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Removal of formaldehyde from polluted air in a biotrickling filter reactor

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ABSTRACT

Optimization of biotrickling filter reactor (BTFR) for the removal of formaldehyde contained in synthetic contaminated air was investigated. The importance of formaldehyde from contaminated air is necessary mainly because it is toxic, carcinogen, and mutagen for humans. Although several studies have been conducted on formaldehyde removal by using BTFR from contaminated air, but optimum conditions for formaldehyde removal is not being studied using a trustable method. The determination of optimum condition to remove formaldehyde was carried out with Taguchi experimental design method. The influence of different factors (pH, nitrogen, phosphorus, formaldehyde concentration) on formaldehyde removal efficiency in BTFR was determined, and the optimum condition for maximizing this response was obtained. The result shows that pH has a greater effect on BTFR efficiency for formaldehyde removal. By operating BTFR in optimum condition, the removal rate increased up to 98%. Thus, the operation of BTFR at optimum condition to remove formaldehyde is considered to be essential in minimizing the pollutants present in the atmosphere.

Keywords: Formaldehyde removal; Contaminated air; Biofilter; Taguchi method

1. Introduction

Rapid development of industries has led to the releasing of large volumes of pollutant effluents directly into the atmosphere. The treatment of air contaminated with toxic compounds resulting from these effluents is of the most importance challenge faced by the researchers. Formaldehyde is a common compound found widely during industrial processes. Due to high consumption of formaldehyde in industries, it is frequently found in the form of contaminated gases [1]. Due to toxicity of formaldehyde to micro-organisms, biological activities can be partly inhibited. Formaldehyde is known as a biodegradable compound, and it can be removed using aerobic [2–4] and anaerobic reactors [5–8]. The last two decades witnessed a significant increase in the installation of biotrickling filter reactors (BTFRs). Although BTFRs have been applied to treat gases contaminated with

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odorous components, these days, they are frequently applied to remove organic pollutants such as volatile organic compounds from contaminated air [9,10]. Nowadays, traditional physical and chemical gas treatment technologies are being replaced with BTFRs as they are more reliable and provide cost-effective technology [11].

The most important subject in biodegradation processes is to optimize the process of achieving maximum removal of pollutants by changing environmental conditions. Experimental design approaches, including the Taguchi method, have found wide applications in determining the effect of factors on the process responses and also to find the optimum conditions of biodegradation processes for organic pollutants [7]. A large number of experiments have to be carried out to optimize processes when classical experimental design methods are applied. Taguchi (1990) developed a new statistical method to solve this problem and later it was called the Taguchi method [12].

The Taguchi method uses a special design of orthogonal arrays to study the entire parameter space with only a small number of experiments. In recent years, Taguchi method is applied as an optimization tool for wide range of studies such as engineering and biotechnology processes. The following three objectives can be achieved when the Taguchi method is applied (i) determination of optimal effective factors for a process or a product (ii) estimation of each effective factor to the contribution of quality characteristics (QCs) (iii) prediction of QCs based on the optimal effective factors. Although some researchers have conducted studies on formaldehyde removal from contaminated air using BTFR, but the determination of optimum condition has not yet been studied using a trustable method such as the Taguchi method. The main objective of this study is to determine the optimum conditions of formaldehyde removal in a BTFR. The nitrogen and phosphorous sources, temperature, and pH were first determined, and later, the optimal values of selected factors and their effectiveness were found experimentally in a BTFR for the removal of formaldehyde from contaminated air using Taguchi method.

