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Improving Biotreatment Efficiency of Hot Waste Air Streams: Experimental Upgrade of a Full Plant

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Biological methods as bio and biotrickling filtration are an energy-efficient and economical alternative to treat biodegradable odorants and volatile organic compounds (VOCs) in order to obey stringent releases regulations that have arisen during the last few decades. In this work a plant upgrade case study, employing these techniques, is presented. It refers to a critical situation in which off air streams, characterized by medium odorous compounds loads and high temperatures, were treated using a biofilter only. In that context, sufficient removal efficiencies were not achieved. Therefore, it has been proposed to replace the existing biofilter by a biotrickling one implementing a minimal number of structural plant modifications.

1. Introduction

Over the past decades, impact of air pollution on human health and environment has received an increasing interest from public, government and industry. As a result, stringent regulations governing the releases of volatile organic compounds (VOCs), reduced sulfur compounds (RSCs) and hazardous air pollutants (HAPs) have arisen, leading to both the development of new air pollution control technologies and the improvement of existing ones (Shareefdeen and Singh, 2004).
The treatment of off-gases has been practiced for years basing on non-biological methods such as condensation, activated carbon adsorption, absorption and incineration. Such methods show different efficiency degree according to the main characteristics of the air stream to be treated.
In recent years biofiltration, that was originally aimed at removing low odorous concentrations (Kennes and Veiga, 2010), has received increased attention as a low-cost, energy-efficient and effective method to treat waste air streams containing low/medium concentrations of biodegradable compounds. Different industrial sectors including rendering, food processing, flavour manufacturers and composting facilities have selected biofilters to convert gas phase chemical compounds into common biological degradation products such as carbon dioxide, water and mineral salts. In biofilters, as the contaminated air is passed through a bed of media, the contaminants and oxygen are first transferred to the biofilm formed on the surface of the media particles and, then, metabolized by bacteria. In order to sustain microbial growth, moisture is provided by saturating the processed air before it enters the biofilter unit. Moisture is also provided by intermittent and occasional spray irrigation of the media. The media within a biofilter are normally composed of natural material (e.g. peat, wood bark, soil, compost,

pine leaves, coconut fibre, etc.) or synthetically manufactured media (e.g. lignite, lava rock, activated carbon, etc.).
Even biotrickling filtration is one of the many promising biological techniques for odour and VOC control. The trickling liquid provides moisture, salts, metabolites and supplemental nutrients to the process culture; moreover it is a convenient mean to control pH. Biotrickling filters show several advantages over biofiltration technologies for air pollution control: little bed height limitation (up to 2 - 3 m); smaller footprints; packing longevity over 10 y; lower pressure drops due to high media porosity; easy control of temperature, pH, salt concentration and metabolites accumulation; wider range of treatable pollutants (Shareefdeen and Singh, 2004). For all these features biotrickling filters can be used both in combination and in substitution of conventional biofilters to improve odour control and diminish VOCs emission.
In this work a plant upgrade case study is presented. It refers to a critical situation in which off air streams, characterized by medium VOCs loads and high temperatures (over 45 °C), were treated using a biofilter only. In that context, sufficient removal efficiencies were not achieved. Therefore, the existing biofilter has been replaced by a biotrickling filter taking care to implement a minimum number of structural plant modifications.

2. Case Study Overview

At the end of 2009, a composting plant aimed to process municipal organic solid wastes (about 20,000 kg h^{-1}) has been realized. This plant was very close (about 75 m) to another facility that often suffered from odor impacts caused by the first one.
Off-gases coming from the stabilizer (a reactor dedicated to the aerobic fermentation of the solid wastes), rich of VOCs and odorous compounds, needed for a further treatment before being discharged directly into the atmosphere. Therefore, off air streams were conveyed to a pre-humidifying chamber and, then, to a biofilter using bioactivated wood barks as organic carrier. The most critical aspect of the contaminated air stream coming from the stabilizer was its relatively high temperature: about 48 °C. Such a temperature was too high to permit air stream to be treated into a simple biofilter because the employed bacterial population is generally mesophilic (good degrading performances between 10 - 40 °C). To face such a problem, external clean air (about 30 % v/v of the contaminated one) was mixed with the exhausted gases coming from the stabilizer, just before entering the biofilter, hence lowering the inlet air temperature to about 41 °C. A simple plant sketch with process streams characteristics, before the modification, is reported in Figure 1a.
Despite dilution effect and biotreatment, the biofilter outlet concentration of VOCs and odorous compounds was not satisfying and periodic complains from nearby facility workers were registered.

