Removal of ammonia in raw water taken from Surabaya river using biofiltration process

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Abstract. Research on the removal of ammonia in raw water taken from the Surabaya river has been carried out. The study was conducted by operating a biofilter pilot plant consisting of lamella type-sedimentation tanks and biofilter reactors filled with honeycomb type-plastic media. The results of the study showed that the biofilter process which consists of sedimentation tanks and biofilter reactors can reduce turbidity, ammonia, and nitrite inside raw water. Generally, the longer the retention time in the biofilter pilot plant is, the greater the efficiency is in eliminating turbidity, ammonia, and nitrite in the raw water. With the total retention time of 81 minutes in the biofilter test model, the efficiency of turbidity, ammonia, nitrate, and nitrite was 33.83%, 57.84%, 16.40%, and 59.41%, respectively. Based on the results of the research above, it was found that with the ammonia load of 1 until 5 gr/m². Hours (24 until 120 gr/m².day), it efficiently reduced the ammonia compounds by 30%-50%. In the chlorination process, chlorine compounds can react with ammonia compounds in the raw water, so by decreasing the concentration of ammonia, the consumption of chlorine will be reduced, resulting in lower production costs. With the reduced use of chlorine compounds, the potential formation of micropollutant compounds due to the chlorination process will also be reduced.

1. Introduction
Surabaya river is a source of raw water for the communities in Surabaya and surrounding areas, including industrial, PDAM of Surabaya City. Along with the rapid growth of the residential area and the development of the industrial sector, it results in a higher level of pollution, and as a consequence the quality of the water does not meet the quality standards required as the raw water for drinking water. The most potential parameter which causes pollution is ammonia pollutants. The concentration of ammonia pollutants in the Surabaya river has exceeded class I water quality standards based on Government Regulation (PP) No. 82 of 2001, namely for the raw water for drinking [1].

Ammonia concentration in Surabaya river (sampled from Ngagel) from 2015 to 2017 ranges from 0.09 mg/l to 1.87 mg/l. The monthly ammonia concentrations for 2015 to 2017 are 0.45 mg/l, 0.94 mg/l and 1.27 mg/l, respectively [2]. Based on Government Regulation (PP) No. 82 of 2001, class I water quality standards which are meant for allotment of raw drinking water, the maximum concentration of ammonia in raw drinking water is 0.5 mg/l [3].

Free ammonium and ammonia cations are in equilibrium in water, depending on pH and temperature. At 20°C, ammonium ions predominate in drinking water below pH 9.3, while free ammonia is mainly found at pH above 9 [4]. pH adjustment can be used to influence the shape of ammonia in water [5].

Ammonia in water is very active with chlorine forming chloramine compounds. Comparison of the three forms of chloramine shows that (mono-chloramine, dichloramine, and trichloramine) is very dependent on the pH of the water. Monochloramine is more dominant at pH >8.5. Monochloramine and dichloramine both exist at pH between 4.5 and 8.5 and trichloramine is formed at pH<4.5. Monochloramine is the dominant substance formed in the atmosphere for the pH in the water treatment process (pH 6 – 9) [6].
For pure water, the dose of chlorine 1 mg/l produces residual chlorine 1 mg/l. But if there is ammonia in the water, the chlorine compound can react with ammonia to form chloramine. The peak of monochloramine formation occurs when the ratio of chlorine to ammonia is between 4:1 and 6:1, then decreases to a minimum called a breakpoint. Breakpoint, when chloramine is oxidized to nitrogen gas, occurs when the ratio of chlorine to ammonia-N is between 7.5:1 and 11:1 [7]. In the process of drinking water treatment, it is desired that only monochloramine will be formed because dichloramine and trichloramine cause an unpleasant taste in water.

With the increasing concentration of ammonia in raw water, the need for chlorine compounds for disinfection is very large. If the concentration of ammonia in raw water can be reduced, the chlorine demand will decrease so that the cost of producing drinking water will also be cheaper. Also, if the concentration of ammonia is high, there will be additional affixing of chlorine, so the possibility of micropollutants such as trihalomethane, chlorophenol, and others is also becoming more intense.

Trihalomethane (THM) such as chloroform, dichloromethane, bromodichloromethane, dibromochloromethane, bromoform, 1,2 dichloroethane, and carbon tetrachloride are chlorine compounds produced due to the process of chlorination of water. These compounds are carcinogens. This encourages US EPA to determine the limit of maximum (MCL) THM content of 100 µg/l [8].

Removal of ammonia can be achieved via physicochemical treatment and biological treatment. Chlorination, ion exchange, and membrane filtration are known as common physicochemical treatment methods applied in the Drinking Water Treatment Plant [9-11]. However, biological approach has gained much attention nowadays and is considered as a future technology for drinking water production. In biological treatment, ammonia will undergo an oxidation process to form nitrite by the ammonia-oxidizing bacteria (AOB) or ammonia-oxidizing archaea (AOA) and further oxidized to nitrate by nitrite-oxidizing bacteria (NOB). Most of the microorganisms detected are Nitrosomonas and Nitrobacter type of bacteria [12-14].

