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Performance of different biological waste air purification processes in treatment of a waste gas mix containing *tert*-butyl alcohol and acetone: A comparative study

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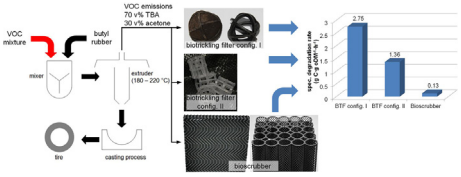
HIGHLIGHTS

- Biodegradability of a VOC mix of 70v %TBA and 30v% acetone in waste air was tested.
- Performance levels of two biotrickling filters and a bioscrubber were compared.
- The bioscrubber showed the highest total elimination capacity ($80.2\ g\ C\ m^{-3}\ h^{-1}$).
- BTF I showed the highest specific elimination capacity ($2.75\ g\ C\ g\ oDM^{-1}\ h^{-1}$).
- Two novel strains TBA100 and TBA200 able to degrade TBA were isolated.

GRAPHICAL ABSTRACT



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ABSTRACT

Two biotrickling filters and a bioscrubber system, equipped with different package materials and inoculums, were compared in regard of performance in treatment of a VOC mixture containing 70 v% of *tert*-butyl alcohol (TBA) and 30 v% of acetone. The average absolute elimination capacity at highest test loadings was $80.2\ g\ C\ m^{-3}\ h^{-1}$ for the bioscrubber (BIOdek® and BIO-NET® as package material; *Mycobacterium austroafricanum* TBA100 and *Aquincola tertiaricarbonis* TBA200 as inoculum) and 25.2–27.5 $g\ C\ m^{-3}\ h^{-1}$ for the biotrickling filters (configuration I, Bio-airSPHERES®, no further inoculation; configuration II, Hilfow™ rings, *Mycobacterium austroafricanum* TBA100 as inoculum). However, normalizing elimination capacities to either biomass or the cell count of TBA degraders the biotrickling filter of configuration I ranked first ($2.75\ g\ C\ g\ oDM^{-1}\ h^{-1}$ and $12.91\ g\ C\ 10^{12}\ cells^{-1}\ h^{-1}$) and the bioscrubber last, due to a largely sized regeneration unit ($0.13\ g\ C\ g\ oDM^{-1}\ h^{-1}$ and $0.40\ g\ C\ 10^{12}\ cells^{-1}\ h^{-1}$). Even though TBA is hardly biodegradable, its biodegradation could be done without expensive formation of inoculums using Bio-airSPHERES® as composite package material. Performance levels were enhanced and stabilized by repetitive dosage of fertilizer in all systems.

This is both the first report about biodegradation of a waste gas containing TBA and application of Bio-airSPHERES® for biodegradation of high VOC concentrations.

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1. Introduction

tert-Butyl alcohol (TBA) is a widely used high production volume chemical with a national production volume of nearly 1902 kilotons in 2012 in the United States [1]. It is applied as an organic solvent for industrial and pharmaceutical uses, and as such, it is added to lacquers, paint removers, industrial cleaners, nail enamels and polishes, wax pastes for leather conservation, leather finishes, marble, ceramic, linoleum, plastic and varnished wood floors, as dehydrating agent and denaturant for ethanol [2]. TBA is further used in coatings on metal and paperboard food containers and in the manufacturing of flotation and defoaming agents, flavors and fruit essences, artificial musk and perfumes [3]. It is also applied in the synthesis of methyl methacrylate as monomer for the production of miscellaneous plastic products, and as intermediate in the synthesis of oil soluble polyester resins and antioxidants, isobutylene, *tert*-butyl chloride, *tert*-butylphenol, methyl *tert*-butyl ether (MTBE), ethyl *tert*-butyl ether (ETBE), and *tert*-butyl hydroperoxide [4]. Prior to 2006, TBA was also used as octane booster and oxygenate in gasolines [3]. Due to its melting point of 25.7 °C, TBA is generally used in mixture with other solvents like methanol, ethanol, acetone, or other isomers of butanol.

Owing to its tremendous use in multiple applications, TBA may be directly released to the environment through various waste streams [2,3]. The Toxics Release Inventory Program National Analysis Report estimated a TBA release of more than 450 tons into the soil and of 837 tons into the air and water bodies in the United States in 2014 [5]. Beside these emissions, as a likely degradation product of MTBE, ETBE, *tert*-butyl hydroperoxide, and *tert*-butyl acetate, TBA is detectable in multiple legacy areas and aquifers in concentrations of $0.18–4.4\ 10^{6}\ \mu g\ L^{-1}$ [6–9].

