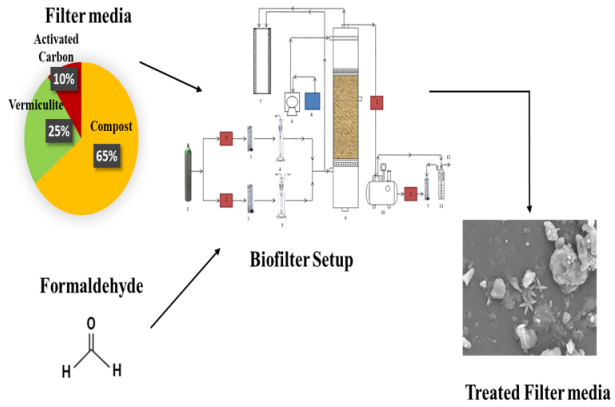
Global NEST Journal, Vol 26, No 3, 05622

Formaldehyde degradation using biofilter loaded with compost, vermiculite and activated carbon: performance and optimization

R. Malathi and J. Jeyanthi*
[1]Department of Civil Engineering, Government College of Technology, Coimbatore - 641013, Tamil Nadu, India
Received: 01/12/2023, Accepted: 11/01/2024, Available online: 17/01/2024
*to whom all correspondence should be addressed: e-mail: j.jeyanthigct@gmail.com
https://doi.org/10.30955/gnj.005622

Graphical abstract



Abstract

Formaldehyde is one of the typical indoor pollutants causing the prevalence of acute health issues among those residing in indoor environments. Biofiltration is a commonly adopted technique to treat indoor air pollutants. In this investigation, a biofilter was fabricated and compactly filled with compost, vermiculite and activated carbon (65:25:10) as a media composition for enhanced formaldehyde degradation. The influence of formaldehyde inlet concentration (0.01 – 1 gm^{-3}) and EBRT (30 – 180 s) was studied to evaluate the biofilter performance. Their progression was assessed with dynamic operations in five phases with two stages each. The formaldehyde removal was achieved between the range of 32% to 98% with regular intervals of EBRT at 30, 60, 90, 120, 150 and 180 seconds, which depend upon varying their flow rate and inlet loading rate. The biofilter performance lasted for over 202 days with a 98% formaldehyde removal and 23 g/m^3h^{-1}biofilter capacity under the maximum EBRT (180 s) and inlet formaldehyde concentration (1 gm^{-3}). The experimental findings demonstrated the efficacy of biofilter in withstanding substantial formaldehyde loading rate while maintaining an acceptable EBRT. The primary microbe responsible for formaldehyde degradation was identified to be as gram-negative bacteria.

Keywords: Biofilter; EBRT; Elimination capacity; Formaldehyde; Inlet concentration; Removal efficiency.

1. Introduction

Indoor air pollutants have increased with the extension of industrial activities, manmade activities and degradation of natural resources. The escalation of pollutants can be attributed to inadequate air circulation and the rising concentration of certain indoor pollutants emitted from sources under specific temperature and humidity conditions. Typical indoor pollutants found inside a room are carbon-monoxide (CO), oxides of nitrogen (No_x), particulate matter (PM) and volatile organic compounds (VOCs) (Talaiekhozani *et al*., 2021). Among these indoor pollutants, volatile organic compounds have been produced more in modern homes and from all manufactured products (i.e., detergent, perfume, cosmetic products, repellents, manure etc.) used for regular work (Zhong *et al*., 2020; Wu *et al*., 2003). Moreover, the primary ingredients of many household products contribute to indoor air pollutants when they form vaporized gases at room temperature. Frequently found VOCs indoors are toluene and styrene (Dewidar *et al*., 2022), xylenes (Li *et al*., 2020), benzene and ethylbenzene (Sun *et al*., 2020) and formaldehyde (Mohammed *et al*., 2015). The predominant VOCs are formaldehyde; released into the atmosphere from various industries such as wood-based products, resin manufacturing, fuel burning, adhesive, and cosmetic products (Halios *et al*., 2022). Exposure to formaldehyde causes eye-burning sensation, throat irritation, and other illnesses like vomiting sensation, difficulty breathing, coughing, wheezing, skin rashes, and allergic reactions. It is colorless and emits strong-smelling gas (Rezaei *et al*., 2015; Prado *et al*., 2004).

New technologies have emerged to control indoor air pollutants, such as the Photo catalyst method (Robert & Nallathambi, 2021; Huang *et al*., 2016), Adsorption (Robert & Nallathambi, 2021; Lu *et al*., 2010), Phyto-remediation (Shao *et al*., 2020), Membrane bio-filter (Robert & Nallathambi, 2021), Biodegradation (Lu *et al*., 2012) and Bio-filtration (Sheoran *et al*., 2022; Dorado *et al*.,2010). Among these technologies, bio-filtration is the most predominant technology for controlling organic and

inorganic indoor air pollutants. The biofilters are loaded with materials such as compost, perlite, wood chips, etc., that have the ability for microbial degradation ofthe organic pollutants and convert them into carbon dioxide and water substances (Fulazzaky *et al*., 2016). Studies reported on the biofilter performance were developed using a laboratory-scale biofilter model. The control of concentration is a primary concern in the biofilter treatment while utilizing it for the VOC removal from the polluted gas stream (Prado *et al*., 2004). According to Zilli *et al*. (2001), the choice of filter media should be determined by cost-effective technology and poorly insoluble materials in the waste air stream to degrade VOC concentration. The main advantages of filter media are that it uses low-cost materials, wastes organic products, is enriched with more nutrients and carbon content, has more water-holding capacity, and is easily decomposed. However, the filter media's drawbacks exist, which are creating odour and fly nuisance, lengthy process and more land area required.

