REVIEW PAPER



Advanced Oxidation Processes for Water and Wastewater Viral Disinfection. A Systematic Review

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Received: 26 January 2021 / Accepted: 6 June 2021 / Published online: 14 June 2021 © The Author(s), under exclusive licence to Springer Science+Business Media, LLC, part of Springer Nature 2021

Abstract

Water and wastewater virological quality is a significant public health issue. Viral agents include emerging and re-emerging pathogens characterized by extremely small size, and high environmental stability. Since the mainly used conventional disinfection methods are usually not able to achieve complete disinfection of viral and other microbial targets, in real water and wastewater matrices, effective strategies for the treatment, use and reuse of water and the development of next-generation water supply systems are required. The scope of the present systematic review was to summarize research data on the application of advanced oxidation processes (AOPs) for viral disinfection of water and wastewater. A literature survey was conducted using the electronic databases PubMed, Scopus, and Web of Science. This comprehensive research yielded 23 records which met the criteria and were included and discussed in this review. Most of the studies (14/23) used only MS2 bacteriophage as an index virus, while the remaining studies (9/23) used two or more viral targets, including phages (MS2, T4, T7, phiX174, PRD-1, S2, \phiB124-14, \phicrAssphage) and/or Adenovirus, Aichivirus, Norovirus (I, II, IV), Polyomavirus (JC and BK), Sapovirus, Enterovirus, Coxsackievirus B3, Echovirus, and Pepper mild mottle virus. The vast majority of the studies applied a combination of two or more treatments and the most frequently used process was ultraviolet light-hydrogen peroxide (UV/H₂O₂) advanced oxidation. The review is expected to highlight the potential of the AOPs for public health protection from the waterborne viral exposure.

Keywords Advanced oxidation processes \cdot Water \cdot Wastewater \cdot Treatment \cdot Disinfection \cdot Virus

Introduction

General Issues

The contamination of water resources is an issue of global concern and the need for provisions of clean water is increasingly becoming demanding (Kokkinos et al., 2020). Moreover, water scarcity has been identified as a major problem of this century (Schmitz et al., 2016). Limited and

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frequently deteriorated freshwater resources, along with an increasing water consumption, combined with the climate change has led to water reclamation concepts, which support the reuse of treated wastewater worldwide for different purposes (e.g. agricultural, industrial, potable, etc.) (Giannakis et al., 2017a; Gomes et al., 2019; Schmitz et al., 2016). Water microbiological quality issues have been extensively investigated, since waterborne outbreaks have been recorded in both developing and developed countries, and contaminated water may seriously affect economic activities (primary production, tourism, etc.) (Kokkinos et al., 2011, 2020). Bacteria, viruses and protozoa are the cause of many emerging and new waterborne infectious diseases, and the most common cause is fecal pollution of human and/or animal origin (Kokkinos et al., 2020). More stringent water quality regulations are adopted and, nowadays, it is well recognized that chlorination and other conventional disinfection methods are often not able to achieve complete disinfection of bacterial, viral and protozoa microbial targets, in real water and wastewater matrices (Galeano

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et al., 2019; Giannakis et al., 2017a; Nieto-Juarez & Kohn, 2013). The prevalence of human enteric viruses in water and wastewater may pose a serious threat to public health, and new and more efficient treatment methods are required to ensure the microbiological quality of water and specifically the abatement of enteric viruses to levels that pose no significant risk to human health (Gerba et al., 2018; Nieto-Juarez & Kohn, 2013). Interestingly, the vast majority of the viral agents, which are transmitted via the fecal-oral route are non-enveloped, highly stable under environmental conditions, characterized by extremely small size, and include emerging and re-emerging pathogens with the most relevant of them belonging to the families of Adenoviridae, Caliciviridae, Hepeviridae, Picornaviridae and Reoviridae (Rodríguez-Lázaro, et al., 2012). The enteric viruses of human stool and urine belong to more than 140 types (Kokkinos et al., 2011). Surprisingly, untreated wastewater has been identified as the most diverse viral metagenome examined so far, with most sequence reads having little or no sequence relation to known viruses, indicating that most of the viruses have not yet been characterized (Cantalupo et al., 2011). The current global outbreak of SARS-CoV-2, the virus that causes COVID-19, has highlighted the urgent need to investigate the fate and transport of coronavirus and other enveloped viruses in urban sewage and drinking water, and also to develop rapid, sensitive, specific and inexpensive methods for water virological analysis (Foladori et al., 2020; La Rosa et al., 2020).

Advanced Oxidation Processes

Nowadays, it is well recognized that the existing biological and physicochemical treatment methods are unable to deal with the complete abatement of viral agents in hydric resources. In spite of its inherent weaknesses such as the production of disinfection by-products (e.g. trihalomethanes), chlorination is still the most common disinfection technique (Giannakis et al., 2017a). However, the adoption of advanced treatment technologies such as advanced oxidation processes (AOPs) are a promising approach for the improvement of water and wastewater treatment (Feitz, 2005). AOPs have emerged as promising, environmentally friendly and efficient alternative disinfection methods to conventional ones, to control water microbiological quality. They rely on the in situ formation of chemical oxidants to disinfect water and degrade diverse harmful organic contaminants (Giannakis et al., 2017a; Marjanovic et al., 2018; Nieto-Juarez & Kohn, 2013; Shabat-Hadas et al., 2017). AOPs are, in practice, redox technologies including different processes such as ozonation, ozonation coupled with H₂O₂ and/or ultraviolet (UV) radiation, Fenton and alike reactions, photocatalysis activated by semiconductors such as TiO₂, sonolysis, electrochemical oxidation, and various combinations of them (Fig. 1). They are based on the production of highly reactive oxygen species (ROS), characterized by the non-selectivity on the target and can be used as pre- or post-treatment to a biological process (Galeano et al., 2019; Kokkinos et al., 2020; Monteiro et al., 2015). The principal oxidizing agent is the hydroxyl radical, being the second most powerful oxidant after fluorine. However, other ROS may also be produced (e.g. hydroperoxyl radicals, superoxide radical anions, etc.) (Giannakis et al., 2017a, 2017b, 2017c). Hydroxyl radicals are frequently produced by the homolytic cleavage of the O-O bond of hydrogen peroxide with UV light (Bounty et al., 2012; Shabat-Hadas et al., 2017). A wellstudied AOP is the photo-Fenton process, in which hydroxyl radicals are produced by light, iron and hydrogen peroxide (Nieto-Juarez et al., 2010, Marjanovic et al., 2018). It is an environmentally friendly, simple, low-cost process which has been shown to be effective in the abatement of structurally simple, complex or resistant microbes (Giannakis et al., 2017a). Indeed, AOPs have shown a high disinfection potential against a wide range of microorganisms like virus, protozoa, spore-forming bacteria, fungus, and yeasts, mainly through the action of ROS such as singlet and triplet oxygen, anion-radical superoxide, hydroxyl and hydroperoxyl radical, and hydrogen peroxide. ROS are known oxidants of different types of molecules, such as proteins, lipids, and also nucleic acids. Specifically for nucleic acids, ROS may exert their action at different levels (e.g. change the nucleotides, break the phosphodiester bond, enhance the formation of pyrimidine dimers, change the tridimensional structure, and affect DNA replication) (Galeano et al., 2019). Different factors, such as the use of sunlight (a renewable source of energy), the minimal needs of hydrogen peroxide and consequently reduced costs, and the application of a low-cost catalyst such as Fe³⁺, which is frequently present in natural waters, render photo-Fenton a process of great potential for viral abatement in water matrices (Ortega-Gomez et al., 2015). Sulfate radicals, which are characterized by a redox potential close to that of hydroxyl radicals (2.6 V), may be used as an alternative to hydroxyl ones, since they are more selective against various target contaminants and pollutants. Different activating factors (light, heat, transition metals, etc.) may promote the production of sulfate radicals from chemicals such as peroxymonosulfate and peroxydisulfate (PDS) (Marjanovic et al., 2018). Although during the last years, the photo-Fenton and alike processes have been used as a green alternative to chemical disinfection of waters and wastewaters, they have still to be completely clarified (Giannakis et al., 2017b, Giannakis, 2018). Different studies have revealed the order of microbial resistance to solar photo-Fenton process as follows, bacteria < viruses < spores (fungi or bacteria) (Giannakis et al., 2016). The assessment of the disinfection using light-driven AOPs is based on the use of different approaches such as computational fluid dynamics,



chemical actinometry or biodosimetry (Shabat-Hadas et al., 2017). The concurrent abatement of pathogens and chemical pollutants by solar-enhanced AOPs has been reviewed by Tsydenova et al. (2015)

To face the limitations of the homogeneous Fenton processes, which are associated with the pH-dependent solubility and stability of ferrous and ferric species, the use of an alternative iron source, i.e. iron (hydr)-oxide particles, has been assessed in a process called heterogeneous Fenton-like. The effectiveness of such a process for viral inactivation catalyzed by colloidal iron around neutral pH under sunlight has been proved (Nieto-Juarez & Kohn, 2013; Nieto-Juarez et al., 2010). Interestingly, iron-bearing particles may support viral disinfection by different ways, i.e. physical removal by adsorption onto particles, by Fentonlike processes, photocatalysis or disintegration upon adsorption (Nieto-Juarez & Kohn, 2013). More research is needed though on viral inactivation by photo-Fenton process (Giannakis et al., 2017b, 2017c).

