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A comparative study of the effect of compost/woodchips mixture, natural zeolite and zeolite/activated carbon mixture as packing materials on the biofilter performance

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ABSTRACT

The removal of formaldehyde from contaminated air was investigated via three laboratory-scale biofilters packed with different materials: a mixture of compost and woodchips (I), the natural clinoptilolite zeolite particles in the original form (II), and the mixture of zeolite/activated carbon (III). The biofilters were inoculated using aerobic sludge. The average removal efficiencies of 97.5%, 90%, and 93.5% were obtained at a 100 s empty bed residence time (EBRT) and 20 mg/m^3 inlet concentration of formaldehyde for the biofilter of configurations I, II, and III, respectively. Also, the performance of the reactors was investigated at different EBRTs of 20, 30, 60, and 100 s, and the maximum elimination capacity of 2840 mg/m^3.h was achieved at the lowest EBRT (20 s) for the biofilter of configuration II. Increasing the inlet formaldehyde concentration from 20 mg/m^3 to 80 mg/m^3 led to the maximum formaldehyde removal efficiency of 82% for the biofilter of configuration III. Therefore, a comparison of the results of the biofilters' performances showed that the biofilter of configuration III had the best performance, which was validated by obtaining a higher mass transfer coefficient. However, the biofilter of configurations II and III achieved steady-state conditions in a shorter time.

1. Introduction

Due to industrial development, the distribution of hazardous compounds into the environment has increased. It is one of the most important issues encountered in the past decades. One of the pollutants gaining more attention is formaldehyde [1]. Formaldehyde is a colorless gas, which is easily ignited at room temperature and is solvable in water. This compound is dangerous due to its high toxicity [2], relatively high emission rate into the environment, and negative effects on human health and ecological systems [1]. Formaldehyde is a pollutant that may be found everywhere. In 1975, it was recognized for the first time that formaldehyde was produced from the chipboards which were employed in building materials [3]. It is often released from industrial processes such as oxidation and photo-oxidation. Also, it is a product of the secondary reactions of hydrocarbons with O_3 [4]. It can also be the result of processes such as burning, paper production, artificial resins, neopan fabrication, and the chemical industry; plus, it can be released from neopan, carpets, curtains, paper products, pesticides, and some of the adhesives in indoor sources [5]. Formaldehyde in high concentrations can cause nausea, vomiting, bellyache, diarrhea [6], and at higher levels, it can even cause death. Therefore, it is necessary to consider efficient methods for removing it from the air. These methods are divided into two classes: physicochemical and biological [5]. The physicochemical methods include adsorption, absorption, condensation, and combustion [7]. But these methods have some disadvantages, such as high energy consumption, low removal efficiencies, and damaging risks [3]. Recently,

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biological techniques such as bio-scrubber, biofilter, bio-trickling filter, and membrane bioreactor have been used more widely; they are effective due to their lower operational costs, non-production of secondary pollutants, and reliability when compared to that of physicochemical methods [7]. In this regard, the improvement and application of biological techniques are under the radar of researchers. Among the biological techniques, biofilters have received more attention due to their optimal removal of odor compounds and volatile organic compounds. [8,9]. The utilization of biofilters for the treatment of waste gases began in the early 1960s [10]. Biofiltration is a process that uses a porous packed bed in which contaminated airflow passes through and immobilizes the microorganisms. The contaminated airflow is treated by transporting the pollutants from the gas flow to the biofilm through their metabolization [11]. The removal of formaldehyde from the air with biofilters has been investigated in the literature [4-7,12-14]. The selection of the packing materials, as a biofilter medium, is an important issue in the optimization of biofiltration efficiency. Generally, the packing material must have some properties such as a high moisture holding capacity, high porosity, high surface area, ability to not clog, low pressure drop, enough resistance, and low cost [15]. Previous studies have focused on the traditional packing materials that have been used most frequently as biofilter beds such as soil, peat, compost, woodchips, and barks. Each of these materials has advantages and disadvantages that can be found in detail elsewhere [16]. Recently, Dobslow et al. (2019) used Bio-airSPHERES as composite packing material which could biodegrade the pollutants that were hard to remove [17]. Among the suitable materials for the biofilter bed, zeolite and activated carbon are cheap and easily available, and therefore, have received more attention. Zeolites with three-dimensional structures are hydrated compounds of aluminosilicates. They are hydrophilic materials that can gain and lose water reversibly and have a good capacity for cation exchange. The important properties of the zeolites include high porosity for the airflow, high specific surface area, a wide range of moisture holding capacity, contain various nutrient components, high adsorption, and capable of supporting microbial growth [18]. In these regards, natural zeolites, especially clinoptilolite, are suitable candidates as a biofilter medium (bed). Zeolite particles as packing material have many advantages: good immobilization of the microorganisms, high surface area, availability, low cost, having a porous structure, and high mechanical stability that prevents bed crushing and compaction. Still, they have some disadvantages like abrasion, damage to pumps by particles, and high mechanical clogging by particles over time.

