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Review

Ozone and Photocatalytic Processes for Pathogens Removal from Water: A Review

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Abstract: The search for alternative water sources is pushing to the reuse of treated water coming from municipal wastewater treatment plants. However, this requires that tightened standards be fulfilled. Among them is the microbiological safety of reused water. Although chlorination is the mostly applied disinfection system, it presents several disadvantages, such as the high doses required and the possibility of formation of dangerous by-products. Moreover, the threat of antibiotic resistance genes (ARGs) spread throughout poorly treated water is requiring the implementation of more efficient disinfection systems. Ozone and photo assisted disinfection technologies are being given special attention to reach treated water with higher quality. Still, much must be done to optimize the processes so that cost-effective systems may be obtained. This review paper gives a critical overview on the application of ozone and photo-based disinfection systems, bearing in mind their advantages and disadvantages when applied to water and municipal wastewater. Also, the possibility of integrated disinfection systems is considered.

Keywords: disinfection; pathogens; ozone; photo-aided systems; photocatalytic processes; water reuse; wastewater treatment

1. Introduction

Water scarcity is one of the main challenges mankind is facing nowadays. Water quality protection aiming to reduce impacts over ecosystems and human health will lead to even tighter legislation regarding treated municipal wastewater disposal into the natural water courses. Moreover, special care must be taken if wastewater reuse is to be considered. In fact, water problematic is leading to a new paradigm where liquid effluents are starting to be envisaged as water sources. Besides, their discharge into aquifers makes them a secondary source of potable water [1]. A water reuse system is only viable if wastewater disinfection is effective due to the public health issues that are related with microorganisms.

Human contamination of water sources, mainly due to faecal contamination, still represents a major public health concern, especially in highly populated areas. Domestic wastewater will always contain microorganisms, either from human or animal origin, some of them pathogenic to humans. Therefore, the treatment of such wastewaters should assure that such pathogens are eliminated,

or reduced until levels with no or low level of impact in public health, before its discharge into a receiving water or, and especially, if reuse purposes are foreseen.

Bacteria, namely those present in human gastrointestinal tract, represent the microorganism group more frequently studied and evaluated to determine the microbiological quality of different types of waters [2]. Nevertheless, special attention should also be done to other pathogens, such as viruses and parasitic protozoa, which could also infect human host either through the ingestion of contaminated water or food, or direct contact with contaminated recreational water [3]. Human pathogenic viruses, such as noroviruses (NoVs), adenoviruses (AdVs), enteroviruses (EVs), and human polyomaviruses (PyVs) are excreted in high concentrations in the faeces and urine of infected individuals (up to 10^{11} viruses/g of faeces, and up to 10^{7} viruses/mL of urine), with or without symptoms [4,5]. High concentrations of these viruses will, therefore, be present in wastewaters, which if not properly eliminated will end up infecting susceptible hosts. In fact, viruses that are present in wastewaters represent a major microorganism group risk to public health. Despite its high concentrations in wastewaters, they are also more resistant to conventional treatment systems and have a greater infectivity than the majority of bacteria and protozoa [6]. Therefore, an effective disinfection system should be able to assure its removal or reduction.

Another special concern is the presence of large amounts of antibiotic resistant bacteria in wastewater treatment plants [7–12]. Indeed, the overuse of antibiotic is leading to the increased and threatening spread of antibiotic resistant bacteria. A municipal wastewater treatment plant (MWWTP) represents a favorable place to the occurrence of such spread, since the high density of bacteria in such environment promotes horizontal gene transfer, including the transfer of antibiotic resistant genes (ARG) from resistant bacteria to other bacteria, initially susceptible [8,9,11]. The spreading of antibiotic resistance is a serious threat for public health, since it hinders antibiotics development and diseases treatment [13]. Thus, also for this point it is required that suitable disinfection systems are developed to avoid their release into the environment [14].

Chlorination is the most common disinfection process. However, the production of dangerous by-products, such as halo-organics, as well as the large chlorine doses that are usually required for an effective disinfection constitute major drawbacks for such technology [15]. On the other hand, some pathogens, such as *Cryptosporidium parvum*, are relatively immune to free chlorine at the dosages typically used for water disinfection [16].

Alternatively, ozone is able to inactivate a wide range of pathogens, such as bacteria, including its spores, viruses, protozoa, and prion protein [17]. The use of ozone as a disinfection step also improves the efficiency of downstream technologies. For example, it reduces fouling in the membrane processes [18]. The main drawback is associated with the possibility of producing dangerous by-products during the partial oxidation of dissolved organic compounds. Generally, the ozone dose that is required for an effective disinfection is higher than the one leading to organic compounds degradation. Thus, during disinfection, also chemical micropollutants, are removed [17]. Still, the operating conditions must be carefully selected since ozonation processes efficiency varies significantly due to the characteristics of the reclaimed water [19]. Thus, for example, the ozone transferred dose must be optimized so that a suitable disinfection is achieved and no toxic by-products are formed during the treatment. Moreover, research is focused on optimizing ozonation action over pollutants. This involves the addition of catalysts as well as radiation sources.

According to the Environmental Protection Agency (EPA) [20], ozone is more efficient than chlorine regarding disinfection, involving shorter reaction times without regrowth of microorganisms. The remaining ozone is quickly decomposed into oxygen and thus does not constitute a problem. However, it is a complex technology comprising gas-liquid contact systems, ozone generation, and corrosion-resistant materials, which increases the capital and power costs. Besides, it is not economically viable when the water comprises high amounts of solids and organic matter due to the high ozone demand. Regarding by-products formation, one should bear in mind the possibility of toxic bromate formation when the water is rich in bromide. Moreover, several reports proved that ozone is

effective for viruses removal [3,21,22], either by promoting protein coagulation or oxidation of nuclei acids [23]. Besides, its capacity of polishing the wastewater by removing some of the chemical oxygen demand makes it an interesting water disinfection process [24]. Of course, to be viable, the amount of organic matter in the water to be disinfected must not be high.

Photo-assisted technologies may also be envisaged as interesting alternatives for water disinfection. It is well known that UV disinfection is a common process that is used for pathogens inactivation [22,25–30]. However, its application usually requires secondary residual disinfectants, such as chlorine [28,31,32], so that no regrowth is possible in the water supply network. Nevertheless, the formation of disinfection by-products (DBP), which could be mutagenic and carcinogenic, during chlorination demand the improvement of already known treatment or finding more promising processes [27,32]. Furthermore, one of the most important concerns of disinfection is the resistance of some organisms to disinfectant as well as the requirement of much higher doses for deactivation. In spite of all disadvantages, including high costs of UV equipment, UV disinfection is still wildly applied around the world. Although it has to be mentioned that solar water disinfection (SODIS) as well as photocatalytic disinfection are developing very well lately [33–37]. However, potential to use sunlight [37–40] or low cost LED lamps [29,41–43] to initiate the disinfection process in the presence of catalyst seems to give the biggest advantage over other methods. Moreover, photocatalytic disinfection leads to inactivation of viruses, bacteria, and its spores and protozoa maintaining residual disinfectant effect [22,36,42]. Thus, there is still plenty of room for disinfection processes development and optimization.

