

Practical experience with an industrial biofilter treating solvent vapour loads of varying magnitude and composition

Heather J. Campbell^a and Michael A. Connor^b

^a Containers Packaging, 240 Geelong Road, West Footscray, Vic., 3012, Australia

^b Chemical Engineering Department, University of Melbourne, Parkville, Vic., 3052, Australia

Abstract: A biofilter was installed to reduce levels of solvent vapours in air exhausted from a rotogravure printing press. This air was first humidified and then passed through two filter beds in series which contained a mixture of mushroom compost, activated carbon, lime and polystyrene beads. The printing press employed different inks for which the solvents included ethanol, ethyl acetate, n-propyl acetate, acetone, methyl ethyl ketone and toluene. The amount and composition of the solvent vapours in the exhaust air depended on the printing tasks undertaken. Breaks in the supply of solvent to the filter occurred during job changeovers and press shut down. The performance of the biofilter under such varying operating conditions was studied. Maintaining moisture in the bed proved critical. Overall solvent removal efficiency was affected by the nature of the solvent mixture, the relative solubility of individual solvents and their relative biodegradabilities. No significant lag period was evident in biofilter performance when sudden changes in solvent vapour composition occurred. It was inferred that the activated carbon's adsorptive properties were at least partly responsible for this. Inferences drawn from observations during plant operation were used to gain insights into the relative importance of various physical, chemical and biological processes.

Keywords: biofilter, industrial, printing, rotogravure, solvent

INTRODUCTION

A major problem in the area of air pollution control is finding economic ways of treating gas flows containing low levels of pollutants. Where such pollutants are readily biodegradable, there is steadily growing interest in removing these pollutants with biofilters. In many such situations, biofilters have substantial cost advantages over competing processes. The magnitude of such advantages will vary from site to site but representative figures for a number of applications are available. For example, biofilters employed for control of solvent vapours have a capital cost of between 25% and 50% of that of competing processes such as incineration or adsorption on activated carbon. In addition, their operating costs are also low, in the range of 10 to 25% of those for the abovementioned processes (ref.1).

At the heart of a biofilter is a bed of pre-prepared support materials in which resides a mixed consortium of micro-organisms. The support materials usually include an organic material such as compost, which provides the nutrient requirements of the micro-organisms. The bed is kept at a moisture content of 40 to 60% (ref.2) to maintain an environment that is moist enough to meet the requirements of the micro-organisms and yet not wet enough to lead to the development of anaerobic conditions. Such conditions develop when enough moisture is present to fill the smaller pores within the bed, thereby creating regions to which gaseous oxygen cannot readily penetrate.

The pollutant-containing gases are passed through the above bed. During their passage the pollutants diffuse to the boundary between the gaseous phase and the aqueous film around the bed support materials. There they transfer to the aqueous phase and become available to the micro-organisms within. Microbial degradation of the pollutants ensues, producing carbon dioxide and water, new cells, and liberating heat. Gas velocities through the bed, and residence times within it, need to be such as to give the required level of pollutant removal. A typical biofilter has the structure shown schematically in Fig. 1 (ref.2).

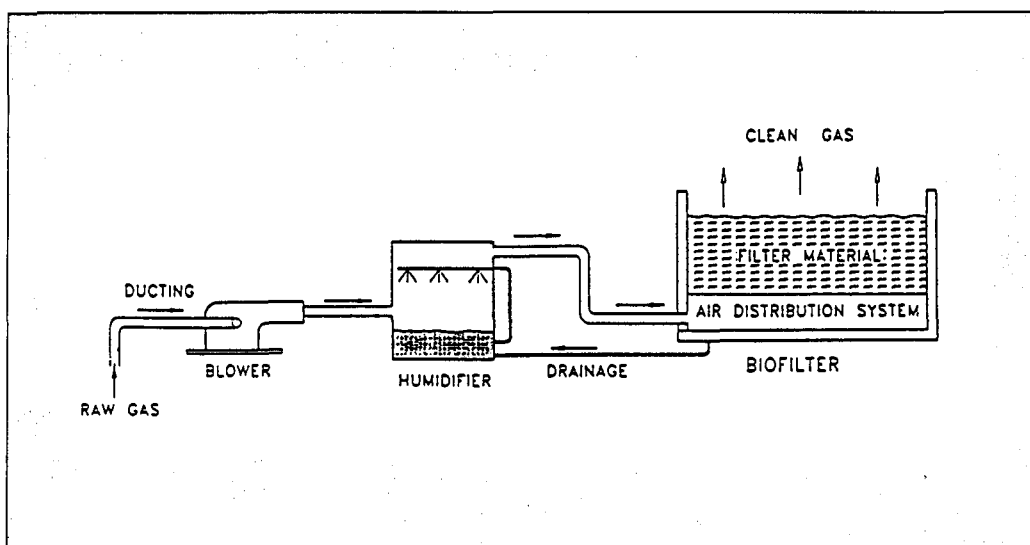


Fig.1 A typical biofilter (after Leson and Winer [1991])

The conoflex packaging plant

A biofilter similar in overall concept to that shown in Fig.1 was installed in 1992 at the Conoflex Packaging printing plant in Preston, Victoria. It was supplied by Clean Air TechniQ, and developed in the Netherlands (refs.3,4 & 5). Its purpose was to reduce levels of solvent vapours in the exhaust air from a rotogravure printing press. Gravure printing uses an engraved rotating cylinder to transfer ink to the web being printed. This web is a film of paper, polyester, polypropylene, aluminium foil, metallised polyester, metallised polypropylene, polystyrene, or combinations thereof. Once the ink has been applied, the film passes through an oven where the solvent is driven off by air heated to 80°C. The film then travels through to the next printing unit where the next colour is laid down. At Conoflex Packaging there are eight colour presses allowing up to eight colours or coatings to be printed. The exhaust air from the ovens of one of the printing presses (known as PF12) plus that from the associated floor sweep exhaust system, which captures solvent vapours from the working environment, are conveyed together to the biofiltration unit.

A wide range of solvents is used in the printing process at the Conoflex Packaging Plant. Most of the ink systems use a combination of at least two solvents. Solvents in regular use are ethanol, ethyl acetate, methyl ethyl ketone, n-propyl acetate, toluene, icinol and acetone. The two principal solvents used are ethanol and ethyl acetate. The majority of jobs printed on PF12 consist of complex designs utilising a number of solvents. The 30000 m³hr⁻¹ of exhaust gas from this machine can contain from 5ppm to 1200ppm of solvents depending on the printing task in hand and the stage reached in the printing process. For example, during the changeover period between printing jobs, very little solvent vapour is carried through to the biofilter. Breaks in the supply of solvent to the micro-organisms in the biofilter also occur when the press is shut down at weekends, and over public holidays.

