Control of VOC emissions from a flexographic printing facility using an industrial biotrickling filter

F. Sempere, V. Martínez-Soria, J. M. Penya-roja, A. Waalkens and C. Gabaldón

ABSTRACT

The study of an industrial unit of biotrickling filter for the treatment of the exhaust gases of a flexographic facility was investigated over a 2-year period with the objective to meet the volatile organic compound (VOC) regulatory emission limits. Increasing the water flow rate from 2 to 40 m³ h⁻¹ improved the performance of the process, meeting the VOC regulation when 40 m³ h⁻¹ were used.

An empty bed residence time (EBRT) of 36 s was used when the inlet air temperature was 18.7 °C, and an EBRT as low as 26 s was set when the inlet temperature was 26.8 °C. During this long-term operation, the pressure drop over the column of the bioreactor was completely controlled avoiding clogging problems and the system could perfectly handle the non-working periods without VOC emission, demonstrating its robustness and feasibility to treat the emission of the flexographic sector.

INTRODUCTION

Legal requirements to preserve air quality and to avoid harmful conditions for fauna, flora, and citizens are becoming more restrictive. Non-methane VOCs are among the most important pollutants that are released into the atmosphere due to different human activities, with a high percentage of the total VOC emission coming from industrial sources. In the European Union-27 (EU), 8963 million tonnes of VOCs were released in 2007. Reductions in the transport sector together with the limits imposed by the EU in the emission and use of solvents allowed reduction of VOC emissions; however, 10 of 27 Member States still have to meet their respective emission ceilings established for 2010 by the EU National Emission Ceilings Directive (European Environmental Agency 2009). Therefore, when VOC emissions cannot be reduced, they should be treated with economical and environmentally sustainable technologies. In this sense, biological treatments such as biotrickling filters become an alternative to physicochemical treatments when emissions with high flow rates and low VOC concentrations are involved, owing to the lack of secondary pollutant production and their lower operational costs. Bioprocesses utilize microbial metabolic reactions to treat contaminated air converting the pollutants to carbon dioxide, water vapour and organic biomass (Devlin et al. 1999). Vapour-phase biotreatments have been also classified as Best Available Technology for the abatement of low VOC concentration waste gas streams in the chemical sector by the European IPPC Bureau (European Commission 2003).

The graphic sector, in which the flexographic industry represents the 17% of the European printing technologies, contributed around 1.7% of the total European turnover in 2003 (Ernst Young & Intergraf 2007). In the flexographic sector, solvent based inks are considered as standard because of their ease of use and the quality of the final product. The use of these inks together with the cleaning operations produce VOC emissions characterized by high air flow rates and low VOC concentrations. For the flexographic sector, the VOC Solvent Emissions Directive (European Commission 1999) establishes an emission limit value (ELV) of 100 mg C Nm⁻³ as the average of all the readings, with none of the hourly averages exceeding this limit by more than a factor of 1.5.

In this study an industrial biotrickling filter has been evaluated to control the VOC emissions from the flexographic facility Papierindustrie Maasmond (The Netherlands) that produces packaging and labels mainly...
for the food industry, and it represents one of the few cases in which this technology has been applied at industrial scale in Europe. This work has been done in parallel with laboratory- and pilot-scale studies. The experiments at the laboratory scale were designed with the objective of transferring both the operational conditions and operational protocols to the industrial scale (Sempere et al. 2008, 2009). The operational protocols developed at the laboratory were also tested at a pilot plant of a biotrickling filter installed in a furniture manufacturing facility (Martínez-Soria et al. 2009). The study presented herein comprises the performance of the unit over a 2-year period, including the start up of installation and the evaluation of the effect of the water flow rate on the performance of the process that allowed the regulatory limits to be met.

METHODS

The biotrickling filter unit (VOCUS™ biotrickling filter system) was supplied by Pure Air Solutions (The Netherlands). Flow scheme and pictures of the unit installed at Papierindustrie Maasmond are shown in Figure 1.

