LARGE EDDY SIMULATIONS OF REYNOLDS AND SCHMIDT NUMBERS EFFECTS ON MASS TRANSFER OF GASEOUS COMPOUNDS FROM QUIESCENT LIQUID SURFACES

Enilene Regina Lovatte, enilene.lovatte@ifes.edu.br
Bruno Furieri, bruno.furieri@ifes.edu.br
Melina Barbosa Peixoto, melina.peixoto@ifes.edu.br
Instituto Federal do Espírito Santo, Av. Vitória, 1729, Jucutuquara, Vitória, 29046-020, Brazil

Neyval Costa Reis Junior, neyval@gmail.com
Jane Meri Santos, jmerisantos@yahoo.com.br
Universidade Federal do Espírito Santo, Av. F. Ferrari, 514, Goiabeiras, Vitória, 29075-910, Brazil

Harerton Dourado, harerton@terra.com.br
Faculdades Integradas de Aracruz, Rua Prof. Berilo Basílio dos Santos, Centro, Aracruz, 29194-910, Brazil

Abstract. *The present work aims to study the volatilization of compounds from quiescent liquid surfaces exposed to atmospheric flow using Large Eddy Simulation (LES). Among the gaseous compounds investigated we included an odorant gas of environmental interest, the hydrogen sulphide (H2S). The H_2S is commonly dissolved in stabilization ponds of Wastewater Treatment Plants. The mass, momentum and chemical species conservation equations are numerically solved employing a commercial CFD code (Ansys Fluent). We investigated the performance of LES with dynamic subgrid model in the region near the gas-liquid interface. Numerical simulations were performed with Reynolds number based on the friction velocity equal to 150, 640 and 1280 and Schmidt numbers equal to 1, 200 and 580 allowing specific investigation of the effects of these parameters governing the mass transfer phenomenon. The validation of LES was carried out by comparisons with DNS results. The methodology is appropriate in understanding the mass transport and turbulence structures visualization (coherent structures) that directly affect the process. The study demonstrated that LES is suitable for understanding the basic principles of the mass transfer in quiescent surfaces, allowing investigation of the effect of high Reynolds and Schmidt numbers. The influence of Reynolds number is mainly associated with reduced thickness of the diffusive sublayer due to the increase of scalar mixing in the bulk. The thickness of the diffusive sublayer near the free surface greatly decreases in cases where the Schmidt number increases.*

Keywords*: CFD, LES, mass transfer, odorous compound, Reynolds number, Schmidt number*

1. INTRODUCTION

The environmental problematic associated to odorous compounds emissions is a huge concern in our cities due to some sources as many industrial sites or wastewater treatment plants (Hudson, 2008). The annoyance caused by the pollution of these structures near the population is significant and a quantification of contaminants transferred to the atmosphere such as hydrogen sulphide (H_2S) is very important. The calculation of contaminant mass transfer may be performed experimentally or numerically (Kumar, 1998, Nezu and Wolfgang, 1996, Sowka et al., 2014). However, experimental measurements of the variables of interest, such as the overall mass transfer coefficient, are difficult to be achieved due to the small thickness of the concentration boundary layer near the interface. The concentration boundary layer thickness is a parameter to be quantified for the comprehension of scalar transfer through gas-liquid interfaces. Numerical simulations allow an accurate investigation of the phenomena for scales even smaller than the thickness of an interface. They are performed much more simply than experiments which would require a lot of effort to measure such a thin layer.

Reynolds Averaged Navier-Stokes (RANS), Large Eddy Simulation (LES) and Direct Numerical Simulation (DNS), classified here before by computational cost and accuracy, may perform the numerical modelling of fluid flow and scalar transport. The practical unfeasibility of DNS in flows with high Reynolds numbers has motivated the investigation of LES and its various sub-grid scale models available to describe the liquid flow and the mass transfer of gaseous substances near free surfaces accurately (Salvetti et al., 1997, Shen et al., 2001, Abba et al., 2003, Yu-Hong et al., 2003).

The turbulent structures near free surface are associated to mass transfer. LES allow the study of turbulent flows with high Reynolds number (Re) being compatible with practical applications. In addition, we are also able to solve problems with large Schmidt numbers (Sc), which means compounds with odorous characteristics, having very thin concentration boundary layer that requires very refined mesh near the interface. For the Schmidt number corresponding

to the odorous compound, the turbulent structure in the liquid flow controls the scalar transfer at the free surface. The authors have not found any work in recent literature that associate turbulence near free surface to existing odorous compound transfer as the hydrogen sulphide is investigated in the present paper.

