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Review

Recent developments in biological waste gas purification in Europe

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Abstract

Polluted air has become an increasing environmental and health concern. Legislation controlling the emissions of air pollutants (VOC, toxics and odor) has proliferated. In recent years, biological techniques have been applied more frequently to control these emissions, because they eliminate many of the drawbacks of classical physical–chemical techniques. Different waste gases require different strategies for optimal and sustainable purification. Biological treatment provides an expanding variety of opportunities for economical and environmentally friendly solutions for many waste gas emissions. In Europe, a significant body of knowledge and experience has been generated on biological waste gas purification. Examples of some interesting developments and new applications of biological waste gas treatment systems in Europe during the last 5 years are presented in this paper.

Keywords: Biological treatment; Air pollutants; Waste gases; Biofiltration

1. Present situation in Europe

The Netherlands and Germany were the first countries in which large numbers of biological waste gas treatment systems were constructed [1–3]. In the 1980s, the biofiltration market grew rapidly and the 1980s and 1990s were the golden era of R&D on biological waste gas technology in Europe. In recent years, biological techniques have been applied more frequently to control these emissions, because they eliminate many of the drawbacks of classical physical–chemical techniques. The disadvantages of the traditionally used air treatment techniques are high-energy costs (incinerators), the use of chemicals, which can be costly to purchase or dispose of and require special operational safety procedures (chemical scrubbers) and the production of waste products (spent chemical solutions or spent activated-carbon).

The authors estimate that at the moment there are probably over 7500 biological waste gas treatment systems and related systems installed in Europe, of which half are installed at sewage treatment and composting plants. A significant body of knowledge and experiences have been generated on biological air purification in Europe, and many improvements are still being made. Examples of some interesting developments and new applications of biological waste gas treatment systems are presented.

2. Thermophilic air treatment

In the mid-1990s, trials were carried out with laboratory- and pilot-scale biofilters at temperatures between 50 and 70 °C [4]. Thermophilic biofilters contain thermostable packing materials and are inoculated with thermophilic microorganisms. These biofilters can eliminate the same types of volatile compounds as non-thermophilic biofilters apart from a few exceptions (e.g., ammonia) [4]. In 1997, a full-scale 60 °C thermophilic biofilter was installed at a cocoa factory. The biofilter had a volume of 13 m^3 and was loaded with a gas flow rate of 1000 m^3 wastegas/h. The gas was prehumidified before entering the biofilter. Odor elimination efficiencies as high as 97% were attained and fat aerosols did not clog the filterbed as the fat stayed liquid and flowed down the bed (see Table 1). The lower efficiency of measurement 2 was probably caused by the

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Table 1
Removal efficiencies of a thermophilic biofilter at a cocoa factory

Measurement	Inlet odor concentration (OU_E/m^3)[a]	Outlet odor concentration (OU_E/m^3)[a]	Removal (%)
1	206000	7150	97
2	134000	66000	51
3	300500	14150	95

Source: TNO, The Netherlands.
[a] OU_E/m^3: odor units per cubic metric of air, measured according to the European standard EN13725.

chemical disinfection of the production facilities, which also affected the biofilter. Because of the satisfactory results, a second thermophilic biofilter was started in 1999 at the same company (unpublished results, TNO, The Netherlands).

A thermophilic bioscrubber was installed at a wood plate factory [5]. The installation, operated in downflow and in which mist is sprayed in an upward direction, contains no packing media and removes formaldehyde, organic acids and wood particles. The inlet airstream has temperatures up to 65 °C and a flow rate of 400,000 m^3/h. The installation has successfully operated since 1999.

Table 2
Removal efficiencies of odors of a multi-layer synthetic media biotrickling system with a footprint of 4 m^2 operated at the design airflow (1700 m^3/h) and operated at an increased (20%) airflow

Pollutant	Inlet odor concentration (OU_E/m^3)[c]	Outlet odor concentration (OU_E/m^3)[c]	Removal (%)
Odor[a]	7800	310	96.0
Odor[b]	5700	540	90.5

[a] At designed air flow of 1700 m^3/h.
[b] At 20% increased airflow of 2050 m^3/h.
[c] OU_E/m^3: odor units per cubic metric of air, measured according to the European standard EN13725.