Some studies have also been done to evaluate the ability of activated carbon in treating pollutants, especially formaldehyde [19]. Activated carbon is a conventional packing material that exhibits excellent performance in the biofilters. Moreover, microorganisms can grow on the activated carbon and form a biofilm. Biofiltration simulations resulted in high absorbance media for the activated carbon rather than medium absorbance media produced by materials such as peat and compost. It should be noted that in the biofilters packed with the activated carbon, the adsorbed pollutants prepared more available sources for microbial growth and enhanced the elimination capacity [19]. Some studies have investigated the biofiltration of VOCs with activated carbon [19-21] and zeolite [18,22-24], alone or mixed with other packing materials as the biofilter medium. However, to the best of the authors' knowledge, no study has been done on the assessment of the performance of the mixture of zeolite/activated carbon in the removal of formaldehyde using the biological method. According to the excellent properties of activated carbon and zeolite in the pollutant biofiltration, it is suggested that the application of activated carbon mixed with zeolite as a biofilter bed can enhance biofilter efficiency. The present study aims to use novel packing materials, namely zeolite particles and a zeolite/activated carbon mixture, in the biological formaldehyde removal process. These materials have the potential to enhance the performance of the biofilters compared to the compost/woodchips mixture that is frequently used [25] in the biological formaldehyde treatment.

2. Materials and methods

The formaldehyde solution (37%) and the components of the nutrient solution containing NH_4Cl, KH_2PO_4, $MgSO_4$, $MnSO_4$, $CaCl_2$, $FeSO_4$, and $ZnSO_4$ were purchased from Merck (Merck Co. Germany). The sodium sulfite as an adsorbent solution of the formaldehyde and the HCl/NaOH solution used to set the nutrient solution pH were provided by Merck (Merck Co. Germany). The source of microorganisms, aerobic sludge from wastewater, which has regularly been used in the biological removal of air pollutants [6], came from a septic tank of the University of Sistan and Baluchestan. The zeolite particles used in this study were obtained from the Semnan mines (Semnan province, Iran) with a particle size of 1-3 mm; the compost was provided by the Isfahan compost plant (Isfahan, Iran). The small woodchips were made using a woodchipper machine. The activated carbon powder with a particle size of 0.075 mm was provided by the Kimia Carbon Markazi Company (Arak, Iran).

2.1. Experimental setup

The biofilters were made of Plexiglas with a 13.6 cm internal diameter and an effective height of 33 cm. Three biofilters (reactors) were packed with the compost/woodchips mixture (50/50 %v/v), zeolite particles, and a mixture of zeolite/activated carbon particles (50/50 %v/v), respectively. Table 1 shows the properties of the packing materials. The biofilters were equipped with inlet and outlet ports for sampling and analyzing the concentration of formaldehyde in both the inlet and outlet air streams. The airflow was produced by a compressor and injected into the formaldehyde solution. The contaminated air flowed from the bottom to the top of the bed. A peristaltic pump sprayed the nutrient solution from the top of the reactor. A perforated Plexiglas distributor was located on the bottom of the bed to ensure uniform distribution of the gas stream. The principal role of humidification in the biofilters was to guarantee an aqueous phase for microorganisms. In fact, the removal of gas pollutant occurred in the liquid phase; therefore, the biofilter bed should be frequently humidified and the optimum moisture content should be kept in the range of 40-60%. The lower moisture in the biofilter bed caused the bed dryness, and a higher one induced the excess pressure drop. In this way, the inlet air to the biofilter should be humidified before entering the biofilter, and 500 ml/day of the nutrient solution should be added to the biofilter medium by a nozzle spray. On the other hand, these helped to provide sufficient moisture in the biofilter bed, which was important for the efficient performance of the biofilter. The moisture content of the biofilter bed was determined daily and kept almost constant. The sludge from the wastewater was used to inoculate the reactors. The experiments were carried out in the empty bed residence time (EBRT) of 100 s at room temperature (25±3 °C). In all of the reactors, the moisture content of the medium was kept in 40-60% to provide a suitable environment for microbial growth. Figure 1 shows the schematic diagram of the experimental setup.