Reboux et al. (2006) indicated that the values of Reynolds numbers based on friction velocity having interest in practical applications are of the order 103 and Hasegawa and Kasagi (2009) pointed out that the Schmidt numbers values range from 1 to 200. Various authors performed LES to solve fluid flow and scalar transport near interface gas-liquid, including Calmet and Magnaudet (2003), Shen and Yue (2001) and Dong et al. (2003). These authors investigated mass transfer of compounds with very low solubility, which means high diffusivity and low Schmidt number. In fact, only the transfer of hydrogen sulphide is taken into account.

The aims of the present work are the investigation of turbulent fluid flow near a quiescent surface and the quantification of the overall mass transfer coefficient as a function of Schmidt number by means of LES technique with Smagorinsky Dynamic Model (SDM). The results are compared with DNS data (Nagaosa, 1999) in order to validate both numerical and turbulent models. The numerical simulations were realized for a fixed Reynolds number based on friction velocity (Re=1280) and three Schmidt numbers (Sc =1, 100 and 580). Schmidt number equals to 580 is equivalent to hydrogen sulphide (an odorous compound) dissolved in water. Sc=1 and Sc=100 are not associated to any existing odorous compound dissolved in water, but they are also discussed in the present work permitting a better understanding of the influence of the Schmidt number on mass transfer.

2. METODOLOGY

2.1 Governing Equations and Turbulence Modelling

The use of Computational Fluid Dynamic (CFD) allows a more detailed description of the fluid flow and mass transfer and allows a better understanding of the physical phenomena involved in the processes. In order to estimate the rate of volatilization of dissolved compound from quiescent liquid surfaces, various computational simulations were performed in which the commercial computational code FLUENT developed by ANSYS was used. The FLUENT code uses the Finite Volume Method (FVM) to solve the conservation equations of mass, momentum and mass of chemical species.

The LES turbulence model equations describe the movement of large eddy structures. The sub-grid scale (SGS) model is used to represent the effects of unresolved small scales of fluid motion (small eddies). Smagorinsky dynamic sub-grid scale model (SDM) is used and the non-dimensional governing equations are given as:

$$\frac{\partial\left(\bar{u}_i\right)}{\partial x_i}=0 \tag{1}$$

$$\frac{\partial\left(\bar{u}_i\right)}{\partial t}+\frac{\partial\left(\bar{u}_i\bar{u}_j\right)}{\partial x_j}=-\frac{\partial\bar{P}}{\partial x_i}+\frac{1}{\mathrm{Re}}\frac{\partial^2\bar{u}_i}{\partial x_j\partial x_j}-\frac{\partial\tau_{ij}}{\partial x_j} \tag{2}$$

$$\frac{\partial\bar{C}}{\partial t}+\frac{\partial\left(\bar{C}u_j\right)}{\partial x_j}=\frac{1}{\mathrm{Re}\,Sc}\frac{\partial^2\bar{C}}{\partial x_i\partial x_j}-\frac{\partial q_j}{\partial x_j} \tag{3}$$

where, $\tau_{ij=}R_{ij}-1/3R_{kk}\delta_{ij}$, $R_{ij}=\overline{u_iu_j}-\bar{u}_i\bar{u}_j$, $q_j=\overline{Cu_j}-\bar{C}\bar{u}_j$, overbar "¯" denotes the resolved variable, the Reynolds number Re is defined as $\mathrm{Re}=u*\delta/\mathrm{v}$, and the Schmidt number Sc as $Sc=\nu/D$, here ν is the kinematic viscosity and D is the molecular diffusivity of scalar, and $\bar{\mathrm{P}}$ is the resolved pressure, which contains a term $R_{kk}/3$, t is the time, u_i,u_j,u_k are the components of the velocities in the directions i,j,k respectively, x_i,x_j,x_k identify the coordinates in the directions i,j,k, respectively. $\bar{C}$ is resolved concentration.

Reynolds and Schmidt numbers are the main dimensionless parameters governing the present study. We evaluate the influence of these parameters on the rate of volatilization of an odorous compound present in the liquid phase. The Reynolds number gives a measure of the importance of inertial forces (associated with the advection effect) compared to viscous forces. Reynolds number characterizes a flow condition (laminar or turbulent). The Reynolds number based on friction velocity considered in this work is 1280.

