Turbulent gas flux measurements near the air–water interface in an oscillating-grid tank

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Abstract. Measurements of interfacial flux of oxygen into water were conducted in a tank in which turbulence was produced through oscillating grids. The experimental profiles of the turbulent mass fluxes close to the air-water interface were compared with predictions using the turbulent diffusivity as a power series expansion of the distance to the interface. As usual, truncations of the power series were used: (1) with one term (usual power law) and (2) with two terms of the series. The different experimental profiles, when plotted in nondimensional form against the distance to the interface, showed the same shape close to the interface, until the first flux peak was reached. The exponents of both approximations for the diffusivity (truncations of the power series expansion) were quantified using the position of this first peak. Considering only one term of the power expansion, the value of 2.0 was obtained from the experimental data. Considering two terms of the power expansion, the exponent was found to be greater than 2.0, and the value of 3.0 was then used, which resulted in a better visual agreement between experimental and predicted nondimensional profiles for the region close to the interface.

Key Words: Turbulent fluxes, turbulent diffusivity, grids turbulence, power expansion for diffusivity

1. Introduction

The understanding of the gas transfer mechanism is important in geophysical, environmental and industrial processes. Turbulence is present in most of the natural and industrial processes, introducing difficulties in the precise quantification of mass fluxes. So, basic studies are still necessary, and approximations that allow for adequate quantifications are welcomed by engineers who need them to support decisions.
The total transport of a passive scalar across the air-water interface is usually written as

\[ j = -D \frac{dC}{dz} + \langle c'w' \rangle \]  

(1)

where the turbulent flux is expressed through the product of the fluctuations of concentration and velocity \((c',\text{ and } w',\text{ respectively})\), \(D\) is the molecular diffusivity, \(C\) is the mean concentration, and \(z\) denotes the direction normal to the interface (positive entering into the water). The complete quantification of the physical mechanisms that control the gas transfer process requires the understanding of the molecular (first term on the right hand side) and the turbulent transports (second term on the right hand side) near the air-water interface.

The aim of the present paper was to study the behavior of turbulent mass fluxes very close to the interface, in order to contribute to the understanding of the physical mechanisms that control the air-water gas transfer process in a bottom-shear-induced turbulent environment. In this sense, theoretical propositions that use the turbulent diffusivity \(D_t\) and the mean concentration as power series of the distance to the surface were considered. Equation 1 is then presented as:

\[ j = -(D + D_t) \frac{dC}{dz} \]  

(2)

The so called “small eddy model” is used here, in which the turbulent flux through the boundary layer is viewed as resulting from a cascade of growing eddies. Following the descriptions of Coantic (1986) and Münsterer (1996), the mean concentration can be written as:

\[ C = C_0 + \sum_{i=1}^{n} \frac{1}{i!} \left. \frac{\partial^i C}{\partial z^i} \right|_{z=0} z^i \]  

(3)

The first derivative at the position \(z = 0\) is given by:

\[ \left. \frac{\partial C}{\partial z} \right|_{z=0} = -\frac{j}{D} \]  

(4a)

From equations 1 and 4a, the first successive derivatives are given by:

\[ D \left. \frac{\partial^2 C}{\partial z^2} \right|_{z=0} = \left. \left( w' \frac{\partial c'}{\partial z} + c' \frac{\partial w'}{\partial z} \right) \right|_{z=0} = 0 \]  

(4b)

\[ D \left. \frac{\partial^3 C}{\partial z^3} \right|_{z=0} = \left. \left( w' \frac{\partial^2 c'}{\partial z^2} + 2 \frac{\partial w'}{\partial z} \frac{\partial c'}{\partial z} + c' \frac{\partial^2 w'}{\partial z^2} \right) \right|_{z=0} = a_3 \]  

(4c)
For rigid walls, continuity imposes \([\partial w'/\partial z]_{z=0}=0\), so that \(\alpha_3=\beta_4=0\), and probably \(\alpha_4 \neq 0\). In this case, the result for the turbulent diffusivity is \(D_T = \left(\frac{D\alpha_4}{12j}\right)z^4\) + higher order terms. For free surfaces, it is generally assumed that \(\alpha_3 \neq 0\), resulting in \(D_T = \left(\frac{D\alpha_3}{4j}\right)z^2\) + higher order terms. The choice of the most adequate exponent of \(z\) needs the use of experimental results. Considering comparisons between measured and predicted mean concentration profiles, Herlina and Jirka (2008) tested the small eddy and the renewal models and concluded that the turbulent mechanism can change between both models, depending on the agitation conditions (or the turbulence level) of the liquid side. In the present study the analysis is conducted considering directly the measured turbulent mass fluxes for the quantification of the exponent of \(z\).

