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Abatement of volatile organic sulfur compounds in odorous emissions from the bio-industry

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Abstract

Compounds of interest in this work are methanethiol (MeSH), dimethyl sulfide (Me_2S), dimethyl polysulfides (Me_2S_x) and carbon disulfide (CS_2) since these volatiles have been identified as predominant odorants in the emission of a wide range of activities in the bio-industry (e.g. aerobic waste water treatment plants, composting plants, rendering plants). In these processes, the occurrence of volatile organic sulfur compounds is mainly related to the presence of anaerobic microsites with consecutive fermentation of sulfur containing organic material and/or to the breakdown of the latter due to thermal heating. Due to the chemical complexity of these low-concentrated waste gas streams and the high flow rates to be handled, mainly biotechnological techniques and scrubbers can be used to control the odour emission. When using biofilters or trickling filters, inoculation with specific microorganisms and pH-control strategies should be implemented to optimise the removal of volatile organic sulfur compounds. In scrubbers, chemical oxidation of the volatile organic sulfur compounds can be obtained by dosing hypochlorite, ozone or hydrogen peroxide to the scrubbing liquid. However, optimal operational conditions for each of these abatement techniques requires a further research in order to guarantee a long-term and efficient overall odour abatement.

Abbreviations: AC – activated carbon; EC – elimination capacity; ECU – European Currency Unit; IBA – isobutyraldehyde; Me_2S – dimethyl sulfide; Me_2S_2 – dimethyl disulfide; MeSH – methanethiol; VOSC – volatile organic sulfur compounds; WWTP – waste water treatment plant

1. Introduction

Odour nuisance is caused by the emission of chemical compounds. Nasty smells are a nuisance for around 20 per cent of the population of the Netherlands (Walpot 1992). A comparative inquiry of the odour nuisance in Flanders in 1993 and 1997 suggests that the amount of complaints in this area is even growing (Van Langenhove & Beckers 1997). In this paper, the contribution of volatile organic sulfur compounds (VOSC) to the odour pollution in the bio-industry will be discussed. In Table 1, it is illustrated that VOSC have a very low odour treshold value and a very negative hedonic value and, as such, will contribute to odour pollution even upon the emission of very small amounts. In order to illustrate the widespread occurrence of these volatiles in the bio-industry, some emission sources and reported concentrations for VOSC are given in part 2. As a result of their widespread occurrence and odorous characteristics, odour abatement technologies in the chemically complex emissions of the bio-industry often focus on the removal of VOSC. In part 3, an overview of current abatement technologies for VOSC is given.

Table 1. Properties of some VOSC (OT = odour threshold value; $H_{25\,^\circ C}$ = dimensionless Henry coefficient at 25 °C $((\mathrm{mol.m}^{-3})_{air}/(\mathrm{mol.m}^{-3})_{water})$

Compound	OT (ppbv)	$H_{25^\circ C}$ (–)	Hedonic value
MeSH	0.9–8.5[1]	0.10[3]	decayed cabbage[4]
Me_2S	0.6–40[1]	0.07[3]	decayed vegetables[4]
Me_2S_2	0.1–3.6[1]	0.04[3]	putrification[5], foul[6]
CS_2	9.6[2]	0.65[3]	vegetable sulfide, aromatic[4]

Superscripts refer to references: [1] De Zwart & Kuenen 1992; [2] De Vos et al. 1990; [3] Przyjazny et al. 1983; [4] Verschueren 1983; [5] Bonnin et al. 1990; [6] Miller & Macauley 1988.

2. Bio-industrial emission sources of volatile organic sulfur compounds

In the bio-industry, biological organic matter (e.g. proteins, carbohydrates, fats) is produced or converted into valuable products. VOSC-emission in these processes is mainly related to the heating and/or the anaerobic decay of the sulfur-containing amino acids methionine and cysteine and derivates (De Zwart & Kuenen 1992). These emission sources will be discussed in 2.1. and 2.2.

2.1. Anaerobic processes

Under anaerobic conditions, microbiological fermentation of sulfur-containing compounds results in the emission of VOSC. Devai & Delaune (1995) reported that the production of MeSH, Me_2S and CS_2 in anaerobic salt marsh soil strongly increases with decreasing soil redox potential (E_h). A major increase in H_2S-emission was measured at $E_h < -100$ mV, while a significant increase in CS_2 emission was observed at E_h between -70 and -100 mV. Most sources of VOSC in the bio-industry as a result of anaerobic conditions are fundamentally aerobic processes where, however, the occurrence of anaerobic microsites cannot be avoided for one or another reason.

