

Jean-François Vinuesa\*, Fernando Porté-Agel  
Saint Anthony Falls Laboratory, University of Minnesota, Minneapolis, United States of America

## 1. INTRODUCTION

Large-eddy simulation (LES) is becoming an important tool for the study of turbulent transport and mixing of reacting chemical species in the convective boundary layer (CBL). The scales associated with turbulent motions in the boundary layer range from the Kolmogorov dissipation scale (on the order of a millimeter) to the boundary layer depth (on the order of a kilometer). The largest eddies are responsible for the turbulent transport of scalars and momentum whereas the smallest ones are mainly dissipative. 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 CBL 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 (Schuman, 1989; Sykes *et al.*, 1994; Molemaker and Vilà-Guerau de Arellano, 1998; Petersen, 2000; 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. Thus, the heterogeneous distribution of reactants at the subgrid scales is neglected. Such an assumption is only justified when the chemistry is slow with respect to the subgrid mixing. This condition may not be always satisfied, especially for highly reactive compounds such as  $OH$  or  $HO_2$ . For instance, Krol *et al.* (2000) noticed that some reactants, such as  $OH$  when reacting with  $RH$  or with  $NO_2$ , can be classified within the fast chemical regime. Therefore, large-eddy simulations with realistic atmospheric chemical schemes may require a SGS model for the subgrid reactant covariance.

Following the analysis of Schumann (1989), the subgrid effects of the chemical terms are usually omitted. However, Meeder and Nieuwstadt (2000) proposed to compute the subgrid covariance by solving an extra equation for this quantity and using closure models for the third order moments. In this paper, we present a subgrid model for the reactant covariance based on the scale similarity approach. We introduce a dynamic procedure to calculate the model coefficient based on information contained in the resolved scales. Such a procedure does not require any parameter specification or tuning. That is of

\* Corresponding author address: Jean-François Vinuesa, St. Anthony Falls Laboratory, Mississippi River at 3rd Avenue SE, Minneapolis, MN 55414, USA; e-mail: [vinue001@umn.edu](mailto:vinue001@umn.edu).

great interest since the model coefficient may depend on the chemical regime, i.e. on how fast the chemistry is with respect to the time-scale of the smallest resolved eddies.

## 2. THE DYNAMIC SIMILARITY MODEL

### 2.1 Model formulation

The filtered governing equation for the concentration of a reacting scalar that is solved in LES reads

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

where  $\tilde{A}$  is the spatially filtered (at scale  $\Delta$ ) concentration of the reactant  $A$ , here involved in the second-order reaction  $A + B \xrightarrow{k} Product$ . The effect of the unresolved scales on the evolution of the filtered scalar concentration appears through the subgrid-scale flux  $Q_{A,i}$  and the subgrid-scale reactant covariance  $\chi$ .

The SGS flux is defined as

$$Q_{A,i} = \widetilde{u_i A} - \tilde{u}_i \tilde{A}, \quad (2)$$

and the SGS reactant covariance  $\chi$  that accounts for the mixing of the reactant at subgrid scales is

$$\chi = \widetilde{AB} - \tilde{A}\tilde{B}. \quad (3)$$

Subgrid-scale models originally designed for potential temperature or moisture are currently used to parameterize the subgrid reactant flux whereas  $\chi$  is usually neglected.

Next, similarity arguments are used to derive an expression for the sub-grid covariance of reactants  $\chi$ . Similarity models assume that the structure of turbulent eddies at scales smaller than  $\Delta$  is similar to that at resolved scales slightly larger than  $\Delta$ . By assuming similarity between resolved and subgrid scales, the subgrid covariance is set to be proportional to the resolved covariance at slightly larger scales (typically between  $\Delta$  and  $2\Delta$ ). The subgrid covariance can then be expressed as

$$\chi = C_{sim}^{\Delta} (\overline{\widetilde{AB}} - \overline{\tilde{A}\tilde{B}}), \quad (4)$$

where the overline represents spatial filtering at scale  $2\Delta$  and  $C_{sim}^{\Delta}$  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 the so-called dynamic procedure. For the scalar covariance, the dynamic procedure is based on the identity

