Best available technologies and treatment trains to address current challenges in urban wastewater reuse for irrigation of crops in EU countries

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Abstract

Conventional urban wastewater treatment plants (UWTPs) are poorly effective in the removal of most contaminants of emerging concern (CECs), including antibiotics, antibiotic resistant bacteria and antibiotic resistance genes (ARB&ARGs). These contaminants result in some concern for the environment and human health, in particular if UWTPs effluents are reused for crop irrigation. Recently, stakeholders' interest further increased in Europe, because the European Commission is currently developing a regulation on water reuse. Likely, conventional UWTPs will require additional advanced treatment steps to meet water quality limits yet to be officially established for wastewater reuse. Even though it seems that CECs will not be included in the proposed regulation, the aim of this paper is to provide a technical contribution to this discussion as well as to support stakeholders by recommending possible advanced treatment options, in particular with regard to the removal of CECs and ARB&ARGs. Taking into account the current knowledge and the precautionary principle, any new or revised water-related Directive should address such contaminants. Hence, this review paper gathers the efforts of a group of international experts, members of the NEREUS COST Action ES1403, who for three years have been constructively discussing the efficiency of the best available technologies (BATs) for urban wastewater treatment to abate CECs and ARB&ARGs. In particular, ozonation, activated carbon adsorption, chemical disinfectants, UV radiation, advanced oxidation processes (AOPs) and membrane filtration are discussed with regard to their capability to effectively remove CECs and ARB&ARGs, as well as their advantages and drawbacks. Moreover, a comparison among the above-mentioned processes is performed for CECs relevant for crop uptake. Finally, possible treatment trains including the above-discussed BATs are discussed, issuing end-use specific recommendations which will be useful to UWTPs managers to select the most suitable options to be implemented at their own facilities to successfully address wastewater reuse challenges.

Keywords: activated carbon, advanced oxidation processes, antibiotic resistance, contaminants of emerging concern, disinfection, ozonation

List of abbreviations

ARB= antibiotic resistant bacteria

ARGs= antibiotic resistance genes

AOPs= advanced oxidation processes

BAC= biological activated carbon

CBZ= carbamazepine

CECs= contaminants of emerging concern

CPC= compound parabolic collector

DBPs= disinfection by products

DCF= diclofenac

DOC= dissolved organic carbon

ERY= erythromycin

FRC= free residual chlorine

GAC= granular activated carbon

$HO^{\bullet}$= hydroxyl radical

LRV= Log removal value

MDR= multi drug resistant

MF= microfiltration

NDMA= N-nitrosodimethylamine

NF= nanofiltration

PAC= powdered activated carbon

RO= reverse osmosis

TMP= transmembrane pressure

UF= ultrafiltration

SMX= sulfamethoxazole

UWTPs = urban wastewater treatment plants

1. Introduction

Wastewater reuse is one of the most important alternatives to conventional water sources to address water scarcity. As a matter of fact, around 1.2 billion people live in areas affected by serious water scarcity conditions (United Nations, 2014) and 1.8 billion people are expected to be living in countries or regions affected by water scarcity by 2025, according to United Nations reports (United Nations, 2014; FAO, 2014). Wastewater reuse for irrigation in agriculture is by far the most established end-use for reclaimed water (Dreschel et al., 2010a), in low-income countries as well as in arid and semi-arid ones (Dreschel et al., 2010b). However, whilst solving water scarcity, wastewater reuse can generate public health risks if treatment, storage and piping are not adequate. The main risk, in particular in low-income countries, is related to consumption of raw or undercooked vegetables contaminated with pathogenic microorganisms stemming from the use of untreated or poorly treated wastewater for crop irrigation (Fuhrimann et al., 2016). In countries of higher income level, wastewater reuse for irrigation is regulated, at least in some of them (Paranychianakis et al., 2015), and concerns tend to shift from microbial risk (effective disinfection processes are typically included in the treatment train) to contaminants of emerging concern (CECs), such as pesticides, pharmaceuticals, illicit drugs, synthetic and natural hormones, personal care products, and resistant microorganisms (i.e. antibiotic resistant bacteria and genes (ARB&ARGs)). However, neither the release of CECs from urban wastewater treatment plants (UWTPs) into the environment (except for Switzerland) nor their occurrence in wastewater for agricultural reuse has been regulated so far. CECs monitoring in UWTPs effluents to reuse for crop irrigation is one of the main debated issues among scientists, policy makers and stakeholders at EU level (Christou et al., 2017a, Piña et al., 2018, Rizzo et al., 2018; Deng et al., 2019) even in relation to the regulation for wastewater reuse which is about to be approved by the Parliament (European Parliament, 2019).

109 According to scientific literature, conventional treatment trains in UWTPs are poorly effective
110 to comprehensively remove CECs (Petrie et al., 2015; Falas et al., 2016; Krzeminski et al.,
111 2019), which can finally be released into the environment, constituting a particular concern
112 when effluents are reused for crop irrigation. To be able to meet stringent limits for wastewater
113 reuse as well as to effectively remove CECs, advanced treatment steps should be implemented
114 in conventional UWTPs (Krzeminski et al., 2019; Rizzo et al., 2019a). However, while the
115 effect of biological processes (Boshir Ahmed et al., 2017; Tiwari et al., 2017; Krzeminski et
116 al., 2019) and advanced treatment technologies (Miklos et al., 2018; von Gunten, 2018;
117 Roccaro, 2018; Marron et al., 2019; Rizzo et al., 2019a; Siegrist et al. 2019) on chemical CECs
118 has been reviewed in different papers, less information is available about ARB&ARGs and,
119 most importantly, on possible treatment trains combining several processes to successfully
120 address these challenges.
121
122 This review paper gathers the efforts of a group of international experts, members of the
123 NEREUS COST Action ES1403[1] "New and emerging challenges and opportunities in
124 wastewater reuse" (Fatta-Kassinos et al., 2015), who for three years have been constructively
125 discussing the effect of the best available technologies (BATs) for urban wastewater treatment
126 on CECs and ARB&ARGs. Accordingly, the objective of this paper is to introduce and discuss
127 the BATs for advanced treatment of urban wastewater, as well as possible treatment trains to
128 control the release of CECs, including ARB&ARGs, to produce wastewater for safe and
129 sustainable reuse practices in agriculture. In particular, the capability of ozonation, activated
130 carbon adsorption, chemical oxidants/disinfectants, UV radiation, advanced oxidation
131 processes (AOPs) and membrane filtration to abate CECs and ARB&ARGs are discussed

[1] COST Action ES1403 New and emerging challenges and opportunities in wastewater reuse (NEREUS), http://www.nereus-cost.eu.

including the advantages and drawbacks of these processes. Moreover, a comparison among the above-mentioned processes is performed for CECs relevant for crop uptake. It is noteworthy that only results from investigations at pilot or full-scale on real wastewater were considered. Subsequently, possible treatment trains including the above-discussed BATs are presented and recommended for possible application in the EU and other developed countries. Finally, possible advantages, drawbacks and recommendations of the proposed treatment trains are summarized.

2. Overview of the BATs for advanced treatment and reuse of urban wastewater: CECs abatement, effect on ARB&ARGs and process drawbacks

The occurrence of CECs into the environment is related to different human activities (Verlicchi et al., 2015; Bilal et al., 2019a, b) and it has been associated to biological adverse effects on living organisms such as toxicity, endocrine disruption and antibiotic resistance in microorganisms (Manaia, 2017; López-Pacheco et al., 2019; Ma et al., 2019). Specifically, several CECs have been found to increase the risks for human-health, because they finally cause imbalance to hormonal and male/female reproductive systems and different disorders, namely metabolism, neurological, and immunological ones (López-Pacheco et al., 2019; Pedrazzani et al., 2019; Rueda-Ruzafa et al., 2019).

In 2015, the European Commission established the EU Watch List (Decision 2015/495/EU) to monitor 17 CECs in water. The target CECs belong to different categories including antibiotics, estrogenic hormones, non-steroidal anti-inflammatory compounds, pesticides and herbicides, UV filters, and they were selected according to their potential to cause damage to aquatic environments and to pose a significant risk at European Union level, but for which monitoring data are insufficient to come to a conclusion regarding the actual posed risk.

UWTPs are recognized among the main anthropogenic sources for the release of CECs and ARB&ARGs into the environment, therefore, taking into account the environment and human health concerns related to their occurrence in UWTPs effluents and into the environment, different advanced treatment technologies have been investigated so far to find effective solutions to minimize their release. In the following sub-paragraphs, the BATs for advanced

161 treatment of urban wastewater are introduced to evaluate their effect one CECs and
162 ARB&ARGs. Possible advantages and drawbacks of these processes are also discussed
163 according to the relevant scientific literature.

164

165 ## 2.1 Ozonation

166 ### 2.1.1 Abatement of CECs

167 The oxidation capacity of the ozone process relies on the strong oxidation potential of both,
168 molecular ozone and HO radicals ($HO^{\bullet}$) (2.07 and 2.8 V against standard hydrogen electrode,
169 respectively). While ozone reacts selectively with compounds containing electron-rich moieties
170 (such as olefins, deprotonated amines or activated aromatics), $HO^{\bullet}$ exhibit a low selectivity and
171 fast reaction with a wide range of organic and inorganic compounds (von Sonntag, 2007).
172 Ozonation and other oxidation-based processes were originally applied for disinfection
173 purposes in drinking water treatment, but have been widely investigated for the abatement of
174 different CECs from urban wastewater since more than 10 years (Ternes et al., 2003). Based on
175 the reaction rate constants with ozone and $HO^{\bullet}$, CEC abatement can be predicted in municipal
176 wastewater (Lee et al. 2013). Hollender et al. (2009) and Bourgin et al. (2018) investigated the
177 abatement of 220-550 micropollutants at two full-scale UWTPs upgraded with ozonation
178 (followed by sand filtration). Compounds such as sulfamethoxazole, diclofenac, or
179 carbamazepine with high apparent second-order rate constants at pH 7 ($k_{O3,pH7}>10^3$) were
180 abated by more than 80% at a specific ozone dose of 0.4 g O_3/g dissolved organic carbon
181 (DOC). Compounds more refractory to oxidation by ozone ($k_{O3,pH7}=10^2$-10^3), such as
182 bezafibrate and benzotriazole, were abated by 80% only at a higher ozone dose (~0.6 g O_3/g
183 DOC). The high efficiency of ozonation in the abatement of CECs from wastewater was also
184 confirmed in other studies on a smaller group of compounds (e.g., Antoniou et al. 2013;
185 Magdeburg et al. 2014). After ozonation, a biological post-treatment (sand filter or biological

activated carbon (BAC) filter) is recommended to elimine possible negative ecotoxicological effects or by-products generated during ozonation (Von Gunten, 2018; Bacaro et al. 2019).

