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### Determination of mass transfer coefficients for packing materials used in biofilters and biotrickling filters for air pollution control. 1. Experimental results

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#### Abstract

Gas film mass transfer coefficients  $(k_G a_t, k_G a_w)$  and liquid film mass transfer coefficients  $(k_L a_w)$  for packing materials used in biofilters and biotrickling filters for air pollution control were determined experimentally. Lava rock, polyurethane foam cube (PUF), Pall ring, porous ceramic beads, porous ceramic Raschig rings and compost-woodchips mixtures were investigated. The experiments were performed at gas velocities ranging from 100 to  $8000 \text{ m h}^{-1}$  and liquid velocities of  $0.1-12 \text{ m h}^{-1}$ , i.e., a wide range that covers most biofilters and biotrickling filters.  $k_G a_t$  in biofilter packings ranged from about 500 to  $2500 \,\mathrm{h}^{-1}$ , while  $k_G a_w$  and  $k_L a_w$  in biotrickling filters ranged from 100 to  $8000 \,\mathrm{h}^{-1}$ , and 1 to  $300 h^{-1}$ , respectively, depending on the packings and the conditions. This is markedly lower than mass transfer coefficients usually observed for conventional wet scrubbing. The gas film mass transfer coefficient ( $k_G a_t$ ) of 50:50% vol compost-woodchips mixture, a common biofilter packing, was greater than this of a 20% vol compost and 80% woodchips mixture, though the mass transfer was not increased by increasing further the volume fraction of compost. All compost mixtures exhibited a greater gas film mass transfer coefficient than lava rock or other synthetic materials. The mass transfer coefficients of compost mixtures was also influenced by packing method and it was directly proportional to the surface area of the bulking agents added. The gas film mass transfer coefficient  $(k_G a_w)$  of five biotrickling filter packing materials increased linearly with gas velocity. The effect of liquid on the gas film mass transfer coefficient was not significant. Of all the biotrickling filter packings, the porous ceramic beads had the highest gas and liquid film mass transfer coefficients followed by lava rock, porous ceramic rings, 1 in Pall ring and PUF cubes. The liquid film mass transfer coefficient ( $k_L a_w$ ) was directly proportional to liquid velocity and the effect of gas velocity was negligible. Several correlations allowing prediction of mass transfer coefficients are presented in Part 2 of this paper. © 2007 Elsevier Ltd. All rights reserved.

Keywords: Mass transfer; Absorption; Packed-column; Biotrickling filter; Biofilter; Environmental engineering

#### 1. Introduction

Biological treatment is increasingly used to control air pollutants as it is cost-effective, especially for the treatment of low concentration of pollutant in large air streams. The most frequently used biotechnologies for air pollution control are biofiltration and biotrickling filtration (Devinny et al., 1999; Kennes and Veiga, 2001). Biofiltration refers to the treatment of gaseous pollutants or vapors in a packed bed of damp material (usually compost mixed with some bulking agent) on which pollutant-degrading microorganisms thrive. In biotrickling filters, a free aqueous phase is trickled over the packing (usually an inert material with a large surface area) to provide optimum conditions to a biofilm of pollutant-degrading bacteria (Cox and Deshusses, 1998). Thus, the pollutant undergoing treatment will be transferred directly to the biofilm in a biofilter, while it may be transferred first to the trickling liquid in a biotrickling filter.

External mass transfer, diffusion and biodegradation kinetics are the main factors affecting the performance of biofilters and biotrickling filters. A better understanding of these limitations is important in order to enhance the performance and design of these bioreactors (Kim and Deshusses, 2005).

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These have been researched by several investigators. For example, Barton et al. (1999) discussed means to increase treatment in kinetically or mass transfer limited bioreactors for air pollution control and found out that the removal was substrate limited deep in the biofilm, but it was kinetically limited near the aqueous/biomass phase. Kirchner et al. (1992) concluded that a trickle-bed bioreactor was limited by oxygen diffusion in the biofilm, biological degradation reaction and external mass transfer. Picioreanu et al. (2000) developed a 2D model to explain the transport of substrate into biofilms and found that the substrate flux to the biofilm was greatly influenced by external mass transfer rate. Kim and Deshusses (2003) observed that the performance of biotrickling filters treating low concentrations of H<sub>2</sub>S at very short gas contact times depended on the air velocity, indicating that external mass transfer was probably limiting. Kan and Deshusses (2006) developed foamed emulsion biofilter having high surface area for mass transfer  $(2400 \text{ m}^2 \text{ m}^{-3})$  and its performance was 2–1000 times higher than other biotreatment.

