The effect of nutrient supplementation on the biofiltration removal of butanal in contaminated air

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Abstract. Butanal is one of the odorous compounds produced in the animal-rendering and food-processing industries and also in sewage-treatment plants. It shows the necessity for complementing such plants with systems for off-gas treatment. Biofiltration using simple packing material was tested for the removal of butanal. Excellent results were obtained when the filters operated at optimal humidity and were supplemented with inorganic nutrients. Without nutrients, butyric acid was detected in the effluent gas, which may explain the lower efficiency of filters without nutrients. Under optimal conditions an elimination of around $90 \ g \cdot m^{-3} \cdot h^{-1}$ was reached.

Introduction

In recent years, biotechnological methods have increasingly been applied in the controlled processing of wastes. With respect to the purification of polluted air, biofiltration is a frequently applied technique for odour abatement (Carlson and Leiser 1966; Helmer 1974; Rands et al. 1981; Furusowa et al. 1984; Ottengraf 1986; Van Langenhove 1985; Lee and Shoda 1989; Diks and Ottengraf 1991; Weckhuysen et al. 1991). This technique is based on the ability of micro-organisms to degrade organic/inorganic pollutants to water, carbon dioxide and mineral salts (Brauer 1984; Bardtke 1987). In a biofilter, odorous air is directed through a fixed bed of materials in which develops a mixed microbial population able to grow and to degrade volatile odour compounds. Materials used for bed formation may contain sufficient nutrients to support growth on polluted air components, but this may not always be possible, especially when synthetic materials are used as filling material. Thus, using biological filters running in parallel, the effects of nutrient enrichment of the biofilter, were studied.

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Butanal with a low odour threshold of $13 \ \mu g \cdot m^{-3}$ (Gemert and Nettenbreijer 1977) was chosen as a model compound. It is released by animal-rendering and food-processing industries and also during the treatment of waste-waters, which enforces the idea that biological waste-water treatment plants should be equipped with systems for polluted air treatment (Weckhuysen et al. 1991). Butanal and other aldehydes are formed by the breakdown of fatty acids and the Strecker degradation of amino acids (Van Langenhove 1987). In this study, wood bark was used as packing material, because of its excellent air permeability characteristics, its availability and its low cost (Van Langenhove et al. 1986).

Materials and methods

Experimental set-up of the biofilter. A scheme of the experimental set-up is given in Fig. 1. The biofilter consists of three plexiglas columns with a diameter of 0.10 m and a height of 0.33 m, each filled with wood bark as packing material. To prevent chanelling of the air flows, the material was tamped down. The three columns were connected with each other to form a filter bed of 0.99 m height. Before entering the filter, the air was humidified and a second air stream was saturated with butanal by passage in a wash bottle with butanal. The butanal-saturated air and the humidified air were mixed before entering the biofilter columns. Four measurement points at different filter-bed heights were used for air analysis. Air flows were measured by a home-made anemometer.

Determination of microbial growth. Moulds, yeasts and bacteria were determined by growth on nutrient agar (Difco Laboratories, Detroit, Mich., USA) with pimaricin (for bacteria) or gentamycin (for yeasts and fungi). Butanal-consuming micro-organisms were determined by growth on a nutrient medium containing in $g \cdot l^{-1}$: agar, 20; $(NH_4)_2SO_4$, 1; NaCl, 0.9; KH_2PO_4, 0.3; $K_2HPO_4 \cdot 3 H_2O$, 1.2; $CaCl_2$, 0.005; $MgSO_4$, 0.05; $FeCl_3$, 0.01; butanal, 0.1. Growth was evaluated after 5 days at 30° C.

Composition of the nutrient solution. The biofilter nutrient solution contained in $g \cdot l^{-1}$: Na_2HPO_4, 0.8; NaH_2PO_4, 0.2; $CaSO_4 \cdot 2 H_2O$, 0.05; $MgSO_4 \cdot 7 H_2O$, 0.5; $(NH_4)_2SO_4$, 1.0; $FeSO_4 \cdot 7 H_2O$, 0.01.

