



Large-eddy simulation of pollutant dispersion from a ground-level area source over urban street canyons with irreversible chemical reactions

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Abstract. In this study, the dispersion of chemically reactive pollutants is calculated by large-eddy simulation (LES) in a neutrally stratified urban canopy layer (UCL) over urban areas. As a pilot attempt, idealized street canyons of unity building-height-to-street-width (aspect) ratio are used. Nitric oxide (NO) is emitted from the ground surface of the first street canyon into the domain doped with ozone (O₃). In the absence of ultraviolet radiation, this irreversible chemistry produces nitrogen dioxide (NO₂), developing a reactive plume over the rough urban surface. A range of timescales of turbulence and chemistry are utilized to examine the mechanism of turbulent mixing and chemical reactions in the UCL. The Damköhler number (*Da*) and the reaction rate (*r*) are analyzed along the vertical direction on the plane normal to the prevailing flow at 10 m after the source. The maximum reaction rate peaks at an elevation where Damköhler number *Da* is equal or close to unity. Hence, comparable timescales of turbulence and reaction could enhance the chemical reactions in the plume.

1 Introduction

Air pollutant dispersion in the UCL is mainly governed by air flows and turbulent mixing, which are indeed affected by both the atmospheric conditions and the rough urban surfaces at the bottom. Because most air pollutants are chemically reactive, the complicated reactions among different species cannot be neglected in air quality assessment. This paper thus attempts to shed some light on the relation between the turbulence and reactions in a plume.

2 Methodology

The computational domain ($L = 72$ m, $W = 5$ m, $H = 8$ m) consists of 36 repeated street canyons placed perpendicular to the prevailing flow (Fig. 1). The street canyon aspect ratio equals unity ($h = w = 1$ m) so the flows fall into the skimming flow regime (Oke, 1988). The spatial domain is discretized into 15 million hexahedral elements. The first grid point is located less than 6 wall units from the nearby solid boundaries.

The UCL flow is assumed to be incompressible and isothermal. The LES is used instead of the Reynolds-averaged Navier-Stokes turbulence models because of its accuracy (Tominaga and Stathopoulos, 2011). The resolved-scale Navier-Stokes equations are

$$\frac{\partial \bar{u}_i}{\partial t} + \frac{\partial \bar{u}_i \bar{u}_j}{\partial x_j} = -\frac{\partial \bar{\pi}}{\partial x_i} + \nu \frac{\partial^2 \bar{u}_i}{\partial x_j \partial x_j} - \frac{\partial \tau_{ij}^a}{\partial x_j} - \Delta P_x \delta_{i1} \quad (1)$$

where $\bar{\pi} (= \bar{p}/\rho + \frac{2}{3}k_{\text{SGS}})$ is the modified pressure with the subgrid-scale (SGS) kinetic energy k_{SGS} , and $\tau_{ij}^a (= -2\nu_{\text{SGS}}\bar{S}_{ij})$ is the anisotropic SGS stress tensor with the SGS eddy viscosity $\nu_{\text{SGS}} (= C_k k_{\text{SGS}}^{1/2} \Delta)$, in which $C_k (= 0.07)$ is an empirical constant and $\Delta (= V_{\text{cell}}^{1/3})$ is the filter size. The flow is driven by the pressure gradient $\Delta P_x (= 0.0004 \text{ m s}^{-2})$ so the freestream velocity u_∞ is close to unity. The kinematic viscosity ν equals $0.0001 \text{ m}^2 \text{ s}^{-1}$, hence, the Reynolds number $Re (= u_\infty h/\nu) = 11\,570$ which is high enough for turbulent flows. Based on the friction

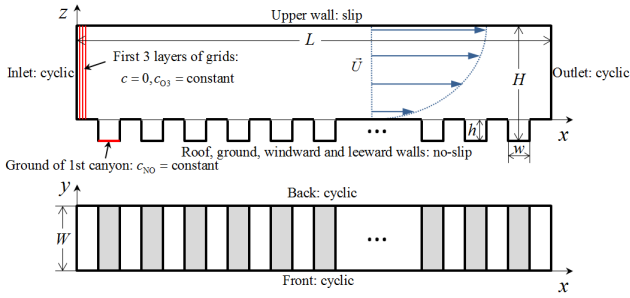


Figure 1. Computational domain and boundary conditions.

velocity u_τ , the friction Reynolds number $Re_\tau (= u_\tau h/\nu)$ equals 529.

