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A comparative study of mass transfer coefficients of reduced volatile sulfur compounds for biotrickling filter packing materials

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HIGHLIGHTS

- Mass transfer coefficients (K_Ga_e) were determined for biotrickling filter packing.
- K_Ga_e of sulfur compounds depend on both air velocity and liquid velocity.
- Correlation between K_Ga_e and key parameters describing packing was failed.
- K_Ga_e to pressure drop was used for selecting proper packing regarding mass transfer.

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ABSTRACT

Biofiltration is a cost-effective technology for reduction of odor emissions from intensive pig production, but removal of volatile sulfur compounds may be limited by low mass transfer. Among these volatile sulfur compounds, hydrogen sulfide and methanethiol are essential odorants emitted from animal houses according to the low odor threshold values. In order to get a better understanding of the limitations for optimization and further development of biofiltration for odor control, a study of overall mass transfer coefficients of volatile sulfur compounds for packing materials potentially used for biotrickling filters is presented. Mass transfer coefficients were obtained by taking advantage of online PTR-MS experiments of breakthrough curves, combined with a developed mass transfer model. The experiments were performed at different air and liquid velocities for selected packing materials. The results demonstrate that the overall mass transfer coefficients of volatile sulfur compounds depend on the velocity of both air and liquid. When the mass transfer coefficients are normalized to (divided by) the corresponding pressure drop, the results indicate that the filter packed with ceramic saddles or cellulose pads have higher normalized mass transfer coefficients, compared to the other materials investigated in this study. Correlating the mass transfer coefficients to characteristics of different packing materials did not succeed and as an alternative empirical relationship between mass transfer coefficients and the gas and liquid velocities were established.

1. Introduction

Odor emissions from intensive animal production and related activities are becoming a severe nuisance to neighbors in the local surroundings [1–3]. Besides the nuisance, the odor emissions may have negative effects to human health [4,5]. Biofiltration has been recognized as a cost-effective technology for reducing the odorants from livestock buildings [6–10]. Whereas the water-soluble odorants (e.g., phenols and carboxylic acids) have been observed to be removed efficiently in biotrickling filters, low removal efficiencies have been observed for volatile sulfur compounds of methanethiol (MT) and dimethyl sulfide (DMS) [9–11], which are in low concentrations (low range of ppbv) but recognized as potential key odorants in the ventilation air to be treated [10,12]. For H_2S, another key odorant, only low to medium removal efficiencies (with concentrations ranging typically from less than one hundred to a few hundred ppbv) were observed in the biotrickling filters dealing with odorants from pig facilities [10,11]. It should be noted that, some previous studies on biotrickling filters indicated that H_2S (with relatively high concentrations of at least 5–10 ppmv) could achieve very high removal under acidic or neutral conditions

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Nomenclature

A, B, C	the longest, the intermediate and the shortest axis of packing
a	specific surface area ($m^2\ m^{-3}$)
a_e	effective specific interfacial area ($m^2\ m^{-3}$)
k_1, k_2, k_3	empirical coefficient (–)
C_g, C_l	gas and liquid contaminant concentration ($mol\ m^{-3}$)
d_c, h_c	cylinder diameter and height for cylinder shape material (cm)
D_p	dispersion-diffusion coefficient ($m^2\ s^{-1}$)
d_p	diameter of packing particles (cm)
H	dimensionless Henry's law constant ($mol_{gas}\ m_{gas}^{-3}/mol_{liquid}\ m_{liquid}^{-3}$ or –)
K_Ga_e	overall volumetric air–water mass transfer coefficient (s^{-1})
M_p	mass of packing under dry condition (g)
M_w	mass of packing under wet condition (g)
Q	air flow rate ($m^3\ s^{-1}$)
r	radius of the filter cross section (cm)
u	pore air velocity ($m\ s^{-1}$)
v_g, v_l	gas velocity ($m\ s^{-1}$) and liquid velocity ($m\ h^{-1}$)
V_{tot}	total volume in Eq. (1) (cm^3)
V_w	volume of water in Eq.(1) (cm^3)
Z	the absorption column depth (m)
Greek letters	
α, β	empirical coefficient (–)
ε_{ex}	external porosity (–)
θ_a	active volumetric water content ($m^3\ m^{-3}$)
ρ_b	the mean dry bulk density ($g\ cm^{-3}$)
ρ_d	the media solid density ($g\ cm^{-3}$)
ρ_p	the mean particle density ($g\ cm^{-3}$)
ϕ	the total porosity (–)
ψ	the sphericity factor of packing material
ω	water holding capacity (–)

