

## Biodegradation of formaldehyde from contaminated air using a laboratory scale static-bed bioreactor

Yaghoub Hajizadeh, Mohsen Rezaei

Environment Research Center, Isfahan University of Medical Sciences (IUMS), Isfahan, Iran, and Department of Environmental Health Engineering, School of Health, IUMS, Isfahan, Iran

#### Address for correspondence:

Dr. Yaghoub Hajizadeh, Environment Research Center, and Department of Environmental Health Engineering, School of Public Health, IUMS, Isfahan, Iran. Email: y\_hajizadeh@hlth.mui.ac.ir

## ABSTRACT

Aims: The objective of the present study was to evaluate the performance of an aerobic fixed-bed bioreactor (FBR) enriched with microorganisms of sewage sludge in biodegradation of formaldehyde in air stream with various retention times and airflow rates in laboratory scale.

**Materials and Methods:** An aerobic biofilter 60 cm in height and 14 cm internal diameter made of steel was constructed and packed with a mixture of pumice and compost as a medium and utilized in this study. The microorganism's growth, which is derived from the sludge of a municipal wastewater treatment plant, was initiated by adding nutrient. During the first few days of run, the airflow containing different concentrations of formaldehyde (from  $24 \pm 3$  to  $224 \pm 5 \text{ mg/m}^3$ ) was introduced to the reactor to ensure biological adaptation. Sampling was performed through a series of two impingers containing adsorbent, and analyzed by chromotropic acid assay using DR-5000.

**Results:** The maximum removal and elimination capacity of formaldehyde was yielded at  $0.48 \pm 0.06 \text{ g/m}^3/\text{h}$  inlet loading rate and 180 s of empty bed retention time (EBRT). These values for stabilized days were almost 88% and 0.42 g/m<sup>3</sup>/h, respectively.

**Conclusion:** The results showed that by increasing the inlet concentration of formaldehyde and reducing the EBRT, the formaldehyde removal capacity of the system decreases. Aerobic bioreactor with appropriate bed volume and compatible with inlet pollutant mass flow rate in optimum retention time will admissibly degrade and reduce the formaldehyde concentration from contaminated gas phase, such as gases produced in municipal wastewater treatment facilities.

Key words: Biodegradation, biofiltration, formaldehyde, fixed bed reactor

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## INTRODUCTION

Emission of toxic compounds into the environment has been widespread in the recent decades that are due to industrial advancement and increased use of organic fuels.<sup>[1]</sup> Volatile organic compounds (VOCs) are group of toxic pollutants that release to the environment either man-made or naturally made.<sup>[1-3]</sup> These compounds degrade slowly and accumulate in the ecosystem, thus they cause serious damage of the

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This article may be cited as: Hajizadeh Y, Rezaei M. Biodegradation of formaldehyde from contaminated air using a laboratory scale static-bed bioreactor. Int J Env Health Eng 2014;3:4. environment and exposed human societies.<sup>[4]</sup> In the past decades industries released a large amount of these pollutants into the ecological systems.<sup>[5]</sup> Among all, formaldehyde (HCHO) because of its harmful effects on human health and high emission level in atmosphere, considers as one of the important VOCs emitted from industries.<sup>[1]</sup> Twenty-one million tons of formaldehyde is producing yearly.<sup>[6]</sup> Health organizations defined exposure limit with formaldehyde to 0.1 ppm.<sup>[7]</sup> Based on Occupational Safety and Health Administration (OSHA) guideline, the admissible amount of formaldehyde in indoor and outdoor air limited at 0.75 and 0.5 ppm, respectively.<sup>[8]</sup>