2.1.2 Effect on ARB&ARGs

Mechanisms for disinfection or inactivation of bacteria by ozone exposure include the disruption of bacterial cell walls (leading to the release of intracellular constituents), damage of nucleic acids (breaking aromatic structure), and breakage of carbon-nitrogen bonds of proteins leading to depolymerisation (Alexander et al., 2016, Michael-Kordatou et al., 2018). The inactivation efficiency by ozonation depends on the susceptibility of the target organism and ozone exposure, which is a function of the wastewater characteristics and transferred ozone dose. Unlike CECs, the effect of ozonation on ARB&ARGs has not been investigated systematically and thoroughly so far. Alexander et al. (2016) observed diverse patterns of resistances and susceptibilities of opportunistic bacteria and accumulations of some ARGs during ozone treatment (0.9 ± 0.1 g O_3/g DOC) of treated wastewater. Ozone affected microorganisms in different ways, with a high susceptibility of enterococci (almost 99% reduction) compared to *Pseudomonas aeruginosa*, that displayed only minor changes in abundance after treatment. The investigated ARGs demonstrated an even more diverse pattern with 2 orders of magnitude reduction of erythromycin resistance gene (*erm*B) but a simultaneous increase in the abundance of ARGs (*van*A, bla_{VIM}) within the surviving wastewater population. Ozonation operated at high contact time (40 min) with an ozone dose of 0.25 g O_3/g DOC was capable of inactivating total as well as antibiotic (sulfamethoxazole and trimethoprim) resistant *Escherichia coli* (*E. coli*), with the simultaneous reduction of the abundance of the examined genes (Iakovides et al., 2019). Accordingly, the studies published so far confirm that the ozonation process is effective in the inactivation of ARB and to some extent in the removal of ARGs (Lüddeke et al., 2014; Zhuang et al., 2015; Alexander et al.,

211 2016; Zheng et al., 2017; Sousa et al., 2017), but it seems that the process may also select for
212 bacterial population (Alexander et al., 2016; Sousa et al., 2017; Czekalski et al., 2016).
213 Regrowth of ARB during biological sand-filtration following ozonation was found to partly
214 compensate inactivation during ozonation (Czekalski et al., 2016). Moreover, mobile genetic
215 elements may reach pre-treatment levels after some days of storage (Sousa et al., 2017), which
216 can be of concern for wastewater reuse practice where treated effluents may be stored for some
217 days before use (Iakovides et al., 2019).

218

219 2.1.3 Formation of oxidation by-products

220 Ozonation can result in the formation of biologically potent (e.g. toxic, mutagenic) oxidation
221 by-products. Among them, N-nitrosodimethylamine (NDMA) and bromate are of particular
222 concern for human health because they are potentially carcinogenic. Therefore, NDMA and
223 bromate need to be measured to test the feasibility of ozonation as an option for advanced
224 wastewater treatment at a specific location (Schindler Wildhaber et al., 2015). Only if the
225 concentrations expected after dilution of discharged effluents are clearly below (potential)
226 drinking water standards (10 µg/L for bromate, 10 ng/L for NDMA, Bourgin et al., 2018),
227 ozonation is considered suitable. Bromate results from the reaction of O_3 and HO• with
228 bromide. NDMA can be formed from the reaction of amine precursors (e.g. containing
229 hydrazine, sulfamide, and dimethylamino functional groups) with generally low yields but that
230 can reach up to ≥ 50% in exceptional cases (Kosaka et al. 2009; Schmidt and Brauch 2008; von
231 Gunten et al. 2010; Krasner et al. 2013, Sgroi et al., 2014). Because precursors are mostly
232 unknown or unidentified in wastewater, the formation of NDMA cannot be excluded a priori.
233 NDMA can also already be present in the UWTP influent.

To minimize the release of biodegradable compounds including e.g. transformation products of CECs formed during ozonation, a subsequent treatment by biologically active sand filtration (or adsorption) is recommended. For the evaluation of the water quality after ozonation, specific and unspecific toxicity of the treated wastewater needs to be measured with bioassays (Schindler Wildhaber et al., 2015).

2.1.4 Application at full-scale as advanced treatment of urban wastewater

Ozonation is well established in drinking water treatment, but only recently has been applied at full-scale as advanced treatment of urban wastewater in Europe for the removal of CECs before discharge into the environment. In particular in Switzerland, ozonation is considered as one of the BATs to meet the requirement of the new Swiss water protection Act (micropollutants removal by 80% relative to the raw wastewater; Eggen et al. 2014, Bourgin et al. 2018), which requires an upgrade of selected UWTPs until 2040. A website of the Swiss Water Association provides updated information on European UWTPs that are planning or running full-scale advanced treatment for CEC removal (www.micropoll.ch).

The occurrence of organic matter (measured as DOC) and other readily oxidizable compounds (such as nitrite) in the effluent of biological treatment affect ozone exposure and should be considered when defining the ozone dose for the abatement of CECs. An ozone dose in the range of 0.4 – 0.6 g O_3/g DOC (in the absence of nitrite) was found to be suitable to efficiently abate micropollutants (Hollender et al. 2009, McArdell et al. 2015, Bourgin et al. 2018). Cost evaluations are shown later (section 2.2.3) in comparison to treatment with activated carbon. In the US and in Australia, ozonation followed by a BAC filter has been successfully applied as low-cost potable reuse option (Gerrity et al. 2014; Reungoat et al. 2012; Stanford et al. 2017);

2.2 Activated Carbon adsorption

2.2.1 Removal of CECs

Unlike oxidation, adsorption is a separation process which does not result in the formation of by-products. Activated carbon is the most used adsorbent in water treatment for the removal of organic and inorganic pollutants dissolved in water. Activated carbon treatment for the removal of CECs from wastewater has been widely investigated (Boehler et al., 2012; Grassi et al., 2013; Rizzo et al., 2015; Ahmed, 2017; Kovalova et al., 2013, Michael et al., 2019). Packed bed adsorption reactors with granular activated carbon (GAC) as adsorbent material are commonly used in drinking water treatment. Due to process costs, their application at full-scale as advanced urban wastewater treatment only recently has attracted the interest of UWTPs managers and professionals, as the concern for possible effect on human health and environment of CECs has increased (Rizzo et al., 2019a; Siegrist et al. 2019). Its advantage compared to powdered activated carbon (PAC) is that operationally it is easier to use, and it can be recovered and regenerated when its adsorption capacity is exhausted. However, the process requires an adequate monitoring strategy, since adsorption competition results in a reduced CEC removal or even desorption of less adsorbable CECs with increasing treated bed volumes due to a decrease in available adsorption sites. PAC can be applied as a post-treatment or dosed into the biological unit in UWTPs and, due to its smaller particle size (higher specific surface area), is more efficient compared to GAC in the removal of water pollutants and specifically CECs (Nowotny et al., 2007, Boehler et al., 2012).

2.2.2 Effect on ARB&ARGs

Even though adsorption is not a disinfection process and not designed to remove bacteria and mobile genetic elements, a contribution to the reduction of antibiotic resistance in wastewater

effluent can be expected due to possible entrapment of ARB&ARGs inside the pores of adsorbent particles (Zhang et al., 2017; Ashbolt et al., 2018; Bürgmann et al. 2018).

2.2.3 Application at full-scale as advanced treatment of urban wastewater

Activated carbon adsorption has been recently applied at full-scale for advanced treatment of urban wastewater as alternative to ozonation, particularly in Switzerland and Germany, for the removal of CECs before effluent discharge into the environment (Rizzo et al., 2019a). Depending on DOC and operation technology, a dose of 10-20 mg/L PAC can be recommended to protect the aquatic environment (Boehler et al. 2012). A post-treatment is also needed in PAC treatment for separation of residual PAC material. The use of GAC-packed reactors is more restricted since it does not allow to react to certain conditions (e.g. rainy periods), whereas PAC dose can be increased (Siegrist et al., 2019). However, GAC in combination with other treatment is used successfully for many years, but just for direct potable reuse application (Vaidya et al. 2019; Piras et al., 2020). As far as operation costs are concerned, feasibility studies conducted in the state of North Rhine-Westphalia (Germany) in the years 2009–2016 resulted in similar median costs (0.04 €/m^3) for ozonation (16 plants), PAC (11) and GAC (9) processes (Figure SI4 in Rizzo et al., 2019a), with highest variability for GAC treatment. Overall costs, including investment and operation, vary substantially with the size of the UWTP. For mid-scale plants (~50.000 PE), the costs are in the range of 0.10 to 0.15 €/m^3 treated wastewater, decreasing further with increasing plant size even below 0.05 €/m^3, with PAC treatment being slightly more expensive than ozonation (Figure 4, Rizzo et al. 2019a). Consistently with the numbers determined in Germany, overall costs for PAC (0.10-0.15 CHF/m^3, 1 CHF being 0.88 € on January 18th, 2019, for dosing 10 mg/L PAC in a large plant with 590,000 p.e.) were estimated to be higher than for ozonation (0.04-0.06 CHF/m^3, for

306 dosing 5 mg/L ozone in a large plant) in Switzerland (McArdell et al., 2015, Abegglen et al.
307 2012).

308

309 ## 2.3 Chemical oxidants/disinfectants

310 Chlorination is by far the most common method of wastewater disinfection, but the concern for
311 human health and the environment related to the formation of toxic by-products (e.g.,
312 trihalomethanes, haloacetic acids and related contaminants) is increasing the interest towards
313 alternative chemical disinfectants, such as peracids. Among them, peracetic acid (PAA) already
314 finds different applications at full-scale in UWTPs, particularly in Italy (Formisano et al., 2016;
315 Di Cesare et al., 2016a) and in the USA (Bell and Wylie, 2016; Stewart et al., 2018).
316 Accordingly, chlorination and PAA disinfection are discussed in the subsequent sub-
317 paragraphs. Neither of the two technologies is applied for CEC abatement as they are not
318 economic and produce problematic effluents.

