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Biodegradation of Triethylamine-laden Emissions in a Compost-based Biofilter

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Abstract: Biofiltration of Triethylamine (TEA) vapor from waste gas was evaluated in this study. Experiments were conducted on a 6 L three-stage biofilter containing a mixture of compost and wood chips (40:60 v/v) as the filter medium. Seed inoculum was obtained from municipal activated sludge and an initial acclimation period of three weeks was allowed prior to data acquisition. TEA elimination rate pattern was evaluated by varying loading rates ($6\text{--}116\text{ g m}^{-3}\text{h}^{-1}$) and hydraulic detention times (40-60 sec) while maintaining operating temperature at $30\pm 1^\circ\text{C}$ and humidity at 50-55%. Results showed that Organic Loading Rates (OLR) of up to $114.4\text{ g m}^{-3}\text{h}^{-1}$ can be handled without any apparent indication of maximum elimination capacity and substrate inhibition. The elimination capacity of this biofilter could reach up to $72\text{ g m}^{-3}\text{h}^{-1}$. When the loading of TEA exceeded this critical value, substrate inhibition occurred and the elimination capacity decreased. However, the requirement of keeping the pressure drop below 1000 Pa m^{-1} to avoid operational problems warranted lower than maximum capacity operation. The optimal OLR values of $90\pm 1.4\text{ g m}^{-3}\text{h}^{-1}$ is suggested for Hydraulic Retention Time (HRT) value of 48 sec. Under these conditions, elimination capacity of $71\pm 3\text{ g m}^{-3}\text{h}^{-1}$ and removal efficiency $81\pm 1.4\%$ were achieved.

Key words: Biofiltration, triethylamine, biodegradation, air pollution

INTRODUCTION

Studies on biofiltration over the last several decades have primarily been focused on odorous compounds such as hydrogen sulfide, ammonia, mercaptanes, etc.^[1-3]. However, adoption of stricter emission policies in recent years have greatly increased the inventory of compounds subject to regulation. Furthermore, increasing costs of chemicals and disposal of hazardous wastes have provided further incentive for development and optimization of biological treatment methods. Biofiltration has emerged as one of the cost effective biological air pollution control technologies for treatment of Volatile Organic Compounds (VOCs) emitted from chemical and process industries^[4].

In biofiltration, the gas to be treated is passed through a packed bed of biomass supported on suitable materials such as compost, peat, wood chips, or inert materials. When waste gases pass through the reactor, target organic pollutants diffuse into the biofilm with subsequent biological oxidation into less harmful substances such as CO_2 , H_2O and minerals. A consortium

of microbial populations are known to play an important role in this process but current understanding of the mechanisms and specific microbial enzymes involved is limited^[5]. In general natural biomass support materials provide sufficient inorganic nutrients for microorganisms. As such, addition of nutrients is not required. The required moisture is provided by saturating influent stream before it enters the reactor and/or by supplying liquid water intermittently^[6].

Amines are widely used as catalysts in casting operations. They are also the major pollutants in the gaseous emissions of chemical manufacturing factories. During the production of casting cores with the so called cold-box-process, polyurethane is used as a binder in the sand core. Considerable amount of amine vapor is used in this process and is partly liberated to the ambient air. Tertiary amines, such as Triethylamine are the main gaseous catalysts comprising the majority of nitrogenous emissions^[7]. Previous studies have suggested suitable biodegradation potential of amines^[8,9]. As such, biofiltration seems to be an appropriate method to treat waste gases containing these pollutants.

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This study was aimed to examine the response of biofilter systems to the variations of inlet concentration, moisture content and loading (organic and hydraulic) conditions. Specifically, the effect of inlet concentration and organic loading rate on the performance of biofilter in treating TEA in a pilot plant system is studied.

MATERIALS AND METHODS

Experimental setup: Experiments were conducted in a laboratory scale reactor shown in Fig. 1. The column had an inner diameter of 5 and an effective height of 100 cm. Perforated Plexiglas plates (pore diameter = 3 mm) placed between sections acted as a support for the packing material as well as for flow redistribution. A 5 cm space in between the sections allowed for representative gas sampling. Provision of two sampling ports at midpoint within each section allowed temperature measurements as well as bed media access.

The main air stream was prepared by sending compressed air through an activated carbon adsorber for residual oil capture. A side stream of purified air was sent through a 1 L bottle containing pure liquid TEA. The rest was humidified and mixed with the exiting side stream containing pollutant vapor. Air flow rates were

appropriately controlled using pressure regulators and flow meters to generate feed air with the needed concentration. Variation of humidity in the influent gas stream and biofilter material was controlled by changing water temperature in the humidifier.

