Optimization of nutrient supply in a downflow gas-phase biofilter packed with an inert carrier

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Abstract.

Several methodologies were tested to supply nutrients to a downflow biofilter packed with perlite and used to treat toluene-polluted air. Despite the presence of an inorganic carrier, elimination capacities of up to around 60 g/m3 per hour could be maintained when a basal medium, containing nitrogen, phosphorus and potassium, was supplied once every fortnight or even once a month rather than once a week. Experimental results also indicated that the addition of vitamins or trace minerals to the basal aqueous medium hardly improved biofilter performance. Furthermore, the nutrient supply could be combined with a biomass control strategy, using air sparging, without any adverse effect on biofilter performance compared to supplying nutrients alone, and limiting the accumulation of excess biomass on the packing material. The performance of the biofilter was not significantly affected by temperature fluctuations between 25 and 33 °C.

Introduction

Over the past century, the emission of volatile compounds into the atmosphere has considerably increased. Most of these emissions are of anthropogenic origin. They include volatile organic compounds (VOCs) and volatile inorganic compounds (VICs), most often released from mobile sources, energy plants or industry (Van Agteren et al. 1998; Ciccioli 1993).

VOCs have traditionally been treated by physical or chemical methods, such as absorption, adsorption, condensation and incineration (Kennes et al. 2001). However, over the past few decades the development of biotreatment technologies has led to a decrease in the relative significance of these traditional approaches. The advantages of a biological treatment include high elimination efficiencies associated with low cost and limited negative environmental effects (Ottengraf 1987; Kennes and Thalasso 1998). In spite of these advantages, further research is needed in order to better understand and optimize the performance of biotreatment technologies.

Most biological air-pollution control strategies were initially based on the use of soil biofilters or biofilters filled with natural organic supports such as peat, compost, or wood bark. Recent studies have resulted in the development of new reactors and new support materials. Several types of inert carriers have been tested in biotrickling filters, and the use of such carriers in conventional biofilters without the continuous supply of a (trickling) aqueous phase has been reported recently (Kennes et al. 1996; Cox et al. 1997; Veiga and Kennes 2001). The present research focused on optimizing the composition and the frequency of supply of a nutritive aqueous solution fed occasionally to a biofilter packed with an inert carrier, namely perlite. The effect of combining the nutrient supply with air sparging – a biomass control strategy – and the influence of the temperature on biofilter performance were also evaluated.

Materials and methods

Biofilter set-up and operating conditions

Two identical downflow laboratory-scale biofilters were used. One contained a water jacket allowing temperature regulation during studies on the effect of temperature on biofilter performance. The biofilters were packed with 4.4 l perlite, an inert material previously used and characterized and very stable for long periods of time (Kennes et al. 1996; Cox et al. 1997; Veiga and Kennes 2001). The filter bed was sieved, retaining perlite grains with a 4–6 mm diameter. The void fraction of the bed before biofilm growth was 0.4. The cylindrical glass-reactors contained four equidistant sampling ports (Fig. 1). The feed was produced by mixing two different air streams. The stream with the highest flow rate was led to a carboy containing water and used as a humidification chamber, while the other one flowed through a flask containing toluene. All flow rates were regulated with flow meters. The overall air flow rate was maintained constant at 0.15 m^3/h throughout the study. Both reactors were protected from daylight in order to avoid the growth of algae.