Biofilter design and development

The first stage in the design and development of a biofilter for the Preston plant involved a small biofiltration pilot plant being connected to one of the exhaust stacks from the printing press PF12 and its performance observed for three months. Results from this trial indicated that a biofiltration plant could be used successfully to treat the solvent laden air emissions from PF12. The results obtained also helped to determine the design of the full size Bioton biofiltration unit installed later.

The VOC (volatile organic carbon) or solvent loading rate assumed in designing the full size Bioton unit was 35 kg/hr, of which it was expected that 80% i.e. 28 kg/hr, would be removed. This 80% removal level was determined by the requirements of the Environment Protection Authority of Victoria (EPAV). Gas was

assumed to be exhausted from around press PF12 at $35000 \text{ m}^3\text{hr}^{-1}$; this gas was expected to be at a temperature of 10 to 40°C and a relative humidity of 10-20%. The concentration of particulates in the exhaust gas was assumed to be less than 10 mgm^{-3} . For design purposes, solvents were assigned to one of two classes. Class 1 solvents were regarded as being more easily biodegradable; these solvents and their maximum expected concentrations (mgm^{-3}) in the exhaust gas were: ethanol (700), ethyl acetate(400), n-propyl acetate (200), acetone (50) and icinol (50).

The maximum expected total concentration of Class 1 solvents in the exhaust gas at any one time was 1000 mg/m^3 . The two Class 2 solvents were methyl ethyl ketone (MEK) and toluene; these had maximum expected individual concentrations of 100 mgm^{-3} and 50 mgm^{-3} respectively. The maximum total concentration of Class 2 solvents expected in the exhaust gas at any one time was 100 mgm^{-3} .

The concept of biodegradability used above in allocating solvents to Class 1 or Class 2 reflects the ease with which solvents are first absorbed into an aqueous layer and then degraded by the microbial community present. For example, acetone and methyl ethyl ketone are regarded as having comparable degradation rates in the environment (ref.6). However, acetone's greater solubility in water means that its apparent degradability in biofilters is superior to that of methyl ethyl ketone, hence its assignment to Class 1 and methyl ethyl ketone to Class 2.

The Bioton biofilter installation, as designed, consisted of a packed tower known as the humidifier and used to humidify the exhaust air from PF12 (Fig.2). This air is forced up the tower through a packing of polyethylene rings against a descending stream of water. A recirculation pump is used to recirculate water through the packed tower. The biofilter unit is a $10\text{m} \times 17.5\text{m} \times 5\text{m}$ insulated box containing two one metre deep beds of "biomass media", one above the other. The humidified air is drawn down through the two layers of biomass media, where solvent vapours are removed. The beds consist of a mixture of mushroom compost, activated carbon, lime and polystyrene beads. Immobilised on these materials are bacteria which are able to utilise the solvents both as a source of energy and as a source of organic carbon for cell synthesis. Load cells are placed under the beds to measure the mass of the beds and hence determine their moisture content. Overhead sprays can be activated to add water to the biomass media when the moisture content drops below a level determined by an algorithm in the computer control system. Pressure transducers located at each level within the biofilter are also used to help determine the "condition" of the filter bed. The exhaust stack consists of an induced draught fan on the stack to draw the air through the packed tower and biofilter. It then exhausts

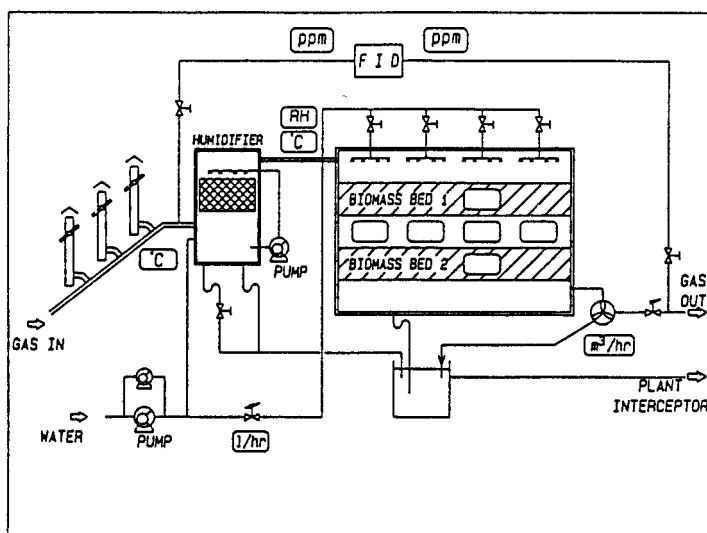


Fig.2 The biofilter installation at the Conoflex Packaging Plant

the treated air through a seven metre high stack into the atmosphere (Fig.2).

The Bioton unit is heavily instrumented. This instrumentation is necessary to protect the bacteria from conditions which would adversely affect their performance, as well as to give an on-going indication of the efficiency of the process. The bacteria are protected by a series of shutdown procedures linked to changes in temperature, pressure drop, and humidity. To obtain the required information to actuate shutdown procedures, temperature probes, humidity probes, and pressure transducers are used. In addition two flowmeters are installed, one indicates whether the internal water sprays are operating while the other indicates the flow rate of air through the exhaust stack. The efficiency of the process is calculated from gas chromatographic measurements using a flame ionisation detector (FID) to determine VOC (volatile organic compounds) concentrations in the inlet and outlet gas.

Factors Affecting the Efficiency of the Biofilter

Various factors that can affect the removal efficiency of a biofilter are discussed below as they relate to the installation at Preston.

Physical and chemical structure of the filter bed

As indicated earlier, the filter bed is made up of a combination of mushroom compost, activated carbon, lime, polystyrene beads and bacteria. The mushroom compost constitutes the bulk of the bed material. It provides a porous structure through which air can flow easily. It is also the main source of the nutrients, such as nitrogen, phosphorus and potassium, that are required by the bacteria. During shutdown periods this material can also act as a source of carbon for the bacteria.

The role of the polystyrene beads is to assist in maintaining aeration and gas flow through the beds. Without them the compost would compact over time, reducing the fraction of pore space within the bed, and hence the ease with which air can move through the bed. This would result in an increased pressure drop across the bed, a consequent increase in the energy required to force the air through the bed, and a greater likelihood of channelling.

