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An overview on ultraviolet persulfate based advances oxidation process for removal of antibiotics from aqueous solutions: a systematic review

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ABSTRACT

Pollution of water resources with pharmaceuticals along with the failure of conventional water treatment methods to remove resistant pollutants increased the attention to advanced water treatment techniques. The present study reviewed the literature of ultraviolet persulfate based advanced oxidation process (AOP) for the removal of antibiotics. Results showed that the removal of antibiotics in UV/ PS systems was pH dependent and the sulfate radicals were dominant in the neutral situation. Also, complete degradation of Ciprofloxacin (CIP) occurred at pH 7 in 60-min reaction time at the peroxymonosulfate [PMS]/[CIP] molar ratio of 20 in wastewater medium. Analyzing the antibiotic degradation pathway in UV/PS system suggested that regions with the highest electron density values were most susceptible to react with reactive oxygen species from which decomposition began. Finally, based on the literature review, the removal of antibiotics needs to be considered by UV/PS processes in the real aqueous medium. It is also necessary to study the scavenging effect of mineral scavengers, as well as the study of organic scavengers can provide significant information in this area. Both toxicity test and determining the bacterial resistance of new degradation compounds can be useful to clarify the degradation pathway.

Keywords: Antibiotic; Aqueous; Advances oxidation process; Persulfate technology

1. Background

Antibiotic are prescribed to kill bacteria and other microscopic pathogens. They help the body to cope with illnesses; hence, they are used to treat many diseases. The presence of antibiotic compounds in ecosystems became a concern since the mid1990s when these compounds were widely used. [1]. Given that the main activity of antibiotic compounds is against bacteria, the presence of these compounds causes growth inhibition or disappearance of species of microorganisms that play a key role in ecosystems [2,3].

Also, studies warned that constant contact of microorganisms with antibiotics promotes highly resistant microbial species, which could be a potential hazard to human health [4]. However, the simultaneous presence of several antibiotics can develop the synergistic effects and increase the range and severity of such effects [2,5]. Antibiotic compounds are present in all environmental compartments including sewage, water, sediment and soil [6–8]. Due to the attachment of antibiotics to suspended solids, biological degradation of antibiotics is very slow in water and soil environments, and the probability to sediment evaporation is very small [9,10]. Presence of these compounds has been reported from several days to several months in aquatic environments, sediments and soil [6]. Antibiotics affect the environment based on their chemical structures by various potentially toxic properties such as the effect on algae and microorganisms in environments [11]. Some common classes of antibiotics based on chemical or molecular structures include beta-lactams, macrolides, tetracyclines, quinolones, aminoglycosides, sulphonamides, glycopeptides and oxazolidinones. Table 1 presents general characteristics and structures of these classes as well as well-known antibiotics in each group [12].

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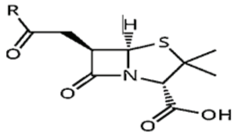
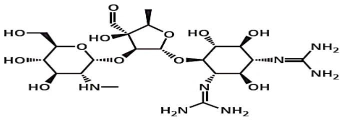
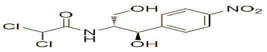
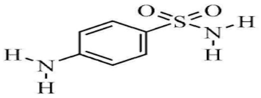
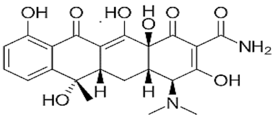
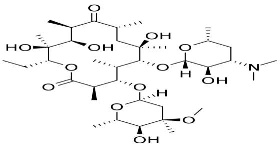
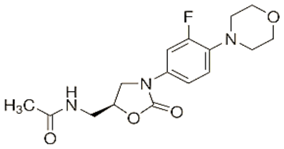
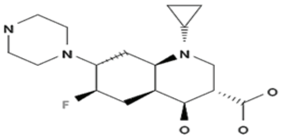
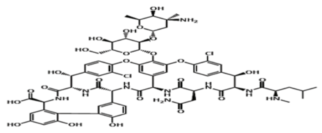
Table 1
Classifiction of antibiotics

Classes of antibiotics	General structure characteristics	Examples	3D Structure	References
β-Lactams	*All contain a beta-lactam ring*	Penicillins Cephalosporins Monobactams Carbapenems	 Penicillin core	[13]
Aminoglycosides	*All contain aminosugar substructures.* The aminoglycosides are compounds of usually 3-amino sugars connected by glycosidic bonds	Streptomycin Neomycin Kanamycin Paromomycin	 Streptomycin	[14]
Chloramphenicol	*Distinct individual compound*	Chloramphenicol	 Chloramphenicol	[15]
Sulfonamides	*All contain the sulfonamide group*	Prontosil, Sulfanilamide Sulfadiazine Sulfisoxazole	 Sulfanilamide	[12]
Tetracyclines	*All contain 4 adjacent cyclic hydrocarbon rings*	Tetracycline Chlortetecycline Oxytetracycline Demeclocycline	 Tetracycline	[16]
Macrolides	*All contain a 14-, 15-, or 16-membered macrolide ring*	Erythromycin Azithromycin Clarithromycin	 Erythromycin	[17]
Oxazolidinones	*All contain 2-oxazolidone somewhere in their structure*	Linezolid Posizolid Tedizolid Cycloserine	 Linezolid	[18]
Quinolones	*All contain fused aromatic rings with a carboxylic acid group attached*	Ciprofloxacin Cinoxacin, Norfloxacin, Ofloxacin Temafloxacin Sparfloxacin, Nalidixic acidEnoxacin	 Ciprofloxacin	[19]
Glicopeptides	*Consist of carbohydrate linked to a peptide formed of amino acids.* Naturally, glycopeptides are made of a cyclic peptide of 7 amino acids, to which are bound 2 sugars, hence the name glycopeptides	Vancomycin Teicoplanin	 Vancomycin	[20]

Due to the undesirable effects caused by antibiotics, removal of these compounds and preventing their entrance into the environment seems to be necessary. The first source allowing antibiotics to enter the environment is effluent of wastewater treatment plants [21].

