



Evaluation of the factors influencing the performance of a natural zeolite-based biologically aerated filter

Beena P. Nambiar*, Renu Pawels, G. Madhu

School of Engineering, Cochin University of Science and Technology, Kochi, Kerala, India, Tel.: +91 9633115118;
emails: beenasaji@gmail.com (B.P. Nambiar), renupawels@gmail.com (R. Pawels), profmadhugopal@gmail.com (G. Madhu)

Received 31 March 2023; Accepted 13 September 2023

ABSTRACT

Biological aerated filters (BAF) are attached growth aerobic systems used for the treatment of wastewater. This system can establish the physical filtration of solids along with the biological decomposition of organic matter. The performance of the BAF depends on the type of support media and its ability to meet objective of the treatment process. Natural zeolites, with properties such as ion exchange, larger surface area, capability to form biofilm, adsorption, regenerative properties etc., are a promising filter material in BAF systems. This paper investigates and discusses the effect of various factors like hydraulic retention time (HRT), carbon to nitrogen (C/N) ratio, aeration rate, depth of filter and particle size of natural zeolite which influence the performance of BAF. Based on the experimental results and optimization study using response surface methodology, carried out on the BAF system with varying process variables like HRT, C/N ratio, aeration rate and depth of filter, the natural zeolite media with particle size 1–3 mm range showed improved performance as compared to the coarser particles, that is, 3–5 mm and 5–10 mm. On conducting a final run using natural zeolite particle size of 1–3 mm under optimized value of HRT = 3.25 h, C/N = 6.2, aeration rate = 69 L/h, depth of filter = 83 cm, the BAF system observed chemical oxygen demand, biological oxygen demand, NH₃-N, turbidity and total dissolved solids removal percentage of 97%, 96%, 99%, 98% and 58%, respectively.

Keywords: BAF; Natural zeolite; Bio-film; Particle size; Response surface methodology; Central composite design

1. Introduction

Most of the natural sources of water like lakes, rivers and groundwater are increasingly getting contaminated and becoming less suitable for consumption due to the discharge of wastewater. A variety of treatment methods and systems including rainwater harvesting, reuse of wastewater, desalination etc. are receiving more attention and becoming popular for the treatment of wastewater nowadays. Among those systems, biological aerated filter (BAF) technology has the distinctive ability to remove solids, carbonaceous material and nitrogenous compounds in single unit [1]. The BAF reactors make use of granular media which allows separation

of solids and secondary or tertiary biological treatment in one unit [2]. They generate high performance even with their compact size and low cost. Its important features like stability in operation, minimum noise and odour during operation, and small space requirements make BAF a useful technology [3]. The BAF system typically consists of a medium that removes organic matter by biofilm formation and entrapment of suspended solids [4].

In a BAF system, selecting the appropriate granular media is very important since the quality of effluent depends on the capability of the media to maintain a high amount of active biomass and a variety of microbial populations [5]. The characteristics of the media, like surface area, surface roughness and porous nature also influence the biofilm

* Corresponding author.

formation and concentration. The media should be inert, stable, have a capacity to remove organic matter, strong to support self-weight and can grow biofilm on the surface [6]. So, selection of an appropriate support media forms a challenge in the designing and operation of the BAF system in addition to the aeration and backwash systems. It ensures that the effluent quality parameters attained are as per the regulated standard. The operation of the BAFs may be in an up-flow or down-flow manner. The BAF operating in the down-flow manner provides efficient supply of oxygen to biofilm attached to the support media of the biofilter [3].

In the present study natural zeolite is used as the filter media in the BAF system. Natural zeolites are hydrated aluminosilicates with honeycomb structure [7]. The three-dimensional structure of zeolite has tetrahedrons of SiO_4 and AlO_4^- connected with one another by common oxygen atoms. The isomorphous replacement of Si^{4+} ions by Al^{3+} ions create a negative charge density within the minute pores and acts as good cation exchange sites [8]. They are highly selective cation exchange materials as far as wastewater contaminants like ammonium ion are considered. They have adsorption properties along with the ability to form biofilm by bacterial attachment. They possess high porosity and large specific surface area for adsorption and biofilm attachment. Some organic matter which are present in dissolved or colloidal forms are larger molecules and cannot penetrate the fine pores of zeolite and get adsorbed on their surface. Microorganisms like bacteria gets trapped in between the intraparticle pores. Thus, the large surface area helps in maintaining a high amount of active bacterial population for the decomposition of organic matter present in the wastewater. The performance of BAF system is also influenced by the shape, size and surface roughness of the media. The rough surface of zeolite media provides a greater surface for the attachment of bacteria and formation of biofilm [9,10]. Also, the irregular surface of the zeolite media improved the performance as compared to any regular spherical media. In addition, natural zeolites have ability to undergo filtration for removal of organic matter and other solids from the deep, submerged reactor. The properties of the natural zeolite like inertness, low-cost, non-metallic and non-toxic nature makes it a very economical support media for wastewater treatment process. These physical features of the natural zeolite make it a suitable material as support media in BAFs. An additional benefit of using natural zeolites is their regenerative properties. The efficiency of natural zeolite-based BAF decreases only after operation for a long duration of time. Regeneration methods like heating regeneration, acid regeneration or NaCl/KCl regeneration can be applied to the zeolite media [11].

An efficient aeration system is necessary for BAFs with any type of support media. This system maintains the dissolved oxygen for the growth of the biofilm by bacterial activity. The activity of the aerobic bacteria increases significantly depending on the aeration provided to the system. The average percentage removal of various parameters like chemical oxygen demand (COD), biological oxygen demand (BOD), $\text{NH}_3\text{-N}$ etc. depends on the activity of bacteria and in turn on the volume of aeration provided [12].

The BAF is provided with a backwash system to remove the accumulated solids and excess of biomass on to the

surface of the support media so as to prevent the clogging of media and for maintaining the active population of the bacteria on the biofilm. Backwash system also helps to minimize the energy requirement in the treatment process [10]. However, if backwashing is done excessively, it can cause reduction of biomass resulting in poor performance of BAF. If the backwash frequency is less, the biochemical processing performance decline and the removal of both BOD and ammonia nitrogen also reduces [13,14]. Usually, for BAF system, backwashing is carried out every 24–48 h when used for secondary treatment [15].

The primary objective of the present study is to evaluate the capacity of the natural zeolite on the removal of organic matter from the wastewater including COD, BOD, $\text{NH}_3\text{-N}$ (ammonia-nitrogen), turbidity and TDS (total dissolved solids) and the influence of hydraulic retention time (HRT), carbon to nitrogen ratio (C/N ratio), aeration rate, depth of filter and zeolite particle size on the removal capacity. An optimization study was conducted on the BAF system using the central composite design (CCD) and response surface methodology (RSM) of Minitab 19 which involves the use of statistical approaches of modelling and analysing mathematical problems. RSM is a design method capable of providing a large amount of information using small number of experiments performed in an economical manner [16,17]. RSM determines the best value of independent variables to optimize responses for an operation [18]. It illustrates the output behaviour of the responses in the experiment and analyses it to interpret the level of optimization of the independent variables. The CCD and RSM employed in the present situation helped in the design of experiments, analysis and optimization of its responses in terms of percentage removal of COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS under the influence of varying values of HRT, C/N ratio, aeration rate, depth of filter and zeolite particle size.

2. Materials and methods

2.1. Experimental set-up

To determine the feasibility of the natural zeolite-based biologically aerated filter (NZBAF) system, a laboratory scale bio-filter in the form of a transparent rigid column made of glass was built. The reactor had a square cross-section of size 15 cm × 15 cm and height of 120 cm. An inlet was provided at a distance of 5 cm from the top and an outlet at 5 cm from the bottom of the filter. A perforated plate was used below the inlet of the biofilter column to uniformly distribute influent and another one was kept above the effluent outlet at the bottom to evenly withdraw the backwash water. The zeolite media was filled inside the biofilter between the two perforated plates which holds the media in place. Initially the depth of the media was maintained to 50 cm from the bottom of the perforated plate and was later varied based on the experimental needs. Two cubical glass tanks of cross-section size 30 cm × 30 cm and height of 30 cm were used, one as an equalizing tank for supplying influent and another one served as the settling basin for collecting effluent. The influent was pumped to the biofilter column using a submersible pump. An aeration system, consisting of an aerator and air diffusers were introduced

to provide aeration at equal intervals along the depth of the filter and rate of aeration was monitored. A backwash system was provided and operated at regular intervals to remove the suspended matter and excess amount of biomass accumulated so as to prevent the system from clogging. The effluent collected in the settling basin served the purpose of backwash water. A schematic diagram of the NZBAF is shown in Fig. 1.