Activated carbon is added to the filter bed to enable a greater fraction of the solvent vapours entering the bed during peak pollutant loads to be captured. This captured material subsequently desorbs from the activated carbon, for example over weekends or during other periods when solvent loads are low. Much of it is then degraded by the bacteria present. The overall effect is for average solvent removal efficiencies to be increased.

The filter beds were inoculated with specific bacteria and an activated sludge mixture. As most bacteria have an optimum pH range of 6 to 8, lime was included in the biomass to act as a buffering substance.

Oxygen availability to the bacteria

Aerobic bacteria oxidise the solvents to CO₂ and H₂O. Thus it is extremely important that the bed does not become anaerobic which can easily occur if the bed becomes too wet or if excessive compaction occurs. The formation of cracks within the bed also reduces efficiency as the cracks provide a path of least resistance for the air passing through the bed. This deprives bacteria of oxygen and reduces the solvent capture.

Moisture content of the filter media

The moisture content of both the biomass and the polluted gas stream are extremely important. The bacteria live in an aqueous layer around the surfaces of solid material in the biofilter bed. They can only utilise the solvent when it is in this water phase.

The presence of adequate moisture is therefore essential for the combined survival and effectiveness of the bacteria. This moisture can be lost in two ways. The first occurs if the gas entering the filter bed is not saturated; under such conditions, given the large interfacial area between the air and the aqueous layer on the bed support material, considerable loss of moisture by evaporation can occur. It is for this reason that the

Conoflex biofilter is designed to provide gas to the filter beds at a relative humidity of close to 100%. The second cause of moisture loss is related to the exothermic nature of the microbially mediated solvent oxidation processes. If not removed, this heat could raise the temperature of the bed to unacceptably high levels. In practice, however, bed temperatures are maintained through evaporation of moisture and the associated loss of excess heat as latent heat of vaporisation. To replace the moisture lost in this way extra water needs to be added periodically to the biomass beds. The amount added is controlled by a computer which utilises measured data on changes in the weights of the beds and in the pressure drop across these beds to determine when extra water is required. The computer is programmed so as to maintain the dry material percentage of the bed at about 55 percent. The importance of regulating the moisture content of the beds is well illustrated by work done by ClairTech in Holland which showed that a dry material percentage variation of as little as 10% can make a substantial difference to the solvent removal efficiency of the filter beds. It was determined that an average dry material percentage of 40% gives higher removal efficiencies than other values of the dry material percentage. However removal efficiencies can decrease rapidly as dry material percentages drop below 40%, i.e. as the bed moisture content increases above 60%. Since the Bioton unit controls bed moisture content by adding water, it is safer to operate at a dry material percentage of 55% rather than risk over moistening the bed.

The moisture content of the bed is also very important as far as the physical characteristics of the bed are concerned, and these in turn affect air flow rates. If the bed becomes saturated the pressure drop across the bed increases and it becomes more difficult to draw solvent laden air through the bed. Excessive water may also lead to sludge/slime formation as bacteria will reproduce profusely given enough water and nutrients. This slime tends to build up on the biomass in the bed, and in the humidifier, the effluent gas demister and on the concrete flooring of the biofilter building. If the biomass becomes too dry it will develop cracks and start to crumble. In severe cases it forms a fine dust. At this point the biofilter needs to be shut down and the biomass reconditioned. This involves physically turning over material and wetting it. For a biofilter the size of Conoflex's this can involve four people for up to a month, as was learned through bitter experience.

Temperature of the filter bed

The temperature of the biomass, and thus the temperature at which the bacteria operate, is important to the performance of the biofilter. The bacteria utilised at Conoflex are mesophiles, having an effective operating temperature of between 10°C and 40°C. The temperatures at which the bacteria can most effectively utilise the solvents (VOC) are 32°C and 37°C. The biodegradation rate approximately halves for every 10°C drop in temperature from the optimum of approximately 37°C (ref.7). In practice, the operating temperatures of the Conoflex biofilter (measured in the air layer above the biomass) were from 20°C to 26°C but can drop to just above 10°C overnight after the printing press (PF12) has been shut down. From a temperature viewpoint, therefore, the system is operating at less than maximum efficiency.

VOC (solvent) inlet load rates

Both the amount and type of VOC material fed to a biofilter over a given period have an effect on the efficiency of the biofilter, as calculated using FID measurements. It is now well established that the elimination capacity of a biofilter, expressed in terms of grams removed per hour per unit volume of bed, is dependent on the concentration of the target substance in the air being treated (ref.3). Above a limiting concentration, specific to the substance involved, the elimination capacity is independent of the gas phase concentration; in this case the rate of removal is reaction limited. Below this limiting concentration, removal rates are diffusion limited and elimination capacities are proportional to the gas phase concentration. The limiting concentration for a given substance in a particular biofilter bed is determined by its solubility in water and its biodegradability. Typical critical concentrations of ethyl acetate and toluene, two of the solvents used at the Conoflex plant, are 200 mg m⁻³ and 1500 mg m⁻³ respectively (ref.3). It can be inferred from the work of Liu *et al.* (ref.8) on the removal of toluene vapours using activated carbon beds that toluene's high critical concentration is linked to its low solubility in water. Once available to bacteria, toluene can be readily broken down, as researchers such as Shareefdeen and Baltzis (ref.9) have shown.

AIMS OF THE STUDY

The performance of the biofilter was studied under the varying conditions imposed on it as a result of the operating practices at the plant. The specific aims of the study were firstly, to investigate the general operation of a biofilter in the gravure printing environment and to assess its operational difficulties. Although pilot plant trials indicated that a biofilter could reduce solvent emissions to required levels it was important that the operating parameters of a full scale unit were fully understood. Secondly, to determine as far as practical constraints permitted, the effect on biofilter solvent removal efficiency of inlet and bed temperatures, bed moisture content, the composition of the solvent mixture in the exhaust gas, frequency of changes in solvent mixture composition, solvent loadings and lengthy shutdowns.

The underlying reasons for studying the first three of the factors listed above were discussed earlier. The fourth factor was investigated to establish whether there was any noticeable correlation, adverse or favourable, between changes in the biofilter's solvent removal efficiency and the frequency with which the composition of the solvent mixture was changed.

