

VOL. 78, 2020



DOI: 10.3303/CET2078045

#### Guest Editors: Jeng Shiun Lim, Nor Alafiza Yunus, Jiří Jaromír Klemeš Copyright © 2020, AIDIC Servizi S.r.l. **ISBN** 978-88-95608-76-1; **ISSN** 2283-9216

# Potential Risk of Antibiotics Pollution in Aquaponic System and Control Approaches

Zhengxuan Dong<sup>a</sup>, Sha Sha<sup>a</sup>, Chunjie Li<sup>a,\*</sup>, Haslenda Hashim<sup>b</sup>, Yueshu Gao<sup>a</sup>, Peiying Ong<sup>c</sup>, Chew Tin Lee<sup>b</sup>, Zhenjia Zhang<sup>a</sup>, Wei-Min Wu<sup>d</sup>

<sup>a</sup>School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China <sup>b</sup>Faculty of Chemical & Energy Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru. Johor. Malaysia <sup>c</sup>Innovation Center in Agritechnology For Advanced Bioprocessing (ICA), Universiti Teknologi Malaysia, Malaysia <sup>d</sup>Department of Civil & Environmental Engineering, Center for Sustainable Development and Global Competitiveness, William & Cloy Codiga Resource Recovery Research Center, Stanford University, Stanford, CA 94305-4020, USA cjli@sjtu.edu.cn

Aquaponic system, which integrates recirculating aquaculture system (RAS) and hydroponics together, maintains high stocking density of fish. In order to eliminate the risk of fish disease, antibiotics has been inevitably used due to human and environmental concern. This paper aims to analyse the potential risk of antibiotics pollution in aquaponic system and propose the control approaches. The removal of residual antibiotics from aqueous phase of aquaponics can be more efficient than the open system. An aquaponic system with comprehensive antibiotics pollution control strategy is proposed by combining the operational units of UV-photolysis and biofilter adsorption/biodegradation, which is expected to work efficiently and cost-effectively. The biofilter is developed by containing immobilized biofilm incorporated with activated carbon. The antibiotics are then adsorbed onto the activated carbon and biodegraded by the microbes in the immobilized media of the biofilter within 12 h. It concludes that aquaponic system with the integrated approaches is promising in effective utilization of antibiotics and removal of the residual antibiotics comparing with the conventional open aquaculture system such as pond, lake or reservoir.

## 1. Introduction

Aquaculture has been growing rapidly around the world for its capacity to provide sufficient quality protein stably and economically. According to the data issued by Food and Agriculture Organization (FAO) of United Nation (UN), global fish production peaked at about 171 Mt in 2016, with aquaculture representing 47 % of the total (FAO, 2018). China is the country producing the largest amount of aquaculture products in the world i.e. 49.2 Mt of aquaculture food fish products was produced in 2016, contributing to 61.5 % of total world output (FAO, 2018).

With high stocking density and monotonous ecological structure, aquaculture is always plagued by diseases caused by pathogens (Kim et al., 2018). The use of antibiotics at therapeutic doses to control/prevent the disease has been widely implemented in aquaculture industry, especially in developing countries where regulation and legislation on the use of antibiotics are not well established. The subtherapeutic doses of antibiotics are also supplemented in animal feed and water to promote growth and improve feed efficiency although this behaviour has been prohibited in many countries and organizations (Liu et al., 2017).

No antibiotics are specifically designed for aquaculture. Authorized products developed for veterinary medicine are used in aquaculture. The applied dosage of antibiotics varies with the patterns of aquaculture system and breeding areas. Developed countries, especially those in European, have more strict standards and systematic monitoring systems compared to the developing countries. Sweden has banned the use of antibiotics as animal feed additives since 1985 (Romero et al., 2012), and the United Kingdom only authorized five antibiotics for use, i.e., amoxicillin, oxolinic acid, oxytetracycline, sarafloxacin, and trimethoprim-sulfadiazine (Liu et al., 2017). In the developed countries, most of the forbidden antibiotics were detected in

Paper Received: 15/04/2019; Revised: 07/09/2019; Accepted: 10/09/2019

meat products, indicating that they are still in use. In the developing countries which provide more than 90% of aquaculture products, the pollution of antibiotics in aquaculture is more serious. It is predicted that the rapid increase in the amount of antibiotics will have a great negative impact on the development of intensive aquaculture in developing countries. In China, 13 antibiotics have been authorized for use in aquaculture, including doxycycline, enrofloxacin, florfenicol, flumequine, neomycin, norfloxacin, oxolinic acid, sulfadiazine, sulfamethoxazole, sulfamonomethoxine, thiamphenicol, and trimethoprim (Liu et al., 2017). Further actions have been taken to limit the use of antibiotics in aquaculture.

