

Removal of hexane in biofilters packed with perlite and a peat–perlite mixture

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Summary

Removal of hexane from air–hexane mixtures in biofilters packed with different solid media under nitrogen supplementation was performed for 70 days. Two columns containing perlite and a mixture of peat and perlite were used. The solid media were supplemented with nitrogen source up to 1 kg/m³ per week for high nutrient supplementation and 0.2 kg/m³ per month for low nutrient supplementation. A high rate of hexane removal: 95 g/m³ h was achieved under high nutrient supplementation, high air flow rate and high hexane concentration. However, the percentage of hexane removal decreased with increasing air flow rate and hexane inlet concentration. For high nutrient supplementation the type of solid medium did not significantly affect the biodegradation capacity. With low nutrient supplementation, the highest removal rate was achieved in the column containing the peat–perlite mixture. The column containing perlite had a significantly lower pressure drop (20 Pa/m) than the 2400–2930 Pa/m observed for the column containing the mixture. Perlite offers an opportunity of running a biofiltration process at a lower and stable pressure drop if the nutrient supplementation is managed properly.

Introduction

Volatile organic compounds (VOCs) are major atmospheric pollutants responsible for smog formation and other harmful health effects such as cancer. The VOCs can be removed from flue gases through application of cost effective biofiltration technology. This technology is based on the ability of microorganisms immobilized on a solid support to convert VOCs in a gas stream to water, carbon dioxide, and biomass (Ottengraf 1986; Wani *et al.* 1997). Biofiltration is increasingly receiving attention in the removal of VOCs from waste gases due to lower operating costs and simpler management than physical and chemical treatments. Many types of biofiltration methods for the removal of VOCs have been performed during the last decade. Most conventional media used in laboratory, pilot-scale and industrial biofilters include peat, compost, humus, and soil (Valentin 1990; Shoda *et al.* 1991a, b; Bohn 1992; Kiared *et al.* 1996; Mallakin & Ward 1996; Martin *et al.* 1996; Wu *et al.* 1999). These organic materials decompose with time resulting in compaction of biofilter beds and eventual failure of biofiltration system. Recent developments in biofiltration have concentrated on improving the mechanical structure of the solid media and reduction of pressure drop,

which is the main operating cost of biofiltration (Bohn 1992; Togna *et al.* 1994; Wu *et al.* 1999; Cho *et al.* 2000).

Nutrient supplementation is considered as an effective method that can be used to increase biofilter performance by supplying necessary nutrients for the growth of microbes capable of degrading the target pollutant (Kiared *et al.* 1996; Mallakin & Ward 1996; Smith *et al.* 1996; Wu *et al.* 1999). However, when an organic carrier such as peat is used, nutrient supply causes over-growth of biomass such as fungi or actinomycetes, which causes clogging of the biofilter resulting in the decrease of VOC removal rate (Abumaizar *et al.* 1998; Gribbins & Loehr 1998).

The objective of this study was to explore the influence of two levels (high and low) of supplementary nutrients on hexane removal using biofilters packed with perlite and a mixture of 50% peat and 50% (v/v) perlite.

Materials and methods

Source of microorganisms

Activated sludge from the waste water treatment plant of the City of Waterloo, Ontario, Canada was used as the

source of microorganisms. Microorganisms were acclimatized to hexane in liquid culture before inoculating the biofilters. Acclimatization involved sub-culturing of the activated sludge broth into a mineral medium containing hexane as the sole carbon source. The composition of mineral medium (g/l) was as follows: K_2HPO_4 , 1; KH_2PO_4 , 0.1; $(NH_4)_2SO_4$, 1; $MgCl_2 \cdot 6H_2O$, 0.5; $FeSO_4 \cdot 7H_2O$, 0.02; and trace element solution, 2 ml. Trace element solution was prepared by dissolving into 1 l of deionized water, 200 mg each of $CaCl_2$, $MnSO_4 \cdot 5H_2O$, $CuSO_4 \cdot 5H_2O$, $ZnSO_4 \cdot 7H_2O$, and $(NH_4)_6Mo_7O_{24} \cdot 4H_2O$. The medium pH was adjusted to 7.0 using 1 M sodium hydroxide and 1 M hydrochloric acid.

