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Pilot-Scale Biofilter Treatment of Hazardous Air Pollutants

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파이로트-규모 바이오필터의 유해성 대기오염물질 처리

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요약

폴리우레탄, 폴리에스테르, 바이페놀, PVC 외 각종 농약 등을 생산하는 울산의 모 화학공장에서, 다양한 종류의 휘발성유기화합물질들(VOCs)이 배출되고 있다. 평균적인 휘발성유기화합물질의 배출 농도는 7283 ppm으로, 톨루엔, 페놀을 포함하여 Trimethyl-pentene, trimethyl-hexene, dimethyl-cyclohexane 등이 검출되었다. Trimethyl-pentene, trimethyl-hexene, dimethyl-cyclohexane 등은 인화성이 강하며 화재를 일으킬 위험성이 매우 큰 것으로 알려져 있고, 특히 톨루엔과 페놀의 경우는 호흡이나 피부접촉 등을 통한 인체로의 유입이 있을 경우 유독성을 나타내게 된다. 이러한 VOCs 제거를 위하여 겨울철 기간에 파이로트-규모의 바이오필터 적용 실험이 진행되어 졌다. 본 연구의 목적은 바이오필터 운영이 진행되는 가운데 온도, 함수비, 하중, 압력손실 등의 제한요소들이 미디어 내부에서 변화하는 상황에 대한 관찰 및 평가에 있다. 이러한 제한요소들은 바이오필터의 디자인과 오염물질 제거에 심대한 영향을 미치게 된다. 바이오필터는 옥외에 설치되어 총 44일간 운영되어 졌는데, 외부 영하온도의 영향을 최소화하기 위하여, 7 cm 두께의 파이버-글래스 소재 단열설비가 반응기 외부에 설치되었고 또한 150°C의 스팀이 바이오필터 반응기와 단열설비 사이에 제공되어 졌다. 바이오필터 반응기 내부에는 23개의 온도 측정 센서와 함수비 센서, 공기샘플포트, 습도계 등이 각기 다른 장소에 설치되어 온도, 함수비 등의 제한요소 영향연구가 진행되었다. 미디어 내부 같은 높이의 서로 반대되는 위치에서 온도차가 13.7도에서 −8.3도까지 차이가 나는 것으로 관찰되었으며, 미디어 높이 위치의 변화에 따라서도 21도에서 2도까지 차이를 나타냈다. 바이오필터 함수비는 실험기간 동안 지속적으로 변화가 발생하였는데, 스팀이 제공되는 동안에는 미디어 함수비가 훨씬 빠른 속도로 증가됨이 관찰되어 졌다.

Key words : biofilter, temperature, pilot-scale, VOC, steam heating, water content

INTRODUCTION

Volatile organic compounds (VOCs), many of which are priority pollutants, may result in serious short-term and long-term effects on human health and the environment (Zhu *et al.*, 2004). Recently, VOCs and offensive odors discharged by petrochemical factories and industrial waste treatment factories emerged as new sources of pollution, which became an increasing annoyance to Korean residents

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living nearby. As a result, regulations in Korea became increasingly stringent and it has stimulated research efforts to develop more efficient and more cost-effective control technologies. Biofiltration is an emerging and promising air pollution control technology. Both organic and inorganic air pollution compounds that are toxic to humans and volatile organic compounds from a variety of industrial and public sector sources have been effectively removed with biofiltration (Sorial *et al.*, 1997; Zhu *et al.*, 2004; Qi *et al.*, 2005; Yang and Allen, 2005). Important advantages of biofiltration systems over other air-pollution-control technologies include low capital and operating costs, low energy requirements, and the absence of residuals and by-products requiring further treatment or disposal.

Research on the biofiltration system has been performed to treat VOCs from chemical manufacturing factory, located in Ulsan Industrial Complex, Korea. The company produces variety of chemical products, including PVC, polyurethane, polyester, biphenol, agrochemicals, etc. A pilot-scale biofilter was installed outdoor in the factory and tested from the middle of January until early March. Some of the key rate-limiting factors (like temperature, water content, loading rate, and pressure drop) significantly affect the biofilter performance and design. The objective of this paper is to observe the fluctuations of the rate-limiting factors in a biofilter and investigate the effects of these factors on the biofilter performance. The experiment was performed during winter time with the pilot-scale biofilter insulated and steam-heated.

