Biotechniques for air pollution control: past, present and future trends

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ABSTRACT. Biofilters were first constructed in the nineteen fifties, but the application of biological waste gas treatment technologies first became popular in the nineteen eighties due to air pollution legislation. Simultaneously, R&D in this field boosted. In the last 15 years a range of new technologies was developed to extend the operational envelop of biological waste gas treatment and to solve operational problems. The author's prediction is that the markets for biofilters, biotrickling filters and bioscrubbers will all grow, however, the relative share of bioscrubbers will decrease and that of biotrickling filters will increase. A trend is the use of biofilters that have properties in between biotrickling filters and biofilters (frequent water addition but not continuous, inert packing material, nutrient addition). More attention will be given to robustness and long term operational stability. The problem of filter clogging still needs a good solution, therefore R&D on this matter (although an old subject) is still justified. Another trend is the diversification of equipment for specific gas streams. Biofilters will not just be copied to treat new gas streams, but increasingly designed taylor made. Within 3 years, Asia will be the most important market.

1 HISTORY (1920 – 1990)

In 1923 Bach proposed to use a biological filter for the treatment of odorous gases from a wastewater treatment plant, but the first evidences of the application of such biofilters only appeared in the nineteen fifties when soil based biofilters were used to treat gases from untreated sewage at sewage treatment plants in the USA (Pomeroy, 1957). In the nineteen sixties soil biofilters were introduced in Europe, e.g. for the treatment of odorous gas from composting plants (Ottengraf, 1986). In 1971, Frechen used a biofilter based on compost as polishing step after a scrubber to treat air from a composting works in Duisburg (Ottengraf, 1986). From that time the numbers of biofilters installed slowly increased, most in Germany but also in the USA and the Netherlands. In the nineteen seventies biofilters were also installed in German animal rendering plants and intensive stock breeding. The first biofilter in the Netherlands was installed in 1978 (Cornelisse *et al.*, 1979).

In 1973 Thistlethwayte and coworkers described a column filled with air as the continuous phase and packed with river gravel or glass balls that was trickled with a

nutrient solution and inoculated with activated sludge. In this pioneer laboratory scale biotrickling filter waste air was flowing in upward direction.
Presumably the first bioscrubbers were installed in the nineteen seventies in German foundries for the removal of amines, phenol, formaldehyde and odour from waste gases (Fischer, 1990; Schippert, 1994).
An early report on the use of peat in German biofilters is from 1977 (Zeisig *et al.*, 1977). Mixtures of compost, peat and $CaCO_3$ (Cornelisse *et al.*, 1979) were used in Dutch wastewater treatment plants. In that period heather was used as well. Patents on closed biofilters (Figure 1) are from 1976 and 1978. These inventions were driven by the need to keep the biofilters compact and free of weather influences (as experienced in the open biofilters).

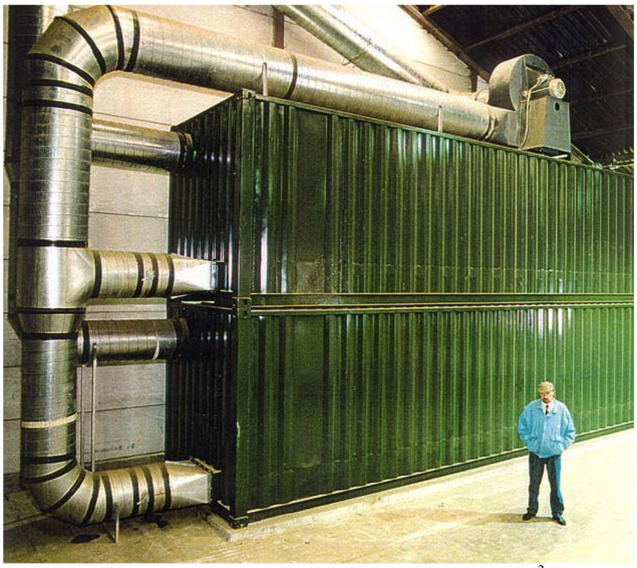

Figure 1. Two-story closed biofilter for treatment of 20,000 Nm^3 waste gas/h.

