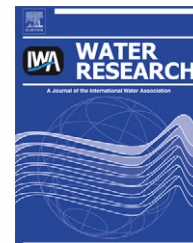




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Review

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.

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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 (Jones et al., 2001; Heberer, 2002; Miao et al., 2004; Batt et al., 2007; Gulkowska et al., 2008; 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; Diaz-Cruz et al., 2003; Brown et al., 2006; Kümmerer, 2009; Czekalski et al., 2012; Le Corre et al., 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 (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 toward *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* (Macrì 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 (*V. fischeri*), a freshwater flea (*D. magna*) and the Japanese medaka fish (*Oryzias latipes*). In this study, *D. 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 UWWTP 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 (ARG) 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-EMA). 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. Between 50 and 5000 $\mu\text{g L}^{-1}$, enrofloxacin induced hormetic effect in plants, with a dose-dependant stimulation or toxicity on the 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 UWWTPs

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

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

2.1. Effect of biological treatment on antibiotics' removal

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

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

Hydrophobic (or non-polar) antibiotic residues are expected to occur at higher concentration in primary and secondary sludge than hydrophilic ones because they have a greater affinity to solids and hence, concentrate in the organic-rich sewage sludge (Le-Minh et al., 2010). Antibiotics can also be removed from aqueous solutions onto solid

particulates by ion exchange, complex formation with metal ions and polar hydrophilic interactions (Diaz-Cruz et al., 2003). Antibiotics that are sorbed to flocs, suspended solids and activated sludge, are removed from the aqueous phase by sedimentation and subsequent disposal of excess sludge. The affinity of antibiotics sorbed to sludge is most often represented by sludge sorption constants K_d ($L\ kg^{-1}$). The higher K_d values the higher sorption of the compounds to sludge. A review on K_d values of several antibiotics is provided in Kovalova et al. (2012). It is important to note that the sludge is often used as fertilizer on agriculture fields, but in several European countries this is forbidden and the sludge is incinerated. Using sludge as fertilizer can therefore be considered as another input pathway for various antibiotics into the environment.

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

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

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

The SRT is related to the growth rate of microorganisms. High SRTs allow the enrichment of slowly growing bacteria and therefore, provide greater diversity of enzymes, some of which are capable of degrading the antibiotic compounds (Jones et al., 2007; Le-Minh et al., 2010). High SRT can be reached with a membrane bioreactor (MBR), where the suspended activated sludge is retained in the reactor by utilizing a membrane for solid/liquid separation instead of a settling tank as used in CAS. Commonly, micro- or ultrafiltration membranes are used in MBRs, which do not retain the antibiotics on the filter. Some studies have been performed to investigate if higher SRTs enhance the elimination of antibiotics, which will be discussed in detail below (Joss et al., 2005; Göbel et al., 2007; Radjenovic et al., 2009b; Tadkaew et al., 2011; Kovalova et al., 2012).

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

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

2.1.1. β -Lactams

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

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%**)	Li et al., 2009
	(<34.4); 77.2–383	CAS + chlorination/nd CAS _{Shatin} (ne) CAS _{Stanley} (82%**)	Li and Zhang, 2011
		Disinfection (91%**)	
		Final (97%**)	
	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%**)	Li et al., 2009
		CAS + chlorination/nd (100%**)	
	658–1718; 65.7–525	CAS _{Shatin} (53%**)	Li and Zhang, 2011
		CAS _{Stanley} (91%**)	
		Disinfection (99%**)	
		Final (100%**)	
Penicillin G	29	na (<LOD**)	Gulkowska et al., 2008
	10	300 (29%*)	Watkinson et al., 2009
Penicillin V	160	Primary/10 (94%*)	Watkinson et al., 2007
		CAS/20 (87.5%*)	
	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%**)	Li and Zhang, 2011
		CAS _{Stanley} (ne)	
		Disinfection (ne)	
		Final (ne)	
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%**)	Göbel et al., 2005;
		CAS/10–30 (–18 to 38%**)	Göbel et al., 2007
		MBR (38, 60, 57%*) [_{SRT 16, 33, 60–80 days}]	
		FBR (~24%*)	
	26–117	CAS/36–69 (<0%*)	Clara et al., 2005
		MBR/(nd, 31, 42) [_{SRT = 10, 27, 55 days}]	
		(100, 52, 64%*)	
	500	500 (0%*)	Watkinson et al., 2009
	3.5–25.3	CAS/14.2 ± 1.1 (43.9%**)	Li et al., 2009
		CAS + chlorination/2.9 ± 0.0 (17.1%**)	
	810 ± 420	540 ± 70 (33%**)	Ternes et al., 2007
	102 ± 32; 164 ± 31;	CAS + chlorination/36 ± 21 (65%*)	Xu et al., 2007
	75 ± 14; 156 ± 29	Oxidation ditch + UV/278 ± 46 (<0%*)	
		CAS/35 ± 8 (53%*)	

(continued on next page)

Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference	
Azithromycin	50 35.6–135; 4.2–141	Chemically enhanced + Chlorination/37 ± 11 (76%*) (40, 60, 55%*) _[STR = 16, 33, 60–80 days] CAS _{Shatin} (46%**) CAS _{Stanley} (40%**) Disinfection (18%**) Final (53%**)	Joss et al., 2005 Li and Zhang, 2011	
	Na 600	<(5)–31 MBR/RO MBR (89.5 ± 7.7%**) _[SRT > 40 days] RO (99.6 ± 0.4%**) CAS-UF/RO UF (81.4 ± 10.1%**) RO (99.9 ± 0.1%**) MBR (>50%**) _[STR > 100 days] (77%**) _[SRT = 44–72 days] MBR (57%**) _[SRT = 15 days] MBR (81%**) _[SRT = 30 days]	McArdell et al., 2003 Sahar et al., 2010	
	500–1000 10 ⁴ 5*10 ⁴	MBR (>50%**) _[STR > 100 days] (77%**) _[SRT = 44–72 days] MBR (57%**) _[SRT = 15 days] MBR (81%**) _[SRT = 30 days]	Abegglen et al., 2009 Reif et al., 2008 Tambosi et al., 2010	
	152 90–380	96 (37%*) Primary/80–320 (10–33%**) CAS/40–380 (–26 to 55%**) MBR (<0, 5, 25%*) _[SRT 16, 33, 60–80 days] FBR (12.5%*)	Gros et al., 2006 Göbel et al., 2005; Göbel et al., 2007	
	4.5–53 1150 _(UWTP I) ; 660 _(UWTP II) ; 1680 _(UWTP III)	4–23 (11–57%*) 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%*)	Loganathan et al., 2009 Fatta et al., 2010	
	139 500–1000 110–142 55	MBR (21%**) _[STR = 30–50 days] MBR (>50%**) _[STR > 100 days] MBR-RO (75%**) _[STR = 45 days] Primary/nd (100%*) CAS/20 (64%*)	Kovalova et al., 2012 Abegglen et al., 2009 Dolar et al., 2012 Watkinson et al., 2007	
	60 1150 ± 70 59–1433 330–660	3400 (<0%*) 60 ± 4 (95%*) 12–32 (99%**) Primary/160–440 (11–14%**) CAS/150–460 (–45 to 20%**) MBR (54, 40, 90%*) _[SRT 16, 33, 60–80 days] FBR (~10%*)	Watkinson et al., 2009 Yang et al., 2004 Lin et al., 2009 Göbel et al., 2005; Göbel et al., 2007	
	319 105.7–724.2 460 ± 100 Na 1500	CAS/117 (13%**) (<LOQ)–610.6 (16%*) 210 ± 40 (54%**) 57–328 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%**) MBR (50%**) _[STR = 30–50 days] MBR (>50%**) _[STR > 100 days] MBR-RO (87%**) _[STR = 45 days] 145–290 (79%**) CAS/52 (0%**) Secondary UWTP I/200 (47%*) UWTP II/250 (11%*) UWTP III/420 (40%*)	Zuccato et al., 2010 Spongberg and Witter, 2008 Ternes et al., 2007 McArdell et al., 2003 Sahar et al., 2010	
	Erythromycin	2555 500–1000 700–2720 71–141 12 380 _(UWTP I) ; 280 _(UWTP II) ; 700 _(UWTP III)	MBR (50%**) _[STR = 30–50 days] MBR (>50%**) _[STR > 100 days] MBR-RO (87%**) _[STR = 45 days] 145–290 (79%**) CAS/52 (0%**) Secondary UWTP I/200 (47%*) UWTP II/250 (11%*) UWTP III/420 (40%*)	Kovalova et al., 2012 Abegglen et al., 2009 Dolar et al., 2012 Roberts and Thomas, 2006 Zuccato et al., 2010 Fatta et al., 2010

Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
Erythromycin-H ₂ O		Outlet	
		UWTP I/30 (92%*)	
		UWTP II/400 (<0*)	
		UWTP III/<LOD (100%*)	
	830 ± 270	620 ± 440 (25%**)	Ternes et al., 2007
	751 ± 109;	CAS + chlorination/430 ± 73 (43%*)	Xu et al., 2007
	1978 ± 233;	Oxidation ditch + UV/2054 ± 386 (<0*)	
	253 ± 22;	CAS/216 ± 34 (15%*)	
	469 ± 38	Chemically enhanced	
		+ chlorination/259 ± 20 (45%*)	
	1000	MBR/RO	Sahar et al., 2010
		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%**)		
32–80	MBR-RO (80%**) _[STR=45 days]	Dolar et al., 2012	
10 ⁴	(91%**) _[SRT=44–72 days]	Reif et al., 2008	
470–810	510–850 (–12 to 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 to 4%**)	Göbel et al., 2005;	
	CAS/50–140 (–22 to 7%**)	Göbel et al., 2007	
	MBR (32, 26, 90%*) _[SRT 16, 33, 60–80 days]		
	FBR (~25%)		
16.7–51.3	CAS/96.3 ± 6.0 (55.6%**)	Li et al., 2009	
	CAS + chlorination/37.9 ± 0.6 (26.1%**)		
200 ± 10	80 ± 5 (60%*)	Yang et al., 2004	
258–409; 169–374	CAS _{Shatin} (15%**)	Li and Zhang, 2011	
	CAS _{Stanley} (26%**)		
	Disinfection (24%**)		
	Final (43%**)		
na	<(20)–199	McArdell et al., 2003	
820	CAS (35.4 ± 50.5%**)	Radjenovic et al., 2009b	
	MBR HF-UF (25.2 ± 108.9%**) _[SRT > 60 days]		
	MBR FS-MF (43.0 ± 51.5%**) _[SRT > 60 days]		
188	MBR (<60%**) _[STR=30–50 days]	Kovalova et al., 2012	
242–6755;	Trickling filter beds/292–2841 (0%**)	Kasprzyk-Hordern et al., 2009	
144–10025	CAS/23–2772 (50%**)		
603	CAS/454 (25%**)	Zuccato et al., 2010	
Spiramycin			
Sulfonamides			
Sulfamethoxazole			
	500	Primary/570 (<0%*)	Watkinson et al., 2007
		CAS/200 (60%*)	
	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	Clara et al., 2005
		MBR/(56, nd, nd) _[SRT = 10, 27, 55 days]	
		(61, 100, 100%*)	
	5450 _(GZ-UWTP1) ;	GZ-UWTP ₁	Peng et al., 2006
	7910 _(GZ-UWTP2)	Primary/9460 (<0*)	
		Secondary/nq	
		Tertiary/nd	
		GZ-UWTP ₂	
		Primary/nq	
		Secondary/nq	
		Tertiary/nd	
	(<80)–674	(<80)–304 (42%**)	Lindberg et al., 2005
	20	70 (<0**)	Bendz et al., 2005

