

## CHAPTER 4 - BIOTRICKLING FILTERS

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### 1. Introduction

As mentioned in Chapter 3, biotreatment for air pollution control started in the 1950s using soil beds. The technology has since matured and industrial applications of biofilters are numerous. More recently, research has focused on a variation of biofilters called *biotrickling filter*, or *trickling biofilter*. Biotrickling filters work in a similar manner to biofilters, except that an aqueous phase is trickled over the packing, and that the packing is usually made of some synthetic or inert material, like plastic rings, open pore foam, lava rock, etc. The trickling solution contains essential inorganic nutrients such as nitrogen, phosphorous, potassium, etc. and is usually recycled.

Biotrickling filters are more complex than biofilters but are usually more effective, especially for the treatment of compounds difficult to degrade or compounds that generate acidic by-products, such as H<sub>2</sub>S (Oh and Bartha, 1997; Mpanias and Baltzis, 1998; Cox and Deshusses, 2000<sup>b</sup>). Biotrickling filters can also be built taller than biofilters which reduces the footprint. It is also easier to control the conditions because of the free liquid phase, so that difficult applications are better handled by a biotrickling filter than by a biofilter. These include but are not limited to the treatment of high concentrations of contaminants, treatment of hot gases and/or of acid producing contaminants, treatment of air streams containing grease or particles, etc. Biotrickling filters are more recent than biofilters, and have not yet been fully deployed in industrial applications, but the prospective future applications are promising. The advantages and disadvantages of biofiltration and biotrickling filtration are compared to those of conventional air pollution control techniques in Table 4.1.

### 2. Biotrickling Filtration Fundamentals

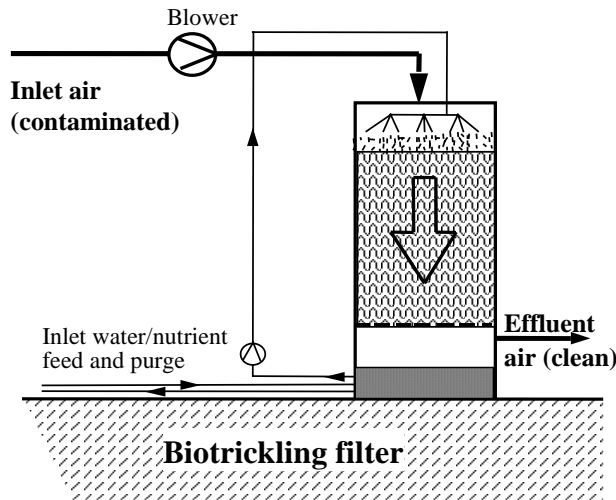
#### 2.1 BIOTRICKLING FILTRATION PRINCIPLE

The basic mechanisms of biotrickling filtration are shown in Figures 4.1 and 4.2. Contaminated air is passed co or counter-currently through a packed bed of inert materials on which a pollutant-degrading biofilm has established. At this time, there is no clear agreement as to whether co-current or counter-current is better. Mathematical modeling of kinetically limited systems (Diks and Ottengraf, 1991<sup>b</sup>, Zuber, 1995) suggests that co-current is better, since it minimizes stripping near the air exit, but experiments have failed to demonstrate that this was indeed true. For mass transfer limited systems, the flow direction may not matter (Lobo *et al.*, 1999) Constraints specific to the equipment construction might prevail until further demonstration that one or the other operating mode is preferred.

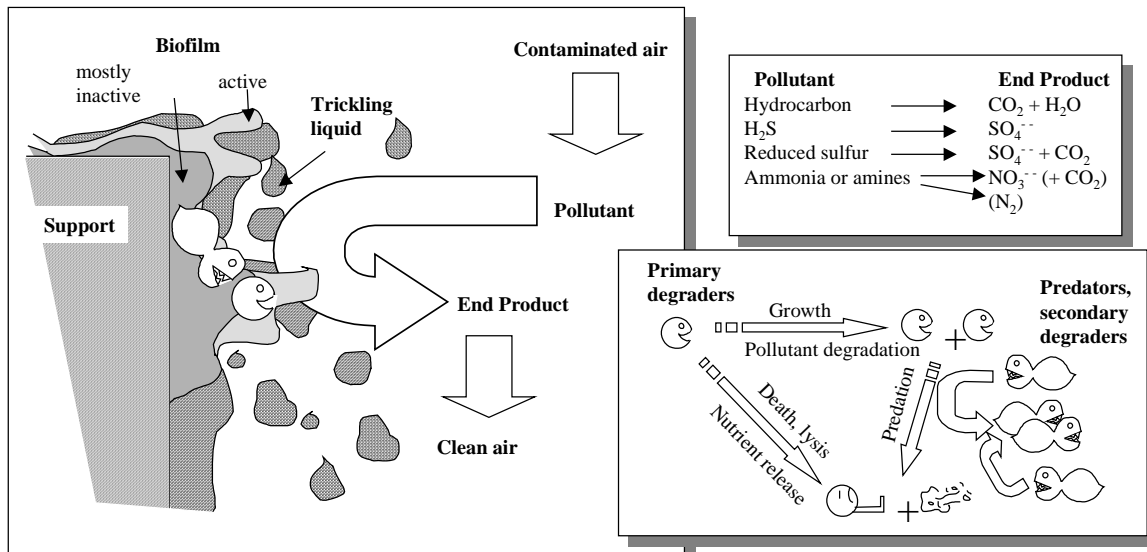
**Table 4.1.** Comparison of biofiltration, biotrickling filtration and conventional air pollution control techniques.

<b>Control Technology</b>	<b>Advantages</b>	<b>Disadvantages</b>
Biotrickling Filtration	<ul style="list-style-type: none"> <li>– Simple and low cost technology</li> <li>– Medium capital low operating costs</li> <li>– Effective removal</li> <li>– Treats effectively acid producing contaminants</li> <li>– Low pressure drop</li> </ul>	<ul style="list-style-type: none"> <li>– More complex to construct and operate than biofilters</li> <li>– Clogging by growing biomass if too much nutrient is added and high concentrations of VOCs are treated</li> </ul>
Biofiltration	<ul style="list-style-type: none"> <li>– Simple and low cost technology</li> <li>– Low operating and capital costs</li> <li>– Effective removal of low concentrations</li> <li>– Low pressure drop</li> <li>– No further waste streams produced</li> </ul>	<ul style="list-style-type: none"> <li>– Relatively large footprint requirement</li> <li>– Medium replacement every 2-5 years</li> <li>– Less suitable for high concentrations than biotrickling filters</li> <li>– Moisture and pH sometimes difficult to control</li> <li>– Particulate matter may clog the bed</li> </ul>
Wet Scrubbing	<ul style="list-style-type: none"> <li>– Medium capital costs</li> <li>– Can operate with particulate in gas stream</li> <li>– Relatively small footprint</li> <li>– Ability to handle variable loads</li> <li>– Well proven technology</li> </ul>	<ul style="list-style-type: none"> <li>– Very high operating costs</li> <li>– Reduced performance by scale deposit</li> <li>– Need for complex chemical feed systems</li> <li>– Does not remove most VOCs</li> <li>– Requires toxic and dangerous chemicals</li> </ul>
Carbon Adsorption	<ul style="list-style-type: none"> <li>– Short retention time/small unit</li> <li>– Consistent, reliable operation</li> <li>– Moderate capital costs</li> </ul>	<ul style="list-style-type: none"> <li>– High to extremely operating costs</li> <li>– Carbon life reduced by moist gas</li> <li>– Creates secondary waste streams (spent carbon)</li> <li>– Medium pressure drop</li> </ul>
Incineration	<ul style="list-style-type: none"> <li>– Effective removal of compounds irrespective of nature and concentration</li> <li>– Suitable for very high loads</li> <li>– Performance is uniform and reliable</li> <li>– Small footprint</li> </ul>	<ul style="list-style-type: none"> <li>– High operating and capital costs</li> <li>– High flow/ low concentrations not cost-effective</li> <li>– Usually requires additional fuel</li> <li>– Creates a secondary waste stream (NO<sub>x</sub>)</li> <li>– Scrutinized by the public</li> </ul>

Contrary to biofilters, biotrickling filters do not require the air to be pre-humidified or pre-conditioned prior to treatment. In most cases, the air will rapidly reach water saturation. If the air contains small amounts of dust or particulate matter, possibly grease, they will be leached via the medium purge. The medium feed consists of mineral nutrients, and can vary from a full basal salt medium with trace elements and pH buffer, as is usual in laboratory studies and in selected small field applications, to a very crude mix of fertilizers containing the necessary nitrogen, phosphorous and potassium. In wastewater treatment plants, industrial water (e.g., chlorinated secondary effluent), containing small amounts of dissolved organic materials and some residual nitrogen, has been used successfully to feed H<sub>2</sub>S degrading biotrickling filters (Morton and Caballero, 1996). It should be noted though that H<sub>2</sub>S degrading biotrickling filters host autotrophic organisms and have usually a low nutrient requirement.



**Figure 4.1.** Schematic of biotrickling filter setup (co-current and forced draft shown).



**Figure 4.2.** Basic principles of biotrickling filtration.

The feed rate and nutrient concentration is based on the expected nutrient requirements of the process culture. Based on typical growth media recipes, biotrickling filters often receive less nutrients than stoichiometrically required. This is because the nutrients are internally recycled by cell death, endogenous biomass digestion, and predators such as protozoa and rotifers. Nutrient limitation can also be a deliberate choice of the operator, in an attempt to limit bacterial growth and slow down biomass accumulation, as discussed in Section 5.5. Interestingly, the nutrient deficiency can be as high as a factor 20 of the theoretical amount required without severe consequences as far as pollutant removal rates are concerned. The determination of the feed rate and feed composition should also consider keeping the concentration of any salt or metabolite below any possible inhibitory concentration, although as will be discussed in Section 5.5, some investigators have on purpose allowed the salt concentration to rise, again, in an attempt to limit biomass growth.

Biotrickling filters differ from biofilters in that there is a free liquid phase. In most cases, the trickling solution is recycled, although in a few applications with very low trickling, a one pass of the liquid has been used. Overall, a wide variety of conditions have been experienced for trickling rates usually ranging from 0.05 to 20 m/h. The trickling rate influences the wetting of the packing and therefore the mass transfer of the pollutants to the biofilm. The trickling rate also influences the pressure drop over the packed bed. The fact that biotrickling filters work with only partial wetting of the biofilm, suggests that pollutants can transfer from the gas to the biofilm directly, or they can transfer from the gas to the trickling liquid, and then to the biofilm. While the latter path is generally assumed, mechanisms and rates for the direct transfer from the gas to the biofilm has been vastly ignored (Cox and Deshusses, 1998).

