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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.

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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.

Table 1
Main characteristics of the packing materials used in this study (a: specific surface area ($m^2\ m^{-3}$); ρ_b: bulk density ($g\ cm^{-3}$); ρ_p: particle density ($g\ cm^{-3}$); ρ_d: media solid density ($g\ cm^{-3}$); ϕ: total porosity (%); ε_{ex}: external porosity (%); ω: water holding capacity (g)). Numbers in brackets indicate one standard deviation.

Material	Leca®	Cellulose	Lava rock	IVP[a]	AP4-60	Woodchips	BIO[b]	CYR[c]	Saddles[d]	DMT-HS	MOL[e]
Main composition	Clay[f]	Cellulose	Lava rock	Carbon[g]	Carbon[h]	Woodchips	PE virgin	Steatite C221	Stoneware	Plastic	Diatom[i]
Shape	Beads	Structure	Irregular	Cylinder	Pellet	Irregular	Cylinder	Cylinder	Saddle	Structure	Beads
Dimension (mm)	8–10	Structure	8–16	4 * 6	4	Irregular	9 * 7	ca.6[j]	¾″	Structure	3–6
a ($m^2\ m^{-3}$)	372 (1.2)	383[k] (–)	458 (8.4)	926 (10.1)	900 (9.3)	532 (6.8)	800 (–)	800 (–)	338 (–)	432[l] (–)	1081 (9.8)
ρ_b ($g\ cm^{-3}$)	0.25 (0.001)	0.02 (–)	0.96 (0.008)	0.53 (0.006)	0.42 (0.004)	0.21 (0.009)	0.14 (0.001)	0.94 (0.004)	0.65 (0.006)	0.13 (–)	0.48 (0.005)
ρ_p ($g\ cm^{-3}$)	0.39 (0.002)	1.2 (–)	1.55 (0.03)	0.98 (0.008)	1.01 (0.007)	0.35 (0.02)	0.20 (0.001)	1.61 (0.01)	1.64 (0.01)	0.95 (–)	0.63 (0.005)
ρ_d ($g\ cm^{-3}$)	2.5 (–)	1.2 (–)	2.24 (0.07)	1.47 (0.02)	1.36 (0.03)	0.52 (0.03)	0.72 (0.03)	2.69 (0.07)	2.62 (0.06)	0.95 (–)	1.57 (0.05)
ϕ (%)	91 (0.4)	98	57 (1.8)	64 (0.9)	69 (1.5)	58 (3.3)	80 (3.4)	65 (1.7)	75 (1.7)	87	69 (2.1)
ε_{ex} (%)	37 (0.2)	98	38 (0.7)	46 (0.5)	58 (0.6)	39 (2.2)	28 (0.2)	41 (0.3)	60 (0.5)	87	23 (0.2)
ω (g/g)	0.76 (0.006)	1.47 (–)	0.18 (0.002)	0.56 (0.003)	0.50 (0.003)	1.05 (0.01)	0.61 (0.005)	0.03 (0.001)	0.11 (0.001)	0.31 (–)	0.95 (0.004)
Supplier	Weber[m]	Munters[n]	VDL[o]	Calgon[p]	Calgon	Local[q]	Hansa[r]	Hansa	Hansa	DMT[s]	Damolin[t]

[a] IVP 4 * 6.
[b] Bioflow 9.
[c] Cylindrial ring.
[d] Ceramic saddles.
[e] Molergranulat or "Mo-clay", a specific type of mineral clay granulates.
[f] Lightweight expanded clay.
[g] Impregnated coal based.
[h] Coal-based extruded.
[i] 30% Clay, 70% Diatom.
[j] Height: 4.3–4.9 mm, OD: 5.7–6.3 mm, ID: 3.3–3.7 mm.
[k] Data obtained from [47].
[l] Data obtained from DMT Milieutechnologie BV, Netherlands.
[m] Weber A/S Randers, Denmark.
[n] Munters A/S, Denmark.
[o] VDL Siergrind BV, Netherlands.
[p] Calgon Carbon Corporation.
[q] Local Supplier, Randers, Denmark.
[r] Hansa Engineering AB, Sweden.
[s] DMT Milieutechnologie BV, Netherlands.
[t] Damolin A/S, Denmark.

2. Materials and methods

2.1. Packing material

11 different packing materials were selected in this study for the comparative test of mass transfer coefficients (Table 1). The characterization of the materials was done according to the methods described in [30,31]. The mean dry bulk density (ρ_b, $g\ cm^{-3}$) for each material except for cellulose and DMT-HS was determined in triplicates by filling a 20 L bucket with each dry material and the according weight. For cellulose and DMT-HS, the materials were cut into cylindrical pieces with diameter of 25 cm, in order to tightly fit into the absorption column of ID 25 cm (thus no shortcuts for air flow on the edges) before these were dried and weighted. Thus, ρ_b is determined according to the weight and calculated volume of the cylinder pieces included. The media solid density (ρ_d, $g\ cm^{-3}$) for each material except for Leca®, cellulose and DMT-HS was determined through water displacement method [32]. The randomly selected number of dry particles (10–50 depending on particle size of each material) were weighed (M_p, g) and carefully immersed into a beaker with a known volume of deionized water (V_w). Then the total volume level was read (V_{tot}). The ρ_d was then obtained as follows:

$$\rho_d = \frac{M_p}{V_{tot} - V_w} \tag{1}$$

the ρ_d for Leca® could not been determined this way due to its floating on the surface of water and The ρ_d for Leca®, cellulose and DMT-HS was assumed to be 2.5, 1.2 and 0.95 for clay, cellulose and plastic materials, respectively.

The mean particle density (ρ_p, $g\ cm^{-3}$) for Leca®, MOL and AP4-60 (AP4) was obtained by the total weight (M_p, g) and the particle volume for randomly selected individual particles (for Leca®, $n = 10$ and for MOL and AP4, $n = 20$) of the packing material assuming smooth surface and ellipsoid shape. Then the ρ_p was calculated according to:

$$\rho_p = \frac{M_p}{\sum\left(\frac{4}{3}\pi A\ B\ C\right)} \tag{2}$$

where A is the longest axis; B is the intermediate axis and C is the shortest axis (with A, B, and C are perpendicular to each other). For IVP 4 * 6 (IVP), Bioflow 9 (BIO) and Cylindrial Ring (CYR) (all are in cylinder shape), the ρ_p was determined according to (for IVP, $n = 20$, for BIO and CYR, $n = 10$):

$$\rho_p = \frac{M_p}{\sum\left(\frac{1}{4}\pi d_c^2 h_c\right)} \tag{3}$$

where d_c is the cylinder diameter (cm) and h_c is the cylinder height (cm). For cellulose and DMT-HS, the ρ_p was assumed to be equal to ρ_d, respectively. For lava rock, woodchips and ceramic saddles, the ρ_p was assumed as below:

$$\left.\begin{aligned} \rho_p &= \frac{M_p}{\frac{1}{5}\times\frac{M_p}{\rho_b} + \frac{4}{5}\times(V_{tot} - V_w)} \quad \textit{(for lava rock and woodchips)} \\ \rho_p &= \frac{M_p}{\frac{1}{5}\times\frac{M_p}{\rho_b} + \frac{4}{5}\times(V_{tot} - V_w)} \quad \textit{(for ceramic saddles)} \end{aligned}\right\} \tag{4}$$

