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On the Transport of Chemically Reactive Pollutants over Urban Roughness in the Atmospheric Boundary Layer

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1. Introduction

Elevated pollutant concentrations are commonly observed in urban areas, such as street canyons, threatening human health. While most practical dispersion models assume inert pollutants, emissions from traffic exhaust are chemically reactive. Their dynamics are further complicated by atmospheric turbulence, geometry/orientation of buildings, thermal stratification and chemical kinetics, etc. Reactive flue gases, including nitrogen oxides (NO_x) and volatile organic compounds (VOCs), are among the most important pollutants that would cause a series of public health problems (Cheng et al., 2013, Mahiyuddin et al. 2013). There is thus a need for improved understanding of the dynamics of chemically reactive pollutants. In this paper, turbulent dispersion of reactive pollutants in the atmospheric boundary layer (ABL) over hypothetical urban areas is numerically investigated using large-eddy simulation (LES). Their transport behavior in and over idealized street canyons of unity aspect ratio in isothermal conditions is studied. The worst scenario is considered in which the prevailing flow is perpendicular to the street axes. In pseudo-steady, fully developed turbulent flows, nitric oxide (NO) is emitted from the ground surface in the first street canyon into the urban ABL doped with ozone (O_3). By looking into the pollutant concentrations and dispersion characteristics over the buildings, it is realized that, apart from pollutant removal, ABL turbulence plays an important role in the mixing of chemicals, which in turn substantially affects the reaction rates. To contrast the effect of chemistry and dispersion, we compare the time scales of reaction and diffusion of pollutants by switching on and off and NO_x-O_3 chemistry. It is found that chemical reaction reduces (increases) the diffusion time scale near the roof level of the rough surface at the bottom (near the domain top in the far field). There exists a branching point in the z direction for different effect caused by chemical reactions. Below this point, the diffusion time scale of NO is increased by the reaction, and vice versa.

2. Methodology

LES with the one-equation subgrid-scale (SGS) model is adopted to handle the flows and the dispersion of chemically reactive pollutants over an array of idealized, identical street canyons. A (spatial) box filter is applied to decompose the variables into their resolved-scale (denoted by overline) and SGS components. The governing equations thus consist of the continuity

$$\frac{\partial \overline{u}_i}{\partial x_i} = 0 \tag{1}$$

and the momentum conservation

$$\frac{\partial \overline{u}_i}{\partial t} + \frac{\partial}{\partial x_j}\overline{u}_i\overline{u}_j = -\Delta P \delta_{i1} - \frac{\partial \overline{p}}{\partial x_i} + \left(\nu + \nu_{SGS}\right)\frac{\partial^2 \overline{u}_i}{\partial x_j \partial x_j} \tag{2}$$

in resolved scales. Tensor notation is used where $\overline{u}_i$ are the velocity components, t the time, x_i the Cartesian coordinates, ΔP the (background) pressure gradient (driving the prevailing flow), $\overline{p}$ the kinematic pressure, ν the kinematic viscosity, ν_{SGS} the SGS kinematic viscosity and δ_{ij} the Kronecker delta.

The following chemical reactions

$$\mathrm{NO_2} + h\nu \xrightarrow{j_1} \mathrm{NO} + \mathrm{O} \tag{3}$$

$$\mathrm{O} + \mathrm{O_2} + \mathrm{M} \xrightarrow{k_2} \mathrm{O_3} + \mathrm{M} \tag{4}$$

and

$$\mathrm{NO} + \mathrm{O_3} \xrightarrow{k_3} \mathrm{NO_2} + \mathrm{O_2} \tag{5}$$

are included to handle the reversible pollution chemistry. Here, NO_2 is the nitrogen dioxide, O the ground-state oxygen atom and M the third-party molecule (usually nitrogen N_2 or oxygen O_2) to absorb the excess vibration energy. We assume that the rate constants remain unchanged throughout the LES in which $j_1 = 8.9\times10^{-3}$ sec^{-1}, $k_2 = 3.64\times10^{-13}$ ppb^{-1} sec^{-1} and $k_3 = 4.43\times10^{-4}$ ppb^{-1} sec^{-1}.

In the reversible chemistry, we use the Damköhler number Da to compare the turbulent diffusion and chemical reactions. It is defined by the ratio of diffusion-to-reaction time scales, as follows

$$Da = \tau_d / \tau_r \tag{6}$$

where τ_d and τ_r are the time scales of diffusion and reaction, respectively.