even for empty bed residence time (EBRT) of a few seconds [13,14]. Such short EBRT (typically from <1 s to <10 s) are also commonly applied to the biological air filters with relatively open filter beds dealing with ventilation air from livestock buildings, in order to keep the filter size small while obtaining low pressure drop [8,15–17]. Consequently, the mass transfer resistance of the sulfur compounds from the gas phase to the liquid phase is considered to be an important limitation for obtaining better removal of volatile sulfur compounds [9,11,18,19]. A recent study on partition coefficients of these sparingly soluble compounds observed uptake rates in biotrickling filter liquids to be as low as in water, which makes the removal of these compounds in biotrickling filters a further challenge [20].

Presently, only limited data on mass transfer coefficients at conditions suitable for biotrickling filters is available but there has been an emerging interest for characterizing the mass transfer in biotrickling filters in recent years [19,21–27]. While most of the existing data is based on measurements of oxygen or toluene, determination of mass transfer coefficients by using target pollutants has been recommended [22,24]. In fact, the mass transfer coefficients may dependent on the physical and chemical properties of the compound as well as the characteristics of the packing materials and operating conditions [28]. Still, predicting mass transfer coefficients by correlations can be valuable as an alternative to experimental estimation. However, a few recent studies indicated that simple power law relationships between the mass transfer coefficients and air/liquid velocities gave much better performance than traditional correlations (for example the Onda equations [29]), probably due to the fact that the hydrodynamics used in these correlations are dramatically different than typical operating conditions in biotrickling filters [22,24,27]. Based on a power function, Kim and Deshusses [22] proposed a simple empirical expression with two empirical constants which are unique for each packing material suitable for biofilters/biotrickling filters (include compost + woodchips, polyurethane foam cubes, lava rock, pall rings, porous ceramic beads or raschig rings). These authors observed an overestimation of mass transfer coefficients with a factor of 3–20 by the Onda equation [22]. Dorado et al. [24] evaluated the overall mass transfer coefficients for selected packing materials for biotrickling filters (polyurethane foam cubes, lava rock, stainless-steel Dixon rings and clay pellets), by comparing commonly used correlations (including the Onda equation) together with the recent relevant study (at that moment) by Kim and Deshusses [22]. However, no correlation was found to be satisfied with the accurate prediction of mass transfer coefficients they measured and thereby obtaining experimental data fitted to simple mathematical expression was recommended in their study [24]. Further, a recent study by San-Valero [27] investigated the mass transfer coefficients of isopropanol (in the packing of random packing of Flexiring 25 mm and structure packing PAS Winded Media) and oxygen (in the packing of above ones plus a random packing of Fefilltech 15 mm), and compared the performance of mass transfer coefficient prediction by a few correlations including the Onda equation and expressions from Kim and Deshusses, as well as a self-established power expression from their study. While the Onda equation seems to predict the oxygen mass transfer coefficients accurately, all other correlations failed except the power expression established in their study [27]. The authors therefore also recommended to use experimental overall mass transfer coefficients with simple power expressions to air or liquid velocities.

Thus the purpose of this study is to experimentally determine the mass transfer coefficients of volatile sulfur compounds under different biotrickling filter operation conditions (e.g., packing, air velocity and liquid velocity), in order to obtain a better understanding of the limitation and to reach further improvement regarding the removal of these compounds as well as further development of biotrickling filters dealing with odorant emissions from livestock production. In a previous study, a method for estimation of mass transfer coefficients of volatile sulfur compounds was demonstrated through the fast and sensitive measurement of breakthrough curves by PTR-MS combined with a numerical model [30]. The method was further validated by application on strictly selected fractions of one material (Leca®) and a correlation between mass transfer coefficients and particle size, air velocity, material specific surface area and contaminant chemical properties was demonstrated [31]. The correlations established were based on simple power functions, in concordance with the previous studies [22,24], except the fact that the particle sizes, specific surface area and chemical properties were also included in the fitting procedure, befitted from the strictly selected fractions.

In the present study, different selected packing materials potentially used for biotrickling filters were characterized firstly. The mass transfer coefficients of volatile sulfur compounds were determined under various air and liquid velocities. Simple power mathematical model were then tested for prediction of mass transfer coefficients.