2. Materials and methods

2.1. Pilot-scale bioreactor

The research was conducted with a laboratory-scale BTFR consisting of biofilter bath with polyurethane pipe as supporting material; a nutrient solution tank, formaldehyde storage tank, compressor, peristaltic pump, flow mater, control air valve, air temperature controller, and mechanical o'clock switch (Fig. 1) were also included in the biofilter reactor. Biofilter bath was about 8 cm in diameter, 66 cm in height, and 3.319 L volume. The percentage of void space in the biofilter bath equaled to 90%. The synthetic contaminated air stream that was ventilated using air-pressure generated by a compressor "Asian Star AP-1,000" was supplied from the formaldehyde storage tank to biofilter bath (Fig. 1). A flow meter of IF series-DWYER was used to monitor the air flow rate. The aerobic granular sludge was used as inoculums for the start up of the BTFR coming from a municipal wastewater treatment plant of Isfahan city, Iran. The experiment, examined the effects of pH, temperature, nitrogen, and phosphorous concentration on the performance of BTFR. In this continuous-flow culture system, the nutrient solution regularly flowed pass the biofilter bath, the mechanical o'clock switch regulated the fed nutrient solution at a flow rate of 50 L/h, and the peristaltic pump circulated the nutrient solution through the top of the biofilter bath, 15-min period for every hour.

The experiment was conducted initially to develop micro-organisms on the supporting material during the adaptation phase of 90 days and continued further to collect the data of monitoring the different nitrogen and phosphorous sources during the experimental period of 60 days and the best nitrogen and phosphorus source were recognized. Finally, to find the optimum combination of operation and environmental factors on the efficiency of formaldehyde removal,

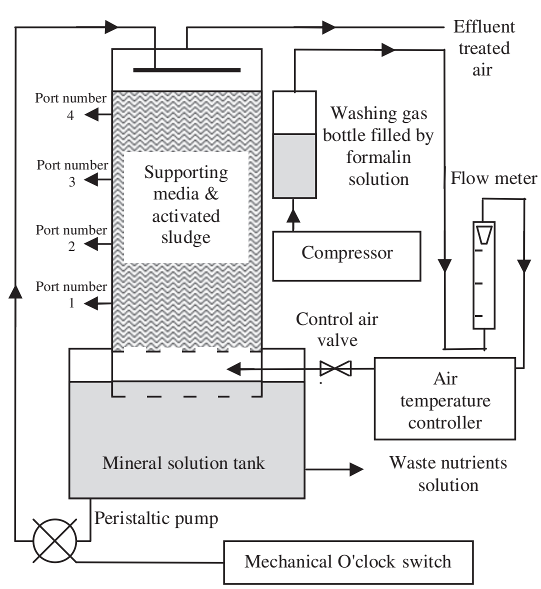

Fig. 1. Schematic model of BTFR.

BTFR was continually conducted, at different pH, temperature, nitrogen, and phosphorous concentration based on designed experiments using Taguchi method for a period of 95 days.

2.2. Culture medium and synthetic wastewater

To have an effective BTFR operation, it was important to maintain a healthy environment for the micro-organisms to thrive at the supporting material and therefore needed some food to support the growth of different microbes. The nutrient solution consisted of sufficient quantity of minerals, that is, 0.1 g/L $MgSO_4$, 0.01 g/L $CaCl_2.2H_2O$, 0.001 g/L $FeSO_4$, 1 g/L NH_4Cl, 0.5 g/L KH_2PO_4, 0.5 g/L K_2HPO_4 and 0.001 g/L $MnSO_4$ and had the function to conserve the biofilter bath in a wet environment during the experiments [13,14]. The nutrient solution was aerated continuously to ensure that dissolved oxygen (DO) was higher than 5 mg/L. The optimum pH of the nutrient solution for all the experiments was set at 7 ± 0.5.

Table 1
Selected factor and their levels

Factors	Levels Level (1)	Level (2)	Level (3)	Level (4)
pH	3	5	7	9
Nitrogen concentration (mg/L)	0.2	0.5	1	1.5
Phosphorous concentration (mg/L)	0.1	0.3	0.5	1
Temperature (℃)	22	26	40	45

2.3. Micro-organisms adaptation

During the adaption phase, a mixture of glucose and formaldehyde was added into the nutrient solution to serve as a carbon source. Starting with a low formaldehyde level, high amount of glucose was added into the nutrient solution, giving an organic matter concentration of about 1 g L^{-1} COD. An increased formaldehyde concentration decreases the glucose concentration for generation of the product of a similar organic matter fraction in the nutrient solution. After an adaptation phase of 90 days, formaldehyde as a sole carbon source was used for feeding the BTFR and then replaced by the injection of air contaminated with formaldehyde into the biofilter.