In this paper, the diffusion time scale is defined as

$$\tau_d = \frac{\sigma_z^2}{4K_\phi} \tag{7}$$

where σ_z is the plume dispersion coefficient after the ground-level area source in the first street canyon and

$$K_\phi = \frac{\langle w''\phi'' \rangle}{\partial \langle \bar{\phi} \rangle / \partial z} \tag{8}$$

the eddy diffusivity by closure assumption. Here, angular brackets and double primes represent the ensemble average and the deviation from the ensemble average, respectively. The physical meaning of diffusion time scale τ_d is indeed the time for diffusion acting on a pollutant concentration gradient over certain distance in crosswind in the plume. The length scale of vertical dispersion coefficient σ_z is used in this paper.

The reaction time scales are different for the chemicals. For NO, it is measured by

$$\tau_{\mathrm{NO}} = \frac{1}{k_3[\mathrm{O}_3]} \tag{9}$$

and for O_3 is

$$\tau_{\mathrm{O}_3} = \frac{1}{k_3[\mathrm{NO}]} \tag{10}$$

They are the retention time of pollutants due to removal by chemical reactions only. Hence, the Damköhler numbers of NO and O_3 are, respectively,

$$Da_{\mathrm{NO}} = \frac{\tau_d}{\tau_{\mathrm{NO}}} = \frac{\sigma_z^2}{4K_{\mathrm{NO}}} k_3[\mathrm{O}_3] \tag{11}$$

and

$$Da_{\mathrm{O}_3} = \frac{\tau_d}{\tau_{\mathrm{O}_3}} = \frac{\sigma_z^2}{4K_{\mathrm{O}_3}} k_3[\mathrm{NO}] \tag{12}$$

3. Computational Domain

Figure 1 depicts the three-dimensional (3D) LES spatial domain employed in this paper. Its height is 12h that consists of 36 identical, idealized street canyons. The building height h and street width b are equal so the building-height-to-street-width (aspect) ratio h/b is equal to 1. The flow in the ABL core is driven by the background pressure gradient that is perpendicular to the street axes, representing the worst scenario of pollutant removal.

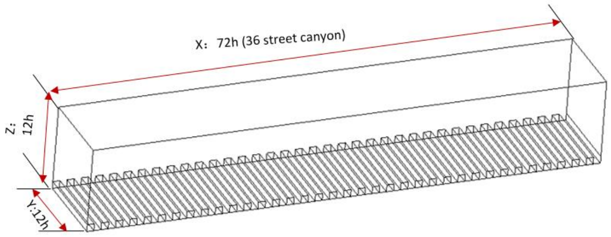

Fig. 1 computational domain

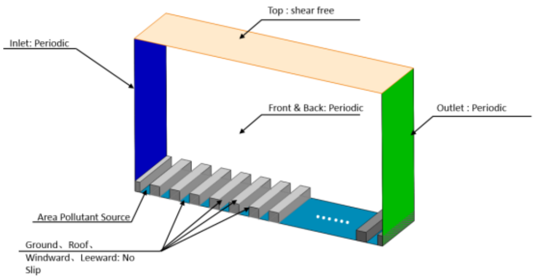

Fig. 2 Boundary conditions

Figure 2 shows the boundary conditions (BCs) of the current LES. The infinitely large array of street canyons is constructed by periodic BCs in the horizontal extent. The prevailing wind enters the spatial domain from the upstream inlet with background O_3 of concentration 1 ppb. An area source of NO of concentration 1 ppb is continuously emitted from the ground surface of the first street canyon to simulate vehicular emission.

Each street canyon comprises of 12, 96, and 12 grid cells in the streamwise (x), spanwise (y), and vertical (z) directions, respectively. There are 864 (x) × 96 (y) × 72 (z) grid cells in the ABL core over the street canyons. The entire spatial domain is thus discretized by over 6.4 million grid cells.

3. Results and Discussion

Figure 3 shows the vertical dispersion coefficient of NO σ_z as a function of streamwise distance x measuring from the pollutant source. Indeed σ_z is the length scale measuring the vertical extent of the pollutant plume. The larger the dispersion coefficient, the wider is the pollutant plume coverage.