Schmidt number associates diffusion and momentum. It measures the ratio of hydrodynamic and mass diffusion layers' thickness. The Schmidt number considered for the odorous compound studied in the present paper (hydrogen sulphide) dissolved in water equals to 580. Others Schmidt numbers tested in our work (1 and 100) were calculated modifying the diffusivity coefficient and keeping constant the kinematic viscosity of water. In fact, they do not represent any odorous compound.

In Equations (2) and (3), τ_{ij} and q_j represents SGS turbulent stress and mass flux, respectively, which need to be modelled by SGS models and are given as Equations (4) and (5). It is assumed that the mass concentration is a passive scalar that does not influence the flow dynamics.

$$\tau_{ij} = -2G\overline{\Delta}^2 \left|\overline{S}\right| \overline{S}_{ij} \tag{4}$$

and

$$\tau_{ij} = -\frac{G\overline{\Delta}^2}{Sc_t} 2\left|\overline{S}\right| \frac{\partial \overline{C}}{\partial x_j} \tag{5}$$

where

$$\overline{S}_{ij} = \frac{1}{2}\left[\frac{\partial \overline{u_i}}{\partial x_j} + \frac{\partial \overline{u_j}}{\partial x_i}\right] \quad , \quad \overline{S} = \left|\sqrt{2\overline{S}_{ij}\overline{S}_{ij}}\right| \tag{6}$$

The model coefficients of G and Sc in the Equation (4) e (5) are calculated as proposed by Germano et al. (1993). After introducing a test filtering with width $\overline{\overline{\Delta}}$ to Equations (1) to (3), the coefficients G and Sc, can be dynamically determined, as:

$$G = -\frac{1}{\overline{\Delta}^2} \frac{\left\langle L_{ij} M_{ij} \right\rangle_s}{\left\langle M_{ij} M_{ij} \right\rangle_s} \qquad Sc_t = -G\overline{\Delta}^2 \frac{\left\langle F_i F_i \right\rangle_s}{\left\langle H_i F_i \right\rangle_s} \tag{7}$$

where

$$M_{ij} = 2\alpha^2 \left|\overline{\overline{S}}\right| \left[S_{ij} - \frac{1}{3}\overline{\overline{S}}_{kk}\delta_{ij}\right] - \overline{m}_{ij} \qquad m_{ij} = 2\left|\overline{S}\right| \left[S_{ij} - \frac{1}{3}\overline{S}_{ij}\delta_{ij}\right] \tag{8}$$

$$L_{ij} = \overline{\overline{u}_i \overline{C}} - \overline{u}_i \overline{C} \qquad F_i = \alpha^2 \left|\overline{\overline{S}}\right| \overline{B}_i - \left|\overline{S}\right| \overline{B}_i \qquad B_j = \frac{\partial \overline{C}}{\partial x_j} \tag{9}$$

Herein, $\alpha = \overline{\overline{\Delta}} / \overline{\Delta}$ and $\langle\ \rangle$ denotes spatial averaging.

2.2 Numerical Simulation Set-up

The computational domain is an open-channel flow of height 2δ in which turbulence is supposed to be fully developed. Streamwise (x) and spanwise (z) directions are periodic. A constant streamwise pressure gradient equals to $dP/dx = -\rho u^{*2}/2\delta$ drives the flow, where u^* is the friction velocity. The boundary condition at the bottom is set as no slip $(\mathrm{u,v,w} = 0)$ and free slip condition $(\partial u_i / \partial x_i = 0)$ is set at the top boundary (called herein free surface). For the chemical species conservation equation, the boundary conditions are set as zero on the bottom and equal to unity on the free surface. The entire domain presents concentration equals to 1 as initial condition.

The size of the computational domain is 2πδ (longitudinal dimension) × πδ (transversal, horizontal dimension) and δ (transversal, vertical dimension) conform showed Figure 1. The mesh is uniform in x and z direction, while in the y direction, the mesh is refined near the free surface and bottom for a better prediction of scalar concentration gradient. The Reynolds number (based on friction velocity on the wall) equals 150 in the reference DNS investigation. In this case, the computational mesh used contains 77, 62 and 89 points in x, y and z directions (longitudinal, normal and transverse to the main flow). The requirement of an excessively refined computational mesh and a minor time step size requires high computational resources.

