DYNAMIC MODELS FOR SUBGRID-SCALE TRANSPORT AND MIXING OF REACTANTS IN ATMOSPHERIC TURBULENT REACTING FLOWS

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1. INTRODUCTION

Large-eddy simulation (LES) can provide valuable highresolution spatial and temporal information required to study the effects of turbulent transport and mixing on the chemical transformation of reacting scalars in the atmospheric boundary layer (ABL). LES consists of explicitly resolving all scales of turbulent transport larger than the grid scale (on the order of tens of meters in the ABL), while the smallest (less energetic) scales are parameterized using a subgrid-scale (SGS) model.

Numerical studies on the effect of turbulence on chemistry in the ABL using LES have been mostly restricted to moderately fast reacting flows involving a second-order reaction between a pollutant emitted at the surface and one entrained from the free troposphere (e.g., Schuman, 1989; Sykes et al., 1994; Molemaker and Vilà-Guerau de Arellano, 1998; Vinuesa and Vilà-Guerau de Arellano, 2003). In these studies, it is assumed that chemical species are perfectly mixed within the grid volume of the LES. However, this assumption may not be always satisfied, especially for highly reactive compounds. Thus, large-eddy simulations with realistic atmospheric chemical schemes may require a SGS model for the subgrid reactant covariance.

In this study, we develop and test a new SGS model for the subgrid-scale reactant covariance. The model is a scale-dependent version of the dynamic scale-similarity model recently introduced by Vinuesa and Porté-Agel (2005). It has some desirable characteristics that make it a robust candidate for LES of ABL reacting flows: (*a*) the model uses a dynamic procedure to calculate the value of the coefficient based on the dynamics of the resolved scales, thus not requiring any parameter tuning; and (*b*) it accounts for scale dependence that can potentially be associated with the effect of chemistry on the reactant covariances.

2. SUBGRID-SCALE MODELING FOR THE REACTANT COVARIANCE

In large-eddy simulation, the filtered conservation equation for a reacting scalar involved in a second-order reaction is

$$\frac{\partial \widetilde{A}}{\partial t} + \widetilde{u}_i \frac{\partial \widetilde{A}}{\partial x_i} = -\frac{\partial Q_{A,i}}{\partial x_i} - k\left(\widetilde{A}\widetilde{B} + \chi\right)$$
(1)

where \widetilde{A} is the spatially filtered (at scale Δ) concentration of the reactant A, which reacts following

$$A + B \xrightarrow{k} Product \tag{2}$$

The effect of the unresolved scales on the evolution of the filtered scalar concentration appears through the subgrid-scale (SGS) flux $Q_{A,i}$ and the subgrid-scale reactant covariance χ . The SGS flux is defined as

$$Q_{A,i} = u_i A - \widetilde{u}_i \widetilde{A},\tag{3}$$

and the SGS reactant covariance χ that accounts for the mixing of the reactant at subgrid scales is

$$\chi = \overline{AB} - \overline{AB}.$$
 (4)

2.1 The dynamic similarity model

By assuming scale similarity between resolved and subgrid scales, the subgrid covariance χ can be modeled as proportional to the resolved covariance at larger scales (typically between Δ and 2Δ). The subgrid covariance can then be expressed as

$$\chi = C_{sim}^{\Delta} \left(\overline{\widetilde{A}} \overline{\widetilde{B}} - \overline{\widetilde{A}} \, \overline{\widetilde{B}} \right), \tag{5}$$

where the overbar represents spatial filtering at scale 2Δ and C_{sim}^{Δ} is the similarity coefficient.

