AIR POLLUTION ABATEMENT: JOINT COMBINATION OF PACKING MATERIALS IN BIOFILTERS

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Closed areas with heavy traffic such as road tunnels and underground parking lots usually present high levels of atmospheric pollution. Thus, the poor-quality air in those enclosures constitutes a health threat for commuters, regular users and workers.

The presence of a fluctuating number of volatile organic and inorganic compounds in those confined spaces encourages the utilization of biological reactors (such as biofilters) for their removal. Biofilters are considered a cost-effective, reliable and safe alternative for treating moderately high loading rates of biodegradable pollutants present in air. As far as the system operation is concerned, the contaminated air stream passes through a bed containing a packing material where the biomass is attached, and thus, the contaminants are degraded by the action of the microorganisms. The nature of the packing material is a key factor for the successful application of any biofilter. Thus, this work focuses the influence of using a joint combination of different packing materials on the performance of volatile organic compounds (VOC) abatement. The contaminated inlet gas flow was generated in the laboratory by carefully mixing biodegradable volatile compounds usually found in motor vehicle exhaust gases.

Keywords: *Biofiltration; Packing material; Air pollution*

REDUCCIÓN DE LA CONTAMINACIÓN ATMOSFÉRICA: COMBINACIÓN DE MATERIALES DE SOPORTE EN BIOFILTROS

El aire ambiente en lugares cerrados con elevado tráfico como túneles y garajes subterráneos presenta en muchas ocasiones una elevada contaminación. Por lo tanto, es necesario tratar dichas atmósferas para proteger la salud de los usuarios y empleados de dichos recintos.

La aplicación de tratamientos biológicos (concretamente por medio de biofiltros) para la depuración de los contaminantes orgánicos e inorgánicos habituales en estos recintos es una alternativa prometedora debido a su bajo coste de operación y su reducido impacto ambiental. El funcionamiento de estos biofiltros consiste en hacer pasar una corriente gaseosa contaminada a través de un lecho fijo constituido por un material de empaquetamiento donde está retenida la biomasa responsable de la degradación final de los contaminantes. La adecuada selección del material de empaquetamiento es necesaria para asegurar la eficacia de degradación en los biofiltros. Por lo tanto, este trabajo se centra en estudiar las posibles combinaciones de varios materiales de empaquetamiento en la eliminación de compuestos orgánicos volátiles (COV) mediante biofiltración. El gas contaminado que se alimenta al reactor se ha generado en el laboratorio mediante la mezcla de los compuestos volátiles usualmente encontrados en las emisiones de los vehículos motorizados.

Palabras clave: *Biofiltración; Material de soporte; Contaminación atmosférica*

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1. Introduction

Air pollution is one of the main environmental problems in many countries. Based on a dataset of 48 countries spanning the period 1990 to 2006, it was concluded that individuals do not grow accustomed to air pollution and that air pollution significantly reduces current life satisfaction (Menz, 2011).

Among the most frequently present categories of air pollutants, volatile organic compounds (VOCs) have been recognized as one of the most important groups of air toxins that play key roles in atmospheric chemistry.

There is no clear or unanimous definition of a VOC: The US EPA defines VOCs as substances with vapor pressure greater than 0.1 mm Hg; the Australian National Pollutant Inventory affirms it is any chemical based on carbon chains or rings with a vapor pressure greater than 2 mm Hg at 25 °C, and the EU considers it to be any chemical with a vapor pressure greater than 0.074 mm Hg at 20 °C. Chemicals such as CO, CO_2, CH_4, and sometimes aldehydes, are often excluded.