Several authors (Van Langenhove et al. 1985; Bonnin et al. 1990; Hwang et al. 1994; Laplanche et al. 1994; Hwang et al. 1995; Brennan et al. 1996) identified VOSC together with hydrogen sulfide as being the major odour stimuli in sewer pipes and aerobic waste water treatment plants (WWTPs). According to Paillard & Blondeau (1988), highly odorous emissions are generated in WWTPs when E_h decreases below -50 mV. According to Laplanche et al. (1994), the initial stages in the treatment (pumping, pretreatment,...) and the sludge treatment line (thickener, dehydration,...) contribute to 30 and 65%, respectively, of the odorous emissions. Gerards et al. (1996) measured concentrations of 30 ppmv Me_2S and 50 ppmv H_2S in the waste gas from an aerobic brewery WWTP, while Mansfield et al. (1992) identified Me_2S ($<$ 1 ppmv) at a WWTP. Hwang et al. (1995) reported that the dissolved concentration of H_2S, CS_2, MeSH, Me_2S and Me_2S_2 in a WWTP was higher in the primary effluent than in the influent due to anaerobic conditions in the primary settling tank and the sludge recycle. However, more than 95% of these sulfur compounds were eliminated in the subsequent activated sludge process due to biodegradation and air stripping.

Also aerobic composting facilities cause odour problems as a result of microbial activity in anaerobic microsites (Van Durme et al. 1992). According to Wilber & Murray (1990), anaerobic conditions in addition to elevated temperatures during the composting process (e.g. in very wet zones or due to a poor mixing of the starter material with the amendments) favour VOSC formation, while their emission is strongly decreased upon aeration. During the production of compost to be used as a mushroom cultivation substrate, H_2S, COS, MeSH, CS_2, Me_2S, Me_2S_2 and Me_2S_3 are the main odorous compounds emitted with concentrations ranging from 24 to 840 ppbv (Derikx et al. 1990). The total sulfur emission was reported to be 8.3 g of sulfur per tonne (fresh weight) of compost. According to Derikx et al. (1991), the production of H_2S, COS, MeSH and Me_2S in these plants is due to a biological process, contrary to the formation of CS_2 and Me_2S_2. In a pilot experiment, Smet et al. (1998b) measured CS_2, Me_2S, Me_2S_2 and methyl propyl disulfide during aerobic composting of biowaste, with a maximum Me_2S concentration of 3.2 ppmv (corresponding to a maximum Me_2S emission of 1.1 $\mathrm{g.tonne^{-1}d^{-1}}$) during the thermophilic composting phase (Figure 1). The total emission of VOSC during this experiment was 9.2 g of sulfur per tonne (fresh weight) of compost. Pöhle & Kliche (1996) measured Me_2S_2 concentrations up to10 ppmv during the aerobic composting of biowaste, and suggest to use Me_2S_2, together with limonene, as indicator compounds for the odour emission in these biowaste composting plants. Upon the storage of dewatered sludge, Sereno et al. (1993) mentioned that MeSH, Me_2S, Me_2S_2 were the primary odorous constituents. Bonnin & Sudry (1992) reported MeSH concentrations of 0.5-3 ppmv in a dewatering sludge room. Koe & Tan (1986) identified Me_2S_2 and Me_2S_3 in the off-gas of a sludge thickener, while

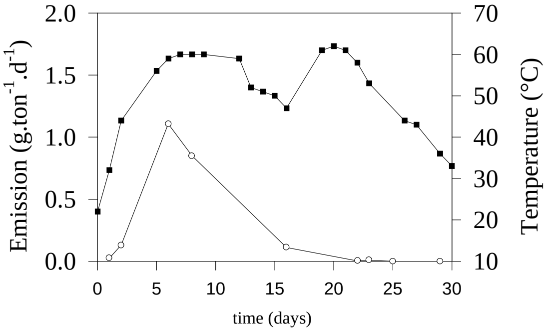

Figure 1. Emission (g.tonne^{-1}.d^{-1}) of dimethyl sulfide (○) and temperature of the composting material (■) versus time (days) during the aerobic composting of biowaste (after Smet et al. 1998b).

Wilber & Murray (1990) detected up to 10 ppmv of Me_2S and Me_2S_2 at a sludge composting facility.

According to Hobbs et al. (1997), MeSH, Me_2S, Me_2S_2 and Me_2S_3, together with 11 other odorants, contribute to the typical pig slurry odour. In animal breeding stations, Termonia et al. (1985) identified Me_2S, Me_2S_2, Me_2S_3, Me_2S_4, carboxylic acids, phenols and indole as being the main odorants. When anaerobic fermentation of the animal waste proceeded, enhancement of the concentration of dimethyl polysulfides occurred.

2.2. High temperature processes

In rendering plants, dead animal bodies and slaughterhouse waste are converted into valuable products such as glue, bone- and fishmeal, inedible grease and tallow (Van Langenhove et al. 1982). During the cooking process ($T > 105$ °C), obnoxious odours are generated, mainly attributable to the presence of VOSC, amines, acids, alcohols and aldehydes (Van Langenhove et al. 1982; Chélu & Nominé 1984; Prokop & Bohn 1985). In the non-condensable gaseous effluent collected from continuous rendering cookers, Chélu & Nominé (1984) measured average concentrations of 30 ppmv H_2S and 30 ppmv MeSH (with peak concentrations up to 800 ppmv H_2S and 200 ppmv MeSH), while McNeillie (1984) reported MeSH concentrations of 300 ppmv in these gases. Van Langenhove et al. (1992) measured in the raw cooking gases of a rendering plant 2-6 ppmv Me_2S_2 and 0.2-2 ppmv Me_2S_3, while Oberthür (1992) reported Me_2S_2 concentrations of 3-18 ppmv. According to the latter, the emission of VOSC in rendering plants is mainly due to the hydrolysis of feathers. According to Lecomte et al. (1995), 10 to 1000 g of sulfur- and nitrogen compounds are emitted to the atmosphere per tonne of treated material and per hour in a rendering factory, mainly as H_2S, MeSH, ethanethiol and NH_3.