As regards the fifth factor, solvent load, the Conoflex plant was designed to treat 35 kg h⁻¹ of solvent, but instantaneous solvent feed rates vary widely around this design level. In view of EPAV requirements, an important objective of the investigation was to determine how such variations affected the biofilter performance. As discussed earlier, previous work has shown that in a standard biofilter the relationship of biofilter performance and loading rate may be diffusion or reaction limited (ref.2). However the exact form of the relationship depends on the properties of the bed and the solvents involved. The presence of activated carbon in the Conoflex biofilter bed would also be expected to have an effect particularly in the case of toluene where 40 to 80 fold improvements in toluene removal rates when using an activated carbon biofilter in place of the normal compost-based beds have been reported (ref.8).

The sixth of the above factors was of interest as for most of 1993 and 1994 printing press PF12 was only operated 20 hours per day, five and a half days per week. In addition the entire plant was closed from 2 to 3 weeks immediately after Christmas. Although one of the advantages often put forward for biofiltration is the ease with which normal operations can be resumed after short to medium length stoppages, there was interest in establishing that this was the case for the Conoflex plant.

DATA COLLECTION

The Conoflex biofilter is controlled by a PLC with inputs from a number of sensors. The readings from the temperature probes, F.I.D., exhaust air flow meter and water flow meter were not used to control the process. Their purpose was to indicate what was happening on the plant. The pressure readings and load cell weights, however, did have a control function, being used to control the moisture content of the biological beds. If the moisture content was too low then the water sprays above the biomass beds were automatically activated. The PLC computer control system is designed to control the moisture percentage of the bed to 55% ± 10%, the preferred range for biofilter operation (ref.2). The Flame Ionisation Detector (F.I.D.) measured the total organic carbon (TOC or solvent) content in the inlet gas stream entering the biofilter and in the gas stream leaving the biofilter through the exhaust stack. The F.I.D. unit was switched every minute to measure the inlet and outlet gas concentration.

All data from the measuring instruments were stored by the PC, with the records being updated continuously. Because of the sheer volume of data, sequences of values spaced ten or twenty minutes apart were used. While this frequency was believed to be adequate to characterise most changes in biofilter performance, it is possible that certain short-term responses were obscured. The practice followed in obtaining data for analysis was as follows. For an "event" of potential interest, values of inlet gas temperature, bed temperature, and inlet and outlet F.I.D. readings were transcribed onto a record sheet together with the date, time, and details of the corresponding job(s) on the press. From this data the ink system(s) in use was determined and this information with the calculated removal efficiencies were added to the record sheet.

To draw conclusions from measurements obtained during specific "events" (such as a change from one printing

“job” to the next) certain assumptions had to be made. The most important of these were that the gas humidity and flow rate did not change over the period in question and neither did the dry matter percentage of the filter bed.

In practice humidity readings remained constant but readings of air flow and dry matter percentage varied. As the main fan was not driven by a variable speed motor, the air flow through the Bioton should have been reasonably constant. The observed variations in air flow rate were unexpected. A partial explanation for this was provided when it was later established that the pilot tube used to measure air flow rate was very close to the top of the exhaust stack and was affected by alterations in external wind speed. The dry matter percentage generally remained close to the set point of 55%, except in some extreme cases which are discussed later.

To supplement and verify the results obtained using FID readings, samples were also taken from the inlet and outlet gas streams periodically and analysed on a Perkin-Elmer Sigma 2000 gas chromatograph (GC) to provide information on the differing extents to which individual solvents were removed by the biofilter. The GC as set up for the company's quality control checks allowed the percentage of each solvent in the gas mixtures to be determined rather than the total mass of solvent present. Nevertheless, where conditions were such as to permit comparison of FID and GC measurements, the results were found to be reasonably consistent.

DISCUSSION OF OBSERVATIONS

General Operation of the Biofilter

The biofilter at the Conoflex plant was beset by mechanical problems during its first year of operation. These were principally a consequence of inadequate maintenance and they affected the efficiency of the biofilter considerably. The mechanical problems that occurred fell mainly into three categories: instrument malfunctions or breakdowns; water spray system problems; structural problems.

The instrument malfunctions and breakdowns consisted mainly of problems with one load cell and the Flame Ionisation Detection unit. The inaccurate readings from one load cell meant that the moisture content of approximately one quarter of the top bed was difficult to control automatically. This section of the bed was often too wet or too dry, resulting in the biomass beds not operating to full capacity. Repairs to the FID unit meant that inlet and outlet solvent concentrations for the biofilter could not be obtained for periods of up to two months at a time. This severely restricted the usefulness of data collected from the biofilter over these periods.

The water spray system controlled the application of the additional water to the biomass beds that was required to maintain the beds' moisture content. There were two main problems with this system: the pressure pump that supplied the water to the spray heads, and the spray lines (plastic tubing and spray nozzles) themselves. When it was discovered (January 1994) that the pressure pump had not been working properly and that the beds had dried out, each had to be completely re-moistened in sections using hoses. They were dug out, re-moistened and put back into place. This procedure meant that the biofilter was not operational again until March 1994. An ongoing problem with the spray system was the blocking of the nozzles. In-line filters were installed to reduce the chance of the nozzles becoming blocked, but they still needed to be checked every few months.

There have been several major structural problems with the biofilter which have resulted in it being out of operation for periods of up to two months. The corrosion of the main fan on the biofilter caused the longest shutdown period as it required two months to build and install a completely new fan.

All of the abovementioned problems with operating the biofilter were mechanical. Some necessitated the shutting down of the biofilter while those related to the water spray system merely reduced the biofilter's effectiveness. In all cases the result was that data of potential interest were unavailable or unrepresentative of normal operating conditions.

Effect of Inlet and Bed Temperatures on Solvent Removal Efficiency

The temperature of the gas stream from the printing process before it enters the humidifier was always below 55°C and the temperature ahead of the humidifier was as low as the external ambient temperature when the gravure printing press was not operating. During 1993 and the first six months of 1994 the gravure printing press only operated twenty hours a day for 5½ days a week. This meant that the temperature of the gas stream into the humidifier could drop to below 5°C overnight. Since late 1994 the gravure printing press has operated twenty-four hours a day, 5½ days a week. This has reduced the effect of temperature fluctuations upon the biofilter.

As described earlier, the air from the printing process work area passed through a packed bed humidifier before reaching the two biomass beds. Unfortunately the humidifier acts as a very efficient 'cooling tower'. As a consequence, the temperature of the air in the biofilter never exceeded 26°C and the operating temperature range for the Conoflex biofilter was normally 19°C to 26°C when the printing press was operating. According to available information for biofilters, rates of biodegradation might be expected to almost double over this temperature range. In an attempt to determine whether there existed any relationship between temperature and removal efficiency, various data relationships were explored. Figure 3 is an example of data examination. In preparing this plot, measurements were grouped into 3 series, depending on the biofilter temperature at the time.