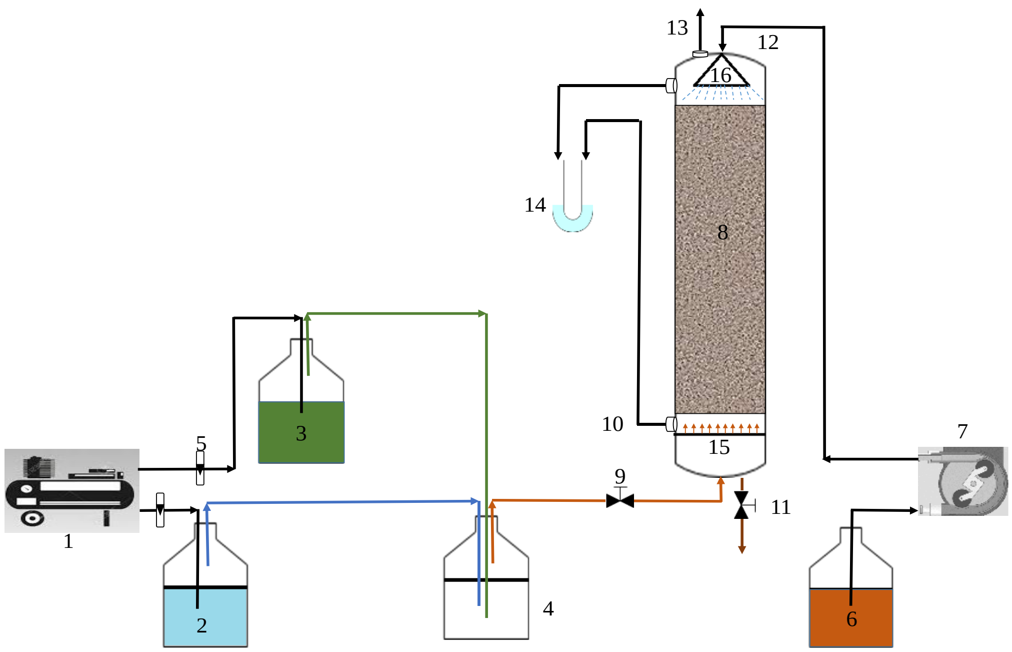

Fig. 1. Schematic diagram of experimental setup: Air compressor (1), Formaldehyde container (2), Humidifier (3), Mixing tank (4), Flowmeter (5), Nutrient solution (6), Peristaltic pump (7), Biofilter bed (8), Air sampling valve (9), Outlet gas stream (10), Effluent nutrient solution (11), Influent nutrient solution (12), Filter air outlet (13), Monometer (14), Distributor (15), Nozzle spray (16).

Table 1. Properties of packing materials

Packing	Compost	Woodchips	Zeolite	Activated carbon
Particle size (mm)	4-6	4-6	1-3	0.075
Dry bulk density (g/cm^3)	1.56	6.69	2.11	1.67
pH	6.65	8.75	7.75	6.50
Moisture content (%)	4.04	5.04	9.27	4.62
Porosity (%)	45	47	51	36
Surface area (m^2/g)	3.650	5.200	25.703	930.000

2.2. Nutrient solution

The nutrient solution consisted of the mineral materials that were dissolved in the distilled water. Table 2 shows the quantities of mineral materials in the nutrient solution.

Table 2. Mineral materials in the nutrient solution

Materials	**Concentration (mg/ml)**
NH_4Cl	1.670
KH_2PO_4	2.330
$MgSO_4$	0.067
$MnSO_4$	0.010
$CaCl_2$	0.167
$FeSO_4$	0.017
$ZnSO_4$	0.010

A suitable pH of the nutrient solution and aerobic sludge is necessary for the microbial activities. In this regard, an optimum pH of 7 was set using HCl or NaOH solutions.