Due to the introduction of air pollution legislation in Germany and the densely populated Netherlands, in the nineteen eighties, the number of biofilters installed exponentially increased in these countries. The cost-effectiveness of these systems as compared with physico-chemical treatment technology was recognized. In this decade also R&D in this area boosted. That time, compost based biofilters only treated 25 m^3 air/m^3 filterbed/h. The need to combine high activity and low gas pressure drops led to the addition of support materials to compost. In the Netherlands, TNO developed a mixture of course and fine fractions from sieved compost (Don and Feenstra,1982) and later, in collaboration with composting company VAM, a mixture of compost and composted wood bark. Gas loading rates of 200 $m^3/m^3/h$ were in principle possible in such systems, but to reach an acceptable elimination efficiency 100 $m^3/m^3/h$ was recommended, a design criterion that became a standard. The Technical University of Eindhoven developed a mixture of compost and polystyrene particles (Ottengraf, 1983). Today, mixtures of compost and coarser material still are the most used packing material for biofilters.
In the nineteen eighties, international biofilter conferences were organized, mathematical models for biofilters were made, more insight in the microbiology of

degradation of xenobiotics was made available, special biotrickling filters, bioscrubbers and biofilters were developed for intensive stock breeding (ammonia), for gases loaded with volatile solvents, and for gases containing H_2S and organosulfur compounds, more knowledge on the effect of operational parameters on performance was gained, the microbiology was studied, design methods were made public, multi-story biofilters were introduced, and more types of packing material were tried (e.g. wood pieces). In addition, new concepts such as bubble columns, membrane bioreactors for gas treatment, and the use of bioscrubbers with two liquid phases (organic/aqueous) were tried. At the end of the nineteen eighties, biological waste gas treatment had become a science and the technology was not solely applied for odour treatment at sewage, composting, and rendering works and intensive stock breeding anymore. It also was introduced in food and chemical industry, foundries, wood processing, surface coating and kitchens.

2 DEVELOPMENTS (1990 – 2005)

2.1 General

Around 1990 the optimism about the possibilities of biological waste gas treatment reached its top. In these years, more feed back was given by biofilter users, and the reports were not always positive. Biofilters based on compost/wood bark sometimes suffered from operational instability, hydrophobic compounds were not removed very well and highly loaded biofilters and biotrickling filters eventually clogged. As a result, efforts were made to solve these problems. In the nineteen nineties R&D budgets were increased and mainly spent on (1) solving operational problems and improving existing concepts, and (2) extending the operational envelop of biological waste gas treatment (hot gases, flue gases, indoor air, hydrophobic compounds, poorly biodegradable compounds). A few interesting examples are given below.

2.2 Biotrickling filters

In the last 15 years a long list of reports on elimination of individual compounds, defined mixtures and compounds from complex waste gases by biotrickling filters has been published. The use of biotrickling filters for removal of H_2S, organo-sulfur compounds and NH_3 seems most successful, because pH and other reaction conditions can be controlled very well. Clogging is not a problem in these applications, because of the use of autotrophic micro-organisms (which have a low biomass yield factor). Biotrickling filters based on heterotrophic micro-organisms, however, are used for high concentrations of organic compounds, in particular chlorinated hydrocarbons (because of pH control). In principle such biotrickling filters can work with high volumetric elimination capacities, but after a few months of operation this leads to clogging of the filterbed with biomass. In nineteen nineties people have tried to solve this problem using various methods: decreasing growth yield using potassium or phosphorus limitation, or high salt concentrations, predation by higher organisms, removing biomass by washing with caustic soda or by mechanical forces. The author has the impression that an elegant method still not has been found.

2.3 Thermophilic biofiltration

In the mid-nineteen nineties trials were carried out with lab- and pilot-scale biofilters at temperatures between 50 ºC and 70 ºC (Heslinga and Groenestijn, 1997). Thermophilic biofilters contain thermostable packing materials and are inoculated with thermophilic micro-organisms. These biofilters can eliminate the same types of volatile compounds as non-thermophilic biofilters apart from a few exceptions (e.g. ammonia) (Heslinga and Groenestijn, 1997). In 1997 a full scale 60 ºC thermophilic biofilter was installed at a cocoa factory. Odor elimination efficiencies as high as 97% were attained and fat aerosols did not clog the filter bed as the fat stayed liquid and flowed down the bed. Cox *et al.* (2001) tested a laboratory scale biotrickling filter for removal of ethanol from air at 53ºC. Elimination capacities as high as 220 g ethanol/m^3/h were possible, while the low biomass yield factor at this temperature helped to limit biomass accumulation. A thermophilic bioscrubber was installed at a wood plate factory (Sperka and Dussing, 2003). The installation in which mist was sprayed in an upward direction contained no packing media and removed formaldehyde, organic acids and wood particles. The inlet air stream had temperatures up to 65°C and a flow rate of 400,000 m^3/h. The installation has successfully operated since 1999.

