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

**Zhangquan WU & Chun-Ho LIU***
**Department of Mechanical Engineering**
**The University of Hong Kong**

**Session: Air Quality in Urban Boundary Layer : Processes**
**21 July 2015, 11:00am - 12:30pm**
**Toulouse, France**

******Corresponding Author*: Chun-Ho LIU; Department of Mechanical Engineering, 7/F Haking Wong Building, The University of Hong Kong, Pokfulam Road, HONG KONG;
*Tel*: +852 2859 7901; *Fax*: +852 2858 5415; liuchunho@graduate.hku.hk

# Outline

- Background
- Objectives
- Methodology
- Results
- Conclusion & Outlook

# Urban Climate

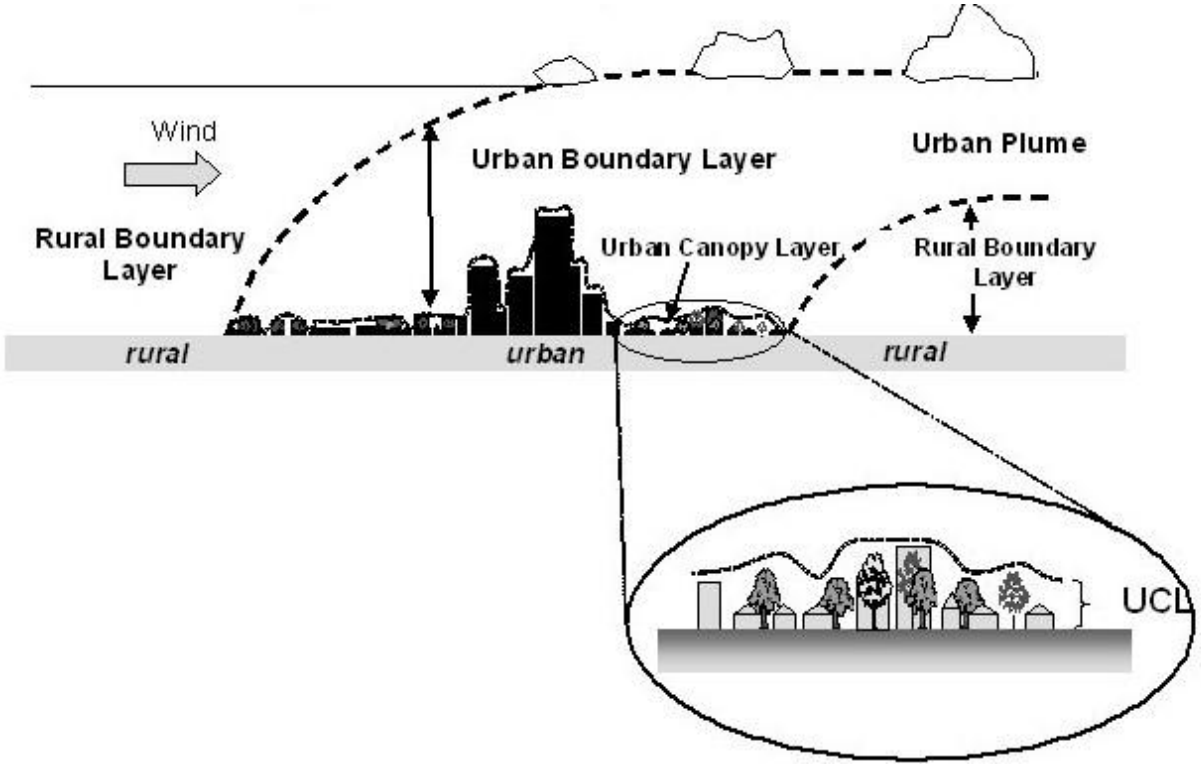



Fig.1

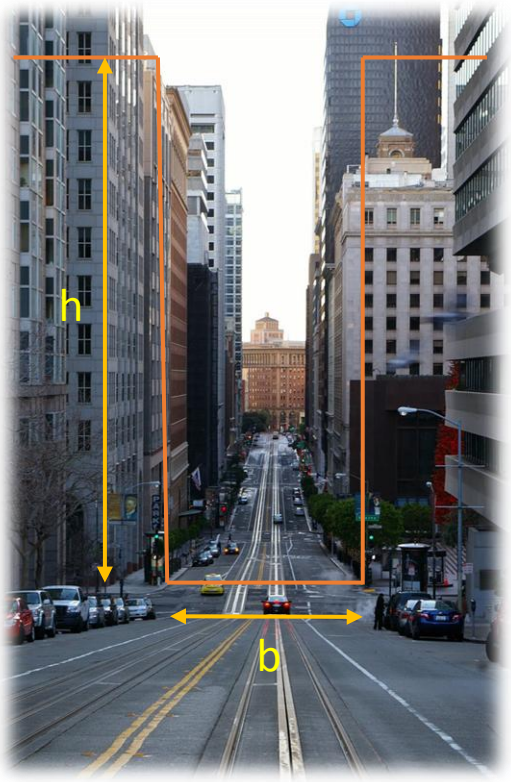



Fig.2

- High pollution levels in urban street canyons due to the increased traffic emissions
- The main traffic-related pollutants are CO, NOx, hydrocarbons, and particles