The total porosity ϕ and the external porosity ε_{ex} were then determined as follows:

$$\phi = 1 - (\rho_b \rho_d^{-1}) \quad (5)$$

$$\varepsilon_{ex} = 1 - (\rho_b \rho_p^{-1}) \quad (6)$$

The specific surface area ($a, m^2 m^{-3}$) for saddles, BIO and CYR were obtained from the supplier. The specific surface area of Leca® was estimated by the method described in [31]. For Woodchips, a was estimated assuming a rectangular shape. For lava rock, IVP, AP4 and MOL, a was determined as below [24]:

$$a = \frac{6(1 - \varepsilon_{ex})}{\psi d_p} \quad (7)$$

where d_p is the particle diameter and ψ is the sphericity factor of each packing material. The sphericity factor ψ was calculated as follows [33]:

$$\psi = \sqrt[3]{\frac{BC}{A^2}} \quad (8)$$

The particle diameter d_p was obtained as below:

$$d_p = \frac{\sum_1^n (A + B + C)}{3n} \quad (9)$$

Water holding capacity ω was determined as below:

$$\omega = \frac{M_w - M_p}{M_p} \quad (10)$$

where M_w is the weight of the material under wet condition (saturated with water and then drained for 2 h).

2.2. Experimental set-up

A lab-scale absorption column (25 cm ID, stainless steel) was used for the experiments in this study as shown in Fig. 1. Each material except for cellulose and DMT-HS was wetted to its maximum water holding capacity by immersion in deionized water for at least 3 days followed by a 2 h draining period, before placing it into the absorption column forming approximately 100 cm (±3 cm) height of packing. For cellulose and DMT-HS, cylindrical pieces of dry material were pushed carefully into the column to a height of roughly 90 cm. The air flow rate was controlled by a CUBUFAN 160 EC ventilation pump (Jenk, Brøndby, Denmark) and recorded via a VA400 thermal mass flow sensor (CS instruments, Tannheim, Germany). For the low trickling flow rates (0.02 m h^{-1} and 0.37 m h^{-1}), the deionized water was pumped by a metering pump (C30, Iwaki pumps, Japan) to the top of the absorption column (ca. 35 cm above the packing), mixed with pressurized air (ca. 0.5 bar) and injected into the air stream against the air flow direction through a jet nozzle (Hardi 1553-08, Nørre Alslev og Taastrup, Denmark) with a Syntal swivel (Hardi 1154, Nørre Alslev og Taastrup, Denmark). For the high trickling flow rates (1.83 m h^{-1}, 3.67 m h^{-1} and 6.11 m h^{-1}), the pressurized tap water was used instead of deionized water (pre-tests showed that there was no difference of obtained breakthrough curves and mass transfer coefficients between deionized water and tap water) and injected into the air stream (against air flow direction) via a SPRAJET nozzle (B 1/4 HHSJ-9007, Spraying Systems Co., USA). The trickling flow was controlled through a rotameter and the pressure drop was measured by a manometer. Teflon (FEP) tubing was used for all sampling and gas injection lines. Two 5 μm Teflon filters (Millipore Millex-LS, Billerica, USA) were placed at the inlet of the gas sampling lines.

2.3. Experimental procedure

A pulse of single air pollutant (H_2S, MT, DMS, dimethyl disulfide namely DMDS or dimethyl trisulfide namely DMTS) provided via a gas cylinder (1000 ppmv, Nitrogen based, AGA A/S, Denmark) was introduced to the ambient air stream (0.7 m after the blower and 2.6 m before the inlet) via solenoid valves (Burkert model 0124, Burkert, Germany) connected to the external device interfaces (24 V) from a high-sensitivity PTR-MS (Ionicon Analytik, Innsbruck, Austria) (Fig. 1). The PTR-MS was operated under standard conditions with a drift tube voltage, temperature and pressure range of 600 V, 60 °C and 2.1–2.2 mbar, respectively. The E/N number was maintained at ca. 135 Td. The transmission efficiencies were estimated through a gas mixture containing 14 aromatic compounds (P/N 34423-PI, Restek, USA). For H_2S, the humidity dependence was calibrated during the experiments by applying the method described in [12]. The PTR-MS was thus utilized to measure the inlet and outlet profiles with a procedure of 5–5–3 (5 inlets, 5 outlets and 3 inlets) for individual volatile sulfur compound under various air flow and liquid flow rates. The dwell time for each compound was 100 ms during single experiment.

While passing through the absorption column, the contaminant in the gas phase will be subject to advection, diffusion, dispersion and mass exchange with the liquid phase. Thus the gas phase concentration could be described as below [30]:

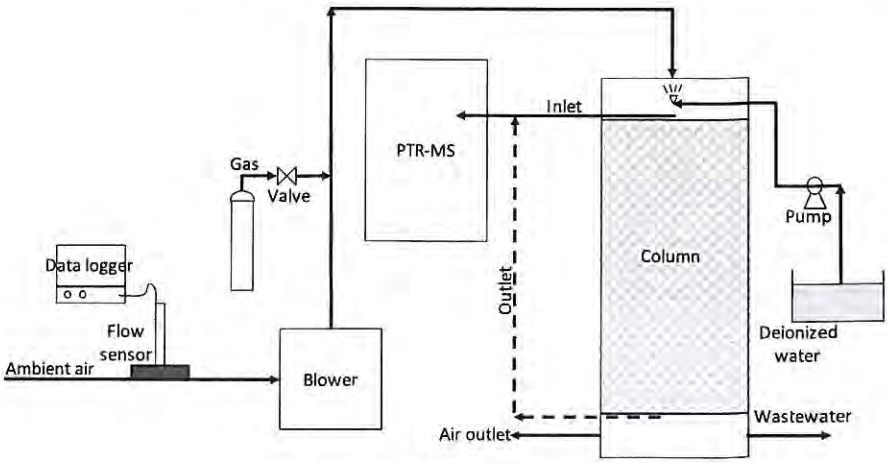

Fig. 1. Scheme of the experimental set-up.

$$\frac{\partial C_g}{\partial t} = D_p \frac{\partial^2 C_g}{\partial z^2} + u\frac{\partial C_g}{\partial z} - K_G a_e (C_g - C_l H) \quad (11)$$

where C_g is the gas phase concentration (mol m^{-3}); D_p is the dispersion–diffusion coefficient (m^2 s^{-1}); z is the column depth (m); u is the pore air velocity (m s^{-1}); $K_G a_e$ is the overall volumetric air–water mass transfer coefficient (s^{-1}); C_l is the liquid phase concentration (mol m^{-3}); H is the dimensionless Henry's law constant (mol_{gas} m^{-3}_{gas}/mol_{liquid} m^{-3}_{liquid} or –).

In addition, the water phase concentration is given as below assuming no advective and diffusive/dispersive transport in the water phase:

$$\frac{\partial C_l}{\partial t} = K_G a_e (C_g - C_l H) \cdot \frac{\varepsilon_{ex}}{\theta_a} \quad (12)$$

where θ_a is the active water content (m^3 m^{-3}).

The pore air velocity u is estimated as described below:

$$u = Q/(\pi \cdot r^2 \cdot \varepsilon_{ex}) \quad (13)$$

where Q is air flow rate (m^3 s^{-1}) and r is the radius of the filter cross section (m).