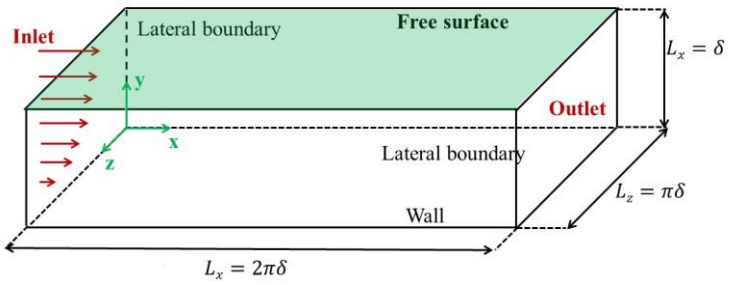


Figure 1: Computational domain

2.3 Governing Parameters

Reynolds and Schmidt numbers (Equations 10 and 11) are the main dimensionless parameters governing the present study. We evaluate the influence of these parameters on the rate of volatilization of an odorant compound present in the liquid phase. The Reynolds number is dimensionless which gives a measure of the importance of inertial forces (associated with the convective effect) compared to viscous forces. This number is used to characterize the type of flow (laminar or turbulent). For a small value of the flow, Reynolds displays small instabilities. The Schmidt number is the ratio of diffusion processes and the moment of mass. Measures the ratio of the thicknesses of the layers hydrodynamics and mass diffusion.

$$\mathrm{Re} = \rho \frac{ul}{\mu} \tag{10}$$

$$Sc = \frac{\nu}{D} \tag{11}$$

where u is the characteristic velocity, ρ is the density of the fluid, l is the characteristic length and μ is the dynamic viscosity of the fluid. The Reynold number considered in this work is 1280. ν and D respectively are the kinematic viscosity and diffusivity of the chemical species that diffuses the fluid.

For a given temperature, a compound, which presents different diffusivity when dissolved in various fluids (e.g. water and air) presents different Schmidt numbers. The Schmidt number considered for hydrogen sulphide (H_2S) in this study is 580.

3. RESULTS AND DISCUSSIONS

3.1 Validation of LES result

The validation of numerical modelling was performed by comparing our results with DNS data from Nagaosa (1999). Figures 1(a) to 1(d) present the validation obtained under the same conditions simulated by Nagaosa (1999) with a less refined mesh using the turbulence model LES-SDM. A very good agreement was found between present data and those obtained by DNS. There is no experimental work in literature that has carried out measurements able to validate our numerical results. For all profiles, the abscissa shows the dimensionless vertical coordinate in wall units.

$$y^+ = \frac{u^* u^+}{\nu} \quad \text{and} \quad y^+ = \frac{u}{u^+} \tag{12}$$

where y is the vertical coordinate, u^* is friction velocity and ν is the kinematic viscosity of water. The ordinate axis in Figure 2(a) is the non-dimensional mean streamwise velocity. Log and linear regions of the vertical profiles were very well represented by LES calculations as we can see in Figure 2(a). In Figure 2(b) one can notice the validation of first order turbulence quantities. Velocity fluctuation components in streamwise and spanwise directions increase toward the free surface, while the velocity fluctuation of vertical components decreases, showing that the anisotropy of the velocity field becomes significant in the region. Consequently, the anisotropy of velocity field becomes significant in the region near the free surface. The mean vertical profile of scalar concentration is also validated in these simulations (Figure 2 (c)). Vorticity fluctuations (Figure 2 (d)) play a very important role in the description of turbulent flow. It is interesting to observe that our modelling (numerical and turbulence) provide trustful results even if we apply a coarse mesh (compared to that used in DNS) and if we implement a sub-grid model inherent to LES.

The profiles found in the present study are in excellent agreement over the entire height of the channel with Nagaosa study DNS data and Handler (2003). Only a slight difference is observed in the range between 30 and 100. The very near the free surface area (for example) the profiles are perfectly matched. The vertical profile of a turbulent flow channel is indicated by the product of average concentration and the floating fluctuation of the vertical velocity component. Figure 2 (e) shows this standardized profile. The Figure 2 (f) shows the RMS profile flotation concentration given by the standard equation. It is possible to observe two peaks along the time being this the highest given boundary conditions established in the background (non-slip) and top (free surface). This always was observed in the chart obtained from DNS data Nagaosa and Handler (2003).

3.2 Turbulence structures

In Figure 3, the areas demarcated by the letters A, B and C identify pairs of vortices (with positive and negative vertical vorticity) in each of the dimensionless time, illustrating the new position of each of these pairs in the calculation domain measurement in which time progresses. The pairs of vortices become deformed over time. The structure highlighted by the region A extends in the longitudinal direction while the structure highlighted by the letter C basically keeps the same for the four moments shown The evolution of structures also reflects the timing of the flow. The structure of the reappearing in time out of the field at time (d). Further analysis shows that most of the vortices moves preferably in the main flow direction, these being important structures in understanding transport of scalar next free surface.

