Thermophilic treatment of acetaldehyde emission in a biotrickling filter

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Extended Abstract 595578

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INTRODUCTION

In 2015, the ethanol industry in the US hit a production milestone of 1 million barrel per day. Ethanol is the major type of biofuel produced and its production is expected to continue to increase[1]. However, hazardous air pollutants (HAPs) such as acetaldehyde, formaldehyde, and acrolein are emitted from distilled dry grain solubles (DDGS) dryers, fermentation tanks and distillation columns during production[2]. Acetaldehyde is considered the major HAP of concern. Federal regulations limit HAP emissions to 10 tons per year of any individual HAP and 25 tons per year for total HAPs for an ethanol plant to be classified as an 'Area Source'[3]. Air pollution control equipment are essential to keep the facility in compliance. The EPA has identified CO_2 scrubbing and regenerative thermal oxidation (RTO) as the Best Available Control Technologies[3]. RTOs and scrubbers are usually used to control the dryers and fermentation, respectively. Both technologies are utility intensive and require large water and energy inputs. At an average ethanol plant producing annually 55 million gallons of denatured ethanol and 164,491 tons of DDGS, the RTO will be sized at about 18 MMBtu/hr. burning natural gas at about 155 MMSCF/yr.

An appealing alternative for the treatment of dilute HAPs is biofiltration[4]. Traditional biofilters were evaluated for the removal of HAPs generated at an ethanol plant with limited success[5]. Acetaldehyde and formaldehyde fumes were individually biodegraded in 10 seconds empty bed resident time (EBRT). However, long-term treatment leads to pH decline and deteriorating performance thereafter. In another study, acetaldehyde was successfully degraded in a mixture of toluene and ethanol in a two-stage biofilter and 95% removal was maintained at 15 seconds EBRT[6]. Ethanol and acetaldehyde had removal yields over 97% at an elimination capacity (EC) of 14.67 $g/m^3/h$ at 100 ppmv and 92-98% (EC 10.3 $g/m^3/h$) at 70 ppmv, respectively[6]. A study on the biofiltration of a mixture of HAPs found that acetaldehyde had more biodegradation potential than ethanol[7].

A key challenge facing biofiltration is inconsistent loadings; changes in flow rate or concentration adversely affect removal as micro-organisms are unable to quickly adapt[8]. Shutdown periods in which no loading is supplied to a BTF may cause deterioration of the

biofilm resulting in poor performance during startup. Hydrophobic compounds are not well suited to degradation in a biofilter[9]. These challenges are not present at ethanol plants; therefore, BTF technology is an attractive alternative. Acetaldehyde, formaldehyde, and ethanol, which are the typical compounds, are all soluble. They are produced continuously since ethanol plants usually perform only one scheduled maintenance per year.

DDGS dryers generate a hot air stream that usually ranges between 100-140 °C. After sending the stream through a baghouse or cyclones for particulate control, the stream is cooled down to about 60 °C[5]. Thermophilic bacterial growth is not usually encountered in a BTF. A comparison of thermophilic and mesophilic BTFs have shown that thermophilic treatment might be favorable; toluene was removed up to 90% at loading rates below 100 $g/m^3/h$[10], H_2S was removed up to 950 ppmv at 1.2 minutes residence time[11], and MTBE was removed up to 99% at 330 $g/m^3/h$[12]. Sludge drying exhaust was treated with over 90% for VOCs, NH_3, and SO_2[13].

In this study the effect of temperature on the removal of acetaldehyde is studied in a biotrickling filter. Three different temperatures were examined; BTF 'A' was operated at room temperature, BTF 'B' was operated at 40°C, and BTF 'C' at 60°C. All BTFs were operated at a loading rate of 42 g m^{-3} hr^{-1} corresponding to 200 ppmv. The loading rate of 42 g m^{-3} hr^{-1} is the minimum loading rate in which the ability of BTF 'C' to biodegrade acetaldehyde was significantly impaired based on a previous study.

THREE BTFS OPERATING AT DIFFERENT TEMPERATURES

Methods

Figure 1 shows a full schematic of one of the three experimental apparatuses used for this study. In each BTF media consisting of (0.3" - 0.5") pellets of diatomaceous earth (Celite 6mm R-635 Bio-Catalyst Carrier; Celite Corp., Lompoc, CA), was housed in a three-inch internal diameter glass column. The media has a mean pore diameter of 20 µm, BET surface area of 0.27 m^2/g, and a bed density of 513 kg/m^3. It consists mainly of SiO_2 with a significant fraction of Al_2O_3.

The beds were seeded with microorganisms. The bed operated at 20°C (BTF 'A') was submerged overnight in return activated sludge obtained from the local wastewater treatment plant. 2 g/L of glucose was added to the sludge beforehand. BTFs 'B' and 'C' were seeded with a slurry of cooking compost. A 1:2 compost to water volume ratio was used and 2 g/L of glucose was once again included.

