



# Remediation of wastewater contaminated by antibiotics. A review

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## Abstract

The rising presence of antibiotics in the environment induces the production of resistant genes and thus threatens animal health because there are actually few alternative antibiotics. Here, we review recent trends on antibiotic research in water. For that, we analyzed countries, institutes, journals and keywords of 5420 articles on antibiotics in water or wastewater published between 2000 and 2017. Findings show that China is the first contributor and that the USA has the highest h-index of 104. The major removal techniques are adsorption, photolysis and photocatalysis, biodegradation, ozonation and electrochemical oxidation. New materials and technologies, such as ionizing beam, are actually studied to improve efficiency and decrease cost. Conversion of wastewater into fuels such as H<sub>2</sub> and methane is also a current research topic.

**Keywords** Antibiotics · Bibliometrics · Adsorption · Photocatalysis · Biodegradation

## Introduction

The use of antibiotics has considerably increased since the last century. According to statistics, global antibiotic consumption has increased from 21.1 billion in 2000 to 34.8 billion defined daily doses in 2015 (Klein et al. 2018). Antibiotics are often used to treat human and animal diseases. They are also applied as feed additives in livestock husbandry and as ripening agents in agriculture. Instead of being fully metabolized in human and animal bodies, most of the antibiotics are excreted into wastewater through urine and feces, and the antibiotics in water or wastewater can only be partially degraded by conventional water treatment. Actually, municipal, agricultural and industrial wastewaters are the main sources of antibiotics and their by-products in the environment. In addition, the sewage treatment plant

effluents contribute the most (Boyd et al. 2003; Carvalho and Santos 2016; Makowska et al. 2016).

Antibiotics that have the potential to propagate antibiotic resistance genes in the environment and are hard to be degraded have been regarded as emerging pollutants. Although some microbial organisms in the environment are originally antibiotic-resistant, the large-scale use of antibiotics undoubtedly accelerates the proliferation of antibiotic-resistant microorganisms and the spread of antibiotic-resistant genes. Even trace antibiotic concentrations have a substantial impact on the abundance of resistant genes in the environment (Peak et al. 2007; Knapp et al. 2010; Johansson et al. 2014).

In this study, the development history and research trends regarding antibiotic wastewater were explored through bibliometrics, which combines mathematics, statistics and philology for quantitative research on the literature (Ho and Kahn 2014). This method has been applied to various fields, including medicine (Zhou et al. 2018); geography (Meerow et al. 2016); operation research and management science (Fahimnia et al. 2015); energy and fuel (Imran et al. 2018); and environmental science and ecology (Ma et al. 2018; Ren et al. 2018). Through this method, 5852 publications related to antibiotics in water or wastewater published between 2000 and 2017 were obtained from the SCI-Expanded database, and their author keywords, countries, journals and other information were analyzed. On this basis, research trends and hot spots in this field were explored for

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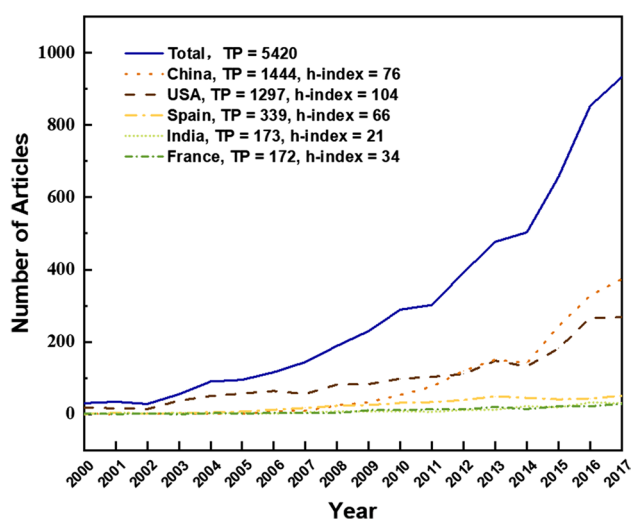
the comprehensive understanding of research status and providing potential research topics in the future.

## Methods

The SCI-Expanded database was selected as data sources, and the keywords '(antibiotic\*)' and '(wastewater or water or aquatic)' were used for the searching of titles, abstracts and keywords. A total of 5852 publications published between 2000 and 2017 were obtained on January 20, 2018 and analyzed in terms of document type, output, journal, country, institute, author keyword and h-index by Microsoft Excel 2016. The impact factor (IF) was obtained from the 2018 edition of the Journal Citation Reports. In the Research hot spots section, numerous articles were utilized, and the keyword co-occurrence analysis was employed from a quantitative viewpoint for the collation of research hot spots related to antibiotics in water or wastewater. Meanwhile, Pajek 5.05 and BibExcel 1.0.0.0 were used for the analysis of keyword co-occurrence.

## Publication performance

Of the 5852 publications, 98.75% were published in English, while 0.44% were in Chinese, ranking second. In addition, among all the 5852 publications, 5420 articles accounted for the highest percentage (92.58%) of all document types. As shown in Fig. 1, the number of publications has rapidly grown in this century because of the rapid increase in antibiotics consumption and the increased attention of people to environmental pollution (Friedrich 2018).



**Fig. 1** Number of articles on antibiotics in water or wastewater (from 2000 to 2017). TP: total publications

Articles coming from the USA, Spain and France accounted for 63.3% of all the articles. As shown in Fig. 1, the research on antibiotic wastewater was firstly conducted in the USA, and the number of articles regarding this topic has steadily increased since then. China began to present a rapid growth in 2008, and the number of articles from China even exceeded that of the USA in 2012 possibly because China accounted for nearly half of the world's total antibiotic consumption and Chinese environment and residents have been exposed to a serious threat from these compounds. Although the number of Chinese articles ranked first, the h-index of the USA was in the first place, which indicated the largest influence of the USA in this field. Spain, India and France have also demonstrated gradual development with relatively slower speed compared with that of China and the USA.

The number of articles from top 20 productive institutes accounted for 15.64% of the total (Table 1). The majorities of the institutes come from China, and 214 papers were published by Chinese Acad Sci, putting the institute first; besides, its h-index ranked first. The rest of the top five institutes were Tongji University, University of Porto, Tsinghua University and Nanjing University.

## Characteristics of journals

Approximately 39.61% of the 5420 articles were from the top 15 productive journals (Table 2). The impact factors of most of these journals were higher than those of other journals in the field of environment, indicating that antibiotic in water or wastewater has been thoroughly researched and has been considered a high-profile environmental issue. The top three journals are Chemosphere, Science of the Total Environment and Water Research (Table 2). Since 2012, the number of articles on Environmental Science and Pollution Research has rapidly grown and even exceeded Chemosphere in 2017, indicating that the journal highly considers the issue of antibiotic wastewater.