Exposure to the high concentrations of formaldehyde causes nausea, vomiting, diarrhea, abdominal pain,[3,9-11] gastroenteritis, sleeping disorders, damage to the optic nerve,<sup>[10]</sup> eye irritation, and in long-term exposure the risk of cancer would be inevitable.<sup>[9-12]</sup> Also exposing to the high concentrations of it can cause death.<sup>[10]</sup> This gas is one of the chemical compounds, which is typically used in chemical processes<sup>[10]</sup> and may be mainly produced by activities such as burning,<sup>[1]</sup> paper production,<sup>[13]</sup> manufacture of synthetic resins,<sup>[1,13]</sup> neopan production, released gas from formaldehyde production, chemical industries and partly from photochemical oxidation of hydrocarbons, methylated compounds and other organic compounds, and also in secondary reactions of hydrocarbons with ozone in atmosphere.<sup>[10-12]</sup> Furthermore, increased use of methanol fuel can also play a role in emission of formaldehyde into the air.<sup>[11]</sup> Examples of indoor source of formaldehyde can be three-ply boards, neopan, carpets, curtains, paper products, tobacco smoke, pesticides, and specific adhesives.<sup>[3,14]</sup> Formaldehyde in indoor air considered as a main air pollutant.<sup>[14]</sup>

There are several methods to remove pollutants from air, which are in two categories, physicochemical methods and biological methods. Physicochemical methods that are almost conventional including adsorption, for example, activated carbon, absorption, catalytic combustion, gas condensate containing pollutant, and modern biological methods in which biofiltration is the most common method. The conventional methods are mostly not cost effective and require transformation of gas phase of the pollutants to other phases such as solid or liquid phases. In addition, these methods are associated with the production and emission of secondary hazardous toxic substances.<sup>[1,3,10,15]</sup> In contrast, biological methods are widely applicable because, compared with traditional methods, they are cost effective and do not associate with producing secondary pollutants.<sup>[1,4,11,13,16]</sup> Other strengths of the biofiltration methods are elimination of different concentrations of VOCs (1-1000 ppm) and low cost of installation.<sup>[9]</sup>

Applications of biological technologies due to their natural processes are the most reliable methods of removal of environmental pollutants.<sup>[16]</sup> The principal of biological technologies are based on the ability of some microbial

species in using volatile organic compounds as an energy source in cellular respiration phase and carbon source in growth phase.<sup>[1,4,9,17,18]</sup> In these systems microbial agents, mostly bacteria and fungi, are grown on porous bed and air containing pollutant passed through the bed. In the next step the microorganisms<sup>[16]</sup> that are suspended in the biofilter, for example, activated sludge or attached to the bed<sup>[19]</sup> break down the pollutant compounds to simple substances. The final products of this process are carbon dioxide<sup>[16]</sup> and water.<sup>[4]</sup>

The microorganisms that can degrade formaldehyde are Pseudomonas putida, Trichosporon penicillatum, Pseudomonas cepacia, Pseudomonas alcaligenes, Methylobacterium extorquens, Halomonas spp.,<sup>[13]</sup> Pseudomonas pseudoalcaligenes, [13,20] Methylococcus spp., Vibrio spp., and methylotrophic yeast.<sup>[17,18]</sup> Effective parameters in removal activity of pollutants by biofiltration can be oxygen saturation, temperature (optimally 20-30°C), the biofilm thickness, pollutant's diffusion into the biofilm, pH, C/N ratio, salinity, humidity (optimally 30%-60%)<sup>[21]</sup> and so on.<sup>[9,18,22]</sup> Biological systems that are currently being used for contaminated air treatment include bioscrubbers, biofilters, trickling filters,<sup>[16,23]</sup> and membrane bioreactor.<sup>[4]</sup> In previous years bioreactors have been used in treating the unpleasant odors<sup>[24]</sup> of municipal wastewater treatment facilities, composts, and bread preparation and storage places. However, recently this method is being used for treatment of gaseous pollutants, such as formaldehyde in laboratory scale and developing significantly in industrial applications.<sup>[1,10,13]</sup> The present study aimed to develop and improve a biofiltration system to remove or minimize formaldehyde from synthetic gas flow containing defined amount of the pollutant. The effects of different operational parameters on the performance of the system were also studied.

## **MATERIALS AND METHODS**

This laboratory scale experimental study was carried out by designing and construction of an aerobic reactor with continuous airflow.

