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Evaluation of sewage sludge and yard waste compost as a biofilter media for the removal of ammonia and volatile organic sulfur compounds (VOSCs)

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ABSTRACT

A biofiltration treatment without addition of water and inoculation has been studied to eliminate exhaust odors formed during the aerobic composting of a mixture containing sewage sludge and yard waste. Odorous compounds present in the air stream have been identified as well as their temporal variation for 27 days of composting. The main emitted pollutants are volatile organic sulfur compounds (VOSCs) with a total concentration of 3.28 ppmv and ammonia at a concentration above 70 ppmv at the 5th day of composting. An EBRT of 60 s was retained to design the experimental biofilter (EB) for a volumetric air flow rate of $15\,m^3\,h^{-1}$. The behaviour of a mature compost containing sewage sludge and yard waste as packing material was studied for the simultaneous removal of ammonia and VOSCs. A removal efficiency (RE) close to 100% was obtained for ethanethiol, dimethylsulfide and dimethyl disulfide whatever the inlet mass loads are. For inlet mass loads between 0.4 and $2.5\,g\,m^{-3}\,h^{-1}$, the mean RE value is 94% for NH_3. In the case of H_2S and methanethiol, after composting during 12 days, the concentration at the outlet at the biofilter was higher than in initial conditions. Emissions remain lower than the levels of toxicity but are above the olfactory thresholds determined for these compounds. A relationship between this emission, the participation of sulfates on the filter medium and the concentration of NH_3 has been determined.

1. Introduction

Biofiltration is one of the earliest biological process developed for the elimination of gaseous compounds and was often used for odor removal in the wastewater treatment industry. Biofiltration is becoming more and more popular because it is a "green" technology which does not use chemicals and does not produce wastes potentially dangerous for the environment. This process is essentially based on the ability of microorganisms to transform organic and inorganic pollutants into less toxic and odorless compounds. Studies have showed that biofilters may be more suitable than other technologies such as wet scrubbers for removing both volatile organic compounds (VOCs) and odor emissions [1]. Nowadays, the number of industrial biological filters for gas treatment installed worldwide has been estimated to be close to 15,000 and half of them are used in wastewater treatment and composting plants [2]. In a biofilter, a contaminated/odorous gas flow passes through a porous and moist medium and the pollutants are sorbed at the surface where they are degraded by microorganisms. The moisture levels in a biofilter are often maintained by a pre-humidification of the inlet gas stream [3]. The by-products formed during the biological degradation are mainly water, carbon dioxide, inorganic salts, some volatile organic compounds (VOCs) and a microbial biomass [4]. The filter bed of porous medium is usually composed of materials such as compost, pine bark, wood chips, peat, inorganic material or a combination of these products. The quality of this filter medium has been reported as one of the key factors in the estimation of the performance of the biofilter. Recently, compost-based media have been extensively used because of their low cost and because they can contain several natural microbial communities that are able to biodegrade various pollutants [5–9].

However, the composition of the compost is highly dependent on the nature of substrates originally available for bioconversion [10]. A bad quality of the substrates (yard waste, sludge, etc.) could give a poor elimination of malodorous compounds during treatment by biofiltration. The objective of this study is thus to quantify this impact on a platform of composting at the Recyclage Organique Mobile (R.O.M) company located at Bordères sur l'Echez (France). A biofiltration treatment has been installed to eliminate the exhaust odors from the composting of sewage sludge and yard waste. In addition to the economical benefit for this company, the originality of this study is that the filter medium which is going to be used is a mature compost based on sewage sludge and yard waste (compost SS/YW) produced by this company.

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Composting is a recycling technology of organic solid wastes that converts organic matter into a stable, humus-like material that could be used as an organic fertilizer [11]. During this process, bacteria are responsible for 80–90% of the biological degradation of the organic matter contained in the sewage sludge. Organic matter is degraded into a stable humus which is a hygienic product, and allows to dry sewage sludge [12].

However, composting of sewage sludge and yard waste produced bad smells. Formerly tolerated, odors are now considered as an unbearable pollution for people at the same level of noise or air pollution.

Composting of organic wastes is responsible for air emission of compounds and the chemical composition of these emissions gives an indication about both the evolution of the process [13] and the quality of the compost. The chemical families of emitted compounds depend directly on the composting process [14].

The biological process of fermentation during composting is responsible for the formation of volatile organic sulfur compounds (VOSCs) which are mainly mercaptan (i.e. methanethiol (MeSH), ethanethiol (EtSH), sulfides (i.e. hydrogen sulfide (H_2S), dimethyl sulfide (Me_2S), disulfides (i.e. dimethyl disulfide (Me_2S_2)), and other sulfur-based compounds. The anaerobic conditions associated with elevated temperatures occurring during the composting process favour the formation of VOSCs, while their emission is strongly decreased upon aeration. For example, the total emission of VOSCs was reported to be 9.2 g of sulfur per tonne (fresh weight) of compost [15]. These compounds are responsible for complaints because they are detectable at very low concentrations (i.e. odor threshold for H_2S: $0.14\,\mathrm{mg\,m^{-3}}$ [16]) and can be dangerous.

Other compounds such as ammonia and amines are also produced by composting and are formed by biological degradation of urea, proteins and amino acids [17]. Ammonia emissions produced during composting of the organic fraction of the municipal solid wastes vary between 18 and 150 g [NH_3] per tonne of waste [18] while ammonia concentrations up to $700\,\mathrm{mg\,m^{-3}}$ have been reported in exhaust gases from composting of sewage sludge [19].

The products formed during biological decomposition of organic matter are different and depend on the aeration conditions. For example, in anaerobic conditions, degradation of lignin led to the formation of terpene [20] while the bacteria reduce amino acids such as methionine into mercaptans [21]. The anaerobic degradation of organic disulfides gives mercaptans and hydrogen sulfide (H_2S) [22].

Other odorous compounds like aldehydes, alcohols, acids and ketones are also responsible for odor pollution.

This study have identified the odorous compounds emitted during composting. They vary temporally and depend on the experimental conditions used (type of solid waste or the composting technology). It is therefore necessary to determine the chemical composition of air emitted by the compost material before treatment by biofiltration.

