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Review

Pollution Control of Industrial Mariculture Wastewater: A Mini-Review

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Abstract: With the rapid development of intensive mariculture, lots of mariculture wastewaters containing residual feed and excrements are discharged into marinelands, leading to coastal pollution. Recently, the environmental problems caused by the discharge of mariculture wastewater have been paid much attention, as have other breeding industries in China. In fact, organic solid waste accounts for most of the pollutants and can be reduced by precipitation or filtration technologies, after which the supernatant can be easily treated by ecological methods. Some national guidelines and relevant local standards have been issued to strictly control the mariculture wastewater, but there are still few effective technologies for mariculture wastewater treatment due to its high salinity and extremely low pollutant concentration. This paper aims to propose feasible pollution control methods of mariculture wastewater according to the wastewater characteristics from different mariculture modes. For raw ammonia-based wastewater, it should be sequentially treated by precipitation, nitrification and denitrification and ecological methods, which would target solid waste, organic carbon/nitrogen and phosphorus removal, respectively. For the nitrate-based wastewater, this just needs denitrification filters and ecological methods for nitrate and phosphorus removal. After an overview of pollution control strategies for different types and scales of industrial mariculture wastewater treatment, some challenges are also mentioned.

Keywords: marine pollution; algae; denitrification filter; constructed wetlands

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1. Introduction

With the rapid development of the mariculture industry in recent years, lots of mariculture wastewater has been directly discharged into coastal waters, leading to eutrophication of receiving water bodies and a decline in the output and quality of mariculture products. Since 2002, China has been the world's largest trading country in aquatic products, and its export volume of aquatic products has consistently been first in the world [1]. According to the Fishery Statistical Yearbook 2021 [2], the mariculture output of China has increased year by year from 2016 to 2020 (Figure 1). In 2020, the total output of marine products reached 52.24 million tons, among which the mariculture yield was 21.35 million tons, accounting for 40.87% of the total marine output.

The mariculture area has always been a small-scale ecological environment system, i.e., it is mainly concentrated in the enclosed, calm and narrow bays where few seawaters exchange with open seas [3]. Although the pollutant concentration of mariculture wastewater itself is lower than that of municipal and industrial wastewaters, the high flow of untreated wastewater can easily exceed the environmental load of the enclosed waters [4]. In particular, mariculture solid wastes, containing lots of metabolic by-products, residual feed, excrement and others, cause continuous release of inorganic nitrogen and phosphorus from the sediment [5,6]. The closed polluted waters and bottom sediments are difficult to exchange with distant seawater, leading to the deterioration of nearshore waters.

With the rapid development and expansion of mariculture, the continuous discharge of mariculture wastewater has had a great impact on the surrounding environment, including nutrient enrichment, drugs pollution, and the spread of farmed fish genes, parasites, and diseases [7]. Therefore, it is now necessary to focus on this problem and quickly restore the nearshore environment.

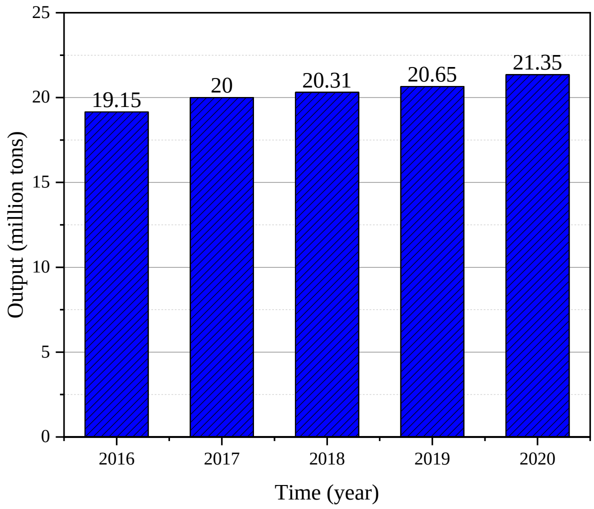

Figure 1. Annual mariculture output of China from 2016 to 2020.