As far as pollutant degradation is concerned, most of the degradation occurs in the biofilm, although in selected cases, significant activity can occur in the recycle liquid, as reported by Cox *et al.*, 2000. Examination of Figure 4.2 reveals that pollutant elimination is the result of a complex combination of mass transfer (diffusion, convection) and biological processes, such as growth, death and lysis, predation. These are complicated by the fact that the biofilm houses multiple species in a complex architecture. All of these are affected by the operating conditions of the system and also affect the pollutant removal and the general dynamic of the biotrickling filter. Under these circumstances, a complete description of all the phenomena involved in a biotrickling filter is not yet available. While an improved fundamental understanding would be desirable for reactor design and operation, the lack of fundamental understanding does not prevent application of the technology.

## 2.2 DEFINITIONS, PERFORMANCE REPORTING

As in the case of conventional biofilters, operation and performance of biotrickling filters for air pollution control is generally reported in terms of removal efficiency, or pollutant elimination capacity as a function of the pollutant loading, or the gas empty bed retention time (EBRT). These terms extensively defined in Chapter 3 are briefly recalled below (Equations 4.1-4.4).

$$\text{Removal} = \text{RE} = \frac{C_{\text{in}} - C_{\text{out}}}{C_{\text{in}}} \times 100 \quad (\%) \quad (4.1)$$

$$\text{Pollutant Elimination Capacity} = \text{EC} = \frac{(C_{\text{in}} - C_{\text{out}})}{V} \times Q \quad (\text{g m}^{-3} \text{ h}^{-1}) \quad (4.2)$$

$$\text{Empty Bed Retention Time} = \text{EBRT} = \frac{V}{Q} \quad (\text{s or min}) \quad (4.3)$$

$$\text{Pollutant loading} = L = \frac{C_{\text{in}}}{V} \times Q \quad (\text{g m}^{-3} \text{ h}^{-1}) \quad (4.4)$$

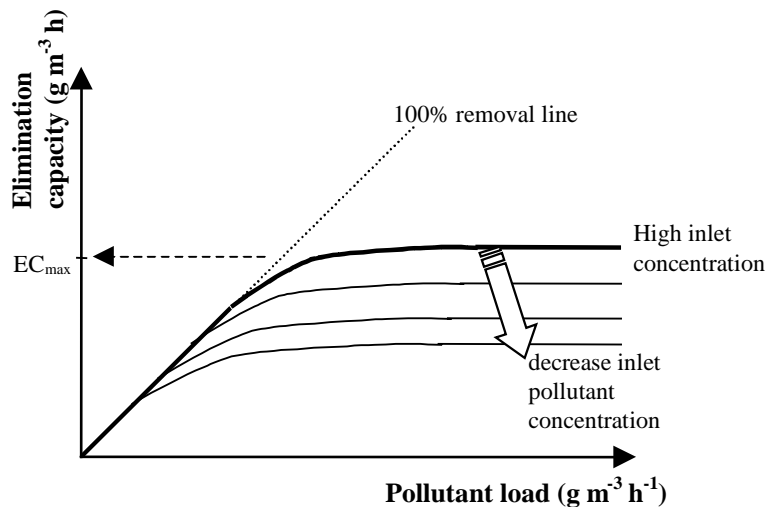
where  $C_{\text{in}}$  and  $C_{\text{out}}$  are the inlet and outlet pollutant concentrations (usually in  $\text{g m}^{-3}$  or in dilution to threshold for odour removal), respectively,  $V$  is the volume of the packed bed ( $\text{m}^3$ ) and  $Q$  is the air flow rate ( $\text{m}^3 \text{ h}^{-1}$ ). Pollutant concentrations are usually reported as mass

per volume; conversion of volumetric to mass concentrations is done using the ideal gas law which reduces to Equation 4.5 at room temperature.

$$\text{Concentration (g m}^{-3}\text{)} = \frac{\text{Concentration (ppm}_v\text{)} \times \text{molecular weight of pollutant (g mol}^{-1}\text{)}}{24,776} \quad (4.5)$$

It should be stressed that the elimination capacity and the loading are calculated using the volume of the packed bed and not to the total volume of the reactor. Depending on the reactor design, the volume of the packed bed volume will be about 40-90% of the total reactor volume. Also, the EBRT is calculated on the basis of the total volume of packed bed (Equation 4.3) and not the void space in the packing. The actual gas residence time will be lower depending on the porosity of the packing, the dynamic liquid hold-up and the amount of biomass attached to the packing. The porosity of packing ranges from about 50% (lava rock) to 95% (all random or structured packings), the liquid holdup is usually less than 5% of the bed volume, and biomass may occupy 5% to 30% of the bed volume. Hence, the actual gas residence can be less than half the EBRT.

A typical elimination capacity vs. pollutant loading curve is shown in Figure 4.3. Note that it is similar to curves obtained for conventional biofilters. It is usual to report the performance as a function of the load, i.e., inlet concentration  $\times$  air flow, rather than the concentration. This enables comparison of systems of different sizes operated under different conditions. One underlying assumption is that the performance depends only on the pollutant load, hence, that low concentrations-high flowrates conditions lead to similar elimination capacities than high concentrations-low flowrates. This assumption is generally valid because the pollutant concentrations commonly encountered in biotrickling filters are often high enough for the micro-kinetic to be of zero order. As schematically shown in Figure 4.3, this is no longer true at very low pollutant concentrations (typically below 0.05 - 0.1 g m<sup>-3</sup>), in particular for pollutants with high Henry's law coefficients, because first order kinetics will prevail in the biofilm resulting in a reduction of the maximum elimination capacity..



**Figure 4.3.** Schematic of a typical elimination capacity vs. load curve for a biotrickling filter.

Examination of Figure 4.3 reveals that there are essentially three operating regimes, as for the case of conventional biofilters:

1. Low loading, also called first order regime. The elimination capacity and the loading are identical and the pollutant is completely removed. The biotrickling filter is operated well below its maximum elimination capacity. The performance increases proportionally with the loading.
2. Intermediate range. Breakthrough of the pollutant occurs. With higher inlet concentrations or higher air flow rates, the elimination capacity increases, but to a lesser extent than the loading.
3. High loading, also called zero order regime. The biotrickling filter is operated at its maximum elimination capacity. Increases in loading do not result in further increases in elimination capacity, the removal efficiency decreases. As indicated on the figure, if the concentration is very low, the maximum elimination capacity is reduced accordingly.

For the evaluation of biotrickling filter performance, one should consider both the maximum elimination capacity and the removal efficiency. For practical reasons, academic research is mainly concerned with the maximum elimination capacity or with high performance, which occur at relatively high pollutant concentration and often less than ~90% removal efficiency. On the other hand, reactor design for industrial application often needs to meet a certain discharge requirement, or achieve a high removal percentage. Thus there might be some challenges in extrapolating research data for reactor design.

### 2.3 BIOTRICKLING FILTER CONSTRUCTION

To be cost effective, biotrickling filters need to be both inexpensive and reliable. This is achieved by careful material selection, reactor design, and reactor operation and monitoring. Basic construction considerations are discussed in this section.

The biotrickling filters are always in-vessel type reactors, although conceptually, one could consider open-bed biotrickling filters, in a similar manner to open bed biofilters (see Chapter 3). The vessel needs to provide the structural strength to support the bed. It can be constructed out of fiberglass or fiberglass reinforced plastic, plastic polymers (PE, PP, less frequently PVC), metal, or concrete. Fiberglass or fiberglass reinforced plastic are commonly used in air scrubbers and provide a good structural strength and excellent resistance to various chemicals. Reactors can be build relatively tall (10-15 m). Fiberglass or fiberglass reinforced plastic reactors are relatively heavy and can be expensive. Polyethylene (PE) or polypropylene (PP) vessels offer the convenience of lightweight materials, low costs and good chemical resistance. The tensile strength of PE or PP is somewhat limited, so that special considerations are required for tall reactors. Biotrickling filters can also be constructed out of carbon steel, stainless steel or other metals. Usually, this is a costly option. A special care should be paid to corrosion resistance, in which case an epoxy coating, or a similar corrosion preventing coating is warranted. Biotrickling filters can possibly be made out of stackable containers to save space, as for some biofilters commercially available (Sabo *et al.*, 1995). Finally, biotrickling filters could be constructed in concrete (custom or precast cinder blocks), although to our knowledge this has not yet been attempted. Here again, special consideration for corrosion resistance is warranted, especially for the treatment of hydrogen sulphide or if low pH is expected. In all cases, in a

similar manner to biofilters, thermal insulation may be required around the vessel for reactors installed in cold climate.

In most cases, the bottom of the reactor is used as a reservoir for the trickling liquid which eliminates the need for a separate tank. The bottom of the reactor is also the preferred location for any liquid heating which is best done using a submerged electric heater. In line heat exchangers are not recommended. In addition to be more costly, heat exchangers would be subject to rapid fouling by biomass which lowers heat transfer and increases liquid pressure drop. The biotrickling filter vessel includes various duct work and liquid piping. For air ducting, fiberglass, PE, galvanized or stainless steel is common. In most cases, inexpensive PVC piping is adequate for the liquid. The concentrations of pollutants are generally too low to alter the PVC. However, a special care should be placed on the liquid distribution system on top of the bed, as improper distribution will affect the reactor's performance. A convenient way to distribute the liquid onto the packing is via a series of spray nozzles placed so that a homogeneous distribution is achieved. Non-clogging nozzles are recommended, and monthly inspection of the nozzles should be performed. The recycle flow is ensured by a pump, usually a high efficiency centrifugal pump. In most cases, the pump will be relatively small (<3 HP). If trickling is intermittent, a timer is installed, or the pump is remotely operated by the central process control software. In key applications, two pumps should be installed, one as a backup in case the other fails. Having a reliable liquid distribution and pumping is key, since the biotrickling filter will rapidly fail in absence of liquid trickling.