2.2. Preparation of natural zeolite

To understand the influence of particle size on the performance of NZBAF, the natural zeolite was broken into small pieces and were graded into coarse (5–10 mm), medium (3–5 mm) and fine (1–3 mm) particles. The important physical characteristics of the coarse, medium and fine grained natural zeolite were then calculated in the laboratory and is as shown in Table 1.

2.3. Aeration system

The aeration system here consists of an air compressor for supplying air and an air diffuser to make the air supply smooth and homogenous within the NZBAF. The system under study is provided with an air compressor and 4 air diffusers which are provided at 4 different levels of the

media at equal intervals. The performance of the system under varying rates of aeration is studied for better removal efficiency.

2.4. Backwash system

For the NZBAF system under study, backwashing on a weekly basis was sufficient to prevent the clogging of the bioreactor. The treated water from the settling basin was pumped in an up-flow direction for backwashing of the zeolite media.

3. Analytical methods

3.1. Preparation of synthetic wastewater

The synthetic wastewater having similar characteristics as that of real greywater was prepared based on the composition shown in Table 2. The physicochemical parameters like COD, BOD, NH₃-N, turbidity and TDS were analyzed as per the standard methods [19] shown in Table 3. The dissolved oxygen, temperature, and pH were regularly monitored during the treatment process.

Table 1 Characteristics of coarse, medium and fine grained natural zeolite

| Characteristics | Coarse grain (5–10 mm) | Medium grain (3–5 mm) | Fine grain (1–3 mm) |
|---------------------|------------------------|-----------------------|---------------------|
| Bulk density (kg/L) | 1.054 | 1.102 | 1.187 |
| Void ratio | 1.28 | 1.11 | 1.17 |
| Porosity | 0.56 | 0.55 | 0.54 |
| Specific gravity | 2.40 | 2.49 | 2.57 |

Table 2 Composition of the synthetically prepared wastewater

| Chemical product | Quantity |
|---|----------|
| Distilled water, mL | 1,000 |
| Glucose (C ₆ H ₁₂ O ₆), mg/L | 300 |
| Sodium acetate trihydrate (C ₂ H ₃ NaO ₂ ·3H ₂ O), mg/L | 400 |
| Ammonium chloride (NH ₄ Cl), mg/L | 225 |
| Disodium hydrogen phosphate (Na ₂ HPO ₄), mg/L | 150 |
| Potassium dihydrogen phosphate (KH ₂ PO ₄), mg/L | 75 |
| Magnesium sulphate (MgSO ₄), mg/L | 50 |
| Cow dung, ml/L | 0.2 |

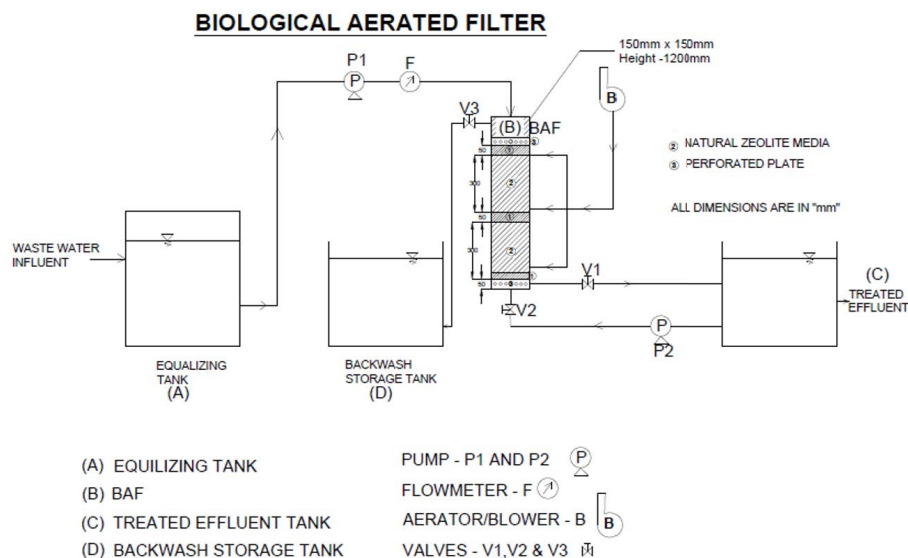


Fig. 1. Schematic diagram of NZBAF.

Table 3
Standard methods and instruments for analysis of various water quality parameters

| Parameter | Method | Instrument |
|--------------------------|--|------------------------|
| Chemical oxygen demand | Potassium dichromate reflux method | Reflux apparatus |
| Biological oxygen demand | Dissolved oxygen determination by Winkler method | BOD incubator |
| Ammonia nitrogen | Nesslerization method | Spectrophotometer |
| Turbidity | Nephelometric method | Nephelometer |
| Total dissolved solids | Electrometric method | Water quality analyzer |

3.2. Bacterial inoculation and acclimation

The biofilm formation on the surface of the biofilter media is influenced by its distinctive properties such as porosity, surface area and roughness. To reduce the start-up time of the NZBAF, the prepared synthetic wastewater was seeded with the activated sludge obtained from an effluent treatment plant and the nutrients required by the bacteria (including peptone, beef extract and NaCl). The bacteria present in the natural zeolite support media of the bio-filters were allowed to acclimatize for a period of 21 d. Aeration was provided at a rate of 15 L/h. After this period the COD in the effluent was constant and low which indicated that the bio-film was firmly formed.

After the process of bacterial acclimatization, the NZBAF was supplied with fresh set of synthetically prepared wastewater and its performance was studied by optimizing the operation of the NZBAF with natural zeolite of particle size 5–10 mm under varying conditions of HRT, C/N ratio, aeration rate and depth of filter. The study was further continued by operating the NZBAF with particle size of 3–5 mm and 1–3 mm and the observation made is discussed.

4. Results and statistical analysis

4.1. Performance of NZBAF using natural zeolite of different particle size, that is, 5–10 mm, 3–5 mm and 1–3 mm under varying operational conditions:

4.1.1. Hydraulic retention time

The influence on the performance of NZBAF under varying values of HRT on the removal of different process parameters were studied while all other variables such as C/N ratio, aeration rate and depth of filter were kept constant. The NZBAF was run under HRT values of 1, 2, 3, 4 and 5 h while maintaining the C/N ratio to 3, aeration rate to 15 L/h, depth of filter to 50 cm. The effluent characteristics were then determined after 1, 2, 3, 4 and 5 h of operation of NZBAF using coarse grained (5–10 mm), medium grained (3–5 mm) and fine grained (1–3 mm) particles.

The performance of biofilter in terms of COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS removal was then observed for all particle sizes. For particle size of 5–10 mm, the COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS were recorded as 72%, 81%, 85%, 68% and 17%, respectively. For 3–5 mm particle sizes the recorded values were 81%, 83%, 88%, 73% and 22% and for particle size of 1–3 mm the values obtained were 85%, 86%, 95%, 76% and 30%, respectively. Fig. 2a–c shows the

percentage removal of the various process parameters at varying HRTs for different particle sizes.

The increase in HRT, showed the increase in the removal of the carbonaceous and nitrogenous organic matter. The results indicated that percentage removal of the various process parameters was of little significance after 3 h. So, the optimum HRT for the further experiments was fixed as 3 h.

4.1.2. C/N ratio

The NZBAF was run with varying values of C/N ratios, that is, 4, 5, 6, 7 and 8 while maintaining the value of HRT to the optimal value of 3 h, aeration was provided at the rate of 15 L/h and the depth of the filter was maintained to 50 cm. At the end of each operation, the characteristics of the effluent were determined.

The results showed that maximum percentage removal was obtained for 1–3 mm particles as compared to 5–10 mm and 3–5 mm particles at a C/N ratio of 6. For particle size of 5–10 mm, the COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS were recorded as 78%, 83%, 85%, 75% and 21%, respectively. For 3–5 mm particle sizes the recorded values were 81%, 83%, 88%, 73% and 22%. The values obtained for 1–3 mm particles were 85%, 85%, 89%, 78% and 30%, respectively for COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS at a C/N ratio of 6. Fig. 3a–c shows the percentage removal of the various process parameters at varying C/N ratios for different particle sizes.