319

320 ### 2.3.1 Chlorination

321 Wastewater disinfection by chlorine is typically performed by chlorine gas (in medium – large
322 UWTPs) or hypochlorite (either calcium or sodium). Limited studies have focused on the
323 abatement of CECs by chlorine, which was found to be quite poor, in particular if compared to
324 oxidation/disinfection processes with higher oxidation potential such as ozone and other AOPs
325 (Anumol et al., 2016; Hua et al., 2019). For example, Li and Zhang (2011) reported abatement
326 of antibiotics during wastewater treatment with chlorine in the range of 18% (roxithromycin)
327 to 40% (trimethoprim), while cephalexin and ampicillin were abated by 99% and 91%,
328 respectively. However, the chlorine dose was not reported in this study, and cephalexin and
329 ampicillin are beta-lactam antibiotics that hydrolyze very quickly, so these results do not allow

to discriminate hydrolysis contribution from chlorine oxidation effects. Contrasting results are documented in the scientific literature for sulfamethoxazole (SMX). Whilst Gao et al. (2014) observed an almost complete abatement of SMX (initial concentration in the range 0.05–2 mg/L) within 15 min contact time and 2.0 mg/L of chlorine, de Jesus Gaffney et al. (2016) observed only 20% abatement (pH 6–7, 2 mg/L of free chlorine) of SMX after 2 h contact time. However, when reaction kinetics of SMX were investigated in different water matrices, the results achieved in real wastewater ($[SMX]_0 = 2.0 \times 10^{-6}$ M), pH 7.3, free residual chlorine (FRC) 11 mg/L) confirmed the substantial degradation of SMX observed in deionized water (half-life of 23 s was measured under pseudo-first-order conditions ($[FRC]_0$ = 20 μM (1.4 mg/L)) (Dodd and Huang, 2004). This expectation is supported by existing observations at full-scale UWTPs, where 89.6% SMX abatement was observed (Renew and Huang, 2004). Despite the fact that single compounds are degraded by chlorination, a broad abatement of CECs cannot be achieved; for example, poor or no abatement of diclofenac or carbamazepine was observed (Hua et al. 2019).

Chlorination can result in the formation of toxic by-products, including trihalomethanes and haloacetic acids (Richardson et al., 2007). Moreover, in effluents with incomplete nitrification, chlorine combines with ammonia to form chloramines or so-called combined chlorine. Chloramine chemistry is complex and will not be discussed further here, but it is noteworthy that chloramines are weaker oxidants and disinfectants compared to free chlorine. NDMA is a typical disinfection byproduct when chloramines are generated in wastewater effluents (Sgroi et al., 2018). It can be concluded that chlorination is not an option for CECs abatement and could produce an adverse effect on effluent organic composition when used for disinfection.

The effect of chlorination on ARB is being investigated since the 70's (Grabow et al., 1976). Although the chlorination process was found to effectively decrease antibiotic resistant *E. coli* in wastewater, it may select bacterial population by increasing antibiotic resistant *E. coli* strains

compared to the corresponding total population (Fiorentino et al., 2015). However, when the effect of chlorination on ARGs was investigated, different results were observed. For example, ARGs *ere*A and *erm*B persisted in chlorinated (15 mg Cl_2 min/L) urban wastewater samples (Yuan et al. 2015) and chlorination was found to be effective in ARGs removal (3.16 Log for *sul*I and 3.24 Log for *tet*G after 120 min treatment) only at non-realistic chlorine concentration (160 mg/L) (Zhuang et al., 2015). On the opposite, Zheng and colleagues (2017) observed that chlorination can reduce ARGs (*tet*A, *tet*M, *tet*O, *tet*Q, *tet*W, *sul*I and *sul*II) abundance to some extent (less than 1 Log unit for *tet*A) even under realistic operating conditions (5 mg/L of chlorine, 30 min contact time). Moreover, Yoon et al. (2017) observed 4 Log reduction of ARGs concentration (two differing amplicons located in the commercially available plasmid pUC4K i.e., amp^R and kan^R) with 33-72 (mg·min)/L chlorine dose at pH 7 in urban wastewater. In particular, intracellular ARGs showed lower rates of damage compared to the extracellular ARGs, possibly due to the protective roles of cellular components. However, when process efficiency was investigated in full-scale UWTPs, chlorination did not prove to have significant contribution to ARGs (*tet*A, *tet*W, *tet*O, *erm*B, *qnr*S, bla_{TEM} *sul*I) removal (Munir et al., 2011; Gao et al., 2012; Di Cesare et al., 2016b).

2.3.2 Disinfection with peracetic acid

PAA is a strong and broad-spectrum disinfectant, with a high reduction-oxidation (redox) potential and strong biocidal effects on bacteria. Because of the formation of toxic by-products in chlorination, PAA is increasingly replacing chlorine in UWTPs as it shows a broad-spectrum efficiency and comparable way of application (Antonelli et al., 2013; Formisano et al., 2016; Di Cesare et al., 2016a).

In spite of no significant formation of disinfection by products (DBPs) resulting from wastewater disinfection by PAA when low doses are used (<5-10 mg/L) (Nurizzo et al., 2005), PAA was found to be toxic for bacteria and crustaceans, even at concentrations lower than the ones commonly used in wastewater disinfection (2-5 mg/L). But when PAA was compared to other disinfection processes, a lower toxicity against aquatic organisms was observed. In particular da Costa et al. (2014) compared PAA (5 mg/L, 20 min contact time), UV light (average UV dose at 254 nm 670.8 mJ/cm^2, 120 s contact time), ozone (29.9 mg/L, 5 min contact time), and sodium hypochlorite (2.5 mg/L, 20 min contact time) against *Ceriodaphnia silvestrii*, *Daphnia similis*, *Chironomus xanthus*, and *Danio rerio* and toxicities after treatment were in the order of free chlorine > ozone > UV > PAA after the respective disinfection treatments had been applied to secondary effluent.

Due to its lower oxidation potential compared to ozone and hydroxyl radicals, possible abatement of CECs in wastewater by PAA has not attracted the interest of the scientific community. As matter of fact, PAA effect on CECs has been investigated only as control test compared to UV/PAA process (Rizzo et al., 2019b). Unlike carbamazepine (no abatement observed even after 300 min contact time), diclofenac was effectively oxidized by 2 mg PAA/L already after 60 min (80% abatement), while SMX was abated at a lower percentage (52% after 300 min). As PAA effect on ARB is of concern, the limit of detection was achieved within 15 min treatment in groundwater inoculated with an antibiotic resistant *E. coli* strain by 1 mg/L and 2 mg/L of PAA (Rizzo et al., 2019b). However, the water matrix strongly affects bacterial inactivation efficiency. As a matter of fact, Huang et al. (2013) observed lower inactivation in reclaimed water with a higher PAA initial dose (20 mg/L). In particular, inactivation was higher for ampicillin-resistant bacteria (2.3 Log) than for total heterotrophic bacteria (2.0 Log) and tetracycline resistant bacteria (1.1 Log) after 10 min treatment. Moreover, the regrowth of chloramphenicol-and tetracycline-resistant bacteria, as well as total heterotrophic bacteria was

more than 10-fold compared to those in the untreated wastewater sample (22 h stilling culture after exposure to 2 or 5 mg PAA /L as for 10 min). Di Cesare et al. (2016a) evaluated the fate of diverse ARGs, heavy metal resistant genes and of a mobile element (the class I integron) in three UWTPs using different disinfection processes. In 2 (*sul*II and *tet*A) out of 4 (*erm*B and *qnr*S) of the quantified ARGs, a decrease was observed after PAA treatment.

2.4 UV radiation

UV radiation (250-270 nm) is widely used for urban wastewater disinfection either for effluent discharge or reuse (Munir et al., 2011; Di Cesare et al., 2016a). UV radiation can damage DNA, resulting in the inhibition of cell replication and, in case of lethal doses, in loss of the ability of reproduction. The effectiveness of a UV disinfection system depends on the characteristics of the wastewater, the UV fluence (intensity × irradiation time), the type of microorganisms and reactor configuration. Since turbidity and suspended solids drastically decrease UV disinfection efficiency, conventional depth filtration should be used before UV disinfection (not necessary when applied following a membrane biological reactor (MBR)).

2.4.1 Abatement of CECs

UV radiation is not at all or is poorly effective in the abatement of most of CECs from water and wastewater, but it can abate some antibiotics and other CECs at very high UV doses (Kim et al., 2009; Rizzo et al., 2019b). For example, an almost complete abatement of tetracyclines and ciprofloxacin was achieved but only at high UV doses (11,000-30,000 mJ/cm^2) (Yuan et al., 2011) and high abatement efficiencies (86-100%) were also observed for sulfonamides (SMX and sulfadimethoxine) and quinolones (norfloxacin and nalidixic acid) (Kim et al.,

2009). Iodinated X-ray contrast media were abated by more than 90% at 720 mJ/cm^2 (Kovalova et al. 2013).

2.4.2 Effect on ARB&ARGs

The effect of UV radiation on ARB&ARGs in urban wastewater has been increasingly investigated in the last years at lab and full-scale (Munir et al., 2011; McKinney and Pruden, 2012; Rizzo et al., 2013; Guo et al., 2013; Zhuang et al., 2015; Di Cesare et al., 2016a). Process efficiency strongly depends on the applied UV dose and target ARB&ARGs, and possibly this is the main reason to explain differences between lab- and full-scale evidences.

Efficient removal of heterotrophic bacteria harboring resistance to erythromycin and tetracycline was observed (Guo et al., 2013) (equivalent Log reduction being 1.4 and 1.1 at a UV dose of 5 mJ/cm^2). As UV dose was further increased to 20 and 50 mJ/cm^2, respectively, ARB were below the detection limit (1 CFU/mL).

