Open Access Library Journal
2020, Volume 7, e6161
ISSN Online: 2333-9721
ISSN Print: 2333-9705

UV-C/H_2O_2 and Sunlight/H_2O_2 in the Core of the Best Available Technologies for Dealing with Present Dares in Domestic Wastewater Reuse

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How to cite this paper: Ghernaout, D. and Elboughdiri, N. (2020) UV-C/H_2O_2 and Sunlight/H_2O_2 in the Core of the Best Available Technologies for Dealing with Present Dares in Domestic Wastewater Reuse. *Open Access Library Journal*, **7**: e6161.
https://doi.org/10.4236/oalib.1106161

Received: February 13, 2020
Accepted: March 13, 2020
Published: March 16, 2020

Abstract

Traditional urban wastewater treatment plants (UWTPs) are deficiently efficient in eliminating most contaminants of emerging concern (CECs), comprising antibiotics, antibiotic resistant bacteria and antibiotic resistance genes (ARB & ARGs). Such pollutants lead to some worry for nature and human health. This work discusses the performance of the best available technologies (BATs) for dealing with urban wastewater (UWW) to eliminate CECs and ARB & ARGs. Ozonation, activated carbon adsorption, chemical disinfectants, UV radiation, advanced oxidation processes (AOPs) and membrane filtration are debated with a view to their potential to efficaciously reduce CECs and ARB & ARGs. Probable treatment trains involving the BATs are compared. In spite of the huge improvements acquired in terms of applying AOPs and understanding their mechanisms in removing ARB & ARGs, transformation products (TPs) of the antibiotics existing may be generated, which may be less bio-decomposable, more poisonous and biologically strong, juxtaposed to the parent compounds. Therefore, attempts have to be concentrated on defining the structure of such TPs and proving if these retain their core moieties, responsible for the antimicrobial activity of the antibiotic, probably comprising antimicrobial resistance to the surrounding microbes.

Subject Areas

Environmental Sciences

Keywords

Urban Wastewater (UWW), Contaminants of Emerging Concern (CECs), Antibiotic Resistant Bacteria and Antibiotic Resistance Genes (ARB & ARGs), Advanced Oxidation Processes (AOPs), Best Available Technologies (BATs), Transformation Products (TPs)

1. Introduction

In several regions in the world, water lack has provoked diverse health and economic issues [1] [2]. Such circumstances are awaited to worsen because of the climate modification and extra stress parameters [3] [4]. Recovered water arriving from urban wastewater treatment plants (UWTPs) is seen as one of the major actions for diminishing the water emergency, since it may be an appropriate solution to water supply for the irrigation of crops [5]. Such procedure has in the past few years been encouraged by the European Union, which has suggested a regulation fixing the minimum quality criteria for recovered water designated for agricultural irrigation. The regulation admits the necessity of evaluating the hazard related to contaminants of emerging concern (CECs) and antimicrobial resistance [6] [7] [8]. The rise and diffusion of antimicrobial resistance is certified as one of the main Global Health dares of the present century through thorough control of grave problem areas, comprising UWTPs, viewing to decreasing its spread [9]. Through the scientific publications, it is well noted that antibiotic chemicals existing in levels under clinical breakpoints (like in the example of wastewater) may select for resistant bacterial strains [10]; however, the lateral gene transfer and diffusion of antibiotic resistance genes (ARGs) [11], may be promoted in the UWTPs, due to the elevated microbial density and supplementary selection pressures [12]. With the aim to counter antimicrobial resistance diffusion in nature, it is consequently requested to define and/or improve techniques capable to efficiently eliminate both the antibiotics and the resistance determinants at the UWTP, prior reuse or elimination of the effluent [12].

Targeting at the microbial demobilization, disinfecting wastewater can constitute a chance to restrict the liberation of antibiotic-resistant bacteria (ARB) into nature and play a part in the reduction of the environmentally-related danger of diffusing resistance determinants. Adopting UV-driven techniques, which are frequently implemented in UWTPs for killing pathogens, might be helpful for the sake of such aim. Irradiating with either a light source (frequently realized with low- or medium-pressure mercury vapor lamps) or natural sunlight, remains a conceivable method of reducing micro-pollutants and dissolved effluent organic matter (dE_fOM) existing in urban wastewater (UWW) effluents. The UV radiation may disfigure DNA, conducting to the curb of cell replication and, in situation of fatal injections, to a deprivation of reproducibility. While accepting UV for disinfecting wastewater has expanded considerably during the previous two decades, investigating on the capability of the UV technology to reduce ARB & ARGs is at most progressing throughout the last few years [13] [14].