3. Waste gas treatment at municipal wastewater treatment plants

To reduce odor emission from municipal wastewater treatment plants, tanks containing untreated sewage or sludge are covered and ventilated. The ventilation gas is treated. In 2000, an inventory in The Netherlands on biological waste gas treatment at these plants was conducted [6]. Approximately, 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 biofilters packing materials could be found: lava rock (38% of the cases), coconut fibre (31%), compost (30%) and synthetic media (1%). For new wastewater installations, chemical scrubbers are rarely used anymore. Over the last 10 years, gas treatment has become obligatory for all new sewage plants and that compost-based biofilters are being replaced by lava rock biofilters or synthetic packing biofilter systems. Compost-based biofilters yield too many problems with acidification, drying and packing renewal. Biofilters with lava and synthetic media are wetted by the effluent of the sewage plant that contains all necessary nutrient minerals and they have a long-packing lifetime (at least 10 years). As a result, the operational stability appears to be better. Biofilters with lava are preferred because of greater experience with its use, but biofilters with synthetic media are more promising for the future because of their lower weight, smaller size and robustness. These synthetic biofilter systems have also been installed during the last couple of years in other parts of the world with great success [7–9]. An example is shown in Table 2, which is discussed in more detail by Webster [10].

4. Sulfur emissions from industries

4.1. H_2S removal from industrial gases

Biogas, natural gas, synthesis gas and Claus process tail gas mostly contain H_2S. In large-scale (>15 tons S/day) gas treatment, amine absorbers and Claus plants can be used to remove H_2S. For smaller quantities, liquid redox systems, based on reaction with iron chelates, are used. Bioscrubbers may be an alternative for these redox systems. A bioscrubber was developed for the removal of H_2S from aerobic gases. It comprises an absorber in which H_2S is absorbed in water and an aerated bioreactor in which sulfide is biologically converted into elemental sulfur [11]. A similar system can be applied to clean anaerobic gases. According to Janssen et al. [12,13], this technology is competitive in the 0.1–15 tons S/day range. The reactions in the absorber and the bioreactor are, respectively,

$$H_2S + OH^- \rightarrow HS^- + H_2O$$

$$HS^- + \tfrac{1}{2}O_2 \rightarrow S^0 + OH^-$$

An early report is given by Dijkman [14] in which this type of bioscrubber for 400 m^3 biogas/h is described. The absorber was a packed spray tower operated at a relatively high pH and the bioreactor was a 72 m^3 submerged fixed-film reactor operated at neutral pH. The bioscrubber removed more than 99% of the H_2S introduced (10,000–15,000 ppm in influent gas and 20–120 ppm in effluent gas). According to the author, compared with a caustic soda scrubber, 90% of the amount of NaOH required can be saved if a bioscrubber is used. Full-scale bioscrubbers for biogas treatment were installed worldwide during the last 5 years.

Table 3
Efficiencies of a full-scale bioreactor treating 1200 m^3/h waste gases from an anaerobic wastewater treatment system at a brewery [17]

Pollutant	Inlet concentration	Outlet concentration	Removal (%)
Hydrogen sulfide (ppm)	800	1.7	>99
Other reduced sulfur (ppb)	>2780	<399	>86
Odor concentration (MOU_E/h)[a]	7000	200	97[b]
	5000	30	>99[c]

[a] OU_E/h: odor units per hour, measured according to the European standard EN13725.
[b] After start-up.
[c] Half year after start-up.

Besides biogas, high-pressure natural gas can be treated in a similar way. Pilot-plant experiments have been carried out with this type of gas. The pilot-plant comprised an absorption column, operating at pressures between 5 and 53 bar, a flash vessel, 0.4 m^3 aerated bioreactor, operating under atmospheric pressure and a plate settler for the separation of sulfur and water. The sulfur slurry can be concentrated in a decanter centrifuge, yielding a sulfur cake with 40% water and dry matter consisting of 95–99% S^0 [15]. Various options exist to make sulfur of different purities and reuse it in agriculture or sulphuric acid manufacturing. After a demonstration phase [14], the first full-scale high-pressure unit was started in Canada in 2001. Outlet sulfur concentrations were typically 4 ppm or lower and the H_2S removal efficiency was always above 99.5%.