In spite of the importance of mass transfer aspects in biofilters and biotrickling filters, no systematic study was ever conducted to determine mass transfer coefficients in these reactors. When mass transfer coefficients were needed e.g., for modeling purposes, investigators (Mpanias and Baltzis, 1998; Baltzis et al., 2001; Kim and Deshusses, 2003) have relied on the large body of information obtained for traditional packing materials used for gas absorption, stripping or distillation (Onda et al., 1968; Piche et al., 2001; Sherwood and Holloway, 1940, Shulman et al., 1955a,b). However, these mass transfer correlations cannot necessarily be applied to biofilters and biotrickling filters. This is because they were developed for different packing materials and may not apply to the very different operating conditions encountered in biofilters and biotrickling filters. Typical wet scrubber operating conditions are superficial gas velocities ranging from 1000 to 10,000 m h<sup>-1</sup> and trickling liquid velocities ranging from 10 to 150 m h<sup>-1</sup>. On the other hand, gas superficial velocities in biofilters and biotrickling filters usually range from 60 to about 1000 m h<sup>-1</sup>, and occasionally up to  $6000 \text{ m h}^{-1}$  (Gabriel and Deshusses, 2003), while the liquid superficial velocity in biotrickling filters rarely exceeds  $10 \text{ m h}^{-1}$ .

Thus, the objectives of this study were to determine gas film mass transfer coefficients for various packings used on biofilters, and to determine both gas film and liquid film mass transfer coefficients for selected biotrickling filter packings. A wide range of gas and liquid trickling velocities was used to increase the applicability of the data.

#### 2. Materials and methods

# 2.1. Experimental setup for gas film mass transfer coefficient $(k_G a_w, k_G a_t)$

For the determination of  $k_G a_w$  (index *w* here denotes the wetted area), absorption of CO<sub>2</sub> in trickling caustic water was used because CO<sub>2</sub> reacts immediately in caustic water, hence the main resistance to mass transfer lies in the gas film (Linek et al., 1984). A schematic of the experimental setup is shown in Fig. 1. A bench-scale absorption column (PVC, 15 cm ID ×160 cm high, Harrington Plastics, Chino, CA) was filled with selected packing materials (see list in Table 1).The packing height was either 10 cm (for lava rocks, expanded clay, some compost beds) or 40 cm (Pall rings, polyurethane foam (PUF) and some compost–woodchips mixtures) depending on the pressure drop. A metered air stream resulting in air velocities ranging from 100 to 8000 m h<sup>-1</sup> was supplied by a centrifugal blower (Broan, Hartford, WI for high velocities, Roton,



Fig. 1. Schematic of the experimental setup for  $k_G a_w$ .

Table 1 Characteristics of the various packing materials used in this study

	Lava rock	Pall ring	Polyurethane Foam cube	Porous ceramic beads (Porcelite <sup>TM</sup> )	Porous ceramic Raschig rings (CBP <sup>TM</sup> )	Compost-woodchips mixtures
Operating mode	BF and BTF	BTF	BTF	BTF	BTF	BF
Main composition	Lava rock	Polypropylene	Polyurethane	Porous Ceramic	Porous Ceramic	Compost & woodchips
Shape	Irregular	Cylinder	Cube	Beads	Raschig rings	Irregular
Dimension (mm)	1-40	25	40	4	13	See methods
Surface area $(m^2 m^{-3})$	Unknown	210	600	2500	360	Unknown
Porosity (%)	49	90	97	38	54	40-60%
Supplier	Classic Stone Corp.	Jaeger	EDT, Germany	Aisin Takaoka, Japan	Aisin Takaoka, Japan	Whittier, Kellogg