Until now, there is restricted information at hand on the capacity of light-driven techniques to altogether eliminate antibiotics, ARB & ARGs from wastewater [15] [16] [17]. More methodical exploration of the running factors of the light-driven techniques and their influence on the global performance of the methods to eliminate these micro-pollutants is needed. In addition, light-driven techniques merged with hydrogen peroxide (H_2O_2), producing supplementary hydroxyl radicals ($^{\bullet}OH$), generating from the division of H_2O_2, may moreover decrease the micro-pollutants existing in wastewater effluents, considerably improving the performance of the technology [18]. The supremacy of the UV/H_2O_2 upon the traditional UV disinfection for demobilizing ARB in wastewater is obviously proved in the scientific publications ($^{\bullet}OH$ may importantly ameliorate the oxidation potential of the chemical system, leading to alterations in the bacterial cell structure) [19]; however, in the situation of ARGs, extended period of UV/H_2O_2 treatment appears to be requested for their efficient reduction. Plus, the probability of employing natural sunlight instead of UV lamps, to catalyze the production of $^{\bullet}OH$ throughout the method, may lead to a low-cost usage [5].

Nevertheless, transformation products (TPs) of the antibiotics existing may be generated, which may be less bio-decomposable, more poisonous and biologically strong, juxtaposed to the parent compounds [20]. Therefore, attempts have to be concentrated on defining the structure of such products; however, as well proving if these retain their core moieties, responsible for the antimicrobial activity of the antibiotic, probably comprising antimicrobial resistance to the surrounding microbes.

2. Effect of UV-C/H_2O_2 and Sunlight/H_2O_2 on Eliminating Antibiotics, Antibiotic Resistance Determinants and Toxicity Present in Urban Wastewater (UWW)

In such background, the likely usage of UV-C/H_2O_2 and sunlight/H_2O_2 techniques as tertiary treatment of UWW deserves investigation. Consequently, Michael *et al.* [5] focused on the effect of UV-C/H_2O_2 and sunlight/H_2O_2 oxidation methods on: 1) the decomposition of two antibiotics (ciprofloxacin [CIP] and sulfamethoxazole [SMX]), when occurring as a mixture in UWW; 2) the demobilization of *Escherichia coli* and *Pseudomonas aeruginosa* involving colonies of such species still cultivable in the existence of sub-minimal inhibitory levels (sub-MIC) of CIP and SMX and 3) the removal of the 16S rRNA gene and ARGs encoding resistance to β-lactams (*bla*$_{TEM}$, *bla*$_{OXA-A}$, *bla*$_{SHV}$, *bla*$_{CTX-M}$, *mec*$_{A}$), sulphonamides (*sul*1, *sul*2), quinolones (*qnrS*), glycopeptides (*van*A) and tetracyclines (*tet*M) in UWW. Michael *et al.* [5] examined the two techniques at pilot-scale, employing real UWW effluents spiked with the antibiotics; at the same time, they dedicated supplementary trials to determining the main photo-transformation products of CIP and SMX. To assess the biological potency of the treated flow, they implemented a chronic toxicity test. Choosing CIP and SMX as the aimed antibiotics to be tested, was founded on their high consumption, their common presence in UWTPs effluents [21] and the dominance of bacteria harboring resistance to these products in the wastewater effluents [22] [23]. Fluoroquinolones, involving CIP, are seen by WHO as greatly significant antibiotics for human medicine [24]; simultaneously, CIP is comprised in the Watch List of substances for EU-wide monitoring [25], because of its consistency with the European One Health Action Plan against antimicrobial resistance [26]. SMX is a sulphonamide antibiotic largely utilized as prophylactic and therapeutic medication for treating human and animal diseases and benefiting agricultural productivity. The occurrence of such products in the wastewater has been illustrated to be considerably related to augmented fluoroquinolone and sulphonamide resistance genes and resistant bacteria in UWTPs effluents [27] [28].

Michael *et al.* [5] considered their work as the first research showing complete information concerning not only the decomposition of antibiotics throughout UV-C/H_2O_2 and sunlight/H_2O_2 techniques and the estimation of the treatments in reducing resistance determinants (bacteria, completely viable and cultivable in the occurrence of sub-MIC of the target antibiotics and ARGs), but as well the clarification of the principal TPs of CIP and SMX (to explore if the methods may oxidize the antibacterial moieties of the antibiotics, the quinolone ring and the amino group of CIP and SMX, respectively) and the evaluation of the treated effluents in matter of poisoning. These researchers assessed UV-C/H_2O_2 and sunlight/H_2O_2 techniques in a combined way and estimated if their implementation lets secure disposal/reuse of treated UWW to nature.