As in the case of biofilters, a careful selection of the blower is a key to cost effective operation. In large applications, a variable speed drive is recommended. While these are much more expensive than single speed drive, they allow for adjusting the flow and the head to compensate for the building up of biomass and the slow increase of pressure drop over time. This saves on energy costs otherwise wasted in dampers or other flow regulating devices. The blower can be installed upstream (forced draft) or downstream (induced draft) of the biotrickling filter. In any cases, sufficient space should be allocated above and below the bed, for the air distribution and the air exit, and for the liquid distribution. This is particularly true for large reactors, where poor air distribution can cause significant short-circuiting and decrease pollutant removal (Chitwood *et al.*, 2000). Depending on the reactor configuration (co or counter-current) and on the air velocity, installation of a demister will be required to prevent carry over of water droplets. It can consist in a simple water knock-out drum or a section of bare packing, or a specially designed non-clogging demister.

The biotrickling filter will usually include a tank for the nutrient solution, which is best stored as a concentrated solution to avoid bacterial growth in the stock solution, and a tank for caustic if the reactor includes a pH control. These are usually made of PE or PP and their volume should be such as to ensure at least a two-week supply. Nutrients or caustic are fed manually or by pump based on a timer, or pumped as triggered by a process controller.

The degree of sophistication of the controls will usually determine the nature of the ancillary equipment as well as reactor operation and maintenance. A minimum of control is desired to ensure safe operation. This includes installation of a low level switch at the bottom of the reactor to prevent the recycle pump to run dry. If deemed necessary, the biotrickling filter may be equipped with sophisticated sensors and control routines using logic statements to detect abnormal operation, and propose or implement remedial action via feedback controllers (Webster *et al.*, 1999). For instance, the medium feed and purge might be controlled using the input of conductivity probe (as an indicator of the overall salt concentration) installed in the recycle liquid, and the input of a level switch. However, it

should be stressed sophisticated controls are not always necessary. The low-tech alternative for the above will consider an operator removing a given volume of the recycle liquid on a given schedule (daily or weekly) and replace it with water and a mineral nutrient mix, often fertilizer grade. The choice of sophisticated control or mostly manual operation depends on the application, the size of the reactor, the regulatory agency and the available budget.

Monitoring the reactor performance is often defined in the permitting of the biotrickling filter. It can be on-line integrated with the process control, or off-line by grab samples. Monitoring is an important part of operation and maintenance. Proper monitoring helps in troubleshooting and in optimizing the operation of the reactor. Most important is the monitoring of pollutant removal. For VOCs, this is best done using a flame ionization detector, while for H<sub>2</sub>S, and ammonia, special meters are available. Monitoring tubes can also be conveniently utilized. For odour treatment, odour panels are still the best measurement although they are expensive. The measure of the odour removal efficiency is usually based on the odour dilution-to-threshold, although the character of the odour should also be considered. Inlet and outlet should always be measured. The frequency of the measurement depends on the regulatory requirement, the equipment used and the operating conditions of the biotrickling filter (steady conditions or rapid transients).

Other parameters to consider in a monitoring plan include airflow rate, reactor temperature, pressure drop, biomass content, and parameters pertaining to the recycle liquid, such as flow rate, pH, ionic strength, biological oxygen demand, and possibly dissolved oxygen in the case of the treatment of high pollutant concentrations. Structural inspection and proper functioning of the biotrickling filter components (e.g., spray nozzles, feed lines, etc.) are also important. The challenge of defining a good monitoring plan will be to schedule the necessary sampling that allows for a good control of the biotrickling filter and an early detection of possible problems, while minimizing maintenance and analytical costs.

### 3. Biotrickling Filter Performance

#### 3.1 EXAMPLES OF POLLUTANTS TREATED IN BIOTRICKLING FILTERS

Over the past decade, many pollutants have been tested for treatment in biotrickling filters, mostly in the laboratory. Effective removal has been reported for aliphatic and aromatic hydrocarbons, oxygenated hydrocarbons, chlorinated hydrocarbons and inorganic compounds. Typical examples are presented in Table 4.2. Most pollutants contain carbon and they serve as a carbon and energy source for aerobic, heterotrophic microorganisms. Other biological processes observed or applied in biotrickling filters include:

- Oxidation of inorganics as a source of energy. An example is hydrogen sulphide, which is oxidized to sulfuric acid by *Thiobacillus* species (Section 3.3). These species are in general autotrophic, using carbon dioxide from air as the carbon source for growth. Another example is the oxidation of ammonia to nitrate. Effective removal of ammonia in a nitrifying biotrickling filter has been demonstrated by Van Groenestijn and Lake, 1999. In a second denitrification reactor, the produced nitrate was reduced to nitrogen gas as the final product of the combined system.
- Cometabolism. An example is the removal of trichloroethylene in biotrickling filters (Sun *et al.*, 1998). Cometabolism requires the presence of an inducing growth substrate, which may be already present in the waste gas or has to be added artificially.

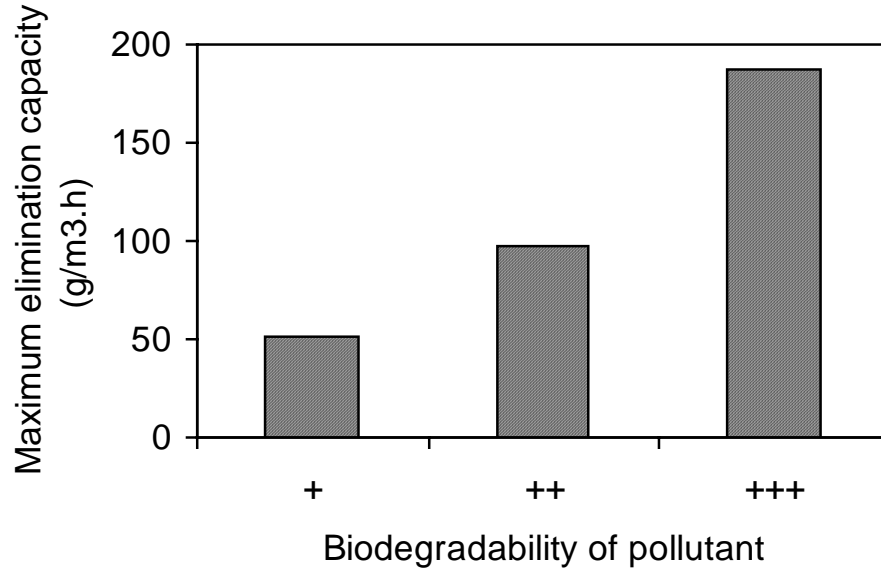
- Anaerobic removal of compounds that are persistent under aerobic conditions (Section 4.2).

Biotrickling filtration, as any other biological technique, is successful only if the pollutant is biodegradable and if it is available in concentrations sufficiently high to support a thriving process culture. Quantitative criteria for pollutant biodegradability have not been established, hence, the pollutants in Table 4.2 are classified in three groups of increasing biodegradability. The pollutant availability to the process culture is related to the pollutant mass transfer into and in the biofilm. Table 4.2 lists the dimensionless Henry coefficient as an approximation of the pollutant availability. The following general trends can be discerned:

- The startup or lag phase increases with decreasing biodegradability. Whereas biotrickling filters treating easily biodegradable compounds may operate at their maximum elimination capacity within a few days, effective removal of compounds such as MTBE may only be observed after several months of operation. As discussed in Section 4.4, the startup phase may be shortened by inoculation of the biotrickling filter with microorganisms with a high specific activity towards degradation of the pollutant of interest.
- If the gas/liquid partition is favourable (low  $H$ ), the elimination capacity increases with higher biodegradability of the pollutant. This is illustrated in Figure 4.4. For biotrickling filters that are kinetically limited, maximum elimination capacities of 200-300  $\text{g/m}^3\cdot\text{h}$  can be obtained for easily biodegradable pollutants. Performance at higher pollutant loads is more likely to be limited by factors other than the biodegradability, for instance by oxygen limitation (Section 4.2) or by rapid accumulation of biomass (Section 5).
- Irrespective of the biodegradability, removal of hydrophobic compounds such as alkanes may be limited by the mass transfer rate. As a rule of thumb, mass transfer limitation can be expected to play an increasingly important role for compounds with a Henry coefficient greater than 0.5-1.

### 3.2 TOLUENE AS MODEL POLLUTANT; COMPARISON OF BIOTRICKLING FILTER PERFORMANCES

Toluene can be considered as moderately water-soluble and easily biodegradable with a maximum elimination capacity in biotrickling filters of about 80  $\text{g/m}^3\cdot\text{h}$  (Table 4.2). Over the past ten years, many researchers have used toluene as their model pollutant. This enables a direct comparison of how reactor design and operation affect the overall performance. It is worthwhile to mention that biotrickling filter performance is very reproducible provided that the reactors are operated under identical conditions. In a comparative study with twenty identical, toluene-degrading biotrickling filters operated in parallel, the average maximum elimination capacity was 79  $\text{g/m}^3\cdot\text{h}$  with a standard deviation of 5.7  $\text{g/m}^3\cdot\text{h}$  (Cox and Deshusses, 2000<sup>a</sup>).



**Figure 4.4.** Maximum elimination capacities averaged for pollutants grouped by increasing biodegradability.

Table 4.3 summarizes the design, operation and performance of toluene-degrading biotrickling filters in five independent studies conducted by different laboratories. The maximum elimination capacity varied from 18 to 120 g/m<sup>3</sup>.h, which greatly exceeds the variability observed for the elimination capacity of identical biotrickling filters as discussed above. Not unexpectedly, parameters related to the mass transfer of the pollutant (e.g., specific surface area of packing, superficial liquid velocity), the microbiology (e.g., type of inoculum, supply of nutrients), the design (e.g., reactor dimension and type of packing) and the operation of biotrickling filters (e.g., gas residence time, inlet concentration, cocurrent versus countercurrent operation) affected the performance. Currently, research is attending to identify and quantify parameters crucial for the optimum biotrickling filter performance. Some is discussed in next sections.

Due to the complexity of the process, the design engineer has at present only a few objective criteria for developing full-scale biotrickling filters that go beyond the thumb-of-rule principle. Despite efforts to model biotrickling filter performance (e.g., Diks and Ottengraf, 1991<sup>2</sup>; Pedersen and Arvin, 1995; Mpanias and Baltzis, 1998; Okkerse *et al.*, 1999; Lobo *et al.*, 1999), these have not yet proven to be very helpful for design. Therefore, it is still common practice to perform pilot-plant studies before designing and constructing full-scale reactors. This pilot tests are important because conditions at field locations are usually not constant, but subject to changes of the temperature, waste gas composition and/or concentration of pollutants. This may have a marked effect on the performance of biotrickling filters, which is often not considered in the constant environment of the laboratory (Webster *et al.*, 1999).