### **Biofilter system**

In this study conventional biofiltration method was chosen, because of, unlike the other biofiltration methods, it is the ability to treat a wide variety of pollutants, lower pressure drop, and there is no wastewater production.<sup>[4]</sup> A stainless steel reactor with height of 60 cm and internal diameter of 14 cm was constructed. A mixture of pumice (80% v/v) and maturated compost (20% v/v) (obtained from Isfahan compost plant) with pH of approximately 8, and 6 L in volume was used as a bed to support the growth of the microorganisms. To uniformly distribute formaldehyde gas through the biofilter, a perforated stainless steel plate was placed at 8 cm from the bottom of the reactor. The inlet and outlet pipes of the reactor were also made of stainless

steel. Three types of chambers were placed before the reactor, including humidifier chamber, formaldehyde vaporproducing chamber, and a chamber for mixing formaldehyde vapor with air. Air flow required by the system was provided by an air compressor, and two flow meters were used to control the output flow of the air. The activated carbon column was used to prevent the entry of probable interference pollutants with the outlet air from the compressor. The schematic of the bioreactor is shown in Figure 1.

#### Materials and reagents used

To provide formaldehyde vapor, 37% formaldehyde solution purchased from (Merck Co. Germany) was utilized. Nutrients applied with their approximate values, which have been injected into the biofilter for strengthening the growth of microorganisms were as follows:  $NH_4Cl$  (500 mg/day),  $KH_2PO_4$  (700 mg/day), MgSO\_4 (20 mg/day), MnSO\_4 (3 mg/ day), CaCl\_4 (50 mg/day), FeSO\_4 (5 mg/day), and ZnSO\_4 (3 mg/day). NaOH was used to adjust the pH of the nutrient and activated sludge. Microorganism's population was provided from south Isfahan activated sludge unit of municipal wastewater treatment plant. The chemical compounds needed for analyzing the formaldehyde also were provided from Merck Company, including 39% sodium bisulfate, 98.5% chromotropic acid, and sulfuric acid 96%.

#### Set up and operation of the biofilter reactor

In the first steps of the biofilter system operation, the air flow from a compressor after flowing through a column of activated carbon directed into an airflow divider and then, the needed airflow controlled by flow meters entered into a 37% formaldehyde-containing chamber and a humidifier chamber. In the next step, the mixture of formaldehyde and water vapor released from the related chambers entered into a mixing

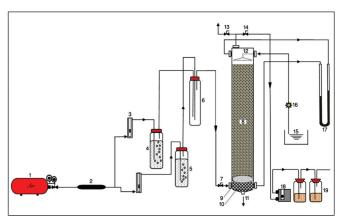


Figure 1: The schematic of the bioreactor: (1) Air Compressor, (2) Activated carbon column, (3) Flow meter,
(4) Humidifier, (5) Tank of formaldehyde, (6) Mixing tank,
(7) Air sampling valve, (8) Biofilter bed, (9) Plate distributor of air, (10) Uniform air distribution platform, (11) Liquid outlet valve of the reactor, (12 Distributor of nutrient,
(13) Filtered air outlet, (14) Sampling valve, (15) Nutrient reservoir, (16) Peristaltic pump, (17) Manometer, (18) Personal sampling pump, (19) Impingers sampling

chamber. The humidifier and mixing chambers were used for diluting and adjusting of the formaldehyde concentration introduced into the reactor. The contaminated air mixture just before entering into the reactor was daily sampled and analyzed to ensure the exact formaldehyde concentration in the inlet. The mixed air with defined concentrations of formaldehyde was entered from the bottom of the biofilter and exit from the top after a defined retention time (from 30 s to 3 min). This study was carried out at  $23 \pm 2^{\circ}$ C. Activated sludge aerated continuously in a chamber at a rate of 3 L/min for 20 days. Also, for preparation and adaptation of microorganisms to new conditions before inoculation, a defined amount of formaldehyde (1-3 mL/L) was added to the chamber containing activated sludge, daily.