Temperature control of the bed material was achieved by circulating water around the exterior of reactor wall. Heated element was used for temperature control in the water tank. During the steady state operational period of the study, bed temperature and humidity were maintained at $30 \pm 1^\circ\text{C}$ and 50-55%, respectively.

Filter media and microbial culture: Filter media was prepared by blending of sieved compost and wood chips. Municipal compost (equivalent diameter 2-5 mm) with a C:N:P ratio of 100:7:2, 37.8% organic matter and a pH value of 6.8 was obtained from a local composting facility. Wood chips (2-5 mm) were added as bulking material to produce a 60:40 v/v ratio of compost-wood chip. The inoculum consisted of municipal activated sludge from the local regional wastewater treatment plant. The following nutrient and buffering solution was also added to the activated sludge (g dm^{-3}): KH_2PO_4 , 5; K_2HPO_4 , 2.5; potassium, 0.2; sodium, 0.64; calcium, 5; magnesium, 2; chloride, 3.7; phosphorus, $1.15^{[10]}$.

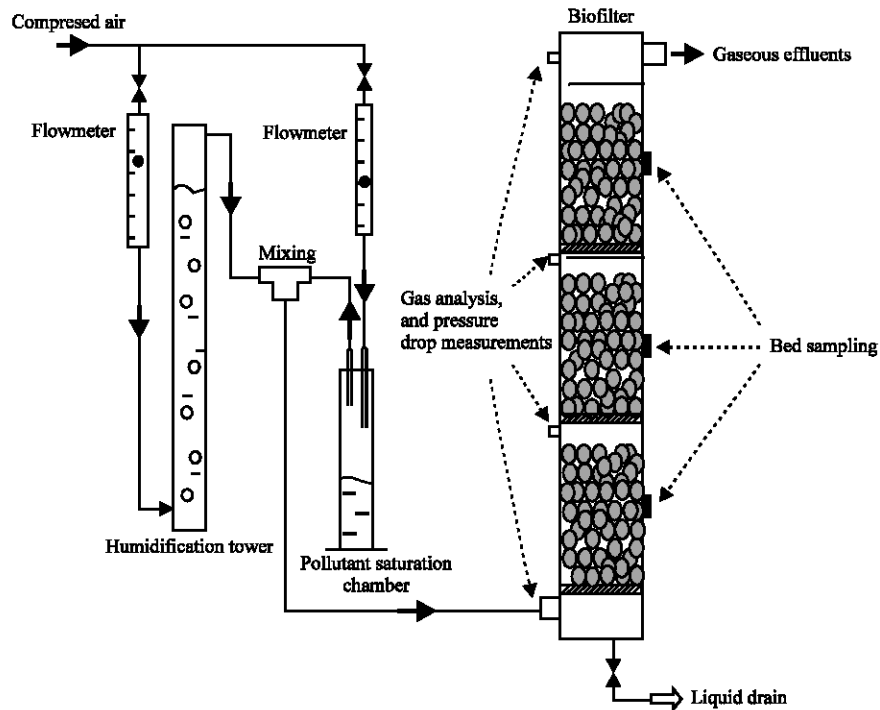


Fig. 1: Schematic diagram of the experimental system

Operational and performance parameters: The critical parameters include Empty-Bed Residence Time (EBRT), Mass Loading Rates (MLR), Removal Efficiency (RE) and Elimination Capacity (EC). EBRT is the time a parcel of air will remain in a empty biofilter and overestimates the actual treatment time. MLR define the amount of contaminant entering the biofilter per unit area or volume of filter material per unit time. Both terms are normalized, allowing for comparison between reactors of different sizes.

RE and EC are used to describe the performance of a biofilter. RE is the fraction of contaminant removed by biofilter and EC is the mass of the contaminant degraded per unit volume of filter material per unit time. Removal efficiency is an incomplete descriptor of biofilter performance because it varies with contaminant concentration, air flow and biofilter size and reflects only the specific conditions under which it is measured. The EC is normalized with respect to volume by definition and allows for direct comparison of the results of two different biofilter systems.

Analytical methods: Gas samples were collected at the inlet, outlet and in the 5 cm plenum between the sections. The amount of TEA was measured by UV spectrophotometer (UV/VIS-911, GBC CO, Australia) at a wavelength of 215 nm. For measuring pH, 1 g of biofilter bed material and 20 mL distilled water were blended and agitated for 10 min and measured by a pH meter (691-Metrohm, Switzerland). Moisture of biofilter bed material was measured by weight loss of 2 g solid sample after being dried at 106°C for 24 h.