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Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
	(<50)–1250 250–640 230–570	(<50)–370 (17.8–100% ^{**}) 250 (67% ^{**}) Primary/90–640 (–21 to (–5)% ^{**}) Secondary/130–840 (–138 to 60% ^{**}) MBR (38, 40, 37% [*]) _[SRT 16, 33, 60–80 days] FBR (~62.5% [*])	Karthikeyan and Meyer, 2006 Carballa et al., 2004 Göbel et al., 2005; Göbel et al., 2007
	246 3000 146.5–355.5	CAS/46 (81% ^{**}) 200 (93% [*]) CAS/46.6 ± 2.6 (68.2% ^{**}) CAS + chlorination/15.3 ± 0.3 (95.7% ^{**}) (65–96% ^{**})	Zuccato et al., 2010 Watkinson et al., 2009 Li et al., 2009
	500–10000 na	UWTP I Secondary/(<60)–640 Chlorination/(<50)–70 UWTP II Secondary/100–1600 UV/330–2140	Yu et al., 2009 Renew and Huang, 2004
	13–155 na	4–39 (69–75% [*]) 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)	Pailler et al., 2009 Batt et al., 2007
	820 ± 230 16 ± 5; 118 ± 17; 10 ± 3; 25 ± 7	620 ± 90 (24% ^{**}) CAS + chlorination/16 ± 7 (0%) Oxidation ditch + UV/78 ± 13 (34% [*]) CAS/12 ± 3 (<0 [*]) Chemically enhanced + chlorination/9 ± 4 (64% [*])	Ternes et al., 2007 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% [*]) (7% ^{**})	Sahar et al., 2010
	3476 500–1000 5*10 ⁵	MBR (75–90% ^{**}) _[STR > 100 days] MBR (88.5, 96.9, 99.3, 99.5% ^{**}) _[STR = 3, 10, 30, 60 days]	Kovalova et al., 2012 Abegglen et al., 2009 Xia et al., 2012
	20–268 10 ⁴ 5*10 ⁴	MBR-RO (69% ^{**}) _[STR = 45 days] MBR (52% ^{**}) _[SRT = 44–72 days] MBR (55% ^{**}) _[SRT = 15 days] MBR (86% ^{**}) _[SRT = 30 days]	Dolar et al., 2012 Reif et al., 2008 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] (92, 75, 68% [*]) _[STR = 16, 33, 60–80 days] MBR (81% ^{**}) _[STR = 30–50 days]	Göbel et al., 2005; Göbel et al., 2007 Joss et al., 2005 Kovalova et al., 2012

Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
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 and Witter, 2008
	3.2–54.7; 17.8	CAS _{Shatin} (100%**)	Li and Zhang, 2011
		CAS _{Stanley} (100%**)	
		Disinfection (ne)	
		Final (100%**)	
	3	MBR/RO	Sahar et al., 2010
	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%*)		
Sulfadiazine	500–1000	MBR (75–90%**) _[STR > 100 days]	Abegglen et al., 2009
	5100 _(GZ-UWTP1) ;	GZ-UWTP ₁	Peng et al., 2006
	5150 _(GZ-UWTP2)	Primary/4180 (19%*)	
		Secondary/nd	
		Tertiary /nd	
		GZ-UWTP ₂	
		Primary/nd	
		Secondary/nd	
		Tertiary/nd	
		CAS/16.2 ± 0.0 (72.8%**)	Li et al., 2009
	CAS + chlorination/nd		
	CAS + chlorination/36 ± 13 (50%*)	Xu et al., 2007	
	CAS _{Shatin} (100%**)	Li and Zhang, 2011	
	CAS _{Stanley} (87%**)		
	Disinfection (4%**)		
	Final (88%**)		
	MBR (-23%**) _[STR = 30–50 days]	Kovalova et al., 2012	
	MBR (75–90%**) _[STR > 100 days]	Abegglen et al., 2009	
	MBR	Xia et al., 2012	
	(93.8, 97.5, 99.6, 99.7%**) _[STR = 3, 10, 30, 60 days]		
Sulfathiazole	40	Primary/nd (100%*)	Watkinson et al., 2007
		CAS/nd (100%*)	
	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
	1560	60 (>93%*)	Choi et al., 2007
Sulfachloropyridazine	70	(<30) (>57%*)	Yang et al., 2005
	460	(<30) (>93%*)	Choi et al., 2007
Sulfadimethoxine	2000–10000	(61–96%**)	Yu et al., 2009
	(<(LOQ)–2.6)	(<LOQ)–1.9 (27%*)	Spongberg and Witter, 2008
	(1.0)–26	(1.0)–9.0 (65%*)	Pailler et al., 2009
	60–150	Primary (-29 to 20%**)	Göbel et al., 2005;
	CAS (-107 to 72%**)	Göbel et al., 2007	
	MBR (60, 48, 55%*) _[SRT 16, 33, 60–80 days]		
	FBR (72%*)		
Sulfasalazine	500–1000	MBR (75–90%**) _[STR > 100 days]	Abegglen et al., 2009
	60	Primary/15 (75%*)	Watkinson et al., 2007
	CAS/nd (100%*)		
Sulfamonomethoxine	100	150 (<0%*)	Watkinson et al., 2009
	3110	(<30) (>99%*)	Choi et al., 2007
Sulfisoxazole	(<LOQ)–22.1)	(<LOQ)–11.9 (46%*)	Spongberg and Witter, 2008
Sulfadimidine	25 ± 12; 696 ± 212	CAS + chlorination/12 ± 6 (52%*)	Xu et al., 2007
		Oxidation ditch + UV/346 ± 54 (50%*)	
Quinolones			
Norfloxacin	na	210	Costanzo et al., 2005

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Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
Ciprofloxacin	210	Primary/145 (31%*) CAS/15 (93%*)	Watkinson et al., 2007
	110–460	85–320 (–20 to 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
	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 and Witter, 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

Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
Enrofloxacin	100	Primary/20 (80%*) CAS/5 (95%*)	Watkinson et al., 2007
Ofloxacin	40	50 (<0%*)	Watkinson et al., 2009
	115–1274	53–991 (2–88%**)	Lin et al., 2009
	470 (22.5 ± 2.5; 30.0 ± 3.0; 19.5 ± 3.0; 9.0 ± 1.5; 10.0 ± 1.0) _[UWTP1-UWTP5]	110 (77%**) (56%**)	Brown et al., 2006 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
	539.80 137 ± 58; 359 ± 52; 80 ± 12; 368 ± 23	183.10 (66%*) CAS + chlorination/41 ± 8 (70%*) Oxidation ditch + UV/137 ± 28 (62%*) CAS/48 ± 7 (40%*) Chemically enhanced + chlorination/165 ± 15 (55%*)	Castiglioni et al., 2008 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
Nalidixic acid	nd–2900	MBR-RO (0%**) _[STR=45 days]	Dolar et al., 2012
	200	Primary/ nd (100%*) CAS/1 (100%*)	Watkinson et al., 2007
Pipemidic acid	26–372	40–200 (37–46%**)	Lin et al., 2009
	200	450 (<0%*)	Watkinson et al., 2009
Flerofloxacin	54	12 (78%*)	Xiao et al., 2008
Lomefloxacin	28	5.8 (79%*)	Xiao et al., 2008
Gatifloxacin	98	17 (83%*)	Xiao et al., 2008
Moxifloxacin	111	56 (50%*)	Xiao et al., 2008
Trimethoprim	44	17 (61%*)	Xiao et al., 2008
	930	Primary/480 (48%*) CAS/30 (97%*)	Watkinson et al., 2007
	120–320	120–230 (~–17 to 62%**)	Gulkowska et al., 2008

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Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
	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 to 31% ^{**}) CAS/80–400 (–40 to 20% ^{**}) MBR (28, 33, 87% [*]) _[SRT 16, 33, 60–80 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;	CAS _{Shatin} (13% ^{**})	Li and Zhang, 2011
	136–172	CAS _{Stanley} (42% ^{**}) Disinfection (40% ^{**}) Final (65% ^{**})	
	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

Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
Tetracyclines			
Tetracycline	35	Primary/nd (100%*) CAS/20 (43%*)	Watkinson et al., 2007
	96–1300	180–620 (–88 to 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 and Witter, 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%**) Final (39%**)	Li and Zhang, 2011
	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

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Table 1 – (continued)

Antibiotic group Antibiotic	Initial concentration (ng L ⁻¹)	Effluent concentration (ng L ⁻¹)/ (% Removal efficiency)	Reference
	6.8–13.3	14.9–32.5 (<0%*)	Sponberg and Witter, 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.

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.

* 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.

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-H₂O. Erythromycin-H₂O 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., 2009a,b) 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; Sponberg and Witter, 2008) up to 80% (Dolar et al., 2012; Lin et al., 2009a,b). For clarithromycin and erythromycin-H₂O 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 500–10,000 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 deconjugate 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.

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

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

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

2.1.4. Trimethoprim

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

The removal rates for chlortetracycline as reported by Li 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 (Drewes, 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 behavior 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 analyzed by Li 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 cut-off (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% was 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 the following steps: (i) solute transport in the bulk-adsorbate movement by the stagnant liquid film surrounding the adsorbent, (ii) film diffusion-adsorbate transport along the film, (iii) pores diffusion-adsorbate diffusion through the porous structure to the active sites (molecular diffusion in the pore and/or in the adsorbent surface), (iv) adsorption-interaction between adsorbate and porous structure (Homem and Santos, 2011).

The removal effectiveness of the activated carbon adsorptive treatment system depends on the properties of the adsorbent (e.g. specific surface area, porosity, surface polarity and physical shape of the material), and the characteristics of the compound (e.g. shape, size, charge and hydrophobicity). Moreover, the sorption efficiencies of antibiotics to activated carbon may be significantly altered by the initial concentrations of the target compounds, the pH, the temperature and the presence of other species in the solution (Aksu and Tunç, 2005). Non-specific dispersive interactions (e.g. van der Waals interactions) are the dominant mechanism of removal for organic compounds, including antibiotics, in activated carbon adsorption systems, removing most non-polar antibiotics with $\log K_{ow} > 2$. However, electrostatic interactions between ionic antibiotics and the charged groups on the surface of activated carbon can result in removal of polar antibiotics (Snyder et al., 2003). The removal of antibiotics by activated carbon has been reported during wastewater treatment in some studies (Adams et al., 2002; Westerhoff et al., 2005; Putra et al., 2009; Rivera-Utrilla et al., 2009; McArdell et al., 2011; Boehler et al., 2012). A post-treatment with powdered activated carbon (PAC) after biological treatment has been mostly investigated. The concentrations of several antibiotics in wastewater with PAC dosages between 10 and 20 mg L^{-1} have been reduced by 49–99% after 4 h contact time (Adams et al.,

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) Wastewater from plant manufacturing AMX (China)	280 ng L ⁻¹ na	MF/RO plant: receives ~10% of CAS effluent 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	MF: nd; RO: nd 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%)	Watkinson et al., 2007 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
Sulfadiazine	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
	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 %)	Kosutić 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 %)	Kosutić 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 %)	Kosutić et al., 2007
Sulfathiazole	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
	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
Sulfamerazine	CAS effluent (Australia)	40 ng L ⁻¹	MF/RO plant: receives ~10% of CAS effluent	MF: nd; RO: nd	Watkinson 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
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

(continued on next page)

Table 2 – (continued)

Advanced treatment process Antibiotic Group	Membrane filtration				
	Type of wastewater (location)	Initial concentration	Treatment process	Results/findings (Removal efficiency)	Reference
Sulfadimethoxine	Missouri River water (Jefferson City)	50 $\mu\text{g L}^{-1}$	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	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%)	Kosutić et al., 2007
Enrofloxacin					
Norfloxacin	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
	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%)	Kosutić et al., 2007
Tetracyclines	Missouri River water (Jefferson City)	50 $\mu\text{g L}^{-1}$	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 ⁻¹	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
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%)	Kosutić et al., 2007
Lincosamides	Waste liquor from the crystallization unit in a pharmaceutical company (Chi Feng, Inner Mongolia, China).	1000 mg L ⁻¹	RO: SEPA CELL flat sheet membrane apparatus; membrane area of 155 cm ² . UF: 0.3 MPa; UF membranes of different molecular weight cut-off (3,10, 30, 50 K Da)	< 80 mg L ⁻¹ (>92%)	Li et al., 2004
	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
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		ACTIVATED CARBON ADSORPTION			
Antibiotic Group	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	HCl 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 Tunç, 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 ₂ O	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 ₂ O	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

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Table 2 – (continued)

Advanced treatment process		ACTIVATED CARBON ADSORPTION			
Antibiotic Group	Type of wastewater (location)	Initial concentration	Treatment process	Results/findings (Removal efficiency)	Reference
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. Missouri River water (Jefferson City).	na 50 µg L ⁻¹	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
Tetracyclines Tetracycline	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	AC dose = 10 mg L ⁻¹ (55%) AC dose = 20 mg L ⁻¹ (65%) AC dose = 50 mg L ⁻¹ (>90%) PAC dose = 23 mg L ⁻¹ (>83%) PAC dose = 43 mg L ⁻¹ (>83%)	Adams et al., 2002 McArdell et al. 2011
Nitroimidazoles Metronidazole	na	na	Four carbonaceous adsorbents: Single walled carbon nanotubes (SWNT); Multi-walled carbon nanotubes (MWNT); Pulverized activated carbon (AC) and nonporous Graphite (G).	Adsorption efficiency: G/ SWNT > MWNT >> AC	Ji et al., 2009
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
OZONATION					
Advanced treatment process Antibiotic Group	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 ₂ O ₂ ([H ₂ O ₂] = 2–40 mM); pH = 10.5	COD removal O ₃ : (10–56%) O ₃ + H ₂ O 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%) O ₃ /pH 11: (52%)	Arslan Alaton and Dogruel, 2004
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 ₂ O ₂ ([H ₂ O ₂] = 20 mM)	(a) (80% in 60 min) (b) (100% in 60 min)	Balcioğlu 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 ₂ O ₂ ([H ₂ O ₂] = 20 mM)	(a) (>99% in 60 min) (b) (100% in 60 min)	Balcioğlu 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., 2009a
	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 ⁻¹ DOC (77%) O ₃ dose = 0.62 g g ⁻¹ DOC (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 ⁻¹ DOC (>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