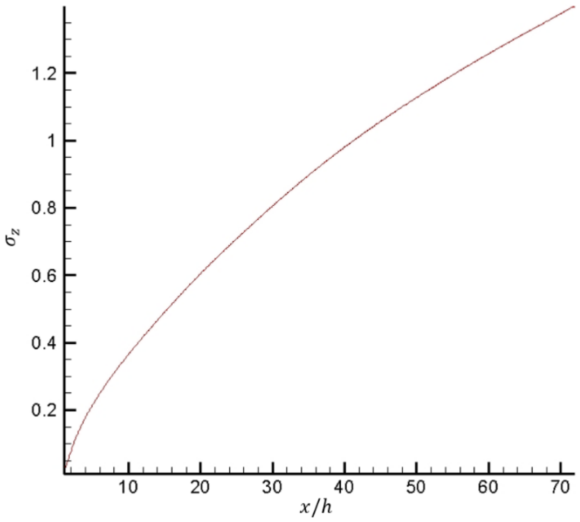

Fig. 3 Dispersion coefficient of NO

Figure 4 depicts the diffusion time scale τ_d of NO (normalized by the diffusion time scale near the roof level τ_{d0} at $z = 1.1h$). Near the first street canyon ($x = 6h$), σ_z is relatively small because the plume is not yet fully developed. The plume dispersion is thus limited near the roof level. The pollutant concentration gradient is small near the domain top so the diffusion time scale remains small from $z = 3h$ and thereafter (Figure 4). To help analyze the plume characteristic, the streamwise domain is divided into near field ($x \leq 6h$) and far field ($x > 6h$). In the far field, the vertical coverage is notable and the diffusion time scale also increases. The normalized diffusion time scale exhibits linear characteristic within the plume ($z/\sigma_z \leqslant 10$) that decreases with increasing distance from the plume centerline. Figure 5 compares the profile of vertical pollutant flux $<w''\phi''>$ at different streamwise locations. A local maximum of vertical pollutant flux is observed near the roof level at different streamwise locations. A similar trend is also observed in the gradient of mean pollutant concentration (Figure 6).