Analytical methods. The moisture content of the filter material was measured by the difference in weight before and after drying
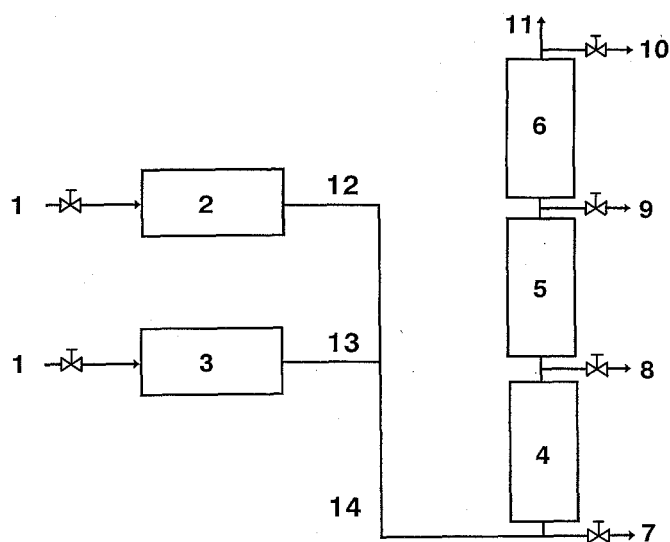
Fig. 1. Experimental set-up of the biofilter: *1*, air inlet; *2*, humidification unit: wash bottle and a bubble column with water in series; *3*, wash bottle with butanal; *4-6*, three biofilter sections filled with wood bark; *7-10*, measurement point at filter-bed heights of 0.00 m, 0.33 m, 0.66 m and 0.99 m, respectively; *11*, air outlet; *12*, humidified air; *13*, butanal-saturated air; *14*, humidified air polluted with butanal

at 110° C. The wood bark pH was measured using a mixture of 1 vol. wood bark and 9 vol. distilled water, after a 5-min mixing time. Butanal concentrations were measured according to Sawicki et al. (1961) using a 0.05% aqueous 3-methyl-2-benzothiazolone hydrazone hydrochloride (MBTH) solution. Butyric acid was measured gas chromatographically (model: Carlo Erba 2350; column: Chromosorb 101) after adding metaphosphoric acid to the alkaline absorption solution (pH 10.0).

Results

Influence of biofilter humidity

To study the influence of the humidity of the biofilter on the elimination efficiency the biofilter was filled successively with wood bark of 15.2, 32.9, 47.8 and 57.4% humidity. The filter-bed height was limited to 0.2 m and the biofilter was charged with 10 ppm (v/v) butanal at a volumetric load of $100 \, m^3 \cdot m^{-2} \cdot h^{-1}$. The butanal concentrations in the outlet were measured after 8 h, 1 and 3 days. The results are expressed as the elimination efficiency (η), calculated as follows:

$$\eta = \left(1 - \frac{Ci}{Co}\right) * 100 \, (\%) \tag{1}$$

where Ci = the butanal concentration measured at height i in the biofilter and Co = the butanal input concentration.

The results in Table 1 show that there is a minimum humidity for the biofiltration process. A humidity of 32.9% and lower gave no permanent butanal elimination. With a humidity of 47.6%, a constant elimination efficiency was obtained after 1 day of filtration. This indicates that the minimum humidity is between 32.9 and

Table 1. Influence of the humidity of the filter material on the butanal elimination efficiency of a biofilter

Humidity of the filter material (%)	Elimination efficiency (%) after		
	8 h	1 day	3 days
15.2	0	0	0
32.9	3	0	0
47.8	30	19	19
57.4	48	40	43

47.6%. This value is higher than the 30% cited by Bardtke (unpublished data) but is certainly depends on the composition of the filter material. In the beginning, only an adsorption and/or absorption phenomenon was observed. Evidence for this hypothesis is found in the measurements with a humidity of 32.9%. After 8 h an elimination efficiency of only 3% was obtained, but after 1 day no elimination was observed. This suggests that there is no regeneration of the absorption and/or adsorption sites on the wood bark by micro-organisms. On the contrary, when the humidity was higher than 47.8% permanent elimination efficiency was obtained after 3 days. The elimination efficiency rises with increasing humidity but increasing humidity results in lower air permeability and higher operating costs. This suggests the existence of an optimal humidity. For hydrogen sulphide this humidity was 65% according Van Langenhove et al. (1986). In the following experiments we have used a humidity of 57% because of its good characteristics for butanal removal and air permeability.

