



Antibiotic removal processes from water & wastewater for the protection of the aquatic environment - a review

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ABSTRACT

Currently the serious problem of contamination by antibiotics is a reality. The scientific evidence of its negative effects on the aquatic environment and human health are numerous and unquestionable. Therefore, it is essential to intensify research into effective and efficient processes for removing antibiotics from the aquatic environment. In this paper, on the one hand, a review of the concentrations detected in all types of waters of some antibiotics is developed. In concrete of Ciprofloxacin (CIP), Erythromycin (ERY), Levofloxacin (LEV), Metronidazole (MET), Norfloxacin (NOR), Ofloxacin (OFL), Sulfamethoxazole (SMX) and Trimethoprim (TIM). Of the publications consulted, it can be noted that the most detected is SMX, while those with the highest concentrations are CIP, SMX and TIM. On the other hand, some of the main methods to eliminate antibiotics from the aquatic environment are defined and classified. The methods are compared, indicating their advantages and disadvantages. Combined processes are also mentioned as a good alternative. Finally, the removal percentages achieved by each method in some representative publications are detailed. In this regard, it can be said that the methods with the best elimination percentages (range 80–100%) are biological methods (Biological Aerated Filter, Anaerobic Digestion & Biological Activated Carbon Filter) and membrane technology (Nanofiltration & Reverse Osmosis). While those with the worst results (under 80%) are chemicals (Coagulation-Flocculation) and constructed wetlands (Horizontal Subsurface Flow Constructed Wetlands).

1. Introduction

Although the use of antibiotics is known to have existed since the ancient Egypt and in the Middle Age, and that there were numerous contributions from different authors since the 19th century, Alexander Fleming can be pointed out as an important figure in the invention of them. He was a British scientist who in 1928 accidentally discovered, in one of his forgotten colonies of *Staphylococcus aureus*, that a fungus (*Penicillium notatum*) inhibited its growth. The concerning molecule was purified and called penicillin [1].

In the modern medicine, the word “antibiotic” initially referred to any agent with biological activity against living organisms, while now it alludes to substances with antibacterial, antifungal or antiparasitic activity. One of the possible modern definitions pretends to qualify antibiotics as chemotherapeutic agents that inhibit or eliminate the growth of microorganisms, such as bacteria, fungi, or protozoa [2].

There are several different kinds of antibiotics, and they can be classified based on their chemical structure, action mechanism, action spectrum, and the route of administration. Out of these classifications the most popular one is their mechanism of action and based on it the

Abbreviations: Reverse Osmosis, RO; Wastewater Treatment Plant, WWTP; Water Treatment Plant, WTP; Drinking Water Purification Plant, DWPP; Solid Phase Extraction, SPE; Coagulation-Flocculation, C-F; Powdered Activated Carbon, PAC; Granular Activated Carbon, GAC; Advanced Oxidation Processes, AOPs; Electrochemical Oxidation, EO; Ozonation Process, OP; Fenton Process, FP; Free Water Surface Flow Constructed Wetlands, FWS CWs; Ultraviolet, UV; Microfiltration, MF; Ultrafiltration, UF; Nanofiltration, NF; Biological Aerated Filter, BAF; Anaerobic Digestion, AD; Up flow Anaerobic Sludge Blanket, UASB; Anaerobic filter, AF; Anaerobic Baffled Reactor, ABR; Sequencing Batch Reactor, SBR; Membrane Bioreactor, MBR; Biological Activated Carbon Filter, BAC; Horizontal Subsurface Flow Constructed Wetlands, HSF CWs; Strengths, Weaknesses, Opportunities, Threats, SWOT; Hydraulic Retention Time, HRT; Hydraulic Loading Rate, HLR; Conventional Activated Sludge, CAS; Ciprofloxacin, CIP; Erythromycin, ERY; Levofloxacin, LEV; Metronidazole, MET; Norfloxacin, NOR; Ofloxacin, OFL; Sulfamethoxazole, SMX; Trimethoprim, TMP; Vertical Subsurface Flow Constructed Wetlands, VSF CWs.

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most common groups are: β -lactams, sulfonamides, monobactams, carbapenems, aminoglycosides, glycopeptides, lincomycin, macrolides, polypeptides, polyenes, rifamycin, tetracyclines, chloramphenicol, quinolones, and fluoroquinolones [3].

Another way is to divide them into the following groups: Beta-lactams that include Cephalosporins, Penicillin, Monobactams, Carbapenems; Macrolides; Lincosamines; Tetracyclines; Aminoglycosides; Amphenicols; Peptides including Polypeptides, Glucospeptides, Lipopeptides, Polymyxins; Oxazolidinones; Nitro-derivatives; Fusidanos; Phosphonates; Pleuromulins; Quinolones; Sulfonamides; Diaminopyridines [4].

In Fig. 1 the diagram explains through the human food chain the connection between the use of antibiotics and human health. It starts with the normal intake of antibiotics, followed by the contamination of food and drinking water and finally leading to infections by drug resistant bacteria which are more difficult to treat [5].

There are other ways to explain the entry routes of antibiotics to the environment that have an impact on human health. We can classify them in two groups: antibiotics used in human medicine and the antibiotics used in veterinary medicine.

One of the gateways of contamination of waters by human antibiotics is the following. Humans take antibiotics and then excrete them. The excretions reach the Wastewater Treatment Plant (WWTP), and after their treatment they generate biosolids and effluent discharges that cause pollution of the soil and the aquatic environment (Surface toilet and groundwater).

Moreover, in veterinary medicine, the antibiotics are given to livestock and then excreted. Excretions generate manure storage tanks and lagoons that cause contamination of the soil and the aquatic environment (Surface water and groundwater). Within this section, an especially representative case that directly causes pollution of the aquatic environment (Surface toilet and groundwater) is the use of antibiotics in aquaculture [6].

There are also unconventional pathways of contamination of environment, for example due to the lack of recycling of expired drugs. The recycling of medicines can be a possibility against this route of entry. For this, it is necessary to study different methodologies to recycle active drugs from expired pharmaceutical products. Studies admit that when a drug expires, it can contain 90% or even more of the active pharmaceutical ingredients. Therefore, suitable chromatographic methods and analytical techniques could be adopted for the isolation and eventual quantification of the active ingredients in order to successfully recycle

them into useful products. This approach would be ecological [7].

In all cases, the contamination of the soil and the aquatic environment (Surface water and groundwater) leads to direct and indirect routes that then get to human beings. Directly, because the contaminated waters access the Water Treatment Plant (WTP) where the drinking water comes from. Indirectly, because the contaminated waters are used for irrigation which drives to the contamination of crops that humans and livestock animals then eat. These agents are then discharged into the sea, contaminating the fish we eat. This water is also used as drinking water for livestock animals which humans then eat too [6].

In addition to the human consumption of antibiotics, there are many other anthropogenic activities such as the use of antibiotics in agriculture and aquaculture, and non-human applications of antibiotics and waste disposal, which generate large environmental resistance reserves [8] and virulence genes with their respective organisms that host them [9].

Multiple genetic and genomic studies of wastewater treatment plants have shown that these are important deposits of resistant genes and organisms [10,11]. Frequently these genes are transported as genomic islands in transmissible plasmids and represent sources of resistance [12].

The main entry routes of pharmaceutical residues (including antibiotics) into the aquatic environment are excretions after use, poor disposal of unused medicines and the waste generated after their production. From the routes of entry, they arrive to the WWTP [13].