During the study at the beginning of the operation, 280 mL of nutrients and buffer was added, four times a day, to the biofilter by peristaltic pump, but later the procedure was done manually from above the reactor for better and uniform distribution of the nutrients. The quantity of the nutrients and supernatant were determined experimentally by trial and error method in order to maintain optimum moisture (30%-50%) of the bed needed for microbial growth. At the beginning for the adaptation of microorganisms, biofilter was operated for 25 days with 120 L/h airflow rate containing  $24 \pm 3 \text{ mg/m}^3$  inlet formaldehyde concentration and empty bed retention time (EBRT) of about 3 min. After adaptation of the system and stability of the removal efficiency in this retention time, the removal efficiency was tested by maintaining a constant input concentration with EBRT in 30, 60, 90, 112, 150, and 180 s, then results were analyzed. At the end of this step, it was observed that in both 150 and 180 s of EBRT the removal efficiency was same and at a maximum level. Therefore these retention times have been considered as optimal retention times for removal of formaldehyde and 150 s has been chosen for further procedures of the study. Then, the airflow containing  $48 \pm 5.5, 77 \pm 4.5, 124 \pm 3, 176 \pm 3.5, and 224 \pm 5 \text{ mg/}$ m<sup>3</sup> concentrations of formaldehyde, each separately for the time periods needed to achieve an approximately constant rate of formaldehyde removal (the time period when the variation of removal efficiency was not observed) entered into the reactor.

#### **Sampling and measurement**

Sampling and measurement of formaldehyde was performed according to the National Institute for Occupational Safety and Health (NIOSH) Method 3500.<sup>[25]</sup> According to this method, sampling of the inlet and outlet air of the reactor was done by individual personal sampling pump, whose inlet was attached to the reactor and outlet was attached to the two impingers containing 1% sodium sulfite adsorbent. Formaldehyde concentrations in liquid adsorbent were measured daily by colorimetric method using spectrophotometer (DR-5000-HACH LANGE Co.Germany) at 580 nm. Moisture content of the medium was determined by gravimetric method using 105°C oven. Pressure drop of the bed was measured continuously by a manometer placed in the inlet and outlet of the reactor. The pH of nutrients and supernatant was measured and adjusted using a pH meter (1500-Cyberscan Co. USA).

Because majority of grown microorganisms in biofilm are bacteria,<sup>[9]</sup> to identify the predominant bacteria for degradation of formaldehyde in this study, enriched, selective, and differential media were used. The process of the identification of predominant bacteria was as follows: First, in sterile condition biofilm samples were cultivated in the primary enriched media, including nutrient agar, BHT broth, blood agar, and chocolate agar for isolating the bacteria from the biofilm in the form of single colonies. Second, the smears were prepared from colonies and stained by Gram's method of staining<sup>[26,27]</sup> and then stained slides were examined under oil immersion ×100 objective using a light microscope.

Bacterial streaking culture has done on MacConkey agar and EMB agar for isolation of predominant colonies of Gramnegative from Gram-positive. Gram-stained smears of two predominant colonies from the two media were examined by microscope.

For this regard, a row of five tubes containing triple sugar iron agar, sulfide-indole-motility medium, Simon's citrate, Urea, MR-VP media, and lysine decarboxylase and *ornithine* decarboxylase tests were used. In addition, the 72-h O-nitrophenyl- $\beta$ -d-galactopyranoside test was used to identify the lactose fermenters from lactose nonfermenters.<sup>[27]</sup>

## RESULTS

For the microorganisms to adapt to the system, in the first stage, reactor operated for 25 days by  $24 \pm 3 \text{ mg/m}^3$  of formaldehyde with 0.48  $\pm$  0.06 g/m<sup>3</sup>/h loading rate. Results

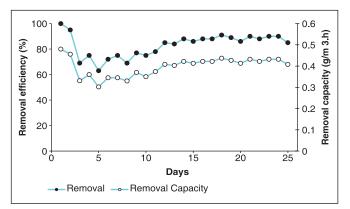


Figure 2: The variations in removal efficiency of formaldehyde during the microorganism's adaptation period with 180 sec. of EBRT and  $24 \pm 3 \text{ mg/m}^3$  of inlet concentration in this stage are shown in Figure 2. Removal efficiency of formaldehyde with the mentioned inlet concentration on the 1st day of inoculation was 100%. By passing days from operation of reactor the removal capacity reached an almost constant level with an average of 0.42 g/m<sup>3</sup>/h (88% removal efficiency). This average value was considered as the maximum removal capacity of the system.

