Treatment of complex gaseous emissions emitted by a rendering facility using a semi-industrial biofilter

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Abstract

BACKGROUND: To improve process design, and scale-up of gas biofilters, a thorough understanding of compounds degradation mechanisms within model engineering biofilters is needed. The aim of this study is then to investigate the spatial distribution of pollutants removal within an experimental semi-industrial biofilter fed with industrial emissions from a rendering plant.

RESULTS: A stratification pattern of pollutants removal has been highlighted: nitrogenous compounds, esters, volatile fatty acids (VFA), alcohols, ketones and aldehydes are completely or mostly removed especially in the biofilter layers near the gas inlet. In return, the removal of sulphur compounds seems to begin when nitrogenous and oxygenated compounds are almost fully degraded. A sequential degradation of sulphur compounds is also observed: hydrogen sulphide is eliminated in the biofilter section near the gas inlet while methylmercaptan (removal efficiency 87%) and dimethyl disulphide (DMDS) (removal efficiency 73%) are mainly eliminated in the second half of the column.

CONCLUSION: An on-site semi-industrial biofilter used for waste gas abatement in an animal-rendering plant exhibited spatial distribution of the compounds removal efficiency. The removal of organic sulphur compounds is not complete. Hence, this work emphasizes the importance of improving gas biofilters to achieve complete elimination of pollutants causing unpleasant odours.

Keywords: exhaust gas; rendering plant; semi-industrial biofilter; performance; spatial distribution

INTRODUCTION

From 2007 to 2013, the US rendering industry produced more than 8 million metric tons of rendered animal products, including tallow, meat and bone meal, poultry-byproduct-meal and feather mea1.[1] About 2.8 million tons of mainly meat by-products were treated in German animal-rendering plants in 1999 (German Federal Environment Ministry).[2] In 2013, around 2.8 million metric tons of raw materials were processed by the French rendering industry (Syndicat des Industries Françaises des Coproduits animaux (SIFCO), www.sifco.fr). Although the products of these processes are no longer being used for animal feed production, rendering plants are still necessary for biologically safe waste treatment.[2] During rendering, the heating of animal tissues liberates a variety of odorous organic and inorganic compounds, creating a significant source of odour nuisance.[3] The odour concentration of the encountered flow is generally between 20 000 and 1 100 000 OU_E (European Odour Unit) m^{-3}.[3,4] The composition of rendering gaseous emissions consists of a wide range of inorganic and organic compounds with about 300 identified compounds.[5] Moreover, it has been reported that the number of odorous compounds ranged between 20 and 50.[3,5,7,8] The odorous compounds that have been identified in gaseous emissions from rendering plants include hydrogen sulfide (H_2S), ammonia (NH_3), organic sulphides (dimethylsulphide, diethyl sulphide), disulphides (dimethyl disulphide, diethyl disulphide), mercaptans (methanethiol), aldehydes (especially C-4 to C-7 aldehydes), amines (trimethylamine, C-4 amines), quinoline, dimethyl pyrazine, other pyrazines, indole, skatole and C-3 to C-6 organic acids. In addition, lesser amounts of C-4 to C-7 alcohols, ketones, aliphatic hydrocarbons, and aromatic compounds are potentially emitted.[5] More recently, other authors[4] have identified and quantified the chemical compounds emitted by rendering plants causing odorant nuisance. Among the identified compounds, the most concentrated pollutants at the biofilter inlet were H_2S, methanethiol, 3-methylbutanal, 2-methylpropanal and ethanal with concentrations generally above 10 mg m^{-3}. The treatment of these pollutants needs then to be efficient in order to reduce the olfactory impact of rendering plant. Over the last few decades, biofilters have become a popular means of odour control, particularly suitable for the treatment of malodorous gaseous effluents characterized by high flow rates (more than