Some filter media support materials that are utilized in enhancing the biofilter performance are woodchip (Rezaei *et al.,* 2015), compost and perlite (Parseh *et al*., 2021), barks (Sheoran *et al*., 2022), leaves, soil, peat (Zilli *et al*., 2001), ceramics (Saravanan *et al*., 2015), agriculture waste (Singh *et al*., 2006), industrial waste (Saravanan *et al*., 2015) and some other inert materials (VanOsdell *et al*., 1996) are used in these applications. The filter media may contain either single or mixed materials (i.e., composite), whichenhances biofilter performance (Kumar *et al*., 2019; Rezaei *et al.,* 2015). Furthermore, studies on biofilters packed with compost-based media and activated carbon have supported the increased adsorption of higher concentrated pollutants (Ondarts *et al*., 2012). Compost as a filter media is a natural medium that is very efficient in biofiltration. It can produce more microorganisms and natural nutrients. It is a readily available and economical material with a water retention capacity. Similarly, vermiculite as a filter media holds more water content and maintains a constant moisture content in the biofilter column. Activated carbon as a filter media, even in its lower proportionate, can be used as a potential adsorbent to remove formaldehyde from real-time experiment studies. It can be used as a steady state condition of biofilter.

The present study employed an assortment of compost, vermiculite and activated carbon as filter media packing materials. Compost is an economical material with nutrient-rich organic content, a wide surface area and a porous surface that can provide a favourable environment for microbial growth. Moreover, it can absorb gases, thereby aiding in mitigating gaseous pollutants (Yuan *et al*., 2019; Gallastegui *et al*., 2011). Likewise, vermiculite is a mineral enriched with silica, capable of absorbing more water, thereby aiding in maintaining the saturated moisture content for longer in a biofilter. Moreover, it can absorb organic pollutants and act as a biocarrier in the biofilter (Wen *et al*., 2016). Similarly, activated carbon is an inert material providing enough surface area to enhance the degradation of gaseous formaldehyde. It serves as a carbonaceous substrate that facilitates the proliferation of microorganisms. It also helps maintain the biofilter media's stability (Kumar *et al*., 2019).

The present communication reported the design of the laboratory-scale biofilter model and its performance in biodegrading formaldehyde gases. The biofilter is compactly packed with compost (65%), vermiculite (25%) and granular activated carbon (10%).Biofilter performance was assessed by evaluating the amount of degradation of synthesized formaldehyde gases with the effect of varying inlet concentration and EBRT (Empty Bed Residence Time) under different loading conditions. The morphology of treated filter media was assessed to visualize the microbial growth and the grown microbial population was measured.

2. Materials and Methods

2.1. Reagents used

An experimental lab-scale biofilter model utilized formaldehyde 37% solution in the vapor form purchased from Merck. The 2,4- Dinitrophenyl Hydrazine (Derivative agent), Ortho Phosphoric acid, Sodium Sulfite (absorbance solvent), Potassium hydroxide pellets andHydrochloric acid (0.1N) & Sodium hydroxide (0.1N) for neutralizing the pH value of activated carbon were also purchased from Merck. Ultrapure water (Milli-Q) of 18.2 MΩ cm resistivity was utilized for preparing the reagents that are used throughout the research.

2.2. Experimental biofilter setup

Experimental studies were conducted in the Biofilter (laboratory scale model), the schematic diagram illustrated in Fig. 1. The Biofilter comprises a 60 cm long transparent column with an internal diameter of 9 cm made up of Poly acrylic material. The formaldehyde vessel is incorporated with a humidified vessel that contains formaldehyde solution and water, respectively and zero air was fed into both vessels. The formaldehyde gas is fed into the biofilterthrough the PVC hose pipe. Filter media was filled with a height of 30 cm and enclosed with a 15 cm depth for the inlet and outlet provided at the bottom and top, respectively. A stainless-steel sieve plate (1mm $\times$ 1mm $\times$ 1mm) was placed on both sides of the filter media, which supports the filter media package and circulates the air stream with uniform distribution throughout it.

Sampling ports were provided at the filter media portion. Similarly, the manometer was connected at the biofilter'sinletand exit to monitor pressure drops continuously. In the experimental studies, pressure drops were set as equal in inflow and outflow of cm of H_2O. Temperature and filter media characteristics were also analyzed. Formaldehyde vapor was obtained from the biofilter outlet using a pump with an attached impinger and their flow was controlled using a rotameter setup. Impinger was filled with 1% sodium sulfite solution to absorb formaldehyde gases (Hajizadeh & Rezaei, 2014). Formaldehyde is removed from the biofilter column using filter media; the polluted stream gas (i.e., formaldehyde) utilizes the filter media using the stability of microbial metabolism activities. Inside the biofilter column, formaldehyde gas is absorbed by filter media in the given

designed EBRT, and treated gases are expelled from the outlet chamber. Finally, pollutant removal efficiency with the degradation rate under operating conditions is analyzed. The biofilter was operated at room temperature throughout the entire system. The nutrients were sprinkled in the filter media every day after achieving steady state condition of quantity depending on the rate of microbial production and moisture content. An impinger is provided at the outlet to collect outlet air from the bioreactor. Excess drain water is discharged from the filter media through the bottom chamber.

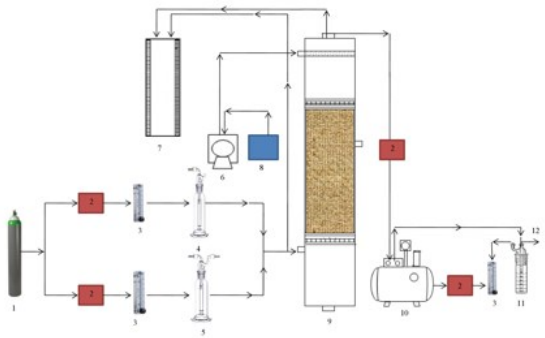


Figure 1. Schematic sketch of laboratory scale Biofilter model: 1. Zero Air Cylinder, 2. Flow Control Valve, 3. Flow regulator, 4. Humidifier, 5, Formaldehyde solution, 6. Peristaltic pump, 7.U-tube manometer, 8. Nutrient Tank, 9. Bioreactor, 10. Pump, 11. Impinger and 12. Sample collector

For longer-run maintenance of the biofilter column, it is important to ensure that the filter media is not clogged due to the production of microbial population. Moreover, it forms a biofilm on the filter media's surface, creating an odour nuisance during their operation. When the nutrients are added to the biofilter column, the filter media absorbs the required quantity, and the remaining liquid content in the inlet chamber can be removed using a drain valve.