Nanoparticle-mediated photocatalysis is another interesting approach for water disinfection, and TiO_2 has been found to be effective against different pathogens such as viruses, bacteria, and fungi (Prasse & Ternes, 2010) Zhu et al. (2020)



Fig. 2 Donut chart visualization of the grouping of the reviewed studies into the main categories of Advanced Oxidation Processes. The vast majority of the studies applied chemical and photochemical processes, while limited cases on the application of electrochemical and other AOPs were also found

reviewed the formation of nanostructured manganese oxides and their application in wastewater remediation; these oxides can catalyze peroxides to produce ROS in aqueous phase, thus initiating in situ chemical oxidation and AOPs. Duan, (2019) performed a proof-of-concept study to unravel the principles in developing fine-tuned and high-performance transition metal (TM)@carbon composites for advanced catalytic oxidation for water decontamination.

During the last decades, the UV technology is upgraded through its application in AOPs (Timchak & Gitis, 2012). The use of UV irradiation for water disinfection of enteric pathogens is an emerging technique characterized by high efficiency and the absence of the formation of disinfection by-products (Chu et al., 2012). Microbial disinfection by UV irradiation is mainly exerted by UV-induced photochemical reactions of the genetic material. Direct (or endogenous) inactivation involves the absorbance of UVB light by the viral genome, which causes its degradation. Although viral proteins may also absorb UVB radiation to a lesser extent, the contribution to viral inactivation has still to be clarified (Mattle et al., 2015). While UVC/UVB are strongly absorbed by DNA and have additive disinfection effects, UVA cannot induce DNA damage and has no direct photochemical reactions on DNA. However, it may produce reactive intermediates such as ROS (e.g. hydroxyl and superoxide radicals, hydrogen peroxide, etc.), which in turn may damage microbial targets (e.g. proteins, DNA, etc.) (Mamane et al., 2007; Song et al., 2019). Compared to DNA, RNA is known to be more susceptible to oxidative damage by exposure to UV (Galeano et al., 2019). In indirect (or exogenous) inactivation, UVB/UVA and visible light are known to be absorbed by different water sensitizers (e.g. organic matter, nitrate, iron-containing complexes, etc.), which produce reactive species with viral inactivation potential. The type of virus, as well as the solution conditions, are both critical factors, which dictate the relative contribution of direct and indirect inactivation (Mattle et al., 2015). The UV treatment disinfection efficacy may be impeded by absorbing particles and microorganisms that are captured in aggregates of particulate matter (Kosel et al., 2017). One of the most widely used AOP is the UV/H_2O_2 . Such a process has been shown to be highly efficient for the treatment of burdened wastewater like that of meat processing industry. (Yapıcıoğlu, 2018).

Another new advanced technique is hydrodynamic cavitation, which exerts its disinfective action through chemical (production of hydroxyl radicals) and physical mechanisms (pressure gradients, shock waves, shear forces, very high local temperatures). Although the exact mechanisms of viral disinfection have not yet been clarified, it may be hypothesized that hydrodynamic cavitation may cause structural damages at different components of a viral particle (coat, host recognition receptors, capsid, genome) (Kosel et al., 2017). We have recently reviewed the main categories of nanomaterials used in catalytic processes (carbon nanotubes/ graphitic carbon nitride (CNT/g- C_3N_4), composites/graphene-based composites, metal oxides and composites, metal–organic framework and commercially available nanomaterials) and discussed their application in the removal of different classes of pollutants, as well as for the elimination of bacterial, viral and protozoan microbial targets, from water and wastewater matrices (Kokkinos et al., 2020).

Since AOPs are typically energy-intensive processes, the so-called electrical energy per order (i.e. the energy needed to reduce the concentration of contaminant by an order of magnitude in a unit volume of water, EEO) is a useful measure to classify them in three groups as follows: <1 kWh m⁻³ (e.g. O_3/H_2O_2 , O_3/UV , UV/H_2O_2 , etc.), 1–100 kWh m⁻³ (e.g. photo-Fenton) and > 100 kWh m⁻³ (e.g. UV-based photocatalysis) (Miklos et al., 2018). The energy efficiency and treatment cost associated with the use of AOPs are important parameters for the practical assessment of their disinfection efficiency (Chen, 2021).

The wide application of AOPs is expected to enhance the removal of emerging contaminants and pathogens from water and wastewater (Sherchan et al., 2014). Accodring to Gerba et al. (2018), although advanced water treatment trains have been applied for the reduction of viral loads, it is of pivotal importance to understand viral inactivation processes by different treatments, improve current estimates and identify new research tasks (Gerba et al., 2018). A donut chart of the grouping of the reviewed studies into the main categories of AOPs is shown in Fig. 2.

Use of Indexes and Applicability

Viruses with diameters from 20 to 300 nm are an important threat to water safety and a significant microbial target, which should be taken into consideration in water treatment processes. Their diversity and polymorphic nature have been recognized and are continuously unraveled (Cantalupo et al., 2011; Giannakis et al., 2017b; Marjanovic et al., 2018) Enteric viruses are listed as emerging biological contaminants on the United States Environmental Protection Agency (USEPA) Contaminant Candidate List (Gomes et al., 2019). Water treatment technologies are required to achieve a 99.99% (4-log) reduction in sample viral concentrations after treatment, to be considered efficient by USEPA and Health Canada (Monteiro et al., 2015).

Bacteriophages have been used as indicators of water microbiological quality due to their specificity and resistance, compared to the traditional bacterial indicators, and they are valuable viral indicators for the assessment of different treatment techniques (Table 1) (Ghernaout, 2020; Ortega-Gomez et al., 2015). The USEPA has recommended the use of coliphages as surrogates of human enteric viruses

Method	Results	Reference
Solar disinfection enhanced by moderate addition of iron and sodium peroxydisulfate	 Solar/heat CP with natural Fe < solar/PDS Solar/ Fe < CP in presence of NOM < CP The abatement was achieved at minute-range residence times 	Marjanovic et al., 2018
Heterogeneous Fenton-like processes catalyzed by iron (hydr)oxide particles	1) Adsorption onto α -FeOOH, Fe3O4, Fe(OH)3 particles caused virus inactivation of 7%, 22%, and 14%, respectively, while first-order inactiva- tion rate constants were $6.6 \times 10 - 2$, $8.7 \times 10 - 2$, 0.55 and 1.5 min – 1, respectively 2) Additional inactivation of viruses which were adsorbed on particles was shown by the action of sunlight and H2O2, while the inactivation for suspended viruses was insignificant. In the absence of sunlight or H2O2, no inactivation was recorded apart that resulted by the adsorption, with the exception of Fe3O4, which inactivated viruses via a dark Fenton-like process	Nieto-Juarez, Kohn 2013
Ultraviolet reactor in combination with hydrogen peroxide	 Ultraviolet radiation caused a decrease of 5.3–5.8 log10 of MS2, and a 1.7–2.8 log10 decrease in viral RNA copy number MS2 abatement was increased by the addition of H2O2 (at 2.5 or 5 ppm with UV at different flow rates), with a reduction of more than 7 log10, while qPCR showed only a 3–4-log10 reduction in viral RNA copy number 	Sherchan et al., 2014
Ozone treatment of reverse osmosis concentrates	Ozonation caused a 5-log abatement in concentrate samples at 1.18 mg O3/mg DOC	King et al., 2020
Ozone, ozone/H2O2	Dissolved ozone concentration integrated over time values of 1 mg-min/L caused inactivations higher than 6-log	Gamage et al., 2013
Ultraviolet light-emitting diodes	UVA pretreatment followed by UVC inactivation was found to cause no amelioration of MS2 inac- tivation, in comparison to <i>E.coli</i>	Song et al., 2019
UV/H2O2 followed by free chlorine	No synergy was recorded for the treatment by UV irradiation followed by free chlorine, while increased inactivation was found when H2O2 was added in the primary UV disinfection step	Cho et al., 2011
Hydrodynamic cavitation	Reductions of viral infectivity higher than 4-logs were recorded	Kosel et al., 2017
UV/H2O2 flow through system followed by free chlorine	A 5.78 log abatement was achieved with a flow rate at 50 L/h, while a 4.49 log removal was achieved with a flow rate of 100 L/h	Chu et al., 2012
N-doped TiO2-coated Al2O3 photocatalytic membrane reactors (PMRs)	 In natural surface water the viral abatement under irradiation was found to be 4.9±0.1 log Complex virus-PMR interactions were found even before the exposure to light Electrostatic forces in addition to photocata- lytic inactivation dictate the viral abatement in a complex water matrix by PMR. While alkaline water pH causes a limited interaction and a reduced viral abatement by PMR, the addition of Ca2 + results to higher MS2 abatement 	Horovitz et al., 2018