Solid media

Solid media used in this research were composed of peat and perlite in two different combinations of 100% (v/v) perlite, and a mixture of 50% peat and 50% (v/v) perlite. Sphagnum peat (high-moor peat) and perlite were purchased at a local flower store in Waterloo, Canada. Laboratory evaluation of this peat showed that it had a bulk density of 166 kg/m^3 and pH value of 3.6. Perlite of horticultural grade was purchased at a local horticulture store in Waterloo, Canada, it had a bulk density of 100 kg/m^3 .

Biofiltration process

The biofiltration system is shown in Figure 1. Four biofilter columns each with internal diameter of 11.5 cm and a height of 95 cm, were used in this study. Two columns were packed with 100% perlite and the other two were packed with the mixture. The height of the solid media in the columns was 75 cm. Two columns of different packings, perlite (P) and the mixture (PP), were used for high nutrient supplementation. The same was performed for low nutrient supplementation. The solid media were inoculated with a mixed culture that had been acclimatized to hexane as the only carbon source. The supplementary nutrient solution was added to the solid medium at the top of the biofilter while a counter current air flow through the columns was introduced to assist the distribution of nutrients throughout the solid media. The nutrient solution collected at the bottom was recycled to the top of the column using a peristaltic pump. After 5 min, counter current air flow was stopped and excess nutrients solution was drained from the column before continuing with the biofiltration process. The drained solution was reintroduced into their respective columns twice a week to retard drying of solid media. To avoid microbial washout the first nutrient solution was introduced together with the inoculum.

Commercial organic-based fertilizer, nitrite, manufactured by Elmira, Ontario, Canada, with N:P:K ratio of 14:7:14 was used as a source of supplementary nutrients. At high nitrogen supplementation, 1 kg nitrogen per m^3 of solid media was added to the biofilters at the beginning of the first and second weeks. In subsequent

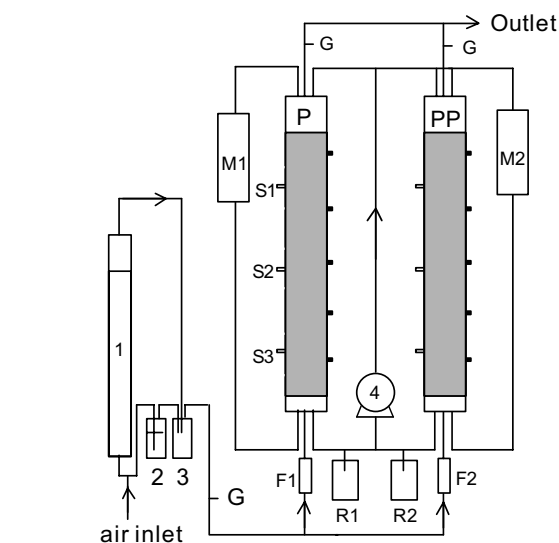


Figure 1. Schematic diagram for biofiltration of *n*-hexane: (1), humidification column; (2), evaporation unit of *n*-hexane; (3), mixing unit; (4), recycle pump, P, perlite column; PP, mixture (perlite 50%: peat 50%); column; F1–F2, flowmeters; G, gas sampling points; M1–M2, manometers; R1–R2, reservoirs; S1–S3, solid sampling points.

weeks nitrogen addition was decreased to 289 g/m^3 to reduce excessive biomass growth. The experiment was conducted at air flow rates of $20 \text{ m}^3/\text{m}^2 \text{ h}$ for the first 14 days, then changed to 100, 30, and $20 \text{ m}^3/\text{m}^2 \text{ h}$ after 14, 28, and 46 days, respectively. Hexane inlet concentration was between 100 and 800 ppm. At low nitrogen supplementation, 0.2 kg nitrogen per m^3 of solid medium was added to the biofilters at the beginning of the experiment and after 30 days. Subsequent addition was made whenever hexane removal dropped below 50%. In this run, the air flow rate through the column was maintained at $20 \text{ m}^3/\text{m}^2 \text{ h}$. The drained liquid containing unused nitrogen was recycled back to the columns twice a week.