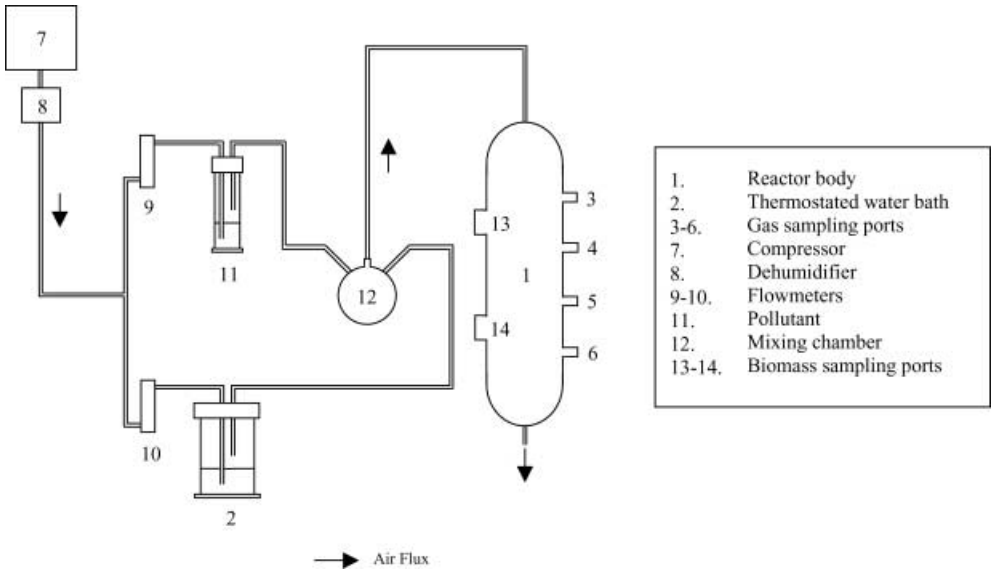

Fig. 1 Bioreactor scheme

A mixed culture obtained from a biofilter used in a previous study (Veiga et al. 2001) was used as inoculum. This reactor was originally inoculated with a defined consortium containing a Pseudomonas sp., a Bacillus sp. and the fungus Trichosporon beigelii and is described elsewhere (Veiga et al. 1999). A sample of the filter bed withdrawn from the biofilter was cultured in a flask containing 1 l medium and 0.2 ml toluene. The flask was maintained at 30 °C and shaken at 200 rpm before using its contents as inoculum.

Inoculation was performed under non-sterile conditions.

A nutrient solution containing (g/l) KH_2PO_4, 4.5; K_2HPO_4, 0.5; NH_4Cl, 2.0; and $MgSO_4 \cdot 7H_2O$, 0.1 was added periodically to the biofilters. In most experiments a vitamin solution and a trace mineral solution were added as well (Kennes et al. 1996). In the first assays and in control experiments, the vitamin solution and the trace mineral solution were added at a volume of 2 ml/l, each, unless otherwise indicated. A total volume of 2 l nutrient solution was usually poured on top of the biofilter and then drained off through the bottom of the reactor. A variable volume of aqueous medium, often around 20–25%, was retained in the biofilter.

Analytical methods

Gas-phase toluene concentrations were measured by gas chromatography on an HP-6890 gas chromatograph (GC) (Agilent Technologies, Spain) equipped with a flame ionization detector (FID) and a 30 m×0.32 mm HP-5 capillary column; the conditions were the same as for experiments carried out simultaneously with alkylbenzene mixtures (Veiga and Kennes 2001). The initial temperature of analysis was increased from 60 °C to 90 °C, with an increase of 5 °C/min. Samples were injected using a 0.5 cm^3 gas-tight syringe. Calibrations were carried out by injecting known amounts of toluene into a closed flask of known volume. After total evaporation of the liquid, a gas sample was injected into the GC.

Total solids and volatile solids were occasionally analyzed to calculate the moisture content and the amount of biomass growing on the packing material, according to Standard Methods (APHAAWWA- WEF 1998). A Warburg manometer was used to determine the pressure drop.

Performance parameters

The parameters used to express the performance of the bioreactors are defined below. They are a function of the inlet (Cin) and outlet(Cout) toluene concentrations, the volume occupied by perlite in the reactor (V) and the total gas flow rate (Q):

$$\mathrm{TL(TolueneLoad.\,g/m^3.h)} = \frac{C_{in}.Q}{V}$$

$$\mathrm{EC(EliminationCapacity.\,g/m^3.h)} = \frac{(C_{in} - C_{out})}{V}.Q$$

$$\mathrm{EE(EliminationEfficiency,\%)} = \frac{(C_{in} - C_{out})}{C_{in}}.100$$

Results

Preliminary studies and start-up

Before reactor inoculation, an experiment was performed aimed at measuring the degree of toluene adsorption on the filter bed. An air stream containing variable amounts of toluene was fed to the packed biofilter, and the concentrations of pollutant were measured at the reactor outlet. The results (Fig. 2) showed that there was no detectable adsorption, which led to the conclusion that, after inoculation, any reduction in toluene concentration along the biofilter would be due exclusively to biodegradation.