Nutrient supplementation

The influence of nutrient supplementation has been investigated using two identical biofilters with wood bark as packing material. To the first biofilter (Biofilter 1) 14 days daily, later weekly, 30 ml of the nutrient solution were supplied. The second biofilter (Biofilter 2) was operating without nutrient supply. These biofilters were filled with wood bark at pH 6.5 and a humidity of 57.4% to obtain 0.99 m of filter material. During 12 weeks the biofilters were loaded with butanal (10 ppm, v/v) at a volumetric load of $100 \, m^3 \cdot m^{-2} \cdot h^{-1}$. The butanal concentrations were measured at four biofilter heights (0.00 m, 0.33 m, 0.66 m, 0.99 m). The elimination efficiencies for the different filter-bed heights were calculated using Eq. 1.

The elimination efficiency measurements for Biofilter 1 are given in Fig. 2A. Although there were some fluctuations in the removal efficiencies, two periods can be distinguished. In a first period the efficiency increased up to a maximum at 21 days. It may reflect the adaptation of the micro-organisms to the substrate. During the second period the efficiencies were clearly different for the three sections of the biofilter. Most of the butanal was removed in the first section. After 84 days the total remained excellent at around 97%, with fluctuations probably due to changes in back pressure (Van Langen-
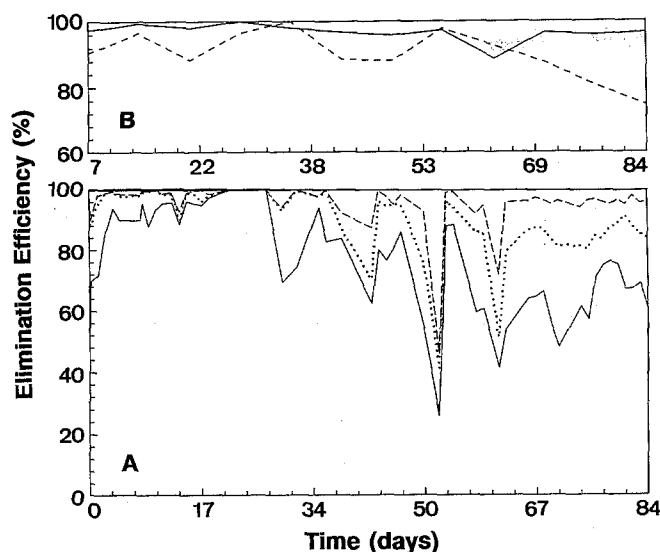
Fig. 2. A Elimination efficiency (%) at different filter-bed heights: 0.33 m (*solid curve*), 0.66 (*dotted curve*) and 0.99 m (*dashed curve*) with a biofilter with nutrient supplementation. **B** Comparison of the global and mean elimination efficiency (%) between biofilters with (*solid curve*) and without (*dashed curve*) nutrient supplementation

hove et al. 1986). Figure 2B compares the global elimination efficiency (week means) of both biofilters. It can be seen that the first biofilter performed better than the second (with exceptions after 35 and 56 days). During the whole period a mean elimination efficiency of 97% was obtained for Biofilter 1 and 86% for Biofilter 2.

Colonization of the biofilters

During these experiments a film of micro-organisms was formed on the packing material. These micro-organisms were scraped off the wood bark, suspended in a Ringer solution and plated out on nutrient agar. Different bacterial strains, moulds and yeasts were present. To ensure that these micro-organisms were able to degrade butanal, scrapings were also inoculated on a nutrient medium with butanal as sole carbon source. Growth on this medium was found for bacteria, moulds and yeasts but no further identification was attempted. The results are summarized in Table 2. Comparing the growth on the two media, it can be concluded that only a part of the micro-organisms scraped off the wood bark are able to use butanal as sole carbon source.