Since this coefficient may depend on the chemical regime, and to avoid tuning or a-priori specification, it is evaluated directly from the resolved scales by using a dynamic procedure. For the scalar covariance, the dynamic procedure is based on the Germano identity

$$\Upsilon = \Xi - \overline{\chi} = \overline{\widetilde{A}\widetilde{B}} - \overline{\widetilde{A}}\,\overline{\widetilde{B}},\tag{6}$$

where Υ is a resolved covariance that can be determined using the resolved scales. $\Xi = \widetilde{AB} - \widetilde{A} \widetilde{B}$ is the subgrid covariance at a test-filter scale (2 Δ). Using the similarity model, Ξ can be expressed as

$$\Xi = C_{sim}^{2\Delta} \left(\widehat{\widetilde{A} \widetilde{B}} - \widehat{\widetilde{A}} \widehat{\widetilde{B}} \right), \tag{7}$$

where the hat represents the filtering procedure applied at scale 4Δ .

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Substitution of (5) and (7) into (6) leads to the system

$$\Upsilon = C_{sim}^{\Delta} X, \tag{8}$$

where

$$X = \frac{C_{sim}^{2\Delta}}{C_{sim}^{\Delta}} \left(\widehat{\widetilde{A} \, \widetilde{B}} - \widehat{\widetilde{A}} \, \widehat{\widetilde{B}} \right) - \overline{\left(\widetilde{\widetilde{A} \, \widetilde{B}} - \overline{\widetilde{A}} \, \widehat{\widetilde{B}} \right)}.$$
(9)

It is important to note that the original dynamic model assumes scale invariance of the model coefficient at the filter and test filter scales, i.e.,

$$C_{sim}^{\Delta} = C_{sim}^{2\Delta} = C_{sim}.$$
 (10)

Minimizing the error associated with the use of the similarity model in (6) over all three vector components results in

$$C_{sim}^{\Delta} = \frac{\langle \Upsilon X \rangle}{\langle X X \rangle},\tag{11}$$

where the brackets $\langle \rangle$ represent the averaging operation. At every time step and vertical position in the flow, the similarity coefficient is computed based on the resolved concentration using (11). The SGS covariance χ is then calculated from (5) and used to obtain the total chemical term as shown in (1).

2.2 The scale-dependent dynamic similarity model

Without assuming that $C_{sim}^{2\Delta} = C_{sim}^{\Delta}$ we can still apply the dynamic model given by (6), (9) and (11). Note that this change introduces a new unknown $\beta \equiv C_{sim}^{2\Delta}/C_{sim}^{\Delta}$. For scale-invariant situations, $\beta = 1$. In order to compute C_{sim}^{Δ} using (11) we need to estimate β . A dynamic value for β can be obtained using a second test-filter at scale $\hat{\Delta} > \overline{\Delta}$. For simplicity, and without loss of generality, we take $\hat{\Delta} = 4\Delta$, and denote variables filtered at scale 4Δ by a caret (^). Writing the Germano identity between scales Δ and 4Δ yields

 $\Upsilon' = C_{sim}^{\Delta} X', \tag{12}$

where

$$\Upsilon' = \widetilde{\widetilde{A}}\widetilde{\widetilde{B}} - \widetilde{\widetilde{A}}\widetilde{\widetilde{B}}$$
(13)

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$$X' = \left[\frac{C_{sim}^{4\Delta}}{C_{sim}^{\Delta}} \left(\stackrel{?}{\widetilde{A}} \stackrel{?}{\widetilde{B}} - \stackrel{?}{\widetilde{A}} \stackrel{?}{\widetilde{B}} \stackrel{?}{\widetilde{B}} \right) - \left(\stackrel{?}{\widetilde{A}} \stackrel{?}{\widetilde{B}} - \stackrel{?}{\widetilde{A}} \stackrel{?}{\widetilde{B}} \right) \right],$$
(14)

where the overbrace (\frown) denotes spatial filtering at scale $8\Delta.$

Again minimizing the error as in Section 2.1 yields, besides (11), another equation for C_{sim}^{Δ} :

$$C_{sim}^{\Delta} = \frac{\langle \Upsilon' X' \rangle}{\langle X' X' \rangle},\tag{15}$$