The physico-chemical analysis of the exhaust gas from the sewage sludge and yard waste composting was the first step of this work. Then, we have particularly studied the biofiltration treatment without addition of water and inoculation of the filter bed composed of a mature compost of sewage sludge and yard waste (compost SS/YW). The objective of this work is to better understand the behaviour of a mature compost SS/YW as packing material for the simultaneous removal of ammonia and VOSCs (EtSH, MeSH, Me_2S, Me_2S_2 and H_2S) exhausted by composting of sewage sludge and yard waste.

2. Materials and methods

The treatment system used in this study is shown in Fig. 1.

2.1. Composted organic wastes

The sewage sludge used in this study was collected at the level of the secondary treatment of a urban water treatment plant (Lyonnaise des Eaux, Suez, France). This sludge mainly contains organic matter and nutrients, such as nitrogen, sulfur, phosphorus which will be used for microbial growth of the biological degradation (fermentation). The presence of these compounds is responsible for the biological fermentation process that emit unpleasant odors. These odors are naturally emitted during the composting process, especially at the beginning of the process and during handling of sewage sludge. Thus, in a sustainable growth approach, we have chosen to treat them by biofiltration without addition of water and inoculation.

The composting process consists of a mixture of sewage sludge (1:3 on a weight basis) and co-products (yard waste, 2:3 on a weight basis) in presence of oxygen. The mixture was performed in a greenhouse (Fig. 1) in order to remove odors from air emitted during the fermentation process. The waste gas is sucked through a fan (flow of $500\,\mathrm{m^3\,h^{-1}}$) and headed towards the biofilters (Fig. 1) using PVC pipes (Pum Plastics, Tarbes, France). Another fan was installed to ensure a forced ventilation of the windrow. For that, ambient air is sucked back through drilled PVC pipes and distributed at the base of the windrow.

2.2. Biofilter set-up

The first biofilter, mentioned as “industrial” in Fig. 1 and noted BI thereafter has been oversized ($5\,\mathrm{m} \times 2.5\,\mathrm{m} \times 3\,\mathrm{m}$) in order to remove all the odorous compounds and to avoid olfactory nuisance for the residents.

The biofilter, mentioned as “experimental” in Fig. 1 and noted EB thereafter has been installed in order to study the different phenomena occurring during the biological biodegradation of pollutants and to better control the operational parameters. To design a biofilter, several parameters have to be taken into account, the air flow rate, the biofilter layer height, the empty bed residence time of the pollutants (EBRT) and the concentration of the pollutants.

Empty bed residence time (EBRT) and true residence time τ are calculated according to Eqs. (1) and (2), respectively:

$$\mathrm{EBRT} = \frac{V_f}{Q} \tag{1}$$

$$\tau = \text{true residence time} = \frac{V_f}{Q} \times \theta \tag{2}$$

where Q is the air flow rate ($\mathrm{m^3\,h^{-1}}$), V_f the filter bed volume ($\mathrm{m^3}$) and θ the porosity ($1\,\mathrm{m^3}$ packing $\mathrm{m^{-3}}$ reactor).

The true residence time (τ) corresponds to the time necessary to the gas to pass through the porous medium. Typical values of EBRT for treating different pollutants in conventional biofilters usually range from 20 to 180 s [23], with a nominal EBRT of 60 s. Higher EBRT are required for soil-based media systems [24]. It is well known that the process of biofiltration is not cost effective for high EBRT values. These high EBRT values could moreover lead to a low removal efficiency and accumulation of dangerous intermediate products.

The gas flow rate is measured by a thermo-anemometer (VT 200, KIMO). Characteristics of the air (temperature, relative humidity) were also measured using a hygroscopic transmitting sensor and temperature, TH 300, KIMO.

The height of the filter medium does not generally exceed 1–1.5 m to limit the energetic costs necessary to balance the strong pressure losses (clogging).

To design EB, the main considered parameters are:

- concentration of the pollutant (ppmv or $\mathrm{mg\,m^{-3}}$);

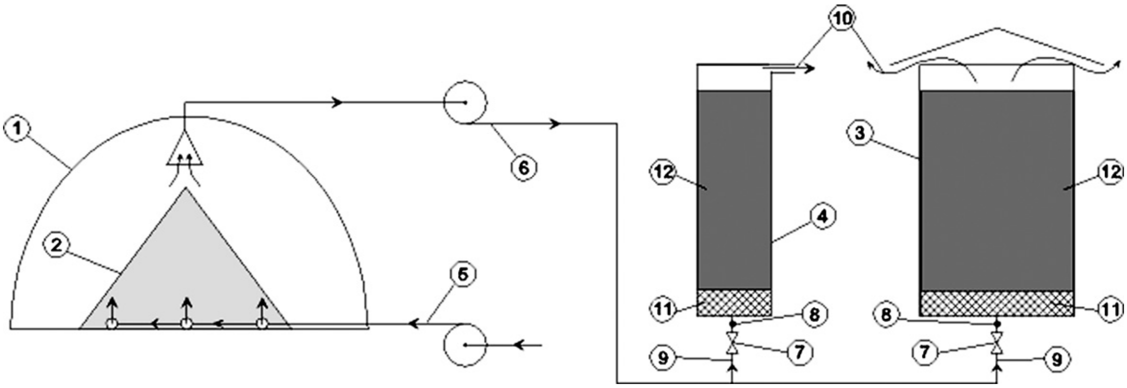

Fig. 1. Treatment installation at the R.O.M platform: 1, greenhouse; 2, sewage sludge/yard waste compost; 3, industrial biofilter (BI); 4, experimental biofilter (EB); 5, ventilation system of the windrow; 6, extraction system of waste gas; 7, valve; 8, measuring points (flow rate, temperature, relative humidity); 9, waste gas; 10, clean gas; 11, shredded wood; 12, compost SS/YW.

- EBRT (s);
- height of the filter medium (m).

The removal efficiency (RE) is the operational parameter generally used to evaluate the functioning of the biofilter as a function of the time. RE is calculated by Eq. (3):

$$\mathrm{RE}(\%) = \frac{C_i - C_o}{C_i} \times 100 \qquad (3)$$

where C_i is the inlet concentration (ppmv) and C_o, the outlet concentration (ppmv).