The UV dose also affects the removal of ARGs. UV doses ranging from 200 to 400 mJ/cm^2 (at least one order of magnitude higher than those for the inactivation of host bacterial cells) were required to remove 3 or 4 Log units of ARGs, namely *amp*C, *mec*A, *tet*A and *van*A (McKinney and Pruden, 2012). Actually, also lower UV doses (5-10 mJ/cm^2) were found to be effective in the removal of ARGs (namely *ere*A, *ere*B, *erm*A, *erm*B, *tet*A, *tet*O) but starting from lower initial ARGs copies per mL (Guo et al., 2013). The relative abundance of selected ARGs increased with low doses of UV (Zhuang et al., 2015). Less than one order of magnitude removal of five tetracycline resistance genes (*tet*A, *tet*M, *tet*O, *tet*Q, *tet*W) and two sulfonamide resistance genes (*sul*I, *sul*II) were observed in UV disinfection (UV fluence 10-160 mJ/cm^2) of wastewater samples taken from the secondary sedimentation tank of a UWTP in Hangzhou, China (Zheng et al., 2017). The removal efficiency of the five *tet* genes was between 52.0%

450 and 73.5% at the lower fluence UV disinfection (40 mJ/cm^2 or less), and between 79.7%, and
451 92.0% at high fluence (160 mJ/cm^2). Lower removal efficiencies were observed for *sul*I, *sul*II
452 (78.1% and 71.1% respectively, at the higher fluence).

453 In full-scale monitoring (5 UWTPs in the USA), UV radiation employed for disinfection did
454 not prove to have a significant contribution to ARGs (*tet*w, *tet*O, *sul*I) and ARB reduction
455 (Munir et al., 2011). These results were confirmed in a subsequent study at full-scale, where no
456 significant difference in ARGs (namely, *erm*B, *qnr*S and *tet*A) was observed before and after
457 UV disinfection, while for *sul*II even an increase was observed after disinfection (Di Cesare et
458 al., 2016a).

459

460 2.5 Advanced oxidation processes

461 Advanced oxidation processes (AOPs) rely on the formation of hydroxyl radicals that can abate
462 a wide range of CECs (Rizzo, 2011; He et al., 2020) as well as inactivate microorganisms
463 (Dunlop et al., 2010; Fiorentino et al., 2015). A possible classification of AOPs includes two
464 groups: homogeneous processes (e.g., UV/H_2O_2, UV/Fe/H_2O_2, O_3, O_3/H_2O_2 etc.) and
465 heterogeneous (solid semiconductors + light source, e.g., UV/TiO_2, UV/ZnO) photocatalytic
466 processes. Homogeneous processes have been widely investigated as advanced treatment of
467 urban wastewater effluents and either are already applied at full-scale (e.g., O_3, see section 2.1)
468 or are characterized by short-/mid-term perspective application (e.g., UV/H_2O_2, UV/Fe/H_2O_2)
469 as opposed to heterogeneous photocatalytic processes (Rizzo et al., 2019a; Maniakova et al.,
470 2020). The main reason why heterogeneous photocatalytic processes are not ready for full-scale
471 application as advanced urban wastewater treatment are related to photocatalyst preparation
472 costs, photocatalyst quantum yield (effectiveness) and reactor configuration (Iervolino et al.,
473 2020). In particular, heterogeneous photocatalytic processes can be operated under two main

474 configurations: (i) with the photocatalyst suspended in the reactor (i.e., slurry system) or (ii)
475 attached to a support (i.e., immobilized system). Due to the higher specific surface area
476 available, a slurry system is more effective than an immobilized one, but a subsequent
477 expensive separation process (e.g., coagulation, filtration, membrane) is necessary to recover
478 the photocatalyst before effluent discharge or reuse (Fernández-Ibáñez et al., 2003).
479 Immobilized photocatalytic systems have relatively lower quantum efficiency than slurry ones,
480 which results in longer treatment time and consequently larger water volume to treat (Spasiano
481 et al., 2015). Some homogeneous photo-driven AOPs can also be operated under natural
482 sunlight (solar/H_2O_2 or solar/Fe/H_2O_2) thus saving energy costs (Klamerth et al., 2010; Ortega-
483 Gomez et al., 2014; Ferro et al., 2015; Giannakis et al., 2016) and this can be considered as an
484 attractive option for small UWTPs in areas with sufficient sunlight.

485

486 2.5.1 Abatement of CECs

487 Due to their high redox potential hydroxyl radicals oxidize a wide spectrum of organic
488 contaminants, accordingly, AOPs successfully degrade several organic micropollutants
489 (Klavarioti et al, 2009; Rizzo, 2011). The most common AOPs studied are UV/H_2O_2, O_3/H_2O_2,
490 O_3/UV, Fenton (Fe/H_2O_2), photo-Fenton (UV/Fe/H_2O_2) and heterogeneous photocatalysis (e.g.,
491 UV/TiO_2, UV/ZnO). Although UV/H_2O_2, is more efficient than UV alone to abate CECs, still
492 more energy is needed compared to ozonation (Rizzo et al., 2019a). O_3/H_2O_2 does not improve
493 abatement of CECs compared to ozone alone in UWTP effluents, since effluent ozonation can
494 be considered an intrinsically AOP due to the high $HO^{\bullet}$ generation potential of the organic
495 matrix (Buffle et al., 2006), at the same time $HO^{\bullet}$ are scavenged by the matrix (Acero and von
496 Gunten, 2001; Kovalova 2013). Fenton and photo-Fenton processes are typically effective
497 under acidic conditions (pH 3) and the abatement of three antibiotics, namely SMX,
498 erythromycin (ERY) and clarithromycin, from urban wastewater was investigated (Karaolia et

al., 2017). SMX and ERY were efficiently abated from UWTP secondary effluents by solar photo-Fenton in continuous flow operation with >80% abatement at a hydraulic residence time of 20 min in non-concentrating raceway pond reactors (Arzate et al., 2017). Nonetheless, this operation mode at full-scale would result in additional process cost and salinity increase because pH has to be first decreased and subsequently neutralized before effluent discharge or reuse. However, photo-Fenton has also been successfully investigated under almost neutral pH conditions and solar radiation for the abatement of CECs from urban wastewater with the addition of complexing agents. As a matter of fact, the (solar driven) photo-Fenton process allowed to effectively decrease CECs from urban wastewater under so-called mild conditions, i.e. under low Fe (< 5 mg/L) and H_2O_2 (< 20 mg/L) concentrations and pH 5-6, thus avoiding the necessity for final separation of soluble iron species from the treated wastewater (Klamerth et al., 2010; De la Obra et al., 2017). The use of organic chelating agents makes the process feasible and effective even under neutral pH conditions (De Luca et al., 2014; Fiorentino et al. 2018; Soriano-Molina et al., 2018). Unlike photo-Fenton, solar-UV/H_2O_2 process can be operated at neutral pH without chelating agents, and it can successfully abate some CECs, but longer reaction time compared to photo-Fenton is needed (Ferro et al., 2015).

2.5.2 Effect on ARB&ARGs

AOPs can successfully inactivate ARB in urban wastewater (Karaolia et al., 2014; Rizzo et al., 2014a; Fiorentino et al., 2019). As a matter of fact, sunlight/H_2O_2 process resulted in a total inactivation of multi drug resistant (MDR) *E. coli* (resistant to a mixture of three antibiotics: ampicillin, ciprofloxacin and tetracycline), after 90 min of treatment (Fiorentino et al., 2015). Noteworthy, longer treatment time (120 min) was necessary to achieve a complete inactivation of the total *E. coli* population, despite the percentage of MDR *E. coli* ((total *E. coli* – MDR *E. coli*)x100/total *E. coli*)) increased as total *E.coli* population decreased with treatment time.

However, the release of mobile genetic elements from bacterial cells, that may take place after disinfection process, and the potential to transfer antibiotic resistance through horizontal mechanism, have been poorly investigated. Photo-driven AOPs have recently been investigated to evaluate if they can be more effective in the removal of ARGs than conventional disinfection processes, such as chlorination and UV radiation. Ferro et al. (2016) investigated the effect of UV/H_2O_2 (broad-band spectrum UV lamp with main emission in the range 320-450 nm), under realistic conditions for wastewater treatment (natural pH (7.6) and 20 mg H_2O_2/L), on antibiotic resistance transfer potential in urban wastewater. The investigated process resulted in bacterial inactivation and a decrease of ARGs in intracellular DNA after 60 min treatment, but UV/H_2O_2 did not remove ARGs effectively. Actually, an increase up to 3.7×10^3 copies/mL ($p > 0.05$) of bla_{TEM} gene was observed in total DNA after 240 min treatment, while no difference ($p > 0.05$) was found for *qnr*S gene between the initial (5.1×10^4 copies/mL) and the final sample (4.3×10^4 copies/mL). In UV/H_2O_2 process (pH 7, 50-130 mJ/cm^2), 4 Log reduction of ARGs (*amp*^{R} and *kan*^{R}) concentration was observed in urban wastewater (Yoon et al., 2017). According to the results previously discussed for the chlorination process, intracellular ARGs showed lower rates of damage compared to extracellular ARGs due to cell protective roles and significant $HO^{\bullet}$ radical scavenging by cellular components. Zhang et al. (2016a) showed that UV/H_2O_2 can effectively remove ARGs (2.8-3.5 logs removal of *sul*1, *tet*X, and *tet*G, within 30 min treatment) but only under conditions that seem unrealistic for full-scale implementation (pH 3.5 and 340 mg H_2O_2/L), moreover UV fluence was not provided.

Solar driven photo-Fenton process is effective in the inactivation of ARB Karaolia et al., 2017; Fiorentino et al., 2019). When the process (5 mg Fe^{2+}/L, 50 mg H_2O_2/L, pH 3) was operated at pilot scale through a compound parabolic collector (CPC) based reactor, on the effluent of an MBR, a complete inactivation of the low initial bacterial population (*E. coli* = 2 CFU/100 mL, *P. aeruginosa* = 4 CFU/100 mL, *Klebsiella* spp. = 3 CFU/100 mL), including antibiotic-tolerant

549 and susceptible bacteria, was observed, after 54 min of solar radiation intensity normalized time
550 (Karaolia et al., 2017). On the other hand, repair of *P. aeruginosa* was observed, with 2
551 CFU/100 mL growing on the selective media 24 h after solar Fenton oxidation. Solar photo-
552 Fenton process was also investigated in raceway pond reactors, at neutral pH conditions (20 mg
553 Fe^{2+}/L, 50 mg H_2O_2/L), in real urban wastewater and an effective inactivation of *E. coli* and
554 *Enterococcus sp.* cefotaxime resistant bacteria was observed (detection limit (1 CFU/mL)
555 achieved after 30-40 min, 3.2-4.7 kJ/L) (Fiorentino et al., 2019). However, both solar driven
556 photo Fenton processes (CPC reactor at pH 3 and raceway ponds at neutral pH) did not
557 effectively remove the target ARGs..