The objective of this research was to conduct a field trial of the biofilter technology application to remove ammonia pollutants in the raw water taken from the Surabaya river. The research was conducted by operating a biofilter pilot plant consisting of lamella-type sedimentation tanks and biofilter reactors filled with honeycomb type-plastic media.

2. Material and method

2.1. Material
This research was conducted by operating a pilot plant of model test biofilter consisting of a lamella-type sedimentation tank with an effective volume of 14.2 m³, and a biofilter reactor unit with an effective volume of 18 m³ filled with 9 m³ honeycomb type plastic media (50% of the effective volume of the biofilter reactor). The biofilter media is made of PVC sheet with a specific area of ± 200 m²/m³. The dimensions of the sedimentation tank and biofilter reactor can be seen in table 1. The pilot plant model test biofilter is also equipped with a submersible pump-type feed pump, flow rate control valve, rotameter and air blower.

| Table 1. Dimensions of sedimentation tank and biofilter reactor. |
|---------------------------------------------------------------|
| **Dimensions** | **Sedimentation Tank** | **Biofilter reactor** |
| Type            | Lamella Settling       | Fixed bed             |
| Width           | 2.0 m                  | 2.0 m                 |
| Length          | 3.5 m                  | 4.5 m                 |
| Water depth     | 2.5 m                  | 2.0 m                 |
| Free Space Height| 0.3 m                 | 0.6 m                 |
| Effective Volume| 14.2 m³                | 18 m³                 |
2.2. Treatment Process
Raw water taken from the Surabaya river is pumped using a submersible pump and flowed through the rotameter then flowed into the inlet of the sedimentation tank. Inside the inlet pipe, before entering the rotameter, is equipped with a bypass valve to regulate the flow of water entering the sedimentation tank. Rotameter is equipped to see the raw water that flows into the settling basin. The sedimentation tank uses the lamella system, which functions to settle suspended solids that can interfere with the performance of the biofilter process.

The overflow from the sedimentation tank flowed to the inlet of the biofilter reactor while continuously being aerated using an air bower. Inside the biofilter reactor is filled with honeycomb-type plastic media, which functions as a place for the growth of microbes in the water. After a few days, microbes will grow attached to the surface of the media in the biofilter reactor. As long as it passes through the biofilter media, ammonia and also organic pollutants in the raw water will come into contact with microbes that are attached to the surface of the biofilter, and a biodegradation process will occur. The organic pollutants in the raw water will be degraded into water and carbon dioxide compounds, and the ammonia compounds will be oxidized into nitrate. Besides, the iron or manganese will also be oxidized into iron oxide or manganese oxide which is water-insoluble.

Overflow from the biofilter reactor is processed water from the biofilter, which then enters the reservoir, and overflow from the reservoir has then flowed to the existing water treatment plant. The process diagram used in this study can be seen in figure 1, while the pilot plant unit that has been installed can be seen in figure 2.

![Figure 1](image1.png)

Figure 1. The process diagram of the biofilter test model pilot plant used in this study.

![Figure 2](image2.png)

Figure 2. Installation of Biofilter Test Model pilot plant.

2.3. Sampling and Analysis of Water Quality
Water sampling is carried out every day, including raw water entering the sedimentation tank and the water that comes out of the biofilter reactor. Sampling was done at the same time every day whenever possible. Water samples were inserted into the sampling bottle that has been prepared. The water quality analysis was carried out in the PDAM Surabaya laboratory. The pollutant parameters analyzed
include acidity (pH), dissolved oxygen concentration (DO), turbidity, ammonium (NH₄⁺), nitrate (NO₃⁻), and nitrite (NO₂⁻).

2.4. Experiment Time
The biofilter pilot plant performance test was conducted for about 5 months starting in February until June 2018. The experiment was done starting from the seeding process, continued by a performance test of the biofilter test model under conditions of varying retention time in the reactor. The scenarios for the retention time of the biofilter test model to improve raw water quality can be seen in table 2.

| Experiment | Flowrate (m³/hr) | HRT Sedimentation Tank (Min) | HRT Biofilter reactor (Min) | Total HRT Model Test Biofilter (Min) |
|------------|------------------|-----------------------------|-----------------------------|-----------------------------------|
| Seeding    | 6                | 142                         | 180                         | 322                               |
| Experiment-1 | 9              | 93                          | 120                         | 213                               |
| Experiment-2 | 18             | 49                          | 60                          | 109                               |
| Experiment-3 | 24             | 49                          | 45                          | 81                                |
| Experiment-4 | 36             | 24                          | 30                          | 54                                |

Note:
The effective volume of Sedimentation Tank : 14.2 m³;
The effective volume of Biofilter reactor : 18 m³

3. Results and Discussion
For the seeding process, the pilot biofilter test model started operating on February 1, 2018 with a water discharge of 6 m³ per hour, while the retention time in the sedimentation tank is 2.37-hours (142-minutes) and the retention time is 3-hours (180-minutes) in the biofilter reactor. The seeding process was performed for almost one month until March 1, 2018.