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Table 2 – (continued)

Advanced treatment process		OZONATION			
Antibiotic Group	Type of wastewater (location)	Initial concentration	Treatment process	Results/findings (Removal efficiency)	Reference
Clarithromycin	Secondary effluent (German)	$0.21 \pm 0.02 \mu\text{g L}^{-1}$	Ozonation-UV treatment plant $\text{O}_3 = 100 \text{ g h}^{-1}$, O_3 dose = $5\text{--}15 \text{ mg L}^{-1}$, 2 diffuser/PVC bubble columns	O_3 dose = $5\text{--}15 \text{ mg L}^{-1}$ ($\geq 76\%$)	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 < $50 \mu\text{M}$ (100%)	Rosal et al., 2010
Erythromycin	CAS effluent (Regensdorf, Switzerland)	206 ng L^{-1}	O_3 dose = $1.6\text{--}5.3 \text{ mg L}^{-1}$ ($0.36\text{--}1.16 \text{ g g}^{-1}$ DOC), Retention time=8–15 min, full scale six compartment reactor	O_3 dose = 0.40 g g^{-1} DOC (94%) O_3 dose = 0.62 g g^{-1} DOC (97%) O_3 dose = 0.79 g g^{-1} DOC (99%)	Hollender et al., 2009
	Secondary effluent (German)	$0.62 \pm 0.24 \mu\text{g L}^{-1}$	Ozonation-UV treatment plant $\text{O}_3 = 100 \text{ g h}^{-1}$, O_3 dose = $5\text{--}15 \text{ mg L}^{-1}$, 2 diffuser/PVC bubble columns	O_3 dose = $5\text{--}15 \text{ mg L}^{-1}$ ($\geq 92\%$)	Ternes et al., 2003
	CAS effluent (Alcala de Henares, Madrid)	72 ng L^{-1}	AirSep AS-12 PSA oxygen generation unit	O_3 dose < $90 \mu\text{M}$ (100%)	Rosal et al., 2010
Erythromycin-H ₂ O Sulfonamides	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 = $1.6\text{--}5.3 \text{ mg L}^{-1}$ ($0.36\text{--}1.16 \text{ g g}^{-1}$ DOC), Retention time = 8–15 min, full scale six compartment reactor	O_3 dose = 0.61 g g^{-1} DOC (>64%)	Hollender et al., 2009
	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
Sulfamethoxazole	Secondary effluent (Kloten-Opfikon, Switzerland)	$1 \mu\text{M}$	Batch experiments, O_3 dose = $0.5\text{--}5.0 \text{ mg L}^{-1}$ 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)	$2 \mu\text{g L}^{-1}$	O_3 dose=0–5 mg L^{-1} ; flow = $200 \pm 10 \text{ L h}^{-1}$ (only column 1).	O_3 dose $\geq 2 \text{ mg L}^{-1}$ ($\geq 90\%$)	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 < $220 \mu\text{M}$ (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 = $1.6\text{--}5.3 \text{ mg L}^{-1}$ ($0.36\text{--}1.16 \text{ g g}^{-1}$ DOC), Retention time=8–15 min, full scale six compartment reactor	O_3 dose = 0.40 g g^{-1} DOC (87%) O_3 dose = 0.62 g g^{-1} DOC (96%) O_3 dose = 0.79 g g^{-1} DOC (96%)	Hollender et al., 2009
Sulfamethazine	Secondary effluent (German)	$0.62 \pm 0.05 \mu\text{g L}^{-1}$	Ozonation-UV treatment plant $\text{O}_3 = 100 \text{ g h}^{-1}$, O_3 dose = $5\text{--}15 \text{ mg L}^{-1}$, 2 diffuser/PVC bubble columns.	O_3 dose = $5\text{--}15 \text{ mg L}^{-1}$ ($\geq 92\%$)	Ternes et al., 2003
	Missouri River water (Jefferson City)	$50 \mu\text{g L}^{-1}$	O_3 dose = 7.1 mg L^{-1}	$0.3 \text{ mg L}^{-1} \text{ O}_3$ at 1.3 min (> 95%)	Adams et al., 2002
Sulfathiazole	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
	Missouri River water (Jefferson City)	$50 \mu\text{g L}^{-1}$	O_3 dose = 7.1 mg L^{-1}	$0.3 \text{ mg L}^{-1} \text{ O}_3$ at 1.3 min (> 95%)	Adams et al., 2002
Sulfamerazine	Missouri River water (Jefferson City)	$50 \mu\text{g L}^{-1}$	O_3 dose = 7.1 mg L^{-1}	$0.3 \text{ mg L}^{-1} \text{ O}_3$ at 1.3 min (> 95%)	Adams et al., 2002
Sulfachloropyridazine	Missouri River water (Jefferson City)	$50 \mu\text{g L}^{-1}$	O_3 dose = 7.1 mg L^{-1}	$0.3 \text{ mg L}^{-1} \text{ O}_3$ at 1.3 min (> 95%)	Adams et al., 2002
Sulfadimethoxine	Missouri River water (Jefferson City)	$50 \mu\text{g L}^{-1}$	O_3 dose = 7.1 mg L^{-1}	$0.3 \text{ mg L}^{-1} \text{ O}_3$ 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 < $50 \mu\text{M}$ (100%)	Rosal et al., 2010
	CAS effluent (Regensdorf, Switzerland)	125 ng L^{-1}	O_3 dose = $1.6\text{--}5.3 \text{ mg L}^{-1}$ ($0.36\text{--}1.16 \text{ g g}^{-1}$ DOC), Retention time=8–15 min, full scale six compartment reactor	O_3 dose = 0.40 g g^{-1} DOC (98%) O_3 dose = 0.62 g g^{-1} DOC (97%) O_3 dose = 0.79 g g^{-1} DOC (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., 2009b
	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 ⁻¹ DOC (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

FENTON OXIDATION

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 (China)	Na	Fenton oxidation after extraction (dichloromethane) [FeSO ₄ ·7H ₂ O]=10 g L ⁻¹ ; [H ₂ O 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 ₂ O 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.	Trovó 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 ₂ O ₂] = 20 mM; [Fe(II)]=1 mM; [Fe(III)] = 1 mM.	COD removal Photo-Fenton: (56%) Photo-Fenton-like: (66%)	Arslan Alaton and Dogruel, 2004

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Table 2 – (continued)

Advanced treatment process		FENTON OXIDATION			
Antibiotic Group	Type of wastewater (location)	Initial concentration	Treatment process	Results/findings (Removal efficiency)	Reference
	Pharmaceutical wastewater (China)	na	Microwave power=100–500 W; radiation time = 2–10 min; pH = 1–11; [H ₂ O ₂] = 3200–19000 mg L ⁻¹ ; [Fe ₂ (SO ₄) ₃] = 2000–8000 mg L ⁻¹	Dark Fenton: (61%) Dark Fenton-like: (46%) TOC removal Photo-Fenton: (51%) Photo-Fenton-like: (42%) Dark Fenton: (33%) Dark Fenton-like: (18%) Optimum conditions: Microwave power = 300 W; radiation time = 6 min; pH = 4.42; [H ₂ O ₂] = 1300 mg L ⁻¹ ; [Fe ₂ (SO ₄) ₃] = 4900 mg L ⁻¹ COD removal: (57.53%) TOC removal: (>40%) Degradation: (55.06%)	Yang et al., 2009
Quinolones	Secondary effluent (Almería, Spain)	100 µg L ⁻¹	Pilot compound parabolic collector plant (CPC), [Fe ²⁺] = 5 mg L ⁻¹ , [H ₂ O ₂] = 50 mg L ⁻¹ , t _{30W} = 102 min	(100%)	Klamerth et al., 2010
Ofloxacin	(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 ₂ O ₂] = 1.357–8.142 mmol L ⁻¹	[Fe ²⁺] = 5 mg L ⁻¹ , [H ₂ O 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 ₂ O 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 ₂ O 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 ₂ O ₂] = 75 mg L ⁻¹ , t _{30WT,n} = 20.1 min	(100%)	Michael et al., 2012b
Tetracyclines	CAS effluent (Araraquara, Brazil)	24 mg L ⁻¹	Black light (15 W) and solar irradiation [H ₂ O 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		HETEROGENEOUS PHOTOCATALYSIS WITH TiO ₂			
Antibiotic Group	Type of wastewater (location)	Initial concentration	Treatment process	Results/findings (Removal efficiency)	Reference
β-Lactams	Antibiotic wastewater (AW)	138 ± 5 mg L ⁻¹	UV/H ₂ O ₂ /TiO ₂ 2000 mL of AW; [TiO ₂] = 0–1000 mg L ⁻¹ ; [H ₂ O ₂] = 50–350 mg L ⁻¹ ; T = 22 ± 2 °C UV lamp (6 W, λ ≈ 365 nm) UV/H ₂ O ₂ /TiO ₂ /SBR 1.5 L of AW; 65 days at HRT 24 hr	UV/H ₂ O ₂ /TiO ₂ [TiO ₂] = 1000 mg L ⁻¹ [H ₂ O ₂] = 250 mg L ⁻¹ 30 min, pH = 5 (100%) UV/H ₂ O ₂ /TiO ₂ /SBR [TiO ₂] = 1000 mg L ⁻¹	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	[H ₂ O ₂] = 250 mg L ⁻¹ (57 % of COD); (53% of DOC) 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 ₂ O ₂ /TiO ₂ 2000 mL of AW; [TiO ₂]=0–1000 mg L ⁻¹ ; [H ₂ O ₂] = 50–350 mg L ⁻¹ ; T = 22 ± 2 °C UV lamp (6 W, λ ≈ 365 nm) UV/H ₂ O ₂ /TiO ₂ /SBR 1.5 L of AW; 65 days at HRT 24 hr	UV/H ₂ O ₂ /TiO ₂ [TiO ₂] = 1000 mg L ⁻¹ [H ₂ O ₂] = 250 mg L ⁻¹ 30 min, pH = 5 (100%) UV/H ₂ O ₂ /TiO ₂ /SBR [TiO ₂] = 1000 mg L ⁻¹ [H ₂ O ₂] = 250 mg L ⁻¹ (57 % of COD); (53% of DOC)	Elmolla and Chaudhuri, 2011
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 ₂ O 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 ₂ O 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 ₂ O ₂] = 1.357–8.142 mmol L ⁻¹	[TiO ₂] = 3 g L ⁻¹ , 120 min (60%) [TiO ₂] = 3 g L ⁻¹ , [H ₂ O 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 Penicillin	Effluent from Varese UWTP Antibiotic formulation effluent (Turkey)	18 ng L ⁻¹ na	UV-light treatment UV light (λ = 253.7 nm, 1.73×10^{-4} Einstein (Ls) ⁻¹); 60 min; pH=7; [H ₂ O ₂]=0–40 mM	(100%) COD removal UV/pH 7: (0%) UV + H ₂ O ₂ (40 mM)/pH 7: (11%) UV + H ₂ O ₂ (30 mM)/pH 7: (22%) TOC removal UV/pH 7: (0%)	Zuccato et al., 2010 Arslan Alaton and Dogruel, 2004

(continued on next page)

Table 2 – (continued)