Having the average inlet and outlet profiles as model inputs, the $K_G a_e$ can thus be estimated through the fitting process as described in [30]. For IVP and AP4, however, it was not possible to determine the $K_G a_e$ by this method due to the strong adsorption of the volatile sulfur compounds to the two materials (thus no well-defined peak in the outlet profile). Thus, these two materials were excluded for the mass transfer determination experiments, but the characterization of these two materials were still included in Table 1 for general information.

3. Results and discussion

3.1. Characterization of the packing materials

The specific surface area (a), water holding capacity (ω) and bed porosity (ϕ and ε_{ex}) are traditionally considered to be important parameters regarding mass transfer. The various selected packing materials in this study covered a wide range of specific surface areas and bed porosities as shown in Table 1. While the MOL has the highest (a), the bed porosity is relatively lower than the others. The structured packing materials (cellulose and DMT-HS) have relatively high (ε_{ex}) but they cannot be compared directly to those from the random packing materials due to the different ways of packing plus different considerations of particle density (for structure packing assuming the particle density = media solid density). For the random packing, the packing of ceramic saddles has the highest external porosity (ε_{ex}) among the others but the specific surface area is relatively low, which was obtained from the supplier. A close look of the ceramic saddles showed very small pores on the surface, which was suspected not be considered when the specific surface area was determined. Generally, both the specific

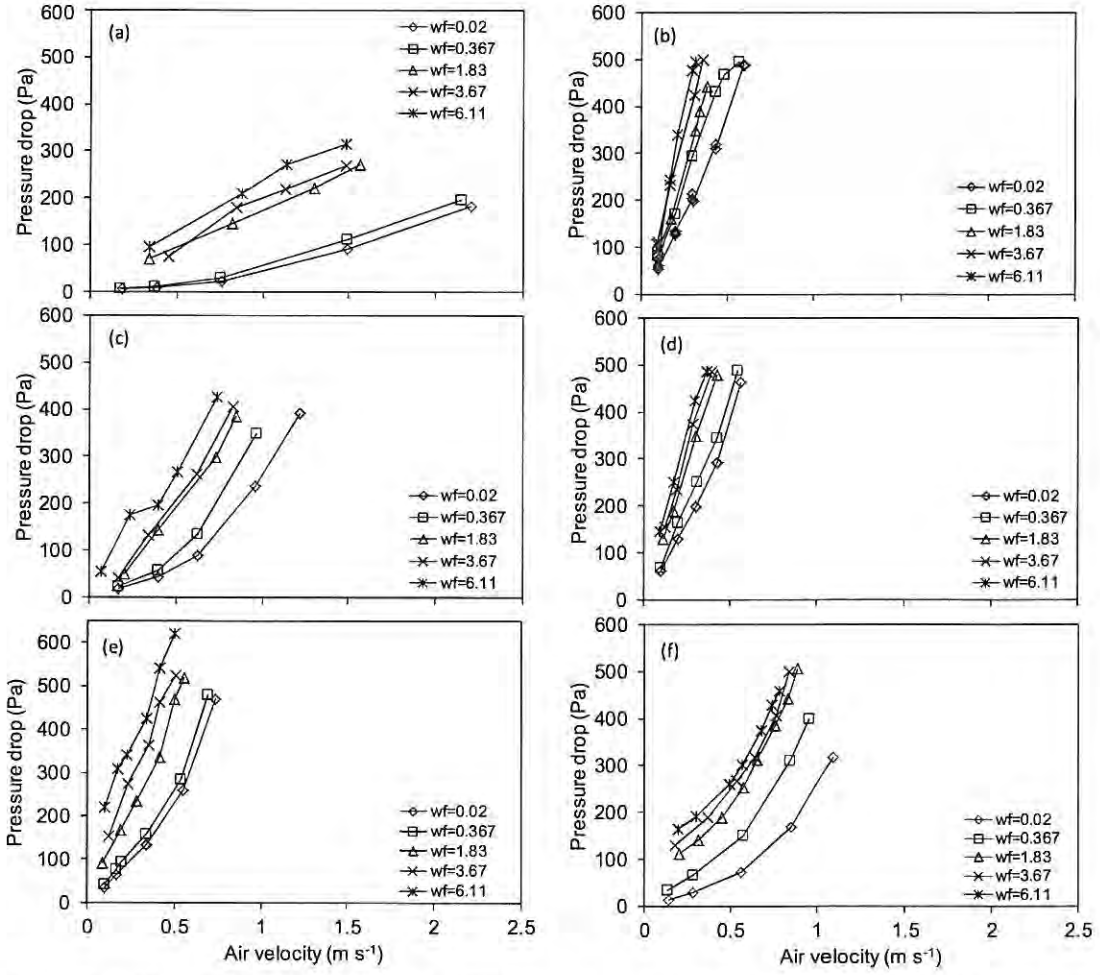

Fig. 2. Pressure drop over the bed for packing materials: Cellulose (a), Leca® (b), DMT-HS (c), woodchips (d), bioflow 9 (e) and ceramic saddles (f). wf: liquid velocity (m h^{-1}).

surface area and the media pressure drop are negatively correlated with the size of the packing material [34].

3.2. Pressure drop

Pressure drop was observed to increase with increased air velocity for all selected packing materials (Fig. 2 and Fig. S1). Cellulose and DMT-HS, which are structured packing materials, introduced the lowest pressure drop for the filters under the same level of air velocity and liquid velocity when compared to random packing materials. Among the random packing filters, ceramic saddles filter showed the lowest pressure drops (Fig. 2). The packing of MOL and CYL (Fig. S1), on the other hand, gave the highest pressure drops compared to any other packing material selected here (not