In thermal sludge treatment plants, methyl sulfides, thiophenes, pyrazines and indole are responsible for sludge treatment odours (Schamp & Van Langenhove 1986, Frechen 1994). Poels et al. (1987) identified Me_2S, Me_2S_2 and Me_2S_3 in the exhaust gas of a thermal (max. 130 °C) drying unit for chicken manure. Other emissions of odorous sulfur compounds due to high temperature processes occur in the food and feed production technology, e.g. during the roasting of coffee (Schamp & Van Langenhove 1986).

3. Abatement technologies

Both biotechnological (3.1.) and physico-chemical (3.2.) technologies can be applied to treat the emission of VOSC. According to Frechen (1994), nearly 80% of all German WWTPs treat their emissions with a biotechnological method (biofilter: 59%, bioscrubber: 6% or treatment in an activated sludge basin: 13%), while the other abatement techniques used include chemical scrubbing (16%), adsorption on activated carbon (3%) and thermal oxidation (3%). According to the operators of these WWTPs, highest odour removal efficiencies are obtained using thermal oxidation, biofiltration and chemical scrubbing (Frechen 1994). According to Laplanche et al. (1994), the high flow rates to be handled at WWTPs result in the preferential use of biotechnological and scrubbing techniques in these plants. For similar and economical reasons, all aerobic biowaste composting plants in Flanders use biofilters for odour pollution abatement.

In the following, an overview of current abatement technologies for VOSC-containing waste gases will be given, with focus on biofiltration and chemical scrubbing.

3.1. Biotechnological methods

Both biofilters, biotrickling filters and bioscrubbers can be used for treatment of off-gases containing biodegradable compounds. An overview of these techniques, their operational limits and applicability was given by Van Groenestijn & Hesselink (1993). In this section, major attention will be given on the optimisation of the biofiltration of VOSC.

3.2.1. Biofiltration of VOSC

Introduction

Among the biotechnological waste gas techniques, biofiltration is the most common one. Contrary to bioscrubbers and biotrickling filters, biofilters are regularly used to treat off-gases in the bio-industry, e.g. in composting and rendering plants. In a biofilter, the gas to be treated is humidified and forced to flow through a bed packed with an organic carrier material (compost, peat, bark, or a mixture of these) on which micro-organisms are attached as a biofilm. In a biofilter, compounds with a dimensionless air-water partition coefficient H up to 10 ((mol.m^{-3})$_{air}$/(mol.m^{-3})$_{water}$) can be removed because the gas residence time (30-60 s) and the specific gas/liquid surface area (300-1000 m^2.m^{-3}) in a biofilter are high (Van Groenestijn & Hesselink 1993). Next to this, the apolar fraction in the organic carrier material promotes sorption and subsequent biodegradation of apolar compounds. As a drawback of this method, however, large reactor volumes are required to obtain high gas residence times mentioned and the operational control of the technique is limited due to the absence of a recirculating water phase.

Microbiological degradation of VOSC in a biofilter

Contrary to the biological removal of H_2S and numerous volatile organic compounds, the removal efficiency for VOSC in biotechnological waste gas treatment systems is reported to be rather low and variable (Tanji et al. 1989; Phae & Shoda 1991; Kasakura & Tatsukawa 1995). Van Langenhove et al. (1992) reported fluctuating biofiltration efficiencies of 0-81% for methyl sulfides at a rendering plant, whereas carbonyl compounds were very efficiently ($>$ 98%) removed. According to Cho et al. (1991a), H_2S-oxidizing micro-organisms are inherent in raw biofilters contrary to the micro-organisms degrading VOSC. Smet et al. (1996a) observed very low elimination capacities ($<$ 0.01 kg.m^{-3}.d^{-1}) for Me_2S in a wood bark and compost lab-scale biofilter. In order to improve these treatment efficiencies, numerous aerobic micro-organisms with degradative properties towards MeSH, Me_2S and Me_2S_2 were isolated (De Zwart & Kuenen 1992; Brennan et al. 1996; Smet et al. 1998a). Among these, mainly methylotrophic *Hyphomicrobium* spp. and autotrophic *Thiobacillus* spp. have been used to inoculate biotechnological waste gas treatment systems. These micro-organisms convert the methylated sulfur compounds into sulfate and carbon dioxide. De Zwart et al. (1997), however, described the coexistence of *Methylophaga sulfidovorans* and *Thiobacillus thioparus* T5 in a continuous culture, where *M. sulfidovorans* fully converted Me_2S to thiosulfate which was subsequently further oxidised to sulfate by *T. thioparus* T5.