The mass load is the mass of the gas considered as the pollutant per unit volume of the filter medium and per unit of time. This parameter is calculated according to Eq. (4):

$$\text{Mass load}\,(\mathrm{g\,m^{-3}\,h^{-1}}) = \frac{Q}{V_f} \times C_i \qquad (4)$$

where Q is the air flow rate ($\mathrm{m^3\,h^{-1}}$); V_f, the filter medium volume ($\mathrm{m^3}$) and C_i the inlet concentration ($\mathrm{g\,m^{-3}}$).

Ammonia is a malodorous pollutant, usually emitted during aerobic composting of organic materials such as sludge [25]. Lau et al. [11] have measured concentrations of NH_3 between 0.2 and 50 ppmv (0.14 and 35 $\mathrm{mg\,m^{-3}}$) during composting of sludge and have obtained a removal efficiency of 95% with an EBRT of 55 s and an elimination capacity (EC) of 4.33 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$ (Table 1). Williams and Miller [26] have also mentioned that NH_3 is commonly present with concentrations higher to 35 ppmv (25 $\mathrm{mg\,m^{-3}}$) in composting plants. Chen et al. [27] have obtained efficiency removal of 97–99% for ammonia by biofiltration treatment with a packing material containing a non-inoculated compost, for concentrations ranging from 28 to 85 ppmv (20–60 $\mathrm{mg\,m^{-3}\,NH_3}$) with an EC of 12 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$ and for an EBRT of 60 s (Table 1). A biofilter containing compost, sludge and pieces of hard plastics reached a removal efficiency of more than 99.9% with an inlet concentration of ammonia of about 51 ppmv (35.7 $\mathrm{mg\,m^{-3}\,NH_3}$) and an empty bed residence time of 60 s [5]. Taking into account these studies, an average ammonia concentration of 50 ppmv (35 $\mathrm{mg\,m^{-3}}$) and an EBRT of 60 s were selected to design EB.

With a flow rate of 15 $\mathrm{m^3\,h^{-1}}$ and an empty bed residence time of 60 s, the volume of the filter medium is estimated at 0.25 $\mathrm{m^3}$. Taking into account a height of the filter medium of 0.5 m which is high enough to avoid channelling and dead volumes, the diameter of the biofilter research can be estimated at 0.8 m.

The values of the superficial velocity, the empty bed residence time, the true residence time and the operating conditions of the EB are given in Table 2 as well as the operating conditions used to run the Experimental Biofilter (EB).

The base of these biofilters (Fig. 1) is composed of wood chips that diffuse the gas to be treated to all the surfaces of the filter medium, made up of mature compost SS/YW (6 months age).

Table 1

Comparison of the overall performances of the biofilter used herein with other studies (adapted from Taghipour et al.[5]).

Authors	Bed media	Acclimation time (day)	EC	RE (%)	EBRT (s)
This study	Compost SS/YW	0	2.52 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	94	60
Galera et al. [58]	Rock wool-compost	nd[a]	6.44 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$ 12.05 $\mathrm{g\,H_2S\,m^{-3}\,h^{-1}}$	78.6 68.1	63–132
Pagans et al. [61]	Mature compost	0	8.29 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$ 7.17 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$ 61.3 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$ 21.7 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	98.8 95.9 89.5 46.7	86
Kapahi and Gross [62]	Compost, oyster shells and perlite	nd	10.6 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	96.4	nd
Gao et al. [63]	Compost-like biomass mixture	nd	0.18 $\mathrm{g\,H_2S\,m^{-3}\,h^{-1}}$	98.9	46
Lau et al. [11]	50% mature compost + 25% screened compost + 25% soil	nd	4.33 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	95	55
Schlegelmilch et al. [64]	Screened yard waste compost	7	4.37 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	100	80
Liang et al. [8]	Compost and activated carbon	15	0.02–0.391 $\mathrm{g\,NH_3\,kg^{-1}}$ media $\mathrm{day^{-1}}$	95–99.6	31.9–79
Pinnette et al. [9]	Compost, bark mulch, wood chips	nd	1 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	nd	nd
Taghipour et al. [5]	Compost, sludge, hard plastics	10	9.85 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$ 9.44 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	99.9 99.9	60 >30
Chen et al. [27]	Compost	1	12 $\mathrm{g\,NH_3\,m^{-3}\,h^{-1}}$	97–99	60

[a] nd: not determined.

Table 2
EB operating conditions.

Volumetric air flow rate ($m^3\,h^{-1}$)	15 ± 1
Superficial velocity ($m\,s^{-1}$)	0.50 ± 0.03
Empty bed residence time of pollutants (s)	60 ± 4
True residence time (s)	25 ± 4
Temperature (°C)	21.0 ± 0.3
Relative humidity of the inlet gas (%)	91 ± 3
Relative humidity of the outlet gas (%)	93 ± 3

Physico-chemical and bacteriological analysis were performed on these media at the beginning ($t = 0$ day), at the middle ($t = 12$ days) and at the end ($t = 27$ days) of the biofiltration treatment. This final time is the time necessary to obtain fermentation of the SS/YW mixture (composting). To achieve this, 8 samples of SS/YW mixture (5 g) were collected (1 sample every 20 cm in diameter (i.e. 4 samples) and 1 sample every 25 cm depending on the height of the filter medium (i.e. 2 samples)). Then all these samples have been mixed to form a representative sample, that was analysed within 24 h at the laboratory of the Compagnie d'Aménagement des Côteaux de Gascogne (CACG, Tarbes, France) according to the standardised method PR NF U44-51.

No analysis of the liquid phase was performed because there was no water addition and then no leachate was produced.

The quantity of VOSCs that can be adsorbed by the filter medium used in the industrial site was determined. The adsorption capacity of the filter medium has been studied under dynamic conditions with a column containing the same filter medium with in EB without the microbial activity responsible for oxidation reactions. Sterilization was performed by autoclaving (30 min at 120 °C), heating in an oven (48 h at 145 °C) and the material is then kept in a desiccators for 3 h. The waste gas continuously flushed through the system is a mixture of H_2S and MeSH with concentrations equivalent to 119 ppbv (168 $\mu g\,m^{-3}$) and 64 ppbv (128 $\mu g\,m^{-3}$), respectively.

