sustainability

Review

Sources of Antibiotic Contamination in Wastewater and Approaches to Their Removal—An Overview

Antony V. Samrot [1,*], Samraj Wilson [2], Ram Singh Sanjay Preeth [3], Pandurangan Prakash [3], Mahendran Sathiyasree [3], Subramanian Saigeetha [4], Nagarajan Shobana [3], Senthilkumar Pachiyappan [5] and Vinod Vincent Rajesh [6]

1 School of Bioscience, Faculty of Medicine, Bioscience and Nursing, MAHSA University, Jenjarom 42610, Selangor, Malaysia
2 Department of Botany, St. John's College, Palayamkottai, Tirunelveli 627002, Tamil Nadu, India
3 Department of Biotechnology, School of Bio and Chemical Engineering, Sathyabama Institute of Science and Technology, Chennai 600119, Tamil Nadu, India
4 Department of Biotechnology, School of Biosciences and Technology, Vellore Institute of Technology, Vellore 632014, Tamil Nadu, India
5 Department of Chemical Engineering, Saveetha Engineering College, Thandalam, Chennai 602105, Tamil Nadu, India
6 MSU College, Naduvakurichi, Sankarankovil Taluk, Tenkasi District, Tirunelveli 627756, Tamil Nadu, India
* Correspondence: antonysamrot@gmail.com

Abstract: In the practice of medicine, antibiotics are extremely important and are employed in the treatment of infections. A lot of antibiotics are consumed by humans and excreted via urine and feces into sewage systems and treatment plants. These are considered to be non-biodegradable, and over the years they accumulate in the aquatic environment. The presence of antibiotics in water resources causes the emergence of antibiotic-resistant bacteria, posing a serious threat to the health of human beings. Water bodies must be adequately treated before being discharged to prevent the spread of antibiotic resistance. In the present article, the sources of antibiotics and strategies used for their effective removal, such as ultrafiltration, microfiltration, nanofiltration, membranous biological reactor treatment, Advanced Oxidation Process (AOP), Reverse Osmosis (RO) and Nano sorbents, are discussed. Conventional wastewater treatment plants are not able to eliminate antibiotics deposition/resistance genes effectively and efficiently. In this regard, the adsorption method is the most effective way of removing antibiotics from wastewater from various sources.

Keywords: antibiotics; recycling strategies; municipality; hospital; wastewater

check for updates

Citation: Samrot, A.V.; Wilson, S.; Sanjay Preeth, R.S.; Prakash, P.; Sathiyasree, M.; Saigeetha, S.; Shobana, N.; Pachiyappan, S.; Rajesh, V.V. Sources of Antibiotic Contamination in Wastewater and Approaches to Their Removal—An Overview. *Sustainability* **2023**, *15*, 12639. https://doi.org/10.3390/su151612639

Academic Editor: Athanasia K. Tolkou

Received: 26 January 2023
Revised: 10 March 2023
Accepted: 21 March 2023
Published: 21 August 2023

1. Introduction

Water, a renewable but limited resource available in nature, is essential to human existence as well as environmental sustainability. In certain developing nations, chemical pollution of surface waterways, mostly owing to industrial and agricultural emissions, poses a serious threat [1]. Most developing nations respond to public health protection needs through pathogen management and water management [2–4]. Antibiotics are considered the most essential drugs in hospitals to treat/prevent diseases. The improper disposal of these antibiotics in various ways causes them to reach water bodies [5]. Continual exposure to and sub-inhibitory concentrations of antibiotics pave way to bacterial resistance to numerous antibiotics, and form multidrug-resistant bacteria that not only cause resistance but interfere in the bio–geo–chemical cycle [6,7]. Besides hospital waste, municipal wastewater is considered as one of the biggest "hotspots" for antibiotics. [8]. Feces and carcasses of human and animal sources are the major sources of the introduction of antibiotics, as well as antibiotic-resistant microbes, into water ecosystems. These bacteria can transmit their genes to waterborne pathogenic microorganisms too, thus inducing resistance [9,10]. Thus, antibiotic resistance could become a critical issue in modern medicine, as it poses the threat of catastrophic epidemics [11]. It becomes highly essential to ensure adequate processing before release into the environment, or else these substances can cause severe pollution and disturb the natural balance. Nevertheless, eliminating antibiotics from wastewater is a major challenge [12]. The most common conventional methods used in removing these antibiotics are chemical precipitation, ion exchange, biosorption, reverse osmosis, nanofiltration, etc. [13]. Each process's characteristics, as well as biological and chemical oxygen demand, water quality conditions, and environmental factors, influence the antibiotic elimination efficiency [14]. Irrespective of all of this, antibiotics are not entirely eliminated in wastewater treatment facilities, as they have been found in stagnant wastewater ponds, municipal sewage, hospital sewage, surface water, and groundwater [15–17]. At present, membrane technology is considered a promising method for removing antibiotics from effluents [12]. An efficient treatment or conservation method would have a positive impact, playing a key role in delivering noteworthy benefits in terms of economic development, either directly or indirectly [18]. In this paper, various sources of antibiotics, and the different strategies for their removal, are outlined in order to help prevent the emergence of antibiotic-resistant genes and bacteria that give rise to new diseases, and to help combat the adverse effects on humans and the environment.

2. Antibiotics in Water

Antibiotics are categorized into 11 types based on their chemical structure, including aminoglycosides, lactams, glycopeptides, macrolides, oxazolidinones, polymyxins, quinolones, streptogramins, sulfonamides and tetracyclines [19]. Tetracyclines, sulfonamides, and macrolides are common antibiotics used in human and veterinary medicine. These antibiotics are used to ensure and protect the health of animals, as well as to diagnose and treat infectious disorders in humans [20]. The most popular and oldest antibiotic is Penicillin (a category of lactams), which contains naturally occurring penicillin G, extended spectrum penicillin–piperacillin and aminopenicillins–ampicillin [21]. The most commonly found antibiotics in effluent, sewage sludge, sediments, soils, and foods are sulfonamides, macrolides, and quinolones. Thus, antibiotic pollution has become a severe matter of concern in many rivers and seas, as it can bioaccumulate in aquatic creatures, leading to a loss of biodiversity [22,23]. The presence of antibiotics could pose threats to the food chain, limiting soil decomposition, threatening marine/fresh water/terrestrial creatures, and increasing bacterial resistance [24,25].

2.1. Source

As mentioned earlier, antibiotic-based pollution is being found in soil/water resources/sewage [26,27]. Since it is harmful to human health and ecosystems, addressing its presence has to be considered a top priority, as it causes bacterial resistance and makes treatment against any infection ineffective. These drug-resistant bacteria could enter humans/higher animals through contaminated water or through food made using that water, and impact the gut microflora within the hosts [28] leading to several health ailments. Finding the source of contamination would help us to prevent the entry of antibiotics into humans or any living system. The sources of antibiotics are discussed below.

2.1.1. Domestic Wastewater

Antibiotics enter the municipal and surface waterways thanks to inadequate treatment or incorrect disposal by users [29], and the major sources of antibiotics are industries (slaughter house/poultry) and domestic household waste (Figure 1). The antibiotics taken by human beings and animals reach the intestines, are excreted out, and then reach the wastewater treatment plants [30]. Although municipal wastewater is the greatest source of pharmaceuticals entering the environment, other sources include the inadequate treatment of manufacturing effluents and the direct dumping of unused medicine. The concentrations and types of antibiotics depend on the chemicals used in the industry or by the consumer.

After reaching the treatment plants, the antibiotics undergo some modification (either physical or chemical), and their destiny depends on the type of treatment and process. Antibiotics based on the major six classes—tetracyclines, β-lactams, quinolones, macrolides and sulfonamide—are identified commonly in activated sludge, sewage, digested sludge, and effluents as pseudo-persistent pollutants [19]. Municipal wastewaters contain the highest quantities and most diverse populations of micro- and macro-organisms, which increases the emergence of drug-resistant organisms. These organisms might be commensals, pathogens, or environmental bacteria/protozoan/fungi, but they are not unexpected to have antibiotic-resistant characteristics [31,32]. Despite the presence of heavy metals, pesticides, endocrine disruptors, and hormones in wastewater, dealing with the development of resistant pathogenic microbes is the most important issue here [33,34]. The global prevalence of antibiotics in wastewater treatment plants is outlined in Table 1. The concentrations and types of antibiotics are more diverse in developed countries compared to underdeveloped countries.

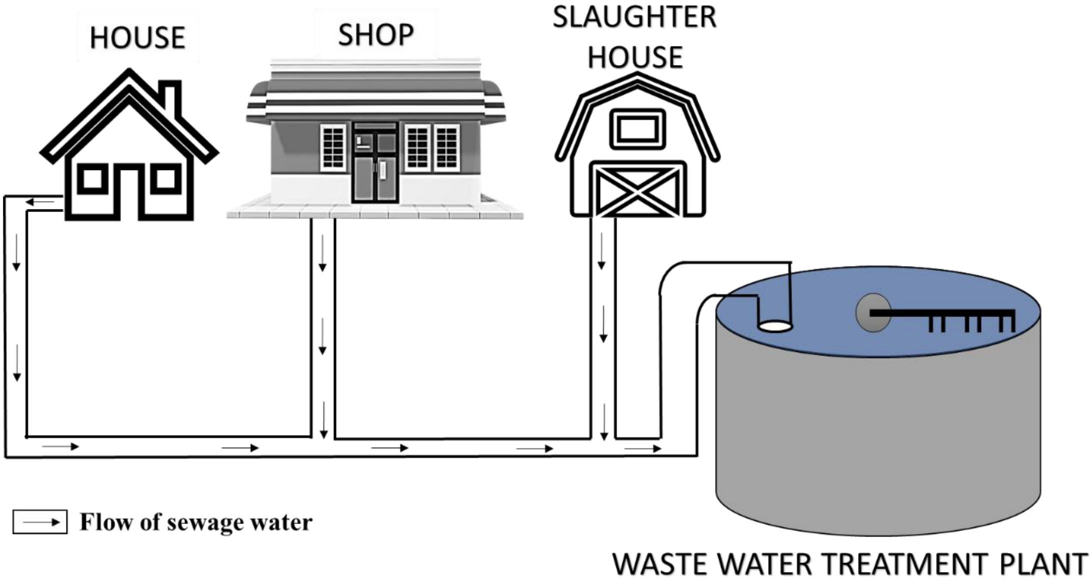

Figure 1. Effluent discharge of municipalities into wastewater treatment plants.

Table 1. The antibiotics found in various regions of world.

S. No.	Antibiotics	Concentration (ng/L)	Region	References
1.	Cefalexin	38.4	WWTP of Portugal	Mozaz et al. [35]
	Trimethoprim	69.1		
	Ciprofloxacin	231.4–584.9		
	Ofloxacin	89.7–184.9		
	Clindamycin	8.5–86.6		
	Azithromycin	178.9–597.5		
	Clarithromycin	74.2–313.2		
	Pipemidic Acid	10.8–20.1		
	Sulfamethoxazole	7.1–30.2		
	Sulfapyridine	4.7–84.5		
	Tetracycline	147.5–231.2		

Table 1. *Cont.*

S. No.	Antibiotics	Concentration (ng/L)	Region	References
2.	Tetracycline	70–370	Wastewater treatment facilities Wisconsin, USA	Riyami et al. [21]
	Trimethoprim	120–550		
	Sulfamethoxazole	50–370		
	Macrolides	300		
	Fluoroquinolones, in the form of ciprofloxacin	40–140		
3	Ofloxacin,	96–7870	Sewage treatment plants (STPs), Hong Kong, South China	Leung et al. [36]; Riyami et al. [21]
	Norfloxacin	35–4000		
	Cefalexin	180–4000		
	Erythromycin	250–4000		
	Sulfamethoxazole	5–300		
	Trimethoprim	60–450		
4.	Ciprofloxacin	1270	Al-Wihda (W1) and Al-Rasheed water treatment plants (R1) Baghdad City, Iraq	Mahmood et al. [37]
	Levofloxacin	177		
	Amoxicillin	1500		
5.	Metronidazole	2600	Rural hospital in Vietnam	Lien et al. [5]
	Sulfamethoxazole	9800		
	Trimethoprim	7700		
	Ceftazidime	1200		
	Ciprofloxacin	42,800		
	Ofloxacin	4600		
	Spiramycin	1700		
6.	Ampicillin	12,680	Yamuna, Delhi, India	Mutiyar and Mittal. [38]
	Ciprofloxacin	8000		
	Gatifloxacin	1220		
	Sparfloxacin	140		
	Cefuroxime	220		
7.	Ciprofloxacin	2200–236,600	Hospital in Ujjain, India	Diwan et al. [39]
	Norfloxacin	6400–29,600		
	Levofloxacin	5000–8800		
	Ofloxacin	4500–7500		
8.	Amoxycillin	90	Watersheds of Southeast Queensland, Australia	Watkinson et al. [40]
	Cephalexin	4100–10,000		
	Nalidixic acid	20–40		
	Enrofloxacin	60–100		
	Clindamycin	4–90		
	Lincomycin	6–1700		
	Roxithromycin	50–400		
	Doxycycline	130–200		
	Sulfamethoxazole	100–300		
	Trimethoprim	300		

Table 1. *Cont.*

S. No.	Antibiotics	Concentration (ng/L)	Region	References
9.	Metronidazole	0.17	WWTPs in Durban, South Africa	Faleye et al. [41]
	Erythromycin	0.14		
	Ofloxacin	0.22		
	Trimethoprim	0.11		
	Azithromycin	0.11		
10.	Tetracycline	37	WWTPs located in Turku, Tampere and Helsinki, Finland	Kortesmäki et al. [42]
	Carbamazepine	47–417		
	Sulfadiazine	326–1069		
	Trimethoprim	170–490		
	Clarithromycin	50–327		
	Roxithromycin	60–145		
11.	Sulfapyridine	90–150	Wastewater treatment plant Kloten-Opfikon Zurich, Switzerland	Göbel et al. [43]
	Azithromycin	170–380		
	Clarithromycin	380–600		
	Sulfamethoxazole	430–570		
12.	Norfloxacin	72–155	Sewage treatment plants, Sweden	Lindberg et al. [44]
	Ofloxacin	287		
	Ciprofloxacin	90–205		
	Sulfamethoxazole	144–674		
	Doxycycline	2480		
13.	Tetracycline	286.7–582.5	Linan City, Eastern China	Li et al. [45]
	Sulfonamides	909.9–2741.3		
	Sulfamethazine	1159		
14.	Sulfamethoxazole	136.7–426.0	WWTPs and the downstream water adjacent to WWTP, Eastern China	Li et al. [46]
15.	Amoxicillin	0.11	Essonne district, France	Dinh et al. [47]
	Sulfamethoxazole	2.1		
	Ofloxacin	17.9		
	Vancomycin	3.6		
	Norfloxacin	12.1		
	Ciprofloxacin	5.8		
16.	Roxithromycin	44.7–130.2	Han River, Republic of Korea	Choi et al. [48]
	Trimethoprim	27–89		
	Chloramphenicol	54		
17.	Sulfamethoxazole	65–200	Girona Catalonia, Spain	Ekwanzala et al. [49]
	Clindamycin	184–1465		
	Azithromycin	85–113		
	Ciprofloxacin	5329–7494		

Table 1. *Cont.*

S. No.	Antibiotics	Concentration (ng/L)	Region	References
18.	Clindamycin	41	Sewage Treatment Plant in Dresden Kaditz, Germany	Ekwanzala et al. [49]; Rossmann et al. [50]
	Doxycycline	249		
	Azithromycin	285		
	Ciprofloxacin	422		
19.	Oxytetracycline	0.641	WWTPs in metropolitan regions of Porto Alegre, Capital of Rio Grande do Sul State, Brazil	Bisognin et al. [51]
	Metronidazole	0.023		
	Sulfamethoxazole	0.980		
	Paracetamol	13.64		
	Tetracycline	0.042		
	Ciprofloxacin	0.385		
20.	Carbamazepine	379	WWTPs in the Po Valley, in Northern Italy	Verlicchi et al. [52]
	Metronidazole	42		
	Ofloxacin	400		
	Norfloxacin	210		

WWTPs—wastewater treatment plants.