Michael *et al.* [5] established that the UV-C/H_2O_2 technique was able to remove CIP and SMX (90 min, 0.9 kJ/L), while sunlight/H_2O_2 method was only apt to reduce CIP (CIP was removed in 60 min and 8 kJ/L, whilst SMX was decreased only by 46% after 300 min and 42 kJ/L). Identical findings were reached for the two techniques, if the matrix was SS, except from the shorter periods needed for antibiotics' removal (because of absence of dE_fOM in the SS). This shows the supremacy of UV-C/H_2O_2 over sunlight/H_2O_2 method for eliminating antibiotics, regardless of the matrix utilized. The generation of recalcitrant organic intermediates was obvious from the reality that total mineralization was not obtained by any technique. The findings of the chronic toxicity bioassay implemented, employing the *V. fischeri* bacterium, illustrated that the poisoning is possibly extracted from the oxidation of the dE_fOM itself. Therefore, in matter of poisoning, which appeared to be bigger during UV-C/H_2O_2, the method is affected with a disadvantage.

The mechanisms of the photo-transformations of the two antibiotics determined, illustrated that all the TPs identified for CIP and SMX still retain the core quinolone and amino moieties, respectively, which are in charge of the antibacterial activity of the compounds. This is an important remark, as more researches have to be performed to define the suitable injection or accumulated energy, which will be apt to oxidize the antimicrobial moiety of the TPs, assessing simultaneously the effect of the TPs on antimicrobial resistance diffusion [5].

Both treatments were observed apt to demobilize *E. coli* and *P. aeruginosa* in saline solution (SS) and UWW, comprising the colonies of such species cultivable in the occurrence of sub-MIC of CIP and SMX, noting though, a quite big difference in the dose/accumulated energy requested by each method (UV-C/H_2O_2: 8 min, 0.1 kJ/L; sunlight/H_2O_2: 120 - 150 min, 16 - 20 kJ/L). Further, following 48 h of post-treatment storage of the sunlight/H_2O_2 treated samples, bacterial regrowth happened, proposing that the treatment was not only longer, but as well it did not furnish total and lasting disinfection. ARGs presented various behavior throughout the two treatments, since specific genes were reduced to levels under the quantification limits and others were persistent throughout the treatment. Plus, the UV-C/H_2O_2 depicted its supremacy over the sunlight/H_2O_2 technique, since throughout the implementation of the former, all the *bla* and *qnrS* genes were removed, whilst in the utilization of the latter, none of the tested genes were eliminated. Nevertheless, the acquired results proved the failure of both methods to avoid the diffusion of ARGs to nature. Demobilizing the investigated microbes and eliminating ARGs were faster than the decomposition of the target antibiotics. Since more understanding is being collected about the inherent unfavorable influences of the ARB & ARGs following their liberation to nature, awareness has to be addressed so that the techniques used at the UWTPs attain both the elimination of antibiotics and their TPs, and the removal of the antimicrobial resistant bacterial and gene loads, whilst inhibiting post-treatment bacterial regrowth.

3. Best Available Technologies (BATs) and Treatment Trains (TTs) for Urban Wastewater (UWW) Reuse

Recently Rizzo *et al.* [29] furnished a high-tech discussion of the best available technologies (BATs) and proposed likely advanced treatment choices to render wastewater reuse safer, especially concerning the elimination of CECs and ARB & ARGs. Numerous elements touch the selection of the most appropriate treatment procedure (like water quality, local regulation/restrictions, process costs, type of crop, irrigation method, soil type, environmental footprint, social acceptance, etc.). However, Rizzo *et al.* [29] performed an effort to assess the probable BATs for the advanced treatment of UWW involving their benefits and hurdles.

Rizzo *et al.* [29] concluded that a single advanced treatment technique is not enough to reduce the liberation of CECs and ARB & ARGs and render wastewater reuse for crop irrigation safer, but a judicious integration of them (**Figure 1**) and an appropriate monitoring program (**Table 1**) would be indispensable. This conclusion appears from the realization that every treatment process possess its proper weaknesses/drawbacks, for instance:

- A biological post-treatment [52] [53] [54] to eliminate oxidation by-products may be needed when ozonation or AOP is employed as advanced treatment [37] [55] [56];
- Ozonation and AOPs need toxicity monitoring due to probable generation of problematic oxidation reaction products;

- Adsorption techniques must be pursued by an efficient disinfection method (*i.e.*, UV disinfection);
- If PAC is utilized, a posterior filtration or membrane process has to be added to eliminate the adsorbent particles;
- Chemical disinfection is not efficacious in dealing with CECs and ARGs; therefore, it has to be combined with more advanced treatment techniques. Over and above, probable generation of DBPs (*i.e.*, chlorination by-products [57] [58] [59] [60] [61]) must be taken into account, and the next treatment for their elimination is requisite;
- NF or RO membrane technology needs a pre-treatment (*i.e.*, sand filtration) to avoid blocking and a potential solution for the recycling of membrane concentrate.