In the next step, different EBRTs were applied in order to define the optimum retention time of formaldehyde polluted air, and the removal efficiency was compared with constant inlet formaldehyde concentration for the EBRTs. This phase of the experiment was done with three replicates (one per day) and the mean efficiency considered as the removal efficiency. Average removal efficiency in retention times of 150 and 180 s were almost similar, about 88% and 87%, respectively [Figure 3]. Because of similar efficiency in two primary EBRTs, 150 s with 144 L/h was chosen and applied in the present study.

By choosing the suitable EBRT in the second step, the study carried out in the third stage with different concentrations of pollutant. After elapsing the days needed to stabilize the removal efficiency, 4 days were chosen as the benchmark of stabilized efficiency for each concentration. As shown in Figure 4a, in operating days with inlet formaldehyde concentration of  $48 \pm 5.5$  mg/m<sup>3</sup> (loading mass  $1.15 \pm 520.132$  g/m<sup>3</sup>/h), formaldehyde removal capacity increased and its concentration in the reactor outlet was decreased simultaneously. In the 1st day, removal capacity and efficiency dropped slightly to 0.864 g/m<sup>3</sup>/h and 75%, respectively. The removal capacity and efficiency, in which in 4th to 7th day reaches to 1 g/m<sup>3</sup>/h and 87.5%, respectively, and did not vary tangibly during the mentioned days.

In the next phase, concentration of formaldehyde increased to  $77 \pm 4.5 \text{ mg/m}^3$  (with  $1.85 \pm 0.108 \text{ g/m}^3/\text{h}$ ). As it is shown in Figure 4b, the removal capacity and outlet formaldehyde concentrations in the new loading rate in different days

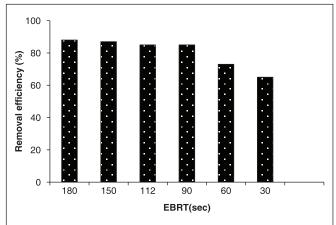


Figure 3: The average removal efficiency of formaldehyde in different EBRTs with  $24 \pm 3 \text{ mg/m}^3$  of inlet concentration

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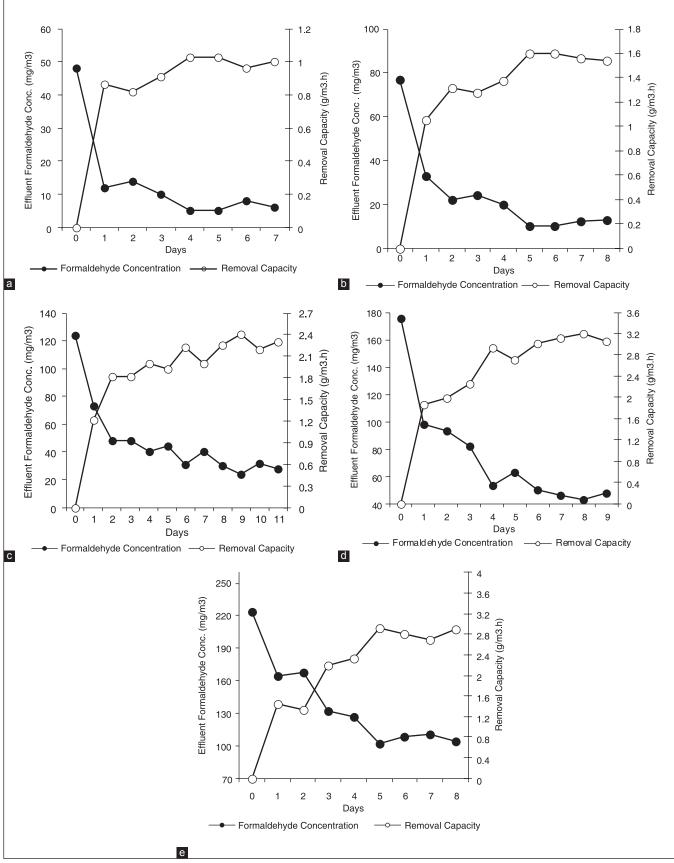