Advanced treatment process		PHOTOLYSIS WITH UV			
Antibiotic Group	Type of wastewater (location)	Initial concentration	Treatment process	Results/findings (Removal efficiency)	Reference
Macrolides				UV + H ₂ O ₂ (40 mM)/pH 7: (10%) UV + H ₂ O ₂ (30 mM)/pH 7: (6%)	
Clarithromycin	Effluent from secondary sedimentation and sand filter (Japan)	110–656 ng L ⁻¹	3 UV lamps ($\lambda=254$ nm; intensity=1.025 mW cm ⁻²); 3 reactors in series (R ₁ -R ₃); Air flow rate=0.5 L min ⁻¹ ; [H ₂ O ₂]=7.8 mg L ⁻¹	UV: (24–34%) UV + H ₂ O ₂ : (>90%)	Kim et al., 2009
Erythromycin	Effluent from Varese UWTP Effluent from secondary sedimentation and sand filter (Japan)	319 ng L ⁻¹ 110–656 ng L ⁻¹	UV-light treatment 3 UV lamps ($\lambda=254$ nm; intensity=1.025 mW cm ⁻²); 3 reactors in series (R ₁ -R ₃); Air flow rate=0.5 L min ⁻¹ ; [H ₂ O ₂]=7.8 mg L ⁻¹	(0%) UV: (24–34%) UV + H ₂ O ₂ : (>90%)	Zuccato et al., 2010 Kim et al., 2009
Azithromycin	Effluent from Varese UWTP Effluent from secondary sedimentation and sand filter (Japan)	12 ng L ⁻¹ 110–656 ng L ⁻¹	UV-light treatment 3 UV lamps ($\lambda = 254$ nm; intensity = 1.025 mW cm ⁻²); 3 reactors in series (R ₁ -R ₃); Air flow rate = 0.5 L min ⁻¹ ; [H ₂ O ₂] = 7.8 mg L ⁻¹	(0%) UV: (24-34%) UV+ H ₂ O ₂ : (>90%)	Zuccato et al., 2010 Kim et al., 2009
Spiramycin	Effluent from Varese UWTP	603 ng L ⁻¹	UV-light treatment	(17%)	Zuccato et al., 2010
Sulfonamides				(48%)	Ryan et al., 2011
Sulfamethoxazole	Effluent from Blue Lake WWTP; Metro WWTP and Lake Josephine (USA) Effluent from secondary sedimentation and sand filter (Japan)	1 μ M 42–187 ng L ⁻¹	Photolysis experiments (Suntest CPS + solar simulator with a UV-Suprax optical filter, 765 W m ⁻²) 3 UV lamps ($\lambda = 254$ nm; intensity = 1.025 mW cm ⁻²); 3 reactors in series (R ₁ -R ₃); Air flow rate = 0.5 L min ⁻¹ ; [H ₂ O ₂] = 7.8 mg L ⁻¹	UV: (89–100%) UV + H ₂ O ₂ : (>90%)	Kim et al., 2009
Sulfamethazine	Effluent from Varese UWTP	246 ng L ⁻¹	UV-light treatment	(0%)	Zuccato et al., 2010
Sulfathiazole	Missouri River water (Jefferson City).	50 μ g 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
Sulfamerazine	Missouri River water (Jefferson City).	50 μ g 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
Sulfachlorpyridazine	Missouri River water (Jefferson City).	50 μ g 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 μ g 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
	Effluent from secondary sedimentation and sand filter (Japan)	42–187 ng L ⁻¹	3 UV lamps ($\lambda = 254$ nm; intensity = 1.025 mW cm ⁻²); 3 reactors in series (R ₁ -R ₃); Air flow rate=0.5 L min ⁻¹ ; [H ₂ O ₂] = 7.8 mg L ⁻¹	UV: (89–100%) UV + H ₂ O ₂ : (>90%)	Kim et al., 2009
Trimethoprim					
	Missouri River water (Jefferson City).	50 μ g 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 μ M	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 ₂ O ₂ : two G15T8 germicidal lamps (General Electric, Fairfield, CT, USA), UV = 300–700 mJ cm ⁻² ; [H ₂ O ₂] = 0–20 mg L ⁻¹ .	UV dose = 300 mJ cm ⁻² ; [H ₂ O ₂] = 20 mg L ⁻¹ (21-67%) UV dose = 500 mJ cm ⁻² ; [H ₂ O ₂] = 20 mg L ⁻¹ (32-92%) UV dose = 700 mJ cm ⁻² ; [H ₂ O 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 ($\lambda = 254$ nm; intensity = 1.025 mW cm ⁻²); 3 reactors in series (R ₁ -R ₃); Air flow rate = 0.5 L min ⁻¹ ; [H ₂ O ₂] = 7.8 mg L ⁻¹	UV: (15%) UV+ H ₂ O ₂ : (>90%)	Kim et al., 2009
Oxytetracycline	Secondary wastewater (Beijing, China)	50 μ M	11 W low-pressure Hg vapor lamp ($\lambda = 254$ nm), photon flow = 4.5×10^{-5} E m ⁻² s ⁻¹ ; UV dose=(0-320) $\times 10^2$ mJ cm ⁻² ; 500 mL WW, [H ₂ O ₂] = 1 mM,	UV UV dose = 30528 mJ cm ⁻² (100%) UV/H ₂ O 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 ($\lambda = 254$ nm), photon flow = 4.5×10^{-5} E m ⁻² s ⁻¹ ; UV dose = $(0-320) \times 10^2$ mJ cm ⁻² ; 500 mL WW, [H ₂ O ₂] = 1 mM,	UV UV dose = 22896 mJ cm ⁻² (100%) UV/H ₂ O ₂	Yuan et al., 2011
Chlorotetracycline	Effluent from secondary sedimentation and sand filter (Japan)	4–17 ng L ⁻¹	3 UV lamps ($\lambda = 254$ nm; intensity = 1.025 mW cm ⁻²); 3 reactors in series (R ₁ –R ₃); Air flow rate = 0.5 L min ⁻¹ ; [H ₂ O ₂] = 7.8 mg L ⁻¹	UV UV dose = 7632 mJ cm ⁻² (100%) UV + H ₂ O ₂ ; (>90%)	Kim et al., 2009
Quinolones Norfloxacin	Effluent from secondary sedimentation and sand filter (Japan)	4–148 ng L ⁻¹	3 UV lamps ($\lambda = 254$ nm; intensity = 1.025 mW cm ⁻²); 3 reactors in series (R ₁ –R ₃); Air flow rate = 0.5 L min ⁻¹ ; [H ₂ O ₂] = 7.8 mg L ⁻¹	UV; (86–100%) UV + H ₂ O ₂ ; (69%)	Kim et al., 2009
Ofloxacin Ciprofloxacin	Effluent from Varese UWTP Secondary wastewater (Beijing, China)	463 ng L ⁻¹ 50 µM	UV-light treatment 11 W low-pressure Hg vapor lamp ($\lambda = 254$ nm), photon flow = 4.5×10^{-5} E m ⁻² s ⁻¹ ; UV dose = $(0-320) \times 10^2$ mJ cm ⁻² ; 500 mL WW, [H ₂ O ₂] = 1 mM,	(19%) UV UV dose = 11448 mJ cm ⁻² (100%) UV/H ₂ O ₂ UV dose = 7632 mJ cm ⁻² (100%) (0%)	Zuccato et al., 2010 Yuan et al., 2011
Nalidixic acid	Effluent from Varese UWTP Effluent from secondary sedimentation and sand filter (Japan)	513 ng L ⁻¹ 4–148 ng L ⁻¹	UV-light treatment 3 UV lamps ($\lambda = 254$ nm; intensity = 1.025 mW cm ⁻²); 3 reactors in series (R ₁ –R ₃); Air flow rate = 0.5 L min ⁻¹ ; [H ₂ O ₂] = 7.8 mg L ⁻¹	UV UV; (86–100%) UV + H ₂ O ₂ ; (>90%)	Zuccato et al., 2010 Kim et al., 2009
Lincomamides Lincomycin Glycopeptides Vancomycin	Effluent from Varese UWTP Effluent from Varese UWTP	9.7 ng L ⁻¹ 41 ng L ⁻¹	UV-light treatment UV-light treatment	(0%) (28%)	Zuccato et al., 2010 Zuccato et al., 2010

NOTES. nd: Not detected. na: Not available. GAC: Granular activated carbon. PAC: Powdered activated carbon.

2002; Westerhoff et al., 2005). In a study on hospital wastewater treatment, macrolides, fluoroquinolones, trimethoprim and clindamycin were removed well at PAC dosages of 20–40 mg L⁻¹, while sulfamethoxazole and metronidazole showed poor removals (McArdell et al., 2011). PAC can also be added directly into the biological reactor, where higher concentrations of carbon are required. Serrano et al. (2011) added 1 g L⁻¹ into a sequential membrane bioreactor and found elimination of 42–64% for erythromycin, 71–97% for roxithromycin whereas no significant removal was obtained for TMP. Putra et al. (2009) compared the adsorption capacity of activated carbon and bentonite and reported that 94.67% of amoxicillin was removed from wastewater using activated carbon at a dose as high as 30 g L⁻¹.

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

2.4. Advanced oxidation processes (AOPs)

Advanced Oxidation Processes (AOPs) are quite efficient novel methods for water and wastewater treatment (Legrini et al., 1993; Klavarioti et al., 2009; Malato et al., 2009). These processes involve the use and generation of powerful transitory species, principally the hydroxyl radical (HO•) (Goslich et al., 1997; Andreozzi et al., 1999). HO• are powerful oxidizing agents leading to oxidation and mineralization of organic matter (Litter, 2005), while this species is characterized by lack of selectivity of attack. This property is of great importance in wastewater treatment because radicals attack the oxidizable part of organic molecules with rates usually in the order of 10⁶–10⁹ M⁻¹ s⁻¹ (Andreozzi et al., 1999). Several studies have reported the effective AOPs treatment for removal of antibiotics in wastewater effluents (Adams et al., 2002; Arslan Alaton et al., 2004; Saritha et al., 2007; Naddeo et al., 2009; Elmolla and Chaudhuri, 2011). It is worth noting the fact that most studies do not include information on the by-products formed during the application of oxidation or any information related to the antibiotic activity of the by-products. Therefore, AOPs should be carefully monitored and ecotoxicological investigations should be accompanied to investigate the formation of potentially toxic transformation products (Hollender et al., 2009; Rizzo, 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} ($0.3\text{--}0.4 \text{ g g}^{-1} \text{ DOC}$) 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} ($0.40 \text{ g O}_3 \text{ g}^{-1} \text{ DOC}$) (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 $2.96 \text{ g L}^{-1} \text{ h}^{-1}$. In a study of Arslan Alaton et al. (2004) the COD of an antibiotic formulation effluent containing penicillin ($\text{COD} = 830 \text{ mg L}^{-1}$) 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 ($\text{pH} = 11$), whereas the removal efficiency was much lower under acidic conditions ($\text{pH} = 3$) (COD removal max = 15%; TOC removal max = 2%). Many authors (Balcioglu and Otker, 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 amenable to oxidative attack (Dickenson et al., 2009).

Research conducted so far demonstrates that ozonation is a promising approach to degrade antibiotics. According to Table 2, ozonation was found to be an effective process for removing β -lactams, macrolides, sulfonamides and trimethoprim, quinolones, tetracyclines and lincosamides. The energy consumption for upgrading a Swiss municipal wastewater treatment plant with ozonation was evaluated by Hollender et al. (2009). For an ozone dose of $0.6 \text{ g O}_3 \text{ g}^{-1} \text{ DOC}$ (effluent DOC $\sim 5 \text{ g m}^{-3}$), 0.035 kWh m^{-3} wastewater was consumed, which is 12% of the total energy consumption of a typical nutrient removal plant (0.3 kWh m^{-3} wastewater). Additionally, $0.01\text{--}0.015 \text{ kWh m}^{-3}$ was needed for pure oxygen production. Ozone treatment performance may be enhanced if ozone is combined with UV irradiation, hydrogen peroxide or catalysts (usually iron or copper complexes) (Klavarioti et al., 2009). However, optimal process and operating conditions have yet to be determined for the various water and wastewater types as well as for the different types of antibiotics (Yargeau and Leclair, 2008).

2.4.2. Fenton oxidation

Fenton's oxidation is a homogeneous oxidation process and is considered to be a metal-catalyzed oxidation reaction, in which iron acts as the catalyst (Tekin et al., 2006; Saritha et al., 2007). The main disadvantage of the process is the low pH value required in order to avoid iron precipitation that takes place at higher pH (Melero et al., 2007; Santos et al., 2007).

Trovó et al. (2008) observed that amoxicillin degradation was not influenced by the source of the irradiation during the photo-Fenton process and the removals of the antibiotic obtained were 89 and 85% under black light and solar irradiation, respectively. A similar study by Bautitz and Nogueira (2007) showed that tetracycline was removed by 80% during the photo-Fenton treatment using two types of iron and irradiation. Moreover, in a study by Arslan Alaton and Dogruel (2004) adequate COD and TOC removal rates were achieved during the photo-Fenton and photo-Fenton-like treatment of a formulation effluent containing penicillin. Trimethoprim was completely removed during solar-Fenton process in the study of Michael et al. (2012a) and it was found that the presence of organic carbon and higher salt content in the simulated wastewater and real secondary effluent, led to lower mineralization though per dose of hydrogen peroxide compared to ultrapure water. It is important to highlight that a new approach aimed at performing photo-Fenton treatment at neutral pH has been proposed by Klammerth et al. (2010) and De la Cruz et al. (2012). The efficiency of the modified photo-Fenton system is based on the reaction of dissolved organic matter (DOM) present in wastewaters with Fe^{2+} leading to the formation of soluble iron-complexes. However, contaminants degradation and mineralization tend to be slower at neutral pH than at pH 3.0.

Michael et al. (2012b) investigated the application of a solar photo-Fenton system for the degradation of antibiotics at low concentration level ($\mu\text{g L}^{-1}$) in secondary treated domestic effluents at a pilot-scale. The examined antibiotics were ofloxacin and trimethoprim and the pilot treatment plant

consisted of a compound parabolic collector reactor. The results demonstrated the efficiency of the process in removing enterococci, resistant to these two antibiotics, while the compounds themselves were completely eliminated. The total cost of a full-scale unit for the treatment of $150 \text{ m}^3 \text{ day}^{-1}$ of secondary wastewater effluent was estimated to be 0.85 € m^{-3} . This value was found to be in agreement with a previous study of the photo-Fenton process in a pilot-scale set-up (Jordá et al., 2011).