The relationship between loading rate and removal efficiency was investigated by many researchers (Hwang *et al.*, 2002; Morales *et al.*, 2003; Morgan-Sagastume *et al.*, 2003; Qi *et al.*, 2005; Yang and Allen 2005). According to previous studies, removal efficiency decreases as the loading rate increases. Determination of maximum loading rate is an essential part of full-scale biofilter design. Devinny and Hodge (1995) experienced a GAC biofilter upset when the biofilter was overloaded with ethanol. According to Devinny and Hodge, biofilter upset caused by overload can result in the reactor performance being degraded and toxic intermediates being carried out of the biofilter.

Van Lith *et al.* (1997) said the moisture content is the operating parameter most relevant to performance and most sensitive to changes. Hwang *et al.* (2002), Morales *et al.* (2003), Morgenroth *et al.* (1996), Leson and Smith (1997), and Morgan-Sagastume *et al.* (2003) also pointed out the importance of water content in their papers. Excessive water in a biofilter may cause elevated pressure drops as water displaces air in the interparticle spaces (van Lith and Leson, 1997; Gostomski *et al.*, 1997; Swanson and Loehr, 1997; Auria *et al.*, 1998), diffusional limitations in the bed for compounds with low water solubility (van Lith and Leson, 1997; Gostomski *et al.*, 1997; Swanson and Loehr, 1997), oxygen transfer problems due to reduced air/water interface per unit biofilm volume (Swanson and Loehr, 1997), creation of anaerobic zones that promote odor formation and slow degradation rates (Swanson and Loehr, 1997; Auria *et al.*, 1998), and Nutrient removal from the biofilter medium (Swanson and Loehr, 1997; Auria *et al.*, 1998).

Insufficient water content may cause a decrease in the microbial degradation rate in the biofilm layer (Gostomski *et al.*, 1997; Swanson and Loehr, 1997; Auria *et al.*, 1998) and cracking to occur within the biofilter medium. The air may then be short-circuited through these cracks, decreasing performance (Gostomski *et al.*, 1997; Swanson and Loehr, 1997; Auria *et al.*, 1998).

As the temperature of biofilter rises, chemical and enzymatic reactions in the microbial cell proceed at more rapid rates and growth becomes faster in a biofilter media. However, above a certain temperature, proteins, nucleic acids, and other cellular components may be irreversibly damaged. Four groups of microorganisms in relation to their temperature optima can be broadly distinguished: psychrophiles, with low temperature optima, mesophiles, with midrange temperature optima, thermophiles, with high temperature optima, and hyperthermophiles, with very high temperature optima (Madigan *et al.*, 1997).

Sorial (1997) investigated temperature effects on toluene removal with peat biofilter. He achieved a removal efficiency of 99% at 32.2°C with a toluene loading rate at 0.45 kg COD/m^3-day and EBRT of 2 minutes. In contrast, at a temperature of 11.1°C, the removal efficiency was 58% with the same operating conditions. Sorial concluded that the optimal temperature for the biofilter operation was 32.2°C.

Lackey (1998) reported significantly higher styrene removal efficiency at biofilter during the summer season than winter. The average temperature of inlet air stream to biofilter was 32.2°C for the summer season, while 13.8°C for the winter season. According to Lackey, optimum inlet gas temperature ranges between 20 and 40°C.

Media pressure drop depends on structure and moisture content of filter material, off-gas face velocity, and bed depth. Pumps and controls contribute smaller amounts of pressure drop. For a given residence time and media, single-bed biofilters show the lowest pressure drop. Systems with two beds in series have twice the face velocity and effective bed height and at least four times the pressure drop of a single layer system (van Lith and Leson, 1997).

MATERIALS AND METHODS

Biofilter media

Composting material was mainly used as the biofilter media. Composting material was manufactured from a local composting facility (Jung-Won Farm, Changwon, Korea). Two different types of hardwood saw dust and cow manure were used for the raw material of the compost. It took about 30 days for the composting facility to produce the composting material. The pH of the compost was 7.2 ($\pm$0.1).