VOC compounds are commonly found in the atmosphere at terrestrial level in different urban and industrial atmospheres. The hundreds of existing atmospheric VOCs are produced both by anthropogenic activities (such as motor vehicle exhaust, motor vehicle fuel evaporative losses, different industrial processes, petroleum storage and distribution and refineries, surface coating and solvent use, domestic wood heaters, biomass burning, environmental tobacco smoke, use of solvent, glues and cleaners in arts and crafts, landfills and agricultural activities) and by natural biogenic processes (such as emissions from trees and vegetation, forest fires produced by natural causes, or anaerobic marshy bog processes, among others).

VOCs pose a potential threat to human health. This health hazard is a consequence of the great mobility and capacity of VOCs to be inhaled by people working or living in places with high concentrations. In the literature, several VOC exposure-monitoring studies have reported that indoor VOC concentrations are generally higher than outdoor ones (Wang et al., 2009).

This finding is crucial since many people in developed societies spend most of their time indoors. For instance, Jones (1999) stated that US residents on average spent 88 % of their day inside buildings, and 7 % in a vehicle.

Although short-term exposure to particular concentrations of some VOCs present in the air is not considered acutely harmful to human health, long-term exposure may result in mutagenic and carcinogenic effects. Median personal exposures to several VOCs have been associated with excess lifetime cancer risk in the 10^{-4}–10^{-5} range, considerably exceeding the US guideline (D'Souza et al., 2009).

Exposure to VOCs can cause such acute and chronic effects as respiratory damage, and can therefore increase, for example, the risk of asthma. The systemic toxic effects of VOCs are also significant. Among these, renal, hematological, neurobiological and hepatic disorders, as well as mucosal irritations, are the most common. Experience of eye, nose or mouth irritation has been reported at 5000–25000 µg VOC m^{-3} (Guieysse et al., 2008). They can also affect the nervous, immune and reproductive systems. Classic neurological symptoms associated with VOCs are feelings of fatigue, headaches, dizziness, lethargy and depression (Ras et al., 2009).

Finally, bad odors should necessarily be included among the adverse effects of VOCs, as they can cause diverse indirect health effects such as bad mood, nausea and vomiting, hypersensitive reactions, loss of appetite and even alterations in the respiratory model. In fact, odor nuisance is so annoying that some authors have estimated reductions in house prices of up to 30 % in properties situated one mile away from the odor source (Estrada et al., 2011).

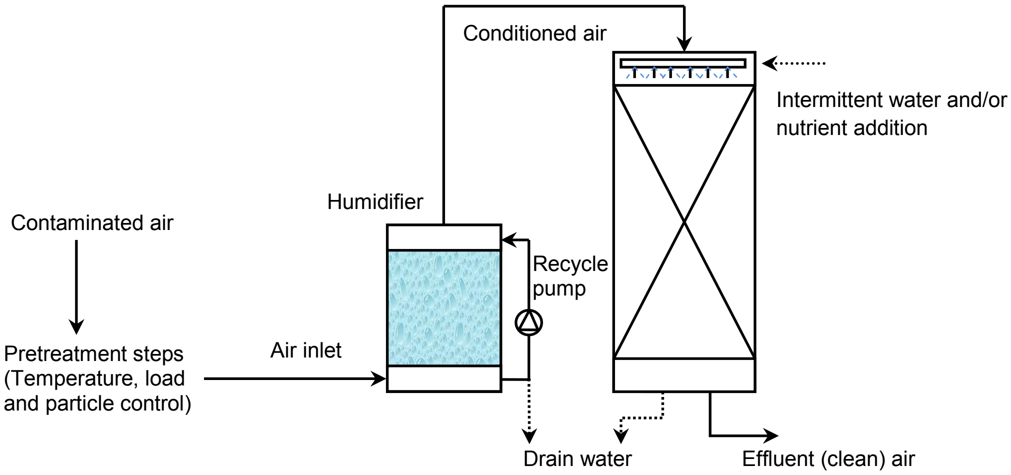
2. Biological systems for waste gas treatment

The growing emphasis on sustainability over the past twenty years has boosted the application of biological processes for waste gas treatment and VOC removal, turning them into a robust and feasible alternative for the removal of air emission streams with relatively low pollutant concentrations at high flow rates.