2.3. Analytical methods

This study analyzed the performance of the biofilters via three parameters, including the removal efficiency (RE), elimination capacity (EC), and loading rate (LR), by using the pollutant´s concentrations in the gas-phase in the inlet and outlet gas stream. The NIOSH (National Institute for Occupational Safety and Health) method was used for calculating the formaldehyde concentration [7]. According to this method, the sampling of the inlet and outlet airflows were done individually and injected into a sodium sulfite solution (2%) as a formaldehyde absorbent medium during a half-hour period. Finally, the formaldehyde concentration was obtained via a UV-VIS spectrophotometer (T80+ model) in a 260 nm wavelength. The RE, EC, and LR can be calculated as follows:

$$\mathrm{RE}\ (\%) = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \tag{1}$$

$$\mathrm{EC} = \frac{F\,(C_{in} - C_{out})}{V} \quad (g/m^3\,.\,h) \tag{2}$$

$$\mathrm{LR} = \frac{C_i \times F}{V} \quad (g/m^3\,.\,h) \tag{3}$$

where F is the flow rate of the gas (m^3/h), C_{in} is the VOC concentration (g/m^3) at the inlet, C_{out} is the VOC concentration (g/m^3) at the outlet streams, and V is the volume of the packing material (m^3). The pressure drop of the bed was determined continuously by using a manometer that was connected to the inlet and outlet air flow. The moisture content of the packing materials was determined by gravimetric analysis. A mercury thermometer was applied for displaying the operating temperature of the biofilter. The pH of the nutrient solution and activated sludge was obtained with a pH-meter (R-pH model).

2.4. Mass transfer modeling in biofilters

The performance of a biofilter depends on the gas-phase mass transfer rate, which is an important part of the overall mass transfer rate. Determining the external mass transfer rate can help to design a biofilter. Moreover, it is necessary to determine the volumetric mass transfer coefficient in the gas-phase. In this regard, various correlations have been proposed to specify this parameter for special applications. A mass balance over a control volume of the biofilter bed was induced to Eq. (4) [26] with the assumption that the biological degradation reactions and liquid diffusion are so fast that the formaldehyde concentration at the gas interphase is nearly zero.

$$C_g.u.A\big|_x - C_g.u.A\big|_{x+dx} = k_g.a\,(C_g - 0)A.dx \tag{4}$$

Eq. (4) leads to Eq. (5) and Eq. (6) after rearranging:

$$\frac{1}{C_g}dC_g = -k_g.a.\frac{1}{u}.dx \tag{5}$$

$$\ln C_g = -(k_g.a).t_r + C \tag{6}$$

Where, u (m/s), A (m^2), C_g (mg/l), $k_g.a$ (1/s), and t_r (s*)* are gas velocity, biofilter cross-sectional area, formaldehyde concentration in gas-phase, volumetric mass transfer coefficient, and the residence time, respectively.

3. Results and discussion

3.1. Adaptation phase

This study investigated different packing materials in three biofilter configurations. Table 3 shows the configurations of the studied biofilters. In the first phase of the experiment (adaptation phase of the microorganisms), the performances of the biofilters were analyzed by introducing 20 mg/m^3 of inlet formaldehyde concentration at an EBRT of 100 s to each of the reactors. During the first days of the experiment, as illustrated in Figure 2, the RE obtained was about 100% for all of the systems; after three days, it declined to about 50%, indicating the pollutant absorption on the packing materials. After the adaptation of microorganisms to the formaldehyde and the propagation of microbial biomass, the RE increased, and finally, it was constant in the last days. The average removal efficiencies of 97.5%, 90%, and 93% were obtained at 100s empty bed residence time for the biofilter of configurations I, II, and III, respectively, while steady-state conditions were attained. The achievement of steady-state conditions in the biofilter of configurations II and III was faster than another biofilter. In a comparative study by Prado et al. (2004), the performance of three reactors packed with different inert packing materials demonstrated similar results for all of them, which is consistent with our obtained results [6]. Table 4 summarizes the results of the performance of the biofilters.