The water content was determined by heating 5–10 g of compost to 103 °C. The principle of determining the water activity (a_w) is to identify at constant temperature the relationship between the water content of a sample thermodynamically equilibrated and the relative humidity of the ambient air set (HygroLab, Rotronic). The density of the compost (ρ) was determined by weighing a volume of 1 L of compost. Compost porosity (ε) is measured by the method proposed by Hodge and Devinny [28]. The granulometric distribution was established by sieving (sieve machine AS 200 Restsch) 500 mL of compost. Diameters of the meshes used were 1, 1.4, 2, 2.8, 4, 5.6, 7.1, and 9 mm.

2.3. Analytical methods

Composting of yard waste and sewage sludge was performed under a greenhouse, the outlet of the gas effluent being obtained by a system which transports the ambient air inside this greenhouse. Gas samples were collected and transported in Tedlar bags (3 bags of 3.8 L per sample) and were analysed within 48 h after sampling.

Sampling was performed the 5th day after composting, i.e. during the period for which emissions and smells were very important. This choice was made because emissions are most important during the thermophilic step (temperatures above 45 °C), especially in the case of ammonia [29]. During measurement of the temperature in the windrow compost, the maximum temperature ($T = 59$ °C) was reached during the 5th day.

The method used required preconcentration of the sample by micro-extraction on solid phase (SPME) before analysis by gas chromatography coupled to a mass spectrometer (GC Varian 3800/MS Varian 1200 Quad) [30,31]. This procedure allows to identify aldehydes, alcohols, acids, ketones, aromatic and terpenic compounds. The quantitative analysis of these compounds was obtained by comparison with the response of the GC/MS for toluene.

The compounds with low molecular weight, such as ammonia and hydrogen sulfide are not detected by this method. Moreover, ammonia and amines are also not easily identified by GC/MS because of their strong capacity to be retained on the column used. This is why at the inlet and at the outlet of the biofilter, ammonia is determined by colorimetric tubes (Dräger). EtSH, MeSH, Me_2S, Me_2S_2 and H_2S, were measured using a specific chromatographic analyzer (electrochemical cell, AirMEDOR). The electrolyte is a 10% chromic acid solution. During elution of sulfur compounds, an oxydo-reduction reaction occurs creating a potential difference at the electrodes. This difference was used to provide composition of VOSCs. This automatic method allows the determination of VOSCs at very low concentrations (at the ppbv level).

3. Results and discussion

3.1. Properties of exhaust air

The chemical analysis of the waste gas has been performed to determine families and concentrations of pollutants, types of odors, the odor threshold value and the corresponding threshold limit value.

Chemical analysis have showed that ammonia is the main emitted compound (above 70 ppmv or 49.6 $mg\,m^{-3}$) during composting. This pollutant was also found in large quantities in a composting sludge plant [11]. According to Rappert and Müller [32], ammonia is produced by degradation of dimethylamine (Me_2A) which is often present in municipal sewage or used in the production of pesticides and herbicides. Dimethylamine is easily degraded by microorganisms under aerobic conditions. The concentration is significantly higher than the threshold limit value fixed at 5.2 ppmv, this implies the presence of a real danger, particularly for workers of the industrial site.

Among sulfur compounds, the main VOSCs emitted by composting of the SS/YW mixture are MeSH and Me_2S with a maximum concentration (C_{max}) of 1780 and 1335 ppbv, respectively at the 5th day, as well as H_2S with $C_{max} = 62$ ppbv at the 27th day. Composting also releases EtSH (16 ppbv) and Me_2S_2 (45 ppbv) at the 5th day (Fig. 2).

MeSH is the major compound in the exhaust gas, this can be explained by the degradation of amino acids containing sulfur (cysteine, methionine, homocysteine and taurine) under anaerobic and aerobic conditions.

H_2S and MeSH were selected because their removal is often considered as an indicator of odor control in wastewater treatment applications [33]: in this waste gas ($d = 5$ days), H_2S was not detected, which is representative of good aeration conditions of the windrow at the beginning of composting. At the contrary, concentrations of Me_2S (1.34 ppmv), Me_2S_2 (0.15 ppmv) and MeSH (1.78 ppmv) are above the odor thresholds values (0.001–0.26, 0.004–0.008, and 0.02–0.025 ppmv, respectively), but they remain below the thresholds limit values (ND[1], 10 and 5.1 ppmv, respectively).

Evolutions of the VOSCs emitted by composting of urban sludge are different (Fig. 2). Indeed, compounds such as EtSH (from 16 to 0 ppbv) and Me_2S_2 (from 45 to 0 ppbv) decrease gradually during the process of composting and they are no more present in the gaseous effluent at the 10th day of composting. Me_2S also follows this behaviour with a concentration at the end of composting at the ppbv level. On the contrary, the H_2S concentration increases during composting to reach a mean value of 130 ppbv. Similarly to H_2S, MeSH is strongly emitted at the beginning of the compost-

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[1] Not defined.