## Research hot spots

The 25 most frequently used keywords were grouped into three 6-year periods during 2000–2017. Their rankings and percentages are shown in Table 3.

In addition, Fig. 2 shows the chart of author keywords co-occurrence analysis, which demonstrates the correlation degree between the keywords. The thicker and darker the line is, the stronger the correlation is.

**Table 1** Top 20 productive institutes during 2000–2017

Institute	h-index	TP	TPR (%)	SPR (%)	CPR (%)	FPR (%)
Chinese Acad Sci, China	21	214	1 (4.0)	1 (3.1)	1 (8.0)	1 (5.1)
Tongji Univ, China	10	51	2 (0.9)	8 (0.9)	5 (1.8)	2 (1.2)
Univ Porto, Portugal	7	49	3 (0.9)	3 (1.3)	3 (1.9)	4 (0.9)
Tsinghua Univ, China	8	46	4 (0.9)	8 (0.9)	2 (2.0)	3 (1.0)
Nanjing Univ, China	8	45	5 (0.8)	5 (1.0)	8 (1.4)	7 (0.8)
Univ Barcelona, Spain	10	40	6 (0.7)	2 (1.5)	9 (1.3)	14 (0.4)
Zhejiang Univ, China	10	37	7 (0.7)	16 (0.5)	11 (1.1)	8 (0.8)
Istanbul Tech Univ, Turkey	9	35	8 (0.7)	16 (0.5)	13 (0.9)	9 (0.8)
Dalian Univ Technol, China	5	34	9 (0.6)	11 (0.8)	14 (0.6)	13 (0.6)
Univ Hong Kong, China	6	30	10 (0.6)	12 (0.7)	6 (1.5)	11 (0.6)
Univ Girona, Spain	7	28	11 (0.5)	20 (0.3)	4 (1.9)	10 (0.7)
Univ Granada, Spain	3	28	11 (0.5)	4 (1.2)	20 (0.3)	20 (0.2)
Harbin Inst Technol, China	7	28	11 (0.5)	16 (0.5)	7 (1.5)	4 (0.9)
Jiangsu Univ, China	9	28	11 (0.5)	15 (0.5)	12 (1.1)	6 (0.9)
Univ Minnesota, USA	4	27	15 (0.5)	5 (1.0)	17 (0.4)	19 (0.2)
Shandong Univ, China	4	27	15 (0.5)	5 (1.0)	17 (0.4)	16 (0.4)
Nankai Univ, China	8	26	17 (0.5)	19 (0.4)	10 (1.2)	11 (0.6)
Univ Zagreb, USA	3	25	18 (0.5)	8 (0.9)	19 (0.4)	18 (0.2)
Univ Sao Paulo, Brazil	3	24	19 (0.4)	14 (0.6)	15 (0.5)	15 (0.4)
Univ Aveiro, Portugal	4	24	19 (0.4)	13 (0.7)	16 (0.5)	17 (0.3)

TP, the number of total publications; TP R (%), rank and the percentage of the total publications; SP R (%), rank and the percentage of single-institute publications; CP R (%), rank and the percentage of inter-institute collaborative publications; FP R (%), rank and the percentage of first-author institute publications; and RP R (%), rank and the percentage of corresponding-author institute publications

**Table 2** Top 15 productive journals during 2000–2017

Journal name	TP	Percent	IF 2018	Total cites 2018	Eigenfactor score 2018
Chemosphere	283	5.22	4.427	66,954	0.06839
Science of the Total Environment	267	4.93	4.610	71,249	0.10865
Water Research	261	4.82	7.051	76,647	0.07280
Environmental Science and Technology	193	3.56	6.653	145,022	0.18373
Environmental Science and Pollution Research	171	3.15	2.800	23,733	0.05124
Journal of Hazardous Materials	162	2.99	6.434	88,496	0.06852
Water Science and Technology	129	2.38	1.247	19,181	0.01074
Chemical Engineering Journal	126	2.32	6.735	73,657	0.10438
Journal of Chromatography A	116	2.14	3.716	61,361	0.05092
Environmental Pollution	90	1.66	4.358	38,318	0.04232
Bioresource Technology	79	1.46	5.807	101,191	0.10945
Desalination and Water Treatment	75	1.38	1.383	10,851	0.01767
Plos One	65	1.20	2.766	582,877	1.86235
Applied and Environmental Microbiology	65	1.20	3.633	100,091	0.07189
Frontiers in Microbiology	65	1.20	4.019	25,111	0.08495

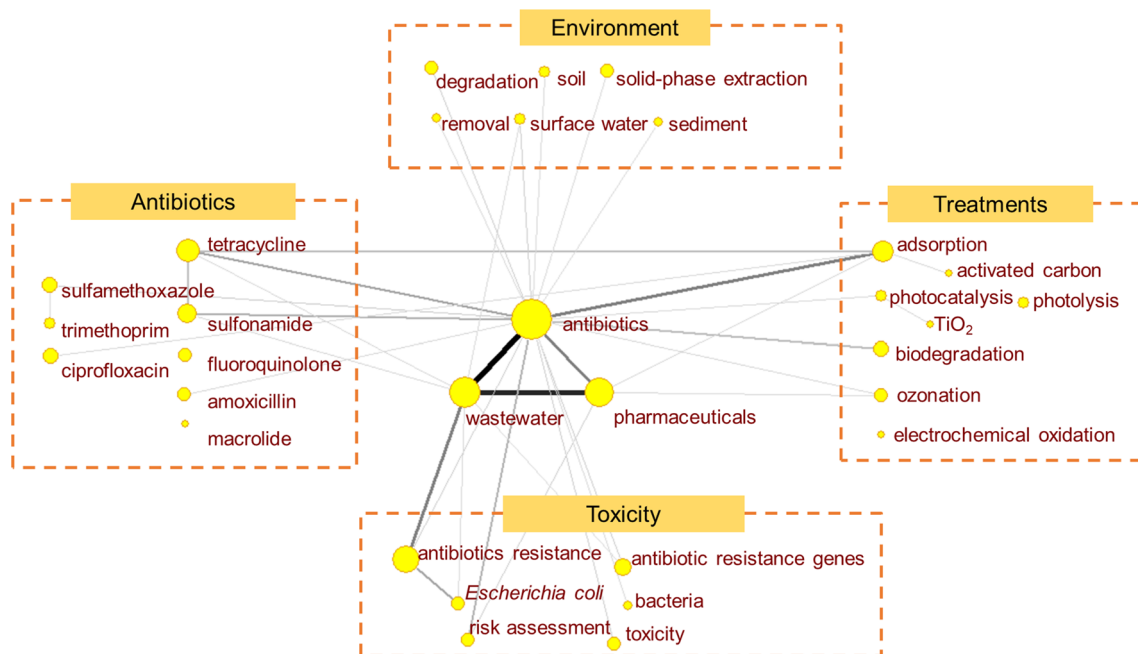
On the basis of the above information, the paper provides a brief introduction to the research hot spots of typical ones,

toxicity and environment behavior of antibiotics in water or wastewater and then elaborates on current status of treatment technologies.