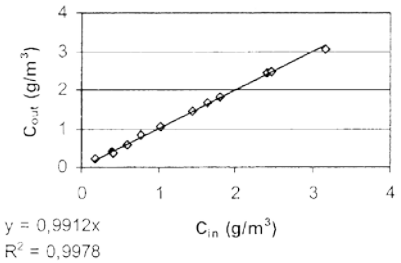

Fig. 2 Outlet vs inlet concentrations of pollutant before inoculation

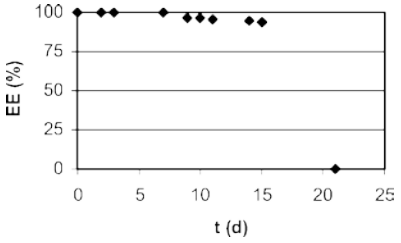

Fig. 3 Performance of the biofilter during the start-up period

Just after inoculation, the toluene supply was initially maintained around 10 g/m^{3}h. The elimination efficiency (EE) remained close to 100% during more than 2 weeks, and then dropped between days 15 and 21. This was most likely caused by a lack of nutrients since no nutrient solution was added to the biofilter during the first 3 weeks of operation, except on day 0 (Fig. 3). The high performance observed immediately after seeding the system is the result of using an adapted biocatalyst (Veiga and Kennes 2001). On day 22, 2 l nutrient solution were poured on top of the reactor and drained off after 30 min, which again resulted in a fast increase in EE up to 100%. From this day on, 2 l nutrient solution were added weekly to the biofilter and then drained off, in order to maintain a constant EE close to 100%. On day 41, the toluene load (TL) was directly increased to 26 g/m^3 per h. The EE remained above 98%. Thus, after the start-up period, a near maximal EE could be maintained by the feeding of an aqueous nutrient solution only once a week. The effect of modifying the feed frequency is described below.

Effect of temperature on biofilter performance

In a first stage, the biofilter was successively operated at three different temperatures. Each experiment lasted 1 month and the nutrient solution was supplied weekly. The goal of this experiment was to determine the effect of operating temperature on the performance of a biofilter inoculated with mesophilic microorganisms. Hence, temperatures chosen for this study were in the mesophilic range. The assays were performed at: (1) a constant temperature of 25 °C, (2) fluctuating room temperature (between 25.3 and 29.5 °C), and (3) a temperature of 33 °C. The TL ranged from 25 to 40 g/m3 per h. Figure 4 shows that basically identical performances were obtained between 25 and 33 °C, under conditions of either fluctuating or constant temperature since the EE values were similar in all three cases, always reaching above 95% at TL up to 30 g/m^3 per h, and remaining between 80 and 95% at higher TL. All further experiments were thus undertaken at room temperature.

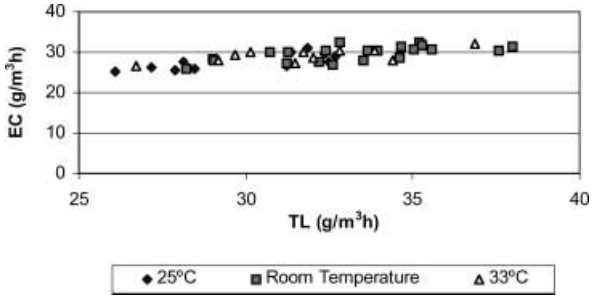

Fig. 4 Influence of temperature on biofilter performance at different toluene loads