Bearing in mind the growing interest in innovative disinfection technologies and the especial focus on ozone and photo-based systems, this paper presents a critical review concerning ozone and photocatalytic processes for pathogens disinfection. Special emphasis was placed on recent research on ozone-based technologies as well as light driven technologies, including UVC, solar, and photocatalytic disinfection. In fact, these processes show interesting features, since undesired disinfection by-products formation can be avoided if the operating conditions are carefully selected. To the best of our knowledge, there is no recent review comparing these two disinfection approaches. Moreover, the focus is not only bacteria removal but also protozoa and viruses. In fact, these pathogens may present even higher infectivity than bacteria, therefore, their presence in treated water may not be neglected.

2. Ozone Based Disinfection Systems

2.1. Single Ozonation

Ozone is a highly reactive molecule with an oxidation potential of 2.07 V [44]. Moreover, it has the capacity of, under some conditions, lead to secondary oxidants (such as hydroxyl radicals) with even higher reactivity [45]. This shows its potential as a disinfectant agent, since, due to these characteristics, it has the capacity of reacting with several cellular constituents, such as the cell wall and DNA structures.

Ozonation was able to remove up 2 log units of cultivable fungi, 16S rRNA and *intl1* genes, about 4 log units of heterotrophs, enterobacteria, and enterococci (Table 1). Moreover, the antibiotic resistance genes (ARG) can be depleted below the detection limit. The presence of dissolved organic matter in real municipal wastewater reduces the disinfection ability of the process when compared with its performance when applied to a synthetic effluent. In fact, organic compounds may quench some of the oxidant species that will not be available for the microbial cells [46]. Even if the synthetic water presented higher COD than the real effluent, disinfection was less efficient when applied to the actual stream. Thus, the lower efficiency of the disinfection process was attributed to other species that are presented in the actual secondary wastewater, such as suspended solids, which may consume oxidants. Besides, the higher complexity of the biota in real streams may be another explanation. Although the high efficiency of the process, regrowth tests showed that, in general, there was the prevalence of

antibiotic resistance and integrase genes amongst the survival microorganisms. Thus, some care must be taken on the selection and optimization of tertiary treatments to avoid ARG in reused water [8,10,46]. Table 1 compares some works dealing with water disinfection through ozone. In all of the reported results, real wastewater was tested with different characteristics. The required ozone dose for an efficient disinfection is highly dependent upon the water characteristics. For example, Xu et al. [15] verified that the optimal ozone dose for disinfection could range from 2–15 mgO_3/L, depending upon the effluent organic matter load.

Table 1. Transferred ozone dose (TOD) required for effective disinfection for different water qualities and sources.

Type of Water	Water Characteristics	Pathogens Removal	Ozone Dose	Reference
Secondary municipal effluent	COD = 42–49 mg/L TSS = 4.5–6 mg/L *E. coli* = 1.8×10^3 CFU/mL	*E. coli* total removal	0.3 mg/L (transferred dose)	[10]
Synthetic and Actual secondary urban effluent	Synthetic effluent: COD = 300 mg/L Actual effluent: COD = 25–50 mg/L TSS = 5.2 mg/L	Synthetic effluent: ~3.3 log fungi ~6.5 log bacteria Actual effluent: ~2 log fungi ~3–4 log bacteria	225 mg/L (injected dose)	[46]
2 secondary and 1 tertiary municipal effluent	COD = 30–71 mg/L TSS = 2.3–18 mg/L *E. coli* = 2.7–4.3 log CFU/100 mL *Clostridum* = 3.0–5.5 log CFU/100 mL	Viruses always removed *Clostridium* the most difficult pathogen	2–15 mg/L (transferred dose, depending water quality)	[15]

COD—Chemical oxygen demand; TSS—Total suspended solids.

Thus, the transferred ozone dose (TOD) needed for an efficient disinfection is highly dependent upon the effluent quality. Xu et al. [15] verified that, for achieving the standards for water reuse for irrigation, the required TOD could range from 2 to 15 mg/L. At those conditions, besides reaching a biologically suitable water, also some $UV_{254\,nm}$ and colour reduction of the water was obtained. This means that some organic contaminants that were present in the wastewater were also partially depleted. This may be a disadvantage, since the presence of organic compounds, such as humic acids (which are usually found in natural water), reduces disinfection efficiency since a lower amount of oxidant will be available for microorganisms [47]. However, this shows the capacity of ozone on polishing reused water, bearing in mind both its chemical and biological characteristics.

Zhuang et al. [48] compared several technologies for ARG removal: chlorination, ozonation, and UV. From their results, it was concluded that chlorination is more effective. This was attributed to the fact that chlorine penetrates the cell and inactivates ARGs directly, while ozonation reacts first with the cell wall. In fact, even if antibiotic resistant bacteria (ARB) can be totally removed by ozonation, some viable ARG may still persist if low dosages of oxidant are applied. It should be still stressed that 40 mg/L of chlorine were required during chlorination. Oh et al. [13] refer that ozonation is a better methodology when compared with chlorination, since more than 30 mg/L of chlorine were required for the removal of 90% of ARB and ARG, which is impracticable. Whereas, ozone dose needed was only 3 mg/L. Nevertheless, the prevalence of ARG after the processes was not discussed. Another important point is related with organic by-products that may be generated during the disinfection. In what regards chlorination, organo-chlorate compounds are of special concern. Thus, this assessment is also required to discuss the applicability of a disinfection process. Implementation and operating costs must be also considered so that the recovered water price is not prohibitive.

Cryptosporidium parvum is an intestinal parasite that may lead to severe problems, especially to people with immune system deficiencies. There are several reports regarding waterborne outbreaks of this protozoan. This species is resistant to chlorine at drinking water treatment levels. Ozone is much more effective on the inactivation of this pathogen, with a kinetics following a first lag-phase followed by a first order kinetics [49]. Hunt and Mariñas [47] verified that ozone reaction rate with *Escherichia coli* is several thousand orders of magnitude higher than with *C. parvum*. Thus, drinking water disinfection systems must be drawn for *C. parvum* inactivation rather than *E. coli*.

Giardia lamblia and *Giardia muris* are other types of intestinal protozoan that are sensitive to ozone [50]. However, due to the high diversity of results reported by the authors, attributed to different ozonation protocols, the use of only a chemical disinfection for the removal of *Giardia* from drinking water was not recommended. In fact, the results that are reported are for the inactivation in organic free water. The presence of other contaminants also demanding ozone may lead to lower disinfection efficiencies.