**Table 3** Top 25 most frequently used author keywords

Author keyword	2000–2017		2000–2005		2006–2011		2012–2017	
	TP	R (%)	TP	R (%)	TP	R (%)	TP	R (%)
Antibiotics	934	1 (20.6)	61	1 (26.1)	255	1 (24.7)	490	1 (15.0)
Wastewater	810	2 (17.9)	28	2 (12.0)	149	3 (14.5)	488	2 (14.9)
Pharmaceuticals	495	3 (10.9)	27	3 (11.5)	169	2 (16.4)	221	3 (6.8)
Antibiotic resistance	425	4 (9.4)	25	4 (10.7)	81	4 (7.9)	214	4 (6.5)
Tetracycline	271	5 (6.0)	10	8 (4.3)	72	5 (7.0)	161	5 (4.9)
Adsorption	265	6 (5.8)	8	12 (3.4)	33	11 (3.2)	159	6 (4.9)
Sulfonamides	218	7 (4.8)	7	16 (3.0)	59	6 (5.7)	95	8 (2.9)
Antibiotic resistance genes	177	8 (3.9)	1	149 (0.4)	2	311 (0.2)	149	7 (4.6)
Biodegradation	133	9 (2.9)	11	6 (4.7)	39	8 (3.8)	65	16 (2.0)
Ciprofloxacin	125	10 (2.8)	4	24 (1.7)	27	14 (2.6)	94	9 (2.9)
<i>Escherichia coli</i>	124	11 (2.7)	11	6 (4.7)	35	10 (3.4)	57	22 (1.7)
Sulfamethoxazole	120	12 (2.6)	2	65 (0.9)	26	16 (2.5)	92	10 (2.8)
Surface water	120	12 (2.6)	4	24 (1.7)	25	17 (2.4)	61	19 (1.9)
Ozonation	109	14 (2.4)	8	12 (3.4)	39	8 (3.8)	29	42 (0.9)
Photocatalysis	105	15 (2.3)	1	149 (0.4)	13	38 (1.3)	68	15 (2.1)
Solid-phase extraction	105	15 (2.3)	3	37 (1.3)	55	7 (5.3)	33	36 (1.0)
Toxicity	93	17 (2.1)	2	65 (0.9)	30	12 (2.9)	61	19 (1.9)
Photolysis	90	18 (2.0)	1	149 (0.4)	25	17 (2.4)	29	42 (0.9)
Risk assessment	89	19 (2.0)	4	24 (1.7)	15	31 (1.5)	70	12 (2.1)
Fluoroquinolones	89	19 (2.0)	8	12 (3.4)	29	13 (2.8)	79	11 (2.4)
Amoxicillin	88	21 (1.9)	3	37 (1.3)	21	19 (2.0)	64	17 (2.0)
Degradation	88	21 (1.9)	3	37 (1.3)	16	29 (1.6)	69	13 (2.1)
Advanced oxidation processes	88	21 (1.9)	3	37 (1.3)	19	23 (1.8)	69	13 (2.1)
Oxytetracycline	83	24 (1.8)	6	17 (2.6)	17	26 (1.7)	62	18 (1.9)
Sediment	82	25 (1.8)	4	24 (1.7)	17	26 (1.7)	41	27 (1.3)

TP, total publications; R (%), ranking and percentage of author keywords

**Fig. 2** Analysis of keywords co-occurrence

## Typical antibiotics

After statistical analysis, the five most investigated classes of antibiotics are tetracyclines, sulfonamides, quinolones,  $\beta$ -lactams and macrolides, and articles of these antibiotics have been increasing since 2000. Tetracyclines, sulfonamides and quinolones have always remained in the top three from 2000 to 2017. Articles on sulfonamides grew fastest, whereas those on macrolides were the slowest (Fig. 3).

Table 3 shows that sulfamethoxazole has been studied the most in all sulfonamides probably because the consumption of sulfamethoxazole was reportedly the biggest, and the concentration in wastewater treatment plant was usually from ng/L to  $\mu$ g/L (Achermann et al. 2018). Moreover, fluoroquinolones are the most researched quinolones. On the contrary, macrolides have received less attention. The 12 most studied compounds of the five classes are listed in Table 4.

## Toxicity and environment behavior

It is reported that the major hazard of residual antibiotics in the environment is to promote the development of antibiotic-resistant bacteria, pose a threat to human health and incur damage to aquatic flora and fauna at variable extents (Cizmas et al. 2015; Daghri and Drogui 2013; Kim et al. 2007; Zhang et al. 2017). However, the production of antibiotic-resistant genes is regarded to be the most important aspect of environmental toxicity posed by antibiotics. Thus, among the 25 author keywords, ‘antibiotic resistance genes’ represents the most remarkable change from 149th (0.4%) in 2000–2005 to seventh (4.6%) in 2012–2017 (Table 3). The characteristics of antibiotic-resistant genes, including persistence, transfer and conversion, have remained the important topics of research on antibiotics in aquatic environment and will continue to gain attention in the foreseeable future.

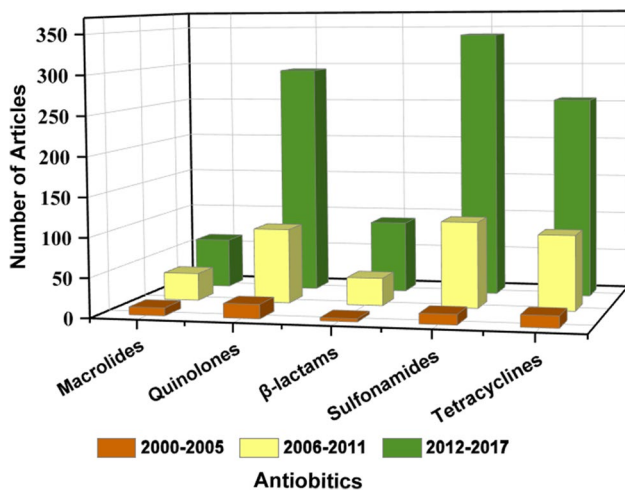


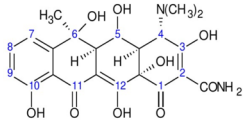
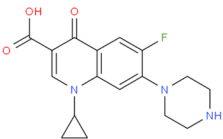
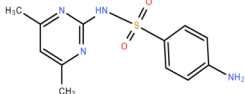
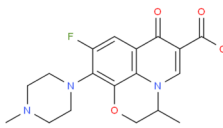
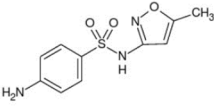
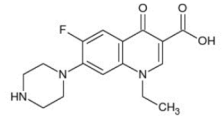
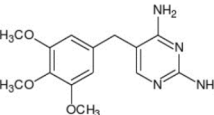
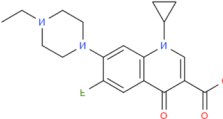
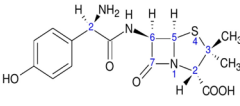
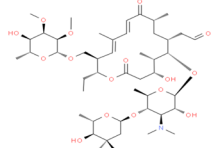
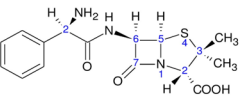
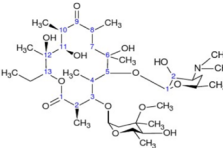
Fig. 3 Growth trends of the top five antibiotics