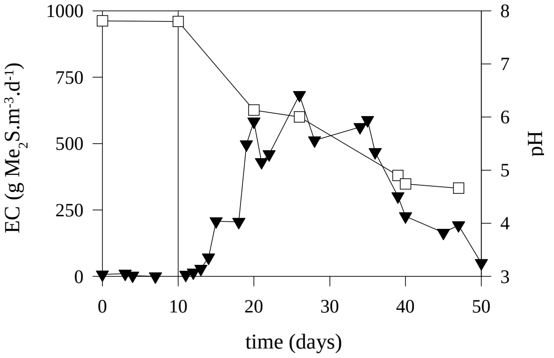

Figure 2. Elimination capacity (EC) of a compost biofilter for dimethyl sulfide (Me_2S) (g.m^{-3}.d^{-1}) (▼) and pH of the compost material (☐) before (days 0-10) and after (days 10-50) inoculation with *Hyphomicrobium* MS3 (after Smet et al. 1996a).

Contrary to the methylated sulfides, CS_2 supports only autotrophic growth, narrowing the range of bacteria that will use this compound as carbon and energy source (Plas et al. 1993). According to Smith & Kelly (1988), CS_2 oxidation by *Thiobacillus thioparus* TK-m proceeds by hydrolytic cleavage into COS and H_2S, while the COS formed undergoes a similar hydrolysis to CO_2 and H_2S. In this biodegradation pathway, the oxidation of H_2S to sulfate is the only energy-yielding process.

Table 2 overviews the Me_2S removal capacity in lab-scale biofilters and biotrickling filters, inoculated with some of these isolated micro-organisms. In Figure 2, the effect of inoculation on the elimination capacity of a biofilter for Me_2S is shown. In Table 2, the pronounced effect of the type of biofilter carrier material on the Me_2S-removal is illustrated for the inoculum *Hyphomicrobium* MS3. The better performance of the compost biofilter was due to the higher nutrient content and buffering capacity of this material in comparison with wood bark (Smet et al. 1996a).

Long-term stability of the biofiltration process for VOSC

Figure 2 illustrates the decrease in performance of the *Hypomicrobium* MS3-inoculated biofilter after a 30-day period due to acidification (accumulation of the metabolite H_2SO_4) in the biofilter. When the pH of the carrier material dropped below 5, the EC for

Table 2. Removal capacities (kg $Me_2S.m^{-3}.d^{-1}$) of biotechniques for Me_2S. The performance of the reactors is expressed as the maximum EC (EC_{max}) or as the removal efficiency (%) at the reported loading rate (after Smet et al., 1998a)

Reactor[a]	Inoculation	Removal capacity (kg $Me_2S.m^{-3}.d^{-1}$)	Reference
BTF (polypropylene)	*T. thioparus* TK-m	0.23 (95%)	Tanji et al. (1989)
BF (peat)	night soil sludge	0.08 (EC_{max})	Hirai et al. (1990)
BF (peat)	*T. thioparus* DW44	0.10 (EC_{max})	Cho et al. (1991b)
BF (peat)	*Hyphomicrobium* I55	0.11 (EC_{max})	Zhang et al. (1991)
BTF (polyurethane)	*Hyphomicrobium* VS	0.33 (EC_{max})	Pol et al. (1994)
BF (bark)	*Hyphomicrobium* MS3	0.03 (EC_{max})	Smet et al. (1996a)
BF (compost)	*Hyphomicrobium* MS3	0.68 (EC_{max})	Smet et al. (1996a)
BF (compost/dolomite)	*Hyphomicrobium* MS3	1.68 (EC_{max})	Smet et al. (1998c)

[a] BF = biofilter; BTF = biotrickling filter.

Me_2S in the biofilter was less than 50% of its maximum value. Regeneration of the acidified biofilter by trickling tap water or a phosphate buffer solution over the reactor was not effective (Smet et al. 1996b). Indeed, since the protons produced in the biofilter displaced the nutrient cations (e.g. Na^+, K^+,...) from the cation exchange sites on the compost material, 95% of the $SO_4{}^{2-}$ was leached as the corresponding sulfate salts and not as sulfuric acid. As a result of this, essential microbial nutrients were leached and the pH of the carrier material remained low. However, mixing limestone powder into the Me_2S-degrading compost biofilter was a successful approach to controlling the pH. A stoichiometric neutralisation reaction (molar ratio $CaCO_3/H_2SO_4$= 1.1) was observed between the $CaCO_3$ added and the metabolite of the Me_2S degradation. No sulfate toxicity was observed in the limestone-supplemented *Hyphomicrobium* MS3-inoculated biofilter, since the toxic sulfate concentration for *Hyphomicrobium* MS3 (= 30-40 g $SO_4{}^{2-}.L^{-1}$) was about 17 times higher than the maximum solubility of $CaSO_4$ (Smet et al. 1996b). For full-scale applications, however, mixing the carrier material with a neutralising agent at regular time periods is not a very practical way to control the acidification. Bonnin et al. (1994) obtained very high (1440-1680 $g.m^{-3}.d^{-1}$) elimination capacities (EC) in a biofilter for the acidifying inorganic volatiles H_2S and NH_3 using a pure carbonate material (mearl) as a biofilter carrier material. Smet et al. (1998c) used a similar carbonate carrier material (dolomite) for biofiltration of Me_2S. While direct inoculation of the dolomite particles with *Hyphomicrobium* MS3 was not successful, start-up of Me_2S-degradation in this biofilter was observed when the dolomite particles were mixed with 33% (wt/wt) of *Hyphomicrobium* MS3-inoculated compost. Under optimal conditions, EC up to 1680 g $Me_2S.m^{-3}.d^{-1}$ were obtained for the compost/dolomite biofilter (Table 2). No reduction in activity due to acidification was observed in the biofilter over a 235-day test period because of direct on-site neutralisation of the sulfuric acid with the carbonate in the dolomite material. However, nutrient limitation resulted in a gradual decrease of EC. In relation to this, stable EC for Me_2S were obtained when nitrogen was dosed to the biofilter at a Me_2S-C/NH_4Cl-N ratio of about 10.