Different viruses have been found in raw and treated sewage. Furthermore, several outbreaks of virus infections have been associated with the consumption of treated water, despite its compliance with the current legislation regarding microbiological quality of water. This indicates that actual organisms used for monitoring the microbiological quality of water, mainly enteric bacteria, are not enough to assure such quality regarding virological criteria [51]. Therefore, the development of new or complementary approaches for water treatment should also ensure the elimination of human pathogenic viruses.

Several studies have evaluated the application of ozone for wastewater treatment and its efficiency in removing human viruses. The majority of such studies points to ozone as an effective treatment approach against waterborne viruses, with a reduction to very low or even undetectable levels of adenoviruses, polyomaviruses, cocksakievirus, noroviruses, astroviruses, and parvoviruses being reported [3,15,21,22].

It is assumed that ozone treatment will conduce to the oxidation of viral capsid proteins, which will result either in the destruction of such viral structure or in its inability to bind to the cellular receptor, preventing, therefore, viral infection of susceptible cells [3,52].

CT, i.e., the disinfectant residual concentration in water times the contact time can be used to check pathogens susceptibility to inactivation by a certain reactant. Table 2 summarizes some CT values for 2 log removal of bacteria (*E. coli*), protozoa and virus by ozone disinfection.

Table 2. CT values required by ozonation for 2 log inactivation of selected pathogens from water.

Pathogen	CT (mgO_3 min/L)	Reference
E. coli (bacteria)	6.0×10^{-3}	[53]
C. parvum (protozoa)	3.08 (25 °C)	[49]
G. lamblia (protozoa)	0.65	[50]
G. muris (protozoa)	0.24	[50]
Echovirus (virus)	1.9×10^{-3}	[21]
Adenovirus (virus)	4.1×10^{-3}	[21]
Coxsackievirus (virus)	8.0×10^{-3}	[21]

All of the results from the literature were obtained for synthetic wastewaters. Bacteria and virus are much more susceptible to ozone oxidation than protozoa, since lower ozone doses are required to achieve bacteria and virus removal. Thus, ozone is an interesting disinfectant for both bacteria and virus inactivation. This means that disinfection systems that are based on ozone must be designed bearing in mind protozoa removal since these will be the limiting pathogen in the process.

One of the disadvantages of ozone application is the production of bromate during disinfection. This is an important concern when high ozone exposures are required for a proper inactivation of the pathogens, such as protozoa, which are more resistant than bacteria and virus [54]. However, some control options are available to reduce bromate production, such as decreasing pH or the

addition of ammonia [55]. Driedger et al. [54] verified that ozone inactivation of *Bacillus subtilis* spores (as an indicator for *C. parvum* oocysts) was insensitive to pH and to the presence of ammonia. This way, operating at pH 6, it was possible to reduce bromate formation by 50% while maintaining disinfection efficiency.

The control on ozone dosage may avoid the formation of by-products during disinfection. Thus, monitoring procedures must be implemented to follow up disinfection and avoid ozone overdosing. CT, i.e., the disinfectant residual concentration in water times the contact time is largely applied for those purposes [56,57]. However, according to Zhang et al. [58], that value should be obtained taking into account the reactor hydrodynamics by solving transport equations to obtain CT (CFD-CT) so that ozone overdosing does not occur. Zhang et al. [59] also verified that a CFD model comprising kinetic-based models was able to predict ozone and bromate concentrations within a disinfection reactor. This way, such models can be an important tool to predict and optimize these complex disinfection systems.

In fact, CFD models may encompass deviations to the ideal behavior by incorporating short-circuits in the contactor and inadequate mixing, which will lead to different retention times. This way, using those models, data coming from bench and pilot scale units can be extrapolated to full scale units. EPA (1986) [60], proposes that dose-response relationships obtained at pilot-scale should be used for the process design. Those dose-response functions are usually function of ozone dose (Equation (1)):

$$\log \frac{N}{N_0} = \beta_0 + \beta_1 \log C_{ozone} \quad (1)$$

where N and N_0 are the surviving and initial bacteria concentrations, respectively, C_{ozone} is the ozone dose, and β_0 and β_1 are model parameters that are a function of the wastewater organic matter load [61].

Generally, ozone systems are considered as expensive mainly due to the low ozone usage efficiency. Since ozone reactions with bacteria are characterized by a fast kinetics, ozone mass transfer is usually the limiting step of the process [15,47]. In fact, although usually homogeneous second order kinetic laws are attributed for the inactivation process, ozone must diffuse to the membrane and then penetrate it. For example, regarding the inactivation of *E. coli*, some authors attribute it to ozone degradation of the cell surface with a consequent leakage of the cell constituents [62]. However, other refer that ozone is able to diffuse the membrane and damaging the cytoplasmatic constituents, such as DNA [63].

The application of micro-bubble injection may improve ozone mass transfer, since higher $K_l a$ values are achieved. Moreover, the energy that is generated due to hydrodynamic cavitation may lead to the production of free radicals with a higher oxidant power than ozone. Zhang et al. [64] tested a micro-bubble ozonation system for the inactivation of *Bacilius subtilis* and concluded that increasing ozone inlet concentration reduces the bubble diameter, leading to higher ozone usages efficiencies. Thus, higher *B. subtilis* inactivation is achieved. However, one must bear in mind that increasing the ozone generator capacity for reaching higher ozone inlet concentrations will very much increase the investment costs, since the ozone system will be much more expensive. For example, while for an ozone production capacity of 5 kg/h, the investment cost may be 230,000 €, for a capacity of 50 kg/h the investment may reach 1,450,000 € [19]. In this context, deep economic analysis embracing investment and operating costs must be performed before proposing a disinfection alternative. However, it is likely that future water requirements will imply the application of more efficient and more expensive treatment processes to safeguard ecosystems and human life. Moreover, also the reactor configuration may be optimized to improve gas/liquid mass transfer. For example, Zhang et al. [58] verified that energy consumption (while improving disinfection performance) could be reduced in a reactor with baffle configuration increasing the number of vertical baffles. Thus, proper design and control may lead to suitable ozone based disinfection systems.

2.2. Hydrogen Peroxide Aided Ozonation

Ozone action may be enhanced by coupling it with hydrogen peroxide or persulfate [13]. While hydrogen peroxide promotes the production of hydroxyl radicals, persulfate leads to the strong oxidant sulfate ion radical. Among the tested additives, (hydrogen peroxide, persulfate, and monopersulfate) monopersulfate leads to the highest efficiency on enhancing ozonation for the removal of ARB and ARG. In fact, CT (concentration × time) for 2 log decay of ARB and ARG decreased from 31 and 33 (mg min)/L, respectively, during ozonation to 11.97 and 14.97 (mg min)/L when monopersulfate was used as additive. This enhancement was attributed to the higher oxidation power of sulfate radical and ability to penetrate into the bacteria cells [13].