**Table 4.2.** Examples of pollutant removal in biotrickling filters.

Pollutant	Biodegradability <sup>b</sup>	Henry coefficient <sup>c</sup>	Lag phase	EC (g/m <sup>3</sup> .h)	Reference
<i>Aliphatic hydrocarbons</i>					
Hexane	++	74	50 days	7.5	Plaggemeier <i>et al.</i> , 1997
Heptane	++	83.2	NA	24	Schindler and Friedl, 1995
<i>Aromatic hydrocarbons</i>					
Styrene	++	0.11	15 days	32	Pol <i>et al.</i> , 1998
Styrene <sup>a</sup>	++	0.11	NA	35	Webster <i>et al.</i> , 1999
Toluene	+++	0.28	10 days	80	Cox and Deshusses, 1999 <sup>a</sup>
<i>Oxygenated hydrocarbons</i>					
Propionaldehyde	+++	0.0024	NA	300	Kirchner <i>et al.</i> , 1992
Acetone	+++	0.0016	NA	500	Kirchner <i>et al.</i> , 1992
Methyl ethylketone	+++	0.0024	NA	40	Chou and Huang, 1997
Methanol	+++	0.00019	5 days	100	Allen <i>et al.</i> , 2000
n-butanol	+++	0.00035	NA	100	Heinze and Friedrich, 1997
Diethyl ether	+	0.028	2 weeks	60	Zhu <i>et al.</i> , 1996
Methyl- <i>tert</i> -butyl ether	+	0.023	7 months	45	Fortin and Deshusses, 1999
<i>Chlorinated hydrocarbons</i>					
Dichloromethane	+++	0.093	1 week	200	Hartmans and Tramper, 1991
	+++	0.093	1 week	157	Diks and Ottengraf, 1991 <sup>b</sup>
Chlorobenzenes	++	0.18	NA	300	Oh and Bartha, 1994
	++	0.18	NA	60	Mpanias and Baltzis, 1998
<i>Nitrogen and sulfur compounds</i>					
Nitrobenzene	+	0.00098	4 weeks	50	Oh and Bartha, 1997
Carbon disulphide <sup>a</sup>	++	0.39	NA	220	Hugler <i>et al.</i> , 1999
Nitric oxide	+	NA	6 weeks	25	Chou and Liu, 2000
Hydrogen sulphide <sup>a</sup>	+++	0.94	2 weeks	100	Kraakman <i>et al.</i> , 1998

<sup>a</sup> Pilot-plant study.

<sup>b</sup> From poorly (+) to highly biodegradable (+++) (Deviny *et al.*, 1999).

<sup>c</sup> Dimensionless Henry coefficient. Major sources: Nirmalakhandan and Speece, 1988, and Lide, 1997.

**Table 4.3.** Selected laboratory studies on toluene removal in biotrickling filters.

Parameter	Pedersen and Arvin, 1995	Cox and Deshusses, 1999 <sup>a</sup>	Smith <i>et al.</i> , 1996	Schönduve <i>et al.</i> , 1996	Weber and Hartmans, 1996
<i>Reactor design</i>					
Reactor, ID × H (cm)	9x70	15x130	15x114	15x25	30x100
Packing	Steel Pall rings	PP Pall rings	Celite pellets	PP Pall rings	Pall rings
Specific surface area (m <sup>2</sup> /m <sup>3</sup> )	317	220	1190	NA	110
Inoculation	Adapted consortium	<i>Pseudomonas corrugata</i>	Sample of reactor	Adapted consortium	Bacterial consortium      Fungal consortium
<i>Reactor operation</i>					
Gas flow	Upflow	Downflow	Downflow	Upflow	Upflow
Empty bed gas residence time (s)	32-160	56	60-120	17-132	36
Toluene inlet concentration (g/m <sup>3</sup> )	0.19-1.61	0.5-3.5	0.93	0.2-3.8	0.7
Superficial liquid velocity (m/h)	3.3-4.7	7.9	0.05	22.6	1.6
Nutrient feed	Batch	Continuous	Semi-continuous <sup>a</sup>	Batch	Batch
Nutrient composition	Semi- full medium	Full medium	Full medium	Full medium N-NH <sub>4</sub> <sup>+</sup> Full medium N-NO <sub>3</sub> <sup>-</sup>	Complete, limiting concentrations
Liquid residence time (day)	NA	0.5	-	NA	NA
<i>Reactor performance</i>					
Load (g/m <sup>3</sup> .h)	6-150	35-225	60	~50-1300	70
Elimination capacity (g/m <sup>3</sup> .h)	35	71-83	60	120      33	18      34

<sup>a</sup> No liquid recycling, but once-through pass of fresh medium.

### 3.3 PILOT-PLANT STUDIES AT US WASTEWATER TREATMENT PLANTS

Although biotrickling filter research in the US has trailed Europe over the past two decades, recently rapid progress has been made especially in the area of odour control at wastewater treatment facilities. Waste gases at these facilities contain hydrogen sulphide ( $\text{H}_2\text{S}$ ) as the principal odour-causing agent in concentrations up to 100 ppm as well as lower concentrations (0-100 ppb) of various VOCs and chlorinated hydrocarbons. Chemical scrubbers are currently employed to reduce the odour problem. Although they are effective in removing  $\text{H}_2\text{S}$ , the high consumption of chemicals (caustic soda, hypochlorite) and their ineffectiveness in controlling VOCs are drawbacks of increasing concern. Demonstration projects, many of them located in Southern California and conducted by universities, wastewater treatment facilities and industry, focus on using biofilters and biotrickling filters as alternatives for chemical scrubbers.

In 1993, a biotrickling filter study was done at the Los Angeles County Sanitation Districts (Morton and Caballero, 1996). Greater than 98% and 99% removal of respectively  $\text{H}_2\text{S}$  and odour was demonstrated in a pilot unit of  $0.3 \text{ m}^3$  with lava rock as the packing material. The gas residence time was varied between 12 and 30 sec, which is considerably more than the typical detention time in chemical scrubbers (2-5 sec). A constant pH of 2.0-3.0 was maintained by continuously supplying secondary effluent water and purging the produced sulfuric acid. The secondary effluent water also proved to be good source of nutrients for growth and activity of  $\text{H}_2\text{S}$  oxidizing bacteria. Because of concerns of clogging of the lava rock, a second experiment was set up using a porous plastic packing. The  $\text{H}_2\text{S}$  removal efficiency was lower, presumably because of mass transfer limitation due to a lower specific surface area of the packing. The study of Morton and Caballero, 1996 illustrates the importance of packing material selection.

Co-treatment of  $\text{H}_2\text{S}$  and VOCs has been studied in a number of projects. For optimization of both removals, there may be a conflict of pH optima.  $\text{H}_2\text{S}$  is in general oxidized by *Thiobacillus* species with an optimum pH of about 2. VOCs are degraded by heterotrophs that generally prefer a neutral pH, although VOC removal in low-pH,  $\text{H}_2\text{S}$ -oxidizing biofilters and biotrickling filters has been observed (Torres and Basrai, 1998; Chitwood *et al.*, 1999; Cox and Deshusses, 2000<sup>b</sup>). Chitwood *et al.*, 1999 investigated a two-stage process at the Ojai Valley Sanitary District. This process consisted of an acidic reactor with lava rocks for  $\text{H}_2\text{S}$  removal, followed by a pH-neutral biofilter for VOC removal. The acidic reactor had a very low intermittent trickling rate. Overall performance and removal efficiency of the two-stage process seemed slightly better than combined removal in a single stage biofilter (Chitwood *et al.*, 1999). Other studies have focused on biotrickling filters operated at a neutral pH by automated caustic soda addition, for simultaneous removal of  $\text{H}_2\text{S}$  and VOCs. A pilot-scale biotrickling filter at the County Sanitation Districts of Orange County removed greater than 87% of  $\text{H}_2\text{S}$  but removal of VOCs was disappointingly low at 11% (Torres and Basrai, 1998). Low VOC loadings and frequent systems upsets were the presumed causes for poor VOC removal. Nevertheless, this study shows the potential of pH-neutral biotrickling filters for the simultaneous removal of  $\text{H}_2\text{S}$  and VOCs.

## 4. Factors Influencing Biotrickling Filter Performance

### 4.1 TEMPERATURE

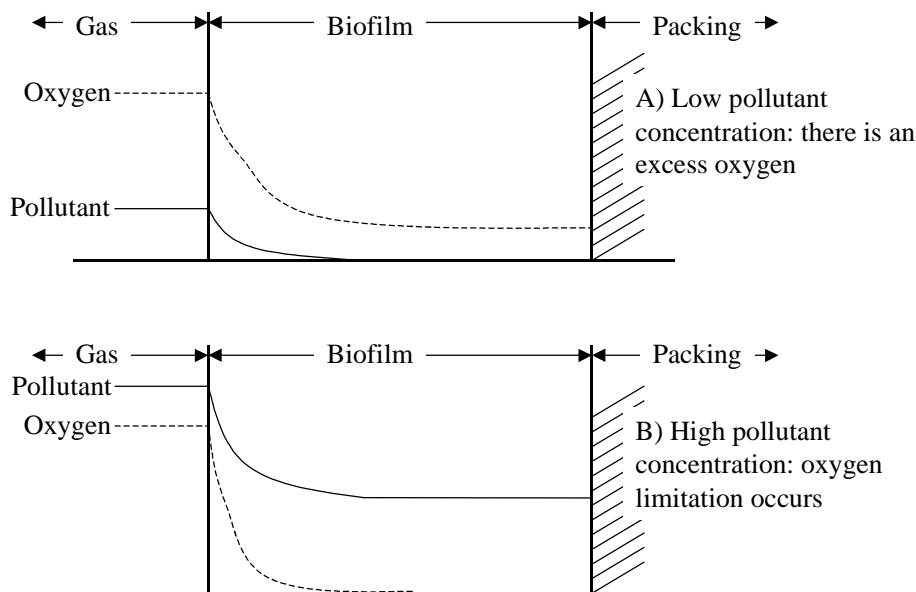
Most biotrickling filters in industry are subject to changing temperatures throughout the day as well as seasonal variations, which both are not experienced in the laboratory. Remarkably few studies have however investigated the influence of the temperature on the performance of biotrickling filters. In general, biotrickling filters are operated at temperatures between 10 and 40°C, which typically is the temperature range for growth of mesophilic microorganisms.