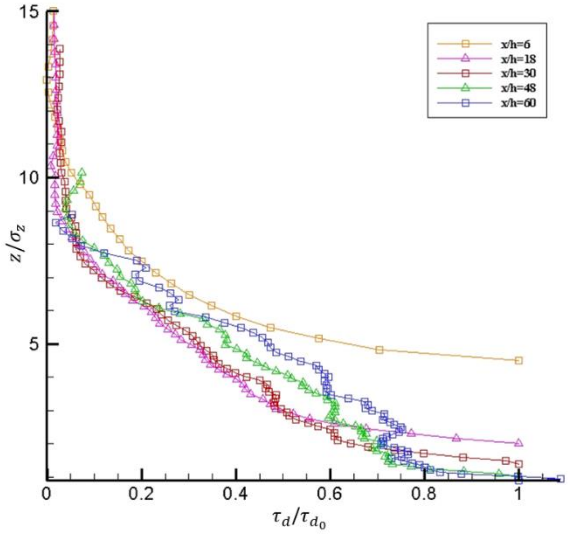

Fig. 4 Diffusion time scale for NO

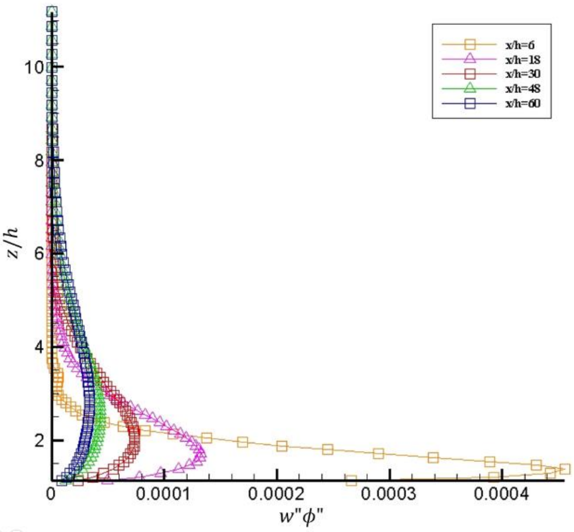

Fig. 5 <$w''\phi''$> of NO

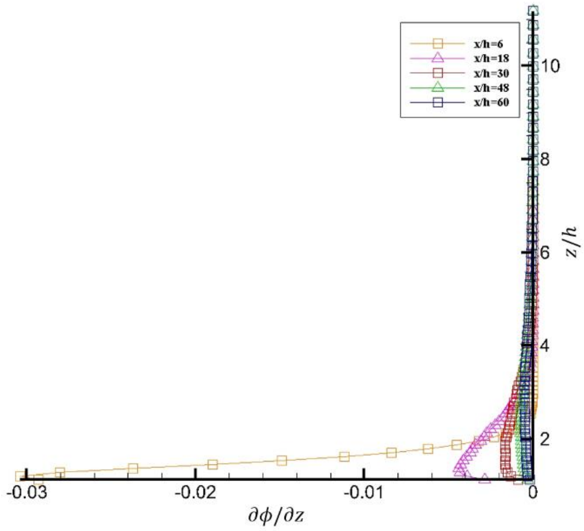

Fig. 6 $\partial\langle\bar{\phi}\rangle/\partial z$ of NO

Figure 8 depicts the reaction time scale of NO which is normalized by the reaction time scale at the domain top τ_{NOr}. In the near-wall region, O_3 is consumed by the chemical reaction Equation (5). The O_3 concentration is lower than that in the upper domain. Therefore, the reaction time scale of NO in the near-wall region is longer than that in the upper domain. In the far field the gradient of reaction time scale in the vertical direction become larger than that in the near field because of O_3 deficit. The increasing speed of reaction time scale of NO in the near-wall region (below $z/\sigma_z = 5$) varies with x/h. The reaction time scale of NO increases faster in the far field than does in the near field. Figure 8 compares the profiles of Damköhler number of NO Da_{NO} which exhibits the characteristic similar to that of the diffusion time scale. The diffusion time scale is much smaller compared with the reaction time scale. In this case, Da_{NO} is smaller than 1, signifying that the turbulent dispersion is faster than its chemical counterpart. Hence, slow chemical reactions occur in the rather well-mixed condition of NO and O_3 in the plume.

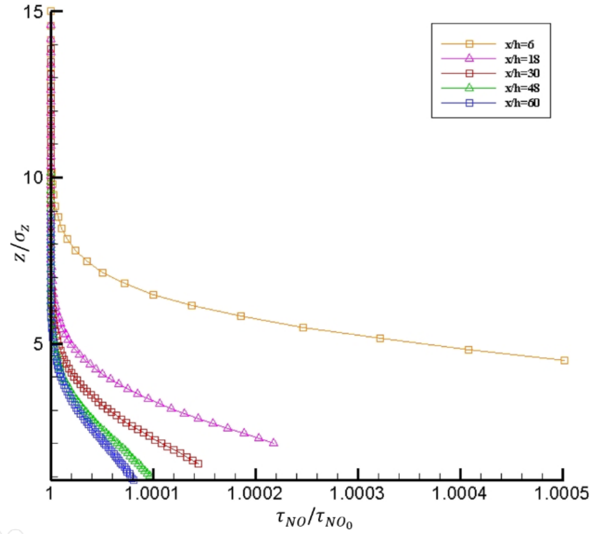

Fig. 7 Reaction time scale of NO

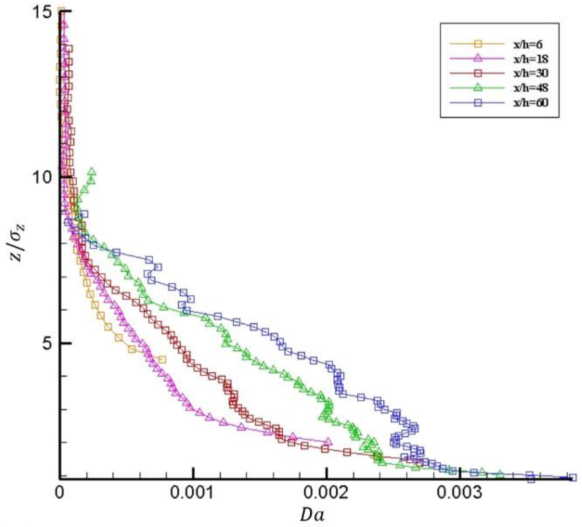

Fig. 8 Damköhler number of NO

We use a passive scalar as the reference for reactive NO to investigate the effect of chemical reactions on the plume dispersion. The initial conditions and BCs of passive scalar are the same as those of NO but no chemical reaction is involved. The relative reduction in diffusion time scale

$$\text{Relative Reduction }\% = \frac{\tau_{\text{passivescalar}} - \tau_{\text{NO}}}{\tau_{\text{passivescalar}}} \times 100\% \qquad (13)$$

at $x = 30h$ is plotted in Figure 9. Near the roof level, the chemical reaction tends to reduce diffusion time scale. For example, at $x = 30h$, below $z = 2\sigma_z$, τ_{NO} is smaller than $\tau_{\text{passive scalar}}$, implying that the chemical reaction reduces the diffusion time scale of pollutant. There exists a level for this effect over which the diffusion time scale is increased by chemical reaction. Over $z = 2\sigma_z$ to the domain top, τ_{NO} is larger than, $\tau_{passive\ scalar}$, signifying that the chemical reaction increases the diffusion time scale of pollutant. The trajectory of that level increases in the wall-normal direction z with increasing streamwise distance x measuring from the pollutant source. For instance, at $x = 30h$, the branching point is at $z = 2h$ (Figure 9). Moreover, at $x = 54h$, the branching point is at $z = 2.88h$ (Figure 10). Figure 11 illustrates the trajectory of branching point as a function of streamwise distance x in which a linear relation is clearly observed.