2.4 Biofilters at municipal wastewater treatment plants

To reduce odour emission from municipal wastewater treatment plants, tanks containing untreated sewage or sludge are covered and ventilated. The ventilation gas is treated. In the year 2000 an inventory in the Netherlands on biological waste gas treatment at these plants was conducted (Stowa, 2000). 80 – 90% of the municipal wastewater treatment plants used gas treatment systems. Of these systems 78% were biological systems, 11% chemical scrubbers and 2% activated carbon adsorbers and 9% treated the odorous gases by introducing these in the aeration tank (scrubbing). Four types of biofilter packing materials could be found: lava rock (38% of the cases), coconut fibre (31%), compost (30%) and synthetics (1%). Lava biofilters were introduced in the first half of the nineteen nineties. For new wastewater installations chemical scrubbers are rarely used anymore. Over the last 10 years gas treatment has become obligatory for all new Dutch sewage plants and compost-based biofilters are being replaced by lava rock biofilters or synthetic packing biofilter systems. These synthetic biofilter systems have also been installed during the last couple of years in other parts of the world (Van Durme *et al.*, 2002; Kraakman, 2004). In particularly the USA, waste air sometimes is treated by introduction in the activated sludge basin via the bubble aeration system.

2.5 Biofilters based on the action of fungi

Biofilters in which fungi are the dominant type of biomass have been developed to overcome a few typical problems. Fungi are more resistant to acid and dry conditions and take up hydrophobic compounds from the gas phase more easily than wet bacterial biofilms. Fungi based biofilters have been developed to remove styrene (Cox, 1995), toluene, xylene, ethylbenzene (Kennes *et al.*, 1996; Groenestijn *et al.*, 2001), ethane, 1,3-butadiene (Groenestijn *et al.*, 1995), alpha-pinene (Groenestijn and Liu, 2002) and hexane (Spigno *et al.*, 2003). At elimination capacities higher than 100 g/m^3/h clogging of the filterbed with fungi can occur. To solve this problem, mites (Figure 2) can be introduced that predate the fungi (Woertz *et al.*, 2002).

Figure 2. Mites as predators in a fungal biofilter (photograph made by Wageningen University).

2.6 Systems with two liquid phases

To treat gases that contain extremely hydrophobic compounds, such as alkanes, systems with two liquid phases (aqueous and oil) seem to work. Poppe and Schippert (1992) used bioscrubbers with silicon oil and water to remove hexane from gases, while Cesario *et al.* (1992) used bubble columns (air lift loop reactors), Budwill and Coleman (1997) used biofilters in which silicon oil was poured and Groenestijn and Lake (1999) used biotrickling filters.

2.7 Bioscrubbers to remove H_2S from biogas and natural gas

Bioscrubbers that remove H_2S from air or anaerobic gases (natural gas, biogas) have been developed during the last 15 years. In these systems H_2S is absorbed into alkaline water using a scrubber. The water is subsequently transported to an aerated bioreactor in which a *Thiobacillus* species converts sulphide into elemental sulphur. The sulphur and sludge is separated from the water and the water is returned to the scrubber (Dijkman, 1995; Janssen *et al.*, 1999, 2000). The sulphur can be used in agriculture. Full-scale bioscrubbers for biogas treatment were installed worldwide during the last 6 years. A first full scale application for high pressure natural gas was started in Canada in 2001.

2.8 Flue gas

Bioscrubbers for the removal of SO_x from flue gas are described by Cetinkaya and coworkers (2000). SO_2 is absorbed in water containing sodium bicarbonate yielding sodium bisulphite. In a first bioreactor this bisulphite is biologically reduced to sulphide using hydrogen gas as an electron donor. In a second bioreactor sulphide is oxidized into elemental sulphur under controlled aeration.

NO_x can be removed from flue gas by bioscrubbers as well. In the absorbers NO_2 is dissolved in water and NO reacts with Fe(II)EDTA to form a complex. In the anoxic bioreactor biological denitrification takes place using an electron donor such as ethanol, yielding dinitrogen gas as a product (Cetinkaya *et al.*, 2000). No full scale bioscrubbers have been constructed yet. Other researchers have tested biofilter and biotrickling filters in which NO_x is oxidized (nitrification) or reduced (denitrification). Such denitrification takes place in case also electron donors are added (e.g. VOCs).