$$\Upsilon = \Xi - \bar{\chi} = \overline{\widetilde{AB}} - \overline{\tilde{A}\tilde{B}} \quad (5)$$

where  $\Upsilon$  is a resolved covariance that can be determined using the resolved scales.  $\Xi = \overline{AB} - \overline{A}\overline{B}$  is the subgrid covariance at a test-filter scale generally equal to twice  $\Delta$ . Using the similarity model to compute  $\Xi$  and substituting (4) in (5) gives

$$\Upsilon = C_{sim}^{2\Delta} \left( \widehat{\overline{AB}} - \widehat{\overline{A}\overline{B}} \right) - \overline{C_{sim}^{\Delta} \left( \overline{AB} - \overline{A}\overline{B} \right)}, \quad (6)$$

where the hat represents for the filtering procedure applied at scale  $4\Delta$ . Assuming scale invariance of the model coefficient, i.e.,  $C_{sim}^{2\Delta} = C_{sim}^{\Delta}$  gives

$$\Upsilon = C_{sim}^{\Delta} X, \quad (7)$$

with

$$X = \widehat{\overline{AB}} - \widehat{\overline{A}\overline{B}} - \overline{\left( \overline{AB} - \overline{A}\overline{B} \right)}. \quad (8)$$

Minimizing the error associated with the use of the similarity model in (5) results in

$$C_{sim}^{\Delta} = \frac{\langle \Upsilon X \rangle}{\langle X X \rangle}, \quad (9)$$

where the brackets  $\langle \rangle$  represent averaging over horizontal planes. At every time step and vertical position in the flow, the similarity coefficient is computed based on the resolved concentration using (9). The SGS covariance  $\chi$  is then calculated from (4) and used to obtain the total chemical term as shown in (1). It is important to note that this procedure is equivalent to use

$$-k \left( \widetilde{AB} + \chi \right) = -k \left( 1 + \frac{\chi}{\widetilde{AB}} \right) \widetilde{AB} = -k_{eff} \widetilde{AB} \quad (10)$$

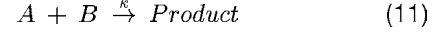
where  $k_{eff}$  is an effective reaction rate that includes the effect of the subgrid covariance, computed here with the dynamic similarity model.

## 2.2 Numerical simulations

The code used is a modified version of the three-dimensional LES code described by Albertson and Parlange (1999), Porté-Agel *et al.* (2000), and Porté-Agel (2004). A chemical solver has been implemented following Vilà-Guerau de Arellano and Cuijpers (2000). We simulate a dry convective atmospheric boundary layer driven by thermal and shear forcing. We prescribe a uniform surface sensible heat flux of  $0.05 \text{ K.m.s}^{-1}$ . The initial potential temperature profile has a constant value of  $311 \text{ K}$  below 700 meters and it increases by  $0.6 \text{ K}$  each hundred meters above that height. The computational domain is of size  $(L_x, L_y, L_z)$  and it is divided into  $N \times N \times N$  uniformly spaced grid points. The height of the domain is equal to 1500 meters and  $L_x = L_y = 2\pi L_z$ . High and low resolutions (HR and LR) are used with  $N = 64$  and  $N = 16$ , respectively. A higher resolution simulation has also been performed showing no significant differences with respect to the HR experiment. Periodic lateral boundary conditions are assumed. The maximum time step used in the calculation is  $0.5 \text{ s}$ . The simulations cover a 1.5 hours period and the statistics presented here

are obtained averaging the results over the last hour. The convective velocity scale  $w_*$ , the friction velocity  $u_*$ , the CBL height  $z_i$  (defined as the height where the sensible heat flux is minimum) are equal to  $1.07 \text{ m s}^{-1}$ ,  $0.3 \text{ m s}^{-1}$ , and  $775 \text{ m}$ , respectively.

