary (-29-20%**) CAS (-107-72%**) MBR (60, 48, 55%*)[SRT 16, 33, 60-80 days] FBR (72%*)	*Göbel et al., 2005; Göbel et al., 2007*

	500-1000	MBR (75-90%**)[STR>100 days]	*Abegglen et al., 2009*
Sulfasalazine	60	Primary / 15 (75%*) CAS / *nd* (100%*)	*Watkinson et al., 2007*
	100	150 (<0%*)	*Watkinson et al., 2009*
Sulfamonomethoxine	3110	(<30) (>99%*)	*Choi et al., 2007*
Sulfisoxazole	(<LOQ)-22.1)	(<LOQ)-11.9 (46%*)	*Spongberg et al., 2008*
Sulfadimidine	25±12; 696±212	CAS + chlorination / 12±6 (52%*) Oxidation ditch + UV / 346±54 (50%*)	*Xu et al., 2007*
Quinolones			
Norfloxacin	*Na*	210	*Costanzo et al., 2005*
	210	Primary / 145 (31%*) CAS / 15 (93%*)	*Watkinson et al., 2007*
	110-460	85-320 (-20-78%**)	*Gulkowska et al., 2008*
	431±45	Primary / 383±61 (11%*) Secondary / 69±15 (84%*) Tertiary / 51±7 (88%*)	*Golet et al., 2003*
	(18±2.5; 27±3.0; 19.0±1.5; (<5.5))[UWTP1-UWTP5]	(>70%**)	*Zorita et al., 2009*
	66-174	(<7)-37 (87%**)	*Lindberg et al., 2005*
	339	85 (75%*)	*Xiao et al., 2008*
	388± 112	57±12 (82±3%**)	*Golet et al., 2002*
	220	250 (<0%*)	*Watkinson et al., 2009*
	nd-59.5	CAS / 13.9±0.5 (76.6%**) CAS + chlorination / *nd*	*Li et al., 2009*
	229±42; 179±41; 54±10; 263±36	CAS + chlorination / 44±19 (81%*) Oxidation ditch + UV / 62±13 (65%*) CAS / 27±6 (50%*) Chemically enhanced + chlorination / 85±12 (68%*)	*Xu et al., 2007*
	5933	MBR (47%**)[STR=30-50 days]	*Kovalova et al., 2012*
Ciprofloxacin	90	138.2 (<0%*)	*Costanzo et al., 2005*
	4600	Primary / 6900 (<0%*) CAS / 742 (84%*)	*Watkinson et al., 2007*
	427±69	Primary / 331±53 (22%*) Secondary / 95±15 (78%*) Tertiary / 71±11 (83%*)	*Golet et al., 2003*
	(320±10; 310±20; 94.0±12.0; 28.0±5.5; 31.5±4.0)[UWTP1-UWTP5]	(>90%**)	*Zorita et al., 2009*
	90-300	7-60 (87%**)	*Lindberg et al., 2005*
	(<50)-310	(<50)-60 (22.2-100%**)	*Karthikeyan and Meyer, 2006*
	80	27 (66%*)	*Xiao et al., 2008*
	434±93	72±14 (82±3%**)	*Golet et al., 2002*
	513	CAS / 147 (71%**)	*Zuccato et al., 2010*
	1100	*nd* (100%*)	*Watkinson et al., 2009*
	99.2-720.0	CAS / 73.3±3.0 (89.8%**) CAS + chlorination / 7.6±0.7 (92.3%**)	*Li et al., 2009*
	11.4-377.2	88-109.9 (71%*)	*Spongberg et al., 2008*
	na	UWTP I Secondary / (<30)-100 Chlorination / (<20) UWTP II Secondary / 80-370 UV / (<20)	*Renew and Huang, 2004*
	na	Amherst (Primary / 1100±100; CAS / 450±1; Nitrification / 450±4; Tertiary / 450±3; Final / 540±5) East Aurora (Primary / 610±30; Secondary / 290±30; Tertiary / 220±9; Final / 220±7) Holland (Primary / 1400±300; Secondary / 590±10; Tertiary / 450±60; Final / 340±60) Lackawana (Primary / 920±50; Secondary / 460±10; Final / 270±20)	*Batt et al., 2007*
	1674.20	626.50 (63%*)	*Castiglioni et al., 2008*
	555-1033; 98.6-235	CAS_{Shatin} (18%**) $CAS_{Stanley}$ (55%**) Disinfection (18%**) Final (66%**)	*Li and Zhang., 2011*
	31980	MBR (51%**)[STR=30-50 days]	*Kovalova et al., 2012*
Enrofloxacin	100	Primary / 20 (80%*) CAS / 5 (95%*)	*Watkinson et al., 2007*

	40	50 (<0%*)	*Watkinson et al., 2009*
Ofloxacin	115-1274	53-991 (2-88%**)	*Lin et al., 2009*
	470	110 (77%**)	*Brown et al., 2006*
	(22.5±2.5; 30.0±3.0; 19.5±3.0; 9.0±1.5; 10.0±1.0)$_{[UWTP1-UWTP5]}$	(56%**)	*Zorita et al., 2009*
	5560$_{(GZ-UWTP1)}$; 3520$_{(GZ-UWTP2)}$	GZ-UWTP$_1$ Primary / 5700 (<0*) Secondary / 860 (85%*) Tertiary / 740 (87%*) GZ-UWTP$_2$ Primary / *nq* Secondary / *nd* (100%*) Tertiary / *nd* (100%*)	*Peng et al., 2006*
	7-287	7-52 (86%**)	*Lindberg et al., 2005*
	1208	503 (58%*)	*Xiao et al., 2008*
	463	CAS / 235 (49%**)	*Zuccato et al., 2010*
	104.4- 335.9	CAS / 556.4±28.7 (-65.6%**) CAS + chlorination / 2.1±0.3 (98.0%**)	*Li et al., 2009*
	na	UWTP I Secondary / (<30)-350 Chlorination / (<20)-50 UWTP II Secondary / 140-260 UV / 100-210	*Renew and Huang, 2004*
	122620$_{(UWTP\ I)}$; 34740$_{(UWTP\ II)}$; 59380$_{(UWTP\ III)}$	Secondary UWTP I / 3020 (87%*) UWTP II / 5930 (83%*) UWTP III / 3330 (94%*) Outlet UWTP I / 1290 (94%*) UWTP II / 4820 (86%*) UWTP III / 1900 (97%*)	*Fatta et al., 2010*
	539.80	183.10 (66%*)	*Castiglioni et al., 2008*
	137±58; 359±52; 80±12; 368±23	CAS + chlorination / 41±8 (70%*) Oxidation ditch + UV / 137±28 (62%*) CAS / 48±7 (40%*) Chemically enhanced + chlorination / 165±15 (55%*)	*Xu et al., 2007*
	478-1042; 188-327	CAS$_{Shatin}$ (26%**) CAS$_{Stanley}$ (59%**) Disinfection (39%**) Final (74%**)	*Li and Zhang, 2011*
	10500	CAS (75.8±13.8%**) MBR HF-UF (91.3±10.8%**)$_{[SRT>60\ days]}$ MBR FS-MF (95.2±2.8%**)$_{[SRT>60\ days]}$	*Radjenovic et al., 2009b*
	nd-2900	MBR-RO (0%**)$_{[STR=45\ days]}$	*Dolar et al., 2012*
Nalidixic acid	200	Primary/ *nd* (100%*) CAS / 1 (100%*)	*Watkinson et al., 2007*
	26-372	40-200 (37-46%**)	*Lin et al., 2009*
	200	450 (<0%*)	*Watkinson et al., 2009*
Pipemidic acid	54	12 (78%*)	*Xiao et al., 2008*
Flerofloxacin	28	5.8 (79%*)	*Xiao et al., 2008*
Lomefloxacin	98	17 (83%*)	*Xiao et al., 2008*
Gatifloxacin	111	56 (50%*)	*Xiao et al., 2008*
Moxifloxacin	44	17 (61%*)	*Xiao et al., 2008*
Trimethoprim			
	930	Primary / 480 (48%*) CAS / 30 (97%*)	*Watkinson et al., 2007*
	120-320	120-230 (~-17-62%**)	*Gulkowska et al., 2008*
	259-949	203-415 (~22-56%**)	*Lin et al., 2009*
	1172	290 (75%*)	*Gros et al., 2006*
	590	180 (69%**)	*Brown et al., 2006*
	99-1300	66-1340 (3%**)	*Lindberg et al., 2005*
	140-1100	(<50)-550 (50-100%**)	*Karthikeyan and Meyer, 2006*
	80	40 (49%**)	*Bendz et al., 2005*
	213-300	218-322 (3%**)	*Roberts and Thomas., 2006*
	210-440	Primary / 80-340 (-13-31%**) CAS / 80-400 (-40-20%**) MBR (28, 33, 87%*)$_{[SRT\ 16,\ 33,\ 60\text{-}80\ days]}$ FBR (~20%*)	*Göbel et al., 2005; Göbel et al., 2007*
	400	Primary (~20%**) Secondary (76±24%**)	*Sui et al., 2010*