CT is the most commonly used parameter to control ozone systems, namely for disinfection [65]. However, sometimes it is not possible to measure dissolved ozone, especially when ozone quickly decomposes, as is the case where its action is aided by H_2O_2 [66]. Gamage [67] compared several parameters to describe *E. coli*, MS2 bacteriophage and *Bacillus subtilis* spores inactivation through ozone and ozone/H_2O_2. Those parameters included CT, ozone to total organic carbon (O_3:TOC) ratio, differential UV_{254} absorbance (ΔUV_{254}), and differential total fluorescence (ΔTF). O_3:TOC, ΔUV_{254}, and ΔTF were able to predict disinfection as alternatives to CT.

3. Light Application for Disinfection

Disinfection by light is one of the most widely used method around the world [25,26]. The conventional method is based on UV light. However, solar light is also efficient for the deactivation of biohazards [68,69] and research regarding its application is increasing using several combinations (Figure 1).

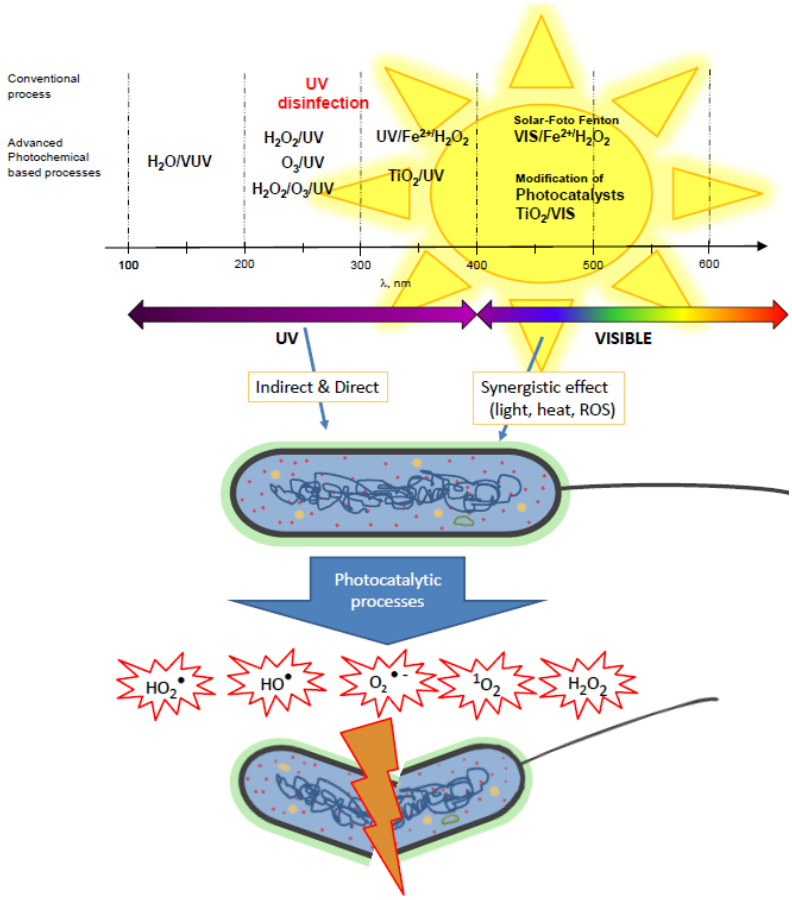

Figure 1. The schematic range of electromagnetic radiation used in photochemical disinfection processes and its effects on patogens (based on [35,51,69–75]).

3.1. UV Disinfection

The UV disinfection that was developed after the discovery that UV wavelengths between 200 and 300 nm (especially 260 nm) exhibits the best germicidal effectiveness due to the relative absorbance of DNA molecules [76,77]. UV light can be used as a broad disinfectant spectrum over a wide temperature range. Furthermore, it can be used to inactivate protozoa, bacteria, bacteriophage, yeast, viruses, fungi, and algae [78–82].

UV light disinfection efficiency depends on the type of microorganism considered. It is known that viruses and bacteria spores are the most resistant to inactivation by UV radiation, followed by intestinal protozoa, such as *Cryptosporidium* and *Giardia*, and finally, by bacteria [78,83].

UV light is the least effective at inactivating viruses and spores, followed by bacteria and it is the most effective against protozoa such as *Cryptosporidium* and *Giardia*. However, some types of microbes (e.g., *Deinococcus radiodurans*) are resistant for low-dose irradiation, and therefore for an effective inactivation, high-dose irradiation is required [84,85]. Although, UV disinfection inactivates microorganisms through the damage of their DNA, some microbes are able to repair it via photoreactivation, which allows inactivated microorganisms to re-contaminate water [46,86,87]. Also, some viruses, such as human adenoviruses and polyomaviruses, have a dsDNA genome type, which enable its repair by host cell machinery during viral replication, and, by this manner, they are able to overcome DNA damage that is induced by UV radiation [51,72].

It was found that UV dose (fluence) of 1.2 mJ/cm^2 was enough to inactivate *E. coli* O157:H7 to 6-log reduction, but 30 mJ/cm^2 UV irradiation was required when photoreactivation was considered [88]. The minimum UV dose for disinfection of drinking water is 40 mJ/cm^2 according to European recommendations [89–91]. That UV dose is able to reduce vegetative bacteria by 4–8 logs, while 3–6 logs reduction are achieved regarding viruses inactivation [92]. In the United States of America (USA), a UV dose of 186 mJ/cm^2 is required so that up to 4-log virus inactivation occurs [93].

However, even if the disinfection effectiveness of UV light is very high, in the United States, free chlorine or chloramines must be added as secondary residual disinfectants that are capable of maintaining a pathogen free distribution system [28,31,32]. According to NSF/ANSI standard 55 (for point-of-use (POU) and point-of-entry (POE) $UV_{254\,nm}$ systems), class "a" required at least 40 mJ/cm^2 but only 16 mJ/cm^2 is needed for supplemental disinfection (class "b") [94]. It gives guarantees that both class "a" and "b" POU as well as POE systems are effective in providing a 4-log inactivation of the gram-negative microbes [30]. However, only the class "a" systems are effective against *Bacillus anthracis* spores, providing 2-log inactivation [30].