Oral administration is the common route to apply antibiotics mixing with feed, especially for intestinal parasites and systemic diseases in aquaculture. Dosages of antibiotics administrated in this method vary greatly. Less than 30 mg antibiotics were used per kg of fish. Bath treatment (immersion in short or long baths) with the dosages of 1-25 mg/L and pond sprinkle with 0.05-0.1 mg/L are two other ways to use antibiotics. Injection is an occasionally used method to use antibiotics in aquaculture (Liu et al., 2017). Among the residue of antibiotics in Chinese aquatic products, 32 compounds were found. Quinolones are the most frequently detected category in aquatic products, followed by sulfonamides and macrolides. Compared to the maximum residue limits (MRL) of antibiotics in aquatic products enacted by the Chinese government, none of these compounds exceed the maximum residue limits (MRLs). However, it should be noted that some residual antibiotics are approaching the MRLs (Liu et al., 2017).

The growing concern on the residual antibiotics in surface water may cause a risk to human health by promoting antibiotic resistant bacteria (ARB) and antibiotic resistance genes (ARG). Long-term exposure experiments showed that antibiotics at low level could lead to resistance in intestinal bacteria, and subsequently ARGs can be conjugated to human pathogens through horizontal gene transfer, resulting in infections for human (Sharma et al.,2016). Control and elimination of residual antibiotics from discharge waster of aquaculture would be one measure to reduce the risk of antibiotics.

In this study, the potential risk of antibiotics pollution in aquaponic system is reviewed and a novel technical approach integrating UV, adsorption and biodegradation is proposed for the removal of residual antibiotics in aquaponics.

# 2. Aquaponics and potential antibiotic pollution

## 2.1 Concept of aquaponics

Aquaponics refers to the system that combines recirculating aquaculture system with hydroponics in a symbiotic environment. In this system, the effluent of fish rearing section provides nutrients in the horticultural part of the system where plants uptake the nutrients provided by the fish waste and clean the recycled water before being returned to the fish rearing section. Aquaponics has been considered as a promising method of industrialized food production in urban, as well as in those areas where land and water resources are scarce (Li et al., 2018).

Water quality is of significance for cultivation of fish. An aquaponic system includes water treatment units for the control of water quality including mechanical and biological filtration units, ultraviolet (UV) disinfection device and plant in hydroponics (Li et al., 2018). Mechanical filtration is used for the removal of solid pollutants (feed residue and fish faeces), which is essential for the long-term operation of the aquaponic system. Biological filtration converts ammonia to nitrate under the synergy of ammonia oxidizing bacteria and nitrate oxidizing bacteria, which makes it a prominent feature in aquaponic system. The heterotrophic microorganisms decompose organic substance generated in the aquaculture tanks into CO<sub>2</sub>, ammonia and phosphorus compounds in biological filtration unit. UV disinfection device is designed for disinfection and pathogen control. Plants in hydroponics play an important role in water quality control by absorbing the nitrogen and phosphorus nutrients (Li et al, 2019).

The major challenge in aquaponics is to use the nutrient input efficiently, to minimise the waste and achieve a zero-discharge recirculating system. The goals are to enhance the efficiency of nutrient recovery from dissolved maters and the sludge produced from aquaculture tank, and to balance the nutrient generation and demand by the plants (Li et al., 2018).

Another challenge in aquaponics is pest and disease management, including plant pests, plant disease, fish parasite and fish disease. Conventional pesticides are prohibited to use because of toxicity risk to the fish and to the desired biofilm (Luprano et al., 2015). Antibiotics is not allowed in aquaponics because it may impact on plant growth and quality and the maintenance of the nitrification biofilm and other nutrient solubilizing microorganisms. UV treatments are used to reduce the level of harmful disease-causing bacteria in both the aquaculture and crops. UV treatment is a valid method to sustain high hygienic standards in the aquaponic system.

However, UV light has poor penetrating power so its efficacy against pathogens is restricted by proximity of generated radiation to the intended treatment as well as product translucency and surface variability,

especially when aquaculture section is in high stocking density. The use of antibiotics might be inevitably employed to control special diseases of the fish, which is the same strategies used in the conventional aquaculture system. Once antibiotics is used, they must be strictly controlled and follow the identical code applied to veterinary medicine. To date, no report has been published on systematic research work on the control of residual antibiotics in aquaponics.