2.1.2. Effluent from Hospitals

Contaminated water from hospitals contains pharmaceutical materials, disinfectants, organic halogens, radioisotopes, X-ray contrasted media, heavy metals, cytostatic agents, residues of chemical compounds used in laboratories, and microorganisms of all kinds. Anti-neoplastic medications, among many of the other compounds used in medical therapy, are thought to pose a danger to the environment because of their mutagenic, teratogenic, and carcinogenic potential, whereas cytostatic medicines are subject to severe production and handling regulations [53–55]. Therefore, hospital wastewater is extremely dangerous, as it is a major source of bacteria and genes resistant to antibiotics [27,56] (Figure 2). Generally, wastewater discharged by hospitals cannot be treated effectively using conventional wastewater treatment technologies, and there is a possibility of these contaminants being discharged into the ecosystem either partially or completely [27,57]. The contribution of hospitals to the pharmaceutical load in wastewater is hard to determine, as contraceptives and medicines are widely utilized by the general public. Various initiatives are now underway to assess and screen hospitals, surveying their potential role as sources of drugs and other substances giving rise to multi-drug-resistant bacteria [58]. Compared to common wastewater, hospital wastewater contains more than 25% antibiotics, with gene concentrations ranging between approximately 0.4 log and 1.8 log [59]. A range of compounds, in addition to pharmaceuticals, are used in hospitals and surgeries for medical purposes, such as in diagnostics and disinfectants [60]. In hospital effluent, many antibiotics, including enrofloxacin, ciprofloxacin, oxalinic, ofloxacin, norfloxacin, sulfapyridine, trimethoprim, and metronidazole, have been found in higher concentrations [61,62].

2.1.3. Effluent from Slaughter Houses

Wastewater from animal slaughterhouses is considered as another significant source of resistant bacteria, as clinically administering antibiotics to animals plays an important role in their dissemination in the environment [63]. In recent times, antimicrobial resistance in bacteria linked to livestock has increased hugely due to the improper and frequent use of antibiotics in treatments, and physical growth hormones are manipulated in animal husbandry, which has encouraged the emergence of new resistances [64,65]. According to Bachmann et al. [66], large volumes of effluents from the slaughtering process may contain bacteria that are resistant to antimicrobials. Because of their high contents of lipids, proteins, fibers, organic contents, pathogens, and medications of veterinary use, meat processing effluents are regarded as detrimental on a global scale [67]. For example, oxytetracycline and procaine penicillin G are two of the medications that are often used to treat cattle in central African nations. These medications are excreted by the animals through their feces or urine, and then enter the sewage water system [68].

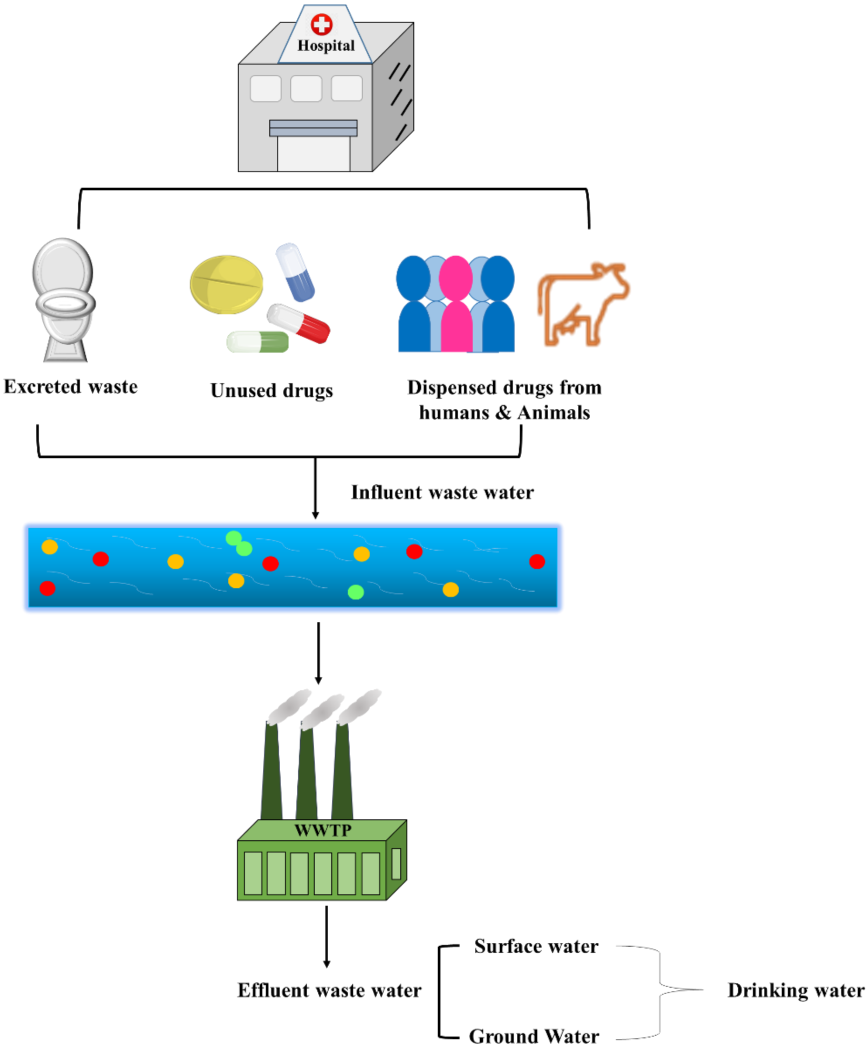

Figure 2. Sources of effluents from hospitals.

3. Runoffs

Antibiotic consumption patterns in developing countries are influenced significantly by the widespread overuse and misuse of drugs, due to their over-the-counter availability. In consequence of their extensive use, as well as their continued drainage via surface runoff and release from wastewater treatment plants, antibiotics can be found in a number of environmental sectors in these countries [69]. This causes antibiotic pollution, resulting in greater antibiotic concentrations in the environment. Thus, antibiotics are being introduced to the environment by a variety of sources, including veterinary waste, human waste streams, livestock husbandry waste, hoarded animal dung, etc. These resources could unleash antibiotics into surface and groundwater, which most often is catalyzed by moderate to heavy rainfall and then begin to move. This has thus become the most common cause of antibiotic contamination in the environment. Furthermore, this runoff is capable of transmitting contaminants from manure to surface water systems in fertilized agricultural fields. Thus, using manure, insecticides and pesticides in cropland also increases the likelihood of the release of bacterial resistance or resistance genes into surface water. In addition, seasonal factors such as snow, precipitation, nutrients, and stream velocity also influence the quantity and the mobility of pollutants [70]. The partitioning of antibiotic losses between runoff and sediment is considerably influenced by antibiotic type [71]. Due to their high water affinity or solubility in water, antibiotics have spread in the environment in a much quicker and more widespread manner [72,73]. In such a context, when effluents from healthcare institutions are dumped straight into a wastewater system without being treated beforehand, it adds to the problem, and this then emerges as one of the major reasons for antibiotic contamination [74]. As mentioned earlier, antibiotics disposed of as solid trash can be washed into surface water bodies by rain, or they can permeate into groundwater from dump sites or be taken to nearby water bodies, as illustrated in Figure 3 [75,76].

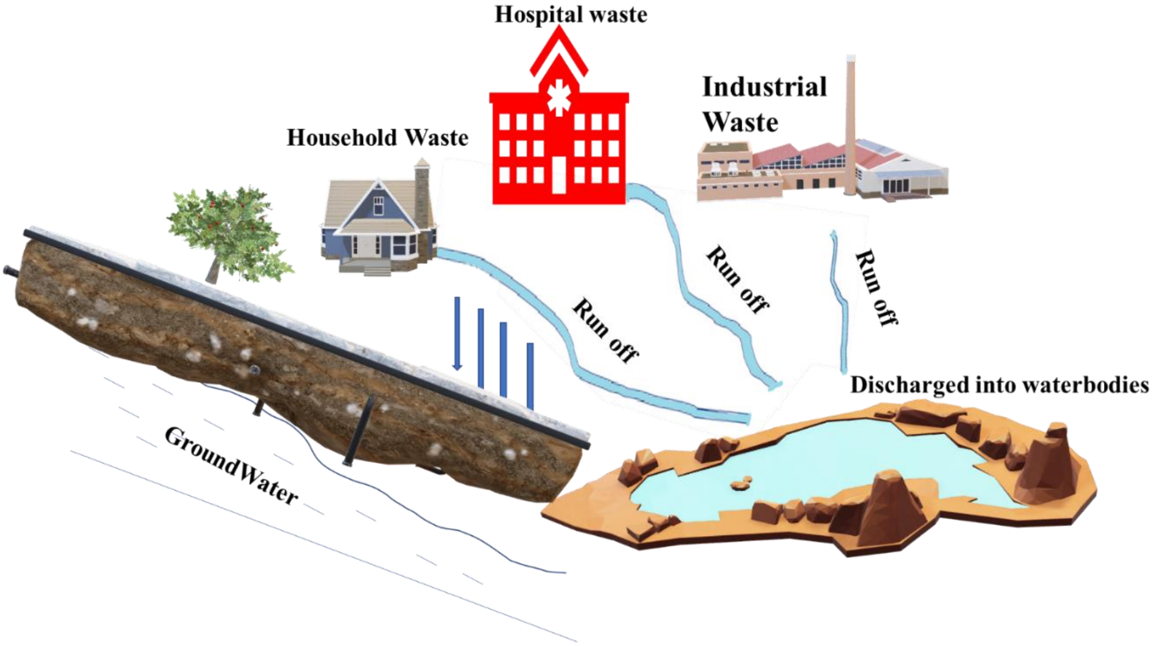

Figure 3. Antibiotic contamination by runoffs.

4. Impact of Antibiotics in Wastewater

The most utilized therapeutic drugs in hospitals include contrast media analgesics, anti-inflammatory drugs, laxatives, and antibiotics. The significant and leading sources of these chemicals, are antibiotics, which enter the multiple process of municipal sewers, digested sludge, activated sludge and then to urban bio-solids. If these antibiotics are not removed during water treatment, they can pose a major threat to aquatic life when discharged [77–80]. There are reports that antibiotics have been found in wastewater treatment plant effluent, lakes, rivers, groundwater, and even drinking water. Sulfonamides, tetracyclines, and macrolides are the most commonly identified antibiotic classes present in swine effluent, with concentrations of 324.4, 388.7, and 72 g/L, respectively [81,82]. As the accumulation of these antibiotics is observed under conditions of sub-inhibitory concentrations, the bacteria can slowly become resistant to those antibiotics, and this resistance then starts spreading among bacteria. When the concentrations of antibiotics are high, they show toxicity against aquatic species and adversely affect physical growth, the development of internal organs, reproduction and lifespan, causing ecological imbalance. It has also been demonstrated that macrolides and sulfonamides could inhibit the growth and development of algae, and could also damage plant cells and their photosystems and lower the rate of carbon dioxide oxidation [83]. The impacts of antibiotics are further discussed in detail under the following subheadings.

4.1. Emergence of Antibiotic Resistance

Antibiotic resistance is the mechanism by which an organism learns to live in the presence of antibiotics, by making antibiotics compatible, by lysing the antibiotics, or by modifying the antibiotic-binding site, and they thus protect themselves from the effects of antibiotics [84]. All the aforementioned mechanisms are made possible if the genes of the organisms are mutated. In recent decades, antibiotic resistance genes have evolved and spread across pathogenic and non-pathogenic bacteria [85]. In wastewater treatment plants, a wide range of bacteria, antibiotics, metals, and other contaminants interact with the environmental bacteria, making them a hotspot for the evolution of antibiotic resistance [86–88]. There is thus an increased chance of the emergence of antibiotic-resistant bacteria/antibiotic resistance genes following inadequate wastewater treatments, which are well reported by researchers in the field [89]. Similarly, multi-drug resistance in *Acinetobacter* sp. linked to wastewater treatment has also been reported [90]. The presence of antibiotics increases the chance of quorum sensing and riboswitches, and leads to increased phenotypic variability and increased virulence [91]. Mutation and gene transfer by conjugation, and transformation or transfection from bacteria to bacteria, can increase the virulence of pathogens, and this could cause serious healthcare issues [91–94]. In a study, it was reported that resistance to aminoglycosides, tetracyclines, and macrolides is conferred by point mutations in ribosomal proteins, while resistance to rifampicin is conferred by point mutations in the RNA polymerase [92]. In a study, over 35 mutations were observed in multidrug-resistant *S. aureus* and *M. tuberculosis*. The genome sequencing of antibiotic-resistant *M. tuberculosis* strains revealed 29 independent alterations in a multi-drug resistant strain and 35 mutations in an extremely drug-resistant strain [95]. Moreover, seven amino acids of *E. coli*'s gyrA gene product were reported to cause antibiotic resistance [96].