Since conventional wastewater treatment systems are not primarily designed to deal with drug contaminants, the presence of these toxic and resistant compounds in water resources limits the use of conventional water and wastewater treatment processes for the removal of these resistant compounds [22,23]. Therefore, in recent years, water treatment technologies exploring methods that are capable to remove these recalcitrant. In the meantime, advanced oxidation processes are considered a lot due to their high efficiency and, also lack of creating secondary contamination and removal of various types of pollutants [24,25].

Advanced oxidation in a broad sense is the set of the chemical processes in which the chemical structure of the contaminants is broken down by production of free radicals such as hydroxyl, hydroperoxyl, and superoxide which have high oxidative properties [24,26,27].

The main feature of advanced oxidation processes is the use of hydroxyl radical which is known as a strong and non-selective oxidizing agent for most organic compounds [28,29]. In some cases, higher selective advanced oxidation processes which are based on sulfate radical generation can be appropriate for removal of resistant contaminants. Persulfate based advanced oxidation involves the production of highly reactive hydroxyl and sulfate radicals which have the relative selectivity for oxidation [30].

Persulfate can be activated via heat, transition metals, ultraviolet (UV) light, etc., and forming the highly reactive sulfate radical with high oxidation potential varies between +2.5 and +3.1 V [21,31]. Sulfate activation methods using thermal and ultraviolet light, ultrasound (US) electrolysis and metal ions are shown in the following equations [32–34].

$$S_2O_8^{2-} + \text{heat} / \text{UV} \rightarrow SO_4^{\cdot-} \tag{1}$$

$$S_2O_8^{2-} + e^- \rightarrow SO_4^{2-} + SO_4^{\cdot-} \tag{2}$$

$$S_2O_8^{2-} + M^{n+} \rightarrow SO_4^{2-} + SO_4^{\cdot-} + M^{(n+1)+} \tag{3}$$

$$S_2O_8^{2-} + \text{US} \rightarrow SO_4^{\cdot-} \tag{4}$$

Because of the disadvantages of other activation methods such as more sludge production, metal ion removal difficulties, high energy consumption and due to gentle reaction conditions in ultraviolet activation method, this activation method has attracted a *great deal* of interest [35]. As selectivity of sulfate radical toward some target species is stronger than hydroxyl radicals, oxidation based on sulfate radical generation may offer advantages over hydroxyl based oxidation [21]. Sulfate radical is effective in the wide ranges of pHs, less consumed by organic compounds and also is more stable and soluble in aqueous media compared to hydroxyl radical [36,37]. For the above-mentioned reasons, persulfate based advanced oxidation process is used to remove a large number of pollutants, including dyes [38,39], Pharmaceuticals [40–43], Bisphenol S (BPS) [44] and other resistant compounds [45,46]. Therefore in the present literature review, we investigated many studies conducted on the topic and different processes based on ultraviolet persulfate oxidation that were used to remove antibiotics from aqueous solution. This review aimed at gathering the related results of ultraviolet persulfate based AOP studies, determining the most appropriate experimental condition including pH, time, PS concentration and also the most studied antibiotics and remaining drawbacks in the field of using UV/PS based AOP for degradation of antibiotics. Also, the experimental media were considered to clarify the effect of experimental media on persulfate based AOP efficiency.

2. Method

This systematic review aimed to pursue the ultraviolet persulfate based advances oxidation process for removal of antibiotics in aqueous solutions. In this regard, relevant studies were retrieved from international databases of Scopus, Science Direct, Web of sciences, PubMed that explored from 2000 until September 2018. The search strategy was developed using key words such as Antibiotic, advances oxidation process, ultraviolet, persulfate, removal, aqueous in titles, abstracts and keywords. To increase the search sensitivity and providing higher records, additional records were identified by hand searching and review of the referenced list of retrieved papers. Search evaluation was conducted randomly by one of the authors who had no role in the search. Titles, abstracts or the full text of all articles, documents, and reports admitted to the study were extracted and the duplicates were removed. Subsequently, the remained articles were carefully reviewed and related articles were selected for further study. All the studies that investigated the removal of antibiotics by ultraviolet activated persulfate advanced oxidation process were captured and included in this systematic review. Fig. 1 shows the search and analyzing procedures [47,48].

Most studies compared the removal of the antibiotics using sole-UV, persulfate without activation and UV-persulfate. It was reported that antibiotics removal by UV/PS system was significantly stronger than UV and persulfate without activation [49,50,53,54,56,59,60]. However, evaluation of UV and persulfate for removal of ampicillin, oxacillin, cloxacillin, cephalexin, cefadroxil, levofloxacin, norfloxacin, ciprofloxacin by Serna-Galvis revealed that UV light degraded ciprofloxacin (~42%), norfloxacin (25%), levofloxacin (1%), oxacillin (88%), cloxacillin (60%), Ampicillin (15%) after 10 min; meanwhile, the removals of cephalexin and cefadroxil achieved 84% and 58%, respectively. It was shown that less than 3% of antibiotics were removed using persulfate after 10 min. Based on this study, persulfate showed low participation in antibiotics degradation and higher antibiotics removal rates were observed for UV/PS compared to UV_{254} and persulfate without activation [50]. Also, Lin et al. reported that no ciprofloxacin was degraded using $S_2O_8^{2-}$ and photolysis of ciprofloxacin was not significant under UV irradiation after 30 min [49].