Setting (11) equal to (15) yields

$$\langle \Upsilon X \rangle \langle X' X' \rangle - \langle \Upsilon' X' \rangle \langle X X \rangle = 0.$$
 (16)

which has two unknowns, $\beta = C_{sim}^{2\Delta}/C_{sim}^{\Delta}$ and $\gamma = C_{sim}^{4\Delta}/C_{sim}^{\Delta}$. In order to close the system, a relationship between β and γ is required. Thus, a functional form of the scale dependence of the coefficient

needs to be postulated. As in Porté-Agel et al. (2000), we assume a power law of the form $C_{sim} \sim \Delta^{\alpha}$. For such a power-law behavior, β does not depend on scale and is equal to $\beta = 2^{\phi}$. Note that this assumption is much weaker than the standard dynamic model, which corresponds to the special case $\phi = 0$. We stress that one does not need to assume the power-law to hold over a wide range of scales, but only between scales Δ and 4Δ . A consequence of the assumed local powerlaw is that $C_{sim}^{2\Delta}/C_{sim}^{\Delta} = C_{sim}^{4\Delta}/C_{sim}^{2\Delta} = \beta$, and thus $\gamma = C_{sim}^{4\Delta}/C_{sim}^{\Delta} = \beta^2$. With this substitution (16) only contains the unknown β , and can be rewritten as a fifth order polynomial on β . One can show that only the largest root is physically viable. A Newton-Raphson method is used to find that root. Once β has been computed, it is used in (14) to compute X' which in turn is used in (15) to obtain C_{sim}^{Δ} .

3. NUMERICAL EXPERIMENTS

The large-eddy simulation code is a modified version of the code described by Albertson and Parlange (1999), Porté-Agel et al. (2000), and Vinuesa and Porté-Agel (2005). The code uses a mixed pseudospectral finite-difference method and the subgrid-scale stresses and fluxes are parameterized with scale-dependent dynamic models. We simulate the same dry convective atmospheric boundary layer described in Vinuesa and Porté-Agel (2005). A uniform sensible heat flux of $0.05 \ K.m.s^{-1}$ is prescribed at the surface. The initial potential temperature has a constant value of 311 K throughout the boundary layer, with an overlying inversion of strength $0.006 \ K.m^{-1}$ above 700 meters. The size of the computational domain is $(2\pi L_z, 2\pi L_z, L_z)$ with $L_z = 1500$ m and it is divided into $N \times N \times N$ uniformly spaced grid points. Three resolutions are used, corresponding to N equal to 32, 64, and 96. Periodic lateral boundary conditions are assumed. The maximum time step used in the calculation is $0.5 \ s$ and the simulations cover a 1.5 hours period. Averaged over the last hour, the convective velocity scale w_* and the friction velocity u_* are equal to $1.07 ms^{-1}$ and $0.3 ms^{-1}$, respectively. The stability parameter z_i/L is equal to -41.

The chemical set-ups are based on a second-order irreversible reaction (2). The reactant *A* is uniformly emitted at the surface with a flux of 0.25 $ppb \ m \ s^{-1}$ and no initial concentration in the CBL. *B* is not emitted and its initial profile has a constant concentration of 2 ppb both in the CBL and in the free troposphere. Two values of the rate coefficient *k* are considered: $2.1 \ x \ 10^{-2} \ ppb^{-1}s^{-1}$ and $4.75 \ x \ 10^{-3} \ ppb^{-1}s^{-1}$. In the following, these experiments are referred to as fast (or F) and slow (or S), respectively. Statistics of the simulated fields are obtained by averaging over the last hour for the S chemical set-up and over the second half-hour for the F one.