4.2. Emergence of New Disease

As antimicrobial-resistant genes are potentially transferable from one microbial community to another via conjugation/transformation, many studies suggest that wastewater could be an important source of antimicrobial resistance in microorganisms [97,98] (Figure 4). The development of antimicrobial resistance in infectious disease-causing bacteria can be described in three steps: 1. The entry of the gene into environmental bacteria in a wastewater treatment plant. 2. The passing of the gene between environmental bacteria in water bodies where the treated water is discharged. 3. Entry into the host, causing disease (now antibiotics cannot work effectively) [99]. Water from wastewater treatment plants could be considered a source of the emergence of new diseases [100]. Approximately 700,000 people die every year from infections caused by resistant organisms, and that number is expected to rise to over 10 million by 2050, according to the UK Review on Antimicrobial Resistance [101]. Some of the pathogens containing genes of resistance to many types of antibiotics, such as methicillin-resistant *Staphylococcus aureus* (MRSA), vancomycin-resistant *Enterococci* (VRE), and *E. coli* carrying extended-spectrum beta-lactamases (ESBL), are known as "superbugs" [91]. Further, the term "superbug" refers to microbes with increased morbidity and mortality due to mutations that provide resistance to different types of antibiotics, including aminoglycosides, fluoroquinolones, trimethoprim, etc. [102]. Carbapenemases-encoding genes are considered to have originated in environmental microorganisms, and NDM-1-carrying bacteria have been found in public drinking water in New Delhi [103]. Likewise, high rates of vancomycin- and ampicillin-resistant *E. faecium* are found in hospital effluents in the east of England, United Kingdom [104]. Additionally, a study by Cahill et al. showed that Carbapenemases-producing *Enterobacteriaceae* (CPE) are a type of AMR found in high numbers in hospital wastewaters in Ireland [105]. In a report, nearly 70% of *S. aureus* strains were found to be resistant to erythromycin and penicillin [91]. More severe diarrhea and higher mortality rates will result, especially if antibiotic resistance increases, which prevents effective treatment [106]. In a 2013 report, the Center for Disease Control and Prevention (CDC) stated that the use of antibiotics in the production of animal feed has encouraged an increase in antibiotic-resistant *Campylobacter*, which caused gastroenteritis in around 2 million individuals in the USA [107–109].

4.3. Immune Response

The immune system is an effective line of defense against infection, and protects against disease organisms [110,111]. Even though antibiotics have several beneficial characteristics, they do have a darker side, as they could disturb normal microbiota/immunity [112]. As such, they can also cause allergic hypersensitivity responses, toxic consequences, nephropathy, carcinogenicity, hepatotoxicity, mutagenicity and antibiotic resistance, among other problems [113–115]. Continuous exposure to antibiotics could lead to enterocolitis, as the normal state of the intestine is disturbed by antibiotics [116]. Antibiotics consumed by females before or during pregnancy could affect the child [117]. Newborn mice born from antibiotic-treated parents died when treated with higher vaccinia viral loads, whereas mice born from normal parents withstood the viral exposure, suggesting that antibiotics could reduce the level of immunity [118].

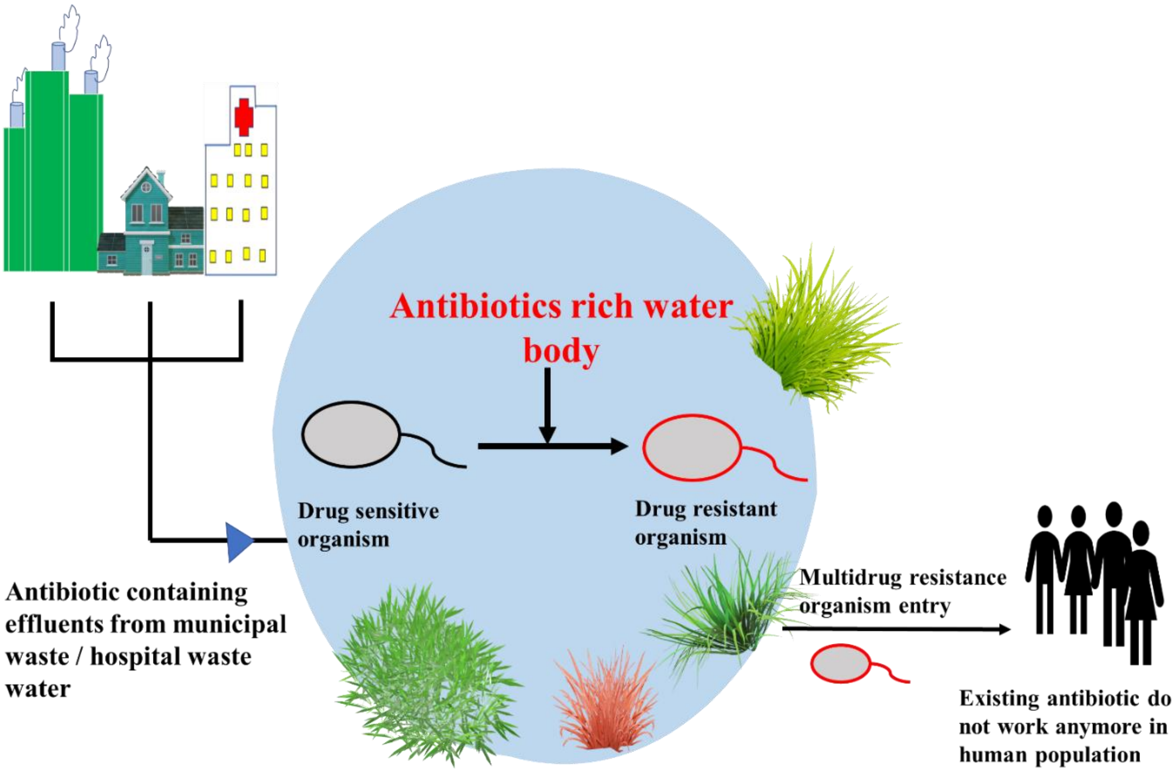

Figure 4. Emergence of new disease due to antimicrobial resistance genes (ARGs).

5. Strategies of Antibiotics Removal

It is well understood that antibiotics should be removed from water, but they are resistant to conventional treatments and purification techniques [119,120]. There are a few =methods that can be considered successful in removing antibiotics from water, and these are discussed in detail in the below section.

5.1. Membrane Technology

Pharmaceutical companies and wastewater treatment plants use conventional processes including filtration, flocculation, sedimentation, etc., but they are not effective in removing antibiotics [120]. One of the better technologies for removing antibiotics is membrane technology, which has the benefits of high productivity, ease of use, and low cost (Figure 5). The three primary concepts that govern the treatment using membranes are adsorption, sieving, and electrostatic phenomena. Membrane separation processes rely on hydrophobic interactions between the membrane and the analyte. The size of the pore and the size of the molecules determine the separation of materials via the membrane [121]. The pore sizes differ from 2 nm to 100 nm depending on the type of filter chosen [120]. Various membrane technologies, including microfiltration, ultrafiltration, nanofiltration, and reverse osmosis, which operate through electrostatic and physical forces, are used for removing antibiotics/antibiotic resistance genes from aqueous solutions such as water [119,122]. Thin, light-weight separation membranes with low filtering times and higher adsorption capacities are of greater use [123]. Ceramic-based membranes are more advantageous than polymeric membranes as they can tolerate microbial degradation and high-pressure situations [124]. Membrane filtration is more widely used in municipal and industrial water/wastewater treatment, as it provides a cost-effective solution for long-term water reuse [125]. A variety of antibiotics, such as Chlorotetracycline, Rifampicin, Cefixime, Oxy tetracycline, sulfa drugs, Doxycycline, Tetracycline, etc., are effectively removed using reverse osmosis [126–128]. Arefi-Oskoui et al. [127] and Javad et al. [128] reported that adding nanoparticles or nanocomposites to membranes does increase their potential ability to remove antibiotics. Membrane technology, however, only changes the state of the antibiotic. If direct or indirect release of concentrations in ecosystems is not prohibited, antibiotics can still recirculate and thus pose a risk of harm in specific regions [129,130].

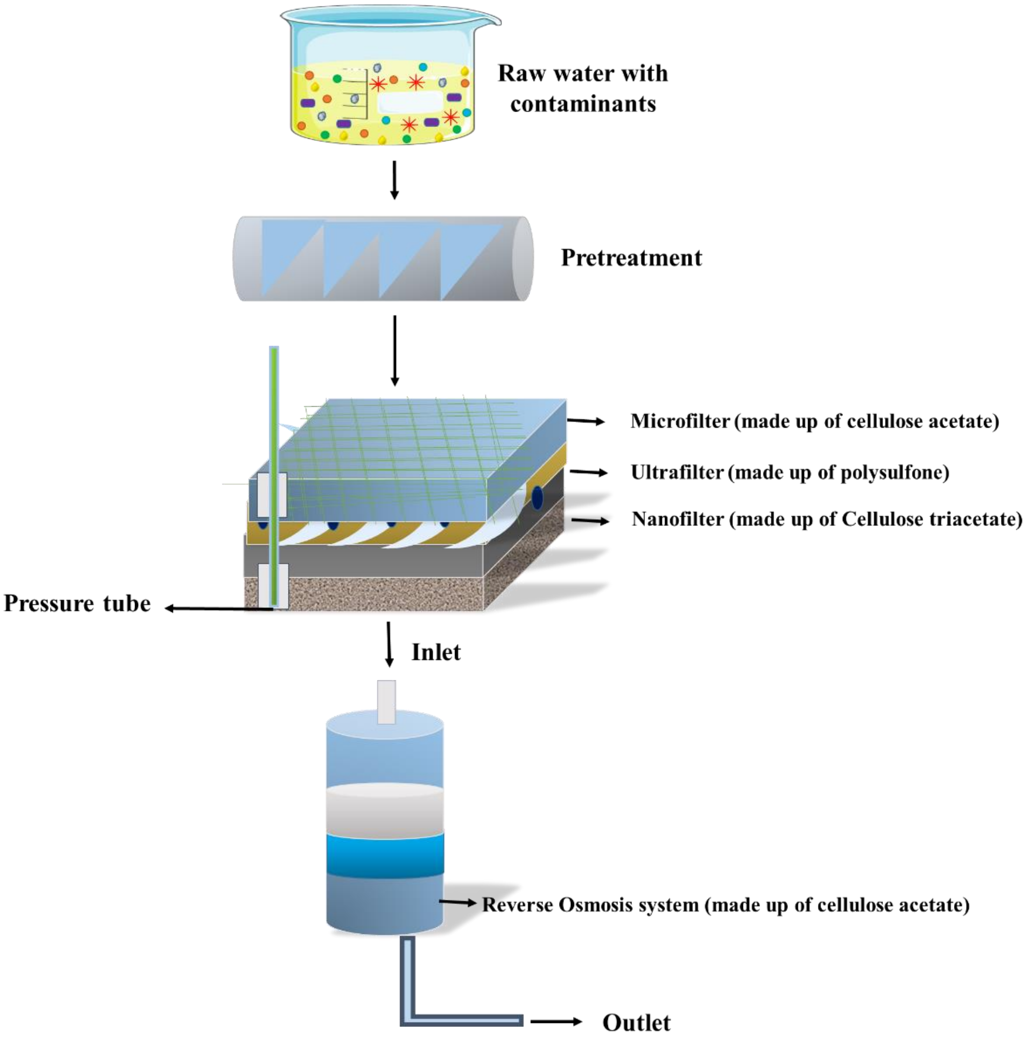

Figure 5. Membrane technology.

5.1.1. Biological Aerated Filter (BAF) Process

For the removal of municipal secondary effluents, such as antibiotics, ammonia, nitrogen, carbon, and total nitrogen, Biological Aerated Filters (BAFs) have been employed [131].

Due to their high organic loading, low cost, and lower energy consumption, BAFs have been used in wastewater treatment plants. The filter media supports the growth of microorganisms and the setup includes three phases: a stable segment that acts as the guide media for the growth of microorganisms, a liquid segment in which the stable fabric is submerged, and a gas segment formed by injecting the air into the reactor [132,133]. As a result of its high biofilm adhesion ability and wide specific area, it enhances the sludge adsorption, resulting in a high antibiotic removal efficiency [134]. The major advantage of BAF systems is that they are compact and take only one-third of the space needed by conventional activated sludge systems. When compared to trickling filters, a BAF system maintains a high biomass concentration, and is resistant to organic and hydraulic load shocks. In addition, the system does not require any further treatment, because the suspended particles in wastewater are filtered using submerged media [135]. A study carried out by Chen et al. [133] reported the removal of around 80–90% of antibiotics by sulfonamides and diaminopyrimidines, such as sulfamonomethoxine, sulfachloropyridazine, sulfamethazine, trimethoprim, and a class of fluoroquinolones including norfloxacin, ofloxacin, lincosamides such as lincomycin, and macrolides including leucomycin and tetracyclines such as oxytetracycline. These are also efficiently used to remove sulfonamides at a rate of 51%, fluoroquinolones at 20% and ciprofloxacin with an efficiency of above 95% [132].

5.1.2. Ultrafiltration

Ultrafiltration (UF) is determined by a variety of parameters, which include load and particle size (Figure 5). UF is not an efficient technique for separating organic streams, but holds the ability to sustain species with molecular weights ranging between 300 and 500,000 Da and pore sizes ranging from 10 to 1000 Å [119]. Ultrafiltration membranes are widely used for water filtration, which removes harmful bacteria, macromolecules, and suspended matter with the consumption of less energy. However, there are certain drawbacks, which include the inability to remove any dissolved inorganic pollutants from water and the requirement of high pressure for regular cleaning [121]. According to Li et al. [136], a membrane system composed of reverse osmosis and ultrafiltration is more efficient for processing oxytetracycline waste and recovering oxytetracycline from wastewater, with a 60% recovery ratio and 80% purity levels. According to a study conducted by Liang et al. [137], the use of membrane technologies such as ultrafiltration and reverse osmosis achieves 99.79% effectiveness in the removal of ARGs and bacteria. In comparison to the conventional approach, ultrafiltration has a high turbidity rate, as well as filtration accuracy, and the effluent water quality is stable and reliable [138,139].

5.1.3. Microfiltration

Microfiltration is a filtration method in which the pores have a diameter of 1 to 10 µ. These pores cannot be crossed by microorganisms. This approach is considered as the initial step in the pre-treatment prior to nanofiltration and reverse osmosis [121]. This method is used to separate colloidal particles via a screening mechanism that is employed to retain substances larger than the pore size [119]. The membrane material in MF can be either organic or inorganic. Polymers such as polyvinylidine fluoride, polysulfone, cellulose acetate and polyamide are used in organic membranes, while porous alumina and metals are used in making inorganic membranes. This process is good for isolating suspensions and emulsions, and can be used to remove sediment, algae, protozoa, and bacteria, while allowing monovalent ions such as Na^+, Cl^-, dissolved organic materials, and tiny colloids and viruses to pass through the membrane [140]. Microfiltration provides the benefit of increased oxidizable material removal, with a rate of around 90%. One of the most significant drawbacks of MF is that it cannot remove impurities (dissolved particles) smaller than 1 mm in size. MF, however, appears to suppress harmful microbes in water when applied in conjunction with disinfection [121]. Qiu et al. [141] reported the removal of antibiotics such as enrofloxacin, sulfamethazine, cefalexin, lomefloxacin, amoxicillin and ampicillin with an efficiency of 58.9 to 100% using a hybrid microfiltration–forward osmotic membrane bioreactor, but were not able to remove low-molecular weight antibiotics such as sulfonamides and trimethoprim.