Table 1 (continued)

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Iron- and copper catalyzed Fenton systems (H2O2, metal concentrations, HO• production, and sunlight)	 Viral reduction was first-order in relation to H2O2 The inactivation rate constant k_{obs} in the Cu/ H2O2 system, increased with added Cu up to 2.5 μM, and then was stabilized The inactivation in the Fe/H2O2 system was dictated by colloidal iron Sunlight irradiation influenced only the Fe/ H2O2 system, causing a 5.5-fold increase in k_{obs} (up to 3.1 min-1), 4) HO• generation could not account for the recorded abatement in the Fe/ H2O2 system 	Nieto-Juarezet et al., 2010
Photo-Fenton process (Fe species and concentra- tion, solar irradiance, pH and microbial competi- tion)	 In comparison to their Fe(III) counterparts, Fe(II) salts, resulted in a faster abatement, in any combination of H2O2 concentration, sunlight irradiance or starting pH (6–8) Starting with Fe(II) resulted to more iron in solution longer than Fe(III), which is responsible for higher inactivation kinetics Exposure to 600 W/m2 (30 min) in presence of Fe(III) and H2O2 (1:1 ratio) resulted in a 4-log MS2 reduction. 4) MS2 reduction was moder- ately reduced in presence of the bacterial host, indicating a limited competition for the oxidants in the bulk 	Giannakis et al., 2017a, 2017b, 2017c
Advanced oxidation processes (AOPs), UVC, UVC/H2O2, and UV/Fenton based	 A reduction of the viral load by approximately 4-log was recorded for UVC and UVC/H2O2 treatment, after 2 or 1.5 min of exposure, respec- tively Iohexol delayed kinetics of abatement to approximately 80% for MS2, in all studied matri- ces, while a value of 60% was recorded by the addition of H2O2 MS2 bacteriophage was found to cause an aver- age 45% reduction of the bacterial inactivation in all studied matrices, and similarly, <i>E.coli</i> was shown to delay the inactivation of MS2, except in synthetic wastewater matrices 	Giannakis et al., 2018
Heterogeneous photo-Fenton process	 Low concentrations of iron oxides in wastewater without H2O2 (wüstite, maghemite, magnetite) was found to cause restricted semiconductor- mediated MS2 inactivation The isoelectric point of the iron oxides and the active surface area are important parameters of the process, as was demonstrated by the working pH and the size of the oxide particles A significant enhancement of the abatement pro- cess was shown after the addition of low amounts of Fe-oxides (1 mg L - 1) and H2O2 (1, 5 and 10 mg L - 1), leading to heterogeneous photo- Fenton processes on the surface of the oxides Photo-dissolution of iron in the bulk, lead to homogeneous photo-Fenton, which was sup- ported by the complexation by the dissolved organic matter in the solution 	Giannakis S et al., 2017b

due to their interesting characteristics (similarity of structural, morphological, and inactivation resistance characteristics) (Schmitz et al., 2016). It is well recognized that the assessment of disinfection processes cannot be based on classic faecal bacterial indicators, since they are too sensitive to disinfectants compared to viral and protozoan pathogens (Sommer et al., 2004).

MS2 bacteriophage is an F+specific, single-stranded RNA phage, with similarities in size (27.5 nm) and structural complexity to some human enteric viruses. It is small, spherical (icosahedron), non-enveloped, and characterized by high resistance to chemical disinfectants and/or environmental stressed conditions (e.g. temperature, osmotic pressure, and desiccation). Interestingly, it seems to be more resistant to inactivation compared to many waterborne viruses of public health concern with similar basic features, such as size, morphology, structure and overall behaviour (Jolis, 2002). Due to its resistibility characteristics and its high numbers in sewage, it is frequently used as an index of wastewater contamination and a quantitative marker for the assessment of the function of water treatment plants and filtration devices as well as for the effectiveness of antiviral/antiseptic agents (Table 1) (Kosel et al., 2017). It is characterized by a negative charge under a wide pH range (isoelectric point ~ 3.9) and it is relatively hydrophobic (Horovitz, 2018). It is also non-pathogenic and easily propagated and purified in laboratory, up to elevated viral titers, which are frequently required for inactivation experiments. (Kosel et al., 2017, Giannakis et al., 2017a, 2017b, 2017c, Giannakis, 2018, Horovitz et al., 2018) A 5-log reduction of MS2 is used in California for the assessment of disinfection systems for non-potable reuse applications (King et al., 2020). It has been extensively used as a surrogate for human enteric viruses in studies of assessment of the AOPs potential for viral abatement. Its inactivation requires a high oxidation power because is mainly based on the denaturation of capsid proteins, whose structure is simple and rigid (Horovitz et al., 2018; Mamane et al., 2007). MS2 has been proposed as a reference virus for the assessment of solar disinfection (SODIS) processes, since it is expected to support the standardization and interpretation of diverse experimental setups, focusing on different water matrices and using different light sources and reactor setups (Cho et al., 2011; Gamage et al., 2013; Giannakis et al., 2017a; Marjanovic et al., 2018; Mattle et al., 2015; Sherchan et al., 2014).

Like MS2, PRD-1 (a coated, double-stranded DNA bacteriophage with an approximate diameter of 65 nm), PMMoV (a rod-shaped, 18×312 nm RNA virus used as a non-pathogenic fecal indicator), bacteriophages related to non-pathogenic human gut microbiota (e.g. ϕ crAssphage and ϕ B124-14, spherical, double-stranded DNA viruses with diameters of approximately 50 nm and 75 nm, respectively), as well as adenoviruses and polyomaviruses have been proposed as indicators for contamination and microbial-source tracking and have been used for the evaluation of viral disinfection in advanced water treatment systems (Table 2). (Papp et al., 2020; Schmitz et al., 2016; Sommer et al., 2004; Vaidya et al., 2019).

Adenovirus has been recorded to be the most resistant known pathogen to disinfection by UV light, a finding which affected the regulations on the application of UV disinfection for virus inactivation in surface and groundwater by USEPA (Bounty et al., 2012). Direct or indirect inactivation processes are responsible for the solar disinfection of viruses. Similarly to MS2, adenovirus is sensitive to ROS but relatively resistant to direct inactivation, while phiX174 (single-stranded DNA) was found to display the contrary behaviour (Mattle et al., 2015) Gerba et al. (2018) stressed out the need to use the appropriate viruses for the assessment of individual treatment processes. For example, they suggested the use of adenoviruses for the evaluation of UV light disinfection, while reoviruses have been proposed for the assessment of chlorination. (Gerba et al., 2018).

It should be taken in consideration that, although viral inactivation may be reached by targeting the viral capsid through direct reaction of disinfectants with its constituents, the diffusion of disinfectants inside other microorganisms through an outer membrane may be required. In such case, disinfectants with a short lifetime such as those produced by photo-Fenton processes, may not be efficient. (Nieto-Juarezet et al., 2010) Some studies focused on the MS2 inactivation by the photo-Fenton process, but complete viral abatement was not succeeded (Ortega-Gomez et al., 2015). The main MS2 inactivation pathway during photo-Fenton treatment has to do with the generation of oxidants in the bulk, while bacterial inactivation is principally attributed to oxidative-stress inside the cell and, to a lesser extent, to that from the bulk concentration of hydroxyl radicals. (Giannakis, 2018) Moreover, due to the absence of repair enzymes and molecular processes for the repair of nucleic acids damages, MS2 is not able to repair UV-induced RNA damages through photoreactivation, in the contrary to bacteria such as E. coli. (Song et al., 2019).