Moreover, along with degradation pathways, the characteristics of antibiotics as inducers of antibiotic-resistant genes are also regarded as critical issues. These characteristics include the migration and transformation of antibiotics in different environments. As mentioned above, wastewater treatment plants are the main sources of antibiotics in the environment. As shown in Fig. 2, antibiotics, which exist in surface water, soil and sediment, migrating from the aquatic environment have attracted a lot of attention. Antibiotics in surface water can be adsorbed by the sediment, and antibiotics have been detected in the sediment (Bagnis et al. 2018; Zhang et al. 2018a; Liu et al. 2018). Sung-Chul and Kenneth found that the concentrations of tetracyclines, sulfonamides and macrolides in sediments were even higher than in water at the same sites of Cache La Poudre River (Sung-Chul and Kenneth 2007). As for groundwater, it may be contaminated by antibiotics due to infiltration and leakage. In addition, domestic sewage containing antibiotics can directly go into the soil or pass through it via composting, irrigation, etc., and antibiotics in soil can go back to the aquatic environment via the surface runoff. Chen et al. (2015a) ever reported that fluoroquinolones were the most commonly detected antibiotics in soil/groundwater, which got into the environment mainly due to the discharge of wastewater treatment plant effluents and landfill leachates. Above all, the migration and diffusion of antibiotics from the aquatic environment matter to the whole contamination of the environment.

## Transformation of antibiotics in aquatic environment

Antibiotics in aquatic environment experience a lot of transformation processes, including biodegradation, hydrolysis, photolysis, adsorption, etc. However, due to the influence of human activities, there still exist a lot of antibiotics in the water body. The presences of antibiotics has been confirmed in China, the USA, Europe and so on (Zuccato et al. 2010; Ojemaye and Petrik 2019; Szymańska et al. 2019). The concentrations of antibiotics detected in seawater ranged from 0.21 to 5000 ng/L. The pH, salinity, ionic strength and conductivity of water may affect the behavior of the antibiotics in the marine environment (Ojemaye and Petrik 2019). The concentrations of antibiotics in rivers are usually in the range of tens to hundreds ng/L, while antibiotic contamination in groundwater is relatively slight, with the concentration generally below tens ng/L (Carvalho and Santos 2016). Antibiotics also exist in drinking water. For example, 23 antibiotics have been detected in the drinking water in the Chinese countryside, the most common of which are fluoroquinolones and macrolides, and the concentrations ranged from 0 to 368.21 ng/L. Generally, the concentration of antibiotics in rural drinking water is higher than that in cities, because the countryside has a relatively poor ability for drinking water

**Table 4** Top 12 representative compounds among the five antibiotic classes

Class	Representative compounds	Structural formula	Class	Representative compounds	Structural formula
Tetracyclines	Oxytetracyclines			Ciprofloxacin	
	Sulfamethazine			Ofloxacin	
Sulfonamides	Sulfamethoxazole		Quinolones	Norfloxacin	
	Trimethoprim			Enrofloxacin	
	Amoxicillin			Tylosin	
β-lactams	Ampicillin		Macrolides	Erythromycin	

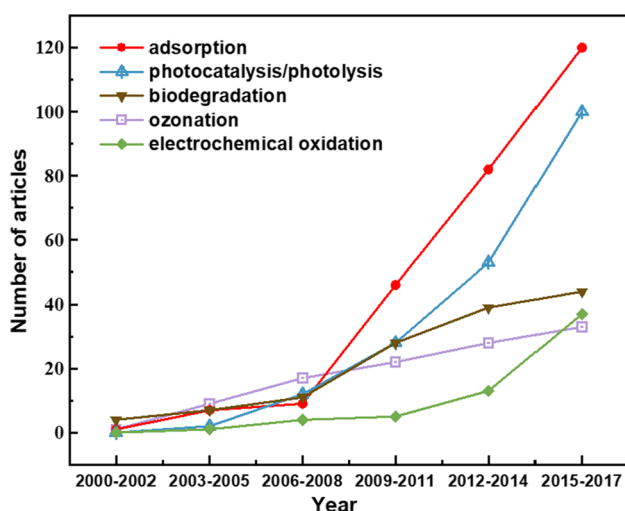
treatment (Wang et al. 2016; Meng et al. 2019). The antibiotics detected in different aquatic environments are not the same. However, quinolones usually get the highest detection rate. In addition, macrolides and sulfonamides also pose a greater threat to the environment (Zuccato et al. 2010; Carvalho and Santos 2016; Liu et al. 2018).

### Most investigated treatment technologies

Table 3 reveals that the five most investigated treatment technologies of antibiotics in water or wastewater are adsorption, photolysis/photocatalysis, biodegradation, ozone and electrochemical oxidation. The number of articles on these technologies shows a growth trend.

The numbers of articles on adsorption, photolysis/photocatalysis, biodegradation, ozonation and electrochemical oxidation are 265, 195, 133, 110 and 60, respectively.

Adsorption is speculated to be the most used technology, and average growth rate of its article number is the fastest. Additionally, the total number of articles on photolysis/photocatalysis is large and maintained a fast growth rate. By contrast, studies on biodegradation, ozonation and electrochemical oxidation are fewer than the two technologies. Unlike ozonation and biodegradation, the development of electrochemical oxidation has a positive trend and shows better development perspective (Fig. 4). In 2015–2017, the number of articles on electrochemical oxidation surpassed ozonation, and the total number of articles



**Fig. 4** Growth trends of the top five treatments

on electrochemical oxidation was forecasted to rank third, which is only behind adsorption and photolysis/photocatalysis in the near future.