Figure 4: Outlet formaldehyde concentration from the rector (the filled circle) the formaldehyde removal capacity (empty circle) in different time periods for stabilizing the removal efficiency in different inlet concentrations. (a)  $48 \pm 5.5 \text{ mg/m}^3$ ; (b)  $77 \pm 4.5 \text{ mg/m}^3$ ; (c)  $124 \pm 3 \text{ mg/m}^3$ ; (d)  $176 \pm 3.5 \text{ mg/m}^3$ ; (e)  $224 \pm 5 \text{ mg/m}^3$ 

have been tested. The removal capacity in the 1<sup>st</sup> day was 1.05 g/m<sup>3</sup>/h. The outlet levels of formaldehyde dropped in the next few days and in the last 4 days decreased to a constant level of 13 mg/m<sup>3</sup> [Figure 4b]. The average removal capacity and efficiency during these 4 days was 1.57 g/m<sup>3</sup>/h and 85.4%, respectively.

In the next step, the inlet concentration of  $124 \pm 3 \text{ mg/m}^3$  (with 2.98  $\pm 0.72 \text{ g/m}^3/\text{h}$ ) was applied and the results are shown in Figure 4c. The mean capacity and removal efficiency during 8th to 11th day were 2.3 g/m<sup>3</sup>/h and 77%, respectively.

After the stabilization of removal efficiency, the inlet formaldehyde concentration increased to  $176 \pm 3.5 \text{ mg/m}^3$ with a mass loading rate of  $4.22 \pm 0.084 \text{ g/m}^3$ /h. Primary removal efficiency on the 1st day showed a greater drop than the previous steps. The removal capacity and efficiency on the 1st day started from  $1.87 \text{ g/m}^3$ /h and 44.3%, respectively, and by increasing the adaptation of microorganisms, they slightly increased. Average removal capacity and efficiency in the last 4 days (6<sup>th</sup> to 9<sup>th</sup> day) increased to 3.09 g/m<sup>3</sup> and 73.4\%, respectively [Figure 4d].

Finally, the last step of the study carried out by  $224 \pm 5 \text{ mg/m}^3$  of inlet concentration with mass loading of  $5.38 \pm 0.12 \text{ g/m}^3/\text{h}$  and the stabilization occurred during 8 days [Figure 4e]. The removal capacity and efficiency on the 1<sup>st</sup> day were 1.44 g/m<sup>3</sup>/h and 26.8%, respectively, and also the mean removal capacity and efficiency from 4th to 8<sup>th</sup> day were recorded as 2.83 g/m<sup>3</sup>/h and 52.6%, respectively. The results on all operation days are shown in Figure 5.

Figure 6 shows the mean removal efficiency in 4 days of stabilized efficiency during the reactor operation days for different inlet formaldehyde.

# Identification of predominant microorganisms in degradation of formaldehyde

The Gram's staining method isolates the bacteria into two groups, gram-positive which appeared as purple-blue bacteria, whereas gram-negatives were observed as pink-red colored ones [Figure 7]. In our study the majority of bacteria were Gram-negative, whereas limited numbers of colonies were gram-positive. It can be concluded that most of the formaldehyde degradation was done by Gram-negative bacteria, which grew and formed biofilm.

The main predominant colonies were observed as pinkcolored *Coccobacilli*, whereas the other colonies were containing pink-colored *bacilli*, which later identified as *Proteus* spp. At this stage, the number of bacterial colonies counted were approximately 10<sup>6</sup> CFU per gram of bed.

These two types of colonies were differentiated by different differential media and biochemical tests, such as IMViC test (Indole, Methyl red, Voges — Proskauer, and Simon's citrate).<sup>[28]</sup> According to the results of these biochemical tests and differentiating media on isolated Gram-negative bacteria, the predominant colony was identified as *Citrobacter freundii*.