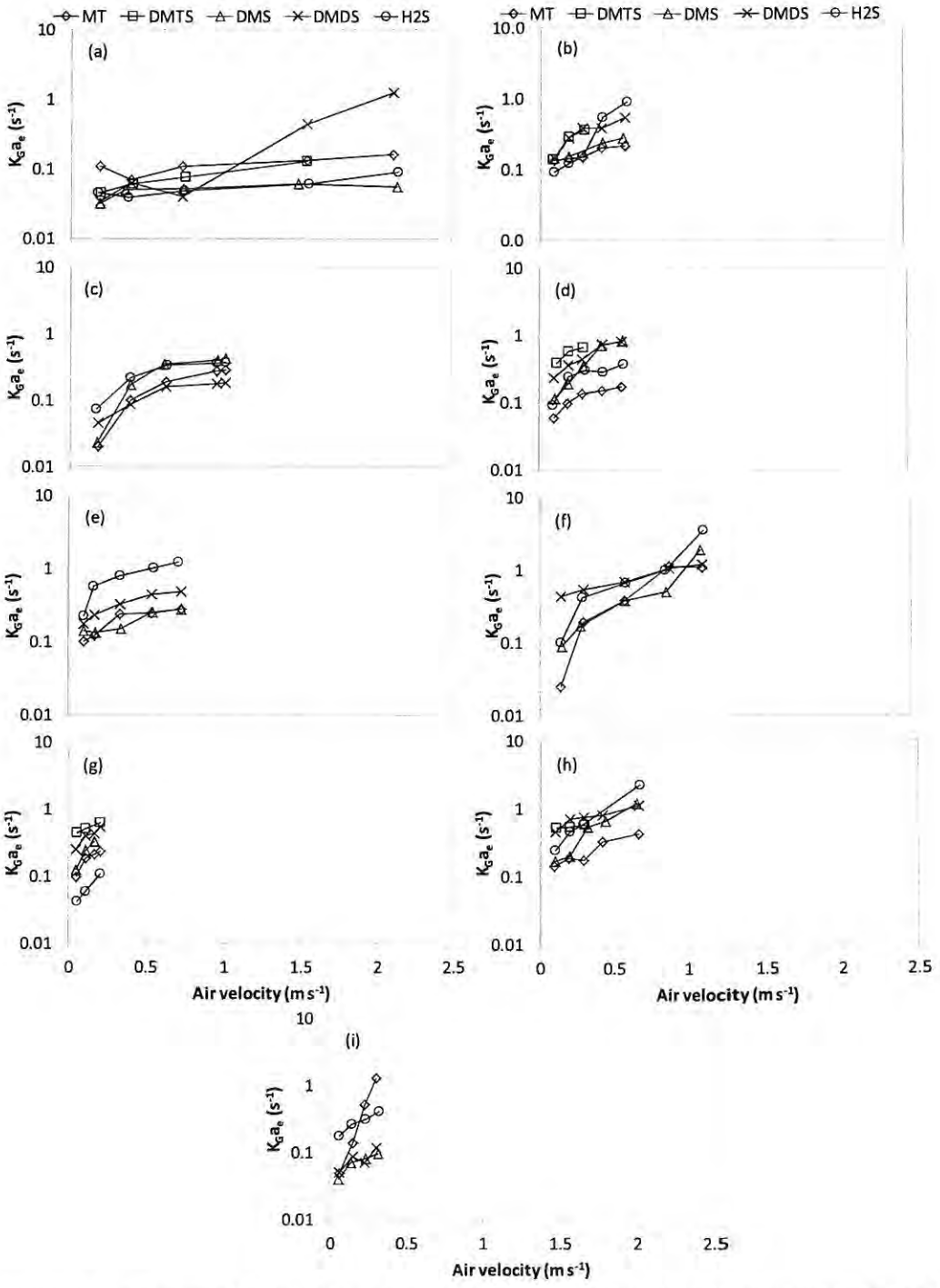

Fig. 3. Mass transfer coefficient (K_Ga_e) as a function of air velocity for packing materials: Cellulose (a), Leca® (b), DMT-HS (c), woodchips (d), Bioflow 9 (e), ceramic saddles (f), molergranulat (g), lava rock (h) and cylindrial ring (i) at liquid velocity = 0.02 m h^{-1}.

including IVP and AP4). All the packing materials also showed increasing pressure drop at increasing liquid velocity except for MOL, for which the pressure drop was already high even with the lowest liquid velocity and only increased gently with increased liquid velocity (Fig. S1).

3.3. Experimental data of K_Ga_e

The dependency of K_Ga_e on air velocity was mainly tested at a liquid velocity level of 0.02 m h^{-1} with a few cases tested at a liquid velocity level of 0.37 m h^{-1}. The dependence of K_Ga_e on liquid velocity, on the other hand, was performed at medium air velocity for each packing material. As shown in Fig. 3, the K_Ga_e generally increased for all packing materials with increased air velocity. For the packing of Leca,® while the K_Ga_e of MT, DMS and dimethyl disulfide (DMDS) increased moderately with increasing air velocity, the K_Ga_e of H_2S increased strongly when the air velocity was higher than 0.5 m s^{-1} (Fig. 3b). The K_Ga_e of H_2S for BIO (Fig. 3e) and lava rock (Fig. 3h) also increased rapidly with increased air velocity while K_Ga_e for other compounds increased more gently. This trend was also observed at a liquid flow rate of 0.37 m h^{-1} for Leca® (Fig. 4b). It should be noted that due to the

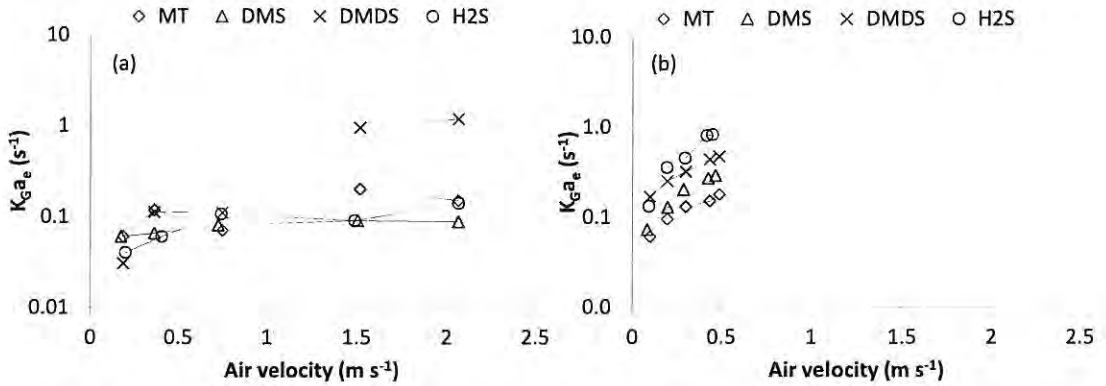

Fig. 4. Mass transfer coefficient (K_Ga_e) as a function of air velocity for packing materials: Cellulose (a) and Leca® (b) at liquid velocity = 0.37 m h^{-1}.

