18th International Conference on
Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes
9-12 October 2017, Bologna, Italy

PARAMETERIZATION STUDY OF CHEMICALLY REACTIVE POLLUTANT DISPERSION USING LARGE-EDDY SIMULATION

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Abstract: Dispersion and transport of pollutant emitted from vehicles over urban areas largely affect pedestrian-level air quality. Poor ventilation inside street canyons often results in the accumulation of pollutants which is harmful to human health. Most vehiclar exhausts are chemically reactive that evolve to their secondary counterparts in the atmospheric boundary layer (ABL). Therefore, the conventional Gaussian plume model, which assumes inert pollutants, should be used with caution. The pollutant transport process from the ground level to the ABL is dominated by advection, diffusion and chemistry processes. In this study, turbulent dispersion of reactive pollutants in the ABL over hypothetical urban area of an array of idealised street canyons is investigated using large-eddy simulation (LES). Nitric oxide (NO) is emitted from the first street canyon into the urban ABL doped with ozone (O_3). Firstly, we use the source depletion analogy instead of the conventional Gaussian plume model to estimate the plume shape. Inaccuracy is caused by the dominated NO oxidation in the near-wall region. Regression to the LES output shows that the vertical dimensionless NO profiles exhibit Gamma γ-distribution for a range of background O_3 concentrations, unveiling a new parameterization of reactive plume dispersion over urban areas. Apart from the plume characteristic, budget analysis of the pollutant transport is conducted. Different levels of background O_3 are used to contrast the importance of individual chemistry terms to the pollutant transport. It is found that three terms (advection, diffusion and chemistry) vary with different streamwise location and coupling with each other.

Key words: *hypothetical urban areas, large-eddy simulation (LES), ozone O_3 titration and reactive nitric oxide NO plume transport, budget analysis***.**

INTRODUCTION

Vehiclar exhaust has become one of the major anthropogenic pollutants in urban areas. Elevated pollutant concentrations are commonly found in urban areas especially in metropolises because of the heavy traffic. Nitrogen oxides (NO_x) are some of the major pollutants in the atmospheric boundary layer (ABL) whose high concentrations would adversely affect the health of inhabitants. The problem is more obvious in cities like Hong Kong where the street-level ventilation is weakened by the high-rise buildings. A comprehensive understanding of the mechanism of the pollutant transport and removal helps formulate effective air quality management strategy. The conventional Gaussuan plume model (Roberts 1923) is commonly adopted for regulatory purposes to safeguard public health whose pollutant (passive and inert) concentration after a continuous line source placed in uniform crossflows in isothermal conditions is

$$C(x,z)=\frac{Q}{(\pi/2)^{1/2}U\sigma_z(x)}\left[\exp\left\{\frac{1}{2}\left(\frac{z-z_s}{\sigma_z(x)}\right)^2\right\}+\exp\left\{\frac{1}{2}\left(\frac{z+z_s}{\sigma_z(x)}\right)^2\right\}\right] \tag{1}$$

where $c(x, z)$ is the pollutant concentration, x the streamwise coordinate, z the vertical coordinate, Q the pollutant emission rate (per unit width), U the (uniform) wind speed, z_s the emission height and $\sigma_z(x)$ the (vertical) dispersion coefficient in term of x. Its results, however, must be interpreted cautiously because of the inert-pollutant assumption and the complicated near-ground turbulent transport processes in the atmospheric surface layer (ASL; Britter and Hanna 2003).

In order to elucidate the chemically reactive pollutant transport mechanism in urban areas, large-scale computer models have been widely adopted by many researchers. Most studies focused on the pollutant transport inside the street canyons (Baik et al., 2007; Chung et al., 2012). The coupling mechanism between turbulence and chemistry is not well understood yet. This study extends the transport process to the entire urban boundary layer that elucidates the physical and chemistry processes in the pollutant transport.