According to Table 2, the removal of Ciprofloxacin was more considered than other antibiotics. Ciprofloxacin is used to treat a number of bacterial infections. Ciprofloxacin is listed by the World Health Organization as an essential

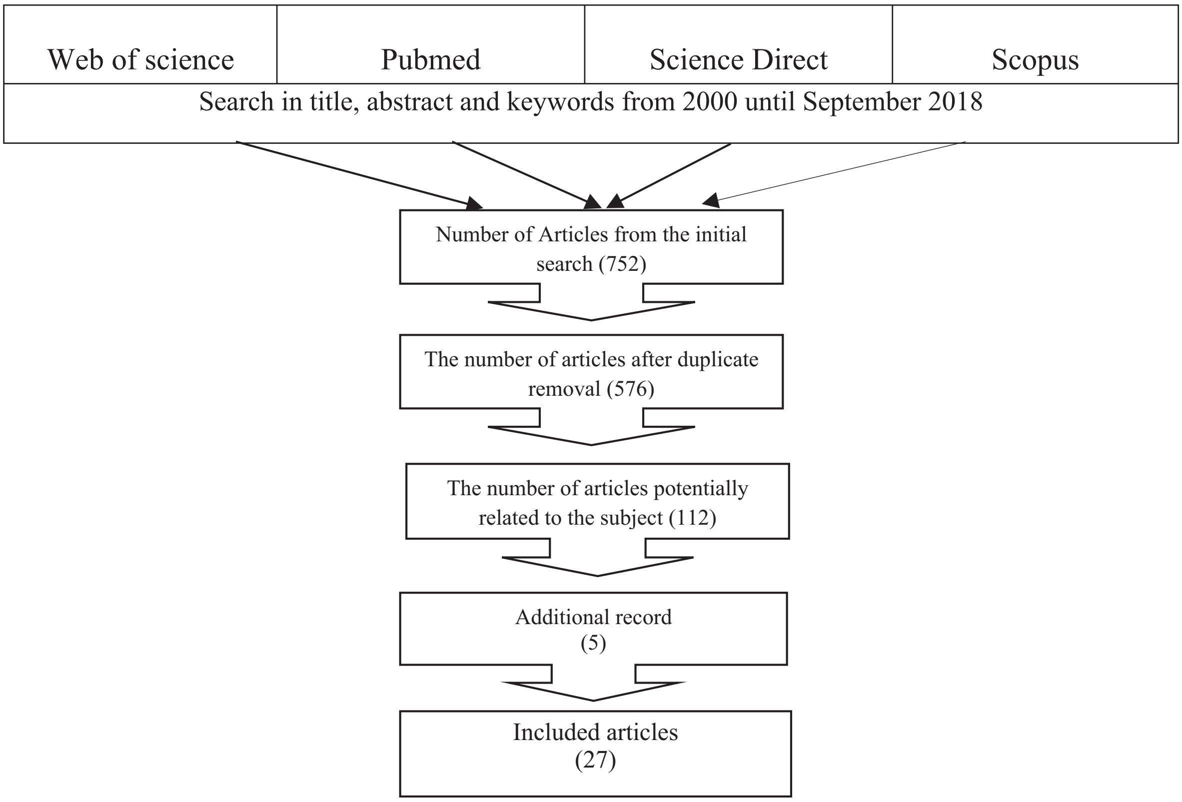


Fig. 1. PRISMA flow diagram showing flow of citations reviewed.

medicine and also it is the most effective and safe medicines needed in a health system which is not very expensive. As shown in Table 1, Ciprofloxacin is a broad-spectrum antibiotic in the fluoroquinolone class. These antibiotics are not completely removed at conventional WWTPs, so their continuous introduction into the environment makes them ‘pseudo-persistent’ compounds which is necessary to be treated from the environmental media [76]. Comparing the results of studies revealed that the ciprofloxacin was completely degraded in wastewater at pH 7 in 60 min at a [PMS]/[CIP] molar ratio of 20 [51]. In another study, the efficiency of degradation of ciprofloxacin was reported 95% using a $Na_2S_2O_8$ concentration of 1.92 g/L within 30 min [49]. Also, it was shown that UV_{280}/PS treatment induced a moderate degradation of Ciprofloxacin with high transformation efficiency and low mineralization [53]. Besides the effect of both experimental parameters and aqueous matrix, the power of the UV lamp and its intensity were influential and the best results obtained using UV_{254} 15 W that shows ciprofloxacin removal is also dependent to the power of UV light. However, it is essential to consider the possibility of using a high-power UV lamp in real conditions with larger quantities of water or wastewater. Next to the quinolones, the sulfonamide antibiotics group was studied more than other antibiotics. Based on studies presented in Table 2, sulfonamides can be eliminated efficiently in UV/PS system [23,56,58,59,64,71,74]. As soil sorption of sulfonamides is low (the soil partitioning coefficient values are 0.6–7.4l kg^{-1}), they spread rapidly in the environment. Also, sulfonamides are not easily adsorbed by activated carbon and are classified as photo and thermally stable substances with the degradation half-life of DT50 > 1 y [77]. Therefore, with the mentioned properties of sulfonamides, the effective elimination of these compounds using advanced oxidation methods is of interest. Other results from Table 2 revealed that few studies were conducted using real wastewater media, 4 studies used natural water including surface water or drinking water as experimental media and all other studies prepared a synthetic solution by distilled water. The results of studies showed that the composition of aqueous matrix affected the antibiotics removal efficiency, e.g. Erythromycin degradation rate decreased in the order of ultrapure water (k_{app}¼ 0.55 min^{-1}) > bottled water (k_{app}¼ 0.26 min^{-1}) > humic acid solution (k_{app}¼ 0.05 min^{-1}) > wastewater effluents (k_{app}¼ 0.03 min^{-1}). It should be mentioned that the anionic spices of water affect the antibiotics removal and the main inhibitory effect was seen for the carbonated ones and only chloride anions showed different behavior in radical scavenging. Chloride may accelerate the removal efficiency by reaction with SO_4^{-} in the UV/PS process to form the chlorine radicals (Cl·) which are strong oxidants (E_o(Cl·/Cl$^-$) = 2.4 V) or inhibit the process as SO_4^{-} reacts with Cl$^-$ much faster than the organic compounds that decrease the SO_4^{-} reacting with a certain organic compound [78].