The closure term τ_{ij}^a is modeled by the SGS k -equation eddy-viscosity model

$$\frac{\partial k_{SGS}}{\partial t} + \frac{\partial \bar{u}_j k_{SGS}}{\partial x_j} = -\tau_{ij}^a \frac{\partial \bar{u}_i}{\partial x_j} + \frac{\partial}{\partial x_j} \left(\nu_{eff} \frac{\partial k_{SGS}}{\partial x_j} \right) - C_\varepsilon \frac{k_{SGS}^{3/2}}{\Delta} \quad (2)$$

where $\nu_{eff} (= \nu + \nu_{SGS})$ is the effective viscosity and $C_\varepsilon (= 1.05)$ is another empirical constant.

The chemical reaction considered is the irreversible O_3 titration



where $k_3 (= 44.05e^{-1370/T} \text{ ppm}^{-1} \text{ s}^{-1})$ is the temperature $T (= 298.15 \text{ K})$ dependent reaction rate constant.

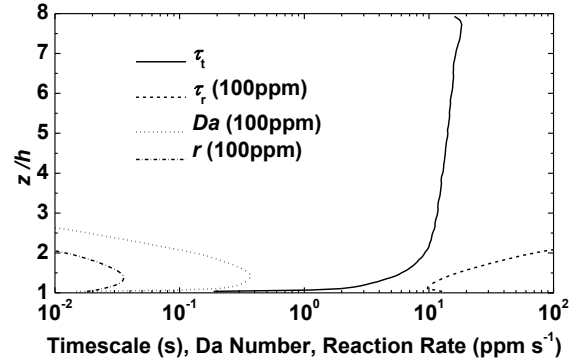
The concentrations of chemical species are calculated by the advection-diffusion equations

$$\frac{\partial \bar{c}_{NO}}{\partial t} + \frac{\partial \bar{c}_{NO} \bar{u}_j}{\partial x_j} - \left(\frac{\nu}{Sc} + \frac{\nu_{SGS}}{Sc_t} \right) \frac{\partial^2 \bar{c}_{NO}}{\partial x_j \partial x_j} + k_3 \bar{c}_{NO} \bar{c}_{O_3} = 0 \quad (4)$$

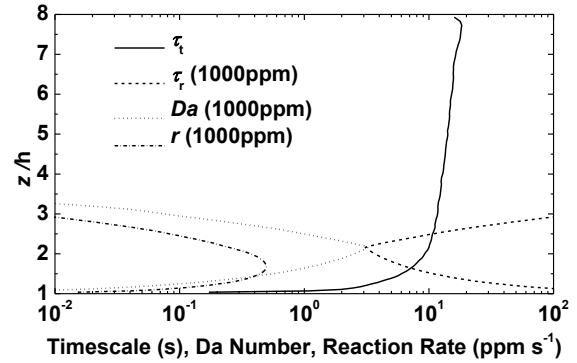
$$\frac{\partial \bar{c}_{O_3}}{\partial t} + \frac{\partial \bar{c}_{O_3} \bar{u}_j}{\partial x_j} - \left(\frac{\nu}{Sc} + \frac{\nu_{SGS}}{Sc_t} \right) \frac{\partial^2 \bar{c}_{O_3}}{\partial x_j \partial x_j} + k_3 \bar{c}_{NO} \bar{c}_{O_3} = 0 \quad (5)$$

where $Sc (= 0.72)$ is the Schmidt number, $Sc_t (= 0.72)$ is the turbulent Schmidt number.

The time derivatives are discretized by the second-order backward differencing, the gradient terms of pressure and velocity are discretized by the central differencing and the convection terms are discretized by the total variation diminishing (TVD) scheme. In the diffusion terms, the central differencing and the explicit non-orthogonal correction are separately used for the approximation schemes of the diffusion coefficients and the surface normal gradients. The LES model was validated by the experimental data of Builtjes (1983).