METHODOLOGY

In view of the inaccuracy of the conventional Gaussian plume model for chemically reactive pollutant, we switch to source depletion analogy. Deposition is the removal of airborne pollutants from the plume onto ground surfaces (Bergin et al., 1999). It is an interfacial process governed by aerodynamic parameters such as friction velocity u_* and roughness length z_0. The pollutant source in the Gaussian plume model Equation (1) is depleted in the streamwise direction x in the following manner

$$\frac{d}{dx}Q_{(x)} = -v_d(x) \times \frac{Q(x)}{(\pi/2)^{1/2} U \sigma_z(x)} \exp\left\{-\frac{1}{2}\left(\frac{z_s}{\sigma_z(x)}\right)^2\right\} \tag{2}$$

where $v_d\ (x)$ is the deposition velocity. Integrating Equation (2) from the source at $x = 0$ to x yields the source-deplection equation

$$\int_{Q_0}^{Q_{dp}(x)} \frac{1}{Q(x)} dQ_{(x)} = \ln\left[\frac{Q_{dp}(x)}{Q_0}\right] = -\frac{1}{(\pi/2)^{1/2} U} \int_0^x \frac{v_d(x)}{\sigma_z(x)} \exp\left\{-\frac{1}{2}\left(\frac{z_s}{\sigma_z(x)}\right)^2\right\} dx \tag{3}$$

where Q_0 is the initial undepleted pollutant source and $Q_{dp}(x)$ the depleted pollutant source in the streamwise direction x in response to ground-level deposition. Instead of inert pollutants, the irreversible O_3 titration

$$O_3 + NO \xrightarrow{k_3} NO_2 + O_2 \tag{4}$$

is considered where O_2 is oxygen molecule and k_3 (= 44.05 exp [−1370/Θ] ppm^{-1} sec^{-1}) the temperature-dependent chemical reaction rate constant. Isothermal conditions at Θ =293.15 K are assumed.

LES of the open-source CFD code Open-FOAM 2.3.0 (OpenFOAM 2015) is used in this paper. The LES model for hypothetical urban area consists of a number of idealized urban street canyons fabricated by identical square ribs of size h (Figure 1). The spatial domain sizes 72h (length) × 12h (width) × 12h (height) that is composed of 36 idealized street canyons of the same geometry (Figure 1). The street width b is the same as the building height h so the building-height-to-street-width (aspect) ratio is equal to unity. The flows thus fall into the skimming flow regime. The prevailing flows in the urban ASL are driven by the (background) pressure gradient ΔP_x perpendicular to the street axes, representing the worst scenario of pollutant removal from street canyons. The domain extent is 12h in the homogeneous spanwise y direction. The infinitely long streamwise x domain is constructed by periodic boundary conditions (BCs) in the horizontal extent. Wall BCs are applied on all the solid boundaries and a shear-free BC along the domain top. The prevailing wind enters the spatial domain from the upstream inflow doped with constant background ozone $[O_3]_0$ at different concentrations (Table 1). An area source of nitric oxide with constant concentration $[NO]_0$ is placed on the ground surface of the first street canyon from which a reactive pollutant is continuously emitted into the computational domain, simulating vehicular exhaust in urban areas. Neumann BCs are applied on the remaining solid boundaries. An open BC of pollutants is prescribed at the downstream outflow so all the chemical species are removed from the computational domain without any reflection.

Table 1. Scenario analysis.

Case No.	**1**	**2**	**3**	**4**	**5**	**6**
NO /ppb	1,000	1,000	1,000	1,000	1,000	1,000
O_3 /ppb	1	10	50	100	200	500

For the pollutant transport, the advection-diffusion equation with chemistry source/sink is given by

$$\frac{\partial \phi}{\partial t} + U_i \frac{\partial \phi}{\partial x_i} = K \frac{\partial^2 \phi}{\partial x_i^2} + S_\phi \tag{5}$$

where ϕ is concentration of chemical species, U_i the wind speed, x_i the Cartesian coordinate, K the mass

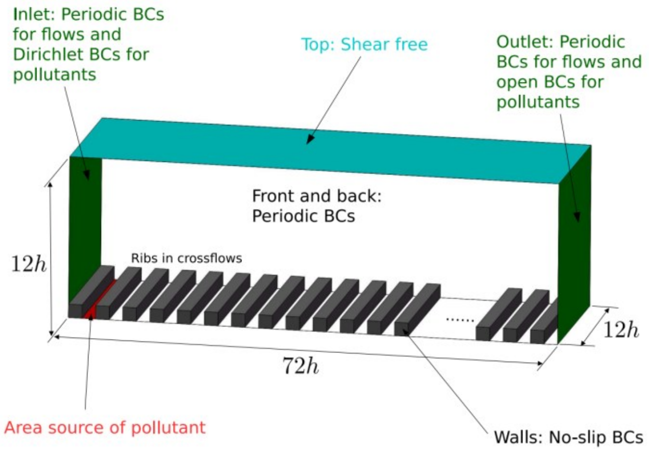

Figure 1: Spatial domain of the LES.