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

2.4.3. Heterogeneous photocatalysis with TiO_2

Heterogeneous photocatalysis by TiO_2 semiconductor is achieved usually by the illumination of a suspension of TiO_2 in aqueous solution with light energy greater than its bandgap energy. This leads to the formation of high energy electron-hole pairs (e^-/h^+) which can migrate on the surface of the catalyst and can either recombine producing thermal energy, or participate in redox reactions with the compounds that are adsorbed on the catalyst's surface (Herrmann et al., 1993; Schiavello, 1993; Robertson, 1996). The valence holes are strong oxidants and are able to oxidize various contaminants, as well as water, resulting in the formation of HO^\bullet while the conduction band electrons are good reductants reducing the dissolved oxygen to $\text{O}_2^{\bullet-}$ (Munter, 2001).

The study of Elmolla and Chaudhuri (2011) examined the feasibility of using combined TiO_2 photocatalysis (UV/ TiO_2 / H_2O_2) and sequencing batch biological reactor (SBR) process for the treatment of an antibiotic wastewater containing amoxicillin and cloxacillin. The complete removal of these compounds was observed at TiO_2 and H_2O_2 doses of 1000 and 250 mg L^{-1} , respectively. Amoxicillin was also completely removed from urban wastewater treatment plant effluent using $[\text{TiO}_2] = 0.8 \text{ g L}^{-1}$ after 120 min of treatment as reported by Rizzo et al. (2009). Ofloxacin in wastewater samples was removed by 60% using $[\text{TiO}_2] = 3 \text{ g L}^{-1}$ (Michael et al., 2010) while Hapeshi et al. (2010) reported that the DOC of a solution contained ofloxacin at 10 mg L^{-1} was reduced by 79% after 120 min of photocatalytic treatment using $[\text{TiO}_2] = 250 \text{ mg L}^{-1}$ and $[\text{H}_2\text{O}_2] = 0.07 \text{ mmol L}^{-1}$.

Besides some drawbacks of the heterogeneous photocatalysis (e.g. the rather small quantum yield of the process; the relatively narrow light-response range of TiO_2 ; the need of post-separation and recovery of the catalyst particles from the reaction mixture in aqueous slurry systems), TiO_2 seems to possess some interesting features, such as high chemical stability in a wide pH range, strong resistance to chemical breakdown and photocorrosion, commercial availability and good performance. The catalyst is also cheap and can be reused (Andreozzi et al., 1999; Malato et al., 2009). The

properties of antibiotics to be treated such as pK_a and molecular structure will determine not only the efficiency of their photocatalytic degradation but also the mechanisms of the oxidation products formation (i.e. contribution of HO^\bullet radical and valence band holes oxidation pathway).

2.4.4. Sonolysis

Ultrasound irradiation or sonolysis is a relatively new process in water and wastewater treatment and therefore, has unsurprisingly received less attention than other AOPs. This is also reflected by the small number of publications concerning the treatment of pharmaceutical compounds. Ultrasound enhances chemical and physical changes in a liquid medium through the generation and subsequent destruction of cavitation bubbles. These bubbles grow over a period of a few cycles to an equilibrium size for the particular frequency applied. It is the fate of these bubbles when they collapse in succeeding compression cycles that generates the energy for chemical and mechanical effects (Parsons, 2004). The sonochemical degradation in aqueous phase involves several reaction pathways and zones such as pyrolysis inside the bubble and/or at the bubble-liquid interface and hydroxyl radical-mediated reactions at the bubble-liquid interface and/or in the liquid bulk. Pyrolytic reactions inside or near the bubble as well as solution radical chemistry are the two major pathways of sonochemical degradation (Emery et al., 2005).

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

It is important to note that there is limited literature (Hernández-Sancho et al., 2010; Mahamuni and Adewuyi, 2010; Jordá et al., 2011; Hollender et al., 2009; Michael et al., 2012b) dealing with advanced wastewater treatment process economics although this aspect is a very important issue.

2.5. Effect of disinfection on antibiotics removal

2.5.1. Chlorination

Limited studies have focused on the removal of antibiotics during wastewater treatment with chlorine. Chlorination is by far the most common method of wastewater disinfection and is used worldwide for the disinfection of pathogens before discharge into receiving streams, rivers or oceans. From the chlorinated species, hypochlorite (ClO^-) has the highest standard oxidation potential ($E_0 = 1.48 \text{ V}$), followed by chlorine gas ($E_0 = 1.36 \text{ V}$) and chlorine dioxide ($E_0 = 0.95 \text{ V}$) (Homem and Santos, 2011). The two major disadvantages of using chlorine based disinfectants are (i) the safety hazards associated with storage, transportation and handling of chlorine, and (ii) the potential formation of disinfection by-products.

The effective removal of antibiotics by chlorination from wastewater requires sufficient free chlorine concentration and contact time. For example, cephalixin which was

removed by 91% in activated sludge treatment at the Stanley WWTP was further removed in the following disinfection process by 99%, resulting in a total removal of 100% in the Stanley WWTP whole treatment process (Li and Zhang, 2011). Li and Zhang (2011) also reported that during chlorine disinfection process roxithromycin was eliminated by a further 18% (total removal 53%), erythromycin-H₂O by 24% (total removal 43%), sulfamethoxazole by 27% (total removal 73%) and trimethoprim by 40% (total removal 65%).

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 (¹O₂), hydroxyl radicals (HO•) or alkyl peroxy radicals (•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 11,000–30,000 mJ cm², 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 (18,868 M⁻¹ cm⁻¹).

Another study of photolysis was conducted by Arslan Alaton and Dogruel (2004) in which penicillin in the form of formulation effluent with total COD = 1555 mg L⁻¹ was treated under UV irradiation or UV combined with H₂O₂. In this study, the removal efficiency was very low compared to the others described above (COD removal max = 22% and TOC removal max = 10% with 30 and 40 mM of peroxide respectively) and this may be attributed to the complexity of the formulation effluent (high COD and TOC values). Zuccato *et al.* (2010) also reported complete elimination of amoxicillin in Varese WWTP with UV-light treatment. The addition of H₂O₂ to UV has proven to be more efficient in removing antibiotics than UV alone, and

lower fluence doses need to be applied for the same removal (Kim *et al.*, 2009; Rosario-Ortiz *et al.*, 2010; Yuan *et al.*, 2011).

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

3. Concluding 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-Minh *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 behavior of antibiotics under both conventional sewage treatment and advanced treatment processes and to gain more knowledge on the elimination processes

within the UWTs including sorption onto sewage sludge. Furthermore, studies should provide all basic treatment plant operational parameters since these are essential for later comparison or assessments.

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

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

Finally, evaluation of the negative impacts (i.e. antibiotic bacteria and resistance genes evolution, toxicity on organisms and plants) caused by the presence of antibiotics in the

environment is considered as a necessity in order to reduce the risk for humans.

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Appendix A

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 (Stage 1: CAS; Stage 2: 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)	Pre-treatment; 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

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Supporting Information – (continued)

Reference	Location	Main treatment steps
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 Golet et al., 2003	Glatt Valley Watershed (Switzerland) Zurich–Werdholzli (Switzerland)	CAS Primary treatment (screens; combined grid; fat removal tank; primary clarification); CAS (SRT = 11 days); Denitrification; Flocculation–filtration.
Gros et al., 2006 Gulkowska et al., 2008	Croatia Hong Kong and Shenzhen (China) (Wan Chai, Shatin, Tai Po, Stonecutters Island, Nan Shan)	CAS 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 Kasprzyk-Hordern et al., 2009 Kovalova et al., 2012	Wisconsin (USA) South Wales (England) Switzerland	CAS Cilfynydd: Trickling filter beds; Coslech: CAS 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·m ⁻² ·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

Supporting Information – (continued)

Reference	Location	Main treatment steps
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). 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.
Reif et al., 2008	Spain	Primary treatment (screening and sedimentation); CAS; Tertiary treatment; Disinfection (UWTP I: chlorination; UWTP II: UV).
Renew and Huang, 2004	California (UWTP I) and Arizona (UWTP II) (Georgia)	Primary treatment (coarse screening; preliminary clarification); CAS and trickling filter system; High-pressure 254 nm UV disinfection.
Roberts and Thomas, 2006	Howdon (UK)	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
Sahar et al., 2010	Tel-Aviv (Israel)	na
Spongberg and Witter, 2008	Northwest Ohio (USA)	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].
Sui et al., 2010	Beijing (China)	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).
Tambosi et al., 2010	Aachen (Germany)	Primary treatment (screen; aerated grid-removal tank; primary clarifier); CAS; Phosphate removal; Nitrification–denitrification.
Ternes et al., 2007	Braunschweig (Germany)	Primary treatment; CAS (SRT = 12.5 days)
Watkinson et al., 2007	Brisbane (Australia)	na
Watkinson et al., 2009	South-East Queensland (Australia)	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
Xia et al., 2012	China	Primary treatment; Secondary treatment processes
Xiao et al., 2008	Gao Beidian (Beijing, China)	Primary treatment; Secondary treatment processes

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Supporting Information – (continued)

Reference	Location	Main treatment steps
Xu et al., 2007	Guangzhou and Hong Kong (South China) (Kaifuqu, Liede, New Territory, Kowloon)	Kaifuqu: Primary treatment; CAS; Chlorination. Liede: Primary treatment; Oxidation ditch; UV. New Territory: Primary treatment; CAS. Kowloon: Primary treatment; Chemically enhanced; Chlorination.
Yang and Carlson, 2004 Yang et al., 2005	Northern Colorado (USA) Fort Collins (Colorado)	Na 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).

REFERENCES

- Abegglen, C., Joss, A., McArdell, C.S., Fink, G., Schlüsener, M.P., Ternes, T.A., Siegrist, H., 2009. The fate of selected micropollutants in a single-house MBR. *Water Research* 43, 2036–2046.
- Adams, C., Asce, M., Wang, Y., Loftin, K., Meyer, M., 2002. Removal of antibiotics from surface and distilled water in conventional water treatment processes. *Journal of Environmental Engineering* 128, 253–260.
- Aksu, Z., Tunç, O., 2005. Application of biosorption for penicillin G removal: comparison with activated carbon. *Process Biochemistry* 40, 831–847.
- Alturki, A.A., Tadkaew, N., McDonald, J.A., Khan, S.J., Price, W.E., Nghiem, L.D., 2010. Combining MBR and NF/RO membrane filtration for the removal of trace organics in indirect potable water reuse applications. *Journal of Membrane Science* 365, 206–215.
- Andreozzi, R., Caprio, V., Insola, A., Marotta, R., 1999. Advanced oxidation processes (AOP) for water purification and recovery. *Catalysis Today* 53, 51–59.
- Andreozzi, R., Canterino, M., Marotta, R., Paxeus, N., 2005. Antibiotic removal from wastewaters: the ozonation of amoxicillin. *Journal of Hazardous Materials* 122, 243–250.
- Arnold, W.A., McNeill, K., 2007. In: Petrović, M., Barceló, D. (Eds.), *Transformation of Pharmaceuticals in the Environment: Photolysis and Other Abiotic Processes*. *Comprehensive Analytical Chemistry* 50, pp. 361–385. Chapter 3.2.
- Arslan Alaton, I., Dogruel, S., 2004. Pre-treatment of penicillin formulation effluent by advanced oxidation processes. *Journal of Hazardous Materials B112*, 105–113.
- Arslan Alaton, I., Dogruel, S., Baykal, E., Gerone, G., 2004. Combined chemical and biological oxidation of penicillin formulation effluent. *Journal of Environmental Management* 73, 155–163.
- Auerbach, E.A., Seyfried, E.E., McMahon, K.D., 2007. Tetracycline resistance genes in activated sludge wastewater treatment plants. *Water Research* 41, 1143–1151.
- Backhaus, T., Grimme, L.H., 1999. The toxicity of antibiotic agents to the luminescent bacterium *Vibrio fischeri*. *Chemosphere* 38, 3291–3301.
- Balcioğlu, I.A., Otker, M., 2003. Treatment of pharmaceutical wastewater containing antibiotics by O₃ and O₃/H₂O₂ processes. *Chemosphere* 50, 85–95.
- Batt, A.L., Kim, S., Aga, D.S., 2007. Comparison of the occurrence of antibiotics in four full-scale wastewater treatment plants with varying designs and operations. *Chemosphere* 68, 428–435.
- Batt, A.L., Snow, D.D., Aga, D.S., 2006. Occurrence of sulphonamide antimicrobials in private water wells in Washington County, Idaho, USA. *Chemosphere* 64, 1963–1971.
- Bautitz, I.R., Nogueira, R.F.P., 2007. Degradation of tetracycline by photo-Fenton process—solar irradiation and matrix effects. *Journal of Photochemistry and Photobiology A: Chemistry* 187, 33–39.
- Bendz, D., Paxéus, N.A., Ginn, T.R., Loge, F.J., 2005. Occurrence and fate of pharmaceutically active compounds in the environment, a case study: Hoje River in Sweden. *Journal of Hazardous Materials* 122, 195–204.
- Boehler, M., Zwicklenpflug, B., Hollender, J., Ternes, T., Joss, A., Siegrist, H., 2012. Removal of micropollutants in municipal wastewater treatment plants by powder-activated carbon. *Water Science and Technology* 66, 2115–2121.
- Boxall, A.B.A., 2004. The environmental side effects of medication. *EMBO Reports* 5, 1110–1116.
- Brown, K.D., Kulis, J., Thomson, B., Chapman, T.H., Mawhinney, D.B., 2006. Occurrence of antibiotics in hospital, residential, and dairy effluent, municipal wastewater, and the Rio Grande in New Mexico. *Science of the Total Environment* 366, 772–783.
- Carballa, M., Omil, F., Lema, J.M., Llopart, M., Garcia-Jares, C., Rodriguez, I., Gomez, M., Ternes, T., 2004. Behavior of pharmaceuticals, cosmetics and hormones in a sewage treatment plant. *Water Research* 38, 2918–2926.
- Castiglioni, S., Pomati, F., Miller, K., Burns, B.P., Zuccato, E., Calamari, D., Neilan, B.A., 2008. Novel homologs of the multiple resistance regulator marA in antibiotic-contaminated environments. *Water Research* 42, 4271–4280.
- Cha, J.M., Yang, S., Carlson, K.H., 2006. Trace determination of β -lactam antibiotics in surface water and urban wastewater using liquid chromatography combined with electrospray tandem mass spectrometry. *Journal of Chromatography A* 1115, 46–57.
- Choi, K.J., Kim, S.G., Kim, C.W., Kim, S.H., 2007. Determination of antibiotic compounds in water by on-line SPE-LC/MSD. *Chemosphere* 66, 977–984.
- Clara, M., Strenn, B., Gans, O., Martinez, E., Kreuzinger, N., Kroiss, H., 2005. Removal of selected pharmaceuticals, fragrances and endocrine disrupting compounds in a membrane bioreactor and conventional wastewater treatment plants. *Water Research* 39, 4797–4807.
- Costanzo, S.D., Murby, J., Bates, J., 2005. Ecosystem response to antibiotics entering the aquatic environment. *Marine Pollution Bulletin* 51, 218–223.