As discussed earlier, biotrickling filter performance is in general limited by the biological reaction rate or by the mass transfer rate. The temperature has a different effect on either limitation, consequently, temperature effects on the performance vary depending on the particular application. For mass transfer limited reactors, the effect of the temperature should be discussed with respect to the parameters that influence mass transfer. The driving force for mass transfer depends on the Henry coefficient, which will decrease with increasing temperature. On the other hand, the diffusion coefficient will increase, facilitating mass transfer inside the biofilm. These two effects may counterbalance each other. Thus, Diks and Ottengraf, 1991<sup>b</sup> observed no effect when the temperature was increased from 20 to 30°C in a biotrickling filter degrading dichloromethane. With the same pollutant, Hartmans and Tramper, 1991 also observed no changes between 18-30°C when their reactor was operated at a high gas flow rate and low pollutant concentrations (mass transfer limitation). Operation of the same reactor at a low gas flow rate and high pollutant concentration (biological reaction limitation), however, showed increasing performance at higher temperature. This was correlated to increasing activity of the microorganisms (Hartmans and Tramper, 1991). The influence of the temperature on microbial activity has been well studied and, in the sub-optimum range (~10-35°C), can in general be described by the Arrhenius equation. Obviously, temperatures beyond the optimum of the process culture will cause a decline of the performance and even cause cell death if the maximum temperature is exceeded.

Many waste gases in industry have temperatures beyond the mesophilic range, but treatment of those gases has received only little attention (Van Lith *et al.*, 1997). Cooling of hot waste gases would be a solution, but is expensive especially when the gas is saturated with water. The use of thermophilic microorganisms adapted to high temperatures is an interesting area, as it allows for treatment without prior cooling. As an example, effective removal of  $\alpha$ -pinene and methanol at biotrickling filter temperatures of respectively up to 60 and 70°C has been demonstrated (Allen *et al.*, 2000). In our own laboratory, we observed that overall performance of a thermophilic biotrickling filter treating ethanol vapours was comparable to that of a mesophilic control (Cox and Deshusses, 2000<sup>c</sup>). Operation at 53°C resulted in the selective enrichment of a thermophilic population. The ethanol removal rate was not affected by the temperature in the range of 50 to 62°C. Thermophilic treatment is expected to be a promising, new area of application of biotrickling filters.

## 4.2 OXYGEN

The oxygen concentration in most waste gases is in general several orders of a magnitude higher than the pollutant concentration. However, because of the low oxygen water solubility, biotrickling filter performance may be limited by mass transfer of oxygen into the biofilm and/or by diffusion in the biofilm. Kirchner *et al.*, 1992 and 1996 observed significant increases of the elimination capacity when the oxygen partial pressure in the gas was increased, thus demonstrating that at normal oxygen content, VOC removal was limited by oxygen availability. Oxygen limitation occurs when the penetration depth of oxygen is less than that of the pollutant, causing anaerobic layers in deeper parts of the biofilm close to the substratum (Figure 4.5). This has experimentally been confirmed using an oxygen microelectrode technique, showing depletion of oxygen at a depth of about 400  $\mu\text{m}$  into the biofilm of a biotrickling filter treating diethyl ether (Alonso *et al.*, 1998<sup>a</sup>).



**Figure 4.5.** Schematic representation of oxygen and pollutant concentration profiles in the biofilm.

Occurrence of oxygen limitation depends on the utilization rates of oxygen and the pollutant on one hand, and the respective diffusion rates in the biofilm on the other hand. This can, in a first approximation, be calculated using Equation 4.6 (Diks and Ottengraf, 1991<sup>a</sup>):

$$A = D_{\text{O}_2} \cdot v_{\text{poll}} \cdot C_{\text{O}_2,i} / D_{\text{poll}} \cdot v_{\text{O}_2} \cdot C_{\text{poll},i} \quad (4.6)$$

where  $D_{\text{O}_2}$  and  $D_{\text{poll}}$  are the diffusion coefficients of oxygen and of the pollutant in the biofilm,  $C_{\text{O}_2,i}$  and  $C_{\text{poll},i}$  are the concentrations of respectively oxygen and the pollutant at the biofilm interface, while the ratio  $v_{\text{poll}}/v_{\text{O}_2}$  (mg pollutant/mg oxygen) reflects the reaction stoichiometry. Oxygen limitation will occur when  $A < 1$ , whereas  $A > 1$  corresponds to the situation of pollutant mass transfer limitation. Equation 4.6 predicts that oxygen limitation occurs when the pollutant at the gas/biofilm interface exceeds a certain concentration, as

illustrated in Figure 4.5. Indeed, pollutant removal in biotrickling filters becomes more sensitive to oxygen-related parameters at high inlet concentrations of the pollutant (Mirpuri *et al.*, 1997<sup>a</sup>). The ratio of pollutant degradation to oxygen consumption ( $v_{\text{poll}}/v_{\text{O}_2}$ ) simply follows from reaction stoichiometrics. However, a factor often neglected is the oxygen consumption by the secondary population in the biofilm. It has been estimated these secondary processes may significantly contribute to the overall oxygen consumption rate in biofilms (Villaverde and Fernández, 1997; Villaverde *et al.*, 1997), but it is not known to what extent this may limit the removal rate of the primary pollutant.

A final note on the ratio of the interfacial concentrations  $C_{\text{O}_2,i}/C_{\text{poll},i}$ . In the past, it has been convenient to assume that a) no resistance occurred at the gas-liquid or gas-biofilm interface, and b) that the partition coefficient of the pollutant or oxygen was this of the partition with water. While the former assumption can easily be corrected using the two-film theory and calculation on individual mass transfer coefficients, the latter assumption may be more questionable. Recent work by Davison *et al.*, 2000 suggests that the partition coefficient of hydrophobic compounds like propane, but possibly oxygen, to biofilm differ considerably from the partition to water. While the reasons for the deviation are still unclear, this may explain why some hydrophobic compounds are better degraded than predicted and/or why oxygen limitation may not be as frequent as one would predict from Equation 4.6.

Anaerobic zones in the biofilm are unwanted for aerobic pollutant degradation, however, they may facilitate the biodegradation of compounds that require anaerobic conditions for biodegradation. For instance, the removal of PCE from aerobic waste gases in biofilters has been reported (Deviny *et al.*, 1995). To date, it is unclear how to best exploit anaerobic processes in mostly aerobic biotrickling filters.

### 4.3 PACKING MATERIAL

Many different sorts of packing materials have been tested in biotrickling filters. Important requirements to the packing include a large specific area, high porosity, high chemical stability and structural strength, low weight, suitable surface for bacterial attachment and growth, and low cost.

A commonly used packing material is lava rock (e.g., Morton and Caballero, 1996; Fortin and Deshusses, 1999). This material has the advantage of providing a large specific surface, a porous structure that facilitates colonization by microorganisms (Fortin Deshusses, 1999), and a low price. Disadvantages are the low porosity (~50%) and the heavy weight which requires special construction of the reactor. Another disadvantage we recently observed in the laboratory is that lava rock is not chemically inert. A substantial weight loss was observed over several months at a low pH. This might be of importance for biotrickling filter applications with a low pH, e.g., odour treatment and H<sub>2</sub>S removal at wastewater purification plants as discussed in Section 3.3.

Random-dump plastic packings such as Pall rings have been used in many laboratory studies and large-scale reactors. These packings are easy to handle, however, experiments indicate that startup is relatively long presumably because poor biofilm establishment on the surface (Fortin and Deshusses, 1999; Kazenski and Kinney, 2000). Also, the relatively low specific surface area is a disadvantage for achieving a high elimination capacity (Webster *et al.*, 1999). On the other hand, the use of plastic packing may be beneficial because of its stability, low cost and high porosity (Kazenski and Kinney, 2000). Structured packings made from stainless steel or plastic combine a high porosity and large specific surface. Good

performance has been observed with these type of packings (Zuber, 1995; Webster *et al.*, 1998), but they are more expensive.

Activated carbon-based packings have adsorbing properties that are not encountered in other materials. It is often assumed that adsorption is beneficial especially when the pollutant concentration in the waste gas is fluctuating ('peak-dampening'). However, activated carbon particles in biotrickling filters will be covered by a biofilm, which will decrease the absorptive capacity of the carbon. A better option would be using a separate activated carbon unit preceding the biotrickling filter (Weber and Hartmans, 1995).

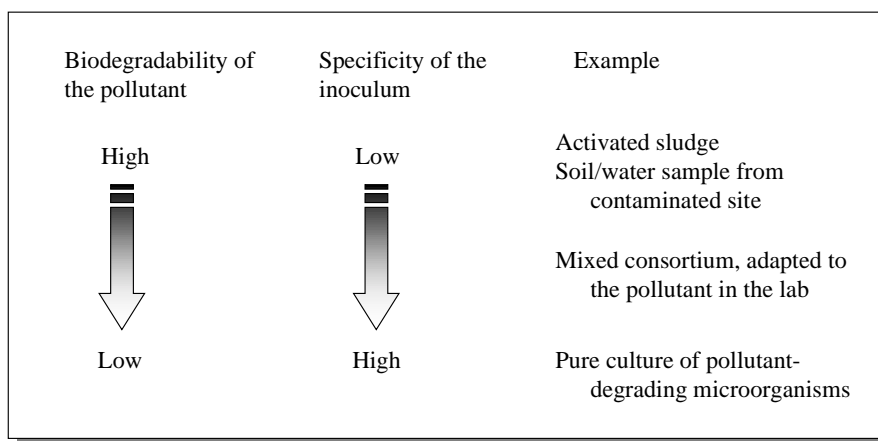
A relatively new packing used in biotrickling filters is open-pore polyurethane foam products. Published data on the performance are scarce, but some major European vendors offer these products, often for biotrickling filters designed for H<sub>2</sub>S removal. Our own experiences in the laboratory (unpublished) indicated improved performance with polyurethane foam cubes over other types of packing, especially at high gas flow rates with low H<sub>2</sub>S concentrations. Due to the open structure and high porosity, the pressure drop over the packing remained low at a relatively high gas velocity. The large specific surface area proved beneficial with respect to mass transfer limitation observed at low H<sub>2</sub>S concentration.

#### 4.4 INOCULATION AND MICROBIAL ECOLOGY

Unlike biofilters with an indigenous microbial population, biotrickling filters need to be inoculated with microorganisms. The following sources are commonly used:

- Activated sludge from wastewater treatment plants.
- Soil or water samples from sites or plants contaminated with the pollutant of interest.
- Consortia that are enriched in the laboratory on the pollutant of interest.
- Pure cultures, degrading the pollutant of interest and obtained either from culture collections or isolated from mixed consortia.
- Samples of biotrickling filters treating the same or a comparable waste gas stream.