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100 000 $m^3 h^{-1}$) and low contaminant concentration (from a few µg m^{-3} to 1–5 g m^{-3}) such as rendering emissions.[2,3,9] Open filterbed biofilters covering more than a 1000 m^2 area can be used.[2,10] The humidified waste gas is generally forced to flow upwards through the filterbed. Gaseous compounds transfer to the liquid phase of the biofilm where they are consumed as carbon, energy or nutrient source by the biomass. Nevertheless, some works contributing to the final objective of improving process design, and scale-up are really scarce. Hence, a thorough understanding of the compounds degradation mechanisms within model engineering biofilters continuously fed with industrial gaseous emissions is needed. When contaminants are introduced simultaneously into a biofilter, process performance is dependent on the interaction between the contaminants.[11]

In the present study, the functional stratification of an experimental semi-industrial biofilter fed with an industrial gaseous effluent was thoroughly investigated by assessing the relative abundances of waste gas compounds using gas chromatography–mass spectrometry (GC-MS) and by quantifying sulphured compound concentrations (H_2S, methylmercaptan, dimethysulphide, dimethyldisulphide (DMDS)) using gas chromatography. We focus on sulphured compounds since these volatiles have been identified as predominant odorants in the emission of a wide range of activities in bio-industry such as rendering plants.

MATERIALS AND METHODS

Inlet stream

A rendering process is defined as a process using high temperature and pressure to convert whole animal and poultry carcasses or their by-products with no or very low value to safe, nutritional, and economically valuable products. Raw material is dehydrated in continuous steam heated cookers. Condenser units are used to reduce the strongest odours which arise from cooking and reducing the temperature of the cooking steam to around 35–40 °C. Remaining odorous gases (direct-fired meal dryers and workshops) can then be transferred to a biofilter bed constructed of materials such as concrete and layered products such as compost, coarse gravel, pinebark, and woodchips.[12] The total flow rate emitted by the rendering plant chosen as study site was 180 000 $m^3 h^{-1}$.This waste gas was treated using a biofilter with a cross-section of 1720 m^2 and a bed height of 1 m.

Pilot-scale biofiltration unit

An outdoor pilot-scale biofilter (1.4 m internal diameter, 2 m height) and packed with sawmill chips (packing volume 3.1 m^3) was carried out for the treatment of the real and humidified (water content >97%) gaseous mixture emitted by the rendering plant. The gas flow rate, empty bed residence time (EBRT) and superficial gas velocity were 154 $m^3 h^{-1}$, 70 s and 100 m h^{-1}, respectively, which was representative of full-scale biofilter operation. At the bottom of the biofilter, a 0.2 m plenum and an above 0.2 m pebbles section were designed to homogenize the gas velocities before entering the filter. After dust removal in a water scrubber, the biofilter was continuously fed (except during maintenance of water scrubbers corresponding to 2–3 h per week), via a propeller (Invertek Drives, Welshpool, Great Britain), by the rendering emissions in upward flow mode. Even if the rendering activity was interrupted during the week-end, the pilot-scale biofilter was continuously fed with building ventilation airstreams. The mean characteristics of the packing material used are given in Table 1. The biofilter was inoculated by spraying packing material with supernatant (60 L) from activated sludge collected at the on-site wastewater treatment plant. The packing material was periodically sprayed with drill water (pH 7.7) (10 s every 5 h, corresponding to an inflow of 22 L m^{-2} day^{-1}) to maintain constant high moisture content. During the experiment, the mean value of pH of the packing material and the pH of percolate waters were around 8 (Table 1). The biofilter operated for 600 days. Due to the size of the reactor, the complexity of the experimental device and the operation costs, only one biofilter could be implemented.

Table 1. Packing material characteristics. These values were obtained for dry packing material

Physical properties	
Specific surface area a_{vs} (m^2 m^{-3})†	632 ± 284
Diameter dp_{sv} (mm)	9.6
Bulk density (kg m^{-3})	330 ± 17
Bed void fraction (%)	67 ± 3
Pressure drop (Pa)‡	< 10
Water-holding capacity (g H_2O g^{-1} material)	0.67 ± 0.04
Chemical property	
pH	8.1 ± 0.3

†The specific surface area (a_{vs}) was estimated by measurement as sawmill chips are parallelepiped-like particles ($a_{vs} = \frac{2lL+2lh+2Lh}{Llh}$ with h: height; l: width and L: length).
‡The pressure drop was measured periodically over the entire bed height of 2 m.