The unit was equipped with a bypass with automatic valves that allowed the air flow rate of the fan that was fed to the VOCUS™ and thus the pollutant load to be controlled. The system was monitored and controlled via modem communication. The biological reactor consisted of a packed column with a volume of 50 m$^3$ of inert packing material (specific surface area >110 m$^2$ m$^{-3}$ and void space > 90%). A nutrient solution containing 19 g N L$^{-1}$ and 4 g P L$^{-1}$ was dosed to the water tank to keep nitrogen concentration in the trickling solution above 10 mg L$^{-1}$. The trickling solution was sprayed and recirculated over the packed bed. The VOC concentration of the inlet and outlet streams of the VOCUS™ were continuously monitored with a total carbon analyser (RS 53-T, Ratfisch Analysensysteme GmbH, Germany) equipped with a flame ionization detector, which was periodically calibrated with propane gas. The system was provided with instrumentation that continuously measured and stored the values of pressure drop over the column, air and water flow rates, temperature, and water level along with the pH and conductivity of the trickling solution. The operational protocols of the system regarding the quality of the recirculation liquid included periodical purges of water, in which a partial volume of the total amount of water was drained. Prior to these purges, samples of water were filtered through a 0.7 μm fibre glass filter, and soluble COD, nitrogen, and phosphorous concentrations of the filtered solution were determined by Hach Lange LCK kits (Hach Company, USA): LCK 514 (COD), LCK 340 (nitrate), LCK 303 (ammonium) LCK 338 (total N), and LCK 348 (phosphate). Activated sludge from the second clarifier of a wastewater treatment plant without previous acclimation was used as the inoculum of the bioreactor.

RESULTS AND DISCUSSION

Characterization of the emission

The production regime of Papierindustrie Maasmond ranges 8 to 12 h from Monday to Friday, therefore, the VOC feeding regime of the VOCUS™ system was subject to these working hours, since during the night and weekend closure periods there was no VOC emission. The characteristics of the emission are shown in Table 1. The emission of the company in terms of air flow rate and VOC concentration was variable during the production hours, associated with the number of printing machines that were running. The company has six printing machines, one of them for the occasional laminating

![Figure 1](http://iwaponline.com/wst/article-pdf/65/1/177/443552/177.pdf)
operations. The VOC composition was also subject to the working of this machine since in its operation only ethyl acetate is used, while the rest of the five printing machines use ethanol (main solvent, >90%), n-propanol, n-propyl acetate, and methoxypropanol. In addition, the laminating operation caused a notable increase in the total VOC concentration of the emission and in the emitted air flow rate.

Table 1 | Characteristics of the emission of Papierindustrie Maasmond

<table>
<thead>
<tr>
<th></th>
<th>Laminating off</th>
<th>Laminating on</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum air flow rate, Nm³ h⁻¹</td>
<td>14,000</td>
<td>20,000</td>
</tr>
<tr>
<td>VOC concentration, mg C Nm⁻³</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Daily average</td>
<td>330–770</td>
<td>615–1,050</td>
</tr>
<tr>
<td>Maximum instantaneous values</td>
<td>1,600</td>
<td>2,500</td>
</tr>
<tr>
<td>VOC mass flow rate, kg C day⁻¹</td>
<td>60–70</td>
<td>95–105</td>
</tr>
<tr>
<td>Compounds</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ethanol</td>
<td>Ethanol</td>
<td></td>
</tr>
<tr>
<td>n-propanol</td>
<td>n-propanol</td>
<td></td>
</tr>
<tr>
<td>n-propyl acetate</td>
<td>n-propyl acetate</td>
<td></td>
</tr>
<tr>
<td>Methoxypropanol</td>
<td>Methoxypropanol</td>
<td></td>
</tr>
</tbody>
</table>

The company is aiming to acquire a solvent-free laminating machine, which implies that the emissions to be treated in the immediate future will correspond to the periods of this work defined as laminating off.

Start-up

The start-up of the VOCUS™ was performed at an EBRT of 60 s and using a continuous trickling of water at 2 m³ h⁻¹, which was changed to an intermittent regime once the inoculum was fixed on the packed bed. The system worked 5 months at these conditions, and data from the last two months showed removal efficiency (RE) values between 50 and 60%. Long start-up periods of bioreactors at industrial scale are typical (Iranpour et al. 2008), since due to the big dimensions of the units it is not viable to perform previous acclimation of the activated sludge to the target pollutants or to use an adapted consortium that shortens the start-up phase, like is done at laboratory scale (Cox & Deshusses 2001). Intermittent and variable emissions also slow down the start-up process. Daily monitoring of the inlet and outlet VOC concentration for typical days during the last months of this start-up stage, together with the pH evolution, are shown in Figures 2 and 3 for operation without and with laminating respectively.