2.4. Experimental design

In this study, four environmental factors (pH, temperature, nitrogen, and phosphorous concentration) were considered as the selected factors (Table 1). The Taguchi design of experiment with four factors, a standard L_{16} orthogonal array was employed (Table 2)

Table 2
Designed experiments and resulted propylene glycol removal

Run	Factors pH	Nitrogen concentration	Phosphorus concentration	Temperature	Percentage of propylene glycol removal Replication #1 (%)	Replication #2 (%)	S/N
1	1	1	1	1	12	18	22.996
2	1	2	2	2	44	35	31.762
3	1	3	3	3	53	63	35.171
4	1	4	4	4	51	54	34.392
5	2	1	2	3	62	50	34.813
6	2	2	1	4	52	42	33.298
7	2	3	4	1	49	59	34.536
8	2	4	3	2	65	63	36.12
9	3	1	3	4	70	66	36.638
10	3	2	4	3	66	74	36.859
11	3	3	1	2	52	50	34.146
12	3	4	2	1	69	65	36.509
13	4	1	4	2	47	40	32.685
14	4	2	3	1	41	39	32.033
15	4	3	2	4	53	59	34.926
16	4	4	1	3	53	49	34.131

using Qualitek-4 (Nutek Inc.) software. L and 16 mean Latin square and the replication number of the experiment. Four–four level factors can be positioned in an L_{16} orthogonal array table. The number in table indicates the level of a factor. Each row of the matrix represents one run at a specified condition. In order to avoid the systematic bias, the sequence in which these runs were carried out was randomized [12]. In the selected coordinate system, the codes (1, 2, 3, 4) represent level (1, 2, 3, 4) based on (Table 1).

2.5. Analytical methods

The analytical methods were used in this study in order to determine the amount of pH, COD and formaldehyde concentration present in water and inlet and outlet of BTFR air. The average formaldehyde concentration in the synthetic contaminated air stream was 490 mg/L. To determine the formaldehyde concentration in the effluent BTFR, air samples were collected every day using the vacuum pump "Champion Air Pump-AAP Model." To determine formaldehyde in BTFR air, first, using scrubber formaldehyde transferred from gas phase to water phase. Second, formaldehyde was measured by the amount of chemical oxygen demand (COD). The COD determined according to the standard methods [15], and pH was measured using digital pH meter.

3. Results and discussion

3.1. Formaldehyde removal from nutrient solution

The adaptation period for the study was carried out within 90 days and the COD level of nutrient solution was monitored during this period. Almost all of the factories that use formaldehyde as one of the chemical feedstock face problems for the presence of formaldehyde in the form of waste gases and wastewater [16]. Due to this reason, finding a suitable method to treat the waste products is very important. In this experiment of formaldehyde removal from nutrient solution, injection of formaldehyde vapor into bio-filter was stopped and circulation of nutrients solution surrounding the biofilms was continuously operated. The COD of nutrient solution was monitored for 79 h. It was seen that initial COD of nutrient solution was equal to 11,900 mg/L where Fig. 2 shows the result of COD monitoring of nutrient solution. Micro-organisms in order to survive needs carbon source, the dissolved formaldehyde present in nutrient solution served as a sole carbon source of the growth of micro-organism in this study. During circulation of nutrient, solution-containing formaldehyde