Low pressure (LP) and medium pressure (MP) lamps are commonly used for UV disinfection [26,78,88,95]. LP lamps emit UV almost mainly at the germicidal wavelength of 253.7 nm, whereas MP lamps emit UV at a wider range of wavelengths, e.g., 200–400 nm. Nowadays, the ultraviolet light-emitting diodes (UV LEDs) are also successfully applied for disinfection purposes [29,41,95]. In a comparison study between LP vs UV LED lamps, it was found that microbial inactivation was not significantly different for *E. coli B* and *MS-2* (4-log$^{LP\ UV}$ required 6.5 mJ/cm^2 and 59.3 mJ/cm^2 while the 4-log$^{UV\ LEDs}$ required 6.2 mJ/cm^2; 58 mJ/cm^2, respectively), but for *B. atrophaeus spores* the dose was much lower for UV LED (18.7 mJ/cm^2) than for LP UV (30.0 mJ/cm^2), according to [43].

UV disinfection has several advantages: very effective for *Cryptosporidium* and *Giardia*; additional chemical are generally not required; contact time is of few seconds; the disinfection by-products (DBP) formation potential, turbidity, pH, TOC does not change during the process; and, it is a system that is easy to operate [26,28]. However, there are many factors that can affect the effectiveness of disinfection. The most important are iron and manganese content (<0.3 ppm and <0.05 ppm), total dissolved solids (TDS < 500 ppm), turbidity (<1 NTU), suspended solids (<10 mg/L), and hardness (<7 gpg) [96,97]. Moreover, sometimes the presence of residual disinfectants is required, so the addition of secondary disinfectants is needed.

Bearing in mind that the Stage 2 Disinfectants/Disinfection Byproducts Rule reduces DBP to total trihalomethanes $\leq$ 80 μg/L and five haloacetic acids $\leq$ 60 μg/L, UV light disinfection could be a very interesting alternative to chlorine dioxide or ozone [98], since the production of such by-products is avoided.

3.2. Solar Disinfection

In 1943, Hollaender published a study about the effect of solar light (long UVA and short VIS (350–490 nm)) on *E. coli* removal, indicating that irradiation of 265 nm directly damages nucleic acids, while short VIS causes the production of toxic compounds that destroy other cell components [99]. The damage over bacteria cells under sunlight irradiation is a consequence of the action of several intracellular light-driven Fenton reactions that can result in DNA and protein damage, as well as disruption of the electron transport chain, leading to an increase in membrane permeability and loss of functionality [37].

Based on this finding, the idea of solar water disinfection, or SODIS, one important point-of-use technology for drinking water treatment, has been developed [100,101]. In many regions of the world, the UVA part of sunlight is employed for disinfection purposes [33,69,100,101]. The germicidal effect of sunlight is attributed to the synergistic effect of solar UV light and mild-thermal heating that is produced during exposure [69,102,103]. However, the SODIS method requires oxygen in the water to create Reactive Oxygen Species (ROS), which are partly responsible for the disinfection [102,104]. Six hours of exposure of unsafe drinking water to the sun in plastic PET bottles results in an inactivation of enteric pathogens (bacteria, protozoa, and viruses) of several orders of magnitude [104–107]. It is reported [108] that, under clear 1.5 atmospheric mass conditions, SODIS is predicted to treat up to 0.057 L/m^2 s, leading to 2-log inactivation of *E. coli*, 0.0016 L/m^2 s for *C. parvum*, and 0.022 L/m^2 s for *MS2*.

3.3. Photocatalytic Disinfection

It is well-known that all frequently used processes for disinfection like: chlorination, ozonation, or even UV light have problems and limitations. The main difficulty is the toxic DBP formation and their impact on water quality in the presence of natural organic matter (NOM) [32,109–114]. In the case of UV disinfection, the problem with DBP does not exist. However UV irradiation can modify the structure of dissolved organic matter (DOM) and increase biodegradability [115,116]. Furthermore, the lack of residual effect of UV disinfection is the biggest disadvantage and chlorination as a secondary disinfectant must be used [28,32]. Moreover, the chlorination after UV treatment could led to increased chloropicrin, chloroform, dichloroacetic acid, trichloroacetic acid, and cyanogen chloride formation [27,32]. A developing alternative that can be considered as an effective low-cost disinfection process is the use of semiconductors as catalysts to promote photo-disinfection. After the successful application of TiO_2-Pt for the inactivation of *Lactobacillus acidophilus*, *Saccharomyces cerevisiae*, and *E. coli* [117], a growing interest was put on photocatalytic disinfection [35,38,40,118–120].

Since this time, the inactivation of *E. coli* via photocatalytic disinfection has been the most studied [39,121–123]. The order of inactivation of *E. coli* and other coliforms under simulated sunlight using TiO_2 as photocatalyst has been presented by Rincón and Pulgarin [36] in the following sequence of efficiency: *E. coli* > Gram-negative > other coliform >>> Enterococcus species. Bogdan et al. [124] showed the wider classification of susceptibility to disinfection (viruses > prions > Gram-negative bacteria > Gram-positive bacteria > yeasts > molds).

Since it is known that photocatalytic disinfection is effective towards several kinds of microorganisms, there is much investigation on this mechanism. Several researchers tried to explain the interaction between generated ROS and biological structure (cells), which influences the inactivation or loss of viability [35,53,73,74]. It is well known that the photoexcitation of semiconductors led to ROS generation (hydroxyl radical ($HO^{\bullet}$), superoxide radical-anion ($O_2^{\bullet -}$), hydroperoxyl radical ($HO_2^{\bullet}$), and hydrogen peroxide (H_2O_2)). Probably due to the findings of a linear correlation between hydroxyl

radicals and the inactivation of *E. coli* [53], most of the publications suggest that the disinfection occurs by the action of $HO^{\bullet}$. However, new findings proved that not only H_2O_2 but also singlet oxygen ($^1O_2$) may play an important role in the photocatalytic disinfection mechanism [35,74]. However, before ROS can act, the adhesion of bacteria to the photocatalyst as well as ROS generation onto the surface has to take place [125]. During photocatalytic disinfection, the attack of ROS on microorganisms firstly occurs at the outside of the organism, and next inside, this action permits destroying the genetic material and inhibiting the metabolic processes [37]. Gomes et al. [126] verified that noble metals doped TiO_2 were interesting photocatalysts for the removal of *E. coli* from water (10^3–10^4 CFU/mL). Pd-TiO_2 and Ag-TiO_2 did not require light to totally remove bacteria. Moreover, no regrowth was observed. This behaviour was related with the bactericide role of Ag and Pd. The authors compared this process with single ozonation and verified that it was able to totally remove *E. coli* in 30 s, corresponding to a transferred ozone dose of 0.16 mgO_3/L (Figure 2). While low bacteria removal was obtained when 0.05 mgO_3/L were used, total depletion was observed when a TOD of 0.16 mgO_3/L was applied. Moreover, no regrowth was detected, which confirms the efficiency of ozone to degrade *E. coli*. Still, the use of Pd-TiO_2 and Ag-TiO_2 seems more suitable, since no energy was required for disinfection.