Two reactants are involved in the irreversible second-order reaction



with a rate coefficient  $k$  of  $2.1 \times 10^{-2} \text{ ppb}^{-1} \text{ s}^{-1}$ . Notice that this latter  $k$  is one order of magnitude lower than the typical reaction coefficient between  $NO$  and  $OH_2$ . The reactant  $A$ , so-called bottom-up reactant, is uniformly emitted at the surface with a flux of  $0.25 \text{ ppb m s}^{-1}$  and its initial concentration in the CBL is set at zero.  $B$  is not emitted and its initial profile shows a constant value of  $2 \text{ ppb}$  both in the CBL and in the free troposphere. Such a chemical set-up allows to clearly distinguish the effect of the SGS flux and SGS covariance on the chemistry since the entrainment of  $B$  is dynamically simulated and depends on its depletion by  $A$  following the chemical scheme (11). It is important to note that inert scalars introduced in the simulations with the same initial conditions and fluxes as  $A$  and  $B$  showed no significant dependence on spatial resolution (LR versus HR). Therefore, any difference in  $A$  and  $B$  concentrations between LR and HR simulations are expected to be due to limitations in the treatment of the chemistry.

The ratio between the turbulent and the chemical time-scales, i.e. the turbulent Damköhler number  $Da$ , is used to estimate the influence of turbulence on chemical transformations (Schuman, 1989; Sykes *et al.*, 1994 and Vilà-Guerau de Arellano, 2003). For reacting flows with  $Da \ll 1$ , the reactants are homogeneously mixed in the boundary layer. There is therefore little influence of the turbulent structures on the chemistry. The reacting flow is in a so-called slow chemistry regime. High above the threshold value of  $Da = 1$ , the flow shows a fast chemistry regime behavior. The chemistry is so active that the chemical species are almost not affected by the turbulent structure of the CBL and more particularly by the turbulent transport. Between these limiting behaviours, e.g. for moderate chemistry, the turbulent mixing is expected to have a non-negligible impact on chemical transformations. By calculating the Damköhler number for  $A$  ( $Da_A$ ), we found that the bottom-up reactant  $A$  is in a fast chemical regime ( $Da_A \approx 16$ ). In such a flow the behavior of the reactant is dominated by the chemistry through the chemical term included in the governing equation for its concentration.

## 3. RESULTS AND DISCUSSION

Figure 1 shows the vertical profiles of the mean reactant concentrations for the two different resolutions (LR and HR) from simulations that use no model for the SGS covariance in the chemical term, i.e.,  $\chi = 0$ . One can notice that these profiles show important concentration differences. These differences can be larger than 50% (see

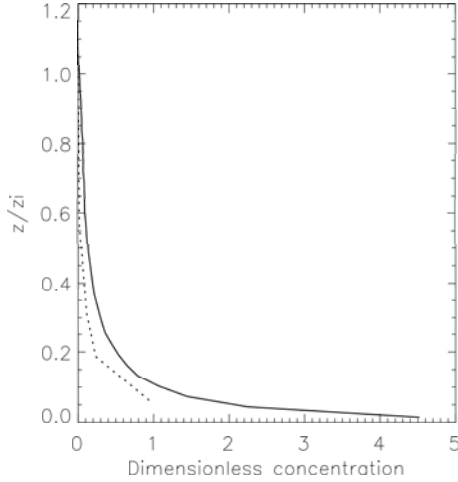


FIG. 1: Vertical profile of dimensionless bottom-up reactant mean concentration. HR and LR experiments are represented by solid and dashed lines, respectively. The values are made dimensionless by the concentration scale  $a_*$  defined as the ratio of the surface flux of  $A$  to the convection velocity scale.

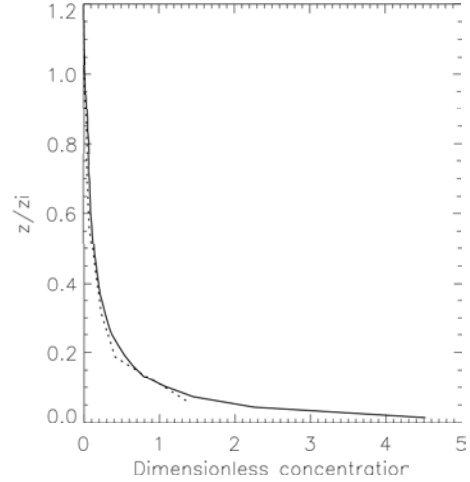


FIG. 2: Vertical profile of dimensionless bottom-up reactant mean concentration. HR and LR experiments are represented by solid and dashed lines, respectively. In the LR experiment, the dynamic similarity model is used to compute the subgrid reactant covariance  $\chi$ .

also figure 4). As mentioned earlier, the results from a higher resolution experiment (not shown here) were found to be similar to the ones reported for HR. This suggests that for this chemical set-up, the subgrid segregation between the reactants has little influence on the reactant concentration for resolutions higher than  $64 \times 64 \times 64$ .