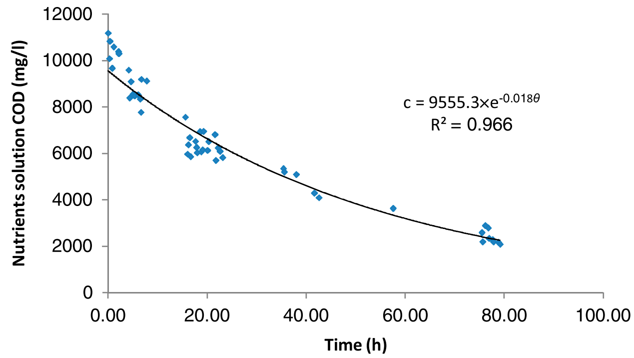

Fig. 2. Formaldehyde removal from nutrient solution in a batch system with respect to time.

surrounding biofilm micro-organisms could absorb formaldehyde from liquid phase, thereby caused decrease in the amount of COD present in nutrient solution. The time taken for the formaldehyde removal by the micro-organisms present in the nutrient solution was 79 h, and the decrease in COD observed was from 11,900 to 2,100 mg/L. The results of this study also showed that the average formaldehyde removal rate within the time period of this study (79 h) was 37.3 mg/h per each liter of BTFR reactor. Fig. 2 represents a nonlinear regression pattern between formaldehyde removal and time. This regression can be described using Eq. (1):

$$c = ae^{b\theta} \tag{1}$$

where a and b are constant coefficients and are equal to 9555.3 and 0.018. c is COD in nutrient tank and is measured in mg/L. θ is time in h and e is neper number which is equal to 2.718.

The results of this experiment proved that BTFR is applicable for formaldehyde removal from wastewater and polluted air at same time. Based on the regression equation, after 509 h, COD of nutrient solution is equal to zero which is considered to be a long period of time for it to be used in the system for the complete removal of wastewater contaminated with formaldehyde. So for this reason, BTFR can be considered suitable only for pretreating of wastewater contaminated with formaldehyde.

3.2. Optimization of nitrogen and phosphorous sources

The approach named one factor at the time to find the best nitrogen and phosphorus sources was used for $(NH_4)_2[Fe(SO_4)_2]$, NH_4Cl, NH_4NO_3, $Ca(NO_3)_2$, KNO_3 and $CaHPO_4$, $NaHPO_4$, K_2HPO_4 and KH_2PO_4

to conduct BTFR for formaldehyde removal efficiency. It can be seen in Figs. 3 and 4 the best sources of nitrogen and phosphorus determined are KH_2PO_4 and $(NH_4)_2[Fe(SO_4)_2]$. After identification of best source, the best concentration was determined during the later part of the study. The amount of nitrogen and phosphorus sources was controlled in relation to formaldehyde concentration so that the C/N/P ratio could be kept at 100:5:1 throughout the experiment [17].

In comparison with $(NH_4)_2[Fe(SO_4)_2]$ other nitrogen sources of NH_4Cl, NH_4NO_3, $Ca(NO_3)_2$ could not play the role of procreative micro-organism in the system. This however did not correspond to the results of Li et al. [18] for a different treatment system, in which NH_4Cl and $NaNO_3$ were found as suitable nitrogen source. There is no consensus among researchers in determining what type of nitrogen source is suitable for biological reactors. For instance, Nitric oxide (NO) and nitrogen dioxide (NO_2) were reported by Mancinelli and McKay [19] as the best nitrogen sources for their biological reactors.

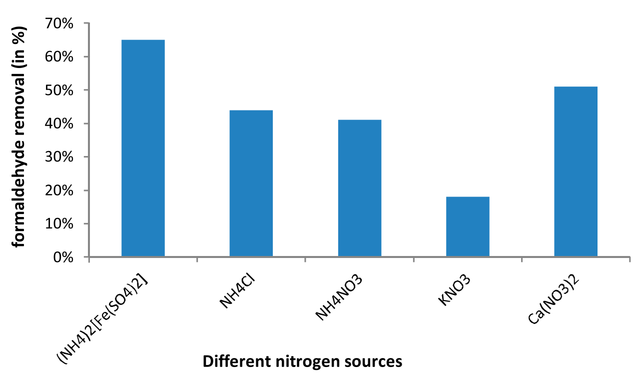

Fig. 3. Effect of using different phosphorus sources on formaldehyde removal efficiency by BTFR.