From an environmental point of view, the incorporation of antibiotics, metabolites and antibiotic resistance genes into the natural environment is of great concern. Studies have shown that contaminants based on antibiotic residues can influence microbial populations through bacteriostatic and bactericidal effects, leading to the disappearance of key microbial groups associated with ecological activities or affecting their physiological functions. The detection, monitoring and characterization of these components in the aquatic environment are important to evaluate their toxic, teratogenic and mutagenic effects in ecosystems [14].

The problem is that these WWTP are not prepared to eliminate these type of waste products. In this regard, there are studies whose experimental results indicate the limitations of primary treatment methods when it comes to breaking down pharmaceutical products [15]. All the above suggests the need of new treatments focused on the removal of antibiotics from the different waters.

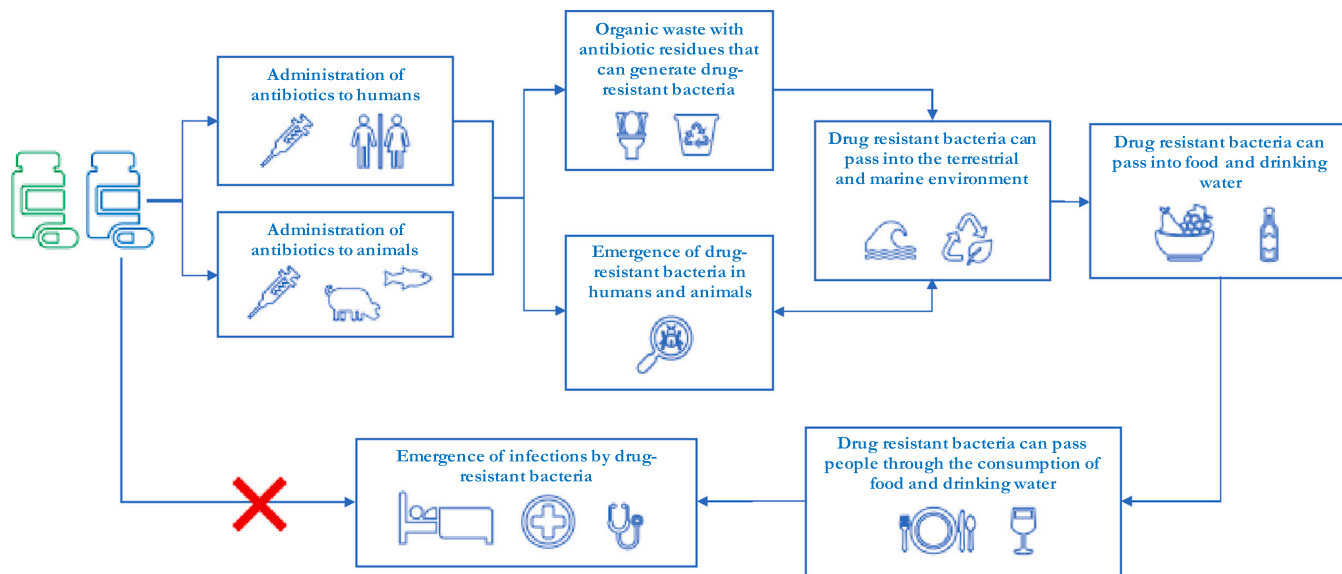


Fig. 1. Connection between the use of antibiotics and human health: Food chain. Source: Adapted to WHO, 2017.

Regarding the high consumption of antibiotics, it is important to underline that in Europe, according to data from FEDESA in 1997 approximately 10,000 t/year of antibiotics were consumed, of which half were used in medicine and the other half in animal health [16]. In Spain, the consumption of antibiotics has fallen but remains high. In 1985 the overall consumption of antibiotics was 21.9 DHD (Doses defined by 1000 Habitants and Day), while in 2000 it was 20.4 DHD [17].

Statistics on the use of Anti-infective for systemic use in the primary care sector in Europe in 2017 show that the consumption measured in DHD in central-southern Europe ranges between 19,317 and 32,148. It can be classified into three blocks: In the first range (19,317, 23,594) there are countries such as Italy; in the second range (23,594, 27,871) we find countries such as Spain or France; in the third range (27,871, 32,148) we find countries such as Greece or Romania. On the lower end, in northern Europe, in countries such as England, Sweden, Norway, Denmark or Germany, consumption is fixed in the range: (10,763, 19,317) [18].

The objectives of this bibliographic review are summarized in, on the one hand, to determine the concentrations of a series of high-consumption antibiotics in developed countries, as well as the analytical techniques used. On the other hand, classify and define some of the main antibiotic elimination processes, including single and combined. Finally, determine for the antibiotics mentioned, the percentages of elimination of some of the main previous processes. Compare and discuss these percentages are also included.

2. Occurrence (in water)

Regarding analysis techniques, it begins by showing in Table 1 the evolution that analysis techniques have followed in the last 50 years.

Determination of pharmaceuticals in different water samples can be performed by various chromatographic techniques, including High Performance Liquid Chromatography-Ultraviolet (HPLC-UV) [19], High Performance Liquid Chromatography-Diode Array Detection (HPLC-DAD) [20,21], Liquid Chromatography-Mass Spectrometry (LC-MS), Liquid Chromatography-Mass Spectrometry/Mass Spectrometry (LC-MS/MS) [22–24] and Gas Chromatography-Mass Spectrometry (GC-MS) [25,26].

Currently, due to their high sensitivity and selectivity, methods based on mass spectrometry are the most applied approach to determine sulfonamides residues at low concentrations (mg/kg₁ or mg/kg₁).

Table 1
Evolution of analysis techniques. Source: [53].

Time period	Analysis technique	Abbreviation
Before 1970	Thin-Layer Chromatography	TLC
1970–1980	Gas Chromatography - Electron Capture Detector	GC-ECD
	High Performance Liquid Chromatography	HPLC
1980–1990	Gas Chromatography - Mass Spectrometry (Selected Ion Monitoring)	GC-MS (SIM)
	High-performance Liquid Chromatography (Diode Array Detection - Ultraviolet)	HPLC (DAD-UV)
	Gas Chromatography - Mass Spectrometry (Electron Capture Detector.)	GC-MS (ECD)
1990–2000	Liquid Chromatography - Mass Spectrometry	LC-MS
	Liquid Chromatography - Mass Spectrometry (Quadrupole-quadrupole-Quadrupole)	LC-MS (QqQ)
	Liquid Chromatography - Mass Spectrometry (Quadrupole-quadrupole-Linear Ion Trap)	LC-MS (QqLIT)
	Ultra-Performance Liquid Chromatography	UPLC
	Ultra-Performance Liquid Chromatography - Time of Flight	UPLC-TOF
2000–2010	Ultra-Performance Liquid Chromatography - Orbitrap	UPLC-ORBITRAP
	Ultra-Performance Liquid Chromatography - Electro-Spray Ionization - Mass Spectrometry	UHPLC-ESI-MS

Within methods, the use of liquid Chromatography-Electrospray-Quadrupole Linear ion trap mass spectrometry (HPLC-ESI-QqLIT-MS/MS) permits analysis with high specificity and adequate limits of detection [27–32].