558 The effect of heterogeneous photocatalysis with TiO_2 on ARB&ARGs has been investigated in
559 slurry and immobilized systems. According to the results observed for homogenous photo-
560 driven AOPs, even heterogeneous photocatalytic processes, while effective in the inactivation
561 of different antibiotic resistant bacterial populations (Tsai et al., 2010; Rizzo et al., 2014a, Rizzo
562 et al., 2014b; Dunlop et al., 2015; Zammit et al., 2019) may not be effective in the removal of
563 some ARGs (Karaolia et al., 2018).

564

565 2.6 Membrane filtration

566 Membrane separation processes include microfiltration (MF), ultrafiltration (UF),
567 nanofiltration (NF) and reverse osmosis (RO), which may be operated separately or in
568 combination with other processes as a part of integrated technologies such as MBR. NF and
569 RO are effective in the removal of both organic and inorganic CECs (Bellona et al., 2004;
570 Alturki et al., 2010; Garcia et al., 2013), while MF or UF are typically used as pre-treatment of
571 either NF or RO to control membrane fouling as well as for disinfection and solids removal.
572 NF and specifically RO provide the opportunity to reduce the effluent salinity, which can be

573 necessary depending on the downstream application of the treated effluent. However, a waste
574 stream containing the separated salts and other pollutants is generated as well.

575

576 2.6.1 Removal of CECs

577 Removal of CECs by membrane processes is primarily based on size exclusion, although
578 electrostatic interactions between charged solutes and negatively charged membranes typically
579 have an important role in the removal (Bellona et al., 2004). Hydrophobic trace contaminants
580 have been shown to absorb to membrane surfaces reducing the rejection of these contaminants
581 through both RO and NF. This has been shown to be particularly relevant in NF processes.
582 Several other factors typically also affect the removal of the target CECs (such as phenolic
583 aromatic compounds) by membrane processes (Bellona et al., 2004). Depending on the type of
584 membrane, the range of rejections of CECs by both RO and NF is quite broad, but the rejection
585 can be higher than 99% for high rejection RO membranes (Krzeminski et al., 2017). However,
586 in these membrane processes the CECs are accumulating in the rejected concentrate. The
587 discharge of the concentrate to the environment can be problematic, as the original salt and
588 pollutant load of the secondary effluent, while not having increased in absolute mass, is now
589 concentrated typically by a factor of 3 to 7, depending on the permeate water recovery
590 percentage of the membrane process. The presence of the contamination in concentrated form
591 can also be an opportunity for targeted treatment since pollutants are more effectively treated
592 by advanced oxidation processes (usually governed by first order kinetics) as initial
593 concentration increases (Miralles-Cuevas et al., 2016).

594 Full-scale applications of RO technology are reported in potable reuse treatment trains, e.g. the
595 Orange County Groundwater Replenishment System (California, USA), NEWater facilities at
596 the Bedok, Kranji, Ulu Pandan and Changi facilities in Singapore and the Torreele Reuse

597 Facility in Belgium (Raffin et al., 2013; Gerrity et al. 2013). RO is also used in direct potable
598 reuse treatment trains, along with MF or UF, in Cloudcroft (New Mexico) and Big Spring
599 (Texas) in USA (Gerrity et al. 2013). NF typically removes CECs in the 300-1,000 molecular
600 weight (MW) range, rejecting selected salts and most organic constituents and microorganisms,
601 operating at higher recovery rates and lower pressures than RO processes. Accordingly, and
602 when feasible, NF can be used instead of RO to save some energy, chemical and concentrate
603 disposal costs (Yangali-Quintanilla et al., 2010). While offering very high removal efficiencies
604 for CECs, specifically RO, on the downside these technologies exhibit high energy
605 consumption.

606

607 2.6.2 Effect on ARB&ARGs

608 As the separation principle is purely based on size, the removal of ARB can be expected to
609 behave very similar to the removal of those not carrying antibiotic resistance. MF and UF are
610 commonly applied barriers for pathogens, with MF being very effective against protozoa and
611 bacteria, while due to a larger pore size, it is not very effective in removing viruses. UF removes
612 all three classes of pathogens to a very high extent (2 to 4 Log removal values (LRV)) (Hai et
613 al. 2014). NF and RO membranes present in theory an even smaller pore size and should be
614 “perfect filters”. In fact, > 6 LRV virus removal has been observed at pilot-scale. However, due
615 to the modular engineering approach system breaches cannot be per se excluded and finding
616 appropriate surrogate measurements remains a challenge to ensure disinfection during
617 operation, at least at levels beyond e.g. the removal of electrical conductivity (Pype et al, 2016).

618 The effect of membrane filtration, in particular NF and RO, on ARB&ARGs, thus far, has been
619 little discussed in the literature as the existing studies have focused mostly on MBRs and MF

and UF membranes (Munir et al. 2011; Riquelme Breazeal et al., 2013; Rizzo et al., 2013; Yang et al., 2013; Sun et al., 2016; Threedeach et al., 2016; Li et al., 2019).

As previously mentioned, membranes can remove bacteria due to membrane retention, thus contributing to reducing the spread of multiple antibiotic resistant strains (Verlicchi et al. 2015). For example, filtration of ARGs spiked UWTP effluent through the 100, 10 and 1kDa membranes in the lab-scale stirred ultrafiltration cell reduced *vanA* and bla_{TEM} ARGs by 0.9, 3.5 and 4.2 Log, respectively (Riquelme Breazeal et al., 2013). The removal of plasmid-associated ARGs improved further at the presence of colloidal material in the water matrix and the colloids influence became more apparent as the membrane pore size decreased. The DNA removal was attributed to membrane retention and following mechanisms: i) size exclusion of the DNA, ii) size exclusion of DNA-colloid complexes, or iii) interactions with the membrane material (Riquelme Breazeal et al., 2013).

Arkhangelsky et al. (2008, 2011) studied, in lab-scale dead-end membrane cell, penetration of plasmid DNA through UF membranes and demonstrated that despite electrostatic repulsion and a significant size difference between plasmid and pore sizes, DNA can penetrate through the UF membrane, indicating that UF did not provide absolute barrier for DNA retention. Also, Riquelme Breazeal et al. (2013) observed that 1 kDa membrane did not completely retain plasmid and pointed out that the effective size of DNA is smaller than predicted by molecular weight because DNA is a long, thin and flexible molecule. Although the penetration mechanism is not yet clear, Arkhangelsky et al. (2011) suggested that plasmid stretches into long hair-shaped flexible strands and penetrates pores based on 'snake-like' movement due to hydrodynamic pressure (transmembrane pressure, TMP) with gradual pore blocking. The proposed penetration mechanism is in accordance with the findings of other studies on DNA (Marko et al., 2011; Travers, 2004). In addition, plasmid transportation levels are linearly correlated to the TMP.

Böckelmann et al. (2009) studied three artificial recharge systems in Europe. Combination of UF and RO proved to be an efficient barrier for the elimination of ARGs. ARGs *tetO* and *ermB* detected in UWTP effluent at concentrations of $1.05x10^7 \pm 3.54x10^6$ gene copies/100mL and $1.92x10^5 \pm 1.06x10^4$ gene copies/100mL, respectively, were removed during the UF-RO process applied in the Torreele Reuse Facility. Noteworthy, *tetO* were detected again, at low concentrations, in subsequent sampling points: in the infiltration water before transport ($5.92x10^3 \pm 1.39 x10^3$ gene copies/100mL) and in the groundwater after infiltration ($3.13x10^3 \pm 1.52x10^3$ gene copies/100mL). In a recent work, a wastewater reuse treatment train including MBR with MF membranes followed by RO provided up to 3.8 Log removal of the ARGs down to absolute abundance of $4.03x10^4$ copies/mL (Lu et al., 2020). MF was capable of 2-3 Log removal of ARGs whereas subsequent RO provided additionally up to 1.5 Log removal. Another recent full-scale study investigating the removal of ARGs in a full-scale wastewater treatment plant including biological and physicochemical treatment located on a swine farm showed very high removals for ARGs in both, NF and RO. The removals achieved depended on the ARG and ranged from 5 to 8 Log removals compared to raw sewage (Lan et al., 2019).

Above 99.2% removal of free DNA from UWTP effluent by NF membrane in the lab-scale system was reported (Slipko et al., 2019). Similar removal rates were observed both in water and in effluent. According to the authors, besides size exclusion mechanism, electrostatic repulsion plays also important role in removal of free DNA in NF and RO.