- Czekalski, N., Berthold, T., Caucci, S., Egli, A., Bürgmann, H., 2012. Increased levels of multiresistant bacteria and resistance genes after wastewater treatment and their dissemination into Lake Geneva, Switzerland. *Frontiers in Microbiology* 3 (106), 1–18.
- Davies, J., Davies, D., 2010. Origins and evolution of antibiotic resistance. *Microbiology and Molecular Biology Reviews* 74 (3), 417–433.
- De la Cruz, N., Giménez, J., Esplugas, S., Grandjean, D., de Alencastro, L.F., Pulgarín, C., 2012. Degradation of 32 emergent contaminants by UV and neutral photo-fenton in domestic wastewater effluent previously treated by activated sludge. *Water Research* 46, 1947–1957.
- Diaz-Cruz, M.S., Lopez de Alda, M.J., Barcelo, D., 2003. Environmental behavior and analysis of veterinary and human drugs in soils, sediments and sludge. *TrAC-Trends in Analytical Chemistry* 22 (6), 340–351.
- Dickenson, E.R.V., Drewes, J.E., Sedlak, D.L., Wert, E.C., Snyder, S.A., 2009. Applying surrogates and indicators to assess removal efficiency of trace organic chemicals during chemical oxidation of wastewaters. *Environmental Science and Technology* 43 (16), 6242–6247.
- Dodd, M.C., Buffle, M., von Gunten, U., 2006. Oxidation of antibiotic molecules by aqueous ozone: moiety-specific reaction kinetics and application to ozone-based wastewater treatment. *Environmental Science and Technology* 40, 1969–1977.
- Dodd, M.C., Kohler, H.E., Gunten, U.V., 2009. Oxidation of antibiotic compounds by ozone and hydroxyl radical: elimination of biological activity during aqueous ozonation processes. *Environmental Science and Technology* 43, 2498–2504.
- Dodd, M.C., 2012. Potential impacts of disinfection processes on elimination and deactivation of antibiotic resistance genes during water and wastewater treatment. *Journal of Environmental Monitoring* 14, 1754–1771. <http://dx.doi.org/10.1039/c2em00006g>.
- Dolar, D., Gros, M., Rodriguez-Mozaz, S., Moreno, J., Comas, J., Rodriguez-Roda, I., Barceló, D., 2012. Removal of emerging contaminants from municipal wastewater with an integrated membrane system, MBR-RO. *Journal of Hazardous Materials* 239–240, 64–69. <http://dx.doi.org/10.1016/j.jhazmat.2012.03.029>.
- Drewes, J.E., 2008. Removal of Pharmaceutical Residues During Wastewater Treatment. In: *Comprehensive Analytical Chemistry* 50. Chapter 4.1.
- Drewes, J.E., Bellona, C., Oedekoven, M., Xu, P., Kim, T.U., Amy, G., 2006. Rejection of wastewater-derived micropollutants in high-pressure membrane applications leading to indirect potable reuse. *Environmental Progress* 24 (4), 400–409.
- Elmolla, E.S., Chaudhuri, M., 2011. The feasibility of using combined TiO₂ photocatalysis-SBR process for antibiotic wastewater treatment. *Desalination* 272, 218–224.
- Emery, R.J., Papadaki, M., Freitas dos Santos, L.M., Mantzavinos, D., 2005. Extent of sonochemical degradation and change of toxicity of a pharmaceutical precursor (triphenylphosphineoxide) in water as a function of treatment conditions. *Environment International* 31, 207–211.
- Fatta-Kassinos, D., Hapeshi, E., Achilleos, A., Meric, S., Gros, M., Petrovic, M., Barcelo, D., 2010. Existence of pharmaceutical compounds in tertiary treated urban wastewater that is utilized for reuse applications. *Water Resources Management* 25, 1183–1193.
- Fatta-Kassinos, D., Meric, S., Nikolaou, A., 2011a. Pharmaceutical residues in environmental waters and wastewater: current state of knowledge and future research. *Analytical and Bioanalytical Chemistry* 399, 251–275.
- Fatta-Kassinos, D., Vasquez, M.I., Kümmerer, K., 2011b. Transformation products of pharmaceuticals in surface waters and wastewater formed during photolysis and advanced oxidation processes – degradation, elucidation of byproducts and assessment of their biological potency. *Chemosphere* 85, 693–709.
- Froehner, K., Backhaus, T., Grimme, L.H., 2000. Bioassays with *Vibrio fischeri* for the assessment of delayed toxicity. *Chemosphere* 40, 821–828.
- Gao, P., Munir, M., Xagorarakis, I., 2012. Correlation of tetracycline and sulfonamide antibiotics with corresponding resistance genes and resistant bacteria in a conventional municipal wastewater treatment plant. *Science of the Total Environment* 421–422, 173–183. <http://dx.doi.org/10.1016/j.scitotenv.2012.01.061>.
- Göbel, A., Mc Ardell, C.S., Joss, A., Siegrist, H., Giger, W., 2007. Fate of sulfonamides, macrolides, and trimethoprim in different wastewater treatment technologies. *Science of the Total Environment* 372, 361–371.
- Göbel, A., Thomsen, A., Mc Ardell, C.S., Joss, A., Giger, W., 2005. Occurrence and sorption behavior of sulfonamides, macrolides, and trimethoprim in activated sludge treatment. *Environmental Science and Technology* 39, 3981–3989.
- Golet, E.M., Alder, A.C., Giger, W., 2002. Environmental exposure and risk assessment of fluoroquinolone antibiotic agents in wastewater and river water of the Glatt Valley Watershed, Switzerland. *Environmental Science and Technology* 36, 3645–3651.
- Golet, E.M., Xifra, I., Siegrist, H., Alder, A.C., Giger, W., 2003. Environmental exposure assessment of fluoroquinolone antibiotic agents from sewage to soil. *Environmental Science and Technology* 37, 3243–3249.
- Goslich, R., Dillert, R., Bahnmann, D., 1997. Solar water treatment: principles and reactors. *Water Science and Technology* 35 (4), 137–148.
- Gros, M., Petrovic, M., Barceló, D., 2006. Development of a multi-residue analytical methodology based on liquid chromatography-tandem mass spectrometry (LC-MS/MS) for screening and trace level determination of pharmaceuticals in surface and wastewaters. *Talanta* 70, 678–690.
- Guardabassi, L., Wong, D.M.A., Dalsgaard, A., 2002. The effects of tertiary wastewater treatment on the prevalence of antimicrobial-resistant bacteria. *Water Research* 36, 1955–1964.
- Gullberg, E., Cao, S., Berg, O.G., Ilbäck, C., Sandegren, L., Hughes, D., Andersson, D.I., 2012. Selection of resistant bacteria at very low antibiotic concentrations. *PLoS Pathogens* 7, 1–9.
- Gulkowska, A., Leung, H.W., So, M.K., Taniyasu, S., Yamashita, N., Yeung, L.W.Y., Richardson, B.J., Lei, A.P., Giesy, J.P., Lam, P.K.S., 2008. Removal of antibiotics from wastewater by sewage treatment facilities in Hongkong and Shenzhen, China. *Water Research* 42, 395–403.
- Hapeshi, E., Achilleos, A., Vasquez, M.I., Michael, C., Xekoukoulotakis, N.P., Mantzavinos, D., Kassinos, D., 2010. Drugs degrading photocatalytically: kinetics and mechanisms of ofloxacin and atenolol removal on titania suspensions. *Water Research* 44, 1737–1746.
- Heberer, T., 2002. Occurrence, fate, and removal of pharmaceutical residues in the aquatic environment: a review of recent research data. *Toxicology Letters* 131, 5–17.
- Hernández-Sancho, F., Molinos-Senante, M., Sala-Garrido, R., 2010. Economic valuation of environmental benefits from wastewater treatment processes: an empirical approach for Spain. *Science of the Total Environment* 408, 953–957.
- Herrmann, J.M., Guillard, C., Pichat, P., 1993. Heterogeneous photocatalysis: an emerging technology for water treatment. *Catalysis Today* 17, 7–20.