Kent, OH for low velocities). A large diameter aluminum flexible duct (15 cm ID, Grainger, Oakland, CA) was used for the gas outlet to avoid possible pressure drop downstream of the bed. The influent air was spiked with a metered stream of CO<sub>2</sub> using a mass flow controller (Porter Instrument Company, Inc., Hatfield, PA) to reach a concentration not exceeding  $5000 \, \text{ppm}_{v}$ . The air was fed to the bottom of the absorption column while a 1 M NaOH solution was trickled from the top and recycled using a peristaltic pump (Masterflex, Cole-Parmer, Vernon Hill, IL). For the highest trickling rates, a centrifugal pump was used (TEEL, Dayton Electronics, Niles, IL). The caustic solution was uniformly sprayed on the top of the bed using a BETE WL 1/4 nozzle (BETE, Greenfield, MA) for low liquid velocities and using a BETE MP 125M nozzle for high liquid velocities. The pH of the caustic solution was monitored using a pH meter (Accumet, Fisher Scientific, Pittsburgh, PA) during the experiments. The solution was replaced when the pH dropped below 13, as this was found (results not shown) to be the threshold pH below which the absorption of CO<sub>2</sub> was dependent on NaOH concentration.

At the beginning of each experiment, a known amount of packing material was placed in the column and 7 L of 1 M NaOH was prepared. For the effect of gas velocity on mass transfer, the gas velocity was increased from 100 to 8000 m h<sup>-1</sup> at selected liquid velocities (0.1, 6.3, and 10 m h<sup>-1</sup>). For the effect of liquid velocity on mass transfer, the tricking velocity was varied between 0.1 and 12 m h<sup>-1</sup> at four gas velocities (100, 720, 2500 or 4700 m h<sup>-1</sup>). Pressure drop was measured using a manometer at each experimental condition. The temperature was maintained at 20 °C  $\pm$  2 °C. During each experiment, the steady state concentrations of CO<sub>2</sub> in the inlet and the outlet air streams were measured and gas film mass transfer coefficient ( $k_G a_w$ ) was calculated as follows:

$$k_G a_w = \frac{U_G}{V_b} \ln \left( \frac{C_{G,\text{in}}}{C_{G,\text{out}}} \right). \tag{1}$$

Compost and woodchips mixtures as well as lava rocks were used to determine the gas film mass transfer coefficients ( $k_G a_t$ ) for commonly used biofilter packings. Since there is little or no trickling liquid during biofiltration, the packing materials had to be conditioned with caustic as follows. Compost (99.8 wt% of total compost passing a 9.8 mm sieve, 68% passing a 2 mm sieve, 54% passing a 1 mm sieve, mixture of organic blend of shavings, sawdust and bark) was mixed with woodchips (average size 25 mm, forest bark) previously soaked in 3 M NaOH for 10 min and allowed to drain for 30 min. Then the initial moisture content of the packing was adjusted to about 50% (wet basis). The same pre-soaking method applied to lava rock. During the experiments, careful management of packing moisture was required and experiments were conducted with either humid air or intermitted spraying of the biofilter bed with a caustic solution. This is discussed in Section 3. The compost volumetric fraction range was 20%, 50% or 100% by volume. The compost or compost woodchips mixtures were used for less than a week so that long-term changes in physical properties were avoided (Morgan-Sagastume et al., 2003). For mass transfer coefficient determinations, the gas velocity was varied from 100 to  $8000 \text{ m h}^{-1}$ . The mass transfer coefficient was calculated following Eq. (2), similar to Eq. (1), except that the area considered is the total surface area  $(a_t)$  of the packing material, instead of the wetted surface area:

$$k_G a_t = \frac{U_G}{V_b} \prod_{b} \left( \frac{C_{G,\text{in}}}{C_{G,\text{out}}} \right).$$
(2)

### 2.2. Experimental setup for liquid film mass transfer coefficient $(k_L a_w)$

For the determination of  $k_L a_w$ , absorption of oxygen, which is sparingly soluble in water, was used (Linek et al., 1984) so that the main resistance to mass transfer was in the liquid phase. The setup (Fig. 2) used the same main equipment used for the determination of  $k_G a_w$  described above. The experimental setup included on-line monitoring of dissolved oxygen in the influent and effluent water. Dissolved oxygen in the liquid was stripped prior to be introduced in the trickling filter by continuously sparging nitrogen during the experiment. Steady state inlet and outlet dissolved oxygen concentrations were used to calculate  $k_L a_w$  using the following equation:

$$k_L a_w = \frac{U_L}{V_b} \ln\left(\frac{C_G/H - C_{L,\text{in}}}{C_G/H - C_{L,\text{out}}}\right).$$
(3)

#### 2.3. Analytical methods

The concentrations of  $CO_2$  in the influent and effluent air streams were determined with a non-dispersive infrared  $CO_2$  gas sensor (Vernier Software & Technology, Beaverton, OR).