Biofilter performance at a lower frequency of nutrient supply

After start-up of a biofilter, the periodic supply of a liquid nutrient solution remains a prerequisite to maintaining a high efficiency during long periods of time, if the support material is inorganic. As mentioned above, a weekly supply of an aqueous nutrient solution maintains a high steady EE in biofilters packed with perlite. Moreover, head loss in the system is minimal despite the dominant growth of filamentous fungi which normally leads to a rapid increase in pressure drop. In many of our biofilters, besides bacteria and fungi, the natural invasion of other organisms such as mites is often observed as well (Fig. 5). The next set of experiments was dedicated to further optimizing the strategy followed for the nutrient supply, by determining, on the one hand, whether the frequency of nutrient supply could be reduced and, on the other hand, if a poorer medium could be used.

After day 165, the nutrient solution was added to the reactor every fortnight instead of once a week. Initially, the TL and EE were, respectively, 65 g/m^3 per h and >99%. The TL was lowered to 6 g/m^3 per h on starting the experiment and then gradually increased to 65 g/m^3 per h, while the rest of the operation parameters were kept constant. As can be seen in Fig. 6, the biofilter functioned with a very high efficiency throughout the experiment, with the EE always above 95% even at the highest load, corresponding to the highest inlet concentration of 1.9 g/m^3. These results show that liquid medium can be supplied to a biofilter packed with perlite once every fortnight only, without affecting performance, even at relatively high TL.

Fig. 5 Invasion of mites in a biofilter packed with perlite and presenting heavy fungal growth

Performance of the biofilter under vitamin and trace mineral limitation

Studies have been published on the influence of a decrease in nutrient supply on biofilter performance, with most experiments focusing on the nitrogen source (Zhu et al. 1996; Deshusses and Cox 1999; Jorio et al. 2000; Moe and Irvine 2001). The present study was undertaken 8 months after start-up and was aimed at determining the effect of lowering the concentration of vitamins and trace minerals in the nutrient solution on biofilter performance. Although such compounds are sometimes used in laboratory-scale studies, they should be avoided in pilotscale or full-scale reactors or replaced by cheaper mixtures. As will be shown below, the addition of vitamins and trace minerals is basically not required as high EE values can also be maintained in their absence. In a preliminary experiment the effect of vitamin limitation on biofilter performance was determined by lowering the concentration of vitamins in the nutrient solution to 10% of the original concentration. The biofilter was operated for 1 month under this condition after running for 2 weeks with no vitamin limitation (control) to check biofilter performance before beginning the study. The same conditions were used as in the previous set of experiments, i.e. lowering the frequency of nutrient supply to once every fortnight. Increasing TLs of between 30 and 65 g/m^3 per h were applied. Figure 7 shows the relationship between TL and EC, during the control period and the period with reduced vitamin concentration. As can be observed in the figure, the difference in EE was minimal.

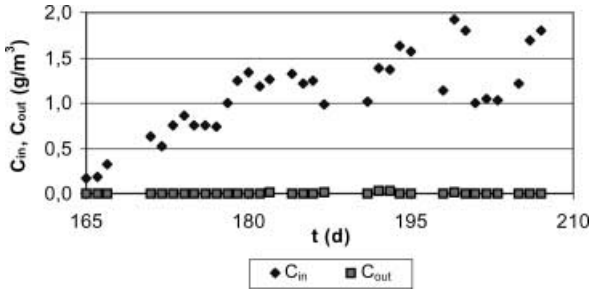

Fig. 6 Influence on biofilter performance of reducing the frequen-cy of nutrient supply from once a week to once every fortnight, at high inlet toluene concentrations

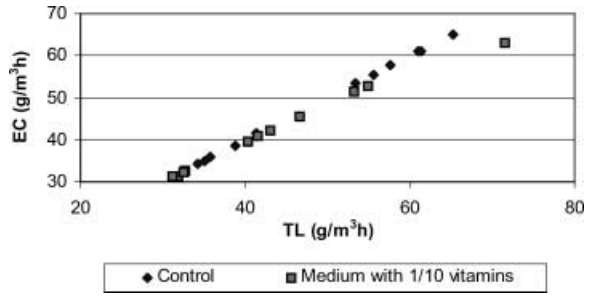

Fig. 7 Performance of the biofilter under vitamin limitation

In the next experiment, the effect of trace mineral limitation was evaluated. The experiment was carried out under conditions similar to the previous one. Again, the biofilter was first operated for 2 weeks without trace mineral limitation (control), while during the second part of the study the trace minerals were reduced to one-tenth of the original concentration. At TLs between 40 and 65 g/m^3 per h, EE values were always above 90% (data not shown) as in the case of vitamin limitation, suggesting that lowering the supply of trace minerals hardly affects biofilter performance.