The further increase in C/N ratio to 7 or 8 was not quite effective in the percentage removal of any of the parameters like COD, BOD, $\text{NH}_3\text{-N}$, turbidity or TDS.

4.1.3. Aeration rate

The system was now operated with varying aeration rate of 15, 40, 65, 90 and 100 L/h maintaining the optimum HRT of 3 h and C/N ratio to 6. The depth of filter was kept as 50 cm. The zeolite particle sizes of 1–3 mm showed better performance as compared to 5–10 mm and 3–5 mm particle sizes at aeration rate of 65 L/h.

Fig. 4a–c shows the percentage removal of the various process parameters at varying aeration for different particle sizes. For particle size of 5–10 mm, the COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS removal were recorded as 86%, 80%, 90%, 88% and 29%, respectively. For 3–5 mm particle sizes the observed values were 87%, 87%, 92%, 90% and 42%. For 1–3 mm particle sizes, the maximum removal

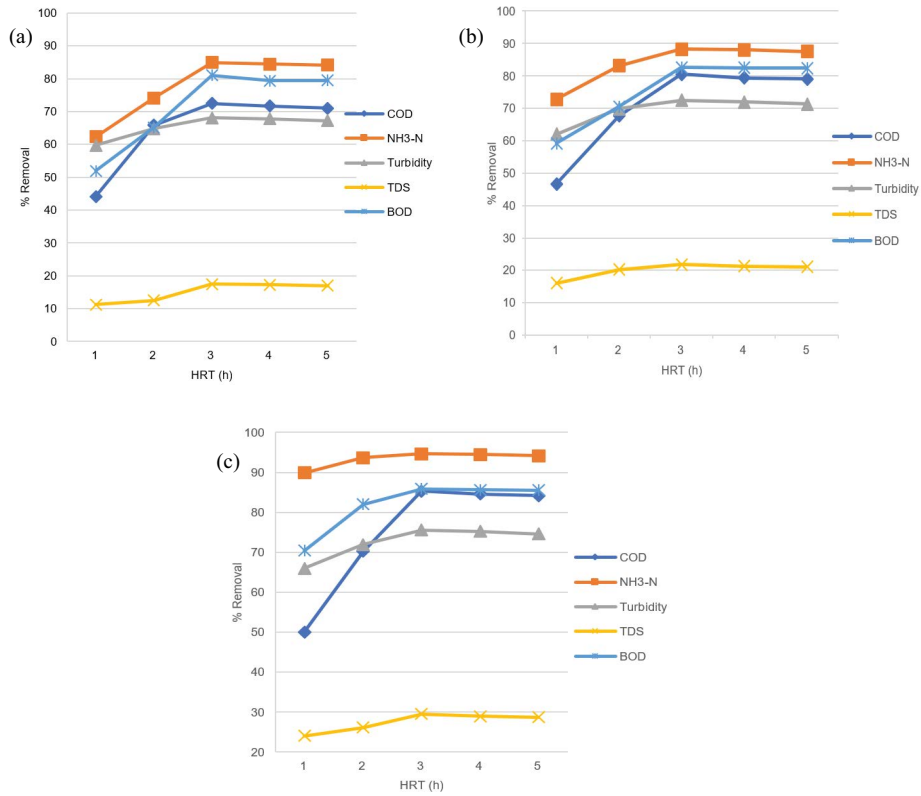


Fig. 2. Effect of varying hydraulic retention time on (a) 5–10 mm, (b) 3–5 mm and (c) 1–3 mm.

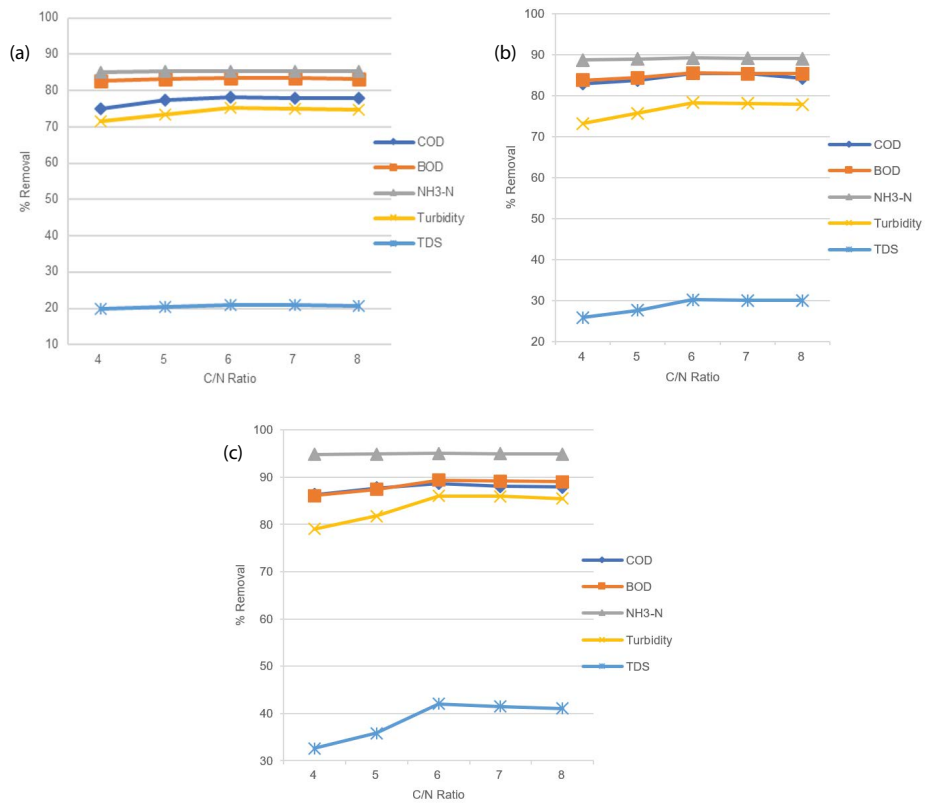


Fig. 3. Effect of varying C/N ratio on (a) 5–10 mm, (b) 3–5 mm and (c) 1–3 mm.

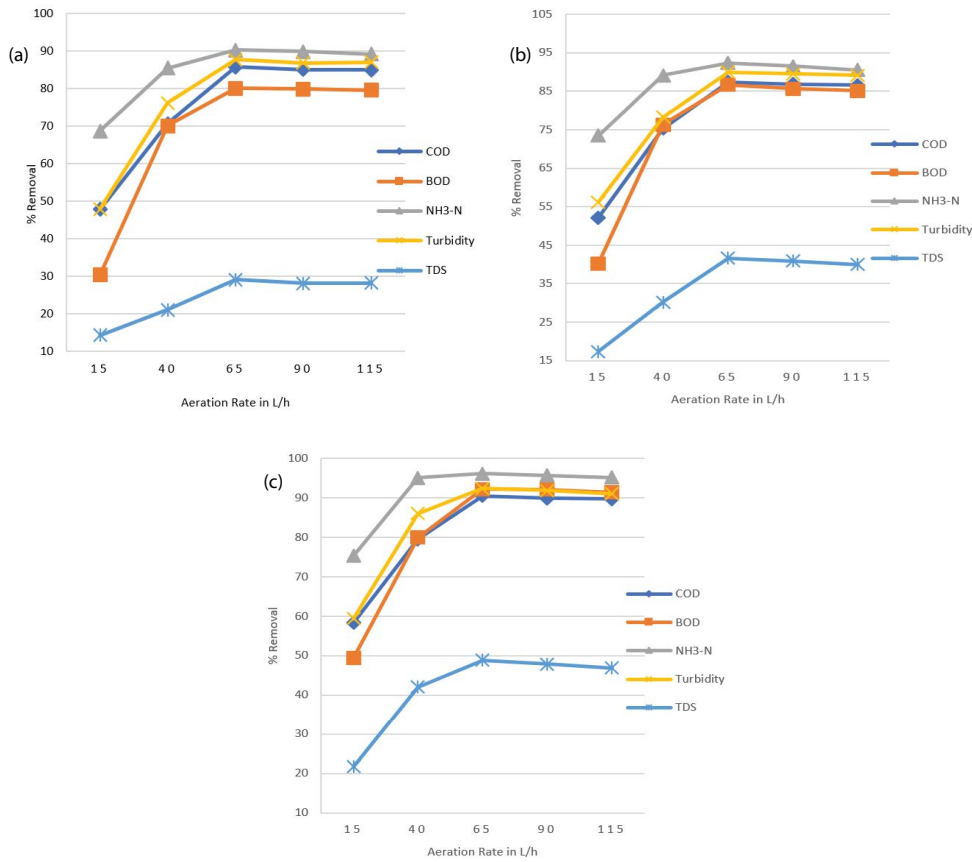


Fig. 4. Effect of varying aeration rate on (a) 5–10 mm, (b) 3–5 mm and (c) 1–3 mm.

percentage attained for COD, BOD, NH₃-N, turbidity and TDS was 90%, 92%, 96%, 92% and 49%, respectively.

