

Remediation of Antibiotics from the Environment

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Abstract—Nowadays there is a vast increase in the production and use of antibiotics by humans. Antibiotics are also added in animal feed, agriculture, horticulture which has led to a vast amount of release of antibiotics in the environment. This has led to a risk to the environment because antibiotics are continuously added to the soil, ground water, surface water and also they exist as residuals in different environmental compartments. Antibiotics produce antibiotic resistant bacteria which have adverse effects on animal, human health and ecosystem. Due to their antimicrobial nature, biodegradation of antibiotics is difficult and they persist in the environment. The common antibiotics present in environment are ciprofloxacin, tetracycline, sulphonamide, erythromycin, clarithromycin, sulfamethazine, augmentin and many others. The main sources of antibiotic contamination in environment is wastewater released from antibiotic producing industries, hospitals, use of antibiotics by humans, antibiotics added in animal feedings. Antibiotics can be removed through traditional methods such as flocculation, sedimentation, filtering and biological processes but these are not effective enough so new techniques were developed such as ozonation, fenton and photo-fenton reactions, chlorination, photolysis, and photo-catalysis which were successful in remediation of antibiotics from wastewater.

Keywords: antibiotics, wastewaters, phytoremediation, tetracycline, ozonation

1. INTRODUCTION

Antibiotics are used for protection of human beings against various microbial diseases, to increase the productivity of livestock. They are also used in agricultural farms for various purposes. Although they are of huge importance to plants, animals and human beings, they can be taken as major contaminants present in environment[1]. Common antibiotic contamination found in environment are ciprofloxacin, streptomycin, tetracycline, sulphonamide, erythromycin, clarithromycin, sulfamethazine, augmentin and many others. Concentration of various antibiotics in different components of environment is highly variable. The increased concentration of antibiotics in environment has led to development of antibiotic resistant microbes[2, 3]. As a result antibiotics are not as effective against diseases as they were earlier. Some of the antibiotics against which microbes have developed resistance are MRSA (methicillin-resistant Staphylococcus aureus), VRE (vancomycin-resistant Enterococcus), ESBL (extended spectrum beta-lactamase), VRSA (vancomycin-resistant S. aureus) and MRAB

(multidrug-resistant A. baumannii)[3]. Microbes which are resistant to multiple antibiotics are called Multiple drug resistant microbes. Being antimicrobial in nature, they cannot be degraded naturally by microbes.

2. SOURCES OF ANTIBIOTICS CONTAMINATION

A recent survey has shown that antibiotic consumption has increased to about 10, 000 tonnes per year in Europe and a major portion of it is passing through human body and animals unchanged[1]. From humans and animals they are passed on to waterbodies, ground water, soil and sediments. Major source of antibiotic contamination is waste released from hospitals, veterinary centres, industries producing antibiotics, sewage waste from households, animal feeds and their excreta, agricultural farms where antibiotics are used to increase plant biomass.

Table 1: Impact of Antibiotics on Humans and Animals

	Humans	Plants
Antibiotics	Beneficial bacteria within the intestine is killed, tendonitis, diarrhoea, impaired kidney function, increase of resistance in human bacterial pathogens	Various antibiotics completely inhibit the growth of symbiotic nitrogen-fixing bacteria, non-symbiotic nitrogen fixing bacteria Sulfur -oxidizing bacteria and many plant pathogenic microorganisms. Root and shoot growth, seed germination were inhibited.

3. METHODS FOR REMEDIATION OF ANTIBIOTICS

Traditional methods

Coagulation and Flocculation. In the coagulation technique, coagulants are added to wastewaters, coagulants are opposite in charge to the suspended solids such as antibiotics. Coagulants are added to neutralise the charges on the organic substances suspended as a result the charge gets neutralised and the suspended particles are able to stick to each other or accumulate together and also a larger particle called microflocs are formed. The next step is flocculation in which the size of suspended particles increases from submicroscopic to visible particles. Collisions occur and larger

particles are formed, when optimum sized flocs are formed water can be further sent for sedimentation process[6].

Sedimentation and filtration. After the flocculation process, the large floc particles and water move through large basins, since the flocs are large in size they settle down at the bottom and water flows out for the further filtration process. In the filtration process, water is allowed to flow above a layer of sand. As the water moves the remaining organic particles i.e. antibiotics are trapped in the sand and water moves out[7].

Biological processes. In this system the activated sludge technology is used. This method involves the organic compound degradation in activated sludge tank using aerobic and anaerobic systems. This method has a drawback of its own since the contaminants can be toxic and can kill the microorganisms used in this process.

New Techniques

Chlorination. It refers to chemical oxidation using chlorinated species (chlorine gas, hypochlorous acid, chlorine dioxide and sodium hypochlorite) in wastewater treatments. This technique oxidizes antibiotics to less toxic and readily biodegradable compounds. The oxidation of three β -lactams (amoxicillin, penicillin G and cefadroxil) with chlorine dioxide have been studied which concluded that cefadroxil and amoxicillin were highly reactive to ClO_2 while penicillin G reacted slowly. A total degradation was attained after 2 hrs for penicillin G and 1 min for the other compounds[8]. Degradation of sulfonamides, carbadox and trimethoprim with Cl_2 was also studied which concluded that in the removal of the studied antibiotics, oxidation with chlorine was effective (>90%)[9]. This technique has proven to be efficient in the degradation of antibiotics which are present in sources with low amount of organic matter, such as drinking water. Also, pH influences the degradation rates. In order to prevent the formation of halogenated species, which are potentially carcinogenic, advanced oxidation processes have replaced this technique.