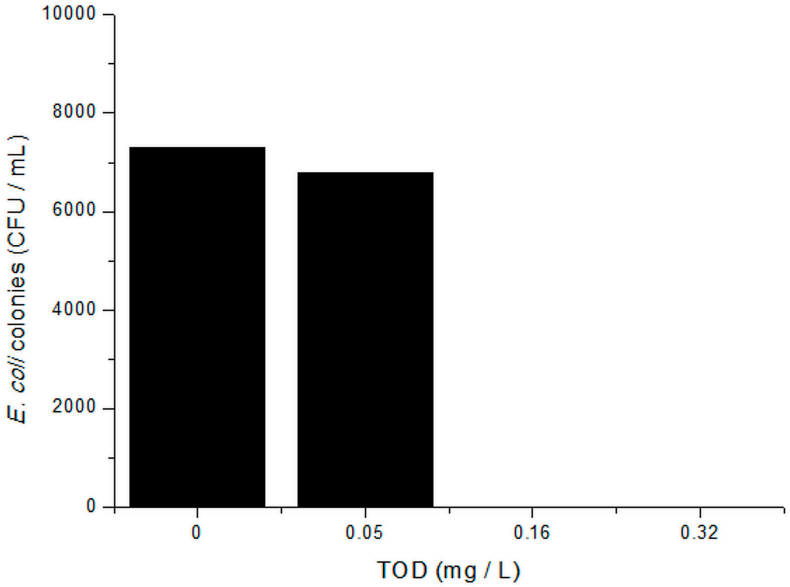

Figure 2. *E. coli* in synthetic wastewater as function of TOD during single ozonation (based on [126]).

Based on the satisfactory results, the solar photocatalytic disinfection (SPC-DIS) started to develop and focus on more resistant microorganisms, like: *Cryptosporidium parvum* [119,127,128], *Bacillus subtilis* [118], protozoa [118], photogenic fungi ([38,118], and MS2 [129]. However, according to Lonnen et al. [101], SPC-DIS was ineffective against the cyst stage of *Acanthamoeba polyphaga*.

The biggest challenge of photocatalytic disinfection is the generation of enough ROS to overcome the defence systems of the microorganisms to reach complete inactivation and avoiding bacterial recovery or regrowth [37]. To guarantee no bacterial regrowth before water consumption, the effective disinfection time (EDT) that is required for total killing of bacteria has to be determined [36]. According to the definition, EDT is the necessary time required for the total inactivation of bacteria without regrowth during a subsequent dark period referenced at 24 or 48 h after ending phototreatment [36]. This parameter is used as an indicator of the solar disinfection processes (including photocatalytic, solar-Fenton) instead of the UV dose that is required to achieve a certain level of disinfection [36,130–132]. Moreover, photocatalytic disinfection has a remarkable advantage, which is the so called "residual disinfecting effect". This effect leads bacteria counts to continue to decrease under dark conditions after the photocatalytic process as well as under fluorescence light irradiation where photoreactivation

should happen [36,131,133]. Furthermore, 24–60 h after the TiO_2-based photocatalytic treatment no bacteria growth was observed and this effect was mostly dependent on the irradiation intensity rather than on the dose [36,131,133]. This "residual disinfecting effect" was also observed [42] when UV-LED lamp was used for irradiation of TiO_2. The explanation of "residual disinfecting effect" after photocatalytic oxidation could be the production of H_2O_2 in water during the photoprocesses. This reactant may persist in water for a few hours after the treatment [134]. Possibly, H_2O_2 could lead to the generation of other reactive oxygen species, inhibiting bacteria reproduction acting as "after-illumination residual disinfectant" [42].

Table 3 resumes some results that were obtained from literature regarding bacteria and ARG removal from water (from different sources) through heterogeneous photocatalysts.

Dunlop et al. [135] tested P25 efficiency under UVA light for the inactivation of *E. coli*. It was verified that the reduction of ARG genes was lower for real wastewater, which was attributed to ROS scavenging by the organic and inorganic components of the effluent. It was highlighted that, in addition to bacterial re-growth, ARG transfer may increase if treatment is not continued to the point of complete pathogen inactivation prior to discharge.

TiO_2 activity under simulated sunlight was enhanced when it was doped with N, which led to higher *E. coli* inactivation efficiency [136]. The process did not significantly affect the resistance of *E. coli* strain to TET and VAN genes as irradiation time increased, but a decreasing trend in the resistance to CIP and sensitivity to CEF was observed. The higher efficiency of N-TiO_2 is related with the semi-conductor band-gap decrease when N is incorporated. This leads to a higher amount of electrogenerated electron-holes. Also, metal (Mn/Co) doped-TiO_2 exhibited better activity when compared to the commercially available TiO_2 on the removal of *Klebsiella pneumoniae* [137]. Karaolia et al. [11] showed the complete and irreversible removal of *E. coli* after 3 h of photocatalytic oxidation using simulated sunlight. Specific genes were degraded, while others, such as sul1, ermB resistance genes, and enterococci detected via the 23S rRNA gene sequences, were persistent throughout the treatment (suggesting that their removal is more challenging when compared to the removal of the other sequences examined). TiO_2 photocatalytic treatment displayed no reduction of ecfX, but the photocatalytic treatment with TiO_2-rGO-PH and TiO_2-rGO-HD successfully reduced it.

The effect of the water matrix was evaluated by Guo et al. [138] on the removal of *Staphylococcus aureus* and *Pseudomonas aeruginosa* through UVC/TiO_2 and concluded that disinfection efficiency was independent from the aqueous matrix. A similar conclusion was withdrawn by Guo et al. [138] on the damaging of ARGs during photocatalytic oxidation aided by H_2O_2. Moreover, the removal of mecA and ampC ARG was significantly improved by 2.7–3.4 and 2.7–3.2 log units in comparison to H_2O_2 and H_2O_2/UV. In fact, Moreira et al. [139] verified that P25/H_2O_2 and solar/H_2O_2 are suitable processes for ARGs removal from an effluent coming from an urban wastewater treatment plant.

Taking into account all the advantages from photodisinfection, the most important is the effectivity of the applied process as well as the time that is required for inactivation. According to Cho et al. [127], the $HO^{\bullet-}$ driven disinfection (Fenton as well as photocatalytic process) required few magnitude lower CT than ozone, ClO_2, and free chlorine (Table 4). Based on the lowest CT, the possible residual disinfection effect as well as possibility of application solar light or low-cost LED lamp, the photocatalytic disinfection seems to be the best technology for the removal of pathogens from water.

Table 3. Application of heterogeneous photocatalytic processes for water disinfection.