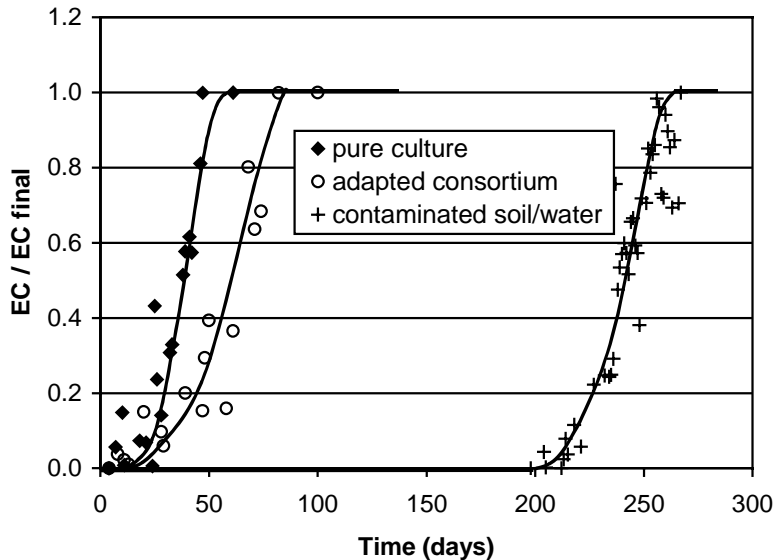
Selection of the inoculum source becomes increasingly important when the pollutant is more difficult to degrade (Figure 4.6).



**Figure 4.6.** Inoculation guide for biotrickling filters.

For easily biodegradable pollutants or for complex waste gases such as odours, a general source such as activated sludge is often sufficient. Activated sludge contains a wide

spectrum of bacteria, capable of degrading many different compounds, and is thus a good choice for odour-treating biotrickling filters. Use of adapted consortia or pure cultures with high biodegradation potential is favoured when treating gases containing poorly biodegradable pollutants. An example is the treatment of MTBE (Figure 4.7). Effective removal of MTBE after inoculation with exposed soil and water samples was observed only after seven months of operation. The startup phase could be significantly reduced by inoculation with an adapted consortium. Startup was the fastest when using a concentrated pure culture isolated in the laboratory and capable of degrading MTBE at a high rate.



**Figure 4.7.** Start-up of MTBE-degrading biotrickling filters inoculated with different sources of microorganisms (Cox and Deshusses, unpublished data for the pure culture and adapted consortium, Fortin and Deshusses, 1999 for the contaminated soil and water inoculum data).

As discussed in Section 2.1, a complex ecological community exists in biotrickling filters. A broad variety of bacteria, yeasts, fungi, protozoa, nematodes, algae and other higher organisms is generally observed (e.g., Hugler *et al.*, 1996; Schönduve *et al.*, 1996; Cox and Deshusses, 1999<sup>a</sup>). This can be attributed to various factors:

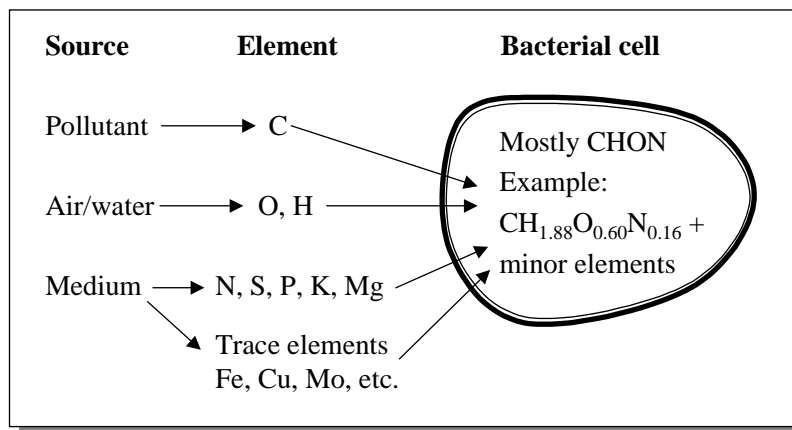
- Biotrickling filters are open systems, hence, invasion by and establishment of microorganisms and other organisms from the environment is unavoidable.
- Biofilm growth is coupled to the production of polymers, cell lysis products and metabolites from the primary pollutant-degrading population. These products serve as sources of carbon and energy for the secondary population. Analysis of the biofilm indicate that these alternative carbon sources may constitute a significant part of the biofilm (Arcangeli and Arvin, 1992; Pedersen *et al.*, 1997), which may explain the existence of a thriving secondary population in biotrickling filters. Apart from this, the cells in the biofilm (the primary and secondary population) itself may serve as food source for predating organisms such as protozoa (Cox and Deshusses, 1999<sup>a</sup>).
- Although biotrickling filters in mathematical modeling are considered to be homogenous reactors, the reality is quite different. Heterogeneity with respect to parameters affecting

microbial activity (e.g., pollutant, oxygen and nutrients concentrations, pH, metabolic products) may exist over the height of the reactor as well as locally inside the biofilm. Consequently, different microbial populations may develop depending on the local conditions. Biofilm heterogeneity in biotrickling filters has been demonstrated by various microscopical techniques (Hugler *et al.*, 1996; Møller *et al.*, 1996; Schönduve *et al.*, 1996; Pedersen *et al.*, 1997).

The use of molecular techniques has improved the understanding of the dynamics of microbial populations in biotrickling filters. The structure of microbial communities as determined by DNA finger-printing appears to be more complex than found with classical techniques such as plating of biofilm samples on microbiological media (Allen *et al.*, 2000). DNA finger-printing also allows for a comparison of microbial communities in different biotrickling filters and adaptations of communities to changing conditions in the reactor. Of particular interest are the studies of Pedersen *et al.*, 1997 and Møller *et al.*, 1996. They used epi-fluorescence microscopy and a 16S rRNA targeting probe to study the establishment and *in-situ* activity of a *Pseudomonas putida* species in the biofilm of a toluene-degrading biotrickling filter. One important outcome of these studies is that the specific activity of *Pseudomonas putida* in the biofilm was lower than as found in batch cultures with the pure species. This demonstrates the potential risk of overestimating the biological degradation rate in biotrickling filters if biokinetic parameters derived from batch or continuous cultures are used.

#### 4.5 NUTRIENTS

In order to maximize the pollutant elimination capacity, nutrients should be present at concentrations high enough to maintain an active, growing culture. In general, the nutrient requirement is qualitatively the same as the elemental composition of biomass. With the waste gas pollutant serving as a source of carbon and energy (i.e., VOCs), essential elements other than carbon should be supplied to the biotrickling filter (Figure 4.8). Biotrickling filter performance is maximized when these are added in excess, i.e., the pollutant in the waste gas is the limiting substrate.



**Figure 4.8.** Utilization of nutrients by heterotrophic bacteria with the pollutant as the carbon source for growth.

The amount of nutrients to be added can be estimated from biomass yield coefficients on each of the essential elements (Pirt, 1975). In the following calculations it is assumed that the waste gas pollutant is the carbon source for microbial growth. Since the pollutant is the limiting substrate, the amount of biomass formed is proportional to the amount of pollutant degraded. The first step is to calculate the expected biomass accumulation rate ( $X_{acc}$  in g dry biomass/m<sup>3</sup> reactor.h) from the desired or design value for the elimination capacity (g C-pollutant/m<sup>3</sup> reactor.h) and the biomass to carbon yield coefficient (g dry biomass/g carbon):

$$X_{acc} = EC \times Y_{X/C} \quad (4.7)$$

For a growing cultures,  $Y_{X/C}$  is approximately 1.0-1.15. As discussed below, the overall biomass yield of the mixed process culture in a biotrickling filter is lower because of predation by higher organisms and other secondary processes. To prevent limitation of growth by element E, the minimal supply rate ( $SR_E$  in g E/m<sup>3</sup> reactor.h) can be calculated according to:

$$SR_E = X_{acc}/Y_{X/E} \quad (4.8)$$

where  $Y_{X/E}$  is the biomass yield coefficient on element E (g dry biomass/g element E). Typical examples of biomass yield coefficients are listed in Table 4.4. Note that these are for pure cultures growing in bioreactors, recommended values for biotrickling filtration are probably close to those listed as the upper limit.

**Table 4.4.** Examples of biomass yield coefficients on some essential nutrients.

Element	$Y_{X/E}$ (g dry biomass/g element)	
	Lower limit	Upper limit
Nitrogen	8.3	20
Phosphorous	23	285
Potassium	25	111
Sulphur	38	3,300
Calcium	111	28,000
Iron	200	50,000
Copper	5,000	100,000

The above calculation assumes that nutrients supplied are irreversibly incorporated into a continuously growing primary culture. Cell death and lysis, and the presence of a secondary population and predators in the biotrickling filter is thus neglected. These processes result in nutrient-recycling, causing a larger availability of nutrients. The supply rate of nutrients can be decreased accordingly, but this is very difficult to quantify. Experiments with toluene-degrading biotrickling filters have indicated that nutrient recycling by secondary processes may reduce the required nutrient supply rate by a factor two (Cox and Deshusses, 2000<sup>c</sup>). Not unexpectedly, the recycle of nutrients by predation and secondary processes is irrelevant when excess nutrient is provided. Under these circumstances rapid growth of biomass and reactor clogging is often experienced. This is further discussed in Section 5.

For full-scale biotrickling filters it is often not practical or too expensive to feed a chemically defined medium. Alternative nutrient sources include agricultural fertilizers and

secondary effluent water from wastewater treatment plants. Although these sources may not have the most ideal composition, they have shown to be satisfactory in supporting sustained pollutant elimination in pilot-scale biotrickling filters (Webster *et al.*, 1999).

#### 4.5 LIQUID RECYCLING

A distinctive feature of biotrickling filters is the recycling of a liquid phase over the packing. The recycle liquid supplies water and nutrients to the biofilm and it removes metabolic products that would otherwise accumulate in the biofilm in possibly toxic concentrations. Continuous monitoring and adjustment of chemical parameters in the recycle liquid allow for better control of microbial activity in the biofilm, which is an important advantage of biotrickling filters over biofilters.