Fig. 2. Schematic of the experimental setup for  $k_L a_w$ .



Fig. 3. Effect of moisture supply method on  $k_G a_I$ . Compost mixture with woodchips (20:80, v/v) was used as packed bed. Error bars indicate the experimental uncertainties.

The reproducibility of the meter after careful calibration was about 1.5%. The CO<sub>2</sub> gas sensors were placed at the inlet and outlet of gas flow. The air velocity ( $\pm 3\%$ ) was measured by a hot wire thermo-anemometer (Extech instruments, Melrose, MA), except for the lowest air flows which were measured using a rotameter (Dwyer Instruments, Inc., Michigan city, IN). The concentration of dissolved oxygen ( $\pm 0.05 \text{ mg L}^{-1}$ ) was determined with a Clark-type polarographic dissolved oxygen probe (Vernier Software & Technology, Beaverton, OR). The moisture content of the packing was measured by determining the weight of packing samples before and after drying at 80 °C. An analysis of the uncertainties of the mass transfer determinations revealed that the greatest contributing factor was the gas flow rate measurement followed by gas concentration measurements. Other uncertainties included bed height fluctuations and inherent variations in gas mixing, liquid distribution, packing wetting, etc. When all these factors were included, the accuracy of the mass transfer coefficients was determined to be about  $\pm 8\%$ .

#### 3. Results and discussion

### 3.1. Determination of gas film mass transfer coefficient $(k_G a_t)$ for biofilter packings

#### 3.1.1. Method validation

Compost, compost and woodchips mixtures, and lava rock were used for the determination of  $k_G a_t$  in common



Fig. 4. Pressure drop over the 20:80% vol compost-woodchips bed for different moisture management methods.



Fig. 5. Effect of packing nature (compost and woodchips mixtures) and density on gas film mass transfer coefficient ( $k_G a_t$ ). Loosely packed bed had a density of 220 kgm<sup>-3</sup>, while the densely packed density was 330 kgm<sup>-3</sup>. Error bars indicate the experimental uncertainties.

biofilter packings. Two methods for maintaining proper moisture in the biofilter beds were evaluated. The first was to use humid air close to saturation, while the second involved intermittent spraying of various amounts of 3 M NaOH through a nozzle at the top of the column during mass transfer coefficient determinations. The objective was to maintain the moisture content of the compost–woodchips mixture to its initial value of 50% and provide stable operation without water logging on the gas–solid interfaces. Results for the gas film mass transfer coefficient ( $k_G a_t$ ) and pressure drop over the bed for selected conditions are shown in Figs. 3 and 4, respectively.  $k_G a_t$  values ranged from 500 to 4000 h<sup>-1</sup> and increased with air velocity. As will be discussed later, these gas film mass transfer coefficients were much greater than those obtained for synthetic material such as Pall ring or PUF cubes. The mass transfer coefficients and pressure drops obtained with humid air and the lowest intermittent spraying (67 mL per data point) were very similar (Figs. 3 and 4, respectively). Both operating modes provided stable operation and no leachate was observed. The moisture content of the packing remained between 43% and 57% (wet basis). This was not the case when intermittently spraying 200 mL per data point, i.e., 11% of (empty) bed volume. Excess liquid leached out of the system, and although higher mass transfer coefficients were observed (Fig. 3), operation resulted in markedly higher pressure drops (Fig. 4) due to water logging conditions. Therefore, all further experiments were conducted with intermittent spraying of 70 mL of caustic solution per data point.



Fig. 6. Effect of packing nature and density on pressure drop.



Fig. 7. Effect of compost and woodchips volume ratio on the mass transfer coefficient. Error bars indicate the experimental uncertainties.