![Figure 2](http://iwaponline.com/wst/article-pdf/65/1/177/443552/177.pdf)

**Figure 2** | Laminating off. (a) Monitoring of inlet and outlet VOC concentration, and (b) pH evolution.

![Figure 3](http://iwaponline.com/wst/article-pdf/65/1/177/443552/177.pdf)

**Figure 3** | Laminating on. (a) Monitoring of inlet and outlet VOC concentration, and (b) pH evolution.
As it can be observed, this phase was characterized by high VOC outlet concentrations, with hourly average values above the 1.5 factor of the ELV. The pH decreased during the VOC feeding period, and this effect might be related to a higher pollutant inlet load (IL) than the pollutant elimination capacity (EC) of the system, remaining intermediate metabolic compounds such as acetic acid that lowered the pH of the trickling water. This same effect has been observed in the treatment of overloaded ethanol biofilters packed with activated carbon, in which the degradation of acetic acid was slower than its formation, causing a pH decrease and affecting the performance of the process (Devinny & Hodge 1995).

Without lamination the decrease of the pH was relatively low, and a recovery of the pH during shutdown periods was observed (Figure 2(b)). However, when the laminating machine was running, pH values under 5 were obtained (Figure 3(b)). This effect can be associated with the presence of ethyl acetate in the emission, as has been observed by Koutinas et al. (2005) in the degradation of ethyl acetate in a biotrickling filter. These authors reported that the amount of NaOH added to control the pH was close to the stoichiometric amount needed to neutralize the acetic acid formation.

**Effect of the water flow rate on the performance of the VOCUS™**

The operation of the VOCUS™ continued with the increase in the water flow rate from 2 to 40 m³ h⁻¹. This decision was adopted taking into account that the results obtained at the laboratory scale showed a better performance when the water flow rate was increased (Sempere et al. 2008). The increase in the water flow rate resulted in a better nutrient distribution, increasing biomass content over the packed column; thus allowing operating the bioreactor at lower EBRTs and treating higher VOC mass flow rates with higher RE values and a better control of the pH. The performance of the VOCUS™ during this period longer than 6 months is shown in Figure 4 in terms of EC versus IL, in which data obtained during operation without and with the laminating machine running have been plotted separately.

As it can be observed in Figure 4(a), without laminating operation a better performance of the VOCUS™ was obtained in each increase in the water flow rate, reaching at 40 m³ h⁻¹ a maximum EC of 122 g C m⁻³ h⁻¹ for an IL of 158 g C m⁻³ h⁻¹, with a RE of 89%. These values are similar to those obtained by Popov et al. (2004) with an industrial unit of biotrickling filter equipped with a pH control system installed at a flexographic facility with a three shift production regime, study in which an EC of 144 g C m⁻³ h⁻¹ for an IL of 161 g C m⁻³ h⁻¹ is reported. Only using water flow rates above 8 m³ h⁻¹ the VOCUS™ could handle ILs up to 50 g C m⁻³ h⁻¹ with high REs, but it was necessary a value of 40 m³ h⁻¹ to obtain RE values higher than 90% for the ILs needed to be treated at Papierindustrie Maasmond, that is up to 90 g C m⁻³ h⁻¹.

When the laminating machine was running (Figure 4(b)) a wide variability of results were obtained, which were attributed to the variation of the pH of the trickling liquid caused by the presence of ethyl acetate in the emission. These results suggest that if the laminating machine ran more frequently, a chemical pH control would allow a more stable performance to be obtained. Regarding the influence of the water flow rate in the performance of the VOCUS™, ILs up to 90 g C m⁻³ h⁻¹ were treated with water flow rates of 8, 20 and 40 m³ h⁻¹. It was observed that water flow rates greater than 20 m³ h⁻¹ allowed a better recovery of the pH during the shutdown periods, so higher ILs were treated in comparison with using water flow rates up to 8 m³ h⁻¹, achieving a maximum EC of approximately 97 g C m⁻³ h⁻¹ for an IL of 133 g C m⁻³ h⁻¹.
Table 2  Long-term operational and performance parameters of the VOCUS™ using water flow rate at 40 m$^3$ h$^{-1}$ and operating without lamination