2.1. The effect of persulfate concentration on antibiotics degradation

Antibiotics degradation improved with the increase of $Na_2S_2O_8$ concentration. Guo et al. reported that an increase of PS dosages significantly promoted the degradation of flu-

Table 2
Review of conducted studies in the field of removing antibiotics using ultraviolet activated persulfate process

Author	AOP removal method	Antibiotic	media	Experimental condition	Results	Highlights
[49]	UV_{254}/PDS	Ciprofloxacin (CIP)	Synthetic solution	UV lamp: 8 W (1 mW/cm^2) No adjusted pH PS: 0.24 to 3.84 g/L CIP: 10 mg/L	An efficiency of degradation of ciprofloxacin was 95% within 30 min using an $Na_2S_2O_8$ concentration of 1.92 g/L in the absence of inorganic anions.	The dominant generated free radical in the UV/$S_2O_8^{2-}$ process was $SO_4^{\bullet-}$.
[50]	UV_{254}/PDS	Ampicillin, Oxacillin, Cloxacillin, Cephalexin, Cephadroxyl, Levofloxacin, Norfloxacin, Ciprofloxacin	Synthetic solution simulated hospital wastewater	UV lamp: 60 W (398 µW cm^{-2}) Initial pH = 6.5 Antibiotics: 40 µmol L^{-1} PS: 500 µmol L^{-1}	Direct photolysis caused degradation of oxacillin and cephalexin, while reactive oxygen species and photolysis were involved in ciprofloxacin removal.	Thioether, β-lactam and piperazyl were the most reactive moieties to ROS and $SO_4^{\bullet-}$.
[51]	UV_{254}/PDS UV_{254} /PMS UV/H_2O_2	Ciprofloxacin	Synthetic solution Waste water	UV lamp: 15 W CIP = 50 µ M H_2O_2 = PDS = PMS = 1 mM pH = 3–8	The most efficient process was UV/PMS followed by UV/PDS and UV/H_2O_2. CIP was fully degraded in wastewater at pH 7 in 60 min for a [PMS]/[CIP] molar ratio of 20.	Sulfate radical anion attacks prompted transformations at the piper-azinyl ring through a one electron oxidation mechanism as a major pathway while hydroxyl radical attacks were mainly responsible for quinolone moiety transformations as a minor pathway. Sulfate radical generation allows for CIP antibacterial activity elimination
[52]	UV_{254}/PDS	Norfloxacin Enrofloxacin	Synthetic solution	UV lamp: 11 W pH = 3–11 Persulfate concentration: (0.05 mM–0.5 mM) Antibiotics concentration: (0.0026–0.052 mM)	It was demonstrated that the UV/persulfate process has a great effect on the mineralization (up to 61.2–62.5%) and toxicity control (14.6–32.6%) for the fluoroquinolone (FQs) antibiotics.	Degradation mainly occurs on the C–F bond, carboxylic acid and piperazine group. Toxicity of FQs and related products can be controlled by UV/PS process
[53]	UV_{280}/PDS	Ciprofloxacin	Synthetic solution	UV lamp: 0.023 mW cm^{-2} CIP: 1 mg L^{-1} (3 mM). PS: 10–200 mg L^{-1} pH 6.8–7.2	The 280 nm UV/PS treatment induced a moderate degradation of CIP, which had high transformation efficiency and a low mineralization.	Moderate UV/PS treatment can reduce the antibacterial activity of CIP by incomplete mineralization.
[54]	UV_{254}/PDS	Oxytetracycline(OTC)	Synthetic solution	UV lamp: 15 W, Fluence rate : 0.1 mW cm^{-2} OTC: 10 M PS: 0–2 mM pH = 2–11.5	This study suggests that UV_{254}/PS is a promising treatment technology for the control of water pollution caused by emerging contaminants such as OTC. The most effective destruction of OTC by UV/PS was achieved at near neutral pH.	Presence of HCO^{3-} or Cu^{2+} enhanced the removal of OTC in UV/PS system. Total organic carbon was degraded slowly during the mineralization of OTC. Four different reaction pathways by $SO4^{\bullet-}$reaction including hydroxylation (+16 Da), demethylation (–14 Da), decarbonylation (–28 Da) and dehydration (–18 Da)

(*Continued*)

Table 2 (*Continued*)
Review of conducted studies in the field of removing antibiotics using ultraviolet activated persulfate process