2.3. Compositions of Filter media

Packed materials were chosen in three manners: organic, inorganic and inert. Filter media is prepared as a mixture of proportions (65:25:10) of Compost, Vermiculite and Activated carbon (Mathur *et al*., 2007). The compost was collected from the Environmental Sludge Management System, Government College of Technology, Coimbatore. Before filling the biofilter, the compost was kept in air-tight plastic bags at indoor room temperature in its indigenous saturated condition. It was further sieved according to the required size (4 mm to 7 mm). In these experimental studies, compost is preferred due to the increase in microbial accumulation rate before and after treatment with the ability to degrade formaldehyde concentration (Rezaei *et al*., 2015). Vermiculite is siliceous, absorbs more water, and maintains the saturated moisture content for longer in the biofilter. The vermiculate's particle size is adopted between 2mm to 4mm (Długosz & Banach, 2018). Activated Carbon (AC) was prepared with different mixing compositions (0.5:1, 1:1 & 1.5:1) of potassium hydroxide and coconut charcoal. Finally, the activated mixture was washed with NaOH and HCl until a pH value of neutrality was attained. The dried AC was sieved in the required size (4 mm to 5 mm). AC was added with compost and vermiculite mixtures due to its adsorbent capability toward formaldehyde (Ondarts *et al*., 2012). The filter media's particle size is averaged as 4 mm. Different sieve sizes are used in the filter media, such as Compost (8.00 mm, 6.3 mm and 5.60 mm), Activated carbon (5.60 mm and 4.00 mm) and Vermiculite (4.00 mm, 3.35 mm and 2.36 mm). Compost and vermiculite are easily available natural and sustainable materials with low cost and small proportionate of activated carbon is added to enhance its effective use as an adsorbent of pollutant. The physical and chemical characteristics studies on filter media were determined to reveal their suitability for pollutant removal.

2.4. Mineral media solution

An adequate quantity of nutrient solution was sprinkled periodically on the upper portion of the filter media twice daily using a peristaltic pump. It was supplied to maintain moisture content and microbial growth rate for increased formaldehyde degradation (Rene *et al*., 2005). The nutrient solution's composition used periodically concerning the filter media's moisture content is as follows: Dipotassium hydrogen phosphate (0.8 g/L), Potassium dihydrogen phosphate (0.2 g/L), Gypsum (0.05 g/L), Magnesium (II) sulphate heptahydrate (0.5 g/L), Ammonium sulphate (1.0 g/L) and Ferrous sulphate (0.01 g/L). The nutrient solution's pH was maintained at 6.5 to 7.5 under standard room temperature (Lu *et al*., 2012).

2.5. Instrumental analysis

The effluent formaldehyde samples were measured using a Gas Chromatography (Agilent 8860) equipped with an FID fused silica capillary column of GC length 30 m, diameter 0.25 mm, and film 0.25 µm, with carrier gas as Helium. Initially, the column was operated at oven temperature is 60°C for 3 min and raised upto 230°C. Similarly, the detector and injector were continued at 280°C and 250°C, respectively. The samples were filtered using membrane filtration (pore size 0.45 µm) before injecting the samples. A moisture analyzer (Wensar HMB100) determined the moisture content of the bed filter media and it further confirmed with the conventional oven drying method. Both studies showed similar values for filter media's moisture content. A U-tube manometer monitored the pressure variations of the biofilter chamber during the continuous operating condition. The pH meter (Hanna pHep HI98107) measured the pH of the filter media and the sampling solution. The temperature was monitored continuously and measured using a mercury thermometer (1°C/ min). CO_2 concentration was measured using a CO_2 analyzer sensor (Testo 440 dP). The CO_2 generated during the biofilter's operation was analyzed at their inlet and outlet chamber. Filter media characteristics studies were observed from their textural morphology using Scanning Electron Microscopy (SEM) and from ultimate and physical analysis.

2.6. Evaluation of biofilter and their design parameters

The biofilter's efficiency was evaluated from the obtained removal efficiency (%) of formaldehyde and biofilter

media's elimination capacity (g/m^3/h)) (Ondarts *et al.*, 2012), which are measured using the following equations.

$$EBRT = \frac{Va}{Q} \quad (1)$$

$$\tau = EBRT * \xi \quad (2)$$

where Q is the gas phase formaldehyde flow rate (m^3/h) and V is the filter bed volume (m^3). τ is the actual filter bed residence time measured (min) and ξ is the filter bed porosity.

The experimental designed parameters studied were the inlet loading rate (IL, g/m^3/h), Removal efficiency (RE, %) and elimination capacity (EC, g/m^3/h). It was used for determining the relationship between the inlet and outlet formaldehyde concentration, Q and Va.

$$IL = \frac{Q * C_i}{V} \quad (3)$$

$$EC = \frac{Q*(C_i - C_o)}{Va} \quad (4)$$

$$RE = \frac{C_i - C_o}{C_i} * 100 \quad (5)$$

The biofilter's performance studies were conducted for around 6.6 months at various operating conditions with an arrangement of up-flow air flow movement with a mixture of compost-based materials. Investigation of the biofilter's performance was assessed in five phases: Phase I, II, III, IV and V, with 2 stages in each phase. At each phase level, the inlet formaldehyde concentration was varied (0.01, 0.03, 0.1, 0.5 and 1.0 gm^{-3}). Similarly, the EBRT was varied (30 – 180 s) for each stage under each phase with an interval of 30 s under dynamic operating conditions of volumetric flow rate and inlet loading rate. Each phase operated continuously until a steady-state condition was achieved. In real-time, formaldehyde was used with lower concentrations in the working environment or industrial area because of health concerns. A lower formaldehyde concentration was chosen for this research based on the real-time work.

3. Results and discussion

3.1. Characterization of filter media

The physical properties of filter media (Table 1) were analyzed to study the suitability of biofilter system utilization. The physical properties of compost (dry weight) were observed to have 40% moisture content of majority of other materials, 1.8 specific gravity and an organic content of 75% or more. The mechanical behavior of filter media was achieved by maintaining their size to be 2 – 4 mm. The void fraction of the filter media materials (> 50%) was considered suitable for the biofilter operation, which might induce low-pressure drops.