High performance liquid chromatography (HPLC) is the most common method used for separation and determination of these compounds because most pharmaceuticals are non-volatile [33]. As the residue of pharmaceutical compounds is usually present at very low concentrations in the environmental water, a sample preparation and pre-concentration step are necessary before analysis [34,35]. Several procedures have been reported for the pre-concentration of pharmaceuticals from water matrices including Solid Phase Extraction (SPE) [22,36], Liquid-Liquid Extraction (LLE) [37], Quick, Easy, Cheap, Effective, Rugged & Safe (QuEChERS) method [38], Magnetic Solid Phase Extraction (MSPE) [39], Hollow Fiber Liquid Phase Microextraction (HFLPM) [40] and salting-out assisted liquid-liquid extraction for Non-Steroid Anti-Inflammatory Drugs (NSAIDs) [41–46]. Among those commonly used methods, solid-phase extraction (SPE) was the most extensively used technique [47–49].

As a conclusion to say that the main technique of analysis to detect these concentrations is liquid chromatography-tandem mass spectrometry [50,51], previously, the samples must be treated to preconcentrate them using Solid Phase Extraction (SPE) [30,52].

Regarding antibiotics, eight types were selected. This selection is due to the search for amply spectrum antibiotics, under the reasoning that this should lead to wide use and therefore large discharge to the aquatic environment. In addition, we learned from the Spanish Agency for Medicines and Health Products that, for example, fluoroquinolones have increased their use in Spain by 26% in 12 years (between 1997 and 2009).

The selection includes Quinolones (*Ciprofloxacin*, *Levofloxacin*, *Norfloxacin* and *Ofloxacin*) were chosen because they are antibiotics that are used for the treatment of a wide spectrum of urinary, respiratory, genital, gastrointestinal, skin, bone and joint bacterial infections. Nitroimidazoles (*Metronidazole* and *Trimethoprim*) were selected because they are antimicrobials with bactericidal action that present a moderately broad antibacterial spectrum. They are used in infections of the urinary tract, ears, lungs, intestines and liver. Macrolides (*Erythromycin*) are used to treat infections caused by bacteria in the respiratory and urinary tracts, as well as ear, intestinal, gynecological, dermatological, and sexually transmitted infections. Sulfonamides (*Sulfamethoxazole*) is an antibiotic that, combined with the Trimethoprim, is used to treat multiple types of bacterial infections: ear, urinary tract, pneumonias and intestinal.

After reviewing the bibliography and limiting the study to the analysis of these eight types of antibiotics whose descriptions are shown

Table 2
Descriptions of antibiotics selected. Source: own elaboration.

Name	Class	Abbreviations	Excreted unchanged in human urine
Ciprofloxacin	Quinolones - Fluoroquinolones	CIP	60% ^a
Erythromycin	Macrolides	ERY	5% ^a
Levofloxacin	Quinolones - Fluoroquinolones	LEV	85% ^b
Metronidazole	Nitroimidazoles	MET	60–80% ^c
Norfloxacin	Quinolones - Fluoroquinolones	NOR	30% ^a
Ofloxacin	Quinolones - Fluoroquinolones	OFL	80% ^a
Sulfamethoxazole	Sulfonamides	SMX	12% ^a
Trimethoprim	Nitroimidazoles	TMP	60% ^a

^a [54–57].

^b Medication datasheet: LEVOFLOXACINO STADA 500 mg.

^c Medication datasheet: METRONIDAZOL I.V. BRAUN 5 mg/ml – CIMA.

in Table 2, we will highlight part of the most representative information in each case: authors; location; water source; maximum concentrations detected in ng/l (see Table 3).

From the total of tests cited in Table 3, two conclusions can be drawn. On the one hand, it should be noted that the antibiotics LEV, SMX and TMP have the highest concentrations, while MET is the one with the least, as can be seen in Fig. 2.

On the other hand, regarding the percentage of times that each antibiotic was studied, it can be seen in Fig. 3 that the antibiotics SMX and TMP are detected in more than 80% of the studies. Next are CIP, ERY, NOR and OFL, with percentages between 30% and 50%. Finally, the least detected is LEV, which does not reach 5%.

3. Removal

3.1. Processes

Some of the single treatment processes for the elimination of emerging pollutants in general and antibiotics for different types of water can be grouped into six large blocks, and each in turn is divided into different elimination techniques as shown in Fig. 4.

First are the chemical processes, where the Coagulation-Flocculation method (C—F) is the most prominent. They can be applied at different stages of water treatment: pretreatment of industrial effluents before entering municipal sewers [95]; primary treatment of urban wastewater

Table 3
Concentrations of antibiotics detected in waters in international studies. Source: Several.

Localization	Water source	Maximum concentration detected (ng/l)								Reference
		CIP	ERY	LEV	MET	NOR	OFL	SMX	TMP	
Brisbane; Australia	Wastewater	6900	–	–	–	210	–	570	930	[58]
Bosnia and Herzegovina, Croatia & Serbia	Sewage	2610	420	–	–	2940	–	11600	2550	[59]
Northwest Ohio; USA	Wastewater	377	–	–	–	–	–	472	–	[60]
Merthyr Tydfil,	River Taff	–	121	–	11	–	–	8	120	[61]
Pontypridd and Cardiff,	WWTP Cilfynydd,	–	6755	–	1583	–	–	150	6796	
United Kingdom	River Ely	–	72	–	24	–	–	4	183	
	WWTP Coslech	–	10025	–	962	–	–	274	4673	
Madrid; Spain	Sewage	13625	2310	–	165	–	5286	530	197	[62]
North América	Sewage	1100	–	–	–	–	–	2800	7900	[63]
Europe	Sewage	3353	–	–	–	–	–	794	1264	
Asia and Australia	Sewage	720	–	–	–	–	–	1400	321	
North América	Rivers - canals	–	–	–	–	–	–	211	212	
Europe	Rivers - canals	–	–	–	–	–	–	4	78.2	
Asia and Australia	Rivers - canals	1300	–	–	–	–	–	2000	150	
Ontario, Canada	Source water	–	145	–	–	–	–	284	25	[64]
	Drinking water	–	155	–	–	–	–	2	15	
Rhône-Alpes region; France	Surface Water	–	–	–	0.3	–	3.2	1.9	0.9	[65]
	Groundwaters	–	–	–	0	–	0	3.0	1.4	
	Drinking water	–	–	–	0.4	–	3.2	11	1.4	
Northeast; Spain	Drinking water	–	33	–	–	–	–	149	22.8	[66]
USA	River	–	–	–	–	–	–	38.1	9.1	[67]
Ulsan, Korea	Wastewater	–	–	–	–	–	–	216	277	[68]
Madrid, Spain	Rivers	224	3847	–	1834	<10	552	952	690	[69]
Valencia; Spain	Wastewater	3850	120	–	–	1070	960	640	160	[70]
Stonecutters; Hong Kong	Sewage	–	684	–	–	861	1263	155	119	[57]
Tai Po; Hong Kong	Sewage	–	557	–	–	67	336	143	119	
Sha Tin; Hong Kong	Sewage	–	982	–	–	286	742	40	136	
20 states; USA	Sewage	320	–	–	–	–	660	2900	370	[71]
Taiwan	Surface Water	–	–	–	–	–	–	60	2.1	[72]
Gran Canaria; Spain	Sewage	20321	–	14154	–	2366	–	–	–	[73,74]
Gran Canaria; Spain	Sewage	–	300	–	630	–	140	130	100	[75]
Manipal STP; India	Sewage	–	–	–	–	–	–	2260	2080	[76]
Portugal	Drinking water	–	5	–	–	–	–	1.3	–	[77]
Japan	DWPP	–	–	–	–	–	–	19	–	[78]
Volos; Greek	Sewage	591	–	–	35	–	–	80	96	[79]
Malaysia	Rivers	299.88	–	–	–	–	–	114.24	–	[80]
Canada	Wastewater	–	–	–	–	–	–	2750	–	[81]
United Arab Emirates	Wastewater	1028	951	–	–	–	1012	228	–	[82]
Mexico	Surface water	–	–	–	–	–	–	1220	395	[83]
	Wastewater	–	–	–	–	–	–	2010	790	
Mexico	Wastewater	2570	1140	–	–	193	1120	6570	1610	[84]
Mexico	Treated Water	65.8	–	–	–	–	293	1215	395	[83,85]
Brazil	Wastewater	33.7	–	–	–	37.7	–	376	65.1	[86,87]
Brazil	Rivers	–	–	–	–	51	–	106	484	[88]
Brazil	Hospital effluent	119	–	–	–	–	–	27800	6650	[89]
Costa Rica	Surface water	740	–	–	–	1744	335	56	122	[90]
Ecuador	Rivers	–	–	–	–	–	–	–	610000	[91]
Kenya	Surface water	1300	–	–	–	2200	–	49700	–	[92]
	Wastewater	3000	–	–	–	2900	–	49300	5600	
Pakistan	Wastewater	32.57	–	6.64	–	–	2.56	16.09	–	[93]
Shanghai, China	Rivers	34.2	6.9	–	–	2.6	28.5	764.9	–	[94]
	Total data	24	19	2	11	15	17	49	41	
	Maximum	20321	10025	14154	1834	2900	5286	49700	610000	
	Minimum	3	5	7	0	3	0	1	1	
	Mean	2563	1507	7080	477	989	749	3452	17387	