Phage T4 is one of the biggest double-stranded DNA bacterial viruses, and it has also been used as a viral indicator in numerous studies (Ghernaout et al., 2020). Similarly, T7 is a double-stranded DNA bacteriophage with many applications, especially in studies of solar UV disinfection assessment (Mamane et al., 2007). Endogenous inactivation by UVC/UVB irradiation has been found to depend on the type and size of the viral genome (Nelson et al., 2018). Hence, MS2 which is an RNA virus, shows a different UV inactivation behaviour compared to the T series viruses, which are DNA viruses (Mamane et al., 2007).

Compared to lab-made strains, naturally occurring pathogens are characterized by elevated resistance to oxidative processes (Giannakis et al., 2016). It must be pointed out that wild strains should also be considered to successfully assess the performance of engineered treatment processes; moreover, field scale systems should also be used for the confirmation of laboratory and pilot plant experimental data.

MS2 and T4 bacteriophages	Degradation of free and nano-bound RhB by direct UV photolysis, UV/H2O2 AOP and solar light- induced photocatalysis	 Minor Rhodamine B degradation was shown by direct UV and solar light A nearly linear Rhodamine B degradation was found in the presence of a solid catalyst by UV/ H202 and photocatalysis/photosensitization A significant adsorption of soluble (free) Rhoda- mine B was recorded on bismuth-based catalyst vs. no adsorption of virus-bound (nano-bound) Rhodamine B on this catalyst or of any form of the dye on titanium dioxide Virus-bound Rhodamine B showed high potential as an indicator of advanced oxidation process 	Shabat-Hadas et al., 2017
MS2 bacteriophage, and phages T4, and T7	UV/H2O2 advanced oxidation	 Bacteriophages T4 in phosphate buffered saline (PBS) were sensitive to > 295 nm filtered UV irradiation (without H2O2), while MS2 was very resistant addition of H2O2 at 25 mg/l in the presence of filtered UV irradiation over a 15 min reaction time did not result in any additional disinfection of virus T4 an additional one log inactivation for T7 and 2.5-logs for MS2 were obtained 	Mamane et al., 2007
MS2 bacteriophage, and phages phi X 174 and T4	Advanced UV/H2O2 oxidation process	 The viral abatement was not affected by the presence of dyes but the addition of hydrogen peroxide improved it. The addition of 0.2 M H2O2 at 70 mJ/cm2 enhanced the MS2 abatement by two logs, while was found to have no effect on phi X 174 and T4 phages. The presence of viruses caused the reduction of the bleaching of fluorescent dyes due to restricted availability of hydroxyl radicals and their preferential involvement in virus inactivation 	Timchak & Gitis, 2012
MS2 bacteriophage, and phages ϕ X174, and PRD-1	Ozone and hydrogen peroxide	The ozone/hydrogen peroxide process was found to have a significant microbicidal effect. MS2 bacte- riophage, and phages $\varphi X174$, and PRD-1 showed a reduction of approximately 6-log	Sommer et al., 2004
MS2 bacteriophage and Pepper mild mottle virus (PMMoV)	Two pilot-scale advanced water treatment plants were compared. The two treatment approaches included a carbon-based treatment process (flocculation/ sedimentation, ozone-biofiltration, granular acti- vated carbon (GAC) adsorption, ultraviolet (UV) disinfection; 20,000 L/day) and a membrane-based treatment process (ultrafiltration, reverse osmo- sis, UV-hydrogen peroxide advanced oxidation; 160,000 L/day)	In both advanced water treatment processes a > 8-log removal of MS2 and > 6-log removal of Pepper mild mottle virus was achieved	Vaidya et al., 2019

Table 2 (continued)			
MS2 bacteriophage and Echovirus	Photo-Fenton process (effects of reactant concentra- tion, H2O2, Fe2 +, Fe3 +, and solar irradiance)	 The solar exposure/Fe3 + treatment was strongly dependent on the concentration of iron and the intensity of solar irradiance A complete inactivation was recorded (from 10⁶ PFU mL-1 to the detection limit) with 1 mg L-1 of Fe3 + and 60 min of solar irradiance (45 W 40 m-2) The abatement of MS2 with the photo-Fenton process (solar exposure/H2O2/Fe2+/3+) performed with Fe2 + (detection limit reached at 20 min and 50 min, respectively). Echovirus complete inactivation by the photo-Fenton process was slower (reached after 120 min) probably due to the competition between the present organic matter in this analysis and Echovirus, for oxidative species 	Ortega-Gomez et al., 2015
Bacteroides bacteriophages $\phi B124$ -14, $\phi crAssphage, and Pepper mild mottle virus (PMMoV)$	Membrane Bioreactor (MBR) advanced treatment and full advanced treatment (FAT) train	 PMMoV, φB124-14, and φcrAssphage were detected in the MBR feed at concentrations of approximately 10³ gene copies (gc)/mL, 10⁵ gc/mL, and 10⁶ gc/mL, respectively Only PMMoV was detected above the limit of quantification in the MBR filtrate (25 ± 8 gc/mL) Viral Log removal values were found to be 1.4±0.5 for PMMoV, > 3.9±0.3 for φB124-14, and > 6.2±0.3 for φcrAssphage 	Papp et al., 2020
Adenovirus, and bacteriophages MS2, φX174	Solar Disinfection	1) $\varphi X174$ was more sensitive to the direct inactiva- tion with the highest quantum yield (1.4 × 10 – 2), compared to MS2 (2.9 × 10 – 3) or adenovirus (2.5 × 10 – 4) 2) Second-order rate constants were found to range from 1.7 × 10 ⁷ to 7.0 × 10 ⁹ M – 1 s – 1 and showed the following sequence: MS2> adenovi- rus > phiX174 3) A predictive model was used to assess the solar disinfection of MS2 and phiX174 in a natural water sample and approximated that of adenovirus within a factor of 6 4) Viral abatement was mainly performed by direct processes, although indirect inactivation by ¹ O2 supported also the disinfection of adenovirus and MS2	Mattle et al., 2015

Table 2 (continued)			
Adenovirus, and somatic and male-specific coliphage	Advanced oxidation-based Net-zero water (NZW) pilot system (septic tank, membrane bioreactor (MBR), aluminum electrocoagulation (EC), flocculation, vacuum ultrafiltration, peroxone or UV-hydrogen peroxide advanced oxidation, chlorine disinfection, and point of use granular activated carbon (GAC) filtration)	No viruses were found in the treated water, although adenovirus genetic material was detected probably due to the presence of inactive viral particles in hydraulic dead zones	Gassie et al., 2016
Adenovirus	Low-dose UV/H2O2 advanced oxidation	 A UV dose of approximately 200 mJ/cm² from a low-pressure source (emitting at 253.7 nm) was needed for a 4-log reduction of adenovirus Addition of H2O2 (10 mg/L) caused a 4-log viral abatement at a dose of 120 mJ/cm² 	Bounty et al., 2012
Adenovirus (human adenovirus type 5)	Photo-electro-oxidation process	 A 60 min treatment by the photo-electro-oxidation process caused an adenovirus reduction of 7 log10. Exposure of 75 min was required for the com- plete abatement of DNAse-treated samples, while completely non-viable adenoviruses were obtained after 30 min 	Monteiro et al., 2015
Norovirus genotype I and II, and JC virus	Single and catalytic ozonation (by a volcanic rock)	Catalytic ozonation caused the complete abatement of all target viruses (Norovirus genotype I and II and JC virus), while JC virus could not be eliminated even after 150 min of treatment by single ozonation	Gomes et al., 2019
Eleven (11) different virus types (pepper mild mottle virus, aichi virus, noroviruses genogroup I, II, and IV, enterovirus, sapovirus, rotavirus group-A, adenovirus, and JC and BK polyomaviruses)	Advanced Bardenpho technology	 Advanced Bardenpho wastewater treatment was proved to be more efficient compared to conven- tional treatments, Aichi virus was found to be a conservative index virus for the assessment of viral removal in waste- water treatment Bardenpho processes were the major sources of virus removal probably because of virus sorption to solids 	Schmitz et al., 2016
Enterovirus	Treatment train consisting of ozone, biological acti- vated carbon, microfiltration, reverse osmosis, and ultraviolet light with an advanced oxidation process	1) Performance distribution functions (PDFs) evi- denced treatment that consistently surpassed the 12-log thresholds for virus, (as required for potable reuse in California) 2) Application of free chlorine disinfection during the treatment, achieved a median annual infection risk by enterovirus of 1.5×10^{-14} (no failures) and a maximum annual value of 2.1×10^{-5} (assuming one 24-h failure per year)	Pecson et al., 2017

irradiation caused a TCID50 decrease of approxi-

mately~4-Log10

2) Simultaneous application of 3 mg/L peracetic acid with 20 mJ/cm² low-pressure ultraviolet