2.7 Comparison among BATs for the removal of CECs relevant for crop uptake

During the last years, several classes of CECs have been proven to taken up through roots and translocated to the aerial parts of crop plants irrigated with treated wastewater, grown under hydroponic or greenhouse control conditions, as well as soils irrigated with treated wastewater

669 in real agricultural systems. The uptake is largely dependent on CECs' bioavailability in soil
670 pore water near the rhizosphere and thus on their physicochemical properties and the properties
671 of the soil environment. Once taken up, the transport of CECs within the plant vascular
672 translocation system (xylem and phloem) mainly depends on their lipophilicity and electrical
673 charge, as well as the physiology and transpiration rate of crop plants and environmental
674 conditions (i.e. drought stress), (Nereus COST Action ES1403, Deliverable 11). Accordingly,
675 different crops have different potential for CECs uptake, for example, uptake potential is
676 generally higher for leafy vegetables compared to fruit vegetables or cereal crops. The main
677 biotic factors that may affect the uptake of CECs by plants are the plant itself (including the
678 species, the variety and cultivar, the genotype, and the physiological state of the plant), and the
679 soil fauna, which constitute the main cause for the biodegradation and biotransformation of
680 CECs within the soil (Ahuja et al., 2010; Goldstein et al., 2014). Climatic conditions and other
681 environmental perturbations (such as temperature, wind speed, UV radiation, salinity, drought,
682 environmental pollution, etc.) constitute the main abiotic factors that influence the potential for
683 CECs uptake by crop plants (Dodgen et al., 2015; Zhang et al., 2016b). The majority of studies
684 with regard to CECs uptake, either conducted in controlled laboratory or greenhouse conditions
685 or under field or simulated conditions, employed mostly (a) vegetables (leafy vegetables such
686 as lettuce and cabbage, fruit vegetables such as tomato and cucumber, and root vegetables such
687 as carrot and radish) and (b) cereals and fodder crops (i.e. maize, wheat, alfalfa). Experimental
688 results revealed that the potential for CECs uptake by crop plants decreased in the order of leafy
689 vegetables > root vegetables > cereals and fodder crops > fruit vegetables. Though, the uptake
690 of CECs by important crop plants, such as fruit trees, has not yet been evaluated. Fruit trees,
691 such as citrus, bananas, apple and other fruit bearing trees, have high net irrigation requirements
692 and evapotranspiration rates, which may render them as plants with moderate to high potential
693 for CECs uptake (similar to that of fruit vegetables), (Christou et al. 2019). Therefore, the

694 recommendation on the BAT should consider both the soil and the type of the crop species to
695 be irrigated by reclaimed water.

696 Consistently with the aim of the present review paper, a comparison among the above-
697 mentioned BATs was performed according to the chemical CECs relevant for crop uptake by
698 considering results from investigations at pilot or full-scale on real wastewater. According to
699 the list compiled by NEREUS COST Action ES1403, 27 CECs are relevant for crop uptake
700 (Krzeminski et al., 2019). The Action also applied selected criteria to establish a prioritised list
701 with CECs which include the following: 1) high frequency of detection in treated effluents,
702 which is related to high patterns of use and recalcitrance during the wastewater treatment
703 process, 2) environmental, agricultural and/or health concern; at least one of the following
704 criteria should be met by the target CECs: a) DT_{50} (time necessary to degrade the 50% of the
705 original contaminant concentration) in soil > 14 d, b) phytotoxicity at environmental relevant
706 concentrations, c) promote a selection pressure to soil microbiota, d) potential human health
707 effects according to threshold contaminant concentration criteria, 3) significant uptake rate by
708 crops (usually bioconcentration factors (RCF = [root]/[growing medium]; LCF =
709 [leaf]/[growing medium]; FCF = [fruit]/[growing medium]) higher than 1). The list of
710 prioritised CECs includes carbamazepine (CBZ), diclofenac (DCF), enrofloxacin, SMX, 17^{α}-
711 ethinyl estradiol, lamotrigine and trimethoprim, (Nereus COST Action ES1403, Deliverable 7;
712 Boxall et al., 2012; Calderón-Preciado et al., 2012; Christou et al., 2017b; Goldstein et al., 2014; Miller
713 et al., 2016; Tanoue et al., 2012; Wu et al., 2015; Zhang et al., 2016b). However, out of 27 crop
714 relevant CECs only for 3 compounds, namely CBZ, DCF and SMX, literature was found on
715 their removal from wastewater matrices during different advanced technologies (Table 1). For
716 SMX, high removal efficiencies (>80-100%) were observed during RO and NF, UV radiation,
717 chlorination (HOCl), ozonation and other AOPs, while lower efficiencies (<64%) were
718 observed for PAA and PAC treatment. High DCF removal efficiencies (80-100%) were
719 observed during RO and NF, UV radiation, PAA treatment, ozonation and other AOPs, good

720 removals (≅ 70%) for PAC, lower (60%) for chlorination. Finally, high CBZ removal
721 efficiencies (90-100%) were observed for PAC, ozonation, and RO, a wide range of efficiencies
722 (>24-100%) for AOPs and NF, depending on the process and operating conditions, UV
723 radiation resulted in a poor efficiency (16%), and no removal was observed for chlorination and
724 PAA treatment under the investigated conditions.

725

726 **Table 1**

727

728 **3. Multi-barrier approach for a safe treated wastewater reuse in agriculture**

729 3.1 Treatment trains for a safe reuse

730 To make wastewater reuse safe for crop irrigation, a multi-barrier approach to wastewater
731 treatment is necessary. These barriers should include typical processes for urban wastewater
732 treatment (namely, primary mechanical pre-treatment, possible primary settling, biological
733 treatment etc.) and advanced treatments. Possible options of treatment trains (TTs) providing
734 different effluent qualities are presented in Figure 1.

735 As matter of fact, no specific regulation on CECs (except in Switzerland) and ARB&ARGs is
736 in force that can justify a prioritization for these contaminants with respect to more traditional
737 parameters (in particular bacteria indicators such as total coliforms and *E. coli*) regulated in
738 different countries and guidelines for wastewater reuse. In particular, as ARB are of concern,
739 total *E. coli* population was suggested to be a good indicator for the inactivation of the antibiotic
740 resistant fraction (Fiorentino et al., 2015).

741 The minimum treatment scheme for safe reuse should include a conventional depth filtration
742 downstream of a biological process (or an UF membrane as in case of MBR, Fig.1, b), followed

743 by a disinfection unit with UV radiation (Fig.1, a). This TT should effectively allow to address
744 typical parameters (e.g., biochemical oxygen demand (BOD), chemical oxygen demand (COD),
745 total suspended solids (TSS), *E. coli* etc.) set in wastewater reuse regulation and guidelines.

746 Chemical disinfection (in particular by chlorine) (Fig.1, c) is cheaper compared to other
747 disinfection options but the formation of DBPs should be considered, and the TT may become
748 expensive compared to other options if DBPs are removed before reuse.

749 It has to be noted that, chemical disinfectants (such as chlorine and PAA) as well as an MBR
750 with UF membrane and UV radiation are poorly effective in the removal of CECs.

751

752 **Figure 1**

753

754 Therefore, if (i) the corresponding limit for bacterial indicators is so stringent that UV
755 disinfection is not sufficient and/or (ii) CECs contamination should be effectively minimized,
756 other, more effective treatment technologies need to be considered (Fig.1, d-g).

757 Among AOPs, ozonation and photochemical processes showed interesting results in the
758 removal of CECs and ARB. In particular, in the short term, ozonation and UV/H_2O_2 processes
759 are more attractive options (Fig.1, d) compared to other photo-driven AOPs to abate CECs as
760 well as to effectively inactivate bacteria (Rizzo et al., 2019a) because:

761 1. their efficiency has been confirmed by different works available in scientific literature.
762 However, ozonation needs considerably less energy compared to UV/H_2O_2 treatment
763 for the same CEC abatement level and shows full-scale application;
764 2. other homogeneous photocatalytic processes (such as photo-Fenton) may request
765 additional costs (e.g., pH adjustment, chelating agents' addition) and/or have not yet

766 been exhaustively investigated (e.g., UV/free chlorine, UV/PAA, sulfate radical based
767 AOPs);
768 3. heterogeneous photocatalytic processes still have serious technological barriers for full-
769 scale application.

770 It is important to note that ozonation and AOPs typically ask for a biological post-treatment,
771 *i.e.* a biological sand or activated carbon filtration, to remove biodegradable oxidation by-
772 products and transformation products (Fig.1, d). Rapid depth filtration or alternatively a
773 dissolved air flotation treatment may be used as pre-treatment method just before AOP in the
774 event that residual suspended solids should interfere with subsequent processes.

775 Adsorption to GAC in packed reactors followed by UV disinfection (unlike O_3 and UV/H_2O_2,
776 adsorption is not a disinfection process) is another option to improve the quality of effluent
777 wastewater before reuse (Fig.1, e). In order to prevent GAC packed reactors from a fast
778 clogging and increase back flushing intervals, cloth or rapid sand filtration may be used to
779 remove suspended solids before the adsorption process.

780 If PAC adsorption is used in combination with the biological process (by adding PAC into the
781 biological treatment) or as a separated unit thereafter, either depth filtration and/or MF/UF
782 membrane processes should be used to remove residual PAC particles before discharge
783 (Fig.1, f). As in GAC treatment, a UV disinfection may have to be installed.

784 Finally, membrane filtration with NF or RO followed by UV disinfection is another possible
785 option for advanced treatment of wastewater before reuse (Fig.1, g). Pre-treatment by sand
786 filtration can be used to remove suspended solids to control membrane fouling, although it is
787 more common to filter settled effluent directly with MF or UF membranes. MF and UF
788 membranes also provide suitable pre-treatment for the NF or RO step (in such a case final
789 disinfection by UV radiation is not necessary for crop irrigation). It is worthy to mention that

790 RO treatment would be additionally beneficial for crop irrigation because of the removal of
791 salts from the effluent. However, for membrane technologies to become sustainable there is
792 need for a deep study of the adequate treatment and/or disposal of concentrates on a case by
793 case basis. Implementation of effective concentrate treatment has the potential to enhance
794 treatment efficiency, move towards a near zero-liquid discharge and avoid unwanted discharge
795 of CEC.

796

797 3.2 Advantages, drawbacks and recommendations of the treatment schemes

798 The main objective of this discussion and analysis is to suggest the "best available technologies
799 able to minimize the release of microcontaminants including ARB&ARGs, and biological risk,
800 and fulfill requirements for a safe reuse for crop irrigation". Important issues for all TT
801 discussed before are summarized in Table 2. Accordingly, and considering that no exhaustive
802 comparative studies addressing CECs and ARB&ARGs removal by advanced treatment
803 methods are available in scientific literature (Rizzo et al., 2019a), a comparative economic
804 evaluation would be questionable. In particular, advanced treatment methods have been
805 compared in terms of either CECs removal, costs, disinfection efficiency, ARB and ARGs
806 removal, formation of DBPs and oxidation reaction products, and final toxicity, but the whole
807 impact on the environment through the simultaneous evaluation of all these issues has not been
808 investigated (Rizzo et al., 2019a). A recommendation needs to be case-specific, taking into
809 account possible regional regulations on wastewater reuse for crop irrigation, intake and
810 required water quality, and local climate conditions, and the relative importance of each aspect
811 needs to be carefully evaluated.