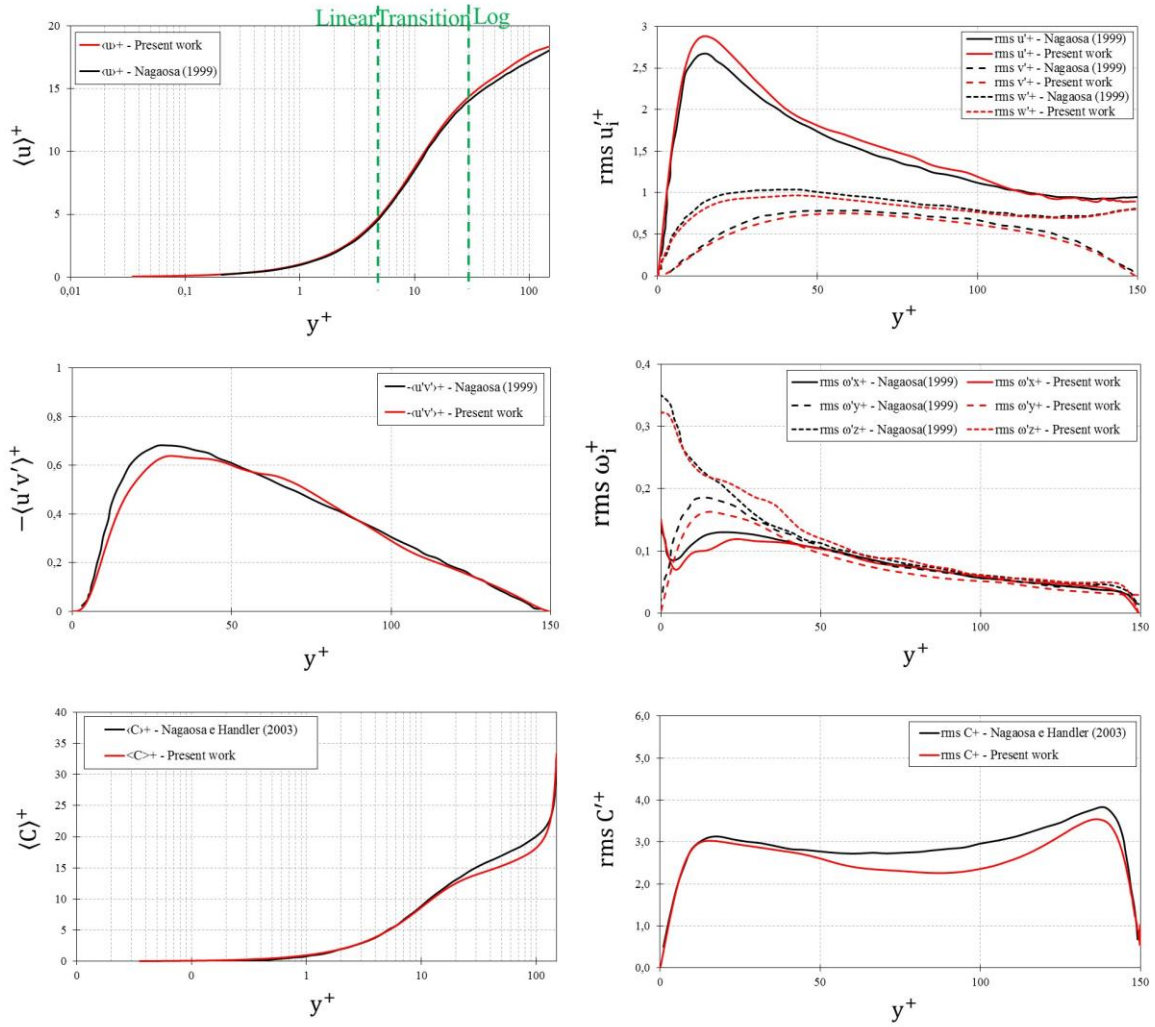


Figure 2 - Vertical profile of mean longitudinal velocity; (b) Vertical profile of velocity RMS; (c) Vertical profile of mean scalar concentration; (d) Vertical profile of vorticity RMS (e) Vertical profile of mean concentration and (f) Vertical profile of concentration RMS.