#### 3.1.2. Effect of packing on gas film mass transfer coefficient

Both the nature and the density of the biofilter packing are expected to affect the mass transfer coefficient. Mass transfer coefficients for different mixtures of woodchips and compost, loosely packed or compressed to simulate a denser packing are compared in Fig. 5. The loosely packed bed had lower mass transfer coefficients than the densely packed bed. This was expected as the densely packed bed had 50% more compost and/or woodchips per bed volume, resulting in a greater interfacial area for mass transfer. The denser packing resulted in significantly higher pressure drops as shown in Fig. 6, while compost–woodchips composite beds had markedly lower pressure drops. Fig. 6 shows pressure drop over wider range of gas

velocities than many previous studies, thereby allowing a trend analysis with respect to air velocity. While it is often assumed that pressure drop in packed beds increases with the square of the velocity as described by Ergun's equation, Fig. 6 shows no such quadratic trend. The reason for this is probably related to the heterogeneous nature of compost–woodchips beds. The same phenomenon had been observed by other researchers (Van Langenhove et al., 1986; Delhomenie et al., 2003) and suggests that Ergun's equations should only be carefully applied to biofilter beds.

In order to compare mass transfer coefficients for an equal mass of packing, the  $k_G a_t$  of the denser packing reported in Fig. 5 were divided by 1.5, i.e., the packing mass ratio (data not



Fig. 8. Effect of compost and woodchips volume ratio on pressure drop.



Fig. 9. Mass transfer coefficient (k<sub>G</sub>a<sub>t</sub>) of a lava rock bed compared to a 20% vol compost-woodchips mixture. Error bars indicate the experimental uncertainties.

shown). The recalculated mass transfer coefficients of dense or loosely packed beds were not significantly different for a given compost–woodchips ratio. The compost bed had a higher mass transfer coefficient than the compost–woodchips bed. The results demonstrate that  $k_G a_t$  was mainly dependent on the surface area of packing (compost has a higher interfacial area than woodchips), and that either compaction had no effect on the interfacial area, or that the reduction of interfacial area was compensated by a greater gas film mass transfer coefficient  $k_G$ caused by the higher interstitial gas velocity in the denser bed. The effect of various woodchips volume fractions was further investigated. Compost itself is a good packing material, but it compacts over time, resulting in excessive pressure drop (Yang and Allen, 1994; Devinny et al., 1999), therefore woodchips are often used as a bulking agent. As shown in Fig. 7, the mass transfer coefficient in beds packed with only 20% vol. compost increased linearly with increasing gas velocity. The results for the 50% and 100% compost mixture also increased with the gas velocity, but significant scattering of the data was observed, both for the mass transfer coefficients and for the



Fig. 10. Comparison of pressure drop of lava rock and compost-woodchips beds.



Fig. 11. Effect of gas velocity (a) and of liquid velocity (b) on mass transfer using 1 in Pall Ring compared to an empty column control. (a) Liquid linear velocity was  $6.3 \text{ m} \text{ h}^{-1}$ . (b) Gas linear velocity was  $1000 \text{ m} \text{ h}^{-1}$ . Error bars indicate the experimental uncertainties.

pressure drop data (Fig. 8). These fluctuations might have been caused by air channeling as described earlier (Yang and Allen, 1994). Channeling is thought to reduce the resistance to air flow by relocating compost in the bed, thereby reducing the surface area for mass transfer. On the contrary, experiments with the 20% vol compost mixture were very stable over the entire range of gas velocities.

Porous lava rock is frequently used as a packing material for biofilters, because it is more durable than organic materials such as compost, woodchips, peat, or fiber and it allows flushing with water, when necessary. It also has a high capacity for microorganisms and is less expensive than synthetic packing materials (Cho et al., 2000). In Fig. 9, the mass transfer coefficients obtained for lava rock beds are compared to those of a 20% vol compost mixture. The gas film mass transfer coefficients in lava rock beds were about half those observed in the compost–woodchips beds. This was attributed to the lower interfacial area of the lava rocks. As before, the mass transfer coefficient increased linearly with the gas velocity. Pressure drop was markedly lower in the lava rock bed (Fig. 10), because of the greater porosity of the lave rock beds.