Biofiltration of VOSC in mixed waste gases

Upon supplementation of isobutyraldehyde (IBA) (56-450 ppmv) as a second gaseous substrate to a *Hyphomicrobium* MS3-inoculated biofilter, Smet et al. (1997) observed sequential degradation profiles of first IBA and subsequently Me_2S in physically separated sections of the biofilter. It was found that the inoculum first degraded the carbonyl compound in the presence of both Me_2S and IBA. Complete purification of a waste gas stream containing both IBA and Me_2S should be performed using two-stage biofilters with IBA-degradation in the first and Me_2S-degradation in the second inoculated biofilter. The addition of toluene (± 50 ppmv) or limonene (20-100 ppmv) as a second substrate was not affecting the Me_2S-degradation rate in the biofilter (Smet et al. 1997). Zhang et al. (1991) reported a decrease in Me_2S-removal efficiency in a *Hyphomicrobium* I55-inoculated peat biofilter from 80% to 25% by the co-presence of hydrogen sulfide and methanethiol due to a preferential degradation of the latter. Also Hirai et al. (1990) reported that the removal of VOSC can be inhibited by the presence of H_2S. In a peat biofilter inoculated with *Thiobacillus thioparus* DW44, however, the elimination capacity for Me_2S was inhibited by the presence of methanethiol, but accelerated by the presence of hydrogen sulfide (Cho et al. 1991b).

Following supplementation with a 20 ppmv NH_3-concentration for 24 hours, Maes (1997) reported that the Me_2S degradation in a *Hyphomicrobium* MS3-inoculated compost/dolomite biofilter increased slightly from 270 $g.m^{-3}.d^{-1}$ to 295 $g.m^{-3}.d^{-1}$. However, the addition of 150 ppmv NH_3 to this biofilter during a 6-day period resulted in a complete inhibition of the Me_2S-removal (Figure 3). In this Figure, it can be seen that the pH of the carrier material strongly increased as a result of sorption of NH_3. About 10 days after this NH_3-supplemention, the biofilter recovered and even surpassed its initial Me_2S-removal rate. It was stated that NH_3 becomes toxic for *Hyphomicrobium* MS3 at a concentration between 20 and 150 ppmv. At lower concentrations, however, the pH-neutralising and nutritional effect of NH_3 is stimulating the Me_2S-degradation in the biofilter (Maes 1997). In a VOSC-loaded *Thiobacillus thioparus* DW44-inoculated peat biofilter, the presence of NH_3 (5-20 ppmv) in the waste gas resulted in a pH-correction because of its chemical reaction with the metabolite sulfuric acid (Cho et al., 1992). On the other hand, several authors (Bremner & Bundy 1974; Juliette et al. 1993) reported that VOSC inhibit the nitrification of ammonia, since they are a substrate for ammonia monooxygenase in *Nitrosomonas* spp. According to Bremner & Bundy (1974), carbon disulfide is a very effective inhibitor of nitrification, while Juliette et al. (1993) reported Me_2S and Me_2S_2 to be weak inhibitors.

3.2.2. Biotrickling filtration and bioscrubbing of VOSC

Introduction

A bioscrubber consists of a scrubbing tower where the waste gas is contacted with a water phase, while the clean-up of the contaminated water phase occurs in a bioreactor. In a biotrickling filter, the waste gas is forced through a packed bed filled with a chemically inert carrier material allowing microbial colonisation. Contrary to biofilters, bioscrubbers and biotrickling filters are only capable of removing compounds with an air-water partition coefficient lower than 0.01 (Dolfing et al. 1993; Van Groenestijn & Hesselink 1993). Taking into consideration Table 1, it can be expected that the removal of VOSC will be critical. However, a big advance of these liquid systems is that the recirculating water phase allows a better control of the reaction conditions (pH, temperature, addition of nutrients, removal of accumulated salts).

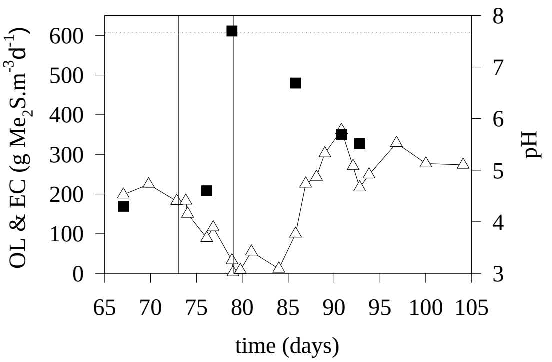

Figure 3. Organic loading rate (OL) (dashed line) and elimination capacity (EC) (△) of a compost biofilter for dimethyl sulfide (Me_2S) ($g.m^{-3}.d^{-1}$) and pH of the compost material (■) before (day 65–73), during (days 73–79) and after (day 79–105) addition of 150 ppmv NH_3 during a 6-day period.