Table 1. Physical properties and ultimate analysis of filter media

Properties	Media Composition (65:25:10)		
	Compost	Vermiculite	AC
Physical Properties			
Moisture content (%), M.C	40	30	6
Wet filter media weight (g), W_1	826	308	101
Dry filter media weight (g), $W_2 = W_1/(1+M.C)$	590.00	236.92	95.28
Diameter of biofilter column (cm), D	9	9	9
Height of filter media (cm), H	19.5	7.5	3
Volume of filter media (cm^3)	1240.53	477.13	190.85
Specific Gravity, G_s	1.8	1.6	-
Particle density	-	-	0.8
Void fraction of filter media (%)	52.44	50.34	50.07
Chemical Properties			
Carbon	40.25	25.88	42.77
Oxygen	57.08	45.34	51.06
Hydrogen	2.1	2.0	1.28
Nitrogen	0.3	3.55	0.09
Sulphur	0.27	1.54	-

The ultimate analysis of compost, vermiculate and AC (Table 1) was examined from the Elemental analyzer. The presence of oxygen and carbon was noted in vermiculate (Długosz & Banach, 2018) as well as AC (Mohammed *et al.*, 2015) and compost (Mathur *et al.*, 2007).Therefore, the filter media holds enough oxygen content for microbial growth, thereby increasing the formaldehyde degradation. The carbon content in the filter media enhances the adsorption of formaldehyde gases. Moreover, the nitrogen and sulphur content in the compost enhances the microorganism growth. Therefore, the ultimate analysis of filter media showed their suitability to be a significant composite filter media material. Consequently, the removal efficiency was increased.

3.2. Dynamic operation of biofilter

The biofilter's operational process was segmented into five separate phases, as shown in Fig. 2. The results were observed from every phase to evaluate performance studies of the biofilter column on two different stages. Inlet concentration and EBRT were gradually increased in each phase and stage, respectively. Table 2 shows the observed operational condition of experimental results.

3.2.1. Phase I

Under Phase I, the inlet loading rate of formaldehyde gas was sustained at 1.2 g/m^{3}h^{-1}, varying the flow rate of formaldehyde at 0.19, 0.20, 0.21 and 0.22 m^3/h. With the above condition, EBRT of 30 seconds and formaldehyde concentration of 0.01 gm^{-3} was started, as revealed in Fig.2. Varied gas flow rate and inlet loading rate influenced thebiofilter's RE and EC under dynamic conditions. Physical adsorption (70%) was achieved on the fourth day in the filter media, representing their pore size capability. Chemically, the formaldehyde breakdown occurred after an hour of filter media's effective contribution in maintaining moisture and temperature. Moisture content was maintained at 40% to 60% throughout the bioreactor column. All the test procedures were conducted under non-sterilized conditions. The stability of formaldehyde concentration was studied in Phase I during the experimental duration of 47 days. The RE and EC of formaldehyde are 92% and 1.105 g/m^3/h, respectively, for 30 s EBRT (Fig. 3). However, as Phase I neared completion, the removal efficiency decreased to70% on the 45th day.

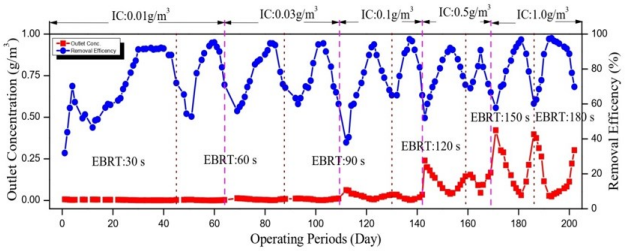


Figure 2. Impact of EBRT on Removal Efficiency and Outlet Concentration (inlet concentration – 0.01 to 1.0 gm^{-3})

Table 2. Operational phases of biofilter

Operational Phase	Stages	Formaldehyde flow rate (m^3/h)	EBRT (s)	Formaldehyde concentration (gm^{-3})	Removal efficiency (%)	Operating period (days)
I	I	0.19 - 0.22	30	0.01	92.1	1-47
	II	0.10 - 0.11	60		95.2	48-68
II	I	0.10-0.11	60	0.03	94.7	69-91
	II	0.07 -0.08	90		94.7	92 -111
III	I	0.07	90	0.1	94.0	112-131
	II	0.05	120		97.0	132-142
IV	I	0.05	120	0.5	92.0	143-160
	II	0.04	150		90.8	161-170
V	I	0.04	150	1.0	96.8	171 -186
	II	0.03 – 0.04	180		97.6	187-202

Moreover, the biofilter's operating conditions were rearranged by varying EBRT due to a drop-down of removal efficiency in Stage I of Phase I. Subsequently, EBRT was increased to 60 seconds while maintaining constant inlet concentration. The increased EBRT instigated decreased gas flow (0.11 m^3/h) and inlet loading rates (0.6 g/m^{3}h^{-1} (Fig. 3). This result showed good RE due to increased EBRT, favoring bacterial acclimatization. However, on the 60th day, RE and EC were obtained as 95% and 0.571 g/m^{3}h^{-1}, respectively (Fig. 3). Stage II of Phase I lasted 48 to 68 days.

Based on the experimental findings, lower inlet concentration and minimum EBRT were not effectively operating in the biofilter, which could not produce sufficient substrate of microbes. Moreover, the flow rate was reduced from 0.22 m^3/h to 0.11 m^3/h. During this period, it was observed that the filter media exhibited significant formaldehyde adsorption when subjected to a longer duration of exposure at a low loading rate of 0.6 g/m^{3}h^{-1}.

3.2.2. Phase II

Phase II lasted from 69 to 111 days. The formaldehyde concentrationfed into the bioreactor increased to 0.03 gm^{-3}with a maintained EBRT of 60 seconds in Stage I. During this stage, the gas flow rate was sustained constantly, thoughthe loading rate was increased from 0.6 to 1.8 g/m^{3}h^{-1}. The abrupt upsurge in the loading rate reduced removal efficiency from 95.2% to 56%. Subsequently; itobtained an efficiency of 94.7% on the 12th day after reaching a steady-state condition.