diffusivity and S_ϕ the source term. Using advection timescale $T_0 = L_0 / U_0$, chemistry timescale $\tau_{\mathrm{NO}}^{-1} = -k_3[O_3]_0$ and $Da = T_0 / \tau_{\mathrm{NO}}$ (Relative time between physics and chemistry), finally we can get the dimensionless transport equation of NO

$$\frac{\partial \hat{\phi}_{\mathrm{NO}}}{\partial \hat{t}} + \hat{U}_i \frac{\partial \hat{\phi}_{\mathrm{NO}}}{\partial \hat{x}_i} = -Da_{\mathrm{NO}} \hat{\phi}_{\mathrm{NO}} \hat{\phi}_{\mathrm{O_3}} \,. \tag{6}$$

Take the ensemble average and apply pseudo-steady state condition for 2D flows over street canyons

$$-\langle \hat{u} \rangle \frac{\partial \langle \hat{\phi}_{\mathrm{NO}} \rangle}{\partial \hat{x}} - \langle \hat{w} \rangle \frac{\partial \langle \hat{\phi}_{\mathrm{NO}} \rangle}{\partial \hat{z}} - \frac{\partial}{\partial \hat{x}} \langle \hat{u}'' \hat{\phi}''_{\mathrm{NO}} \rangle - \frac{\partial}{\partial \hat{z}} \langle \hat{w}'' \hat{\phi}''_{\mathrm{NO}} \rangle - Da_{\mathrm{NO}} \langle \hat{\phi}_{\mathrm{NO}} \rangle \langle \hat{\phi}_{\mathrm{O_3}} \rangle - Da_{\mathrm{NO}} \langle \hat{\phi}''_{\mathrm{NO}} \hat{\phi}''_{\mathrm{O_3}} \rangle = 0 \tag{7}$$

The first two terms are the advection terms, the third and fourth terms in the middle are the turbulent diffusion terms, and the last two terms are the chemistry terms.

RESULTS

Figure 2 depicts the flow properties in the urban ASL over the rib-type hypothetical urban surface described previously. Similar to that of open-channel flows, the ensemble-averaged streamwise velocity $\langle \overline{u} \rangle$ increases rapidly with height over the roughness elements, converging to the prevailing wind speed u_∞ at the domain top. The mean-wind-speed profile (Figure 2a) agrees well with our previous wind tunnel measurement (Ho and Liu 2016). The streamwise $\langle u''u'' \rangle^{1/2}$ (Figure 2b) and the vertical $\langle w''w'' \rangle^{1/2}$ (Figure 2c) fluctuating velocities are normalized by the friction velocity u_*. A good agreement of fluctuating velocities, especially in the lower urban ASL, between the current LES and the previous wind tunnel measurements is observed. As shown in Figure 2d, both our previous wind-tunnel measurements (Ho and Liu 2016) and the current LES results exhibit the conventional characteristics of ensemble averaged vertical momentum flux $\langle u''w'' \rangle$ that decreases with increasing height. Nitrogen conserves in the sensitivity tests regardless of the background ozone concentration so the nitrogen oxides NO_x plume can be taken as passive-scalar dispersion. Figure 3 depicts the dimensionless profiles of passive-scalar concentration $\langle \overline{c}_{\mathrm{NO}_x} \rangle$ as functions of height z at different streamwise locations x. The current LES-calculated passive scalar concentration agrees well with the theoretical Gaussian solution Equation (1).

For the chemically reactive plume, peaks of nitric oxide concentration are elevated in the near-wall region (Figure 4). The conventional Gaussian plume model is no longer suitable. In view of the dominated UCL chemistry, we attempt to modify the Gaussian plume model by source depletion analogy (Arya 1998) to handle ozone titration Equation (4).