Further increase in aeration rate to 90 L/h or 115 L/h was less effective in the removal of any of the process parameters to an appreciable value.

4.1.4. Depth of filter

The effect of the variation in the depth of filter was studied by operating the system at depth of filter of 50, 60, 70, 80, 90 and 100 cm. The HRT was kept at the optimal value of 3 h, C/N ratio was maintained to 6 and aeration rate to 65 L/h. The fine-grained particles (1–3 mm) showed improved performance as compared to medium and fine-grained particles at a depth of 80 cm. Fig. 5a–c show the percentage removal of the various process parameters at varying filter depths for different particle sizes.

For particle size of 5–10 mm, the COD, BOD, NH₃-N, turbidity and TDS were obtained as 86%, 80%, 90%, 88% and 29%, respectively. For 3–5 mm particle sizes the recorded values were 87%, 87%, 92%, 90% and 42%. The NZBAF with fine grains of 1–3 mm showed maximum removal efficiency of 93%, 94%, 98%, 96% and 56%, respectively for COD, BOD, NH₃-N, turbidity and TDS, respectively.

It was observed with the increase in the depth of filter the performance of the NZBAF increased but increase beyond 80 cm reduced the effectiveness of the performance.

4.1.5 Particle size

The particle size comparison on the performance of NZBAF depending on the optimal value of HRT, C/N ratio, aeration rate and depth of filter are shown in Table 4 and Fig. 6. The results show that performance of the NZBAF improved with the decrease in the particle size of the media. The removal of COD, BOD, NH₃-N turbidity and TDS was at the highest value when the NZBAF contained the natural zeolite media of particle size 1–3 mm as compared to coarser particle sizes of 5–10 and 3–5 mm.

4.2. Optimization of the operational condition using CCD and RSM

The effects of the four significant independent variables or design variables viz., HRT, C/N ratio, aeration rate and depth of filter were analysed using Minitab 19 software and its CCD to build a RSM. To explain the performance of the NZBAF depending on the removal of COD, BOD, NH₃-N, turbidity and TDS in percentages for the various particle sizes, the CCD with four-factors consisting of 31 experimental runs was employed.

4.2.1. Analysis of variance results

For BAF media using natural zeolite of particle sizes 1–3 mm, the results were analysed and interpreted using

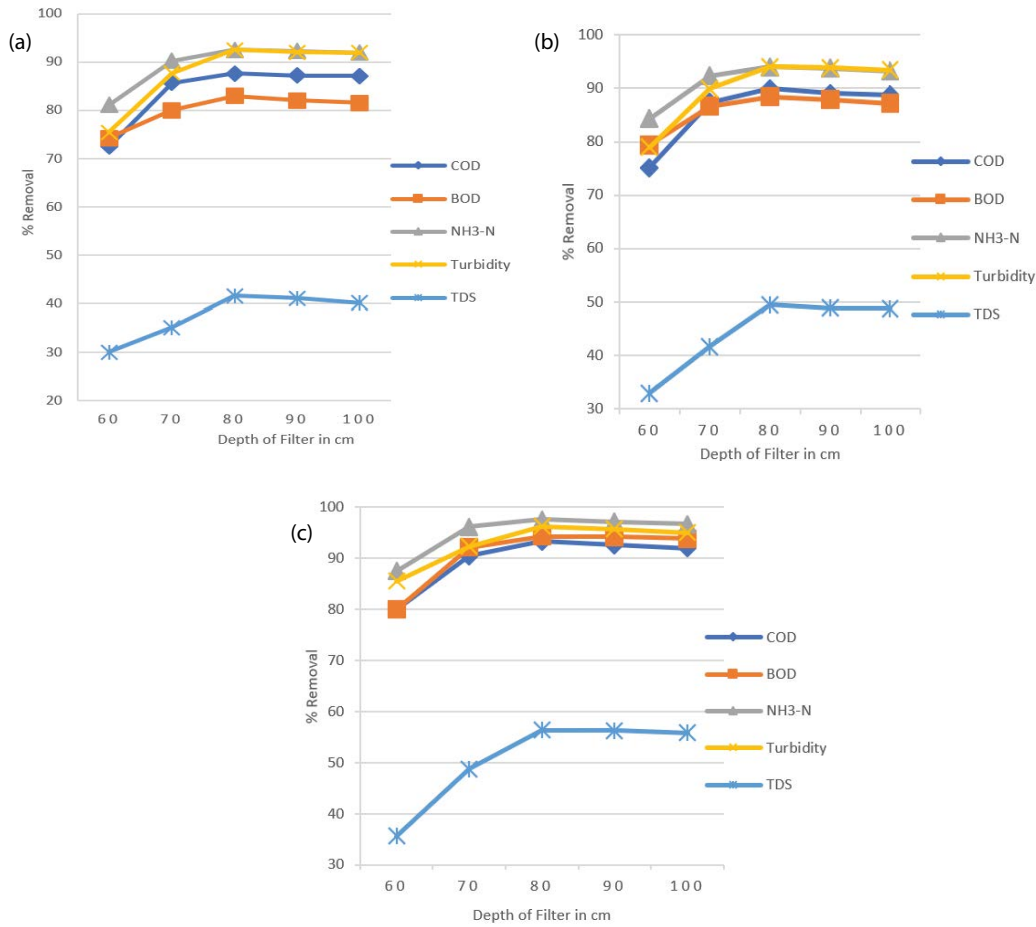


Fig. 5. Effect of varying depth of filter on (a) 5–10 mm, (b) 3–5 mm and (c) 1–3 mm.

Table 4

Particle size comparison for the removal of chemical oxygen demand, biological oxygen demand, NH₃-N, turbidity and total dissolved solids at optimum values of hydraulic retention time, C/N ratio, aeration rate and depth of filter

| Particle size | Chemical oxygen demand | Biological oxygen demand | NH ₃ -N | Turbidity | Total dissolved solids |
|---------------|------------------------|--------------------------|--------------------|-----------|------------------------|
| 10–5 mm | 88% | 83% | 92% | 92% | 42% |
| 5–3 mm | 90% | 88% | 94% | 94% | 49% |
| 3–1 mm | 93% | 94% | 98% | 96% | 56% |

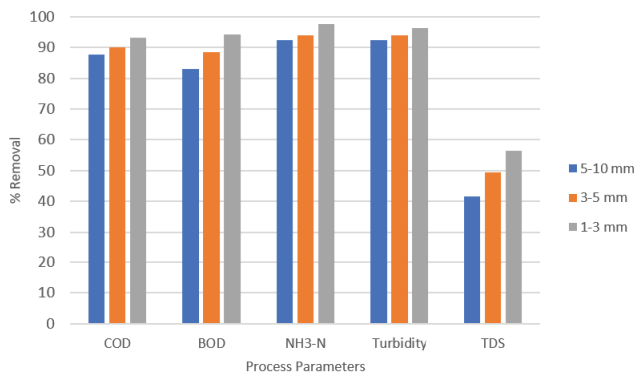


Fig. 6. Performance of NZBAF based on particle sizes.

analysis of variance (ANOVA) and the results are as shown in Fig. 7a–e for COD, BOD, NH₃-N, turbidity and TDS removal, respectively.

The tabulated variables such as the *p*-value illustrates the significance of the model in statistical terms. The ANOVA tables for all process variables indicate that the *p*-value for the model is less than the level of significance of $\alpha = 5\%$ or 0.05 (i.e., 0.000 for COD removal, 0.003 for BOD removal, 0.000 for NH₃-N removal, 0.007 for turbidity removal and 0.000 for TDS removal).