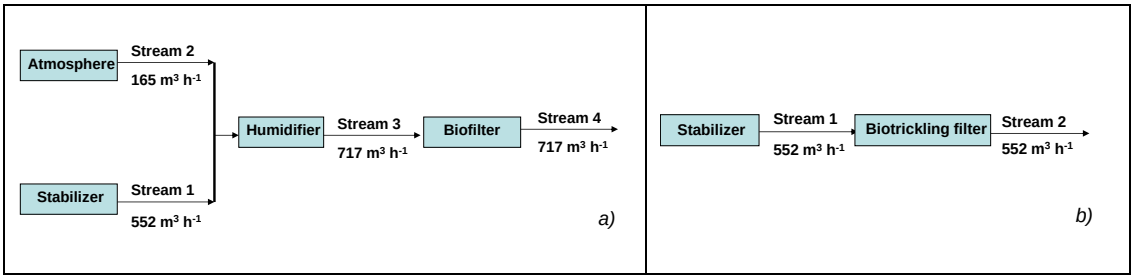

Figure 1: Municipal solid waste treatment plant layout. a) Layout before the modification; b) Layout after the substitution of the biofilter with a biotrickling filter.

3. Plant Upgrade Procedure: Materials and Methods

In order to make acceptable VOCs and odorous emissions, the following upgrade procedure of the existing plant has been proposed.

1-Analyzing the composition of the process streams in order to identify which types of contaminants are present and in which amount. Such analysis has been carried out in accordance with NIOSH standards procedures (O'Connor, 1994). Samples were collected onto suitable sorbent tubes using SKC sample pumps. Following desorption, the samples were analyzed by Gas Chromatography Mass Spectrometry (GC-MS). This system consists of a Varian Gas Chromatograph equipped with a high resolution capillary column interfaced with a Finnigan Matt ITS40 Mass.
2-Evaluating, theoretically, odor and VOCs concentrations at a distance in correspondence of which either first habitations arise or nearby factories workers usually operate. Such an evaluation has been performed using Equation 1 (which has been derived hypothesizing a Gaussian dispersion model for continuous emissions, punctual emission source, 3 m discharge elevation, perfect contaminants reflection from the ground and no rising limit into the atmosphere) in correspondence of a receptor position equal to (x,0,0) and implementing the most conservative aerial dispersion conditions: that is, Pasquill stability class F (Perry and Green, 1999) with wind velocity at the emission source, u_H, equal to 2 m s^{-1}.

$$C_i(x,y,z) = \frac{\dot{m}_i}{2\pi \cdot u_H \cdot \sigma_y \cdot \sigma_z} \cdot \exp\left(-\frac{y^2}{2\sigma_y{}^2}\right) \cdot \left[\exp\left(-\frac{(z-H)^2}{2\sigma_z{}^2}\right) + \exp\left(-\frac{(z+H)^2}{2\sigma_z{}^2}\right)\right] \quad (1)$$