## 2.2. Behavior of antibiotics in aquaponics

When antibiotics is applied, it is vital to achieve optimal manipulation of the system on the basis of ecology and cost-effectiveness. Antibiotics is normally employed with the bait dosage used as feed additives (Figure 1a). Except for metabolization in fish, the residual antibiotics distributes in each unit of aquaponics by the recirculating water flow. Antibiotics may negatively impact on the biological filtration and hydroponic unit (Figure 1b).

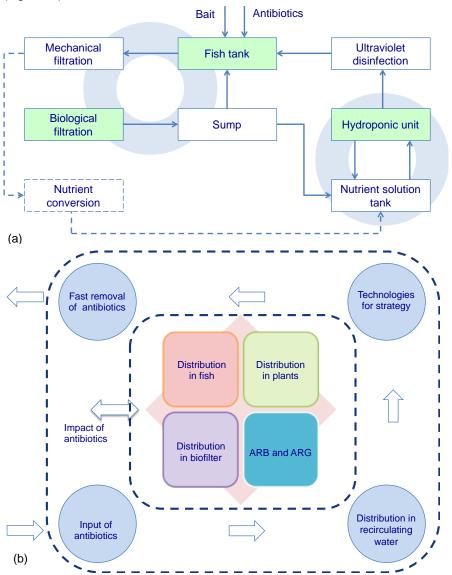


Figure 1: Distribution of antibiotics in aquaponics (a) Process of aquaponics, (b) Distribution of antibiotics. ARB: antibiotic resistant bacteria; ARG: antibiotic resistance genes

It has been confirmed that plants could accumulate antibiotics in hydroponics. Although bioconcentration factor is not high enough compared to other organic pollutants such as DDT. Plants could not cause acute toxicity to human, it can still induce the production of ARG in the aqueous environment. The veterinary antibiotics may have an adverse effect on the roof of the plant, as well as the activity of antioxidant enzymes.

The residue antibiotic could also have negative influence on biofilter, especially for some antibiotics belonging to broad-spectrum bacteriostatic agents. The recirculating aquaculture system could be a reservoir for ARG (Li et al., 2017), and nitrate oxidizing bacteria was fragile when facing certain antibiotics (Schmidt et al., 2012). Certain antibiotics induce immunomodulation (Romero et al., 2012), but it depends on many factors and rarely happen in practice. Therefore, low concentrations of antibiotics will not induce fish stress response under normal circumstances.

# 3. Strategies for residual antibiotics control in aquaponics

## 3.1 Proper way for antibiotics employment in aquaponics

If antibiotics have to be employed in aquaponics for controlling special fish diseases, proper procedures should be established to avoid the distribution of residual antibiotics in the recirculating water flow, especially to prevent residual antibiotics from contacting plants in the hydroponic unit.

Different antibiotics have their own reaction mechanism. Only when a certain concentration and duration time are reached, a good bactericidal effect can be obtained. For instance, oxytetracycline, which is poorly absorbed by fish, requires a high dosage rate of 100-150 mg/d/kg fish for 10-15 d, while florfenicol requires only 10-15 mg/d/kg fish for 3-5 d (Liu et al., 2017).

Antibiotics mixed with feed (oral administration) are added in the fish tank daily and continuously for several days. After each dosage, the water recirculating has to be suspended for several hours. During this period, the feed with antibiotics can be consumed and metabolized by fish and the residual antibiotics are released into the water. The residual antibiotics should be contained in the fish tank, to avoid entering into other units of aquaponics. The next step is to remove the residual antibiotics as soon as possible (within several hours) using integrated technologies.

## 3.2 Technologies for the removal of residual antibiotics in aquaponics

A suitable methodology is required to eliminate antibiotic in recycled water in order to achieve successful antibiotics control in aquaponics (Fig.1b). Physical, chemical and biological technologies can achieve the removal of the residual antibiotics. Physico-chemical technologies include coagulation, adsorption, membrane filtration, chlorination, UV irradiation, Fenton reaction, ozonation, nanotechnology and photocatalytic oxidation (Sharma et al.,2016). Biological technologies involve various anaerobic and aerobic bioreactors, including aerobic membrane bioreactor (MBR), sequencing batch biofilter granular reactor (SBBGR), and constructed wetlands etc.

The antibiotic removal should employ cost-effective solutions. Although advanced oxidation can convert antibiotic molecules into simple compounds or even mineralize them completely, this type of processes is expensive and difficult to maintain for the removal of all compounds including antibiotics at industrial scale.

Adsorption is an effective method for antibiotic removal from contaminated water. It is relatively inexpensive and unaffected by the potential toxicity as for biologically based processes (Ahmed et al., 2015). The efficiency of adsorption processes is highly affected by the type of adsorbent, adsorbate properties, and the compositions of waste stream. The most widely used adsorbents with effective removal are activated carbon (AC), carbon nanotube (CNT) and biochar (BC) (Yu et al., 2016).