## **DISCUSSION**

As shown in Figure 2, elimination efficiency of formaldehyde with the primary inlet concentration on the 1st day of inoculation was 100%. This high elimination rate was probably due to adsorption of formaldehyde by the bed on the 1st day of inoculation. On the 2nd day, the system faced a drop of almost 30% in the removal efficiency, but it gradually increased with time again. However, in the first 10 days of biofiltration, acceptable removal efficiency did not yield [Figure 2]. The low efficiency of the system could be due to less adaptation of the microorganisms with the inlet formaldehyde and less microbial mass at the beginning. Humidity of above 60% wet during the first 5 days of the study due to high water flow rate (0.5 L) containing nutrient into the reactor could probably reduce the growth speed of bacteria, which are the main factors for degradation of organic pollutants, such as formaldehyde. Given the importance of bed moisture in microorganisms' growth by experimental and trial and error method, the nutrient flow rate was reduced to 280 mL. This level of nutrient flow used in this study was different from other studies due to different reactor volume.[11]

The results of the present study, despite the similar used concentrations, are slightly different from that of Xu *et al.*'s report. They yield maximum (100%) removal efficiency with 25 L of bed volume and primary inlet formaldehyde concentration of 20 mg/m<sup>3</sup>. This removal level was accomplished in minimum airflow of 112 L/h with at least about 0.36 g/m<sup>3</sup> loading.<sup>[11]</sup> Thus in their study EBRT was higher compared with the present study, which can be the cause of different results in the two studies. Results in Figure 3 show that microbial mass is able to degrade formaldehyde in 30 s less than primary retention time, 180 s, without tangible reduction in removal efficiency. However, in lower EBRTs removal efficiency gradually decreased and eventually in 30 s of EBRT the removal efficiency declined to 65%.

Therefore, due to very low level and unacceptable removal efficiency in retention times less than 30 s, tests did not continue in retention times less than that. According to the results, the highest removal efficiency was accomplished in 180 s of EBRT. It can be suggested that in higher EBRTs, higher removal efficiency may be achievable, but in this study due to low reactor volume, higher EBRTs were not applicable. Thus the higher EBRTs in future studies is suggested. It can be deduced that, reasonably, the low removal efficiency at low EBRT is due to reduction of the amount of formaldehyde penetration between the biofilm layer and the gas phase. Some other studies support the idea of decreasing removal efficiency by shortening the EBRT. Prado *et al.* studied removal efficiency of methanol gases

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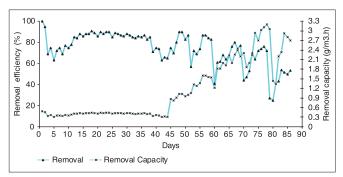


Figure 5: Removal Efficiency and capacity in total of operation days

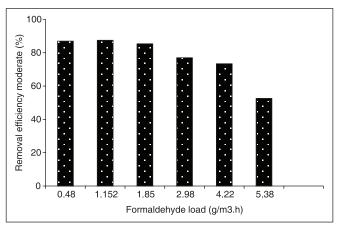


Figure 6: The mean removal efficiency in 4 days of stabilized efficiency during the reactor operation days for different inlet formaldehyde concentrations with 150 sec. of EBRT



Figure 7: Gram-positive and gram-negatives bacteria grown on the primary enriched media

and formaldehyde by two types of reactors, conventional bioreactor and trickling filter in EBRTs of 20.7, 30, 46.5, and 71.9 s. They found that in 30, 46.5, and 71.9 s of EBRTs the removal efficiency was the same, but in 20.7 s of EBRT the efficiency was lower.<sup>[10]</sup>

Efficiency loss in the first phase of the third stage, which is due to the variation of inlet loading from  $0.48 \pm 0.06$  to  $1.152 \pm 0.132$  g/m<sup>3</sup>/h causing shock to the microorganisms.

