

1 Best available technologies and treatment trains to address current challenges in urban 2 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.

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55 Keywords: activated carbon, advanced oxidation processes, antibiotic resistance,

56 contaminants of emerging concern, disinfection, ozonation

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58	List of abbreviations
59	ARB= antibiotic resistant bacteria
60	ARGs= antibiotic resistance genes
61	AOPs= advanced oxidation processes
62	BAC= biological activated carbon
63	CBZ= carbamazepine
64	CECs= contaminants of emerging concern
65	CPC= compound parabolic collector
66	DBPs= disinfection by products
67	DCF= diclofenac
68	DOC= dissolved organic carbon
69	ERY= erythromycin
70	FRC= free residual chlorine
71	GAC= granular activated carbon
72	HO•= hydroxyl radical
73	LRV= Log removal value
74	MDR= multi drug resistant
75	MF= microfiltration
76	NDMA= N-nitrosodimethylamine
77	NF= nanofiltration
78	PAC= powdered activated carbon
79	RO= reverse osmosis
80	TMP= transmembrane pressure
81	UF= ultrafiltration
82	SMX= sulfamethoxazole
83	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 (Fuhriemann 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).

According to scientific literature, conventional treatment trains in UWTPs are poorly effective to comprehensively remove CECs (Petrie et al., 2015; Falas et al., 2016; Krzeminski et al., 2019), which can finally be released into the environment, constituting a particular concern when effluents are reused for crop irrigation. To be able to meet stringent limits for wastewater reuse as well as to effectively remove CECs, advanced treatment steps should be implemented in conventional UWTPs (Krzeminski et al., 2019; Rizzo et al., 2019a). However, while the effect of biological processes (Boshir Ahmed et al., 2017; Tiwari et al., 2017; Krzeminski et al., 2019) and advanced treatment technologies (Miklos et al., 2018; von Gunten, 2018; Roccaro, 2018; Marron et al., 2019; Rizzo et al., 2019a; Siegrist et al. 2019) on chemical CECs has been reviewed in different papers, less information is available about ARB&ARGs and, most importantly, on possible treatment trains combining several processes to successfully address these challenges.

This review paper gathers the efforts of a group of international experts, members of the NEREUS COST Action ES1403¹ “New and emerging challenges and opportunities in wastewater reuse” (Fatta-Kassinos et al., 2015), who for three years have been constructively discussing the effect of the best available technologies (BATs) for urban wastewater treatment on CECs and ARB&ARGs. Accordingly, the objective of this paper is to introduce and discuss the BATs for advanced treatment of urban wastewater, as well as possible treatment trains to control the release of CECs, including ARB&ARGs, to produce wastewater for safe and sustainable reuse practices in agriculture. In particular, the capability of ozonation, activated carbon adsorption, chemical oxidants/disinfectants, UV radiation, advanced oxidation 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

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

2.1 Ozonation

2.1.1 Abatement of CECs

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

activated carbon (BAC) filter) is recommended to eliminate 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 O₃/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 (*ermB*) but a simultaneous increase in the abundance of ARGs (*vanA*, *bla_{VIM}*) within the surviving wastewater population. Ozonation operated at high contact time (40 min) with an ozone dose of 0.25 g O₃/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 (Lüddecke et al., 2014; Zhuang et al., 2015; Alexander et al.,

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

2.1.3 Formation of oxidation by-products

Ozonation can result in the formation of biologically potent (e.g. toxic, mutagenic) oxidation by-products. Among them, N-nitrosodimethylamine (NDMA) and bromate are of particular concern for human health because they are potentially carcinogenic. Therefore, NDMA and bromate need to be measured to test the feasibility of ozonation as an option for advanced wastewater treatment at a specific location (Schindler Wildhaber et al., 2015). Only if the concentrations expected after dilution of discharged effluents are clearly below (potential) drinking water standards (10 µg/L for bromate, 10 ng/L for NDMA, Bourgin et al., 2018), ozonation is considered suitable. Bromate results from the reaction of O₃ and HO• with bromide. NDMA can be formed from the reaction of amine precursors (e.g. containing hydrazine, sulfamide, and dimethylamino functional groups) with generally low yields but that can reach up to ≥ 50% in exceptional cases (Kosaka et al. 2009; Schmidt and Brauch 2008; von Gunten et al. 2010; Krasner et al. 2013, Sgroi et al., 2014). Because precursors are mostly unknown or unidentified in wastewater, the formation of NDMA cannot be excluded a priori. 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 O₃/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 €/m³) 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 €/m³ treated wastewater, decreasing further with increasing plant size even below 0.05 €/m³, 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/m³, 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/m³, for