50% assay (TCID50) reduction to concentra-

tions $\leq 50 \text{ mg/L}$ at a contact time of 15 min

Kibbee & Ormeci, 2020

) Coxsackievirus B3 was found to be highly sensi-

No different peracetic acid formulations (15 and 22%) and low-pressure ultraviolet irradiation

20 mJ/cm², while it was very resistant to peracetic acid with $\leq 1 \text{ Log } 10$ tissue culture infectious dose

tive to low-pressure ultraviolet irradiation at

(Giannakis, 2018, Gerba et al., 2018) Numerous confounding factors affect the correct comparison of different treatment processes (e.g. season, wastewater origin, sampling and analysis methods, etc.) (Schmitz et al., 2016). Moreover, the biological diversity within families and genera of viruses and specific virus types (e.g. serotypes or genotypes) is a significant source of uncertainty (Gerba et al., 2018). More research is needed for the elucidation of the viral behaviour in natural surface waters and engineered systems in an attempt to enhance the disinfection of hydric resources (Ghernaout et al., 2020; Nelson et al., 2018).

Miklos et al. (2018) evaluated in their critical review the application of AOPs for water and wastewater treatment. They reviewed the major reaction mechanisms and formation of by-products, and also performed a critical comparison of different AOPs based on electrical energy per order (EEO) values. Chen et al. (2021) have recently reviewed AOPs for water disinfection. However, authors focused almost exclusively on bacterial inactivation and very limited data on viral abatement were reported. The current literature survey was performed with the view to summarize research data on the application of AOPs for viral disinfection of water and wastewater matrices and to identify the most frequently applied processes as a single or combined treatment(s). Overall, we aimed to highlight the potential of the AOPs for viral abatement and public health protection.

Materials and Methods

Data Sources and Search Strategy

Scopus, PubMed, and Web of Science were the selected databases used to find peer-reviewed articles dealing with the application of AOPs for viral disinfection of water and wastewater. Database searches were conducted between April and July 2020. However, a continuous reviewing of the databases has been performed during the whole time period of the preparation of the manuscript. The searches were conducted by using the following search terms and their variations in combination: "advanced oxidation processes AND water AND virus", "AOPs AND water AND virus", "advanced oxidation processes AND wastewater AND virus", "AOPs AND wastewater AND virus". The titles, abstracts and key words of articles included in the online databases were searched for these search terms.

Data Selection Criteria

This systematic review included both experimental and nonexperimental studies from 2000 onward. All study design types were included, while non-English language studies, conference abstracts, editorials and letters to the editor were

Table 2 (continued)

Coxsackievirus B3

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excluded. The selection criteria developed a priori were as follows: i) The type of water/wastewater matrix; ii) The type of target virus (indicator and/or pathogenic); iii) The type of AOP. Eventually, twenty three articles fulfilled the inclusion criteria and were analyzed in the systematic review. The outcome variables were extracted independently by two investigators into a spreadsheet (Excel, Microsoft) with a standardized approach and any disagreement was resolved by discussion.

Disinfection Studies

Photo-Fenton and Related Processes

For the photo-Fenton process, irradiation with sunlight or an artificial light source is required to enhance the treatment efficiency of the respective dark reaction by the photoreduction of Fe^{3+} to Fe^{2+} and the additional production of hydroxyl radicals. A drawback of this process is the additional cost for artificial irradiation (Brienza & Katsoyiannis, 2017). To overcome the limitations associated with homogeneous Fenton processes (Table 3), including the need to operate at highly acidic conditions, production of large amounts of iron-containing sludge, need to re-adjust the treated effluent to near-neutral conditions, heterogeneous Fenton processes are used based on transition metal catalysts (e.g. zero valent iron, iron minerals, metal oxides, etc.). (Chen et al., 2021).

Marjanovic et al. (2018) studied the enhancement of solar disinfection by the simultaneous addition of iron and PDS. They evaluated PDS activation by three individual factors (i.e. iron, heat and solar light), as well as their possible combinations. Authors reported that the abatement of MS2 bacteriophage was achieved in few minutes, when triple activation was applied (Marjanovic et al., 2018). In another study, the fate of MS2 coliphage in heterogeneous solar Fenton-like processes catalyzed by different iron (hydr)oxide particles was studied. Adsorption to goethite (α -FeOOH), magnetite (Fe₃O₄) and amorphous iron(III) hydroxide (Fe(OH)₃) was found to cause virus inactivation of 7%, 22%, and 14%, respectively. No inactivation was recorded in the absence of sunlight or H_2O_2 with the exception of magnetite, where partial MS2 inactivation occurred through a dark Fenton-like process. The findings of that study proved that heterogeneous Fenton-like processes can support viral abatement from water by physical removal, and inactivation by adsorption and a particle-mediated (photo-)Fenton-like process. (Nieto-Juarez & Kohn, 2013) Ortega-Gomez et al. (2015) also studied the inactivation of MS2 by solar photo-Fenton process, and assessed the effects of reactant concentrations (H₂O₂, Fe^{2+} , Fe^{3+}) and solar irradiance on the process. Complete MS2 inactivation (from 10⁶ PFU/mL to the detection limit)

was achieved with 1 mg/ L Fe³⁺ after 60 min of solar irradiance at 45 W/m², in the absence of H₂O₂. After the addition of 1 mg/L H₂O₂, complete inactivation was achieved after 20 min at 30 W/m². Additionally, it was observed that the process with Fe³⁺ was faster than with Fe²⁺, showing that the source of iron may be a significant inactivation parameter. MS2 and echovirus inactivation by the photo-Fenton process was also tested in natural water samples. Echovirus was more resistant than MS2 and, in either case, inactivation rates were reduced presumably due to the competition between the natural organic matter and the virus for oxidative species. (Ortega-Gomez et al., 2015).

In another study of Nieto-Juarez et al. (2010) regarding MS2 inactivation by iron- and copper catalyzed Fenton-like systems, it was found that viral reduction followed a first-order rate in relation to H_2O_2 . For the Cu/H₂O₂ system, the rate constant increased with increasing copper concentration up to 2.5 μ M, above which it reached a plateau, thus implying that only soluble copper was responsible for viral inactivation. On the other hand, inactivation in the Fe/H₂O₂ system was dictated by colloidal iron. Moreover, solar irradiation influenced only the Fe/H₂O₂ system; oxidants such as ferryl species could have also been implicated, complementing the action of hydroxyl radicals (Nieto-Juarez et al., 2010).

Giannakis et al. (2017b) investigated the application of heterogeneous photo-Fenton process for MS2 disinfection in wastewater matrices. They reported that low concentrations of iron oxides (wüstite, magnetite, maghemite) in wastewater without H₂O₂ resulted in restricted semiconductor-mediated MS2 inactivation. The dissolved organic matter was proved to be a catalyst poison since it competes with MS2 for the active sites and oxidants. A significant enhancement of the abatement process was shown after the addition of low amounts of H₂O₂, leading to heterogeneous photo-Fenton processes on the surface of the oxides. The use of nanosized iron oxides caused an increase of the active sites of the catalyst which in turn increased the active photo-Fenton sites positively affecting viral abatement. Moreover, partial photo-dissolution of iron in the bulk led to homogeneous photo-Fenton, which was supported by the complexation by the dissolved organic matter in the solution. According to the authors, the financial feasibility of such systems based on magnetically separable and reusable catalysts will affect the implementation of photo-Fenton treatment processes in combination with UVA, medium pressure UV lamps or solar light. In another study of photo-Fenton process assessment Giannakis et al. (2017c), evaluated the effect of principal parameters, such as Fe species and concentration, solar irradiance, pH and microbial competition on MS2 disinfection in wastewater. They found that ferrous materials were more effective than their ferric counterparts, resulting in faster abatement at any combination of H₂O₂ concentration,