Heated water was circulated around the bed exterior and connected to a precision thermostat (Atbin Co.) to control temperature within 1°C. Temperature was maintained at 30°C and measured using alcohol in a glass thermometer with a range from -10 to 110 and a scale division of 1°C. Gas flow rate was measured using flow meter (Omega FI-2016) with units of l/min. A water-filled manometer with a minimum division length reading of 1 mm water column was used to measure pressure drop across the column.

RESULTS AND DISCUSSION

Startup: Depending on biofilter conditions, the startup procedure may last for a few days to a few months. It is the acclimation time required to establish optimal biological removal. Additionally, by slowly increasing the mass loading rate on the system at startup, toxic shock may be avoided as microbes acclimate gradually.

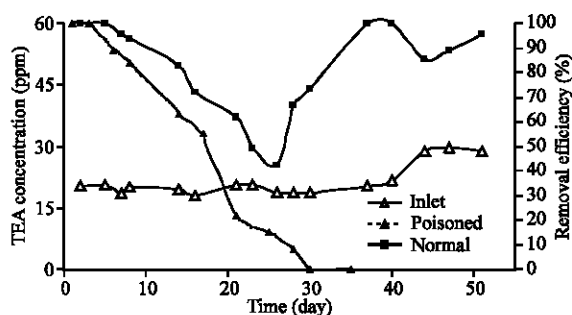


Fig. 2: Time variation of removal efficiency during the start up of the biofilter

At the startup of biofilter in this study, influent TEA concentration was adjusted to 20 ppm at an organic loading rate of $6 \text{ g m}^{-3}\text{h}^{-1}$ and relative humidity of 50-55%. Also, water temperature in humidifier was adjusted to $28 \pm 2^\circ\text{C}$. Superficial gas velocity was 57.3 m h^{-1} , corresponding to a residence time of 48 sec.

Contaminant molecules may be simply dissolved in the water, but they may also be adsorbed on the surface of the medium, taken up by living cells, adsorbed with in organic matter in the biofilm or medium, or collected at the surface of water. For highly soluble contaminants, the dissolved form may be dominant, but for more hydrophobic contaminants, the major reservoir may be material adsorbed on the surface of the medium and absorbed within the organic matter^[11]. In order to understand the process of the biofilter and the physical adsorption capacity of the filter material, at the startup, experiment was performed in active and in poisoned (sterilized by HgCl_2 solution) biofilters. It was found that at the beginning of biofiltration (about 2 weeks), probably adsorption is the main mechanism to remove TEA, but after a lapse of 25 days of the beginning of operation, efficiency decreased from 100 to 42% and again, increased to about 100% during the period of 11 days (Fig. 2).

Adsorption on solid media in biofilter was studied by Freundlich model in which it is assumed sites for adsorption are not limited. As shown in Fig. 3, k_f (Freundlich coefficient) and $1/n$ (a constant) were 10.5 and 0.4522, respectively according to the following equation:

$$x/m = 10.5 * C_e^{0.4522}$$

Where, C_e = Equilibrium concentration of adsorbate in fluid and x/m = mass of adsorbate per mass unit of adsorbent.

In general, higher substrate concentrations will improve treatment efficiency up to a point where biological kinetics drive the degradation rate to zero order.

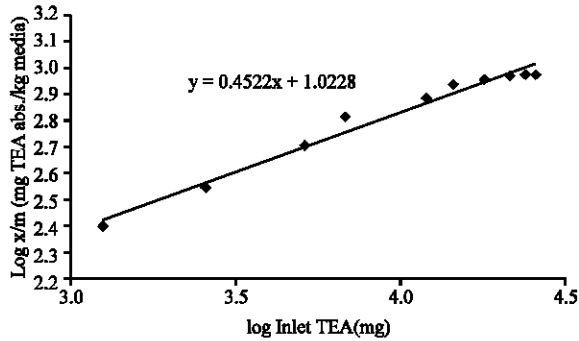


Fig. 3: Adsorption capacity of biofilter medium based on Freundlich model in the poisoned biofilter