For the measurements, the Particle Image Velocimetry (PIV) and the Laser Induced Fluorescence (LIF) techniques were applied to acquire the instantaneous velocity and concentration fields, respectively. The LIF technique was used with an oxygen quenching method that enables quantification of the dissolved oxygen concentration distribution. Besides having the advantages of being non-intrusive and instantaneous, the PIV and LIF techniques may be coupled together, providing simultaneous measurements of velocity and concentration fields which then allow the computation of molecular and turbulent mass fluxes.

2. Experimental methods

2.1 Oscillating-Grid Tank

Figure 1a is a sketch of the oscillating-grid tank used in this study, made of Perspex, with a 0.50 m x 0.50 m square cross-section and a height of 0.65 m. A grid with 6.25 cm mesh size with a solidity of 36% was used, fulfilling the criteria of Hopfinger and Toly (1976). The grid was positioned 20.0 cm above the bottom of the tank to minimize secondary motion. The experiments were conducted at the Institute for Hydromechanics of the University of Karlsruhe, Germany. Full descriptions of the oscillating-grid tank may be found in Herlina (2005) and Janzen (2006). The stroke \(S\) of the grid was maintained at 8.0 cm during the experiments, and the frequency \(f\) varied from 1.2 to 3.2 Hz. The water depth above the grid, \(h\), was maintained at 28.0 cm. The data acquisition began only 10 min after the onset of the oscillation, because the oscillating grid turbulence is sensitive to the initial conditions (Cheng and Law 2001). Table 1 presents the range of
experimental parameters of this study, where $Re$ is the “equipment Reynolds number” defined as $fS^2/v$. Janzen et al. (2010) showed that this Reynolds number is proportional to the one obtained through the product between the turbulence scales of velocity and length at the interface, being thus adequate to characterize the changes in the turbulence level at the interface. The experimental setup of the PIV and LIF measurements is shown in Figure 1b.

2.2 Concentration measurements

A LIF technique was employed to obtain dissolved oxygen concentration fields close to the air-water interface. Pyrene butyric acid (PBA) was used as the indicator of the dissolved oxygen concentration in water (Vaughan and Weber, 1970). The change in the fluorescence lifetime, $\tau$, and the fluorescence intensity, $F$ (also called quenching), is quantitatively described by the Stern-Volmer equation:

$$\frac{F_0}{F} = \frac{\tau_0}{\tau} = 1 + K_{SV}C$$

(5)

$F_0$ and $\tau_0$ are the intensity and lifetime without quencher, respectively, and $K_{SV}$ is the Stern-Volmer quenching constant. Hence, the quencher concentration (dissolved oxygen) can be determined by measuring the intensity of the emitted fluorescence (in this case from the pyrene butyric acid).

Figure 1b shows the LIF setup used in this study (nitrogen laser). The tank was filled with $2 \times 10^{-5} \text{ mol/L}$ PBA. The water in the tank was bubbled with nitrogen to remove the oxygen through stripping. An initially dissolved oxygen concentration of about 0.7 mg/L was achieved after 20 minutes of bubbling. Because the solubility of the oxygen is low, the experimental conditions imposed the saturation concentration at the interface. A pulsed nitrogen laser (MNL 801) with a mean energy power of 0.4 mJ and a wavelength emission of 337.1 nm was used to excite the PBA solution. The laser beam was guided into the centre of the tank through a UV-mirror and a combination of lenses. A FlowMaster CCD camera (1 024 x 1 280 pixels and 12 bit), with a macro objective was used to obtain images of approximately 9.5 mm x 11.9 mm from a distance of about 30 cm. The measurements were made with a resolution of approximately 9 $\mu$m. The PBA fluorescence intensity lies between 370-410 nm. An optical band-pass filter was

<table>
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<th>Run no.</th>
<th>$f$ (Hz)</th>
<th>$S$ (cm)</th>
<th>$Re$</th>
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mounted in front of the camera in order to ensure that only the fluorescent light
could pass through. The maximum number of successive images was limited to
300. Nine hundred images were taken for each run, which were averaged to obtain
the temporally averaged field. This field was then horizontally averaged to obtain
the mean profiles. A series of image-processing steps were performed on the raw
images, including noise removal, water surface detection, correction of laser
attenuation, and correction of optical blurring near the interface. The image
processing procedures followed the method described by Woodrow and Duke
(2002) and Herlina and Jirka (2004). Based on the discussion of Herlina (2005),
who used the same LIF setup of the present study, the standard deviation of the
oxygen concentration images suggests a value of about 5% for the resolution of
this set-up.