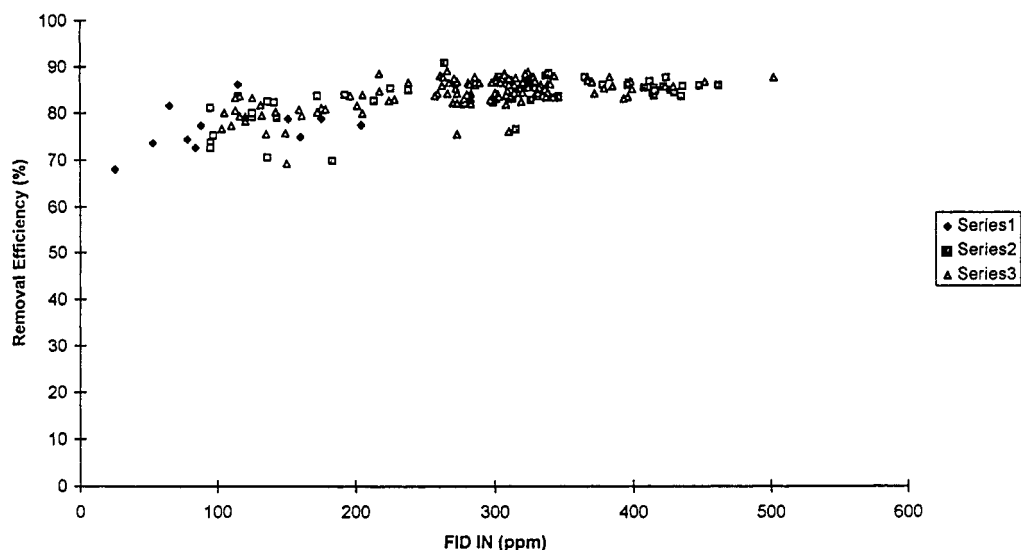


Fig.3 Solvent removal efficiency versus inlet FID reading for Coffee Bag jobs (23-3-94 to 29-3-94).

Series 1 represents data for biofilter temperatures of 19°C to 21°C.

Series 2 represents data for biofilter temperatures of 21.1°C to 23°C.

Series 3 represents data for biofilter temperatures of 21.3°C to 25°C.

In Fig. 3, as in all other cases, the scatter of results was such that no conclusive evidence existed of the dependence of removal efficiency on temperature. This would appear to conflict with the inference drawn from previous research that for a temperature rise of 19°C to 26°C the biodegradation rate would nearly double. A likely explanation, given the short periods over which the observations were made, was that the frequency of temperature fluctuations in the bed was too rapid for any significant temperature-related changes in microbial activity to occur.

Another difficulty that rapidly became apparent was that, as indicated earlier, the temperatures displayed as "Biofilter Temperature" were measured in the air space directly above the top biomass bed. These temperatures represent the air temperature not the actual biomass bed temperature, which would be expected to be significantly higher as a result of heat released during solvent oxidation.

The effect of fluctuations in air temperature upon the bacteria in the biofilter would be expected to be heavily

damped because of the smallness of typical gas/solid heat transfer coefficients. An idea of the slowness of the response of bed temperature to changes in air temperature can be gained from the fact that it takes around 12 hours after the printing press is stopped for bed and air temperatures to equilibrate. An inlet gas hotter than the biomass beds will still transfer some heat to the beds but changes in the biomass bed temperature and thus the bacteria operating temperature would appear to be more closely related to the rate of heat release associated with solvent oxidation. Most of this heat is used to evaporate moisture, which leaves in the gases exiting the biofilter bed. A bed temperature greater than the air temperature is needed to achieve this; the magnitude of the difference between the two temperatures is largely determined by the rate of moisture evaporation necessary to keep the beds in thermal equilibrium.

Effect of the Moisture Content of the Beds upon the Solvent Removal Efficiency

The moisture content of the bed affects the biofilm layer in the biomass beds substantially. If the beds are too wet or too dry the bacteria cannot fully utilise the solvent vapours. As noted earlier, a dry material percentage variation of 10% around an average value of 55% markedly influences the solvent removal efficiency of the biomass.

Figure 4 illustrates variations in moisture content over a 12 month operation period. Moisture content was determined by taking samples of biomass from four different sections of each bed. The samples were then weighed, dried at 105°C and reweighed to determine the moisture content. As can be seen from this graph, the measured moisture contents varied widely, with a substantial number of readings being either well above or well below the desired operating range (i.e. 55 ± 10%). The moisture contents appear to have stabilised in November and December. However, during this period the biofilter was not operating as the fan was being rebuilt. Therefore, the sprays were turned on for two hours per day during this period to stabilise the moisture content of the biomass beds. The results obtained before November show wide variations in moisture contents, almost certainly linked to evaporative losses like those described in the previous section. It is highly likely that a similarly wide range of moisture contents would have occurred in 1993. Trying to determine any direct correlation between moisture content and solvent removal efficiency for the Conoflex biofilter was therefore counter-productive.