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

2.3 Chemical oxidants/disinfectants

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

2.3.1 Chlorination

Wastewater disinfection by chlorine is typically performed by chlorine gas (in medium – large UWTPs) or hypochlorite (either calcium or sodium). Limited studies have focused on the abatement of CECs by chlorine, which was found to be quite poor, in particular if compared to oxidation/disinfection processes with higher oxidation potential such as ozone and other AOPs (Anumol et al., 2016; Hua et al., 2019). For example, Li and Zhang (2011) reported abatement of antibiotics during wastewater treatment with chlorine in the range of 18% (roxithromycin) to 40% (trimethoprim), while cephalexin and ampicillin were abated by 99% and 91%, respectively. However, the chlorine dose was not reported in this study, and cephalexin and 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 ($[FRC]_0 = 20 \mu\text{M}$ (1.4 mg/L)) (Dodd and Huang, 2004). This expectation is supported by existing observations at full-scale UWTs, 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 *ereA* and *ermB* persisted in chlorinated (15 mg Cl₂ min/L) urban wastewater samples (Yuan et al. 2015) and chlorination was found to be effective in ARGs removal (3.16 Log for *sulI* and 3.24 Log for *tetG* 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 (*tetA*, *tetM*, *tetO*, *tetQ*, *tetW*, *sulI* and *sulII*) abundance to some extent (less than 1 Log unit for *tetA*) 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 UWTs, chlorination did not prove to have significant contribution to ARGs (*tetA*, *tetW*, *tetO*, *ermB*, *qnrS*, *bla*_{TEM} *sulI*) 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 UWTs 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/cm², 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 (*sulIII* and *tetA*) out of 4 (*ermB* and *qnrS*) 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 \times 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/cm²) (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/cm² (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/cm²). As UV dose was further increased to 20 and 50 mJ/cm², 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/cm² (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 *ampC*, *mecA*, *tetA* and *vanA* (McKinney and Pruden, 2012). Actually, also lower UV doses (5-10 mJ/cm²) were found to be effective in the removal of ARGs (namely *ereA*, *ereB*, *ermA*, *ermB*, *tetA*, *tetO*) 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 (*tetA*, *tetM*, *tetO*, *tetQ*, *tetW*) and two sulfonamide resistance genes (*suII*, *suIII*) were observed in UV disinfection (UV fluence 10-160 mJ/cm²) 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%

and 73.5% at the lower fluence UV disinfection (40 mJ/cm² or less), and between 79.7%, and 92.0% at high fluence (160 mJ/cm²). Lower removal efficiencies were observed for *suII*, *suIII* (78.1% and 71.1% respectively, at the higher fluence).

In full-scale monitoring (5 UWWTPs in the USA), UV radiation employed for disinfection did not prove to have a significant contribution to ARGs (*tetw*, *tetO*, *suII*) and ARB reduction (Munir et al., 2011). These results were confirmed in a subsequent study at full-scale, where no significant difference in ARGs (namely, *ermB*, *qnrS* and *tetA*) was observed before and after UV disinfection, while for *suIII* even an increase was observed after disinfection (Di Cesare et al., 2016a).

2.5 Advanced oxidation processes

Advanced oxidation processes (AOPs) rely on the formation of hydroxyl radicals that can abate a wide range of CECs (Rizzo, 2011; He et al., 2020) as well as inactivate microorganisms (Dunlop et al., 2010; Fiorentino et al., 2015). A possible classification of AOPs includes two groups: homogeneous processes (e.g., UV/H₂O₂, UV/Fe/H₂O₂, O₃, O₃/H₂O₂ etc.) and heterogeneous (solid semiconductors + light source, e.g., UV/TiO₂, UV/ZnO) photocatalytic processes. Homogeneous processes have been widely investigated as advanced treatment of urban wastewater effluents and either are already applied at full-scale (e.g., O₃, see section 2.1) or are characterized by short-/mid-term perspective application (e.g., UV/H₂O₂, UV/Fe/H₂O₂) as opposed to heterogeneous photocatalytic processes (Rizzo et al., 2019a; Maniakova et al., 2020). The main reason why heterogeneous photocatalytic processes are not ready for full-scale application as advanced urban wastewater treatment are related to photocatalyst preparation costs, photocatalyst quantum yield (effectiveness) and reactor configuration (Iervolino et al., 2020). In particular, heterogeneous photocatalytic processes can be operated under two main