	4300	250 (94%*)	*Watkinson et al., 2009*
	128.7-161.2	CAS / 66.2±0.7 (48.6%**) CAS + chlorination / 10.8±1.1 (93.3%**)	*Li et al., 2009*
	1000	(74%**)	*Yu et al., 2009*
	na	UWTP I Secondary / 30-1210 Chlorination / (<40) UWTP II Secondary / 270-1220 UV / (<40)-1760	*Renew and Huang, 2004*
	na	Amherst (Primary / 7900±400; CAS / 7600±500; Nitrification / 2500±300; Tertiary / 2600±200; Final / 2400±200) East Aurora (Primary / 7000±1000; Secondary / 300±30; Tertiary / 270±20; Final / 210±9) Holland (Primary / 2300±500; Secondary / 580±20; Tertiary / 570±10; Final / 540±50) Lackawana (Primary / 2100±400; Secondary / 590±3; Final / 360±40)	*Batt et al., 2007*
	$50_{(UWTP\ I)}$; $140_{(UWTP\ II)}$; $350_{(UWTP\ III)}$	Secondary UWTP I / <LOD (100%*) UWTP II / 90 (36%*) UWTP III / 60 (83%*) Outlet UWTP I / <LOD (100%*) UWTP II / <LOD (100%*) UWTP III / <LOD (100%*)	*Fatta et al., 2010*
	1100±260	340±80 (69%**)	*Ternes et al., 2007*
	100-154; 136-172	CAS_{Shatin} (13%**) $CAS_{Stanley}$ (42%**) Disinfection (40%**) Final (65%**)	*Li and Zhang., 2011*
	204	CAS (40.4±25.4%**) MBR HF-UF (47.5±22.5%**)$_{[SRT>60\ days]}$ MBR FS-MF (66.7±20.6%**)$_{[SRT>60\ days]}$	*Radjenovic et al., 2009b*
	30	MBR/RO MBR (96±4%*)$_{[SRT>40\ days]}$ RO (97.2±2.8%*) CAS-UF/RO UF (66.4±20.5%*) RO (93.2±6.8%*)	*Sahar et al., 2010*
	930	MBR (96%**)$_{[STR=30-50\ days]}$	*Kovalova et al., 2012*
	10^4	MBR (36%**)$_{[SRT=44-72\ days]}$	*Reif et al., 2008*
	$5*10^4$	MBR (55%**)$_{[SRT=15\ days]}$ MBR (86%**)$_{[SRT=30\ days]}$	*Tambosi et al., 2010*
	464-6769; 1514-4673	Trickling filter beds / 625-3052 (40%**) CAS / 385-1218 (70%**)	*Kasprzyk-Hordern et al., 2009*
Tetracyclines			
Tetracycline	35	Primary / *nd* (100%*) CAS / 20 (43%*)	*Watkinson et al., 2007*
	96-1300	180-620 (-88-73%**)	*Gulkowska et al., 2008*
	46-234	16-38 (66-90%**)	*Lin et al., 2009*
	200	(<30) (>~85%*)	*Yang et al., 2005*
	110	(<30) (>73%*)	*Choi et al., 2007*
	240-790	(<50)-160 (67.9-100%**)	*Karthikeyan and Meyer, 2006*
	100	20 (80%*)	*Watkinson et al., 2009*
	134.5-270.8	CAS / 89.4±4.2 (67.0%**) CAS + chlorination / *nd* (100%**)	*Li et al., 2009*
	29.3-38.9	(<LOQ)-34.4 (12%*)	*Spongberg et al., 2008*
	(1.0)-85	(1.0)-24 (72%*)	*Pailler et al., 2009*
	na	Amherst (Primary / 1100±100; CAS / 410±20; Nitrification / 170±10; Tertiary / 170±2; Final / 160±1) East Aurora (Primary / 320±30; Secondary / 75±3; Tertiary / 61±9; Final / 61±3) Holland (Primary / 580±20; Secondary / 240±20; Tertiary / 220±40; Final / 210±2) Lackawana (Primary / 430±200; Secondary / 240±20; Final / 290±30)	*Batt et al., 2007*
	221-353; 59.8-110	CAS_{Shatin} (24%**) $CAS_{Stanley}$ (36%**) Disinfection (13%**)	*Li and Zhang, 2011*

		Final (39%**)	
	$5*10^5$	MBR (83.6, 89.7, 92.6, 93.6%**)[STR=3, 10, 30, 60 days]	*Xia et al., 2012*
Chlortetracycline	270	60 (~78%**)	*Yang et al., 2005*
	970	40 (>96%*)	*Choi et al., 2007*
	200	250 (<0%*)	*Watkinson et al., 2009*
	155; 178	CAS_{Shatin} (85%**) $CAS_{Stanley}$ (82%**) Disinfection (6%**) Final (83%**)	*Li and Zhang., 2011*
	$5*10^5$	MBR (82.9, 84.4, 81.5, 77.6%)[STR=3, 10, 30, 60 days]	*Xia et al., 2012*
Doxycycline	65	Primary / 40 (78%*) CAS / 20 (69%*)	*Watkinson et al., 2007*
	210	70 (~67%**)	*Yang et al., 2005*
	220	30 (86%*)	*Choi et al., 2007*
	(<64)-2480	(<64)-915 (~70%**)	*Lindberg et al., 2005*
	650	150 (77%*)	*Watkinson et al., 2009*
Oxytetracycline	240	(<30) (>88%*)	*Choi et al., 2007*
	350	70 (80%*)	*Watkinson et al., 2009*
	(1.0)-7.0	(1.0)-5.0 (29%*)	*Pailler et al., 2009*
	53.5-107; *nd*	CAS_{Shatin} (44%**)	*Li and Zhang, 2011*
	$5*10^5$	MBR (79.7, 84.4, 87.9, 88.6%**)[STR=3, 10, 30, 60 days]	*Xia et al., 2012*
Minocycline	380	(<30) (>92%*)	*Choi et al., 2007*
Democlocycline	270	30 (89%*)	*Choi et al., 2007*
Meclocycline-Sulfosalicylate	500	180 (64%*)	*Choi et al., 2007*
Lincosamides			
Lincomycin	80	Primary / 70 (12.5%*) CAS / 50 (37.5%*)	*Watkinson et al., 2007*
	9.7	CAS / 6.1 (37%**)	*Zuccato et al., 2010*
	500	300 (40%*)	*Watkinson et al., 2009*
	3.9	3.70 (5%*)	*Castiglioni et al., 2008*
Clindamycin	5	Primary / 5 (0%*) CAS / 5 (0%*)	*Watkinson et al., 2007*
	60	70 (<0%*)	*Watkinson et al., 2009*
	6.8-13.3	14.9-32.5 (<0%*)	*Spongberg et al., 2008*
	983	MBR (-18%**)	*Kovalova et al., 2012*
Polyether ionophores			
Monensin	190	Primary / 10 (95%*) CAS / 1 (99.5%*)	*Watkinson et al., 2007*
Salisomycin	300	*nd* (100%*)	*Watkinson et al., 2009*
Glycopeptides			
Vancomycin	41	CAS / 40 (2%**)	*Zuccato et al., 2010*
	(<36.5)-60.6; *nd*	CAS_{Shatin} (52%**)	*Li and Zhang, 2011*
Aminoglycosides			
Gentamicin	400-7600	200-1300 (50-83%*)	*Löffler and Ternes, 2003*
Nitroimidazoles			
Metronidazole	*nd*-1140	MBR-RO (95%**)[STR=45 days]	*Dolar et al., 2012*
	158-1583; 347-962	Trickling filter beds / 60-421 (21%*) CAS / 129-561 (23%**)	*Kasprzyk-Hordern et al., 2009*
	1000-2000	(<30%**)	*Jelic et al., 2011*
	3388	MBR (45%**)[STR=30-50 days]	*Kovalova et al., 2012*

NOTES

- CAS: Conventional activated sludge treatment; MBR: Membrane bioreactor; FBR: Fixed bed bioreactor; SRT: Sludge retention time; HRT: Hydraulic retention time
- **(*)** Removal efficiencies, not reported by authors in the cited study, are calculated from the average influent and effluent concentrations which were stated in the study.
- **(**)** Removal efficiencies reported by authors in the cited study
- Value in the parenthesis is the limit of detection (LOD)
- Negative removal values result from an observed increase of loads from inflow to outflow of wastewater treatment.
- LOQ: Limit of Quantification
- *nd*: not detected; *na*: not available; *ne:* not evaluated; *nq:* not quantified