2.3 Velocity measurements

The velocity field was measured using a PIV technique. Figure 1b also shows
the PIV setup used in this study. The light source was a 25 mJ NdYag laser. A
CCD camera with a resolution of 1 024 pixels x 1 280 pixels was used to record the
images. The field of view of the camera was 2.3 cm wide and 2.9 cm high.
Polyamid particles were used as flow field tracers. After capturing and storing
the images in the computer, the PIV software DaVis® was applied to each pair of
images (cross-correlation) to obtain velocity vector fields. The interrogation area chosen to evaluate the vectors was 32 pixel x 32 pixels with a 50% overlap. The size of the interrogation area affects the data resolution, while the overlap of interrogation areas provides inherent correlations among the adjacent vectors. Nine hundred images were taken for each run. The current PIV system was evaluated to be less than 0.5 pixels (also mentioned by Herlina 2005; and Janzen 2006).

3. Theoretical considerations

3.1 Turbulent mass flux

Following the direct analogy to the molecular diffusive transport, equations 1 and 2 furnish:

\[ <c'w'> = D \frac{dC}{dz} \]  

(6)

King (1966) suggested the general form

\[ D_r = az^n \]  

(7)

where \( a \) and \( n \) are constants. This form coincides with the small eddy model predictions, in which \( n \) assumes the values of 2 or 3, for free and rigid surfaces, respectively. In this study, the exponential approximation of the normalized concentration, \( C^+ = \exp(-z^+) \), is used in equation 6 to represent the \( <c'w'> \) profiles with the general exponent \( n \). The normalized concentration is given by \( C^+ = (C - C_B)/(C_S - C_B) \), being \( z^+ = z/\delta_e \) the normalized depth. \( C_S \) and \( C_B \) are the superficial and the bulk liquid concentrations, respectively. The regression constant \( \delta_e \) is a characteristic length usually related to the diffusive layer at the surface and defined as the depth where the value of the normalized concentration is equal to 1/e = 0.37. The exponential curves were used, for example, by Chu and Jirka (1992) and Herlina and Jirka (2008). Substituting the exponential approximation into Eq. (6), and normalizing by the maximum value \( <c'w'>_{\text{max}} \), we obtain

\[ \frac{<c'w'>}{<c'w'>_{\text{max}}} = \left( \frac{z^+}{n} \right) \exp(-z^+ + n) \]  

(8)

This first approximation allows quantification of the position of the peak of the turbulent flux, given by

\[ z_{\text{max}} = \frac{\delta_{ew}}{\delta_e} = n \quad \text{or} \quad \delta_{ew} = n\delta_e \]  

(9)
That is, from equation 8 the position of the peak, \( \delta_{cw} \), is proportional to the thickness of the diffusive layer, \( \delta_e \), being the value of the proportionality constant given by \( n \). Because \( \delta_{cw} \) and \( \delta_e \) can be quantified from the measured data, \( n \) is easily obtained.

To verify more directly the exponents of \( z \) that can be used to express the turbulent fluxes \( \langle c'w' \rangle \), series expansions of the concentration and velocity fluctuations were used, respectively, as

\[
c' = c'_0 + \sum_{i=1}^{n} \frac{1}{i!} \frac{\partial^i c'}{\partial z^i} \bigg|_{z=0} z^i
\]

\[
w' = w'_0 + \sum_{i=1}^{n} \frac{1}{i!} \frac{\partial^i w'}{\partial z^i} \bigg|_{z=0} z^i
\]

At the interface \( c'_0 = w'_0 = 0 \), and the mean product of equations 10 and 11 is given by

\[
\langle c'w' \rangle = \left( \frac{\partial c'}{\partial z} \bigg|_{z=0} \frac{\partial w'}{\partial z} \bigg|_{z=0} \right) z^2 + \frac{1}{2} \left( \frac{\partial^2 c'}{\partial z^2} \bigg|_{z=0} \frac{\partial^2 w'}{\partial z^2} \bigg|_{z=0} \right) z^3 + \frac{1}{6} \left( \frac{\partial^3 c'}{\partial z^3} \bigg|_{z=0} \frac{\partial^2 w'}{\partial z^2} \bigg|_{z=0} \right) z^4 + \ldots
\]

As can be seen, equations 12 and 13 suggest theoretical values of \( n = 2 \) and 3 for free surfaces and rigid walls, respectively, similarly to those obtained for power series applied to the mean concentration (equations 4). The two gradients in the first term of equation 12 may oscillate between positive and negative values. This term can be zero only if both gradients are independent. However, literature suggests dependence between them for free surface flows, so that the mean product does not vanish (see, for example, Hasegawa and Kasagi, 2009).