The next experiment was aimed at evaluating the effect of completely eliminating any vitamin and trace mineral supply. Therefore, two biofilters were started up and operated

in parallel under similar conditions. The biofilter from the previous experiment was unpacked and clean, unused perlite was mixed with colonized perlite from that system in a ratio 70:30; this mixture was then used in two identical biofilters. This procedure was followed for several reasons. Since the biofilter from the previous studies had operated for already almost 1 year, unpacking and mixing the filter bed minimized compaction or channeling phenomena that could have adversely affected the performance. Also, the unpacking procedure followed by manual mixing allowed any residual liquid present in the filter bed to easily leach out, while a relatively large amount of biomass remained attached on the carrier material. Finally, by running two biofilters in parallel, the results could more readily and accurately be compared. The same amount packing material was introduced in each biofilter. In one of the reactors (biofilter 1) 2.5 l nutritive medium was poured on top of the biofilter and then drained off on day 0. In the other reactor (biofilter 2) the same procedure was followed except that the medium supplied on day 0 did not contain any vitamins or trace minerals. No aqueous phase was added to thebioreactors during the first 50 days of operation. The initial moisture and organic matter content found in two filter-bed samples taken from each reactor at the beginning of the experiment were very similar (Table 1).

Table 1

Initial moisture and organic matter contents in biofilters1 and 2

	Biofilter 1	Biofilter 2
Moisture (%)	54.8	55.2
Organic matter (%)	6.3	5.9

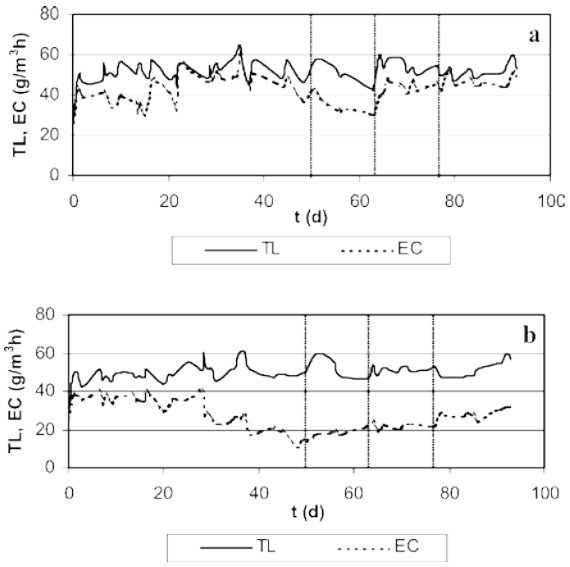

Fig. 8 Performance of two biofilters with (**a**) or without (**b**) addi-tion of vitamins and trace minerals on day 0. *Dotted lines* Days when the nutrient solution was supplied to the biofilters