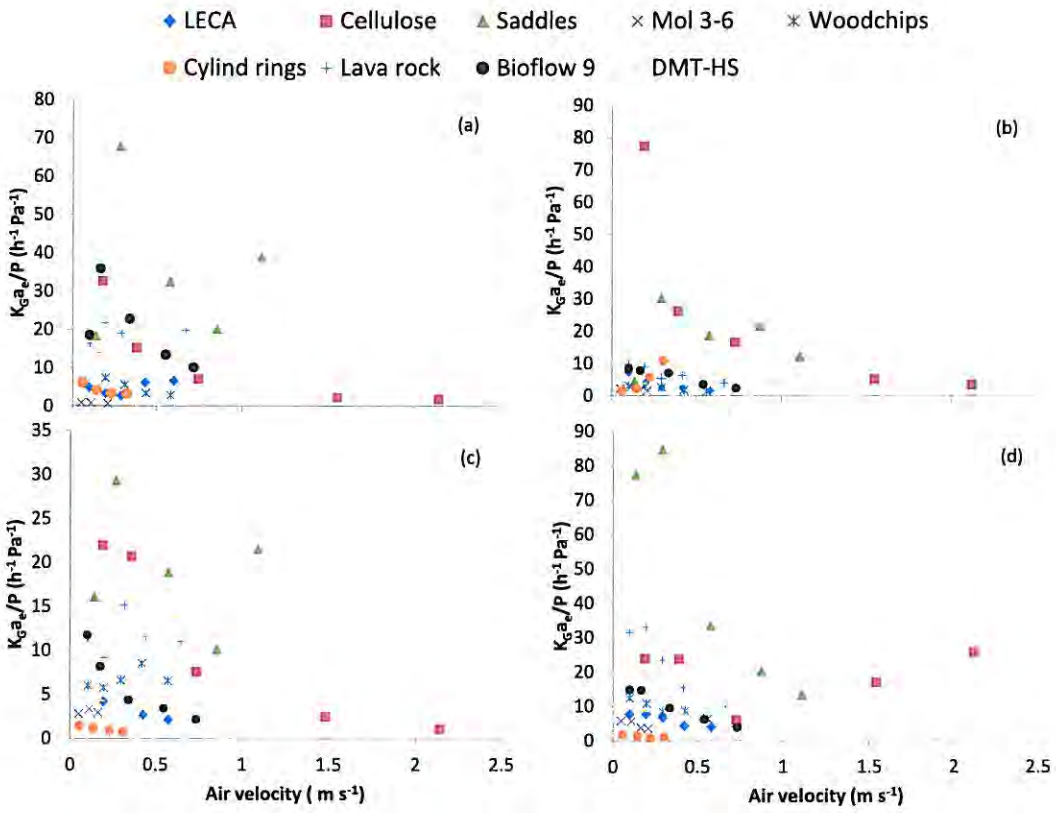

Fig. 5. Normalized mass transfer coefficient (K_Ga_e) to bed pressure drop (P) as a function of air velocity for H_2S (a), MT (b), DMS (c) and DMDS (d) at liquid velocity = 0.02 m h^{-1}.

dissociation of H_2S in water (into HS^- depending on pH), combined with the potential proton transfer chemical reaction of H_2S especially in Leca® [35], it is likely that the K_Ga_e of H_2S was overestimated. However, it is rather difficult to determine the pH on the liquid surface (and thus the gradient of pH) and the reactivity of H_2S with the packing materials, and thus it is difficult to estimate exactly how much overestimation was made for the K_Ga_e of H_2S. In overall, measured K_Ga_e of the sulfur compounds are comparable for all packing materials, although the structured packing especially the cellulose packing material showed a bit lower values compared to the others at the same air velocity. The dependence of K_Ga_e on air velocity at a level of liquid velocity of 0.37 m h^{-1} for cellulose and Leca® basically showed a trend similar to that obtained at a liquid velocity of 0.02 m h^{-1}.

Increased K_Ga_e at increasing air velocity have been reported in numerous studies [23,24,27,31,36–38] and may be explained partly by the increased turbulence within the packed bed and the occurrence of reduced stagnant air zones unavailable for mass transfer [39]. The liquid turbulence may also be increased significantly during the increase of air velocity. Further, a few studies have indicated that the effective interfacial area for mass transfer can be enlarged by the increased gas flow causing higher instabilities of liquid flow, leading to ripples, waves and droplets creation at the sheets [36,37,40,41]. This effect may be particularly important for materials with lower a (e.g., saddles, Leca® and lava rock) than the materials with higher a (e.g., MOL, CYL and BIO) [37].

Fig. 5 summarized the K_Ga_e of volatile sulfur compounds normalized by (divided by) the corresponding pressure drop for

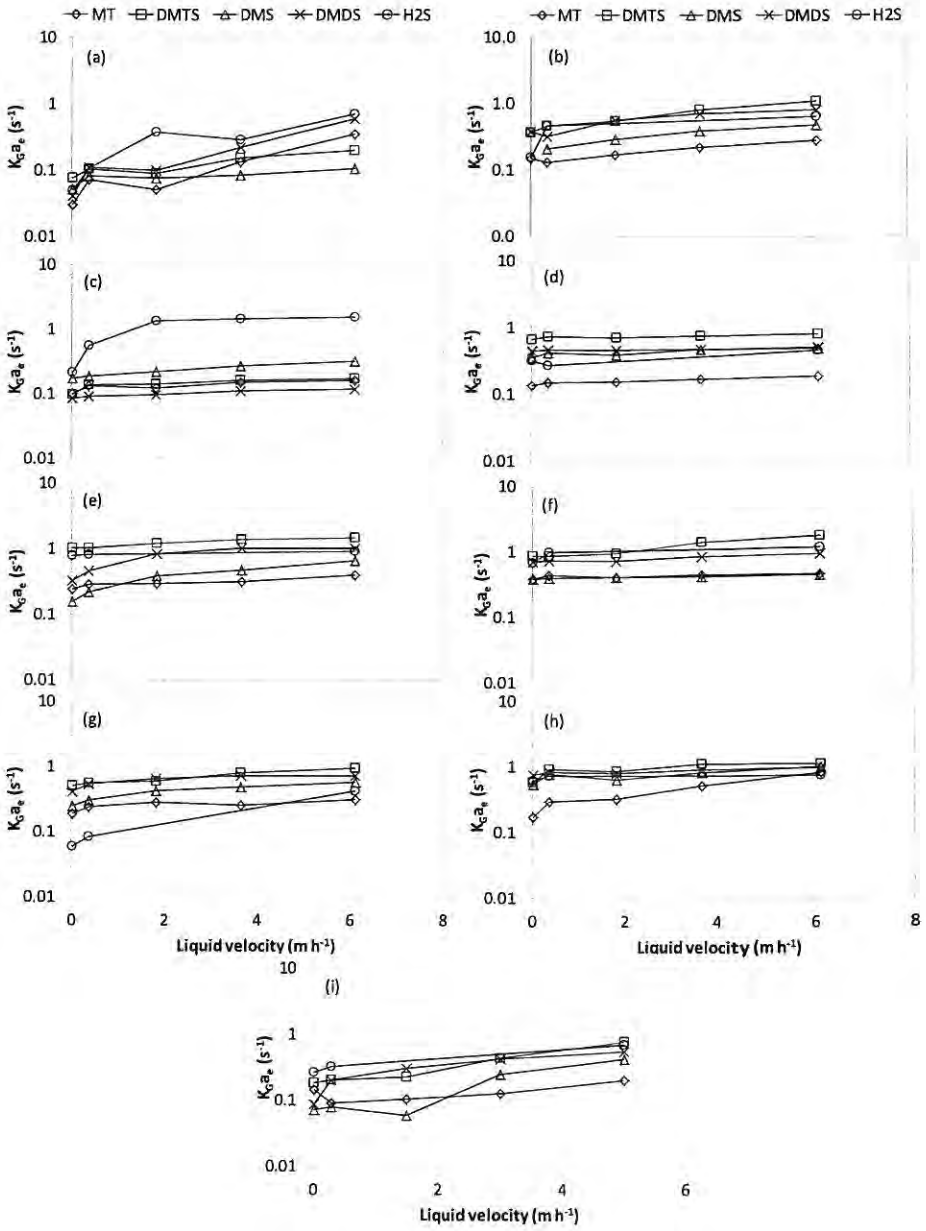

Fig. 6. Mass transfer coefficient (K_Ga_e) as a function of liquid velocity for packing materials: Cellulose (a; air velocity = 0.8 m s^{-1}), Leca® (b; air velocity = 0.3 m s^{-1}), DMT-HS (c; air velocity = 0.4 m s^{-1}), Woodchips (d; air velocity = 0.3 m s^{-1}), Bioflow 9 (e; air velocity = 0.34 m s^{-1}), Saddles (f; air velocity = 0.57 m s^{-1}), molergranulat (g; air velocity = 0.11 m s^{-1}), lava rock (h; air velocity = 0.3 m s^{-1}) and cylindrial ring (i; air velocity = 0.14 m s^{-1}).