5.1.4. Nanofiltration

One of the main techniques involves using nanoparticles [142–144]. Nanofiltration is an effective method for the removal of large organic materials and average-sized minerals, but is not applicable to low-molecular weight organic substances such as methanol, because these membranes operate at very low pressures (Figure 6) [119]. Membranes used for nanofiltration are made of cellulose acetate blends or polyamide composites. The modified versions of UF membranes include sulfonated polysulfone. In the nanofiltration process, the wastewater is fed into the inlet and the incoming stream is separated into two parts: the permeate, which is the filtered component, and retentate, or concentrate, which is the non-filtered fraction that is rejected. NF is capable of successfully removing organic matter, and works on the principle of the application of hydrostatic pressure, which is used to convey a molecular mixture to the membrane's surface. When pressure is applied, solvents and low-molecular weight solutes pass through the membrane, while other components are trapped. This is sufficient to eliminate ions that contribute significantly to osmotic pressure, resulting in reduced operating pressures. As soluble fractions cannot be eliminated by nanofilter membranes, highly contaminated fluids require a pretreatment process [140]. Nanofiltration membrane's removal rates depend on the target antibiotic molecule; where macrolides have been removed with an efficiency rate of 80 to 95%. In a study using self-made polyethersulfone membrane nanofilters, the removal efficiency of the antibiotic amoxicillin was found to be 99.09% at a temperature of 298 K and pressure of 2 MPa [145,146].

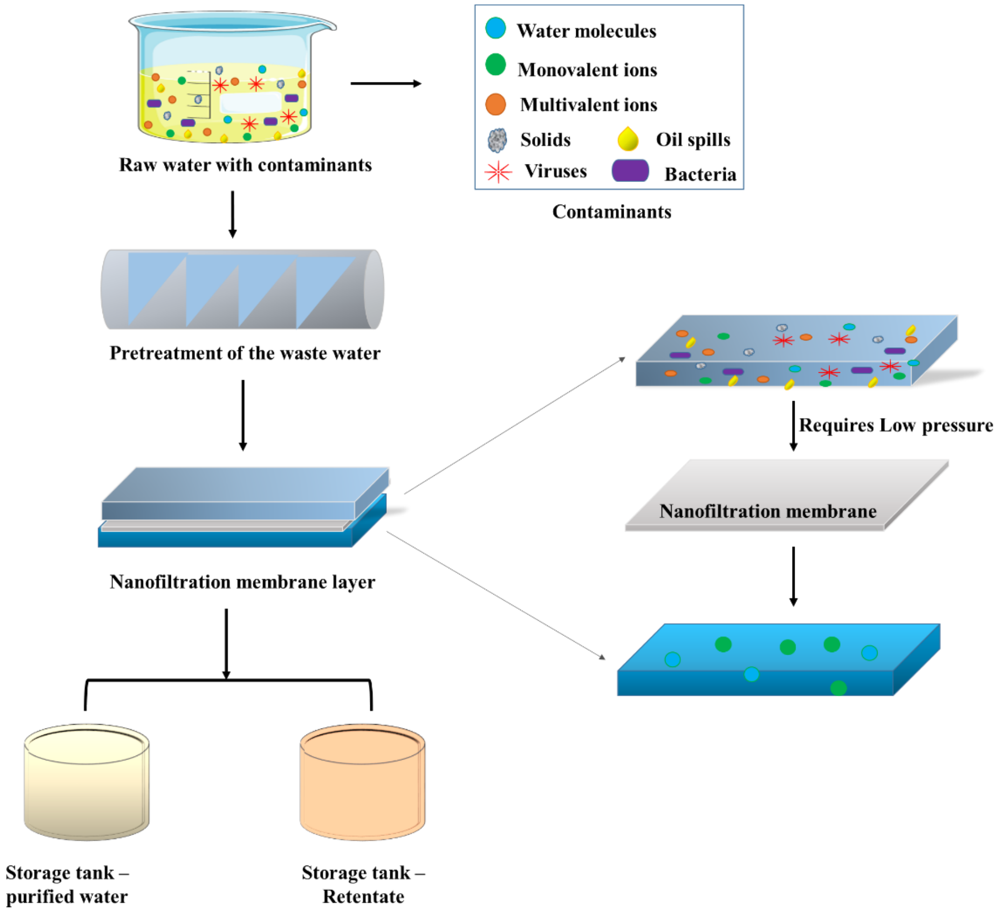

Figure 6. Nanofiltration.

5.1.5. Reverse Osmosis

Reverse osmosis (RO) is a method whereby large ions and molecules are typically removed from liquid effluents using the process of diffusion, which involves the application of pressure to the solution on one side of a semipermeable membrane. The impurities are confined on the pressured side of the membrane, while the clean effluent flows against the concentration gradient [147]. The reverse osmosis technique can be used to remove bacteria, viruses, dissolved solid particles, and microbial agents present in wastewater. This method is mostly employed to desalinate water [119]. The main physiochemical properties considered for the process of reverse osmosis are porosity and mechanical resistance. In this context, these membranes must be chemically and microbially resistant, as well as mechanically and structurally durable over long periods of time, and they can be eliminated by the use of polymer membranes [147]. For amoxicillin, the removal rate via RO membranes ranges from 73.52% to 99.36%, while for ampicillin, the removal rate and permeate flux range from 75.1% to 98.8% [148]. Further, the removal rates of antibiotics such as trimethoprim dexamethasone, febantel, ciprofloxacin and sulfamethoxazole using a hybrid of reverse and nanofiltration were observed to be 97%, according to Dolar et al. [149]. Despite the apparent benefits of utilizing an RO membrane, it is still unclear whether it can act as a complete barrier to enhance the removal of organic micropollutants. Several top scientists have identified RO as the most effective and promising method for removing organic micropollutants [150,151].

5.1.6. Membranous Biological Reactor

Membranous Biological Reactors are a form of biological wastewater treatment system that blends membrane separation technology with biological technology. They have a chamber in which biological matter is separated by a membrane, a micro-filter with a size of roughly 1–10 μm [119]. This is a wastewater treatment or resource recovery method that incorporates biological processes such as activated sludge with membrane processes such as ultrafiltration, nanofiltration, and microfiltration [130]. The use of a Membranous Biological Reactor has many advantages, including a long sludge holding period, ease of operation, low sludge output, and excellent nitrification performance [134]. Filtered wastewater derived from these reactors has a comparable quality to that of secondary sewage treatment, and exhibits similar microfiltration output. These reactors are excellent for use in wastewater treatment and reuse in both urban and industrial settings. These reactors are demonstrated to be effective for use in both aerobic and anaerobic wastewater filtration [119]. The MBR process has emerged as an effective wastewater treatment method due to its ability to fill in the gaps left by traditional activated sludge processes, such as their inability to deal with fluctuations in effluent flow rates and composition, and their inability to comply with higher effluent discharge limits related to reuse. In comparison to traditional treatment systems, membrane bioreactors also save a lot of space [130]. In a study, the removal of 15 organic micropollutants using a laboratory-scale membranous biological reactor was evaluated, and it was discovered that most organic micropollutants were removed at an efficiency rate of 80–92% [152]. It was also found that 100% of roxithromycin and norfloxacin and 99% of sulfadimidine could be removed using membrane bioreactors, while accelerating the degradation of antibiotics and decreasing their antibacterial activity could be achieved through pretreatment with bioelectrochemical systems [56,81]. As a result, for the majority of pharmaceutical chemicals, discharge concentrations of Membranous Biological Reactors were considerably lower than those using traditional techniques [120].

5.1.7. Advance Oxidation Process

The recalcitrant nature of effluents containing antibiotic residues makes conventional biological treatments ineffective in removing these chemicals. One of the approaches used in these processes is advanced oxidation [147]. In advanced oxidation processes (AOPs), organic molecules in wastewater, which are difficult to handle biologically, are oxidized and converted to simpler products [153]. This approach is based on the production and use of free radicals, such as hydroxyl, as potent oxidizers with objectives such as the destruction of substances that do not completely oxidize with commonly used oxidizers such as chlorine. By breaking chemical bonds or through mechanisms such as electron transfer, addition, and substitution, the OH^- decomposes organic pollutants into small molecules of organic matter, as well as CO_2 and H_2O, more easily [134,154]. This approach includes ozonaion, Fenton oxidation, photo Fenton, oxidation of electrochemical and UV/H_2O_2 methodologies [119,134]. The removal efficiencies of antibiotics such as Amoxicillin, Ampicillin, Ciprofloxacin, Doxycycline, Erythromycin, Norfloxacin, Ofloxacin, Tetracycline, Vancomycin and Roxithromycin using AOPs are 73–100%, 99–100%, 87–100%, 70–99%, 90–100%, 80–100%, 94–100%, 70–95% and 89–99%, respectively. In comparison with the conventional methods of wastewater treatment, AOP has the following advantages: (i) no sludge production, (ii) lesser retention time for pollutant removal, and (iii) the ability to mineralize contaminants at a faster pace [155]. The major drawback of AOP is its high cost, which is related to the use of expensive chemicals such as hydrogen peroxide (H_2O_2) and the significant energy consumption related to the generation of O_3 or UV radiation [1].

5.2. Adsorption

Understanding antibiotic absorption on adsorbents requires knowledge of their physical and chemical characteristics. Adsorption is the deposition of elements from a gas or liquid phase onto the surface of an adsorbent, which can be physical or chemical [156]. Because of its great efficiency, low cost, ease of operation, and design simplicity, adsorption technology has seen widespread use (Figure 7) [65]. Adsorption is a typical method used for removing metal ions from a variety of industrial effluents [157]. Magnetic nanoparticles, silver nanoparticles, activated carbon, nanotubes, and polymer nanocomposites have all been utilized as adsorbents to remove various pollutants, including heavy metals, which are extremely toxic even at low concentrations [121,158,159]. Adsorbents such as activated carbon can be used in an adsorption filtering process to remove high levels of antibiotic compounds, but the effectiveness of antibiotic absorption is influenced by several factors such as (i) activated carbon, (ii) target compound concentration, (iii) dissolved oxygen concentration, and (iv) pH and temperature [119]. Natural zeolite, bentonite, activated carbon, carbon nanotubes, and biochar are used to remove antibiotics from aquatic environments by adsorption [160]. The phenomenon of adsorption involves (i) the movement of the adsorbate via the stationary liquid film around the adsorbent—solute transfer in the bulk, (ii) the diffusion of film and transport of adsorbate along the film, (iii) the diffusion of pores and adsorbate through the porous structure into the active sites, and (iv) adsorption between the adsorbate and porous structure. The antibiotic adsorption capacity is further influenced by electrostatic and hydrophobic interactions between antibiotics and sludge. The mechanisms of adsorption can be further identified using an isotherm model such as Langmuir, Freundlich, Tempkin, etc. [161]. Antibiotics are classified as positive, neutral, or negative, depending on the pH of the solution, and the removal efficiency depends upon the electrostatic interaction between antibiotics and charged sorbents [132]. The effectiveness of MBR in treating water contaminated with antibiotics is increasingly being investigated. Membranous Biological Reactors (MBRs) have several advantages, including a higher solids retention time (SRT), reduced sludge generation, and a higher concentration of mixed liquid-suspended solids when compared to Conventional Activated Sludge Systems (CAS) [162]. The efficient removal of antibiotics such as Sulfamonomethoxine (SMM) (11%), Sulfamethoxazole (SMX) (6.5%), and Sulfadimethoxine (SDM) (19%) from wastewater using sorbents has been reported by researchers [162].

Nanoparticle Interaction

Nanoparticles are particles with sizes ranging from 1 to 100 nm that have a wide variety of uses owing to their unique physio-chemical qualities [163]. Nanoparticles such as iron oxide nanoparticle, superparamagnetic iron oxide nanoparticles and silver nanoparticles have been used in the removal of water pollutants [142,163–165]. When nanoparticles are used to combat antibiotic resistant, two alternative methods are available. Firstly, antibiotics are mixed with a functionalized nanomaterial, and the nanomaterial enters the ARB, releasing a huge number of damaging ions. Combining antibiotics with nanomaterials provides synergistic effects in the second strategy, which means they combat ARGs jointly rather than individually. A combination of nanomaterials and molecular antibiotics has gained popularity due to its effectiveness in eliminating multidrug-resistant pathogenic bacteria and attacking a wide range of ARBs and ARGs [144]. Carbon nanotubes are a member of the carbon family, and are notably a new type of adsorbent that has demonstrated a fantastic ability to cast off many types of pollution [166]. Carbon nanotubes are prospective solutions that can be used to tackle environmental pollution because of their large specific surface area, extensive pores, hollow structures, and strong and effective interactions with pollutant molecules [167]. As such, to be effective as an absorbent material, graphene, which has a large specific surface area and pore volume and thus offers adequate adsorption sites for contaminants in wastewater treatment, should be used [168].

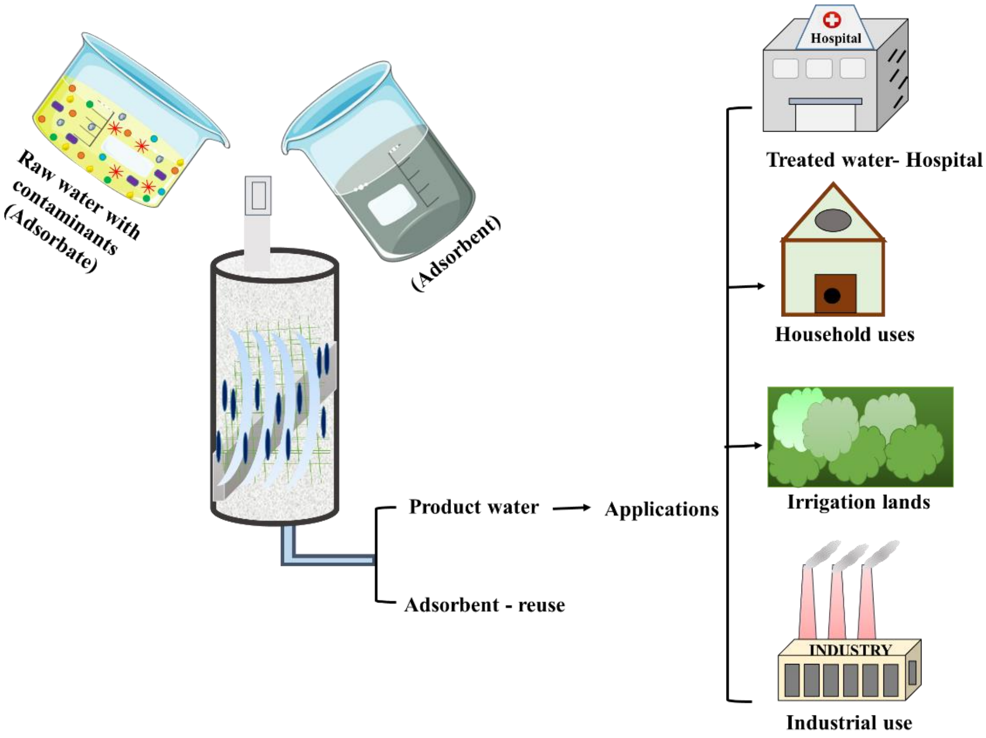

Figure 7. Process of adsorption.