Further, the raw water discharge was changed to 9 m³ per hour, so that the retention time in the sedimentation tank becomes 1.55-hours (93-minutes) and the retention time in the biofilter reactor becomes 2-hours (120-minutes). The experiment was conducted for three weeks starting from March 2 to March 20, 2018.

Afterwards, the raw water discharge was changed again to 18 m³ per hour, so that the retention time in the sedimentation tank becomes 0.78-hours (49-minutes) and the retention time in the biofilter reactor becomes 1-hour (60-minutes). Experiments with 18 m³ raw water flow are carried out for about one month starting from March 21, 2018, to April 19, 2018.

Then, the experiment was conducted by changing the raw water flow to 24 m³ per hour, so that the retention time in the sedimentation tank was 36-minutes and the retention time in the biofilter reactor was 45-minutes. Experiments with the raw water discharge of 24 m³ per hour were done for approximately one month starting from 20 April 2018 until 17 May 2018. Afterwards, the discharge of raw water was changed to 36 m³ per hour, so that the retention time in the sedimentation tank becomes 24-minutes and retention time in the biofilter reactor becomes 30-minutes. Experiments with the raw water flow of 36 m³ per hour were performed for approximately two weeks, starting from May 18, 2018, to May 31, 2018. During the study, analysis of the water quality inlet and outlet of the biofilter test model was conducted, and water quality analysis began on February 5, 2018.

3.1. Turbidity
The turbidity concentration of raw water (inlet) and treated water (outlet), and the removal efficiency are shown in figure 3. During the experiment with the raw water discharge of 6 m³ per hour, the retention time in the sedimentation tank is 2.37-hours (142-minutes) and retention time is 3-hours (180-minutes) in the biofilter reactor. The inlet turbidity ranged from 70.8 NTU - 455 NTU or 177.99
NTU for average. After going through the biofilter test model process, the turbidity dropped to 32.30 NTU - 195 NTU or the average outlet turbidity of 99.48 NTU. Turbidity removal efficiency ranged from 13.37 - 79.69%, or 41.67% for average.

In the experiments with raw water discharge of 9 m$^3$ per hour, the retention time in the sedimentation tanks is 1.55-hours (93-minutes) and retention time is 2-hours (120-minutes) in the biofilter reactor. Turbidity in raw water (inlet) ranged from 140 NTU - 268 NTU or 189.33 NTU for average. After going through the biofilter test model process, the turbidity dropped to 72.70 NTU - 150 NTU or 114.58 NTU for average. Turbidity removal efficiency ranged from 17.44 - 69.58%, or 36.87% for average.

![Inlet and outlet turbidity and total removal efficiency](image)

**Figure 3.** Inlet and outlet turbidity and total removal efficiency.

In the experiments with raw water discharge of 18 m$^3$ per hour, the retention time in the sedimentation tank is 0.78-hours (49-minutes) and the retention time is 1-hour (60-minutes) in the biofilter reactor. The turbidity in the raw water (inlet) ranged from 53.70 NTU - 357 NTU or 114.38 NTU for average. After going through the biofilter test model process, the turbidity dropped to 32.80 NTU - 196 NTU, with 79.16 NTU for average. The turbidity removal efficiency ranged from 13.21 - 59.03%, or 30.34% for average.

In the experiments with raw water discharge of 24 m$^3$ per hour, the retention time in the sedimentation tank is 36-minutes, and the retention time is 45-minute in the biofilter reactor. The turbidity in raw water (inlet) ranged from 23 NTU - 162 NTU, and the turbidity average was 56.89 NTU. After going through the biofilter test model process, the turbidity dropped to 16.19 NTU - 135 NTU, with 37.66 NTU for average. The turbidity removal efficiency ranged from 16.67 - 45.07%, with 33.83% for average.

In the experiments with the raw water discharge of 36 m$^3$ per hour, the retention time in the sedimentation tank is 24-minutes, and the retention time of 30-minutes in the Biofilter reactor, the turbidity in raw water (inlet) ranged from 11.3 NTU - 31.3 NTU with the average of 22.43 NTU. After going through the biofilter test model process, the turbidity dropped to 8.04 NTU - 24.4 NTU, and the average was 15.84 NTU. Turbidity removal efficiency ranged from 12.05 - 47.39%, or 29.71% for average.
In general, the faster the retention time in the biofilter test model reactor is, the greater the decrease in turbidity removal efficiency will be. Based on the experiment above, the average efficiency of the largest turbidity removal was 41.67% which occurred during the experiment with a discharge of 6 m$^3$ per hour. With a discharge rate of 36 m$^3$ per hour, the average efficiency of turbidity removal decreased to 29.71%. Based on other research conducted by Suprihatin et al., the biofilter process with a hydraulic residence time of 2-hours can reduce the turbidity of raw water turbidity to 55% [15].