More comparative investigations between various advanced treatment techniques on real wastewater, following diverse criteria (*i.e.*, CECs removal, ARB & ARGs, toxicity, DBPs [62] [63] [64] [65] [66], costs) are suggested [29].

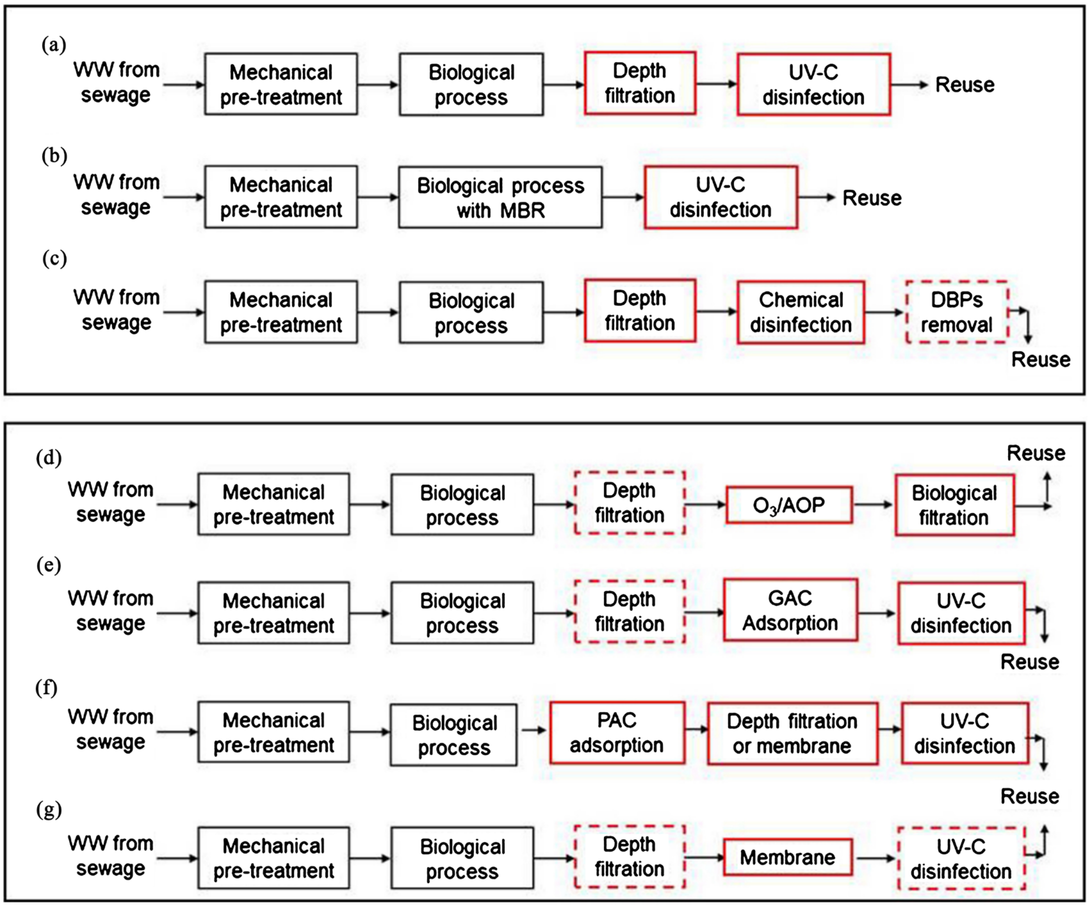

Figure 1. Various choices of treatment trains (TTs) for UWW reuse to deal with conventional factors fixed in wastewater reuse regulation and guidelines (such as biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSSs), *E. coli*, etc.) (a)-(c); and to efficiently eliminate CECs in addition to the usual elements (d)-(g). Advanced treatment in red lines; red dotted lines significate that method usage has to be estimated case by case. "Biological process" followed by "depth filtration" may be replaced by "membrane bioreactor (MBR)" for TTs "d" and "e" [29].