6. Recycling Strategies

The "Era of Wastewater Reclamation, Recycling, and Reuse" began in the early twentieth century as a result of technological improvements in the chemical, physical, and biological processes of wastewater treatment [169,170]. The reuse of wastewater effluent is commonly seen in both developing and industrialized countries such as the United States of America, Japan, China, Korea, and Israel [171–173]. The majority of previous studies have focused on floatation, coagulation, and reverse osmosis or membrane bioreactor processes as potential wastewater treatment options [174]. The use of sequencing anoxic/anaerobic membrane bioreactors, a type of MBR method that removes nitrogen and phosphorus concurrently, has been introduced, and it outperforms the improved Luzack–Ettinger process of phosphorus removal [175]. The percentage of activated sludge in this process was raised from 30 to 70%; Galaxolide, Tonalid, Ibuprofen, Naproxen, 17-Estradiol, and Sulfamethoxazole can be removed in wastewater treatment plants [176]. Activated carbon filtration, with both powdered and granular activated carbon, has been used [177]. The adsorption of micropollutants from wastewater using powdered activated carbon is an excellent method for removing micropollutants such as Sulfamethoxazole, Atenolol, Diclofenac, etc. [178]. Granulated activated carbon is the most cost-effective and frequently used material for removing organic micropollutants such as Fexofenadine, Carbamazepine, Lamotrigine, Oxazepam, Fluconazole, Cetirizine, and N, N-diethyl-meta-toluamide from contaminated drinking water [179].

The law clarifies the governments' and other stakeholders' responsibilities in the fight against antibiotic resistance, including the use and discharge of antibiotics into the environment. Recent systematic reviews laid out 17 policy measures to decrease human antibiotic usage, although it is unclear whether or not most of the policies will have any influence on antibiotic use [180]. Some of the recycling strategies include the construction of vertical flow wetlands, which will help in the removal of antibiotics such as sulfamethazine, ciprofloxacin and oxytetracycline with an efficiency of 68–85%. Similarly, other strategies include the construction of horizontal subsurface flow wetlands, photocatalytic degradation, sonocatalytic irradiation, ultrafiltration using PVC membrane, adsorption by activated carbon, carbon nanotubes, electro-coagulation, advanced oxidation processes in combination with UV/hydrogen peroxide, and advanced oxidation processes using Fenton process, and their efficiencies are shown in Table 2.

Table 2. Various recycling techniques for antibiotics removal.

S. No	Strategy Used	Antibiotics Removed	Efficiency	References
1.	Vertical flow constructed wetlands	Sulfamethazine	68–73%	Huang et al. [134]
		Ciprofloxacin	91–95%	
		Oxytetracycline	82–85%	
2.	Horizontal subsurface flow constructed wetlands	Sulfamethoxazole	4–59%	Huang et al. [134]; Liu et al. [123]
3.	Photocatalytic degradation	Tetracycline	94.96%	Akhil et al. [181]
		Cephalexin	96%	
		Metronidazole	94.5%	
		Sulfamethazine	78%	
4.	Sonocatalytic irradiation	Norfloxacin	69.07%	Akhil et al. [181]
		Sulfanilamide	95.64%	
		Rifampicin	95.3%	
		Azithromycin	98.4%	
5.	Ultrafiltration using PVC membrane	Norfloxacin	80%	Bao et al. [129]; Wu et al. [182]
6.	Adsorption by activated carbon	Cephalexin	74–88%	Ahmed et al. [156]
		Ciprofloxacin	100%	
		Amoxicillin	95%	
		Tetracycline	74–88%	
		Ornidazole	90%	
7.	Adsorption by carbon nanotubes	Sulfamethoxazole	80%	Ahmed et al. [156]
		Sulfamethoxazole	96%	
		Lincomycin	>90%	
		Amoxicillin	86.5%	
8.	Electro-coagulation	Ampicillin	3.6%	Baran et al. [183]
		Doxycycline	96.4%	
		Sulfathiazole	3.3%	
		Tylosin	3.1%	

Table 2. *Cont.*

S. No	Strategy Used	Antibiotics Removed	Efficiency	References
9.	Advanced oxidation processes in combination of UV/hydrogen peroxide	Metronidazole	92%	Anjali and Shanthakumar, [184]
		Ciprofloxacin	93%	
10.	Advanced oxidation processes using Fenton process	Ofloxacin	100%	Carbajo et al. [185]

7. Conclusions

It is well understood that the presence of antibiotics in water is becoming a serious issue, and it has to be sorted out. To conclude, one of the main causes of the introduction of drugs and antibiotic-resistant bacteria into aquatic environments is excrement and corpses from human and animal sources. These bacteria can also pass on their genes to waterborne pathogenic germs, giving them the ability to produce resistance genes. The increased use of antibiotics will definitely result in the emergence of large numbers of new antimicrobial-resistant bacteria, and the diseases caused by them could be untreatable. This review has included a variety of sources of antibiotic pollution, their effects, and a number of innovative methods for removing these antibiotics from wastewater. In order to combat the growing problems of pollution caused by antibiotics, it is critical to not only continuously improve wastewater treatment systems, but also to prevent antibiotic overuse and excessive discharge into water resources. The life expectancy of humans will be shortened if these antibiotics are widely used in water bodies. The treatment of an increasing number of ailments becomes increasingly difficult as antibiotics lose their efficacy. Longer hospital stays, higher healthcare costs, and higher mortality are all consequences of antimicrobial resistance.

Author Contributions: A.V.S.: Conceptualization, Writing—Original Draft, Editing, Visualization, Supervision; S.W.: Writing—Original Draft, Editing; R.S.S.P.: Writing—Original Draft, Editing, Visualization; P.P.: Writing—Original Draft, Editing; M.S.: Writing—Original Draft, Editing; S.S.: Writing—Original Draft, Editing; N.S.: Writing—Original Draft, Editing; S.P.: Writing—Original Draft, Editing; V.V.R.: Writing—Original Draft, Editing. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data used to support the findings of this study are included in the article. Should further data or information be required, these are available from the corresponding author upon request.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Correa, E.M.C.; Maria Franco, A.; Gonzalez, C.F. Advanced Oxidation Processes for the Removal of Antibiotics from Water. An Overview. *Water* **2019**, *12*, 102. [CrossRef]
2. Bateganya, N.L.; Nakalanzi, D.; Babu, M.; Hein, T. Buffering municipal wastewater pollution using urban wetlands in sub-Saharan Africa: A case of Masaka municipality, Uganda. *Environ. Technol.* **2015**, *36*, 2149–2160. [CrossRef] [PubMed]
3. von Sperling, M.; de Lemos Chernicharo, C.A. Urban wastewater treatment technologies and the implementation of discharge standards in developing countries. *Urban Water* **2002**, *4*, 105–114. [CrossRef]
4. Polprasert, C.; Ujang, Z.; Henze, M. *Municipal Wastewater Management in Developing Countries-Principles and Engineering*, 5th ed.; IWA Publishing: London, UK, 2006.
5. Lien, L.T.Q.; Hoa, N.Q.; Chuc, N.T.K.; Thoa, N.T.M.; Phuc, H.D.; Diwan, V.; Dat, N.T.; Tamhankar, A.J.; Lundborg, C.S. Antibiotics in Wastewater of a Rural and an Urban Hospital before and after Wastewater Treatment, and the Relationship with Antibiotic Use—A One Year Study from Vietnam. *Int. J. Environ. Res. Public Health* **2016**, *13*, 588. [CrossRef]

6. Burch, K.D.; Han, B.; Pichtel, J.; Zubkov, T. Removal efficiency of commonly prescribed antibiotics via tertiary wastewater treatment. *Environ. Sci. Pollut. Res.* **2019**, *26*, 6301–6310. [CrossRef]
7. Aydin, S.; Aydin, M.E.; Ulvi, A.; Kilic, H. Antibiotics in hospital effluents: Occurrence, contribution to urban wastewater, removal in a wastewater treatment plant, and environmental risk assessment. *Environ. Sci. Pollut. Res.* **2018**, *26*, 544–558. [CrossRef]
8. Shi, Y.; Liu, J.; Zhuo, L.; Yan, X.; Cai, F.; Luo, W.; Ren, M.; Liu, Q.; Yu, Y. Antibiotics in wastewater from multiple sources and surface water of the Yangtze River in Chongqing in China. *Environ. Monit. Assess.* **2020**, *192*, 159. [CrossRef]
9. Baquero, F.; Martinez, J.L.; Cantón, R. Antibiotics and antibiotic resistance in water environments. *Curr. Opin. Biotechnol.* **2008**, *19*, 260–265. [CrossRef]
10. Calero-Cáceres, W.; Muniesa, M. Persistence of naturally occurring antibiotic resistance genes in the bacteria and bacteriophage fractions of wastewater. *Water Res.* **2016**, *95*, 11–18. [CrossRef]
11. Chandler, C.I. Current accounts of antimicrobial resistance: Stabilisation, individualisation and antibiotics as infrastructure. *Palgrave Commun.* **2019**, *5*, 53. [CrossRef]
12. Huang, A.; Yan, M.; Lin, J.; Xu, L.; Gong, H.; Gong, H. A Review of Processes for Removing Antibiotics from Breeding Wastewater. *Int. J. Environ. Res. Public Health* **2021**, *18*, 4909. [CrossRef] [PubMed]
13. Borbély, G.; Nagy, E. Removal of zinc and nickel ions by complexation–membrane filtration process from industrial wastewater. *Desalination* **2009**, *240*, 218–226. [CrossRef]
14. Tsakona, M.; Anagnostopoulou, E.; Gidarakos, E. Hospital waste management and toxicity evaluation: A case study. *Waste Manag.* **2007**, *27*, 912–920. [CrossRef]
15. Azanu, D.; Styrishave, B.; Darko, G.; Weisser, J.J.; Abaidoo, R.C. Occurrence and risk assessment of antibiotics in water and lettuce in Ghana. *Sci. Total. Environ.* **2018**, *622–623*, 293–305. [CrossRef] [PubMed]
16. Karthikeyan, K.; Meyer, M.T. Occurrence of antibiotics in wastewater treatment facilities in Wisconsin, USA. *Sci. Total. Environ.* **2006**, *361*, 196–207. [CrossRef]
17. Carmona, E.; Andreu, V.; Picó, Y. Occurrence of acidic pharmaceuticals and personal care products in Turia River Basin: From waste to drinking water. *Sci. Total. Environ.* **2014**, *484*, 53–63. [CrossRef]
18. Aghalari, Z.; Dahms, H.U.; Sillanpää, M.; Sosa-Hernandez, J.E.; Parra-Saldívar, R. Effectiveness of wastewater treatment systems in removing microbial agents: A systematic review. *Glob. Health* **2020**, *16*, 13. [CrossRef]
19. Zhang, T.; Li, B. Occurrence, Transformation, and Fate of Antibiotics in Municipal Wastewater Treatment Plants. *Crit. Rev. Environ. Sci. Technol.* **2011**, *41*, 951–998. [CrossRef]
20. Kim, S.; Carlson, K. Quantification of human and veterinary antibiotics in water and sediment using SPE/LC/MS/MS. *Anal. Bioanal. Chem.* **2007**, *387*, 1301–1315. [CrossRef]
21. Al-Riyami, I.M.; Ahmed, M.; Al-Busaidi, A.; Choudri, B.S. Antibiotics in wastewaters: A review with focus on Oman. *Appl. Water Sci.* **2018**, *8*, 199. [CrossRef]
22. Li, W.; Shi, Y.; Gao, L.; Liu, J.; Cai, Y. Occurrence of antibiotics in water, sediments, aquatic plants, and animals from Baiyangdian Lake in North China. *Chemosphere* **2012**, *89*, 1307–1315. [CrossRef] [PubMed]
23. Rosal, R.; Rodríguez, A.; Perdigón-Melón, J.A.; Petre, A.; García-Calvo, E.; Gómez, M.J. Occurrence of emerging pol-lutants in urban wastewater and their removal through biological treatment followed by ozonation. *Water Res.* **2010**, *44*, 578–588. [CrossRef] [PubMed]
24. Chen, K.; Zhou, J. Occurrence and behavior of antibiotics in water and sediments from the Huangpu River, Shanghai, China. *Chemosphere* **2014**, *95*, 604–612. [CrossRef] [PubMed]
25. Yang, S.; Carlson, K. Routine monitoring of antibiotics in water and wastewater with a radioimmunoassay technique. *Water Res.* **2004**, *38*, 3155–3166. [CrossRef] [PubMed]
26. Hubeny, J.; Harnisz, M.; Korzeniewska, E.; Buta, M.; Zieliński, W.; Rolbiecki, D.; Giebułtowicz, J.; Nałęcz-Jawecki, G.; Płaza, G. Industrialization as a source of heavy metals and antibiotics which can enhance the antibiotic resistance in wastewater, sewage sludge and river water. *PLoS ONE* **2021**, *16*, e0252691. [CrossRef]
27. Ebrahimi, S.M.; Reyhani, R.D.; Asghari-JafarAbadi, M.; Fathifar, Z. Diversity of antibiotics in hospital and municipal wastewaters and receiving water bodies and removal efficiency by treatment processes: A systematic review protocol. *Environ. Evid.* **2020**, *9*, 19. [CrossRef]
28. Kumar, S.B.; Arnipalli, S.R.; Ziouzenkova, O. Antibiotics in Food Chain: The Consequences for Antibiotic Resistance. *Antibiotics* **2020**, *9*, 688. [CrossRef]
29. Sun, S.; Yao, H.; Fu, W.; Xue, S.; Zhang, W. Enhanced degradation of antibiotics by photo-fenton reactive membrane filtration. *J. Hazard. Mater.* **2019**, *386*, 121955. [CrossRef]
30. Gao, P.; Munir, M.; Xagoraraki, I. Correlation of tetracycline and sulfonamide antibiotics with corresponding resistance genes and resistant bacteria in a conventional municipal wastewater treatment plant. *Sci. Total Environ.* **2012**, *421–422*, 173–183. [CrossRef]
31. Kumar, A.; Pal, D. Antibiotic resistance and wastewater: Correlation, impact and critical human health challenges. *J. Environ. Chem. Eng.* **2018**, *6*, 52–58. [CrossRef]
32. Teixeira, P.; Costa, S.; Brown, B.; Silva, S.; Rodrigues, R.; Valério, E. Quantitative PCR Detection of Enteric Viruses in Wastewater and Environmental Water Sources by the Lisbon Municipality: A Case Study. *Water* **2020**, *12*, 544. [CrossRef]
33. Magureanu, M.; Piroi, D.; Mandache, N.B.; David, V.; Medvedovici, A.; Bradu, C.; Parvulescu, V.I. Degradation of antibiotics in water by non-thermal plasma treatment. *Water Res.* **2011**, *45*, 3407–3416. [CrossRef] [PubMed]