The unresolved scales can affect the evolution of the reactant concentration through the subgrid flux and the subgrid covariance included in the chemical contribution. We noticed earlier that the reactant  $A$  was in fast chemical regime, meaning that its concentration behavior is dominated by the chemical contribution, and also that inert scalar concentrations show no dependence on resolution. As a result, the differences between the LR and HR experiments are mainly due to the chemical contribution to the reactant concentration behavior.

In the LR experiment, the reactants are not uniformly distributed at the subgrid scales and, consequently, the assumption of perfect mixing at those scales is not valid. In this case, the use of the product of the reaction rate coefficient  $k$  (determined at the laboratory under perfect mixing conditions) with the resolved concentrations as the chemical term in (1) (i.e.  $\chi = 0$ ) leads to an overestimation of the magnitude of the chemical term. As a result, the reactant  $A$  is depleted too fast and important differences are noticed with respect to the HR experiment (figure 1).

In figure 2, the profile of the mean concentration of  $A$  obtained from the HR simulation is compared with that from a low resolution simulation that uses the dynamic similarity model to calculate the subgrid reactant covariance. The profiles for both resolutions look very similar in the bulk with slightly smaller values of the reactant con-

centration near the surface for the LR case. By comparing the results in figures 1 and 2, it is clear that by including a model for the non-homogeneous mixing at subgrid scales, our LES code is able to account for the effect of the unresolved scales on the chemical transformations. We found that the dynamic similarity coefficient  $C_{sim}^{\Delta}$  is very close to 1 throughout the CBL (see the vertical profile of the dynamic similarity coefficient in figure 3).

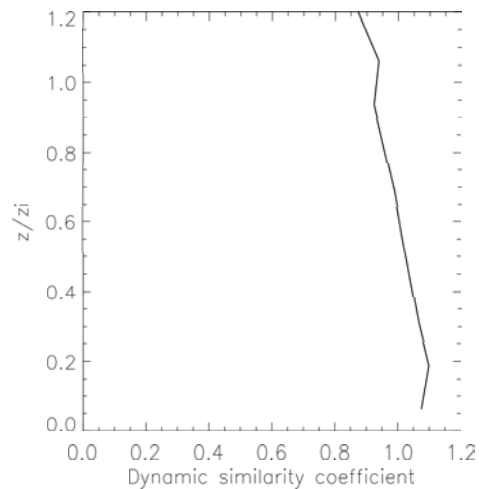


FIG. 3: Vertical profile of the dynamic similarity coefficient.

Figure 4 shows the effect of using the dynamic similarity subgrid covariance model on the normalized difference between the simulated concentrations from the HR

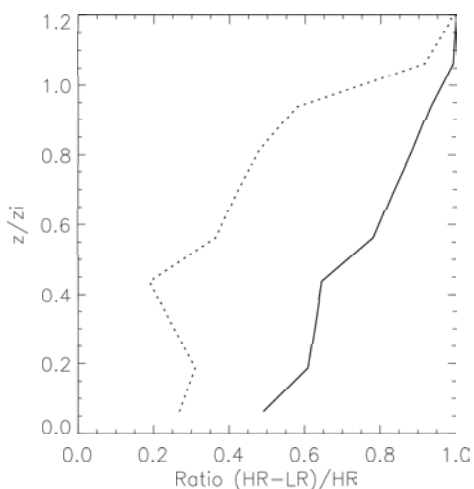


FIG. 4: Vertical profile of the difference between the concentrations of the reactant  $A$  calculated during the HR and LR experiments weighted by the HR concentrations. The two lines correspond to the LR experiments with (dashed line) and without (solid line) SGS model for the reactant covariance  $\chi$ .

and LR simulations. In this figure, the HR results have been interpolated at each LR level by using the HR results at the two nearest HR levels. Despite of the errors associated with such an averaging procedure, one can notice that the effect of the resolution on the simulation results is clearly reduced by using the dynamic similarity model.