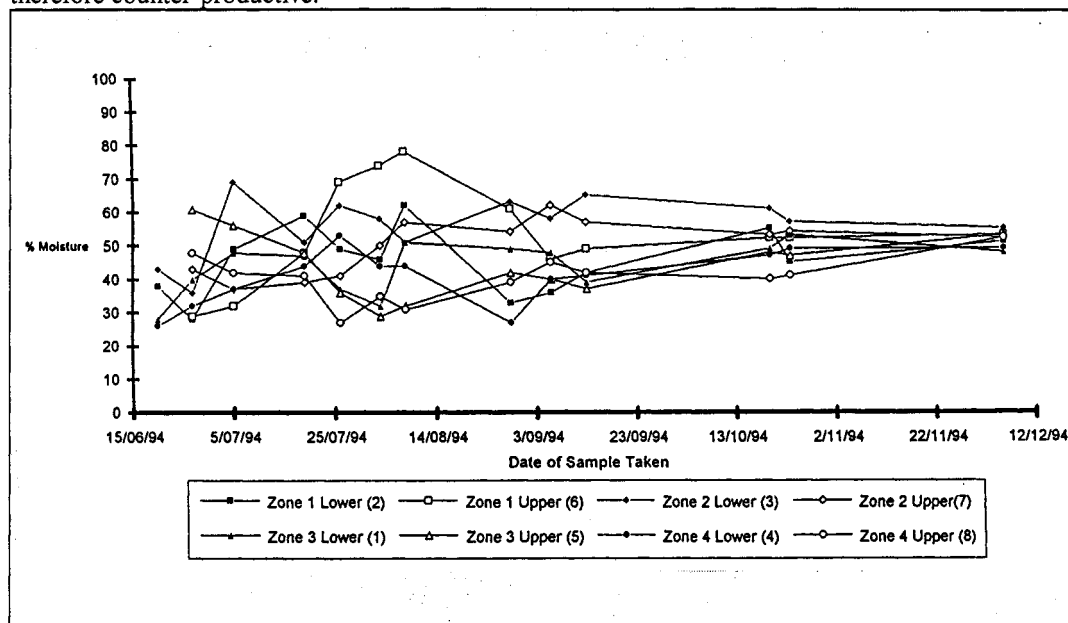


Fig.4 Variations of moisture content in various sections of the Bioton beds

While there was insufficient information to draw correlations between bed moisture content and solvent removal efficiency, evidence was available to show the effect of the bed being excessively dry. Sections of the bed dried out during December 1993 and January 1994. This was only discovered (mid-January 1994) when samples for the inlet gas and outlet gas from the biofilter were analysed and indicated solvent removal efficiencies of 36.4% and 34.9% respectively for two sets of samples taken at this time. From past experience

one would have expected results no lower than 50% removal efficiency. On inspection the biomass beds were found to be extremely dry thus necessitating their reconditioning (see above). It is therefore very clear that if the beds are allowed to become too dry, the solvent removal efficiency is greatly reduced. Our experience indicates that the humidifier alone was insufficient to keep the beds moist. The high solvent loads to which the biofilter was subjected and the significant amounts of heat liberated in the exothermic solvent oxidation reactions increased the drying effect. Gas temperature increases of up to 7°C across the beds have been recorded, increases that would permit a substantial uptake of water from the beds even if the incoming gas were saturated. To combat this additional moisture loss operating procedures were amended to ensure the spray nozzles above the beds were turned on automatically for at least two hours per day. The water supplied in this way helped to supplement moisture added in the humidifier and during automatic operation of the water sprays.

Effect of the Nature of the Solvent Mixture upon the Solvent Removal Efficiency

Conoflex Packaging uses many inks, each of which contain a different solvent mixture. Consequently, whenever there is a change from one print job to the next, there is a corresponding change in solvent load and composition.

The effect of such changes on solvent removal efficiency is illustrated in Fig.5. Here Job 1 employed inks with a solvent base consisting of (approximately) 13.5% ethyl acetate, 40.5% ethanol, 38.8% methyl ethyl ketone and 7.2% minor components. Job 2 employed different inks, with the main solvent base being methyl ethyl ketone (43.5%) and toluene (17.9%). The solvent mixture used in Job 1 contains a large proportion of the highly soluble, readily biodegraded solvent, ethanol, whereas the Job 2 mixture was dominated by Class 2 solvents, of which toluene is particularly hard to capture and degrade. It was therefore not surprising that a decrease in removal efficiency occurred (Fig.5).

After Job 2, the next job (Job 3) used an ink with a solvent base consisting of (approximately) 49% ethanol, 20% ethyl acetate, 26% methyl ethyl ketone and 5% minor constituents. If anything, this solvent mixture would be expected to be more easily removed and degraded than that used in Job 1. The rise in solvent removal efficiency at the end of Job 2 was therefore not unexpected.

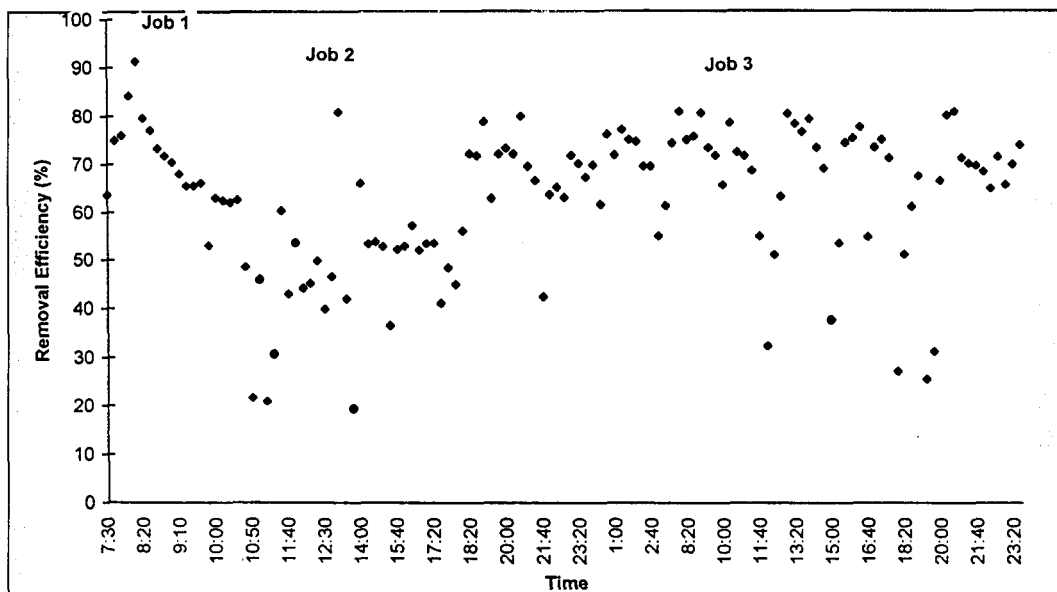


Figure 5: Solvent Removal Efficiency from 8-11-1993 to 9-11-1993

Whilst it is not possible to determine from the results (Fig.5) whether removal of specific solvents was reaction limited or diffusion limited, some indications of what was likely to be occurring can be deduced. Ottengraf (ref.4) found that the elimination capacity of a biomass bed was very similar for ethyl acetate and toluene. Toluene removal however was diffusion limited until the toluene concentration in the inlet gas exceeded 1500 gm⁻³. For ethyl acetate, the corresponding limit was much lower, being 200 gm⁻³. These limits would be

expected to vary somewhat from biofilter to biofilter. However, given that the total solvent load on the Bioton Unit was usually of the order of 1500 gm^{-3} , it can be deduced that toluene removal is likely always to be diffusion limited while ethyl acetate removal is likely to be reaction limited in many situations. As discussed earlier, a key factor contributing to the high gas phase solvent concentration at which toluene removal becomes reaction limited is its low solubility in water. This makes toluene less able to be absorbed into the water film around the biomass compared to other solvents such as methyl ethyl ketone and acetone. Diffusion into the water layer is governed by the solvent driving force (gas phase concentration) and Henry's law constants. The Henry's law constant for toluene is 0.26 while for ethanol and acetone the Henry's law constants are 0.00021 and 0.0016 respectively (ref.9). This indicates just how much more insoluble toluene is than the other solvents present.