Table 1. Benefits, obstacles, and recommendations for each TT in Figure 1 [29].

TT (advanced treatment)	Benefits	Obstacles	Recommendations
a or b (UV)	• Efficient disinfection (comprising ARB [30] demobilization) • No disinfection by-products (DBPs) [31] [32] generation contrasted to chemical disinfection	• Poor/no CECs elimination • Partial elimination of ARGs	• Compliance with local residual bacterial density standards should be evaluated
c (chemical disinfection)	• Efficient disinfection (comprising ARB demobilization)	• Poor/no reduction of CECs and ARGs • Generation of DBPs [33] [34] [35]	• Toxicity trials recommended [36] [37] • DBPs (following the disinfectants utilized) must be controlled [38] [39] [40] [41]
d (O_3/AOP and biological post-treatment)	• Efficient disinfection (comprising ARB demobilization) • CECs reduction: Elevated throughout ozonation and (solar) photo Fenton [42], moderate with UV/H_2O_2 • Full-scale evidence on practicability only for O_3	• Generation of numerous DBPs (*N* nitrosodimethylamine (NDMA), bromate) • throughout ozonation • Production of oxidation transformation products throughout AOP and ozonation [43] [44] [45] • Partial ARGs reduction	• Toxicity trials recommended • NDMA and bromate must be controlled in O_3 treatment
e (GAC and UV)	• Efficient disinfection via UV • Elevated CECs reduction via GAC • Full-scale evidence on practicability	• Poor/no reduction of ARB & ARGs via GAC alone • For UV see above, TT a & b	• Reducing adsorption capacity with elevating bed volume must be considered
f (PAC and UV)	• Efficient disinfection via UV • Elevated CECs elimination via PAC • Full-scale evidence on practicability for CEC removal by PAC	• Poor/no reduction of ARB & ARGs via PAC alone • For UV see above, TT a & b	
g (nanofiltration (NF) or reverse osmosis (RO) membrane filtration, with potential pre-treatment with microfiltration (MF) or ultrafiltration (UF) membranes)	• Efficient disinfection for bacteria (comprising ARB) and protozoa for all membranes; viruses well removed by UF, NF & RO [46] • ARGs well removed by NF and RO [47] • CECs removal from poor (MF, UF) to very good (NF, RO) following membrane Type [48] • RO and partially also NF reduce salinity [49] [50] • For post UV-C see TT a & b	• Poor/no reduction of ARGs at full-scale by MF (for UF some reduction is expected) • Poor CECs elimination for MF and UF • Elevated energy needs for NF and RO • Formation of a substantial concentrate waste stream by NF and RO • For post UV-C see TT a & b	• Effect of membrane features on disinfection, ARB, ARG, and CEC reduction has to be carefully taken into account 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 [51] (e.g. following prior chloramination)

As seen through this work, there is no miraculous BAT for treating wastewater for water reuse in agriculture [67] [68] [69] [70]. An appropriate combination of many techniques would be suggested following each case [71] [72] [73] [74].

4. Conclusions

From this work, the following conclusions can be drawn:

1) Traditional urban wastewater treatment plants (UWTPs) are deficiently efficient in eliminating most contaminants of emerging concern (CECs), comprising antibiotics, antibiotic resistant bacteria and antibiotic resistance genes (ARB &

ARGs). Such pollutants lead to some worry for nature and human health. This work discusses the performance of the best available technologies (BATs) for dealing with urban wastewater (UWW) to eliminate CECs and ARB & ARGs. Ozonation, activated carbon adsorption, chemical disinfectants, UV radiation, advanced oxidation processes (AOPs) and membrane filtration are debated with a view to their potential to efficaciously reduce CECs and ARB & ARGs. Probable treatment trains involving the BATs are compared. In spite of the huge improvements acquired in terms of applying AOPs and understanding their mechanisms in removing ARB & ARGs, transformation products (TPs) of the antibiotics existing may be generated, which may be less bio-decomposable, more poisonous and biologically strong, juxtaposed to the parent compounds. Therefore, attempts have to be concentrated on defining the structure of such TPs and proving if these retain their core moieties, responsible for the antimicrobial activity of the antibiotic, probably comprising antimicrobial resistance to the surrounding microbes.

2) Since more understanding is being collected about the inherent unfavorable influences of the ARB & ARGs following their liberation to nature, awareness has to be addressed so that the techniques used at the UWTPs attain both the elimination of antibiotics and their TPs, and the removal of the antimicrobial resistant bacterial and gene loads, whilst inhibiting post-treatment bacterial regrowth.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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