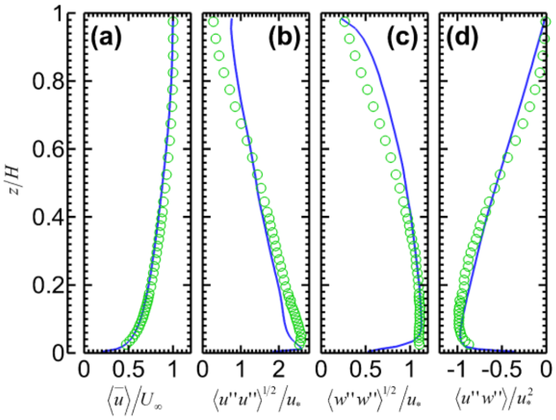

Figure 2: Vertical profiles of (a) mean wind speed; (b) streamwise fluctuating velocity; (c) vertical fluctuating velocity and (d) vertical momentum flux. Solid line: LES data and symbols: wind tunnel data (Ho and Liu 2016).

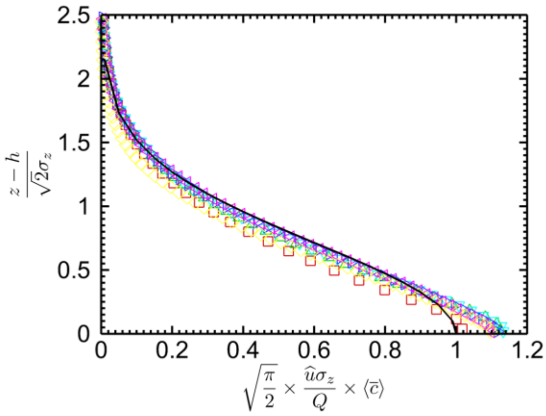

Figure 3: Profiles of dimensionless concentration of passive scalar at x/h =: (a) 15.5 (□), (b) 25.5 (△), (c) 35.5 (▽), (d) 45.5 (▷), (e)55.5 (◁) and (f) 65.5 ◇). Theoretical Gaussian plume profile (——)

For the far-field nitric oxide concentration $\langle \bar{c}_{NO} \rangle$, the analytical Gaussian-form source depletion plume model and the current LES-calculated results exhibit various degrees of disagreement in response to the background ozone concentrations $[O_3]_0$. An obvious discrepancy, which increases with increasing background ozone concentration, is observed in the UCL. Over the mean plume rise the prediction of the newly developed source depletion model is good, suggesting a handy parameterization for the estimate of reactive plume dispersion.

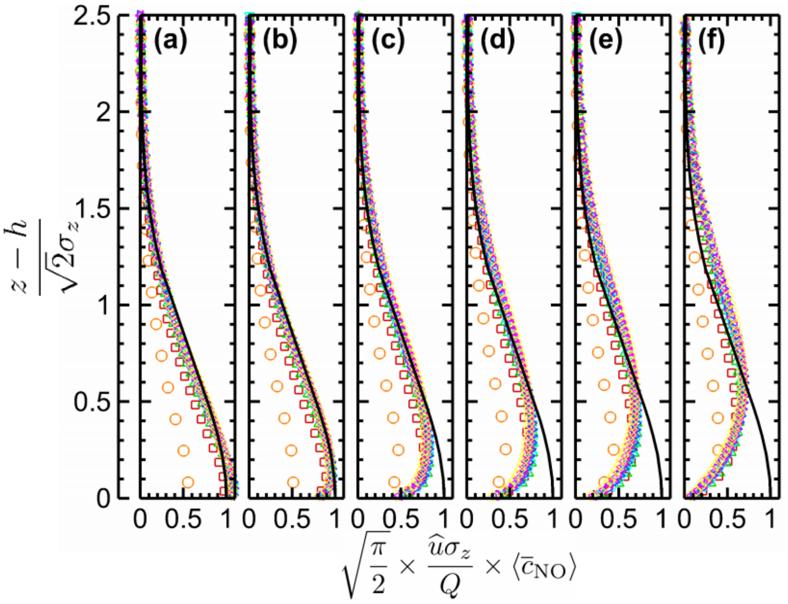

Figure 4: Dimensionless profiles of reactive pollutant nitric oxide concentration in the streamwise direction at x/h = 15.5:□, 25.5:Δ, 35.5:∇, 45.5:▷, 55.5:◁ and 65.5: ◇ expressed in dimensionless wall-normal coordinate $(z-h)/(2^{1/2}\sigma_z)$ for background ozone concentration $[O_3]_0$ of (a) 1 ppb; (b) 10 ppb; (c) 50 ppb; (d) 100 ppb; (e) 200 ppb and (f) 500 ppb. Also shown are the profiles of the theoretical Gaussian plume model: —— Equation (1) and the γ-distribution: - - - - - - Equation (21).