AOP	Advantages	Disadvantages
Homogeneous photo-Fenton	Efficient and rapid oxidation and mineralization Production of reduced quantities of iron sludge com- pared to the dark Fenton reaction Low operating costs Effective at ambient temperatures Suitability for batch treatment	Need to operate at highly acidic conditions Potential effects on downstream processes due to the use of iron and acidic conditions Production of iron-containing sludge
UV/H ₂ O ₂	Efficient against most organic pollutants and for the disinfection of water matrices Fast treatment due to the production of hydroxyl radicals Proved functionality in existing full-scale treatment systems	Potential interference of compounds (e.g. nitrate) to the UV light absorbance Colour and turbidity may impede UV light penetration and decrease its effectiveness Deterioration of lamp performance at high temperatures Hydrogen peroxide is a costly chemical
Ozone-based processes	High efficiency against a variety of pollutants No sludge production Possible combination with various catalysts Use of ozone as a gas and thus no increase of treated water volume	High turbidity, particulate matter and heavy metals may negatively affect effectiveness Need for protection measures due to the use of ozone which is a highly toxic gas Potential generation of toxic by-products High capital and operating costs
Hydrodynamic cavitation	 Potential for incorporation into a continuous flow process Potential scale-up that allows for a cost-effective treatment system High potential if applied in combination with other processes (UV, ozone treatment) reaching a synergistic effect 	Immature technology The combination with other processes (UV, ozone, etc.) may result in an increase of the operating costs
Semiconductor photocatalysis	Titania is the single most suitable and low-cost mate- rial Environmentally friendly process Functionality with light of different wavelengths, including solar irradiation Possible recycle/reuse of the catalysts	Need for detailed preliminary studies on optimum cata- lyst type and concentration Requirement for catalyst separation if added as a slurry (this limitation may be faced by the hybrid photocata- lytic membrane reactor, in which photocatalysis is combined with filtration in a single step) Potential generation of harmful by-products
Photo-electro oxidation	No requirements for extra chemicals Operation at ambient conditions	Low selectivity High operating cost due to electricity consumption

Table 3 The main advantages and disadvantages of different advanced oxidation processes (AOPs)

sunlight irradiance or starting pH (6–8). Initial treatment with Fe(II) caused the presence of more free iron ions in solution compared to Fe(III), which led to higher inactivation kinetics. Even so, exposure for 30 min at 600 W/m² in the presence of Fe(III):H₂O₂ at 1:1 ratio resulted in a 4-log MS2 reduction. Finally, it was shown that MS2 inactivation was moderately reduced in the presence of the bacterial host, indicating a limited competition for the oxidants in the bulk. Although ROS scavengers were present, an effective inactivation of MS2 was recorded (Giannakis et al., 2017c).

In another study, Giannakis et al. (2018) assessed the application of UVC, UVC/H_2O_2 and photo-Fenton methods against *E. coli* and MS2 in ultrapure water, wastewater and urine. Disinfection kinetics were investigated in the presence and absence of iohexol (an iodinated contrast medium). A reduction of the viral load by approximately 4-logs was recorded for UVC and UVC/H_2O_2 treatment after 2 and 1.5 min of exposure, respectively. MS2 bacteriophage was found to cause an average 45% reduction of the bacterial inactivation in all studied matrices, and similarly, *E. coli* was

shown to delay the inactivation of MS2, except in synthetic wastewater matrices. Interestingly, the study showed that a rapid bacterial and viral inactivation was achieved even in the presence of significant quantities of iohexol, as found in hospital effluents. (Giannakis et al., 2018).

Solar Disinfection

Sunlight is known to be effective for waterborne viral abatement via both direct and indirect processes. The sunlight is absorbed by virus, but it is also absorbed by external chromophores, which subsequently generate reactive species finally attacking viral targets (Mattle et al., 2015). Photocatalysis is an attractive "green" disinfection strategy since solar energy is used and there is no need of additional oxidants. It is mainly based on semiconductors that are sensitive to the UVA and/or visible regions of the solar spectrum and, thus, the disinfection processes are based on the photoinduced electron–hole pairs on the excited semiconductor to produce different ROS (Chen et al., 2021). Numerous semiconductors have been applied but titanium dioxide (either as such or in various modified forms) has been extensively used due to its interesting characteristics (chemical inertness, high photoactivity, non-toxicity, low cost) (Brienza & Katsoyiannis, 2017).

In the study of Mattle et al. (2015) focusing on solar viral disinfection, three viral targets were used (MS2, phiX174, adenovirus) to study quantum yields of direct inactivation. Moreover, the second-order rate constants of four reactive species (OH•, ${}^{1}O_{2}$, CO₃•⁻, triplet states) implicated in indirect inactivation process were estimated. φ X174 was more sensitive to direct inactivation, with the highest quantum yield, than the other studied viruses. Second-order rate constants took values from 1.7×10^7 to 7.0×10^9 M⁻¹ s⁻¹ in the order: MS2 > adenovirus > phiX174. A predictive model was used to assess the solar disinfection of MS2 and φ X174 in a natural water sample. Viral abatement was mainly performed by direct processes, although indirect inactivation by ${}^{1}O_{2}$ also participated in the disinfection of adenovirus and MS2 (Mattle et al., 2015).

In the study of Venieri et al. (2015) Mn-, Co- and binary Mn/Co-doped TiO2 catalysts were evaluated for the abatement of MS2 in real wastewater samples under simulated and natural solar irradiation (dopant concentration: 0.02-1 mol wt%, wavelength: > 420 nm, photon flux: $4.93-5.8 \times 10-7$ E/(L s)). Disinfection was found to follow a pseudo-first-order kinetic rate. Metal-doped catalysts caused a 60% MS2 abatement in 60 min under simulated solar irradiation (for an initial MS2 concentration of 10^5 PFU/mL). Interestingly, binary Mn/Co-doped TiO2 catalysts showed the highest photocatalytic activity resulting in almost 99% MS2 disinfection in less that 20 min of irradiation, a finding which underlined the synergistic effect of composite dopants. (Venieri et al., 2015).

UV-Based AOP

Ultraviolet (UV) irradiation has been characterized by high efficiency towards microbial disinfection including viral inactivation. The most common UV sources are mercury lamps, such as low-pressure (LP) and medium pressure (MP) lamps for monochromatic UV at 254 nm, and polychromatic UV with a broad spectrum, respectively (Song et al., 2019).

The study of Song et al. (2019) focused on MS2 and *E.coli* inactivation by sequential UVA and UVC irradiation using light-emitting diodes (UV-LED). UVA pre-treatment followed by UVC treatment did not enhance MS2 inactivation, in comparison to *E.coli*, a finding which was attributed to the limited viral metabolic activities (Song et al., 2019).

Coxsackievirus B3 was found to be highly sensitive to low-pressure UV irradiation at 20 mJ/cm², while it was very resistant to peracetic acid with less than 1 log10 tissue culture infectious dose 50% assay (TCID50) reduction at acid concentrations up to 50 mg/L and a contact time of 15 min. However, the simultaneous application of 3 mg/L peracetic acid with 20 mJ/cm² irradiation caused a TCID50 decrease of approximately ~ 4-log10. Secondary wastewater effluent samples were used for that study (Kibbee & Ormeci, 2020).

UV/H₂O₂ AOP

 UV/H_2O_2 AOP based on the production of hydroxyl radicals due to the photolysis of H_2O_2 has been proved to be an efficient approach for the degradation of organic pollutants and microbial agents in water matrices. However, this AOP is less effective when the wastewater is characterized by high absorbance, while its application is restricted by high operational cost (Table 3) (Rasalingam et al., 2014).

Sherchan et al. (2014) studied the MS2 inactivation in deionized water using a small community ultraviolet light reactor (with energy-efficient, high-output amalgam lamps) combined with hydrogen peroxide. Ultraviolet radiation caused an MS2 decrease of 5.3-5.8 log10 (as assessed by plaque counts) and a 1.7-2.8 log10 decrease in viral RNA copy number (as assessed by quantitative polymerase chain reaction). MS2 abatement was increased by the addition of 2.5–5 mg/L H_2O_2 with a reduction of more than 7 log10, while qPCR showed only a 3-4-log10 reduction in viral RNA copy number, an indication that this technique overestimates the infective viruses (Sherchan et al., 2014). It is well recognized that compared to the traditional culturebased methods, sequence-specific DNA amplification strategies can significantly decrease the required time of microbiological/virological analyses of environmental samples, especially for non- or slow-culturable organisms/virus types. However it is also known that a positive PCR result does not discriminate between infective and inactivated viruses since viral nucleic acid can also be amplified even when no infective virus is present. Thus the interpretation of PCR/qPCR data regarding virus survival should be carefully performed.