Overall, the results of the gas film mass transfer coefficient determinations in biofilter beds show that the packing area plays an important role in the actual value of  $k_G a_t$ . However, the actual interfacial area of compost beds is virtually impossible to determine. The results obtained with different wood-chips volume fractions suggest that the compost area could be determined by a series of mass transfer coefficient determinations with selected bulking agents with known interfacial areas. Further, the increasing trend of mass transfer coefficients with increasing gas velocity using compost mixtures was similar to the trends observed for plastic packings (Onda et al., 1968;



Fig. 12. Effect of gas velocity on gas film mass transfer coefficient ( $k_G a_w$ ) in biotrickling filter. Packing materials: (A) lava rock, (B) PU foam, (C) 1 in Pall ring, (D) porous ceramic beads, (E) porous ceramic rings. Note the different x and y scales between the graphs. Error bars indicate the experimental uncertainties.

Piche et al., 2001), although the mass transfer coefficients observed here are roughly one to two orders of magnitude lower than those observed in conventional packings. The similarity indicates that even though the materials and properties of the packings were completely different, some of the correlations developed for mass transfer for conventional packings could be adapted to biofilter packings. This is presented in Part 2 of this paper (Kim and Deshusses, 2007).



Fig. 13. Effect of liquid velocity on gas film mass transfer coefficient ( $k_G a_w$ ) in biotrickling filter. Packing materials: (A) lava rock, (B) PU foam, (C) 1 in Pall ring, (D) porous ceramic beads, (E) porous ceramic rings. Note the different y scales between the graphs. Error bars indicate the experimental uncertainties.

## 3.2. Determination of gas film mass transfer coefficient $(k_G a_w)$ for biotrickling filter packings

#### 3.2.1. Control experiments

Because of the relatively small size of the equipment and possible influence of the wall surface in mass transfer, control experiments were conducted by spraying deionized water and caustic water in an empty absorption column while the air:carbon dioxide mixture was passed counter-currently. Little CO<sub>2</sub> absorption was observed when deionized water was used while some absorption occurred when 1 M NaOH was sprayed (results not shown). However, absorption in controls



Fig. 14. Effect of liquid velocity on liquid film mass transfer coefficient ( $k_L a_w$ ) in biotrickling filter. Packing materials: (A) lava rock, (B) PU foam, (C) 1 in Pall ring, (D) porous ceramic beads, (E) porous ceramic rings. Note the different y scales between the graphs. Error bars indicate the experimental uncertainties.

was always at least 6 times lower than in experiments with packing. This is illustrated in Fig. 11 which shows the effect of gas and liquid velocity on  $k_G a_w$  in a column packed with 1 in (2.54 cm) Pall rings compared to the empty tower control. The mass transfer coefficient in the control experiments

did not greatly vary with either liquid or gas velocity. The largest effect was a modest increase with increasing the liquid velocity, which was likely due to the increase in wetting of the walls of the absorption column. The mass transfer coefficient decreased when increasing gas velocity over  $1000 \text{ m h}^{-1}$ .



Fig. 15. Pressure drop during the mass transfer experiments for the porous ceramic rings (left) and beads (right).

Overall, the mass transfer coefficients in the controls remained small compared to these in the 1 in Pall Rings bed. The effect of liquid and gas velocity on mass transfer coefficients for different packings is discussed next.

# 3.2.2. Effect of gas and liquid velocities on gas film mass transfer coefficient $(k_G a_w)$ for biotrickling filter packings

Five different packing materials were tested in a biotrickling filter mode and  $k_G a_w$  was determined. Recall that  $a_w$  stands for the specific wetted area. The effect of gas velocity is shown in Fig. 12. The upper range of the superficial gas velocity was determined by the pressure drop in the system and current upper limit of biotrickling filter operation. The mass transfer coefficients ranged from about  $100 h^{-1}$  up to about  $8000 h^{-1}$ , depending on the packing and the conditions. They were the greatest in porous ceramic beads, followed by lava rocks, ceramic rings, plastic rings and PUF. With the exception of the ceramic beads, the  $k_G a_w$  values were slightly lower than the  $k_G a_t$  values found in biofilters (see Section 3.1). Although operating conditions of biotrickling filters and wet scrubbers are markedly different, a comparison of the mass transfer coefficients is warranted. In most cases, biotrickling filters are operated at gas velocities below  $500 \text{ m h}^{-1}$ . At these conditions, the majority of the  $k_G a_w$  values in Fig. 12 is in the 500–2000 h<sup>-1</sup> range. This is significantly lower than is usually observed in conventional wet scrubbers for which  $k_G a_w$  commonly reach values of 15, 000–30,  $000 h^{-1}$  (Piche et al., 2001). This suggests that important gas side mass transfer effects may have been overlooked in biotrickling filters.