Influence of butanal load

Different butanal loads were applied to evaluate the elimination capacity. Figure 3 shows a typical curve for each biofilter. At a critical load (L_c) the so called maximum elimination capacity (EC_{max}) is reached. The results of this experiment are given in Table 3 showing a higher critical load, maximum elimination capacity, butanal removal and carbon removal with nutrient supplementation.

Table 2. Growth of bacteria, yeasts and moulds isolated from the scrapings of the wood bark material

Growth of	Nutrient agar medium (cfu·cm^{-2})	Butanal medium (cfu·cm^{-2})
Bacteria	$\pm 10^8$	$\pm 10^7$
Yeasts and moulds	$\pm 10^7$	$\pm 10^6$

cfu, colony-forming units
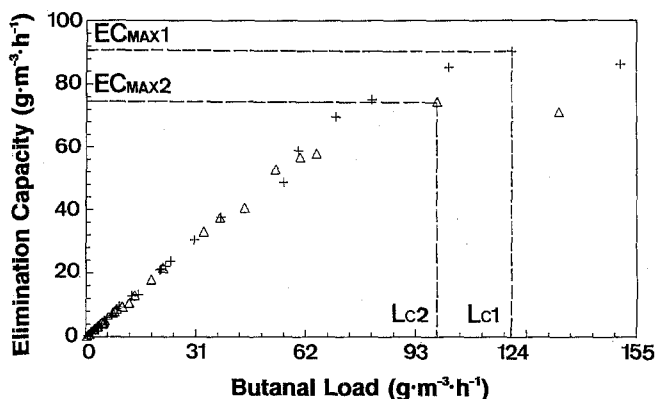
Fig. 3. Elimination capacity (in g·m^{-3}·h^{-1}) of biofilters as a function of the butanal load (in g·m^{-3}·h^{-1}) and maximal elimination capacity (EC_{max}) and critical loads (L_{c1} and L_{c2}) for biofilters with (+) and without (△) nutrient solution

Table 3. The critical load, maximal elimination capacity, butanal removal and carbon removal for biofilters with (1) and without (2) nutrient supplementation

Parameters	Biofilter 1	Biofilter 2
Critical load (g·m^{-3}·h^{-1})	120.0	99.0
Maximal elimination capacity (g·m^{-3}·h^{-1})	90.4	74.5
Butanal removal (kg·m^{-3}·day)	2.2	1.8
Carbon removal (kg·C·m^{-3}·day)	1.6	1.2

Effect of CaCO₃

For pH stabilisation, phosphates, which are absent in waste gas, may act as buffering salts, thus explaining why butyric acid was found in the exit gas from the filter that was not supplied with nutrients. A buffering capacity may also be obtained by adding $CaCO_3$ to the filter material. Thus two filters were operated, one with addition of 100 g $CaCO_3$ (Filter 1) to the amount of material to fill the three sections of the filter, and one without addition (Filter 2). The humidity of the wood bark was 57.6%. With $CaCO_3$ the pH was increased from 6.5 to 7.2. In this experiment butyric acid was chosen as the contaminating compound and introduced at a volumetric load of 100 m^3·m^{-2}·h^{-1} with a concentration of around 6 ppm (v/v). During more then 84 days the butyric acid concentrations were followed at the three biofilter levels. The elimination efficiencies after each stage
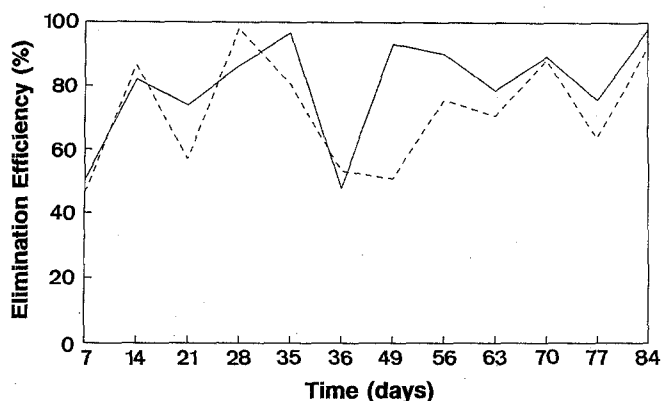
Fig. 4. Comparison of the global and mean elimination efficiency (%) of butyric acid between biofilters with (*solid line*) and without (*dashed line*) $CaCO_3$