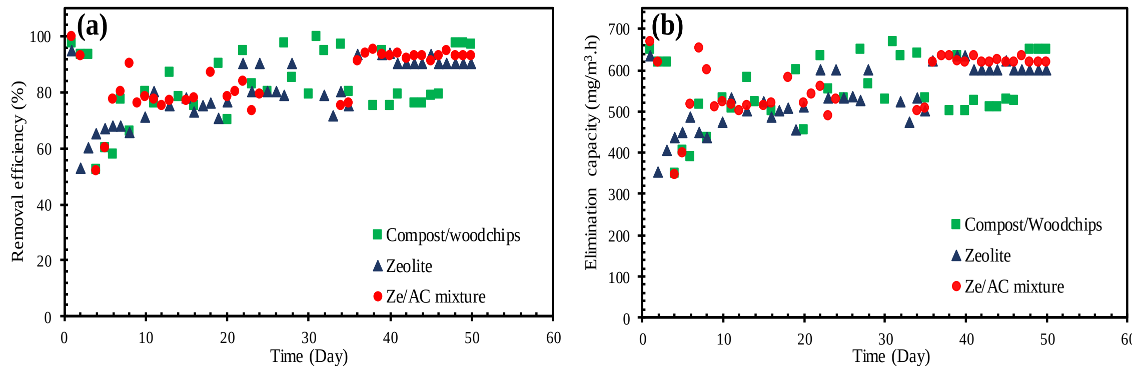

Fig. 2. (a) Removal Efficiency (RE) and (b) Elimination Capacity (EC) of the biofilters.

Table 3. Different configurations of the biofilters.

Biofilter of configuration	Packing material	Bed porosity (%)
I	Compost/Woodchips mixture	60
II	Zeolite particles	50
III	Zeolite/Activated carbon (Ze/AC) mixture	54.8

Table 4. Summary of results for each biofilter

Biofilter configuration	I	II	III
K_G.a (1/S)	0.0096	0.0132	0.0301
RE (%)	97.5	90	93.5
EC (mg/m^3.h)	650	600	623
ΔP (mmH_2O)	8	65	22

3.2. Effect of different EBRTs

The effect of the different EBRTs on the RE and EC of formaldehyde removal was investigated. In the second phase of the experiment, different EBRTs of 20, 30, 60, and 100s were used, while keeping the inlet formaldehyde concentration constant. An average RE was obtained for each of the biofilters in the EBRTs of 20, 30, 60, 100 s, which were tested for four days and achieved steady-state conditions. Figure 3 shows the effect of EBRT on the RE and EC of the systems. The RE increased and EC decreased with an increase of the EBRT, which was due to more contact time between the gas phase and biofilm layer, as well as a higher rate of formaldehyde transfer between them; therefore, mass transfer was enhanced. The maximum RE of 90 and 93.33% was achieved at 100 s of EBRT for the biofilter of configurations II and III, respectively; the maximum EC of 2840 mg/m^3.h was obtained at 20 s of EBRT for the biofilter of configuration II. Figures 3a and 3b show that in lower EBRTs, the zeolite particles perform better than that of the other biofilters. In this way, Lu et al. (2012) found that the RE decreased from 97% to 86.8% when the EBRT dropped from 113 to 22.6 s, while the EC increased [3]. In other work, Prado et al. (2004) presented a decline in the formaldehyde RE by increasing the formaldehyde EC from 41.2±15.8 to 111.8±15.2 g/m^3.h with a reduction of the EBRT from 71.9 to 20.7 s [6].

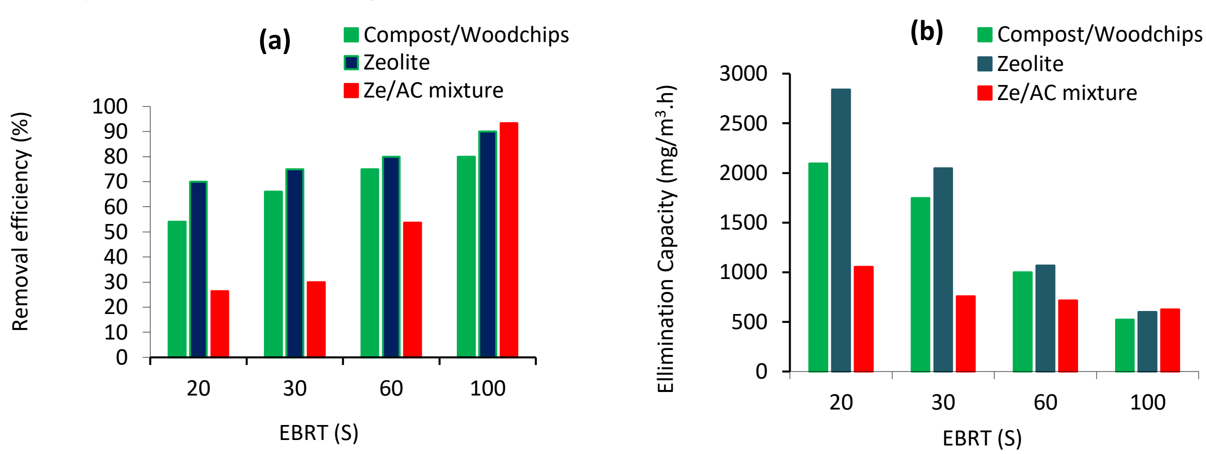

Fig. 3. Effect of different EBRTs on the performance of the biofilters **(a)** RE (%) and **(b)** EC (mg/m^{3}h).