3.2. Dissolved oxygen (DO)

The concentration of dissolved oxygen (DO) in the raw water (Inlet) and treated water (Outlet), and the percentage of the increase in DO concentration are shown in figure 4. The overall DO concentration after the biofilter process has increased due to the aeration process done inside the biofilter reactor.

During the experiments with raw water discharge of 6 m$^3$ per hour, the retention time in a sedimentation tank is 2.37-hours (142-minutes), and the retention time is 3-hours (180-minutes) in the biofilter reactor. The DO concentrations in the raw water (inlet) ranged from 3.35 mg/l - 4.11 mg/l with the average of 3.75 mg/l. After going through the biofilter test model process, the DO concentration rose to 4.17 mg/l - 5.71 mg/l or 4.78 mg/l for average. The percentage increase in DO range from 15.02% - 52.53%, or 28.65% for average.

![Figure 4. Concentration of Dissolved Oxygen (DO) in biofilter inlet and outlet and increase percentage.](image)

In the experiments with raw water discharge of 9 m$^3$ per hour, with retention time in sedimentation tank of 1.55-hours (93-minutes) and retention time of 2-hours (120-minutes) in the biofilter reactor, the DO concentrations in the raw water (inlet) ranged from 2.14 mg/l - 3.57 mg/l and the average DO concentrations were 3.10 mg/l. After going through the biofilter test model process, the DO concentration rose to 2.98 mg/l - 4.38 mg/l, and the DO outlet concentration on average was 3.99 mg/l. The percentage of increase in DO ranged from 18.77% - 39.25%, and the average percentage of increase in DO was 31.22%.

In the experiments with raw water discharge of 18 m$^3$ per hour, with retention time in sedimentation tank of 0.78-hours (49-minutes) and the retention time of 1-hour (60-minutes) in the Biofilter reactor, the DO concentrations in raw water (inlet) ranged from 15.08 mg/l - 23.97 mg/l and
the DO average was 19.59 mg/l. After going through the biofilter test model process, the DO concentration rose to 8.99 mg/l - 17.03 mg/l, and the DO outlet concentration on average was 8.99 mg/l. The percentage of increase in DO range from 13.31% - 52%, with an average of 31.22%.

In the experiments with a raw water discharge of 24 m³ per hour, with the 36-minutes retention time in the sedimentation tank and retention time of 45-minutes in the Biofilter reactor, DO concentrations in the raw water (inlet) ranged from 1.64 mg/l - 2.46 mg/l and the average of DO concentrations was 2.16 mg/l. After going through the biofilter test model process, the DO concentration rose to 2.37 mg/l - 3.52 mg/l and the average DO outlet concentration was 2.97 mg/l. The percentage of DO increase ranged from 13.31% - 52%, with an average of 31.22%.

In the experiments with a raw water discharge of 36 m³ per hour, with the 36-minutes retention time in the sedimentation tank and retention time of 45-minutes in the Biofilter reactor, DO concentrations in the raw water (inlet) ranged from 1.12 mg/l - 2.39 mg/l or 1.89 mg/l on average. After going through the biofilter test model process, the DO concentration rose to 2.57 mg/l - 3.74 mg/l and, and the average DO outlet concentration was 3.01 mg/l. The percentage increase in DO ranges from 25.81% - 3.52%, with an average of 38.30%.

3.3. pH

The raw water pH (inlet) and treated water (outlet) during the experiments are shown in figure 5. During the experiments with raw water discharge of 6 m³ per hour, the retention time in the sedimentation tank is 2.37-hours (142-minutes) and 3-hour (180-minutes) in the biofilter reactor. The pH of raw water ranged from 7.03 to 7.31, with the average pH of 7.18. After going through the biofilter test model process, the pH increased, ranged from 7.51 - 8.20, with an average outlet pH of 7.83. Thus, it has increased by 9.05%.

Figure 5. Inlet and outlet pH graph of the biofilter test model.

In the experiments with the raw water discharge of 9 m³ per hour, with retention time in the sedimentation tank of 1.55-hours (93-minutes), and 2-hours (120-minutes) in the biofilter reactor, the pH of raw water ranged from 7.01 - 7.64, with the average pH of 7.27. After going through the biofilter test model process, the pH increased, ranging from 7.41 - 7.87, with an average outlet pH of 7.70. Thus, it has increased by 5.91%.
In the experiments with the raw water discharge of 18 m³ per hour with retention time in the sedimentation tank of 0.78-hours (49-minutes), and retention time of 1-hour (60-minutes) in the biofilter reactor, the pH of raw water ranged from 7.17 - 7.51, with the average pH of 7.33. After going through the biofilter test model process, the pH increased, ranging from 7.49 - 7.81, with an average outlet pH of 7.60. Thus, it has increased by 3.68%.

In the experiments with the raw water discharge of 24 m³ per hour, with retention time in the sedimentation tank of 36-minutes, and 45-minute in the biofilter reactor, the pH of raw water ranged from 7.09 to 7.45, with an average pH of 7.29. After going through the biofilter test model process, the pH increased ranging from 7.36 - 7.63 with an average outlet pH of 7.55. Thus, it has increased by 3.57%.