Under Stage II, 0.03 gm^{-3} formaldehyde concentration was fed into the bioreactor with varied EBRT from 60 to 90 seconds. During this stage, the bioreactor encountered a reduction in loading rate from 1.8 to 1.2 g/m^3 h^{-1}, while the gas flow rate decreased from 0.11 to 0.07 m^3/h.These reductions lead to an abrupt increase in EBRT, causing a shock effect. At the end of Stage II, a similar efficiency was achieved on the ninth day. These results found that the experiment survived more acclimation periods, which are adequately maintained at a minimalflow rate and loading rate. The elimination rate was almost similar to those two stages of this phase at 1.704 g/m^3/h (Stage I) and 1.136 g/m^{3}h^{-1} (Stage II).

3.2.3. Phase III

Similar experimental procedures were followed during Phase III except for feeding elevated inlet concentration from 0.03 to 0.1 gm^{-3}. The flow rate of formaldehyde is kept

at 0.07m^3/h with a consistent EBRT of 90 seconds. Moreover, the average loading rate of stage I and II is 4 g/$m^3h^{-1}$and 3 g/m^3h^{-1}, respectively. Initially, changes were observed in the biofilter's performance and instability in the microbial growth due to a rapid increase in the inflow concentration. Later on,the same concentration was fed in Stage II, which triggered the removal efficiency upto 94% and 97% on the 123rd and 137th day, respectively. This might be due to the decreased gas flow rate and inlet loading rate, resulting in increased biomass concentration stability. Moreover, the EBRT elevation from 90 to 120 seconds was crucial in declining the inlet loading rate and maintaining appropriate moisture content (52%), thereby increasing removal efficiency in Stage II. Finally, 97% removal efficiency was achieved with a higher degradation rate of microbes.

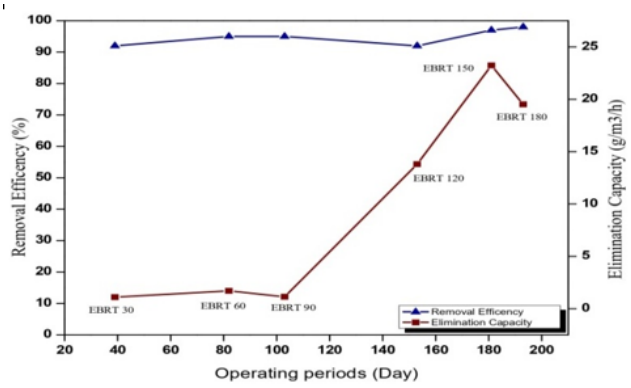


Figure 3. Impact of EBRT on Removal efficiency and Elimination capacity: Inlet concentration 0.01gm^{-3} to 1.0gm^{-3}

3.2.4. Phase IV

In Stage I of Phase IV, the inlet formaldehyde concentration is elevated to 0.5 gm^{-3}and EBRT is maintained at 120 seconds. As a result, the inlet loading rate was increased by 80%, resulting in decreased removal efficiency (92%). The reduction in RE might be due to the sudden impact of a higher inlet loading rate on the filter media. However, the EC of formaldehyde obtained was the maximum of 13.800 g/m^3h^{-1}.

In Stage II, the EBRT was raised to 150 seconds, resulting in varied inlet loading rate and gas flow rate (0.04 m^3/h), further impacting the removal efficiency (90.8%). However, during this Stage II operation, the RE was achieved earlier on the 165th day due to attaining optimum moisture content in filters media throughout this period. Therefore, it was noted that the EBRT's increment might have increased the capacity of the degree of degradation of formaldehyde. Consequently, the microbial growth was more sustained in the next phase with an EBRT of 150 seconds.

3.2.5. Phase V

In Phase V, the formaldehyde's inlet loading rate decreased by 17% from 24g/m^3/h to 20g/m^3/h. As in the above stage, the gas flow rate (0.04 m^3/h) into the biofilter was maintained constantly. The biofilter receives a maximum inlet concentration of formaldehyde (1.0 gm^{-3}). Phase V lasted from the 187th to the 202nd day. It was observed that the inlet loading rate showed an indirect influence on the biofilter's efficiency. For instance, an increased loading rate decreased the biofilter's performance. During this phase, EBRT was maintained at 180 seconds, which aids in attaining 98% removal efficiency and a maximum EC of 19.52g/m^3h^{-1}. In Stage II, 63% removal efficiency was obtained initially and rapidly increased in a short period, possibly due to the increased rate of microbial degradation.

3.3. Inference on Performance Outcome of Biofilter

Biofilter performance was analyzed regarding formaldehyde EC and RE under dynamic conditions in different phases. The results revealed that both parameters were assessed with increased microbial rate. The amount of formaldehyde degradation in the biofilter was evaluated based on the varied EBRT (30 to 180 seconds) in each stage. The increased inlet loading rate negatively impacted formaldehyde degradation. Moreover, the corresponding degradation rate was represented in terms of variation in the lower to higher level inlet concentration. From these observations, the result obtained from the biofilter is more compactable and sensitive for present filter media and other operational inlet loading load and EBRT.The biofilter achieved a maximum RE of 98% when operated with 180 seconds EBRT and 1.0 gm^{-3}initial concentration. Also, the biofilter maintained 52% moisture content under 25℃.

3.4. Influence offormaldehyde loading rate

The biofilter was fed with varied inlet concentrations of formaldehyde from 0.01 to 1.0 gm^{-3} and their corresponding performance was evaluated (Fig. 4). The inlet loading rate of the biofilter is directly impacted by either increased or decreased inlet concentration of formaldehyde (Rene *et al*., 2005). The inlet loading rate (1.2 g/m^3h^{-1}) was observed to be similar for the initial phases (i.e., Phase I and II). Due to the variations in inlet loading rate, there is a corresponding decrease and increase in gas flow rate and filter media's volume. Moreover, the loading rate was observed to have increased due to the population growth of microbes. This increase was attributed to the long-term response of the filtered media, which facilitated the enrichment of bacteria capable of maintaining their survival. Subsequently, the removal of pollutants was supported during this process. Later, increased pollutant removal was achieved earlier with more bacterial survival responses in filtered media.

In each phase, removal efficiency losses were higher in their corresponding Stage I, which may be due to the sudden variations in the inlet loading rate (0.6 – 24 g/m^3h^{-1}) that might have increased the microbial growth.