Ozonation. The ozone is a strong oxidant which has the ability to remove solids from wastewater by oxidation and physical floatation. Ozonation of wastewater results in the development of foam in which significant amount of antibiotics gets trapped[10]. The advantage of this technique is that it can be applied when the compositions of the effluents and flow rates are fluctuating. High cost of equipment and maintenance, extreme pH-dependence and also the energy required to supply the process comprise the disadvantages. Also, in this process, there is a limitation of mass transfer. So, when compared with other oxidative methods, a greater amount of oxidant to treat the same pollutant load is required. For these reasons, this technique does not seem apt for the treatment of water contaminated with antibiotics. Various studies have been done about the ozonation methodology applied to waters contaminated with antibiotics such as lincosamides[11], macrolides[12], quinolones [13],

sulfonamides[11, 14] and tetracyclines [15] and it was observed that for all of them occurrence of degradation was above 76% with a slight increase in the biodegradability of effluents. However, for β -lactam antibiotics, lower degradations were attained.

Fenton and photo-Fenton. This technique uses Fenton's reagent which is a solution of hydrogen peroxide and ferrous ions, which has strong oxidizing properties. The efficiency of oxidation process increases when it conjugates with UV radiation - photo-Fenton[16]. Factors like temperature, pH, catalyst, target-compound and hydrogen peroxide concentrations mainly affect the performance of these processes. This type of process is advantageous as it makes the use of low-cost reagents. Besides, hydrogen peroxide is environmentally safe and easy to handle and iron is abundant and a non toxic element. Several studies have been conducted about this technique applied to different antibiotic classes such as imidazoles[17], quinolones[18], β -lactams[19], lincosamides[20], tetracyclines [20] and sulfonamides[16]. From all these research papers, it can be concluded that the photo-Fenton seems to be more efficient (degradation efficiency above 74%, TOC removal>50%, COD removal>56%) as compared to the Fenton (degradation efficiency above 53%, TOC removal>20%, COD removal>44% and a slight increase in biodegradability). Overall, an improvement can be seen in the Fenton process in the presence of UV light (photo-Fenton).

Photolysis. It is the dissociation or decomposition of chemical compounds by artificial or natural light. It is commonly of two types - indirect and direct photolysis. In the first case, the photo-degradation is caused by photo-sensitizers like hydroxyl or peroxy and oxygen radicals[19]. Direct photolysis involves the absorption of UV light by organic compounds and their reaction with the components of the water matrix or undergoing self-decomposition[21]. Photolysis under solar irradiation is considered one of the promising methods for the degradation of antibiotics in the natural aquatic environment [22]. Photo-degradation of imidazole was studied and it was found out that in the absence of UV/ H_2O_2 , 6-12% removal was attained while in its presence, the removal increased to 58-67%[17]. Several studies were done on the degradation of various antibiotics such as tetracyclines, which is a very light sensitive antibiotic group, quinolones and sulfonamides and it was concluded that tetracyclines obtained high removals (about 80%) while quinolones and sulfonamides achieved very low removals[17, 22]. This method has proved to be less effective when compared with the others described so far. This technology degrades only photo-sensitive compounds and waters with low COD concentrations.

Semiconductor photocatalysis. This technique involves the activation of a semiconductor (mostly TiO_2 due to its low cost, high stability and good performance) by sunlight or artificial. This method requires the presence of three basic components: a source of photon energy, a catalytic photo-sensitive surface

(mainly a semi-conductor which is inorganic, such as TiO₂) and an appropriate oxidizing agent. Several studies have been done about this technique applied to different antibiotic classes such as b-lactam, sulfonamides, tetracycline, lincosamides, quinolones and chloramphenicol and it was concluded that this method was very efficient. The degradation rates for b-lactam and sulfonamides were found to be above 50% [23] and above 80% [24] respectively, and for all others high removals (>98%) can be achieved [25]. This process seems to be effective for the treatment of effluents containing a less amount of organic matter (river, drinking water and groundwater).

Phytoremediation. It is an alternative for advanced oxidation methods. It uses vegetation to sequester, contain, modify, remove or degrade antibiotics. Pistiastratiotes (water lettuce), Myriophyllum aquaticum (parrot feather) and Helianthus annuus (sunflower) have been studied to give significant removal of both tetracycline and oxytetracycline from water. Water lettuce showed highest removal rates, sunflower hairy roots gave intermediate removal rates which depended on growth period whereas parrot feather depicted least removal rates [26].

4. CONCLUSION

The increased persistence of antibiotics in environment has created a selection pressure over bacteria resulting in development of antibiotic resistance which has turned into a global issue [3]. Because of development of multiple drug resistant microbes, a large number of therapies are becoming ineffective. It is really important to develop awareness among people. First of all people should stop taking antibiotics without doctor's recommendation, excessive use of antibiotics to treat the symptoms need to be avoided and apart from these, use of antibiotics against viral infections such as cold and flu should be circumvented.

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