A numerical method for the study of solid-liquid mass transfer in turbulent flows

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Abstract. A numerical method for the study of high-Schmidt number mass transfer close to solid boundaries is presented. Computational Fluid Dynamics with the Large Eddy Simulations and the dynamic Smagorinsky model for the subgris scale stress tensor were used. Periodic boundary conditions were used to obtain proper boundary conditions for the LES simulation. A fine mesh was used in the bulk of the flow and an extremely fine mesh ($y^+ = 1/40$) on a part of the surface where the diffusional flux was evaluated. Model validation shows that the theoretical $y^+$ dependences were obtained for the fluctuations parallel to the surface and a somewhat lower $y^+$ dependence was obtained for the much smaller perpendicular fluctuation. It is also shown that DNS resolution was obtained in the extremely fine mesh. The usefulness of the modeling method is exemplified with a simulation of species concentration and by particle tracking the influence of the turbulence on the fluid elements near the walls at $y^+ < 1$.

1. Introduction

The existence of proper models for the prediction of near-wall flows is of fundamental importance in all fluid flow modeling. Momentum transport between the wall and fluid determines many of the flow properties that usually are of interest, for example the intensity of turbulence, pressure drop, etc. Near-wall momentum transport has been thoroughly investigated by a large number of scientists during the last century, and the models we have today are usually considered to be sufficient in most situations. The outcome of most models is that a combination of viscous (molecular) and turbulent effects contribute to the wall-normal momentum transport. As the wall is being approached, viscous effects dominate, and vice versa. Even though turbulent structures occasionally reach almost all the way in to the wall surface, fluid viscosity evens them out relatively quickly reducing their influence on overall momentum transport. The impact of such very near-wall turbulent structures is thus very limited.

It is possible to extend the same modeling arguments to heat and mass transfer situations. Heat transport from a solid wall is governed by the same fundamental transport mechanisms as momentum transport, and as long as the thermal diffusivity $\alpha$ is numerically comparable with the fluid viscosity $\nu$ ($Pr = \nu/\alpha \sim 1$), it has been established that analog relations between momentum and heat can be used to accurately predict heat transfer rates. The Reynolds and Chilton-Colburn analogies are famous examples of such analogous relations. An identical approach can be used for mass transfer modeling as long as the mass diffusivities $D$ are numerically similar to the fluid viscosity ($Sc = \nu/D \sim 1$).
Problems arise, however, in situations where $Sc \gg 1$. Such cases include many industrially important operations, and almost always when liquid solvents are used (Welty et al., 2001). In liquids it is not unusual that $Sc > 1,000 - 3,000$. In such cases it is no longer possible to use the same modeling arguments as earlier since mass diffusivities are much smaller than the fluid viscosity. This means that mass concentration gradients will be much steeper than its viscous counterparts and the effects of near-wall turbulence on momentum and mass transport will thus be fundamentally different.

Near-wall flow can be studied using Computational Fluid Dynamics (CFD). However, for the complete resolution of the species concentration the flow must be resolved at the Bachelors scale i.e. the Kolmogorov scale $\eta Sc^{1/2}$ (Fox, 2003). In liquids this corresponds to a length scale $> 30$ times smaller than the Kolmogorov scale. Resolving spatially and temporally all the turbulent fluctuations of velocity and concentration would require a prohibitively large amount of computer power, and this will also be case for the foreseeable future. It is therefore necessary to use a coarser grid, and then to model the influence that the unresolved fluctuations have on the resolved quantities using a turbulence model.

Many different turbulence models exist in the literature. In the specific case, the choice of model mainly depends on the specific focus of the simulation at hand and the level of details wanted, the amount of computer resources available, and the time aspect. The study of turbulent structures close to walls requires that a model is used that is capable of resolving these structures accurately. Previous findings (Spalart, 2000; Calmet & Magnaudet, 1997) suggest that the Large Eddy Simulations (LES) approach is appropriate together with the Dynamic Smagorinsky Model (DSM) for this kind of analysis. The standard procedure in using LES is that at least 80% of the kinetic energy of the flow is resolved in the grid (Pope, 2000). This usually corresponds to a near-wall resolution of approximately one (1) non-dimensional wall unit (wu.; wall normal distance scaled with the friction velocity and molecular viscosity). If the mass concentration gradients ($Sc \gg 1$) are to be resolved it would be necessary to go below 1 wu.