<table>
<thead>
<tr>
<th>Operational period</th>
<th>12 months</th>
<th>Summer EBRT &lt; 30 s</th>
<th>Winter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inlet air temperature, °C</td>
<td>22.4 ± 3.6</td>
<td>26.8 ± 1.4</td>
<td>18.7 ± 1.0</td>
</tr>
<tr>
<td>Air flow rate, Nm$^3$ h$^{-1}$</td>
<td>5,557 ± 1,601</td>
<td>6,947 ± 378</td>
<td>5,024 ± 588</td>
</tr>
<tr>
<td>EBRT, s</td>
<td>32 ± 7</td>
<td>26 ± 1</td>
<td>36 ± 9</td>
</tr>
<tr>
<td>Inlet VOC concentration, mg C Nm$^{-3}$</td>
<td>484 ± 179</td>
<td>561 ± 182</td>
<td>499 ± 175</td>
</tr>
<tr>
<td>Outlet VOC concentration, mg C Nm$^{-3}$</td>
<td>53 ± 31</td>
<td>54 ± 28</td>
<td>64 ± 31</td>
</tr>
<tr>
<td>VOC mass flow rate, kg C day$^{-1}$</td>
<td>23 ± 10</td>
<td>30 ± 12</td>
<td>22 ± 7</td>
</tr>
<tr>
<td>IL, g C m$^{-3}$ h$^{-1}$</td>
<td>56 ± 15</td>
<td>78 ± 17</td>
<td>50 ± 10</td>
</tr>
<tr>
<td>EC, g C m$^{-3}$ h$^{-1}$</td>
<td>50 ± 11</td>
<td>71 ± 13</td>
<td>44 ± 9</td>
</tr>
<tr>
<td>RE, %</td>
<td>90 ± 6</td>
<td>91 ± 5</td>
<td>87 ± 7</td>
</tr>
</tbody>
</table>

**Long-term performance of the VOCUS™ at a water flow rate of 40 m$^3$ h$^{-1}$**

After adjusting the water flow rate the system worked over a whole year. The robustness of the system to nonworking periods associated with long time stops of production (3 weeks of production closure) without volatile organic compound emission was demonstrated (data not shown here), as was tested in the experiments performed at laboratory scale (Sempere et al. 2008). Several studies also showed that bioreactors are able to handle transient conditions and starvation periods from hours to several days of duration (Cox & Deshusses 2002; Álvarez-Hornos et al. 2007). Martin & Loehr (1996) indicated that the reacclimation time of a toluene degrading biofilter depended on the duration of starvation and the presence of alternative carbon source. In this sense, Zhang & Bishop (2005) observed in a liquid phase bioreactor that biofilm extracellular polymeric substances coming from the natural secretions of bacteria, cell lysis and hydrolysis products can be used as a substrate by their own producers and by other microorganism when they are starved.

In Table 2 the average operational and performance parameters during this long-term operation are shown under normal emission condition, which is without lamination, in which data corresponding to summer and winter are presented separately.

It can be observed how the higher temperatures of summer allowed the setting of a residence time as low as 26 s to comply with the regulatory limits, obtaining average outlet concentrations of 53 mg C Nm$^{-3}$. In winter, an EBRT of 36 s was required to obtain similar RE values. Soluble COD measurements of the trickling solution showed that the VOCs removed in the partial purge of water were not significant compared to the pollutant fed during the period between two purges. This indicates that the VOCs were biologically removed.

These results demonstrate the technical and economical feasibility of the technology applied to the flexographic sector in comparison with conventional technologies such as RTO (regenerative thermal oxidizer).

**CONCLUSIONS**

This work shows the first study of a long-term performance of an industrial biotrickling filter to treat the VOC emission of a flexographic facility, demonstrating that the VOCUS™ biotrickling filter system is a sustainable and robust technology to control this emission. The start-up of the unit was lower than 3 months using activated sludge without previous acclimation to the VOC mixture to be degraded; this start-up period is typical of industrial bioreactors. The water flow rate was found to be a key parameter in the performance of the process; with an optimal value of 40 m$^3$ h$^{-1}$ to reach average outlet VOC concentration below 65 mg C Nm$^{-3}$. During the summer period, daily average inlet air temperatures of 26.8 °C allowed the EBRT to be decreased from 36 to 26 s, which means increasing the treated VOC mass flow rate and maintaining similar outlet VOC concentrations.

**ACKNOWLEDGEMENTS**

This research was supported by the Ministerio de Educación y Ciencia (Spain, research project CTM 2010-15031 with FEDER funds). Feliu Sempere thanks the Universitat de València for the award of his V Segles Empresa fellowship. The authors would like to give special thanks to Hans Oonk.
(Pure Air Solutions) and Bert Verweel (Papierindustrie Maasmond) for their work and collaboration.

REFERENCES


Devinny, J. S., Deshusses, M. A. & Webster, T. S. 1999 *Biofiltration for Air Pollution Control*. CRC-Lewis Publishers, Boca Raton, USA.


First received 25 May 2011; accepted in revised form 25 August 2011.