In the experiments with the raw water flow of 36 m³ per hour, with retention time in the sedimentation tank of 24-minute, and 30-minute in the biofilter reactor, the pH of raw water ranged from 7.29 - 7.37, with an average pH of 7.34. After going through the biofilter test model process, the pH increased ranging from 7.54 to 7.99 with an average outlet pH of 7.60. Thus, it has increased by 3.54%.

From figure 5, it could be seen that at all hydraulic retention times, the pH of treated water is always higher than the pH of the raw water. This was due to the aeration effect, in which the carbon dioxide dissolved in the water lowered because it is replaced by oxygen gas. Carbon dioxide gas in the water tends to form an equilibrium and produce carbonate ions. With the reduction in carbonate ions due to reduced carbon dioxide, the pH will increase. Even though the pH of treated water rises, it is still in the neutral range (pH 7-8), so it would not be a problem for the processed water. From the overall condition in the study, the pH of the outlet water is still in the neutral range, which ranged between 7-8, while the standard pH is 6-9.

3.4. Ammonia

Ammonia concentrations in the raw water and treated water, as well as their removal efficiency, are shown in figure 6. During the experiments with raw water discharge of 6 m³ per hour, with retention time in the sedimentation tank of 2.3-hours (142-minutes), and 3-hours (180-minutes) in the biofilter reactor, the concentration of ammonia in the raw water (inlet) ranged from 0.99 mg/l - 1.34 mg/l, with the average of 1.14 mg/l. After going through the biofilter test model process, the ammonia concentration dropped to 0.23 mg/l - 1.11 mg/l, with the average of 0.66 mg/l. The ammonia removal efficiency ranged from 9.30 - 76.93%, with an average of 42.96%.

In the experiments with the raw water discharge of 9 m³ per hour, with retention time in the sedimentation tank of 1.55-hours (93-minutes), and 2-hours (120-minutes) in the biofilter reactor, the concentration of ammonia in raw water (inlet) ranged from 0.68 mg/l - 1.24 mg/l, with average of 1.05 mg/l. After going through the biofilter test model process, the ammonia concentration dropped to 0.17 mg/l - 1.14 mg/l, or 0.63 mg/l for average. Ammonia removal efficiency ranged from 1.95 - 85.53%, with average efficiency of 39.1%.

In the experiments with the raw water discharge of 18 m³ per hour, the retention time in the sedimentation tank of 0.78-hours (49-minutes), and the retention time of 1 hour in the biofilter reactor (60 minutes), the concentration of ammonia in raw water (inlet) ranged from 0, 26 mg/l - 1.69 mg/l, with the average of 0.61 mg/l. After going through the biofilter test model process, the ammonia concentration decreased to 0.17 mg/l - 0.96 mg/l, with 0.36 mg/l for average. Ammonia removal efficiency ranged from 2.71 - 87.39%, or 34.95% for average.

In the experiments with the raw water discharge of 24 m³ per hour, the retention time in the sedimentation tank of 36-minutes, and the retention time of 45-minute in the biofilter reactor, the concentration of ammonia in the raw water (inlet) ranged from 0.08 mg/l - 1.91 mg/l, with the average of 0.95 mg/l. After going through the biofilter test model process, the ammonia concentration dropped to 0.03 mg/l - 0.61 mg/l, and the ammonia concentration was 0.31 mg/l. The ammonia removal efficiency ranged from 26.93 - 89.15%, or 57.84% for average.
In the experiments with the raw water discharge of 36 m³ per hour, with retention time in the sedimentation tank of 24-minute sedimentation tank, and 30-minutes in the biofilter reactor, the concentration of ammonia in the raw water (inlet) ranged from 0.08 mg/l - 1.05 mg/l, with the average of 0.84 mg/l. After going through the biofilter test model process, the ammonia concentration dropped to 0.04 mg/l - 0.65 mg/l, with the average of 0.41 mg/l. Ammonia removal efficiency ranged from 33.06 - 73.92%, or the average efficiency was 52.0%.

During the study, the concentration of ammonia in the raw water ranged from 0.08 mg/l to 1.91 mg/l. During the experiment with a discharge of 24 m³ per hour, ammonia removal achieved the highest average efficiency of 57.84%. Whereas in experiments with raw water debit 6-18 m³ per hour, the average ammonia removal efficiency tends to decrease from 42.96 to 34.95%. From those results, it can be seen that the efficiency of ammonia removal occurs in experiments with a discharge of 24 m³ per hour, where the retention time in the sedimentation tank was 36-minutes, and the retention time in the biofilter reactor was 45-minutes, with a total retention time of 81-minutes (1.35-hours). The average concentration of ammonia in the raw water (inlet) is 0.95 mg/l, and the average ammonia concentration after the biofilter test model process (outlet) was 0.31 mg/l, with an ammonia removal efficiency of 57.84%. The quality standard for maximum ammonia concentration for class I water for drinking water is 0.5 mg/l. The reduction of ammonia in water goes through the nitrification process which is a microbiological process in which ammonia compounds are sequentially oxidized to nitrite and nitrate ($\text{NH}_3 \rightarrow \text{NO}_2^- \rightarrow \text{NO}_3^-$). The nitrification process is mainly carried out by two groups of bacteria, namely autotrophic nitrifying bacteria that can form organic molecules using energy obtained from inorganic sources, in this case ammonia or nitrite.