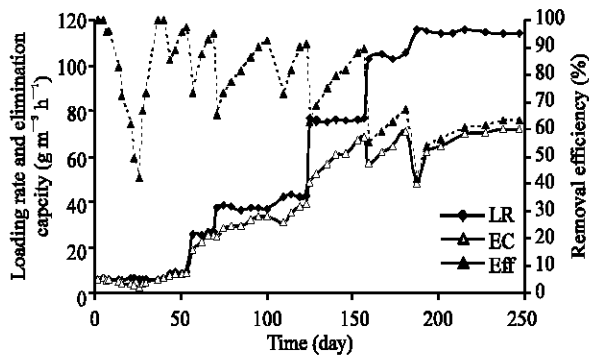


Fig. 4: Removal efficiency and elimination capacity of biofilter at different inlet TEA concentration

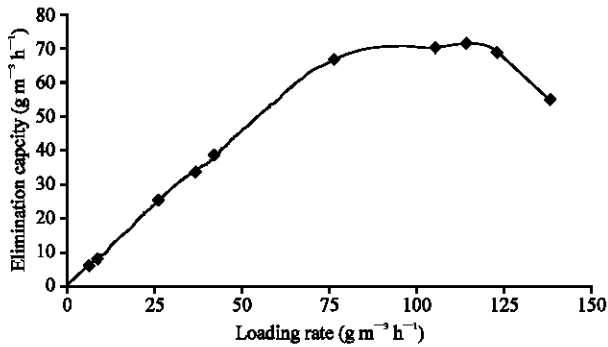


Fig. 5: Elimination capacity of biofilter at different TEA loading rates

In a case of diffusion limitation, higher gas-phase concentration of substrate drives the contaminant into the biofilm more rapidly^[12]. As such, gradual increase during the initial phases of reactor operation is essential for proper microbial growth and increase during the initial phases of reactor operation is essential for proper microbial growth and development. The observed acclimation period in this study was the 3-4 weeks, because microorganisms in municipal activated sludge had not been acclimated to the target pollutant.

Decreasing of removal efficiency was observed with a lag period after increasing the inlet concentration with subsequent increase in the removal efficiency after gradual acclimation of microbes to the pollutant.

Removal efficiency: Inlet TEA concentration was increased up to 385 ppm (1.5 g m^{-3}) stepwise at the HRT of 48 sec, so mass loading changed proportionally. As shown in Fig. 4 at the beginning of each step change in feed concentration, there was a decreasing in RE with gradual recovery with time. For low concentrations RE was higher than 90% (below 90 ppm, 95-100%, below 180 ppm, 90-95%).

When the inlet concentration is increased, initially, the biofilm may be affected by a pseudo shock load with consequent increase TEA in effluent air from the column. At lower concentrations, the degree of drop in RE as a result of increased concentration is lower than at higher concentrations, due to lower microbial population demand. The time to recover from the drop is increasing with increased inlet TEA, due to an apparent increased inhibitory effect at higher concentration. This is more pronounced in regions where the concentration is approaching saturation EC.

Elimination capacity: EC shows what portion of the incoming organic loading is being biodegraded. As the loading rate is increased, a point of saturation or maximum EC corresponding to maximum microbial substrate utilization rate is observed. This limitation is due to the effect of high concentrations on the Monod kinetics of biodegradation^[13]. In some cases, it is known that very high concentration of substrate can become inhibitory^[12].

In order to evaluate EC, OLR can be increased through increased influent concentration or flow rate (reduced HRT). By increasing inlet TEA concentrations while maintaining constant flow rate (HRT=48 sec), OLR was increased. As shown in the Fig. 5, there is a linear relationship between EC and OLR up to an OLR value of $72.3 \text{ g m}^{-3}\text{h}^{-1}$ (inlet TEA concentration of 250 ppm). Beyond this value, a flattening of the curve is observed with eventual decreasing trend for OLR values greater than $120 \text{ g m}^{-3}\text{h}^{-1}$. This is a bit lower than the results reported by Tang et al, 1996 on the onset of inhibitory effects at loading rate of $140 \text{ g m}^{-3}\text{h}^{-1}$ for the compost/chaff biofilter. The difference may be attributable to the higher column length of 100 cm and HRT value of 60 vs. 48 sec for this study.