configurations: (i) with the photocatalyst suspended in the reactor (i.e., slurry system) or (ii) attached to a support (i.e., immobilized system). Due to the higher specific surface area available, a slurry system is more effective than an immobilized one, but a subsequent expensive separation process (e.g., coagulation, filtration, membrane) is necessary to recover the photocatalyst before effluent discharge or reuse (Fernández-Ibáñez et al., 2003). Immobilized photocatalytic systems have relatively lower quantum efficiency than slurry ones, which results in longer treatment time and consequently larger water volume to treat (Spasiano et al., 2015). Some homogeneous photo-driven AOPs can also be operated under natural sunlight (solar/H₂O₂ or solar/Fe/H₂O₂) thus saving energy costs (Klamerth et al., 2010; Ortega-Gomez et al., 2014; Ferro et al., 2015; Giannakis et al., 2016) and this can be considered as an attractive option for small UWTPs in areas with sufficient sunlight.

2.5.1 Abatement of CECs

Due to their high redox potential hydroxyl radicals oxidize a wide spectrum of organic contaminants, accordingly, AOPs successfully degrade several organic micropollutants (Klavarioti et al, 2009; Rizzo, 2011). The most common AOPs studied are UV/H₂O₂, O₃/H₂O₂, O₃/UV, Fenton (Fe/H₂O₂), photo-Fenton (UV/Fe/H₂O₂) and heterogeneous photocatalysis (e.g., UV/TiO₂, UV/ZnO). Although UV/H₂O₂, is more efficient than UV alone to abate CECs, still more energy is needed compared to ozonation (Rizzo et al., 2019a). O₃/H₂O₂ does not improve abatement of CECs compared to ozone alone in UWTP effluents, since effluent ozonation can be considered an intrinsically AOP due to the high HO[•] generation potential of the organic matrix (Buffle et al., 2006), at the same time HO[•] are scavenged by the matrix (Acero and von Gunten, 2001; Kovalova 2013). Fenton and photo-Fenton processes are typically effective under acidic conditions (pH 3) and the abatement of three antibiotics, namely SMX, 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 H₂O₂ (< 20 mg/L) concentrations and pH 5-6, thus avoiding the necessity for final separation of soluble iron species from the treated wastewater (Klammerth 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/H₂O₂ 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/H₂O₂ 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.

524 However, the release of mobile genetic elements from bacterial cells, that may take place after
 525 disinfection process, and the potential to transfer antibiotic resistance through horizontal
 526 mechanism, have been poorly investigated. Photo-driven AOPs have recently been investigated
 527 to evaluate if they can be more effective in the removal of ARGs than conventional disinfection
 528 processes, such as chlorination and UV radiation. Ferro et al. (2016) investigated the effect of
 529 UV/H₂O₂ (broad-band spectrum UV lamp with main emission in the range 320-450 nm), under
 530 realistic conditions for wastewater treatment (natural pH (7.6) and 20 mg H₂O₂/L), on antibiotic
 531 resistance transfer potential in urban wastewater. The investigated process resulted in bacterial
 532 inactivation and a decrease of ARGs in intracellular DNA after 60 min treatment, but UV/H₂O₂
 533 did not remove ARGs effectively. Actually, an increase up to 3.7×10^3 copies/mL ($p > 0.05$)
 534 of *bla*_{TEM} gene was observed in total DNA after 240 min treatment, while no difference ($p >$
 535 0.05) was found for *qnrS* gene between the initial (5.1×10^4 copies/mL) and the final sample
 536 (4.3×10^4 copies/mL). In UV/H₂O₂ process (pH 7, 50-130 mJ/cm²), 4 Log reduction of ARGs
 537 (*amp*^R and *kan*^R) concentration was observed in urban wastewater (Yoon et al., 2017).
 538 According to the results previously discussed for the chlorination process, intracellular ARGs
 539 showed lower rates of damage compared to extracellular ARGs due to cell protective roles and
 540 significant HO[•] radical scavenging by cellular components. Zhang et al. (2016a) showed that
 541 UV/H₂O₂ can effectively remove ARGs (2.8-3.5 logs removal of *sul1*, *tetX*, and *tetG*, within
 542 30 min treatment) but only under conditions that seem unrealistic for full-scale implementation
 543 (pH 3.5 and 340 mg H₂O₂/L), moreover UV fluence was not provided.