Organic carbon, nitrogen and phosphorus in the mariculture wastewater can be easily treated by the traditional biological methods. Microbial biofilm process with high biomass have seemed to be the best choice due to inhibition of high salinity and diluted nutrients [8–10], otherwise high-cost membranes are required to ensure high biomass [11]. Commonly, nitrogen is oxidized to nitrate by nitrification pathway and then to nitrogen gas by denitrification pathway [12,13], or by direct absorption by plants or algae [14,15]. Phosphorus can also be absorbed by microalgae, depending on their photosynthesis to absorb nutrients for the synthesis of organic matter. Organic carbon and phosphorus can also be removed from municipal wastewater by chemically enhanced primary treatment [16], but this will not work well in the mariculture wastewater due to strong ion concentrations. In particular, phosphorus in the mariculture wastewater must be captured by algae. As a lower cost, environment friendly and energy-saving technology, constructed wetland based on microorganisms, plants, or algae should be the best choice for the mariculture wastewater treatment [17,18], but this is limited by seasonal temperature changes and large footprint space. On the other hand, the function of plants or algae is really irreplaceable for the deep purification of wastewater. Therefore, this review proposes several feasible treatment methods according to the pollution status and wastewater characteristics of industrial mariculture in China. Comparative assessment of mariculture wastewater methods will be also reviewed.

2. Current Situation of Mariculture Wastewater Pollution in China

As industrial mariculture has always been a high-density culture, the nutrients in the natural seawater are far from meeting the growth needs of aquatic animals. Therefore, it is necessary to add feed to ensure the nutritional requirements of farmed animals. However, some of the feed cannot be consumed and will be discharged into the sea after gravity settling [19,20]. In the process of manually changing water and cleaning fishponds, organic particles from the wall and bottom of fishponds are directly discharged into the sea. Residual feed, excrements and urea can be broken down by microorganisms, releasing soluble nitrogen and phosphorus, which led to eutrophication of the coastal water [21–23]. In addition, the use of drugs such as levofloxacin, sulfamethoxazole and azithromycin, as well as pesticides such as rotenone, diuron and vitamins, also raises environmental concerns, as serious diseases associated with industrial mariculture farming require regular disinfection and sterilization [21,24,25].

The eutrophication is still a current focus of concern, since it causes a serious degradation of the marine ecosystem. In 2020, a total of 31 red tides caused by the sudden proliferation of algae were found in China's seas, covering a total area of 1748 km^2, among which two toxic red tides were found in the coastal waters of Tianjin and Shenzhen Bay, with a total area of 81 km^2 [26]. Compared with the past decade, the number of red tides in 2020 has decreased significantly. Besides, compared with the average of the past five years, the maximum coverage area of green tide decreased by 54.9%, and the maximum daily biomass decreased from 1.508 million tons to 0.68 million tons in 2020 [27]. Therefore, China has made remarkable achievements in the prevention and control of eutrophication, but the problem of coastal water pollution still remains serious in China.

Nitrogen and phosphorus are the main drivers of eutrophication in the marine. In recent years, with the improvement of China's wastewater discharge standards, the input of exogenous nitrogen into coastal water has gradually decreased, while total phosphorus has become the main pollutant exceeding the standard. [27], something which creates long-term eutrophic conditions because phosphorus is trapped in the water for a long time [28]. Since 2010, the annual average area of red tide has reached 2200 km^2 in China, and most estuaries and gulf ecosystems are in an unhealthy state, losing ecosystem functions [21]. By establishing the regression relationship between the total aquaculture output and the total pollution load, we can show that there is a significant positive correlation between the total pollutant discharge and the total output value of mariculture [29].

The total amount of pollutants produced by mariculture has been estimated based on the production–discharge coefficient manual of aquaculture pollution sources in the first national survey of pollution sources of China and the Chinese Fishery Statistical Yearbook in Liaoning province in 2021 [2]. Results from Table 1 indicate that a large number of pollutants were produced each year from the production of fish products. Commonly, low-strength mariculture wastewater is directly discharged into the offshore waters, so there should be a significant correlation between fishery production and pollutant discharge amounts. With the development of intensive mariculture and the expansion of mariculture scale, the effect of marine pollution caused by mariculture itself is becoming more and more obvious. As such, Chinese governments at all levels have recently upgraded the discharge standards of mariculture wastewater in accordance with local conditions to control mariculture wastewater pollution.

Table 1. Mariculture waste discharge of Liaoning province in 2020.