Biofilter design

A schematic of the biofiltration system is shown in Fig. 1. The body of the biofilter reactor was constructed with steel with an internal diameter of 230 cm and a height of 260 cm. The Biofilter consisted of the following sections, from top to bottom.

1) A headspace for housing the water spray nozzles.
2) A section containing the biofilter media. The height of the biofilter media was one meter and the size of the media was about 4.3 m^3. Conceptually, the biofilter was divided into five different consecutive stages. The height of each stage was 20 cm.
3) A bottom space to collect leachate from the biofilter.

Inlet and outlet relative humidity (R.H.$_1$ and R.H.$_2$) were measured using a DUO humidity indicator and a Capacitive RH Sensor (rbr-Computertechnik, Germany). Relative humidity of inlet air stream was maintained at almost 100% for most of the experimental period. The body of the humidifying water chamber was constructed with steel with an internal diameter of 76 cm, and a height of 130 cm. Water was sprayed through nozzles inside the humidification chamber against the air flow and relative humidity was measured for the entire experimental period.

Inlet and outlet air pressure (A.P.$_1$ and A.P.$_2$) was measured with SS-3011 pressure gauges (Woo-Jin Electronics, Co. Ltd., Korea). Using ThetaProbe soil moisture sensors (type ML2, Delta-T Devices Ltd., Cambridge, U.K.), the water content of the biofilter media at each stage was measured. Water content sensors were inserted inside the biofilter media at different heights and different locations from the center of the media. Table 1 shows the water content sensor codes, corresponding stages, depth, and distances from the center, where the sensors were located inside the media.

Temperatures of the biofilter media at five different consecutive stages (stages # 1, # 2, # 3, # 4, and # 5) were measured using a model 865F thermistor meter and model OL-710-PP probes (Omega Engineering Inc., Stamford, CT). Temperature sensors were spaced throughout the media: 3 to 5 sensors were spaced at 30 cm to 60 cm intervals at various heights throughout the biofilter media. Table 2 shows