Biotrickling filters can be operated with the gas and liquid flowing cocurrently or countercurrently. Cocurrent operation is often advocated, as countercurrent operation would allow stripping of the pollutant from the liquid phase at the gas outlet side of the reactor, thereby causing a lower elimination capacity. This is merely a matter of theoretical concern. Experimental data indicate no major difference between either mode of operation (Diks and Ottengraf, 1991<sup>b</sup>). Moreover, stripping effects are likely to occur only in biotrickling filters overloaded with the pollutant, causing a high pollutant concentration in the liquid phase. In most field applications a near complete removal of the pollutant is usually the objective, in which case the pollutant concentration in the liquid phase will be negligible as well as possible stripping effects.

The rate of liquid recycling is often expressed as the superficial liquid velocity. Velocities typically range from 0.05 up to 20 m/h. Usually an increase of the elimination capacity is observed with a higher superficial liquid velocity. This has been attributed to an increase of the wetted biofilm surface area (Diks and Ottengraf, 1991<sup>b</sup>), or to a reduction of mass transfer resistance in the liquid phase (Hartmans and Tramper, 1991). Treatment of pollutants that release acid end-products is a case that requires special attention. At too low a trickle rate, the liquid may not be able to remove the produced acids fast enough so that the pH could be reduced to the extent of inhibiting microbial activity (Diks and Ottengraf, 1991<sup>b</sup>; Oh and Bartha, 1994). This situation can be assessed by calculating the expected pH at the bottom of the column using the trickle rate, the acid production rate and the trickling liquid buffer capacity. Alternatively, one could measure the pH in the liquid trickling from the bottom of the packed bed.

The upper limit of the superficial liquid velocity is determined by the flooding point of the reactor, which will decrease over time as biomass accumulates. The superficial velocity will also have an effect on the removal of hydrophobic pollutants. Thick layers of recycle liquid covering the biofilm may act as a barrier causing a reduced mass transfer rate of the pollutant from the gas phase into the biofilm (Zhu *et al.*, 1998; Nascimento *et al.*, 2000). This type of mass transfer resistance can be reduced by lowering the superficial liquid velocity. Also intermittent instead of continuous liquid recycling may improve the mass transfer of hydrophobic pollutants and improve their removal in biotrickling filters (Wolff, 1992; Pol *et al.*, 1998).

## 5. Biomass Growth and Long-Term Performance of Biotrickling Filters

With continuous supply of nutrients, the biotrickling filter will accumulate large amounts of biomass. At present, clogging of the reactor over the long run is one of the major obstacles at the industrial scale. This section describes the various stages of biomass accumulation, the consequences for the performance and long-term stability, and methods to control biomass growth.

### 5.1 BIOMASS GROWTH KINETICS AND POLLUTANT ELIMINATION

In general, pollutants are used by the primary culture to produce new biomass and to generate energy for maintenance activities of the existing biomass. The biotrickling filter process can at first instance be considered as a continuous culture (e.g., neglecting secondary processes, local heterogeneities, and mass transfer effects):

$$EC = (\mu/Y + m) \cdot X \quad (4.9)$$

$$\mu = \mu_{\max} \cdot S / (S + K_S) \quad (4.10)$$

Equation 4.9 describes the elimination capacity as a function of the amount of active biomass of the primary culture per volume of reactor ( $X$ ), the specific growth rate ( $\mu$ ), the maintenance energy requirement coefficient ( $m$ ) and the biomass yield on the limiting nutrient ( $Y$ ). Equation 4.10 is the Monod equation for growth on pollutant  $S$  as defined in Chapter 3, which can be extended to include oxygen limitation (i.e., the factor  $O / \{K_{S,O} + O\}$ ), nutrient limitations (element  $E$ ) other than the pollutant ( $E / \{K_{S,E} + E\}$ ) or the presence of inhibitors ( $I / \{1 + I/K_I\}$ ).

The population in biotrickling filters consists of different sub-populations, e.g., the primary culture, the secondary population, predators and other higher organisms. Biomass accumulation in the biotrickling filter is the addition of growth, cell death and lysis, predation by higher organisms and wash-out of detached biomass via the liquid purge each sub-population present in the system. This is shown in Equation 4.11, which expresses the overall biomass accumulation rate in the biotrickling filter as the sum of accumulation rates of sub-populations (e.g., primary culture, secondary population, predators) present in the biotrickling filter.

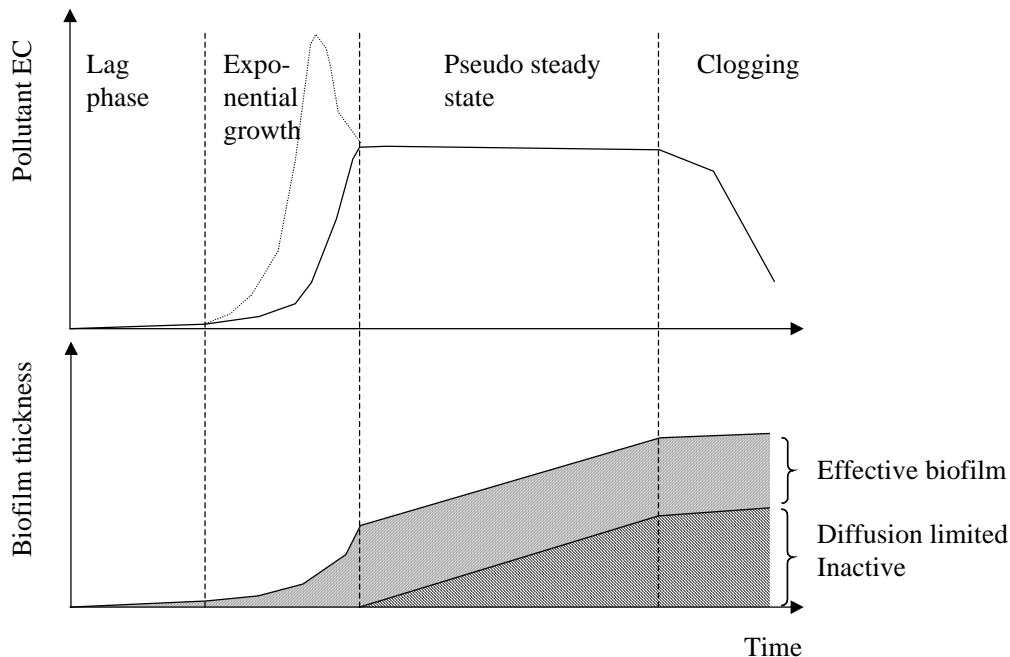
$$\text{Overall rate of biomass accumulation} = \sum \{(\mu-d) \cdot X - \text{predation} - \text{wash-out}\} \quad (4.11)$$

where  $d$  is the specific death rate of the sub-population. Predation and wash-out of biomass have received little attention in biotrickling filter research and these processes have not been quantitatively described as yet. Despite the limitation of being virtually impossible to solve, Equation 4.11 demonstrates that the overall biomass accumulation rate in biotrickling filters depends on the intrinsic microbial growth rate on the one hand, and various mechanisms of biomass reduction on the other hand. This allows for development of various strategies to control the accumulation of biomass, as discussed in Section 5.5.

## 5.2 STAGES OF BIOMASS FORMATION

Biotrickling filters are often operated with sufficient supply of nutrients to maximize the pollutant removal rate. Various stages of biomass accumulation can be distinguished after inoculation of the biotrickling filter with an appropriate source of microorganisms. At first, a lag phase will occur during which the microorganisms adapt to environmental conditions in the reactor and to the pollutant (Figure 4.9). The duration of the lag phase mainly depends on the biodegradability of the pollutant. Lag phases as short as a few hours to a few days have been reported for e.g. ethanol and toluene. On the other hand, removal of methyl-*tert*-butyl ether (MTBE), a gasoline additive with low biodegradability, may only be observed after several months of operation (see Section 4.4).

The exponential growth phase follows the lag phase (Figure 4.9), with rapid biomass accumulation and an exponentially increasing biofilm thickness (Pedersen *et al.*, 1997). Pollutant degradation takes place throughout the entire biofilm and the overall pollutant removal rate increases with the thickness. During this phase, a short-term peak in the elimination capacity can sometimes be observed. This has been linked to a rapid proliferation of suspended biomass in the recycle liquid (Cox *et al.*, 2000). Generally, the pollutant removal rate soon reaches a constant value (Figure 4.9, pseudo steady-state). At this point, diffusion limitation of the pollutant (Okkerse *et al.*, 1999), oxygen (Kirchner *et al.*, 1996; Mpanias and Baltzis, 1998) and/or nutrients (Zhu *et al.*, 1996; Rihn *et al.*, 1997) in the biofilm causes biodegradation to take place only in the upper layer at the gas/biofilm interface. Similar to fixed-film wastewater treatment processes, the effective biofilm thickness in biotrickling filters is as a rule of thumb in the order of 25-300  $\mu\text{m}$  (e.g., Kirchner *et al.*, 1996; Schönduvel *et al.*, 1996; Mirpuri *et al.*, 1997<sup>a</sup>).



**Figure 4.9.** Schematic of the long-term performance of biotrickling filters with continuous supply of nutrients. The upper section shows the development of the elimination capacity over time, whereas biofilm grown is shown in the lower section. (Note that the schematic is not to scale).

Prolonged operation with nutrient supply causes further growth of the biofilm, however, the effective biofilm thickness and the elimination capacity remain constant (Zuber, 1995; Cox and Deshusses, 1999<sup>a</sup>; Okkerse *et al.*, 1999). Prolonged operation therefore primarily results in formation of biomass not actively involved in biodegradation. Thick biofilms in biotrickling filters contain large fractions of inactive or dead cells and inert material (Schönduve *et al.*, 1996; Hugler *et al.*, 1996; Okkerse *et al.*, 1999; Pedersen *et al.*, 1997). Finally, biotrickling filters subject to excessive biomass formation (Figure 4.9, clogging) face clogging with increasing pressure drops across the reactor and a decreasing pollutant removal rate (Cox *et al.*, 1998; Cox and Deshusses, 1999<sup>a</sup>; Okkerse *et al.*, 1999).

### 5.3 BIOFILM ARCHITECTURE AND MASS TRANSFER

The gas/biofilm interfacial area is an important parameter in assessing the mass transfer in biotrickling filters. Usually a planar geometry is assumed, with a flat and homogeneous biofilm and a specific surface area the same as that of the supporting packing. Recently, a few innovative techniques have been developed that demonstrate the heterogeneous structure of the biofilm. Scanning confocal laser microscopy (SCLM) showed a porous biofilm with cell-free channels in a toluene-degrading biotrickling filter (Møller *et al.* 1996). One-dimensional scanning of the biofilm demonstrated a rough surface with significant variation in the local biofilm thickness (Okkerse *et al.*, 1998). Computed axial tomography was used to visualize *in situ* biofilms in toluene-degrading biotrickling filters (Deshusses *et al.*, 1998). This technique allows for quantification of the distribution and sizes of air and water channels and the determination of the surface area without disturbing the biofilm structure as is the case with most *ex situ* techniques. A large variation in pore diameter of air and water channels was found, and the surface area of the biofilm was larger than expected due to a rough surface.