where $C_i(x,y,z)$ is the i-th contaminant concentration in correspondence of position (*x*,*y*,*z*), [mg m^{-3}]; $\dot{m}_i$ is the i-th contaminant mass flow rate, [mg s^{-1}], $\sigma_y = 0.067 \cdot x^{0.90}$ is the standard deviation on *y* axes, [m] and $\sigma_z = 0.057 \cdot x^{0.80}$ is the standard deviation on *z* axes, [m]. These evaluations have been utilized because no contaminants sampler could be located nearby the desired positions.
For each substance, concentration value in (x, 0, 0) has to be compared with the corresponding 50 % odor recognition concentration (ORC) (Nagata and Takeuchi, 1990) to check if odor nuisances can arise.
3-Conveying the streams with the most elevated contaminants load into a suitable biotrickling filtration system. In this case, a single biotrickling filter has been proposed in substitution of the existing bio one (see Figure 1b).
4-Identifying the most critical contaminants to select the most suitable organic carrier and one or more specific bacterial strains to be inoculated into the biotrickling filters. In this case, bioactive shells of blue mussel (*Mytilus Edulis*) have been selected as carrying media. *Mytilus Edulis* shells, being rich of calcium carbonate ($CaCO_3$), exhibit a high buffering power and a strong affinity for slightly acidic compounds that, even if not present into the off air to be treated, are unavoidably produced during microbial metabolic activities. Moreover a selected consortium of microbial population was decided to be further inoculated in order to lower start-up times and, meanwhile, improve biotrickling filters efficiency in terms of total abatable mass.
5-Searching for biotrickling filters optimum process parameters: that is, bioactive media volume capable of treating the required off-gas flow rate and minimum trickling water to ensure good moisture, pH and nutrients for the active microbial population.
6-Checking on the effectiveness of the proposed plant upgrading in terms of odor and VOCs removal efficiency.

4. Results

1- Process streams compositions analysis

Referring to Figure 1a, compositions of stream 1 (i.e., off air from the stabilizer), stream 2 (i.e., external clean air) and stream 4 (i.e., treated air exiting from the biofilter), using an average off air flow rate of 552 m^3 h^{-1}, are reported in Table 1.
As it can be observed, among all odorous compounds, limonene and α-pinene are those ones present in the highest concentration.

Table 1: Contaminants concentrations in stream 1, 2 and 4 before the plant layout modification (Figure 1a). Removal efficiency, estimated odorous compounds concentrations and check on odor detection capacity (reference flow rate, 552 $m^3 h^{-1}$).

Compound	Stream 1 [mg m^{-3}]	Stream 2 [mg m^{-3}]	Stream 4 [mg m^{-3}]	Removal Efficiency [%]	Concentration (75 m, 0, 0) [mg m^{-3}]	Check on odor
Ethanol	3.51	ND	$8.90e^{-1}$	67.04	$9.60e^{-3}$	No Odor
Acetone	8.05	ND	2.90	53.17	$3.13e^{-2}$	No Odor
MEK	4.89	ND	1.60	57.46	$1.73e^{-2}$	No Odor
Toluene	1.56	ND	$8.90e^{-1}$	25.83	$9.58e^{-3}$	No Odor
Ethyl Benzene	$8.90e^{-1}$	ND	$4.70e^{-1}$	31.35	$5.07e^{-3}$	No Odor
m-Xylene	$9.50e^{-1}$	ND	$2.90e^{-1}$	60.32	$3.13e^{-3}$	No Odor
Limonene	$4.31e^{+1}$	ND	$2.46e^{+1}$	26.89	$3.18e^{-1}$	Odor (ORC=$2.12e^{-1}$)
Naphthalene	3.16	ND	1.56	35.82	$1.68e^{-2}$	No Odor
Benzene	$1.49e^{+1}$	ND	6.47	43.78	$6.98e^{-2}$	No Odor
α-pinene	$3.48e^{+1}$	ND	$2.05e^{+1}$	23.37	$2.21e^{-1}$	Odor (ORC=$1.00e^{-1}$)

2- Theoretical evaluation of contaminants concentration at (x, 0, 0)

Looking at the concentration values of all compounds 75 m away from the plant atmosphere discharge, it is possible to see that limonene and α-pinene represented effectively a serious problem for the nearby facility workers because they exceeded their 50 % odor recognition concentration (see Table 1 – ORC values indicated between brackets) even after the biofiltration treatment. Of course, the simplicity of the model employed to evaluate target contaminants concentration does not allow to take into account potential synergic effects (either positive or negative), terrain morphology, obstacles presence and so on. Therefore, it does not assure the complete reliability of concentration data even if more complex dispersion models (Wang et al., 2006) have provided analogous results. Nevertheless, reiterated complains from nearby facility workers have confirmed these theoretical evaluations. Anyway, observing concentration data for such compounds, it is possible to notice that the exceeding of 50 % ORC is moderate (for Limonene $3.18e^{-1}$ [mg m^{-3}] vs. $2.12e^{-1}$ [mg m^{-3}]; for α-pinene $2.21e^{-1}$ [mg m^{-3}] vs. $1.00e^{-1}$ [mg m^{-3}]).