Analysis

Gas concentration

The concentration of *n*-hexane in the gas phase was analysed using a Hewlett Packard (Avondale, Pennsylvania, USA) 5890 Series II gas chromatograph equipped with a flame ionization detector (FID). The column was a RTX-502.2 fused silica megabore column of $30 \text{ m} \times 0.53 \text{ mm}$ and $3.0 \mu\text{m}$ film thickness. Helium was used as the carrier gas at a flow rate of 15 ml/min. The auxiliary gas was nitrogen and the flame ionization gases were hydrogen and compressed air.

Carbon dioxide in the inlet air was measured using an infrared gas analyser (IR) (Backman model 864, Germany). The amount of CO_2 generated in the biofilter was calculated as the difference between the amount of CO_2 in the inlet and exit gas streams.

Ammonia content

Solid phase ammonia content was monitored using an ammonium electrode meter (Orion 9512 BN, Massachusetts, USA). Solid samples of 1 g each were withdrawn from each column at the lower, middle, and top sections. Each sample was homogenized and mixed with 100 ml of deionized water. The mixture was filtered using Whatman No. 2 filter paper. Two millilitres of 1 M NaOH solution was added to the filtrate. The mixture was stirred and the ammonium content was directly measured using the ammonium electrode.

Exact concentrations of the measured *n*-hexane gas, carbon dioxide, and ammonia were obtained from calibration curves prepared prior to the sample analysis.

Pressure drop

The pressure drop was monitored using water manometers connected to both column ends. The manometer scale had a sensitivity of 1 mm H₂O. Before recording the readings, air flow through the packed columns was checked and adjusted to the required flow rate.

Results and discussion

Effect of high nutrient supplementation

Removal of hexane during high nutrient supplementation is illustrated in Figure 2. Under high nutrient supplementation, the biodegradation efficiency was not significantly dependent on the type of the solid medium. Availability of nutrients resulted in a high removal of hexane in both P and PP columns. Within the first 7 days, the percent removal had reached 57 and 73% in column P and PP, respectively. The percent removal strongly depended on air flow rate and inlet concentrations of hexane which varied between 280 and 980 ppm (Table 1). Percentage removal values for both P and PP columns decreased as the air flow rates increased from 20 to 30 and 100 m³/m² h at the same range of hexane inlet concentrations. A similar trend of low percentage of VOC removal with increasing air flow rates was observed during biofiltration of triethylamine in compost biofilters (Lackey *et al.* 1998). The air flow rate affected the residence time of hexane in the biofilter, hence this trend of lower percentage removal at higher air flow rates was expected.

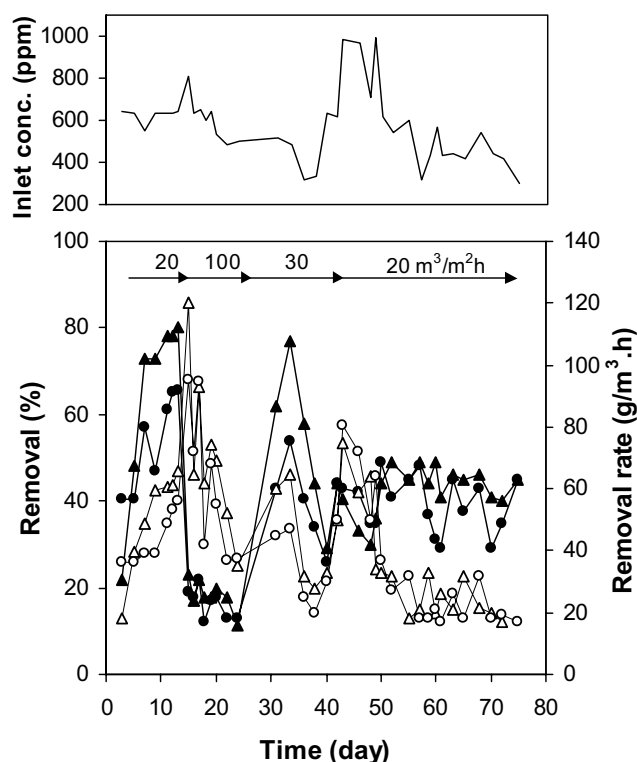


Figure 2. Removal of *n*-hexane in the mixture and perlite biofilters with high nutrient supplementation: (▲) mixture (%), (●) perlite (%), (△) mixture (g/m³ h), (○) perlite (g/m³ h).