3.3. Effect of different inlet formaldehyde concentration

The effect of inlet formaldehyde concentrations from 20 to 80 mg/m^3 on the performance of the biofilter was explored. A variation of inlet concentration led to shocking of the microorganisms. Figure 4 displays the variation of RE and EC in the different inlet formaldehyde loads. According to Figure 4, RE decreased and EC increased with increasing of the inlet formaldehyde concentration. For different inlet formaldehyde concentrations (loading rates), the average RE and EC were analyzed when the systems achieved steady state conditions. Higher toxicity was attained in the medium with increasing of the inlet formaldehyde concentration, which reduced the microbial activity. This explanation was in agreement with the results of Lu et al., 2012 [3]. Also, re 4 shows that with an increase in the formaldehyde concentration, the severity of changes in the RE of the biofilter of configurations II and III had a slower decline than another biofilter. However, in the high concentration of formaldehyde, the RE of the biofilter of configuration III was higher than the other biofilters. In Figure 4, the dashed line represents RE=100% and EC achieved to the maximum values, which is consistent with the results of Hu et al. (2015) [27]. This result confirmed that the biofilter of configurations II and III were perfectly performed. In another study, Wang et al. (2012) observed a similar pattern for the EC with an increase in the inlet toluene loadings. They obtained a desirable performance for the removal of high temperature toluene gas (55 ˚C) with low inlet loading conditions (< 100 g/m^3.h) [28] in a biofilter packed with granular activated carbon, in which a similar result was obtained for the shock loading process. Also, Chen et al. (2012) considered a tubular biofilter for the removal of toluene. They operated at an EBRT of 15 s and obtained an increase in the EC from 18.3 to 83.0 gm^{-3}h^{-1} and a decrease in RE in the range of 99% to 52.2% with an increase in the inlet toluene loads of 18.7 to 149.3 gm^{-3} h^{-1}. Their results showed that high elimination capacities were obtained for high organic loading rates under low pollutant concentrations [29]. Hajizadeh et al. (2017) reached an EC of 0.3, 0.65, and 1.2 g/m^3.h and an average RE of 91, 88, and 84% that corresponded to the concentrations in three ranges of 3.42-6.4, 9.29-12.35, and 18.5-22.3 mg/m^3.h, respectively [30].

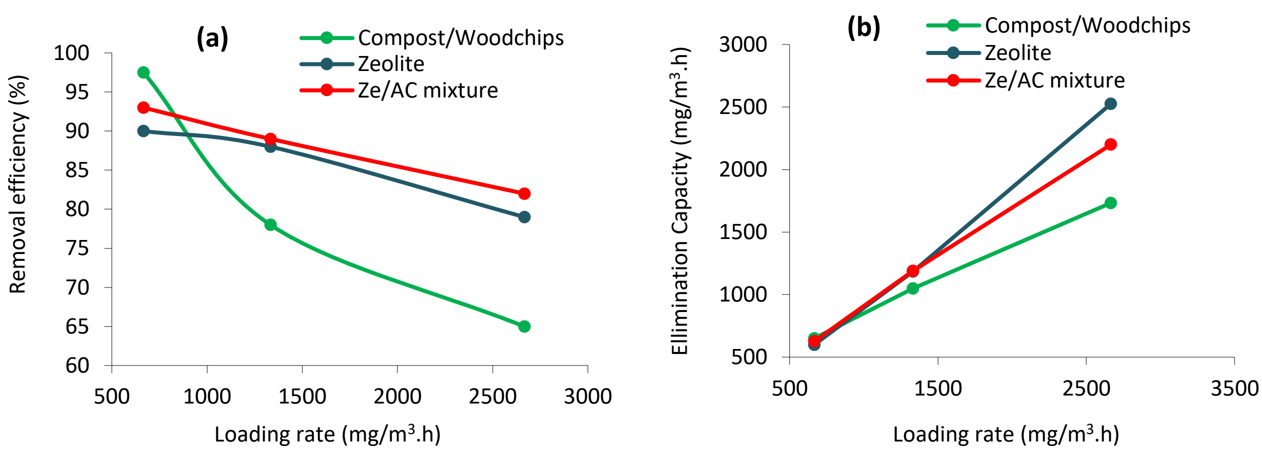

Fig. 4. (a): Removal efficiency (RE) and (b): elimination capacity (EC) versus formaldehyde loading rate for the biofilters.