3- Choice of the suitable biotreatment system

Starting from this scenario, it has been decided to replace the biofilter with a more efficient biotrickling filter to diminish odorous concentrations under their detection threshold values (see Figure 1b).

4- Choice of the microbial consortium

Since the most critical compounds to be abated were limonene and α-pinene, it has been decided to use bioactive shells of *Mytilus Edulis* further inoculated with a selected consortium of *Aspergillus Niger MTCC 961* and *Pseudomonas Putida MTCC 1072* (provided by the Laboratorio di Biologia – Università degli Studi dell'Insubria, Italy) that, according to literature (Prema and Bhattacharyya, 1962, Chatterjee and Bhattacharyya, 2001), shows a good terpenes removal efficiency. It must be underlined that some authors (Lopez et al., 2010) have suggested to employ *Ophiostoma Stenoceras* fungus (optimum operating pH range: 4 - 6) to increase monoterpenes (e.g. α-pinene) elimination capacity.

5- Search for biotrickling filter optimum operating parameters

For what concern biotreatment unit design and determination of process parameters (that is, minimum active media volume, empty bed residence time and trickling water flow rate), it has been decided to re-equip the stainless steel vessel previously used as biofilter (dimensions: 1.25 m x 2.0 m x 1.0 m). Such a unit has been filled with 2.2 m^3 of active media (the maximum loadable amount) supported by a polyvinylchloride net. Consequently, the empty bed residence time (EBRT) has been set equal to about 16 s: the minimum value compatible with the widest range of volumetric specific loads that will be treated by the plant (from 50 to 350 [m^3 h^{-1} m^{-3}]). Then, the irrigation system, previously used to periodically spray fresh water onto the previous active media, has been readapted to supply trickling water continuously under a flow rate controller. Moreover, a purge/re-integration water system has

been realized to maintain optimum water conditions for microbial metabolism and growth. Optimum trickling water flow rate has been set up at 3.36 m^3 h^{-1} (with a purge/re-integration water flow rate of about 0.03 m^3 h^{-1}).

6- Check on the effectiveness of plant upgrade

The revised unit has been then started up (after an inoculation period of 48 h, in order to provide sufficient acclimation time) and tested employing four different off air flow rates: 150, 325, 552 and 700 m^3 h^{-1} (corresponding to a specific volumetric load of about 68, 148, 251 and 318 m^3 h^{-1} m^{-3}, respectively). Such a flow rates choice has been a consequence of the broad range of wastes flow rates that might be supplied over the time. Operating conditions and results about odor concentrations (both experimental – using a field olfactometer - and theoretical ones) are reported in Table 2.

Table 2: Operating conditions and results about odor concentrations for the different off air flow rates which have been tested in this work.

Operating parameters	***Case 1***	***Case 2***	***Case 3***	***Case 4***
Average Off-Gas Flow Rate, [m^3 h^{-1}]	150	325	552	700
Off-Gas Inlet Temperature, [°C]	45.0	48.5	48.0	50.5
Off-Gas Outlet Temperature, [°C]	43.2	45.5	45.2	46.5
Average Pressure Drop, [mmH_2O]	25.0	27.4	26.3	83.0
Trickling Water Flow Rate, [m^3 h^{-1}]	3.36	3.36	3.36	3.36
Water Inlet Temperature, [°C]	42.0	42.0	42.1	42.3
Water Outlet Temperature, [°C]	44.2	44.6	44.3	45.0
Odor Concentrations				
Experimental Odor Concentration – Inlet, [ou m^{-3}]	478.5	761.1	963.2	1140
Experimental Odor Concentration – Outlet, [ou m^{-3}]	19.03	31.09	37.36	49.35
Theoretical Odor Concentration – (75,0,0), [ou m^{-3}]	0.1578	0.2579	0.3099	0.4093
Average Removal Efficiency, [%]	96.0	95.9	96.1	95.7

As it can be noticed, theoretical odor concentrations 75 m away from the plant atmosphere discharge (always evaluated using equation (1)) are minor of the threshold value 3.0 ou/m^3 proposed by Capelli et al. (2011) for the detection of odor nuisance. As an example, Table 3 reports a detailed analysis concerning contaminants concentrations with an off air flow rate of 552 m^3 h^{-1} (case 3).