Another type of bioreactor installation that treats industrial gases containing high-H_2S concentrations aerobically has been developed and built. This bioreactor was used, for example, for the treatment of gases from a vegetable oil refinery that contain up to 2000 ppm H_2S. The pH of the recycled water was maintained around one and fresh water addition was controlled by electro-conductivity. Typically, a removal efficiency of >98% H_2S was found [16]. This method of treatment was also shown to be effective at industrial anaerobic wastewater treatment plants (see Table 3). The bioreactor consists of two or three layers. In the first layer, mainly autotrophic bacteria such as *Thiobacillus* are active and H_2S is removed. In the second layer, heterotrophic microorganisms degrade organic odorous compounds. No chemical addition is required in this process.

4.2. SO_x and NO_x removal from flue gases

SO_x can be removed from flue gases by scrubbing with dilute solutions of caustic soda or limestone. However, in these processes, the costs for chemicals are high and products such as disodium sulfate can be difficult to dispose [18]. A biological alternative was developed [18,13]. In this bioscrubber, the hot gases pass an absorber in the form of a reverse jet wet scrubber in which particulates and SO_x are absorbed. The most important reactions are:

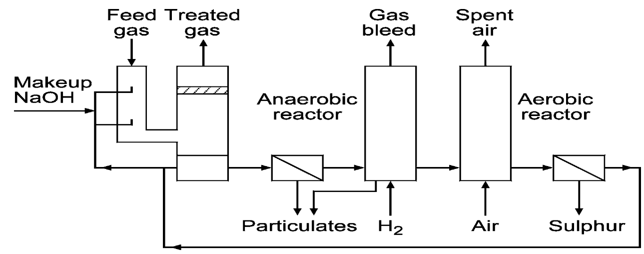

Fig. 1. Flow sheet of a bioscrubber for SO_x removal from flue gases (adapted from Cetinkaya et al. [18]). Inlet and outlet air streams of flue gas are called feed gas and treated gas.

$$SO_2 + NaHCO_3 \rightarrow NaHSO_3 + CO_2$$

$$NaHSO_3 + \tfrac{1}{2}O_2 + NaOH \rightarrow Na_2SO_4 + H_2O$$

The latter reaction consumes only a part of the $NaHSO_3$ formed in the first reaction. The liquid from the absorber is transferred to an anaerobic reactor in which the sulfites and sulfates are biologically converted to sulfide. For this process, an electron donor is required. For large-scale applications, H_2 gas is preferred, while in small-scale plants, methanol and ethanol may also be used (H_2 generation on a small-scale is relatively costly). The reactions involved are:

$$NaHSO_3 + 3H_2 \rightarrow NaHS + 3H_2O$$

$$Na_2SO_4 + 4H_2 + CO_2 \rightarrow NaHS + NaHCO_3 + 3H_2O$$

The conversion of sulfide to sulfur is described in Section 4.1. A simplified flow sheet of the process can be found in Fig. 1.

Pilot-plant experiments have been carried out at a 600 MW power plant that produced 2000,000 m^3 of flue gas/h. The pilot-scale unit treated 6000 m^3/h of flue gas with a temperature of 120 °C. The pilot-plant comprised a 6 m high-absorber tower and a 10 m high-anaerobic ‘Internal Circulation’ reactor, in which gas was circulated by an external compressor to provide good mixing. The aerobic reactor was an airlift loop reactor. The first experiments were carried out using a thermophilic anaerobic reactor (50 °C) and ethanol as electron donor. The start-up took 6 weeks. The aerobic second reactor initially produced mainly sulfate, but after increasing sulfide-loading rates, sulfur was mainly produced. The plant was loaded with 6 kg SO_2/h and the SO_x removal efficiency was 98% [13].

A bioscrubber for the removal of NO_x from flue gases is described by Cetinkaya et al. [18]. The difficulty with NO_x removal from flue gases is that 95% of the NO_x is NO, a compound poorly soluble in water. To overcome this solubility problem, Fe(II)EDTA is used in the scrubber liquid to react with NO. As a result, $Fe(II)[EDTA]NO^{2-}$, a nitrosyl complex, is formed and NO is absorbed from the gas at a high rate, at any temperature. The scrubber liquid containing the absorbed NO_2 and the nitrosyl complex is subsequently transferred to an anoxic bioreactor in which biological

Fig. 2. Bioreactors treating CS_2-emissions at a viscose plant seeded with extremophile acidophilic microorganisms.

denitrification takes place. The nitrogen compounds mentioned are reduced to dinitrogen gas using an electron donor, e.g., ethanol. The Fe(II)EDTA in the liquid can be reused again in the absorber. Removal efficiencies of more than 80% can be achieved. It is possible to simultaneously remove SO_2 and NO_x using two bioscrubbers by simple modifications to an existing wet limestone gypsum plant [18].