Sampling strategy

The performance of each compartment has been evaluated by gas sampling at three dates (7 April, 27 July and 6 October 2011) corresponding to a period of 6 months between 280 and 463 days of operation and to an usual operating activity (confidential data). Due to cost and time reasons, it has not been possible to carry out further gas samples over the total bed height and the biofilter operating period. Gas-sampling ports were located at 0, 0.2, 0.8, 1.6 and 2 m from the gas inlet. This enabled the biofilter performance to be assessed as a function of height. Gas sampling was carried out during functional steady state which corresponds to odour removal efficiency of 80% (data not shown). Gas samples were collected in laboratory-made 40 L Nalophan® bags (for sulphur compounds quantification) or into a 1 L glass bubble (for gas chromatography–mass spectrometry analysis) using a KNF N86KN pump (Midisciences, Rousset, France).

Gas composition assessment

The extraction of pollutants from sampled gases was achieved according to the protocol developed by Lestremau.[13] Briefly, the extraction of pollutants was performed by SPME (Solid Phase MicroExtraction) in static mode using a PDMS/Carboxen (75 µm stableflex) fiber (Sigma-Aldrich, Saint-Louis, MO, USA). The SPME fiber was inserted in the 1 L sampling bulb through a suitable septum and was exposed for 10 min at 20 °C. At the end of sampling, the fiber was retracted in the needle, removed from the bulb and thermal desorption was carried out in the gas chromatograph injection port which was held at 250 °C.

The richness and the relative abundances of pollutants were determined by gas chromatography–mass spectrometry (trace

gas chromatograph coupled with a DSQ mass detector (Thermo Fisher Scientific, Waltham, MA, USA)). An Optima 5-ms Accent analytical column, 0.25 mm internal diameter, 1 μm film thickness, 60 m (Macherey-Nagel, Düren, Germany) was used with carrier gas (helium 6.0) at a flow rate of 1.5 mL min^{-1}. The temperature of the detector was 200 °C. The method used was as follows: 40 °C for 9 min; 10 °C min^{-1} from 40 to 90 °C; 90 °C for 6 min; 5 °C min^{-1} from 90 to 250 °C; 250 °C for 10 min. The mass spectrometer was operated, by electronic impact, in full scan in the range of m/z 20–250.

Quantification of gaseous sulphured compounds

Gaseous concentrations of sulphured compounds were determined by connecting Nalophan® bags to a gas chromatograph CHROMA-S (Chromatotech, Houston, TX, USA) with MXT-624 column (Øi 0.53 mm, length 30 m, stationary phase thickness 3 μm, Restek, Bellefonte, PA, USA) with a carrier gas (compressed air) at 130 hPa, and equipped with a flame photometric detector (FPD). The detector temperature was set at 150 °C, and the oven temperature at 50 °C.[14] The calibration was realized using a certified gaseous mixture (Messer France, Puteaux, France) of hydrogen sulphide, methanethiol, ethanethiol, diethylsulphide, dimethylsulphide, dimethyl disulphide and carbon disulphide with a concentration of 100 mg m^{-3} each. The detection limit of each compound was 150 μg m^{-3}.

Determination of biofilter performance

For each compound *i*, the removal efficiency (RE_i) was calculated at the biofilter height *h*:

$$RE_{i,h} = \frac{C_{i,inlet} - C_{i,h}}{C_{i,inlet}} \quad (1)$$

where $C_{i,inlet}$ is the concentration of the compound *i* at the entrance of the biofilter.