Mariculture Species	Output/t	Discharge Capacity/kg		
		Total Nitrogen	Total Phosphorus	COD (Chemical Oxygen Demand)
Fish	53,776	110,725	70,662	4,379,894
Shrimp	14,093	43,590	20,674	1,543,113
Abalone	2600	52,520	1063	117,263
Sea cucumber	56,380	378,761	205,392	1,725,341

3. Treatment Technologies of Industrial Mariculture Wastewater

Compared with municipal wastewater, mariculture wastewater contains high dissolved oxygen (DO) and salinity, which increases its treatment difficulty by biological methods. Mariculture wastewater is divided into ammonia-based wastewater from traditional culture system and nitrate-based wastewater from recirculating aquaculture system (RAS) [30], so the treatment methods are different. The ammonia-based wastewater is the raw mariculture wastewater after a simple solid separation, and mainly contains ammonia, organic nitrogen, active phosphorus, organophosphorus, organic carbon and some urea released by animals. This kind of wastewater should be treated by nitrification and denitrification processes similar to municipal wastewater treatment [31]. The nitrate-based wastewater contains high nitrate from the RAS wastewater treatment unit, where most of the nitrogen is oxidized to nitrate, and organic carbon is oxidized to CO_2 in the biological aerated filter [30]. RAS facility enables 90%–99% of the water to be recycled, during which all the pollutants are treated, leaving only the nitrate cycle to accumulate. In the effluent of RAS, the accumulated nitrate needs to be removed by a deep denitrification with external organic carbon similar to the advanced treatment of municipal wastewater [32].

At present, the main targets of mariculture wastewater treatment are organic carbon, nitrogen and phosphorus, while the other pollutants, such as chemicals, need to be controlled from the feed sources. That is to say, the main target of mariculture wastewater is to remove most of the organic solid wastes containing the above pollutants. The conventional physical, chemical, biological and ecological technologies are feasible for the treatment of all the biodegradable pollutants. Organic solid wastes can be effectively separated by precipitation or filtration, while the residual organic carbon and nutrients need to be removed by functional microorganisms and/or algae [33–36]. Mariculture wastewater treatment is still in its infancy [37,38], but the application of related technologies has been relatively applied for the treatment of the other wastewater. Herein, according to different characteristics of mariculture wastewater from different culture modes, two wastewater treatment methods are summarized and proposed respectively for the treatment of ammonia-based and nitrate-based wastewater. What needs to be pointed out is that plants or algae are needed to further remove nutrients from the biologically treated wastewater due to the strict discharge standard of mariculture [39].

3.1. Treatment of Raw Mariculture Wastewater

Traditional mariculture is culture fish in open ponds and raceways and needs frequent water exchange to maintain a good water environment. The schematic diagram of fresh mariculture wastewater is shown in Figure 2. The main pollutants of this method are fresh feed and excrements, which can be separated by physical filtration methods. Organic waste can be treated for methane production by co-digestion with waste plants or algae. The supernatant wastewater containing few pollutants can be treated in an ecological pond, and then stored in an ecological reservoir for recycling or discharged after in-depth removal of suspended sludge and phosphorus by an artificial wetland.

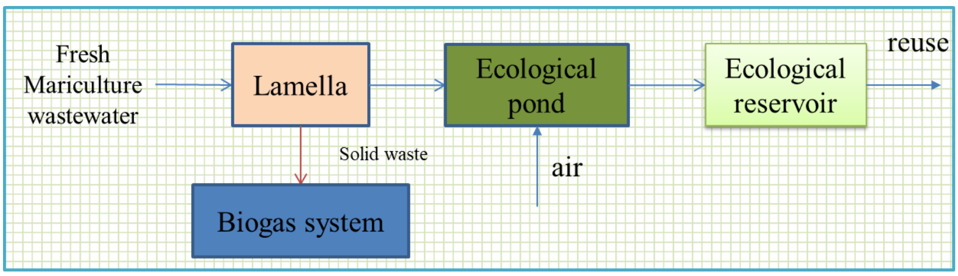

Figure 2. Biological nutrients removal of raw mariculture wastewater.

Inclined tube settling tank (Lamella) is suggested for the removal of the fresh organic solids at a long hydraulic retention time (HRT). After concentration, the waste sludge enters into a sludge pressure filter or centrifugal dehydrator, and the mud cake is transported for centralized disposal and treatment. Anaerobic co-digestion of organic residual feed, excrements, waste sludge and algae can be used to produce methane for energy recovery [40]. The anaerobic digestion liquid is rich in nitrogen and phosphorus, which can be used for the cultivation of algae or irrigation of coastal constructed wetlands.