2.9 Biofiltration of indoor air

In 1994 experiments with breathing-wall biofilters have taken place in Canadian offices. These indoor biofilters were made of porous, constantly wetted lava-rock panels covered with mosses and maidenhair ferns. Fans drew air through this wall and water from it circulated into a terrestrial zone and aquarium and subsequently back to the top of the wall. The system was tested with a range of VOCs introduced in the office air (Anonymous, 1999). Membrane bioreactors in which indoor air is contacted with a wet biofilm have been tested in space (in space station MIR and space shuttle Columbia) by the European Space Agency.

2.10 Other developments

- Alternative technologies to contact polluted gas with micro-organisms in aqueous phases have been tried. Examples are fog and foam in reactor vessels. The advantages of such systems without packing materials are savings on packing and the absence of clogging. The disadvantage is the complicated way the water phase has to be created in and removed from the gas phase. Another example are rotating biological contactors in which disks covered with biofilms rotate in a reactor half filled with an aqueous phase and half with the polluted gas phase.
- Cometabolic bioscrubbers. Compounds such as trichloroethylene can not be biodegraded under aerobic conditions, however, when adding phenol as a cosubstrate degradation takes place.
- The range of packing materials extended by e.g. activated carbon granules, pelletized compost, perlite, expanded clay, polyurethane foam cubes, inert types of wood bark, polyvinyl alcohol / peat composite beads, coconut fibres and textile. Addition of nutrients is required to make certain types of materials fit as a substrate for biofilm growth.
- Combinations of advanced oxidation, such as by UV-radiation or non thermal plasma, and biofiltration have been demonstrated. Compounds poorly biodegradable or poorly soluble in water can be partly oxidized physico-chemically to make them degradable and hydrophilic, which makes biofiltration feasible again.

2.11 Different development in world regions

Since a few years in the Netherlands the market is saturated and Dutch vendors are mainly exporting biofilters, while R&D efforts have decreased. In Germany there is still room for growth, particularly in Eastern Germany. The introduction of biofilters in other countries has been slower, with the USA as the most active place (last 15 years). In the nineteen nineties, next to the introduction of biofilters and biotrickling filters, the USA increased their R&D efforts to high levels. Simultaneously with the fast economic development of Asia, the last 5 years, a considerable number of R&D papers on biological waste gas treatment was generated by Asian countries. In China odour removal at wastewater treatment plants is the most important application of biofilters. For example, at the moment of writing this paper, 5 of the 20 domestic wastewater treatment plants in Beijing use biofilters to reduce odour emission (personal communication Prof. Junxin Liu). In addition, biofilters remove odours from Chinese public toilets and garbage collection houses. Most Chinese biofilters contain compost/wood bark mixtures, but even soil biofilters are still newly constructed. In

China a trend is about to start to find alternatives for compost/wood bark, due to disappointing performance.

2.12 Conferences

In the last 18 years biofilter experts were able to meet at international conferences. The German VDI and DECHEMA and the Dutch Vereniging Lucht organized meetings in 1987, 1989, 1991, 1994, and 1997, while VDI is still organizing meetings with a semi-international character. Los Angeles and surroundings is the place for the 2-yearly USC-CSC-TRG conferences on biofiltration for air pollution control, while the USA Air and Waste Management Association has annual conferences at which biological air treatment is a part of the programme. In 2001 and 2003 the IWA (International Water Association) organized conferences on odour and VOCs with 25% of the programme dedicated to biological treatment technologies. ISEB (Europe) and Forum for Applied Biotechnology (Belgium) always had biofiltration sessions.

2.13 Books

Specialised books on biological air treatment have been published over the past 6 years. Examples can be found in the list of references under Devinny *et al.* (1999) and Kennes and Veiga (2001). The German VDI has published guidelines for biofilters (VDI 3477, 2004) and for bioscrubbers and trickle bed reactors (VDI 3478, 1996). The latter is now being revised and a new version will be published in 2006.