Script means that the antibiotic was not analyzed in that study or that the result is less than the limit of detection.

DWPP: Drinking Water Purification Plant.

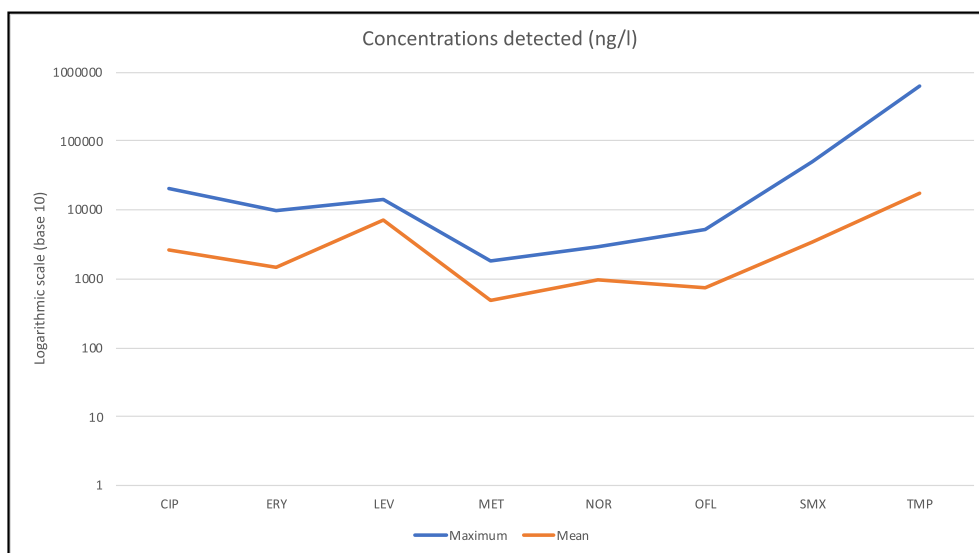


Fig. 2. Summary of antibiotics concentrations detected from the bibliographic review shown in Table 3. Source: Own elaboration.

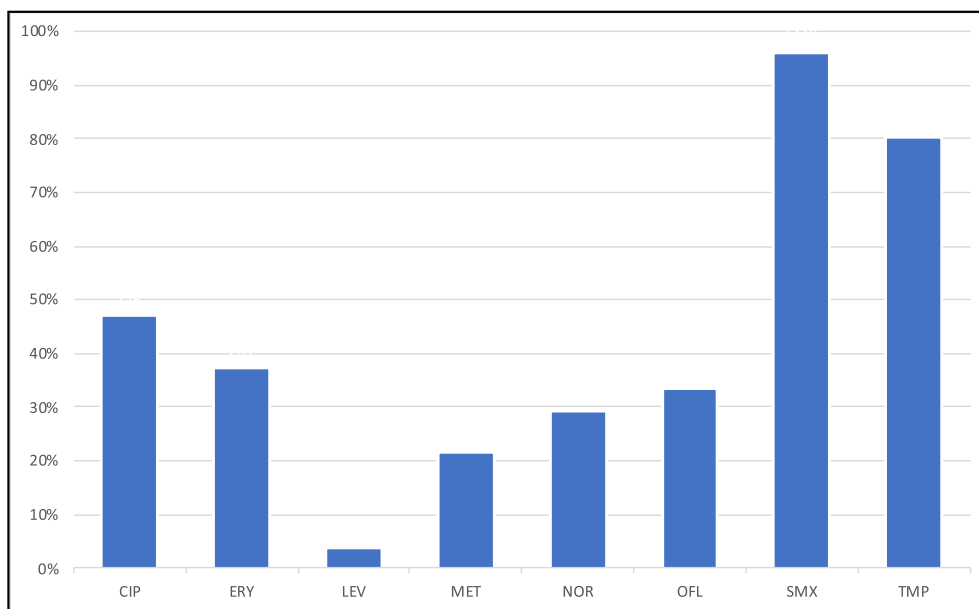


Fig. 3. Percentage of times that each antibiotic was studied out of the total of tests consulted in the bibliographic review shown in Table 3. Source: Own elaboration.

[96]; tertiary urban wastewater treatment [97]; drinking water treatment plants. Interesting studies have been published regarding the detection of pharmacological products in drinking water plants [98], as well as during the primary treatment of municipal wastewater [99]. Regarding the removal of antibiotics in wastewater, there are also published paper [100].

Second group are Adsorption, highlighting Activated Carbon. It is a promising advanced treatment process that can remove many of the pharmacological products that are usually detected in wastewater [101]. There are studies that include the elimination of antibiotics in wastewater and hospital wastewater both using the Powdered Activated Carbon (PAC) [102] and Granular Activated Carbon (GAC) techniques [103]. Another adsorption option is to use cellulose membranes. With this method, in a study with CIP, a 27% elimination was obtained [104]. There are also studies that used zeolites for the adsorption of ciprofloxacin. With this method, elimination percentages between 90% and 97% have been obtained. These are shown to be promising for future

applications for the disposal of this drug in wastewater from the pharmaceutical industry, as well as other drugs with similar structural characteristics [105].

Third are Physicochemical: AOPs, which include Electrochemical Oxidation (EO), technology that produces strong oxidants to degrade pollutants through electrode reactions [106]; the Ozonation Process (OP), which in turn has two mechanisms for ozone-based antibiotic degradation: the direct oxidation of ozone and indirect oxidation through the generation of free radicals [107]; the Fenton Process (FP). In this process, the reagents (H_2O_2 and Fe^{2+}) react with each other to generate OH radicals, which then oxidize and break down the antibiotics [108]; Ultraviolet rays (UV/ H_2O_2). In this method, H_2O_2 decomposes to produce OH with UV irradiation [109].