#### 4. CONCLUSION

In this study, we have developed and implemented a similarity model for the subgrid covariance to be used in LES of turbulent reacting flows in the atmosphere. The model coefficient is computed using a dynamic procedure that is based on the resolved reactant concentrations and that does not require any parameter specification or tuning.

Our results show that even for moderately fast chemistry (reaction rate coefficient set to 10% of the typical reaction coefficient between  $NO$  and  $OH_2$ ), subgrid effects on the chemical transformations of reacting scalars are important in simulations of CBLs. We found that neglecting these effects can lead to an overestimation of the depletion rate of the reactants.

By computing the similarity coefficient at every time step and vertical position in the flow, and using it to compute the SGS reactant covariance included in the chemical term, our LES code is able to account for the effect of the unresolved scales on the chemical transformations. We define an effective reaction rate that incorporates the subgrid covariance effects and that can be used instead of the reaction coefficient obtained in the laboratory under perfect mixing conditions. Further studies will address the

variability of the effective rate coefficient with respect to the chemical regime and the treatment of subgrid reactant covariances in LES.

#### Acknowledgements

The authors gratefully acknowledge the financial support by NASA (NAG5-11801), the National Science Foundation (EAR-0094200 and EAR-0120914 as part of the National Center for Earth-Surface Dynamics) and the computer resources from the Minnesota Supercomputing Institute.

#### REFERENCES

- Albertson, J. D. and M. B. Parlange, 1999: Natural integration of scalar fluxes from complex terrain. *Adv. Water Res.*, **23**, 239-252.
- Krol, M. C., Molemaker, M. J. and J. Vilà-Guerau de Arellano, 2000: Effects of turbulence and heterogeneous emissions on photochemically active species in the convective boundary layer. *J. Geophys. Res.*, **105**, 6871-6884.
- Meeder, J. P. and F. T. M. Nieuwstadt, 2000: Large-eddy simulation of the turbulent dispersion of a reactive plume from a point source into a neutral atmospheric boundary layer. *Atmos. Env.*, **34**, 3563-3573.
- Molemaker, M. J. and J. Vilà-Guerau de Arellano, 1998: Turbulent control of chemical reactions in the convective boundary layer. *J. Atmos. Sci.*, **55**, 568-579.
- Petersen, A. C., 2000: The impact of chemistry on flux estimates in the convective boundary layer. *J. Atmos. Sci.*, **57**, 3398-3405.
- Porté-Agel, F., C. Meneveau and M. B. Parlange, 2000: A scale-dependent dynamic model for large-eddy simulation: application to a neutral atmospheric boundary layer. *J. Fluid Mech.*, **415**, 261-284.
- Porté-Agel, F., 2004: A scale dependent dynamic model for scalar transport in LES of the atmospheric boundary layer. *Boundary-Layer Meteorol.*, **112**, 81-105.
- Schumann, U., 1989: Large-eddy simulation of turbulent diffusion with chemical reactions in the convective boundary layer. *Atmos. Env.*, **23**, 1713-1729.
- Sykes, R. I., Parker, S. F., Henn, D. S. and W. S. Lewellen, 1994: Turbulent mixing with chemical reactions in the planetary boundary layer. *J. Appl. Meteorol.*, **33**, 825-834.
- Vilà-Guerau de Arellano, J. and J. W. M. Cuijpers, 2000: The chemistry of a dry cloud: the effects of radiation and turbulence. *J. Atmos. Sci.*, **57**, 1573-1584.
- Vilà-Guerau de Arellano, J., 2003: Bridging the gap between atmospheric physics and chemistry in studies of small-scale turbulence. *Bull. Amer. Meteorol. Soc.*, **84**, 51-56.
- Vinuesa, J.-F. and J. Vilà-Guerau de Arellano, 2003: Fluxes and (co-)variances of reacting scalars in the convective boundary layer. *Tellus*, **55B**, 935-949.