Among the different biotechnologies, biofiltration can be defined as a maturing technology, taking into account that the number of full-scale biofilter plants has rapidly increased over the past 20 years.

A schematic of a biofilter is presented in Figure 1. The contaminated inlet gas is conditioned (i.e., humidified and heated/cooled) before it is uniformly forced to pass through the biofilter bed. The bacteria are attached to the porous packed biofilter bed.

Figure 1. Scheme of a conventional closed biofilter



The effectiveness of biofiltration lies on the ability of these microorganisms to degrade contaminants. Within the biofilter, biodegradable volatile compounds are adsorbed/absorbed by the bed material and the biologically active biofilm that grows on the porous packed bed particles. Subsequently, the bacteria biologically oxidize contaminants into new biomass and innocuous substances like CO_2, H_2O, NO_3^- and SO_4^{2-}.

Biofilters usually have a height of 1 or 2 m to prevent excessive air velocities through the media, which easily results in high-pressure drop or airflow preferences. They are mainly recommended for the treatment of waste gases with concentrations below 5 g m^{-3}. They are also more suitable for the treatment of relatively low or moderate flow rate: loads do not normally exceed 500 $m^3\ m^{-2}\ h^{-1}$ (Kennes et al., 2009).

In contrast with other media-based biotechniques (such as biotrickling filters or bioscrubbers), in conventional biofilters there is no continuous feed of a liquid phase. Therefore, this bioreactor is especially suitable for the treatment of hydrophobic and poorly water-soluble compounds with a Henry's constant of up to about 1, and it provides the highest removal efficiencies for moderately hydrophobic pollutants (Iranpour et al., 2005).

However, its effectiveness is limited by operating parameters such as moisture content, the structural weakness of most conventional organic packing materials, medium acidification or the accumulation of toxic metabolites due to the biodegradation of chlorinated or sulfured organic compounds. These factors are difficult to control and require the frequent replacement of the packing material.

3. Packing material’s challenge

The proper selection of the packing material used in a biofilter is an important decision for achieving high removal efficiencies and maintaining an optimal performance in long-term operation. The support media’s main function is to provide contact between the gas-phase contaminants and active microbial cultures attached to the material’s surface as a biofilm.

Other functions of the media are to distribute the gas flow evenly within the bed’s cross-sectional area with minimal gas-phase pressure drop, distribute any liquid nutrients sprayed onto the bed surface, and prevent biomass accumulation, which would eventually lead to undesired channeling of the gas and liquid (Dumont et al., 2008).

Packing materials that can be used during VOC biofiltration are grouped into two main categories: organic and inorganic materials. The latter can be divided into natural inorganic materials or entirely synthetic materials.

Organic materials include peat, soil and compost, although wood bark, sugarcane bagasse and nuts shells are also used. Organic materials are generally considered by several authors as the preferred materials; in fact, a recent compilation of data from full-scale installations revealed that approximately 87 % of the biofilters operated with organic packing materials, wood chips and compost being the most popular supports.

The main advantages of these materials are that they are readily available and naturally contain contaminant-degrading microorganisms. Another advantage is that they provide nutrients, such as nitrogen and phosphorous, which are necessary for microorganism growth.

However, these materials undergo structural damage, bed compression, increasing pressure drop and thus decreasing biofilter efficiency. Several authors have recorded bed height losses in the range of 4–14 % when organic (maize stubble) or organic-inorganic mixtures (polyurethane foam–poplar wood chips or compost–perlite) were used (Hernández et al., 2013; Lebrero et al. 2011; Singh et al. 2006). Lebrero et al. (2014) observed acceleration in traditional organic packing materials (compost, wood bark and Macadamia nutshells) biodegradability by 17–26 % in 21 days due to the exposure to VOCs. Therefore, organic packing materials need to be replaced after 2–5 years.