RESULTS AND DISCUSSION

The aim of this study is to investigate the performance of a semi-industrial biofilter fed with a real (i.e. complex) gaseous mixture emitted by a rendering plant. Hence, we focus on compounds removal efficiency along biofilter bed height by comparing 'ionic current signals' for different compounds or families of compounds. As the analysis of volatile sulphur compounds using PDMS (Polydimethylsiloxane)/Carboxen SPME (solid-phase microextraction) fiber is limited by the formation of artefacts such as the reaction of mercaptans to form the corresponding dimers (DMDS), we used another analytical system for this chemical family (direct quantification of gaseous sulphur compounds by connecting the sampling bag to a gas chromatograph equipped with a flame photometric detector). Moreover, the fingerprint of the inlet gas composition has been tentatively assessed (compounds identification and relative abundance expressed in percentage of total emitted compounds in the complex mixture) to check the composition similarity with other such gaseous industrial emissions.

Assessment of the inlet gaseous effluent composition

Waste gas analyses at the biofilter inlet (Fig. 1(A)) reveal that the effluent mainly contains aldehydes (54.8 ± 2.8% of the total emitted compounds) and sulphur compounds (32.8 ± 4.9% of the total emitted compounds). The most abundant aldehydes (10–15 identified compounds) are 2-methyl butanal and 3-methyl butanal followed by 2-methyl propanal. Among ketones, the dominant compounds are acetone, methyl ethyl ketone, propyl methyl ketone and methyl isobutyl ketone. Different esters and VFA (around 8–12 compounds for each chemical family) are detected. The identified alcohols are ethanol, 1-propanol and 3-methyl-1-butanol. The detected nitrogenous compounds are pyrrole, pyrazine (degradation products of DNA bases or amino acids such as proline, histidine) and trimethylamine. Ammonia is not detected. Concerning sulphur compounds (Fig. 1(B)), the dominant compounds are hydrogen sulphide (3.6 ± 2.8 mg m^{-3}) and methylmercaptan (5.1 ± 2.4 mg m^{-3}). According to these data (three dates), the composition of the inlet gas remains globally unchanged corresponding to a fingerprint currently observed for rendering gaseous emissions. The detected compounds in this study have effectively been identified from such gaseous samples.[2–4,6,7] As a given molecule is linked to characteristic fragment ions, the removal of each chemical family and associated compounds within the biofilter could be evaluated.

Spatial distribution of pollutants along the biofilter height

The analysis of Figs 2 and 3 reveals a stratification pattern of pollutants removal. It is apparent that different chemical families with nitrogenous compounds, esters, VFA, alcohols and aldehydes are completely or mostly removed especially in the lower layers of the biofilter (up to around 1 m depth). The ionic current observed for aldehydes at 1.6 m depth is only due to the detection of 3 methyl butanal from one gas sample (7 April 2011) while it was not identified for the others. The removal efficiency of ketones at the biofilter outlet is about 97–98%. The ionic current observed for ketones at 2 m depth is mainly due to the detection of acetone from all gas samples. Hence, among ketones, acetone penetrated down the filters in all layers until breakthrough while the others are eliminated up to 1 m depth. In return, sulphur compounds removal seems to begin when nitrogenous and oxygenated compounds are degraded (from 0.8 m depth to 2 m depth). This functional stratification obtained for an experimental pilot unit fed with a complex exhaust air stream (around 40–50 compounds) corroborates the results observed for duplicated laboratory scale biofilters (about 10 cm across) fed with synthetic gases containing a few compounds only (up to 10 pollutants).[8,15–17] Under long-term stable operating conditions, the treatment of a gaseous mixture (including ammonia and a mixture of volatile organic compounds), representative of composting emissions, revealed the same stratification pattern of elimination capacity within both bioreactors, with better and faster elimination of ammonia and oxygenated compounds compared with organic sulphur compounds.[15] Other work has reported the lower biodegradability of sulphur compounds compared with oxygenated compounds, of ketones compared with esters and aldehydes and of acetone compared with methyl ethyl ketone.[18,19] Friedrich[2] investigated the vertical profile of a full-scale industrial biofilter by assessing the concentrations of some waste gas compounds (aldehydes, ammonia, hydrogen sulphide and dimethyldisulphide) for different depths of the filterbed using gas chromatography–mass spectrometry (GC-MS). The authors stated that the biofilter section near the gas inlet proved to be the most active part for the degradation of ammonia and aldehyde compounds, while dimethyldisulphide concentration revealed no consistent decrease throughout the filter bed. The differences of removal efficiency between different compounds