In the ANOVA for COD removal (Fig. 7a) the *p*-value for HRT, aeration rate and depth of filter are 0.000, 0.001, 0.033, respectively which are below 0.050 and shows their significance on the model. For BOD removal (Fig. 7b) the *p*-value for HRT, aeration rate and depth of filter are 0.000,

Analysis of Variance

| Source | DF | Adj SS | Adj MS | F-Value | P-Value |
|-------------------|----|---------|---------|---------|---------|
| Model | 14 | 3255.74 | 232.55 | 12.96 | 0.000 |
| Linear | 4 | 1493.19 | 373.30 | 20.80 | 0.000 |
| C/N | 1 | 4.28 | 4.28 | 0.24 | 0.632 |
| HRT | 1 | 1119.57 | 1119.57 | 62.37 | 0.000 |
| Aeration | 1 | 271.49 | 271.49 | 15.12 | 0.001 |
| DOF | 1 | 97.85 | 97.85 | 5.45 | 0.033 |
| Square | 4 | 1762.15 | 440.54 | 24.54 | 0.000 |
| C/N*C/N | 1 | 71.46 | 71.46 | 3.98 | 0.063 |
| HRT*HRT | 1 | 1238.88 | 1238.88 | 69.02 | 0.000 |
| Aeration*Aeration | 1 | 676.00 | 676.00 | 37.66 | 0.000 |
| DOF*DOF | 1 | 99.95 | 99.95 | 5.57 | 0.031 |
| 2-Way Interaction | 6 | 0.40 | 0.07 | 0.00 | 1.000 |
| C/N*HRT | 1 | 0.31 | 0.31 | 0.02 | 0.897 |
| C/N*Aeration | 1 | 0.02 | 0.02 | 0.00 | 0.971 |
| C/N*DOF | 1 | 0.00 | 0.00 | 0.00 | 1.000 |
| HRT*Aeration | 1 | 0.07 | 0.07 | 0.00 | 0.953 |
| HRT*DOF | 1 | 0.00 | 0.00 | 0.00 | 1.000 |
| Aeration*DOF | 1 | 0.00 | 0.00 | 0.00 | 1.000 |
| Error | 16 | 287.21 | 17.95 | | |
| Lack-of-Fit | 10 | 287.21 | 28.72 | * | * |
| Pure Error | 6 | 0.00 | 0.00 | | |
| Total | 30 | 3542.94 | | | |

(a)

Analysis of Variance

| Source | DF | Adj SS | Adj MS | F-Value | P-Value |
|-----------------------------|----|---------|---------|---------|---------|
| Model | 14 | 1466.70 | 104.764 | 10.37 | 0.000 |
| Linear | 4 | 573.21 | 143.302 | 14.19 | 0.000 |
| C/N Ratio | 1 | 3.88 | 3.880 | 0.38 | 0.544 |
| HRT | 1 | 398.29 | 398.291 | 39.44 | 0.000 |
| Aeration Rate | 1 | 63.21 | 63.213 | 6.26 | 0.024 |
| DOF | 1 | 107.82 | 107.823 | 10.68 | 0.005 |
| Square | 4 | 870.91 | 217.727 | 21.56 | 0.000 |
| C/N Ratio*C/N Ratio | 1 | 102.32 | 102.316 | 10.13 | 0.006 |
| HRT*HRT | 1 | 495.24 | 495.238 | 49.04 | 0.000 |
| Aeration Rate*Aeration Rate | 1 | 353.12 | 353.118 | 34.97 | 0.000 |
| DOF*DOF | 1 | 153.13 | 153.127 | 15.16 | 0.001 |
| 2-Way Interaction | 6 | 22.58 | 3.764 | 0.37 | 0.886 |
| C/N Ratio*HRT | 1 | 0.28 | 0.278 | 0.03 | 0.870 |
| C/N Ratio*Aeration Rate | 1 | 0.06 | 0.061 | 0.01 | 0.939 |
| C/N Ratio*DOF | 1 | 0.06 | 0.064 | 0.01 | 0.938 |
| HRT*Aeration Rate | 1 | 7.58 | 7.576 | 0.75 | 0.399 |
| HRT*DOF | 1 | 14.04 | 14.044 | 1.39 | 0.256 |
| Aeration Rate*DOF | 1 | 0.56 | 0.559 | 0.06 | 0.817 |
| Error | 16 | 161.58 | 10.099 | | |
| Lack-of-Fit | 10 | 161.58 | 16.158 | * | * |
| Pure Error | 6 | 0.00 | 0.000 | | |
| Total | 30 | 1628.28 | | | |

(b)

Analysis of Variance

| Source | DF | Adj SS | Adj MS | F-Value | P-Value |
|-----------------------------|----|---------|---------|---------|---------|
| Model | 14 | 2258.12 | 161.294 | 6.69 | 0.000 |
| Linear | 4 | 1050.20 | 262.550 | 10.89 | 0.000 |
| C/N Ratio | 1 | 1.59 | 1.591 | 0.07 | 0.801 |
| HRT | 1 | 813.17 | 813.170 | 33.73 | 0.000 |
| Aeration Rate | 1 | 115.19 | 115.194 | 4.78 | 0.044 |
| DOF | 1 | 120.24 | 120.243 | 4.99 | 0.040 |
| Square | 4 | 1206.86 | 301.716 | 12.51 | 0.000 |
| C/N Ratio*C/N Ratio | 1 | 80.77 | 80.769 | 3.35 | 0.086 |
| HRT*HRT | 1 | 615.16 | 615.157 | 25.52 | 0.000 |
| Aeration Rate*Aeration Rate | 1 | 478.50 | 478.499 | 19.85 | 0.000 |
| DOF*DOF | 1 | 350.17 | 350.173 | 14.52 | 0.002 |
| 2-Way Interaction | 6 | 1.06 | 0.177 | 0.01 | 1.000 |
| C/N Ratio*HRT | 1 | 1.06 | 1.061 | 0.04 | 0.836 |
| C/N Ratio*Aeration Rate | 1 | 0.00 | 0.000 | 0.00 | 1.000 |
| C/N Ratio*DOF | 1 | 0.00 | 0.000 | 0.00 | 1.000 |
| HRT*Aeration Rate | 1 | 0.00 | 0.000 | 0.00 | 1.000 |
| HRT*DOF | 1 | 0.00 | 0.000 | 0.00 | 1.000 |
| Aeration Rate*DOF | 1 | 0.00 | 0.000 | 0.00 | 1.000 |
| Error | 16 | 385.75 | 24.109 | | |
| Lack-of-Fit | 10 | 385.75 | 38.575 | * | * |
| Pure Error | 6 | 0.00 | 0.000 | | |
| Total | 30 | 2643.87 | | | |

(c)

Analysis of Variance

| Source | DF | Adj SS | Adj MS | F-Value | P-Value |
|-----------------------------|----|---------|---------|---------|---------|
| Model | 14 | 2825.07 | 201.79 | 7.27 | 0.000 |
| Linear | 4 | 658.55 | 164.64 | 5.93 | 0.004 |
| C/N Ratio | 1 | 38.56 | 38.56 | 1.39 | 0.256 |
| HRT | 1 | 147.61 | 147.61 | 5.32 | 0.035 |
| Aeration Rate | 1 | 288.01 | 288.01 | 10.37 | 0.005 |
| DOF | 1 | 184.37 | 184.37 | 6.64 | 0.020 |
| Square | 4 | 2154.68 | 538.67 | 19.40 | 0.000 |
| C/N Ratio*C/N Ratio | 1 | 328.22 | 328.22 | 11.82 | 0.003 |
| HRT*HRT | 1 | 1398.86 | 1398.86 | 50.37 | 0.000 |
| Aeration Rate*Aeration Rate | 1 | 686.70 | 686.70 | 24.73 | 0.000 |
| DOF*DOF | 1 | 305.15 | 305.15 | 10.99 | 0.004 |
| 2-Way Interaction | 6 | 11.83 | 1.97 | 0.07 | 0.998 |
| C/N Ratio*HRT | 1 | 7.02 | 7.02 | 0.25 | 0.622 |
| C/N Ratio*Aeration Rate | 1 | 0.56 | 0.56 | 0.02 | 0.889 |
| C/N Ratio*DOF | 1 | 0.56 | 0.56 | 0.02 | 0.889 |
| HRT*Aeration Rate | 1 | 3.06 | 3.06 | 0.11 | 0.744 |
| HRT*DOF | 1 | 0.06 | 0.06 | 0.00 | 0.963 |
| Aeration Rate*DOF | 1 | 0.56 | 0.56 | 0.02 | 0.889 |
| Error | 16 | 444.32 | 27.77 | | |
| Lack-of-Fit | 10 | 444.32 | 44.43 | * | * |
| Pure Error | 6 | 0.00 | 0.00 | | |
| Total | 30 | 3269.38 | | | |

(d)

