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Abatement robustness of volatile organic compounds using compact trickle-bed bioreactor: Biotreatment of styrene, ethanol and dimethyl sulfide mixture in contaminated airstream

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ABSTRACT

A wide range of volatile organic compounds (VOCs) chemical characteristics (aromatic, sulfur-containing organics and alcohol) were selected to demonstrate a prospective potential of the biotrickling technology in practical applications through the appropriate selection and combination of biological ‘agents’. The principal objective of this study was to specify operating boundaries of parameters at which the sampled microorganisms were most effective in the biodegradation of gaseous streams containing styrene, ethanol and dimethyl sulfide mixture at dynamic variations of pollutant load. The average conversion factor for the 3-component VOCs mixture was higher than 95% at lower range of the individual pollutant load and basically fell to 80% at middle range *vs.* 55% at the higher contaminant loads; however, the effectiveness of ethanol biodegradation is stable at the entire investigated range of the mass load. The consequences of an unexpected pollutant overload (media clogging) and the time necessary for the subsequent regeneration of the microbial community and restoring the process stability were investigated as well.

1. Introduction

Modern civilization is confronted with a worldwide rise of atmospheric pollution due to the expansion of industrial and agricultural areas as well as urban settlements. Volatile organic compounds (VOCs) known as ‘*compounds having vapor pressure exceeding 0.1 mmHg at standard conditions*’ and inorganic odorous compounds (VICs) pose hazards to the global ecosystem, health of human beings and plants vegetation forming a significant part of indoors/outdoors pollution created by gases emitted from certain solids or liquids (Golhosseini et al., 2013; Huang et al., 2016). The VOCs and odors in the atmosphere originate from anthropogenic activity or the biogenic emissions of certain reactive hydrocarbon derivatives formed as chemical (by-)products of natural transformations as well (Atkinson and Arey, 2003; Hu et al., 2016). The anthropogenic nitrogen and sulfur derivatives, (semi-)volatile compounds and toxic metals constitute a main class of air contaminants with increasingly detrimental impact on human health in the long-term exposure being classified as a potential mutagenic and carcinogenic risk factors (Kampa and Castanas, 2008). Individual VOCs exert some irritant and toxic effects, that depend on the genre of poisonous compounds/mixtures, the contaminant concentrations and the extent/time of exposure to the living organisms (Pérez et al., 2016; Ras-Mallorquí et al., 2007). Moreover, odor-causing mixtures of sulfur- and nitrogen-containing pollutants and their metabolites contribute to the forming of tropospheric and photochemical smog with noticeable impact on global warming effect. As a matter of fact, the atmospheric photooxidation of halogenated VOCs leads to the stratospheric depletion of ultraviolet radiation-absorbing ozone layer and also generates toxic

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organic aerosols (e.g. peroxyacetyl nitrate, PAN) especially harmful to human respiratory system (Atkinson, 2000; Mohamed et al., 2002; Cetin et al., 2003). In the last decades, the indirect impact of air contamination on human health and the surrounding environment were extensively scrutinized and better understood (Rumchev et al., 2004; Curtis et al., 2006). The increasing public awareness of necessity for environmental protection with nuisance-free, breathable air was the main driving force for the increasingly stringent regulations governing release of hazardous air pollutants (HAPs) and reduced sulfur compounds (RSCs). Environmental legislations are constantly pushing industry for reduction in emission of poisonous low-molecular weight gases and developing/optimizing of cost-effective 'green' manufacturing technologies that impose less burden on the ecosystem. Volatile organic compounds (VOCs) as '*any compound containing carbon, excluding carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates and ammonium carbonate*' represented by carbon-based chemicals like vinyl acetate, ethyl acetate, styrene or dimethyl sulfide, however less known to public, play invariably important role in air poisoning (Nielsen et al., 2007; Estrada et al., 2012; Lahel et al., 2016). Efficient eradication of odorous VOCs and VICs from industrial off-gases is a complex process due to very low threshold detection limits (concentration) of these toxic compounds; however a vast range of air treatment technologies was proposed basically falling into three categories: chemical, physical and biological (Fig. 1 of supplementary materials). Generally, two procedures of removing air contaminants (especially VOCs) from industrial gases can be distinguished based on the nature of applied transformations. The first technique comprises transfer and/or concentration of pollutants in a liquid phase (condensation, absorption/scrubbing) or onto solid phase (activated carbon adsorption), whereas the second one is related to the destruction/decomposition of molecules by thermal/catalytic oxidation, flaring or degradation by microorganism community to harmless (by-) products (Smet and van Langenhove, 1998; Spigno et al., 2003). Traditionally, the industrial waste gases were treated by physicochemical methods, which are costly (incinerators), use lots of chemicals (chemical scrubbers) and generate waste products that still require further treatment or disposal (scrubbers). On the other hand, the biopurification of the off-gases containing low concentration of volatile (in)organic compounds is gaining particular attention due to high cost-effectiveness (low investment and operating expenses), treatment efficiency and environmental acceptability as odorant removal/cleaning procedures are based on the natural ability of microorganisms to degrade odorous/toxic contaminants from industrial/municipal airstreams and operate in the mild conditions (at around ambient temperature and atmospheric pressure) (Attaway et al., 2001; Kasperczyk and Urbaniec, 2015; Horel and Schiewer, 2016). Briefly, pollutants diffuse into the liquid phase where indigenous or inoculated species of microorganisms or microbial consortium transform certain inorganic contaminants with the formation of stable odorless (by-)products such carbon dioxide, water, inorganic metabolites or elemental nitrogen and sulfur. As a matter of fact, pollutants cannot be degraded explicitly in the gaseous phase; therefore toxic compounds have to be first transferred with oxygen from flowing airstream to biologically active aqueous phase (solubilized), where the microbes, either growing suspended or attached to a support might attack and decompose contaminants (Tu et al., 2015). In fact, this microbial 'predators' use the pollutants as nutritional source to draw energy and carbon for growth (biomass production) and the maintenance of biological activity realized under aerobic or anaerobic conditions, while other nutrients (minerals and trace elements) should be supplied (Li et al., 2015; Wang et al., 2015). In a broader sense, a single/mixed microbial population or consortium can be regarded as the biocatalysts in the degradation process which continually evolve to meet the changes in pollutant feed and environmental conditions transforming pollutants into biomass and harmless (by-)products.