Figure 8 shows the performance of the two biofilters. As can be seen, the reactors had very similar efficiencies during the first 3 weeks of operation. After day 25, the elimination capacity (EC) of biofilter 1 remained very high (Fig. 8a), while biofilter 2 suffered a gradual depletion in efficiency during the fourth week of operation (Fig. 8b). Since both reactors were run under similar conditions, it is clear that the different behaviors resulted from the presence or absence of vitamins and trace minerals. In biofilter 2, a high performance could be maintained for more than 3 weeks despite the absence of a nutrient supply and water during that period. Only after the sixth week of operation did biofilter 1 begin to suffer a slight drop of EE. The behavior patterns of the two reactors tended to continue until day 50, when they were moistened with tap water, without any nutrient source, to check the effect of water addition on biofilter performance. The addition of water did not significantly improve the performance of either of the reactors (Fig. 8), suggesting that nutrients rather than moisture were limiting. On day 63 both biofilters were fed a nutrient solution that contained only basal medium, without vitamins or trace minerals. The addition of nutrients resulted in an immediate increase in the efficiency of biofilter 1, but, as can be observed in Fig. 8b, had no significant effect on biofilter 2. This suggests that biofilter 2 was affected by a lack of one or more nutrients, which did not occur in biofilter 1, or that the biocatalyst's activity had been strongly or irreversibly affected. Although no data are available on viable cell counts, it is possible that a lack of nutrients over a long period of time in

biofilter 2 resulted in a decrease of active biomass. The absence of a positive response in biofilter 1 following the addition of water demonstrates that the moisture content was high enough and that the concentration of macro-nutrients (nitrogen, potassium, etc.) had been limiting. Therefore, vitamins and trace minerals were not a prerequisite for the stable and efficient operation of the biofilter for more than 2 months, when these compounds are added during the start-up of the biofilter. On day 77, both biofilters were fed the full nutrient solution, containing vitamins and trace minerals. In this case, the efficiency of biofilter 1 hardly increased since it had already reached more than 80% EE and an EC of around 50 g/m^3 per h. The data suggest that perlite-based reactors can operate efficiently for more than 6 weeks at an EC of 50–60 g/m^3 per h without the need for additional chemicals (Fig. 8a). The results also indicate that the addition of macro-nutrients, including nitrogen source, phosphorus and potassium may be reduced to once a month, while maintaining optimal EE and limiting at the same time biomass growth. The addition of vitamins and trace minerals after two and a half months had basically no effect on the performance of biofilter 1. In biofilter 2 (Fig. 8b), although the EC increased from 20 g/m3 per h to about 30 g/m^3 per h, the recovery was less significant and much slower than in biofilter 1, probably as a result of the presence of a less active or damaged biocatalyst.

Effect on biofilter performance of combining nutrient supply with air sparging

Next, the effect on biofilter performance of combining the nutrient supply with a biomass control strategy, to avoid clogging of the filter bed, was evaluated. The bioreactor was operated under constant conditions, and nutrients were supplied on three occasions, every 14-days, in combination with an air-sparging method developed in our laboratory (Kennes and Veiga 2002; Mendoza 2002): on day 1 (S1), day 14 (S2) and day 28 (S3). This approach was chosen because air sparging is one of the most efficient methods for biomass control in perlitebased biofilters (Kennes and Veiga 2002). The method consists of feeding the nutrient solution to the biofilter and sparging air through the system, which allows removal of excess biomass. Immediately after the end of feeding and air sparging, the polluted air flux was restored and Cin and Cout were measured. Figure 9 shows the EE vs time after each nutrient supply/air sparging session, with t=0 the time when nutrient supply/air sparging was started, and t=0.5 (dotted line) the time when these treatments ended and the polluted air flux restored; the TL was around 50 g/m3 per h. In all cases a decrease of EE was observed immediately after the treatments, ranging between 25% and 40%. The time needed for the reactor to recover EE above 80–90% was 8 h or less, suggesting that combining nutrient addition with the biomass control strategy did not adversely affect reactor performance and allowed the simultaneous removal of excess biomass.