Figure 2 shows the organic filter bed material agglomeration. This may cause local anoxic or anaerobic zones within the biofilter, reduction of active points for microbial attachment and a final worsening in the bioreactor performance.

Figure 3 shows bed compression in three biofilters packed with pelletized agro-waste, amended soil and a mixture of both over time.

Inorganic materials were first used as additives to improve the mechanical properties of organic material based biofilters. This group includes natural and manufactured materials such as lava rocks, ceramic rings, glass beads, polyurethane foam, activated carbon, perlite and vermiculite, among others.

Figure 2. Evolution of organic packing material particles (mixture of pelletized agro-waste and amended soil) over time: original packing material before use (left) and particle conglomeration within the biofilter after 130 days of operation (right)

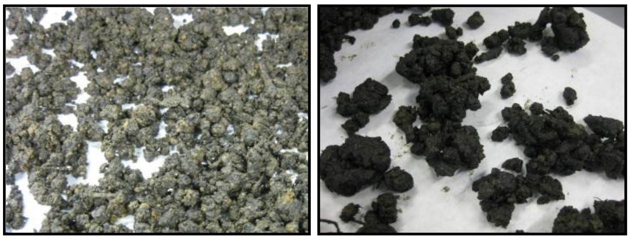
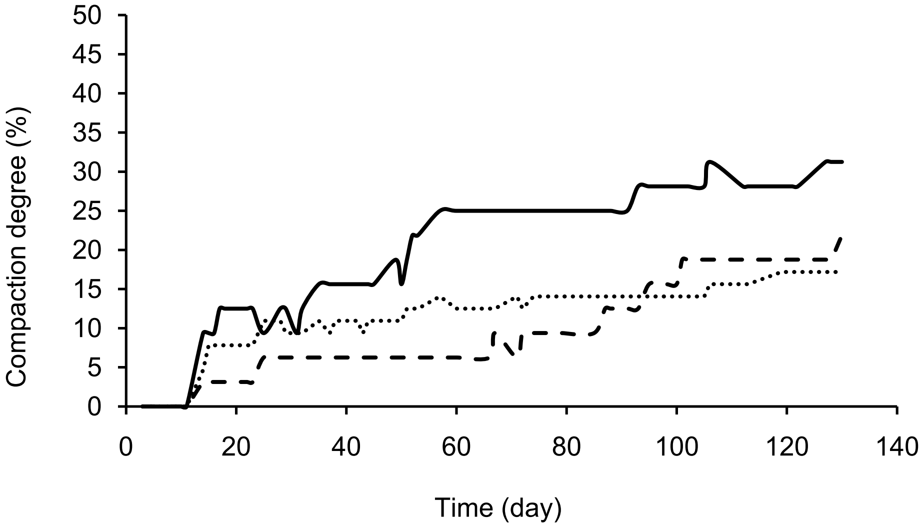


Figure 3. Compaction degree over time within a biofilter packed with amended soil (continuous line), pelletized agro-waste (broken line) and mixture of both (dotted line)



When used during biological processes, they offer several advantages, such as good mechanical resistance in comparison to organic materials. Their physical properties (e.g. porosity and specific surface area) can be more easily adjusted according to the requirements of the bioprocess. Wolstenholme et al. (2013) defined the use of inorganic media as a "greener" solution as it solved problems related to cost minimization, footprint requirement or depleted material management. Several inert packing materials can be either regenerated or reused before requiring replacement. However, plastic and polymeric materials may pose a more serious problem of solid-waste generation than packings such as perlite, ceramic beads or lava rock when the filter bed gets exhausted (Kennes et al., 2009). However, their main disadvantage is that they do not provide any nutrients for the biomass and, in some cases, they are unaffordable.