SUPPORTING INFORMATION

Reference	Location	Main treatment steps
Abegglen et al., 2009	Switzerland	CAS and MBR (aerobic or anoxic). SRT_1>150 days; HRT_1=6.3 days; SRT_2>100 days; HRT_2=3.4 days
Batt et al., 2007	Erie County (New York) (Amherst, East Aurora, Holland, Lackawanaa).	Amherst: Primary treatment; Secondary treatment (Stage1: CAS; Stage2: nitrification); Tertiary treatment (Sand filtration); Chlorination. East Aurora: No Primary treatment; Secondary treatment (Extended aeration; Ferrous chloride addition); Tertiary treatment (Sand filtration); UV radiation. Holland: Primary treatment; Secondary treatment (Rotating biological contactors); Tertiary treatment (Sand filtration); UV radiation. Lackawana: Primary treatment; Secondary treatment (Pure oxygen activated sludge); Chlorination.
Bendz et al., 2005	Kallby (Sweden)	Primary treatment (Bar screening; Grit removal; Primary clarification); CAS; Chemical phosphorous removal; Final sedimentation.
Brown et al., 2006	Rio Grande (Colorado) (Magdalena; Hagerman; Socorro; Portales; Santa Fe; Albuquerque)	CAS
Carballa et al., 2004	Galicia (Spain)	Pre-treatment (coarse screening, bar racks, fine screening and aerated chambers for grit and fat removal); Primary treatment; CAS; Sedimentation tank.
Castiglioni et al., 2008	Varese Olona (Italy)	*na*
Cha et al., 2006	Fort Collins (Colorado)	Pretreatment; Primary treatment; CAS; Chlorination.
Choi et al., 2007	Korea	CAS
Clara et al., 2005	South-East of Austria	Primary treatment (screen; grit chamber); CAS. MBR pilot plant (UF, cross flow); SRT=10-55 days.
Costanzo et al., 2005	Brisbane (Australia)	*Na*
Dolar et al., 2012	Castell-Platja d'Aro (Spain)	MBR-RO pilot plant (8 m^2 of flat sheet membranes; pore size of 0.4 μm): HRT=12.5 h; SRT=45 days RO system: one pressure vessel housing, a double element (Ropur membranes TR70-4021-HF) with an automatic cleaning system; high flow, crosslinked, aromatic polyamide, negative charge spiral wound module.
Fatta et al., 2010	Cyprus	UWTP I: Primary treatment; Secondary treatment (oxidation ditches, secondary settlement); Tertiary treatment (sand filtration); Chlorination. UWTP II: Primary treatment; CAS; Tertiary treatment (sand filtration); Chlorination. UWTP III: Primary treatment; Secondary treatment (phosphorus biological removal, nitrification and denitrification, secondary clarifiers); Tertiary treatment (sand filtration); Chlorination.
Göbel et al., 2005; Göbel et al., 2007	Switzerland (Kloten-Opfikon (UWTP-K); Altenrhein (UWTP-A))	Primary treatment (screen, aerated grit-removal tank, primary clarifier); Secondary treatment (UWTP-K: CAS; UWTP-A: CAS and FBR); Tertiary treatment (sand filtration).MBR (in UWTP-K): operated in parallel to CAS (HRT=13 h). Three different membrane filtration units: MF plate membrane module (0.4 μm); UF hollow-fibre modules (0.1 μm); UF hollow-fibre modules (0.04 μm). SRT_1=16; SRT_2=33 days; SRT_3=60-80 days FBR (in UWTP-A): 8 Biostyr up-flow cells, 3.6 mm Styrofoam beads as biofilm support
Golet et al., 2002	Glatt Valley Watershed (Switzerland)	CAS
Golet et al., 2003	Zurich-Werdholzli (Switzerland)	Primary treatment (screens; combined grid; fat removal tank; primary clarification); CAS (SRT=11days); Denitrification; Flocculation-filtration.
Gros et al., 2006	Croatia	CAS
Gulkowska et al., 2008	Hong Kong and Shenzhen (China) (Wan Chai, Shatin, Tai Po, Stonecutters Island, Nan Shan)	Wan Chai: Primary treatment Shatin: Primary treatment (Screening; Settlement of grit particles; Primary sedimentation); CAS Tai Po: Primary treatment (Removal of solids and grit; Primary sedimentation); Biological treatment; Stonecutters Island: Chemically enhanced primary treatment Nan Shan: Primary treatment
Jelic et al., 2011	Catalonia (Spain)	$UWTP_1$: Pre-treatment, Primary treatment; Secondary treatment (anoxic/aerobic and secondary settling, coagulation/flocculation/lamella clarifier); Tertiary treatment (microfiltration); Chlorination. $UWTP_2$: Pre-treatment, Primary treatment, CAS $UWTP_3$: Primary treatment; Secondary biological treatment (nitrogen and phosphorus removal).
Joss et al., 2005	Switzerland (Kloten-Opfikon (UWTP-K); Altenrhein (UWTP-A))	Primary treatment (screen, aerated grit-removal tank, primary clarifier); Secondary treatment (UWTP-K: CAS; UWTP-A: CAS and a FBR); Tertiary treatment (sand filtration). SRT_1=16-33 days; SRT_2=60-80 days
Karthikeyan and Meyer, 2006	Wisconsin (USA)	CAS

Kasprzyk-Hordern et al., 2009	South Wales (England)	Cilfynydd: Trickling filter beds; Coslech: CAS
Kovalova et al., 2012	Switzerland	Pilot-scale MBR: average influent of 1.2 m^3 day^{-1} pumped directly from the hospital sewer collection system. Sludge concentration= 2 g L^{-1}, SRT=30-50 days, $T_{average}$=29 °C, pH=7.8, conductivity=1100 μS cm^{-1}. Submerged ultrafiltration flat sheet membrane plates (Huber MembraneClearBox, PP carrier, PES membrane, 7 m^3, 15−30 $L \cdot m^{-2} \cdot h^{-1}$, 38 nm pore size, 150 kDa).
Li and Zhang, 2011	Hong Kong (Stanley and Shatin)	Shatin (Anoxic-Aerobic CAS); Stanley (Anoxic-Aerobic CAS and Chlorination)
Li et al., 2009	Hong Kong (Stanley and Shatin)	*na*
Lin et al., 2009	Taipei (Taiwan)	$UWTP_1$: Screening and sedimentation; CAS; UV. $UWTP_2$: Grit removal and screening and sedimentation, deep shaft and step aeration and sedimentation; Chlorination. $UWTP_3$: Screening; Trickling filter and sedimentation; Chlorination. $UWTP_4$: Screening and grit removal and sedimentation; CAS and sedimentation; Chlorination.
Lindberg et al., 2005	Sweden (Stockholm; Gothenburg; Umeå; Kalmar; and Floda)	Chemical removal of phosphorus; Primary clarification; CAS with nitrogen removal (except Umeå and Floda); Secondary clarification.
Löffler and Ternes, 2003	Germany	Hospital wastewater; 0.45-μm polystyrene filters
Loganathan et al., 2009	South-western Kentucky	Large grit removal; Returned Activated Sludge; Post-Clarifier/Pre-Chlorination; Oxidation ditch; Post-Chlorination
McArdell et al., 2003	Switzerland (Kloten-Opfikon; Zurich-Werdhoelzli; and Duebendorf)	Primary treatment; Secondary treatment; Tertiary treatment (sand filtration)
Pailler et al., 2009	Beggen (Luxemburg)	*na*
Peng et al., 2006	Guangzhou (China)	GZ-$UWTP_1$: Sedimentation; CAS; Filtration. GZ-$UWTP_2$: CAS; Filtration; Chlorination.
Radjenovic et al., 2009b	Terrassa (Spain)	Two pilot-scale MBRs were operating in parallel with CAS (SRT>60 days): Hollow-fibre ultra-filtration membranes (HF-UF) (HRT=7.2 h); flat-sheet micro-filtration membranes (FS-MF) (HRT=15 h).
Reif et al., 2008	Spain	MBR: Zenon ZW-10 submerged hollow fibre membrane module (average pore size=0.04 μm; nominal surface area of 0.9 m^2), SRT=44-72 days.
Renew and Huang, 2004	California (UWTP I) and Arizona (UWTP II) (Georgia)	Primary treatment (screening and sedimentation); CAS; Tertiary treatment; Disinfection (UWTP I: chlorination; UWTP II: UV).
Roberts and Thomas, 2006	Howdon (UK)	Primary treatment (coarse screening; preliminary clarification); CAS and trickling filter system; High-pressure 254 nm UV disinfection.
Sahar et al., 2010	Tel-Aviv (Israel)	MBR/RO plant: Two Zenon ZeeWeed 500 UF immersed hollow fiber membranes (total area=2 m^2); RO membrane Filmtec TW30 25-40 (surface area=2.7 m^2). CAS-UF/RO plant: UF (24 modules, 1024 m^2, ZeeWeed-1000 immersed hollow fibers); RO membrane Filmtec BW30-400 (total area= 1295 m^2). SRT > 40 days
Spongberg et al., 2008	Northwest Ohio (USA)	*na*
Sui et al., 2010	Beijing (China)	Primary treatment; Secondary biological treatment (A and D: anaerobic/anoxic/oxic [A^2/O]) CAS; B: anoxic/oxic [A/O]) CAS; C: Oxidation ditch [OD].
Tambosi et al., 2010	Aachen (Germany)	MBR pilot plant receives effluent from the pre-settling tank. MBR-15 (V=260 L): SRT=15 days; HRT=6 h MBR-30 (V=240 L): SRT=30 days; HRT=13 h Hollow-fiber ultrafiltration (UF) membranes (PURON, KMS Germany): area=1.43 m^2; pore size=0.04 μm; polyethersulfone (PES).
Ternes et al., 2007	Braunschweig (Germany)	Primary treatment (screen; aerated grid-removal tank; primary clarifier); CAS; Phosphate removal; Nitrification-denitrification.
Watkinson et al., 2007	Brisbane (Australia)	Primary treatment; CAS (SRT=12.5 days)
Watkinson et al., 2009	South-East Queensland (Australia)	*na*
Xia et al., 2012	China	Lab-scale A/O-MBR (6 L): (i) anoxic unit (AN, 2 L) and (ii) aerobic unit (AO, 4 L). A hydrophilic polyvinylidene fluoride (PVDF) hollow fiber membrane module was used in the AO unit (pore size=0.02 μm; effective filtration area=0.1 m^2). SRT=3-60 days; HRT=6-24 h
Xiao et al., 2008	Gao Beidian (Beijing, China)	Primary treatment; Secondary treatment processes
Xu et al., 2007	Guangzhou and Hong Kong (South China) (Kaifaqu, Liede, New Territory, Kowloon)	Kaifaqu: Primary treatment; CAS; Chlorination. Liede: Primary treatment; Oxidation ditch; UV. New Territory: Primary treatment; CAS. Kowloon: Primary treatment; Chemically enhanced; Chlorination.
Yang et al., 2004	Northern Colorado (USA)	*Na*
Yang et al., 2005	Fort Collins (Colorado)	Pretreatment; Primary treatment; Secondary treatment (secondary clarification); Chlorination.
Yu et al., 2009	Taiwan	Extended sludge age biological technology (HRT=12 h; SRT> 200 days; MLSS=16000 mg L^{-1})
Zorita et al., 2009	Kristianstad (Sweden) ($UWTP_1$-$UWTP_5$)	Primary treatment (screens; grit-aerated chamber); CAS; Chemical removal; Tertiary treatment (Sand filtration).
Zuccato et al., 2010	Italy and Switzerland (Milan, Varese, Como, Lugano)	Pre-treatment; Primary treatment (primary settling); CAS; UV-light treatment (Varese).

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Table 2. Removal of antibiotics from wastewater effluents through advanced treatment processes.