The presence of ammonia in the raw water will require high oxidizing agents (oxidants) and reduce the efficiency of disinfection. The reaction between ammonia and chlorine is very fast, and ammonia can negatively affect the removal of organic and inorganic compounds such as iron, manganese, and arsenic by reducing the chlorine availability for oxidation [16,17]. Free ammonium and ammonia cations are in an equilibrium condition in the water, depending on pH and temperature. At 20°C, ammonium ions dominate in the drinking water which has a pH below 9.3, while free ammonia is mainly found at a pH above 9.3 [4].

**Figure 6.** The concentration of inlet ammonia and biofilter outlets, as well as removal efficiency.
Ammonia in the water reacts with chlorine compounds, forming the chloramine compounds. Comparison of the three forms of chloramine (mono-chloramine, dichloramine, and trichloramine) highly depends on the pH of the water. The chlorination breakpoint is the point at which the need for chlorine is fully met in terms of chlorine addition into the water. When chlorine is added into the water, the chlorine reacts with pollutant compounds, especially ammonia in the water. This ammonia compound will react with the chlorine so that the residue or residual chlorine in the water becomes zero. The chlorination breakpoint requires a chlorine dosage about 8-10 times higher (based on weight) than ammonia concentration to achieve free chlorine residue. This process is a series of reactions in which monochloramine is formed first. The reaction speed of the monochloramine formation depends on pH, temperature and the weight ratio between chlorine and ammonia-nitrogen (Cl\_2: NH\_3-N), generally ranges from 3:1 to 5:1. After monochloramine is formed and the ratio of Cl\_2:NH\_3-N is greater than 5:1, chlorination breakpoint is produced through two stages of reactions, namely the reaction of monochloramine with chlorine to form dichloramine and trichloramine, then decomposition of trichloramine into free nitrogen gas (N\_2). Both stages of the reactions require excessive doses of chlorine [18].

With the increasing concentration of ammonia in the raw water, the chlorine compounds’ requirement for disinfection becomes very high. Also, if the ammonia concentration is high, the chlorine affixation will become high, which causes the possibility of micropollutants forming a chlorinated byproduct compound, such as trihalomethane, chlorophenol, and others to be higher. If the ammonia concentration in the raw water can be lowered, the need for chlorine will be lessened, and the production cost for drinking water also becomes cheaper.

Trihalomethane (THM) such as chloroform, dichloromethane, bromodichloromethane, dibromo-chloromethane, bromoform, 1,2 dichloroethane, and carbon tetrachloride are chlorine compounds produced due to the chlorination process of the water. These compounds are carcinogens. This encourages US EPA to determine the maximum (MCL) THM content limit of 100 µg/l [8].

3.5. Nitrate

The concentration of nitrate in the raw water (inlet) and treated water (outlet), as well as the removal efficiency, are shown in figure 7. During the experiments with the raw water discharge of 6 m\^3 per hour, the retention time in the sedimentation tank is 2.37-hours (142-minutes) and the retention time is 3-hours (180-minutes) in the biofilter reactor. The concentration of nitrate in raw water (inlet) ranged from 1.04 mg/l - 1.21 mg/l, with the average of 1.12 mg/l. After going through the biofilter test model process, the nitrate concentration decreased to 0.84 mg/l - 1.12 mg/l, with an average of 0.94 mg/l. The nitrate removal efficiency ranged from 7.57 - 20.66\%, with an average efficiency of 16.04\%.

In the experiments with the raw water discharge of 9 m\^3 per hour, with the retention time in the sedimentation tank of 1.55 hours (93 minutes) and 2-hours (120-minutes) in the biofilter reactor, the concentration of nitrate in the raw water (inlet) ranged from 0.98 mg/l - 1.3 mg/l with the average of 1.10 mg/l. After going through the process of the biofilter test model, the concentration of nitrate dropped to 0.81 mg/l - 1.04 mg/l, with the average of 0.93 mg/l. The nitrate removal efficiency ranged from 4.74 - 22.42\%, with an average efficiency of 15.27\%.

In the experiments with the raw water discharge of 18 m\^3 per hour, with retention time in the sedimentary tank of 0.78 hours (49 minutes) and 1-hour (60-minutes) in the biofilter reactor, the concentration of nitrate in the raw water (inlet) ranged from 1.02 mg/l - 1.13 mg/l, with the average inlet nitrate concentration of 1.08 mg/l. After going through the biofilter test model process, the nitrate concentration decreased to 0.90 mg/l - 1.07 mg/l, with the average outlet nitrate concentration of 1.02 mg/l. The nitrate removal efficiency ranged from 2.71 - 15.11\%, with an average removal efficiency of 5.66\%. 
Figure 7. The concentration of nitrate in the raw water (Inlet) and treated water (Outlet).