812

813 **Table 2**

4. Concluding remarks

The safety of treated wastewater to be reused for crop irrigation is a relevant issue worldwide. Recently the interest has increased at EU level and stimulated a discussion among policy makers, scientists, professionals, practitioners and other stakeholders, because the European Commission is about to approve a regulation on "Minimum requirements for water reuse" (European Parliament, 2019). Accordingly, the aim of this paper is to provide a technical contribution to this discussion by recommending possible advanced treatment options to make wastewater reuse safer, in particular with regard to the removal of CECs and ARB&ARGs. Different factors affect the choice of the most suitable treatment approach (i.e., water quality, local regulation/restrictions, process costs, type of crop, irrigation method, soil type, environmental footprint, social acceptance, etc.). Nevertheless, an attempt was made in this manuscript by discussing possible BATs for the advanced treatment of urban wastewater including their advantages and drawbacks.

The main conclusion of this work, that gathers the efforts of a group of international experts, members of the NEREUS COST Action ES1403, is that a single advanced treatment method is not sufficient to minimize the release of chemical CECs and ARB&ARGs and make wastewater reuse for crop irrigation safer, but a smart combination of them (Figure 1) and a suitable monitoring program (Table 2) would be necessary. This conclusion stems from the awareness that each treatment method has its own weaknesses/drawbacks, for example:

- a biological post-treatment to remove oxidation by-products may be necessary when ozonation or AOP is used as advanced treatment.;
- ozonation and AOPs require toxicity monitoring because of possible formation of problematic oxidation reaction products;

- adsorption processes should be followed by an effective disinfection process (i.e., UV disinfection) to meet the stringent limits for wastewater reuse;
- if PAC is used, a subsequent filtration or membrane process should be applied to remove the adsorbent particles;
- chemical disinfection is not effective in the removal of CECs and ARGs, thus it should be coupled to other advanced treatment methods. Moreover, possible formation of DBPs (i.e., chlorination by products) should be considered, and a subsequent treatment for their removal may be necessary;
- NF or RO membrane technology would require a pre-treatment (i.e., sand filtration) to prevent clogging and a sustainable solution for the management of membrane concentrate.

Further comparative studies among different advanced treatment methods on real wastewater, using different criteria (i.e., CECs removal, ARB&ARGs, toxicity, DBPs, costs) are recommended. The results will be useful to UWTPs managers to select the most suitable options to be implemented at their own facilities to successfully address wastewater reuse challenges.

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Table 1. Effect of BATs on the abatement of chemical CECs relevant for crop uptake. Only results from investigations at pilot or full-scale on real wastewater are presented (part of these data is extracted from Table 3 and supplementary information of "Rizzo et al., 2019a").

CEC	Process	Scale of study	Water matrix[1]	DOC (mg/L)	CEC initial concentration	Comments	CEC abatement (%)	Reference
Sulfamethoxazole	PAC	Pilot/full	RMW	5-10	171 ng/L (data only from 1 paper)	10-20 mg PAC/L. 0.3-1h contact time.	58-64	Boehler et al. 2012; Margot et al. 2013
	GAC	Pilot	RMW	5.8	145 ng/L	7400 bed volumes treated. 14 min EBCT.	59	Bourgin et al. 2018
	O_3	Pilot/full	RMW	3.5-8.6	-	0.61±0.04 g O_3/g DOC.	94-97	Hollender et al. 2009; Kreuzinger et al. 2015; Bourgin et al. 2018.
	Free chlorine	Full	RMW	-	576 ng/L	Neutral pH, sample taken from the effluent of chlorination unit (dose not provided)	89.6	Renew and Huang, 2004
	PAA	Pilot	RMW	24	100 µg/L	2.0 mg PAA/L, 300 min	52	Rizzo et al., 2019b
	UV	Pilot	RMW	24	100 µg/L	4.58 kJ/L	100	Rizzo et al., 2019b
	Solar photo-Fenton (CPC rector)	Pilot	RMW/SRMW	10.2-42.7	5.5 ng/L – 1879 µg/L	Fe: 5 – 10 mg/L; H_2O_2: 20 – 100 mg/L; pH: 2.8 or higher (5-6).	>80-100	Klamerth et al., 2010; Karaolia et al., 2014,

							2017; Prieto-Rodríguez et al., 2013;
Solar photo-Fenton (Raceway pond)	Pilot	RMW	40	282 ± 36.7 ng/L	Continuous mode. Two liquid depths (5, 15 cm) and three HRTs (80, 40, 20 min); Fe: 5.5 mg/L; H_2O_2: 30 mg/L. pH 2.8.	81-100	Arzate et al., 2017
Photo Fenton	Pilot	RMW	5-7.5[2]	487 ng/L	30 mg H_2O_2/L; 2 mg Fe/L. pH 6-7 (no chelating agents added). 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2.	82	De la Cruz et al., 2013
UV/H_2O_2	Pilot	RMW	5-7.5[2]	487 ng/L	30 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2.	89	De la Cruz et al., 2013
sunlight/TiO_2 (CPC reactor)	Pilot	SRMW	13	100 µg/L	TiO_2 immobilized on glass spheres (0.335 g TiO_2/L). k=0.03 1/min	100	Miranda-García et al. 2011
RO	Pilot	Secondary treated wastewater	-	56 ng/L	Saehan 4040 FL, Flux = 20 L/(m^2.h)	>98	Snyder et al. 2007
		RMW/primary treated wastewater	7.8	15-1800 ng/L	Saehan 4040 FL, Osmonics AK4040, Flux = 17-20 L/(m^2.h)	94-99 (based on 2 studies)	
		Secondary /Tertiary treated	-	805-1030 ng/L	Hydranautics ESPA2	>99	

			wastewater				(based on 2 studies)	
	RO	Pilot	RMW	-	85-122 ng/L	Filmtec TW30 25–40, Flux = 22-31 L/(m^2.h) Filmtec BW30–400, Flux = 45 L/(m^2.h)	98 98	Sahar et al 2011
	RO	Pilot	RMW	-	20-27 ng/L	Ropur TR70-4021-HF	>99	Dolar et al. 2012
	NF RO	Pilot	RMW	-	100-500 ng/L	Filmtec NF90, MWCO 200 Da, Flux = 18 L/(m^2.h) Hydranautics ESPA2, MWCO 100 Da, Flux = 18 L/(m^2.h)	99 100	Mamo et al. 2018
Diclofenac	PAC	Pilot	RMW	7.3(±1.9)	1187 ng/L	10-20 mg PAC/L; 0.3-0.7h contact time.	69	Margot et al. 2013
	GAC	Pilot	RMW	4.4	1008 ng/L	23400 bed volumes treated. 14 min EBCT.	72	Bourgin et al. 2018
	O_3	Pilot/full	RMW	3.5-8.6	-	0.61(±0.04) g O_3/g DOC.	98-100	Hollender et al. 2009; Kreuzinger et al. 2015; Bourgin et al. 2018.
	Free chlorine	Full	RMW	-	-	Neutral pH	60	Anumol et al., 2016
	PAA	Pilot	RMW	24	100 µg/L	2.0 mg PAA/L, 60 min	80	Rizzo et al., 2019b

UV	Pilot	RMW	24	100 µg/L	2.22 kJ/L	90	Rizzo et al., 2019b
Photo-Fenton	Pilot	RMW	5-7.5[2]	925 ng/L	20-50 mg H_2O_2/L; 2-4 mg Fe/L. pH 6-7. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2.	93-100	De la Cruz et al., 2013
Solar photo-Fenton (CPC rector)	Pilot	RMW/SRMW	10.2-36	1 – 5100 µg/L	Fe: 5 – 10 mg/L; H_2O_2: 20 – 60 mg/L; pH: 2.8 or neutral (chelating agent used).	80-100	Klamerth et al., 2010, 2011; Prieto-Rodríguez et al., 2013;
UV/H_2O_2	Pilot	RMW	5-7.5[2]	925 ng/L	20-50 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2.	99-100	De la Cruz et al., 2013
sunlight/TiO_2 (CPC reactor)	Pilot	RMW/SRMW	13-23	414 ng/L-100 µg/L	20 mg/L TiO_2 and supported TiO_2, neutral pH.	80-100	Miranda-García et al., 2011; Prieto-Rodríguez et al., 2012;
RO	Pilot	Secondary treated wastewater	-	37 ng/L	Saehan 4040 FL, Flux = 20 L/(m^2.h)	>97	Snyder et al. 2007
		RMW/primary treated wastewater	7.8	1.1-38 ng/L	Saehan 4040 FL, Osmonics AK4040, Flux = 17-20 L/(m^2.h)	>93% (from 2 pilots)	
		Secondary /Tertiary					

			treated wastewater	-	49-59 ng/L	Hydranautics ESPA2	>98 (from 2 pilots)	
	RO	Pilot	RMW	-	500-580 ng/L	Filmtec TW30 25–40, Flux = 22-31 L/(m^2.h) Filmtec BW30–400, Flux = 45 L/(m^2.h)	95-99 (from 2 pilots)	Sahar et al 2011
	NF	Pilot	Effluent UWTP	-	720 ng/L	Flux = 1-2 LMH, TMP = 0.7 bar	60-65	Röhricht et al. 2009, 2010
	NF RO	Pilot	RMW	-	260-440 ng/L	FILMTEC NF90-4040, 200 Da FILMTEC BW30-4040	87-98 88-96	Cartagena et al. 2013
	NF RO	Pilot	RMW	-	100-500 ng/L	Filmtec NF90 MWCO=200 Da, Flux = 18 L/(m^2.h) Hydranautics ESPA2 MWCO 100 Da, Flux = 18 L/(m^2.h)	100 100	Mamo et al. 2018
Carbamazepine	PAC	Pilot/full	RMW	5-10	221-461 ng/L	10-20 mg PAC/L; 0.3-1h contact time; data from 3 papers.	90-92	Boehler et al., 2012; Margot et al., 2013; Mailler et al., 2015; Karelid et al., 2017.
	GAC	Pilot	RMW	4.4	110 ng/L	23400 bed volumes treated. 14 min EBCT.	72	Bourgin et al. 2018