### Adsorption

Figure 2 shows a high correlation between ‘antibiotics’ and ‘adsorption.’ Furthermore, the relationship between ‘adsorption’ and ‘activated carbon’ is close, indicating that activated carbon is the most investigated adsorbent. This is because the low cost, the well-developed pore structure of activated carbon and, in addition, the rich surface functional groups make it possible to adsorb antibiotics via physical adsorption as well as chemical adsorption. In the adsorption process, activated carbon type, precursors, activation methods, adsorbates and operating conditions (pH, adsorbent dosage and ionic strength) all have impact on adsorption efficiency (Mansour et al. 2018). For example, the selection of granular activated carbon particle size plays an important role in the

removal of specific pollutants, and the smallest sizes exhibited the highest removal efficiency which was probably due to the increased mass transfer (Östman et al. 2019). Various modification methods are available for changing the pore structure and surface chemistry characteristics of activated carbon in order to get an increase in adsorption capacity. As seen in Table 5, the surface area of five typical activated carbons was increased, and No. 3 case introduced amino functional groups to further enhance the adsorption effect simultaneously.

In the research of adsorption, tetracycline is the most studied antibiotics because the keyword ‘tetracycline’ frequently occurs and is closely connected with ‘adsorption’ (Fig. 2). Tetracycline is a broad-spectrum and affordable antibiotic. Polar functional groups, such as carboxyl and acylamino, provide tetracycline with a strong affinity with environmental medium (Figuerola et al. 2004; Pan and Chu 2016), which leads to the strong adsorption of tetracycline in soil, clay and sediments, and tetracyclines usually exist in the soil for a long time because of their strong adsorption and refractory property. Also, adsorption is inferred to be an efficient method for the treatment of tetracycline wastewater. Oberlé et al. (2012) reported that owing to the crucial role of adsorption, the activated sludge system achieved a tetracycline removal rate of 93.2%.

Apart from tetracycline, the keyword ‘ciprofloxacin (CIP)’ of fluoroquinolones appears frequently and is closely related to ‘adsorption.’ The groups in fluoroquinolones, such as carbonyl (C=O), fluorine atom group (F<sup>-</sup>) and carboxyl (–COOH), facilitate soil adsorption through complexation, electrostatic attraction and van der Waals force and contribute to the difficulty of migration (Zhang and Huang 2007). Li et al. synthesized one-step carbonized nanoporous carbon (NPC) derived from zeolitic imidazolate framework-8 (ZIF-8) and used it to remove CIP from water. The optimized NPC-700 (carbonized at 700 °C for 2 h) did not exhibit considerable loss of adsorption after seven cycles with the action of interactions and hydrophobic interactions

**Table 5** Adsorption of some antibiotics by activated carbon (AC)

No	Antibiotic	Different types of AC	Maximum adsorption capacity	Ref.
1	Chlortetracycline	Synthesized from dried pomegranate wood and modified by NH <sub>4</sub> Cl solution	482.5 mg/g	Alahabadi et al. (2017)
2	Tetracycline	Prepared from apricot shell and activated by H <sub>3</sub> PO <sub>4</sub>	308.3 mg/g	Marzbali et al. (2016)
3	Ciprofloxacin/norfloxacin	Derived from bamboo product wastes, magnetized by Fe <sub>3</sub> O <sub>4</sub> and modified by certain amination reactions	293.2 mg/g and 315.7 mg/g, respectively (bamboo-based AC: 172.5 mg/g and 193.4 mg/g, respectively)	Peng et al. (2018)
4	Equilibrium	Prepared from A.L. seed pods and activated by microwave-assisted KOH and K <sub>2</sub> CO <sub>3</sub> activations	137.0 mg/g (KOH) 118.1 mg/g (K <sub>2</sub> CO <sub>3</sub> )	Ahmed and Theydan (2012)
5	Sulfamethazine	Modified by FeCl <sub>3</sub>	17.2 mg/g (AC: 3.1 mg/g)	Liu et al. (2017b)

(Li et al. 2017). Li et al. (2018) coated a layer of silica gel on ZIF-8 nanoparticles, and the maximum CIP adsorption amount reached 516.8 mg/g, which is higher than that of NPC-700 (416.7 mg/g) because of the increased electrostatic interaction.

Sulfonamides and  $\beta$ -lactams are comparably difficult to be adsorbed by solid phases. However, due to the advantages of easy operation and low cost, adsorption is still the most common wastewater treatment technology for the two classes of antibiotic.

Adsorbents that can be used in antibiotic removal include carbon materials, such as activated carbon, biochar and graphene oxide, and nanocarbon materials, such as carbon nanotubes and nanoporous carbon (Chen et al. 2015b, 2019; Mohammadi et al. 2015; Li et al. 2017); other materials include bentonite, chitosan beads, etc. (Adriano et al. 2005; Weng et al. 2017, 2018; Ifebajo et al. 2019).

Bentonite is a newly developed material that is cheaper than activated carbon. Bentonite modified with hexadecyl trimethyl ammonium to adsorb amoxicillin from aqueous solution could reach a removal rate higher than 80% (Zha et al. 2013). Weng et al. (2014, 2017) prepared bentonite-supported nanoscale zerovalent iron and bentonite-supported bimetallic Fe/Ni nanoparticles, and both of them had good performance on amoxicillin adsorption.

Adsorption is a technology with the advantages of low energy consumption, easy operation, no by-product and so on (Crini et al. 2019). However, this technology could only achieve the phase transfer of antibiotics but not the degradation. In the meanwhile, the adsorbent which has good performance on adsorption is often of small particle size and the separation of adsorbent from water would be difficult. Besides, the corrosion resistance, the recovery of the adsorbents and the treatment of waste adsorbents restrict the widespread application and need more investigation.

### Photolysis and photocatalysis

Photolysis/photocatalysis is the second most investigated treatment technology after adsorption. Although photolysis and photocatalysis use light, the mechanisms of the two processes are different. Photolysis has three pathways, namely direct photolysis, indirect photolysis and self-sensitization photolysis. Direct photolysis refers to pyrolysis, heterolysis and photo-ionization of compounds after adsorption of photons. Indirect photolysis is caused by the substances originally present in the environment, which absorb light energy and produce highly reactive substances, such as singlet oxygen ( $^1\text{O}_2$ ), hydroxyl radicals ( $\cdot\text{HO}$ ) and alkyl peroxy radicals (OOR), to degrade organic matter. As self-sensitized photolysis process, organics shift to excited state by absorbing light energy, transferring energy to the ground-state  $^3\text{O}_2$  or  $\text{H}_2\text{O}$  and producing reactive oxygen species, such as

$^1\text{O}_2$  and  $\cdot\text{OH}$ , which degrade the ground-state molecules of the organics (Fatta-Kassinos et al. 2011; Xie et al. 2018). The research on photolysis includes the fate of antibiotics in water and the influence of compound structure, pH and dissolved substances in water on photolysis process, photolysis kinetics, degradation products and toxicity (Boreen et al. 2004; Snowberger et al. 2016; Arsand et al. 2018).