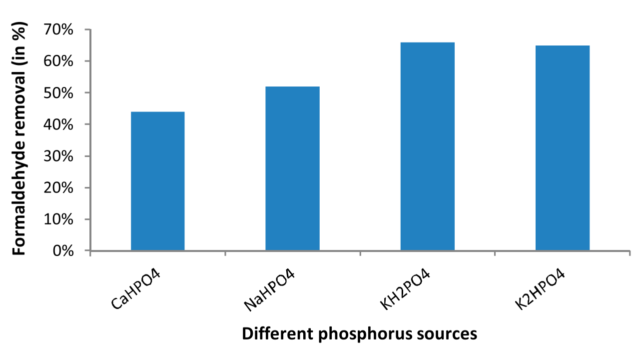

Fig. 4. Effect of using different nitrogen sources on formaldehyde removal efficiency by BTFR.

Similar condition can be found about phosphorus sources. Some researchers such as Uysal and Türkman [20], Zandvoort et al. [21] and Gigras et al. [22] successfully applied KH_2PO_4 as phosphorus source in their bio reactor. Also, mixture of KH_2PO_4 and K_2HPO_4 is reported as a suitable phosphorus source that provides an appropriate buffer conditions in bioreactors. Buffer environment can lead operators to greater degree of control on bioreactors [23].

3.3. Taguchi transformed response

The conventional approach of experimenting with one variable (or one factor) at a time is labor-intensive and time-consuming. Therefore, in this study, Taguchi method was applied to optimize formaldehyde removal using BTFR. Based on our literature review, the main operational parameters and their levels were selected and shown in Table 1. To perform the designed experiments, biotrickling filtration operation was conducted based on (Table 2) experiments was operated until a steady state was achieved. In this study, steady-state condition is defined as four consecutive days of BTFR operation and the difference among formaldehyde removal efficiency was below 4%.

Taguchi experimental design was applied in order to investigate the influence of environmental factors on formaldehyde removal by micro-organisms particularly when the micro-organism gets adapted to the system. The results of repeated formaldehyde measurements at different experimental runs are reported in Table 2.

In Taguchi method, a transformed response called signal per noise (S/N) was used for the analysis of experimental results. The signal-to-noise ratio indicates the value of changes in response to variation of controlled factors with respect to that of errors. Therefore, the higher value of S/N ratio is desirable in all QCs. "Bigger is better" was used as a QC in this study. For example, higher formaldehyde removal can achieve better BTFR efficiency. In order to carry out the statistical analysis for this QC, the S/N ratio can be calculated from Eq. (2) [12]:

$$\frac{S}{N} = -10\log\left(\frac{\frac{1}{y_1^2}+\frac{1}{y_2^2}+\ldots+\frac{1}{y_n^2}}{n}\right) \tag{2}$$

where y_i *is* the formaldehyde removal efficiency at reactor for ith experiment, and n is the number of measurements carried out for each run. The S/N ratio obtained for each experiment is shown in Table 2.

3.4. Main effects

The influences of trends for each factor on S/N (corresponding to formaldehyde removal) are discussed in the following sections and it should be noted that the interpretation of these results is valid just for the range of levels considered for the factors in this study.