34. Magureanu, M.; Piroi, D.; Mandache, N.B.; David, V.; Medvedovici, A.; Parvulescu, V.I. Degradation of pharmaceutical compound pentoxifylline in water by non-thermal plasma treatment. *Water Res.* **2010**, *44*, 3445–3453. [CrossRef] [PubMed]
35. Rodriguez-Mozaz, S.; Vaz-Moreira, I.; Della Giustina, S.V.; Llorca, M.; Barceló, D.; Schubert, S.; Berendonk, T.U.; Michael-Kordatou, I.; Fatta-Kassinos, D.; Martinez, J.L.; et al. Antibiotic residues in final effluents of European wastewater treatment plants and their impact on the aquatic environment. *Environ. Int.* **2020**, *140*, 105733. [CrossRef] [PubMed]
36. Leung, H.; Minh, T.; Murphy, M.; Lam, J.C.; So, M.; Martin, M.; Lam, P.K.; Richardson, B. Distribution, fate and risk assessment of antibiotics in sewage treatment plants in Hong Kong, South China. *Environ. Int.* **2012**, *42*, 1–9. [CrossRef]
37. Mahmood, A.R.; Al-Haideri, H.H.; Hassan, F.M. Detection of Antibiotics in Drinking Water Treatment Plants in Baghdad City, Iraq. *Adv. Public Health* **2019**, *2019*, 7851354. [CrossRef]
38. Mutiyar, P.K.; Mittal, A.K. Occurrences and fate of selected human antibiotics in influents and effluents of sewage treatment plant and effluent-receiving river Yamuna in Delhi (India). *Environ. Monit. Assess.* **2013**, *186*, 541–557. [CrossRef]
39. Diwan, V.; Tamhankar, A.J.; Khandal, R.K.; Sen, S.; Aggarwal, M.; Marothi, Y.; Iyer, R.V.; Sundblad-Tonderski, K.; Lundborg, C.S. Antibiotics and antibiotic-resistant bacteria in waters associated with a hospital in Ujjain, India. *BMC Public Health* **2010**, *10*, 414. [CrossRef]
40. Watkinson, A.; Murby, E.; Kolpin, D.; Costanzo, S. The occurrence of antibiotics in an urban watershed: From wastewater to drinking water. *Sci. Total. Environ.* **2009**, *407*, 2711–2723. [CrossRef]
41. Faleye, A.C.; Adegoke, A.A.; Ramluckan, K.; Fick, J.; Bux, F.; Stenström, T.A. Concentration and reduction of antibiotic residues in selected wastewater treatment plants and receiving waterbodies in Durban, South Africa. *Sci. Total Environ.* **2019**, *678*, 10–20. [CrossRef]
42. Kortesmäki, E.; Östman, J.R.; Meierjohann, A.; Brozinski, J.; Eklund, P.; Kronberg, L. Occurrence of Antibiotics in Influent and Effluent from 3 Major Wastewater-Treatment Plants in Finland. *Environ. Toxicol. Chem.* **2020**, *39*, 1774–1789. [CrossRef] [PubMed]
43. Göbel, A.; Thomsen, A.; McArdell, C.S.; Joss, A.; Giger, W. Occurrence and Sorption Behavior of Sulfonamides, Macrolides, and Trimethoprim in Activated Sludge Treatment. *Environ. Sci. Technol.* **2005**, *39*, 3981–3989. [CrossRef] [PubMed]
44. Lindberg, R.H.; Wennberg, P.; Johansson, M.I.; Tysklind, M.; Andersson, B.A.V. Screening of Human Antibiotic Sub-stances and Determination of Weekly Mass Flows in Five Sewage Treatment Plants in Sweden. *Environ. Sci. Technol.* **2005**, *39*, 3421–3429. [CrossRef] [PubMed]
45. Li, J.; Cheng, W.; Xu, L.; Strong, P.J.; Chen, H. Antibiotic-resistant genes and antibiotic-resistant bacteria in the effluent of urban residential areas, hospitals, and a municipal wastewater treatment plant system. *Environ. Sci. Pollut. Res.* **2014**, *22*, 4587–4596. [CrossRef] [PubMed]
46. Li, J.; Cheng, W.; Xu, L.; Jiao, Y.; Baig, S.A.; Chen, H. Occurrence and removal of antibiotics and the corresponding resistance genes in wastewater treatment plants: Effluent's influence to downstream water environment. *Environ. Sci. Pollut. Res.* **2015**, *23*, 6826–6835. [CrossRef] [PubMed]
47. Dinh, Q.; Moreau-Guigon, E.; Labadie, P.; Alliot, F.; Teil, M.J.; Blanchard, M. Fate of antibiotics from hospital and domestic sources in a sewage network. *Sci. Total Environ.* **2017**, *575*, 758–766. [CrossRef]
48. Choi, K.; Kim, Y.; Jung, J.; Kim, M.-H.; Kim, C.-S.; Kim, N.-H.; Park, J. Occurrences and ecological risks of roxithromycin, trimethoprim, and chloramphenicol in the Han River, Korea. *Environ. Toxicol. Chem.* **2008**, *27*, 711–719. [CrossRef]
49. Ekwanzala, M.D.; Lehutso, R.F.; Kasonga, T.K.; Dewar, J.B.; Momba, M.N.B. Environmental Dissemination of Selected Antibiotics from Hospital Wastewater to the Aquatic Environment. *Antibiotics* **2020**, *9*, 431. [CrossRef]
50. Rossmann, J.; Schubert, S.; Gurke, R.; Oertel, R.; Kirch, W. Simultaneous determination of most prescribed antibiotics in multiple urban wastewater by SPE-LC–MS/MS. *J. Chromatogr. B* **2014**, *969*, 162–170. [CrossRef]
51. Bisognin, R.P.; Wolff, D.B.; Carissimi, E.; Prestes, O.D.; Zanella, R. Occurrence and fate of pharmaceuticals in effluent and sludge from a wastewater treatment plant in Brazil. *Environ. Technol.* **2019**, *42*, 2292–2303. [CrossRef]
52. Verlicchi, P.; Al Aukidy, M.; Jelic, A.; Petrović, M.; Barceló, D. Comparison of measured and predicted concentrations of selected pharmaceuticals in wastewater and surface water: A case study of a catchment area in the Po Valley (Italy). *Sci. Total Environ.* **2014**, *470–471*, 844–854. [CrossRef] [PubMed]
53. Verlicchi, P.; Galletti, A.; Petrovic, M.; Barceló, D. Hospital effluents as a source of emerging pollutants: An overview of micropollutants and sustainable treatment options. *J. Hydrol.* **2010**, *389*, 416–428. [CrossRef]
54. Wegrzyn, A.; Machura, M.; Zabczynski, S. Possibilities for Removal of Contrast Agents from Wastewater. *Ochro-Nasrodowiska* **2015**, *37*, 55–63.
55. Capoor, M.R.; Bhowmik, K.T. Cytotoxic Drug Dispersal, Cytotoxic Safety, and Cytotoxic Waste Management: Practices and Proposed India-specific Guidelines. *Indian J. Med. Paediatr. Oncol.* **2017**, *38*, 190–197.
56. Wang, L.; Liu, Y.; Ma, J.; Zhao, F. Rapid degradation of sulphamethoxazole and the further transformation of 3-amino-5-methylisoxazole in a microbial fuel cell. *Water Res.* **2016**, *88*, 322–328. [CrossRef]
57. Arslan, A.; Veli, S.; Bingöl, D. Use of response surface methodology for pretreatment of hospital wastewater by O_3/UV and O_3/UV/H_2O_2 processes. *Sep. Purif. Technol.* **2014**, *132*, 561–567. [CrossRef]
58. Escher, B.I.; Baumgartner, R.; Koller, M.; Treyer, K.; Lienert, J.; McArdell, C.S. Environmental toxicology and risk assessment of pharmaceuticals from hospital wastewater. *Water Res.* **2011**, *45*, 75–92. [CrossRef]

59. Paulus, G.K.; Hornstra, L.M.; Alygizakis, N.; Slobodnik, J.; Thomaidis, N.; Medema, G. The impact of on-site hospital wastewater treatment on the downstream communal wastewater system in terms of antibiotics and antibiotic resistance genes. *Int. J. Hyg. Environ. Health* **2019**, *222*, 635–644. [CrossRef]
60. Orias, F.; Perrodin, Y. Characterisation of the ecotoxicity of hospital effluents: A review. *Sci. Total. Environ.* **2013**, *454–455*, 250–276. [CrossRef]
61. Khan, N.A.; Ahmed, S.; Farooqi, I.H.; Ali, I.; Vambol, V.; Changani, F.; Yousefi, M.; Vambol, S.; Khan, S.U.; Khan, A.H. Occurrence, sources and conventional treatment techniques for various antibiotics present in hospital wastewaters: A critical review. *TrAC Trends Anal. Chem.* **2020**, *129*, 115921. [CrossRef]
62. Carraro, E.; Bonetta, S.; Bertino, C.; Lorenzi, E.; Bonetta, S.; Gilli, G. Hospital effluents management: Chemical, physical, microbiological risks and legislation in different countries. *J. Environ. Manag.* **2016**, *168*, 185–199. [CrossRef] [PubMed]
63. Savin, M.; Bierbaum, G.; Hammerl, J.A.; Heinemann, C.; Parcina, M.; Sib, E.; Voigt, A.; Kreyenschmidt, J. Antibiotic-resistant bacteria and antimicrobial residues in wastewater and process water from German pig slaughterhouses and their receiving municipal wastewater treatment plants. *Sci. Total Environ.* **2020**, *727*, 138788. [CrossRef] [PubMed]
64. Savin, M.; Bierbaum, G.; Hammerl, J.A.; Heinemann, C.; Parcina, M.; Sib, E.; Voigt, A.; Kreyenschmidt, J. ESKAPE Bacteria and Extended-Spectrum-β-Lactamase-Producing *Escherichia coli* Isolated from Wastewater and Process Water from German Poultry Slaughterhouses. *Appl. Environ. Microbiol.* **2020**, *86*, e02748-19. [CrossRef] [PubMed]
65. Tang, Y.; Huang, H.; Xue, W.; Chang, Y.; Li, Y.; Guo, X.; Zhong, C. Rigidifying induced fluorescence enhancement in 2D porous covalent triazine framework nanosheets for the simultaneously luminous detection and adsorption removal of antibiotics. *Chem. Eng. J.* **2020**, *384*, 123382. [CrossRef]
66. Homeier-Bachmann, T.; Heiden, S.; Lübcke, P.; Bachmann, L.; Bohnert, J.; Zimmermann, D.; Schaufler, K. Antibiotic-Resistant *Enterobacteriaceae* in Wastewater of Abattoirs. *Antibiotics* **2021**, *10*, 568. [CrossRef]
67. Bustillo-Lecompte, C.; Mehrvar, M. Slaughterhouse wastewater: Treatment, management and resource recovery. In *Physico-Chemical Wastewater Treatment and Resource Recovery*; IntechOpen: London, UK, 2017; pp. 153–174. [CrossRef]
68. Masse, M.T.; Aloys, R.J.S.; Betbui, B.T. Profile of Antibiotic Resistant Bacteria Isolated from Slaughterhouse Effluents of Etoudi-Yaounde and Its Receiving Waterbody. *Int. J. Health Sci. Res.* **2021**, *11*, 40–47. [CrossRef]
69. Lucas, D.; Badia-Fabregat, M.; Vicent, T.; Caminal, G.; Rodríguez-Mozaz, S.; Balcázar, J.; Barceló, D. Fungal treatment for the removal of antibiotics and antibiotic resistance genes in veterinary hospital wastewater. *Chemosphere* **2016**, *152*, 301–308. [CrossRef]
70. Beattie, R.E.; Walsh, M.; Cruz, M.C.; McAliley, L.R.; Dodgen, L.; Zheng, W.; Hristova, K.R. Agricultural contamination impacts antibiotic resistance gene abundances in riverbed sediment temporally. *FEMS Microbiol. Ecol.* **2018**, *94*, fiy131. [CrossRef]
71. Davis, J.G.; Truman, C.C.; Kim, S.C.; Ascough, J.C.; Carlson, K. Antibiotic Transport via Runoff and Soil Loss. *J. Environ. Qual.* **2006**, *35*, 2250–2260. [CrossRef]
72. Christy, M.L.; Sampson, M.; Edson, M.; Anthony, O. Antibiotic Use in Agriculture and Its Consequential Resistance in Environmental Sources: Potential Public Health Implications. *Molecules* **2018**, *23*, 795.
73. Tasho, R.P.; Cho, J.Y. Veterinary antibiotics in animal waste, its distribution in soil and uptake by plants: A review. *Sci. Total Environ.* **2016**, *563*, 366–376. [CrossRef] [PubMed]
74. Hocquet, D.; Muller, A.; Bertrand, X. What happens in hospitals does not stay in hospitals: Antibiotic-resistant bacteria in hospital wastewater systems. *J. Hosp. Infect.* **2016**, *93*, 395–402. [CrossRef] [PubMed]
75. Anthony, A.A.; Adekunle, C.F.; Thor, A.S. Residual antibiotics, antibiotic resistant superbugs and antibiotic resistance genes in surface water catchments: Public health impact. *Phys. Chem. Earth* **2018**, *105*, 177–183. [CrossRef]
76. Tarrass, F.; Benjelloun, M.; Benjelloun, O. Recycling Wastewater After Hemodialysis: An Environmental Analysis for Alternative Water Sources in Arid Regions. *Am. J. Kidney Dis.* **2008**, *52*, 154–158. [CrossRef] [PubMed]
77. Meena, V.D.; Dotaniya, M.L.; Saha, J.K.; Patra, A. Antibiotics and antibiotic resistant bacteria in wastewater: Impact on environment, soil microbial activity and human health. *Afr. J. Microbiol. Res.* **2015**, *9*, 965–978.
78. Niemi, L.; Taggart, M.; Boyd, K.; Zhang, Z.; Gaffney, P.P.; Pfleger, S.; Gibb, S. Assessing hospital impact on pharmaceutical levels in a rural 'source-to-sink'water system. *Sci. Total Environ.* **2020**, *737*, 139618. [CrossRef] [PubMed]
79. Pereira, A.M.P.T.; Silva, L.J.G.; Laranjeiro, C.S.M.; Meisel, L.M.; Lino, C.M.; Pena, A. Human pharmaceuticals in Portuguese rivers: The impact of water scarcity in the environmental risk. *Sci. Total Environ.* **2017**, *609*, 1182–1191. [CrossRef]
80. Hernández, F.; Montory, M.; Gómez-Fuentes, C. Occurrence of antibiotics and bacterial resistance in wastewater and sea water from the Antarctic. In Proceedings of the 15th International Conference on Environmental Science and Technology, Rhodes, Greece, 31 August–2 September 2017.
81. Cheng, D.; Ngo, H.; Guo, W.; Liu, Y.; Zhou, J.; Chang, S.; Nguyen, D.; Bui, X.; Zhang, X. Bioprocessing for elimination antibiotics and hormones from swine wastewater. *Sci. Total. Environ.* **2018**, *621*, 1664–1682. [CrossRef]
82. Li, X.; Guo, P.; Shan, Y.; Ke, Y.; Li, H.; Fu, Q.; Wang, Y.; Liu, T.; Xia, X. Determination of 82 veterinary drugs in swine waste lagoon sludge by ultra-high performance liquid chromatography–tandem mass spectrometry. *J. Chromatogr. A* **2017**, *1499*, 57–64. [CrossRef]
83. Sosa-Hernández, J.E.; Rodas-Zuluaga, L.I.; López-Pacheco, I.Y.; Melchor-Martínez, E.M.; Aghalari, Z.; Limón, D.S. Sources of antibiotics pollutants in the aquatic environment under SARS-CoV-2 pandemic situation. *Case Stud. Chem. Environ. Eng.* **2021**, *4*, 100127. [CrossRef]