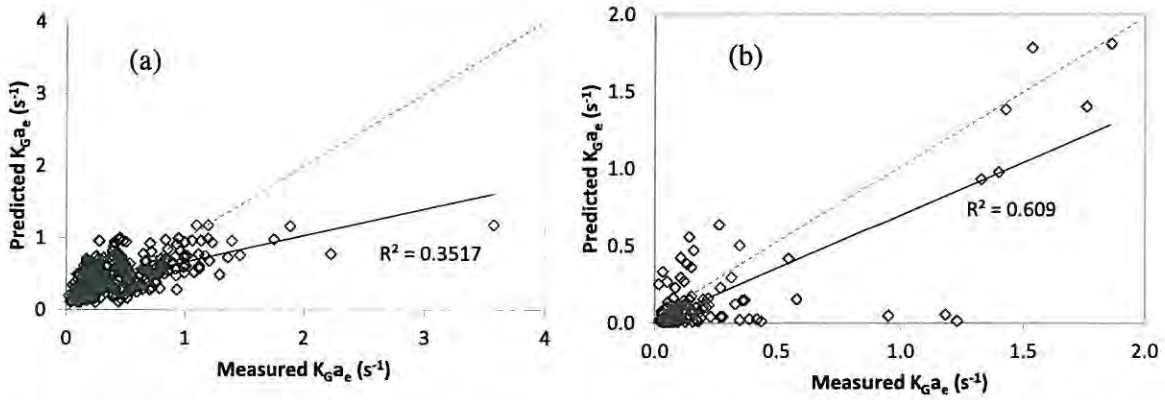

Fig. 7. Predicted mass transfer coefficient (K_Ga_e) as a function of measured mass transfer coefficient (K_Ga_e) for (a) random packing and (b) structure packing.

Table 2
Fitting parameters (k_1, k_2 and k_3) for simple power function model for mass transfer coefficient (K_Ga_e) of sulfur compounds related to air velocity (V_G) and liquid velocity (V_L) Packing materials: (a) Cellulose, (b) Leca®, (c) DMT-HS, (d) woodchips, (e) bioflow 9, (f) ceramic saddles, (g) molergranulat, (h) lava rock and (i) cylindrial ring. chemicals: MT (methanethiol); DMS (dimethyl sulfide); DMDS (dimethyl disulfide); DMTS (dimethyl trisulfide); H_2S (hydrogen sulfide).

		V_G (m/s)	V_L (m/h)	k_1	k_2	k_3	RMSE
(a)	MT	0.2–2.1	0.02–6.1	0.167	0.522	0.139	0.07
	DMS	0.2–2.1	0.02–6.1	0.085	0.199	0.122	0.01
	DMDS	0.2–2.1	0.02–6.1	0.365	1.919	0.090	0.13
	DMTS	0.2–2.1	0.02–6.1	0.124	0.468	0.078	0.03
	H_2S	0.2–2.1	0.02–6.1	0.244	0.901	0.587	0.06
(b)	MT	0.1–0.6	0.02–6.1	0.301	0.436	0.054	0.04
	DMS	0.1–0.6	0.02–6.1	0.751	0.780	0.200	0.05
	DMDS	0.1–0.6	0.02–6.1	1.058	0.604	0.147	0.09
	DMTS	0.1–0.3	0.02–6.1	1.433	0.735	0.144	0.10
	H_2S	0.1–0.6	0.02–6.1	3.45	1.519	0.159	0.06
(c)	MT	0.1–1.0	0.02–6.1	0.510	1.523	0.131	0.02
	DMS	0.1–1.0	0.02–6.1	0.927	1.664	0.174	0.06
	DMDS	0.1–1.0	0.02–6.1	0.218	0.812	0.043	0.01
	DMTS	0.4	0.02–6.1	0.140	0.000	0.096	0.01
	H_2S	0.1–1.0	0.02–6.1	1.894	0.899	0.397	0.14
(d)	MT	0.1–0.56	0.02–6.1	0.325	0.623	0.057	0.01
	DMS	0.1–0.56	0.02–6.1	1.965	1.257	0.036	0.05
	DMDS	0.1–0.56	0.02–6.1	1.137	0.717	0.000	0.05
	DMTS	0.1–0.3	0.02–6.1	1.383	0.548	0.017	0.03
	H_2S	0.1–0.56	0.02–6.1	0.631	0.651	0.054	0.07
(e)	MT	0.1–0.73	0.02–6.1	0.507	0.498	0.101	0.02
	DMS	0.1–0.73	0.02–6.1	0.517	0.375	0.173	0.07
	DMDS	0.1–0.73	0.02–6.1	1.186	0.510	0.193	0.06
	DMTS	0.34	0.02–6.1	1.207	0.000	0.068	0.08
	H_2S	0.1–0.73	0.02–6.1	2.067	0.833	0.045	0.08
(f)	MT	0.1–1.1	0.02–6.1	1.179	1.827	0.021	0.12
	DMS	0.1–1.1	0.02–6.1	0.772	1.103	0.017	0.37
	DMDS	0.1–1.1	0.02–6.1	1.137	0.601	0.010	0.08
	DMTS	0.57	0.02–6.1	1.142	0.000	0.155	0.21
	H_2S	0.1–1.1	0.02–6.1	2.181	1.345	0.096	0.66
(g)	MT	0.05–0.21	0.02–6.1	1.094	0.671	0.105	0.02
	DMS	0.05–0.21	0.02–6.1	2.171	0.824	0.127	0.05
	DMDS	0.05–0.21	0.02–6.1	1.947	0.560	0.116	0.04
	DMTS	0.05–0.21	0.02–6.1	1.314	0.285	0.094	0.08
	H_2S	0.05–0.21	0.02–6.1	0.744	0.724	0.242	0.09
(h)	MT	0.1–-0.66	0.02–6.1	0.751	0.571	0.138	0.12
	DMS	0.1–0.66	0.02–6.1	2.826	1.199	0.116	0.09
	DMDS	0.1–0.66	0.02–6.1	1.438	0.424	0.034	0.05
	DMTS	0.1–0.3	0.02–6.1	1.071	0.137	0.112	0.07
	H_2S	0.1–0.66	0.02–6.1	2.991	1.161	0.000	0.15
(i)	MT	0.06–0.3	0.02–6.1	32.9	2.86	0.000	0.08
	DMS	0.06–0.3	0.02–6.1	0.208	0.514	0.044	0.13
	DMDS	0.06–0.3	0.02–6.1	0.575	0.390	0.340	0.02
	DMTS	0.14	0.02–6.1	0.262	0.000	0.148	0.20
	H_2S	0.06–0.3	0.02–6.1	1.183	0.505	0.145	0.05

various packing materials, under liquid velocity of 0.02 m h^{-1}. The normalized K_Ga_e to pressure drop is especially interesting in practice since pressure drop is decisive for the running cost of the biofiltration and thus should be limited. Thus a low mass transfer coefficient for a material can be compensated by increasing the filter size if this material gives a lower pressure drop. The results showed in general that ceramic saddles and cellulose packed filters gave the highest pressure drop-normalized K_Ga_e for selected sulfur compounds (especially for MT) under the air velocity range applied, while DMT-HS and lava rock packed filters gave second