Analysis of Variance

| Source | DF | Adj SS | Adj MS | F-Value | P-Value |
|-----------------------------|----|---------|---------|---------|---------|
| Model | 14 | 4835.73 | 345.41 | 7.11 | 0.000 |
| Linear | 4 | 741.53 | 185.38 | 3.82 | 0.023 |
| C/N Ratio | 1 | 73.99 | 73.99 | 1.52 | 0.235 |
| HRT | 1 | 350.06 | 350.06 | 7.20 | 0.016 |
| Aeration Rate | 1 | 113.19 | 113.19 | 2.33 | 0.146 |
| DOF | 1 | 204.28 | 204.28 | 4.20 | 0.057 |
| Square | 4 | 4066.64 | 1016.66 | 20.92 | 0.000 |
| C/N Ratio*C/N Ratio | 1 | 1017.26 | 1017.26 | 20.93 | 0.000 |
| HRT*HRT | 1 | 2355.99 | 2355.99 | 48.48 | 0.000 |
| Aeration Rate*Aeration Rate | 1 | 1153.28 | 1153.28 | 23.73 | 0.000 |
| DOF*DOF | 1 | 712.93 | 712.93 | 14.67 | 0.001 |
| 2-Way Interaction | 6 | 27.57 | 4.59 | 0.09 | 0.996 |
| C/N Ratio*HRT | 1 | 23.67 | 23.67 | 0.49 | 0.495 |
| C/N Ratio*Aeration Rate | 1 | 0.22 | 0.22 | 0.00 | 0.948 |
| C/N Ratio*DOF | 1 | 0.25 | 0.25 | 0.01 | 0.944 |
| HRT*Aeration Rate | 1 | 0.18 | 0.18 | 0.00 | 0.952 |
| HRT*DOF | 1 | 1.00 | 1.00 | 0.02 | 0.888 |
| Aeration Rate*DOF | 1 | 2.25 | 2.25 | 0.05 | 0.832 |
| Error | 16 | 777.48 | 48.59 | | |
| Lack-of-Fit | 10 | 777.48 | 77.75 | * | * |
| Pure Error | 6 | 0.00 | 0.00 | | |
| Total | 30 | 5613.21 | | | |

(e)

Fig. 7. ANOVA for (a) chemical oxygen demand, (b) biological oxygen demand, (c) NH₃-N, (d) turbidity, and (e) total dissolved solids removal.

0.024 and 0.005, respectively which denotes the significant effect of these variables. The ANOVA for $\text{NH}_3\text{-N}$ removal (Fig. 7c) has p -value of 0.000, 0.0440, and 0.0400 for HRT, aeration rate and depth of filter, respectively. For removal of turbidity the ANOVA table (Fig. 7d) denotes p -value as 0.035, 0.005 and 0.020 for HRT, aeration rate and depth of filter, respectively and the ANOVA for TDS removal (Fig. 7e) has p -value of 0.016 for HRT. The above p -values implied the significant effect of HRT, aeration rate and depth of filter on the model which indicates that they are the most important factors which significantly influence the percentage removal. The effect of C/N ratio was found to be less important for the removal of contaminants from the wastewater.

The model summary with the R^2 and R^2 adjusted values for COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS removal are shown in Fig. 8a–e, respectively. The coefficient of determination or R^2 value is a statistical tool to evaluate the model accuracy. The higher the R^2 value, the better the model fits the data. A higher R^2 value, that is, 91.89% and 90.08% was achieved for COD and BOD removal respectively and a good R^2 value of 85.41% for $\text{NH}_3\text{-N}$ removal, 86.41% for turbidity removal and 86.15% for TDS removal was obtained. This denotes that the model is a good fit to describe the variation in the percentage removal as a function of the independent variables viz. HRT, C/N ratio, aeration rate and depth of filter.

The responses of RSM were also obtained in the form of contour plots based on the experimental values. The effect of two variables at a time on the responses were also evaluated, that is, C/N ratio vs. depth of filter, aeration rate vs. C/N ratio and depth of filter vs. aeration rate, HRT vs. C/N ratio, aeration vs. HRT and depth of filter vs. HRT and are as shown in Fig. 9a–e, respectively.

4.2.2. Residual analysis

The adequacy of the regression model was examined based on residual plots. Fig. 10a–e shows the residual plots for the percentage removal of COD, BOD, $\text{NH}_3\text{-N}$, turbidity

and TDS, respectively. It shows that almost all data points are normally scattered close to the straight line for COD, BOD, $\text{NH}_3\text{-N}$, turbidity and TDS removals.

5. Optimized condition validation

Applying defined information in final step of optimization, experimental data were compared to theoretical expected values. At optimal conditions, additional experiments were performed to compare the predicted values with experimental results. Based on the optimum conditions obtained from RSM, a final run was conducted with the optimum value of HRT = 3.25 h, C/N = 6.2, aeration rate = 69 L/h, depth of filter = 83 cm. Table 5 and Fig. 11. show the percentage removals of the various process parameters.

6. Discussion

Experiments were conducted using natural zeolite-based BAF technology under laboratory conditions to evaluate the effect of size of media using natural zeolite. As far as particle size is considered, the performance of the biofilter with finer grained natural zeolite of particle size 1–3 mm was superior to that of medium grained (3–5 mm) and coarse grained (10–5 mm) particles.

6.1. Effect of HRT

The efficiency in the removal of pollutants increased when the value of HRT was increased from 1 to 3 h. But the removal was not substantial beyond it. So, optimum HRT was fixed as 3 h. Organic matter removal was at a faster rate initially due to availability of sufficient absorbent sites. Later, due to decrease in active sites, the removal rate slowly decreased [20]. Shorter HRT means smaller footprint, which can save the construction costs of BAFs.

6.2. Effect of C/N ratio

When the C/N ratio was varied from 4 to 6, the percentage removal of the different process parameters increased

Model Summary

| S | R-sq | R-sq(adj) | R-sq(pred) |
|---------|--------|-----------|------------|
| 4.23678 | 91.89% | 84.80% | 53.31% |

(a)

Model Summary

| S | R-sq | R-sq(adj) | R-sq(pred) |
|---------|--------|-----------|------------|
| 4.91013 | 85.41% | 72.64% | 15.96% |

(c)

Model Summary

| S | R-sq | R-sq(adj) | R-sq(pred) |
|---------|--------|-----------|------------|
| 3.17787 | 90.08% | 81.39% | 42.84% |

(b)

Model Summary

| S | R-sq | R-sq(adj) | R-sq(pred) |
|---------|--------|-----------|------------|
| 5.26970 | 86.41% | 74.52% | 21.72% |

(d)

Model Summary

| S | R-sq | R-sq(adj) | R-sq(pred) |
|---------|--------|-----------|------------|
| 6.97082 | 86.15% | 74.03% | 20.22% |

(e)

Fig. 8. Model summary for (a) chemical oxygen demand, (b) biological oxygen demand, (c) $\text{NH}_3\text{-N}$, (d) turbidity, and (e) total dissolved solids removal.