3 PRESENT SITUATION

The author estimates the number of biological filters and scrubbers for gas treatment installed worldwide at least 15,000, of which half at wastewater treatment plants and composting facilities. More than half of this number can be found in Europe. Biofilters are more popular than biotrickling filters, while bioscrubbers can only be found in a small niche markets. The market for biological waste gas treatment is only a few percent of the total market for gas treatment, which is dominated by chemical scrubbers. Problems with operational stability, performance predictability and reproducibility of biological systems still cause some buyers to choose for physico-chemical gas treatment even when the latter is more costly. The situation has been much improved the last 10 years, but more effort is required. The process is hampered by the fact that there are large quality differences amongst biofilter vendors. To cope with that problem a quality certificate has recently been introduced in Germany. In the Netherlands such certificate only exists for bio(trickling) filters in intensive stock breeding.

The activities in the field of biological waste gas treatment are very fragmented: there are many small groups. It is estimated that a few hundred suppliers are active worldwide. There are only a few dedicated companies (only selling biological filters), with not more than 30 employees per company. Most suppliers are part of larger companies in environmental technology. Companies specialized in large plastic products (made of GRP and HDPE) sometimes have absorbed biofilter expertise and started to sell biofilters. Consulting companies mostly have not more than 1 to 2 experts on biological waste gas treatment. The number of university and institute research groups in this field probably is between 100 and 150. Table 1 gives an impression of the location of research groups known by the author. These groups are mostly small as compared with groups active in e.g. biological wastewater treatment. In addition, R&D takes place at companies.

Table 1. Locations of university and institute research groups on biological waste gas treatment (not complete).

Country	City, town, state
Australia	Sydney
Austria	Graz, Schwechat, St. Pölten, Vienna
Belgium	Ghent
Canada	Guelph, Laval, Kingston, Manitoba, Montreal, Ottawa, Sainte-Foy, Sherbrooke, Toronto, Vancouver, Vegreville
China	Beijing, Hangzhou, Nanjing, Shanghai, Tianjin
Czech Republic	Prague
Denmark	Lyngby, Tjele
Finland	Espoo, Kuopio
France	Alès, Nantes, Toulouse
Germany	Braunschweig, Dortmund, Essen, Frankfurt, Freiberg, Hamburg, Munich, Münster, Oberhausen, Osnabrück, Stuttgart, Wiesbaden
India	Madras, Roorkee
Iran	Teheran
Italy	Genova, Piacenza, Rome
Japan	Fujisawa-shi, Yokohama
Latavia	Jelgava, Riga
Mexico	Mexico City
Korea (South)	Busan, Dae-Jeon, Seoul
Russia	Moscow
Singapore	Singapore
Spain	Barcelona, La Coruña, Manresa, Tarragona
Sweden	Lulea, Växjö
Switzerland	Lausanne, Zurich
Taiwan	Hsin Chu, Kaoshiung, Taichung
The Netherlands	Apeldoorn, Nijmegen, Wageningen
USA	Amherst, Anderson, Austin, Baton Rouge, Cincinnati, Clemson, Cleveland, Davis, Fayetteville, Idaho Falls, Las Cruces, Los Angeles, Newark, New Brunswick, Notre Dame, Oak Ridge, Pittsburgh, Riverside, Tennessee, Worcester

4 THE FUTURE

The markets for biofilters, biotrickling filters and bioscrubbers will all grow, however, the relative share of bioscrubbers will decrease and that of biotrickling filters will increase. A trend is the use of biofilters that have properties in between biotrickling filters and biofilters (frequent water addition but not continuous, inert packing material, nutrient addition). The driver for this process is to have more control of the treatment process. More attention will be given to robustness and long term operational stability. Good manuals for operators will play a more important role and these manuals will be more specific for the type of gas and type of equipment. This development will be supported by R&D on the effect of anomalies (such as interruption of gas flow and water flow, and fluctuations of many parameters) on biofilter performance. The problem of filter clogging still needs a good solution, therefore R&D on this matter (although an old subject) is still justified.

Another trend is the diversification of equipment for specific gas streams. Biofilters will not just be copied to treat new gas streams, but increasingly designed taylor made. This trend is caused by the increasing knowledge on the interaction between gas properties and biofilter properties and on a developed customer demand. Such diversification will

manifest in specific micro-organisms used, combinations of treatment technologies (trains of unit operations), packing materials used and water addition regimes.
Market researcher McIlvain Company has estimated the market for biofiltration in 2007. China may have the largest market (135 million US$, which is 2% of the Chinese market for air pollution control equipment), followed by the USA (94 million US$) and further in order of decreasing market size by Japan, South Korea, Germany, France, UK, Italy, Canada and Taiwan. Odour control at wastewater treatment plants will continue be the most important application.

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