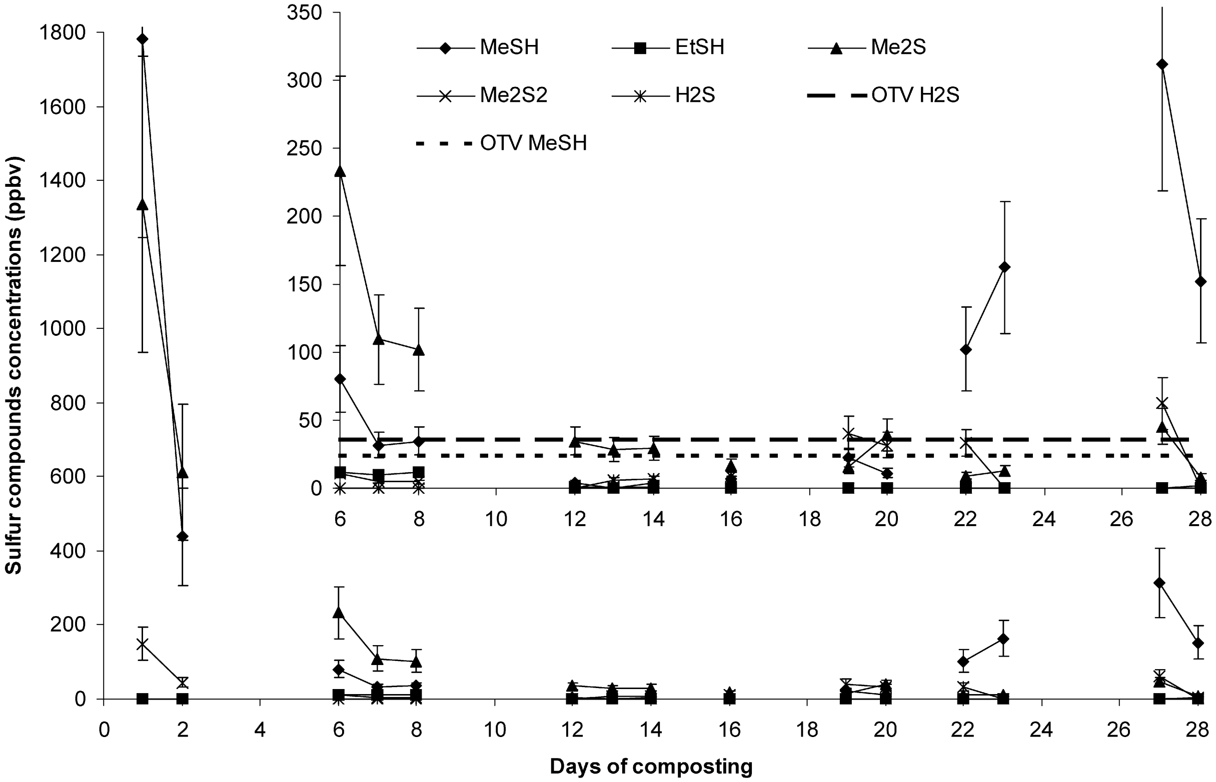

Fig. 2. Evolution of VOSCs emitted during composting (OTV: odor threshold value).

ing process. It is possible that anaerobic zones are created in the windrow at the end of the composting process. Thus, the forced ventilation does not seem sufficient and a mechanical reversal is thus necessary.

In this work, compounds from the ketones families were also measured such as acetone (0.73 ppmv) and 2-butanone (0.75 ppmv).

Independently from ammonia, compounds from family of VOSCs are the major compounds found in the waste exhaust air with a total concentration of 3.28 ppmv. These compounds are produced by microbial degradation of sulfur containing amino acids, methionine, cysteine and derivatives, or by thermal decomposition [32]. Indeed, VOSCs are responsible for 80–90% of odors emitted during composting of sewage sludge. This nuisance is mainly due to the very low odor thresholds value of these products [34].

Terpenes and oxygenated compounds are also present in the waste gas with concentration values of 1.32 and 1.54 ppmv, respectively. Terpene compounds are aromatic by-products formed during decomposition of lignin. Their presence is important in materials derived from wood such as barks of pine [35].

Although the compost used contains more green-waste than sewage sludge, VOSCs and ammonia are the major compounds measured. This is consistent with conventional processes for sludge composting.

Considering the important quantities of emitted NH_3 and VOSCs, these compounds were monitored at the moment for which the biological breakdown of this industrial effluent is observed.

3.2. Properties of filter media

3.2.1. Adsorption capacity

Results obtained are in agreement with those determined for concentrations of 40 ppmv H_2S with an organic material (pig manure and sand) [36]. If the inlet H_2S concentration is 100 ppmv, the adsorption capacity of the filter medium based on activated carbon is 15 mg g^{-1} dry weight whereas this value is only 0.75 mg g^{-1} dry weight for an organic-based filter. Furthermore, this adsorption capacity value is in agreement with values from literature confirming that SS/YW compost is not an effective adsorbent for H_2S and MeSH [37]. Therefore, the low adsorption of this SS/YW compost cannot be responsible for the emission of H_2S and MeSH at the end of the composting process.

3.2.2. Characteristics of the SS/YW compost

Water content is the most frequently used parameter to characterise compost medium since it can be easily measured and quite clear relationships have been reported between the moisture content and the biofilter performances [23]. Water content of the SS/YW compost (54.8%) is a parameter that plays a very important role in the process of biofiltration because microorganisms require water to produce a normal metabolic activity [38]. A water content in the range 50–80% is necessary for organic packing materials [39]. From a thermodynamical point of view, the energy necessary to keep the water molecules is usually well represented by the water activity. According to Gostomski et al. [40], to obtain a system microbiologically active, a water activity lower or equivalent to 0.9 is necessary. However, from studies performed with soil or compost-based biofilter, the ideal relative humidity has to be equal or higher than 99%, which corresponds to a water activity of 0.99. For a peat-based compost, Acuña et al. [41] have shown that water activities of 0.999 and 0.988 which correspond to water levels of 70% and 45%, respectively, are sufficient to promote the bacterial growth. In this study, a water activity of 0.998 was then retained.

The porosity and the bulk density are also two essential parameters for defining a filter medium. First, because the particle size and the void fraction are directly linked to the surface available for the biomass [42]. The determination of these two parameters allows the calculation of the pressure drop through the reactor and then

Table 3
Physico-chemical properties of the SS/YW filter medium obtained from the application of the PR NF U44-051 standard.

	Time (days)		
	d = 0	*d* = 12	*d* = 27
Aerobic microorganisms 30 °C (UFC g^{-1})	8.3×10^7	3.4×10^7	4.5×10^7
pH (±0.2)	6.3	6.1	6.2
Dry mass (±1.5%)	45.2	51.1	54.3
Total organic *N* (%)	2.56	2.79	3.32
N as nitrates (±1 mg kg^{-1})	16,936	14,733	9068
N as ammonium (±1 mg kg^{-1})	24	157	299
Sulfates (g) (±10%)	238	250	141

clogging. The bulk density of the SS/YW compost (400 kg m^{-3}) is lower than values found in the literature (514 [43], 568, 614, and 665 kg m^{-3} [44]). Clogging was however not observed during the 27 days of biofiltration. Porosity of the filter medium is quite low (41%) and is in agreement with values from literature [27,45] even if studies using compost as packing media in biofilters mentioned a higher porosity (close to 50%) [43,44,46].

Measurement of the particle size distribution of the SS/YW compost showed that 57% of the particles have a size higher than 4 mm. The size of the particles of the filter medium also plays a fundamental role on the behaviour of the biofilter according to Delhomenie et al. [44] who suggested a minimum size of 4 mm to minimize the pressure drop through the bed.

3.2.3. Physico-chemical properties of the SS/YW compost

The physico-chemical properties as the function of the time of biofiltration for the filter medium SS/YW used in EB are presented in Table 3.