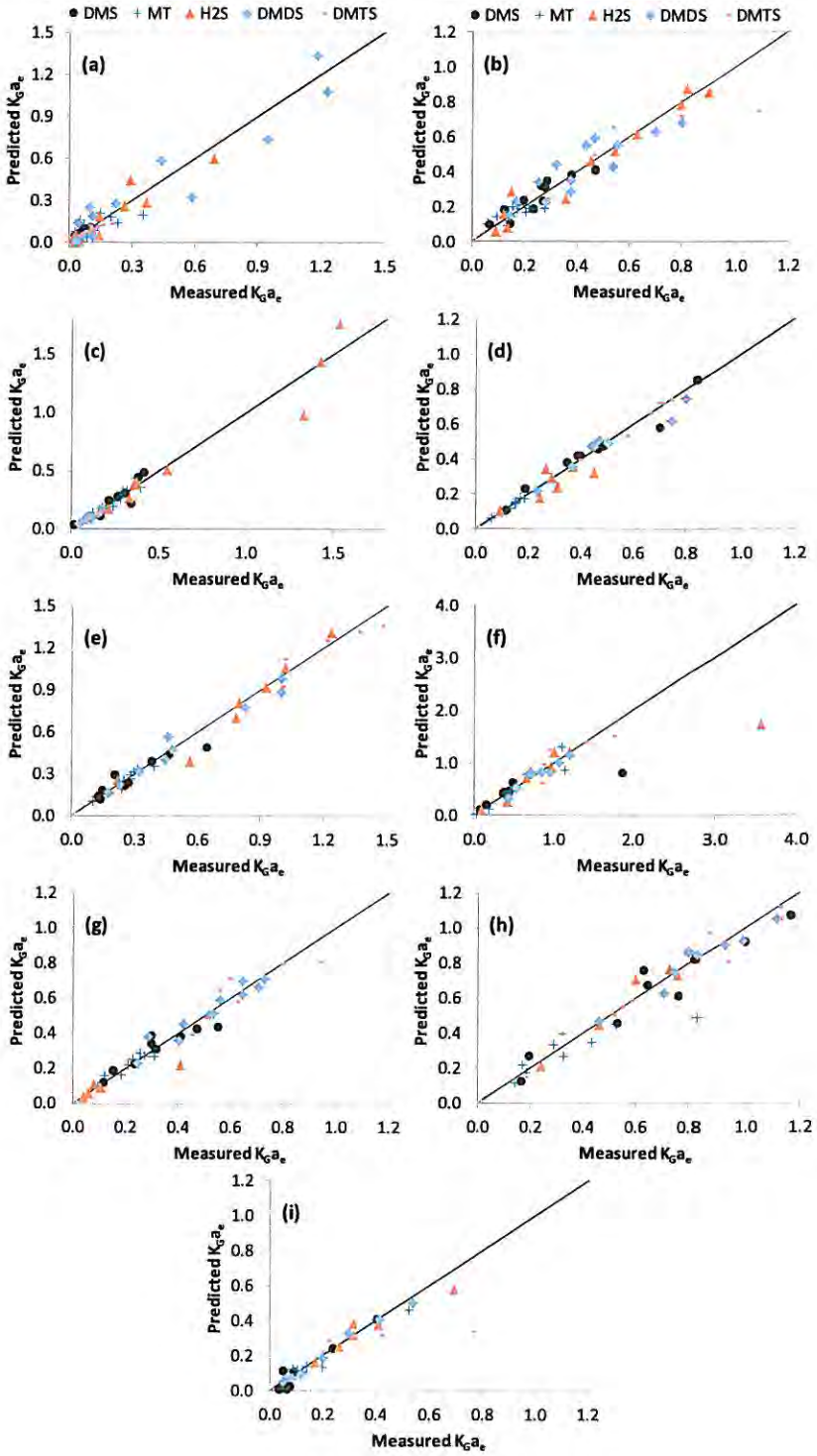

Fig. 8. Comparison of measured mass transfer coefficient (K_Ga_e) to predicted mass transfer coefficient (K_Ga_e) from simple power function model to air velocity and liquid velocity. The dot lines showed the uncertainty of ±20%. Packing materials: Cellulose (a), Leca® (b), DMT-HS (c), woodchips (d), bioflow 9 (e), ceramic saddles (f), molergranulat (g), lava rock (h) and cylindrial ring (i).

highest results, among the other packing filters. The high normalized K_Ga_e obtained for ceramic saddles may be due to the relatively high K_Ga_e especially at high air velocity combined with a moderate pressure drop. The high normalized K_Ga_e obtained for cellulose, on the other hand, is mainly due to the much lower pressure drop through the column compared to any other material. The K_Ga_e of H_2S obtained from BIO also gave comparable results to that obtained from DMT-HS, whereas the K_Ga_e values of DMS obtained from woodchips and BIO were slightly lower than those obtained from DMT-HS, ceramic saddles and cellulose. CYL, MOL and Leca® gave the poorest results, which were mainly due to the high pressure drop through the bed under similar air velocity condition.

Fig. 6 showed the dependence of measured K_Ga_e of volatile sulfur compounds on liquid velocity. In overall, the K_Ga_e increased at increasing liquid velocity. The increase might be partly due to the increased effective interfacial area as the liquid velocity increased [29,42]. While the increase of K_Ga_e was clear in most cases by increased liquid flow, there were a few cases where the influence of liquid velocity on K_Ga_e was less clear especially in the woodchips packed filter (Fig. 6d). Traditionally, the mass transfer is considered to be limited by the liquid-side resistance for relatively sparingly water-soluble compounds [29]. Thus, the increase of liquid velocity typically leads to an increase of the liquid-film mass transfer coefficient and therefore the overall mass transfer coefficient K_Ga_e

Table 3
Fitting parameters (α and β) for simple power function model of mass transfer coefficient (K_Ga_e) related to empty bed residence time (EBRT) of air (at Liquid velocity = 0.02 m h^{-1}).

	H_2S			MT			DMS			DMDS		
	α	β	R^2	α	β	R^2	α	β	R^2	α	β	R^2
Leca®	1.361	−1.334	0.847	0.239	−0.29	0.802	0.379	−0.573	0.999	0.795	−0.715	0.948
Cellulose	0.059	−0.266	0.693	0.121	−0.212	0.462	0.052	−0.214	0.713	0.223	−1.434	0.78
Lava rock	3.184	−1.181	0.974	0.487	−0.585	0.833	1.699	−1.103	0.922	1.275	−0.424	0.946
Woodchips	0.596	−0.7	0.875	0.262	−0.623	0.973	1.731	−1.244	0.97	1.243	−0.744	0.969
Bioflow 9	1.723	−0.79	0.915	0.359	−0.549	0.942	0.287	−0.375	0.774	0.581	−0.518	0.993
Cylindrial Ring	0.686	−0.495	0.987	10.35	−1.99	0.94	0.176	−0.508	0.968	0.171	−0.426	0.724
Saddles	1.941	−1.474	0.918	1.172	−1.811	0.966	0.972	−1.304	0.895	1.056	−0.496	0.931
DMT-HS	0.415	−0.91	0.94	0.32	−1.526	0.966	0.526	−1.635	0.913	0.191	−0.835	0.97
Molergranulat	0.292	−0.68	0.963	0.62	−0.6	0.96	1.444	−0.828	0.999	1.113	-0.495	0.954