Removal of VOSC in a trickling filter

Kasakura & Tatsukawa (1995) reported high (> 90%) elimination efficiencies for both H_2S and VOSC using a two-stage trickling filter with separate degradation of H_2S and the organic sulfur compounds. In the first stage, a low pH allowed efficient H_2S biodegradation, while the organic sulfur compounds were removed in the second stage where the pH was controlled at a value of 6-7. Lee & Shoda (1989) used activated carbon (AC) as a microbial carrier and obtained, upon inoculation with night soil sludge, complete removal of MeSH up to a load of 0.72 g $MeSH.kg^{-1}$ $AC.d^{-1}$. The application of CS_2 degrading *Thiobacillus* spp. in waste gas treatment systems was illustrated by Berzaczy et al. (1988) and Revah et al. (1994), who obtained removal rates up to 1.7 and 6.0 kg $CS_2.m^{-3}.d^{-1}$, respectively, in inoculated biotrickling filters. No results were found for the abatement of VOSC using a bioscrubber.

3.2. Physico-chemical methods

Among the physico-chemical methods, mainly chemical scrubbing is used to control the emission of

VOSC. Other methods include masking, adsorption and incineration.

3.2.1.A. Scrubbing

Scrubbing aims to transfer the pollutant from the gas to the aqueous phase by intense contacting of the polluted air stream with a water phase. The mass transfer depends on the concentration and air/water partitioning (Henry) coefficient of the volatiles (Table 1) and the mass transfer resistance of the scrubber system. Because of the high Henry-coefficient of VOSC, scrubbing these compounds from a waste gas stream with water requires a high liquid flow rate to gas flow rate (L/G). However, the scrubbing efficiency of VOSC can be enhanced by making the water alkaline, by their oxidation to more water soluble compounds or by the use of other scrubbing liquids.

Alkaline scrubbing can be applied for the abatement of MeSH (pK = 9.7) provided the pH of the scrubbing liquid is higher than 11 (Laplanche et al. 1994). As a drawback of this method, CO_2-coabsorption will result in a high alkaline consumption cost and precipitation of $CaCO_3$ and $MgCO_3$ can result in clogging of the scrubber when the pH of the liquid is in the carbonate range ($pH > 10$). Next to this, desorption of MeSH from the liquid is possible due to the reversibility of the reaction.

Hypochlorite is considered to be the most efficient oxidant for the chemical scrubbing of VOSC (Van Durme et al. 1992; Sereno et al. 1993). Depending on the pH of the scrubbing solution, the active compound is hypochlorite (OCl^-, $pH > 6$), hypochlorous acid (HOCl, pH 2-6) or chlorine water (Cl_2, $pH < 2$). According to Laplance et al. (1994), MeSH is oxidised to methane sulfonic acid ($MeSO_3H$) via Me_2S_2 as an intermediate. The authors reported a chemical consumption of 9 moles hyphochlorite and 7.6 moles NaOH for the oxidation of 1 mole MeSH. Since HOCl has a higher oxidation potential towards VOSC in comparison with OCl^-, Muirhead et al. (1993) and Hentz et al. (1990) preferred a pH 6.5 in the hypochlorite scrubber treating compost waste gases. Sereno et al. (1993), however, reported that best odour removal efficiencies at a sludge storage unit were obtained using a pH of about 11 in the hyphochlorite scrubber.

Since NH_3 and lower amines are preferentially chlorinated to more odorous or toxic compounds (chloroamines) rather than being oxidised by hypochlorite, these nitrogen compounds have to be removed from the gas phase before contacting it with hypochlorite, e.g. by an initial acid (pH 2-6) scrubber (Van Durme et al. 1992; Muirhead et al. 1993). It should, however, be ensured (e.g. by using a demister) that droplets from the acid scrubber do not enter the oxidative scrubber, since these droplets can result in interference of the VOSC-removal (Thompson et al. 1995). Next to this, hypochlorite oxidation will produce strong chlorine and burnt organic odours when operated at too low pH-values (Pincince 1992). Sereno et al. (1993) reported production of small quantities of methylene chloride, chloroform, 1,2-dichloroethane and trichloroethylene in a hyphochlorite scrubber at pH 6.1. According to Thompson et al. (1995), the emission of these compounds can be prevented using a subsequent alkaline hydroperoxide scrubber.

Valentin (1993) and Kelly & Butcher (1997) reported very high odour removal efficiencies using a single alkaline hyphochlorite scrubber in combination with a fixed bed nickel-based catalytic reactor, through which the scrubbing liquid is recirculated. In this combination, acid pre-scrubbing and caustic post-scrubbing was not necessary and oxidation reaction rates strongly increased.