In the experiments with the raw water discharge of 24 m$^3$ per hour, with retention time in the sedimentary tank of 36-minutes and 45-minutes in the biofilter reactor, the concentration of nitrate in the raw water (inlet) ranged from 1.01 mg/l - 1.09 mg/l, with the average inlet nitrate concentration of 1.06 mg/l. After going through the biofilter test model process, the nitrate concentration decreased to 0.29 mg/l - 1.08 mg/l, with the average outlet nitrate concentration of 0.88 mg/l. The nitrate removal efficiency ranged from 1.10 - 90.83%, with an average of 16.40%.

In the experiments with the raw water discharge of 36 m$^3$ per hour, with retention time in the sedimentary tank of 24-minutes and the retention time of 30-minutes in the biofilter reactor, the concentration of nitrate in raw water (inlet) ranged from 1.01 mg/l - 1.05 mg/l, with the average of 1.04 mg/l. After going through the biofilter test model process, the nitrate concentration decreased to 0.88 mg/l - 0.99 mg/l, with the average of 0.93 mg/l. The nitrate removal efficiency ranged from 6.10 - 15.35%, with an average of 10.03%. During the experiment with a discharge of 24 m$^3$ per hour, nitrate removal achieved the highest average efficiency of 16.40%. Whereas in experiments with the raw water discharge of 6-18 m$^3$ per hour, the average nitrate removal efficiency tends to decrease, from 16.04 to 5.66%.

From these experimental results, it can be seen that the highest efficiency of nitrate removal occurs in the experiments with a discharge of 24 m$^3$ per hour, with the retention time in the sedimentation tank of 36-minutes and the retention time of 45-minute in the biofilter reactor, or 81-minutes (1.35-hours) in total. The average nitrate concentration in the raw water (inlet) is 2.32 mg/l and the average nitrate concentration after the biofilter test model process (outlet) is 0.88 mg/l, with an efficiency of average nitrate removal of 27.54%. The standard quality of the maximum nitrate concentration for class I water for raw drinking water is 10 mg/l.

3.6. Nitrite
The concentration of nitrite in the raw water (inlet) and treated water (outlet), as well as the removal, are shown in figure 8. During experiments with the raw water discharge of 6 m$^3$ per hour, with retention time in the sedimentation tank of 2.37-hours (142-minutes), and the retention time of 3-hours (180-minutes) in the biofilter reactor, the nitrate concentration in the raw water (inlet) ranged from 0.049 mg/l - 0.078 mg/l, with the of 0.065 mg/l. After going through the biofilter test model process,
the nitrite concentration dropped to 0.007 mg/l - 0.046 mg/l, with the average of 0.037 mg/l. The nitrite removal efficiency ranged from 19.10 - 87.45%, with an average removal efficiency of 43.64%.

In the experiments with the raw water discharge of 9 m$^3$ per hour, with the retention time in the sedimentation tanks of 1.55-hours (93-minutes) and the retention time of 2-hours (120-minutes) in the biofilter reactor, the nitrite concentration in the raw water (inlet) ranged from 0.027 mg/l - 0.145 mg/l, with the average concentration of 0.095 mg/l. After going through the biofilter test model process, the nitrite concentration dropped to 0.019 mg/l - 0.122 mg/l, or 0.071 mg/l on average. The nitrite removal efficiency ranged from 3.79% - 51.45%, with an average removal efficiency of 24.44%.

In the experiments with the raw water discharge of 18 m$^3$ per hour, with the retention time in the sedimentation tanks of 0.78-hours (49-minutes) and the retention time of 1-hour (60-minutes) in the biofilter reactor, the nitrite concentration in the raw water (inlet) ranged from 0.015 mg/l - 0.283 mg/l, with the average of 0.126 mg/l. After going through the biofilter test model process, the nitrite concentration dropped to 0.001 mg/l - 0.184 mg/l, with the average of 0.067 mg/l. The nitrite removal efficiency ranged from 9.24% - 91.16%, with an average removal efficiency of 49.79%.

In the experiments with the raw water discharge of 24 m$^3$ per hour, with the retention time in the sedimentary tank of 36-minutes, and the retention time of 45-minute in the biofilter reactor, the nitrite concentration in the raw water (inlet) ranged from 0.007 mg/l - 0.318 mg/l, or 0.144 mg/l for average. After going through the biofilter test model process, the nitrite concentration dropped to 0.004 mg/l - 0.133 mg/l, with the average of 0.049 mg/l. The nitrite removal efficiency ranged from 13.47% - 96.30%, with an average efficiency of 59.41%.