The ecological system is the key for diluted nutrients, especially phosphorus removal. According to the concentrations of pollutants, a small number of fixed-bed fiber carriers can be installed below the ecological floating bed, enhancing removal of organic carbon and nitrogen. Ammonia is oxidized to nitrite by ammonium-oxidizing bacteria (AOB), and then further to nitrate by nitrite-oxidizing bacteria (NOB), while the nitrate will be denitrified to N_2 by denitrifying bacteria in the inner biofilm [41]. The plants or algae further assist in the removal of phosphorus and nitrogen. The biological fiber carriers are fixed in the water with the planted float system as the shelter, which not only achieves simultaneous carbon, nitrogen and phosphorus removal, but also increases the beauty of the water treatment system. Aeration device is also needed to increase dissolved oxygen for aerobic organisms, though the DO is high in the raw wastewater. Moreover, aquatic plants should be regularly harvested to transfer nutrients. What needs to be pointed out is that the ecosystem relies on the physical interception of residual feed and excrements. Only through a physical separation process to maximize the retention of organic solid waste, the treatment load of the ecological unit can be reduced to ensure a stable operation of the ecosystem.

Microalgae-attached biocarriers can be installed in the shallow water to enhance nutrients removal and algae collection, or some shellfish can also be raised underwater to assist in filtering particulate organic matters or algae [42]. Besides, the other ecological technologies, such as constructed wetland, ecological floating island and artificial submerged bed can also be used for advanced water purification. Finally, the treated wastewater can be stored for recycling in the landscape pond.

For large-scale farmers or industrial mariculture garden areas, the mariculture wastewater can be treated in a centralized way. Wastewater treatment stations are established with households as units, and the specific technological process is shown in Figure 3. After solid–liquid separation by a Lamella sedimentation tank, the concentrated feed and excrement are transferred to an anaerobic digestion system. Bacterial nitrification and denitrification process is used for the removal of nitrogen in the supernatant. The residual nitrate and phosphorus should be further removed in a shellfish and algae pond or ditches/river channels, while plant/algae floating beds are required for phosphorus removal. When the mariculture wastewater meets the discharge standard, it can be optionally discharged or reused.

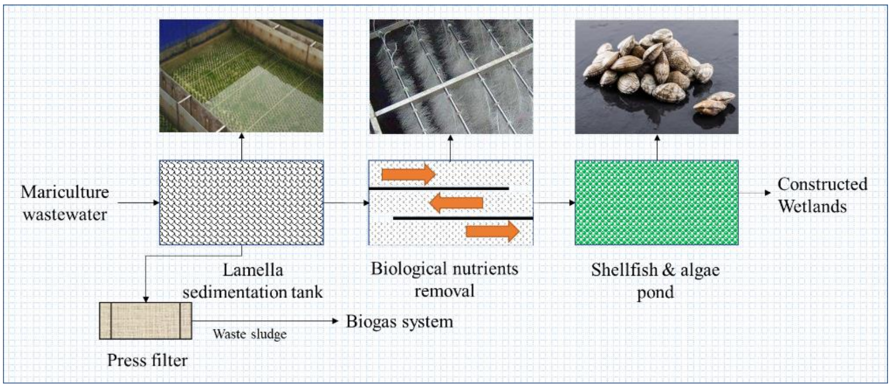

Figure 3. Wastewater treatment technology of large-scale mariculture plants.

For small-scale farmers in mariculture zones, local governments can control the amount of water used by fisheries and centralize wastewater treatment (Figure 4). The mariculture wastewater of each farmer is first treated in its own Lamella sedimentation tank. The residual feed and excrement are transported for centralized disposal, and the supernatant is discharged into the public drainage ditch or pipe network. Only the concentrations of suspended sludge are needed to be controlled at each wastewater outlet. The ecological channel can be filled with biological fillers or large sand gravels, and the bottom is seeded with filter-feeding shellfish. Constructed wetland technology can be selectively used to remove nitrogen and phosphorus. What needs to be emphasized is that the public wastewater treatment facilities should be led by the local government, operated by professional companies, while farmers need to pay a small fee of wastewater treatment.