$$(\text{VOCs}, \text{VICs}) \xrightarrow{\text{microbes}, O_2, H_2O, \text{heat}, \text{minerals}} \text{biomass} + (\text{by}-)\ \text{products} \tag{1}$$

By definition, the efficient biological air treatment is typically limited to low molecular weight and highly soluble compounds with simple bond structure; however an implementation of biotechnology for removal of more complex pollutants from contaminated air was reported (Lebrero et al., 2010). Moreover, biotechnology offers '*clean at the end of pipe*' performance standards that means a very minor secondary emission without shifting pollution problem from air to other environmental compartments such as water or soil.

The proper design of an industrial biopurification process for VOCs treatment and deodorization requires certain information about kinetics and chemistry of given bio-degradation. Hence, the multi-step researches from small, lab-size glass reactors up to larger-scale, semi-industrial installation are necessary to reveal information in the following areas:

- choice of the microbial population or microorganism consortium able to biodegrade the specific contaminants from the waste airstream,
- adaptation of the microflora to the swinging process conditions,
- choice of the immobilization mode,
- removal efficiency of pollutant(s) as a function of process variables (flow, temperature, humidity, pressure, etc.) and chemical compound characteristics (composition, concentration, reactivity, solubility, etc.), especially at changing contaminant load.

Industrial air-phase bioreactors applied for odor and VOCs removal were basically divided into biofilters, biotrickling filters, bioscrubbers and membrane reactors (shown schematically in Fig. 2 of supplementary materials) depending on the water-phase mobility and microorganisms availability (fixed film cultures on a solid phase and/or suspended growth community in liquid) (Mudliar et al., 2010). In biofilters the contaminants are initially transported to the biofilm cultures that are fixed on the surface of the support as the polluted, previously saturated air passes through a bed of media to be subsequently metabolized by microflora (Kauselya et al., 2015). Similarly, in biotrickling filters pollutant is firstly solubilized in the falling liquid and then transferred to the biofilm growing on the support surface; however liquid is continuously recirculating through the packing providing moisture, nutrients, pH control, etc. Formally, the term 'biotrickling filter' means one unit system with a continuous flow of aqueous nutrients passing through a bed of synthetic media with a pretty low residual saturation and water retention capability (Iranpour et al., 2005). The flow of contaminated air is basically directed up- or down-flow that is counter- or co-current with the liquid flow. Bioscrubber is sometimes described as a kind of biotrickling filter, but it comprises a three-phase, fluidized-bed bioreactor system with water recirculated between two reactor units. The gaseous contaminant is eradicated by the absorption in a gas-liquid contactor while the pollutant-laden liquid is then regenerated by the bacterial community with supplementary oxygen (Potivichayanon et al., 2006). If the conditions of the stream exclude the direct contact with biomass the membrane bioreactors can be used, where the pollutants diffuse from the gas-phase to the biofilm through a semi-permeable hydrophobic membrane made of latex or silicon.

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