In general  $k_G a_w$  increased with increasing gas velocity, as expected from mass transfer theories and experiments (although under different conditions) by others (Onda et al., 1968; Piche et al., 2001), although in some cases (e.g., lava rock), mass transfer coefficients seemed to reach a plateau beyond a certain gas velocity. The reason for such a behavior was not firmly identified, but may be related to entrainment of trickling water at the higher gas velocities.

At each gas flow rate, three liquid velocities were investigated so that the effect of wetting could be observed. The gas film mass transfer coefficients were generally low at low liquid velocities, and increased with the liquid velocity. This is because wetting of the packing improved with greater trickling rates. As will be discussed later, wetting in lava rock was not a strong function of gas velocity, whereas it was in other packings. In most cases, only marginal improvements were observed when the trickling rate was increased beyond  $6.3 \,\mathrm{m \, h^{-1}}$ , indicating that wetting was near complete. However, there were some exceptions. The mass transfer coefficients in lava rocks decreased at the highest trickling rate, because the system approached flooding conditions. This was most notable at the highest air flow rates, i.e., at conditions that favor flooding (Trejo-Aguilar et al., 2005). Mass transfer coefficients in the PUF bed exhibited a complex behavior. The  $k_G a_w$  values increased more or less linearly with the gas velocity at the two lowest liquid trickling rates, and further increased with increasing the trickling rate. This is due to the fact that the PUF is a very open and porous packing (see Table 1), with very low static and dynamic holdups. Hence, greater liquid trickling meant a greater wetted area for mass transfer, unlike for the other packing where wetting was essentially complete at lower trickling rates. However, for the PUF at the highest trickling rate, the gas film mass transfer coefficient started to decrease when the gas velocity was increased beyond  $3000 \text{ m h}^{-1}$ . This is because at greater gas velocities, small droplets of trickling liquid were entrained into the counter-current air flow. This resulted in a lower dynamic hold-up and a lower wetted interfacial area for mass transfer.

The effect of liquid velocity on the gas film mass transfer coefficient is further detailed in Fig. 13. Generally gas film mass transfer coefficient increased with increasing liquid velocity at low liquid velocity  $(0.1-4 \text{ m h}^{-1})$  but the trend was changed to essentially constant or slightly increasing values above intermediate liquid velocities (around  $6.3 \text{ m h}^{-1}$ ). The most probable reason for this behavior is that wetting of the packings was incomplete at the low liquid velocities and it increased with



Fig. 16. Effect of gas velocity on liquid film mass transfer coefficient  $(k_L a_w)$  in biotrickling filters. Packing materials: (A) lava rock, (B) PU foam, (C) 1 in Pall ring, (D) porous ceramic beads, (E) porous ceramic rings. Note the different y scales between the graphs. Error bars indicate the experimental uncertainties.

greater trickling rates until the packing was fully wetted (Piche et al., 2001). This is particularly clear in Figs. 13B, D and E, though the liquid velocity at which  $k_G a_w$  became constant depended on the packing material, since each packing has different wetting properties (Linek et al., 1984). Thus wetted

interfacial area and liquid hold-up were the governing factors for the effect of liquid velocity. An excess amount of liquid hold-up could be detrimental to the gas film mass transfer coefficient as shown in Fig. 13B and C. However, overall, the effect of liquid velocity on  $k_G a_w$  remained limited and gas film resistance was much less affected by liquid velocity than by gas velocity.

# 3.3. Determination of liquid film mass transfer coefficient $(k_L a_w)$

The liquid film mass transfer coefficient  $k_L a_w$  was determined for the five packings.  $k_L a_w$  values ranged from about 1 to  $300 \text{ h}^{-1}$  depending on the packing and the conditions (Fig. 14). This is roughly equivalent to  $k_L a_w$  measured in conventional scrubbers (Piche et al., 2001).