A sequential UV treatment, with and without the addition of H_2O_2 , followed by free chlorine MS2 disinfection was studied by Cho et al. (2011) in organic-free water. No synergy was recorded for the treatment by UV irradiation followed by free chlorine, while increased inactivation was found when H_2O_2 was added in the primary UV disinfection step. In this case, a significant synergy was recorded since an additional 1.5-log inactivation was added to the 2 logs occurring by UV treatment and free chlorine. The synergistic effect was based on the additional inactivation achieved during the primary UV disinfection step due to the generation of hydroxyl radicals during H_2O_2 photolysis, and also on the microbial injury during the primary step which facilitated the subsequent secondary chlorine disinfection. Overall the addition of H_2O_2 in the primary step, and thus the conversion of the UV process to an AOP, could significantly enhance the efficiency of UV/free chlorine sequential disinfection processes (Cho et al., 2011). Similarly, a UV/ H₂O₂-Cl₂ integrated flow through system was assessed by Chu et al. (2012) for MS2 disinfection. A 5.78 log abatement was achieved at a flow rate at 50 L/h (corresponding to a hydraulic retention time HRT = 7 s), while a 4.49 log removal was achieved at a flow rate of 100 L/h (HRT = 36 s) (Chu et al., 2012) Mamane et al. (2007) studied the virucidal efficiency of a UV/H₂O₂ system on MS2, T4 and T7 phages, filtering out UV radiation between 200 and 295 nm in order to i) limit the effect of direct UV photolysis, and ii) isolate the effect of hydroxyl radicals. Authors reported that bacteriophages T4 in phosphate buffered saline were sensitive to irradiation > 295 nm in the absence of H_2O_2 , nonetheless, MS2 was very resistant. Overall, sensitivity decreased in the order: T4 > T7 > MS2, with MS2 suffering no inactivation at wavelengths > 295 nm. Interestingly, the addition of 25 mg/L H₂O₂ under UV irradiation for 15 min resulted in 2.5-logs of MS2 inactivation, had a small additional effect on T7 inactivation (of 1 log), but it did not result in any additional T4 disinfection (Mamane et al., 2007). To investigate dye-virus interactions, Timchak and Gitis (2012) used various combinations of phages (MS2, φ X174 and T4) and fluorescent dyes (rhodamine B and fluorescein) to study inactivation and degradation processes. Authors found that viral abatement was not affected by the presence of dyes but the addition of 0.2 M H₂O₂ at 70 mJ/cm² UV dose enhanced MS2 abatement by two logs; however, peroxide addition was found to have no effect on $\varphi X174$ and T4 phages. The presence of viruses caused the reduction of the bleaching of fluorescent dyes presumably due to restricted availability of hydroxyl radicals and their preferential involvement in virus inactivation (Timchak & Gitis, 2012). In another study, Bounty et al. (2012) focused on the improvement of UV adenovirus abatement with the addition of hydrogen peroxide. A UV dose of approximately 200 mJ/cm² from a low-pressure source emitting at 253.7 nm was needed for a 4-log reduction of adenovirus without peroxide. The addition of 10 mg/L H₂O₂ was capable of achieving a 4-log viral abatement at a reduced dose of 120 mJ/cm² (Bounty et al., 2012). Virus-bound rhodamine B was employed as an indicator of advanced disinfection in the study of Shabat-Hadas et al. (2017), who evaluated direct UV photolysis, UV/H₂O₂ and solar light-induced photocatalysis using fluorescencelabelled MS2 and T4 phages (Shabat-Hadas et al., 2017).

A schematic representation of the UV/H2O2 AOP effects on viral targets is shown in Fig. 3.

Ozone-Based AOP

Ozone is a powerful oxidizing agent that can effectively degrade different organic pollutants, as well as disinfect

microorganisms, either by direct electrophilic attack or indirectly through its reaction with water and the subsequent formation of hydroxyl radicals (Rasalingam et al., 2014). Different parameters may affect treatment efficiency (e.g. O_3 concentration, treatment duration, water temperature, pH, scavengers, etc.). Homogeneous and heterogeneous catalytic ozonation processes are applied for disinfection purposes, employing transition metal ions or solid catalysts (activated carbon, metal oxides, etc.), respectively. (Chen et al., 2021). Ozone-based AOPs are characterized by elevated capital and operating costs, and high electric power consumption besides other drawbacks, such as low water solubility of ozone and formation of hazardous by-products (Table 3) (Brienza & Katsoyiannis, 2017; Rasalingam et al., 2014).

King et al. (2020) assessed ozone treatment of pathogens and pollutants in reverse osmosis (RO) concentrates produced during the potable reuse of municipal wastewaters. Approximately 5-log removal was observed at $0.30 \text{ mg O}_3/$ mg of dissolved organic carbon (DOC) when $10^6 - 10^7$ PFU/ mL of MS2 were spiked into RO concentrate samples from facilities where nitrite was not measurable. Samples from facilities, which contained nitrite, required higher O₃ doses to achieve 5-log inactivation of MS2. However, ozonation led to a 5-log abatement of MS2 in all concentrate samples at 1.18 mg O₃/mg of DOC. Due to its higher selectivity, ozone may be a more effective oxidant for controlling pathogens in RO concentrates compared to other AOPs which are based only on hydroxyl radicals, given that the contact time is sufficient before its rapid degradation and partial conversion to hydroxyl radicals (King et al., 2020) Gamage et al. (2013) assessed ozone and ozone/H₂O₂ for MS2 inactivation in secondary wastewater effluents. Time-integrated dissolved ozone concentrations of 1 mg·L⁻¹·min caused MS2 inactivation higher than 6 logs. Various process control indicators, i.e. ozone to DOC ratio, differential UV₂₅₄ absorbance and differential total fluorescence were also assessed in the context of this study and they were found useful for predicting the inactivation of MS2 (Gamage et al., 2013). To assess the disinfection efficiency of ozone and hydrogen peroxide treatment at a pilot plant, different phages (MS2, φ X174, and PRD-1) were used in the study of Sommer et al. (2004). The ozone/hydrogen peroxide process was found to have a significant microbicidal effect. MS2 bacteriophage and phages φX174 and PRD-1 showed a reduction of approximately 6 logs. These findings corresponded to ozone disinfection in controlled batch experiments (residual 0.4 mg/L after 4 min contact time, 20°C) (Sommer et al., 2004). In another study, catalytic ozonation with a volcanic rock led to complete abatement of all target viruses (norovirus genotype I, II, and JC virus) from secondary municipal wastewater, while JC polyomavirus could not be eliminated even after 150 min of treatment by non-catalytic ozonation (Gomes et al., 2019).



Hydrodynamic Cavitation

Sun et al. (2020) have summarized the fundamental principles of hydrodynamic cavitation (HC) and the recent progress achieved in HC disinfection. HC is a promising emerging technology for large-scale disinfection without the need of using chemicals. Interestingly, it may generate extreme conditions (pressures of ~ 1000 bar, local hotspots of ~ 5000 K, high oxidation—hydroxyl radicals) which in turn can be highly destructive for microorganisms in water matrices. Kosel et al. (2017) studied the effect of hydrodynamic cavitation on MS2 infectivity. Reductions of viral infectivity higher than 4-logs were recorded using small scale reactors. Authors underlined the viral disinfection efficiency of wastewater and discussed the potential of using cavitation generators for large-scale applications (Kosel et al., 2017).

Photocatalytic Membrane Reactor

When an AOP such as photocatalysis is combined with membrane filtration, high quality water may be produced by the combination of filtration/photocatalytic inactivation, in a single step treatment Horovitz et al. (2018) assessed MS2 inactivation by a hybrid photocatalytic membrane reactor (PMR), which included a photocatalyst immobilized onto a membrane substrate. Authors used water of different quality to investigate the viral removal efficiency of a N-doped TiO_2 -coated Al_2O_3 PMR and they recorded a viral abatement of $4.9 \pm 0.1 \log (> 99.99\%)$ in natural surface water under irradiation. Complex virus–PMR interactions were found to occur even before the exposure to light. Electrostatic forces, in addition to photocatalytic inactivation, were also responsible for viral abatement in complex matrices. Although alkaline water pH reduced interactions and, consequently, the extent of viral abatement, the addition of Ca²⁺ had a beneficial effect (Horovitz et al., 2018).

Photo-Electro-Oxidation

The main advantage of photo-electro-oxidation processes is that they do not require the use of additional chemicals and/ or disinfectants, while they operate at ambient conditions of pressure and temperature (Table 3). On the other hand, they are frequently characterized by low selectivity and reaction rates. Their application may be restricted by high operational costs associated with the use of UV irradiation and electric power (Monteiro et al., 2015; Rasalingam et al., 2014).