3.4. Pressure drop

Figure 5 displays the effect of different gas flow rates on the pressure drop in the bed. The average pressure drop in the packed bed media can be estimated by the Ergun equation [31]. It can be seen that the experimental data of the pressure drop along the biofilter bed was fitted with polynomial regression as follows:

$$\Delta P = 0.194Q^2 + 2.7883Q + 1.5362 \quad (7)$$
(for compost/woodchips packing)

$$\Delta P = -0.4013Q^2 + 17.859Q + 29.153 \quad (8)$$
(for Zeolite particles packing)

$$\Delta P = 0.4058Q^2 + 1.8464Q + 14.932 \quad (9)$$
(for Zeolite/activated carbon mixture packing)

where ΔP is the pressure drop (mmH$_2$O) and Q is the air flow rate (Lmin^{-1}). These results are consistent with the results of Chou et al. (2009) and Lu et al. (2012) [31], [3]. The pressure drop in the bed of the biofilter depends on several parameters such as the moister in the bed, air flow rate, and the interior packing situation of the biofilter [3]. The pressure dropped with an increase in the moister bed and air flow rate in the bed or clogging of the bed due to packing material compression, and therefore, the energy consumption of the air compressor increased [3]. Figure 5 shows that the pressure drop, due to a lower porosity, in the biofilter of configuration II is higher than that of the other biofilters. Therefore, it suggests that the use of larger particles of zeolite prevents the high-pressure drop in the zeolite biofilter bed. This can be proved with the Ergun equation, which proposes that the pressure drop is inversely proportional to a function of particle diameter and is in agreement with the results of Keyser et al. (2006) [32].

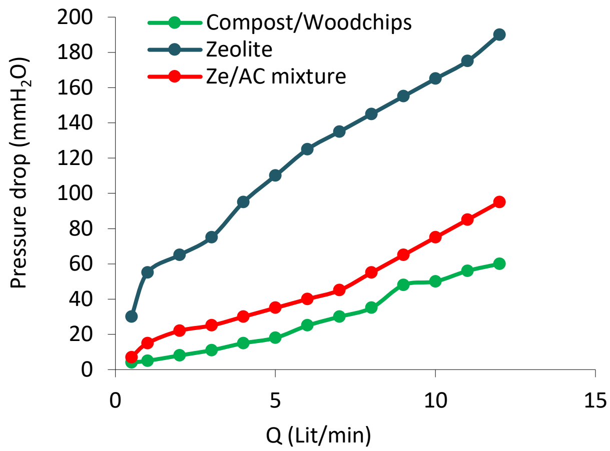

Fig. 5. Pressure drop in different gas flow rate for the biofilters.

3.5. pH effect on the performance of the biofilter

The pH values for an effluent nutrient solution were determined daily. Figure 6 shows the pH changes during the operation of the biofilters. The optimum pH for the removal of formaldehyde was reported to be about 7 [6]. Therefore, the pH in the inlet nutrient solution was set as neutral (pH =7), while the pH of the outlet was decreased during the operation. Because the occurrence of the metabolite reaction in the biofilm layer led to the production of formic acid, the medium became acidic. It was induced by reducing the pH in the effluent nutrient solution. In this study, the pH of the effluent nutrient solution was reduced from 7 to about 5 for the biofilters.

In comparison, similar results were obtained by studies of Cho et al. (1991) on a pilot-scale peat biofilter to remove malodorous gases [33] and Chung et al. (1996) on *a Thiobacillus thioparus* CH_{11} biofilter for H_2S removal [34].

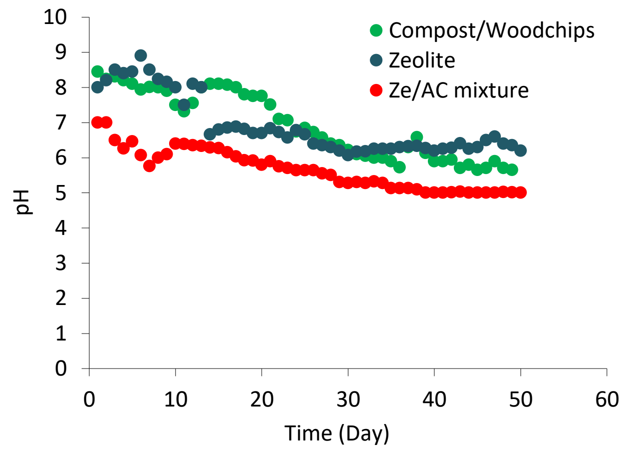

Fig. 6. Variation of pH for the biofilters.