4. RESULTS

4.1 Simulations without SGS covariance model

Figures 1a and 1b show the vertical profiles of the mean concentration of the reactant A for the three different resolutions $(32^3, 64^3 \text{ and } 96^3)$ and the two chemical setups, S and F, respectively. In these experiments, no model for the SGS covariance in the chemical term is used, i.e., $\chi = 0$. For the F chemical set-up, important differences are found in the simulated concentrations obtained with the three resolutions. In particular, the concentration for the $F - 32^3$ case can be as small as 50% compared with the concentration in the $F - 96^3$ case. In the experiments with a number of points lower than 96^3 for F and 64^3 for S, the reactants are not uniformly mixed at the subgrid scales and, consequently, the assumption of perfect mixing at those scales is not valid. In these cases, ignoring the sub-grid covariance associated with heterogeneous mixing at the unresolved scales, leads to an overestimation of the chemical term in equation (1). As a result, the reactant A is depleted too fast and important differences are found in its concentration with respect to the highest resolution experiment.

The vertical profiles of the subgrid-scale Damköhler number Da_{sgs} , defined as the ratio between the characteristic time scale associated with the smallest resolved eddy motions and the chemical time scale, are shown in Figures 2a and 2b. For the S chemical set-up, all experiments show Dasgs smaller than one, indicating that the time scale of chemistry is larger than the one associated with the smallest resolved turbulent eddies. As a result, the reactants are well mixed at the subgrid scales and the unresolved scales do not affect the chemical transformations. For the faster (F) chemical set-up, the calculated Da_{sqs} values are larger than one for $F - 32^3$, and they are $\sim O(1)$ for $F - 64^3$. For these two lowest resolutions, segregation of the reactants at the subgrid scales is expected to affect the chemical transformation. This effect, which is stronger for the lower resolution case (with larger Da_{sqs}), is expected to reduce the amount of chemical transformation of the reactants. Since the simulations do not account for the effect of reactant segregation at the subgrid scales, the depletion rate of the reactant A is overestimated and, as a result, the concentration of A is underestimated. This is in qualitative agreement with the results shown in Figure 1.

4.2 Simulations with SGS covariance model

Next, results from simulations with the dynamic similarity model for the subgrid-scale covariance (section 2.1) and its scale-dependent version (section 2.2) are presented and compared with the results obtained without subgrid-scale model. In Figure 3, the simulated concentration profiles obtained using the two subgrid-scale models for the reactant covariance (i.e., $\chi \neq 0$) are plotted for the 32^3 resolution. The agreement with the 96^3 experiments is largely improved by using the similarity models. It is clear that by including a model for the non-homogeneous



FIG. 1: Vertical profiles of the non-dimensional bottom-up reactant concentration for the S (a) and F (b) chemical set-ups. Three resolutions are considered: 96^3 (solid line), 64^3 (triangles), and 32^3 (diamonds). The values are made dimensionless using a concentration scale defined as the ratio between the surface flux of *A* and the convection velocity.

mixing at the subgrid scales, LES is able to incorporate, at least partially, the effect of the unresolved scales on the chemical transformations. Moreover, accounting for scale-dependence in the dynamic model further improves the results.

In order to quantify the effect of the subgrid scales on the reactant mixing and, in turn, on the overall reaction rate, one can define an effective reaction rate k_{eff} as

$$k_{eff} = k \left(1 + \frac{\chi}{\widetilde{A}\widetilde{B}} \right), \tag{17}$$

where χ is determined at each timestep by using the subgrid scale models for the reactant covariance. This procedure is equivalent to using

$$-k\left(\widetilde{A}\widetilde{B}+\chi\right) = -k\left(1+\frac{\chi}{\widetilde{A}\widetilde{B}}\right)\widetilde{A}\widetilde{B} = -k_{eff}\widetilde{A}\widetilde{B} \quad (18)$$

as the chemical term in the filtered equation solved in LES for the reacting scalars (1).



FIG. 2: Vertical profiles of the subgrid scale Damköhler number for the S chemical set-up (a) and for the F one (b). Three resolutions are considered: 96^3 (solid line), 64^3 (triangles), and 32^3 (diamonds).