Muirhead et al. (1993) obtained good odour removal from composting waste gases with a two-stage packed tower scrubber using acid (pH 2.5) in stage I and hypochlorite (pH 6.7) in stage II. In this system, Me_2S/Me_2S_2 (15 ppmv), mercaptans (2 ppmv), amines (20 ppmv) and ammonia (300 ppmv) were removed up to 99%. The authors reported a high chemical consumption of 4.15 ECU H_2SO_4(1 European Currency Unit $\cong$ 1 US Dollar) and 15.20 ECU NaOCl per tonne of dry biosolids composted. In a similar two-stage scrubber installed at a WWTP, Bonnin et al. (1990) reported a 90% and 99% removal efficiency for nitrogen and sulfur compounds (total sulfur concentration: 14.6 $mg.m^{-3}$), respectively. Thompson et al. (1995) reported very high removal efficiencies (MeSH: 100%; Me_2S: 100%; Me_2S_2: 85%; NH_3: 99.5%) at a composting plant using a 3-stage (acid (H_2SO_4), oxidative (HOCl/pH 6.5) and final wash (NaOH, H_2O_2)) scrubber. Because of the slow reactions, however, the oxidative scrubber had to be dimensioned at a high gas residence time of 13 s. Although high ($> 90\%$) odour removal efficiencies were obtained in both an alkaline and acid hypochlorite scrubber (oxidant concentration 600-2000 ppm) at a sludge storage unit, Sereno et al. (1993) reported the removal of volatile organic compounds to be very poor.

In aqueous solutions at ordinary temperatures, hydrogen peroxide (H_2O_2) converts Me_2S quantitatively to dimethyl sulfoxide (Me_2SO), while dimethyl sulfone (Me_2SO_2) is formed at higher temperatures and excess H_2O_2 (Adewuyl & Carmichael 1986). According to these authors, this reaction is first order with respect to both H_2O_2 and Me_2S and is subject to catalysis by strong acids ($pH < 1$). The catalytic effect of HCl was found to be twice that of H_2SO_4 at 20 °C. In a similar way, MeSH and Me_2S_2 can be oxidised by H_2O_2 to methyl sulfonic acid ($MeSO_3H$) (Adewuyl & Carmichael 1986). McNeillie (1984) reported complete removal of amines and mercaptans in rendering waste gases using an alkaline ($pH > 9.6$) hydrogen peroxide (H_2O_2) solution (0.1-0.8 $g.l^{-1}$) in scrubbing stage I and an acid ($pH < 5.0$) H_2O_2 solution (0.5-0.8 $g.l^{-1}$) in scrubbing stage II. Unspecified catalysts (3-10 $mg.l^{-1}$) were added to promote this oxidation process. According to these authors, the chemical cost for this two-stage peroxide scrubber amounted to only 63% of the corresponding two-stage hypochlorite scrubbing process (McNeillie 1984). According to Sereno et al. (1993), however, peroxide scrubbing at low (pH 5.0) or high (pH 9.6) pH-values is not effective against odorants from a sludge storage unit, but should only be used as a polishing unit after an acid hyphochlorite scrubber.

Because of its decomposition into OH-radicals, ozone (O_3) is a powerful oxidant as well (Laplanche et al. 1994). The decomposition of O_3 is promoted at higher pH-values. In the water phase, sulfur containing compounds are efficiently oxidised with ozone (Anderson 1984; Hwang et al. 1994) while gas phase oxidation reactions are usually too slow to be of interest, except for H_2S (Anderson 1984). According to De Guardia et al. (1996), oxidation products of VOSC with O_3 include sulfonic acids, sulfones, acids and sulfonic anhydrides. As was found for the chlorination of MeSH, the ozonation of MeSH results in the accumulation of methane sulfonic acid ($MeSO_3H$) via Me_2S_2 as an intermediate (Hwang et al. 1994). Since O_3 is a sparingly soluble gas, specialised techniques are required to achieve a high degree of dissolution (Paillard and Blondeau 1988). Laplanche et al. (1994) measured a chemical consumption of 1.5 mole O_3 and 7 mole NaOH for the oxidation of 1 mole MeSH, yielding a significantly lower reagent cost in comparison with hypochlorite oxidation. Recently, De Guardia et al. (1996) described the use of silicone oil as a scrubbing liquid for the absorption of MeSH and Me_2S_2 from the gas phase. The absorbed sulfur compounds were subsequently oxidized with O_3 in the silicone oil, while the metabolites of the oxidation process were extracted from the solvent phase by water injection. The silicone oil used was stable towards O_3.

The use of chlorine dioxide (ClO_2) and potassium permanganate ($KMnO_4$) as an oxidant is more expensive in comparison with hypochlorite. Moreover, ClO_2 produces toxic chlorine in the exit gas, while the use of $KmnO_4$ results in the formation of MnO_2 precipitates which can result in clogging of the reactor.