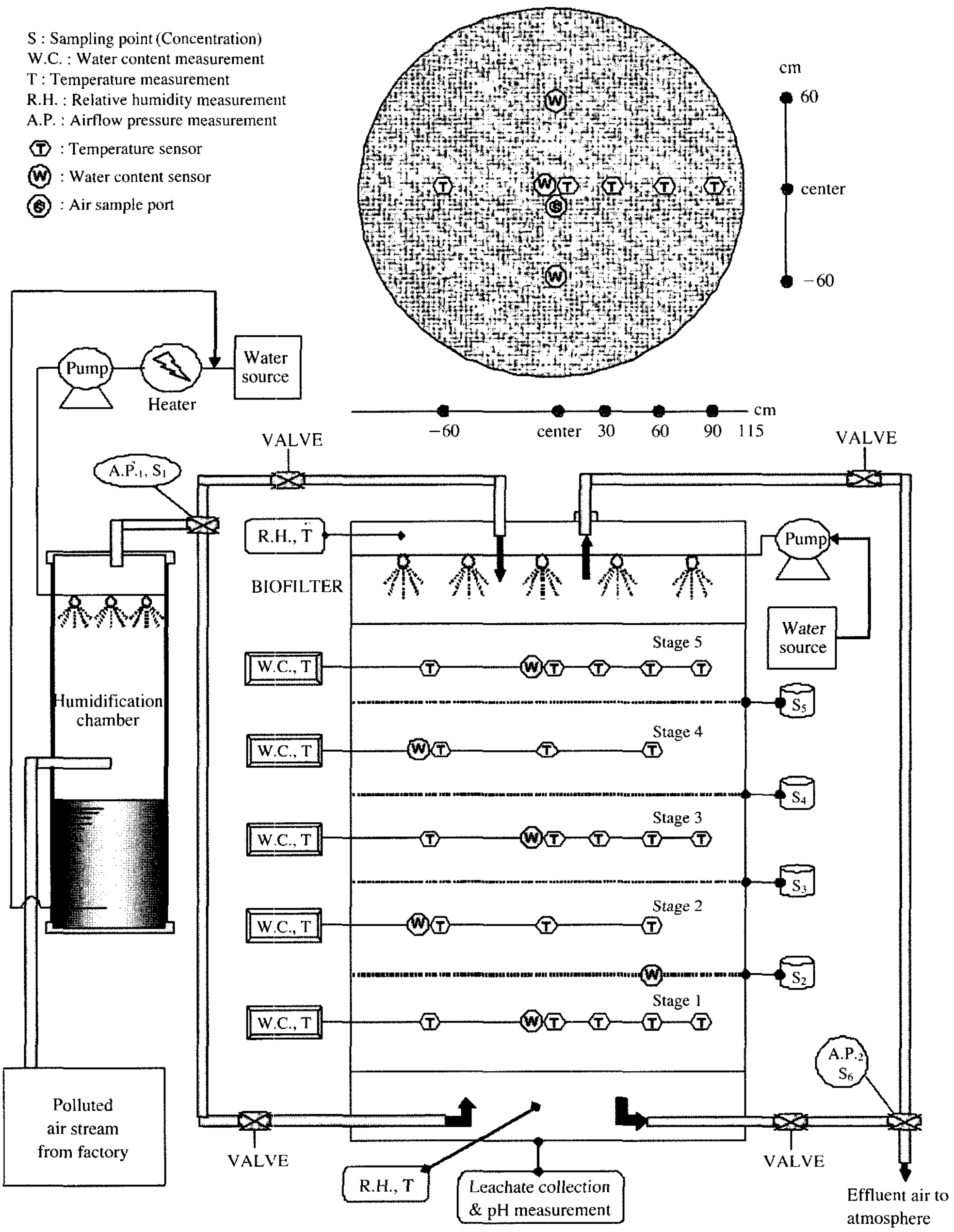

Fig. 1. Schematic of the biofiltration system.

the sensor codes, corresponding stages, depth, and distances from the center, where the temperature sensors were located inside the media.

As the pilot-scale biofilter was installed outdoor and operated during winter time (from middle of January until early of March), temperature management for the biofilter was needed. To overcome the outside freezing cold weather, 7 cm thick fiber glass insulation and 150°C steam heating was used. Seven millimeter diameter copper pipe circulated the main

Table 1. Water content sensor location inside the pilot-scale biofilter

Sensor code	Corresponding stage	Height	Distance from center
St # 1	Stage # 1	10	0
St # 1-2	Border of Stage # 1 & # 2	20	60
St # 2	Stage # 2	30	−60
St # 3	Stage # 3	50	0
St # 4	Stage # 4	70	−60
St # 5	Stage # 5	90	0

Table 2. Temperature sensor location inside the pilot-scale biofilter

Sensor code	Corresponding stage	Height	Distance from center
1-1	Stage # 1	10	90
1-2	Stage # 1	10	60
1-3	Stage # 1	10	30
1-4	Stage # 1	10	0
1-5	Stage # 1	10	−60
2-1	Stage # 2	30	60
2-2	Stage # 2	30	0
2-3	Stage # 2	30	−60
3-1	Stage # 3	50	90
3-2	Stage # 3	50	60
3-3	Stage # 3	50	30
3-4	Stage # 3	50	0
3-5	Stage # 3	50	−60
4-1	Stage # 4	70	60
4-2	Stage # 4	70	0
4-3	Stage # 4	70	−60
5-1	Stage # 5	90	90
5-2	Stage # 5	90	60
5-3	Stage # 5	90	30
5-4	Stage # 5	90	0
5-5	Stage # 5	90	−60

body of the biofilter reactor for steam heating. Total height of the biofilter reactor body was 2.6 meter and the height of media was one meter. Around middle section of the wall of the biofilter reactor where the media attached inward, copper pipe circulation was installed at 10 cm interval. At the lower and upper sections of the wall of the biofilter reactor (where the media is not attached inward directly), it was installed at 20 cm interval. Fiber glass insulation covered the outside of the reactor body over the copper pipe circulation. In the case of humidifying water chamber, only fiber glass insulation was made without steam heating.

For an initial irrigation of the biofilter, about 10.0 m^3 of chlorine-free water were sprayed through the nozzle in the headspace of the reactor. The leachate in the bottom of the reactor was recycled until the whole biofilter media was thoroughly saturated. After the initial water irrigation of the biofilter media, no exterior water was directly added to the reactor for the remainder of the experimental period.