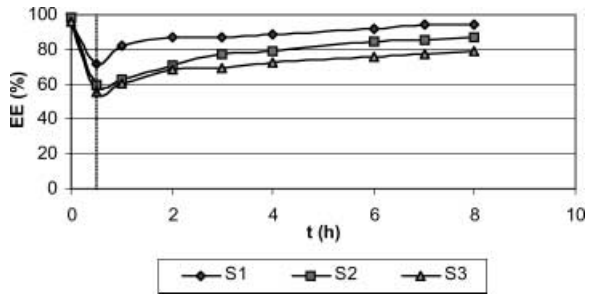

Fig. 9 Effect of a combination of nutrient supply and air sparging on biofilter performance

Discussion

Biological waste-gas treatment was originally carried out in conventional biofilters in which natural filter beds were used, i.e. compost, peat or soil. Such filter beds naturally

contain nutrients required by microorganisms to degrade VOCs. More recently, biotrickling filters have been developed. In such reactors, inert and/or inorganic packing materials are employed (Cox and Deshusses 2001). Therefore, the addition of an aqueous phase containing nutrients is a prerequisite for optimal performance, which is done through the continuous addition of a nutritive trickling phase. Over the past few years, it has been shown that inert materials such as perlite may also be used in conventional biofilters without the need of continuously feeding a nutritive solution, although the occasional addition of nutrients still remains a prerequisite (Groenestijn et al. 1995; Kennes et al. 1996; Cox et al. 1997).

In the present study, undertaken with mesophilic microorganisms, all experiments were done at room temperature since biofilter performance remained basically unchanged between 25 and 33 °C. Other authors have reported similar results. Indeed, Darlington et al. (2001) observed that between 23 and 26 °C, the temperature did not affect the performance of a biofilter degrading toluene, ethylbenzene and xylene, although the biological activity started decreasing at 20 °C. Cox et al. (2001), working over a wider temperature range, found that two biofilters, one operating at 22 °C and the other at 53 °C, had similar ethanol-removal efficiencies. Conversely, some authors have observed a more significant influence of temperature on biofilter performance (Shinabe et al. 1995; Elmrini et al. 2001).

Regarding the effect of the strategy used for the nutrient supply, during the start-up phase, after the initial addition of a nutritive solution, biofilters packed with perlite can be run under near optimal conditions for at least 2 weeks without the need to re-supply nutrients. The same is true after long-term biofilter operation. The addition of vitamins or trace minerals to the macro-nutrients (nitrogen, phosphorus, potassium) present in the nutritive solution does not significantly improve biofilter performance. Although the addition of nutrients every fortnight allows optimal removal efficiencies to be maintained, it appears that this frequency could even be reduced, at least temporarily, to once a month without adversely affecting performance. It was recently reported that it takes about 4 weeks for the water content to drop from 55–60% down to 35–40% in a perlite-based biofilter, while performance drops only at a water content below approximately 35% (Cox et al. 1996; Veiga and Kennes 2001). The present results indicate that not only the water content but also the nutrient content will remain high enough for at least 1 month without any addition of aqueous nutritive solution. Soon after pouring the nutritive solution on top of the biofilter, the solution is drained off. Nevertheless, part of it is retained in the system. In a filter bed, the level of water retention, which was around 20–25% in the present case, is largely dependent on the type of carrier material used (Bohn and Bohn 1999). The water-retention capacity of materials such as perlite acts as a reservoir of nutrients for the microorganisms. This phenomenon combined with the natural lysis of cells explains why a biofilter packed with an inert carrier does not necessarily require the continuous supply of nutrients.

The supply of low concentrations of nutrients at a relatively low frequency slows down biomass accumulation that might otherwise generate large pressure drops. Still, in the

long run, biofilters packed with inert carriers might eventually present channeling and plugging problems. The present results indicate that the nutrient supply can be combined with an air-sparging treatment aimed at removing excess biomass. The combined treatment significantly delays clogging problems (Kennes and Veiga 2002) and does not negatively affect biofilter performance. Only a temporary reduction in EE of a few hours is observed following the combined treatments. A similar decrease in performance has also been reported in response to other biomass control strategies in gas-phase biofilters, e.g. back-washing (Smith et al. 1996; Alonso et al. 1997).

In our biofilters, fungi gradually became dominant. Although the presence of filamentous fungi is expected to favor pressure drop, head loss remained minimal during the course of the experiments. The natural growth of mites (Fig. 5) grazing on fungi may explain this observation, since it was recently reported that the addition of

mites to filter beds colonized by fungi delays clogging problems (Groenestijn et al. 2001).

Acknowledgements

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