In the experiments with the raw water discharge of 36 m$^3$ per hour, the retention time in the sedimentary tank of 24-minute and the retention time of 30-minutes in the biofilter reactor the nitrite concentration in the raw water (inlet) ranged from 0.143 mg/l - 0.446 mg/l, with the average inlet nitrite concentration of 0.261 mg/l. After going through the biofilter test model process, the nitrite concentration dropped to 0.049 mg/l - 0.232 mg/l with the average outlet nitrite concentration of 0.143 mg/l. The nitrite removal efficiency ranged from 25.03% - 76.28%, with an average removal efficiency of 43.20%.

Figure 8. Nitrite inlet and outlet concentrations of biofilter and removal efficiency.
From these results, it can be seen that the highest nitrate removal efficiency occurs in experiments with a discharge of 24 m$^3$ per hour, the retention time in the sedimentation tank of 36-minutes, and the retention time of 45-minutes in the biofilter reactor, with the total retention time of 81-minutes (1.35-hours). The average nitrate concentration in the raw water (inlet) was 0.144 mg/L and the average nitrite concentration after the biofilter test model process (outlet) was 0.049 mg/L with an average nitrite removal efficiency of 59.41%. The quality standard for the maximum nitrite concentration for class I water for drinking water is 0.06 mg/L.

3.7. Recapitulation of research results for biofilter pilot plant (February-May 2018)

Ammonia load is the amount of ammonia compound that enters the reactor per unit volume of the biofilter test model reactor per unit time, which is expressed as the weight of organic matter per media volume unit per day. From the results of the experiments above, it shows that with an ammonia loading of 1 - 5 gr/m$^3$.hours (24 - 120 gr/m$^3$.day), the efficiency of ammonia compound is reduced by 30% - 50%.

The result recapitulation of the pilot biofilter plant research to improve the quality of raw water from the Surabaya river can be seen in Table 3.

| Q (m$^3$/hr) HRT (minute) | Average Removal Efficiency (%) |
|---------------------------|--------------------------------|
|                           | Turbidity | Ammonia | Nitrate | Nitrite |
| Q=6 m$^3$/hr              |           |         |         |         |
| ST: 142                   | 41.67     | 42.96   | 16.04   | 43.64   |
| BF: 180                   |           |         |         |         |
| Q=9 m$^3$/hr              |           |         |         |         |
| ST: 93                    | 36.87     | 39.1    | 15.27   | 24.44   |
| BF: 120                   |           |         |         |         |
| Q=18 m$^3$/hr             |           |         |         |         |
| ST: 49                    | 30.34     | 34.95   | 5.66    | 49.79   |
| BF: 60                    |           |         |         |         |
| Q=24 m$^3$/hr             |           |         |         |         |
| ST: 36                    | 33.83     | 57.84   | 16.40   | 59.41   |
| BF: 45                    |           |         |         |         |
| Q=36 m$^3$/hr             |           |         |         |         |
| ST: 24                    | 29.71     | 52.0    | 10.03   | 43.20   |
| BF: 30                    |           |         |         |         |

Note: ST: Sedimentation Tank  BF: Biofilter

According to the table, generally, the highest ammonia removal efficiency that can be achieved is with retention time (HRT) of 36-minute in the sedimentation tank and retention time of 45-minute in the biofilter reactor. Based on these considerations, for the ammonia removal application with the biofiltration process, the condition with a retention time of 81-minutes in the biofilter pilot plants is chosen. That is, 36-minutes of retention time in the sedimentation tank and 45-minutes of retention time in the biofilter reactor.

4. Conclusion

From the research results on the processing of drinking water raw water with submerged biofilter reactors with aeration using honeycomb plastic media, it can be concluded as follows:

1. The biofilter pilot plant process consists of sedimentation tank and biofilter reactors using honeycomb plastic media can reduce turbidity, ammonia, and nitrite in raw water.
2. The aeration process in the biofilter reactor can increase the dissolved oxygen concentration by 28.65 - 68.09% and increase the pH by 3.54 - 9.05%.

3. In general, the longer the retention time in the biofilter test model is, the greater the turbidity, ammonia, and nitrite removal efficiency in the raw water will be.

4. Based on technical considerations and the processed water quality of biofilter test models as well as construction costs, for the optimum planning basic data, experimental results data with the condition of total retention time (HRT) 81-minutes, with retention time (HRT) of 36-minutes in the sedimentation tank and retention time (HRT) of 45-minutes in the biofilter reactor, were selected as the optimum design.

5. In the total retention time in the 81-minute biofilter test model, the efficiency of turbidity, ammonia, nitrate, and nitrite removal were 33.83, 57.84, 16.40, and 59.41% respectively. From the results of the research above, it was found that with the ammonia load of 1 - 5 gr/m³.hours (24-120 gr/m³.day), the efficiency of ammonia compounds reduction by 30 - 50% was obtained.

6. In the process of chlorination, chlorine compounds can react with ammonia compounds in raw water, so with the decrease in the ammonia concentration, the consumption of chlorine will be reduced that the production costs will also be lower.

7. With the reduced use of chlorine compounds, the potential formation of micropollutant compounds due to the chlorination process will be reduced.

5. References

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