Effect of Turbulence on Atmospheric Chemistry

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At present usually chemical transformations in the atmosphere are calculated using average concentrations neglecting turbulent fluctuations of temperature and concentrations of chemically active species. However because of relatively coarse spatial resolution turbulence could significantly affect nonlinear chemical transformations especially in the regions of high chemical and turbulent activity in the boundary layer, convective cells, and in cloud anvils.

Accounting for an effect of turbulence on the chemical transformations requires constructing a scheme for correct estimation of the chemical turbulent term in the transport equation of the concentrations [*Villa-Guerau de Arellano*, 2003; *Verver et al.*, 1997,]. *Shnaydman et al*. [1998] and *Shnaydman* [2004] proposed to use a turbulent closure based on the system of the hydrodynamic equation with two-equation closure scheme that includes equations of turbulent kinetic energy and dissipation rate. This closure scheme allows us to predict the three-dimensional space distributions of mean and turbulence parameters with a higher accuracy than one-equation parameterizations. Turbulent mixing coefficient ranges for different type of the atmospheric flows from 1-100 m^2/s in the boundary layer to 500-1000 m^2/s in the cloud anvils and convective cells. The transport equation for N reacting species q_i

$$\frac{\partial q_i}{\partial t} + u_\alpha \frac{\partial q_i}{\partial x_\alpha} = \alpha_{ij} q_i q_j \qquad (1)$$

contains a right-hand-side non-linear term responsible for chemical transformations. α_{ij} are elements of a symmetric matrix of rates of chemical reaction between species ***i*** and ***j***. The 3-D advection term could be split into horizontal and vertical parts, respectively

$$u_\alpha \frac{\partial q_i}{\partial x_\alpha} = u_\beta \frac{\partial q_i}{\partial x_\beta} + u_3 \frac{\partial q_i}{\partial x_3}$$

We assume that summation is conducted on all repeating indexes. Indexes $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ equal to 1, 2, 3 and 1, 2, respectively, and indexes ***i*** and ***j*** change from 1 to N. Chemical rate transformations α_{ij} are functions of temperature but here for simplicity we assume that they are constant. Applying a Reynolds averaging to (1) we can get a transport equation for mean concentrations with a chemical term in the right-hand-side of the equation calculated using mean concentrations and covariances of concentration fluctuations

$$\frac{\partial \overline{q}_i}{\partial t} + \overline{u}_\alpha \frac{\partial \overline{q}_i}{\partial x_\alpha} = \frac{\partial}{\partial x_\beta} K_L \frac{\partial \overline{q}_i}{\partial x_\beta} + \frac{\partial}{\partial x_\beta} K_Z \frac{\partial \overline{q}_i}{\partial x_3} + \alpha_{ij} \overline{q}_i \overline{q}_j + \alpha_{ij} \overline{q'_i q'_j} \qquad (2)$$

The left-hand-side of (2) describes advection and turbulent diffusion of mean concentrations. The equations for covariances and variances we obtain from the equations for instant and mean concentrations using turbulent viscosity approximation for turbulent fluxes of chemical species, and for the third mixed moment of component velocity and concentration fluctuations $(\overline{u_i' q_i' q_j'})$

$$\frac{\partial}{\partial t}\overline{q_i' q_j'} + \overline{u}_\alpha \frac{\partial}{\partial x_\alpha}\overline{q_i' q_j'} = 2\left(K_L \frac{\partial \overline{q}_i}{\partial x_\beta}\frac{\partial \overline{q}_j}{\partial x_\beta} + K_3 \frac{\partial \overline{q}_i}{\partial x_3}\frac{\partial \overline{q}_j}{\partial x_3}\right)$$

$$+ \frac{\partial}{\partial x_\beta} K_L \frac{\partial}{\partial x_\beta}(\overline{q_i' q_j'}) + \frac{\partial}{\partial x_3} K_z \frac{\partial}{\partial x_3}(\overline{q_i' q_j'}) + \alpha_{ij}(\overline{q_i'^2 q_j'}) + \alpha_{ij}(\overline{q_i' q_j'^2}) +$$

$$\alpha_{ij}(\overline{q}_j \overline{q_i' q_j'} - \overline{q}_j \overline{q_i' q_j'}) + \alpha_{ij}(\overline{q_j' q_i'^2} + \overline{q_i' q_j'^2}) \qquad (3)$$

$$\frac{\partial \overline{q_i'^2}}{\partial t} + \overline{u}_\alpha \frac{\partial \overline{q_i'^2}}{\partial \partial x_\alpha} = 2\left(K_L \frac{\partial \overline{q}_i}{\partial x_\beta}\frac{\partial \overline{q}_i}{\partial x_\beta} + K_3 \frac{\partial \overline{q}_i}{\partial x_3}\frac{\partial \overline{q}_i}{\partial x_3}\right) + \frac{\partial}{\partial x_\beta} K_L \frac{\partial \overline{q_i'^2}}{\partial x_\beta} +$$

$$\frac{\partial}{\partial x_3} K_z \frac{\partial \overline{q_i'^2}}{\partial x_3} + \alpha_{ij}(\overline{q}_i \overline{q_i' q_j'}) + \alpha_{ij}(\overline{q_i' q_j'^2}) + \alpha_{ij}(\overline{q_j' q_i'^2}), \qquad (4)$$

The equations (3) and (4) for covariances and variances account for the effects of advection, mean- flow-turbulence interaction, turbulent diffusion, and chemical transformations. We neglected the third moments of the concentration fluctuations in (3) and (4) because they are relatively small. This simplification is adopted in the most of the current parameterization schemes. Equations (2)-(4) together with the hydrodynamic equations for mean flow with a corresponding turbulent closure provide a complete description of a transport of chemically reacting species by a turbulent flow. As the next step of this work we plan to include the contribution of the covariance $\overline{\alpha_{ij}' q_i'}$ to take into account the dependence α_{ij} on the temperature and conduct simulations with different spatial resolution using mesoscale and cloud-resolving models to better quantify the effects of turbulence on atmospheric chemical transformations.

References

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