The columns extend for 3' above the top of the packing material, where the acetaldehyde-laden air was introduced at the top to allow uniform mixing. Each BTF is equipped with sampling ports located at packed depths of 3, 13, 23, 33, and 36 inches. All connections are airtight. Air from any sampling port can be directed for analysis to either an Agilent 7820A GC system with an MS detector and 30 m, 0.25 mm I.D. HP-5MS column or a 490 µ-GC equipped with a thermal conductivity detector and a two-channel module. BTFs 'B' and 'C' are heated by a heat-tape wrapped around the packed length of the column. Approximately half of the surface area of the column is covered by the heat-tape. A thermocouple placed through the fifth sampling port allows for temperature control.

A filtered air stream is split and regulated to 8 L/min (corresponding EBRT of 32 seconds) by two mass flow controllers. Liquid acetaldehyde with 99.5% purity obtained from Acros Organics (Pittsburgh, PA) is infused into the air stream through a septum housed in a stainless-steel tee union. A syringe pump and Hamilton Gastight syringes were used to regulate the infusion.

Nutrient/Buffer solution is delivered to the BTF beds intermittently by a pump and timer-controlled solenoid valves. The nutrient solution, which is used for a once-through flow and was not recycled, consists of essential inorganic salts and vitamins necessary to grow micro-organisms. A fresh five-gallon batch of nutrient solution is prepared every five days. The pipe delivering solution to each BTF is controlled by a pressure valve and a misting nozzle.

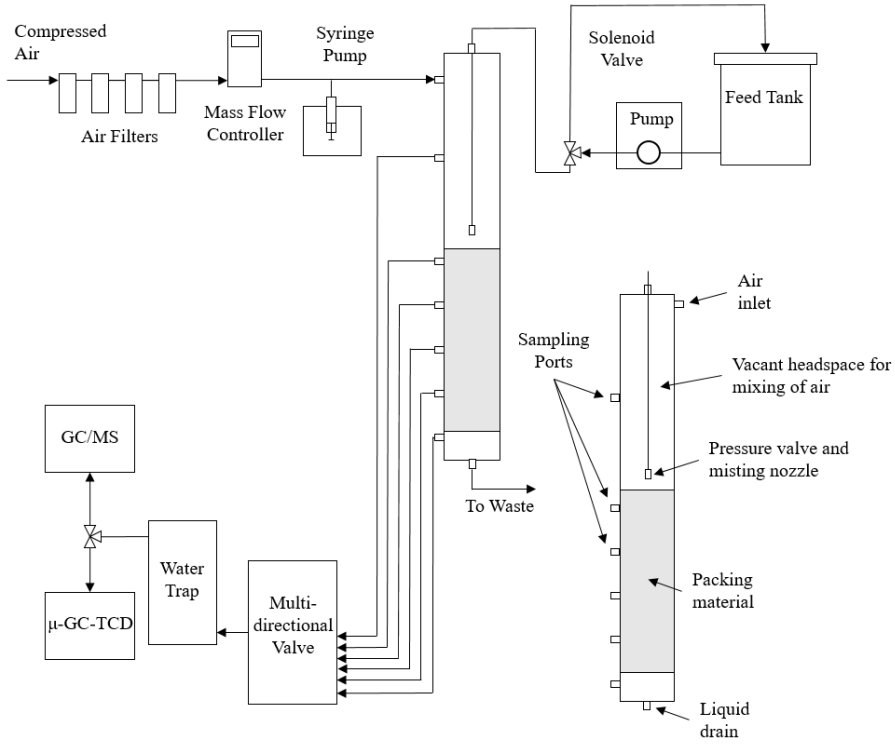

Figure 1: Schematic of one of the three utilized experimental apparatus. The temperature was varied using a heating tape with a temperature controller wrapped around the filter bed

Results and Discussion

The BTFs were operated at a loading rate of 45.3 g m^{-3} hr^{-1} corresponding to a concentration of 200 ppmv. Figure 2a shows the elimination capacity for each of the temperatures. A higher elimination capacity was observed at BTF 'B' in comparison to the other temperatures. Acetaldehyde solubility decreases with increasing temperature; therefore, the elimination capacity is also expected to decrease at higher temperature. Furthermore, the bed seeded with a cooking compost slurry is expected to perform better than those prepared with activated sludge due to a healthier thermophilic community. The compost slurry would perform better at higher temperature since, it contains thermophilic microorganisms that thrives at higher temperatures. This trend suggests that thermophilic conditions are still superior to mesophilic conditions at the optimum temperature range. Considering these trends together may explain the good performance of the 40 °C bed – it was operated at a moderate temperature and contains a cooking compost seed.