**Street canyon**
Street height-to-width ratio
AR=h/b

# Street Canyon

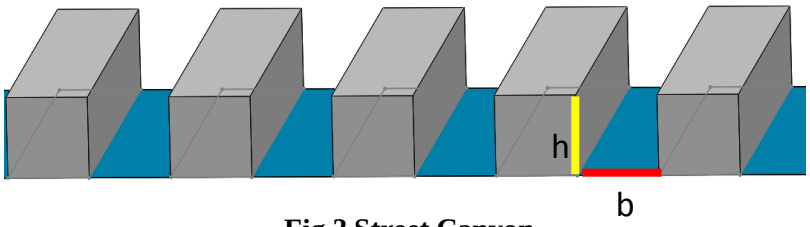



**Fig.3 Street Canyon**

Idealized street canyon is an infinitely long street flanked by high-rise buildings. Its building-height-to-street-width ratio, h/b, is a key geometric factor determining the flow regime.

(1) skimming (h/b > 0.7, isolated recirculations)
(2) wake interference (0.3 < h/b < 0.7, entrainment)
(3) isolated roughness (h/b < 0.3, reattachment and separation)

These regimes characterize the mechanisms of flow, transport and pollutant removal from the street level to the atmospheric boundary layer (ABL) through the urban canopy layer (UCL).

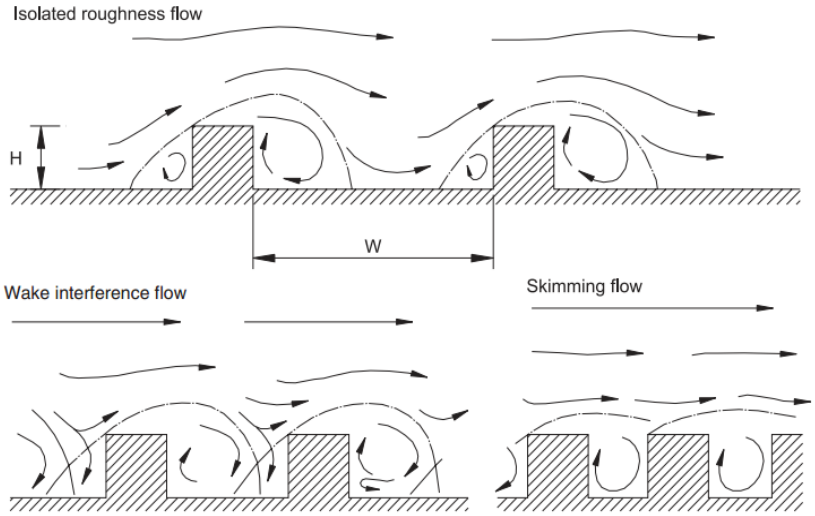



**Fig.4 Flow regime**

# Objectives

- Formulate a CFD model for simple $NO_x$-$O_3$ chemistry
- Look into the pollutant concentrations and dispersion characteristics over the street canyons
- Analyze the characteristics of plume dispersion of passive scalar and chemically reactive pollutant
- Examine the relative contribution between chemistry and turbulent dilution to NO consumption along the plume dispersion using time scale analysis

# Large-Eddy Simulation

- Reynolds-averaged Navier–Stokes equations (RANS)
  - the full range of turbulence scales is simplified to one parameterization.
  - cost-effective but merely captures the important aspects of recirculating flow in street canyons

- Large-Eddy Simulation (LES)
  - calculates explicitly the large-scale energy-carrying turbulence
  - small portions of the fluxes are modeled by subgrid-scale (SGS) models
  - Improved prediction accuracy of the pollutant dispersion compared with that of the $k$–$\varepsilon$ models.

- Direct Numerical Simulation (DNS)
  - computes the whole spectrum of turbulence
  - unaffordable expense of lengthy computation time and costly resources.

Subgrid-scale (SGS) model  One-equation model (OEM)

# Subgrid-scale (SGS) model

Spatial filtering

$$f = \overline{f} + f' \qquad \overline{f}(x_i,t) = \int_\Omega f(x_i,t)G(x_i - \xi_i, \overline{\Delta})d\xi_i$$

$\bar{f}$: the large filtered-scale component

$f'$: the SGS component

Conservation of momentum

$$\frac{\partial u_i}{\partial t} + \frac{\partial}{\partial x_j}u_i u_j = -\frac{\partial p}{\partial x_i} + \frac{1}{\mathrm{Re}}\frac{\partial^2 u_i}{\partial x_j \partial x_j} \quad \Rightarrow \quad \frac{\partial u_i}{\partial t} + \frac{\partial}{\partial x_j}\overline{u}_i\overline{u}_j = -\frac{\partial \overline{p}}{\partial x_i} - \frac{\partial \tau_{ij}}{\partial x_j} + \frac{1}{\mathrm{Re}}\frac{\partial^2 \overline{u}_i}{\partial x_j \partial x_j}$$

