Parameterization Study of Chemically Reactive Pollutant Dispersion using Large-Eddy Simulation

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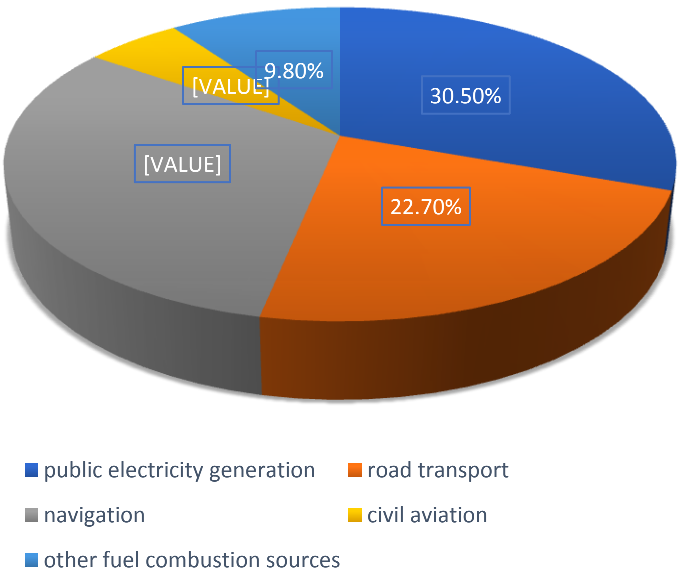
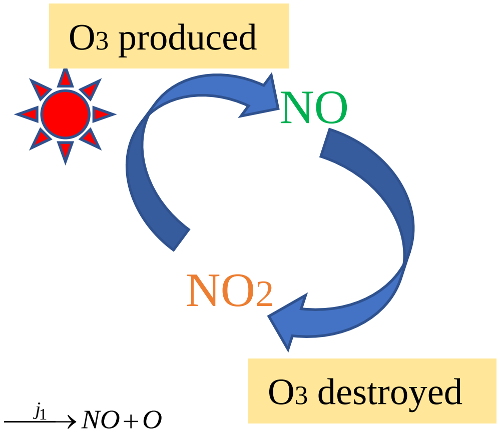
Background

- Buildings and population largely affect the air quality and even the local urban climate in many metropolises.
- Accurate modelling of the interface between an urban surface and the atmosphere will benefit applications such as weather forecasting, air quality and sustainable urban development.
- Hong Kong is a city with high density of population and buildings.

Background





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

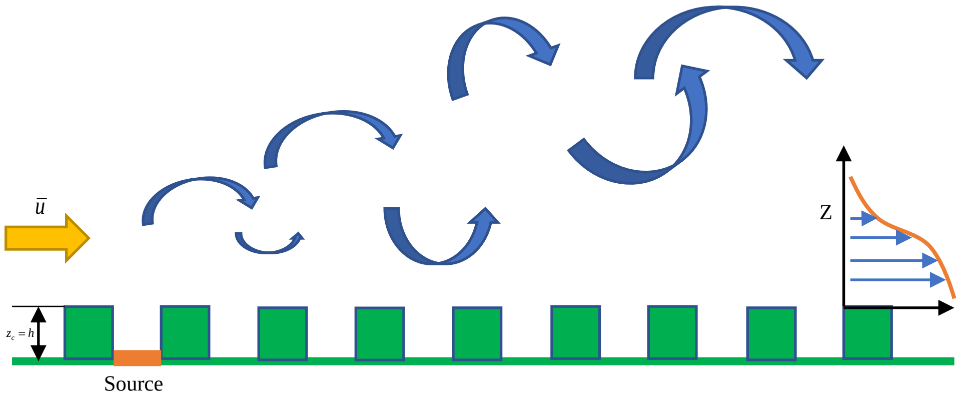
$$O + O_2 + M \xrightarrow{k_2} O_3 + M$$