VOC analysis

Inlet and outlet air samples from each stage of the biofilter (S_1, S_2, S_3, S_4, S_5, and S_6) were collected in one liter Tedlar sample bags. The concentrations of VOCs were measured using a Varian Saturn-3 Gas Chromatography/Mass Spectrometry (GC/MS). Five hundred micro-liters from the Tedlar bag sample were injected into the GC injection port with a Hamilton Gastight #1750 syringe. For species separation, a 0.25-mm ID*30 m DB5 capillary column (J & W Scientific, Folsom, CA) containing 0.25 μm film thickness was used. Helium (He) was used as the carrier gas at a flow rate of 8 mL/min. The injection temperature was 220°C and the transfer line temperature to the Mass Spectrometry was set at 220°C. The total sample run for each VOC analysis was 28 minutes in duration. For the first 5 minutes, the GC oven temperature was maintained at 30°C and then the temperature was increased to 130°C at a rate of 10°C/min, then it was increased to 250°C at a rate of 15°C/min. At 250°C, the GC oven temperature was maintained for 5 minutes. A single point external standard calibration method was used. All results are given in parts per million (ppm) by volume.

Retention time and loading rate to the biofilter

Fig. 2 shows the empty bed residence time for biofilter reactor and humidifying water chamber

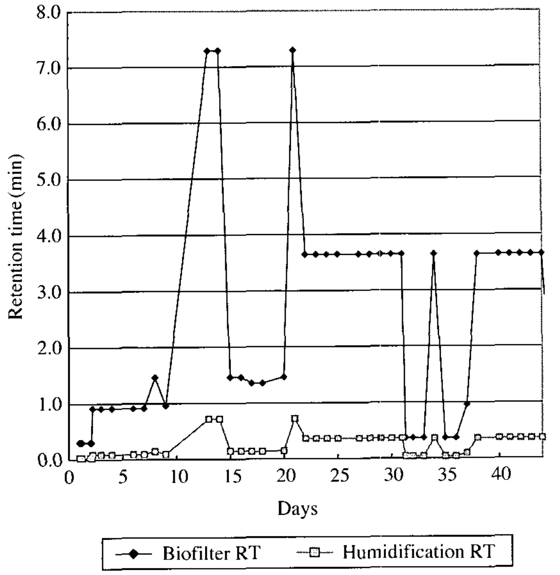

Fig. 2. Residence time for biofilter reactor and humidifying water chamber.

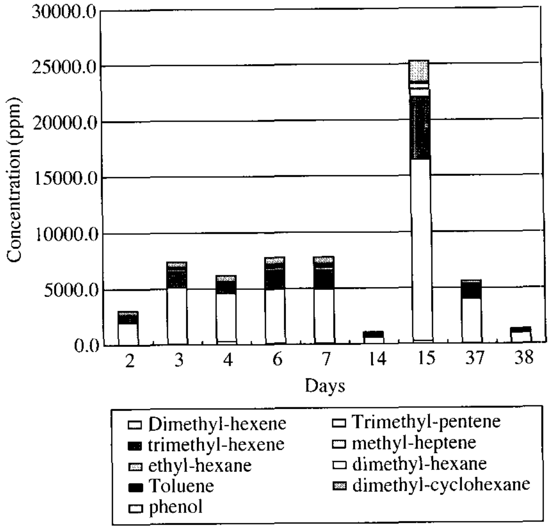

Fig. 3. Daily VOCs measurement of the inlet air flow from chemical manufacturing process.

during the experiments. Volatile organic compounds (VOCs) COD loading rate (kg COD/m^3) to the biofilter was calculated based on the individual stage volume (0.83 m^3). During the experiment, only on day 2 to 7, 14, 15, 37, and 38, the contaminated air stream was provided to the biofilter system. On other days, ambient air was supplied to the system to maintain proper operational condition.

RESULTS AND DISCUSSION

Applied VOC concentration and acclimation period

People exposed to hazardous air pollutants at sufficient concentrations and durations may have an increased chance of getting cancer or experiencing other serious health effects. These health effects can include damage to the immune system, as well as neurological, reproductive, developmental, respiratory and other health problems. Like humans, animals may experience health problems if exposed to sufficient quantities of air toxics over time. People are exposed to toxic air pollutants in many ways such as by breathing contaminated air, eating contaminated food products, drinking water contaminated by toxic air pollutants, ingesting contaminated soil, and touching contaminated soil, dust, or water. Once toxic air pollutants enter the body, some persistent toxic air pollutants accumulate in body tissues.