The importance of the packing material properties depends heavily on the characteristics of the biofiltration system and its operation, which implies that a material may be suitable in certain conditions but inappropriate in others. In general terms, the packing material must provide a favorable environment as far as pH, moisture, temperature, nutrients and oxygen supply are concerned. A perfect material should have the following characteristics:

- A suitable particle size, void fraction (0.5–0.9) and specific surface area (≥ 300 m2 m–3), enabling the filter bed to benefit from a high biofilm surface and facilitating the transfer of the VOCs contained in the air. If the size of the particle is too small, a large specific surface area is available, although resistance to the gas flow is increased; by contrast, if it is too large, it favors gaseous flows, but the number of potential sites for the microbial activity is lower. A threshold diameter-value higher than 4 mm (minimal pellet size) has been suggested by Leson and Winer (1991). Delhoménie et al. (2002) concluded that pellet size and specific surface area were the major limiting factors for the biodegradation process.

- A high water-holding capacity is desirable to maintain the optimal activity of the immobilized microorganisms. Gutiérrez-Acosta et al. (2010) were able to modify the hydrophobic character of polyurethane foam using a tertiary amine as an additive. This organic compound, containing hydroxyl groups, increased the water retention capacity of the synthetic material without the total loss of its hydrophobicity. The water retention capacity of the composite was 34 % w/w, while for the non-modified polyurethane it was 12.5 % w/w. Such a modification contributed to significant biomass growth and attachment and to the establishment of better interactions with pollutant compounds. The time required for the complete biodegradation of hydrocarbons with the modified polymer was at least 30 % shorter than that obtained with the not amended polymer. Table 1 shows the water-holding capacities of different packing materials used in the literature.

Table 1. Water-holding capacities of various packing materials

Material	Water-holding cap. (%)	Material	Water-holding cap. (%)
Towel scrap[a]	301.2	Sawdust[a]	245.4
Tobermolite[e]	109.5	Perlite[b]	42.8
Polyurethane + Tertiary amine[c]	34.0	Polyurethane[b]	33.3
Activated carbon powder[a, g]	31.7	Earthworm casting[a]	31.7
Activated carbon granule[a, h]	31.4	Activated carbon powder[a, i]	30.5
Polypropylene[b]	21.7	Crab shell[a]	18.1
Coconut fibre[f]	17.5	Fibrous peat[f]	12.7
Zeolite[a]	5.4	Compost[d]	1.95
Waste tyre scrap[a]	1.3	Soil[a]	0.9
Wood bark[d]	0.42	Scallop shell[a]	0.4
Pouzzolane[f]	0.3	Macadamia nutshells[d]	0.11

Notes: (a) Kwon et al. (2009); (b) Gutiérrez-Acosta et al. (2012); (c) Gutiérrez-Acosta et al. (2010). This material was made of a modified polymeric foam based on polyurethane with an organic compound (tertiary amine) containing hydroxyl groups; (d) Lebrero et al. (2014); (e) Kim et al. (2014); (f) Anet et al. (2013) (g) from coconut; (h) from coconut; (i) from sawdust.

- The bacteriological characteristics favorable to bacterial development are high inorganic and organic nutrient content, high buffering capacity avoiding large pH fluctuations and the absence of inhibiting compounds.

- A high adsorption capacity in order to buffer intermittent loads. Inlet concentrations of contaminant fluctuate throughout the daytime, the operating schedule or even when an eventual emergency situation occurs. The packing material should act as a buffer to adsorb and soften the pollutant load on the biofilter. Table 2 shows the toluene adsorption capacity of different packing materials. Bioreactors have recorded better yields under stationary conditions.

However, industrial air emissions usually present variable and discontinuous concentrations that could hinder the performance of full-scale biofilters. Operating problems have been reported when the adsorption capacity of packing material is not high enough to achieve effective pollutant load equalization under cyclic or discontinuous operation (Sempere et al., 2010).