84. Vaz-Moreira, I.; Nunes, O.; Manaia, C.M. Bacterial diversity and antibiotic resistance in water habitats: Searching the links with the human microbiome. *FEMS Microbiol. Rev.* **2014**, *38*, 761–778. [CrossRef] [PubMed]
85. Hultman, J.; Tamminen, M.; Pärnänen, K.; Cairns, J.; Karkman, A.; Virta, M. Host range of antibiotic resistance genes in wastewater treatment plant influent and effluent. *FEMS Microbiol. Ecol.* **2018**, *94*, fiy038. [CrossRef]
86. Cacace, D.; Fatta-Kassinos, D.; Manaia, C.M.; Cytryn, E.; Kreuzinger, N.; Rizzo, L.; Karaolia, P.; Schwartz, T.; Alexander, J.; Merlin, C.; et al. Antibiotic resistance genes in treated wastewater and in the receiving water bodies: A pan-European survey of urban settings. *Water Res.* **2019**, *162*, 320–330. [CrossRef] [PubMed]
87. Sharmaa, N.; Sharmab, S.K. Wastewater Treatment Plants as emerging source of antibiotic resistance. In *Green Chemistry and Water Remediation: Research and Applications*; Elsevier: Amsterdam, The Netherlands, 2020; p. 239. [CrossRef]
88. Gwenzi, W.; Kanda, A.; Danha, C.; Muisa-Zikali, N.; Chaukura, N. Occurrence, human health risks, and removal of pharmaceuticals in aqueous systems: Current knowledge and future perspectives. In *Applied Water Science Volume 1: Fundamentals and Applications*; Scrivener Publishing LLC: Beverly, MA, USA, 2021; pp. 63–101. [CrossRef]
89. Singh, R.; Singh, A.P.; Kumar, S.; Giri, B.S.; Kim, K.-H. Antibiotic resistance in major rivers in the world: A systematic review on occurrence, emergence, and management strategies. *J. Clean. Prod.* **2019**, *234*, 1484–1505. [CrossRef]
90. Zhang, Y.; Marrs, C.F.; Simon, C.; Xi, C. Wastewater treatment contributes to selective increase of antibiotic resistance among *Acinetobacter* spp. *Sci. Total. Environ.* **2009**, *407*, 3702–3706. [CrossRef]
91. Kraemer, S.A.; Ramachandran, A.; Perron, G.G. Antibiotic Pollution in the Environment: From Microbial Ecology to Public Policy. *Microorganisms* **2019**, *7*, 180. [CrossRef]
92. Lupo, A.; Coyne, S.; Berendonk, T.U. Origin and Evolution of Antibiotic Resistance: The Common Mechanisms of Emergence and Spread in Water Bodies. *Front. Microbiol.* **2012**, *3*, 18. [CrossRef] [PubMed]
93. Kim, S.; Aga, D.S. Potential Ecological and Human Health Impacts of Antibiotics and Antibiotic-Resistant Bacteria from Wastewater Treatment Plants. *J. Toxicol. Environ. Health Part B* **2007**, *10*, 559–573. [CrossRef]
94. Madigan, M.T.; Martinko, J.M.; Parker, J. *Brock Biology of Microorganisms*, 8th ed.; Prentice Hall International, Inc.: New York, NY, USA, 2021.
95. Mwangi, M.M.; Wu, S.W.; Zhou, Y.; Sieradzki, K.; de Lencastre, H.; Richardson, P.; Bruce, D.; Rubin, E.; Myers, E.; Siggia, E.D.; et al. Tracking the in vivo evolution of multidrug resistance in *Staphylococcus aureus* by whole-genome sequencing. *Proc. Natl. Acad. Sci. USA* **2007**, *104*, 9451–9456. [CrossRef]
96. Khameneh, B.; Diab, R.; Ghazvini, K.; Bazzaz, B.S.F. Breakthroughs in bacterial resistance mechanisms and the potential ways to combat them. *Microb. Pathog.* **2016**, *95*, 32–42. [CrossRef]
97. Golovko, O.; Kumar, V.; Fedorova, G.; Randak, T.; Grabic, R. Seasonal changes in antibiotics, antidepressants/psychiatric drugs, antihistamines and lipid regulators in a wastewater treatment plant. *Chemosphere* **2014**, *111*, 418–426. [CrossRef] [PubMed]
98. Sims, N.; Kasprzyk-Hordern, B. Future perspectives of wastewater-based epidemiology: Monitoring infectious disease spread and resistance to the community level. *Environ. Int.* **2020**, *139*, 105689. [CrossRef] [PubMed]
99. Morse, S. Factors and determinants of disease emergence. *Rev. Sci. Tech.-Off. Int. Des Epizoot.* **2004**, *23*, 443–451. [CrossRef] [PubMed]
100. Crittenden, J.C.; Trussell, R.R.; Hand, D.W.; Howe, K.J.; Tchobanoglous, G. *MWH's Water Treatment: Principles and Design*; John Wiley & Sons: Hoboken, NJ, USA, 2012.
101. Rafraf, I.D.; Lekunberri, I.; Sànchez-Melsió, A.; Aouni, M.; Borrego, C.M.; Balcázar, J.L. Abundance of antibiotic resistance genes in five municipal wastewater treatment plants in the Monastir Governorate, Tunisia. *Environ. Pollut.* **2016**, *219*, 353–358. [CrossRef]
102. Aslam, B.; Wang, W.; Arshad, M.I.; Khurshid, M.; Muzammil, S.; Nisar, M.A.; Alvi, R.F.; Aslam, M.A.; Qamar, M.U.; Salamat, M.K.F.; et al. Antibiotic resistance: A rundown of a global crisis. *Infect. Drug Resist.* **2018**, *11*, 1645–1658. [CrossRef]
103. Rolain, J.; Canton, R.; Cornaglia, G. Emergence of antibiotic resistance: Need for a new paradigm. *Clin. Microbiol. Infect.* **2012**, *18*, 615–616. [CrossRef]
104. Gouliouris, T.; Raven, K.E.; Moradigaravand, D.; Ludden, C.; Coll, F.; Blane, B. Detection of vancomycin-resistant *Enterococcus faecium* hospital-adapted lineages in municipal wastewater treatment plants indicates widespread distribution and release into the environment. *Genome Res.* **2019**, *29*, 626–634. [CrossRef]
105. Cahill, N.; O'Connor, L.; Mahon, B.; Varley, Á.; McGrath, E.; Ryan, P. Hospital effluent: A reservoir for carbapenemase-producing *Enterobacterales*? *Sci. Total Environ.* **2019**, *672*, 618–624. [CrossRef]
106. Burnham, J.P. Climate change and antibiotic resistance: A deadly combination. *Ther. Adv. Infect. Dis.* **2021**, *8*, 2049936121991374. [CrossRef]
107. Chahal, C.; Akker, B.V.D.; Young, F.; Franco, C.; Blackbeard, J.; Monis, P. Pathogen and Particle Associations in Wastewater. *Adv. Appl. Microbiol.* **2016**, *97*, 63–119. [CrossRef]
108. Farooq, R.; Ahmad, Z. Biological Wastewater Treatment and Resource Recovery Spreading of Antibiotic Resistance with Wastewater. In *Biological Wastewater Treatment and Resource Recovery*; IntechOpen: London, UK, 2017; pp. 73–98.
109. Pandey, P.K.; Kass, P.H.; Soupir, M.L.; Biswas, S.; Singh, V.P. Contamination of water resources by pathogenic bacteria. *AMB Express* **2014**, *4*, 51. [CrossRef] [PubMed]
110. Handel, A.; Margolis, E.; Levin, B.R. Exploring the role of the immune response in preventing antibiotic resistance. *J. Theor. Biol.* **2009**, *256*, 655–662. [CrossRef]

111. Marshall, J.S.; Warrington, R.; Watson, W.; Kim, H.L. An introduction to immunology and immunopathology. *Allergy Asthma Clin. Immunol.* **2018**, *14*, 49. [CrossRef] [PubMed]
112. Peterson, C.T.; Sharma, V.; Elmén, L.; Peterson, S.N. Immune homeostasis, dysbiosis and therapeutic modulation of the gut microbiota. *Clin. Exp. Immunol.* **2015**, *179*, 363–377. [CrossRef] [PubMed]
113. Miarov, O.; Tal, A.; Avisar, D. A critical evaluation of comparative regulatory strategies for monitoring pharmaceuticals in recycled wastewater. *J. Environ. Manag.* **2019**, *254*, 109794. [CrossRef]
114. Bos, J.; Zhang, Q.; Vyawahare, S.; Rogers, E.; Rosenberg, S.M.; Austin, R.H. Emergence of antibiotic resistance from multinucleated bacterial filaments. *Proc. Natl. Acad. Sci. USA* **2014**, *112*, 178–183. [CrossRef]
115. Gillissen, G. Side Effects of Antibiotics on Immune Response Parameters and their Possible Implications in Antimicrobial Chemotherapy. *Zent. Bakteriol. Mikrobiol. Hyg.* **1988**, *270*, 171–199. [CrossRef]
116. Cotten, C.M.; Taylor, S.; Stoll, B.; Goldberg, R.N.; Hansen, N.I.; Sánchez, P.J.; Ambalavanan, N.; Benjamin, D.K., Jr.; NICHD Neonatal Research Network. Prolonged duration of initial empirical antibiotic treatment is associated with increased rates of necrotizing enterocolitis and death for extremely low birth weight infants. *Pediatrics* **2009**, *123*, 58–66. [CrossRef]
117. Miller, J.E.; Wu, C.S.; Pedersen, L.H.; de Klerk, N.; Olsen, J.; Burgner, D.P. Maternal antibiotic exposure during pregnancy and hospitalization with infection in offspring: A population-based cohort study. *Int. J. Epidemiol.* **2018**, *47*, 561–571. [CrossRef]
118. Gonzalez-Perez, G.; Hicks, A.L.; Tekieli, T.M.; Radens, C.M.; Williams, B.L.; Lamouse-Smith, E.S.N. Maternal antibiotic treatment impacts development of the neonatal intestinal microbiome and antiviral immunity. *J. Immunol.* **2016**, *196*, 3768–3779. [CrossRef]
119. Derakhshan, Z.; Mokhtari, M.; Babaei, F.; Ahmadi, R.M.; Ehrampoush, M.H.; Faramarzian, M. Removal Methods of Antibiotic Compounds from Aqueous Environments—A Review. *J. Environ Health Sustain Dev.* **2016**, *1*, 43–62.
120. Nasrollohai, N.; Vatanpour, V.; Khataee, A. Removal of antibiotics from wastewaters by membrane technology: Limitations, successes, and future improvements. *Sci. Total Environ.* **2022**, *838*, 156010. [CrossRef] [PubMed]
121. Nqombolo, A.; Mpupa, A.; Moutloali, R.; Nomngongo, P. Wastewater Treatment Using Membrane Technology. In *Wastewater and Water Quality*; Yonar, T., Ed.; IntechOpen: London, UK, 2018; pp. 29–40.
122. Schwermer, C.U.; Krzeminski, P.; Wennberg, A.C.; Vogelsang, C.; Uhl, W. Removal of antibiotic resistant *E. coli* in two Norwegian wastewater treatment plants and by nano- and ultra-filtration processes. *Water Sci. Technol.* **2017**, *77*, 1115–1126. [CrossRef]
123. Liu, M.K.; Liu, Y.Y.; Bao, D.D.; Zhu, G.; Yang, G.H.; Geng, J.F.; Li, H.T. Effective Removal of Tetracycline Antibiotics from Water using Hybrid Carbon Membranes. *Sci. Rep.* **2017**, *7*, srep43717. [CrossRef]
124. Sharma, V.; Kumar, R.V.; Pakshirajan, K.; Pugazhenthi, G. Integrated adsorption-membrane filtration process for antibiotic removal from aqueous solution. *Powder Technol.* **2017**, *321*, 259–269. [CrossRef]
125. Ganiyu, S.O.; van Hullebusch, E.D.; Cretin, M.; Esposito, G.; Oturan, M.A. Coupling of membrane filtration and advanced oxidation processes for removal of pharmaceutical residues: A critical review. *Sep. Purif. Technol.* **2015**, *156*, 891–914. [CrossRef]
126. Yin, F.; Lin, S.; Zhou, X.; Dong, H.; Zhan, Y. Fate of antibiotics during membrane separation followed by physical-chemical treatment processes. *Sci. Total Environ.* **2021**, *759*, 143520. [CrossRef]
127. Arefi-Oskoui, S.; Khataee, A.; Jabbarvand Behrouz, S.; Vatanpour, V.; Haddadi Gharamaleki, S.; Orooji, Y.; Safarpour, M. Development of MoS_2/O-MWCNTs/PES blended membrane for efficient removal of dyes, antibiotic, and protein. *Sep. Purif. Technol.* **2022**, *280*, 11982. [CrossRef]
128. Javad, M.; Mohsen, S.; Vatanpour, V. Performance improvement of PES membrane decorated by Mil-125(Ti)/chitosan nanocomposite for removal of organic pollutants and heavy metal. *Chemosphere* **2022**, *290*, 133335.
129. Phoon, B.L.; Ong, C.C.; Saheed, M.S.M.; Show, P.-L.; Chang, J.-S.; Ling, T.C.; Lam, S.S.; Juan, J.C. Conventional and emerging technologies for removal of antibiotics from wastewater. *J. Hazard. Mater.* **2020**, *400*, 122961. [CrossRef]
130. Ezugbe, E.O.; Rathilal, S. Membrane Technologies in Wastewater Treatment: A Review. *Membranes* **2020**, *10*, 89. [CrossRef] [PubMed]
131. Wu, C.; Zhou, Y.; Sun, X.; Fu, L. The recent development of advanced wastewater treatment by ozone and biological aerated filter. *Environ. Sci. Pollut. Res.* **2018**, *25*, 8315–8329. [CrossRef] [PubMed]
132. Zhu, T.-T.; Su, Z.-X.; Lai, W.-X.; Zhang, Y.-B.; Liu, Y.-W. Insights into the fate and removal of antibiotics and antibiotic resistance genes using biological wastewater treatment technology. *Sci. Total Environ.* **2021**, *776*, 145906. [CrossRef]
133. Chen, J.; Liu, Y.-S.; Zhang, J.-N.; Yang, Y.-Q.; Hu, L.-X.; Yang, Y.-Y.; Zhao, J.-L.; Chen, F.-R.; Ying, G.-G. Removal of antibiotics from piggery wastewater by biological aerated filter system: Treatment efficiency and biodegradation kinetics. *Bioresour. Technol.* **2017**, *238*, 70–77. [CrossRef]
134. Wang, J.; Yang, Z.; Wang, H.; Wu, S.; Lu, H.; Wang, X. Decomposition process of cefotaxime sodium from antibiotic wastewater by Up-flow Blanket Filter (UBF) reactor: Reactor performance, sludge characteristics and microbial community structure analysis. *Sci. Total Environ.* **2021**, *758*, 143670. [CrossRef]
135. Azimi, S.M.; Malakootian, M.; Yaghmaeian, K.; Ahmadian, M.; Rahimi, S. Studying the efficiency of a biological aerated filter (BAF) with oyster Media on improving the quality of effluent produced by treatment plants. *Fresen. Environ. Bull.* **2014**, *23*, 2401–2406.
136. Li, S.-Z.; Li, X.-Y.; Wang, D.-Z. Membrane (RO-UF) filtration for antibiotic wastewater treatment and recovery of antibiotics. *Sep. Purif. Technol.* **2004**, *34*, 109–114. [CrossRef]
137. Liang, C.; Wei, D.; Zhang, S.; Ren, Q.; Shi, J.; Liu, L. Removal of antibiotic resistance genes from swine wastewater by membrane filtration treatment. *Ecotoxicol. Environ. Saf.* **2021**, *210*, 111885. [CrossRef]