Feasibility test for a pilot-scale biofilter was performed to treat VOCs, many of which are hazardous air pollutants, during winter season. The polluted air stream was produced from manufacturing processes of PVC, polyurethane, polyester, biphenol, agrochemicals, etc. The biofilter experiment was conducted for 44 days. Fig. 3 shows VOCs measurement of the inlet air flow to the biofilter system. It shows VOCs concentrations and the compositions of each chemical compound on selected days. Table 3 shows the average concentrations and percent ratio of chemical compounds of VOCs. Total average concentration was 7,283 ppm and trimethyl-pentene, trimethyl-hexene, dimethyl-cyclohexane, methyl-heptene, toluene, phenol and etc. were detected. Trimethyl-pentene, trimethyl-hexene, dimethyl-cyclohexane, and methyl-heptene are flammable and are known to

Table 3. Average VOCs concentrations and percent ratio of the inlet air flow to the biofilter system

Compounds	Concentration (ppm)	Percent ratio (%)
Dimethyl-hexene	91	1.2
Trimethyl-pentene	4,766	65.4
trimethyl-hexene	1,475	20.3
methyl-heptene	177	2.4
ethyl-hexane	9	0.1
dimethyl-hexane	128	1.8
Toluene	105	1.4
dimethyl-cyclohexane	533	7.3
phenol	1	0.0001
Total	7,283.0 (ppm)	100.0 (%)

have dangerous fire risk. Toluene and phenol are toxic to human body when they were received by ingestion, inhalation, and skin absorption. In the case of phenol, it is strongly irritant to tissue and its threshold limit value (TLV) is 5 ppm in air. Threshold limit value for toluene is 100 ppm and its explosive limits in air is 1.27 ~ 7% (Lewis, 1997).

Microorganisms exposed to a new substrate may require a period of acclimation before they begin vigorous degradation. Excessive pollutant loading may cause a significant decrease of removal efficiency in the biofilter (Devinny and Hodge, 1995). Therefore, when new substrate is provided, or significant operational conditions and/or microbial growth environment are changed, an acclimation period is necessary.

Initially, misinformation was provided from the vender and the range of inlet air stream VOCs concentration was expected from about 50 ppm to 300 ppm. However, actual incoming concentration to the biofilter system was measured from 1,000 ppm to 25,000 ppm. The unexpected heavy loading rate became a severe shock for the biofilter to operate properly. As a result, the contaminated air stream from chemical manufacturing process could not be provided to the biofilter all the time. On selected nine days (days 2 to 7, 14, 15, 37, and 38), the contaminated air stream was provided. On the other days, ambient air was supplied to the system.

Table 4. Input and output concentration (ppm), loading rate (kg COD/m^3), and removal efficiency (%) of the biofilter

Days	Input concentration (ppm)	Output concentration (ppm)	Input loading rate (kg COD/m^3)	Output loading rate (kg COD/m^3)	Removal efficiency (%)
2	3,041	1,842	720	439	39
3	7,419	2,524	1,756	597	66
4	6,201	5,200	1,468	1,233	16
6	7,799	6,187	1,844	1,457	21
7	7,764	6,159	1,840	1,454	21
14	3,305	1,080	64	21	67
15	25,255	11,277	3,735	1,681	55
37	5,616	83	332	3	99
38	3,341	556	138	23	83