Despite the decreasing percentage of hexane removal at higher air flow rates, the rate of hexane removal in g/m³ h increased with the air flow rate and concentration (Table 1, Figure 2). For the same range of hexane inlet concentration, rate of hexane removal was 56 g/m³ h at a flow rate of 20 m³/m² h on day 13, the rate increased to 96 g/m³ h within 48 h when the air flow rate was increased to 100 m³/m² h. A similar observation of increasing biofiltration rates with VOC inlet concentration was reported during biofiltration of acetone (Deshusses *et al.* 1997).

After 46 days of high nutrient supplementation, the addition of nutrients did not improve the biofilter performance (Figure 2). High biomass overgrowth was visually observed and the columns were clogged such that supplementary nutrients added could not easily pass down the columns. Percentage hexane removal remained at an average of 40% (25 g/m³ h) for both columns. By day 60, columns under high nutrient

Table 1. Hexane removal at different air flow rates with high nutrient supplementation.

Time scale (day)	5–13	14–27	28–46	47–75
Flow rate (m ³ /m ² h)	20	100	30	20
Inlet concentration (ppm)	560–680	500–720	320–980	280–640
Perlite (%)	40–67	10–24	27–54	30–50
(g/m ³ h)	34–56	36–96	20–80	16–40
Mixture (%)	73–80	11–24	36–75	40–50
(g/m ³ h)	50–67	34–120	27–71	16–40

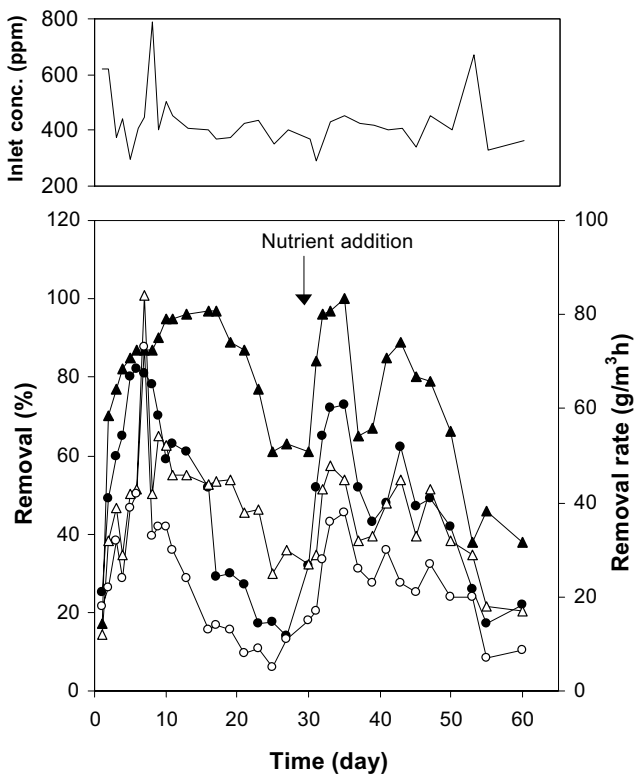


Figure 3. Removal of *n*-hexane in the mixture and perlite biofilters with low nutrient supplementation: (▲) mixture (%), (●) perlite (%), (△) mixture ($\text{g}/\text{m}^3 \text{ h}$), (○) perlite ($\text{g}/\text{m}^3 \text{ h}$).

supplementation developed a high pressure drop which forced the termination of the experimental run. Deshusses (1997) and Ziehr *et al.* (1993) suggested that a continuous supply of nutrients was not desirable because rapid biofilter clogging by excessive biomass growth would occur. It is necessary to properly manage the amount of nitrogen added to the biofilter such that reasonably high biofiltration rates are achieved with a limited microbial growth. A method for managing microbial growth was proposed by Smith *et al.* (1996). Smith reduced microbial growth when nitrogen source was nitrate instead of ammonia.