- Mechanically resistant, chemically inert and stable. A relatively constant volume of pores is advisable to avoid bed compaction and ensure a uniform airflow through the filter bed.
- Low bulk density in order to ensure better hydrodynamic properties and avoid bed compaction. Table 3 shows the bulk density of different packing materials.
- The presence of diverse indigenous microorganisms (including bacteria, actinomycetes, fungi, yeasts, algae and protozoa) excludes the need for inoculation and ensures shorter start-up periods. Nevertheless, this microflora should be carefully monitored, as pathogenic species can also be found (Borin et al., 2006).
- Low cost: the price of the packing material has a significant impact on overall costs, not only because of the high volumes usually required for biofilter construction, but also because packing material replacement is mandatory due to its limited lifespan. Commercial biofilter packing materials typically last from 1 to 15 years, with the longest lifespans found for nutrient-enriched inert materials, and from 1 to 3 years for conventional organic materials (Estrada et al., 2011). Table 4 shows the price of several packing materials.

Table 2. Toluene adsorption capacities of various packing materials (Kwon et al.,2009)

Material	Adsorption cap. (mg toluene g–1 material)	Material	Adsorption cap. (mg toluene g–1 material)
Activated carbon powdera	56.9	Activated carbon powderb	56.6
Activated carbon granulec	56.3	Crab shell	15.7
Waste tyre scrap	11.5	Zeolite	8.2
Earthworm casting	6.6	Soil	5.7
Used briquette	4.3	Sawdust	4.2
Chalk	3.5	Styrofoam	3.2
Sand	2.7	Towel scrap	2.2
Pine needle	1.6	Scallop shell	1.1
Rice straw	0.2	Leaf mould	0.1
Wood	0.0	Pine cone	0.0

[a] From coconut; [b] From sawdust; [c] From coconut

Table 3. Bulk density of various packing materials

Material	Bed bulk density ($g\ ml^{-1}$)	Material	Bed bulk density ($g\ ml^{-1}$)
Macadamia nutshells (wet)[e]	1.09	Compost (wet)[e]	0.93
UP–20[a]	0.92	Compost-Polyethylene[b]	0.825
Pouzzolane[g]	0.783	Expanded schist[g]	0.586
Wood bark[e]	0.35	Tobermolite[f]	0.276
Hydroballs[c, h]	0.27	Polypropylene[d]	0.262
Bark chips[c]	0.24	Peat[c]	0.21
Vermiculite[c]	0.15	Perlite[d]	0.115
Polyurethane[d]	0.073	Coconut fibre[g]	0.041
Heather[g]	0.016		

Notes: (a) Gaudin et al. (2008). UP–20 was a packing material based on urea phosphate; (b) Wu et al. (2009). It was mixture of mature pig compost and a new packing material made of polyethylene; (c) Oh and Choi (2000); (d) Gutiérrez-Acosta et al. (2012); (e) Lebrero et al. (2014); (f) Kim et al. (2014); (g) Anet et al. (2013); (h) Hydroball was a horticultural product manufactured by Haeran Co. (Korea).

4. Packing material airborne emissions: potential biohazard?

Germ and spore emission problem from packing material has hardly been considered in the debates over the benefits and risks of implementation of biological waste air cleaning techniques into industrial use.

Bearing in mind the “physical” function of a biofilter, it should “remove” or “hold” particulate matter, including airborne microorganisms and dust from the waste gas stream. Nevertheless, regarding the biological activity into the bed, the bioreactor could also be a microorganism emission source. Scarce studies have been carried out to clear up this issue.

Becker and Rabe (1997) observed that a compost biofilter reduced the levels of *Aspergillus fumigatus*, but other fungi spores inhabiting the biofilter were released into the emitted air (*Paecilomyces variotii* and *Aureobasidium pullulans*).

Schlegelmilch et al. (2005) concluded that biofilters commonly used at composting facilities were effective to reduce airborne emissions. Thus, the biological waste gas systems were able to retain potentially pathogenic microorganisms which were fed into the bioreactor together with the waste inlet gas. On the contrary, non-pathogenic secondary emissions were released after treatment.