[55]	UV_{254} UV_{254}/H_2O_2 UV_{254}/PDS	Sulfamethoxazole (SMX)	Synthetic solution filtrated waters from two drinking water plants using surface water and ground water	UV lamp: 10 W pH = 3–8 SMX: 20 mM, H_2O_2: 1 Mm PDS: 1 mM.	Due to the electrophilic nature of $SO_4^{•-}$ it shows the higher reactivity towards the anionic form of SMX than the neutral form of SMX. While, $OH^{•-}$ exhibited the comparable reactivity to SMX species	Based on ecotoxicity of transformation products, oxidation of $SO_4^{•-}$ or CO^-_3 with SMX generated more toxic products than those of $OH^{•-}$
[56]	UV_{254}/PMS	Sulfonamides (SA)	Synthetic solution Drinking water	UV lamp: 15 W PS: 0.5–5.0 mg·L^{-1} Each SA: 0.2–2.0 µg·L^{-1} pH = 7.5	The order of SA degradation rates and efficiencies is UV/PMS > UV > PMS. In UV/PMS at an initial PMS concentration of 1.0 mg·L^{-1}, >98.5% decomposition occurs within the first minute, and removal is complete after 5 min. UV/PMS provided more thorough elimination of sulfonamides, as demonstrated by the complete removal of 200 ng L^{-1} of all sulfonamides within 5 min at an initial PMS concentration of 1.0 mg·L^{-1}.	Scavenging experiments showed that sulfate radical ($SO_4^{•-}$) was the predominant species involved in the degradation.
[57]	UV_{254}/H_2O_2 UV_{254}/PDS	β-lactam antibiotics (penicillins, ampicillin,penicillin V, and piperacillin; cephalosporin: cephalothin)	Synthetic solution	UV lamp:15 W, fluence rate: 0.1 mW cm^{-2} PS: 1 mM Antibiotics: 50 µM	This study suggests that UV/H_2O_2 and $UV/S_2O_8^{2-}$ advanced oxidation processes (AOPs) are capable of degrading - β lactam antibiotics decreasing consequently the antibiotic activity of treated waters	The steady-state radical concentrations were estimated, revealing a non-negligible contribution of hydroxyl radicals in the $UV/S_2O_8^{2-}$ system. $HO^{•-}$ reacted faster with the-β lactams than $SO_4^{•-}$ but sharing similar by products.
[58]	UV_{254}/PDS	Sulfamethazine (SMT)	Synthetic solution	UV lamp: 15 W pH = 3–11 SMT: 0.02 mM, PS: 0–0.5 mM	The removal efficiencies appeared to be influenced by such factors as persulfate doses and solution pH. Under the experimental condition, the SMT degradation exhibited a pseudo-first-order reaction pattern. After 45 min of irradiation, SO_4^{2-} in water reached 0.41 mM, corresponding to a 97.5% persulfate factor, thus almost all the persulfate eventually transformed to SO_4^{2-}	Both the highest SMT degradation rate and the lowest mineralization degree were observed at pH 6.5, while the highest mineralization extent was accomplished at pH 11
[59]	UV/PMS UV/PDS UV/H_2O_2	Sulfamethoxazole (SMX)	Synthetic solution	UV lamp: 2.8 kW Medium pressure lamps emitting various UV light outputs ranging from 200 nm to 300 nm. SMX: 5.92–35.54 µM; PMS = PDS = H_2O_2 = 0.5–5 mM pH = 3–11	The rate of SMX degradation during the three UV-based systems was observed in the following order: UV/PMS > UV/PDS > UV/ H_2O_2. The maximum SMX degradation occurred at pH 11, 5, 3 in UV/PMS, UV/H_2O_2 and UV/PDS system, respectively.	Only PMS could be activated by some inorganic anions as compared to PS and H_2O_2. Adding Cl^- and CO_3^{2-} into UV/PMS process can differentially raise the degradation rate. The presence of NO_3^- could improve the SMX degradation in the three UV-based AOPs.

[60]	UV_{254}/PDS	Chloramphenicol (CAP)	Synthetic solution	UV lamp: 75 W, fluence rate: 1.14 W/m^2. CAP = 0.008–0.06 mM PS: 0.5–4 mM pH = 3.0–11.0	It can be concluded that a lower concentration of Cl^- (1~5 mM) accelerated the CAP degradation rate. Although high removal efficiency was obtained in CAP degradation by UV/PS treatment, CAP may not completely be mineralized and it probably converted to other organic byproducts, which might also be poisonous to human beings.	When pH > 7.0, both $SO_4^{-\bullet}$ and $HO^\bullet$ exist in UV/PS system
[61]	UV_{254}/PDS	Chloramphenicol (CAP)	Natural water	UV lamp: 4.9 W, fluence rate: 2.43 mW/cm^2 CAP = 31 µM PS: 0–5 mM Natural conditions	UV/PS system was pH independent compared to ZVI chemically activated PS systems. Full CAP mineralization was reached in 1 h upon increasing [PS] (2.5 mM) along with the UV fluence (874 J). Results showed the superiority of the UV/PS systems compared to thermal and chemical PS-based systems in terms of % CAP mineralization under similar conditions.	Radical quenching experiments showed equal contribution of HO and SO_4 (generated from PS) in CAP degradation apart from photolysis responsible for 65% of CAP oxidation. Fumaric acid (FA) and humic acids (HA) strongly affected the oxidation performance of CAP while Cl, HCO_3, NO_3, NO_2 and SO_4^2 showed moderate implication.
[62]	UV_{254}/PDS	Erythromycin (ERY)	Ultrapure water Bottled water Humic acid solution Conventionally treated wastewater samples	UV lamp: 9 W ERY = 50–200 mg L^{-1} PS: 0–50 mg L^{-1} pH = 3.0–8.0	The results revealed that 10 mg/L of sodium persulfate (SPS) can result in rapid and complete antibiotic degradation within 90 min of irradiation. The contribution of hydroxyl and sulfate radicals on ERY degradation was found to be ca. 37% and 63%, respectively.	ERY degradation rate was strongly affected by the chemical composition of the aqueous matrix and it decreased in the order of: ultrapure water (k_{app}¼ 0.55 min^{-1}) > bottled water (k_{app}¼ 0.26 min^{-1}) > humic acid solution (k_{app}¼ 0.05 min^{-1}) > wastewater effluents (k_{app}¼ 0.03 min^{-1}). This study evidences the potential use of the UV-C/SPS process in producing a final treated effluent with lower phytotoxicity (<10%) compared to the untreated wastewater
[63]	UV_{254}/PDS	Penicillin G (PG)	Synthetic solution	UV lamp: 30 W PG = 0.02–0.14 mM PS: 0.5–3 mM pH = 3.0–8.0	Te results indicated that UV-activated persulfate process can effectively eliminate PG from water. Te highest PG removal efficiency was obtained as 94.28% at pH 5, and the decomposition percentage was raised by increasing persulfate dose from 0.5 to 3 mM and the reaction time from 15 to 90 min	The results of COD removal demonstrated that the UV/PS process may be effectively used for PG decomposition.