The variations in inlet loading rate in response to formaldehyde concentration and EBRT were depicted in Fig. 4. During Phase III, 3.0 g/m^3h^{-1}IL rate yielded an adequate EC of 2.910 g/m^3h^{-1}, which may be due to the increased EBRT of more than 90 seconds (Rezaei *et al*., 2015). Finally, in Phase V, the maximum EC of formaldehyde was achieved to be 19.52 g/m^3h^{-1} in the biofilter at the IL rate of 20 g/m^3h^{-1}. Therefore, the EBRT of more than 90 seconds (i.e., 90 – 180 seconds) and inlet

concentration above 0.1 g/m^3 were the best operating conditions for obtaining efficient and earlier removal efficiency.

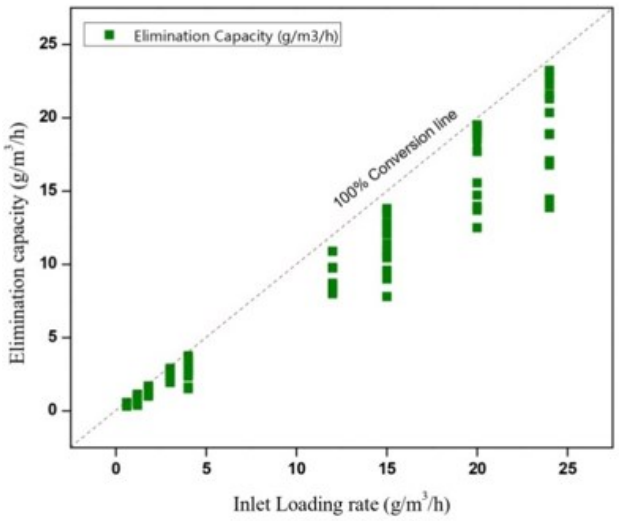


Figure 4. Influence of formaldehyde loading rate

3.5. Behavior of Filter media and microbial characteristics

The filter media porosity of the bioreactor column is estimated to be 50%. The filter media's pH, temperature and moisture content were controlled for the entire bed column and the dynamic operations. The neutral pH was observed throughout the experimental studies. Optimal moisture contents of 49 to 52% may have enhanced bacterial growth conditions with sufficient media volume (Ondarts *et al*., 2012). Mesophyll temperature conditions were designed, which sustained the microbial activities suitable for formaldehyde degradation. Moreover, the average temperature of 22±3℃ was maintained in the biofilter to overcome the diverse effects of temperature throughout the entire bioreactor system. Therefore, temperature fixation was the primary factor concerning microbial populations' consumptive use of oxygen.

The lower RE was achieved while feeding the biofilter with increased inlet concentration, possibly due to the increased pollutant load (Zilli *et al*., 2001). Later, the addition of nutrients to the biofilter yielded increased removal efficiency. The nutrient was sprinkled every day on the filter bed concerning the presence of moisture content and the corresponding quantity was measured. Adding nutrients increased the availability of organic carbon and the corresponding energy released, which were subsequently utilized by microbes for their metabolic activities (Fulazzaky *et al*., 2016).

During the first two months, the filter media absorbed the moisture content and the nutrients added to it gradually increased the essential microbial growth. Approximately 60 to 90 mL of water is added to the top of the filter column per day with the effect of media drying. These consequences were affected by increased compaction, swelling, and shrinking, which are assumed to have detrimental effects on filter media parameters. It also revealed a decreased gas flow rate due to the combined impact of reduced filtered volume and pressure drops.

The microbial growth developed in the filter media was analyzed for the presence of gram-straining bacteria, as reported by Hajizadeh &Rezaei (2014). The results (Fig. 5) represented the presence of more gram-negative bacteria (pink coloured) rather than gram-positive bacteria (purple coloured). Moreover, the gram-negative bacteria were reported to decompose the formaldehyde quickly. Therefore, the filter media designed in the present experimental study developed gram-negative bacteria on the filter media, thereby being efficient in formaldehyde degradation (Rezaei *et al*., 2015).

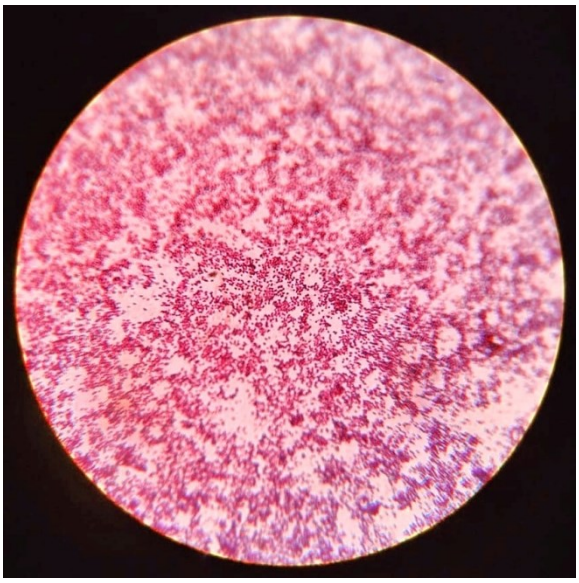


Figure 5. Primary bacterial development

From Fig. 5, it was visualized that quantity of gram-negative bacteria surpasses that of gram-positive bacteria. In bacterial isolation and identification studies, formaldehyde's degradation rate is high in gram-negative presence. Straining techniques were analyzed, and more expected gram-negative bacterial levels were obtained. As the microbial population grows, formaldehyde's degradation rate increases, forming a biofilm layer. The smear process was done using nutrient agar media and the corresponding bacterial growth was observed by microscope. Two predominant bacteria were noticed and contained more than 100 CFU (countable colonies forming units) per gram of filter media bed. Coccobacilli types of bacteria are predominantly present in the majority and only a few other colonies were identified in the treated filter media. Therefore, it can be seen that gram-negative bacteria played an essential part in degrading formaldehyde. Similar trends were reported by Rezaei *et al*.(2015).

The morphology of filter media materials (i.e., compost, vermiculite, activated carbon) and the treated filter media sample was illustrated in Fig. 6. The elongated fibrous structure of the compost can be observed in Fig. 6(a) exhibiting various sizes (3 to 10 µm). These fibers may be composed of cellulose fibers. The porous structure of vermiculite (Fig. 6(b)) was observed, whereas the morphology of activated carbon (Fig. 6(c)) exhibited a flat rough surface with irregular size distribution.