Advanced treatment process Antibiotic Group	MEMBRANE FILTRATION				
	Type of wastewater (location)	**Initial concentration**	**Treatment process**	**Results/findings (Removal efficiency)**	**Reference**
β-Lactams					
Amoxicillin	Simulated wastewater (USA)	10 mg L^{-1}	RO: plate and frame configuration, ACM-LP fully aromatic polyamide low pressure advanced composite membrane	(100%)	*Morse and Jackson, 2004*
	CAS effluent (Australia)	280 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: *nd*; RO: *nd*	*Watkinson et al., 2007*
	Wastewater from plant manufacturing AMX (China)	*na*	Laboratory-scale cross flow RO unit. Two high-pressure cross flow membrane cells (SS316, 155 cm^2) mounted with a flat-sheet polyamide RO membrane. TOC=18925 mg L^{-1} COD=80000 mg L^{-1}	RO_1 TOC=283.9 mg L^{-1} (98.5%) COD=800 mg L^{-1} (99.0%) RO_2 TOC=56.8 mg L^{-1} (99.7%) COD=240mg L^{-1} (99.7%)	*Zhang et al., 2006*
Cefaclor	CAS effluent (Australia)	980 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: *nd*; RO: *nd*	*Watkinson et al., 2007*
Cephalexin	CAS effluent (Australia)	5600 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 100 ng L^{-1}; RO: 40 ng L^{-1}	*Watkinson et al., 2007*
Penicillin V	CAS effluent (Australia)	160 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: *nd*; RO: *nd*	*Watkinson et al., 2007*
Cloxacillin	CAS effluent (Australia)	320 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: *nd*; RO: *nd*	*Watkinson et al., 2007*
Macrolides					
Roxithromycin	CAS effluent (Australia)	100 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 125 ng L^{-1}; RO: 15 ng L^{-1}	*Watkinson et al., 2007*
Tylosin	CAS effluent (Australia)	55 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 10 ng L^{-1}; RO: 5 ng L^{-1}	*Watkinson et al., 2007*
Sulfonamides					
Sulfamethoxazole	*na*	1 mg L^{-1}	RO membranes: Polyamide (XLE); Cellulose acetate (SC-3100). Cross flow membrane unit with a flat-sheet membrane cell Effective membrane area in the cell= 32 cm^2	XLE (70%) SC-3100 (82%)	*Kimura et al., 2004*
	CAS effluent (Australia)	500 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 445 ng L^{-1}; RO: *nd*	*Watkinson et al., 2007*
Sulfadiazine	Model wastewater for veterinary use (Croatia)	10 mg L^{-1}	RO membranes: XLE; HR95PP; TFC-S. NF membranes: NF90; HL Desal, Osmonics Surface area of membranes: 10.8 cm^2	XLE (99.4%) HR95PP (99.4%) TFC-S (100 %) NF90 (99.4 %) HL (88.5 %)	*Kosutic et al., 2007*
Sulfaguanidine	Model wastewater for veterinary use (Croatia)	10 mg L^{-1}	RO membranes: XLE; HR95PP; TFC-S. NF membranes: NF90; HL Desal, Osmonics Surface area of membranes: 10.8 cm^2	XLE (99.3%) HR95PP (98.9%) TFC-S (100 %) NF90 (99.1 %) HL (67.3 %)	*Kosutic et al., 2007*
Sulfamethazine	Model wastewater for veterinary use (Croatia)	10 mg L^{-1}	RO membranes: XLE; HR95PP; TFC-S. NF membranes: NF90; HL Desal, Osmonics Surface area of membranes: 10.8 cm^2	XLE (99.1%) HR95PP (99.3%) TFC-S (100 %) NF90 (99.4 %) HL (96.3 %)	*Kosutic et al., 2007*
	Missouri River water (Jefferson City)	50 μg L^{-1}	Barnstead RO system: Model D2716, Cellulose acetate membrane D2731, Flow: 1.9 L min^{-1}.	(90.3%)	*Adams et al., 2002*
Sulfathiazole	Missouri River water (Jefferson City)	50 μg L^{-1}	Barnstead RO system: Model D2716, Cellulose acetate	(90.3%)	*Adams et al., 2002*

			membrane D2731, Flow: 1.9 L min^{-1}.		
	CAS effluent (Australia)	40 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: *nd*; RO: *nd*	*Watkinson et al., 2007*
Sulfamerazine	Missouri River water (Jefferson City)	50 μg L^{-1}	Barnstead RO system: Model D2716, Cellulose acetate membrane D2731, Flow: 1.9 L min^{-1}.	(90.3%)	*Adams et al., 2002*
Sulfachloropyridazine	Missouri River water (Jefferson City)	50 μg L^{-1}	Barnstead RO system: Model D2716, Cellulose acetate membrane D2731, Flow: 1.9 L min^{-1}.	(90.3%)	*Adams et al., 2002*
Sulfadimethoxine	Missouri River water (Jefferson City)	50 μg L^{-1}	Barnstead RO system: Model D2716, Cellulose acetate membrane D2731, Flow: 1.9 L min^{-1}.	(90.3%)	*Adams et al., 2002*
Sulfasalazine	CAS effluent (Australia)	60 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 55 ng L^{-1}; RO: *nd*	*Watkinson et al., 2007*
Quinolones					
Enrofloxacin	Model wastewater for veterinary use (Croatia)	10 mg L^{-1}	RO membranes: XLE (Dow/FilmTec, Midland MI); HR95PP (Dow/FilmTec, Midland MI); TFC-S (Koch Membrane Systems, Wilmington, MA). NF membranes: NF90 (Dow/FilmTec); HL Desal, Osmonics (GE Infrastructure Water Process Techn.,Vista, CA). Surface area of membranes: 10.8 cm^2	XLE (97.2%) HR95PP (98.8%) TFC-S (100%) NF90 (99.1 %) HL (99.4 %)	*Kosutic et al., 2007*
	CAS effluent (Australia)	100 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 240 ng L^{-1}; RO: 10 ng L^{-1}	*Watkinson et al., 2007*
Norfloxacin	CAS effluent (Australia)	240 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 190 ng L^{-1}; RO: 15 ng L^{-1}	*Watkinson et al., 2007*
Ciprofloxacin	CAS effluent (Australia)	4600 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 170 ng L^{-1}; RO: *nd*	*Watkinson et al., 2007*
Nalidixic acid	CAS effluent (Australia)	200 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 260 ng L^{-1}; RO: 75 ng L^{-1}	*Watkinson et al., 2007*
Trimethoprim					
	Model wastewater for veterinary use (Croatia)	10 mg L^{-1}	RO membranes: XLE (Dow/FilmTec, Midland MI); HR95PP (Dow/FilmTec, Midland MI); TFC-S (Koch Membrane Systems, Wilmington, MA). NF membranes: NF90 (Dow/FilmTec); HL Desal, Osmonics (GE Infrastructure Water Process Techn.,Vista, CA). Surface area of membranes: 10.8 cm^2	XLE (98.6%) HR95PP (98.2%) TFC-S (100%) NF90 (99.2 %) HL (88.8 %)	*Kosutic et al., 2007*
	Missouri River water (Jefferson City)	50 μg L^{-1}	Barnstead RO system: Model D2716, Cellulose acetate membrane D2731, Flow: 1.9 L min^{-1}.	(90.3%)	*Adams et al., 2002*
	CAS effluent (Australia)	930 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 85 ng L^{-1}; RO: 10 ng L^{-1}	*Watkinson et al., 2007*
	Secondary effluent (Beijing, China)	400 ng L^{-}	UF: Dead-end ultrafiltration system (Zenon GE), 6 trains of Zee-Weed 1000 membrane, pore size of 0.02 μm (PVDF), flow=23 L $(m^2\ h)^{-1}$ MF/RO: Spiral-wound cross flow module (Filmtec, DOW).	UF (0-50%) MF/RO (>90%)	*Sui et al., 2010*
Tetracyclines					
Oxytetracycline	Model wastewater for veterinary use (Croatia)	10 mg L^{-1}	RO membranes: XLE (Dow/FilmTec, Midland MI); HR95PP (Dow/FilmTec, Midland MI); TFC-S (Koch Membrane Systems, Wilmington, MA). NF membranes: NF90 (Dow/FilmTec); HL Desal, Osmonics (GE Infrastructure Water Process Techn,Vista, CA). Surface area of membranes: 10.8 cm^2	XLE (99.2%) HR95PP (99.3%) TFC-S (100%) NF90 (99.0 %) HL (99.2 %)	*Kosutic et al., 2007*
	Waste liquor from the crystallization unit in a	1000 mg L^{-1}	RO: SEPA CELL flat sheet membrane apparatus; membrane	< 80 mg L^{-1} (>92%)	*Li et al., 2004*

	pharmaceutical company (Chi Feng, Inner Mongolia, China).		area of 155 cm^2. UF: 0.3 MPa; UF membranes of different molecular weight cut-off (3,10, 30, 50 K Da)		
Lincosamides					
Clindamycin	CAS effluent (Australia)	5 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 10 ng L^{-1} RO: 5 ng L^{-1}	*Watkinson et al., 2007*
Lincomycin	CAS effluent (Australia)	80 ng L^{-1}	MF/RO plant: receives ~10% of CAS effluent	MF: 35 ng L^{-1} RO: 1 ng L^{-1}	*Watkinson et al., 2007*
Advanced treatment process **Antibiotic Group**	**ACTIVATED CARBON ADSORPTION**				
	Type of wastewater (location)	**Initial concentration**	**Treatment process**	**Results/findings (Removal efficiency)**	**Reference**
β-Lactams					
Amoxicillin	Real wastewater (P.T. Coronet Crown)	317 mg L^{-1}	GAC: BET surface area=1092.951 m^2 g^{-1}, pore size < 20A°, dose: 1.5 g per 50 mL solvent	16.9 mg L^{-1} (94.67%)	*Putra et al., 2009*
Penicillin G	*Na*	50-1000 mg L^{-1}	HCI washed PAC: particle size <0.15 mm, BET surface area=1000 m^2 g^{-1}, bulk density=0.46. 0.1 g PAC was treated with 100 ml of PG at a defined pH, temperature and initial PG concentration	$Adsorption_{MAX}$: 375.0 mg g^{-1} (pH: 6.0, 35 ◦C) adsorption (%): 44.0-290.0 (25 ◦C) 39.6-64.4 (35 ◦C) 24.6-51.6 (45 ◦C)	*Aksu and Tunc, 2005*
Macrolides					
Azithromycin	Hospital wastewater after treatment with MBR	110 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (20%) PAC dose=23 mg L^{-1} (100%) PAC dose=43 mg L^{-1} (100%)	*McArdell et al. 2011*
Clarithromycin	Hospital wastewater after treatment with MBR	1280 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (100%) PAC dose=23 mg L^{-1} (100%) PAC dose=43 mg L^{-1} (100%)	*McArdell et al. 2011*
Roxithromycin	Membrane bioreactor operating in a sequential mode (SMBR)	4.5-6 μg L^{-1}	PAC QP: 1.665 g cm^3 real density; 0.25 g cm^3 apparent density; 328.2 m^2 g^{-1} specific surface area.	PAC dose=1 g L^{-1} (71-86%)	*Serrano et al., 2011*
Erythromycin	Membrane bioreactor operating in a sequential mode (SMBR)	6.5-8.5 μg L^{-1}	PAC QP: 1.665 g cm^3 real density; 0.25 g cm^3 apparent density; 328.2 m^2 g^{-1} specific surface area.	PAC dose=1 g L^{-1} (42-64%)	*Serrano et al., 2011*
Erythromycin–H_2O	Four matrices: Colorado River from Lake Mead; Ohio River near Louisville; Passaic River near Totowa; Model water.	*na*	Two PACs: AC800 (Acticarb, Dunnellon, FL) and WPM (Calgon Carbon Corp., Pittsburgh, PA). Contact time=4 h; AC dose=1-20 mg L^{-1}	AC800 dose=5 mg L^{-1} (20%)	*Westerhoff et al., 2005*
Erythromycin & Erythromycin-H_2O	Hospital wastewater after treatment with MBR	10 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (>95%) PAC dose=23 mg L^{-1} (>88%) PAC dose=43 mg L^{-1} (>88%)	*McArdell et al. 2011*
Sulfonamides					