544 Solar driven photo-Fenton process is effective in the inactivation of ARB Karaolia et al., 2017;
 545 Fiorentino et al., 2019). When the process (5 mg Fe²⁺/L, 50 mg H₂O₂/L, pH 3) was operated at
 546 pilot scale through a compound parabolic collector (CPC) based reactor, on the effluent of an
 547 MBR, a complete inactivation of the low initial bacterial population (*E. coli* = 2 CFU/100 mL,
 548 *P. aeruginosa* = 4 CFU/100 mL, *Klebsiella* spp. = 3 CFU/100 mL), including antibiotic-tolerant

and susceptible bacteria, was observed, after 54 min of solar radiation intensity normalized time (Karaolia et al., 2017). On the other hand, repair of *P. aeruginosa* was observed, with 2 CFU/100 mL growing on the selective media 24 h after solar Fenton oxidation. Solar photo-Fenton process was also investigated in raceway pond reactors, at neutral pH conditions (20 mg Fe²⁺/L, 50 mg H₂O₂/L), in real urban wastewater and an effective inactivation of *E. coli* and *Enterococcus sp.* cefotaxime resistant bacteria was observed (detection limit (1 CFU/mL) achieved after 30-40 min, 3.2-4.7 kJ/L) (Fiorentino et al., 2019). However, both solar driven photo Fenton processes (CPC reactor at pH 3 and raceway ponds at neutral pH) did not effectively remove the target ARGs..

The effect of heterogeneous photocatalysis with TiO₂ on ARB&ARGs has been investigated in slurry and immobilized systems. According to the results observed for homogenous photo-driven AOPs, even heterogeneous photocatalytic processes, while effective in the inactivation of different antibiotic resistant bacterial populations (Tsai et al., 2010; Rizzo et al., 2014a, Rizzo et al., 2014b; Dunlop et al., 2015; Zammit et al., 2019) may not be effective in the removal of some ARGs (Karaolia et al., 2018).

2.6 Membrane filtration

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

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

2.6.1 Removal of CECs

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

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

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

2.6.2 Effect on ARB&ARGs

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

The effect of membrane filtration, in particular NF and RO, on ARB&ARGs, thus far, has been 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 $1.05 \times 10^7 \pm 3.54 \times 10^6$ gene copies/100mL and $1.92 \times 10^5 \pm 1.06 \times 10^4$ 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 ($5.92 \times 10^3 \pm 1.39 \times 10^3$ gene copies/100mL) and in the groundwater after infiltration ($3.13 \times 10^3 \pm 1.52 \times 10^3$ 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 4.03×10^4 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

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

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

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

Table 1

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

3.1 Treatment trains for a safe reuse

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

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

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

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

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

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

Figure 1

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

Among AOPs, ozonation and photochemical processes showed interesting results in the removal of CECs and ARB. In particular, in the short term, ozonation and UV/H₂O₂ processes are more attractive options (Fig.1, d) compared to other photo-driven AOPs to abate CECs as well as to effectively inactivate bacteria (Rizzo et al., 2019a) because:

1. their efficiency has been confirmed by different works available in scientific literature. However, ozonation needs considerably less energy compared to UV/H₂O₂ treatment for the same CEC abatement level and shows full-scale application;
2. other homogeneous photocatalytic processes (such as photo-Fenton) may request 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