3.5. Effect of pH

The pH is defined as the negative logarithm of the hydrogen ion concentration in aquatic environments which has a great impact on micro-organism growth. Estévez et al. [24] and Daneshvar et al. [19] could show environmental pH is essentially effective on micro-organisms growth. Most of the micro-organisms require neutral pH conditions for optimal growth with minimum and maximum range between 4 and 9, and it will affect the metabolism process as well as enzymatic mechanism in the cell [17]. The different strains of micro-organisms can be found on BTFR due to the utilization of activated granular sludge, and it was considered important to evaluate the effects of different pH for BTFR efficiency in formaldehyde removal from polluted air. The results showed that formaldehyde removal efficiency increased as the operating pH increased in the range of 3–7 and decreased at pH in the range of 7–9 (Fig. 5). In this study, BTFR showed best performance at neutral pH (7.0). There is no consensus among researchers on which pH is suitable for biofiltration. For instance, Brennan et al. (1996) reported that pH under 5 can exceedingly reduce biofiltration performance [25] or Estévez et al. [24] can find fungal biofilter can be notably active in pH below 3.5. Application of different micro-organism strains is the main reason of this leak of consensus on suitable pH for biofiltration processes.

3.6. Effect of nitrogen concentration

Nitrogen is undoubtedly a vital element for growth and to regulate metabolism of micro-organisms [18]. In this study four different concentrations of $(NH_4)_2[Fe(SO_4)_2]$ (0.2, 0.5, 1.0, 1.5 mg/L) were examined. Although the best nitrogen concentration in nutrient solution on formaldehyde removal was found to be 1 mg/L (Fig. 6), but increasing of nitrogen concentration more than 0.5 mg/L did not have a very significant effect on BTFR performance. Bancroft et al. [26] showed that, heterotrophic bacterial populations in a biological process have a faster growth by the presence of special nitrogen sources in special concentration. As can be seen in Fig. 6 nitrogen concentration has a direct impact on performance of BTFR. There is consensus among researchers regarding what nitrogen concentration is suitable for biological processes. The best nitrogen concentration is directly related to carbon concentration. An optimal ratio of carbon/nitrogen equal to 100/5 was suggested for biological reactors [17], whereas the optimum concentration of nitrogen source obtained during this study is near to the optimum ratio of (100/5).

3.7. Effect of temperature

The BTFR study was carried out under mesophilic condition where the temperature range was between 22 and 45℃. Micro-organisms that can withstand higher temperature called thermophilic [27]. Each micro-organism strain has its own particular range of

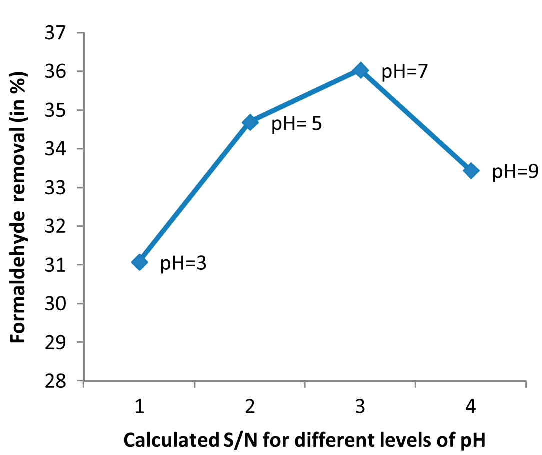

Fig. 5. Effect of pH on formaldehyde removal.

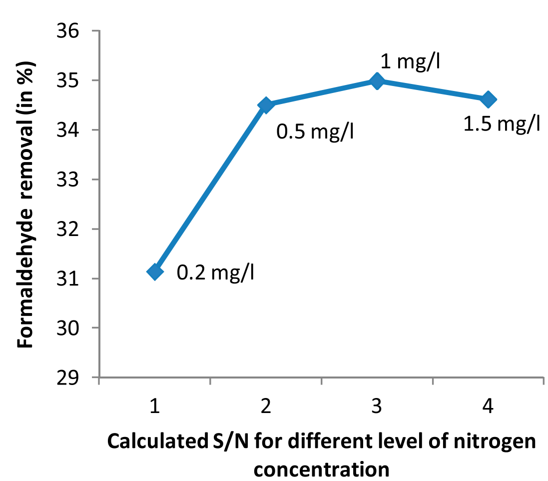

Fig. 6. Effect of various concentrations of $(NH_4)_2[Fe(SO_4)_2]$ on formaldehyde removal.