To model the impact of the sub-grid concentration fluctuations on the resolved concentration field, it is assumed that the sub-grid viscosity ($\nu_{SGS}$) modeled by DSM can be scaled into a sub-grid mass diffusivity ($D_{SGS}$) using a sub-grid scale Schmidt number ($Sc_{SGS}$). $< \phi >$ denotes the space filtered variable $\phi$.

$$< u_j c > = -D_{SGS} \frac{d < c >}{dx_j}$$

$$D_{SGS} = \frac{\nu_{SGS}}{Sc_{SGS}}$$

$c$ is species concentration, and $u_j$ is the velocity in the $j$ direction. Being scalar entities, the SGS viscosity and diffusivity cannot account for the effects of sub-grid scale anisotropic turbulence. In such situations mass and momentum are transported unequally efficiently in different spatial directions. Tavoularis & Corrsin (1985) have investigated the effect of shear on the turbulent diffusivity tensor. The arguments presented there can be analogously applied on $D_{SGS}$. Chumakov (2008) showed that the angle between the SGS mass transport and the gradient of the mean concentration is significant. The consequence of these findings is that the anisotropic turbulence must be resolved in order for the analysis to be correct.

The SGS Schmidt number is not constant, but varies with the molecular Schmidt number and the distance to the wall, see Reynolds (1975) for a comparative presentation of different models. This deviation comes from the anisotropic structures close to the wall. Bergant & Tiselj (2007) found that the turbulent $Sc$-number increases with $Sc$ from unity for $Sc = 1$ to 8 at $Sc = 500$. This uncertainty also makes it very important to have a very fine grid close to the wall and all mass transfer should be resolved in the simulations to obtain reliable results i.e. to use DNS close to the wall.
In this paper we present a method of studying near-wall flow and mass transfer using CFD. The method will be tested by comparing with theoretical models for average flow and turbulence characteristics close to walls. Also, by comparing the transport terms for species we show that the grid resolution is sufficiently fine to neglect the sub-grid turbulent transport. The flow in turbulent boundary layers is complex in many ways and is not fully understood (Hunt & Morrison, 2000). Particle tracking is used to visualize this part of the flow. We also show different ways of interpreting the results from such simulations.

2. Theory

2.1. Large Eddy Simulations (LES)

In Large Eddy Simulations, only the largest eddies are resolved (filtered) in the mesh. The smaller eddies, in the so called sub-grid scale (SGS) region, are not resolved but their effect on the resolved scales are modeled using a SGS model. The motivation behind LES is that smaller scale turbulence is easier to model accurately than the larger eddies, and that a majority of the turbulent kinetic energy resides with the larger eddies. The absence of upstream history-effects makes it possible to model smaller eddies using only local (spatial and temporal) variables, and their higher degree of isotropy opens up for the possibility to model their influence on the resolved variables as being of scalar type.

In this paper, we have chosen to call the flow variables "resolved" and "sub-grid", instead of "filtered" and "residual". There is some controversy regarding this naming convention, see for example Pope (2000).

The governing equations for LES with mass transfer are the filtered Navier-Stokes equations together with the filtered mass balance.

\[
\frac{\partial < u_i >}{\partial t} + < u_j > \frac{\partial < u_i >}{\partial x_j} = -\frac{1}{\rho} \frac{\partial < p >}{\partial x_i} + \frac{\partial}{\partial x_j} \left( \nu \frac{\partial < u_i >}{\partial x_j} - < u_i u_j > \right) \quad (1)
\]

\[
\frac{\partial < c >}{\partial t} + < u_j > \frac{\partial < c >}{\partial x_j} = \frac{\partial}{\partial x_j} \left( D \frac{\partial < c >}{\partial x_j} - q_j \right) \quad (2)
\]

The filtering operation \((G)\) is a filtering kernel.

\[
< \phi(\mathbf{x}) > = \int G(\mathbf{x} - \xi)\phi(\xi) \, d\xi
\]

Filtering can be done in physical space as well as in frequency space, and a large variety of different filters exist (Pope, 2000). In this paper, we use a physical box filter that corresponds to the computational mesh. The sub-grid terms \(< u_i u_j > \) and \(q_j\) on the right hand side of Eqs. 1 and 2 represent the impact of the SGS velocities and concentration on the resolved variables. They must be modeled.

The filter length scale can be thought of as a border that separates the resolved part of the variable from the SGS part. Since the prime focus of this paper is to study high-Sc mass transfer it is important that not only viscous arguments are used when determining this length scale, but also arguments that take into account the behavior of slowly diffusing species \((Sc \gg 1)\). This means that the mesh will have to be fine enough to leave only the smallest concentration fluctuations to be modeled.