and the global elimination efficiencies were calculated, following Eq. 1. After stages 1, 2 and 3 the elimination efficiency reached 38%, 59% and 80%, respectively, for Biofilter 1 and 35%, 59% and 72%, respectively, for Biofilter 2. These results indicate that in all stages of the biofilter the elimination efficiency was equal or higher when $CaCO_3$ was added to the filter material. The global and mean elimination efficiency values of each week are given in Fig. 4. Thus, during the whole period a mean elimination efficiency of 80% and 72% was obtained for Biofilters 1 and 2, respectively. These are rather low elimination efficiencies for carbon compounds but similar results are reported in the literature. According to Van Langenhove et al. (1987) isobutyric acid can be removed by biofiltration with an elimination efficiency between 69% and 98% but after 25 days the elimination became unstable and the biofilter had to be sprinkled with a buffer solution. Thereafter a higher and more stable removal efficiency was obtained. In our experiments, the elimination efficiency stabilised only after 42 days, suggesting an adaptation process of the micro-organisms. It can be concluded that buffering has a low but positive effect on the biofiltration process. Thus in the biofilter without nutrients, butyric acid accumulation might have been the reason for a lower elimination efficiency of butanal.

Discussion

It is concluded that butanal can be removed from odorous air in wood bark biofilters with a high and stable elimination efficiency and capacity. A higher elimination efficiency and capacity was obtained with nutrient supplementation, possibly due to a better nutrient balance for the micro-organisms and pH stabilisation. The first reason can be easily understood because butanal is a carbon source only for micro-organisms. Mixing the filter material with $CaCO_3$ can stabilize the pH and increase the removal efficiency. The elimination efficiency and capacity for butanal and butyric acid are similar to results published for other carbon compounds (Ottengraf 1986).

High elimination efficiencies are needed for odour abatement. Following the Weber-Fechner psychophysical power law (Stevens 1970), odour intensities and the odour compound concentrations are related by the following expression:

$$\left(\frac{I_1}{I_2}\right) = \left(\frac{C_1}{C_2}\right)^n \tag{2}$$

where I_1, I_2 = odour intensity of the odorant, C_1, C_2 = odorant concentration and $C_{1,2} > C_d$ with C_d = odour threshold value of the odourant, and n = odorant dependent parameter. The odour-dependent parameter n varies from 0.12 to 0.87 (Patte et al. 1975; Hall et al. 1983). With a mean value of 0.5 it can be calculated from Eq. 2 that by eliminating the odorant concentration by a factor of 10, the odour intensity will reduce by a factor of 3.16. In order to eliminate the odour of a compound with a low odour threshold value, such as butanal, high elimination efficiencies are needed. To achieve such high efficiencies, nutrient supplementation and/or pH stabilization can be a solution. An additional effect of nutrient supplementation is a higher elimination capacity, removing a higher pollutant concentration under the same operating conditions.

The biofilter area and therefore the cost of the biofilter can be reduced significantly. Bardtke (unpublished data) stated that it was not necessary to add nutrients on biofilters but Adam (unpublished data) found a nutrient supply useful when specific micro-organisms degrade an odour compound on a carrier material. Also a greater elimination capacity was obtained for the degradation of hydrocarbons in the case of nutrient supplementation (Don 1986). Factors such as nutrient balance are also important for the explanation of the effect of nutrient supplementation on the biofiltration of butanal.

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