However, by the adaptation of them with the new condition the removal capacity and efficiency increases again. On the 1st day, similar to the previous concentration removal capacity and efficiency showed a low value, even lower than the previous step. As already mentioned, in the next phase (loading  $1.85 \pm 0.108 \text{ g/m}^3/\text{h}$ ) by increasing the inlet loading from  $1.152 \pm 0.1132$  to  $1.85 \pm 0.108 \text{ g/m}^3/\text{h}$ , the removal efficiency dropped to 57% due to the shock the microorganisms faced. Loading took 8 days of testing and after the 4th day no variation in removal and outlet formaldehyde was observed.

In the next phase, the trend of variation in outlet concentrations were done similar to the previous steps and again low efficiency was seen on the 1st day of operation, which is due to a shock that microorganisms faced by a new concentration of formaldehyde. On the 1st day of operation the removal capacity and efficiency was about 1.22 g/m<sup>3</sup>/h and 41%, respectively, but by adapting the microorganisms to the new concentration, the removal capacity increased and after 8th day maintained in a stable and constant level. Interestingly, this step of the study took more time than the previous steps for stabilizing its removal capacity. In the loading rate of 4.22  $\pm$  0.084 g/m<sup>3</sup>/h, primary removal efficiency on the 1st day showed a greater drop than the previous steps. Finally in the last phase of the third stage, the stabilization occurred during 8 days. According to the results by increasing the inlet loading rate, despite the increase in removal capacity, the removal efficiency decreased slightly [Figure 6] and finally in 5.38  $\pm$  0.12 g/m<sup>3</sup>/h of inlet loading rate, an acceptable removal was not observed. This fact indicates that the biofilter system for higher inlet loading rate with the studied condition is not responsive, although by increasing the EBRT up to 150 s would probably result in higher removal rate.

There are few studies in this field conducted by other researchers; however, the results of Xu *et al.* are slightly different from what we found. They used a biofilter packed with ceramic rings with 3.7 min of EBRT, and tested the removal efficiency in different heights of the bed with inlet concentrations of 5-207 mg/m<sup>3</sup>, and found the removal efficiency more than 97% in inlet concentration of 70 mg/m<sup>3</sup> and more than 90% in inlet concentration of 207 mg/m<sup>3</sup> in the middle part of the reactor.<sup>[11]</sup> Apparently, the main cause of the difference between the results of Xu et al. and ours is probably the results of different EBRTs. Prado et al. investigated the removal of the mixture of formaldehyde and methanol gases with lava rock bed and primary loading rate of 15 and 78.2  $\pm$  2.9 g/m<sup>3</sup>/h and yield maximum of 9.48  $\pm$  1.63 and 36.8  $\pm$  103 g/m<sup>3</sup>/h with 80 s of EBRT, respectively. However, the higher rate of loading and lower EBRT and also competitive degradation of formaldehyde with methanol as a carbon source by microorganisms, are probably compelling reasons for the limited removal efficiency compared with that in the present study.<sup>[10]</sup>

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## CONCLUSIONS

The formaldehyde removal efficiency of the aerobic bioreactor in the preparation and operation days was achieved at an acceptable level. The highest removal efficiency was achieved in  $0.48 \pm 0.06$  g/m<sup>3</sup>/h of inlet loading rate. In addition, the removal efficiency in higher inlet loadings with constant EBRT, showed progressive decrease. It can be concluded that by increasing the inlet loading and reduction in the EBRT, the overall removal efficiency of biofilter system decreases. The aerobic bioreactor with a suitable bed for microorganism growth, optimum EBRT, and applied loading values has an acceptable performance in the degradation and elimination of formaldehyde from the air and can be one of the potential methods for filtering of the polluted air such as the air of municipal wastewater treatment plants. Therefore, it is suggested that in the future studies, a pilot study of this system in the industrial scale or real polluted air be exploited. In this case, the effect of other inlet polluted airflows along with formaldehyde into the reactor can be investigated and also the interaction of different pollutants and microbial resistance against them can be studied to achieve the best leading conditions for biofiltration of polluted air in the industrial scale. Also, it is beneficial to study the effect of different optimum temperatures on the performance and activity of microorganisms in pilot scale in order to simplify and improve the operation of biofilters.

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