3.2.1.B. Masking

Masking agents as e.g. terpenes can be added to discontinuous or small volumes of odorous emissions in order to overcome their environmental nuisance. However, the application of these products becomes dangerous if they mask the odour of high and/or toxic concentrations of volatiles (Paillard & Blondeau 1988). Moreover, when dilution of the masking agent to below its threshold value occurs before the malodorous compounds have reached their odour threshold value, odour problems can arise at a certain distance from the source (Anderson 1984). In addition to masking agents, other deodorisation products are commercially available. Some of these act as chemicals (acid/base reactions) and can be effective against H_2S and MeSH, while others have enzymatic properties. There are also indications that counteracting agents can reduce the intensity of smells (Laffort 1994). In general, however, the effectiveness of these commercial products is not yet well documented. Dosing a neutralising chemical (6-60 ppm) with a fruity, citrus odour to the scrubbing liquid at a dewatered sludge storage, Sereno et al. (1993) obtained a similar (50%) odour removal as using water alone. According to the authors, however, the hedonic value of the waste gas should be taken into account to evaluate these products. Mehmood-ul-Hassan (1997) tested 9 commercially available deodorising products for their activity towards volatiles. None of them was found to be active towards Me_2S, while 4 of them showed some activity towards 2-propanethiol. In the headspace of most of these deodorising products, benzaldehyde was found to be present as a masking agent.

3.2.1.C. Adsorption

Among the available adsorbents (activated carbon, silica gel, zeolites, activated alumina, synthetic resins,...), activated carbon (AC) is most often used for the removal by physical adsorption of volatile organic compounds and hydrogen sulfide (Turk et al. 1989). Physical adsorption is maximised by using AC with a high specific surface (750-1500 $m^2.g^{-1}$) and a significant portion of its total pore volume in the micropore range (less than 2.5 nm diameter) (Turk et al. 1989). Regeneration of the AC should be implemented by desorption of the volatiles. Several regeneration methods for AC are developed and/or currently investigated, such as steam or hot inert gas injection, chemical or solvent washing, induction heating (Joule effect, electromagnetic induction) and biological oxidation (Mocho et al. 1995).

According to Hwang et al. (1994) and Chélu & Nominé (1984), AC provides a high sorption capacity towards hydrogen sulfide and VOSC, while adsorption of volatile nitrogen compounds is poor. Hinokiyama et al. (1991) reported that the presence of H_2S in the waste gas strongly affected the breakthrough time of MeSH on AC. To optimise the adsorption of H_2S and MeSH, AC can be impregnated with NaOH or KOH to neutralise the adsorbed compounds (Turk et al. 1989; Le Bec et al. 1995). These basic compounds, however, also react with CO_2 in the air to form carbonates, thus decreasing the process performance. Turk et al. (1989) observed that an unimpregnated AC, used in conjunction with a small side stream of ammonia gas (7 - 50 ppmv) as a catalyst, was much more efficient in comparison with KOH impregnated AC. Tests with MeSH offered an advantage in capacity for the ammonia/AC process of 800% over unimpregnated AC and of 300-600% over KOH impregnated AC (Turk et al. 1993). Similarly, the addition of a few ppmv of ozone to sulfur-containing process gases prior to the AC adsorption was found to increase the bed life considerably, presumably as a result of the oxidation of the sulfur compounds by the adsorbed O_3 (Dorling 1980; Anderson 1984).

As a very general guide, a residence time of 1 s in the adsorption bed can be applied in odour treatment applications, corresponding to 0.1 kg $AC.(m^3.h^{-1})^{-1}$ (Dorling 1980). For process gases containing dust, water droplets or high humidity ($> 75\%$), AC adsorption is often not useful without some form of pretreatment, e.g. filtering or preheating the waste gas (Dorling 1980; Anderson 1984; Paillard & Blondeau 1988).

3.2.1.D. Incineration

Incineration is a very efficient, but expensive waste gas technology when pollutant concentrations are low and flow rates are high. Thermal incineration requires temperatures up to 700-1000 °C with gas residence times of 0.5-1 s, while catalytic incineration can take place at 300-450 °C (Paillard and Blondeau 1988). However, the catalyst can be poisoned by the presence of sulfur compounds as H_2S and SO_2. Besides this, the thermal oxidation of reduced sulfur compounds may lead to SO_2 emission, which requires an additional posttreatment to prevent environmental deterioration by e.g. acid rain (Tichy et al. 1998). According to Prokop and Bohn (1985), incineration of rendering waste gases in the rendering boiler is a valid treatment option provided the volume of air to be treated is kept minimal and in balance with the boiler size and combustion requirement for the burner.

Conclusions

Because of their very low odour threshold value (ppbv range) and negative hedonic value, the presence of volatile organic sulfur compounds in bio-industrial emission sources should be kept to a minimum or efficient abatement technologies have to be installed. Because of the characteristics of these waste gas streams, mainly biotechnological (biofiltration) and chemical scrubbing (oxidative scrubbing using hypochlorite, hydrogen peroxide, ozone) technologies can be efficient in controlling the emission of VOSC. However, no standard abatement technology is available for VOSC-contaminated waste gases since the optimal solution depends on several factors, as e.g. the qualitative and quantitative composition of the waste gas, the flow rate and temperature of the waste gas, available space on the plant, investment and operational costs of the abatement technology, availability of an incinerator on the plant, continuity of the gaseous emissions, legislation. Moreover, pilot plant experiments should be performed to select for each specific problem the optimal (combination of) abatement technology(ies) and optimal reaction conditions, which can, in combination with a thorough prevention strategy, control environmental problems caused by VOSC.

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