Dry mass (mean value = 50%) and pH (mean value = 6.2) values do not change significantly with the duration of the treatment. On the contrary, values strongly vary for N as nitrates, N as ammonium and S sulfates.

3.2.4. Variation of the chemical forms of nitrogen in the filter medium during biofiltration

The physico-chemical properties of the SS/YW are presented in Table 3. During the biofiltration process, half of the initially present nitrate is removed. This significant decrease is probably due to microbial growth and denitrification leading to a conversion of nitrate into organic nitrogen or N_2. The increase in the percentage of nitrogen as organic form varies between 2.56% and 3.32%, this phenomenon is often identified during an anaerobic process [47]. In a study using biofiltration with ammonia contaminated air streams and compost containing pieces of hard plastics, the concentration of ammonium decreased while the nitrate concentration increased. The operational parameters such as the inlet concentration of NH_3 and the residence time are almost similar (EBRT = 60 s and $C_{NH_3} = 51$ ppmv) to those used in this study. In this case, the nitrifying microorganisms and the biological removal process are dominant [5]. This difference can be due to the pH of the compost used which is between 7 and 8.5 whereas the pH in the SS/YW compost used herein is lower (mean value is 6.2) (Table 3).

The life of the biofilter seems suggest that anaerobic regions were created. Such areas are usually located in bioreactors, particularly in the internal areas of biological flocculation and in the biofilm [47].

In presence of water and at pH 6, ammonia is transformed into ammonium in the biofilm. According to Chen et al. [27] and Baquerizo et al. [48], when the ammonium concentration increases, the pH decreases until acid values. This is not the case in this study that presents constant pH values (6.3–6.2) (Table 3). Chung et al. [49] have suggested that this phenomenon may be due to a chemical reaction of ammonium by sulfate. Indeed, ammonium in excess reacts with the degradation products of sulfur compounds (SO_4^{2-}) [29], this reaction maintaining a constant pH.

Therefore, it seems not necessary to add specific solutions to maintain the pH at a constant value because the SS/YW compost used in this study has a good buffer capacities.

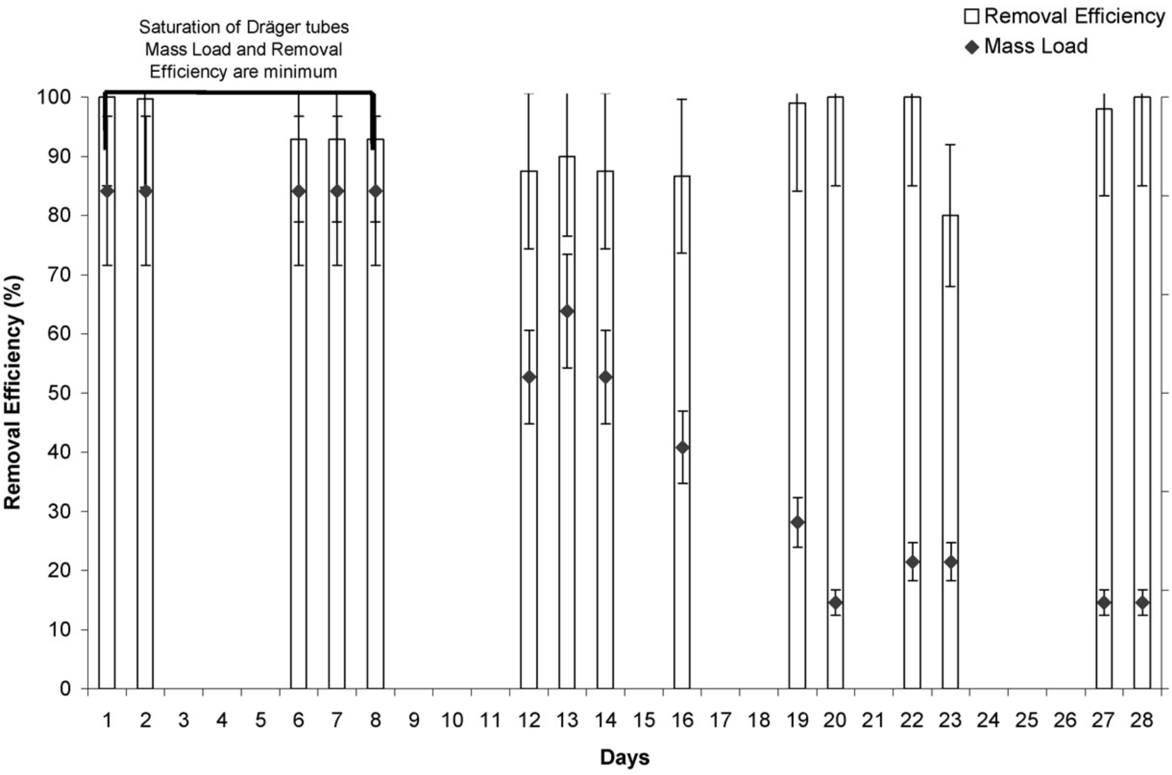

Fig. 3. Removal efficiency and mass load of ammonia.