Fig. 6(d) and (e) represented the morphology of treated filter media collected from the intermediate zone of the biofilter (Rene et *al*., 2005). The microbial population present as biofilm can be observed from those corresponding SEM images. Hence, formaldehyde was adsorbed more consistently in filtered media, producing a high contaminant removal rate in the biofilter.

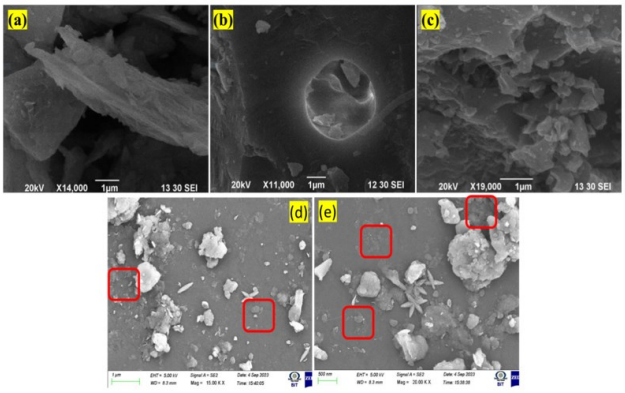


Figure 6. SEM image of (a) Compost, (b) Vermiculite, (c) Activated Carbon, (d) Treated filter media

3.6. Production of Carbon dioxide

The present study increased the formaldehyde concentration and EBRT in each phase. Moreover, the EC and removal efficiency were evaluated based on the formaldehyde degradation rate with respect to CO_2 production.

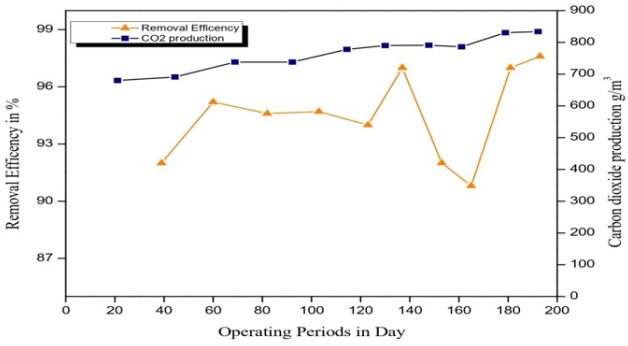


Figure 7. Production of carbon dioxide

The elimination rate was studied from the oxidation of formaldehyde pollutants, which was directly proportional to CO_2 production (Fulazzaky *et al*., 2016; Mathur *et al*., 2007). The oxidation speed was increased with the influence of the filter media of the bioreactor. Variation of CO_2 production was measured from initial to final is 57.8% increased during operation of the bioreactor system. The stability of biomass increased and was measured using carbon and energy sources to release the end products of CO_2 and water.

4. Conclusion

The present study examined the effectiveness of a composite biofilter in degrading the formaldehyde from the simulated air stream. The composite materials of biofilter are composite (nutrient enriched), vermiculite (high water holding capacity) and activated carbon (wide surface area). Formaldehyde degradationgradually decreased with increasing inlet loading and gas flow rates and decreasing EBRT. Increased EBRT resulted in good efficiency (97.6%) and elimination capacity (19.52 g/m^3h^{-1}). Therefore, the filter media utilized provided a suitable support medium for microbial growth and proved their enhanced performance in the degradation of formaldehyde. Laboratory scale models are a more realistic system to measure formaldehyde removal with potential microbial growth. Furthermore, the upper and bottom portion of the biofilter has a high potential for degradation due to optimal circumstances conducive to the proliferation of microorganisms.

Acknowledgment

The authors express their appreciation to the Centre for Research – Environmental Studies, Government College of Technology, Coimbatore, for their provision of essential laboratory facilities.

References

Dewidar, A. A., Sorial, G. A. and Wendell, D. (2022). Simultaneous acidic air biofiltration of toluene and styrene mixture in the presence of rhamnolipids: Performance evaluation and neural model analysis, Biochemical Engineering Journal, 187, 108637.

Długosz, O. and Banach, M. (2018). Sorption of Ag^+ and Cu^{2+} by vermiculite in a fixed-bed column: Design, process optimization and dynamics investigations, Applied Sciences, 8(11), 2221.

Dorado, A. D., Lafuente, F. J., Gabriel, D. and Gamisans, X. (2010). A comparative study based on physical characteristics of suitable packing materials in biofiltration. Environmental technology, 31(2), 193–204.

Fulazzaky, M. A., Talaiekhozani, A. and Abd Majid, M. Z. (2016). Formaldehyde removal mechanisms in a biotrickling filter reactor, Ecological Engineering, 90, 77–81.

Gallastegui, G., Muñoz, R., Barona, A., Ibarra-Berastegi, G., Rojo, N. and Elías, A. (2011). Evaluating the impact of water supply strategies on p-xylene biodegradation performance in an organic media-based biofilter, Journal of hazardous materials, 185(2–3), 1019–1026.

Hajizadeh, Y. and Rezaei, M. (2014). Biodegradation of formaldehyde from contaminated air using a laboratory scale static-bed bioreactor, International journal of environmental health engineering, 3, 18–25.doi: 10.4103/2277-9183.131804

Halios, C. H., Landeg-Cox, C., Lowther, S. D., Middleton, A., Marczylo, T. and Dimitroulopoulou, S. (2022). Chemicals in European residences–Part I: A review of emissions, concentrations and health effects of volatile organic compounds (VOCs), Science of the Total Environment, 839, 156201.

Huang, Y., Ho, S. S. H., Lu, Y., Niu, R., Xu, L., Cao, J. and Lee, S. (2016). Removal of indoor volatile organic compounds via photocatalytic oxidation: a short review and prospect, Molecules, 21(1), 56.

Kumar, M., Giri, B. S., Kim, K. H., Singh, R. P., Rene, E. R., López, M. E. and Singh, R. S. (2019). Performance of a biofilter with compost and activated carbon based packing material for gas-phase toluene removal under extremely high loading rates, Bioresource technology, 285, 121317.