The total removal efficiency of the system fluctuated on the days when polluted air stream was provided (Table 4). Total loading rate to the system was calculated between 60 to 3,700 kg COD/m^3 based on the inlet air flow VOCs concentration. Because of extremely heavy loading rate and severe fluctuation of loading rate (60 to 3,700 kg COD/m^3) to the biofilter system, acclimation period was essential and it took much longer time to manage and reduce the inlet concentration and flow rate to a desirable level. Until day 7, about 1,500 kg COD/m^3 was provided to the biofilter system. As it was too heavy for the biofilter, ambient air was provided from day 8. On day 14 and 15, the polluted air stream was provided to the biofilter system. The loading rate was 65 and 3,700 kg COD/m^3 and removal efficiency was about 67 and 55 percent. Particularly, the loading rate on day 15 was extremely heavy and it made serious shock to the biofilter. Therefore, another period of ambient air supply was made until day 36. On day 37 and 38, polluted air stream was provided (330 and 140 kg COD/m^3) and removal efficiency was 99% and 83% each. Unfortunately, VOC measurement was not available from day 40 until day 44, because of mechanical problem of GC/MS. More acclimation time and test period was needed to determine the maximum loading rate for the pilot-scale biofilter to manage with given operating condition.

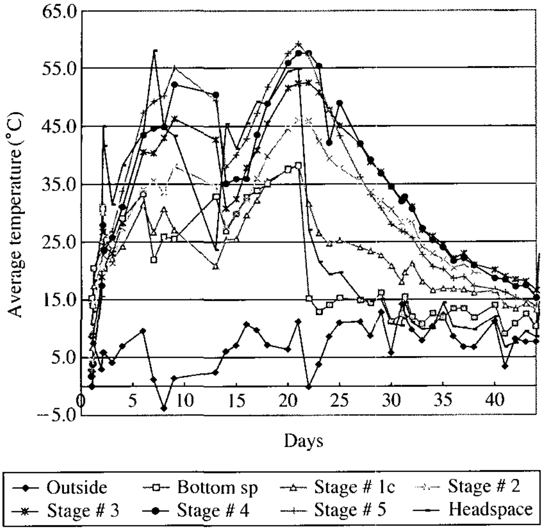

Fig. 4. Average temperature of each stage in the biofilter media.

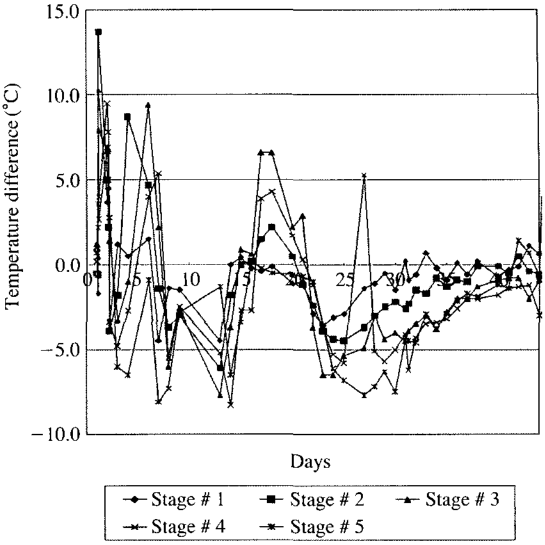

Fig. 5. Temperature differences between location (60 cm) and (−60 cm) from center at each stage.

Temperature

Temperature was measured for the entire experimental periods. The pilot-scale biofilter was installed outdoor and the experiment was performed from middle of January until early of March. Ambient temperature continued to be fluctuated and occasionally dropped below zero. Fig. 4 shows the average temperature of each stage, including the outside ambient temperature. Average temperature of each stage ranged from 6°C to 59°C. For temperature management of the biofilter reactor, 7 cm thick fiber glass insulation and 150°C steam heating was used.

In the case of steam heating, it was provided until day 21 and resulted in general increase of average temperatures inside the biofilter reactor. Without steam, all the temperature measurements inside the biofilter reactor started to be decreased. Among them, the temperatures of headspace and bottomspace decreased most dramatically. With steam heating, temperatures of stage # 5 and headspace were highest and temperature difference between media and outside ambient air was getting greater with proceeded operation. However, without steam, temperatures of stage # 3 were highest and temperature differences with each other generally decreased. Among temperature measurements of the media, stage # 1 was the lowest during the entire experimental periods. With steam heating, temperatures of the top sections of the media were greater than those of bottom sections. Without steam heating, intermediate stages (like stage # 3) generally had higher temperature measurement than those of bottom and top stages. Average temperature differences between stage # 1 and # 4 was 21 (C on day 21, and it was reduced to 2°C on day 44.