A good alternative is the AOPs based on ultraviolet rays (UV / H_2O_2), but its high costs derived mainly from the need for upstream pretreatment and downstream H_2O_2 cooling are a significant inconvenient [110]. On the other hand, ozone is presented as a very interesting

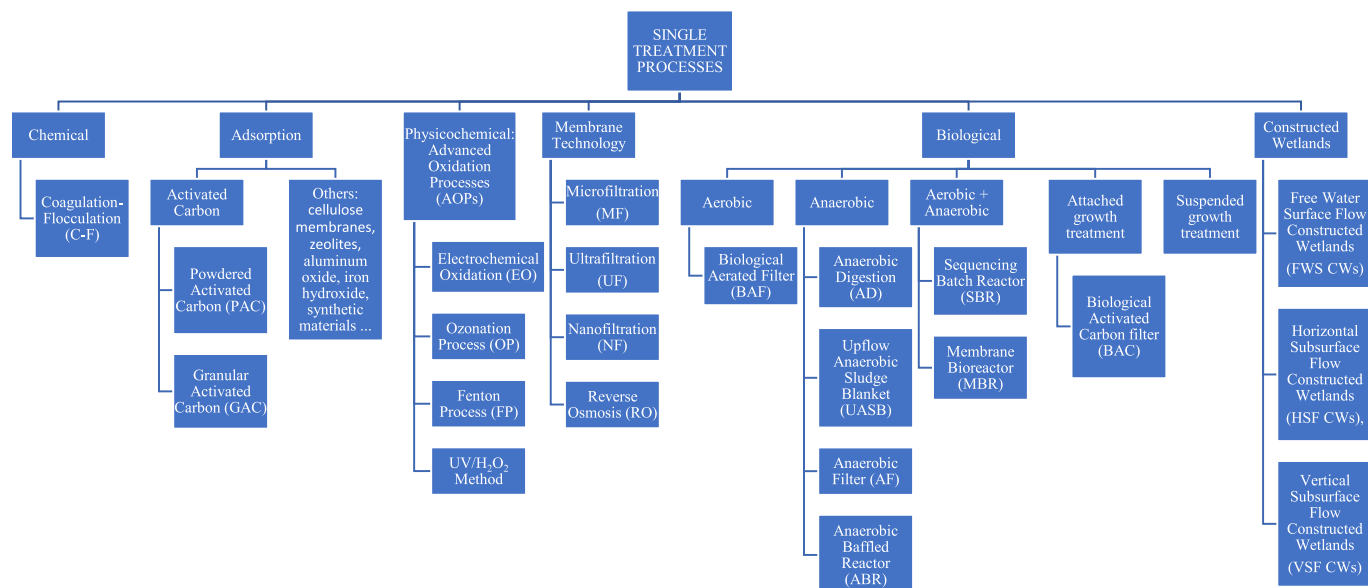


Fig. 4. Single treatment processes for antibiotic removal in different types of waters. Source: Own elaboration.

alternative because its efficiency is like the previous one, but with lower costs due to lower chemical and energy requirements [111]. Antibiotic elimination studies using this process have been reviewed, all of them in wastewater and commonly in pilot plants, but also in municipal treatment plants [110,112].

Fourth are the membrane technology. In this process, pollutants are intercepted as the wastewater passes through small pores in the membrane. They can be divided into Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF) and Reverse Osmosis (RO). In this regard, it should be noted that membrane filtration technologies have been tested on a real scale, in pilot plants and at the laboratory level to remove pharmaceutical products (including antibiotics) from different types of waters [58,113–118].

Fifth are biological treatments, which can be classified as aerobic, anaerobic, and combined aerobic and anaerobic methods. The main aerobic method is the Biological Aerated Filter system (BAF). This is a new type of process used for wastewater treatment that combines oxidation and filtration by biological contact [119]. Includes a solid phase to support microbial growth, a liquid phase to submerge the solid material, and a gas phase for air input [120].

The main anaerobic methods are the Anaerobic Digestion (AD), upflow anaerobic sludge blanket (UASB), anaerobic filter (AF), and anaerobic baffled reactor (ABR) processes. In concrete stand out AD that includes four stages: hydrolysis, acidification, hydrogen production, and acetic acid and methane production [121]. The problem you have is that the treated sewage and sludge residue could still cause damage to the surrounding environment [122]; The main combined aerobic and anaerobic methods are the Sequencing Batch Reactor (SBR) and the Membrane Bioreactor (MBR) processes. The SBR is based on one or more aeration reaction tanks, and the sewage enters the tank in batches. This reactor operates in five sequences: influent feeding, anoxic phase, aerobic phase, sludge settling, and effluent discharge [123]; The MBR processes, which combines modern membrane separation technology and biological technology. Its advantages are long sludge retention time, flexible operation, low sludge production, and high nitrification

performance. Its disadvantages are its high energy consumption and operating costs [124]. Highlight that they can retain more than 5 and 2 log units of bacteria and viruses, respectively. Pilot studies have been developed in hospital wastewater that demonstrate the efficacy of MBR for some pharmacological products, even eliminating more than 95% of them [125–127].

Within the biological ones there are also the attached growth treatment and the suspended growth treatment. In the attached growth treatment, there are the processes Biological Activated Carbon filter (BAC). This process consists of a fixed bed of granular activated carbon that supports the growth of bacteria attached to its surface. They have been used for years to treat drinking water, demonstrating their effectiveness in eliminating natural organic matter [128]. They can be considered an interesting alternative on the one hand, because bio-filtration systems in general tend to be robust, simple to build and require little energy [129]. On the other hand, because BAC filtration costs can be expected to be in the same range as sand filtration [130].

Sixth are Constructed Wetlands. It is an artificial wastewater treatment ecosystem, which uses the combined action of soil, plants and microorganisms to treat wastewater entering the wetland. Wastewater purification is achieved by filtration, adsorption, co-precipitation, ion exchange, plant adsorption, and microbial decomposition [131]. This treatment can be classified as Free Water Surface flow Constructed Wetlands (FWS CWs), Horizontal Subsurface Flow Constructed Wetlands (HSF CWs), and Vertical Subsurface Flow Constructed Wetlands (VSF CWs) according to the direction of water flow [106].

3.1.1. Comparative: SWOT analysis

In this section a comparison of some the antibiotic removal processes from Water & wastewater is made. For this, the selected technique is SWOT analysis because it is a simple tool for strategic analysis that is very widespread in decision-making. It has been decided to carry out a SWOT analysis for each elimination method to decide which techniques should be used according to the case and the intended objective.

SWOT Analysis Coagulation-Flocculation		SWOT Analysis Adsorption: Activated Carbon		SWOT Analysis OP	
Weaknesses	Threats	Weaknesses	Threats	Weaknesses	Threats
It requires the use of chemicals.	Toxic compounds are transferred to the solid phase so the sludge must be treated later.	Compounds with low molecular weight and high solubility are difficult to absorb.	The presence of other compounds competes for available adsorption sites.	It presents limitations in the treatment of organic compounds in highly concentrated wastewater.	Performance can be very diverse in the treatment of organic compounds.
Strengths	Opportunities	Strengths	Opportunities	Strengths	Opportunities
Separate many types of particles the water which achieves multiple objectives.	It is applied at different stages of water treatment as appropriate.	Processes are highly studied in removing emerging contaminants.	Active carbons (PAC and GAC) do not generate toxic products.	It operates in ranges where conventional systems are not feasible.	Do not generate by-products that require further processing.