Sulfamethoxazole	Hospital wastewater after treatment with MBR	3230 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (2%) PAC dose=23 mg L^{-1} (33%) PAC dose=43 mg L^{-1} (62%)	*McArdell et al. 2011*
	Four matrices: Colorado River from Lake Mead; Ohio River near Louisville; Passaic River near Totowa; Model water.	*na*	Two PACs: AC800 (Acticarb, Dunnellon, FL) and WPM (Calgon Carbon Corp., Pittsburgh, PA). Contact time=4 h; AC dose=1-20 mg L^{-1}	AC800 dose=5 mg L^{-1} (20%)	*Westerhoff et al., 2005*
Sulfamethazine	Missouri River water (Jefferson City).	50 µg L^{-1}	PAC dose=0-50 mg L^{-1}; Contact time=4 h	AC dose=10 mg L^{-1} (49%) AC dose=20 mg L^{-1} (85%) AC dose=50 mg L^{-1} (>90%)	*Adams et al., 2002*
Sulfathiazole	Missouri River water (Jefferson City).	50 µg L^{-1}	PAC dose=0-50 mg L^{-1}; Contact time=4 h	AC dose=10 mg L^{-1} (70%) AC dose=20 mg L^{-1} (85%) AC dose: 50 mg L^{-1} (>90%)	*Adams et al., 2002*
Sulfamerazine	Missouri River water (Jefferson City).	50 µg L^{-1}	PAC dose=0-50 mg L^{-1}; Contact time=4 h	AC dose=10 mg L^{-1} (60%) AC dose=20 mg L^{-1} (80%) AC dose=50 mg L^{-1} (>90%)	*Adams et al., 2002*
Sulfachloropyridazine	Missouri River water (Jefferson City).	50 µg L^{-1}	PAC dose=0-50 mg L^{-1}; Contact time=4 h	AC dose=10 mg L^{-1} (58%) AC dose=20 mg L^{-1} (75%) AC dose=50 mg L^{-1} (>90%)	*Adams et al., 2002*
Sulfadimethoxine	Missouri River water (Jefferson City).	50 µg L^{-1}	PAC dose=0-50 mg L^{-1}; Contact time=4 h	AC dose=10 mg L^{-1} (50%) AC dose=20 mg L^{-1} (80%) AC dose=50 mg L^{-1} (>90%)	*Adams et al., 2002*
Sulfadiazine	Hospital wastewater after treatment with MBR	2330 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (0%) PAC dose=23 mg L^{-1} (40%) PAC dose=43 mg L^{-1} (>40%)	*McArdell et al. 2011*
Sulfapyridine	Hospital wastewater after treatment with MBR	251 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (85%) PAC dose=23 mg L^{-1} (95%) PAC dose=43 mg L^{-1} (>95%)	*McArdell et al. 2011*
Quinolones					
Ciprofloxacin	Hospital wastewater after treatment with MBR	15700 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (100%) PAC dose=23 mg L^{-1} (>99%) PAC dose=43 mg L^{-1} (>99%)	*McArdell et al. 2011*
Norfloxacin	Hospital wastewater after treatment with MBR	3140 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (99%) PAC dose=23 mg L^{-1} (>99%) PAC dose=43 mg L^{-1} (>99%)	*McArdell et al. 2011*
Trimethoprim					
	Four matrices: Colorado River from Lake Mead; Ohio River near Louisville; Passaic River near Totowa; Model water.	*na*	Two PACs: AC800 (Acticarb, Dunnellon, FL) and WPM (Calgon Carbon Corp., Pittsburgh, PA). Contact time=4 h; AC dose=1-20 mg L^{-1}	AC800 dose= 5 mg L^{-1} (93%)	*Westerhoff et al., 2005*
	Missouri River water (Jefferson City).	50 µg L^{-1}	PAC dose=0-50 mg L^{-1}; Contact time=4 h	AC dose=10 mg L^{-1} (55%) AC dose=20 mg L^{-1} (65%) AC dose=50 mg L^{-1} (>90%)	*Adams et al., 2002)*
	Hospital wastewater after treatment with MBR	37 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=23 mg L^{-1} (>83%) PAC dose=43 mg L^{-1} (>83%)	*McArdell et al. 2011*
Tetracyclines					
Tetracycline	*na*	*na*	Four carbonaceous adsorbents: Single walled carbon	Adsorption efficiency: G/SWNT	*Ji et al., 2009*

			nanotubes (SWNT); Multi-walled carbon nanotubes (MWNT); Pulverized activated carbon (AC) and nonporous Graphite (G).	> MWNT >> AC	
Nitroimidazoles					
Metronidazole	Hospital wastewater after treatment with MBR	1860 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (3%) PAC dose=23 mg L^{-1} (67%) PAC dose=43 mg L^{-1} (78%)	*McArdell et al. 2011*
Metronidazole	Motril (Granada)	100-600 mg L^{-1}	Three activated carbons (0.1 g): Sorbo (S); Merck (M) and carbon prepared by chemical activation of petroleum coke with KOH (C). S (BET=1225 m^2 g^{-1}); M (BET=1301 m^2 g^{-1}); C (BET=848 m^2 g^{-1})	Adsorption capacity S: 1.92 mmol g^{-1} M: 1.25 mmol g^{-1} C: 1.68 mmol g^{-1}	*Rivera-Utrilla et al., 2009*
Dimetridazole	Motril (Granada)	100-600 mg L^{-1}	Three activated carbons (0.1 g): Sorbo (S); Merck (M) and carbon prepared by chemical activation of petroleum coke with KOH (C). S (BET=1225 m^2 g^{-1}); M (BET=1301 m^2 g^{-1}); C (BET=848 m^2 g^{-1})	Adsorption capacity S: 1.99 mmol g^{-1} M: 1.32 mmol g^{-1} C: 2.04 mmol g^{-1}	*Rivera-Utrilla et al., 2009*
Tinidazole	Motril (Granada)	100-600 mg L^{-1}	Three activated carbons (0.1 g): Sorbo (S); Merck (M) and carbon prepared by chemical activation of petroleum coke with KOH (C). S (BET=1225 m^2 g^{-1}); M (BET=1301 m^2 g^{-1}); C (BET=848 m^2 g^{-1})	Adsorption capacity S: 1.37 mmol g^{-1} M: 1.56 mmol g^{-1} C: 1.04 mmol g^{-1}	*Rivera-Utrilla et al., 2009*
Ronidazole	Motril (Granada)	100-600 mg L^{-1}	Three activated carbons (0.1 g): Sorbo (S); Merck (M) and carbon prepared by chemical activation of petroleum coke with KOH (C). S (BET=1225 m^2 g^{-1}); M (BET=1301 m^2 g^{-1}); C (BET=848 m^2 g^{-1})	Adsorption capacity S: 1.97 mmol g^{-1} M: 1.82 mmol g^{-1} C: 1.89 mmol g^{-1}	*Rivera-Utrilla et al., 2009*
Clindamycin	Hospital wastewater after treatment with MBR	1160 ng L^{-1}	PAC Norit SAE Super, PAC retention time=2 days, dose=8-43 mg L^{-1}, contact time=3-5 days	PAC dose=8 mg L^{-1} (96%) PAC dose=23 mg L^{-1} (>99%) PAC dose=43 mg L^{-1} (100%)	*McArdell et al. 2011*
Advanced treatment process **Antibiotic Group**	**OZONATION** **Type of wastewater (location)**	**Initial concentration**	**Treatment process**	**Results/findings (Removal efficiency)**	**Reference**
β-Lactams					
Cephalexin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=3 mg L^{-1} (100%)	*Dodd et al., 2006*
Penicillin	Antibiotic formulation effluent (Turkey)	*na*	O_3 dose =2500 mg $(L\ h)^{-1}$; pH=2.5–12.0 O_3+H_2O_2 [H_2O_2]=2-40 mM); pH=10.5	COD removal O_3: (10-56%) O_3+H_2O_2 (20 mM): (83%)	*Arslan Alaton et al., 2004*
	Antibiotic formulation effluent (Turkey)	*na*	O_3 dose =2760 mg $(L\ h)^{-1}$; pH=3-11.5	COD removal O_3/pH 3: (15%) O_3/pH 7: (28%) O_3/pH 11: (49%) TOC removal O_3/pH 3: (2%) O_3/pH 7: (23%)	*Arslan Alaton and Dogruel, 2004*