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

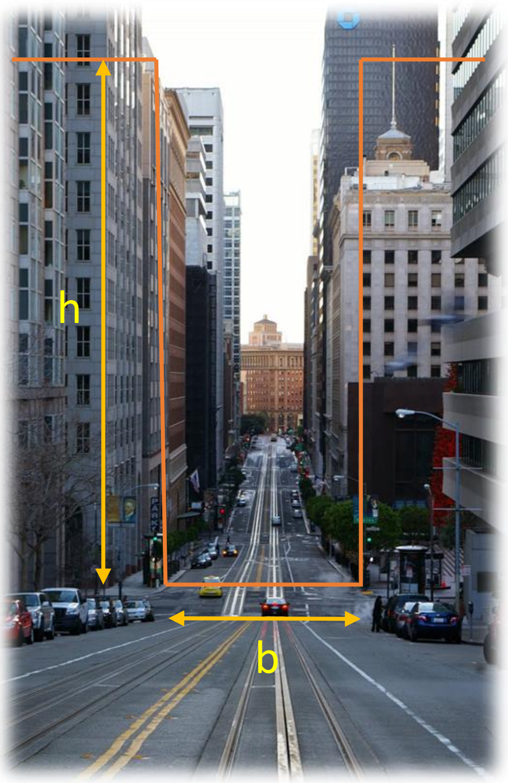
- According to Hong Kong (HK) Environmental Protection Department (EPD), the total emission of NOx in HK in 2013 was about 113,220 tonnes.
- The emission sources of NO_x in HK include public electricity generation, road transport, navigation, civil aviation and other fuel combustion sources, etc.
- Road transport is one of the major sources of NO_x.

- While most practical dispersion models assume inert pollutants, emissions from traffic exhaust are chemically reactive.
- NO can be oxidized by ozone (O_3) in the atmosphere.
- Under sunlight, NO_2 can also be decomposed into NO and O_2
- Chemical reactions in the atmosphere are much more complicated.

Background



- Street canyon is a basic unit commonly used in CFD model to investigate the mechanism of flows over roughness.
- Dynamics are complicated by atmospheric turbulence, geometry/orientation of buildings, thermal stratification and chemical kinetics, etc.
- Oxidation rate of NO is affected by both physical and chemical processes. The physical process is mixing of the plume with the ambient air. The chemical processes are the molecular reactions of NO with different species in the atmosphere.
- There exists a wide range of turbulent eddies which act on plume dispersion. Dispersion and mixing of materials, which in fact constitute the plume, are driven by eddies of all sizes in the atmospheric boundary layer.
  - Large eddies : cause plume meandering.
  - Middle eddies : cause widening and internal mixing of plume on small scales.
  - Small eddies: important for chemical reactions.



Street canyon
Building-height-to-street-width (aspect) ratio
AR = h/b

Objective

- Develop a CFD model for simple NO_x-O_3 chemistry.
- Contrast the plume dispersion characteristics of passive scalar and chemically reactive pollutant.
- Analyze the plume characteristics in scenarios with different O_3 background concentrations using Gaussian plume model.
- Elucidate the contribution of advection, diffusion and chemistry term in pollutant transport.

Methodology

- **Model**
  Large-eddy simulation (LES) with one-equation SGS model

- **Governing equations (filtered)**

  - continuity

$$\frac{\partial \bar{u}_i}{\partial x_i} = 0$$

  - momentum conservation

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

  - Transport equation of pollutants

$$\frac{\partial \bar{\phi}}{\partial t} + \bar{u}_j \frac{\partial \bar{\phi}}{\partial x_j} = \frac{\partial}{\partial x_j}\left(D \frac{\partial \bar{\phi}}{\partial x_j}\right) + S(\bar{\phi})$$

- First step is to handle the **irreversible chemical reaction**

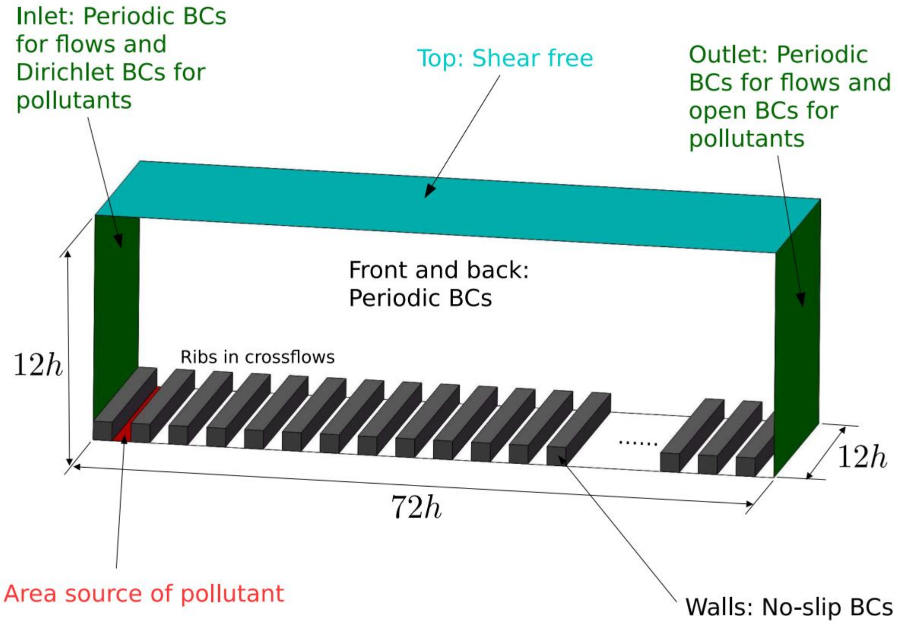
$$\mathrm{NO} + \mathrm{O_3} \xrightarrow{k_3} \mathrm{NO_2} + \mathrm{O_2}$$