Biofilm heterogeneity results in a greater area for mass transfer – and hence, in a faster mass transfer rate – than from planar biofilm geometry would be expected. This has been experimentally confirmed by Pedersen and Arvin, 1997<sup>a</sup>, who showed that the mass transfer coefficient ( $K_{La}$ ) in biotrickling filters was 25-140% larger when the packing was covered by a biofilm.

### 5.4 STEADY-STATE VERSUS NON-STEADY-STATE

Although many mathematical models are based on growing cultures according to a Monod-type equation, they frequently assume a steady state with no accumulation of biomass over time. This obviously is a weakness; steady-state models may accurately predict the performance at a certain point in time, but they fail to predict the long-term performance. The calculations shown in Table 4.5 demonstrate the importance of clogging. For a toluene-degrading biotrickling filter with an elimination capacity of 40 g/m<sup>3</sup>.h it may take only 11 weeks to produce an amount of biomass equal to the reactor volume. However, flooding may already be observed at biomass contents of about 50% by volume of the reactor (Cox *et al.*, 1998). Also as indicated by the calculation in Table 4.5, the pollutant load and elimination capacity as well as the biomass yield are the principal parameters in determining the biomass accumulation rate (Okkerse *et al.*, 1999; Cox *et al.*, 1998). The critical parameter is the biomass yield coefficient, which e.g. for pure cultures of *Pseudomonas* species with excess of nutrients has been found to range from 0.3 to 1.2 g biomass/g toluene (Mirpuri *et al.*,

1997<sup>b</sup>). Secondary processes in biotrickling filters – e.g., cell death and lysis, cryptic growth and predation by higher organisms – enhance nutrient-recycling, thus decreasing the actual growth yield and extending the operating life-time (Diks *et al.*, 1994<sup>a</sup>; Cox and Deshusses, 1999<sup>a</sup>).

Recently, dynamic mathematical models have been developed that account for biomass accumulation over time (Alonso *et al.*, 1997, 1998<sup>b</sup>). These models are useful in identifying the underlying cause of decreasing pollutant efficiency in clogged biotrickling filters. Although one would expect higher removal rates with increasing amounts of biomass (Equation 4.9), overall performance decreases because of diminishing accessibility of the pollutant to the biomass. With increasing thickness, biofilms on adjacent particles will overlap each other, thus decreasing the actual biofilm specific surface available for mass transfer into the biofilm (Alonso *et al.*, 1998).

**Table 4.5.** Example of the estimation of the biomass accumulation rate in biotrickling filters treating toluene vapours. The amount of biomass produced in 77 days is equal to the total reactor volume.

Toluene elimination capacity <sup>a</sup>	40 g toluene/m <sup>3</sup> reactor.h = 36.5 g C-toluene/m <sup>3</sup> reactor.h
Degree of carbon mineralization <sup>b</sup>	69%
C-toluene to C-biomass conversion ratio	31%
C-biomass accumulation rate	11.3 g C-biomass/m <sup>3</sup> reactor.h
Dry biomass accumulation rate <sup>c</sup>	25.7 g dry biomass/m <sup>3</sup> reactor.h
Wet biomass accumulation rate <sup>d</sup>	559 g wet biomass/m <sup>3</sup> reactor.h ~0.013 m <sup>3</sup> biomass/m <sup>3</sup> reactor.day

<sup>a</sup> Average value from Pedersen and Arvin, 1997<sup>b</sup>. <sup>b</sup> Cox *et al.*, 1998. <sup>c</sup> Carbon content in dry biomass of 44% (Cox and Deshusses, 1999<sup>a</sup>). <sup>d</sup> Water content in wet biomass of 4.6% (Cox and Deshusses, 1999<sup>a</sup>).

## 5.5 PREVENTION OF CLOGGING

The first option to prevent clogging is reduction of the biomass accumulation rate (Equation 4.11). According to Equations 9 and 11, this can be achieved by reducing the biomass yield coefficient ( $Y_{X/S}$ ), by increasing the maintenance requirements ( $m_s$ ) or by stimulating predation or the death rate. The challenge is to maintain a high pollutant removal rate (Equation 4.9), however, growth, yield, activity and maintenance are interrelated parameters reflecting general cell metabolism and, as such, they are often difficult to influence independently.

Various strategies to reduce the specific growth rate in biotrickling filters have been investigated in the laboratory (Table 4.6). These include limiting the supply of nutrients essential for growth, the use of nitrate as a nitrogen source instead of ammonium and the addition of compounds such as NaCl in concentrations that partially inhibit microbial growth. In general, these strategies also result in reduction of microbial activity. Hence, a relatively large reactor volume will be required to treat the same volume of waste gas at the same efficiency. An interesting option is the use of fungi, as these type of microorganisms show a higher removal rate than bacteria under nutrient-limiting conditions in toluene-degrading biotrickling filters (Weber and Hartmans, 1996). Predation of biomass by higher organisms such as protozoa is a relatively unexplored area in biotrickling filter research, although the phenomenon is quite known in wastewater treatment systems. Protozoa are natural inhabitants of biotrickling filters, and stimulation of their activity was showed to

**Table 4.6.** Control of biomass accumulation in biotrickling filters.

Biomass control option	Principle	Disadvantage	References
<i>Reduction of the biomass accumulation rate</i>			
Nutrient limitation	Reduction of the biomass yield coefficient	Reduction of the specific microbial activity	Holubar <i>et al.</i> , 1999; Weber and Hartmans, 1996; Cox <i>et al.</i> , 1998 Wübker and Friedrich, 1996
N-NO <sub>3</sub> <sup>-</sup> instead of N-NH <sub>4</sub> <sup>+</sup>	Reduction of the biomass yield coefficient	Reduction of the specific microbial activity	Schönduve <i>et al.</i> , 1996; Smith <i>et al.</i> , 1996
Addition of growth inhibitors	Reduction of the biomass yield coefficient	Reduction of the specific microbial activity	Schönduve <i>et al.</i> , 1996; Diks <i>et al.</i> , 1994 <sup>b</sup>
Use of specific microbial species	Selection of species with low biomass yield and high activity	Selective advantage for species with high biomass yield	Weber and Hartmans, 1996
Protozoan predation	Reduction of the biomass yield coefficient	Protozoan predation does not fully balance microbial growth	Cox and Deshusses, 1999 <sup>a</sup>
Periodical removal of excess biomass			
Backwashing	Biomass removal	40% larger reactor volume needed for full packing fluidization. Requirement for packing media that can be fluidized	Sorial <i>et al.</i> , 1995; Smith <i>et al.</i> , 1996; Smith <i>et al.</i> , 1998
Periodical stirring	Biomass removal	Complicated reactor design and construction.	Laurenzis <i>et al.</i> , 1998; Wübker <i>et al.</i> , 1997
Chemical washing	Biomass removal	Toxicity to microorganisms. Secondary wastes.	Weber and Hartmans, 1996; Cox and Deshusses, 1999 <sup>b</sup>

reduce the biomass accumulation rate (Cox and Deshusses, 1999<sup>a</sup>). The advantage of predation over options mentioned earlier is that the process culture is allowed to grow at the maximum rate, and hence with maximum microbial activity. In fact, stimulation of protozoan predation caused a slight increase of the maximum elimination capacity, which was tentatively attributed to an increased rate of nutrient recycling (Cox and Deshusses, 1999<sup>a</sup>). The predation rate was however not high enough to fully counterbalance microbial growth, and only delaying clogging of the reactor could be achieved.

The second option to prevent clogging is periodical removal of the produced biomass. In this scenario, the biotrickling filter is operated at a high elimination capacity allowing rapid accumulation of biomass. Removal of biomass can be done physically or chemically (Table 4.6). Physical removal of biomass relies on biofilm detachment by high shear forces. This can be done by backwashing of the reactor, or by periodical stirring of the packed bed. Although these techniques result in prolonged, stable biotrickling filter operation, certain drawbacks exist (Table 4.6). It should be noted that shear stress of the trickling liquid during normal operation of the biotrickling filter is not sufficient to remove substantial amounts of attached biomass (Pedersen *et al.*, 1997; Cox and Deshusses, 1999<sup>b</sup>). Chemical removal of biomass is a simple operation as no major changes of the reactor configuration are required. A stable toluene-degrading biotrickling filter has been obtained by periodical washing of the packing with a NaOH solution for 3 hours (Weber and Hartmans, 1996). A post treatment with HCl was needed to restore the pH to a neutral value. Other chemicals such as sodium hypochlorite and hydrogen peroxide may be more effective in removing biomass, but they are also more toxic to the microbial population (Cox and Deshusses, 1999<sup>b</sup>). This could potentially slow down the restart of the reactor.

It is worth to stress that the biomass control strategies discussed in this Section have only been investigated in the laboratory. Very little experience is available at industrial scale, because the few existing full-scale biotrickling filters have all been designed for applications with a low potential for clogging. Cost-effective, long-term operation of biotrickling filters requires finding the optimum between two extreme cases (Deshusses and Cox, 1999):

1. Operation of large-volume, low-performance biotrickling filters that do not or only very occasionally require biomass removal.
2. Operation of small-volume, high-performance biotrickling filters with frequent biomass removal.

It may be expected that with further understanding of microbiological processes in biotrickling filters solutions will be found that combine high microbial activity and a low biomass accumulation rate.

## 6. Conclusion

Biological wastewater treatment has become part of our everyday life. Although, the process is not fully understood, wastewater treatment is a major improvement for water quality. More than 100 years after the introduction of the first wastewater treatment plants, bioreactors for the treatment of gaseous effluents have the potential to play a similar role for air quality. In the past decades, major progress has been accomplished in the development of vapour-phase bioreactors, in particular in biotrickling filters. While the level of understanding of the biotrickling filtration process still remains limited, the evident success

of wastewater biotreatment should be a motivation to pursue active research. Clearly, the full potential of biotrickling filtration for air pollution control has not yet been explored. There is great promise for both effective and environmentally friendly biotreatment of contaminated gases.

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