(Continued)

Table 2 (*Continued*)
Review of conducted studies in the field of removing antibiotics using ultraviolet activated persulfate process

[64]	$UV_{365}/K_2S_2O_8$ $UV_{365}/TiO_2/K_2S_2O_8$ UV_{365}/TiO_2 Solar light/$K_2S_2O_8$ Fe(II)/$K_2S_2O_8$	Sulfaclozine (SCL)	Synthetic solution	UV lamp: 125 W, fluence rate: 4 mW cm^{-2} SCL = 88 mol L^{-1} $[K_2S_2O_8]/[SCL] = 20–500$ pH = 7, 11	Activating persulfate ions with UV, solar irradiation, Fe (II)and UV/TiO_2 systems for $SO_4^{\bullet-}$ generation was found to be efficient for the elimination of sulfaclozine from water. The highest percentage of the elimination of sulfaclozine was obtained in the UV/$TiO_2/K_2S_2O_8$ system	It was found that at pH 7, hydroxyl and sulfate radicals were involved in the degradation of sulfaclozine, whereas at pH 11 no contribution of sulfate radicals was observed
[65]	UVC/PDS UV/H_2O_2	Florfenicol (FLO)	Synthetic solution	UV lamp: 75 W FLO = 20 µM PS = 0.2–4 mM No adjusted pH	UV/PS is a promising technology for water polluted by antibiotics, and the treatment is optimized only after the impacts of water characteristics are carefully considered. UV/PS has been advantageous over UV/H_2O_2 in terms of the FLO degradation efficiency, mineralization extent, and chemical costs	High PS dose and appropriate Fe^{2+} concentration favored the UV/PS oxidation efficiency. In contrast, the presence of nitrate, chloride, bicarbonate, and HA all reduced the FLO degradation efficiency. Bacterial inhibition tests revealed that the treatment is capable of controlling the toxicity for FLO degradation
[66]	UV_C/PDS	Metronidazole	Synthetic solution	UV lamp: 75 W Metronidazole = 10–50 mg L^{-1} PS =0.1–0.9 mM L^{-1} pH = 3, 5, 7	The optimum conditions in this study were obtained at pH = 9, persulfate concentration of 0.7 mmol/l, and contact time of 30 min.	
[67]	UVC/PDS	Tylosin	Synthetic solution	UV lamp: 30 W Tylosin = 20–80 ppm PS = 0.5–20 mM pH = 2.4–9.8	No significant destruction was observed using separate UV illumination. more than 97% of antibiotic content was removed using UV irradiation and peroxydisulfate simultaneously.	Neutral pH (6.7) was monitored as the optimum pH; however, all levels of pH demonstrate satisfactory removal
[68]	UV/PDS	Metronidazole (MNZ)	Synthetic solution	UV lamp: 8 W MNZ = 5–100 ppm PS = 0.1–1 g/L pH = 3–11	UV/S_2O_8 process efficiency is higher in acidic pH values due to production of further radical SO_4 and increases with extended contact time, but the efficiency of the process is reduced by increasing the concentration of MNZ	
[69]	UV_{254}/PDS UV_{254}/H_2O_2	Thiamphenicol (TAP)	Synthetic solution	UV lamp: 20 W TAP = 10–50 µM PS = 0.2–5 mM pH = 3–11	UV/PS process was more efficient in degrading and mineralizing Thiamphenicol. TAP degradation was affected by solution pH, anions and HA. $SO_4^{\bullet-}$ played the main role in the TAP degradation by UV/PS process. When TAP was completely degraded, only part of TAP w$_a$s mineralized with UV/H_2O_2 process, and was totally mineralized with UV/PS process.	HCO_3^- and HA markedly inhibited the degradation with the both processes, NO_3^- had a slight inhibition on the degradation in UV/H_2O_2 system, and Cl^- significantly enhanced the degradation with UV/PS process. The electrical energy per order (EE/O) analysis showed that UV/PS process was a less energy consumption process