2.2. The dynamic Smagorinsky model (DSM)

The SGS stress tensor can be modeled assuming that it is proportional to the resolved rate-of-strain,
The proportionality constant \( \nu_{SGS} \) is an SGS viscosity. This relation has a similar form to the eddy-viscosity concept used in RANS models. \( L \) is a length scale and \( |S| \) is the magnitude of the filtered rate-of-strain \( S_{ij} = 1/2(\partial <u_i>/\partial x_j + \partial <u_j>/\partial x_i) \). \( C_s \) is a model constant.

Equation 3 assumes that the SGS stress tensor is in perfect alignment with the resolved rate-of-strain tensor. In many situations this is found not to be correct (Chumakov, 2008).

There are a large variety of different models for \( \nu_{SGS} \), see for example Abba et al. (2003) where different models were tested a priori. The earliest model (Smagorinsky, 1963) suggests that \( C_s \) is a pre-defined constant. Later investigations revealed that this model was inaccurate for near-wall modeling. Further, Fureby et al. (1997) have shown that LES is fairly insensitive to SGS models, especially in simulations with high density meshes.

In this work, the dynamic Smagorinsky model (DSM) (Germano et al., 1991; Lilly, 1992) is used to model \( \nu_{SGS} \). With DSM \( C_s \) is no longer required to have a single numerical value, but it is allowed to vary, both spatially and temporally. Following in the steps of Lilly (1992) it is possible to establish a relation for \( C_s \) based on filtered velocities. The steps are not presented here. To prevent numerical instabilities, \( C_s \) is clipped at 0 and 0.23.

2.3. SGS modeling for mass transfer
The SGS mass transport is modeled analogously with the SGS stress tensor,

\[
q_j = -\frac{\nu_{SGS} S_{ij}}{S c_{SGS}} \frac{\partial <c>}{\partial x_j} \tag{4}
\]

The rationale behind Eq. 4 is that SGS mass and momentum transport are due to the same mechanisms, i.e. sub-grid scale eddies that move fluid elements from local regions with a relatively high concentration of velocity or species concentration to regions with lower ones. \( Sc_{SGS} \) is a SGS Schmidt number. The model limitations are the same as for the SGS stress modeling, which means that it is assumed that isotropic conditions prevail on the SGS filter scales. As stated earlier, this is known not to be the case close to walls. Thus, it is necessary to increase the mesh density close to walls in order to suppress the SGS contribution to mass transfer. Since only high-Sc flows are considered, this condition implicitly means that the SGS contribution to momentum transport is suppressed to an even higher extent. In fact, the solution in the near-wall region approaches a DNS solution for velocity.

2.4. Particle tracking
Particle tracking is used to study the near-wall flow. Particles are injected at certain positions and follow the flow. The tracking is done in a Lagrangian framework, and the flow-particle interaction is one-way coupled. This means that the particles are influenced by the flow, but not vice versa. Each particle adheres to Newtons second law of motion,

\[
\frac{d u_p}{dt} = F_D(u - u_p) \tag{5}
\]

\( F_D(u - u_p) \) is the drag force, and it is given by \( F_D = 18 \mu/(\rho_p d^2) C_D Re/24 \). \( C_D = a_1 + a_2/Re + a_3/Re^2 \), where the constants \( a_1, a_2, \) and \( a_3 \) are given by Morsi & Alexander (1972). \( Re \) is the particle Reynolds number based on the slip velocity.

The drag law does only take into account the effect due to the resolved velocities. If SGS contributions to particle velocity is to be included, a random movement of the particle must be
added. However, as discussed previously, the mesh density close to the wall should be close to DNS, and the intention is that SGS contributions to particle trajectories should be negligible. Thus, they are not included in the model. These arguments are also supported by the findings in Armenio et al. (1999) that state that the particles are quite insensible to the small-scale velocity field in a well resolved LES.

3. Computational domain and setup

The method is validated on a 20 cm long tube with diameter 3.2 cm. The axial ends are modeled as periodic boundaries, and all other boundaries as no-slip walls. A section of the axial cross-section of the pipe has been removed, see Figure 1, so that the bottom boundary becomes a flat surface. The reason is that this resembles the geometry available for later experimental comparison. Close to the entrance of the pipe, a small area (1 × 1 cm) of the flat bottom has been marked, see Figure 2. This corresponds to a membrane in the experimental setup. The species studied in this paper are introduced to the flow through this area.