				O_3/pH 11: (52%)	
Penicillin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=5 mg L^{-1} (100%)	*Dodd et al., 2006*
Penicillin V	Synthetic wastewater (Turkey)	*na*	(a) O_3 (flow=100 L h^{-1}, O_3 dose=2.96 g L^{-1} h^{-1}); (b) O_3/H_2O_2 ($[H_2O_2]$=20 mM)	(a) (80% in 60 min) (b) (100% in 60 min)	*Balcioglu and Otker, 2003*
Ceftriaxone	Synthetic wastewater (Turkey)	*na*	(a) O_3 (flow=100 L h^{-1}, O_3 dose=2.96 g L^{-1} h^{-1}); (b) O_3/H_2O_2 ($[H_2O_2]$=20 mM)	(a) (>99% in 60 min) (b) (100% in 60 min)	*Balcioglu and Otker, 2003*
Macrolides					
Roxithromycin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=1 mg L^{-1} (55%)	*Dodd et al., 2006*
	CAS and MBR effluent (Kloten-Opfikon, Switzerland)	2 μg L^{-1}	O_3 dose=0-5 mg L^{-1}; flow=200 ±10 L h^{-1} (only column 1).	O_3 dose ≥ 2 mg L^{-1} (≥90%)	*Huber et al., 2005*
	Secondary effluent (German)	0.54±0.04 μg L^{-1}	Ozonation-UV treatment plant O_3=100 g h^{-1}, O_3 dose= 5-15 mg L^{-1}, 2 diffuser/PVC bubble columns	O_3 dose= 5-15 mg L^{-1} (≥ 91%)	*Ternes et al., 2003*
	Secondary wastewater effluent (Spain)	*na*	Batch experiments, O_3 flow=35 L h^{-1}, O_3 dose=20 mg L^{-1}.	(100%)	*Radjenovic et al., 2009*
	CAS and sand filtration (Tokyo)	27.2 ng L^{-1}	O_3 dose=3 mg L^{-1}, Retention time=27 min	(90.9%)	*Nakada et al., 2007*
	CAS effluent (Regensdorf, Switzerland)	9 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g-1 DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.40 g g-1DOC (77%) O_3 dose= 0.62 g g-1DOC (80%)	*Hollender et al., 2009*
Azithromycin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose: 1 mg L^{-1} (62%)	*Dodd et al., 2006*
	CAS and sand filtration (Tokyo)	*nd*	O_3 dose=3 mg L^{-1}, Retention time=27 min	(92.6%)	*Nakada et al., 2007*
	CAS effluent (Alcala de Henares, Madrid)	235 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<50 μM (100%)	*Rosal et al., 2010*
	CAS effluent (Regensdorf, Switzerland)	100 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g-1 DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.61 g g-1DOC (>99%)	*Hollender et al., 2009*
Tylosin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=3 mg L^{-1} (100%)	*Dodd et al., 2006*
	Pharmaceutical effluent (Taiwan)	40 mg L^{-1}	O_3/O_2 mixture, O_3 dose(v/v)=5.3%, flow=1.6 L min^{-1}.	(>99%)	*Lin et al., 2009b*
Clarithromycin	Secondary effluent (German)	0.21±0.02 μg L^{-1}	Ozonation-UV treatment plant O_3=100 g h^{-1}, O_3 dose= 5-15 mg L^{-1}, 2 diffuser/PVC bubble columns	O_3 dose=5-15 mg L^{-1} (≥ 76%)	*Ternes et al., 2003*
	CAS and sand filtration (Tokyo)	228 ng L^{-1}	O_3 dose=3 mg L^{-1}, Retention time=27 min	(84.69%)	*Nakada et al., 2007*
	CAS effluent (Alcala de Henares, Madrid)	39 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<50 μM (100%)	*Rosal et al., 2010*
	CAS effluent (Regensdorf, Switzerland)	206 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g-1 DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.40 g g-1DOC (94%) O_3 dose= 0.62 g g-1DOC (97%) O_3 dose= 0.79 g g-1DOC (99%)	*Hollender et al., 2009*
Erythromycin	Secondary effluent (German)	0.62±0.24 μg L^{-1}	Ozonation-UV treatment plant O_3=100 g h^{-1}, O_3 dose= 5-15 mg L^{-1}, 2 diffuser/PVC bubble columns	O_3 dose =5-15 mg L^{-1} (≥ 92%)	*Ternes et al., 2003*
	CAS effluent (Alcala de Henares, Madrid)	72 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<90 μM (100%)	*Rosal et al., 2010*
	Pharmaceutical effluent (Taiwan)	40 mg L^{-1}	O_3/O_2 mixture, O_3 dose(v/v)=5.3%, flow=1.6 L min^{-1}.	(>99%)	*Lin et al., 2009b*
	CAS effluent (Regensdorf, Switzerland)	36 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g-1 DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.61 g g-1DOC (>64%)	*Hollender et al., 2009*
Erythromycin-H_2O	CAS and sand filtration (Tokyo)	150 ng L^{-1}	O_3 dose=3 mg L^{-1}, Retention time=27 min	(88.7%)	*Nakada et al., 2007*
Sulfonamides					

Sulfamethoxazole	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=3 mg L^{-1} (100%)	*Dodd et al., 2006*
	CAS and MBR effluent (Kloten-Opfikon, Switzerland)	2 μg L^{-1}	O_3 dose=0-5 mg L^{-1}; flow=200 ±10 L h^{-1} (only column 1).	O_3 dose ≥ 2 mg L^{-1} (≥ 90%)	*Huber et al., 2005*
	CAS and sand filtration (Tokyo)	104 ng L^{-1}	O_3 dose=3 mg L^{-1}, Retention time=27 min	(87.4%)	*Nakada et al., 2007*
	CAS effluent (Alcala de Henares, Madrid)	95 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<220 μM (100%)	*Rosal et al., 2010*
	Pharmaceutical effluent (Taiwan)	40 mg L^{-1}	O_3/O_2 mixture, O_3 dose(v/v)=5.3%, flow=1.6 L min^{-1}.	(93%)	*Lin et al., 2009b*
	CAS effluent (Regensdorf, Switzerland)	197 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g-1 DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.40 g g-1DOC (87%) O_3 dose= 0.62 g g-1DOC (96%) O_3 dose= 0.79 g g-1DOC (96%)	*Hollender et al., 2009*
Sulfamethazine	Secondary effluent (German)	0.62±0.05 μg L^{-1}	Ozonation-UV treatment plant O_3=100 g h^{-1}, O_3 dose= 5-15 mg L^{-1}, 2 diffuser/PVC bubble columns.	O_3 dose =5-15 mg L^{-1} (≥ 92%)	*Ternes et al., 2003*
	Missouri River water (Jefferson City)	50 μg L^{-1}	O_3 dose=7.1 mg L^{-1}	0.3 mg L^{-1} O_3 at 1.3 min) (> 95%)	*Adams et al., 2002*
	Pharmaceutical effluent (Taiwan)	40 mg L^{-1}	O_3/O_2 mixture, O_3 dose(v/v)=5.3%, flow=1.6 L min^{-1}.	(95%)	*Lin et al., 2009b*
Sulfathiazole	Missouri River water (Jefferson City)	50 μg L^{-1}	O_3 dose=7.1 mg L^{-1}	0.3 mg L^{-1} O_3 at 1.3 min) (> 95%)	*Adams et al., 2002*
Sulfamerazine	Missouri River water (Jefferson City)	50 μg L^{-1}	O_3 dose=7.1 mg L^{-1}	0.3 mg L^{-1} O_3 at 1.3 min) (> 95%)	*Adams et al., 2002*
Sulfachloropyridazine	Missouri River water (Jefferson City)	50 μg L^{-1}	O_3 dose=7.1 mg L^{-1}	0.3 mg L^{-1} O_3 at 1.3 min) (> 95%)	*Adams et al., 2002*
Sulfadimethoxine	Missouri River water (Jefferson City)	50 μg L^{-1}	O_3 dose=7.1 mg L^{-1}	0.3 mg L^{-1} O_3 at 1.3 min) (> 95%)	*Adams et al., 2002*
	Pharmaceutical effluent (Taiwan)	40 mg L^{-1}	O_3/O_2 mixture, O_3 dose(v/v)=5.3%, flow=1.6 L min^{-1}.	(96%)	*Lin et al., 2009b*
Sulfapyridine	CAS and sand filtration (Tokyo)	492 ng L^{-1}	O_3 dose=3 mg L^{-1}, Retention time=27 min	(93.9%)	*Nakada et al., 2007*
	CAS effluent (Alcala de Henares, Madrid)	50 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<50 μM (100%)	*Rosal et al., 2010*
	CAS effluent (Regensdorf, Switzerland)	125 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g-1 DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.40 g g-1DOC (98%) O_3 dose= 0.62 g g-1DOC (97%) O_3 dose= 0.79 g g-1DOC (97%)	*Hollender et al., 2009*
Quinolones					
Norfloxacin	CAS effluent (Alcala de Henares, Madrid)	38 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<90 μM (100%)	*Rosal et al., 2010*
Ciprofloxacin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=3 mg L^{-1} (100%)	*Dodd et al., 2006*
	CAS effluent (Alcala de Henares, Madrid)	522 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<130 μM (100%)	*Rosal et al., 2010*
Enrofloxacin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=3 mg L^{-1} (100%)	*Dodd et al., 2006*
Trimethoprim					
	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=3 mg L^{-1} (100%)	*Dodd et al., 2006*
	Secondary effluent (German)	0.34±0.04 μg L^{-1}	Ozonation-UV treatment plant O_3=100 g h^{-1}, O_3 dose= 5-15 mg L^{-1}, 2 diffuser/PVC bubble columns	O_3 dose : 5- 15 mg L^{-1} (≥ 85%)	*Ternes et al., 2003*
	Secondary wastewater effluent (Spain)	*na*	Batch experiments, O_3 flow=35 L h^{-1}, O_3 dose=20 mg L^{-1}.	100%	*Radjenovic et al., 2009*
	Missouri River water (Jefferson City)	50 μg L^{-1}	O_3 dose=7.1 mg L^{-1}	0.3 mg L^{-1} O_3 at 1.3 min) (> 95%)	*Adams et al., 2002*
	CAS and sand filtration (Tokyo)	53.5 ng L^{-1}	O_3 dose=3 mg L^{-1}, Retention time=27 min	(96%)	*Nakada et al., 2007*
	CAS effluent (Alcala de Henares, Madrid)	73 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<90 μM (100%)	*Rosal et al., 2010*
	WWTPs in Beijing (China)	400 ng L^{-1}	O_3 dose=5 mg L^{-1}; Contact time=15 min MF/RO: Spiral-wound crossflow module	(>90%)	*Sui et al., 2010*