- Hirsch, R., Ternes, T., Haberer, K., Kratz, K.L., 1999. Occurrence of antibiotics in the aquatic environment. *Science of the Total Environment* 225, 109–118.
- Hollender, J., Singer, H., McArdell, C., 2008. Polar organic micropollutants in the water cycle. In: Hlavinec, P., et al. (Eds.), *Dangerous Pollutants (Xenobiotics) in Urban Water Cycle*. Springer, pp. 103–116.
- Hollender, J., Zimmermann, S.G., Koepke, S., Krauss, M., McArdell, C.S., Ort, C., Singer, H., von Gunten, U., Siegrist, H., 2009. Elimination of organic micropollutants in a municipal wastewater treatment plant upgraded with a full-scale post-ozonation followed by sand filtration. *Environmental Science and Technology* 43, 7862–7869.
- Homem, V., Santos, L., 2011. Degradation and removal methods of antibiotics from aqueous matrices – a review. *Journal of Environmental Management* 92, 2304–2347.
- Huber, M.M., Göbel, A., Joss, A., Hermann, N., Löffler, D., McArdell, C.S., Ried, A., Siegrist, H., Ternes, T.A., von Gunten, U., 2005. Oxidation of pharmaceuticals during ozonation of municipal wastewater effluents: a pilot study. *Environmental Science and Technology* 39, 4290–4299.
- Jelic, A., Gros, M., Ginebreda, A., Cespedes-Sanchez, R., Ventura, F., Petrovic, M., Barcelo, D., 2011. Occurrence, partition and removal of pharmaceuticals in sewage water and sludge during wastewater treatment. *Water Research* 45, 1165–1176.
- Ji, L.G., Chen, W., Duan, L., Zhu, D., 2009. Mechanisms for strong adsorption of tetracycline to carbon nanotubes: a comparative study using activated carbon and graphite as adsorbents. *Environmental Science and Technology* 43, 2322–2327.
- Jiao, S., Zheng, S., Yin, D., Wang, L., Chen, L., 2008. Aqueous photolysis of tetracycline and toxicity of photocatalytic products to luminescent bacteria. *Chemosphere* 73, 377–382.
- Jones, O.A.H., Voulvoulis, N., Lester, J.N., 2001. Human pharmaceuticals in the aquatic environment a review. *Environmental Science and Technology* 22, 1383–1394.
- Jones, O.A.H., Voulvoulis, N., Lester, J.N., 2007. The occurrence and removal of selected pharmaceutical compounds in a sewage treatment works utilising activated sludge treatment. *Environmental Pollution* 145 (3), 738–744.
- Jordá, L.S.J., Martín, M.M.B., Gómez, E.O., Reina, A.C., Sánchez, I.M.R., López, J.L.C., Pérez, J.A.S., 2011. Economic evaluation of the photo-Fenton process. Mineralization level and reaction time: the keys for increasing plant efficiency. *Journal of Hazardous Materials* 186, 1924–1929.
- Joss, A., Kellera, E., Aldera, A.C., Göbel, A., McArdell, C.S., Ternes, T., Siegrist, H., 2005. Removal of pharmaceuticals and fragrances in industrial wastewater treatment. *Water Research* 39, 3139–3152.
- Karthikeyan, K.G., Meyer, M.T., 2006. Occurrence of antibiotics in wastewater treatment facilities in Wisconsin, USA. *Science of the Total Environment* 361, 196–207.
- Kasprzyk-Hordern, B., Dinsdale, R.M., Guwy, A.J., 2009. The removal of pharmaceuticals, personal care products, endocrine disruptors and illicit drugs during wastewater treatment and its impact on the quality of receiving waters. *Water Research* 43, 363–380.
- Katsoyiannis, I.A., Canonica, S., Gunten, U., 2011. Efficiency and energy requirements for the transformation of organic micropollutants by ozone, O_3/H_2O_2 and UV/H_2O_2 . *Water Research* 45, 3811–3822.
- Kemper, N., 2008. Veterinary antibiotics in the aquatic and terrestrial environment. *Ecological Indicators* 8, 1–13.
- Khan, S.J., Ongerth, J.E., 2005. Occurrence and removal of pharmaceuticals at an Australian sewage treatment plant. *Water* 32 (4), 80–85.
- Kim, I., Yamashita, N., Tanaka, H., 2009. Performance of UV and UV/H_2O_2 processes for the removal of pharmaceuticals detected in secondary effluent of a sewage treatment plant in Japan. *Journal of Hazardous Materials* 166, 1134–1140.
- Kim, S., Eichhorn, P., Jensen, J.N., Weber, A.S., Aga, D., 2005. Removal of antibiotics in wastewater: effect of hydraulic and solid retention times on the fate of tetracycline in the activated sludge process. *Environmental Science and Technology* 39, 5816–5823.
- Kim, Y., Choi, K., Jung, J., Park, S., Kim, P.G., Park, J., 2007. Aquatic toxicity of acetaminophen, carbamazepine, cimetidine, diltiazem and six major sulfonamides, and their potential ecological risks in Korea. *Environment International* 33, 370–375.
- Kimura, K., Toshima, S., Amy, G., Watanabe, Y., 2004. Rejection of neutral endocrine disrupting compounds (EDCs) and pharmaceutical active compounds (PhACs) by RO membranes. *Journal of Membrane Science* 245, 71–78.
- Klamerth, N., Rizzo, L., Malato, S., Maldonado, M.I., Agüera, A., Fernández-Alba, A.R., 2010. Degradation of fifteen emerging contaminants at $\mu g L^{-1}$ initial concentrations by mild solar photo-Fenton in MWTP effluents. *Water Research* 44, 545–554.
- Klavarioti, M., Mantzavinos, D., Kassinos, D., 2009. Removal of residual pharmaceuticals from aqueous systems by advanced oxidation processes. *Environment International* 35, 402–417.
- Kolpin, D.W., Furlong, E.T., Meyer, M.T., Thurman, E.M., Zaugg, S.D., Barber, L.B., Buxton, H.T., 2002. Pharmaceuticals, hormones, and other organic wastewater contaminants in U.S. streams, 1999–2000: a national reconnaissance. *Environmental Science and Technology* 36, 1202–1211.
- Kosutic, K., Dolar, D., Asperger, D., Kunst, B., 2007. Removal of antibiotics from a model wastewater by RO/NF membranes. *Separation and Purification Technology* 53, 244–249.
- Kovalova, L., Siegrist, H., Singer, H., Wittmer, A., McArdell, C.S., 2012. Hospital wastewater treatment by membrane bioreactor: performance and efficiency for organic micropollutant elimination. *Environmental Science and Technology* 46, 1536–1545.
- Kümmerer, K., 2009. Antibiotics in the aquatic environment – a review – Part II. *Chemosphere* 75, 435.
- Lange, F., Cornelissen, S., Kubac, D., Sein, M.M., von Sonntag, J., Hannich, C.B., Golloch, A., Heipieper, H.J., Moder, M., von Sonntag, C., 2006. Degradation of macrolide antibiotics by ozone: a mechanistic case study with clarithromycin. *Chemosphere* 65 (1), 17–23.
- Längin, A., Alexy, R., König, A., Kümmerer, K., 2009. Deactivation and transformation products in biodegradability testing of β -lactams amoxicillin and piperacillin. *Chemosphere* 75, 347–354.
- Le Corre, S.K., Ort, C., Kateley, D., Allen, B., Escher, B.I., Keller, J., 2012. Consumption-based approach for assessing the contribution of hospitals towards the load of pharmaceutical residues in municipal wastewater. *Environment International* 45, 99–111.
- Lee, Y., Zimmermann, S.G., Kieu, A.T., von Gunten, U., 2009. Ferrate (Fe(VI)) application for municipal wastewater treatment: a novel process for Simultaneous micropollutant oxidation and phosphate removal. *Environmental Science and Technology* 43 (10), 3831–3838.
- Legrini, O., Oliveros, E., Braun, A.M., 1993. Photochemical processes for water treatment. *Chemical Reviews* 93, 671–698.
- Le-Minh, N., Khan, S.J., Drewes, J.E., Stuetz, R.M., 2010. Fate of antibiotics during municipal water recycling treatment processes. *Water Research* 44, 4295–4323.
- Li, B., Zhang, T., 2011. Mass flows and removal of antibiotics in two municipal wastewater treatment plants. *Chemosphere* 83, 1284–1289. <http://dx.doi.org/10.1016/j.chemosphere.2011.03.002>.
- Li, B., Zhang, T., Xua, Z., Fang, H.H.P., 2009. Rapid analysis of 21 antibiotics of multiple classes in municipal wastewater using

- ultra performance liquid chromatography-tandem mass spectrometry. *Analytica Chimica Acta* 645, 64–72.
- Li, S., Li, X., Wang, D., 2004. Membrane (RO-UF) filtration for antibiotic wastewater treatment and recovery of antibiotics. *Separation and Purification Technology* 34, 109–114.
- Lin, A.Y.C., Yu, T.H., Lateef, S.K., 2009a. Removal of pharmaceuticals in secondary wastewater treatment processes in Taiwan. *Journal of Hazardous Materials* 167, 1163–1169.
- Lin, A.Y.C., Lin, C., Chiou, J., Hong, P.K.A., 2009b. O₃ and O₃/H₂O₂ treatment of sulfonamide and macrolide antibiotics in wastewater. *Journal of Hazardous Materials* 171, 452–458.
- Lindberg, R.H., Wennberg, P., Johansson, M.I., Tysklind, M., Andersson, B.A.V., 2005. Screening of human antibiotic. Substances and determination of weekly mass flows in five sewage treatment plants in Sweden. *Environmental Science and Technology* 39, 3421–3429.
- Litter, M.I., 2005. Introduction to photochemical advanced oxidation processes for water treatment. *Environmental Chemistry* 2 (Pt. M), 325–366.
- Löffler, D., Ternes, T.A., 2003. Analytical method for the determination of the aminoglycoside gentamicin in hospital wastewater via liquid chromatography electrospray-tandem mass spectrometry. *Journal of Chromatography A* 1000 (1–2), 583–588.
- Loganathan, B., Phillips, M., Mowery, H., Jones-Lepp, T.L., 2009. Contamination profiles and mass loadings of macrolide antibiotics and illicit drugs from a small urban wastewater treatment plant. *Chemosphere* 75, 70–77.
- Macri, A., Stazi, A.V., Dojmi di Delupis, G., 1988. Acute toxicity of furazolidone on *Artemia salina*, *Daphnia magna*, and *Culex pipiens molestus* larvae. *Ecotoxicology and Environmental Safety* 16, 90–94.
- Mahamuni, N.N., Adewuyi, Y.G., 2010. Advanced oxidation processes (AOPs) involving ultrasound for wastewater treatment: a review with emphasis on cost estimation. *Ultrasonics Sonochemistry* 17, 990–1003.
- Mahugo-Santana, C., Sosa-Ferrera, Z., Torres-Padrón, M.E., Santana-Rodríguez, J.J., 2010. Analytical methodologies for the determination of nitroimidazole residues in biological and environmental liquid samples: a review. *Analytica Chimica Acta* 665, 113–122.
- Malato, S., Fernández-Ibáñez, P., Maldonado, M.I., Blanco, J., Gernjak, W., 2009. Decontamination and disinfection of water by solar photocatalysis: recent overview and trends. *Catalysis Today* 147, 1–59.
- McArdell, C.S., Molnar, E., Suter, M.J.F., Giger, W., 2003. Occurrence and fate of macrolide antibiotics in wastewater treatment plants and in the Glatt Valley Watershed, Switzerland. *Environmental Science and Technology* 37, 5479–5486.
- McArdell, C.S., Kovalova, L., Siegrist, H., Kienle, C., Moser, R., Schwartz, T., 2011. Input and Elimination of Pharmaceuticals and Disinfectants from Hospital Wastewater. Final project report. Eawag, Dübendorf, Switzerland. accessible at <http://tinyurl.com/eawag-spitalabwasser>.
- Melero, J.A., Calleja, G., Martínez, F., Molina, R., Pariente, M.I., 2007. Nanocomposite Fe₂O₃/SBA-15: an efficient and stable catalyst for the catalytic wet peroxidation of phenolic aqueous solutions. *Chemical Engineering Journal* 131, 245–256.
- Miao, X.S., Bishay, F., Chen, M., Metcalfe, C.D., 2004. Occurrence of antimicrobials in the final effluents of wastewater treatment plants in Canada. *Environmental Science and Technology* 38, 3533–3541.
- Michael, I., Hapeshi, E., Michael, C., Fatta-Kassinos, D., 2010. Solar Fenton and solar TiO₂ catalytic treatment of ofloxacin in secondary treated effluents: evaluation of operational and kinetic parameters. *Water Research* 44, 5450–5462.
- Michael, I., Hapeshi, E., Osorio, V., Pérez, S., Petrović, M., Zapata, A., Malato, S., Barceló, D., Fatta-Kassinos, D., 2012a. Solar photocatalytic treatment of trimethoprim in four environmental matrices at a pilot scale: transformation products and ecotoxicity evaluation. *Science of the Total Environment* 430, 167–173.
- Michael, I., Hapeshi, E., Michael, C., Varela, A.R., Kyriakou, S., Manaia, C.M., Fatta-Kassinos, D., 2012b. Solar Fenton process on the abatement of antibiotics at a pilot plant scale: degradation kinetics, ecotoxicity and phytotoxicity assessment and removal of antibiotic resistant enterococci. *Water Research* 46, 5621–5634.
- Migliore, L., Cozzolino, S., Fiori, M., 2003. Phytotoxicity to and uptake of enrofloxacin in crop plants. *Chemosphere* 52, 1233–1244.
- Morse, A., Jackson, A., 2004. Fate of amoxicillin in two water reclamation systems. *Water, Air, & Soil Pollution* 157 (1–4), 117–132.
- Munter, R., 2001. Advanced oxidation processes-current status and prospects. *Proceedings of the Estonian Academy of Sciences Chemistry* 50, 59–80.
- Naddeo, V., Meric, S., Kassinos, D., Belgiorno, V., Guida, M., 2009. Fate of pharmaceuticals in contaminated urban wastewater effluent under ultrasonic irradiation. *Water Research* 43, 4019–4027.
- Nakada, N., Shinohara, H., Murata, A., Kiri, K., Managakia, S., Sato, N., Takada, H., 2007. Removal of selected pharmaceuticals and personal care products (PPCPs) and endocrine-disrupting chemicals (EDCs) during sand filtration and ozonation at a municipal sewage treatment plant. *Water Research* 41, 4373–4382.
- Paillet, J.Y., Krein, A., Pfister, L., Hoffmann, L., Guignard, C., 2009. Solid phase extraction coupled to liquid chromatography-tandem mass spectrometry analysis of sulfonamides, tetracyclines, analgesics and hormones in surface water and wastewater in Luxembourg. *Science of the Total Environment* 407, 4736–4743.
- Parsons, S., 2004. *Advanced Oxidation Processes for Water and Wastewater Treatment*. IWA Publishing.
- Peng, X., Wang, Z., Kuang, W., Tan, J., Li, K., 2006. A preliminary study on the occurrence and behavior of sulfonamides, ofloxacin and chloramphenicol antimicrobials in wastewaters of two sewage treatment plants in Guangzhou, China. *Science of the Total Environment* 371, 314–322.
- Perez, S., Eichhorn, P., Aga, D.S., 2005. Evaluating the biodegradability of sulfamethazine, sulfamethoxazole, sulfathiazole and trimethoprim at different stages of sewage treatment. *Environmental Toxicology and Chemistry* 24 (6), 1361–1367.
- Putra, E.K., Pranowo, R., Sunarso, J., Indraswati, N., Ismadji, S., 2009. Performance of activated carbon and bentonite for adsorption of amoxicillin from wastewater: mechanisms, isotherms and kinetics. *Water Research* 43, 2419–2430.
- Radjenovic, J., Godehardt, M., Petrovic, M., Hein, A., Farre, M., Jekel, M., Barcelo, D., 2009a. Evidencing generation of persistent ozonation products of antibiotics roxithromycin and trimethoprim. *Environmental Science and Technology* 43, 6808–6815.
- Radjenovic, J., Petrovic, M., Barcelo, D., 2009b. Fate and distribution of pharmaceuticals in wastewater and sewage sludge of the conventional activated sludge (CAS) and advanced membrane bioreactor (MBR) treatment. *Water Research* 43, 831–841.
- Reif, R., Suárez, S., Omil, F., Lema, J.M., 2008. Fate of pharmaceuticals and cosmetic ingredients during the operation of a MBR treating sewage. *Desalination* 221, 511–517.