Photolysis is of great importance to the fate of some antibiotics in the environment, such as tetracycline and fluoroquinolone, which contain aromatic rings, heteroatoms and other functional chromophores (Fatta-Kassinos et al. 2011). The main route of the removal of oxytetracycline in the surface water is direct photolysis (Jin et al. 2017; Chen et al. 2008). However, completely mineralizing tetracycline is difficult due to the aromatic ring system, which contributes to the stability and toxicity of the by-products in the oxidation process. López-Peñalver et al. determined the biotoxicity through the luminescence inhibition rate of *Vibrio fischeri*. When tetracycline was dissolved in surface water, the toxicity of tetracycline was found to increase from 12.6 to 39.9% after 60 min of photolysis (López-Peñalver et al. 2010). Unlike oxytetracycline, norfloxacin undergoes direct photolysis and self-sensitized photolysis (Liang et al. 2015). However, photolysis has its limitation that it only works with photosensitive antibiotics such as tetracyclines.

Catalyst is the main research topic of photocatalysis. Figure 2 shows that  $\text{TiO}_2$  has been the most researched catalyst since 2000 due to its advantages of low cost, nontoxicity and stability (Elmolla and Chaudhuri 2010; Kanakaraju et al. 2014). However,  $\text{TiO}_2$  requires ultraviolet radiation for photocatalytic activation. With ultraviolet light accounting for only 5% of solar energy,  $\text{TiO}_2$  demonstrates its ineffectiveness in the sunlight (Kumar et al. 2019; Chong et al. 2010). In recent years, different modification methods have been applied to  $\text{TiO}_2$  to improve its photocatalytic activity in the visible region, including  $\text{TiO}_2$  doping with metals (such as Cu, Ni and Au), nonmetals (such as N), Co-doping  $\text{TiO}_2$  nanomaterials and surface modification of organic ligands. In addition, improving the adsorption capacity through the modification of the  $\text{TiO}_2$  surface is another way to enhance the photocatalytic activity, and this process includes surface organic modification, doping with carbon-based nanoparticles and stabilization and separation of  $\text{TiO}_2$  particles (Dong et al. 2015). Elmolla and Chaudhuri (2010) found that amoxicillin in aqueous solution cannot be considerably degraded by  $\text{TiO}_2$  under UVA (365 nm), but the addition of  $\text{H}_2\text{O}_2$ , which would be beneficial to the production of  $\cdot\text{OH}$ , resulted in complete degradation of amoxicillin, ampicillin and cloxacillin after 30 min, revealing the high efficiency of UV/ $\text{H}_2\text{O}_2$ / $\text{TiO}_2$  photocatalysis. On the other hand, some visible-light-driven photocatalysts, such as  $\text{g-C}_3\text{N}_4$ , BiOX ( $X = \text{Cl, Br and I}$ ) and  $\text{Ag}_3\text{PO}_4$ , have been developed and modified to improve performance (Yi et al. 2010; Xiang



**Table 6** Degradation of antibiotics by some visible-light-driven photocatalysts

No	Antibiotic	Experimental conditions	Removal (%)	Ref.
1	Tetracycline	AgI/BiVO <sub>4</sub> was synthesized by an in situ precipitation procedure, under visible light, 2 h	94.1	Chen et al. (2016)
2	Ciprofloxacin	Carbon quantum dots (CQDs)/Bi <sub>2</sub> WO <sub>6</sub> were synthesized by a facile hydrothermal method, under visible light, 2 h	87	Di et al. (2015)
3	Ciprofloxacin	Graphene-like BN/BiOBr was prepared by an ionic liquid-assisted solvothermal method, under visible light, 80 min	81.5	Di et al. (2016)
4	Sulfamethoxazole	RGO-WO <sub>3</sub> was synthesized by a facile one-step hydrothermal method, under visible light, 3 h	> 98	Zhu et al. (2017)
5	Amoxicillin	MIL-68(In)-NH <sub>2</sub> /GrO was synthesized by a simple solvothermal method, under visible light, 2 h	> 93	Yang et al. (2017)

et al. 2011; Shabani et al. 2019; Di et al. 2016; Dehghan et al. 2018). Table 6 lists some of the visible-light-driven catalysts used in the treatment of antibiotic wastewater; all of these promoted efficient separation of photoinduced electron–hole pairs.

Recently, photocatalysis has received more and more attention. As one of the advanced oxidation processes, it exhibits high mineralization efficiency for refractory organic pollutants. However, the widely applied photocatalyst, TiO<sub>2</sub>, only responds to ultraviolet light and the newly developed visible-light-response catalysts such as g-C<sub>3</sub>N<sub>4</sub> face a problem of low quantum yield. Therefore, the large-scale application of these photocatalysts still has a long way to go.

### Ozonation

Similar to photocatalysis, ozonation is also one of the advanced oxidation processes, and its mechanisms comprise direct oxidation by ozone and indirect oxidation by ·OH. Alkaline ozonation is found to result in high COD (chemical oxygen demand) and TOC (total organic carbon) removal rates because of the increase in reactive radicals (·OH) and the occurrence of indirect reaction, whereas direct oxidation dominates the reaction under acidic condition and shows weak capability for eliminating pollutants (Arslan-Alaton and Dogruel 2004). The research hot spots of the study on antibiotic removal by ozone alone include optimal process conditions, kinetic mechanisms, degradation pathways, removal of resistant genes and toxicity of degradation product (Almomani et al. 2016; Feng et al. 2016a; Zheng et al. 2017). Hoigné and Bader (1983) reported that deprotonated species were strong nucleophiles and reacted fast with the electrophilic ozone. Furthermore, Huber et al. found that sulfamethoxazole and carbamazepine reacted fast with ozone due to the aromatic amino groups and double bond connecting the two phenyl moieties as reaction sites, respectively. However, roxithromycin reacted slowly due to the predominance of nonreactive protonated amine (Huber et al. 2003).

The sterilization ability of ozone was stronger than that of ultraviolet and chlorination, but its function of disinfection leads to apoptosis and release of antibiotic-resistant genes into the environment. The antibiotic-resistant genes removal efficiency cannot be substantially enhanced by increasing the ozone concentration (Zheng et al. 2017), which is speculated to be the major reason why the proportion of articles on ozone declines in Table 3 from the 12th in 2000–2005 to the 42nd in 2012–2017. The synergy between ozone and other technologies is often studied. Guo et al. showed that the addition of ultrasound during ozonation treatment increased the reaction rate by 6–26% at different pH levels. When degrading sulfadiazine in water, the combined UV/O<sub>3</sub> process demonstrated better performance in degradation efficiency and mineralization than the UV or O<sub>3</sub> treatment alone under the same conditions (Guo et al. 2015, 2016). In addition, the strong ability of ozone to capture electrons promotes the separation of photoinduced electron–hole pairs and the self-decay of ozone to generate ozonide ion radicals (·O<sub>3</sub><sup>-</sup>) and convert them into ·OH, so the organic matter can be effectively degraded. Xiao et al. (2016) reported that the coupling coefficient of ozone with the g-C<sub>3</sub>N<sub>4</sub> photocatalyst was 95.8%.