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

3.2 Advantages, drawbacks and recommendations of the treatment schemes

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

Table 2

814

815 4. Concluding remarks

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

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

- 834 • a biological post-treatment to remove oxidation by-products may be necessary when
835 ozonation or AOP is used as advanced treatment.;
- 836 • ozonation and AOPs require toxicity monitoring because of possible formation of
837 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/SR MW	10.2-42.7	5.5 ng/L – 1879 µg/L	Fe: 5 – 10 mg/L; H ₂ O 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 ₂ O 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 ₂ O 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 ₂ O ₂	Pilot	RMW	5-7.5 ²	487 ng/L	30 mg H ₂ O 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)	
Diclofenac	RO	Pilot	RMW	-	85-122 ng/L	Filmtec TW30 25–40, Flux = 22-31 L/(m ² .h)	98	Sahar et al. 2011
						Filmtec BW30–400, Flux = 45 L/(m ² .h)	98	
	RO	Pilot	RMW	-	20-27 ng/L	Ropur TR70-4021-HF	>99	Dolar et al. 2012
	NF	Pilot	RMW	-	100-500 ng/L	Filmtec NF90, MWCO 200 Da, Flux = 18 L/(m ² .h)	99	Mamo et al. 2018
	RO					Hydranautics ESPA2, MWCO 100 Da, Flux = 18 L/(m ² .h)	100	
	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 ₂ O 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 reactor)	Pilot	RMW/SR MW	10.2-36	1 – 5100 µg/L	Fe: 5 – 10 mg/L; H ₂ O 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 ₂ O ₂	Pilot	RMW	5-7.5 ²	925 ng/L	20-50 mg H ₂ O 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/SR MW	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	Pilot	RMW	-	260-440 ng/L	FILMTEC NF90-4040, 200 Da	87-98	Cartagena et al. 2013
	RO					FILMTEC BW30-4040	88-96	
	NF	Pilot	RMW	-	100-500 ng/L	Filmtec NF90 MWCO=200 Da, Flux = 18 L/(m ² .h)	100 100	Mamo et al. 2018
	RO					Hydranautics ESPA2 MWCO 100 Da, Flux = 18 L/(m ² .h)		
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/SR MW	10-36	70 ng/L- 100 µg/L	Fe: 5 mg/L; H ₂ O 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 ₂ O 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 ₂ O 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 ₂ O ₂	Pilot	RMW	5-7.5 ²	333 ng/L	20-50 mg H ₂ O 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 r	-	147 ng/L	Saehan 4040 FL, Flux = 20 L/(m ² .h)	>99	Snyder et al. 2007
		RMW/primary treated wastewater r	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 r	-	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
100 Da, Flux = 18 L/(m².h)

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

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

TT (advanced treatment)	Advantages	Drawbacks	Recommendations
a or b (UV)	<ul style="list-style-type: none"> • Effective disinfection (including ARB inactivation) • No DBPs formation compared to chemical disinfection 	<ul style="list-style-type: none"> • 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 	<ul style="list-style-type: none"> • Compliance with local residual bacterial density standards should be evaluated
c (chemical disinfection)	<ul style="list-style-type: none"> • Effective disinfection (including ARB inactivation) 	<ul style="list-style-type: none"> • 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) 	<ul style="list-style-type: none"> • Toxicity tests recommended • DBPs (depending on the disinfectants used) should be monitored
d (O ₃ /AOP and biological post-treatment)	<ul style="list-style-type: none"> • Effective disinfection (including ARB inactivation) • CECs abatement high during ozonation and (solar) photo Fenton, moderate with UV/H₂O₂ • Full-scale evidence on practicability only for O₃ 	<ul style="list-style-type: none"> • Formation of some DBPs (NDMA, bromate) during ozonation • Formation of oxidation transformation products during AOP and ozonation • partial ARGs removal 	<ul style="list-style-type: none"> • Toxicity tests recommended • NDMA and bromate should be monitored in O₃ treatment
e (GAC and UV)	<ul style="list-style-type: none"> • effective disinfection by UV • high CECs removal by GAC • full-scale evidence on practicability 	<ul style="list-style-type: none"> • Poor/no removal of ARB&ARGs by GAC alone • for UV see above, TT a & b 	<ul style="list-style-type: none"> • Decreasing adsorption capacity with increasing bed volume should be taken into account
f (PAC and UV)	<ul style="list-style-type: none"> • Effective disinfection by UV 	<ul style="list-style-type: none"> • Poor/no removal of ARB&ARGs by PAC alone 	

	<ul style="list-style-type: none"> • High CECs removal by PAC • Full-scale evidence on practicability for CEC removal by PAC 	<ul style="list-style-type: none"> • For UV see above, TT a & b 	
g (NF or RO membrane filtration, with potential pre-treatment with MF or UF membranes)	<ul style="list-style-type: none"> • 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 	<ul style="list-style-type: none"> • 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 	<ul style="list-style-type: none"> • 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)