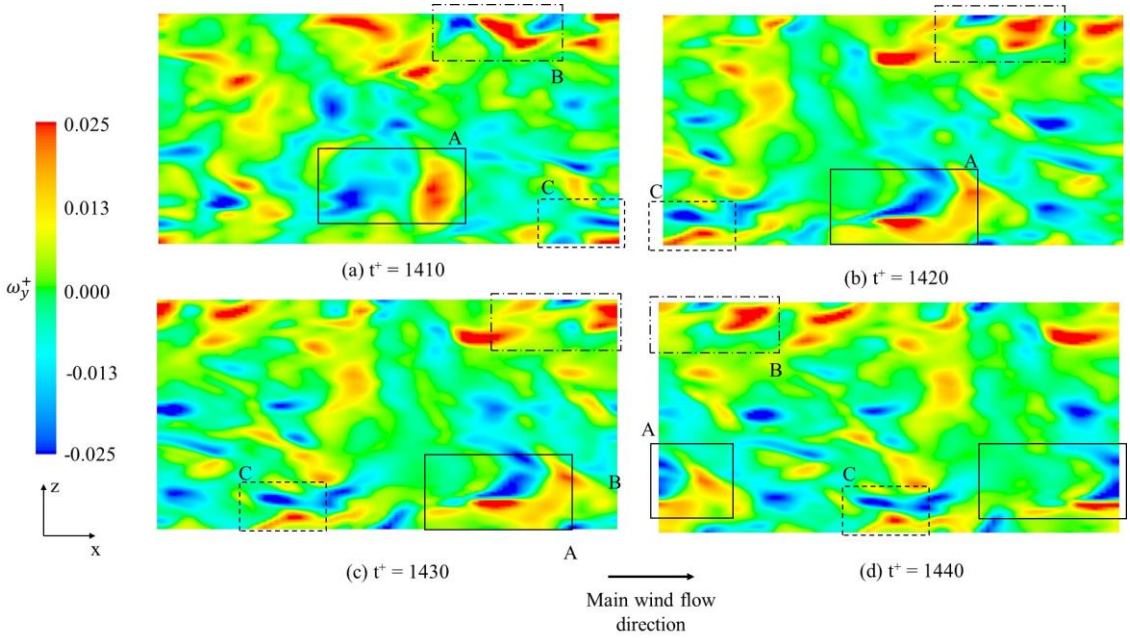
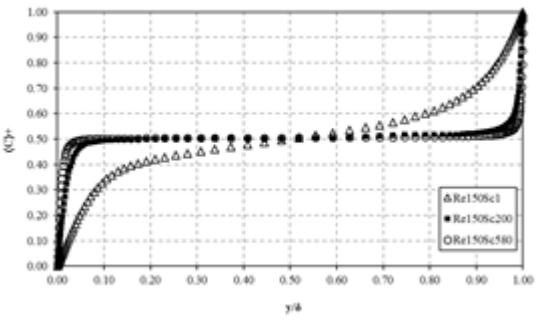


Figure 3 – Distribution of field vertical component of instantaneous vorticity in the XZ plane just below the free surface (y + = 145) in successive times. (a) t $^+$ = 1410, (b) T $^+$ = 1420, (c) = t $^+$ 1430 and (d) = t $^+$ 1 440

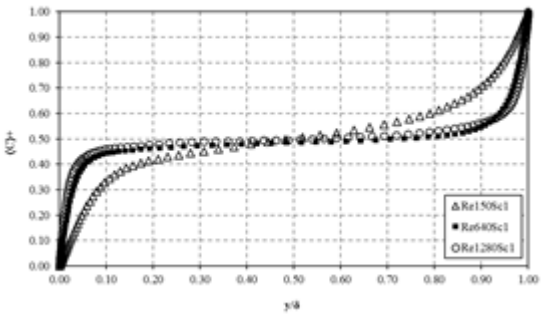
3.2 Analysis of mass transfer behavior for various Schmidt number and Reynolds number

In this study, different cases were tested for analysis of mass transfer phenomena compounds in quiescent liquid surface. Three Schmidt numbers (1, 200 and 580) and three Reynolds number based on the friction velocity (150, 640 and 1280) were investigated. As stated earlier, the main objective of this study is to calculate the transfer of a passive scalar in turbulent flow and analyze the influence of these parameters on mass transfer compound. LES methodology and the dynamic subgrid modelling used were able to correctly predict the mass transfer coefficient variations. The mesh used in the vicinity of the free surface is sufficiently fine to capture the small scales the vertical scalar field, resembling the grid resolution used in DNS studies, as defined by Jiang and Lai (2009).