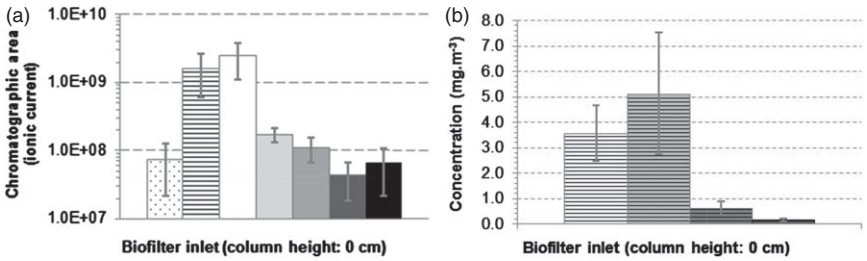

Figure 1. Assessment of the waste gas composition. (A) GC-MS analysis. nitrogenous compounds □, sulphured compounds ▤, aldehydes □, ketones □, esters □, volatile fatty acids ■ alcohols ■. (B) Detailed composition of sulphur compounds: hydrogen sulphide ▤, methylmercaptan ▤, dimethyldisulphide ▤, dimethylsulphide ▤. For each group or individual compound, the results obtained for different gas samples were averaged. h_{total}: total height of the column, i.e. 2 m.

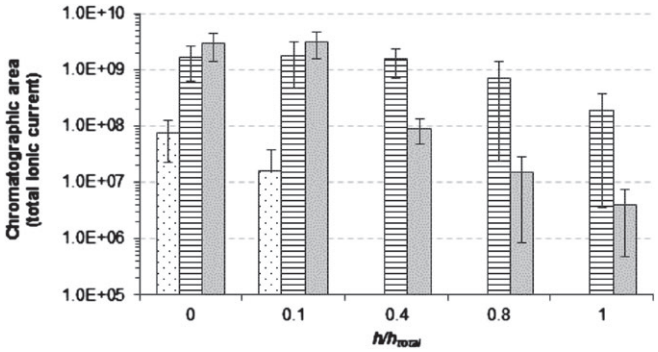

Figure 2. Spatial distribution of chemical families of compounds (nitrogenous □, sulphured ▤, oxygenated □. For each chemical family, the results obtained for different gas samples were averaged. h_{total}: total height of the column, i.e. 2 m.

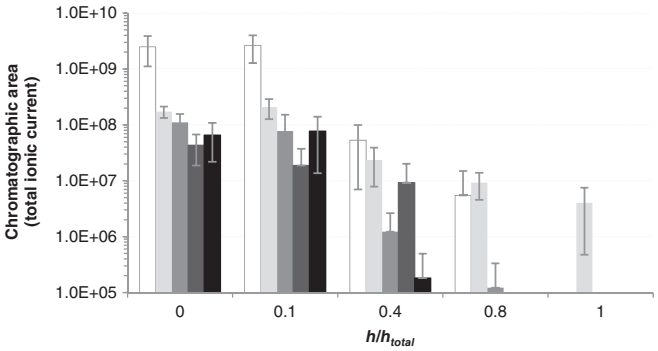

Figure 3. Spatial distribution of oxygenated compounds. Aldehydes □, ketones □, esters □, volatile fatty acids (VFA) ■, alcohols ■. For each chemical group, the results obtained for different gas samples were averaged. h_{total}: total height of the column, i.e. 2 m.

mainly lay in mass transfer characteristics, with a positive correlation between the degradation order and the substrate availability in the liquid phase. In our study, the better elimination of oxygenated (aldehydes, VFA, esters and alcohols) compared with sulphured compounds (methanethiol, dimethyldisulphide) may be due to transfer limitation of poorly soluble sulphur compounds from the gas to the liquid phase (biofilm).[16] This difference can also be attributed to biological factors. This pattern can be explained by competition between substrates: inhibition of hydrophobic compounds (methanethiol, dimethyldisulphide) metabolism by preferential hydrophilic substrates (aldehydes, VFA, alcohols and some esters).[15,20−22] This pattern could also result from competition between bacterial taxa, through the selection of specialized populations able to degrade the biodegradable compounds at sections near the gas inlet and to largely out-compete the other taxa for space, energy and oxygen.[23−25]