Process	Operational Conditions	Most Relevant Results	Bacteria	ARG	References
Photocatalysis/UVA	TiO_2 (P25) $[TiO_2]$ = 0.5 mg/cm^2 immobilised onto borosilicate plate, 2 × UVA lamp (9 W 80 W/m^2) DW and ASE from UWWTP	Bacteria declined from 3 log to 0.5 log (180 min). Gene pair conjugant numbers after treatment in DW showed a four-fold increase. In effluent, a lower reduction in ARG gene pair conjugates was observed.	*E. coli*	9:1 mixture of J-53R (recipient) to HT-99 (donor) (i.e., the ARG recipient within the conjugated pair was present in 10-fold excess)	[135]
photocatalysis/simulated Sunlight	N-doped TiO_2 $[TiO_2]$ = 25–500 mg/L Simulated sunlight (250 W lamp equipped with a UV filter) Effluents from UWWTP	Total inactivation of *E. coli* (60 min) N-TiO_2 lead to higher efficiency than TiO_2	*E. coli*	cip, cef, tet and van according to Kirby–Bauer test.	[136]
photocatalysis/Simulated sunlight	TiO_2 TiO_2-rGO-PH TiO_2-rGO-HD [Cat] = 100 mg/L Solar simulator Newport type 91193 (Xe Lamp (Vis, 1000 W, 63 W/m^2), pH 5.2–6.2 effluents after MBR from WWTP	Total inactivation of *E. coli* (180 min)	*E. coli*	sul1, ampC, ermB, mecA, ecfX, 23S rRNA	[11]
Photocatalysis/UVC	TiO_2 thin film UVC (254 nm, 300 W, 800 W) fluence = 0–120 mJ/cm^2 PBS solution (pH = 7.4) NW from drinking water source (pH = 7.2)	No difference was observed between PBS and NW on bacteria inactivation. 5.8 log mecA reduction and 4.7 log ampC reduction were achieved (120 mJ/cm^2) in the presence of TiO_2 for both matrixes.	*Staphylococcus aureus*, *Pseudomonas aeruginosa*	mecA, ampC	[138]
photocatalysis/simulated and natural sunlight	Mn-, Co- and binary Mn/Co$eTiO_2$ Natural and simulated solar irradiation (150 W), effluents after CAS from WWTP spiked with *K. pneumonia*	Simulated solar irradiation, yielding 6 log reduction after 30 min. Slower under natural solar irradiation (2 log). Only sul1 and ampC remained after Mn/Co-TiO_2 photocatalysis.	*Klebsiella pneumoniae*	tetA, tetM, sul1, blaTEM, ampC	[137]
Photocatalysis/UVC/H_2O_2	TiO_2 thin film UVC (254 nm, 300 W, 800 W) fluence = 0–120 mJ/cm^2 $[H_2O_2]$ = 10–100 mmol/L PBS solution (pH = 7.4) NW from drinking water source (pH = 7.2)	Increasing H_2O_2 concentration increased efficiency.	*Staphylococcus aureus*, *Pseudomonas aeruginosa*	mecA, ampC	[138]
Photocatalysis/natural sunlight TiO_2/H_2O_2 Solar/H_2O_2	TiO_2 (P25), GO-TiO_2, [Cat.] = 200 mg/L $[H_2O_2]$ = 20 mg/L Effluent from UWWTP	P25/H_2O_2 and Solar/H_2O_2 most eficiente processes for ARGs reduction.	Faecal coliforms and enterococci	16S rRNA, intI1, qnrS, blaCTX-M, sul1, blaTEM and vanA	[139]

ASE—autoclaved secondary effluents, DW—distilled water, PBS—phosphate-buffered saline, MBR—membrane bioreactor, NW—natural water, CAS—Conventional Activated sludge process; UWWTP—Urban wastewater treatment plant.

Table 4. CT values required by different disinfection methods for 2 log inactivation of selected pathogens from water.

		CT # (mg min/L) for 2 log Inactivation		
		Escherichia coli	***Bacillus subtilis* spores**	***Cryptosporidium parvum***
HO• radical driven ozone		0.000015 0.006	0.000082 4.1	0.000093 3.5
Chlorine driven	ClO_2	0.08	50	70
	Cl_2	0.13	450	4200

CT—for ozone and chlorine driven disinfection the common used CT while for hydroxyl radical driven disinfection OH radical CT (HO• radical concentration ×contact time).

3.4. Photocatalytic Ozonation

Ozone can also be coupled with these light driven systems, leading to an enhancement of the disinfection efficiency [140]. This coupling leads to an improvement of hydroxyl radicals production due to the synergetic effect between ozonation and photocatalysis [141]. Ozone electrophilic character enhances photogenerated electrons to be withdrawn, which minimizes the electron-hole recombination contributing to a more effective and fast process [142,143].

Moreira et al. [141] verified that photocatalytic ozonation using immobilized TiO_2 and UV LEDS as light source was able to remove different groups of cultivable microorganisms and ARGs. However, some microorganism regrowth after three days storage of the treated water was observed (even if ARG did not increase after that period). Thus, further studies must be performed to optimize this process so that safe reusable water may be reached.

Mecha et al. [144] studied the synergetic effect of the combination between ozonation and photocatalysis on the disinfection of municipal wastewater. The target pathogens were *E. coli*, *Salmonella*, *Shigella*, and *Vibrio cholerae*. Although the contact time was longer when using real wastewater, all species were totally removed with the combined system with treatment time being reduced up to 50–75% when compared with the single systems. Moreover, no regrowth was detected after a period of recovery of 48 h. The main drawback of this integrated system is the operating costs related with ozone production. In fact, the use of sunlight as a radiation source avoids the use of lamps. However, the selection of a suitable reactor configuration is a challenge.

The combination of ozone with catalysts/light/H_2O_2 may be a suitable solution for disinfection (Figure 3). Still, the operating costs that are associated may be prohibitive, especially for low incoming countries.

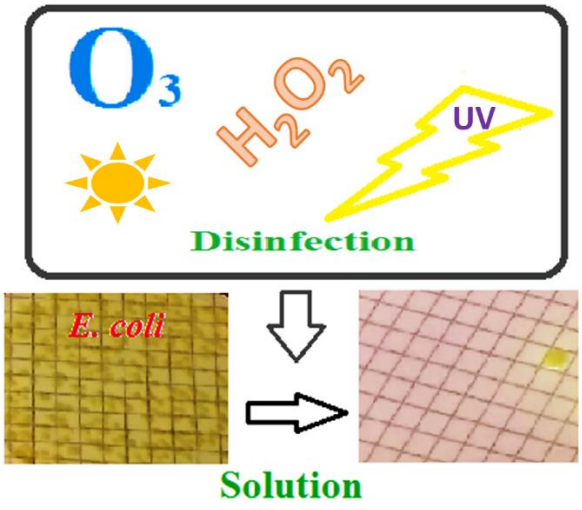

Figure 3. Ozone-aided processes for water disinfection.