O_3	Pilot/full	RMW	3.5-7.6	-	0.61±0.04 g O_3/g DOC.	97-100	Hollender et al. 2009; Kreuzinger et al. 2015; Bourgin et al. 2018.
Free chlorine	Full	RMW	-	-	Neutral pH	No removal	Anumol et al., 2016
PAA	Pilot	RMW	24	100 µg/L	2.0 mg PAA/L, up to 300 min	No removal	Rizzo et al., 2019b
UV	Pilot	RMW	24	100 µg/L	15.12 kJ/L	16	Rizzo et al., 2019b
Solar photo-Fenton (CPC rector)	Pilot	RMW/SRMW	10-36	70 ng/L- 100 µg/L	Fe: 5 mg/L; H_2O_2: 50 – 60 mg/L; pH: 2.8 or neutral (chelating agent used).	>24-100	Klamerth et al., 2010, 2011; Prieto-Rodríguez et al., 2013;
Solar photo-Fenton (Raceway pond)	Pilot	RMW	40	422 ± 54.9 ng/L	Two liquid depths (5, 15 cm) and three HRTs (80, 40, 20 min); Fe: 5.5 mg/L; H_2O_2: 30 mg/L. pH 2.8	86-96	Arzate et al., 2017
Photo-Fenton	Pilot	RMW	5-7.5[2]	333 ng/L	20-50 mg H_2O_2/L; 2-4 mg Fe/L. pH 6-7. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2.	66-94	De la Cruz et al., 2013
UV/H_2O_2	Pilot	RMW	5-7.5[2]	333 ng/L	20-50 mg H_2O_2/L. 5 low pressure mercury lamps (254 nm) of 150 W each, incident light 70 W/m^2.	82-99	De la Cruz et al., 2013

sunlight/TiO_2 (CPC reactor)	Pilot	SRMW	13	100 µg/L	TiO_2 immobilized on glass spheres.	50-80	Miranda-García et al. 2011
sunlight/TiO_2 (CPC reactor)	Pilot	RMW	15-50	56 ng/L	0.2 g TiO_2 powder/L.	65-80	Bernabeu et al. 2011
RO	Pilot	Secondary treated wastewater	-	147 ng/L	Saehan 4040 FL, Flux = 20 L/(m^2.h)	>99	Snyder et al. 2007
		RMW/primary treated wastewater	7.8	181-410 ng/L	Saehan 4040 FL, Osmonics AK4040, Flux = 17-20 L/(m^2.h)	>99 (from 2 pilots)	
		Secondary /Tertiary treated wastewater	-	237-271 ng/L	Hydranautics ESPA2	>99 (from 2 pilots)	
RO	Pilot	RMW		64-99 ng/L	Ropur TR70-4021-HF	>99	Dolar et al. 2012
NF	Pilot	Effluent UWTP	-	640 ng/L	Flat sheet, Flux = 1-3 L/(m^2.h), TMP = 0.3-0.7 bar	12	Röhricht et al. 2009, 2010
NF	Pilot	RMW	-	300-380 ng/L	FILMTEC NF90-4040, 200 Da	78-92	Cartagena et al. 2013
RO					FILMTEC BW30-4040	82-93	
NF	Pilot	RMW	-	100-500 ng/L	Filmtec NF90 MWCO=200 Da, Flux = 18 L/(m^2.h)	79	Mamo et al. 2018

	RO	Hydranautics ESPA2 MWCO 100 Da, Flux = 18 L/(m^2.h)	100

[1]RMW= real municipal wastewater; SRMW= spiked real municipal wastewater; [2]TOC.

Table 2 Advantages, drawbacks and recommendations for each TT in Figure 1.

TT (advanced treatment)	Advantages	Drawbacks	Recommendations
a or b (UV)	• Effective disinfection (including ARB inactivation) • No DBPs formation compared to chemical disinfection	• If local standards for reuse are too stringent for residual bacterial density, UV may not be sufficient • Poor/no CECs removal • Partial removal of ARGs	• Compliance with local residual bacterial density standards should be evaluated
c (chemical disinfection)	• Effective disinfection (including ARB inactivation)	• Poor/no removal of CECs and ARGs • Formation of DBPs • If local standards for reuse are too stringent for DBPs, some disinfectant cannot be used (e.g., chlorine in Italy)	• Toxicity tests recommended • DBPs (depending on the disinfectants used) should be monitored
d (O_3/AOP and biological post-treatment)	• Effective disinfection (including ARB inactivation) • CECs abatement high during ozonation and (solar) photo Fenton, moderate with UV/H_2O_2 • Full-scale evidence on practicability only for O_3	• Formation of some DBPs (NDMA, bromate) during ozonation • Formation of oxidation transformation products during AOP and ozonation • partial ARGs removal	• Toxicity tests recommended • NDMA and bromate should be monitored in O_3 treatment
e (GAC and UV)	• effective disinfection by UV • high CECs removal by GAC • full-scale evidence on practicability	• Poor/no removal of ARB&ARGs by GAC alone • for UV see above, TT a & b	• Decreasing adsorption capacity with increasing bed volume should be taken into account
f (PAC and UV)	• Effective disinfection by UV	• Poor/no removal of ARB&ARGs by PAC alone	

	• High CECs removal by PAC • Full-scale evidence on practicability for CEC removal by PAC	• For UV see above, TT a & b	
g (NF or RO membrane filtration, with potential pre-treatment with MF or UF membranes)	• Effective disinfection for bacteria (incl. ARB) and protozoa for all membranes; viruses well removed by UF, NF & RO • ARGs well removed by NF and RO • CECs removal from poor (MF, UF) to very good (NF, RO) depending on membrane type, • RO and partially also NF reduce salinity • For post UV-C see TT a & b	• Poor/no removal of ARGs at full-scale by MF (for UF some removal is expected) • Poor CECs removal for MF and UF • High energy requirements for NF and RO • Generation of a substantial concentrate waste stream by NF and RO • For post UV-C see TT a&b	• Impact of membrane characteristics on disinfection, ARB, ARG, and CEC removal should be carefully considered in design • Consider AOP instead of UV disinfection if the risk of unknowns and spills is considered high • Consider high UV doses if NDMA can be suspected in the membrane effluent (e.g. following prior chloramination)

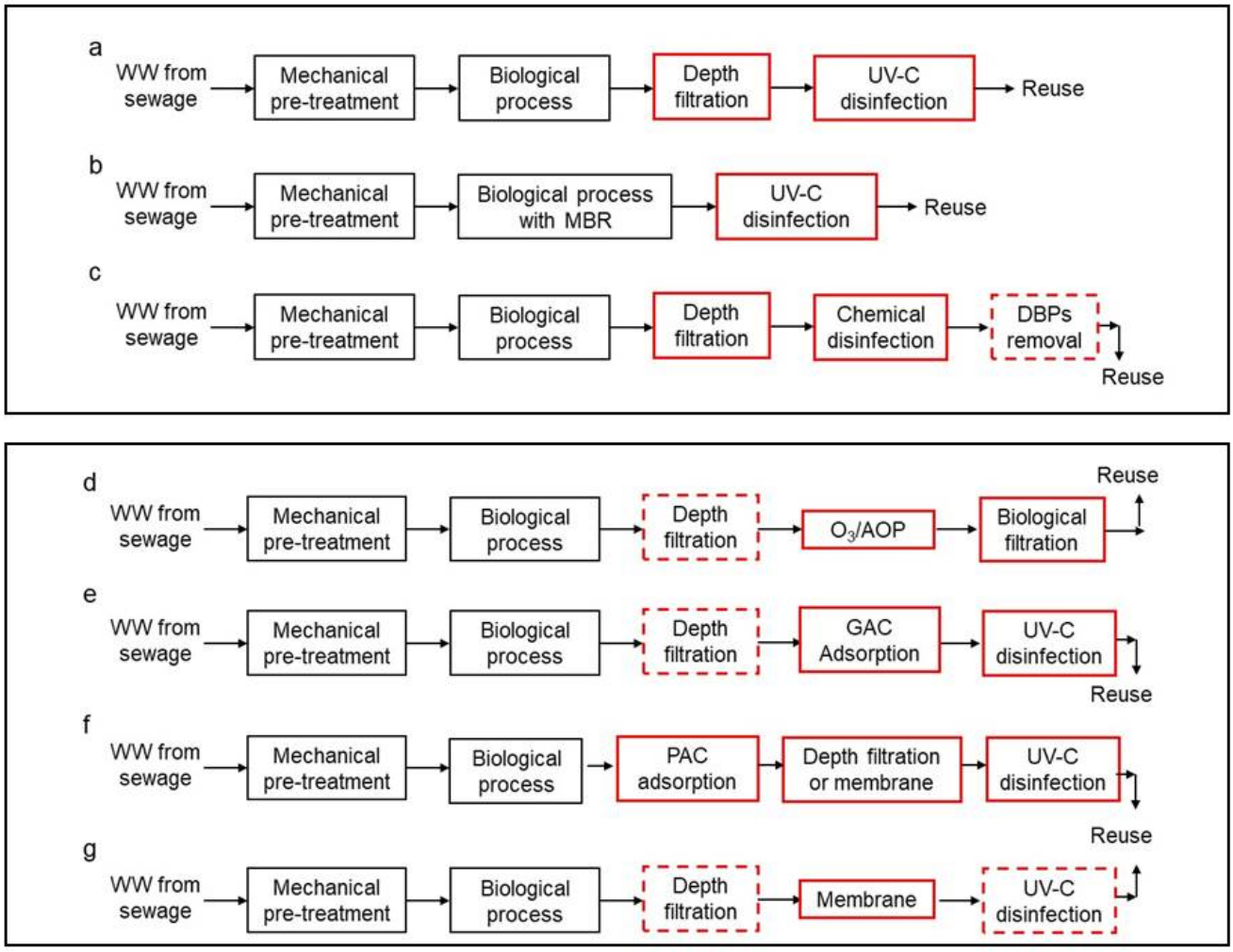

Figure 1. Different options of treatment trains for urban wastewater reuse to address traditional parameters set in wastewater reuse regulation and guidelines (e.g., BOD, COD, TSS, *E. coli* etc.) (a, b, c) and to effectively remove CECs in addition to the typical parameters (d, e, f, g). Advanced treatment in red lines; red dotted lines mean that process application should be evaluated case by case. "Biological process" followed by "depth filtration" may be replaced by "MBR" for treatment trains "d" and "e".