The disinfection processes could result in the inactivation of ARB and to decrease the residual antibiotics or abundance of ARGs. Among three disinfection processes i.e. UV, chlorination and ozonation, Ultraviolet light (UV-C) treatment in recirculating aquaculture has been suggested to reduce pathogens in the water column, without adding any chemicals into the water, thus maintaining fish health and reducing the volume of water exchange (Zheng et al., 2017). UV treatments can decompose enrofloxacin with above 60 % of removal efficiency. The abundance of ARGs decreased as the disinfectant dosages of UV increased (Zheng et al., 2017).

Most antibiotics are refractory for microorganisms. Alexandrino et al. (2017) investigated the biodegradation of the veterinary antibiotics enrofloxacin (ENR) and ceftiofur (CEF) by microbial communities derived from the experimental constructed wetlands. Complete removal of CEF, and 40-55 % removal of ENR was obtained. Overall, this work demonstrated that microorganisms are capable of adapting and responding to the presence of antibiotics. Santos et al. (2019) found that microbes derived from the rhizosphere of the estuarine plants were capable of biodegrading the veterinary antibiotic ENR. Malek et al. (2019) reported that microbial cultures derived from the two estuarine sediments were able to remove up to 98 % of ENR and over 95 % of oxytetracycline (OXY). The presence of ENR and OXY in the culture media resulted in a lower microbial diversity and richness and the predominance of bacterial species.

A combined treatment system attracts more attention to improve the removal of recalcitrant or poorly degraded antibiotics and other trace organic contaminants (Yu et al., 2016). Physico-chemical reactions

responsible for the removal of antibiotics flowing in a biological treatment unit may be a highly integrated treatment system for antibiotics removal in aquaponics.

#### 3.3 Novel technical approach for residual antibiotics removal

As a recirculating system, aquaponic system accumulates a relatively lower level of antibiotics after fish uptakes the majority of feed mixed with antibiotics. Based on the description above, a novel technical approach integrating UV, adsorption and biodegradation is proposed for the removal of residual antibiotics in aquaponics (Figure 2).

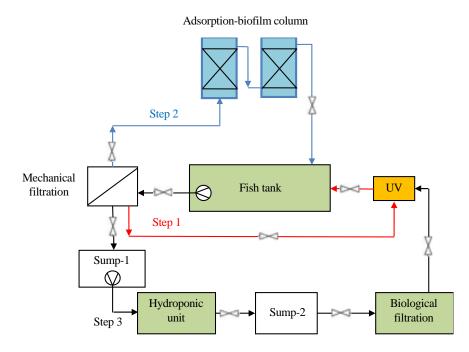


Figure 2: Schematic diagram of technical integration for antibiotic removal in aquaponics

UV unit is used to pre-treatment the residual antibiotics in water. Antibiotics can be converted into substances that are easily biodegraded. The treated water passes through an adsorption-biofilm column which combines adsorption and biodegradation functions. The columns are filled with immobilized biofilm medium particles which contain adsorbents in package or fluidization. Activated carbon (AC), biochar (BC) or carbon nanotube (CNT) can be used as adsorbents to remove the residual antibiotics in water. Microbes in the immobilized biofilm degrade the residual antibiotics and the degraded compounds adsorbed by the adsorbents.

Figure 2 shows the operation of the proposed antibiotics removal in the aquaponic system in three steps. First, once the antibiotics are supplemented in fish tank, water recirculation of the aquaponic system is suspended. After several hours such as 3 h, depending on the antibiotics dosage and consumption efficiency by the fish, step-1 is operated for about 6 h, and the residual antibiotics in the water are decomposed via the UV unit. Step-2 is operated for another 12 h to recirculate water from fish tank via the adsorption/biodegradation columns to completely remove antibiotics by adsorption/biodegradation functions. Step-3 will restart the recirculation water in the aquaponic system. The proposed approaches are expected to provide highly efficient and cost-effective control of residual antibiotics in the aquaponic system. The relevant experiment based on enrofloxacin (ENR) is ongoing.