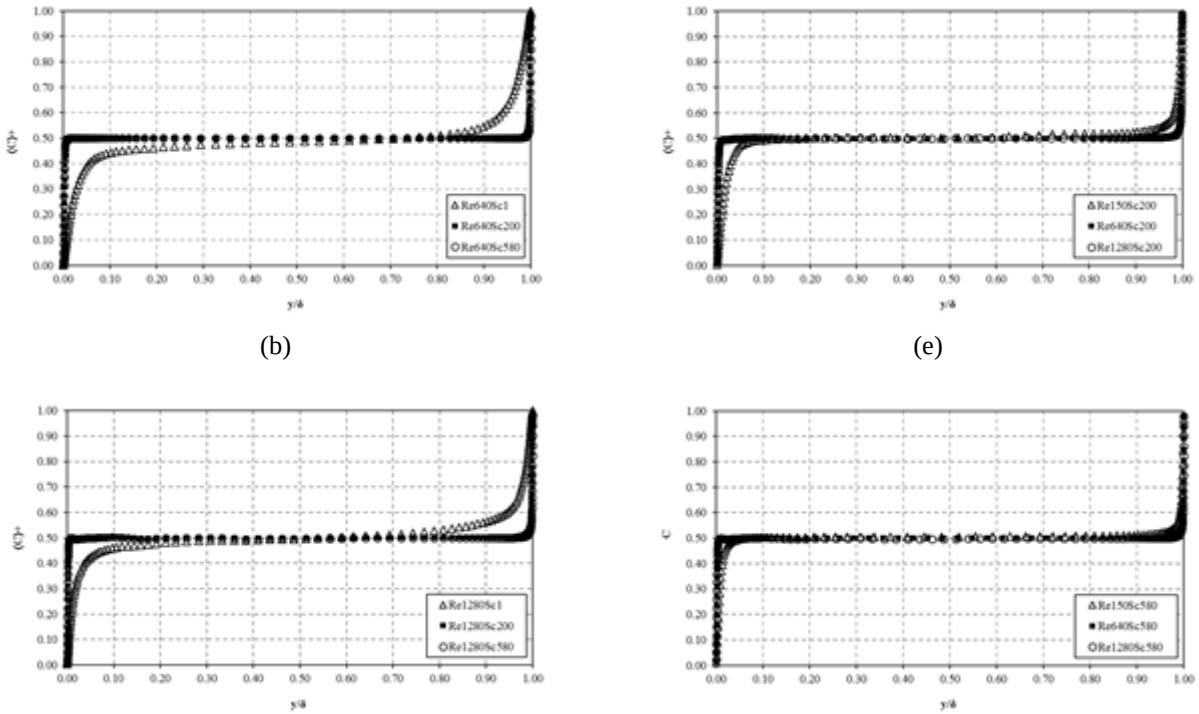
The curves shown in Figure 4 (a) to Figure 4 (f) are related to the mean concentration values obtained in the present study. The curves have similar forms, however, near the free surface due to the high concentration gradient of the curve is remarkably higher for Schmidt number values higher.



(a)



(d)



(b) (e)

(c) (f)

Figure 4 – Vertical profile of mean concentration: (a) Re = 150 varying Schmidt, (b) Re = 640 varying Schmidt, (c) Re = 1280 varying Schmidt, (d) Sc = 1 varying Reynolds, (e) Sc = 200 varying Reynolds and (f) Sc = 580 varying Reynolds.

The trend of the curves is very similar for all three studied Schmidt numbers, however, it can be observed that for higher Schmidt numbers the maximum value of concentration fluctuation is closer to the free surface. In terms of dimensional distance, the peaks are located, respectively, to Schmidt numbers of 1, 200 and 580. Another important analysis to be made, it refers to sudden decay of RMS values of the fluctuation in concentration toward the sinus drainage, Schmidt for larger numbers, which is not easily noticed for the Schmidt number equal to unity and is apparent to Schmidt numbers equal to 200 and 580. As the current discussion concerning the same and different Reynolds numbers Schmidt, reduced diffusivity results in the reduction in the variations of concentration values.