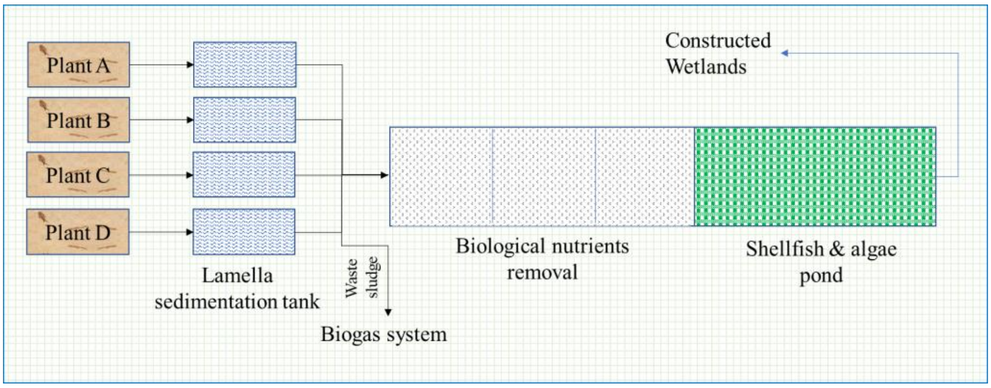

Figure 4. Wastewater treatment technology of small-scale mariculture plants.

3.2. Treatment of RAS Mariculture Wastewater

Compared with the traditional process, the daily exchanging water of RAS is only 5%–10%. RAS wastewater mainly contains high-level nitrate and active phosphorus. Nitrate can be removed using traditional denitrification process with addition of external organic carbon source, such as glucose, acetate or others [43,44]. However, phosphorus cannot be removed under the anoxic conditions, and needs to be further removed by chemically enhanced phosphorus removal or photosynthesis [33]. For the denitrification, it is necessary to overcome the high salt and high DO, which highly inhibit the anaerobic denitrifying bacteria [45,46]. In fact, a large amount of external carbons are oxidized to carbon dioxide using the DO, rather than nitrate as electron acceptor [47]. Besides, a further nitrogen and phosphorus removal is required using ecological methods.

3.2.1. Combined Denitrification Filter and Ecological Method

This specific treatment process is shown in Figure 5. Denitrification filter is usually used to treat the effluent from a secondary sedimentation tank of municipal wastewater treatment plants [48]. External carbon addition should meet the requirement of COD/N above 3.5 due to aerobic oxidation of organic carbon at high DO. Denitrification filter is divided into upflow filter and downflow filter according to hydraulic flow pattern. The upflow filter is divided into a water distribution area, supporting layer, packing layer and clean water area from bottom to top, respectively, while the downflow filter is divided into water distribution area, packing layer, supporting layer and water outlet collection area from top to bottom, respectively. Denitrification filters have good filtration function, and the effluent suspended solids can meet the discharge standard. Normally, traditional denitrification filters need no aeration equipment in the filter, but there is a high concentration of sulfate in seawater. In the presence of organic carbons, sulfate can be reduced to an H_2S gas that has strong toxicity to marine organisms [49]. Denitrification can inhibit the sulfate reduction process, but it is greatly affected by C/N/S ratio [50]. As such, intermittent aeration should be applied to inhibit oxygen sensitive sulfate reducing bacteria in the denitrification filters treating RAS mariculture wastewater, and otherwise the treated wastewater should be treated for the sulfide oxidation to sulfate in a large-volume trickling filter. Finally, the treated wastewater can be stored in an eco-purification pond. Another problem is that phosphorus will accumulate in ecological ponds as the N/P ratio is not suitable for photosynthesis and microalgae are difficult to be separated from the water. In fact, microalgae and the organic solids from Lamella and trickling filter are good biogas production sources.

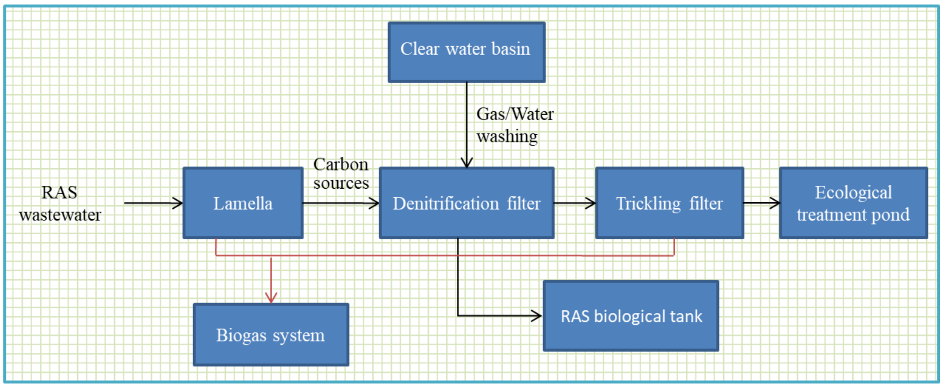

Figure 5. Schematic diagram of a combined denitrification filter and ecological pond.