138. Li, X.; Jiang, L.; Li, H. Application of ultrafiltration technology in water treatment. In *IOP Conference Series: Earth and Environmental Science*; IOP Publishing: Bristol, UK, 2018; Volume 186. [CrossRef]
139. Bourgeous, K.N.; Darby, J.L.; Tchobanoglous, G. Ultrafiltration of wastewater: Effects of particles, mode of operation, and backwash effectiveness. *Water Res.* **2000**, *35*, 77–90. [CrossRef]
140. Bera, S.P.; Godhaniya, M.; Kothari, C. Emerging and advanced membrane technology for wastewater treatment: A review. *J. Basic Microbiol.* **2021**, *62*, 245–259. [CrossRef]
141. Qiu, G.; Chen, H.; Srinivasa Raghavan, D.S.; Ting, Y.P. Removal behaviors of antibiotics in a hybrid microfiltration-forward osmotic membrane bioreactor for real municipal wastewater treatment. *Chem. Eng. J.* **2021**, *417*, 129146. [CrossRef]
142. Samrot, A.V.; Sahithya, C.S.; Selvarani, A.J.; Pachiyappan, S.; Kumar, S. Surface-engineered Super-Paramagnetic Iron Oxide Nanoparticles for Chromium Removal. *Int. J. Nanomed.* **2019**, *14*, 8105–8119. [CrossRef] [PubMed]
143. Samrot, A.V.; Sahithya, C.S.; Purayil, S.K.; Ponnaiah, P. A review on synthesis, characterization and potential biological applications of superparamagnetic iron oxide nanoparticles. *Curr. Res. Green Sustain. Chem.* **2020**, *4*, 100042. [CrossRef]
144. Barancheshme, F.; Munir, M. Strategies to Combat Antibiotic Resistance in the Wastewater Treatment Plants. *Front. Microbiol.* **2018**, *8*, 2603. [CrossRef]
145. Moarefian, A.; Golestani, H.A.; Bahmanpour, H. Removal of amoxicillin from wastewater by self-made Polyethersulfone membrane using nanofiltration. *J. Environ. Health Sci. Eng.* **2014**, *12*, 127. [CrossRef]
146. Abdel-Fatah, M.A. Nanofiltration systems and applications in wastewater treatment: Review article. *Ain Shams Eng. J.* **2018**, *9*, 3077–3092. [CrossRef]
147. Homem, V.; Santos, L. Degradation and removal methods of antibiotics from aqueous matrices—A review. *J. Environ. Manag.* **2011**, *92*, 2304–2347. [CrossRef] [PubMed]
148. Gholami, M.; Mirzaei, R.; Kalantary, R.R.; Sabzali, A.; Gatei, F. Performance evaluation of reverse osmosis technology for selected antibiotics removal from synthetic pharmaceutical wastewater. *Iran. J. Environ. Health Sci. Eng.* **2012**, *9*, 19. [CrossRef]
149. Dolar, D.; Vuković, A.; Ašperger, D.; Košutić, K. Efficiency of RO/NF membranes at the removal of veterinary antibiotics. *Water Sci. Technol.* **2012**, *65*, 317–323. [CrossRef]
150. Sahar, E.; David, I.; Gelman, Y.; Chikurel, H.; Aharoni, A.; Messalem, R.; Brenner, A. The use of RO to remove emerging micropollutants following CAS/UF or MBR treatment of municipal wastewater. *Desalination* **2011**, *273*, 142–147. [CrossRef]
151. Govardhan, B.; Fatima, S.; Madhumala, M.; Sridhar, S. Modification of used commercial reverse osmosis membranes to nanofiltration modules for the production of mineral-rich packaged drinking water. *Appl. Water Sci.* **2020**, *10*, 230. [CrossRef]
152. Cheng, H.; Hong, P.Y. Removal of Antibiotic-Resistant Bacteria and Antibiotic Resistance Genes Affected by Varying Degrees of Fouling on Anaerobic Microfiltration Membranes. *Environ. Sci. Technol.* **2017**, *51*, 12200–12209. [CrossRef]
153. Dave, S.; Das, J. Technological model on advanced stages of oxidation of wastewater effluent from food industry. In *Advanced Oxidation Processes for Effluent Treatment Plants*; Elsevier: Amsterdam, The Netherlands, 2021; pp. 33–49. [CrossRef]
154. Mahdi, M.H.; Mohammed, T.J.; Al-Najar, J. Advanced Oxidation Processes (AOPs) for treatment of antibiotics in wastewater: A review. *IOP Conf. Series Earth Environ. Sci.* **2021**, *779*, 012109. [CrossRef]
155. Akbari, M.Z.; Xu, Y.; Lu, Z.; Peng, L. Review of antibiotics treatment by advance oxidation processes. *Environ. Adv.* **2021**, *5*, 100111. [CrossRef]
156. Ahmed, M.B.; Zhou, J.L.; Ngo, H.H.; Guo, W. Adsorptive removal of antibiotics from water and wastewater: Progress and challenges. *Sci. Total. Environ.* **2015**, *532*, 112–126. [CrossRef]
157. Lakherwal, D. Adsorption of Heavy Metals: A Review. *Int. J. Environ. Res. Devel-Opment* **2014**, *4*, 41–48.
158. Farhana, A.; Selvarani, A.J.; Samrot, A.V.; Alsrhani, A.; Raji, P.; Sahithya, C.S.; Cypriyana, P.J.J.; Senthilkumar, P.; Ling, M.P.; Yishak, S. Utilization of Superparamagnetic Iron Oxide Nanoparticles (SPIONs) Impregnated Activated Carbon for Removal of Hexavalent Chromium. *J. Nanomater.* **2022**, *2022*, 4326939. [CrossRef]
159. Samrot, A.V.; Shobana, N. *Citrus sinensis* cellulose fibres incorporated with SPIONs for effective removal of crystal violet dye. *Biocatal. Agric. Biotechnol.* **2021**, *39*, 102211. [CrossRef]
160. Wang, X.; Yin, R.; Zeng, L.; Zhu, M. A review of graphene-based nanomaterials for removal of antibiotics from aqueous environments. *Environ. Pollut.* **2019**, *253*, 100–110. [CrossRef]
161. Eniola, J.O.; Kumar, R.; Barakat, M.A. Adsorptive removal of antibiotics from water over natural and modified adsorbents. *Environ. Sci. Pollut. Res.* **2019**, *26*, 34775–34788. [CrossRef]
162. Yang, S.-F.; Lin, C.-F.; Wu, C.-J.; Ng, K.-K.; Lin, A.Y.-C.; Hong, P.-K.A. Fate of sulfonamide antibiotics in contact with activated sludge—Sorption and biodegradation. *Water Res.* **2012**, *46*, 1301–1308. [CrossRef]
163. Samrot, A.V.; Angalene, J.L.A.; Roshini, S.M.; Raji, P.; Stefi, S.M.; Preethi, R.; Selvarani, A.J.; Madankumar, A. Bioactivity and Heavy Metal Removal Using Plant Gum Mediated Green Synthesized Silver Nanoparticles. *J. Clust. Sci.* **2019**, *30*, 1599–1610. [CrossRef]
164. Samrot, A.V.; Senthilkumar, P.; Rashmitha, S.; Veera, P.; Sahithya, C.S. *Azadirachta indica* influenced biosynthesis of superparamagnetic iron-oxide nanoparticles and their applications in tannery water treatment and X-ray imaging. *J. Nanostructure Chem.* **2018**, *8*, 343–351. [CrossRef]
165. Samrot, A.V.; Shobana, N.; Sruthi, P.D.; Sahithya, C.S. Utilization of chitosan-coated superparamagnetic iron oxide nanoparticles for chromium removal. *Appl. Water Sci.* **2018**, *8*, 192. [CrossRef]
166. Stafiej, A.; Pyrzynska, K. Adsorption of heavy metal ions with carbon nanotubes. *Sep. Purif. Technol.* **2005**, *58*, 49–52. [CrossRef]

167. Cong, Q.; Yuan, X.; Qu, J. A review on the removal of antibiotics by carbon nanotubes. *Water Sci. Technol.* **2013**, *68*, 1679–1687. [CrossRef]
168. Yu, F.; Ma, J.; Bi, D. Enhanced adsorptive removal of selected pharmaceutical antibiotics from aqueous solution by activated graphene. *Environ. Sci. Pollut. Res.* **2015**, *22*, 4715–4724. [CrossRef]
169. Asano, T.; Levine, A.D. Wastewater reclamation, recycling and reuse: Past, present, and future. *Water Sci. Technol.* **1996**, *33*, 1–14. [CrossRef]
170. Ajibode, O.M. Effect of Residence Time on Microbial and Chemical Quality of Reclaimed Water in Urban Infrastructures. Ph.D. Thesis, The University of Arizona, Tucson, AZ, USA, 2012.
171. Kihila, J.; Mtei, K.M.; Njau, K.N. Wastewater treatment for reuse in urban agriculture; the case of Moshi Municipality, Tanzania. *Phys. Chem. Earth Parts A/B/C* **2014**, *72–75*, 104–110. [CrossRef]
172. Angelakis, A.; Durham, B. Water recycling and reuse in EUREAU countries: Trends and challenges. *Desalination* **2008**, *218*, 3–12. [CrossRef]
173. Yang, H.; Abbaspour, K.C. Analysis of wastewater reuse potential in Beijing. *Desalination* **2007**, *212*, 238–250. [CrossRef]
174. Manouchehri, M.; Kargari, A. Water recovery from laundry wastewater by the cross flow microfiltration process: A strategy for water recycling in residential buildings. *J. Clean. Prod.* **2017**, *168*, 227–238. [CrossRef]
175. Al-Hashimia, M.A.I.; Abbas, T.R.; Jasema, Y.I. Performance of sequencing anoxic/anaerobic membrane bioreactor (SAM) system in hospital wastewater treatment and reuse. *Eur. Sci. J.* **2013**, *9*, 169–180.
176. Tiwari, B.; Sellamuthu, B.; Ouarda, Y.; Drogui, P.; Tyagi, R.D.; Buelna, G. Review on fate and mechanism of removal of pharmaceutical pollutants from wastewater using biological approach. *Bioresour. Technol.* **2017**, *224*, 1–12. [CrossRef] [PubMed]
177. Rodriguez-Mozaz, S.; Lucas, D.; Barceló, D. Full-Scale Plants for Dedicated Treatment of Hospital Effluents. *Hosp. Wastewaters Charact. Manag. Treat. Environ. Risks* **2017**, *60*, 189–208. [CrossRef]
178. Boehler, M.; Zwickenpflug, B.; Hollender, J.; Ternes, T.; Joss, A.; Siegrist, H. Removal of micropollutants in municipal wastewater treatment plants by powder-activated carbon. *Water Sci. Technol.* **2012**, *66*, 2115–2121. [CrossRef]
179. Golovko, O.; de Brito Anton, L.; Cascone, C.; Ahrens, L.; Lavonen, E.; Köhler, S.J. Sorption Characteristics and Removal Efficiency of Organic Micropollutants in Drinking Water Using Granular Activated Carbon (GAC) in Pilot-Scale and Full-Scale Tests. *Water* **2020**, *12*, 2053. [CrossRef]
180. Polianciuc, S.I.; Gurzău, A.E.; Kiss, B.; Ştefan, M.G.; Loghin, F. Antibiotics in the environment: Causes and consequences. *Med. Pharm. Rep.* **2020**, *93*, 231–240. [CrossRef]
181. Akhil, D.; Lakshmi, D.; Kumar, P.S.; Vo, D.-V.N.; Kartik, A. Occurrence and removal of antibiotics from industrial wastewater. *Environ. Chem. Lett.* **2021**, *19*, 1477–1507. [CrossRef]
182. Wu, H.; Niu, X.; Yang, J.; Wang, C.; Lu, M. Retentions of bisphenol A and norfloxacin by three different ultrafiltration membranes in regard to drinking water treatment. *Chem. Eng. J.* **2016**, *294*, 410–416. [CrossRef]
183. Baran, W.; Adamek, E.; Jajko, M.; Sobczak, A. Removal of veterinary antibiotics from wastewater by electrocoagulation. *Chemosphere* **2018**, *194*, 381–389. [CrossRef] [PubMed]
184. Anjali, R.; Shanthakumar, S. Insights on the current status of occurrence and removal of antibiotics in wastewater by advanced oxidation processes. *J. Environ. Manag.* **2019**, *246*, 51–62. [CrossRef] [PubMed]
185. Carbajo, J.B.; Petre, A.L.; Rosal, R.; Herrera, S.; Letón, P.; García-Calvo, E.; Fernández-Alba, A.R.; Perdigón-Melón, J.A. Continuous ozonation treatment of ofloxacin: Transformation products, water matrix effect and aquatic toxicity. *J. Hazard. Mater.* **2015**, *292*, 34–43. [CrossRef] [PubMed]