Another, quite dramatic illustration of the dependence of the removal efficiency upon the particular solvent mix employed is shown in Fig.6. The ink mix used for Job 4 contains approximately 21% ethyl acetate and 76% ethanol while Job 5 uses a mix that is predominantly MEK (43.5%) and toluene (17.9%). (Job 5 was a similar printing task to Job 2 on Fig. 5). The Job 6 mix contains predominantly ethanol (44%), n-propyl acetate (42%) and ethyl acetate (12%).

It should be noted that the drop in efficiency while running Jobs 2 and 5 was not solely due to the difference in ease of capture and biological degradability of the solvents. On inspection of the data it became apparent that the total solvent concentrations being fed to the biofilter were much higher when running the above jobs. This could be expected to contribute to the decreased efficiencies illustrated above.

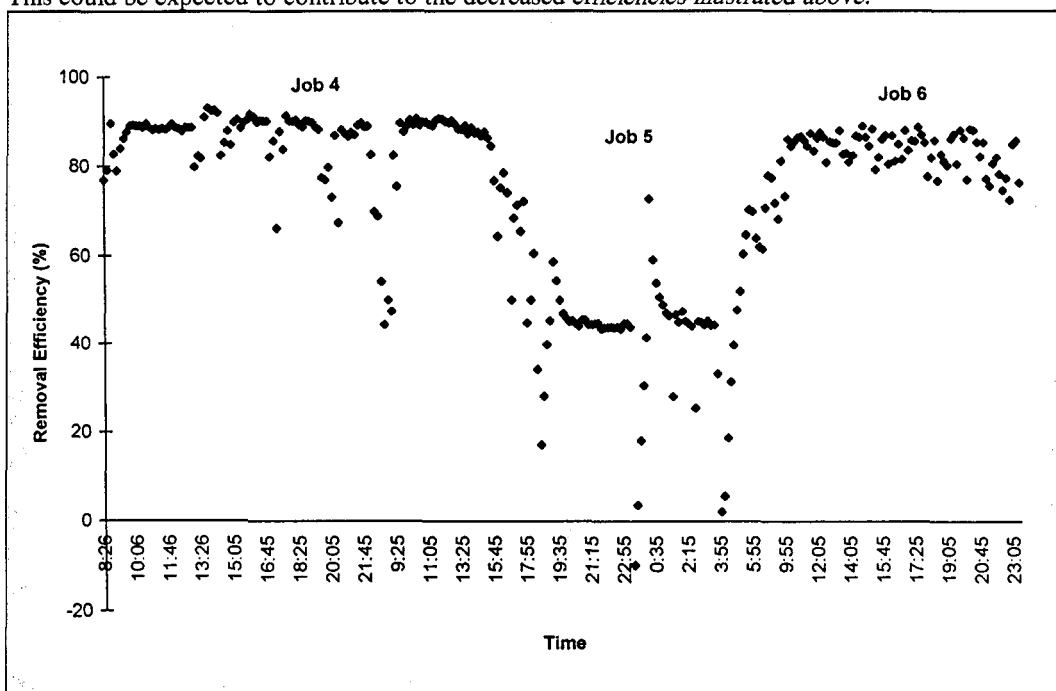


Fig.6. Solvent Removal Efficiency over the period from 19-3-1994 to 23-3-1994

A better understanding of the effect of solvent type on removal efficiencies can be gained from GC analyses of solvent mixes in gases entering and leaving the biofilter. (Unfortunately, no analyses are available for gas streams containing toluene.) These results confirm that methyl ethyl ketone removal efficiencies are poorer than those for the Class 1 solvents used. On the basis of the GC analyses, the solvents were ranked in order of decreasing ease of removal (this combines transfer to the aqueous phase and subsequent biodegradation) as follows: ethanol, n-propyl and iso-propyl alcohol, ethyl acetate, n-propyl acetate, methyl ethyl ketone. It should be noted, however, that this ranking is based on typical solvent mixes and loads at the Conoflex plant. Because such rankings must be dependent on the filter bed characteristics and on whether individual solvent removal rates are diffusion limited or reaction limited, different rankings could result on other plants.

Effect of Frequency of changing between Solvents upon the Solvent Removal Efficiency

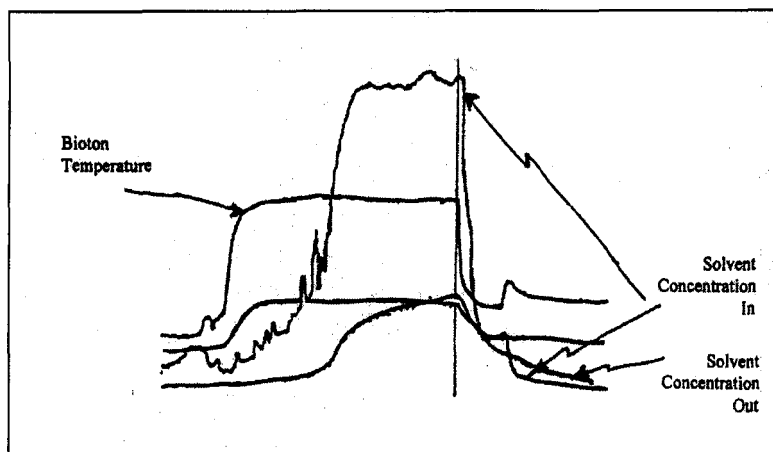


Fig.7. Section of a computer display showing evidence of solvent stripping occurring.

To establish whether any noticeable effect existed, the data was searched to find instances where a fairly rapid succession of jobs occurred and where a number of these used inks of similar composition. Data for such instances reinforce the conclusion reached earlier that the ink/solvent mix used does affect the solvent removal efficiency. However, the marked adverse effect upon solvent removal efficiency of, for example, using ink with a substantial toluene content is only apparent while the job

concerned is being run. There may be a brief transition period as one job gives way to another, but there appears to be no lasting influence of one job on solvent removal efficiencies for subsequent jobs.

It can be postulated that the lack of influence of changes of solvent mix is due to the regularity with which such changes occur on the plant. It would seem likely that because of repeated exposures to different solvent mixtures the biomass has developed a community of micro-organisms containing groups capable of handling each of the solvents regularly encountered. As one job succeeds another it might be expected that a different group of organisms become active. Any lag phase in microbial activity would in such circumstances be very short and not apparent in the data analysed.