Table 3: Contaminants concentrations in stream 1 and 2 after the plant layout modification (Figure 1b). Removal efficiency, estimated odorous compounds concentrations and check on odor detection capacity (reference flow rate, 552 m^3 h^{-1}).

Compound	Stream 1 [mg m^{-3}]	Stream 2 [mg m^{-3}]	Removal Efficiency [%]	Concentration (75 m, 0, 0)	Check on odor
Ethanol	3.51	$1.20e^{-1}$	96.58	$9.95e^{-4}$	No Odor
Acetone	8.05	2.50	68.94	$2.07e^{-2}$	No Odor
MEK	4.89	1.05	78.53	$8.71e^{-3}$	No Odor
Toluene	1.56	$4.30e^{-1}$	72.43	$3.57e^{-3}$	No Odor
Ethyl Benzene	$8.90e^{-1}$	$1.70e^{-1}$	80.90	$1.41e^{-3}$	No Odor
m-Xylene	$9.50e^{-1}$	$1.50e^{-1}$	84.21	$1.24e^{-3}$	No Odor
Limonene	$4.31e^{+1}$	6.47	85.20	$5.37e^{-2}$	No Odor
Naphthalene	3.16	$5.20e^{-2}$	98.35	$4.31e^{-4}$	No Odor
Benzene	$1.49e^{+1}$	$8.00e^{-1}$	94.65	$6.63e^{-3}$	No Odor
α-pinene	$3.48e^{+1}$	6.31	81.86	$5.23e^{-2}$	No Odor

As it can be observed, in all cases, threshold limits are not overcome guaranteeing a sufficient abatement efficiency for odorous compounds. Moreover, such results have been obtained without diluting the off air coming from the stabilizer. The renounce to operate the dilution causes the gaseous stream temperature entering the biotrickling filter to be comprised in the 45 - 50 °C range. Such temperatures are quite high for mesophilic microorganisms (as those ones employed in the biotrickling

filter). But, taking into account that trickling water diminishes the off air temperature under 44 °C by simple heat transfer, acceptable global thermo-hygrometric conditions and pH control are reached. This plant has been monitored for over 90 days, continuously registering off-gas flow rates, pressure drops and biotrickling filter inlet and outlet temperatures (see averaged values in Table 2). The results confirmed the attainment of quasi-pseudo-stationary operating conditions during all the monitoring period.

Finally, in order to characterize the biotrickling system from the microbiological point of view, a sample of filtrating material has been analyzed with its trickling water. Bacterial count has evidenced a microbial concentration of about $4 \cdot 10^8$ cfu g^{-1} (wet weight). Moreover, a quite high microbial activity is confirmed by the relevant weight increase of the active media due to adherent biomass (weight has approximately doubled, increasing from 370 g L^{-1} to 665 g L^{-1}). With regard to trickling water analysis, carried out by GC-MS, the following results have been collected: methanol: 0.5 mg L^{-1}, acetonitrile: 0.51 mg L^{-1}, ethanol: 5.0 mg L^{-1}, acetone: 0.8 mg L^{-1}, isopropanol: 0.45 mg L^{-1}, styrene: 31.2 µg L^{-1}, methyl benzene: 70.8 µg L^{-1}. These concentration levels confirm the significant microbial population metabolic activity. Despite the presence of slightly acidic compound in minor concentrations, a neutral pH has been maintained even after 90 days of experimentations therefore providing a good growth environment for bacteria responsible for the desired degradation.

5. Conclusions

In this work it has been shown that biotrickling filters can represent a sound solution to upgrade treatment systems dedicated to VOCs and odorous compounds removal from off gases coming from organic solid wastes treatment plants. Particularly, it has been observed that biotrickling filters are able to guaranteeing both a better temperature control and removal efficiency than biofilters and, consequently, have to be preferred with respect to these last ones whenever high temperature and medium contaminants load off air streams must be treated.

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