[70]	UV/PMS	Ciprofloxacin (CIP)	Synthetic solution	UV lamp: 2.8 kW Medium pressure lamps (MP) emitting various UV light outputs ranging from 200 nm to 300 nm. CIP = 0.75–6.04 µM PS = 0.1–1 mM pH = 3–11	A high PMS dose, high pH, low CIP concentration and the presence of Cl^-, CO_3^{2-} and NO_3^- promoted CIP removal. Defluorination and transformation of the piperazine ring were found to be the main pathways for degrading CIP during MPUV-only and the MPUV/PMS processes, respectively	MPUV/PMS was effective to reduce the genotoxicity of CIP solutions
[71]	UV_{254}/PDS UV_{254}/TiO_2 GAC-TiO_2 UV_{254}/GAC-TiO_2	Sulfadiazine(SDZ)	Synthetic solution	UV lamp: 28 W. SDZ = 1 mg L^{-1} PS = 50–200 mg L^{-1} pH = No adjusted pH	In UV-C/PS system, near complete degradation of SDZ (99.8%) was observed within 10 min under 50 mg/L of PS and 28 W UV illumination. On the other hand, complete SDZ removal was observed in PS alone system at a dosage of 1000 mg/L but the formation of SO_4^{2-} was found to be a drawback	The comparison of electrical energy consumed (EEO) in different systems revealed that UV-C/ GAC-TiO_2 and UV-C/PS system were energy efficient compared with other systems
[72]	UV_{254}/PDS UV_{254}/TiO_2 UV_{254}/PS/ TiO_2	Metronidazole (MNZ)	Synthetic solution	UV lamp: 32, 64 W. MNZ = 10 mg L^{-1} PS = 100 mg L^{-1} pH = No adjusted pH	The PS dosage for 100 mg L^{-1} was found to be optimal for removing of 10 mg L^{-1} of MNZ in batch reactor operated with 32 W UV power. The sequence of MNZ removal (from highest to lowest) in the presence of anions was (SO_4^{2-} + Cl^- + NO_3^-) > (SO_4^{2-} + NO_3^-) > SO_4^{2-} > (SO_4^{2-} + Cl^-) > Cl^- > NO_3^-.	The presence of SO_4^{2-} ions promoted the MNZ removal whereas NO_3^- and Cl^- ion declined the rate of MNZ removal
[73]	UV/PDS	Penicillin antibiotics (PENs): Pencillin G(PG), Amoxicillin (AMX), Carbenicillin (CBN)	Surface water (SW) Ultrapure water (UW) Waste water (WW)	UV intensity = 5.42×10^{-7} PENs = 0.05 mg L^{-1} PS = 0–5 mM pH = 3–9	The selected PENs could be eliminated effectively by UV/ PDS treatment. Among the three PNEs, PG with the least stable β-lactam ring structure and exhibited higher reactivity than AMX and CBN. The developed model predicted PENs degradation in UW well but underestimated the degradation in SW and WW samples.	Carbonate radical seems to have no effect on PENs removal. However, Cl^- showed inconsistent effects on PENs degradation. Cl^- derived radicals had the highest reactivity towards AMX.

(*Continued*)

Table 2 (*Continued*)
Review of conducted studies in the field of removing antibiotics using ultraviolet activated persulfate process

[74]	UV_{254}/PDS UV_{254}/H_2O_2	Sulfasalazine (SSZ)	Synthetic solution	UV intensity = 15 W SSZ = 29.1 µM PS = 0.1–1.0 mM pH = 7	Although SSZ was resistant to direct UV photolysis, it could be effectively removed by both UV/H_2O_2 and UV/PDS processes. Natural water constituents, such as natural organic matter (NOM) and bicarbonate (HCO_3), influenced the degradation of SSZ differently	The azo functional group of SSZ molecule has predicted as the reactive site susceptible to electrophilic attack by $SO_4^{\bullet-}$by frontier electron densities (FEDs) calculations
[75]	UV_{254}/PDS UV_{254}/H_2O_2	Sulfonamides: Sulfadiazine (SDZ), sulfathiazole (STZ), Sulfamerazine (SM1), Sulfisoxazole (SIZ), Sulfamethazine (SM2), Sulfamethoxypyridazine (SMP), Sulfamonomethoxine (SMM) and Sulfadimethoxypyrimidine (SDM)	Synthetic hydrolyzed urine Synthetic wastewater	UV intensity = 4 W Sulfonamides = 1 µM PS = 0–1 mM	Sulfonamide antibiotics were eliminated more efficiently in synthetic hydrolyzed urine than in synthetic wastewater and UV/PDS was more efficient than UV/H_2O_2 to degrade most sulfonamides.	Energy evaluation showed that UV/PDS costs less energy than LPUV and UV/H_2O_2. By varying UV dose and oxidant dose, the UV/H_2O_2 process can be optimized to achieve higher efficiency than the UV/PDS process in synthetic wastewater

oroquinolones [52]. Although, in a study, further increase of $Na_2S_2O_8$ concentration resulted in saturation, i.e. no obvious increases of degradation efficiency were observed [79]. Liua et al. reported that with initial PS concentration less than 1 mM, k_{obs} well fitted with linear regression, while with the PS initial concentration more than 1 mM, the non-linear model was best fitted which may be due to the radical scavenging effect of excess amount of PS [54]. Based on Cui et al study, an increase of PMS dosage accelerated the rate of sulfonamides degradation without reaching saturation; this UV/PS system was conducted in 0.5–5 mg/L PS concentration [56]. On the other hand, Liua et al. reported that using UV/PS system conducted in 0–2 mM PS concentration, high levels of PS concentration led to saturation and scavenging effects. Also, they concluded that an increase of PS dosage resulted in higher operational costs due to the additional treatment of sulfate anion produced from the photolysis of PS [54]. It should be mentioned that the efficiency of UV/PS system not only depends on the type of target pollutants but also depends on the pollutant/PS concentration ratio [52].

Ao et al. showed that sulfamethoxazole degradation rate dropped with an increase of initial concentration of sulfamethoxazole in UV/PMS, UV/PDS, and UV/H_2O_2 systems. Accordingly, with the increase of sulfamethoxazole concentration, the penetration of photons into the solution will be decreased and the solution acts as an inner filter which inhibited photolysis efficiency of antibiotics. Also, in this situation, the peroxides dissociation decreased and fewer radicals were produced to react with target contaminant [59]; therefore, more reaction time was required for the removal of target pollutants [60].