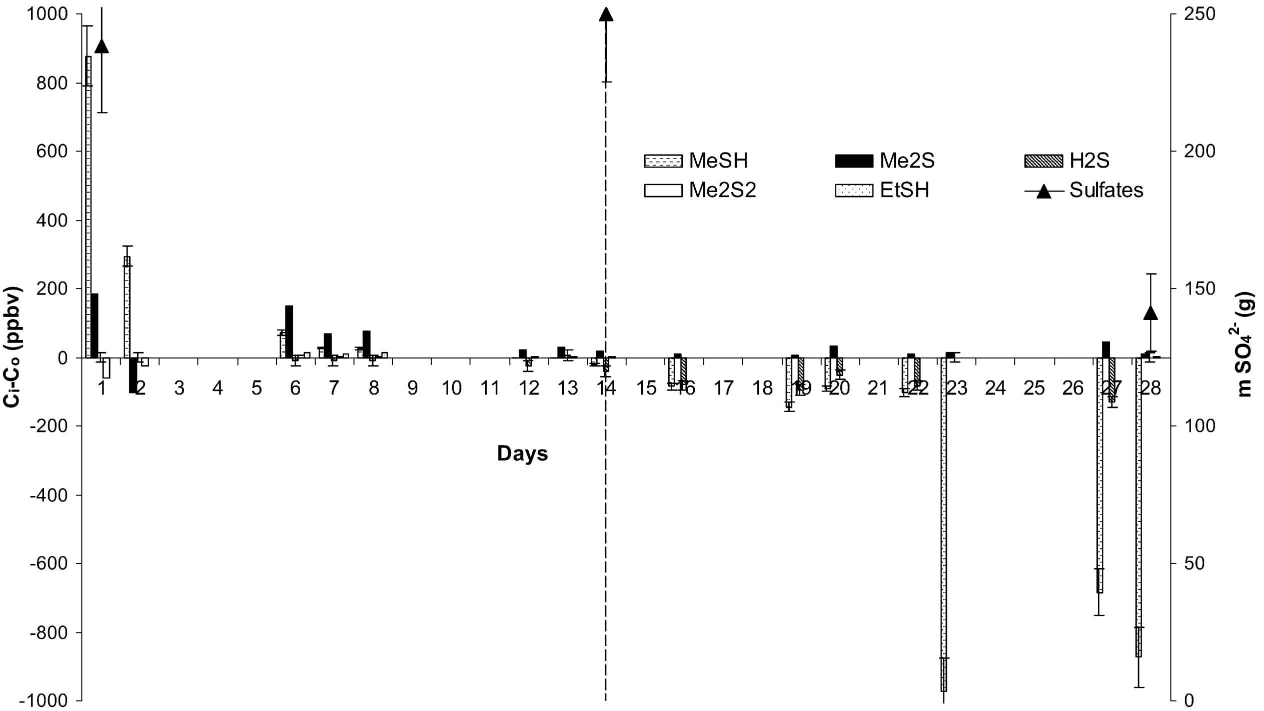

Fig. 4. Evolution of the VOSCs biodegradation and sulfates in the filter medium.

3.2.5. *Participation of sulfates*

During the first 12 days, an increase in the amount of sulfates (Table 3) has been observed, probably because of biodegradation of the sulfur compounds. Indeed, sulfates are products resulting from oxidation of sulfites [50]. The sulfur-oxidizing bacteria (such as *Thiobacillus thioparus*) developed in biofilters use H_2S to form sulfates [49]. In this study, these bacteria are able to use Me_2S, Me_2S_2 and ethylsulfur for their own development. These specific bacteria responsible for the oxidation of sulfur could not be quantified in this study but this is consistent with observations performed in the literature [50]. In the case of a membrane bioreactor containing heterotrophic microorganisms such as *Hyphomicrobium VS*, it was shown that these microorganisms can degrade Me_2S and produce $SO_4{}^{2-}$ [51].

Within 15 days after the first 12 days of biofiltration, half of $SO_4{}^{2-}$ initially present are reduced and this decrease can be correlated with the transformation of $SO_4{}^{2-}$ into other sulfur compounds that are not measured, such as elemental sulfur [52].

3.3. *Ammonia concentration*

The NH_3 emissions measured vary between 10 and 70 ppmv, this last value corresponding to the saturation of the instrument used for the measurement. The NH_3 inlet concentration at the biofilter gradually decreases, corresponding to the end of the process of composting. Indeed, during composting, nitrogen under an organic form is mineralized and transformed into ammonia gas (NH_3) and nitrogen gas (N_2). When this degradation is completed, i.e. at the end of the composting process, there are no more emission of NH_3.

Removal efficiency of ammonia reaches an average of 94% (Fig. 3). Whatever the inlet mass loads are (Eq. (4)), the removal efficiency is high with non-significant variations. Therefore, a biofilter

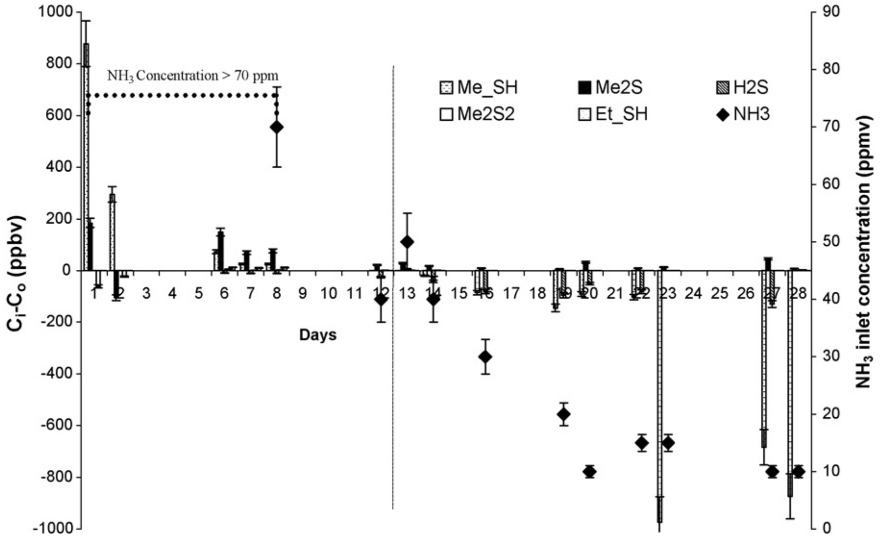

Fig. 5. Evolution of the VOSCs biodegradation and inlet concentration of NH_3.

containing a filter medium compost based on a SS/YW mixture is effective and stable for a NH_3 inlet mass loads ranging between 0.4 and 2.5 g m^{-3} h^{-1}.

For high NH_3 concentrations (50 ppmv), it was shown in the literature that biofiltration is not the best adapted process, taking into account the sensitivity of the nitrifying microorganisms [27,53]. When a biofilter containing organic materials (compost, peat, etc.) is used, it is however recognized that the removal efficiency could reach 100% with an inlet concentration of 213 ppmv [54]. Similar results were observed by Pagans et al. [6]: an ammonia removal efficiency of 96% was obtained in the biofilter containing mature compost that decreases for inlet concentrations higher than 2000 mg m^{-3} (or 2823 ppmv).

3.4. VOSCs biodegradation

3.4.1. Removal efficiency

From 6 or 7 days after the beginning of biofiltration, the removal efficiency of EtSH, Me_2S and Me_2S_2 have reached about 100%, whatever the inlet concentration is.