The sequence graphics of Figure 4 (d) to Figure 4 (f) refers to the vertical profiles mean concentration calculated for different Reynolds numbers and Schmidt numbers: (a) Sc = 1 varying Reynolds (b) Sc = 200 varying Reynolds and (c) varying Reynolds Sc = 580. It is noteworthy that for higher values of Reynolds number the thickness of the sublayer is less diffusive, decreasing as the Reynolds number increases. In Figure 4 (d) where the scalar has Schmidt number equal to 1, increasing the Reynolds number rather modifies the shape of the especially near the free surface profile and the bottom surface. Specifically concerning the present study objectives, the mean concentration gradient in the free surface undergoes significant changes as the Schmidt number increases, these influences the Reynolds number is more noticeable in these profiles but are felt particularly when calculating the transfer coefficient mass, which depends on the concentration gradient.

3.3 Variation in mass transfer coefficient with the Schmidt number and Reynolds number

The mass transfer coefficient in liquid phase is an essential parameter for calculating the flux of the compound that is transferred by volatilization of the liquid phase to the gaseous phase. The value is dependent on the following variables: concentration of the compound within the flow of liquid phase, concentration of the compound within the gaseous phase constant and Henry's law. Figure 6 shows a 3D visualization on how Reynolds and Schmidt numbers have influence on the mass transfer coefficient in liquid phase. From this graphic is instantaneously noticeable that the lowest values of Schmidt numbers and highest Reynolds numbers lead to the highest emission rate.

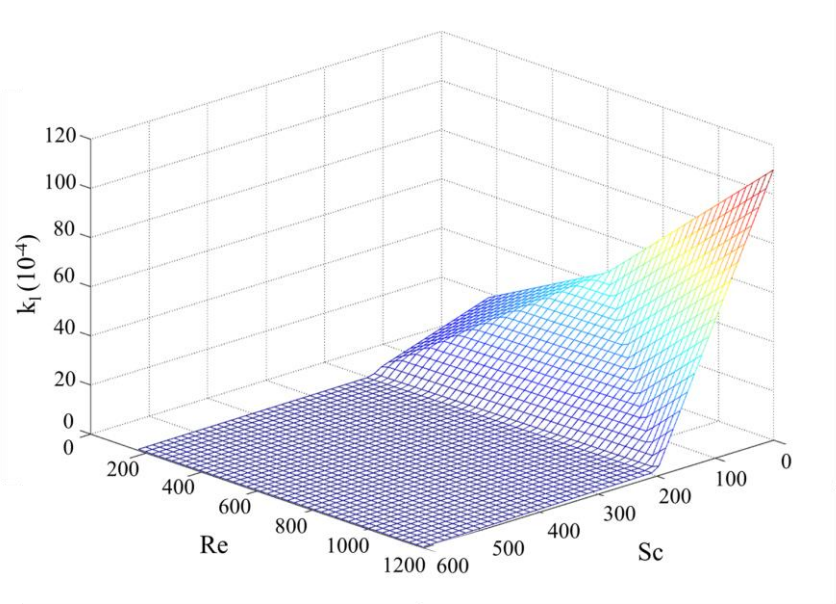


Figure 6 – Variation in mass transfer coefficient with the Schmidt number and Reynolds number

4. CONCLUSIONS

The present paper presented an overview of the mass transfer of chemical species and turbulent fluid flow structures near the air-water interface. The numerical simulations corresponded to open channel where both flow and scalar fields were validated by DNS results found in literature. We investigated the influence of Schmidt numbers on the overall mass transfer coefficients. LES remained a very appropriate methodology to investigate the principles of mass transfer mechanisms through gas-liquid interface in quiescent surfaces. Numerical results shown a strong correlation between vertical velocity fluctuation and scalar fluctuation very near the free surface. The second analysis showed the tested cases varying with Schmidt numbers for a fixed value of Reynolds number based on the friction velocity (1280). The smallest Schmidt number (Sc=1) is associated to higher gradient of scalar concentration and consequently higher volatilization compared to the other tested Schmidt numbers (Sc=100 and Sc=580). The diffusivity is smaller as Schmidt number increases for a same fluid where the compound is dissolved. The transport for higher Schmidt numbers are very much slow. This is associated to high solubility of the compound meaning small volatilization and overall mass transfer coefficient. Numerical simulations greatly help the investigations of emissions of odorous compounds from quiescent surfaces. This relatively easy and trustable way to calculate overall mass transfer coefficients is a very useful tool for dispersion modelling studies that need odorous compounds emission rates as input data for regulatory purposes.

5. ACKNOWLEDGEMENTS

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