2.2. The effect of pH

pH is an important parameter in chemical reactions, especially in photochemical systems. Like other photochemical systems, pH affects the photochemical degradation in UV/PS system. The effect of pH also depends on the characteristics of the target component. Some components appear as different forms in different pHs and this variation leads to different reactivity toward radical species. In addition, the concentration and speciation of reactive radicals are pH dependent. Considering the results from Guo et al., fluoroquinolones removal well correlated with the distribution of deprotonated species of fluoroquinolones. Carboxyl and piperazinyl are two proton binding functional groups of norfloxacin and enrofloxacin which may appear as cationic, neutral and anionic spices at different pHs (pKa value of 6.10–6.22 and 7.70–8.51, respectively). The k_{app} for the degradation of norfloxacin and enrofloxacin followed the similar order of pH 3.0 < pH 5.0 < pH 7.0 < pH 11.0 < pH 9.0. The k_{app} for norfloxacin at pH of 3, 5, 7, 9 and 11 were 0.101, 0.106, 0.147, 0.186 and 0.127 min^{-1}, while they were equal to 0.104, 0.118, 0.153, 0.250 and 0.210 min^{-1} for enrofloxacin, respectively. Guo suggested that, in the thermally activated persulfate system, the acidic condition could promote reactions. Accordingly, not only the antibiotic form and reactive radicals differ in different pHs, but also persulfate in different activation methods have a different behavior [52].

pH also has significant effects on the removal and mineralization of TOC. Gao et al. reported that the highest degradation rate of sulfamethazine ($k = 7.50 \times 10^{-2}$) was observed at pH 6.5. However, the maximum mineralization (maximum complete oxidation means the degradation of sulfamethazine and their intermediate products) occurred at pH 11, followed by pH 3 and, then, 6.5 [58]. Other studies showed that $SO_4^{\bullet-}$ was the dominant species in pH < 7.0 with a major role in Chloramphenicol degradation, but as pH increased the greater amounts of OH^- reacted with $SO_4^{\bullet-}$ to generate $HO^\bullet$, radical spices with lower redox potentials (1.9–2.7 V) than sulfate radical (2.5–3.1 V). However different pH values can differentiate redox potential of hydroxyl and sulfate radicals and, in base condition, the redox potential of hydroxyl radical may be slightly higher than sulfate radical [80,81]. As highlighted before, $SO_4^{\bullet-}$ acted more selective than other radicals such as $HO^\bullet$. If there is more $HO^\bullet$ in the system, less selective characteristic of $HO^\bullet$ leads to complete oxidation of organic items (degradation of antibiotics and their intermediate products) [59,60]. Overall, the best recommendation is to take benefit from both spices of $SO_4^{\bullet-}$ and $HO^\bullet$ by adjusting the pH of the process to achieve the most concentration of $SO_4^{\bullet-}$ followed by generation of more $HO^\bullet$ for the total removal of pollutants and oxidation of intermediate products, respectively.

3. Degradation pathway

The review of studies showed that antibiotics would rapidly decompose in UV/ PS systems. The degradation pathway could be different based on antibiotic structures. In sole-UV systems, the main route of degradation is direct photolysis for some antibiotics. In UV/ PS process, despite the fact that hydroxyl radicals can be formed by the reaction of sulfate radical with hydroxide anion, hydroxyl radical has no important role in the degradation process and sulfate radical is responsible for enhanced degradation of antibiotics [50]. However, Chang Zheng Cui reported that PMS decomposed under UV radiation by cleavage of its peroxide bond and produced one $SO_4^{\bullet-}$ and one $HO^{\bullet-}$ and these radicals rapidly oxidized many organic compounds with constant rates ranging from 106 to 1010 $M^{-1}\cdot s^{-1}$ [56]. As mentioned above, a different structure of the target compound led to a different degradation pathway and, also, the removal and byproducts showed to be different in UV/PS system. In other words, for any compound some groups are more susceptible to reactions with reactive oxygenate species (ROS) and $SO_4^{\bullet-}$, so considering the susceptible groups, the degradation pathway is different. Regions of antibiotic with the highest values of electron density such as piperazinemoiety and the benzene ring of the quinolone, thioether, primary amine, benzene ring ,and β-lactam ring and central secondary amide in β-lactam group antibiotics seemed to be the most susceptible to reactions with ROS and $SO_4^{\bullet-}$ [50].

4. Conclusion

The literature review demonstrated that antibiotics removal was stronger in UV/PS system compared to UV or persulfate alone. Also, UV/PS could achieve the objective of the advanced oxidation process which was a complete degradation of the pollutants and its mineralization. However, the selection of optimal concentration to avoid saturation and loss of persulfate accounts for lower operational cost and related complications. Antibiotics removal in UV/PS systems was pH dependent, and, at the neutral condition, the sulfate radicals were dominant but at higher pHs, the hydroxyl radicals showed good capability toward degradation of antibiotics and their by-products. An analysis of the antibiotic degradation pathway in UV/PS system suggested that regions the highest electron density values were the most susceptible to reactions with reactive oxygenated species and the decomposition started from these reactions. Reviews of the studies showed that most of studies were done in synthetic solution media and the removal of antibiotics by UV/PS processes needs to be investigated in the real aqueous medium. Finally, all the included studies in this systematic review evaluated the scavenging effect of organic compounds and study of inorganic scavengers' effects can provide significant information at this area. Toxicity test and determining bacterial resistance of new compounds of degradation can be useful to clarify the degradation pathway.

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