The removal efficiencies for BTFs 'A', 'B', and 'C' are 84.8%, 97.1%, and 60.9%, respectively. Figure 2b shows the variability of pH, Chemical Oxygen Demand number (COD), and Volatile

Suspended Solids (VSS) with each of the operating temperatures. The pH of the effluent wastewater for each of the operating temperatures had some slight differences. The average measured pH for the influent solution was 8.53. It is expected that the pH will increase due to aerobic degradation of acetaldehyde. At higher concentration other acidic byproducts were formed. Their concentration was increased with elevated influent acetaldehyde concentration decreasing the pH of the effluent liquid. COD is the only source of effluent carbon in the liquid phase. COD composition includes microorganisms, soluble byproducts, and dissolved acetaldehyde. The major byproduct identified was acetate, and the COD contributed by acetate is of great relevance. The COD content for the 60 °C bed was observed to be the maximum. This is due to the low degradation within the bed. The increase in VSS suggests biomass growth greater than the media holding capacity. Moreover, CO_2 concentration at the effluent was recorded at 234 ppmv in the 20°C bed. This corresponds to about half of the influent organic carbon is being converted to CO_2. In the, 40°C bed, the CO_2 effluent concentration increased by 70 ppmv, despite a greater removal of acetaldehyde. The CO_2 concentration in the 60°C bed increased by 76 ppmv. It should be noted that CO_2 solubility decreases significantly with increasing temperature suggesting lower inorganic portion in the effluent liquid at higher temperatures.

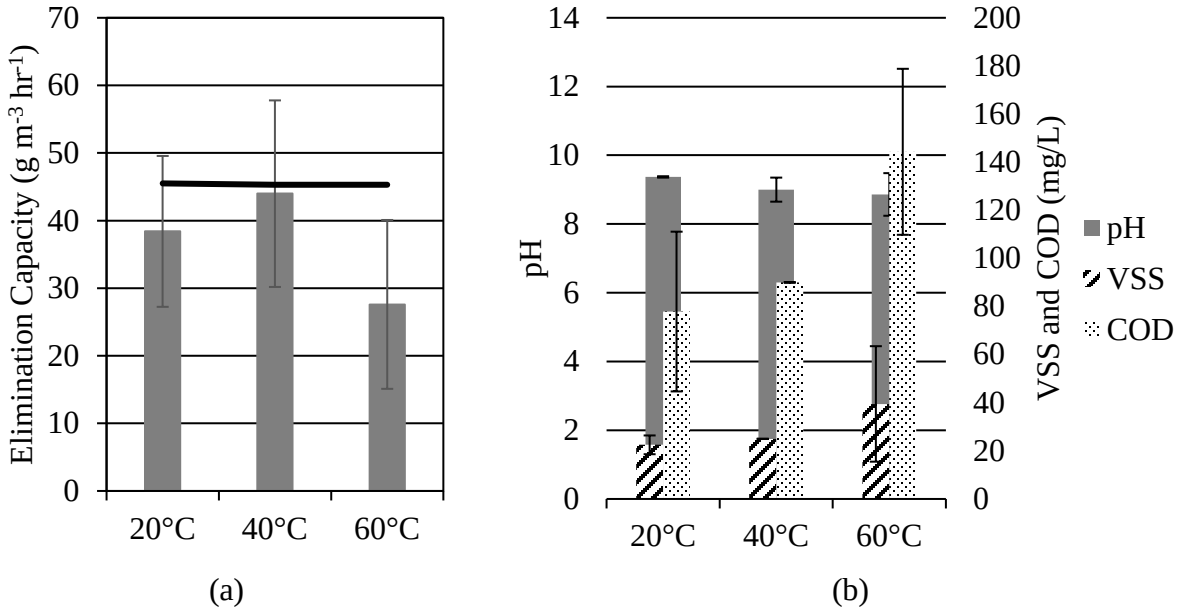

Figure 2: (a) Elimination capacity for each operating temperature. Solid line represents the loading rate. (b) Average pH, VSS and COD for the effluent liquid at each operating temperature. Error bars show one standard deviation.

SUMMARY

This study compared the performance of the BTFs at different temperatures. Treatment at highest temperature was not optimal due to the decrease in acetaldehyde solubility with increasing temperature. In addition, seeding with cooking compost is better than seeding with activated sludge at thermophilic conditions since the cooking compost has a wider variety of active thermophilic bacterial seed. However, the performance of the BTFs will still be expected to decrease with the increase of temperature regarding the type of the seeding. Comparing this to a study conducted by Chen [5], it was found that at 20 ºC the elimination capacity value is 28 g m^{-3} hr^{-1} at a maximum loading rate of 35 g m^{-3} hr^{-1}.[3] Whereas, this study showed a bit higher

elimination capacity at 38.5 g m^{-3} hr^{-1} in the same conditions. Studies conducted by Ryu[11], Cox[14], and Wei[15] explained the thermophilic performance of the BTFs of different compounds such as Trimethylamine, and Ethanol. The studies have observed a high elimination capacity of the treated compound. With acetaldehyde the elimination capacity is lower to the other compounds that were investigated in the previous studies at thermophilic conditions.

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