Operational costs are lower than those of the caustic soda process, while the investment costs are higher. Paybacks within 2 years can easily be achieved at higher sulfur loads [13].

4.3. CS_2 emission from viscose industries

Bioreactors have been developed for the removal of CS_2 from gases. The bioreactor is filled with a synthetic medium comprised of an acid resistant polymer. The bacteria that grow on the packing oxidize CS_2 or mixtures of CS_2 and H_2S into CO_2, H_2O and H_2SO_4. A starter culture with special extremophile acidophilic bacteria is used and operated at a pH < 1. In 1999, a bioreactor was installed at a sponge factory treating 50,000 m^3/h of gas containing an average of 300 ppm CS_2 and 250 ppm H_2S (see Fig. 2). Removal efficiencies of about 90% CS_2 and 95% H_2S were typically measured. The robustness of this system has been studied and described [19]. Full-scale plants have been delivered to other viscose industry facilities (sponges, fibres) and some other industries emitting CS_2 [19,17].

5. Odor and NH_3 emission from live stock industries

Ammonia emissions in Europe [20] are an important contributor to the acid rain that can cause acidification of the environment. Air ventilation from pig stables at livestock industries are subject to more stringent regulation than for other animal housing facilities in many countries. In addition, treatment can be required, because of odor nuisance in the neighbourhood of the livestock industry. Treatment of this ventilation air from stables is possible but difficult, since the amounts of air are relatively high and the concentrations in the air relatively low. Conventional biological waste gas treatment systems (biofilters and bioscrubbers) have been developed for the treatment of ventilation air from live stock industries, but have not proven to be sufficiently cost-effective or robust. The introduction of biotrickling systems made biological treatment more effective and many full-scale systems have been in operation for many years, especially in Germany and The Netherlands. Table 4 shows an example of a full-scale biotrickling filter treating odor and ammonia emissions from a pig stable. Ammonia is treated biologically to form nitrate by nitrification. Some of these biotrickling filters are capable of denitrification of the nitrate to nitrogen gas [21].

Table 4
Full-scale biotrickling filter treating air from a pig stable [21]

Number of pigs (#)	500	
Airstream (m^3/h)	46800	
Empty bed gas residence time (s)	1.4	
pH recirculation water	7.0–7.7	
Pressure drop (Pa)	< 100	
Efficiencies of ammonia removal	Period 1	Period 2
Inlet concentration (ppm)	23	24
Outlet concentration (ppm)	7	7
Removal ammonia (%)	71	73
Efficiencies of odor removal	Period 1	Period 2
Inlet concentration (OU_E/m^3)[a]	1206	2329
Outlet concentration (OU_E/m^3)[a]	232	248
Removal odor (%)	81	89

[a] OU_E/m^3: odor units per cubic metric of air, measured according to the European standard EN13725.

6. Reactors using mechanical forces that prevent biomass clogging

In Europe, much attention has been paid to solve the problem of clogging of biotrickling filters with biomass. The use of high-salt concentrations showed that clogging could be reduced [22]. Experiments with potassium and phosphorus limitation [23] and predation by higher organisms [24] have been carried out to prevent clogging. One of the developments in the struggle to prevent biomass accumulation is system that has been developed to remove biomass by mechanical forces.

A moving bed trickling filter was developed using mechanical forces to remove biomass [25]. These filters are cylindrical towers with gas as the continuous phase. They are filled with small plastic spheres, which are continuously removed at the bottom of the tower, after which they are mechanically cleaned and returned at the top of the tower. The trickling filter can be used for a combined treatment of wastewater and waste gas, and full-scale plants have been installed. The largest plant is running at the site of a chicken slaughterhouse. It treats 640 m^3 wastewater (400 kg COD)/day and 30,000 m^3 waste gas/h using a reactor with

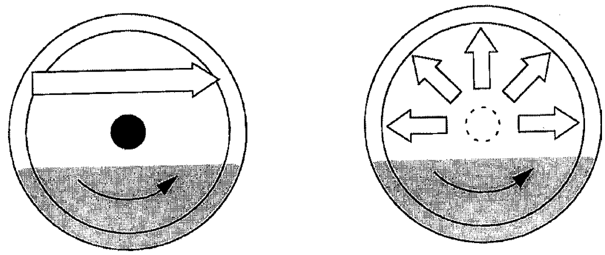

Fig. 3. Gasflow paths through a rotating biofilm reactor [26].

a diameter of 5 m and a volume of 150 m^3. Water and gas both co-currently down the tower. Wastewater-loading rates of 20 kg COD/(m^3 reactor day) can be used and COD removal efficiencies of 85–90% have been attained.