In Figure 4, the vertical profiles of the normalized effective reation rate k_{eff}/k are presented. This ratio is smaller than one due to the fact that the inhomogeneous mixing at the subgrid scales yields a non-zero negative reactant covariance χ . From (17), this produces an effective reaction rate k_{eff} which is smaller than the reaction rate obtained under perfect-mixing laboratory conditions, k. In all cases, k_{eff}/k is closer to one away from the surface and it decreases to smaller values near the surface, indicating that the relative contribution of the SGS chemical term χ to the total chemical transformation becomes larger as the surface is approached. As expected, for all heights k_{eff}/k is also affected by resolution and chemical regime. In particular, the ratio becomes smaller with decreasing resolution and increasing value of k which are associated with a reduction of the relative importance of the subgrid-scale chemical transformations.

4.3 Subgrid-scale model for the reactant fluxes

The above results focus on the effects of non-uniform mixing of the reactants at the subgrid scales on the simulated chemical transformations. However, the evolution of the reactant concentration can also be affected by the subgrid scales through the the subgrid-scale fluxes in (1). The subgrid-scale fluxes in the simulations are parame-



FIG. 3: Vertical profiles of the non-dimensional reactant concentration for the 32^3 resolution simulations for the S chemical set-up (a) and F chemical set-up (b). The solid lines correspond to the 96^3 simulations shown in Figure 1.

terized using eddy-diffusion subgrid-scale models. For the scalar ${\cal A}$ this model is of the form

$$\widetilde{u_i A} - \widetilde{u}_i \widetilde{A} = -\Delta^2 \left[S c_{sgs,A}^{-1} C_S^2(\Delta) \right] \left| \widetilde{S} \right| \frac{\partial A}{\partial x_i}.$$
 (19)

where $|\widetilde{S}| = (2\widetilde{S}_{ij}\widetilde{S}_{ij})^{1/2}$ is the resolved strain-rate magnitude, and \tilde{S}_{ij} is the resolved strain rate tensor. C_S and $Sc_{sgs,A}$ are the Smagorinsky coefficient in the eddyviscosity model and the subgrid-scale Schmidt number for the scalar A, respectively. Usually the subgrid-scale Schmidt number $Sc_{sgs,A}$ and, as a result, the lumped coefficient $Sc_{sas,A}^{-1}C_S^2$ are only determined (calculated or prescribed) for inert scalars, and those values are used for the calculation of the subgrid scale reactant fluxes using (19). It is, therefore, assumed that chemistry does not affect the subgrid scale fluxes and, consequently, $Sc_{sqs,A} = Sc_{sqs}$, where Sc_{sqs} is the subgridscale Schmidt number for an inert scalar, usually potential temperature. Following that approach, in the above simulations, a scale-dependent dynamic model for the SGS heat flux was implemented and the same coefficient used to calculate the reactant scalar fluxes. This scaledependent dynamic SGS scalar flux model has been shown to yield accurate results in the case of inert scalars (Porté-Agel, 2004). However, in a recent study of a re-



FIG. 4: Vertical profiles of the non-dimensional effective reaction rate coefficient calculated by the dynamic similarity model (a and c) and by the scale-dependent dynamic similarity model (b and d). The values are non-dimensionalized using the reaction rate coefficient determined in the laboratory with perfect mixing of the reactants.

acting scalar involved in a first-order reaction, Vinuesa et al. (2006) showed that explicitely calculating $Sc_{sgs,A}$ using a separate scale dependent dynamic procedure allows to account for the effect of the chemical reactivity on the value of the coefficient, which improves the simulation results when $Da_{sgs} \sim O(1)$ or larger. Here, we have tested the impact of calculating both $Sc_{sgs,A}^{-1}C_S^2$ and $Sc_{sgs,B}^{-1}C_S^2$ using separate scale-dependent dynamic models on the results presented above.