3.2.2. Combined Fixed-Bed Denitrification and Ecological Method

This is also a nitrogen denitrified process with external carbon source (Figure 6). Suspended solids need to be removed in a Lamella sedimentation tank. The plug-flow fixed-bed denitrification process is simple to operate compared with a denitrification filter. However, the DO concentrations in the effluent of denitrification tank are very low, so air should be supplied into the ecological pond.

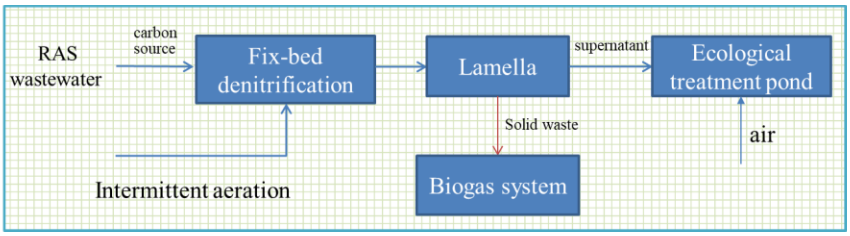

Figure 6. Combined fixed-bed denitrification and ecological treatment system.

4. The Main Challenge of Mariculture Wastewater Treatment

There are several challenges associated with mariculture wastewater treatment due to its high salinity, low temperature and low-concentration pollutants. High salinity and low temperature have a great inhibitory effect on microorganisms, so a long HRT is needed. With the increase of salinity, both microbial abundance and activity gradually decrease in the biofilm, leading to a decrease of nitrogen removal efficiency [51,52]. High salinity also inhibits anaerobic digestion of solid wastes such as residual feed and excrements [53,54]. As such, a larger biological system is needed to compensate for the slow reaction rates, while a larger land use will limit the construction of the wastewater treatment facilities.

Conventional algae or plant purification technology is difficult to work at low temperature as photosynthesis is suppressed or even stopped in winter [55–57]. In general, when the temperature is higher than 10 °C, higher plants can be considered for phosphorus removal. When the temperature is below 10 °C, microalgae can be considered for phosphorus removal. The photosynthetic efficiency of microalgae is 10 times higher than that of terrestrial plants [58]. In view of the low winter temperature, biological and ecological treatment units are not operating, so the discharge standard should also be relaxed.

Slow growth rate of microorganisms is caused by the diluted mariculture wastewater, so some biological treatment processes based on suspended sludge will be of no consideration. Commonly, the attached-growth processes, such as moving bed biofilm reactor (MBBR) and biological filters, are used for the treatment of extremely low-strength wastewater, which can guarantee a high concentration of activated sludge [38,47,59,60]. membrane biological reactor (MBR) is also feasible for the treatment of mariculture wastewater, but the cost is too high.

Low COD/N ratios of mariculture wastewater will affect nitrogen and phosphorus removal efficiency, so external organic carbon needs to be added. With the improvement of sewage discharge standards, the lack of carbon source has become the main bottleneck of mariculture wastewater treatment, similar to a deep nitrogen removal of the secondary effluent of municipal wastewater treatment [48]. What should be pointed out is that excessive carbon sources can easily lead to anaerobic reduction of sulfate, releasing toxic H_2S [61,62].

5. Conclusions

The main characteristics of industrial mariculture wastewater are high salinity, high dissolved oxygen, high solids and low pollutant concentration, which prevents conventional biological treatments from being used. Commonly, physical filtration is the most ideal method for the pre-treatment of fresh mariculture wastewater, and then ecological methods can be used for the deep removal of nutrients. However, in fact, mariculture does not exchange water in time, resulting in part of organic solids dissolved into the water, which should be treated by combined microbial and ecological methods. Although there are many challenges in treating this kind of wastewater, it is really not difficult to treat the pollutants mainly from animals.

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