Effect of low nutrient supplementation

Biodegradation of hexane during low nutrient supplementation is illustrated in Figure 3. Biodegradation of hexane increased after the start of the experimental run in both biofilters due to the availability of the supplementary nutrients and decreased gradually with time

until another addition of the supplementary nutrients. The hexane removal rate in both P and PP columns increased exponentially during the first 7 days of biofiltration, reaching the maximum values of $73 \text{ g}/\text{m}^3 \text{ h}$ for P and $84 \text{ g}/\text{m}^3 \text{ h}$ for PP. The hexane percent removal in the PP column was maintained at the maximum level for 17 days then decreased gradually from 97 to 61% by day 30, when the second nutrient solution was added. From day 30 to day 60, the pattern of percentage and rate of hexane removal was similar to that of the first 30 days (Table 2). Similar biofiltration behaviour was observed when nutrients were re-added to the solid medium during the biofiltration of alcohol mixture (Pearson *et al.* 1991).

However, there was a significant difference in the performance between the two solid media; perlite and the mixture. The total amount of hexane removed in the PP column was significantly higher (40%) than that removed in the P column. The results indicated that within 60 days, higher biodegradation rate was achieved by a combination of both perlite and peat. This result suggested that the mixture provides better properties than those of perlite alone. This was attributed to the fact that the mixture of peat and perlite had a higher nitrogen content, higher water retention, and higher immobilization capacity than perlite alone.

Utilization of nitrogen

Biodegradation of hexane was accompanied by the utilization of nitrogen (Figure 4). Analysis of ammonia nitrogen concentration in the solid media showed that an increased removal rate occurred when a significant amount of ammonia was available in the solid medium and decreased as the ammonia content got depleted. The dependency of biodegradation rates on nitrogen content suggested that hexane removal was mainly due to the microbial growth. In the absence of nitrogen source, hexane was removed through oxidation to provide maintenance energy and limited microbial growth depending on how fast the nitrogen was recycled from dead cells. At the beginning of the low nutrient supplementation experiment, columns received equal amount of ammonia nitrogen per unit volume, but the PP column registered higher ammonia nitrogen ($450 \text{ g}/\text{m}^3$) than the P column ($200 \text{ g}/\text{m}^3$) despite the negligible amounts of liquid drained from the columns (Figure 4). The extra ammonia nitrogen could only have come from the peat. The peat used in this research contained 4% (w/w) total nitrogen. When nutrient

Table 2. Hexane removal at $20 \text{ m}^3/\text{m}^2 \text{ h}$ and average concentration of 400 ppm with low nutrient supplementation.

Time scale (day)		4–30	30–60
Perlite	(%)	16–76	20–72
	($\text{g}/\text{m}^3 \text{ h}$)	6–50	6–40
Mixture	(%)	60–99	38–100
	($\text{g}/\text{m}^3 \text{ h}$)	24–54	15–58

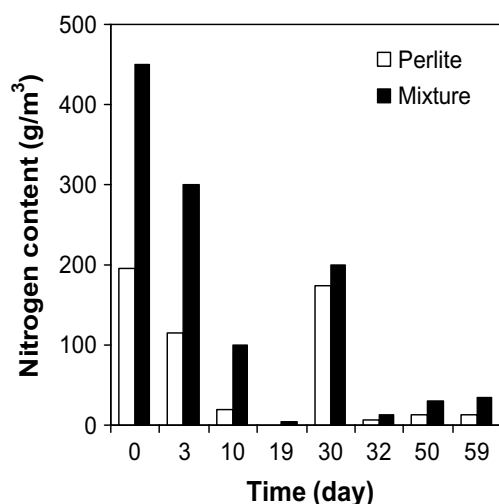


Figure 4. Nitrogen utilization in the mixture and perlite biofilters with low nutrient supplementation.

solution was added to the columns for the second time (on day 30), variation in ammonia adsorption capacity was observed. The amount of immobilized nitrogen was calculated as the difference in ammonia nitrogen contents of the added nutrient solution and the liquid drained from the columns. The mixture showed higher ammonia immobilization capacity (200 g/m^3) than perlite (180 g/m^3). This indicated that the mixture of peat and perlite had higher (20%) adsorption capacity for ammonia than the perlite.