Effect of Solvent Load and Duration upon Bioton Solvent Removal Efficiency

The Bioton was designed to reduce a 35 kg h^{-1} solvent load in the printing press exhaust gas stream by 80%. However, the input into the Bioton is not constant.

It was also noted that for each period of constantly high solvent input, the concentration at the biofilter outlet started off low and then increased to a relatively constant value. This relationship between inlet and outlet solvent concentrations occurred consistently throughout the period of the study. It seemed reasonable to attribute the initially higher solvent removal efficiency at the beginning of a period of high inlet concentrations to an increase in the amount of solvent adsorbed on the activated carbon and absorbed into the biolayer. The pattern followed by the outlet FID reading was similar to that that would be expected as the amount of solvent adsorbed or absorbed moved closer to the level at which it would be in equilibrium with the higher liquid phase solvent level brought about by the increase in gas phase solvent concentration. It follows that the constant solvent removal rate achieved towards the end of a period of high inlet solvent load reflected the upper limit of the capacity of the micro-organisms to utilise the particular solvent mix that is passing through the biofilter under the conditions existing in the biomass at the time.

It was also observed that for lower solvent concentration input values, such as input values less than 500 ppm, fluctuations in output solvent concentration were not as marked. The greater removal efficiencies achieved at these low solvent concentration input values implied that the biofilter had sufficient unused adsorptive capacity to take up most of the solvent during occasions when temporary marked increases in solvent load occurred. The consequence of this was the much closer adherence of the Bioton to design specifications. Another observation of interest apparent on displays such as that shown in Fig.7, is that at certain times, the solvent concentration in the outlet gas from the biofilter was greater than the inlet gas solvent concentration.

This occurred periodically when the inlet solvent concentration dropped sharply after a period of constantly high inlet values. Solvent stripping was occurring and reflected a reversal of the solvent absorption/adsorption process. The sudden drop in solvent inlet concentration was inferred to result in concentration gradient changes that cause solvent to be driven from the biolayer back into the gas film.

The Effect of Run Lengths and Weekend Shutdowns upon the Bioton Solvent Removal Efficiency

During 1993 and most of 1994 the printing press PF12 operated for 20 hrs/day for 5-1/2 days per week. Work undertaken by ClairTech in Holland had suggested that there is an adaptation phase which the micro-organisms go through at the beginning of an operating period following a significant shutdown. However there was no evidence of this adaptation phase in the Conoflex results. No adaptation period was apparent at the beginning of the day shift, at a stage when the printing press had not been operated for at least four hours. In addition, no adaptation phase was evident when the printing press was started on a Monday morning after having been shut down since the preceding Saturday evening. The lack of an adaptation phase at Conoflex was attributed to the presence of the activated carbon in the beds. During normal operation the activated carbon adsorbs substantial amounts of solvent. This solvent would be expected to be released and consumed by the bacteria as liquid phase solvent concentrations fall during periods of low or zero solvent load. The extent of the disruption to microbial metabolic activity should consequently be much less marked than would be the case if no activated carbon were present.

Nevertheless, careful examination of the data suggested that a slight decrease in overall efficiency occurred towards the end of the week but this could not be substantiated in any objective way. Such a decrease, if real, could be attributed to the activated carbon becoming progressively more saturated with less degradable solvents over the course of the week and losing the capacity to respond as effectively to peaks in solvent load. From a practical viewpoint, however, it was clear that there was no significant dependence of solvent removal efficiency on the day of the week concerned.

The Effect of a Holiday Shutdown Period upon the Bioton Solvent Removal Efficiency

Each summer Conoflex Packaging shuts down for a two to three week maintenance period. During this time the biofilter system is also shut down. From knowledge gained by ClairTech in Holland it was expected that once it started to receive solvent again after such a shutdown period, the biofilter would take time to reach a point where removal efficiencies had returned to normal. After the biofilter was restarted, after the Christmas holidays in 1994, it was discovered that the biomass beds were excessively dry (as noted above). Consequently the biofilter was not operational for a further month while this problem was rectified. The biofilter commenced operation again early in March. At this time there was a slightly reduced removal efficiency on the first day compared to that achieved during the next few days. This initially low removal efficiency value could reflect the existence of an adaptation period but the evidence for this is tenuous.

CONCLUSIONS

The data gathered on the biofilter at the Conoflex Packaging plant in Preston, Victoria, show that biofiltration was an effective way of achieving substantial reductions in solvent concentrations in off-gases from a rotogravure printing press. However, to achieve good and consistent results care was needed to ensure that the biofilter was properly constructed, operated and maintained. Of particular importance in achieving high solvent removal efficiencies was maintaining a moisture content of 40 to 60% in the filter beds. Humidification of the inlet gas was insufficient to achieve this. It was also necessary to add water directly to the filter beds to replace moisture losses; these losses were a direct consequence of the heat released in solvent oxidation reactions and play a vital role in stabilising filter bed temperatures.

For individual components of the solvent mix present in the printing press off-gases, it was not easy to predict the likely efficiency of removal. This was because local removal rates in the biofilter may be either diffusion limited or reaction limited, depending on the solvent vapour, load and composition, and presumably also on the distance the gases have travelled through the filter bed. For toluene removal rates in the Conoflex biofilter will always be diffusion limited. However, for other solvents, either other limiting mechanism may be

operating, depending on the circumstances. Despite these uncertainties, it was clear that where toluene was a major constituent of the solvent mix, a substantial reduction in removal efficiency occurred.

The measurements taken on the Conoflex biofilter also suggested that the system adapted rapidly to changes. It was inferred that the regularity with which changes occurred in the solvent loads and concentrations in the printing press off-gases had led to the development of a microbial population capable of handling any combination of solvents used in normal operations. It was also inferred that they were positively assisted in this by the presence of activated carbon, the damping of fluctuations in outlet gas solvent concentrations when inlet solvent loads are low, and the absence of adaptation periods after short to medium length shutdowns both support this.

Whilst it was clear that biofiltration is an effective means of removing a large fraction of biodegradable organic materials present in low concentrations in industrial exhaust air streams, it was also clear that much remains to be learnt about the best ways to build, operate and control biofilter systems. More research is needed to obtain a better understanding of the heat transfer, mass transfer and reaction processes occurring within biofilter beds. Further comprehensive, long-term studies of actual biofilter systems would also be most valuable in improving our understanding of biofilters used to remove solvents from off-gases generated in the printing industry.

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