The fluid is incompressible water at 25 °C, and the flow is driven by a prescribed mass flow which gives an average velocity of 0.5 m/s. The Reynolds number based on the pipe diameter is 16,000.

The mesh consists of tetrahedrons with hexahedral elements to resolve the boundary layer close to the wall. Above the membrane area, the first cell center is centered at $y^+ = 1/40$. The span wise spacing is 1 wu and the stream wise spacing is 5 wu. The total number of cells is about 5 millions.

Initially, the simulation was run until the initial conditions were no longer important before any data was sampled.

4. Method validation and results

4.1. Species transport

The presented method builds on the assumption that the SGS transport of momentum and mass always are negligible in the region of interest. This means that only resolved and diffusional transport mechanisms are allowed to be important. Validation of the method thus has to focus on these aspects.

In Figure 3, the root-mean-square (rms) of the resolved velocities are displayed. As can be seen, the rms-x and rms-z curves show the theoretically expected asymptotic behavior ($y^1$). The rms-y curve shows a somewhat weaker dependence on y than expected ($y^2$). The reason behind
Figure 3. Root-mean-squares (rms) of the resolved velocities fluctuations [m/s].

Figure 4. The SGS viscosity ratio \( \nu_{SGS}/\nu \).

...this is probably that \( v_y \) is numerically much smaller than \( v_x \) and \( v_z \). The existence of an SGS part, yet a very small one, is thus more evident in the rms-y component than in the other two.

Figure 4 shows that the SGS viscosity ratio is approximately \( 10^{-4} \) at \( y^+ = 1 \). The ratio decreases in the wall direction. This means that the SGS momentum transport can be safely neglected in the entire near-wall region.

To extend the arguments to mass transport, a molecular Schmidt number must be introduced. \( \text{Sc} > 1 \) means that \( D < \nu \) and the mass transport due to molecular diffusion is reduced compared to the mass transport due to SGS diffusivity, which is fairly independent on Sc (see also below). As Sc is being increased, eventually it will no longer be possible to neglect the SGS mass transport. We have then reached the limit of the applicability of the method on the given mesh. To continue further, the mesh has to be refined. On the present mesh, a Schmidt number of 1,000 means that the SGS mass transport is approximately 10% of the transport by molecular diffusion at \( y^+ = 1 \). At \( y^+ = 0.5 \) the SGS mass transport is less than 1% of the molecular diffusion for the same Schmidt number. Taking into account that the wall-normal SGS transport is approximately two orders of magnitude smaller than the other two components, this means that the SGS transport acting in the wall-normal direction is approximately 1% of the molecular transport at \( y^+ = 1 \), and 0.1% at \( y^+ = 0.1 \).

The above arguments assume that the SGS Schmidt number equals unity. It has previously been demonstrated (Bergant & Tiselj, 2007) that the turbulent Prandtl number increases as the...
wall is approached, and in particular for high-Pr fluids. For Pr = 500, Prt = 8 at y+ = 0.1. It is difficult to draw any certain conclusion to what effect these findings have on SGS Schmidt number in this work. However, it should be recognized that Sc
SGS can divert from unity.

4.2. Particle trajectories
The method presented here can also be used with particle tracking to visualize the near-wall flow. In the absence of any molecular diffusivity, it suffices to show that the SGS contribution to particle transport is negligible compared to the resolved velocity contribution.

Individual particles are influenced only by the drag force, see Eq. 5. However, the tracked particles are very small (d_p = 0.1 micron) and have the same density as the fluid. Thus, they tend to follow the resolved velocity exactly. To determine the relative importance of the SGS velocities, proper velocity scales must be established for the SGS and resolved velocities.

Removing the axial mean velocity, the particle velocity can be expected to be large in regions with a high degree of fluctuating velocities. This implies that a relevant scale (v
res) for the resolved velocities is the rms of the resolved velocity fluctuations. Jaffrzic & Breuer (2008) suggest the following relation for the SGS viscosity based on SGS kinetic energy k
SGS.

\[ \nu_{SGS} = C_\mu k_{SGS}^{1/2} \Delta \]  

C_\mu = 0.048, and \Delta is taken from Figure 5. Since \nu_{SGS} is already known from the simulations, Eq. 6 can be used to isolate k
SGS. The square root of k
SGS gives the velocity scale v
SGS. If v
SGS \ll v
res the SGS contribution to particle motion can be neglected.