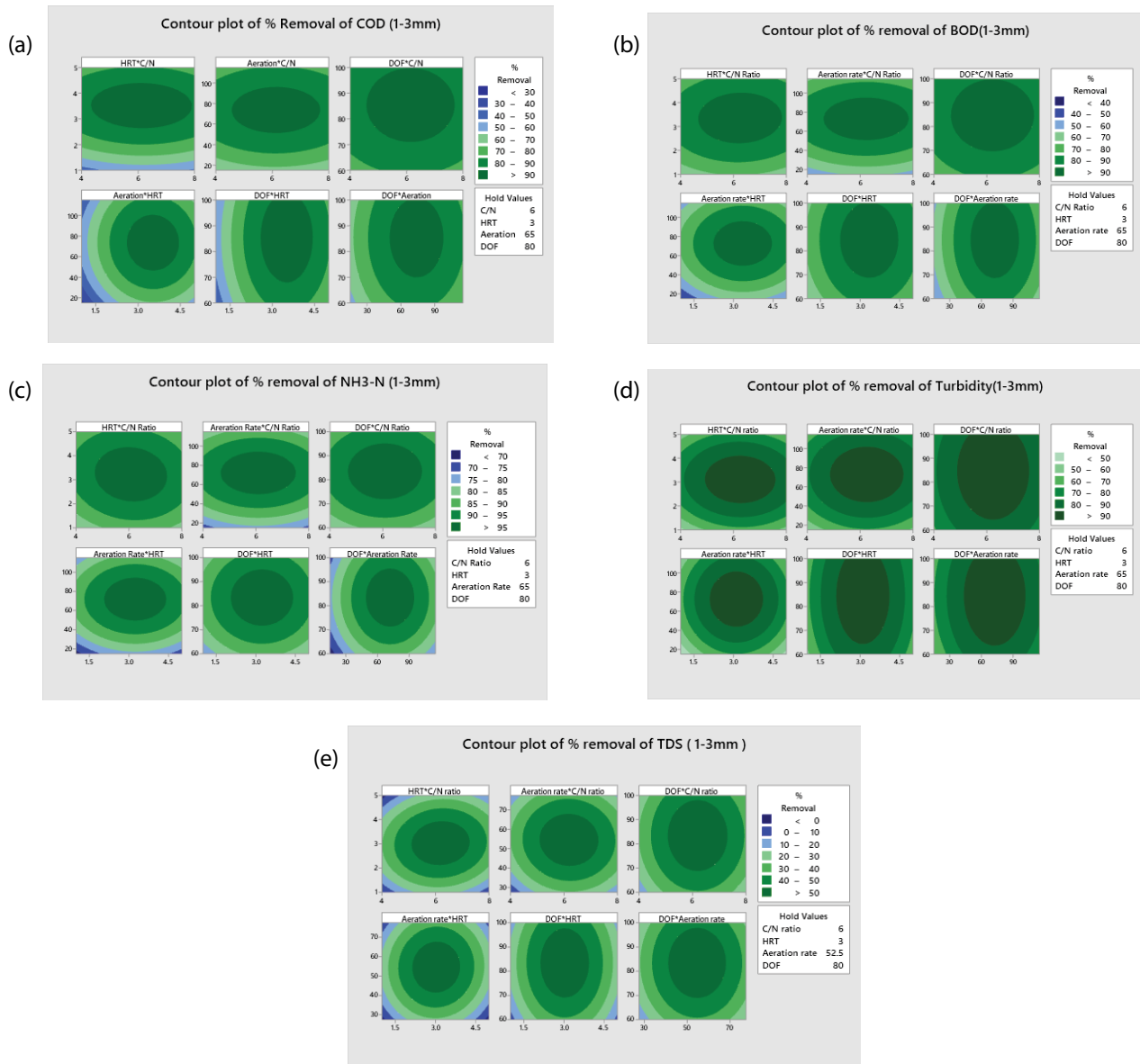


Fig. 9. Contour plot for percentage removal of (a) chemical oxygen demand, (b) biological oxygen demand, (c) NH₃-N, (d) turbidity and (e) total dissolved solids.

but was found to decrease beyond C/N ratio of 6. So, the optimum C/N ratio was fixed as 6. The efficiency in the removal of the carbonaceous and nitrogenous organic matter was due to its good contact with the bacterial biofilm which in turn is due to its porous nature and large surface area [20]. Performance of biofilter decreases with increase of organic loading rate beyond a limit because at higher organic loading rate the activity of the bacteria is prohibited significantly.

6.3. Effect of aeration

All the parameters showed better percentage removal when aeration was provided at the rate of 65 L/h and increasing the rate of aeration to 90 and 115 L/h did not show any appreciable improvement in performance. So,

rate of aeration was fixed to 65 L/h. The bacterial activity increased when sufficient oxygen was available as result of aeration provided to the system.

6.4. Effect of depth of filter

The percentage removal for all the parameters increased with the increase in the depth of filter from 60 to 80 cm. But the percentage removal was not appreciable beyond 80 cm depth of filter. Performance of the biofilter after some time in operation deteriorates with further increase in depth of the media, and accumulates biomass. The pressure built-up in the bed due to the accumulation of organic matter causes uneven distribution of influent and air which ultimately reduces the biological activity of bacteria and efficiency of the BAF [21].

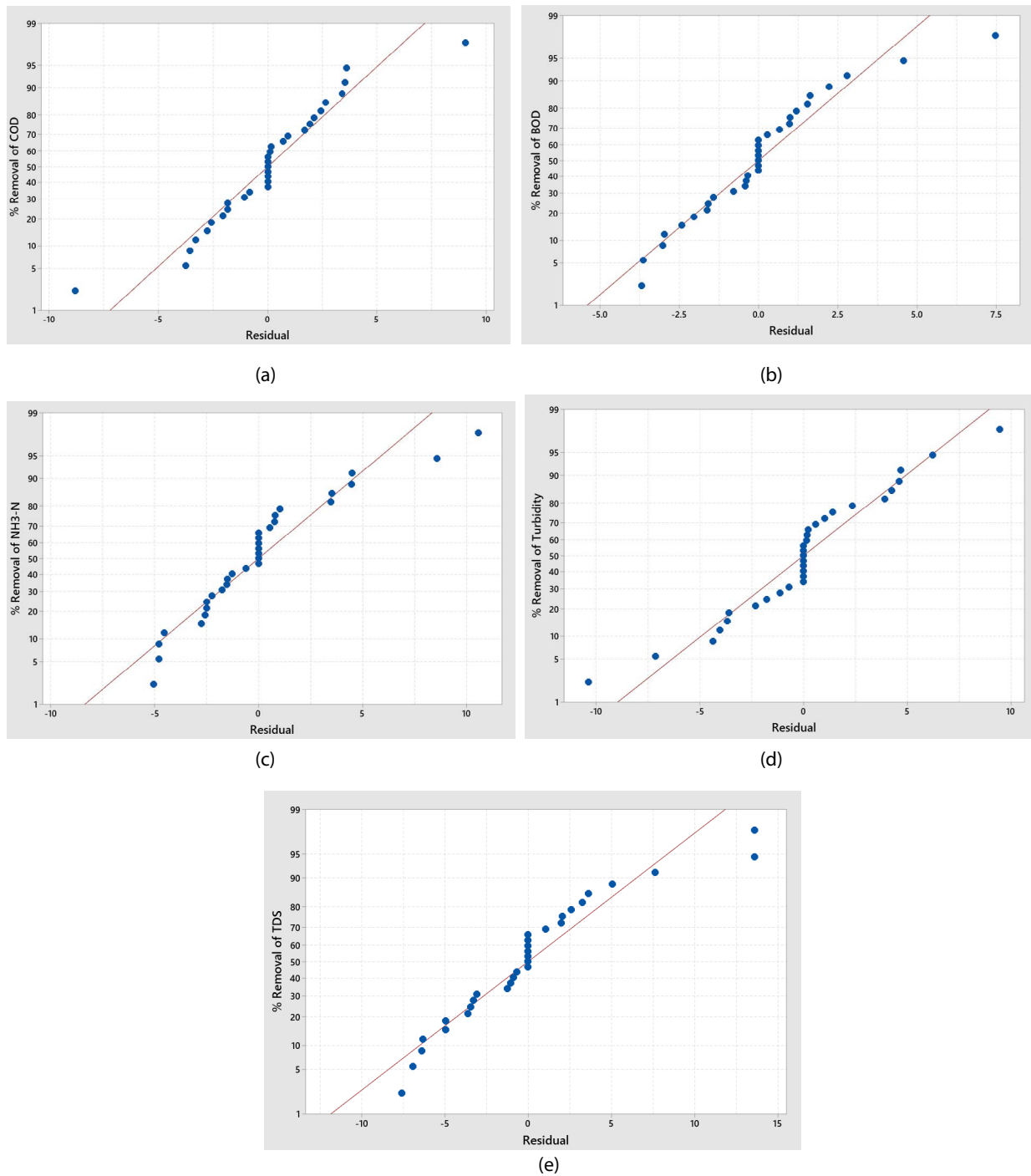


Fig. 10. Residual plot for (a) chemical oxygen demand, (b) biological oxygen demand, (c) NH₃-N, (d) turbidity, and (e) total dissolved solids removal.