SWOT Analysis RO		SWOT Analysis MBR		SWOT Analysis Attached growth treatment processes	
Weaknesses	Threats	Weaknesses	Threats	Weaknesses	Threats
The membranes are designed for the treatment of salt water.	Impossibility of using the membranes in all types of waters.	High installation and maintenance costs. ⁷	Fouling by the layer of sludge that accumulates on the surface of the membrane.	Filters should be changed regularly.	The pollutants are separated from the water but are not destroyed.
Strengths	Opportunities	Strengths	Opportunities	Strengths	Opportunities
It is a mature technology.	It does not require adaptation and presentation good percentages of elimination of emerging pollutants.	Increased quality of the effluent as compared to conventional activated sludge systems.	Easy adaptation to existing active sludge plants.	Efficient in removing some organic compounds from drinking water and wastewater.	Used materials available everywhere.

3.1.2. Removal percentage tests

Unfortunately, there is no specific type of treatment that manages to eliminate several micropollutants due to their different properties. To date, there are no reliable processes that allow the elimination of both bulk substances and micropollutants [72].

A bibliographic review was carried out including some of the main studies that have been performed for the elimination of antibiotics from different types of water (see Table 4). In each case it is specified: the treatment used, the method used if appropriate, the water source analyzed, the most representative specifications of the test, the percentage of elimination obtained and finally the reference of the study.

From the data expressed in the Table 4, it can be concluded that there are multiple techniques that achieve excellent elimination percentages (>90%). Therefore, it appears that the decision to use one or the other method will depend on the facilities already in place at the site in question. In Section 4 we will proceed to discuss the results obtained in Table 4.

3.2. Combined processes

Currently many combined processes are used to remove all types of emerging pollutants, including antibiotics, from waters. Some of the most representative are listed below:

First, nanofiltration combined with ozone-based advanced oxidation processes in wastewater. In a study on the elimination of antibiotics in a wastewater treatment plant in China, UV₂₅₄ photolysis, ozonation and UV/O₃ processes were used to treat the nanofiltration concentrate. The conclusions were on the one hand, nanofiltration efficiently removed antibiotics from the effluent. On the other hand, UV₂₅₄ photolysis was not effective in degrading antibiotics. Lastly, the UV/O₃ process was able to further remove the antibiotics in the nanofiltration concentrate

effectively, in addition, the synergistic effect between O₃ and UV in the degradation of antibiotics is observed. Results show high antibiotic rejections (>98%) in all sets of experiments. Thus, zero discharge of micropollutants from WWTPs is possible through the proposed scheme in the study [139].

Second, in an important study of the processes for removing antibiotics from rearing wastewater, combined treatments were analyzed to improve removal efficiency. This study concludes that the combined treatments show a high efficiency of elimination of antibiotics. They have broad prospects for development and application in the treatment of breeding wastewater [106]. This paper quotes some important authors in this sense:

Ben et al. studied on degradation of antibiotics in swine wastewater using a combined biological-Fenton process. In this method, the SBR was used to perform the biological treatment, and then the Fenton process was used for further treatment. The final removal rates of macrolides and sulfonamide were as high as 99% and 92%–97%, respectively. Therefore, the integration of an AOP and a biological method can effectively remove antibiotics from breeding wastewater [140].

Qian et al. conducted a study in which swine wastewater was pre-treated using an up flow anaerobic sludge layer and SBR process, and then the wastewater was treated with the Fenton process to remove antibiotics. The average antibiotic removal efficiency was 74% [141].

Han et al. showed that the antibiotic removal rate was as high as 92% when the SBR and AD methods were combined to treat swine wastewater [142].

In addition, promising techniques using microalgae or with biofuel cells driven by microalgae bacteria have also been studied [143].

Third, Z AL-Qodah et al. has published a series of very interesting articles on combined processes that include electrocoagulation. A first

Table 4

Percentage of elimination of antibiotics through different single treatments process. Source: Several.

Treatment	Method	Water source (Country)	Specification test	Antibiotic	Removal (%)	Reference
Chemical	C-F	Hospital wastewater (Spain)	Optimum coagulant dose: SMX: 50 ppm FeCl ₃ TMP: – ERY: –	SMX	6.0 ± 9.5 ^a	[100]
				TMP	–32.1 ±	
Adsorption. activated carbon.	PAC	Hospital wastewater (Switzerland)	A pilot-scale hospital wastewater treatment plant consisting of a primary clarifier, membrane bioreactor, and five post-treatment technologies including PAC. Dosage: 8; 23; 43 mg/l	CIP	100, >99,	[102]
				ERY	>99	
				MET	>95, >88,	
				NOR	>88	
				SMX	3, 67, 78	
	GAC	Wastewater (China)	The percent removal that were detectable in the GAC influent. Log K _{ow} values. Full scale.	TMP	90	[103]
				ERY	80	
				CIP	50	
				LEV	70	
				SMX	98 (±0.2)	
Advanced oxidation processes (AOPs)	OP	Water reclamation facility (USA)	Pilot-scale evaluation. Average % removal after O ₃ /H ₂ O ₂ 5 mg/l of applied ozone O ₃ and 3.5 mg/l of H ₂ O ₂ Contact time: 30 min.	TMP	>99	[110]
				SMX	98 (±0.2)	
	UV/H ₂ O ₂	Domestic wastewater (Switzerland)	Municipal wastewater treatment plan UV + H ₂ O ₂ (50 mg/l) First contact time: 10 min. Second contact time: 30 min.	CIP	69, 100	[111]
				MET	52, 100	
				NOR	100, 100	
				OFL	100, 100	
Membrane technology	UF	Wastewater (China)	WWTP with inhabitants served 814 × 10 ³ ; daily flow 400 × 10 ³ m ³ ; hydraulic retention time 11 h; solids retention time 12–15d.	TMP	50	[132]
				NF	NF 90: 99.2 HL: 88.8	
	Membranes: NF 90 (Dow/FilmTec) HL (Desal, Osmonics, GE Infrastructure Water Process Techn).	Synthetic wastewater (Croatia)	pH: 7.4–7.5. Temperature 25 °C. Antibiotics solutions: 10 mg/l.	TMP	NF 90: >99.99	[133]
				Wastewater (Croatia)	pH: 6.29–6.82. Temperature: 25–30 °C. Antibiotic feed: 406 µg/l	
	RO	Wastewater (Israel)	Combine with other method. Membrane: Filmtec TW30 25–40 with 2.7 m ² surface area, and a flux range of 22–31 l/mh at a pressure range of 9.5–10.2 bar.	ERY	MBR/RO: 99.3	[135]
				SMX	97.6	
				TMP	MBR/RO: 97.2	
				ERY	97.6	
				SMX	MBR/RO: 97.2	
				TMP	CAS-UF/ RO: 99.3 (c) CAS-UF/ RO: 97.6 CAS-UF/ RO: 93.2	
Biological: aerobic	BAF	Laboratory wastewater (China)	Operation conditions: Hydraulic retention time (HRT) = 40–48 h Hydraulic loading rate (HLR) = 2.8 cm/h	TMP	98.3	[136]
				NOR	81.7	
Biological: anaerobic	AD	Wastewater (China)	Operation conditions: 1.38–2.16 kg chemical oxygen demand (COD)/m ³ /d, 37 ± 1 °C, hydraulic retention time (HRT) = 16 d	OFL	98.6	[137]
				CIP	85	
Biological: aerobic + anaerobic	MBR	Hospital Wastewater (Switzerland)	Primary clarifier and an MBR. Submerged ultrafiltration flat sheet membrane plates (<i>Huber MembraneClearBox</i> , PP carrier, PES membrane, 7 m ² , 15–30 l·m ⁻² ·h ⁻¹ , 38 nm pore size, 150 kDa). The sludge concentration: 2 g/l, the sludge age: 30–50 days.	CIP	51	[127]
				ERY	<60	
				MET	45	
				NOR	47	
				SMX	7	
Biological: attached growth treatment	BAC	Wastewater (Australia)	Pilot plant. The columns are 3 m high and 22.5 cm internal diameter; they consist of 80 cm filtering bed supported by a 20 cm layer of gravel at the bottom. Contact time: 18 min.	TMP	96	[130]
				ERY	>90	
				SMX	90	
Constructed wetlands	HSF CWs	Aquaculture wastewater (China)	Plant: phragmites communis. Fill material: gravel and zeolite Hydraulic retention time (HRT) = 1d Hydraulic retention time (HRT) = 3d Hydraulic loading rate (HLR) = 25.2 cm/d	SMX	4 (HRT = 1d)	[138]
				ERY	59 (HRT = 3d)	
				TMP	90	
				SMX	90	