As can be concluded from Figure 4 and 5, v
SGS = 4.2 \times 10^{-2} \text{ mm/s at } y^+ = 1, \text{ and } 2.2 \times 10^{-4} \text{ mm/s at } y^+ = 0.1. \text{ These values are to be compared to } v
res = 10 \text{ mm/s at } y^+ = 1, \text{ and } 1 \text{ mm/s at } y^+ = 0.1, \text{ see Figure 3.}

4.3. Results
Only two examples of possible outcomes from the method is presented here. In Figure 6, the instantaneous species concentration at y^+ = 0.5 is depicted, together with spanwise velocity vectors. The high-concentration streaks in the streamwise direction are due to turbulent

Figure 5. Filter length scale (solid line) [m], and test filter length scale (dashed line) [m].
Figure 6. Instantaneous surface concentration on the diffusion area at $y^+ = 0.5$ and span wise velocity vectors colored by vorticity. The Schmidt number is 100. The diffusion area is $1 \times 1$ cm and the velocity vectors extends 3 mm ($y^+ \approx 75$) out from the wall.

structures that have been resolved by the mesh. The SGS contribution to mass transport has shown to be negligible and is not included in the simulation. Figure 7 shows the trajectory and wall-normal velocity of a particle released near the leading edge of the membrane at $y^+ = 0.6$. The distance to the wall is fairly constant until the particle suddenly accelerates away from the near-wall region.

References

Abba, A., Cercignani, A.C. & Valdettaro, L. 2003 Analysis of subgrid scale models. *Comput. Math. Appl.* 46 (4), 521–535.

Armenio, V., Piomelli, U. & Fiorotto, V. 1999 Effect on the subgrid scales on particle motion. *Phys. Fluids* 11 (10), 3030–3042.

Bergant, R. & Tiselj, I. 2007 Near-wall passive scalar transport at high prandtl numbers. *Phys. Fluids* 19.

Calmet, I. & Magnaudet, J. 1997 Large-eddy simulation of high-schmidt number mass transfer in a turbulent channel flow. *Phys. Fluids* 9 (2), 438–455.

Chumakov, S.G. 2008 A priori study of subgrid-scale flux of a passive scalar in isotropic homogeneous turbulence. *Phys. Rev. E* 78.

Fox, R.O. 2003 *Computational Models for Turbulent Reacting Flows*. Cambridge University Press.

Fureby, C., Tabor, G., Weller, H.G. & Gosman, A.D. 1997 A comparative study of subgrid scale models in homogenous isotropic turbulence. *Phys. Fluids* 9 (5), 1416–1429.

Germano, M., Piomelli, U., Moin, P. & Cabot, W.H. 1991 A dynamic subgrid-scale eddy viscosity model. *Phys. Fluids* 3, 1760–1765.

Hunt, J.C.R. & Morrison, J.F. 2000 Eddy structure in turbulent boundary layers. *Eur. J. Mech. B - Fluids* 19, 673–694.
Figure 7. Particle wall distance $w_u$ (above) as a function of time $[\text{ms}]$, and particle wall normal velocity $[1,000 \text{ m/s}]$ (below). The maximum velocity in the wall normal direction is 4.5 cm/s.

Jaffrzic, B. & Breuer, M. 2008 Application of an explicit algebraic reynolds stress model within a hybrid rans-les method. Flow Turbulence Combust 81, 415–448.

Lilly, D.K. 1992 A proposed modification of the germano subgrid-scale closure method. Phys. Fluids A 4, 633–635.

Morsi, S.A. & Alexander, A.J. 1972 An investigation of particle trajectories in two-phase flow systems. J. Fluid Mech. 55, 193–208.

Pope, S.B. 2000 Turbulent flows. Cambridge University Press.

Reynolds, A.J. 1975 The prediction of turbulent prandtl and schmidt number. Int. J. Heat Mass Transfer 18, 1055–1069.

Smagorinsky, J. 1963 General circulation experiments with the primitive equations: I. the basic equations. Mon. Monthly Rev. 91.

Spalart, P.R. 2000 Strategies for turbulence modelling and simulations. Int. J. Heat and Fluid Flow 21, 252–263.

Tavoularis, S. & Corrsin, S. 1985 Effects of shear on the turbulent diffusivity tensor. Int. J. Heat Mass Transfer 28, 265–276.

Welty, J.R., Wicks, C.E., Wilson, R.E. & Rorrer, G. 2001 Fundamentals of momentum, heat, and mass transfer. John Wiley & Sons, Inc.