Ozonation can remove antibiotics effectively via direct or indirect oxidation, but only remove the antibiotic-resistant genes partially. In order to improve the effluent quality, the research on ozonation would focus on the combination of ozone and UV, biodegradation, H<sub>2</sub>O<sub>2</sub> or other technologies.

### Biodegradation

Biodegradation is not only a crucial degradation pathway of antibiotics in the environment but also one of the commonly used treatment technologies for antibiotic wastewater in the sewage treatment plant. The research hot spots of biodegradation include the conditions, effects and mechanisms (Peng et al. 2019; Zheng et al. 2019). Activated sludge process is the most commonly used technology, and its efficiency is mainly affected by the sludge adsorption and biodegradation

performances. Antibiotics such as norfloxacin, ciprofloxacin, ofloxacin and ampicillin in quinolones, ampicillin in  $\beta$ -lactams and tetracyclines are removed mainly by adsorption, while others such as cephalexin in  $\beta$ -lactams are mainly removed by biodegradation (Li and Zhang 2010; Yang et al. 2011).

Sulfonamide antibiotics cannot be completely removed by conventional activated sludge treatment systems. Adamek et al. (2016) found that in aerobic biodegradation of the four selected sulfa antibiotics (sulfanilamide, sulfamethoxazole, sulfadiazine and sulfathiazole), sulfathiazole was the easiest to be biodegraded, and sulfamethoxazole was the most resistant to biodegradation. Alvarino et al. (2016) reported that the maximum mineralization rate of sulfamethoxazole was 3% in heterotrophic aerobic conditions, whereas the mineralization rate was only 2.2% in anaerobic conditions. Furthermore, sulfamethoxazole in freshwater is the most stable compound and difficult to be biodegraded; the biodegradation rate of sulfamethoxazole was 0 after 28 days in freshwater while the concentrations of antibiotics such as phenylbutazone and acetaminophen decreased considerably (Baena-Nogueras et al. 2017). Sahar et al. (2011) compared the removal rates of MBR and traditional activated sludge treatment for antibiotics, such as erythromycin and sulfamethazine, and found that the removal rate of MBR was high probably due to the removal mechanism of biofilm, but not biodegradability.

Ozonation is in close relationship with biodegradation. On the one hand, ozonation can be used as pretreatment of biodegradation to prevent considerable improvement in antibiotic resistance during biological treatment (Liu et al. 2017a). Akme Mehmet Balcioglu and Otker (2003) applied ozone to veterinary antibiotic wastewater, and the BOD<sub>5</sub>/COD ratio increased from 0.07 to 0.38, improving the biodegradability of the wastewater. On the other hand, ozonation can also be used as posttreatment of biodegradation. Wang et al. (2018a, b) found that combining the activated sludge process with sludge ozonation could simultaneously reduce excess sludge and antibiotics and even achieve a removal rate of 86.4–93.6% of the studied antibiotics. De Wilt et al. improved the traditional activated sludge system by designing a biological-ozone-biological treatment process for the treatment of secondary clarified effluent from the wastewater treatment plant. The removal rate of sulfamethoxazole reached 99%, which was much higher than 50% in activated sludge system alone of the same biomass (de Wilt et al. 2018).

As a low-cost technology, the biological wastewater treatment unit plays an important role in degrading pollutants. However, the microbial metabolism may be restrained because of the exposure to antibiotic wastewater, which influences the sewage treatment capability of the biological treatment unit. Pretreatment of the antibiotic wastewater

using technologies such as ozonation is an effective way to reduce the disposal difficulty of biological unit.

## Electrochemical oxidation

Electrochemical oxidation is a rapidly developing treatment technology in recent years. Despite its late start, electrochemical oxidation shows great potential in the field of refractory wastewater treatment because of its mild reaction condition and strong oxidation (Fabińska et al. 2014; Crini and Lichtfouse 2019). The catalytic activity and stability of the electrode materials, especially the anode electrode, determine the electrochemical oxidation ability and efficiency to a great extent (Feng et al. 2016b). Electrode materials, such as Pt electrodes, dimensionally stable anodes and boron-doped diamond electrodes (BDD), are commonly used in the electrochemical oxidation. Among these electrodes, BDD is a material with the highest oxygen evolution overpotential and can maintain weak oxygen evolution reaction at high current density, thereby achieving high degradation efficiency (Chen 2004).

The improvement of anode materials is currently a devoted topic of study on electrochemical oxidation. Xia and Dai degraded levofloxacin using a novel rare-earth La, Y co-doped PbO<sub>2</sub> electrode, and when the current density is 30 mA/cm<sup>2</sup>, the initial pH is 7 and the initial levofloxacin concentration is 500 mg/L, the levofloxacin removal rates by using PbO<sub>2</sub>, La-PbO<sub>2</sub>, Y-PbO<sub>2</sub> and La-Y-PbO<sub>2</sub> electrodes were 83.3%, 90.9%, 85.6% and 95.4%, respectively. (Xia and Dai 2018). Wang et al. (2018a, b) degraded tetracycline by plasma-sprayed Ti/Ti<sub>4</sub>O<sub>7</sub> electrode, and the tetracycline removal rate reached 95.8%. Filling the particle electrodes (such as  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, activated carbon, ceramic, zeolite and kaolin) between the anode and cathode electrodes will form several tiny electrolytic cells in the electrochemical reactor and improve the antibiotic removal efficiency (He et al. 2014; Li et al. 2015; Zheng et al. 2016). The combination of electrochemical oxidation technology with other technologies is another popular subject. Liu et al. (2015) established a forward osmosis process with electrochemical oxidation function (FOwEO), which could effectively remove trace antibiotics from wastewater (> 98%) and concentrate (99%).

Electrochemical oxidation is a highly efficient technology to remove antibiotics from wastewater with no need for external reagent. However, electrochemical oxidation has not been widely used, mainly because of the high cost of electrode materials. In addition, the mass transfer of the electrochemical oxidation reactor is slow when the conductivity of wastewater is low, resulting in low current efficiency and high energy consumption. Therefore, it is necessary to develop low-cost anode material with high catalytic activity and high stability, and new reactors such as three-dimensional electrode reactor should be investigated

to reduce mass transfer resistance and improve current efficiency for the wide application of electrochemical oxidation technology.

## Research perspectives

### Application of new technologies and development of new materials

Municipal wastewater treatment plant, which is a place for antibiotic wastewater-concentrated disposition, cannot completely remove antibiotics by conventional secondary treatment. In this case, the research emphasis of antibiotic wastewater treatment includes the removal efficiency and, at the same time, the cost.

A simple method to improve the removal efficiency of each technology is to look for optimal experimental conditions such as pH and temperature. Except for this, the combination of different technologies, such as ozonation and adsorption, photocatalysis and ozonation and, especially, the combination of biodegradation and advanced oxidation processes (for example, photocatalytic and ozonation), adsorption or membrane, would be highly effective and attract more and more attention.