With a similar motivation rotating systems have been developed. Rotating systems with fixed carriers were originally developed for wastewater treatment. The carriers were fixed vertically to a horizontally mounted and rotating shaft. They are partially (40–60%) immersed in the wastewater. During rotation, the biofilm absorbs the oxygen from the gas phase and the organic material from the wastewater. The rotation leads to good mixing and holds detached biofilm particles in a floating condition. To treat waste gas, the gas usually flows tangentially along the carriers' discs (see Fig. 3). Central introduction of the gas would also make it possible to use a packing instead of discs as support for the biofilm as proposed by Rudolf von Rohr and Ruediger [26]. Gai et al. [27] proposed rotating support packing units and demonstrated the system at pilot-scale. Results are promising and more theoretical and experimental work is underway.

7. Other developments

7.1. Biofilters based on the action of fungi

Biofiltration is found to be cost-effective for off-gases with low concentrations of VOC (<3 g/m^3) [28] and an odor reduction of 99% is possible. However, conventional biofilters based on compost and bacterial activity, face problems with the elimination of hydrophobic compounds such as aromatic compounds, alkenes and alkanes. Because of the low solubility in water, the compounds are poorly absorbed by the bacterial biofilms. Besides that, biofilter operational stability is often hampered by acidification and drying out of the filterbed.

To overcome these problems, biological waste gas treatment systems with fungi on inert packing material have been developed. Fungi are more resistant to acid and dry conditions than bacteria, which is a helpful property when operating biofilters. Moreover, it is hypothesised that the aerial mycelia of fungi, which are in direct contact with the gas, can take up hydrophobic compounds faster than flat aqueous bacterial biofilm surfaces. This is believed to be due to the absence of a water layer between the gas phase and the biomass and the presence of a relatively large surface area of the mycelia of the fungi. Perlite, which is an inert granular porous ceramic material, was selected as one of the most suitable support media for fungal growth and it maintained a high-biofilter volumetric elimination capacity [29]. Water amended with the required nutrients has to be added at regular time intervals to support growth.

A biofilter for styrene has been scaled up and was demonstrated on 1.5 m^3 scale at the site of two polyester factories [30]. Preliminary tests have been carried out with mixtures of toluene, ethylbenzene and xylenes, and with ethene, 1,3-butadiene, alpha-pinene [31–33] and hexane (Groenestijn, unpublished results).

The effect of pH and humidity in the biofilters for gas containing toluene was demonstrated in Ref. [34]. Four biofilters, which differed in pH 4 and 8 and gas relative humidity, were compared. A difference in performance appeared: the filters with the low pH were able to remove more toluene from the gas than the filters with the high pH. The low pH filters developed fungi, while in the other filters mainly bacteria were growing. The addition of medium/water was stopped in all filters in order to simulate drying. The biofilter with a pH 4 and a relatively low relative humidity of the inlet airstream maintained a volumetric activity of about 125 g/(m^3 filterbed h) up to 1000 h running time. The bacterial biofilters had a capacity of not more than 20 g/(m^3 h) during this drying period. The fungal biofilters eliminated up to 99% of the toluene.

However, the high-volumetric activities found are not always beneficial with respect to control of gas pressure drop, i.e., clogging can occur. This problem was solved by the introduction of mites as predators, in biofilters containing fungi, and appeared to be successful to prevent filterbed clogging, while maintaining high-elimination capacities [24]. The pressure drop of a fungal biofilter of 1 m height, loaded with 200 m^3 gas/(m^3 filterbed h) stabilised around only 130 Pa. Unpublished data of a preliminary study on emissions of fungal spores from this type of bioreactor did not show an increase the potential impact on human healths; therefore, effluent filters may not be necessary and were not recommended.