The effect of liquid velocity on mass transfer coefficients is shown in Fig. 14. All packing materials showed a strong linear relationship between  $k_L a_w$  and liquid velocity except for the porous rings at the lowest gas velocity. This finding is consistent with the general trend predicted by liquid film mass transfer correlations by Onda et al. (1968). For comparison between packings, a liquid velocity of  $10 \text{ m h}^{-1}$  and a gas velocity of  $720 \,\mathrm{m}\,\mathrm{h}^{-1}$  were selected. The porous ceramic bead had the highest  $k_L a_w$  followed closely by lava rock and porous ceramic ring, PUF and Pall rings which had 6- to 10-fold lower  $k_L a_w$ . As mentioned,  $k_L a_w$  was essentially constant in the porous ceramics rings at the lowest air velocity (Fig. 14E). The reasons for this are unclear, but may be related to air and water flow path in and around the rings at low air velocity. However, when the pressure drop in the ceramic ring bed is plotted vs. liquid and gas velocities, little differences exist between the data at 100 and  $720 \text{ m h}^{-1}$  (Fig. 15, left). Further research with gas and liquid tracer injections would be necessary to identify the reasons for this behavior. Even so, the results suggest that poor liquid mass transfer may exist in porous rings packed bed operated at low air velocities. Fig. 15 further compares the pressure drop in the ceramic packings. Not unexpectedly, the beads had a significantly higher pressure drop than the rings. This was mostly due to differences in void volume and interfacial area, and resulted in marked differences in liquid film mass transfer coefficient, as shown in Fig. 14E.

The effect of gas velocity on the liquid film mass transfer coefficient was determined (Fig. 16). As found by others for traditional scrubbing packings (Piche et al., 2001),  $k_L a_w$  was not affected by increasing gas velocity, except for the lava rock packing for which a sudden drop of mass transfer occurred when the gas velocity exceeded 700 m h<sup>-1</sup>. The reasons for this behavior are unknown.

#### 4. Conclusions

Mass transfer coefficients for packing materials typically used in biofilter and biotrickling filters for air pollution control were studied. Significant differences in the mass transfer coefficients were obtained between the different packings, indicating that mass transfer characteristics is an important consideration in packing selection. Gas film mass transfer coefficients in biofilter increased linearly with gas velocity when compost was mixed with woodchips. Lava rock had relatively high mass transfer coefficients, although lower than compost–woodchips mixtures. The gas and liquid film mass transfer coefficients were determined for five biotrickling filter packing materials in a range of liquid and gas velocities. Gas film mass transfer coefficients ( $k_G a_w$ ) generally increased with increasing gas velocity, although some packing materials had low mass transfer at low liquid velocity, which supports the fact that gas film mass transfer was directly affected by wetting. For these, the effect of liquid velocity was significant at low liquid velocities, but became negligible when liquid velocity increased. The liquid film mass transfer coefficient ( $k_L a_w$ ) was solely dependent on the liquid velocity.

Overall, this study is the first attempt in systematically determining mass transfer coefficients for packings used in biofilters and biotrickling filters for air pollution control. The results should facilitate the selection of packings and the modeling of bioreactors used for air pollution control.

#### Notation

$a_t$	total specific surface area, $m^2 m^{-3}$
$a_w$	wetted specific surface area, $m^2 m^{-3}$
$C_{G,in}$	gas phase $CO_2$ concentration at inlet, g m <sup>-3</sup>
$C_{G,\text{out}}$	gas phase $CO_2$ concentration at outlet, g m <sup>-3</sup>
$C_{L,in}$	dissolved oxygen concentration at inlet, $g m^{-3}$
$C_{L,out}$	dissolved oxygen concentration at outlet, $g m^{-3}$
$H^{\prime}$	Henry's constant, dimensionless
$k_G a_t$	gas film mass transfer coefficient in biofilter, $h^{-1}$
$k_G a_w$	gas film mass transfer coefficient in biotrickling filter $h^{-1}$
$k_L a_w$	liquid film mass transfer coefficient in biotrickling filter, $h^{-1}$
$U_G$	superficial gas velocity, m h <sup>-1</sup>
$U_L$	superficial liquid velocity, m $h^{-1}$
$V_b$	bed volume, m <sup>3</sup>
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