Table 5
Comparison of predicted values and experimental results for 1–3 mm particles

| Method | Chemical oxygen demand | Biological oxygen demand | NH ₃ -N | Turbidity | Total dissolved solids |
|----------------------------------|------------------------|--------------------------|--------------------|-----------|------------------------|
| Theoretically expected value (%) | 96 | 96 | 98 | 97 | 57 |
| Experimental value (%) | 97 | 96 | 99 | 98 | 58 |

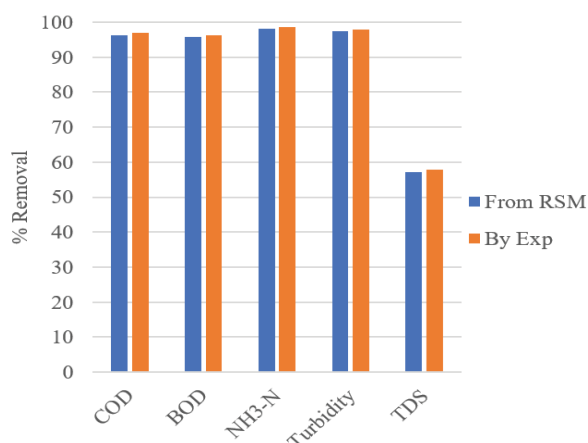


Fig. 11. Percentage comparison of theoretically expected values and experimental values for removal of parameters for zeolite particle size of 1–3 mm.

6.5. Effect of particle size

The NZBAF with fine grain sized particles (1–3 mm) showed good results compared to medium grain sized particles (3–5 mm) and coarse grain sized particles (5–10 mm). Finer particles have more sites for ion exchange, more specific surface area for adsorption and for bio-film formation by bacteria. They also remove minute solids from wastewater by mechanisms like straining, sedimentation, interception etc. It thus helps to reduce the volume of BAF required but the frequency of backwash increases [22]. Thus, BAF with fine grains as support media showed optimal performance under optimal conditions of HRT = 3 h, C/N ratio = 6, aeration rate = 65 L/h and filter depth = 80 cm.

During optimization, the results of the ANOVA using the RSM tool of Minitab 19 showed a lower *p*-value for the independent variables HRT, aeration rate and depth of filter while a higher *p*-value was observed for the variable C/N ratio. This denotes that HRT, aeration rate and depth of filter has significant impact on the removal of COD, BOD, NH₃-N, turbidity and TDS whereas the impact of C/N ratio was comparatively less significant.

After the optimization process using RSM and CCD, a final run was conducted on the NZBAF using natural zeolite of 1–3 mm. It was operated at optimum value of HRT = 3.25 h, C/N = 6.2, aeration rate = 69 L/h, depth of filter = 83 cm and the performance in terms percentage removal of COD, BOD, NH₃-N, turbidity and TDS was recorded as 97%, 96%, 99%, 98% and 58%, respectively.

7. Conclusion

The overall removal efficiency of contaminants from the wastewater in the NZBAF system was affected by the particle size of the natural zeolite media, HRT, C/N ratio, aeration rate and depth of filter. The smaller the media size, greater was the surface area per unit volume available for adsorption, ion exchange and biofilm development which in turn reduces the size of the BAF. The zeolite media with finer particle size (1–3 mm) gave better results than the coarser particles (3–5 mm and 5–10 mm). The optimization study of

the process variables, that is, HRT, C/N ratio, aeration rate and depth of filter was conducted using RSM for optimizing the removal of COD, BOD, NH₃-N, turbidity and TDS and to understand the overall performance of NZBAF system. It is a powerful tool which illustrates the output behaviour of the responses in the experiment and analyses it to interpret the level of optimization of the four process variables. It was observed that HRT, aeration rate and depth of filter had significant effect on the percentage removal as compared to C/N ratio. Finally, an experiment was conducted using natural zeolite of 1–3 mm with optimized values of process variables, that is, HRT = 3.25 h, C/N = 6.2, aeration rate = 69 L/h, depth of filter = 83 cm and the percentage removal achieved was 97% for COD, 96% for BOD, 99% for NH₃-N, 98% for turbidity and 58% for TDS. Thus, the study suggests NZBAF with natural zeolite of particle size 1–3 mm as media is a potential solution to the wastewater treatment. The substantial enhancements in performance in terms of the organic matter and nutrient removal under the optimum conditions was possible mainly due to processes like ion exchange, adsorption, bacterial adherence, etc.

References

- [1] L. Mendoza-Espinosa, T. Stephenson, A review of biological aerated filters (BAFs) for wastewater treatment, *Environ. Eng. Sci.*, 16 (1999) 201–216.
- [2] A.T. Mann, T. Stephenson, Modelling biological aerated filters for wastewater treatment, *Water Res.*, 31 (1997) 2443–2448.
- [3] B.K. Pramanik, S. Fatihah, Z. Shahrom, E. Ahmed, Biological aerated filters (BAFs) for carbon and nitrogen removal: a review, *J. Eng. Sci. Technol.*, 7 (2012) 428–446.
- [4] J.H. Ha, S.K. Ong, R. Surampalli, Impact of media type and various operating parameters on nitrification in polishing biological aerated filters, *Environ. Eng. Res.*, 15 (2010) 79–84.
- [5] W.S. Chang, S.W. Hong, J. Park, Effect of zeolite media for the treatment of textile wastewater in a biological aerated filter, *Process Biochem.*, 37 (2002) 693–698.
- [6] D.M. Leles, D.A. Lemos, U.C. Filho, L.L. Romanielo, M.M. de Resende, V.L. Cardoso, Evaluation of the bioremoval of Cr(VI) and TOC in biofilters under continuous operation using response surface methodology, *Biodegradation*, 23 (2012) 441–454.
- [7] S. Wang, Y. Peng, Natural zeolites as effective adsorbents in water and wastewater treatment, *Chem. Eng. J.*, 156 (2010) 11–24.
- [8] N. Karapinar, Application of natural zeolite for phosphorus and ammonium removal from aqueous solutions, *J. Hazard. Mater.*, 170 (2009) 1186–1191.
- [9] H. Tan, An Evaluation of Biological Aerated Filtration for Wastewater Treatment Through Pilot and Laboratory Scale Experiments, Master's Thesis, Queen's University, Kingston, ON, Canada, 2008.
- [10] N. Widiastuti, H. Wu, M. Ang, D.K. Zhang, The potential application of natural zeolite for greywater treatment, *Desalination*, 218 (2008) 271–280.
- [11] M. Li, X. Zhu, F. Zhu, G. Ren, G. Cao, L. Song, Application of modified zeolite for ammonium removal from drinking water, *Desalination*, 271 (2011) 295–300.
- [12] A.B. Robinson, W.J. Brignal, A.J. Smith, Construction and operation of a submerged aerated filter sewage-treatment works, *Water Environ. J.*, 8 (1994) 215–227.
- [13] W. Xie, Q. Wang, G. Song, M. Kondo, M. Teraoka, Y. Ohsumi, H.I. Ogawa, Upflow biological filtration with floating filter media, *Process Biochem.*, 39 (2004) 767–772.
- [14] Z. Ke, Z. Gang, Z. Yu, K. Zheng, Study on backwash of biological aerated filter, *Appl. Mech. Mater.*, 448 (2014) 429–433.

- [15] G. Bacquet, J.C. Joret, F. Rogalla, M.M. Bourbigot, Biofilm start-up and control in aerated biofilter, *Environ. Technol.*, 12 (1991) 747–756.
- [16] A.B. Jasni, H. Kamyab, S. Chelliapan, N. Arumugam, S. Krishnan, M.F.M. Din, Treatment of wastewater using response surface methodology: a brief review, *Chem. Eng. Trans.*, 78 (2020) 535–540.
- [17] C.W. Lin, C.H. Yen, H.C. Lin, D.T. Tran, Response surface optimization of dissolved oxygen and nitrogen sources for the biodegradation of MTBE and BTEX, *Biodegradation*, 21 (2010) 393–401.
- [18] A. Mojiri, L. Ziyang, R.M. Tajuddin, H. Farraji, N. Alifar, Co-treatment of landfill leachate and municipal wastewater using the ZELIAC/zeolite constructed wetland system, *J. Environ. Manage.*, 166 (2016) 124–130.
- [19] APHA (American Public Health Association), American Water Works Association, and Water Pollution Control Federation, *Standard Methods for the Examination of Water and Wastewater*, 13th ed., American Public Health Association, New York, 1971.
- [20] N. Widiastuti, H. Wu, H.M. Ang, D. Zhang, Removal of ammonium from greywater using natural zeolite, *Desalination*, 277 (2011) 15–23.
- [21] N. Qamaruz-Zaman, N. Yaacof, F.F. Kamarzaman, Chapter 9 – Control of Odors in the Food Industry, C. Galanakis, Ed., *The Interaction of Food Industry and Environment*, Academic Press, Cambridge, 2020, pp. 281–313.
- [22] R. Moore, J. Quarmby, T. Stephenson, The effects of media size on the performance of biological aerated filters, *Water Res.*, 35 (2001) 2514–2522.