The concentration of ammonia nitrogen within the columns decreased faster in the bottom section than in the middle and top sections in the PP column. The nitrogen concentration gradient along the biofilter columns was due to two factors: the bottom section where the polluted gas entered the column had higher hexane removal rates due to high hexane concentration and hence higher utilization of nitrogen than the top sections. Second, the drained water was recycled through the top of the columns, resulting in a high accumulation of nitrogen in the top sections.

CO₂ generation

Measurement of carbon dioxide generation in biofilters was measured for the low nutrients supplementation experiment. The amount of carbon dioxide generated in the biofilters depended on the type of solid medium (Figure 5). More CO₂ was generated in the PP column than in the P column. During the first 5 days of biofiltration, carbon dioxide generated in both P and PP columns increased rapidly from zero to more than 1600 ppm, and decreased gradually to approximately 1000 ppm for the PP column and 200 ppm for the P column by day 30. A second addition of nutrient solution after 30 days of biofiltration revived generation of CO₂ and the pattern was repeated. The pattern of CO₂ generation was similar to the pattern of hexane

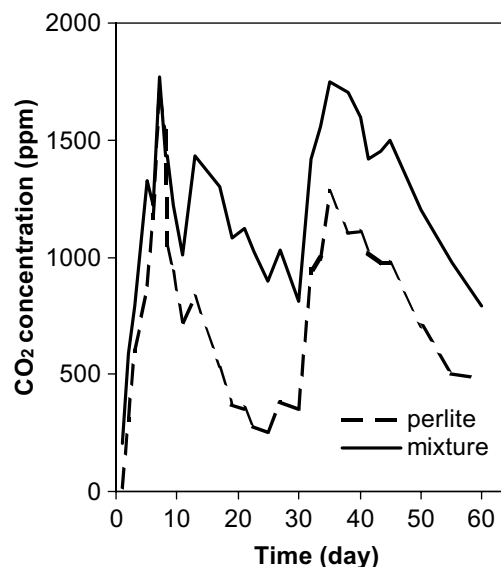


Figure 5. Carbon dioxide profile from the mixture and perlite biofilters with low nutrient supplementation.

removal rates (Figures 3 and 5), showing a strong correlation between the hexane removal rates and CO₂ generation. Higher hexane removal corresponded with high CO₂ generation and vice versa. From the observation, carbon dioxide release from biofilters can be used as a measure of biofiltration performance to indicate the VOC biodegradation rates.

Pressure drop

Figure 6 shows pressure drop trends across the columns which were under high nutrient supplementation. The PP column showed higher pressure drop values than the P column. The pressure drop for the P column increased with air flow rate. During the first 10 days, at $20 \text{ m}^3/\text{m}^2 \text{ h}$, the pressure drop was between 20 and 30 Pa/m, increased to 130–230 Pa/m at $100 \text{ m}^3/\text{m}^2 \text{ h}$, then decreased to 5–100 Pa/m when the air flow rate was reduced to $30 \text{ m}^3/\text{m}^2 \text{ h}$. From day 46, air flow rate in the biofilter remained at $20 \text{ m}^3/\text{m}^2 \text{ h}$ until the end of experiment (day 70). During this period, water added to the column to restore moisture caused pressure drop to increase to 80–120 Pa/m (Figure 6). Similarly for the PP column, the pressure drop was 58–94 Pa/m at $20 \text{ m}^3/\text{m}^2 \text{ h}$, increased to 890–1730 Pa/m at $100 \text{ m}^3/\text{m}^2 \text{ h}$, then decreased to 320–1770 Pa/m at $30 \text{ m}^3/\text{m}^2 \text{ h}$. From day 46 to day 70, air flow rate remained $20 \text{ m}^3/\text{m}^2 \text{ h}$, but the pressure drop increased and remained at 2400–2930 Pa/m after addition of water to restore moisture (Figure 6).