	CAS effluent (Regensdorf, Switzerland)	119 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g^{-1} DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.40 g g-1DOC (97%) O_3 dose= 0.62 g g^{-1}DOC (95%) O_3 dose= 0.79 g g^{-1}DOC (93%)	*Hollender et al., 2009*
Tetracyclines					
Tetracycline	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose=1.5 mg L^{-1} (100%)	*Dodd et al., 2006*
Lincosamides					
Lincomycin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose:=1 mg L^{-1} (70%)	*Dodd et al., 2006*
	CAS effluent (Alcala de Henares, Madrid)	12 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose<50 μM (100%)	*Rosal et al., 2010*
Clindamycin	CAS effluent (Regensdorf, Switzerland)	36 ng L^{-1}	O_3 dose= 1.6-5.3 mg L^{-1} (0.36-1.16 g g^{-1} DOC), Retention time=8-15 min, full scale six compartment reactor	O_3 dose= 0.40 g g^{-1}DOC (95%) O_3 dose= 0.62 g g^{-1}DOC (94%) O_3 dose= 0.79 g g^{-1}DOC (91%)	*Hollender et al., 2009*
Aminoglycosides					
Amikacin	Secondary effluent (Kloten-Opfikon, Switzerland)	1 μM	Batch experiments, O_3 dose=0.5-5.0 mg L^{-1} DOC=5.3 mg L^{-1}	O_3 dose: 1 mg L^{-1} (25%)	*Dodd et al., 2006*
Advanced treatment process	**FENTON OXIDATION**				
Antibiotic Group	**Type of wastewater (location)**	**Initial concentration**	**Treatment process**	**Results/findings (Removal efficiency)**	**Reference**
β-Lactams					
Amoxicillin	Wastewater from plant manufacturing AMX (China)	*Na*	Fenton oxidation after extraction (dichloromethane) [$FeSO_4.7H_2O$]=10 g L^{-1}; [H_2O_2]=2 g L^{-1} TOC=18925 mg L^{-1} COD=80000 mg L^{-1}	TOC=2195.3 mg L^{-1} (88.4%) COD=832 mg L^{-1} (89.6%)	*Zhang et al., 2006*
	CAS effluent (Araraquara, Brazil)	42 mg L^{-1}	Black light at 365 nm and solar irradiation [H_2O_2]=2.0 mM [Ferrioxalate or $Fe(NO_3)_3$]=0.20 mM pH=2.5	Black light: (89% in 1 min) Solar light: (85% in 1 min) AMX degradation was not influenced by the source of the irradiation.	*Trovo et al., 2008*
Penicillin	Antibiotic formulation effluent (Turkey)	*na*	UV light (λ=253.7 nm, 1.73×10^{-4} Einstein $(Ls)^{-1}$); 60 min; pH=3; [H_2O_2]=20 mM; [Fe(II)]=1 mM; [Fe(III)]=1 mM.	COD removal Photo-Fenton: (56%) Photo-Fenton-like: (66%) Dark Fenton: (61%) Dark Fenton-like: (46%) TOC removal Photo-Fenton: (51%) Photo-Fenton-like: (42%) Dark Fenton: (33%) Dark Fenton-like: (18%)	*Arslan Alaton and Dogruel, 2004*
	Pharmaceutical wastewater (China) COD=49912.5 mg L^{-1}; TOC=11540 mg L^{-1}	*na*	Microwave power=100-500 W; radiation time=2-10 min; pH=1-11; [H_2O_2]=3200-19000 mg L^{-1}; [$Fe_2(SO_4)_3$]=2000-8000 mg L^{-1}	Optimum conditions: Microwave power=300 W; radiation time=6 min; pH=4.42; [H_2O_2]=1300 mg L^{-1}; [$Fe_2(SO_4)_3$]=4900 mg L^{-1} COD removal: (57.53%) TOC removal: (>40%)	*Yang et al., 2009*

				Degradation: (55.06%)	
Quinolones					
Ofloxacin	Secondary effluent (Almería, Spain)	100 μg L^{-1}	Pilot compound parabolic collector plant (CPC), $[Fe^{2+}]$= 5 mg L^{-1}, $[H_2O_2]$= 50 mg L^{-1}, t_{30W}=102 min	(100%)	*Klamerth et al., 2010*
	(Lemessos, Cyprus)	10 mg L^{-1} (0.0277 mmol L^{-1})	Batch experiments (300 mL), solar simulator (1 kW Xenon lamp) $[Fe^{2+}]$= 1-5 mg L^{-1}, $[H_2O_2]$= 1.357-8.142 mmol L^{-1}	$[Fe^{2+}]$= 5 mg L^{-1}, $[H_2O_2]$= 2.714 mmol L^{-1} (100% at 30 min)	*Michael et al., 2010*
	Secondary effluent (Cyprus)	100 μg L^{-1}	Pilot scale experiments $[Fe^{2+}]$= 5 mg L^{-1}, $[H_2O_2]$= 75 mg L^{-1}, $t_{30WT,n}$=38.7 min	(100%)	*Michael et al., 2012b*
Trimethoprim					
	Simulated effluent from municipal wastewater treatment plant (SWW) and pre-treated real effluent from municipal wastewater treatment plant (RE) (Almería, Spain)	10 mg L^{-1}	Pilot compound parabolic collector plant (CPC), $[Fe^{2+}]$= 2 mg L^{-1}, $[H_2O_2]$= 2.5 mg L^{-1} (in doses). SWW: DOC=25 mg L^{-1} RE: DOC=10 mg L^{-1}	100 %	*Michael et al., 2012a*
	Secondary effluent (Cyprus)	100 μg L^{-1}	Pilot scale experiments $[Fe^{2+}]$= 5 mg L^{-1}, $[H_2O_2]$= 75 mg L^{-1}, $t_{30WT,n}$=20.1 min	(100%)	*Michael et al., 2012b*
Tetracyclines					
Tetracycline	CAS effluent (Araraquara, Brazil)	24 mg L^{-1}	Black light (15 W) and solar irradiation $[H_2O_2]$= 1-10 mM [Ferrioxalate or $Fe(NO_3)_3$]=0.20 mM pH=2.5	Black light: (80% in 3 min) Solar light: (80% in 3 min)	*Bautitz and Nogueira, 2007*
Advanced treatment process **Antibiotic Group**	**HETEROGENEOUS PHOTOCATALYSIS WITH TiO_2**				
	Type of wastewater (location)	**Initial concentration**	**Treatment process**	**Results/findings (Removal efficiency)**	**Reference**
β-Lactams					
Amoxicillin	Antibiotic wastewater (AW)	138±5 mg L^{-1}	UV/H_2O_2/TiO_2 2000 mL of AW; $[TiO_2]$=0-1000 mg L^{-1}; $[H_2O_2]$=50-350 mg L^{-1}; T=22±2 ºC UV lamp (6 W, λ≈365 nm) UV/H_2O_2/TiO_2/SBR 1.5 L of AW; 65 days at HRT 24 hr	UV/H_2O_2/TiO_2 $[TiO_2]$=1000 mg L^{-1} $[H_2O_2]$=250 mg L^{-1} 30 min, pH=5 (100%) UV/H_2O_2/TiO_2/SBR $[TiO_2]$=1000 mg L^{-1} $[H_2O_2]$=250 mg L^{-1} (57 % of COD); (53% of DOC)	*Elmolla and Chaudhuri, 2011*
	CAS effluent (Salerno, Italy)	10 mg L^{-1}	Batch experiments (300 mL), 125W black light fluorescent lamp (300-420 nm; photon flux= 4.7×10^{-7}einstein s^{-1}) TiO_2 Degussa P25, $[TiO_2]$=0.2-0.8 g L^{-1}	120 min, $[TiO_2]$=0.8 g L^{-1} (100%)	*Rizzo et al., 2009*
Cloxacillin	Antibiotic wastewater (AW)	138±5 mg L^{-1}	UV/H_2O_2/TiO_2 2000 mL of AW; $[TiO_2]$=0-1000 mg L^{-1}; $[H_2O_2]$=50-350 mg L^{-1}; T=22±2 ºC UV lamp (6 W, λ≈365 nm)	UV/H_2O_2/TiO_2 $[TiO_2]$=1000 mg L^{-1} $[H_2O_2]$=250 mg L^{-1} 30 min, pH=5	*Elmolla and Chaudhuri, 2011*