- **Source terms** for NO, NO_2 and O_3

$$\frac{d[NO]}{dt} = -k_3[O_3][NO] \qquad \frac{d[O_3]}{dt} = -k_3[O_3][NO] \qquad \frac{d[NO_2]}{dt} = k_3[O_3][NO]$$

- The open-source CFD tool OpenFOAM is used to simulate. Finite volume method (FVM) is used to solve the governing equations. The implicit second-order-accurate backward differencing is using in the temporal domain.

CFD Model



Case NO.	NO Concentration /ppb	O_3 Concentration /ppb
Case 1	1000	1
Case 2	1000	10
Case 3	1000	50
Case 4	1000	100
Case 5	1000	200
Case 6	1000	500

- The LES model for hypothetical urban area consists of a number of idealized urban street canyons fabricated by identical square bars of size h .
- The spatial domain sizes 72h (length) × 12h (width) × 12h (height) that is composed of 36 idealized street canyons of the same geometry.
- 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 that falls into the skimming flow regime (Oke 1988).
- The prevailing flows in the urban ABL are driven by the (background) pressure gradient ΔP_x perpendicular to the street axes, representing the worst scenario of pollutant removal from street canyons.

Results: Flow Validated by Wind Tunnel Experiment

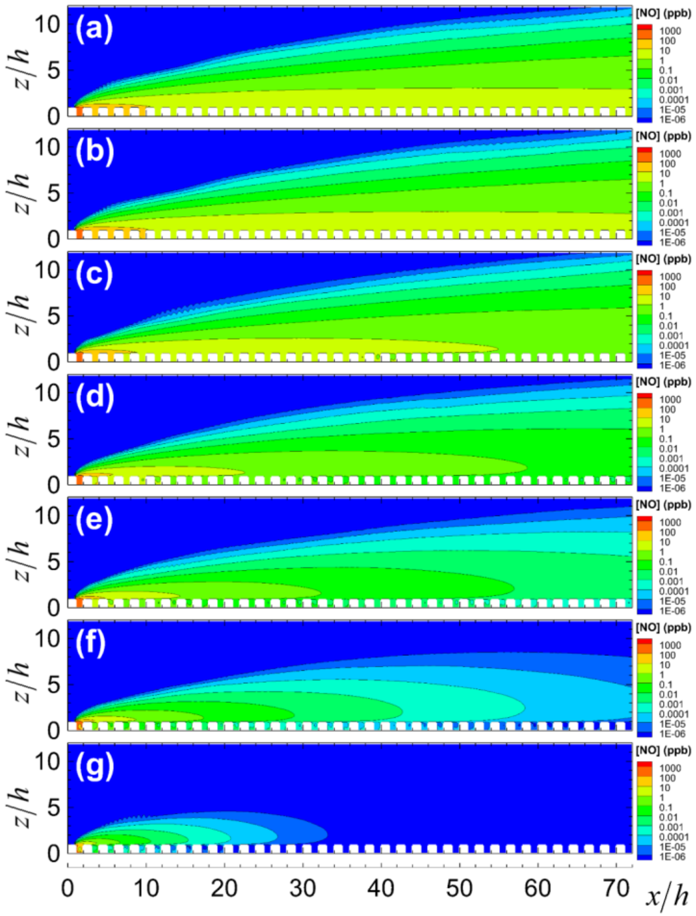


Figure 3. Concentration contours of (a) passive scalar (unit: ppb); (b) NO in case $[O_3]_0$ = 1 ppb; (c) NO in case $[O_3]_0$ = 10 ppb; (d) NO in case $[O_3]_0$ = 50 ppb; (e) NO in case $[O_3]_0$ = 100 ppb; (f) NO in case $[O_3]_0$ = 200 ppb; and (g) NO in case $[O_3]_0$ = 500 ppb.