Concentration profiles of sulphur compounds along the biofilter height

As has been underlined by Lestremau,[26] the analysis of volatile sulphur compounds using PDMS /Carboxen SPME fiber is limited by the formation of artefacts such as the reaction of mercaptans to form the corresponding dimers (DMDS). Nevertheless, because of their very low odour thresholds (around 1 μg Nm^{-3}), these compounds deserve special attention. Hence, the analysis of these compounds has been determined by using an adapted gas chromatography apparatus. The vertical pattern of sulphur compounds is sharpened by determining the concentration of dominant sulphur compounds. The results (Fig. 4) reveal that sulphur compounds removal begins from 0.2 m height (i.e. $h/h_{total} = 0.1$). A sequential degradation of sulphur compounds is also observed: hydrogen sulphide is mostly eliminated up to 1 m depth while methylmercaptan (removal efficiency of 87%) and DMDS (removal efficiency of 73%) are mainly eliminated in the second half of the column. This analysis corroborates the biodegradation order of sulphur compounds: hydrogen sulphide > methylmercaptan > DMDS.[27,28] According to literature, different specialized groups able to degrade hydrogen sulphide and organic sulphur compounds have been identified.[29] This stratification seems then to display the enrichment and dominance of specific populations, favored by specific operating conditions. In the section near the gas inlet, hydrogen sulphide oxidizing bacteria would then out-compete other populations able to use organic sulphur compounds for their growth.[30]

CONCLUSIONS

Little information is available concerning vertical degradation profiles of model engineering biofilters (1.54 m^2 section and 2 m height) fed with industrial gaseous emissions. As has been highlighted in laboratory-scale biofilters treating synthetic gaseous effluents, an on-site biofilter used for waste gas abatement in an animal-rendering plant also exhibited spatial distribution of compounds removal efficiency. These results then revealed that the removal efficiency depends on the type of pollutants. It has been reported, at laboratory scale, that gaseous concentration of compounds is also related to their abatement levels.[31,32] For example, for a bioprocess operator, it remains difficult to maintain the stability of full-scale biofilters, as industrial waste streams are characterized by variability in terms of pollutants concentration.

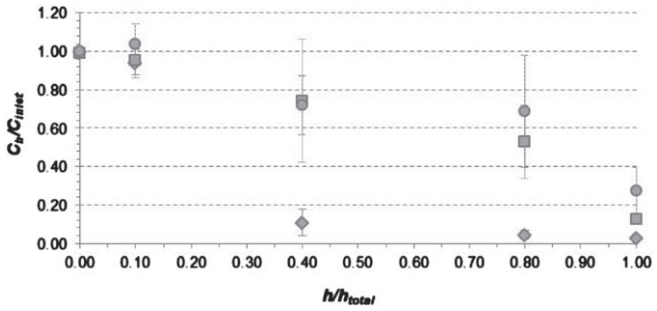

Figure 4. Concentration profiles of sulphur compounds along biofilter height. Hydrogen sulphide ◆, methylmercaptan ■, dimethyldisulphide ●. The concentration ratio C_h/C_{inlet} was plotted against the height ratio h/h_{total}, where C_h is the outlet concentration (mg m^{-3}) at the height h of the biofilter (m), C_{inlet} the inlet concentration (mg m^{-3}) and h_{total} the total height of the column (2 m). For each compound, the results obtained for different gas samples were averaged.

Hence, appropriate efforts (semi-industrial biofilters implementation, reliable sampling strategies) to determine to what extent biofilters are able to cope with a fluctuating environment must be made. Moreover, this work emphasizes the importance of improving gas-biofilter performance to achieve complete elimination of pollutants causing unpleasant odours by developing long-term reliable microbial resource management strategies (using microbial ecology principles) as microbial communities are key players of these complex engineered ecosystems.

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