Tymczyna et al. (2011) also demonstrated the validity of a biofilter fitted to the outlet of the ventilation system of a litter-bed pig house to remove air microbial contaminants. Thus, mean bacterial reduction of 77 % and fungal reduction of 69 % were obtained for inlet concentrations of $8.3{*}10^6$ CFU m^{-3} and $1.9{*}10^5$ CFU m^{-3}, respectively. After the biotreatment process, complete absorption/reduction of *Rhodococcus*, *Brevibacterium*, *Neisseria*, *Pantoea*, *Pseudomonas* bacteria and *Scopularopsis*, *Mucor* and *Paecilomyces* fungi was achieved.

Zilli et al. (2005) obtained emission values in the range of $1.5–4{*}10^3$ CFU m^{-3} for three lab-scale biofilters when comparing the performance of an indigenous bacterial consortium and an inoculum of a cell suspension of pure benzene-degrading strain of Pseudomonas. This range is lower than the occupational threshold limit value proposed for bacterial emissions (10^3 to 104 CFU m^{-3}) by Health and Safety Executive (2003).

Table 4. Price of common packing materials used in biofiltration

Packing material	Cost (€ m^{-3})	Lifespan (years)	Annual cost (€ m^{-3} $year^{-1}$)
Activated carbon[c]	1500	10	150
Coconut fibre[a]	200 – 240	2	100 – 120
CAC[a, f]	450 – 500	10	45 – 50
Lava rock[b]	215 – 250	10	45 – 50
Granulated peat[c]	170	4	42.5
Lava rock[a]	40 – 50	15	3 – 4
SMP[b, d]	125	5	25
Peat with heather[a]	40 – 50	2	20 – 25
Advanced material[a, e]	300 – 360	15	20 – 24
Pine leaves[a]	25 – 35	2	12 – 17
Expanded chist[c]	110	10	11
SBC[a, g]	70 – 90	10	7 – 9
Pouzzolane[c]	50	10	5
Lignite[a]	40 – 50	10	4 – 5
Ceramic Rashig ring[b]	350 – 375	10	3 – 4
Compost[a]	5 – 10	2	2 – 5
PUF[a]	25 – 35	15	2 – 3
Ceramic Pall ring[b]	250 – 315	10	2 – 3

Notes: (a) Price of packing materials according to 2009 prices on the Spanish market published by Dorado et al. (2010); (b) Price of packing materials according to the Chinese market published by Sun et al. (2011); Economical data presented herein have been converted to euro (€) currency, considering that 1 € = 0.125 ¥; (c) Anet et al. (2013). Material costs were obtained from biofilters suppliers for business proposal corresponding to 500 m^3 packing material. (d) SMP was comprised of coral rock, bark, ceramisite, charcoal, and compost (160:120:100:60:15, w:w).The SMP structure was reinforced using a urea-formaldehyde resin to enhance bed porosity and specific surface area; (e) The advanced material is based on a thin layer of compost over a clay pellet; (f) CAC is a commercial activated carbon; (g) SBC is based on a sludge-based carbon.

5. Conclusions

The packing material is the main factor influencing both reactor long-term operating stability and global costs. Its physicochemical properties, such as porosity, water retention capacity, density and pH, have a strong influence on the attachment of microorganisms and development of biofilms. A packing material should ideally be a long-lasting and cheap material to ensure a robust and economic performance and it should contain the nutrients required for microbial growth.

The utilization of packing materials comprising the advantages of both organic and inorganic materials might lead to an improvement in the performance of biofilters. The addition of inert materials together with classic organic ones might extend filter bed durability, increase removal efficiencies during the acclimation period of the microbial consortium and lead to more stable waste gas treatment operations since the buffering capacity of the resulting filter bed is increased.

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