A higher order approximation for the turbulent diffusivity was also considered, using two terms of a power expansion for \( D_r \), which can be viewed as an extension of the small eddy model. The turbulent diffusivity is then expressed as

\[
D_r = cz^n + dz^{n+1}
\]
where \( c, d, \) and \( n \) are constants. Substituting equation 14 and the exponential approximation, \( C^+ = \exp(-z^+) \), into Eq. (6), normalizing by the maximum value \( <c'w'^+> \max \) and considering \( \theta = c/(d\delta_e) \), we obtain a second approximation for \( <c'w'^+> \), in the form

\[
\frac{<c'w'^+>}{<c'w'^+> \max} = \frac{\left(\frac{z^+}{z^+ \max}\right)^n + \frac{(\frac{z^+}{z^+ \max})^{n+1}}{\theta}}{\exp(-z^+) \exp(-z^+ \max)}
\]

With this approximation, the nondimensional position of the peak is given by

\[
z^+ \max = \frac{\delta_{cw}}{\delta_e} = 1 + n - \theta - \sqrt{\left(\frac{1 + n - \theta}{2}\right)^2 + n\theta}
\]

Equations 8 and 15 were tested using the measured turbulent fluxes close to the surface. Equation 14 produces a maximum value of \( D_T \) along \( z > 0 \) for \( d < 0 \) (or \( \theta < 0 \)). The normalized position, \( z^+_\max \), of this maximum value is given by

\[
z^+_\max = \frac{-\theta n}{n+1}
\]

It is expected that, in oscillating grid experiments, transfer parameters like turbulent diffusivities and viscosities do not decrease farther from the interface (closer to the grid). Additionally, Schulz, Janzen and Souza (2006) showed that adequate theoretical solutions for the turbulent kinetic energy and the energy dissipation rate can be obtained using constant turbulent viscosities. So, equation 14 was used for \( 0 < z^+ < z^+_\max \) \( (D_T \) increasing with \( z \) until the peak value was reached), while the maximum value of \( D_T \) was used to calculate \( <c'w'^+> / <c'w'^+> \max \) for \( z^+ > z^+_\max \).

### 4. Results and discussion

#### 4.1 Mean concentration and length scales

Figure 2a presents the measured nondimensional mean concentration \( C^+ \) as function of the normalized depth \( z^+ \). It can be observed that all nondimensional profiles fall together and that exponential curves reproduce adequately the experimental data. Figure 2b shows the length scale \( \delta_e \) and the concentration boundary layer thickness, \( \delta_c \), versus the Reynolds number. The concentration boundary layer thickness is normally defined as the depth where the value of the normalized concentration \( C^+ \) attains a value of 0.01. The concentration boundary layer is then the distance over which the normalized gas concentration varies from the value at the surface, 1, to 0.99 times the bulk value (99% of the difference
between $C_S$ and $C_B$). Figure 2b shows that $\delta_c$ and $\delta_e$ have similar behaviors and decrease with increasing Reynolds number. This behavior agrees with the general understanding that the thickness of the “diffusive region” close to the surface must be lower for higher agitation levels of the liquid.

4.2 RMS concentration

Figure 3a presents the normalized rms concentration profiles, $c^+ = \left[ \langle c'^2 \rangle / (C_S - C_B) \right]^{1/2}$, with a normalized depth ($z^+ = z/\delta_p$). The vertical axis is normalized with the $c'$-peak coordinate. Figure 3c shows that $\delta_c$ and $\delta_e$ have similar behaviors. All profiles show a well defined peak. Steep slopes are observed close to the surface and long tails after the peak. The experimental amplitudes of the peaks vary in the range from 0.14 to 0.17, which are lower than 0.5. These results support Schulz and Janzen’s (2009) predictions, where they presented an analysis of the rms concentration profiles based on simplified random square wave records for the region near the surface. They have shown that the $rms$ concentration may be expressed as $c^+ = (1-\alpha)[(1-c^+)c^+]^{1/2}$ (where $\alpha$, usually named “reduction coefficient”, is a measure of the effect of diffusion over the $C$ records). A value of $\alpha$ close to 1 indicates strong influence of diffusion while a value close to 0 indicates weak influence of diffusion. Figure 3b presents the variation of $\alpha$ with $z^a$. It can be noted that molecular diffusion is dominant at $z^a < 1$ (values of $\alpha$ closer to 1.0).