Concentration gradient: TEA concentration along the bed was continuously monitored throughout the study period. As shown in Fig. 6, the first section has the

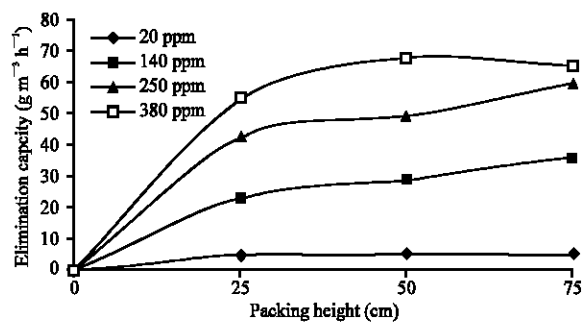


Fig. 6: Effect of inlet TEA concentration on the biodegradation potential along the bed

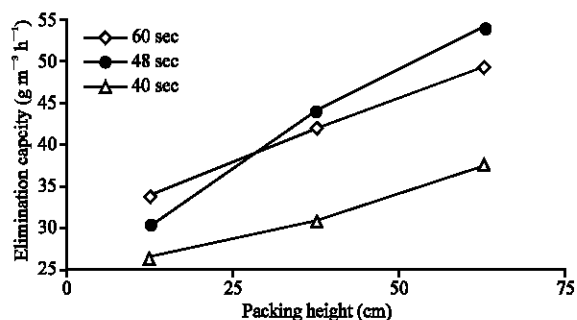


Fig. 7: Effect of the hydraulic retention time on the elimination capacity along the biofilter at constant inlet concentration of TEA

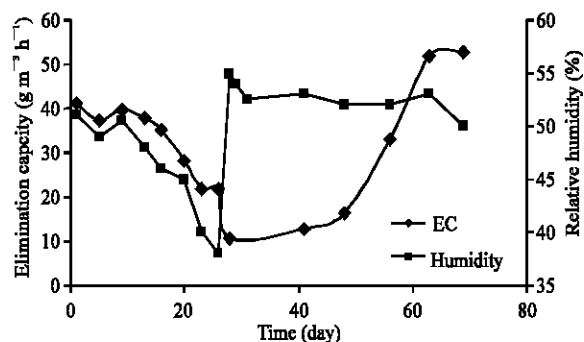


Fig. 8: Effect of the relative humidity on the elimination capacity at constant inlet TEA concentration

highest removal capacity due to exposure to higher relative concentrations of TEA. At low OLR values, up to 75% of the overall removals occur in the initial section. As organic loading was increased, more penetration of substrate occurred and latter sections showed increased EC. Slight reduction in the biodegradation potential of the first section and an increased activity in latter sections can be attributed to overall microbial density limitations and saturation of specific activity for microorganisms in the first section. Dry patches due to higher biological activity and increased water evaporation due to energy

released from biooxidation, observed in the initial portion of the column may also provide additional insight in the reduction of differences in EC for different sections at higher influent concentrations.

Hydraulic retention time: The total mass of TEA entering the biofilter increased with an increase in flow rate. While at the same time, there was a reduced contact time due to lower HRT values ensuing reduced EC values. Maximum EC was observed at HRT of 60 sec which is in agreement with the range of minutes for alcohols to 3 h for 90% removal of trichloroethylene reported by Bohm^[14]. As shown in Fig. 7 the reduction in EC as a result of decreasing HRT to 48 sec was minimal but further reduction of HRT to 30 sec adversely affected the performance. While lower HRT value is favored since it implies more removal per unit volume of reactor, constraints of mass transfer from the gas phase to the biofilm limits optimum operating ranges.

Relative humidity: Biological activity ceases if the moisture content is too low. In addition, cracks develop and hydraulic channelling occurs. Conversely, too much moisture leads to anaerobic zones forming in the bed where oxygen required for bio-oxidation is absent^[12]. For determination of optimum bed moisture, the operation of biofilter was studied when relative humidity varied from 38 to 55% at constant inlet TEA concentration of 220 ppm. The result indicated, when RH decreased from 52 to 38%, the EC dropped from 40 to 10 g m⁻³h⁻¹. Adjusting the moisture content back to the original range of 55% (Fig. 8) resulted in restoration of EC to previous values. The time taken was excessive (37 days) pointing to the fact that physical rewetting may not pose any problem but restoration of biological activity may take much longer.

CONCLUSIONS

Biofiltration of Triethylamine (TEA) vapor from waste gas evaluated under different operational conditions in this study. Several conclusions can be surmised from the results as follows:

- Maximum EC value for TEA removal was observed to be 72 g m⁻³h⁻¹ at an OLR of 114 g m⁻³h⁻¹.
- Up to 75% of TEA removals were observed in the initial section of the biofilter.
- Operation conditions for optimum bioconversion of TEA in biofilter are recommended as follows: moisture content, 50-55%; HRT, 48 sec and a maximum loading rate for 100% and 81±14% RE, are 53±1 and 71±3 g m⁻³h⁻¹, respectively.

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