Other technologies (e.g., ionizing radiation, constructed wetland) bring new breakthroughs in the treatment of antibiotics in water or wastewater. Meanwhile, the toxicity of the intermediate product or by-product requires more research. Constructed wetland, as a sustainable technology, is an economical and efficient technology on the removal of antibiotics. It was reported that the removal efficiency of the vertical flow constructed wetlands for most antibiotics could be more than 70% (Huang et al. 2017; Liu et al. 2019). The ionizing radiation, including gamma ray and electron beam, is based on the reactive species (e.g.,  $\cdot\text{OH}$ , solvated electrons). It can remove most antibiotics from wastewater with the removal rate up to 90%. Also, the combination of ionizing radiation and other technologies such as  $\text{H}_2\text{O}_2$ ,  $\text{Fe}^{2+}$  and biological treatment processes would have a better performance (Wang et al. 2019).

In addition, new materials should be developed and utilized, for example, using cheap and readily available materials, such as minerals (bentonite, montmorillonite, etc.) and solid wastes (fly ash, excess sludge, etc.), or materials with excellent adsorption capacity, such as carbon nanotubes, instead of conventional adsorbents (such as activated carbon) in adsorption; developing new material modification methods to improve the oxidation property of the anode in the electrochemical oxidation; and investigating new photocatalysts which are easy to prepare and sensible to visible light.

### Risk assessment and control of antibiotics and antibiotic resistance genes

With the improvement in environmental safety requirements, investigating the types and concentrations of antibiotics, especially trace antibiotics, is an issue of crucial importance. Based on the quantitative analysis by high-performance liquid chromatography–mass spectrometry, a simple, highly accurate and low-cost detection method should be developed, which is of considerable importance for the research of distribution, migration, transformation, ecotoxicity and joint toxicity of antibiotics in water or other environments. Applying amplification (such as PCR) combined with sequencing methods (such as high-throughput sequencing) to detect the abundance of antibiotic resistance genes and integrating sampling, processing and identification to form a comprehensive detection system are also supposed to be implemented to obtain systematic distribution data of the antibiotic-resistant genes. The studies on diffusion law and mechanism of resistant genes should be conducted from different levels, such as molecules, individuals and communities (Rodgers et al. 2019).

Based on previous studies, it is important to establish a comprehensive and quantitative model of antibiotic risk assessment, which concerns the human microbiome caused by ingested antibiotic residues and the environmental microbiome (Ben et al. 2019). To do this, the standard detection methods for antibiotics and antibiotic-resistant genes should be developed and monitoring systems for different environmental antibiotics and resistant genes should be established. Besides, the relations between the antibiotic level and antibiotic resistance in the environment should be tracked and the early warning system for resistance genes can be established.

In order to control the antibiotics and antibiotic-resistant genes in the environment, perfect pollutant emission standards containing the antibiotics index should be established. Particularly, as the largest producer of antibiotics, China should formulate reasonable emission standards for antibiotic production enterprises as soon as possible and even develop corresponding standards for surface water and soil environment in point source pollution areas. On the other hand, controlling strategies for antibiotics in wastewater should be established, and treatment processes, such as disposal and transfer, should be strictly managed.

### Conversion from waste to energy

It has been discovered that wastewater containing antibiotics can be converted to hydrogen energy during photocatalysis, providing a new source of energy. Catalysts with excellent photocatalytic performance make it possible for the combination of photocatalytic oxidation and photocatalytic reduction (such as  $\text{H}_2$  evolution) in a photocatalytic process (Nie

et al. 2018; Xu et al. 2017). With utilization of these specific catalysts, antibiotics in water can play a role of sacrificial agents while they are being degraded, so as to promote H<sub>2</sub> revolution. Wei et al. reported that amoxicillin wastewater could be converted to hydrogen energy (about 40 μmol/h) under visible light ( $\lambda > 420$  nm) via the 2% bismuth spheres coupled with g-C<sub>3</sub>N<sub>4</sub> (Wei et al. 2019). Meanwhile, Jiang et al. prepared the 0D Ta<sub>3</sub>N<sub>5</sub> nanoparticles anchored on 3D TiO<sub>2</sub> hollow nanosphere composites which could degrade levofloxacin wastewater as well as realize the solar-to-fuel conversion (Jiang et al. 2018).

In addition, the anaerobic biological process of antibiotic wastewater is expected to enhance the methane production. In the treatment process of tetracycline wastewater, Zhang et al. added granular activated carbon/nano-zerovalent iron mediator into expanded granular sludge blanket (EGSB) reactor, the function of which was to promote the interspecies mass and electron transfer. The result showed that the tetracycline removal rate was 81.5% at the stable stage with the methane production rate reaching up to  $199.4 \pm 21.2$  mL/g COD (Zhang et al. 2018b). Except for hydrogen and methane, there is much more to learn about the conversion from antibiotic wastewater to other forms of energy.

## Conclusion

In this study, 5420 articles on antibiotics in water or wastewater are researched, and the conclusions are as follows:

1. The journal ‘Chemosphere’ published most of the articles related to antibiotic wastewater, followed by ‘Science of the Total Environment.’ Among the countries studied on this subject, the USA started the earliest with the highest h-index, reflecting the highest influence in the world. Although China started lately, the rapid development has been shown in recent years, and the Chinese Academy of Sciences is the institution that has the most contribution to this subject in all institutions, with the number of articles and the h-index ranking first.
2. The five most investigated antibiotics in water or wastewater are sulfonamide, quinolone, tetracycline, β-lactam and macrolide. The five most investigated treatment technologies are adsorption, photolysis/photocatalysis, biodegradation, ozonation and electrochemical oxidation. Activated carbon seems to be the most frequently used adsorbent, and various modification methods are available for increasing its adsorption capacity. Antibiotics such as tetracyclines, ciprofloxacin and amoxicillin are often removed from wastewater by adsorption. Modifying TiO<sub>2</sub> by diverse methods and developing new visible-light-driven catalysts are the popular research

topics on photolysis. The improvement in electrodes, especially anode materials, is currently a devoted topic of electrochemical oxidation.

3. In the future, the combination of different technologies in the treatment of antibiotics in water is predicted to receive increasing attention for improving the removal efficiency of antibiotics. In addition, the development of new materials and other treatment technologies (such as ionizing radiation and constructed wetlands) will continue to be a popular subject. In recent years, an increasing number of researchers have been devoted to the determination of trace antibiotics. Besides, the risk assessment of antibiotics and resistant genes will continue to be a hot topic. The formulation and improvement of antibiotic-related standards are also of great importance to mitigate the pollution problem brought by antibiotics. In addition, converting waste (antibiotics in wastewater) to energy attracts more attention with the reduction in energy.

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