			UV/H_2O_2/TiO_2/SBR 1.5 L of AW; 65 days at HRT 24 hr	(100%) UV/H_2O_2/TiO_2/SBR [TiO_2]=1000 mg L^{-1} [H_2O_2]=250 mg L^{-1} (57 % of COD); (53% of DOC)	
Sulfomanides					
Sulfamethoxazole	Final effluent (Lemessos, Cyprus)	10 mg L^{-1}	Batch experiments (350 mL), 9W UVA lamp (Radium Ralutec, 9W/78, 350-400 nm), photon flux= 2.81×10^{-4} einstein min^{-1}. TiO_2 Degussa P25, [TiO_2]=500 mg L^{-1}	~20 min, pH=4.8<pH<5.6 (100%) 60 min, pH=7.5<pH<8.2 (>99%)	*Xekoukoulotakis et al., 2010*
Quinolones					
Ofloxacin	Final effluent (Lemessos, Cyprus)	10 mg L^{-1}	Batch experiments (350 mL), 9W UVA lamp (Radium Ralutec, 9W/78, 350-400 nm), photon flux= 3.37×10^{-6} einstein s^{-1}. TiO_2 Degussa P25, [TiO_2]=250 mg L^{-1}, [H_2O_2]= 0.14 mmol L^{-1}	~85% (Degussa P25; 250 mg L^{-1}; 30 min) ≈Hombicat UV 100 (83%) > Aldrich (73%) > Tronox A-K-1 (67%)> Tronox TR-HP-2 (39%)> Tronox TR(33%) [H_2O_2]=0.07 mmol L^{-1} [TiO_2]=250 mg L^{-1} (79% of DOC)	*Hapeshi et al., 2010*
	Secondary effluent (Lemessos, Cyprus)	10 mg L^{-1} (0.0277 mmol L^{-1}),	Batch experiments (300 mL), solar simulator (1 kW Xenon lamp) TiO_2 Degussa P25, [TiO_2]= 0.25-4.0 g L^{-1}, [H_2O_2]= 1.357-8.142 mmol L^{-1}	[TiO_2]= 3 g L^{-1}, 120 min (60%) [TiO_2]= 3 g L^{-1}, [H_2O_2]= 5.428 mmol L^{-1}, 120 min (67%)	*Michael et al., 2010*
Advanced treatment process **Antibiotic Group**	**SONOLYSIS** **Type of wastewater (location)**	**Initial concentration**	**Treatment process**	**Results/findings (Removal efficiency)**	**Reference**
β-Lactams					
Amoxicillin	Final effluent before disinfection (Salerno, Italy)	2.5-10.0 mg L^{-1}	Ultrasound generator: 20 kHz, titanium horn (d=1.3 cm), 25-100 W L^{-1}	100 W L^{-1} (~40%)	*Naddeo et al., 2009*
Advanced treatment process **Antibiotic Group**	**PHOTOLYSIS WITH UV** **Type of wastewater (location)**	**Initial concentration**	**Treatment process**	**Results/findings (Removal efficiency)**	**Reference**
β-Lactams					
Amoxicillin	Effluent from Varese UWTP	18 ng L^{-1}	UV-light treatment	(100%)	*Zuccato et al., 2010*
Penicillin	Antibiotic formulation effluent (Turkey)	*na*	UV light (λ=253.7 nm, 1.73×10^{-4} Einstein $(Ls)^{-1}$); 60 min; pH=7; [H_2O_2]=0-40 mM	COD removal UV / pH 7: (0%) UV+H_2O_2 (40 mM) / pH 7: (11%) UV+H_2O_2 (30 mM) / pH 7: (22%) TOC removal	*Arslan Alaton and Dogruel, 2004*

				UV / pH 7: (0%) UV+H_2O_2 (40 mM) / pH 7: (10%) UV+H_2O_2 (30 mM) / pH 7: (6%)	
Macrolides					
Clarithromycin	Effluent from secondary sedimentation and sand filter (Japan)	110-656 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (24-34%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
	Effluent from Varese UWTP	319 ng L^{-1}	UV-light treatment	(0%)	*Zuccato et al., 2010*
Erythromycin	Effluent from secondary sedimentation and sand filter (Japan)	110-656 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (24-34%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
	Effluent from Varese UWTP	12 ng L^{-1}	UV-light treatment	(0%)	*Zuccato et al., 2010*
Azithromycin	Effluent from secondary sedimentation and sand filter (Japan)	110-656 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (24-34%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
Spiramycin	Effluent from Varese UWTP	603 ng L^{-1}	UV-light treatment	(17%)	*Zuccato et al., 2010*
Sulfonamides					
Sulfamethoxazole	Effluent from Blue Lake WWTP; Metro WWTP and Lake Josephine (USA)	1 μM	Photolysis experiments (Suntest CPS + solar simulator with a UV-Suprax optical filter, 765 W m^{-2})	(48%)	*Ryan et al., 2011*
	Effluent from secondary sedimentation and sand filter (Japan)	42-187 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (89-100%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
	Effluent from Varese UWTP	246 ng L^{-1}	UV-light treatment	(0%)	*Zuccato et al., 2010*
Sulfamethazine	Missouri River water (Jefferson City).	50 μg L^{-1}	Mercury vapor lamp (254 nm), UV dose=0-10000 mJ cm^{-2}	UV dose=10000 mJ cm^{-2} (85%)	*Adams et al., 2002*
Sulfathiazole	Missouri River water (Jefferson City).	50 μg L^{-1}	Mercury vapor lamp (254 nm), UV dose=0-10000 mJ cm^{-2}	UV dose= 10000 mJ cm^{-2} (100%)	*Adams et al., 2002*
Sulfamerazine	Missouri River water (Jefferson City).	50 μg L^{-1}	Mercury vapor lamp (254 nm), UV dose=0-10000 mJ cm^{-2}	UV dose=10000 mJ cm^{-2} (83%)	*Adams et al., 2002*
Sulfachlorpyridazine	Missouri River water (Jefferson City).	50 μg L^{-1}	Mercury vapor lamp (254 nm), UV dose=0-10000 mJ cm^{-2}	UV dose: 10000 mJ cm^{-2} (83%)	*Adams et al., 2002*
Sulfadimethoxine	Missouri River water (Jefferson City).	50 μg L^{-1}	Mercury vapor lamp (254 nm), UV dose=0-10000 mJ cm^{-2}	UV dose: 10000 mJ cm^{-2} (85%)	*Adams et al., 2002*
	Effluent from secondary sedimentation and sand filter (Japan)	42-187 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (89-100%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
Trimethoprim					
	Missouri River water (Jefferson City).	50 μg L^{-1}	Mercury vapor lamp (254 nm), UV dose=0-10000 mJ cm^{-2}	UV dose: 10000 mJ cm^{-2} (85%)	*Adams et al., 2002*
	Effluent from Blue Lake WWTP; Metro WWTP and Lake Josephine (USA)	1 μM	Photolysis experiments (Suntest CPS + solar simulator with a UV-Suprax optical filter, 765 W m^{-2})	(18%)	*Ryan et al., 2011*
	Tertiary water from Las Vegas, Nevada (LVNV), Rocky Mountain Region of Colorado (RMCO) and Pinellas County, Florida (PCFL).	38-760 ng L^{-1}	Bench scale UV/H_2O_2: two G15T8 germicidal lamps (General Electric, Fairfield, CT, USA), UV=300-700 mJ cm^{-2}; [H_2O_2]=0-20 mg L^{-1}.	UV dose=300 mJ cm^{-2}; [H_2O_2]=20 mg L^{-1} (21-67%) UV dose=500 mJ cm^{-2}; [H_2O_2]=20 mg L^{-1} (32-92%) UV dose=700 mJ cm^{-2}; [H_2O_2]=20 mg L^{-1} (39-92%)	*Rosario-Ortiz et al., 2010*

Tetracyclines					
Tetracycline	Effluent from secondary sedimentation and sand filter (Japan)	4-17 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (15%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
Oxytetracycline	Secondary wastewater (Beijing, China)	50 μM	11 W low-pressure Hg vapor lamp (λ=254 nm), photon flow= 4.5×10^{-5} E m^{-2} s^{-1}; UV dose=(0-320)×10^2 mJ cm^{-2}; 500 mL WW, [H_2O_2]=1 mM,	UV UV dose=30528 mJ cm^{-2} (100%) UV/H_2O_2 UV dose=7632 mJ cm^{-2} (100%)	*Yuan et al., 2011*
Doxycycline	Secondary wastewater (Beijing, China)	50 μM	11 W low-pressure Hg vapor lamp (λ=254 nm), photon flow= 4.5×10^{-5} E m^{-2} s^{-1}; UV dose=(0-320)×10^2 mJ cm^{-2}; 500 mL WW, [H_2O_2]=1 mM,	UV UV dose=22896 mJ cm^{-2} (100%) UV/H_2O_2 UV dose=7632 mJ cm^{-2} (100%)	*Yuan et al., 2011*
Chlorotetracycline	Effluent from secondary sedimentation and sand filter (Japan)	4-17 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (<6.18 μg L^{-1}) (100%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
Quinolones					
Norfloxacin	Effluent from secondary sedimentation and sand filter (Japan)	4-148 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (86-100%) UV+ H_2O_2: (69%)	*Kim et al., 2009*
Ofloxacin	Effluent from Varese UWTP	463 ng L^{-1}	UV-light treatment	(19%)	*Zuccato et al., 2010*
Ciprofloxacin	Secondary wastewater (Beijing, China)	50 μM	11 W low-pressure Hg vapor lamp (λ=254 nm), photon flow= 4.5×10^{-5} E m^{-2} s^{-1}; UV dose=(0-320)×10^2 mJ cm^{-2}; 500 mL WW, [H_2O_2]=1 mM,	UV UV dose=11448 mJ cm^{-2} (100%) UV/H_2O_2 UV dose=7632 mJ cm^{-2} (100%)	*Yuan et al., 2011*
	Effluent from Varese UWTP	513 ng L^{-1}	UV-light treatment	(0%)	*Zuccato et al., 2010*
Nalidixic acid	Effluent from secondary sedimentation and sand filter (Japan)	4-148 ng L^{-1}	3 UV lamps (λ=254 nm; intensity=1.025 mW cm^{-2}); 3 reactors in series (R_1-R_3); Air flow rate=0.5 L min^{-1}; [H_2O_2]=7.8 mg L^{-1}	UV: (86-100%) UV+ H_2O_2: (>90%)	*Kim et al., 2009*
Lincosamides					
Lincomycin	Effluent from Varese UWTP	9.7 ng L^{-1}	UV-light treatment	(0%)	*Zuccato et al., 2010*
Glycopeptides					
Vancomycin	Effluent from Varese UWTP	41 ng L^{-1}	UV-light treatment	(28%)	*Zuccato et al., 2010*

NOTES

- *nd*: not detected
- *na*: not available
- GAC: Granular activated carbon
- PAC**:** Powdered activated carbon