3.6. Mass transfer coefficient

Figure 7 shows the plot of Ln C_g versus time, and the linear fitting of the data produce a line with the slope of the volumetric mass transfer coefficient in the gas-phase.

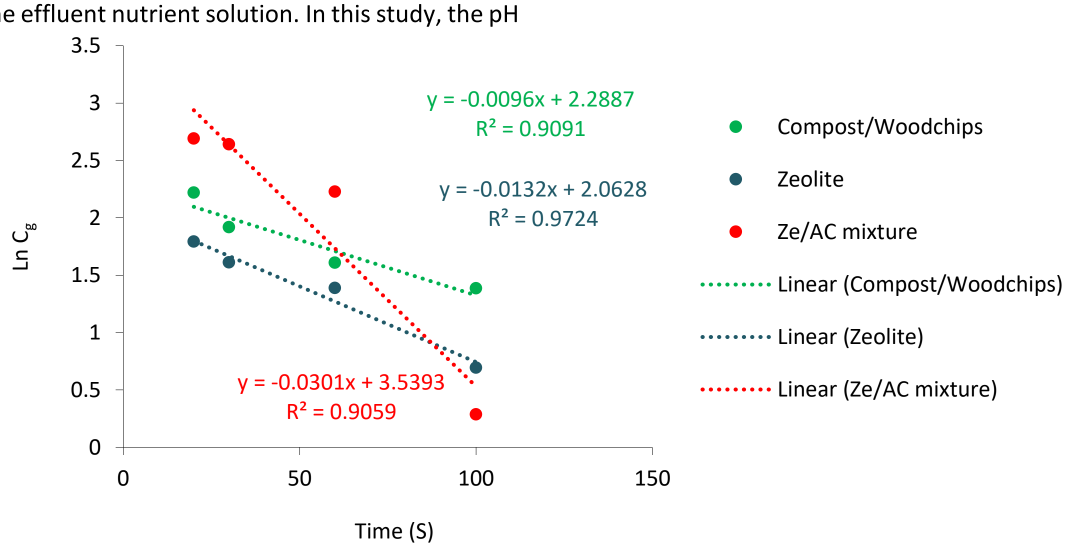

Fig. 7. Ln C_g in different gas retention times.

$$\text{Compost and woodchips biofilter:} \quad \text{Ln Cg} = -0.0096\,t + 2.2887 \tag{10}$$

$$\text{Zeolite biofilter:} \quad \text{Ln Cg} = -0.0132\,t + 2.0628 \tag{11}$$

$$\text{Zeolite /activated carbon mixture biofilter:} \quad \text{Ln Cg} = -0.0301\,t + 3.5393 \tag{12}$$

The slopes of Eqs. (10), (11), and (12) are the volumetric mass transfer coefficients ($K_g.a$) in the gas-phase for the biofilters, which are 0.0096, 0.0132, and 0.0301 s^{-1} for the configurations I, II, and III, respectively. As a result, a higher volumetric mass transfer coefficient presents a more efficient performance of the biofilter in the removal of formaldehyde. Also, the correlation of the volumetric mass transfer coefficient calculation helps to design a similar biofilter [26].

4. Conclusions

In this study, the performance of three biofilters packed with different materials was investigated. The results showed that the formaldehyde could be removed from the contaminated air with a high removal efficiency via all of the biofilters. However, the biofilters packed with zeolite and the zeolite/activated carbon mixture eliminated the formaldehyde from the contaminated air in a shorter time and obtained a higher mass transfer coefficient. In the high loading rates, a maximum RE of 82 % was achieved for the biofilter packed with the zeolite/activated carbon mixture. Also, in the lowest EBRT, a maximum EC of 2840 mg/m^3.h was obtained for the biofilter packed with zeolite. Generally, it was concluded that the performance of the zeolite/activated carbon mixture was better than that of the zeolite as the packing material, which was shown in the adaptation phase and when the concentration of the pollutant was high. But in some cases, e.g., lower EBRT, the zeolite performance was better than that of the zeolite/activated carbon mixture.

Acknowledgments

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