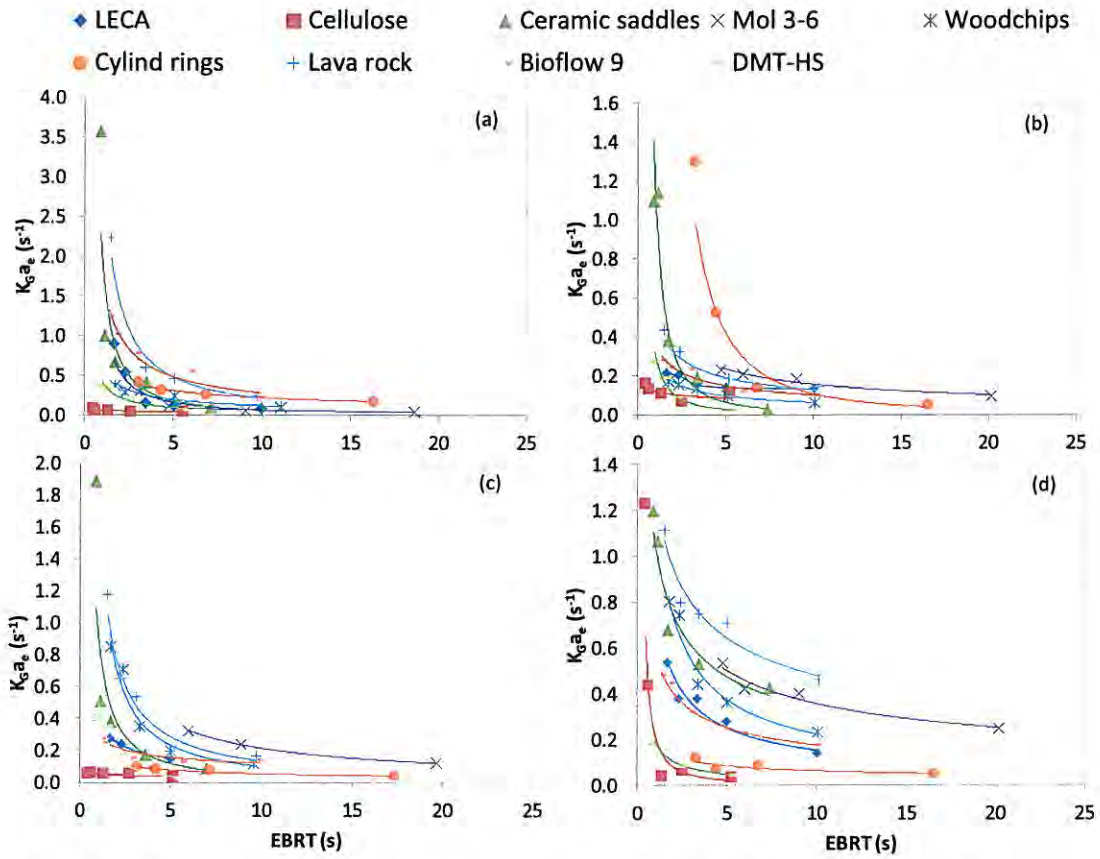

Fig. 9. Mass transfer coefficient (K_Ga_e) as a function of empty bed residence time (EBRT) of air for H_2S (a), MT (b), DMS (c) and DMDS (d) at liquid velocity = 0.02 m h^{-1}.

[21,29,43]. However, the dependence of overall mass transfer on liquid velocity may be compound-specific [28]. In overall, the results in the present study indicate that K_Ga_e is affected by both the air and liquid velocity and further that the impact of these vary across different packing materials. The study in [44] showed a similar trend when they measured the toluene absorption using a viscous absorbent.

3.4. Comparison to simple power function models

The effort of finding correlation models for predicting the observed results were not based on previously established correlation types [29,45,46], since the predicted K_Ga_e by these correlations were strongly influenced by liquid velocity but not air velocity [24,27,44]. Instead, empirical correlations of K_Ga_e as a function of air velocity, liquid velocity, specific surface area, external porosity, water holding capacity and Henry's law constant were investigated. This was done by minimizing root mean square error of a fit to the data of the type $K_Ga_e = eE^x + fF^y + gG^z$, where small letters denote empirical coefficients and capital letters denote investigated parameters. However, the established empirical model including these parameters only gave poor predictions to the measured values for both random packing and structured packing (Fig. 7). In general, the K_Ga_e is considered to be dependent on fluid dynamics, filter characteristics and chemical properties. However, due to the different ways of determination of filter characteristics for different packing materials, those obtained filter characteristics (e.g., specific surface area) might not be as directly comparable as desired when they were determined. Instead, the K_Ga_e was correlated only to air velocity and liquid velocity, in concordance with previous studies [22,27]:

$$K_Ga_e = k_1 V_G^{k_2} V_L^{k_3} \tag{14}$$

where k_1, k_2, k_3 are fitting parameters. The results for the simple power correlations are shown in Table 2. Fig. 8 showed the comparison between measured and predicted K_Ga_e with uncertainty lines of ±20%, including all the obtained data in this study. Good performance was generally observed and uncertainty was within ±20% for most cases with a few exceptions, especially for cellulose, most probably due to the low mass transfer coefficients obtained and unclear trends when air or liquid velocity was increased. It is also possible that the method, which was established based on random packing, is less suitable for structured packing materials, which may have caused larger errors when the mass transfer coefficients were determined. Still, the power correlation to air and liquid velocities shown above could be generally considered as the appropriate expressions for the relationship between the fluid dynamics and the mass transfer in biotrickling filters. The simple mathematical fitting of the experimental data, instead of using empirical correlations, was also recommended in the recent studies by Dorado et al. and San-Valero et al. [24,27].

Considering the insufficient fitting performance above for cellulose packing, instead an even simpler correlation between K_Ga_e and EBRT (under liquid velocity = 0.02 m h^{-1}) was applied for all the packing materials, in concordance with the previous study by Dorado et al. [24]:

$$K_Ga_e = \alpha \times EBRT^{\beta} \tag{15}$$

where α and β are fitting parameters. The results are shown in Table 3 and Fig. 9. An indeed better performance was obtained for cellulose packing, and thus this simple power correlation is also considered to be an appropriate expression, as recommended in [24]. However, it should be noted that such a simple power correlation is fully empirical and is dependent on packing materials and conditions.

4. Conclusions

Experimental determination of overall mass transfer coefficients of volatile sulfur compounds is presented for selected packing materials potentially used for biological air filters. The data is obtained by simulating breakthrough curves obtained by PTR-MS measurements at variable air and liquid velocities. The obtained mass transfer coefficients of sulfur compounds increased clearly with increased air velocity. At the same time, the mass transfer coefficients of sulfur compounds generally also increased with increasing liquid velocity. As a part of this work, key parameters describing the packing materials were determined. However, finding correlations between the mass transfer coefficients and these key parameters were failed for tested materials. The data was instead fitted to simple mathematical expressions only regarding air velocity and liquid velocity or empty bed residence time under a specific liquid velocity. The results showed that these empirical relationships could generally reproduce the experimental determined mass transfer coefficients in this study. Based on the ratio between mass transfer coefficient and pressure drop, two materials (ceramic saddles and cellulose) stood out with relatively high mass transfer coefficient-to-pressure drop ratios, mostly due to the low pressure drops observed for these two packing materials.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.cej.2014.08.070.

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