temperature in which it grows and reproduces best. As the temperature increases, molecules move faster, enzymes speed up metabolism and micro-organisms rapidly increase in size or number. The results show that formaldehyde removal efficiency increased when the operating temperature was in the range of 22–45˚C. As it can be seen from (Fig. 7) formaldehyde removal efficiency increased, with a higher slope obtained in the range of 22–40˚C, but the slope showed slight decrease in the temperature range of 40–45˚C. This temperature range of (40–45˚C) is the optimum border temperature for mesophiles and thermopiles due to this the rate of mesophilic growth is gradually reduced, whereas the rate of thermophilic growth is raised with the increasing temperature of this range. These results are partially similar to the study of Lu et al. [28] for BTFR efficiency in the removal of a mixture of benzene, toluene, ethyl benzene, and o-xylene (BETX) from air. They could find at steady-state condition, BTEX removal efficiency increased as the operating temperature increased in the range of 15–30˚C and decreased at temperatures above 30˚C.

3.8. Effect of phosphorus concentration

Phosphorus, like carbon and nitrogen, is an essential element for all living systems and is required for the synthesis of nucleic acid molecules (DNA, RNA). During this study, the effect of four levels of KH_2PO_4 as sole phosphorus source on BTFR to remove formaldehyde from polluted air was carried out in this study. The results showed that formaldehyde removal efficiency increased when KH_2PO_4 operating concentration was in the range of 0.1–0.5 mg/L and appeared to be stable above 0.5 mg/L (Fig. 8) and it showed 0.5 mg/L of KH_2PO_4 was sufficient to support the growth of micro-organisms in the BTFR when formaldehyde inlet concentration was equal to 490 mg/L. An optimal ratio equal to 100/1 has been suggested for carbon/phosphorus ratio [17] which is not close to our results. In this study, ratio of carbon/phosphorus is 100/0.1 that it is 10 times lower than the suggested optimum ratio. The increasing of this ratio has a negative impact on formaldehyde removal as seen in Fig. 8. This behavior of BTFR can be described by the rising of osmotic pressure where KH_2PO_4 can be hydrolyzed in water to K^+ and HPO_4^-. K^+ can increase the nutrient solution osmotic pressure, lower the microbial activity and result in lower BTFR performance.

3.9. Analysis of variance

The analysis of variance (ANOVA) is a powerful technique in Taguchi method that explores the contribution (%) of factors affecting the response [29]. The strategy of ANOVA is to know how much variations of each factor contribute to the total variation observed in the results. The statistical analysis of the results were carried out using Qualitek-4 (Nutek Inc.) software based on Eq. (3):

$$SS_T = \sum_{i=1}^{n} (\mu_i - \mu_m)^2 \tag{3}$$

where n is the number of experiments in the orthogonal array, μ_i is the mean S/N ratio for the ith experiment and SS_T is total sum of squared deviations from the total mean S/N ratio μ_m.

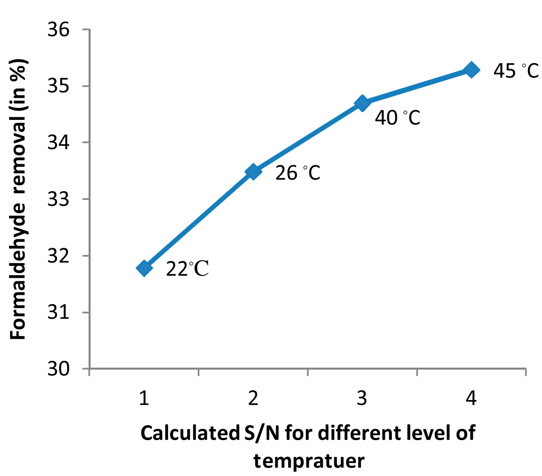

Fig. 7. Effect of different temperature on formaldehyde removal.