In view of the discrepancy observed above, additional effort is sought to improve the parameterization for the dimensionless profiles of nitric oxide (reactive plume dispersion). We focus on the near-wall region below the mean plume rise $z \leq z_r$ where the source depletion model over-predicts the ground-level nitric oxide concentration. Gamma γ-distribution in terms of γ function $\Gamma(\alpha)$

$$\sqrt{\frac{\pi}{2}} \times \frac{\hat{u}\sigma_z}{Q_{dp}} \times \langle \bar{c}_{NO} \rangle = \frac{\left(z/\sqrt{2}\sigma_z\right)^{\alpha-1}}{\beta^{\alpha}\Gamma(\alpha)} \exp\left(-\frac{z/\sqrt{2}\sigma_z}{\beta}\right) \qquad (8)$$

is tested again by regression where α and β are parameters which can be determined by the mean and the variance of the distribution $\mu = \alpha\beta$ and $\sigma^2 = \alpha\beta^2$. The reduction in nitric oxide concentration below the mean plume rise z_r in the near-wall region is predicted well by the Gamma γ-distribution (Figure 4). Over the mean plume rise, both the Gaussian-form source depletion model and the Gamma γ-distribution show consistent dimensionless profiles which are close to the current LES-calculated nitric oxide concentration.

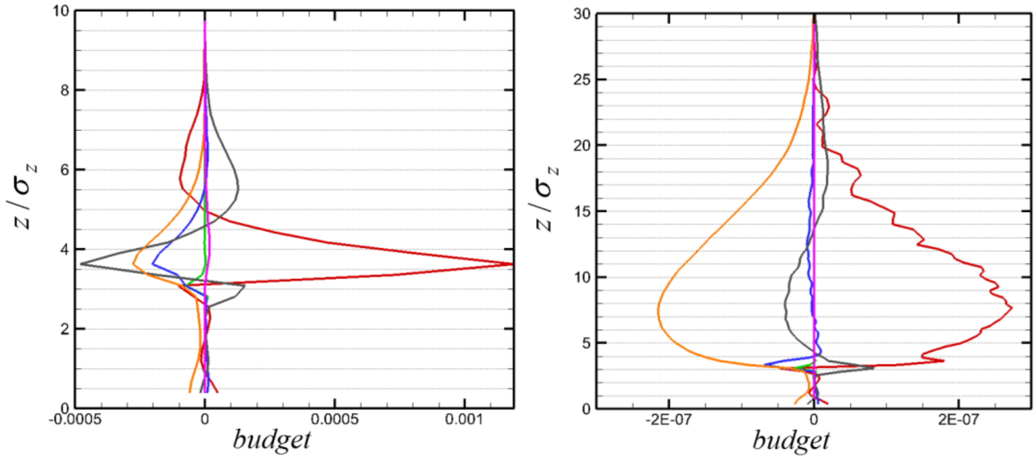

Figure 5: Budget analysis in case 3 in the streamwise direction at x/h = 5.54: (left) and x/h = 65.54 (right). ——**term 1**; ——**term 2**;——**term 3**;——**term 4**; ——**term 5**; and ——**term 6**.

Figure 5 analyzes the budget of the pollutant NO transport at different streamwise location for case 3. In the near field (close to the pollutant source) the advection along x direaction is important for the pollutant transport. Term 2 is the advection in z direction, which is neglectable except at the roof level. Term 6 is the fluctuation part of the chemistry term, which is also neglectable compared with others. Below the roof level, the chemistry term shifts between positive and negative values, implying the complicated mixing and chemistry inside the street canyons. In the far field, the chemistry term becomes more important compared with the advection. The diffusion terms (both in x and z direction) fluctuate along the vertical height. The advection in z direction (term 2) and term 6 is still neglectable compared with other components. The advection, diffusion and chemistry terms are tightly coupled with each other. Last but not least, pollution chemistry cannot be ignored in the pollutant removal from street-level air quality.

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