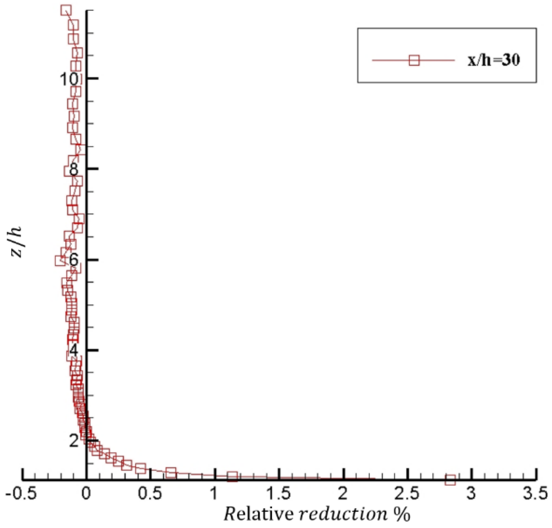

Fig. 9 Relative reduction in diffusion time scale $\frac{(\tau_{Pollutant1}-\tau_{NO})}{\tau_{Pollutant1}} \times 100\%$*. (at* x */h= 30)*

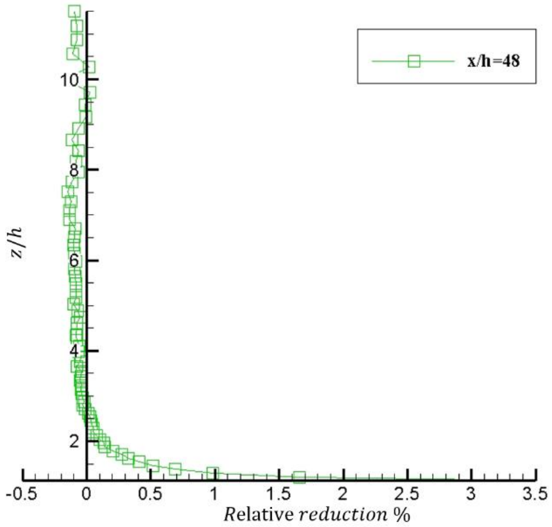

Fig. 10 Relative reduction in diffusion time scale $\frac{(\tau_{Pollutant1}-\tau_{NO})}{\tau_{Pollutant1}} \times 100\%$*. (at* x */h= 48)*

4. Conclusion

A series of LES is performed to examine the dispersion behaviors of reactive pollutants (NO is used in this paper) over hypothetical urban areas. By using the time scales of diffusion and chemistry, the variation of Damköhler number of NO in plume dispersion is reported. It is find that the two time scales are coupled with each other. In particular, pollutant chemistry could increase or decrease the diffusion time scale, i.e. dispersion coefficient. As such, the dispersion and chemistry of pollutant should not be considered separately.

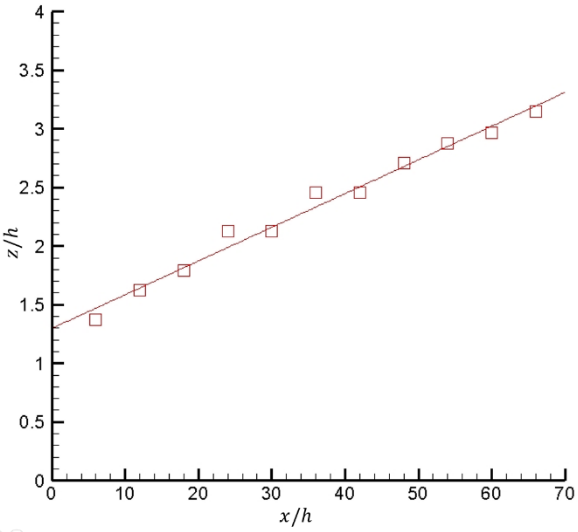

Fig. 11 Critical point in the streamwise x direction

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