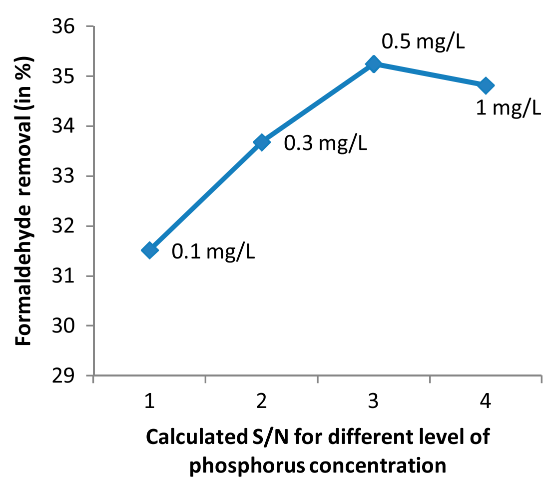

Fig. 8. Effect of phosphorus concentration on formaldehyde removal.

Table 3
ANOVA for factors affecting the formaldehyde removal

Factor	DOF	Sum of squares	Variance	Percent contribution (in %)
pH	3	53.3	17.766	28.765
Temperature	3	28.716	9.572	13.488
Nitrogen concentration	3	38.581	12.86	19.619
Phosphorus concentration	3	33.314	11.104	16.345
Other/Error	3	7.01	2.336	21.783
Total	15	160.923	–	100

Table 3 shows the ANOVA statistical terms representing the significance of four environmental factors affecting the formaldehyde removal. It is implied from the contribution (%) in the last column of ANOVA table that all of the factors are more or less important and significantly affects the response. In the range of levels considered for any factors in this study the following order was observed for the significance of factors

pH > $(NH_4)_2[Fe(SO_4)_2]$ concentration > KH_2PO_4 concentration > temperature.

3.10. Optimum conditions

It is seen from (Figs. 3–6) and the data in ANOVA table, one can estimate the relative optimum conditions at which the maximum rate of micro-organism growth can occur or the formaldehyde removal to be attained. Table 4 indicates the optimum conditions obtained through Taguchi method approach in order to obtain the maximum response for BTFR, using pH 7, 1 mg/L $(NH_4)_2[Fe(SO_4)_2]$, 0.3 mg/L KH_2PO_4 and temperature of 45˚C which are considered suitable. The percent of improvement in response (S/N) with respect to current average of results was also calculated (Table 4). It is predicted that application of optimum conditions would improve the transformed response by 38% over the current average performance. However, more experiments are required in order to find the exact optimum condition for a defined target.

Table 4
Optimum condition for maximum removal of propylene glycol

Factors	Best levels	Contribution (S/N)
pH	3	2.224
Temperature	4	1.474
Nitrogen concentration	3	1.177
Phosphorus concentration	3	1.43
Total contribution from all factors	6.304	
Current grand average of performance (S/N)	33.813	
Expected result at optimum condition (S/N)	40.118	

4. Conclusions

The influence of four environmental variables on the rate of micro-organism growth and formaldehyde removal in biotrickling filter were statistically analyzed using Taguchi experimental design. The results obtained showed that the biofiltration process has higher efficiency to remove formaldehyde from polluted air after optimization. Therefore, Taguchi method can be considered as a suitable method to optimize biological reactors. The formaldehyde removal was dependent on pH, nitrogen, phosphorus concentration, and temperature. All of the factors examined in this study had significant effect on formaldehyde removal. The nutrient solution pH had the largest effect and contribution in formaldehyde removal, and the results of this study can be applied to conduct BTFR to remove formaldehyde from air with maximum efficiency.

Acknowledgment

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Symbols

C — nutrient tank COD (in mg/L)
θ — time (in h)
a and b — constants coefficients
S/N — signal-to-noise ration
y_i — formaldehyde removal efficiency at BTFR for ith experiment
N — number of measurements carried out for each run
SS_t — total sum of squared deviation from the total mean S/N ration
μ_i — mean ration for the ith experiment
μ_m — the total mean S/N ratio

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