4.3 Turbulent mass flux

The spatial evolution of the mean turbulent mass flux measured in this study is presented in Figure 4a. The turbulent flux is normalized by the maximum value $<c'w'>_{max}$ of the first measured peak, and the vertical axis is normalized with the $<c'w'>$-peak coordinate, $\delta_{sw}$, so that $z^* = z/\delta_{sw}$. Strong oscillations are observed for $z^* > 1$, but the behaviors of all profiles are similar very close to the interface.
The values of $z_{\text{max}}$, defined as the ratio between $\delta_{cw}$ and $\delta_e$, are shown in Figure 4b. It can be noted that $z_{\text{max}}$ varied in the range of around 1.5 to 2.3, with mean value close to 2. This conclusion coincides with the comments of Herlina and Jirka (2008). Considering the first approximation, from equation 9 it follows that $n=2$. Figure 4c shows the profile of $\langle c'w' \rangle / \langle c'w' \rangle_{\text{max}}$ given by equation 8 and $n=2$ together with the measured profiles. Considering the second approximation, from equation 16 it follows that, for the measured $z_{\text{max}} \approx 2$, the value of $n$ must be greater than 2 ($n=2$ is an impossible solution in this case). This result suggests that $\partial c'/\partial z|_{z=0}\partial w'/\partial z|_{z=0} \approx 0$ in equation 12, which may eventually be related to some contamination of the surface. Using the value $n=3$ also implies that $\theta=-4$, which imposes a maximum value for the turbulent diffusivity along $z>0$. Figure 4d shows the profile of $\langle c'w' \rangle / \langle c'w' \rangle_{\text{max}}$ given by equation 15 and $n=3$ for $0<z^+<z^+_{\text{DT}}$. The maximum value of $D_r$ was used to calculate $\langle c'w' \rangle / \langle c'w' \rangle_{\text{max}}$ for $z^+>z^+_{\text{DT}}$. The comparison between the measured and predicted profiles in Figure 4, for $0<z^+<1.5$ (closer to the surface), shows that $n=3$ furnishes better superpositions than $n=2$. Equation 17 furnishes $z_{\text{DT}}^+=3$ for the position of the maximum value of $D_r$, so that $z_{\text{DT}}^+/z_{\text{max}}^+ = z_{\text{DT}}/\delta_{cw} \approx 1.5$ for the present data. In all cases, the mass flux vanishes at the bottom of the tank, accumulating oxygen in the
bulk liquid. It is thus expected that $\omega c$ also approaches zero in the bulk liquid, which was observed in the measured data for increasing distances to the interface ($z^+ > 4$).

### 5. Conclusions

Simultaneous PIV and LIF measurements were used to investigate the dependence of air-water gas transfer on the turbulence generated in an oscillating-grid tank. Mean concentration profiles, $rms$ concentration profiles, and turbulent mass profiles were obtained from the measured velocity and concentration fields. The mean concentration profiles were well reproduced by exponential distributions, for which the thickness $\delta_e$ was adjusted for each run. The $rms$ profiles, $c'$, reached maximum peaks below the water surface, where amplitudes varied from 0.14 to 0.17, lower than the theoretical limit of 0.5. Close to the surface ($z^+ < 1$), the reduction coefficient $\alpha$ attained values closer to 1, indicating higher influence of diffusion in this region. It was shown that, when representing the turbulent diffusivity as truncated power series of the distance to the surface, a
single power law truncation produced a power $n = 2$ for the measured data, which approximately reproduces the measured $<c'w'>/<c'w'>_{\text{max}}$ profiles close to the surface. Considering two terms of the power expansion for the turbulent diffusivity, resulted in a power $n$ greater than 2 for the measured data. The value $n = 3$ was used, leading to a predicted $<c'w'>/<c'w'>_{\text{max}}$ profile that superposes well the measured profiles. Although not directly observed, the higher value of the exponent may be related to some contamination of the surface. The mass flux vanishes at the bottom of the tank, accumulating oxygen in the bulk liquid and attaining low values also in the bulk liquid. The measured data and the predictions show that $<c'w'>$ is zero at the interface, grows to a peak value and decreases with increasing distance from the interface.

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**References**


Janzen, J.G., (2006), Gas transfer near the air-water interface in oscillating-grid tanks and properties of isotropic turbulent flows (Portuguese: Fluxo de massa na interface ar-água em tanques de grades oscilantes e detalhes de escoamentos turbulentos isotrópicos oscilantes), Doctoral thesis, 170 pp., University of São Paulo, Brazil.