## 4. Conclusions

Aquaponics, as an intensive food production method, has a high stocking load of fish reared in a compact space, which leads to an increased risk of disease. The use of antibiotics might be inevitable in many cases in order to control or cure fish diseases, resulting in the risk of antibiotics pollution to plants in the hydroponic unit. However, due to the low dosage of antibiotics applied and the nature of a closed system, the control of residual antibiotics in aquaponics could be relatively easy, as compared to the conventional aquaculture in an open system. A novel technical approach integrating UV, adsorption and biodegradation is proposed for the removal of residual antibiotics in the aquaponics. UV unit is used for the pre-treatment of residual antibiotics in

water, to convert the refractory of antibiotics into degraded products that are readily biodegraded. Adsorbent and immobilized biofilm are incorporated in the medium particles and packaged or fluidized in the column reactors to remove residual antibiotics rapidly and completely. The proposed approaches are expected to provide effective control against antibiotics pollution in aquaponic system.

#### Acknowledgments

This research is financially supported by Science and Technology Commission of Shanghai Municipality (No. 18230743000, 16ZR1417400) and National Natural Science Foundation Council of China (No. 21808141).

#### Reference

- Ahmed M.B., Zhou J.L., Ngo H.H., Guo W., 2015, Adsorptive removal of antibiotics from water and wastewater: progress and challenges. Science of The Total Environment, 532, 112-126.
- Alexandrino D.A.M., Mucha A.P., Almeida C.M.R., Gao W., Jia Z., Carvalho, M.F., 2017, Biodegradation of the veterinary antibiotics enrofloxacin and ceftiofur and associated microbial community dynamics, Science of The Total Environment, 581, 359-368.
- FAO (Food and Agriculture Organization of the United Nations). 2018. The State of World Fisheries and Aquaculture 2018 Meeting the sustainable development goals. Rome, Italy.
- Kim Y.B., Jeon J.H., Choi S., Shin J., Lee Y., Kim Y. M., 2018, Use of a filtering process to remove solid waste and antibiotic resistance genes from effluent of a flow-through fish farm, Science of The Total Environment, 615, 289-296.
- Li C., Lee C.T., Gao Y., Hashim H., Zhang X., Wu W.M., Zhang Z., 2018, Prospect of aquaponics for the sustainable development of food production in urban, Chemical Engineering Transactions, 63, 475-480.
- Li C., Zhang B., Luo P., Shi H., Li L., Gao Y., Lee C.T., Zhang Z., Wu W.M., 2019, Performance of a pilotscale aquaponics system using hydroponics and immobilized biofilm treatment for water quality control, Journal of Cleaner Production, 208, 274-284.
- Li S., Zhang S., Ye, C., Lin W., Zhang M., Chen L., Li J., Yu, X., 2017, Biofilm processes in treating mariculture wastewater may be a reservoir of antibiotic resistance genes, Marine pollution bulletin, 118(1-2), 289-296.
- Liu X, Steele J C, Meng X Z., 2017, Usage, residue, and human health risk of antibiotics in Chinese aquaculture: A review[J], Environmental Pollution, 223,161-169.
- Luprano M. L., De Sanctis M., Del Moro G., Di Iaconi C., Lopez A., Levantesi C., 2016, Antibiotic resistance genes fate and removal by a technological treatment solution for water reuse in agriculture, Science of the Total Environment, 571, 809-818.
- Malek H., Alexandrino D.A.M., Fatma A., Boubaker E., Bei L., Zhongjun J., 2019, Biodegradation of oxytetracycline and enrofloxacin by autochthonous microbial communities from estuarine sediments, Science of The Total Environment, 648, 962-972.
- Romero J., Feijoó C.G., Navarrete P., 2012, Antibiotics in aquaculture–use, abuse and alternatives, Health and Environment in Aquaculture, 159.
- Sharma V.K., Johnson N., Cizmas L., Mcdonald T.J., Kim H., 2016, A review of the influence of treatment strategies on antibiotic resistant bacteria and antibiotic resistance genes, Chemosphere 150, 702-714.
- Santos F., Mucha A.P., Alexandrino D.A.M., Almeida C.M.R., Carvalho M.F., 2019, Biodegradation of enrofloxacin by microbial consortia obtained from rhizosediments of two estuarine plants, Journal of Environmental Management, 231, 1145-1153.
- Schmidt S., Winter J., Gallert, C., 2012, Long-term effects of antibiotics on the elimination of chemical oxygen demand, nitrification, and viable bacteria in laboratory-scale wastewater treatment plants, Archives of Environmental Contamination and Toxicology, 63(3), 354-364.
- Yu F., Li Y., Han S., Ma J., 2016, Adsorptive removal of antibiotics from aqueous solution using carbon materials, Chemosphere, 153, 365-385.
- Zheng J., Su C., Zhou J., Xu L., Qian Y., Chen H., 2017, Effects and mechanisms of ultraviolet, chlorination, and ozone disinfection on antibiotic resistance genes in secondary effluents of municipal wastewater treatment plants, Chemical Engineering Journal, 317, 309-316.