^a Mean \pm standard deviation.

^b n.a.: value not available.

paper on the performance of electrocoagulation-assisted biological treatment processes, determines that the electrocoagulation process is simple, cost-effective and efficient when used as a pretreatment prior to biological processes. Advantages are the elimination of toxic materials that could inhibit biocatalysts in the biological treatment stage, the reduction of the high organic load and the reduction of membrane fouling in MBR. Finally, he proposes increasing research and expresses the possibility of expanding its use to an industrial treatment scale [144].

A second paper is a review on combined electrocoagulation processes as a novel approach for enhanced pollutants removal, including the combination of electrocoagulation with chemical coagulation, adsorption, magnetic separation, and reverse osmosis. In this review the author detects the following problems that are reproduced in many investigations: On the one hand, in most of the studies carried out, synthetic wastewater was used instead of real samples. Consequently, the production results may not be representative of the real case systems. On the other hand, no attempt has been cited to increase adsorption, chemical coagulation, magnetic field, and reverse osmosis assisted electrocoagulation systems. Finally, very few studies have considered the application of kinetic models to predict these combined systems. In addition, it proposes using sustainable energy resources to cover the electrical energy required by electrocoagulation processes and thus reduce operating costs. He also expresses that more attempts must be made in search of more sustainable electrodes, apart from Fe and Al [145].

In a third paper the author reviews the main results obtained from studies on the performance of biological treatment combined with electrocoagulation as a post-treatment process. The most representative conclusion is the demonstration that electrocoagulation is an efficient and promising post-treatment process for biological treatment processes, especially for anaerobically treated effluents that need more chemical oxygen demand and removal of color. Besides, the author insists on the need for additional studies that combine biological treatment and electrocoagulation as a post-treatment process and to compare them to select the most profitable [146].

Finally, the author cites some interesting studies regarding using a combined process that consists of two subsequent biological and electrocoagulation steps [147–150], as well as of process of biological with electrocoagulation as a post-treatment process [151,152].

4. Discussion

In this section we proceed to discuss the results obtained in the bibliographic review carried out, pointing out the most representative aspects of the concentrations of antibiotics detected, the most prominent removal methods and the elimination tests consulted.

Regarding the review of the concentrations of antibiotics detected in water in international studies, it should be noted that the most studied antibiotic is sulfamethoxazole, appearing in 96% of the analyzes, while the least studied is levofloxacin, which only appears in the 4%. Too, the antibiotics with the highest concentration are Sulfamethoxazole, Trimethoprim and Ciprofloxacin (all >20,000 ng/l), while the lowest concentration is Metronidazole (1800 ng/l). Another important issue to highlight is that 53% of the studies consulted use wastewater as a source, while for example only 18% of the papers use river water as a source. Therefore, increasing the water analysis in other water sources is very interesting for the scientific community.

As to the main analysis techniques examined, highlight the great variety existing in the physical and chemical foundations on which they are based: membrane process, chemicals, absorption, filters, oxidation... This allows you to choose between one method or another depending on

the water source, the infrastructures developed in the place and the emerging pollutant that you want to eliminate.

Respect of removal percentage tests consulted, emphasize that among the different methods analyzed, those with the best elimination results (Ranges between 80 and 100%) are AOPs: OP; Membrane technology: NF & RO; Biological: BAF, AD & BAC. While the one with the worst result are Chemical: C-F; Constructed Wetlands: HSF CWs.

In general, the following criticisms and evaluations can be made. On the one hand, in biological treatments, the elimination of antibiotics is affected by factors such as process parameters, water quality conditions and environmental factors. This must be considered if this option is chosen. On the other hand, AOPs and combined treatments for the elimination of antibiotics show high efficiency. They have broad development and application prospects in wastewater treatment. Lastly, membrane technologies are effective in removing antibiotics, but it is true that they are rarely used in wastewater today. Choosing this technology is a viable option [106].

Next, the RO will be studied in more detail, not because it is a more suitable technique than the others, but because in locations with water scarcity where RO desalination plants abound (as is the case of the place where this paper is made, Canary Islands, Spain), proving the validity of this technology for the elimination of antibiotics is very encouraging. If the analysis of membrane processes is investigated to a greater extent, it is observed that there are multiple studies that prove the efficacy of RO to eliminate antibiotic concentrations in different types of water. Following, some of the main successful studies, selected for their relevance in terms of rejection of contaminants, are explained in greater detail.

First, the pilot unit of Croatia with wastewaters and an initial concentration of TIM of 406 $\mu\text{g/l}$. The operation conditions were 25 °C of temperature and 5.98 pH. The membranes utilized were XLE of Dow film Tec. The results of rejection were >99.99% [134].

Second, the theoretical/practical case of Gran Canaria (Spain) with synthetic seawater and an initial concentration of CIP of 50, 200 and 500 $\mu\text{g/l}$. The membranes used were RE2521 of CSM Toray Chemical Korea Inc. The operation conditions were in the initial phase 22–30 °C of temperature and 7 \pm 0.2 pH and in the optimization phase 25 °C and 7 \pm 0.2 pH. The results of rejection were in the initial phase: 99.72%, 97.91% and 99.55% respectively for 50, 200 and 500 $\mu\text{g/l}$ and in the optimization phase 95.96%, 99.69% and 99.83% respectively 50, 200 and 500 $\mu\text{g/l}$ [153].

The data of the elimination percentages consulted in the international literature review establish ranges from 93% to 99.99%, repeating values higher than 99% regularly. In summary, RO presents magnificent percentages of antibiotic removal in different types of water. RO is also positioned as a candidate technology, as one of the main allies in the purpose of eliminating concentrations of antibiotics in the aquatic environment, competing with other techniques such as activated carbon adsorption, Ozonation and advanced oxidation processes, attached growth treatment processes or other membrane process.