4. Application to Real Water and Integrated Disinfection Schemes

The efficiency of the disinfection processes seems to depend upon the water composition but also upon the microorganisms characteristics. For example, the physiological state of the bacteria community and composition seems to play an important role during disinfection [145]. Thus, it is important to perform studies using real wastewaters to confirm data coming from spiked water with laboratory growth microorganisms.

Zimmermann et al. [146] developed a model involving hydraulics (data obtained from residence time distribution analysis) and chemical kinetics to predict the behavior of a full scale ozonation reactor. The pollutants (P) (including *E. coli*) removal kinetic was considered to follow a second order rate law when considering the contribution of molecular ozone and hydroxyl radicals [147,148] (Equation (2))

$$ln\frac{[P]}{[P_0]} = -\left(k_{p,O_3} + k_{p,HO\bullet}Rct\right)\int [O_3]dt \quad (2)$$

k_{p,O_3} and $k_{p,HO\bullet}$ are the second order kinetic constants for the reaction between the pollutants and ozone or hydroxyl radicals, respectively. Rct is the ratio between the concentrations of hydroxyl radicals and ozone. Although fast inactivation of *E. coli* was observed in the full scale reactor (reductions from 0.5 to 2.5 log units), the developed model strongly overestimated the reactor performance. This behavior was related with some shielding effect due to sludge flocks that somehow protected *E. coli* from oxidation.

Lee et al. [145] reached up to 99% of autochthonous bacteria removal in real wastewaters (with dissolved organic carbon (DOC) within the range 4.7–7.0 mg/L) using 0.5 $gO_3/gDOC$. The combination between ozonation and $UV_{254\ nm}$ radiation was tested on the disinfection of municipal wastewater [149]. The high initial ozone demand of the effluent (probably related with the chemical oxygen demand of the sample) reduced disinfection efficiency. Transferred ozone doses of 10 mg/L lead to *E. coli* removal. Whereas, a UV dose of 16.5 mJ/cm^2 was necessary to achieve the same result. When pre-ozonated samples (2 to 3 mg/L) were subjected to UV, the dose was slightly reduced to 12.4 mJ/cm^2, so that *E. coli* inactivation occurred. This way, no synergetic effect was observed between these two disinfection processes.

Water disinfection is also an important point when dealing with aquaculture systems. Tighter water control is required when water is recirculating through the system. Without the proper disinfection, opportunistic pathogens would build up in the fish tanks. Ozone is able to reduce up to 99.9% of *Aeromonas liquifaciens*, *A. salmonicida*, *Pseudomonas fluorescens*, and *Yersinta ruckerii*, which are bacterial fish pathogens [150,151]. It was also effective to reduce fish virus IHNV and IPNV [152].

The use of moderate ozone doses followed by UV irradiation led to the total inactivation of heterotrophic bacteria in the water stream [153,154]. It was concluded that most of disinfection was due to UV, but that process efficiency was improved by the former ozonation step, probably due to the increase on the water UV transmittance and reduction on particles. An important point that was discussed by Summerfelt et al. [153] is the way to automatically control ozone dose to fulfill the water ozone demand. The direct dissolved ozone concentration measurement is time consuming and shows high variability. An alternative is to indirectly obtain ozone dose through the water oxidation-reduction potential.

Fan et al. [155] proposed an alternative tap water treatment disinfection sequence. The traditional one involved pre-oxidation, coagulation, sedimentation, sand filtration, granular activated carbon (GAC) filtration, membrane ultra-filtration, and disinfection. The hybrid treatment combines ultrafiltration using ceramic membranes with ozonation in one single step. This step is preceded by coagulation and proceeded by GAC filtration. This method involving a lower number of operations seems adequate to remove turbidity, particles, bacteria, *Cryptosporidium*, and *Giarda* cysts, as well as microcontaminants, such as pharmaceutical and personal care products. Moreover,

the direct integration of ozone with the ceramic membranes reduces fouling and improves ozonation performance since catalytic ozonation will occur inside the membrane pores. According to the authors, such an approach leads to an energy consumption of 0.51 kW/m^3.

The selection of the most suitable technology must bear in mind the costs that are associated to the process and also the specificities of the area where the treatment/disinfection plant will be installed. For example, in remote areas it is also critical that the unit requires low maintenance and low specialized intervention [156]. In this context, ozone-based systems may be an interesting solution due to the high degree of automation and the low amount of chemicals needed for the process. The main disadvantage is related to the need of complex equipment, such as air treatment (to concentrate oxygen), ozone generator, and off-gas ozone destroyer. All of this implies energy costs during the operation. The main advantage of these processes is related with their capability of removing organic compounds as well as disinfecting, which may increase the final water quality. Also, UV systems can be easily automatized. However, in that case, as discussed before, a secondary oxidant will be required to maintain a residual disinfectant effect. Although the high interest in photocatalytic oxidation/ozonation disinfection, the use of catalysts in their powder form is an important disadvantage, since a solid/liquid separation step must be designed [157]. Thus, research in that field should focus on the development of structured catalysts that may be used in fixed beds without losing much of their activity.

Solar water disinfection processes are of special interest since the Sun can be used as a renewable and low-cost radiation source. The simple method of storing water in PET bottles that are left under Sun irradiation has shown important results in the disinfection of water in low-incoming countries [99]. In fact, results show that this methodology already significantly reduced the incidence of dysentery and diarrhea [158,159]. However, the wide spreading of such simple technology seems difficult in developed countries where it will be hardly seen as suitable for water disinfection [104]. Besides, the use of plastic bottles leads to some scepticism regarding the possibility of some chemicals leaching to the water. Research in this field is focused on the development of reactors able to enhance solar disinfection such as the one developed by Polo-López et al. [160] that would allow the process automatization. Still, the activity of such methods is not very much explored on the reduction of viral activity in water and it is verified that usually these pathogens are more resistant to solar disinfection [161]. Thus, aiming to reach safe reusable water, such problematic must be also addressed in future works.

5. Conclusions

Despite the attempts to reduce antibiotic consumption, the global resistance has increased significantly. During the last few years, attention was paid on combating the spread of ARB and ARGs in wastewater treatment plants, which are reservoirs of microbes and resistance genes. However, the efficient reduction of resistant bacteria by conventional wastewater treatment and disinfection methods have been lately investigated. In the full scale, chlorine and UV disinfection do not significantly affect ARGs abundance. Furthermore, during those processes, the regrowth of bacteria could occur. Therefore, evaluation of ARGs fate after advanced disinfection processes, in order to minimize the potential for subsequent antibiotic resistance transfer, should be addressed. The biggest challenge of photocatalytic disinfection is the generation of enough reactive oxygen spices to overcome the defence systems of the microorganisms to avoid complete inactivation and bacterial recovery or regrowth. The application of ozone and photo-based disinfection as a hybrid technology seems to be an interesting option for tertiary treatment of water and municipal wastewater.

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