- Renew, J.E., Huang, C.H., 2004. Simultaneous determination of fluoroquinolone, sulfonamide, and trimethoprim antibiotics in wastewater using tandem solid phase extraction and liquid chromatography-electrospray mass spectrometry. *Journal of Chromatography A* 1042, 113–121.
- Richardson, S.D., Ternes, T.A., 2011. Water analysis: emerging contaminants and current issues. *Analytical Chemistry* 83 (12), 4614–4648.
- Rivera-Utrilla, J., Prados-Joya, G., Sánchez-Polo, M., Ferro-García, M.A., Bautista-Toledo, I., 2009. Removal of nitroimidazole antibiotics from aqueous solution by adsorption/bioadsorption on activated carbon. *Journal of Hazardous Materials* 170, 298–305.
- Rizzo, L., 2011. Bioassays as a tool for evaluating advanced oxidation processes in water and wastewater treatment. *Water Research* 45, 4311–4340.
- Rizzo, L., Meric, S., Guida, M., Kassinos, D., Belgiorno, V., 2009. Heterogenous photocatalytic degradation kinetics and detoxification of an urban wastewater treatment plant effluent contaminated with pharmaceuticals. *Water Research* 43, 4070–4078.
- Roberts, P.H., Thomas, K.V., 2006. The occurrence of selected pharmaceuticals in wastewater effluent and surface waters of the lower Tyne catchment. *Science of the Total Environment* 356, 143–153.
- Robertson, P.K.J., 1996. Semiconductor photocatalysis: an environmentally acceptable alternative production technique and effluent treatment process. *Journal of Cleaner Production* 4 (34), 203–212.
- Rogers, H.R., 1996. Sources, behaviour and fate of organic contaminants during sewage treatment and in sewage sludges. *Science of the Total Environment* 185, 3–26.
- Rosal, R., Rodríguez, A., Perdigón-Melón, J.A., Petre, A., García-Calvo, E., Gómez, M.J., Aguera, A., Fernández-Alba, A.R., 2010. Occurrence of emerging pollutants in urban wastewater and their removal through biological treatment followed by ozonation. *Water Research* 44, 578–588.
- Rosario-Ortiz, F.L., Wert, E.C., Snyder, S.A., 2010. Evaluation of UV/H₂O₂ treatment for the oxidation of pharmaceuticals in wastewater. *Water Research* 44, 1440–1448.
- Ryan, C.C., Tan, D.T., Arnold, W.A., 2011. Direct and indirect photolysis of sulfamethoxazole and trimethoprim in wastewater treatment plant effluent. *Water Research* 45, 1280–1286.
- Sahar, E., David, I., Gelman, Y., Chikurel, H., Aharoni, A., Messalem, R., Brenner, A., 2010. The use of RO to remove emerging micropollutants following CAS/UF or MBR treatment of municipal wastewater. *Desalination* 273 (1), 142–147.
- Santos, A., Yustos, P., Rodriguez, S., Simon, E., Garcia-Ochoa, F., 2007. Abatement of phenolic mixtures by catalytic wet oxidation enhanced by Fenton's pretreatment: effect of H₂O₂ dosage and temperature. *Journal of Hazardous Materials* 146, 595–601.
- Saritha, P., Aparna, C., Himabindu, V., Anjaneyulu, Y., 2007. Comparison of various advanced oxidation processes for the degradation of 4-chloro-2-nitrophenol. *Journal of Hazardous Materials* 149, 609–614.
- Schafer, A.I., Kalinowski, J., Puhler, A., 1994. Increased fertility of *Corynebacterium glutamicum* recipients in intergeneric matings with *Escherichia coli* after stress exposure. *Applied and Environmental Microbiology* 60 (2), 756–759.
- Schiavello, M., 1993. Some working principles of heterogeneous photocatalysis by semiconductors. *Electrochimica Acta* 38 (1), 11–14.
- Schwartz, T., Kohnen, W., Jansen, B., Obst, U., 2006. Detection of antibiotic-resistant bacteria and their resistance genes in wastewater, surface water and drinking water biofilms. *FEMS Microbiology Ecology* 43 (3), 325–355.
- Serrano, D., Suárez, S., Lema, J.M., Omil, F., 2011. Removal of persistent pharmaceutical micropollutants from sewage by addition of PAC in a sequential membrane bioreactor. *Water Research* 45, 5323–5333.
- Snyder, A.S., Westerhoff, P., Yoon, Y., Sedlak, D.L., 2003. Pharmaceuticals, personal care products, and endocrine disruptors in water: implications for the water industry. *Environmental Engineering Science* 20 (5), 449–469.
- Spongberg, A.L., Witter, J.D., 2008. Pharmaceutical compounds in the wastewater process stream in Northwest Ohio. *Science of the Total Environment* 397, 148–157.
- Sui, Q., Huang, J., Deng, S., Yu, G., Fan, Q., 2010. Occurrence and removal of pharmaceuticals, caffeine and DEET in wastewater treatment plants of Beijing, China. *Water Research* 44, 417–426.
- Tadkaew, N., Sivakumar, M., Khan, S.J., McDonald, J.A., Nghiem, L.D., 2011. Effect of mixed liquor pH on the removal of trace organic contaminants in a membrane bioreactor. *Bioresource Technology* 101, 1494–1500.
- Tambosi, J.L., de Sena, R.F., Favier, M., Gebhardt, W., José, H.J., Schröder, H.F., Moreira, R.F.P.M., 2010. Removal of pharmaceutical compounds in membrane bioreactors (MBR) applying submerged membranes. *Desalination* 26, 148–156.
- Tekin, H., Bilkay, O., Ataberk, S.S., Balta, T.H., Ceribasi, I.H., Sanin, F.D., 2006. Use of Fenton oxidation to improve the biodegradability of a pharmaceutical wastewater. *Journal of Hazardous Materials* 136, 258–265.
- Ternes, T.A., Stuber, J., Herrmann, N., McDowell, D., Ried, A., Kampmann, M., Teiser, B., 2003. Ozonation: a tool for removal of pharmaceuticals, contrast media and musk fragrances from wastewater? *Water Research* 37, 1976–1982.
- Ternes, T.A., Bonerz, M., Herrmann, N., Teiser, B., Andersen, H.R., 2007. Irrigation of treated wastewater in Braunschweig, Germany: an option to remove pharmaceuticals and musk fragrances. *Chemosphere* 66, 894–904.
- Thomulka, K.W., McGee, D.J., 1993. Detection of biohazardous materials in water by measuring bioluminescence reduction with the marine organism *Vibrio harveyi*. *Journal of Environmental Science & Health Part A* 28, 2153–2166.
- Trovó, A.G., Melo, S.A.S., Nogueira, R.F.P., 2008. Photodegradation of the pharmaceuticals amoxicillin, bezafibrate and paracetamol by the photo-Fenton process – application to sewage treatment plant effluent. *Journal of Photochemistry and Photobiology A: Chemistry* 198, 215–220.
- Watkinson, A.J., Murby, E.J., Costanzo, S.D., 2007. Removal of antibiotics in conventional and advanced wastewater treatment: implications for environmental discharge and wastewater recycling. *Water Research* 41, 4164–4176.
- Watkinson, A.J., Murby, E.J., Kolpin, D.W., Costanzo, S.D., 2009. The occurrence of antibiotics in an urban watershed: from wastewater to drinking water. *Science of the Total Environment* 407, 2711–2723.
- Westerhoff, P., Yoon, Y., Snyder, S., Wert, E., 2005. Fate of endocrine-disruptor, pharmaceutical, and personal care product chemicals during simulated drinking water treatment processes. *Environmental Science and Technology* 39, 6649–6663.
- Wollenberger, L., Halling-Sorensen, B., Kusk, K.O., 2000. Acute and chronic toxicity of veterinary antibiotics to *Daphnia magna*. *Chemosphere* 40, 723–730.
- Xekoukoulotakis, N.P., Drosou, C., Brebou, C., Chatzisyneon, E., Hapeshi, E., Fatta-Kassinos, D., Mantzavinos, D., 2010. Kinetics of UV-A/TiO₂ photocatalytic degradation and mineralization of the antibiotic sulfamethoxazole in aqueous matrices. *Catalysis Today* 161, 163–168.
- Xia, S., Jia, R., Feng, F., Xie, K., Li, H., Jing, D., Xu, X., 2012. Effect of solids retention time on antibiotics removal performance and microbial communities in an A/O-MBR process. *Bioresource Technology* 106, 36–43.

- Xiao, Y., Chang, H., Jia, A., Hu, J., 2008. Trace analysis of quinolone and fluoroquinolone antibiotics from wastewaters by liquid chromatography – electrospray tandem mass spectrometry. *Journal of Chromatography A* 1214, 100–108.
- Xu, P., Drewes, J.E., Kim, T.U., Bellona, C., Amy, G., 2006. Effect of membrane fouling on transport of organic contaminants in NF/RO membrane applications. *Journal of Membrane Science* 279 (1–2), 165–175.
- Xu, W., Zhang, G., Li, X., Zou, S., Li, P., Hu, Z., Li, J., 2007. Occurrence and elimination of antibiotics at four sewage treatment plants in the Pearl River Delta (PRD), South China. *Water Research* 41, 4526–4534.
- Yang, S., Carlson, K.H., 2004. Solid-phase extraction-high-performance liquid chromatography-ion trap mass spectrometry for analysis of trace concentrations of macrolides antibiotics in natural and waste water matrices. *Journal of Chromatography A* 1038, 141–155.
- Yang, S., Cha, J., Carlson, K., 2005. Simultaneous extraction and analysis of 11 tetracycline and sulfonamide antibiotics in influent and effluent domestic wastewater by solid-phase extraction and liquid chromatography-electrospray ionization tandem mass spectrometry. *Journal of Chromatography A* 1097, 40–53.
- Yang, Y., Wang, P., Shi, S., Liu, Y., 2009. Microwave enhanced Fenton-like process for the treatment of high concentration pharmaceutical wastewater. *Journal of Hazardous Materials* 168, 238–245.
- Yargeau, V., Leclair, C., 2008. Impact of operating conditions on decomposition of antibiotics during ozonation: a review. *Ozone: Science and Engineering* 30, 175–188.
- Yu, T., Lin, A.Y., Lateef, S.K., Lin, C., Yang, P., 2009. Removal of antibiotics and non-steroidal anti-inflammatory drugs by extended sludge age biological process. *Chemosphere* 77, 175–181.
- Yuan, F., Hu, C., Hu, X., Wie, D., Chen, Y., Qu, J., 2011. Photodegradation and toxicity changes of antibiotics in UV and UV/H₂O₂ process. *Journal of Hazardous Materials* 185, 1256–1263.
- Zhang, G., Ji, S., Xi, B., 2006. Feasibility study of treatment of amoxicillin wastewater with a combination of extraction, Fenton oxidation and reverse osmosis. *Desalination* 196, 32–42.
- Zhang, Y., Marrs, C.F., Simon, C., Xi, C., 2009. Wastewater treatment contributes to selective increase of antibiotic resistance among *Acinetobacter* spp. *Science of the Total Environment* 407, 3702–3706.
- Zorita, S., Mårtensson, L., Mathiasson, L., 2009. Occurrence and removal of pharmaceuticals in a municipal sewage treatment system in the south of Sweden. *Science of the Total Environment* 407, 2760–2770.
- Zuccato, E., Castiglioni, S., Bagnati, R., Melis, M., Fanelli, R., 2010. Source, occurrence and fate of antibiotics in the Italian aquatic environment. *Journal of Hazardous Materials* 179, 1042–1048.