Due to the addition of supplementary nutrients, there was biomass overgrowth (visually observation) which increased such that, during the last week of experiment, supplementary nutrient solution could not pass down through both columns. At the end of the experiment, disintegration of peat particles into much smaller ones was visually observed. High pressure drop between day

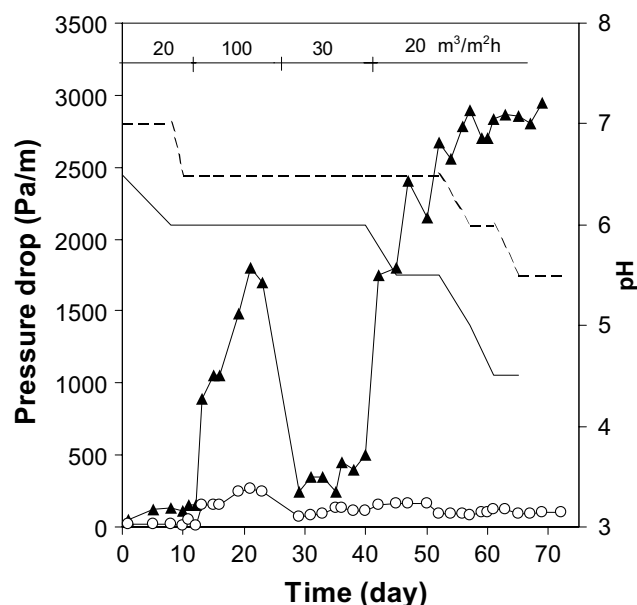


Figure 6. Variation of pressure drop and pH in the mixture and perlite biofilters with high nutrient supplementation: (—▲—) mixture (Pa/m), (—○—) perlite (Pa/m), (—) mixture (pH), (---) perlite (pH).

50 and day 70 coincides with change from high to low pH values; 7 to 5.5 for P column and 6.5 to 4.5 for PP column, suggesting that microbial community might have favoured low-pH tolerant fungi, hence high pressure drop and consequently biofilter clogging and failure. Wu *et al.* (1999) suggested pH values between 7 and 7.2 for bacterial growth and better biofilter performance.

High pressure drop is not desired in biofilter operations due to the associated high running cost. It is therefore necessary that pressure drop remains as low as possible. Under high nutrient supplementation, the P column showed a lower pressure drop than the PP column. Since the biofiltration efficiency of the P column was comparable to that of the PP column, it may be concluded here that perlite offers an opportunity of running biofiltration process at a lower and stable pressure drop if the nutrient supplementation is managed properly.

Pressure drop profiles under reduced supplementary nutrient addition were not shown due to the negligible pressure drop. Throughout the 60 days of the experiment, the P column showed the lowest pressure drop with an average value of 10 Pa/m. Pressure drop for the PP column ranged between 10 and 20 Pa/m. Kiared *et al.* (1996) reported a pressure drop of 600 Pa/m at air flow rate of 70 m³/m² h for biofiltration of acetone.

Our observation indicates that high nutrient supplementation resulted in pressure drop values far higher than recommended values of 100 and 700 Pa/m for practical application (Pearson *et al.* 1991; Ergas *et al.* 1995). On the contrary, low nutrients supplementation had pressure drop far below the recommended range.

Conclusion

Our short-term biofiltration of hexane under high and low nutrient supplementation was dependent on the availability of nutrients. Hexane biodegradation rates were higher during high nutrient supplementation than during low nutrient supplementation. High nutrient supplementation resulted in excess biomass growth and high pressure drop. Under low nutrient supplementation, the pressure drop for the two solid media was low and not significantly different and not much biomass accumulation occurred to cause high pressure drop. To prevent the increasing of pressure drop resulting from excess biomass growth, a low level of nutrient supplementation should be maintained. It is suggested here that if a method to remove excess biomass from biofilters is developed, biofilters can be operated under high nutrient supplementation protocol with a periodical removal of the excess biomass. Such operation will increase overall biofiltration rates and reduce biofilter volumes on a long-term basis. In that case, a mechanically strong non-biodegradable media should be used instead of biodegradable peat.

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