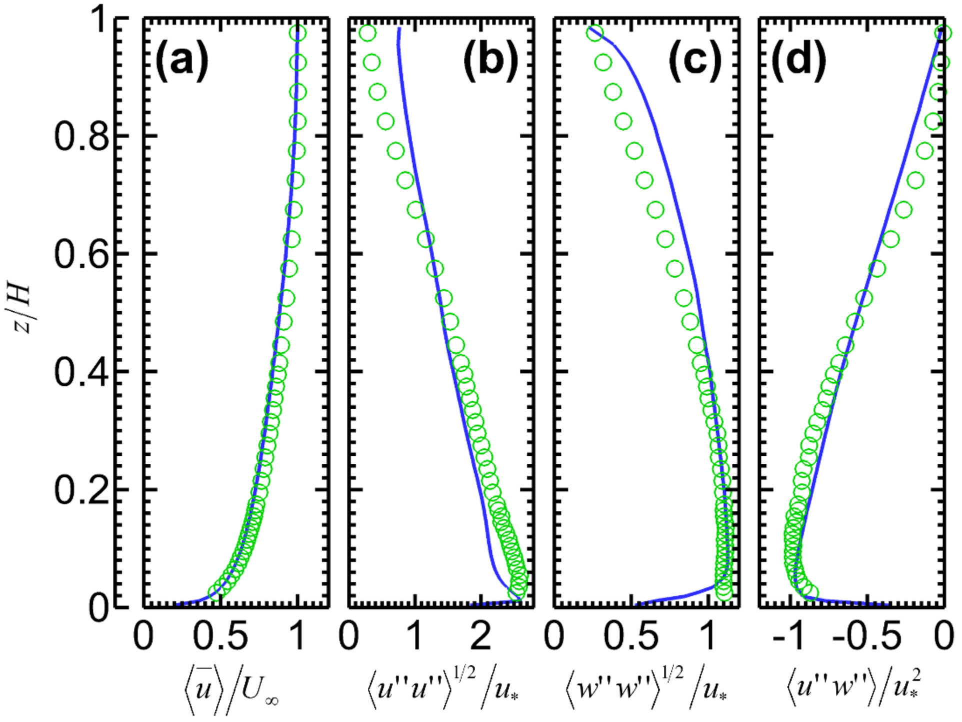
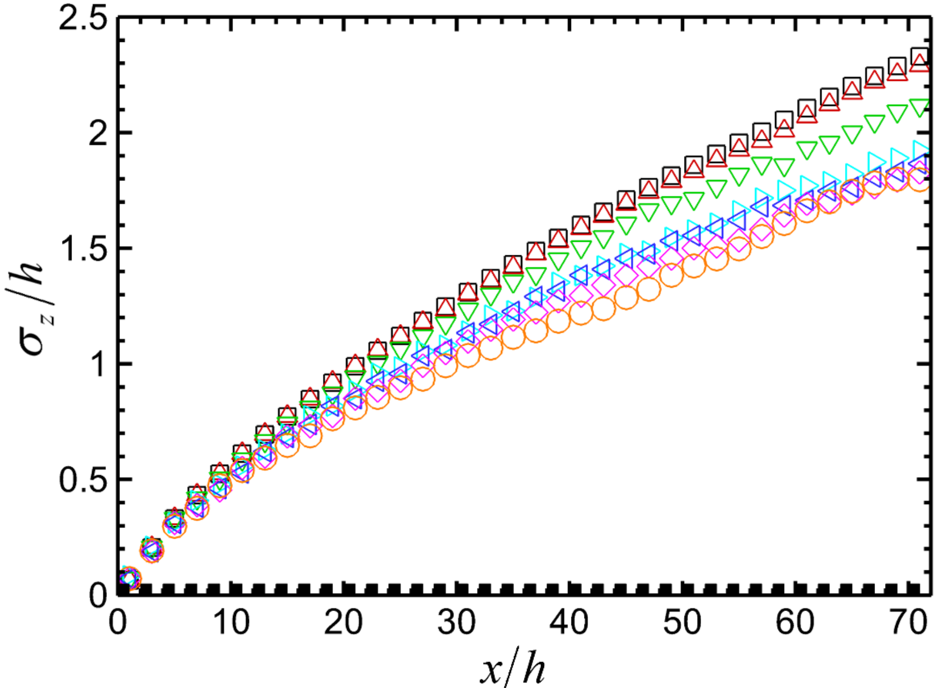
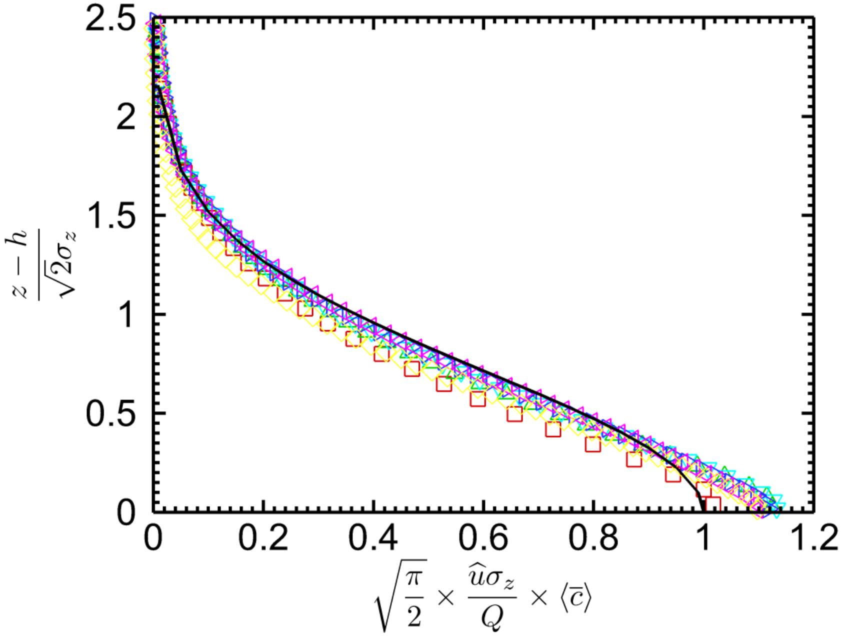