Observation was made whether one side of the media had higher temperature than the opposite side at the same depth. Fig. 5 shows the temperature differences of opposite locations at (60 cm) and (−60 cm) from center of all the stages. In general, temperatures of one side (northern side) of the media are usually higher than those of the opposite side (southern side) at the same depth. From time to time, warmer side of the media was changed at different stages. The range of temperature differences of opposite locations were 13.7°C to −8.3°C during the experiment.

Temperature management of inner media with

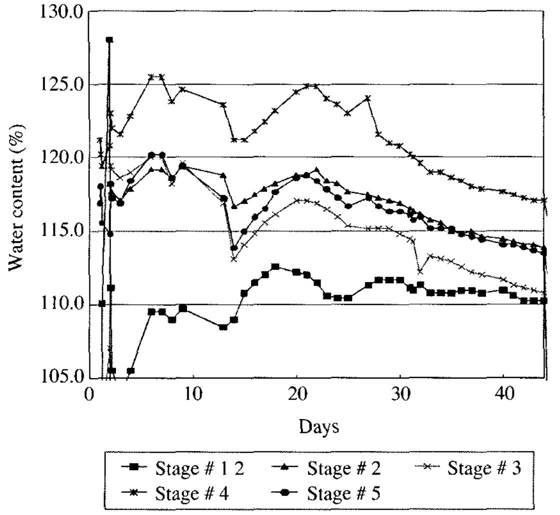

Fig. 6. Water content of the biofilter media.

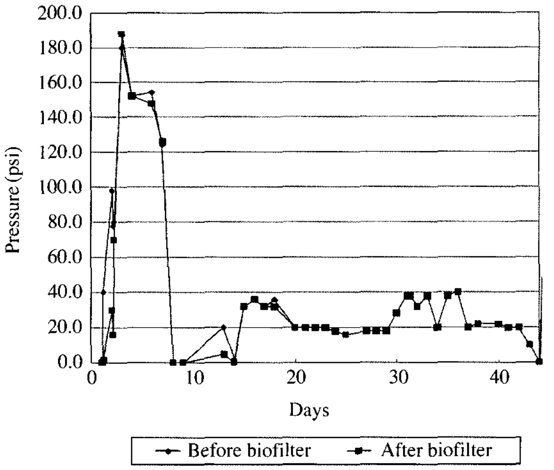

Fig. 7. Pressure measurement before and after biofilter media.

fiber glass insulation and steam heating was feasible even on the freezing cold whether outside. In most cases, fiber glass insulation alone may tolerate even below zero temperatures. With steam heating, outer side of the media was warmer than inner side of the media. Therefore, manipulation method to make the temperature of the entire sections uniform needs to be developed.

Water content

Water content of the biofilter media was measured (Fig. 6). Until day 7, water content of all the stages increased and then reduced until day 14. With polluted air stream provided, water content was increased until day 7. With ambient air stream provided, water content decreased until day 14. From day 15 to day 21, water content of all stages increased again, then they decreased until the last day of the experimental period. With adjusted and reduced air flow to the biofilter system, water contents increased from day 15 until day 21. However, without steam heating, water contents decreased until the last day of the experimental period. This phenomenon could be related with the temperature decrease of the biofilter media. As temperature of media decreases, water holding capacity of the media might also decrease. In general, water content of each stage on the day 44 reduced about ten to five percent compared to the measurements on day 21.

Increased temperature of inlet air stream substantially escalate the amount of water vapor carried by the air flow. With the steam heating, water content of each stage can be increased more rapidly. Therefore, management for water content of the media can be assisted with the steam heating. No leachate was generated at the bottom of the reactor during the entire biofilter operation. Therefore, no nutrient could be removed away from the biofilter media (Striebig *et al.*, 2001; Son and Striebig, 2001).

Pressure drop

Pressure was measured before and after the biofilter media for the entire experimental periods (Fig. 7). Each pressure measurement (before and after biofilter media) fluctuated with different flow rate to the biofilter. Until day 2, pressure drop of the biofilter media was very high and it was measured 68 psi. However, from day 3, pressure drop reduced dramatically and it was measured less than 15 psi for the last of the experimental periods. The initial big pressure drop could be caused by the initial water irrigation. The applied water might block the air stream pathway and with further proceeded biofilter operation, air stream pathway could be readjusted.

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