Li, L., Chai, F., Liang, C., Wang, Y., Zhang, X., Yang, K. and Xiao, B. (2020). Comparison and application of biofilter and

suspended bioreactor in removing gaseous o-xylene, Environmental Research, 188, 109853.

Lu, N., Pei, J., Zhao, Y., Qi, R. and Liu, J. (2012). Performance of a biological degradation method for indoor formaldehyde removal, Building and environment, 57, 253–258.

Lu, Y., Wang, D., Ma, C. and Yang, H. (2010). The effect of activated carbon adsorption on the photocatalytic removal of formaldehyde, Building and Environment, 45(3), 615–621.

Mathur, A. K., Majumder, C. B. and Chatterjee, S. (2007). Combined removal of BTEX in air stream by using mixture of sugar cane bagasse, compost and GAC as biofilter media, Journal of Hazardous Materials, 148(1–2), 64–74.

Mohammed, J., Nasri, N. S., Zaini, M. A. A., Hamza, U. D. and Ani, F. N. (2015). Adsorption of benzene and toluene onto KOH activated coconut shell based carbon treated with NH3, International Biodeterioration & Biodegradation, 102, 245–255.

Ondarts, M., Hort, C., Sochard, S., Platel, V., Moynault, L. and Seby, F. (2012). Evaluation of compost and a mixture of compost and activated carbon as biofilter media for the treatment of indoor air pollution, Environmental technology, 33(3), 273–284.

Parseh, I., Hajizadeh, Y., Jaafarzadeh, N., Goudarzi, G., Shakerinejad, G., Badeenezhad, A. and Fallahizadeh, S. (2021). Removal behavior of gaseous furfural using a biofilter packed with perlite, ripe compost, and oak woodchips, Process Safety and Environmental Protection, 149, 135–143.

Parseh, I., Hajizadeh, Y., Jaafarzadeh, N., Goudarzi, G., Shakerinejad, G., Badeenezhad, A. and Fallahizadeh, S. (2021). Removal behavior of gaseous furfural using a biofilter packed with perlite, ripe compost, and oak woodchips, Process Safety and Environmental Protection, 149, 135–143.

Prado, Ó. J., Veiga, M. C. and Kennes, C. (2004). Biofiltration of waste gases containing a mixture of formaldehyde and methanol, Applied microbiology and biotechnology, 65, 235–242.

Rene, E. R., Murthy, D. V. S. and Swaminathan, T. (2005). Performance evaluation of a compost biofilter treating toluene vapours, Process Biochemistry, 40(8), 2771–2779.

Rene, E. R., Murthy, D. V. S. and Swaminathan, T. (2005). Performance evaluation of a compost biofilter treating toluene vapours, Process Biochemistry, 40(8), 2771–2779.

Rezaei, M., Fazlzadehdavil, M. and Hajizadeh, Y. (2015). Formaldehyde removal from airstreams using a biofilter with a mixture of compost and woodchips medium, Water, Air, & Soil Pollution, 226, 1–10.

Robert, B. and Nallathambi, G. (2021). Indoor formaldehyde removal by catalytic oxidation, adsorption and nanofibrous membranes: A review, Environmental Chemistry Letters, 19, 2551–2579.

Saravanan, V., Rajasimman, M. and Rajamohan, N. (2015). Performance of packed bed biofilter during transient operating conditions on removal of xylene vapour, International Journal of Environmental Science and Technology, 12, 1625–1634.

Shao, Y., Wang, Y., Zhao, R., Chen, J., Zhang, F., Linhardt, R. J. and Zhong, W. (2020). Biotechnology progress for removal of indoor gaseous formaldehyde, Applied microbiology and biotechnology, 104, 3715–3727.

Sheoran, K., Siwal, S. S., Kapoor, D., Singh, N., Saini, A. K., Alsanie, W. F. and Thakur, V. K. (2022). Air pollutants removal using biofiltration technique: a challenge at the frontiers of sustainable environment, ACS Engineering Au, 2(5), 378–396.

Singh, R. S., Rai, B. N. and Upadhyay, S. N. (2006). Performance evaluation of an agro waste based biofilter treating toluene vapours, Environmental technology, 27(4), 349–357.

Sun, Z., Ding, C., Xi, J., Lu, L. and Yang, B. (2020). Enhancing biofilm formation in biofilters for benzene, toluene, ethylbenzene, and xylene removal by modifying the packing material surface, Bioresource technology, 296, 122335.

Talaiekhozani, A., Lotfi Ghahroud, M., &Rezania, S. (2021). Estimation of carbon monoxide, sulfur oxides, nitrogen oxides, volatile organic compounds, and particulate matters emission due to cryptocurrency miners' activity in Iran, Earth, 2(3), 667–673.

VanOsdell, D. W., Owen, M. K., Jaffe, L. B. and Sparks, L. E. (1996). VOC removal at low contaminant concentrations using granular activated carbon, Journal of the Air & Waste Management Association, 46(9), 883–890.

Wen, Z. D., Wu, W. M., Ren, N. Q. and Gao, D. W. (2016). Synergistic effect using vermiculite as media with a bacterial biofilm of Arthrobacter sp. for biodegradation of di-(2-ethylhexyl) phthalate, Journal of hazardous materials, 304, 118–125.

Wu, P. W., Chang, C. C. and Chou, S. S. (2003). Determination of formaldehyde in cosmetics by HPLC method and acetylacetone method, Journal of food and Drug Analysis, 11(1), 5.

Yuan, J., Du, L., Li, S., Yang, F., Zhang, Z., Li, G. and Wang, G. (2019). Use of mature compost as filter media and the effect of packing depth on hydrogen sulfide removal from composting exhaust gases by biofiltration, Environmental Science and Pollution Research, 26, 3762–3770.

Zhong, X. Z., Sun, Z. Y., Wang, S. P., Tang, Y. Q., Kida, K. and Tanaka, A. (2020). Minimizing ammonia emissions from dairy manure composting by biofiltration using a pre-composted material as the packing media, Waste Management, 102, 569–578.

Zilli, M., Palazzi, E., Sene, L., Converti, A. and Del Borghi, M. (2001). Toluene and styrene removal from air in biofilters, Process Biochemistry, 37(4), 423–429.