Spigno et al. [35] isolated fungi capable of absorbing and degrading hexane at a high rate. Introduction of the fungi in a biofilter yielded an elimination capacity of 150 g hexane/(m^3 h). A recently published review paper by Kennes and Veiga [36] shows that many different fungi have the potential to be used in a biofilter system. The use of fungi can increase elimination capacities up to 5–10 times greater than those reported with conventional compost biofilters. Estimated costs for these fungal biofilters for treatment of 48,000 m^3 gas/h with 500 mg VOC/m^3, aiming at 80–90% removal are € 24 investment costs/(m^3 h) and € 0.60 operating costs including capital costs/1000 m^3 treated gas. A full-scale project has not been realized yet, but such a project is in preparation.

Table 5
Infrared measurements as monitoring tool for biofilters

Biofilter A (a good performing biofilter)			
Temperature range on biofilter surface (°C)[a]	5		
VOC concentration emitted from the biofilter surface (average/standard deviation)	Day 1 45 ± 2 ppm	Day 2 62 ± 3 ppm	Day 3 48 ± 19 ppm
Biofilter B (a poor performing biofilter)			
Temperature range on biofilter surface (°C)[a]	20		
VOC concentration emitted from the biofilter surface (average/standard deviation)	Day 1 85 ± 101 ppm	Day 2 85 ± 101 ppm	

[a] The difference between minimum and maximum temperature measured on the biofilter surface.

7.2. Elimination of alkanes from off-gases using biotrickling filters containing two liquid phases

Because of their low solubility in water, removal of alkanes by biofilters is troublesome. Conventional techniques such as biofilters have low elimination capacities for hydrophobic compounds caused by a poor mass transfer from the gas to the aqueous phase. To overcome these problems, a novel biotrickling filter was developed and is characterized by the use of a non-biodegradable organic solvent. In such a biotrickling filter, a mixture of an organic solvent and water is continuously trickled over a packed bed, while the polluted gas passes counter-current to the liquid [37]. The microorganisms exist on the packing material and in the circulating liquid. The alkanes are absorbed in the oil phase of the liquid, transferred to the microorganisms and biodegraded. Laboratory-scale experiments at 20 l scale with hexane as a model pollutant and silicon oil as a solvent demonstrated that 90% elimination efficiency could be reached at a volumetric-loading rate of 100 g hexane/(m^3 filterbed h). The influent gas contained 10 g hexane/m^3 and had a temperature of 29 °C. The method could be a cost-effective way to treat gases containing high concentrations of hexane, other alkanes or other strongly hydrophobic compounds. Biological co-oxidation of other biodegradable pollutants from the gas is to be expected.

7.3. Determination of homogeneous air distribution

An important, often underestimated, process parameter for biofilter operation is the air distribution evenly through the media. The airflow through the media of a biofilter must be even and leakage of the airstream to be treated should be avoided in utilize the biodegradation capacity optimally. Monitoring of this uniform air distribution is often qualitatively performed using smoke, but quantitative measurements, e.g., by using trace gases are often not practical or not possible because of cost or equipment limitations. A monitoring method to detect uneven airflow through a filter has been developed using infrared-measurements [38]. The heat radiation, emitted from the surface of a single stage biofilter, is recorded and presented as a temperature field image. No contact between the measuring device and the unit under measurement (the biofilter) is necessary. In Table 5, results are shown from two conventional biofilters at composting facilities. The VOC concentrations emitted from different places of the biofilter surface changes much less at biofilter A than biofilter B as can be seen from the standard deviation in Table 5. A relatively large standard deviation of the VOC measured from different places of the biofilter surface indicates that the biodegradation capacity in the filter is not optimally used. The temperature range on the surface of biofilter A is much smaller than the temperature range of biofilter B (5 °C versus 20 °C) indicating that the air is not distributed evenly through the biofilter media and that optimizing the airflow distribution through the media of biofilter B will most likely improve its performance.

7.4. Biofilters in space

A biological air purification system for spacecraft is being developed, based on a membrane module in which gaseous pollutants are transferred to a wet biofilm on the other side of the membrane. The regeneration method could permit cabin air to be cleaned without producing much waste (e.g., used active carbon), especially for long-term space missions. Promising tests have already been carried out by the European Space Agency in space station MIR and in January 2003 in the space shuttle Columbia (which disintegrated in the upper atmosphere during re-entry).

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