Figure 2. Vertical profiles of (a) mean wind speed; (b) streamwise fluctuating velocity; (c) vertical fluctuating velocity and (d) vertical momentum flux. Solid line: LES data; symbols: wind tunnel data (Ho and Liu 2016).

Results: Plume Characteristic



Dispersion coefficient σ_z of passive scalar (□) and NO in case $[O_3]_0$ = : 1 ppb (△), 10 ppb(▽), 50 ppb(▷), 100 ppb(◁), 200 ppb(◇), 500 ppb(○).



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 (——)

- To parameterize the plume characteristic, the dispersion coefficient σ_z is adopted to represent the length scale of the plume coverage.
- Dimensionless profiles of passive scalar concentration is depicted as functions of height z at different streamwise locations x. The current-LES calculated passive scalar concentration agrees well with the theoretical Gaussian-form solution.

$$\sigma_z = \sqrt{\frac{\iiint (z - z_c)^2 \langle \overline{\phi} \rangle \, dydtdz}{\iiint \langle \overline{\phi} \rangle \, dydtdz}}$$

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

Results: Plume Characteristic

- For passive scalar, Gaussian form fits well. For the chemically reactive pollutant, Gaussian form cannot explain the region below the plume center. Above the plume center, Gaussian form can still fit well.

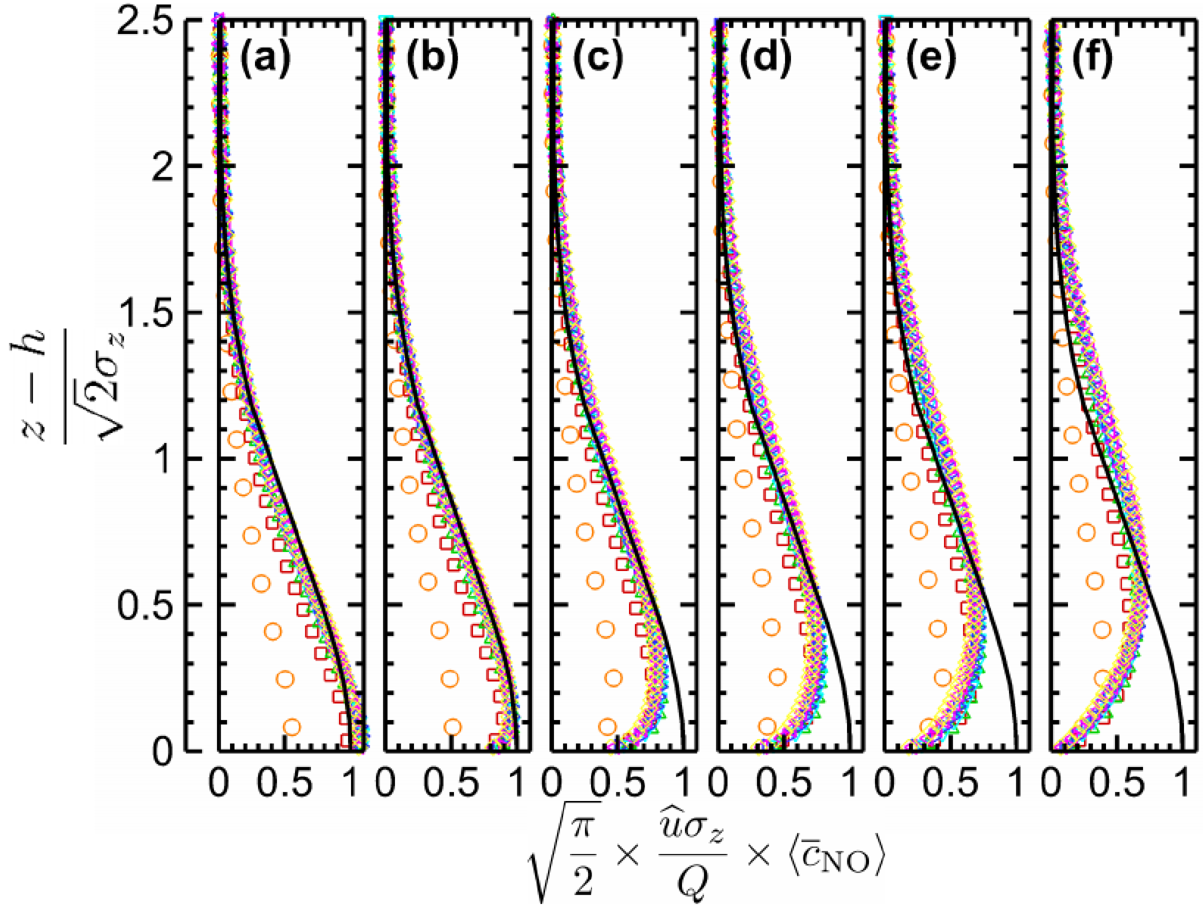


Figure 5: Profiles of dimensionless concentration of reactive pollutant NO at $x/h =$: 15.5 (□), 25.5 (△), 35.5 (▽), 45.5 (▷), 55.5 (◁) and 65.5 (○) for background ozone concentration $[O_3]_0$ = (a) 1 ppb, (b) 10 ppb, (c) 50 ppb, (d) 100 ppb, (e) 200 ppb and (f) 500 ppb. Also shown is the theoretical Gaussian plume profile (——).

NO Transport in Reactive Plume

- The Convection-Diffusion equation with source/sink term:

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

Advection Term | Diffusion Term | Source/Sink Term

- The simple ozone titration is used as the chemistry model:

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

$$\frac{d[NO]}{dt} = -k_3[O_3][NO]$$

$$\frac{d[O_3]}{dt} = -k_3[O_3][NO]$$

Sink of NO and O3

$$\frac{d[NO_2]}{dt} = k_3[O_3][NO]$$

Source of NO2

NO Transport in Reactive Plume

- Take NO as an example

$$\frac{\partial[NO]}{\partial t} + U_i \frac{\partial[NO]}{\partial x_i} = K \frac{\partial^2[NO]}{\partial x_i^2} - k_3[NO][O_3]$$

- Define characteristic scale

Symbol	$[NO]_0$	$[O_3]_0$	L_0	U_0	T_0
Description	Characteristic concentration scale for NO	Characteristic concentration scale for O_3	Characteristic length scale	Characteristic velocity scale	Characteristic time scale for transport

- Dimensionless equation for NO transport

$$\frac{1}{T_0}\frac{\partial[\overline{\overline{NO}}]}{\partial \overline{\overline{t}}} + \frac{U_0}{L_0}\overline{\overline{U_i}}\frac{\partial[\overline{\overline{NO}}]}{\partial \overline{\overline{x_i}}} = K\frac{1}{L_0^2}\frac{\partial^2[\overline{\overline{NO}}]}{\partial \overline{\overline{x_i}}^2} - [O_3]_0 k_3[\overline{\overline{NO}}][\overline{\overline{O_3}}]$$

NO Transport in Reactive Plume

- When advection dominate the NO transport, $T_0 = L_0/U_0$

$$\frac{\partial[\overline{\overline{NO}}]}{\partial \overline{\overline{t}}} + \overline{\overline{U_i}}\frac{\partial[\overline{\overline{NO}}]}{\partial \overline{\overline{x_i}}} = \frac{1}{Sc\,\mathrm{Re}}\frac{\partial^2[\overline{\overline{NO}}]}{\partial \overline{\overline{x_i}}^2} - Da_{NO}[\overline{\overline{NO}}][\overline{\overline{O_3}}]$$