TBA is considered as a poorly biodegradable compound both due to its physicochemical properties (low K_{OC} value of 37 and high water solubility) [10] and its molecular structure, which shows a high steric hindrance towards enzymatic degradation. In early reports only 1–7% of its theoretical BOD level, calculated on the base of the TBA level supplied, was achieved in time spans of 5–14 days [11,12]. In the last 25 years, a couple of microcosm studies of MTBE degradation successfully showed TBA degradation both under anaerobic and aerobic conditions as well as co-metabolic conditions, but mainly focused on the elimination of TBA as contaminant in different water bodies and soil. An overview of studies dealing with aerobic degradation either of MTBE or TBA as sole source of carbon is given in Table 1. A summary of studies examining degradation of MTBE or TBA by mixed cultures is presented in Table S1; co-metabolic degradation of TBA under aerobic conditions is presented in Table S2 of the Supplementary Materials. In contrast, only few studies dealt with the elimination of MTBE and TBA as an intermediate in waste air streams [13–18]. To our best knowledge, no study directly focused on the biodegradation of TBA as contaminant in waste air streams so far.

Composite package materials are of emerging interest due to their advantages towards conventional biotrickling filter and biofilter materials, like low pressure losses, no compaction tendency, high specific surface, as a source of nutrients and supplements, high water storage capacity, and the presence of an established microbial community enabling the start-up of a waste air treatment plant without further inoculation [19,20]. Beside applications in odor abatement, a single study described the use of Bio-airSPHERES® as composite material for removal of VOCs at concentration levels lower than 40 ppm [19].

The main objectives of this study were the investigation of TBA mineralization by two new isolates, able to use TBA as sole source of carbon and energy, and comparative characterization of performance levels in three bioreactors for waste gas treatment under different waste gas conditions. Two biotrickling filters with either Hiflow™ rings or Bio–airSPHERES® as package materials and a bioscrubber equipped with BIOdek® and BIO-NET® structures as packed structures for enhanced mass transfer were analyzed. All three systems were tested with a crude gas mixture containing 70% of TBA and 30% of acetone, a mixture commonly used in the formation and extrusion of butyl elastomers, which can be used as air barriers for tubeless automobile tires.

2. Material and methods

2.1. Isolation and cultivation conditions

Two strains able to use *tert*-butyl alcohol as sole source of carbon and energy were either isolated from the biotrickling filter equipped with Bio–airSPHERES®, containing a mix of clay and biologically highly active compost (*Mycobacterium austroafricanum* TBA100) or from sewage sludge of a waste water treatment plant in Basel, Switzerland (*Aquincola tertiaricarbonis* TBA200), and further cultivated. During isolation of TBA100, 100 g of wet organic filler of the Bio–airSPHERES® were separated from the plastic bodies and suspended in 1.9 L of 0.9% NaCl. 100 µL aliquots either of the suspension or the sewage sludge were plated on solid mineral medium supplied with 15 µL of TBA as sole source of carbon. After three days of cultivation at 30 °C, emerging colonies were repetitively picked and transferred to fresh media until pure strains were obtained.

For re-isolation of bacterial strains from the pilot-scale biotrickling filter equipped with Hiflow™ rings, 2–3 g of wet biomass were scratched off the topmost carrier of the column, suspended in 0.9% NaCl, and plated on solid mineral medium supplied with TBA as sole source of carbon. The composition of the mineral medium was previously described [46].

Determination of cell counts was performed by suspending 2–3 g of wet biomass in 10 mL of 0.9% NaCl and subsequent logarithmic dilution of this suspension. Aliquots of 100 µL of appropriate dilutions were either spread on Difco™ nutrient broth complex medium for total cell counts, or mineral medium plates supplied with TBA or acetone as sole carbon source for specifically counting cells able to degrade these contaminants. The isolates TBA100 and TBA200 were deposited in the DSMZ collection.

2.2. Genetical characterization of the isolates and microbial monitoring strategies

Bacterial isolates, capable of repetitive growth on TBA, were genetically characterized by PCR amplification of the 16S rRNA gene using standard oligonucleotides 27F (5′-AGAGTTTGATCMTGGCT CAG-3′) and 1492R (5′-TACGGYTACCTTGTTACGACTT-3′). PCR products were purified using the peqGOLD Cycle-Pure Kit (VWR Peqlab, Erlangen, Germany). 16S rRNA gene sequencing was carried out by GATC Biotech AG (Constance, Germany) and the results were compared to the GenBank Database at the National Center for Biotechnology Information (https://blast.ncbi.nlm.nih.gov/Blast.cgi). Corresponding gene sequences were deposited in GenBank under accession numbers MH580612 and MH580613.

Further differentiation of TBA200 and *Aquincola tertiaricarbonis* L10 was performed by PCR amplification of the gene sequence of 2-hydroxyisobutyl-CoA mutase using oligonucleotide primers hcmA-F (5′-CAC ACGATCGGGGACTTC-3′) and hcmA-R (5′-TGACCAAGCCGCAGT ACA-3′). Subsequent steps were performed as described.

To identify and quantify the strains TBA100 and TBA200 within the bacterial community of the pilot-scale biotrickling filter equipped with Hiflow™ rings, emerging isolates grown on mineral media plates supplied with TBA were further characterized by BOX-PCR fingerprinting. BOX-PCR was performed with these isolates as well as TBA100 and TBA200 as references using the primer BOX-A1R (5′-CTACGGCAAGG CGACGCTGACG-3′) [47]. Products of BOX-PCR were compared after separation by agarose gel electrophoresis using an extended DNA ladder as reference (T835.1, Carl Roth, Germany).