In the case of H_2S and MeSH, from the 14th day, it is not really a removal efficiency because the effluent at the outlet of the biofilter is higher than at the inlet ($C_o > C_i$) but they are sometimes eliminated. Nevertheless, the outlet concentrations remains below the average exposure value of 5 ppmv for these two compounds (5 times higher for MeSH and 25 times higher for H_2S). Goodwin et al. [33] have also observed that outlet concentrations of H_2S and MeSH higher than the inlet concentrations. This was explained by the anaerobic degradation of Me_2S_2, Me_2S and/or MeSH. Decrease in the removal efficiencies of Me_2S was shown in the case of a peat biofilter inoculated with *Hyphomicrobium* 155 with the formation of H_2S and MeSH [32]. In a peat biofilter inoculated with *Thiobacillus thioparus* DW44, the degradation efficiency for Me_2S was inhibited by the presence of MeSH but was increased in presence of hydrogen sulfide [55]. The presence of H_2S leads to incomplete oxidation of Me_2S, resulting in the formation of H_2S and MeSH. Indeed, Hirai et al. [56] have also reported that the removal of VOSCs can be inhibited by the presence of H_2S.

3.4.2. Participation of sulfates

To study the biodegradation of VOSCs, Fig. 4 presents the consumption (or "the production") of gaseous pollutants and sulfates in the filter medium (SS/YW compost).

Two distinct phases were observed. During the first 12 days, there is biodegradation of VOSCs ($C_i - C_o > 0$) with an increase of sulfates in the filter medium. After this period, the outlet concentration of VOSCs is higher than the inlet value ($C_i - C_o < 0$). At the same time, sulfates in the filter medium decreases. Therefore, a correlation seems occur between the amount of sulfates in the filter medium and the biodegradation of VOSCs.

3.4.3. Ammonia concentration and VOSCs biodegradation

The evolution of the VOSCs biodegradation and the inlet concentration of NH_3 is presented in Fig. 5. Fig. 5 shows that there is also a relationship between the inlet concentration of NH_3 and the production and/or the degradation of H_2S and MeSH.

When the concentration of NH_3 is high (above 30 ppmv), H_2S and MeSH are biodegradable ($C_i - C_o > 0$). This can be explained by the contribution of nitrogen to the development of microorganisms specific to VOSCs biodegradation. However, when concentrations of NH_3 are below 30 ppmv, conditions appear to be unfavorable for the activity of these microorganisms. During quantifying of the contribution of nitrogen, Chung et al. have showed that the H_2S efficiency removal is improved when NH_3 has a concentration two times higher than H_2S [49]. In this study, the concentrations of NH_3 and H_2S are much more important, the concentration of NH_3 is in the range of ppmv while the amount of H_2S is close to the ppbv level.

Smet and Van Langenhove [57] have also shown a correlation between the effectiveness of the removal of VOSCs and the presence of some compounds in the gas mixture to be treated. When 20 ppmv of ammonia is added, the degradation of Me_2S slightly increases, while an addition of 150 ppmv NH_3 inhibits this elimination.

Contradictory experiences are available in the literature. For example, Galera et al. [58] have shown that the ammonia efficiency removal was not affected by the quantity of H_2S but rather their respective proportions.

During the first step of the degradation process, when the NH_3 concentration is higher than 30 ppmv, VOSCs are biodegraded into sulfates. In a second step, the NH_3 concentration decreases with an emission of MeSH and H_2S by the biofilter while they were biodegraded in the first phase. In this second phase, microorganisms responsible for the biodegradation of VOSCs are not in conditions facilitating their activity.

There is a lack of data, particularly about elemental sulfur (S_0), to really understand why an oxidation first occurred followed by a reduction and a release of these compounds. Many hypothesis can be drawn to explain this phenomenon.

The decrease of $SO_4{}^{2-}$ can be related with the production of MeSH and H_2S. Indeed, microorganisms can reduce organic sulfur ($SO_4{}^{2-}$, $SO_3{}^{2-}$, etc.) to hydrogen sulfide [59].

Due to a lack of $NH_{3,}$ the biodegradation of Me_2S and Me_2S_2 can be incomplete, i.e. the production of $SO_4{}^{2(}$, and can therefore give by-products such as MeSH and H_2S.

According to Bentley and Chasteen [60], *Hypophomicrobium and Thiobacillus* sp. bacteria are able to transform VOSCs and particularly Me_2S into MeSH and formaldehyde because of the presence of the enzyme Me_2S monooxygenase under aerobic conditions.

4. Conclusion

- An experimental biofilter (EB) filled with a mature compost containing sewage sludge and yard waste (SS/YW) as filter medium was designed to treat an average ammonia concentration of 50 ppmv (35 mg m^{-3}) with an empty bed residence time of 60 s and a true residence time of 25 s.
- The biological process of fermentation during composting gives rise to the formation of ammonia (70 ppmv or 50 mg m^{-3}) and volatile organic sulfur compounds (VOSCs) with a total concentration of 3.28 ppmv. The ammonia concentration is well above the threshold limit value (5.2 ppmv) which implies a real risk for workers. For the sulfur compounds, the risk may be associated with complaints from residents because the Me_2S (1.34 ppmv), Me_2S_2 (0.15 ppmv) and MeSH (1.78 ppmv) concentrations are above the odor thresholds value.
- A mean removal efficiency of 94% was obtained for ammonia with an inlet mass loads between 0.4 and 2.5 g m^{-3} h^{-1}, corresponding to an inlet concentration between 10 and 70 ppmv.
- For the sulfur compounds, whatever the inlet concentrations are, the removal efficiencies of EtSH, Me_2S, and Me_2S_2 reached 100%. In the case of H_2S and MeSH, the outlet effluent is more concentrated than the inlet effluent. A relationship with ammonia concentration and VOSCs biodegradation has been shown. When the NH_3 concentration is high (above 30 ppmv), H_2S and MeSH are biodegraded. However, when the concentrations are below 30 ppmv, conditions appear to be unfavourable for the activity of the microorganisms specific to the biodegradation of H_2S and MeSH.
- The SS/YW mature compost as packing material of a biofilter is efficient, under specific conditions, for the simultaneous removal

of ammonia and VOSCs exhausted by industrial composting of sewage sludge and yard waste.

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