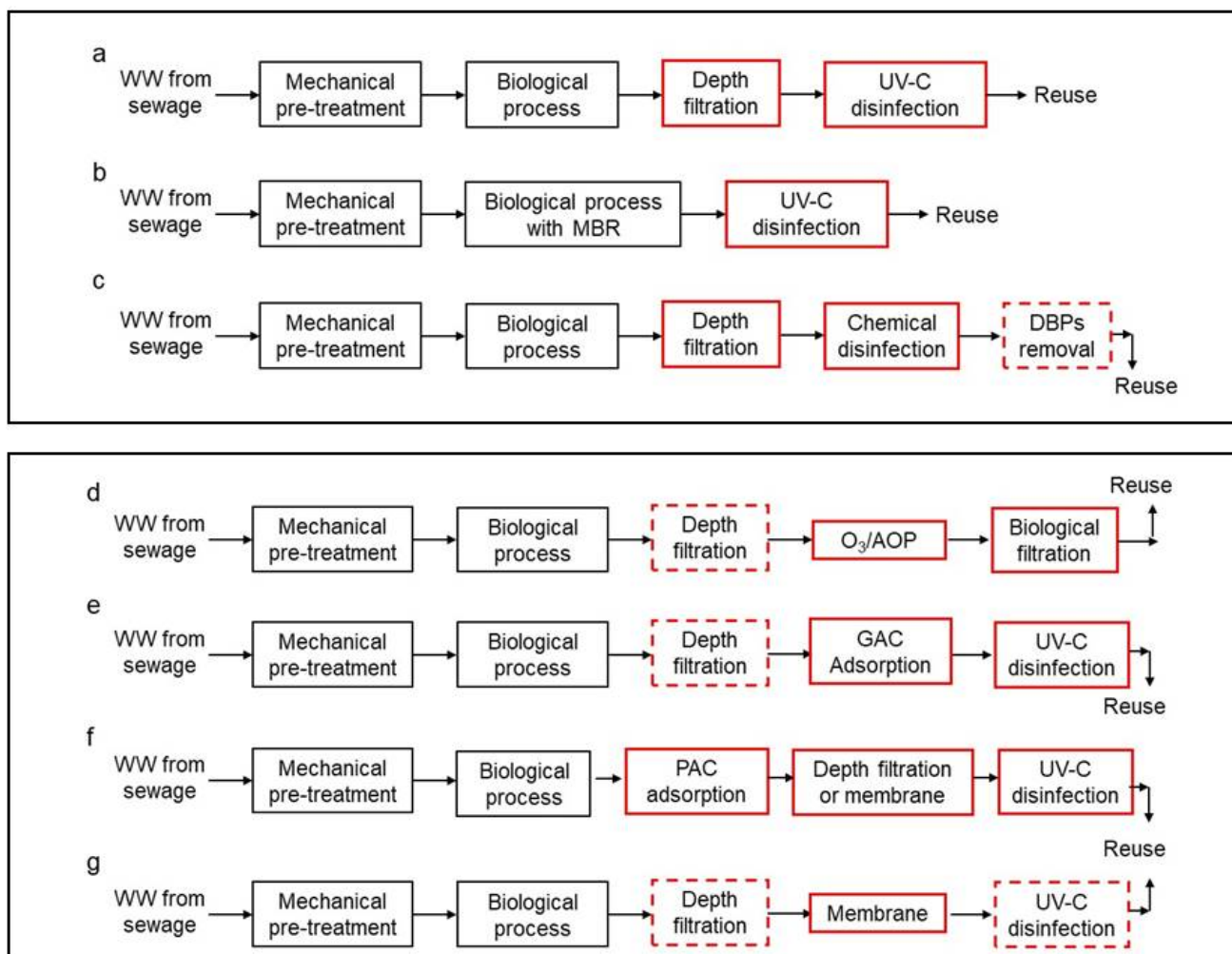


Figure 1. Different options of treatment trains for urban wastewater reuse to address traditional parameters set in wastewater reuse regulation and guidelines (e.g., BOD, COD, TSS, *E. coli* etc.) (a, b, c) and to effectively remove CECs in addition to the typical parameters (d, e, f, g). Advanced treatment 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”.