$Sc = \nu / K$ — Schmidt number

$\mathrm{Re} = U_0 L_0 / \nu$ — Reynolds number

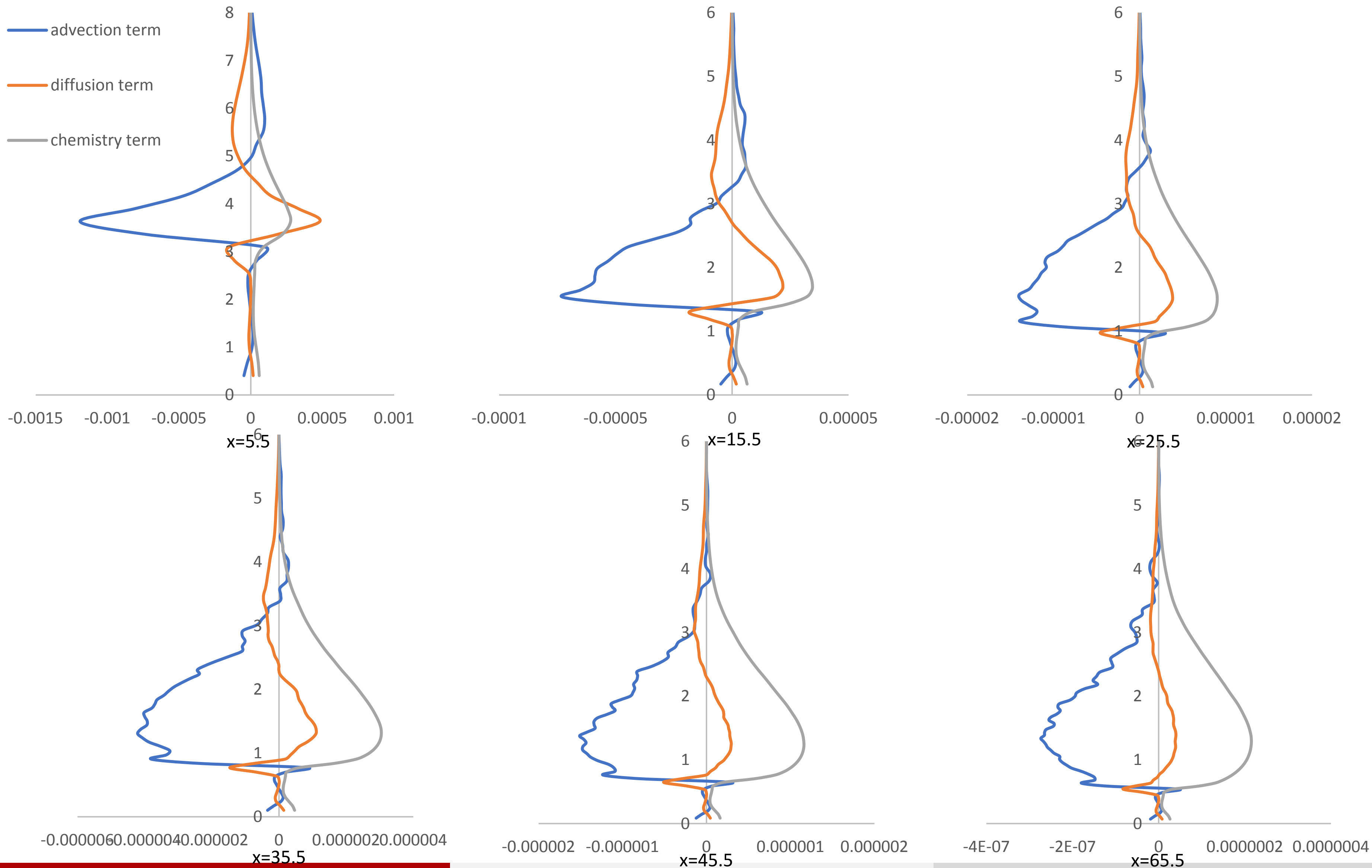
$Da_{NO} = T_0 / \tau_{NO} = L_0 k_3 [O_3] / U_0$ — Damköhler number

- Take the ensemble-average and assume the pseudo-steady state condition:

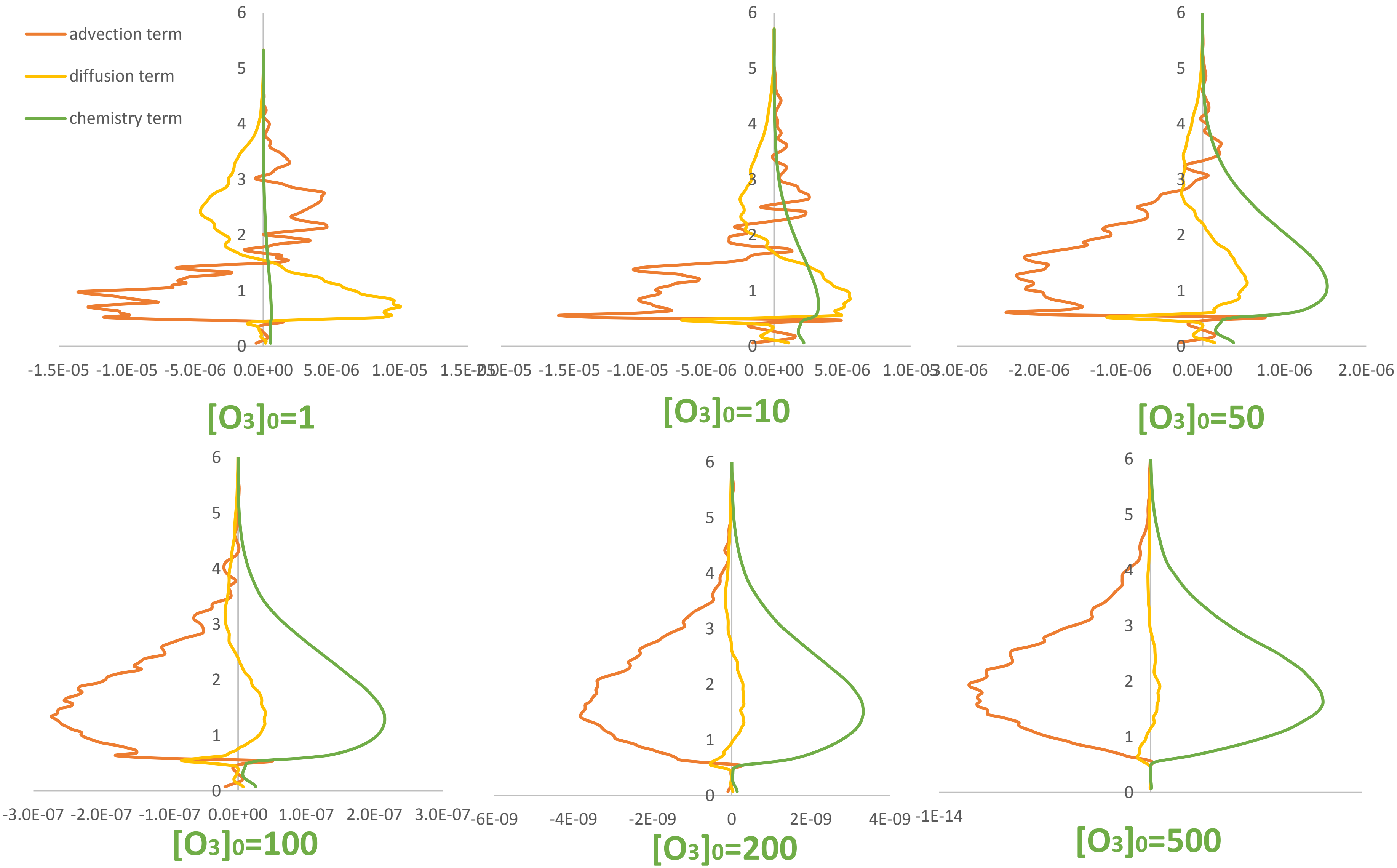
$$\left\langle \overline{\overline{U_i}} \right\rangle \frac{\partial \left\langle [[\overline{\overline{NO}}] \right\rangle}{\partial \overline{\overline{x_i}}} = \frac{K_{NO}}{U_0 L_0}\frac{\partial^2 \left\langle [\overline{\overline{NO}}] \right\rangle}{\partial \overline{\overline{z}}^2} - Da_{NO}\left\langle [\overline{\overline{NO}}] \right\rangle \left\langle [\overline{\overline{O_3}}] \right\rangle$$

Angle brackets ‘< >’ represent the ensemble-averaged value.

Budget Analysis in AR=1,$[O_3]_0$=100 ppb in different *x* location



Budget Analysis in AR=1,with different $[O_3]_0$, at x=66.5



Conclusion

- A NO-O_3 chemistry model is developed to investigate the mechanism of chemically reactive pollutant transport.
- LES data are validated by the wind tunnel experiments.
- Using the modified Gaussian plume model, chemically reactive pollutant still fits in the region above the plume center in the far field.
- Budget analysis shows the correlation between the advection term, diffusion term and chemistry term, implying the significance of chemistry in the pollutant removal.

Thanks!