5. Future perspectives

The future perspectives go through mitigating, preventing, and controlling antibiotic resistance, which is one of the global priorities recognized by the competent authorities. Future studies should include improvement of wastewater management practices and monitoring of environmental water contamination by antibiotics. Multiclass methods are required that allow simultaneous analysis of antibiotics and their by-products at very low concentrations. All to combat concerns with antibiotic mixtures and their effects on human health and the environment derived from chronic low-level exposure [6].

The risk of development and proliferation of resistance to antibiotics is critical and adequate methods to assess this risk must be developed in future lines of research. Studies carried out in rivers of China revealed that the concentration of antibiotics depends on three major conditioning factors: First, economic factors, for example population density and its corresponding release of antibiotics; Second, geochemical factor, for example the texture of the sediments and the content of sedimentary organic carbon; Third, geographic and hydrological factors, for example rainfall and currents [154]. Consequently, the locations for monitoring the concentrations of antibiotics in the aquatic environment cannot be chosen arbitrarily. A strategy is needed to determine the strategic points where monitoring should be performed.

Research in southern Lake Michigan concludes that concentrations of pharmaceuticals can have potential harmful effects on aquatic organisms and humans through exposure to drinking water. This study shows that conducting more in-depth research to quantify potential threats is critical [155].

It is important to emphasize that risk assessments of emerging contaminants (for example antibiotics) are always helpful, but your results should be treated with caution because single compound exposure scenarios are not realistic. Multiple pollutants are reproduced in combination with effects that are unknown and of considerable ecological concern. Therefore, prospects are to carry out more specific analyzes to define possible adverse effects on the aquatic environment and whether synergistic effects between pollutants can occur [156].

As a summary regarding the fight against antibiotic resistance, add that there are authors who establish a series of major future lines of action: First, carry out prevention and awareness campaigns to reduce the consumption of antibiotics, as well as greater control of the elimination of expired drugs; Second, optimize the performance of existing WWTPs through the implementation of tertiary treatment techniques; Third, increase research on new and more innovative techniques for water treatment; Fourth, a strict and uniform regulation on the application of sludge from urban WWTPs and livestock manure as fertilizers in agricultural activities since they are sources of contamination of agricultural soils and, in some cases, of harvested crops; Fifth, conduct more research focused on the ecotoxicological risks associated with pharmaceutical contamination. This would allow deciding which compounds represent a threat to the aquatic environment and therefore must be monitored and included in the lists of priority substances defined in legislative frameworks such as Directive 2013/39/EC [157].

Also, in the future it is necessary to globalize studies. There are knowledge gaps that must be filled through studies of the fate and transport of emerging pollutants in countries located in a wider range of climates (from tropical to arctic). Especially there is a significant lack of information from countries such as Indonesia, India, Canada, Russia, and countries in Africa and South America [63].

An interesting line of investigation for the future is to use renewable sources for the elimination of antibiotics from the aquatic environment. In a study in Almeria (south of Spain), treatment by solar photo-Fenton was effectively used to remove antibiotics from real secondary effluents from two different wastewater treatment plants. In it, 7 out of 10 antibiotics detected in the investigated wastewater samples were effectively eliminated (60–100%). However, it was also shown that the process was ineffective in eliminating antibiotic resistance genes, so it was not possible for them to conclude that the process can effectively minimize the risk of transfer of antibiotic resistance to the environment. Further research on more intensive oxidative conditions is needed [158].

Another line of work for the future may be based on advancing and refining the methods that make it possible to determine a standardized elimination efficiency by compound types (for example, for antibiotics). These proceeds because there is scientific evidence that determines serious difficulties in comparing the efficacy of the different treatment processes because the elimination of organic micropollutants seems to be specific to each compound [159,160].

The prospects also include reducing the costs of eliminating

antibiotics from the aquatic environment. For example, there are relatively recent studies on hospital effluents that establish that total costs range between 4.1 €/m³ and 5.5 €/m³ in the case of incorporating secondary treatment using a membrane biological reactor and advanced oxidation processes [161].

Finally say that in countries with large coastal areas and especially in the ones that have islands with water scarcity, wastewater treatment and desalination are essential to meet their water needs. A good example is the case of Gran Canaria Island (Spain), where desalination represents approximately 45% of the total water produced, being used as follows: agricultural (14%), recreational (9%), urban (73%), tourism (73%) and industrial (60%). The technology used widely for desalination on the island is RO; it is implemented in the 85% of the existing desalination plants. Therefore, it can be said that RO in this island is a mature, reliable, and experienced technology [162]. Taking advantage of this experience and these infrastructures of RO for the objective of eliminating antibiotics from the aquatic environment is an opportunity in this type of location.

In this sense, to say that RO can also be used as one more treatment within a concatenated series of treatments.

This was done in a study in a WWTP in northern Spain in which the concentrations of 77 emerging pollutants (including antibiotics) in raw municipal wastewater and secondary treatment effluent in a WWTP were monitored for two years. In it, a wastewater treatment scheme was used that integrates activated sludge, UF, RO and electro-oxidation to eliminate them. Their results dictated that the amount of micropollutants removed during secondary treatment varied widely by compound. While the UF removal efficiency for the different compounds varied significantly, although it was less than 20% for most. Excellent removal rates were achieved in the reverse osmosis treatment. They rejected more than 99% of all target compounds. Finally, electro-oxidation with boron-doped diamond electrodes removed more than 95% of most of the compounds studied from the RO effluent [163].

6. Conclusions

The human and animal waste that is produced after the consumption of antibiotics causes the contamination of the aquatic environment. This ends up harming human health by generating resistance to antibiotics. To fight against this problem and protect the Aquatic Environment, numerous efforts are being made around the world. The problem is not easy because there are multiple routes of entry for antibiotic residues of human and animal origin into the aquatic environment and the consumption of antibiotics for medical and veterinary use has high levels in developed countries. If means for their elimination from the aquatic environment are not studied, the problem will worsen over time. Because of higher global consumption, large discharges excreted into the aquatic environment, which will be added to the existing concentrations.

In this bibliographic review, the study has focused on analyzing the concentrations detected and the elimination percentages experienced of the following antibiotics: *Ciprofloxacin*, *Erythromycin*, *Levofloxacin*, *Metronidazole*, *Norfloxacin*, *Oxfloxacin*, *Sulfamethoxazole*, *Trimethoprim*. In addition, this study includes countries around the world, as well as different types of waters: Sewage, wastewater (hospital, domestic, synthetic), water reclamation facility, rivers, surface water, drinking water and synthetic seawater.

- i. The most studied antibiotic is *Sulfamethoxazole*, appearing in 96% of the analyzes, while the least studied is *Levofloxacin*, which only appears in 4%. Regarding the concentrations detected, the antibiotics with the highest concentrations are *Sulfamethoxazole*, *Trimethoprim* and *Ciprofloxacin* (all >20,000 ng/l), while the one with the lowest concentration is *Metronidazole* (1800 ng/l).
- ii. Multiple techniques have been experimented with to tackle the removal of concentrations of these antibiotics from the aquatic

environment with different results. Among the different methods analyzed, the ones that present the best results of elimination (Range 80–100%) are: NF & RO; Biological: BAF, AD & BAC. While the one with the worst result (under 60%) are Chemical: C-F; Constructed Wetlands: HSF CWs.

- iii. RO is an effective technique with elimination percentages higher than 93% in all the analyzes reviewed of *Ciprofloxacin*, *Sulfamethoxazole*, *Trimethoprim* and *Erythromycin* in wastewaters (real and synthetic) and seawater synthetic.

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