A 60-min treatment by the photo-electro oxidation process was capable of decreasing human adenovirus type 5 (HAdV-5) by 7 log10 in no DNAse-treated samples. Exposure for 75 min was required for the complete abatement of DNAse-treated samples used to study intact viruses only. Completely non-viable adenoviruses were obtained after 30 min, as was evidenced by integrated cell culture quantitative polymerase chain reaction (ICC/qPCR) analysis (Monteiro et al., 2015).

Commercial Installations

Different commercial installations involving 1) the use of a membrane bioreactor and a full advanced treatment (FAT) train, 2) an advanced Bardenpho process, 3) an advanced oxidation-based Net-zero water (NZW) pilot system, and 4) a full-scale direct potable reuse demonstration facility are summarized below.

Papp et al. (2020) studied water reuse facilities using different microbial surrogates such as bacteroides bacteriophage ϕ B124-14 and ϕ crAssphage, as well as the pepper mild mottle virus (PMMoV). PMMoV, \u03c6B124-14 and *\phi*crAssphage were detected in membrane bioreactor (MBR) feeds at concentrations of approximately 10^3 , 10^5 and 10⁶ gene copies/mL, respectively. Only PMMoV was detected above the limit of quantification in the MBR filtrate $(25 \pm 8 \text{ gene copies/mL})$. Viral log removal values were found to be 1.4 ± 0.5 for PMMoV, > 3.9 ± 0.3 for $\phi B124$ -14 and > 6.2 ± 0.3 for ϕ crAssphage (Papp et al., 2020). The study of Schmitz et al. (2016) compared the advanced Bardenpho to conventional treatment processes by assessing the abatement of eleven different types of viruses. The Bardenpho process is a multiple stage biological nutrient removal process without any addition of chemicals and in the study of Schmitz et al. (2016) was composed of serial compartments (anaerobic, anoxic, oxic, anoxic, and oxic). Interestingly, the advanced Bardenpho wastewater treatment was proved to be more efficient than conventional treatments in reducing pathogenic viruses. The highest mean reduction values were recorded for nine target virus types, and also the highest rate of removal to concentrations below the detection limit was achieved. Mean reduction of viruses as calculated by the mean difference between influent and effluent sample sets were > 2.7 ± 1.6 , > 2.7 ± 0.9 , > 1.7 ± 1.4 , > 2.6 ± 0.6 , $> 3.6 \pm 0.4$, $> 3.0 \pm 0.6$, $> 2.5 \pm 0.5$, $> 1.7 \pm 1.1$, > 3.3 ± 1.2 , $> 3.1 \pm 0.5$, $> 3.4 \pm 1.1$ for pepper mild mottle virus (PMMoV), Aichi virus (AiV), genogroup I, II, and IV noroviruses (GI NoV, GII NoV, GIV NoV), enterovirus (EV), sapovirus (SaV), group-A rotavirus (ARV), adenovirus (AdV), JC and BK polyomaviruses (JCPyV and BKPyV), respectively. Following the advanced Bardenpho treatment all viruses except PMMoV were found at concentrations of $< 4.0 \log_{10}$ copies/L. Aichi virus was found to be a conservative index virus for the assessment of viral removal in wastewater treatment. Bardenpho processes were the major sources of virus removal probably because of virus sorption to solids (Schmitz et al., 2016).

Vaidya et al. (2019) compared the performance of two pilot-scale, advanced water treatment plants with different

treatment processes, a carbon-based and a membrane-based. The carbon-based treatment process involved flocculation/ sedimentation, ozone-biofiltration, granular activated carbon (GAC) adsorption, and ultraviolet (UV) disinfection. The membrane-based treatment process was composed of ultrafiltration, reverse osmosis, and UV-hydrogen peroxide advanced oxidation steps. In both water treatment processes, a > 8-log removal of MS2 and > 6-log removal of Pepper mild mottle virus was achieved (Vaidya et al., 2019). An advanced oxidation-based net-zero water (NZW) pilot system was evaluated in the study of Gassie et al. (2016). The system consisted of a septic tank, membrane bioreactor (MBR), aluminum electrocoagulation (EC), flocculation, vacuum ultrafiltration, peroxone or UV-hydrogen peroxide advanced oxidation, chlorine disinfection, and point-of use granular activated carbon (GAC) filtration. No viruses (somatic, male-specific coliphage and adenovirus) were found in the treated water, although adenovirus genetic material was detected probably due to the presence of inactive viral particles in hydraulic dead zones (Gassie et al., 2016) Pecson et al. (2017) assessed the efficiency of a fullscale, direct potable reuse demonstration facility. The treatment train included ozonation, biological activated carbon, micro- or ultrafiltration, reverse osmosis, and a UV-based advanced oxidation process. Performance distribution functions evidenced treatment that consistently surpassed the 12-log thresholds for virus, as required for potable reuse in California (Pecson et al., 2017).

Table 1 summarizes the studies which have used only MS2 bacteriophage as the index virus, while Table 2 summarizes the studies which have assessed more than one viruses.

Future Aspects

Although constructed wetlands, stabilization ponds and membrane filtration have traditionally been employed for microbial abatement, the use of AOPs as tertiary or disinfection processes capable of dealing with resistant microbial targets, including viruses, has received less attention (Meric & Fatta Kassinos, 2009). The increasing application of AOPs has been recognized as one of the most promising technological strategies for efficient water treatment, as well as the safe discharge or potential reuse of wastewater effluents. Specifically, photo-Fenton processes have been proven to be effective for the treatment of secondary effluents and potable water and are also efficient against micropollutants and different microbial targets (Galeano et al., 2019; Giannakis et al., 2016, 2017a; Gogate & Pandit, 2004). Of course, AOPs suffer from certain limitations, including the production of significant quantities of ferrous sludge associated with Fenton and alike processes, costly chemicals such as

hydrogen peroxide and ozone, elevated installation and operating costs for UV/ozone processes, etc. (Srivastav et al., 2020). Interestingly, such limitations can partly be counterbalanced through process integration, involving the coupling of various AOPs either together or to other physical and/ or biological processes. For instance, the synergy recorded when two or more AOPs are combined is mainly due to the enhancement of free radicals production, which in turn is responsible for increased oxidation rates, but also to the modification of the reactor conditions and/or configuration (Gogate & Pandit, 2004). Although viral abatement in water and wastewater matrices is a tricky task, the application of advanced water treatment trains is expected to support the efficient disinfection goals. The adoption of a multi-barrier techniques' approach is expected to reduce the risks due to the prevalence of viruses for both the environment and public health (Ghernaout et al., 2020).

To promote the application of AOPs in real world scenarios, issues such as i) the investigation of metabolites generated during the process, ii) the examination of biological safety linked to cytotoxic and/or genotoxic effects, and iii) the execution of scaling-up and economic feasibility studies, have to be taken into account (Galeano et al., 2019). AOPs are expected to find broader full-scale applications, according to the trend which is recorded during the last decade. Most probably, diverse technological solutions will be applied locally, according to the specific prevailing conditions (e.g. solar driven processes will be adopted in areas with high solar irradiation levels) (Kokkinos et al., 2020). Omics tools (high throughput sequencing, metagenomics, proteomics, transcriptomics, etc.), are expected to further support the exploitation of the viral diversity in water matrices and also to elucidate the mechanisms of disinfection achieved by treatment methods such as AOPs. They will also expand the research from culturable pathogens and indicators to whole microbial communities (Nelson et al., 2018).

To compare fairly the disinfection potential of various AOPs with particular focus on the virological quality of water matrices, energy consumption and treatment cost, it would be interesting to link the EEO values with the viral load. This could provide a useful tool for the selection of the most effective processes for viral abatement with the higher potential for full-scale applications according to energy and operational cost.

The current Coronavirus Disease 2019 (COVID-19) pandemic has stressed out, among others, the need to better understand wastewater as potential source of epidemiological data, environmental and human health risks, and to effectively disinfect wastewater to reduce the risks to the public and the environment (Carraturo, 2020; Kitajima et al., 2020; Wang, 2020). Limited research data are available on endogenous and exogenous reaction rates of enveloped viruses (Nelson et al., 2018). The implications of SARS-CoV-2 prevalence in water and wastewater matrices have to be clarified, along with its fate during different treatment processes. According to Ghernaout et al. (2020), the combination of UV irradiation with membrane ultrafiltration will successfully remove coronaviruses from water matrices (Ghernaout et al., 2020). The role of AOPs, alone or in combination with other processes in wastewater and drinking water treatment plants in the event of future pandemics may be crucial towards safeguarding public and environmental health.

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