Figure 5b shows the simulated concentrations obtained using separate scale-dependent dynamic models for the reacting scalar fluxes. For comparison, also shown are results obtained with the heat flux eddy diffusivity in the reactant subgrid flux model (Figure 5a). In general, all the results, with and without subgrid-scale covariance models, are improved by using the separate subgrid-scale flux model. It is important to note that using the scale-dependent dynamic similarity model for the reactant covariance together with the scale-dependent dynamic SGS flux model leads to resolution-independent concentration profiles, even for the most challenging case, i.e. the $F - 32^3$ case corresponding to the lowest resolution and the fastest chemical regime.

5. SUMMARY

Numerical experiments were carried out to study the effects of assuming instantaneous and homogeneous mixing of reacting scalars at the subgrid scales in large eddy simulation of reacting atmospheric boundary layer flows. Using a simple chemical mechanism with two reactants transported in opposite directions, the effect of the unre-



FIG. 5: Vertical profiles of the non-dimensional reactant concentration for the $F - 32^3$ simulations not using (a) and using (b) separate scale-dependent dynamic models for reactant scalar fluxes.

solved scales on the chemical transformation is analysed. The relative magnitude of this effect is found to be well characterized by the subgrid scale Damköhler number Da_{sqs} , which is defined as the ratio between the characteristic time scale associated with the smallest resolved eddy motions and the chemical time scale. In particular, the subgrid scales are found to affect the chemical transformations when $Da_{sgs} \sim O(1)$ or greater. The effect is stronger for larger Dasgs associated with reduced resolution and/or increased chemical reactivity. When this effect is not accounted for in the simulations, unrealistic differences between the simulation results obtained at different resolutions are found. These differences can be explained considering that the reactants are segregated at the subgrid scales, leading to a reduction in the effective reaction rate. Neglecting these effects can lead to substantial overestimations of the reactant depletion rates and, as a result, underestimations of their concentrations.

Two models for the subgrid-scale reactant covariance χ , i.e. quantity that accounts for the inhomogeneous mixing and segregation of reactants at subgrid scales, are presented and tested, and the results compared with the ones obtained without SGS covariance model. The first model is the dynamic scale-similarity model introduced by Vinuesa and Porté-Agel (2005), which assumes scale invariance of the model coefficient. The second model is a modified version that allows for possible scale dependence of the model coefficient. Both models are free from parameter tuning since the model coefficients are computed as the simulation progresses using dynamic procedures based on the information contained in the resolved scales. Simulation results show that both models are able to account in part for the effect of the segregation of the scalars at the subgrid scales, considerably reducing the resolution dependence of the results found when no subgrid covariance model is used. The scale-dependent dynamic version yields better results than its scale-invariant counterpart.

Finally, another potential effect of the subgrid scales on the evolution of the reactant concentration is studied: the subgrid-scale model for the reactant fluxes. A scale-dependent dynamic model for the SGS flux is used, which has been shown to give accurate simulation results for inert scalars. Two scenarios are considered: one in which the model is applied using the same eddy diffusivity obtained for temperature, a common practise in this type of simulations; and a second scenario in which a separate dynamic model is used to calculate a different eddy diffusivity for the reacting scalars. Our results show important differences between the two cases when $Da_{sqs} \sim O(1)$ or greater. It is interesting to note that simulations that use both the scale-dependent dynamic model for the subgrid-scale covariance and the one for the subgrid-scale flux yield simulations show virtually no resolution dependence even for relatively fast chemical regimes, which highlights the potential of these tuningfree models to capture the effect of the subgrid scales.

Future work will extend the implementation of the proposed scale-dependent dynamic models for subgridscale reactant covariances and fluxes to complex chemical mechanisms involving several reactants. In that case, increased computational cost associated with solving additional transport equations for all the reactant is likely to limit the spatial resolutions of the LES, placing additional burden on the subgrid-scale models. Dynamic subgridscale models like the ones presented in this paper are expected to provide tuning-free, more reliable simulations of those flows.

Acknowledgments

This research was supported by NSF (grants EAR-0537856 and EAR-0120914 as part of the National Center for Earth-surface Dynamics). Computer resources were provided by the Minnesota Supercomputing Institute.

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