(a) $C_{NO} = 100 \text{ ppm}$, $X = 10 \text{ m}$



(b) $C_{NO} = 1000 \text{ ppm}$, $X = 10 \text{ m}$

Figure 2. Vertical profiles of turbulent timescale τ_t , reaction timescale τ_r , Damköhler number Da and reaction rate r (averaged in the spanwise direction).

The roof, ground, windward and leeward walls are no-slip boundaries. The upper wall is slip. The inlet and outlet are cyclic so the flow circulates from the outlet to the inlet. The front and back surfaces are cyclic so the spanwise domain is infinite. The NO concentration on the ground of the first street canyon is constant (1, 10, 100 or 1000 ppm) in each configuration. In the first 3 layers of grids after the inlet, the O_3 concentration is kept constant (1 ppm) while the concentrations of the other species (NO , NO_2 and O_2) are equal to zero in order to prevent the species at the outlet from periodically re-entering the spatial domain.

The time step increment is 0.01 s so that the Courant number (≈ 0.3) is less than 1. The initial calculation needs more than 1000 s for fully developed flows. The time for statistics collection after the initial spinning up is 400 s and the time interval for data sampling is 0.1 s.

3 Timescale analysis

The total lifetime is the ratio of turbulent kinetic energy and dissipation rate. The dissipation lifetime is a tenth of the total lifetime (Pope, 2000). Therefore, the turbulence timescale is

$$\tau_t \approx 0.1 \frac{k_t}{\langle \varepsilon_f \rangle} \quad (6)$$

where $k_t \left(= \frac{1}{2} \langle u_i'' u_i'' \rangle \right)$ is the turbulent kinetic energy and $\varepsilon_f \left(= \nu \frac{\partial \bar{u}_i}{\partial x_j} \frac{\partial \bar{u}_i}{\partial x_j} \right)$ is the dissipation rate of the filtered kinetic energy (Pope, 2000). Here $\langle \varepsilon_f \rangle$ represents the spatio-temporal average of the filtered dissipation rate.

Because reaction rates are determined by the slowest one, the reaction timescale is equal to the largest timescale between NO and O₃

$$\tau_r = \max(\tau_{\text{NO}}, \tau_{\text{O}_3}) \quad (7)$$

where $\tau_{\text{NO}} = \frac{1}{k_3 \langle \bar{c}_{\text{O}_3} \rangle}$ and $\tau_{\text{O}_3} = \frac{1}{k_3 \langle \bar{c}_{\text{NO}} \rangle}$, are the reaction timescales of NO and O₃, respectively.

The chemical reaction mechanism is measured by the dimensionless Damköhler number

$$Da = \tau_t / \tau_r \quad (8)$$

That is the ratio of turbulence timescale to reaction timescale. When $Da > 1$, the chemical reaction is faster than the turbulent mixing. When $Da < 1$, the turbulent mixing is stronger than the chemical reaction (Meeder and Nieuwstadt, 2000).

When the maximum Da is less than 1 (Fig. 2a), the reaction rate $r \left(= k_3 \langle \bar{c}_{\text{NO}} \rangle \langle \bar{c}_{\text{O}_3} \rangle \right)$ peaks at the height where Da reaches the maximum. The maximum Da is at the location where the turbulence timescale is closest to the reaction timescale, implying that comparable timescales of turbulence and reaction are crucial to the reaction rate.

When the maximum Da is larger than 1 (Fig. 2b), there are two points at which $Da = 1$. The height of the maximum reaction rate is very close to that of the lower $Da = 1$ point where the timescales of turbulence and reaction are comparable to each other. The upper $Da = 1$ point also has comparable timescales, however, its reaction rate is delayed by the weaker turbulent mixing (longer turbulence timescale).

4 Conclusions

Pollutant plume dispersion with irreversible chemistry over idealized urban roughness is performed by LES. With increasing wall-normal distance from the bottom roughness elements, the turbulence timescale increases. On the contrary, the reaction timescale decreases initially then increases quickly. The height of maximum reaction rate is observed where Da number is equal or close to unity, implying that comparable timescales of turbulence and reaction is one of the factors enhancing chemical reactions.

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