The continuity

$$\frac{\partial u_i}{\partial x_i} = 0 \quad \Rightarrow \quad \frac{\partial \overline{u}_i}{\partial x_i} = 0$$

The scalar transport equation

$$\frac{\partial c}{\partial t} + \frac{\partial u_i c}{\partial x_i} = \frac{1}{\mathrm{Re}\,Sc}\frac{\partial^2 c}{\partial x_i \partial x_i} \quad \Rightarrow \quad \frac{\partial \overline{c}}{\partial t} + \frac{\partial \overline{u}_i \overline{c}}{\partial x_i} = -\frac{\partial \sigma_i}{\partial x_i} + \frac{1}{\mathrm{Re}\,Sc}\frac{\partial^2 \overline{c}}{\partial x_i \partial x_i}$$

$t$: time
$u_i$: velocity components
$p$: kinematic pressure
Re: Reynolds number

$Sc = \nu / D$ Schmidt number

$\tau_{ij} = \overline{u_i u_j} - \overline{u}_i \overline{u}_j$ SGS stress

# Chemistry Model

$$\frac{\partial C_a}{\partial t} + U_j \frac{\partial C_a}{\partial x_j} = \frac{\partial}{\partial x_j}\left(D \frac{\partial C_a}{\partial x_j}\right) + S_a(\boldsymbol{C})$$

→ Source term

Here $C$ denotes a vector of all the reacting species in the flow.

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

$$\frac{\delta[O_3]}{\delta t} = j_1 - k_3[O_3][NO]$$

In the presence of sunlight

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

$$\frac{\delta[NO_2]}{\delta t} = k_3[O_3][NO] - j_1$$

independent of solar radiation

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

$$\frac{\delta[NO]}{\delta t} = j_1 - k_3[O_3][NO]$$

# Geometry Information

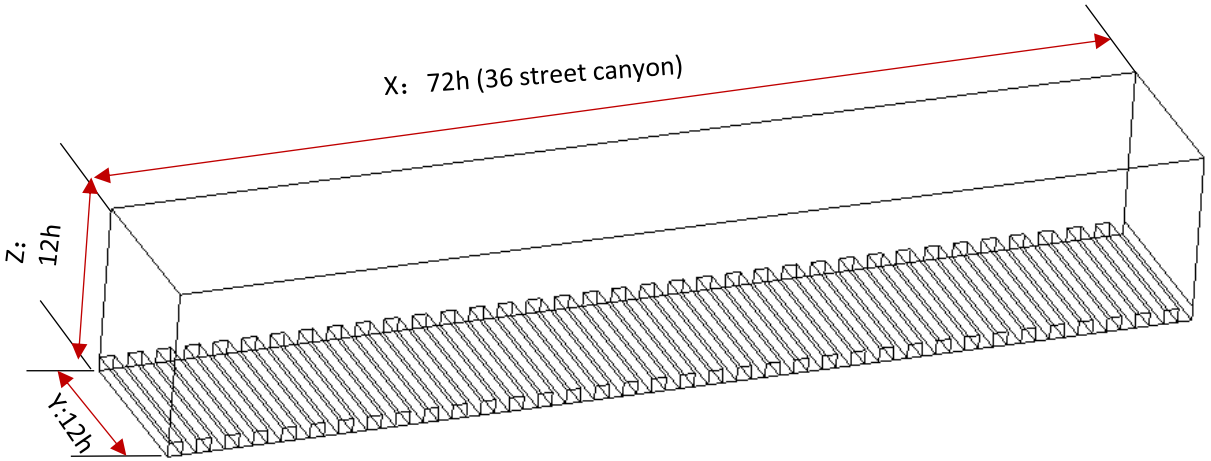



**Fig.5 Computational Domain**

| Description | Parameter | Value |
|---|---|---|
| Domain Length | X | 72h |
| Domain Width | Y | 12h |
| Domain Height | Z | 12h |
| Building Height | h | h |
| Street Width | b | h |
| Number of Street Canyons | n | 36 |

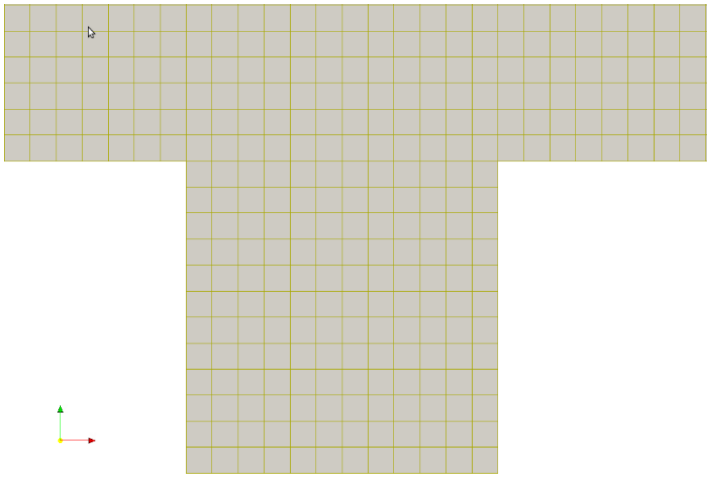
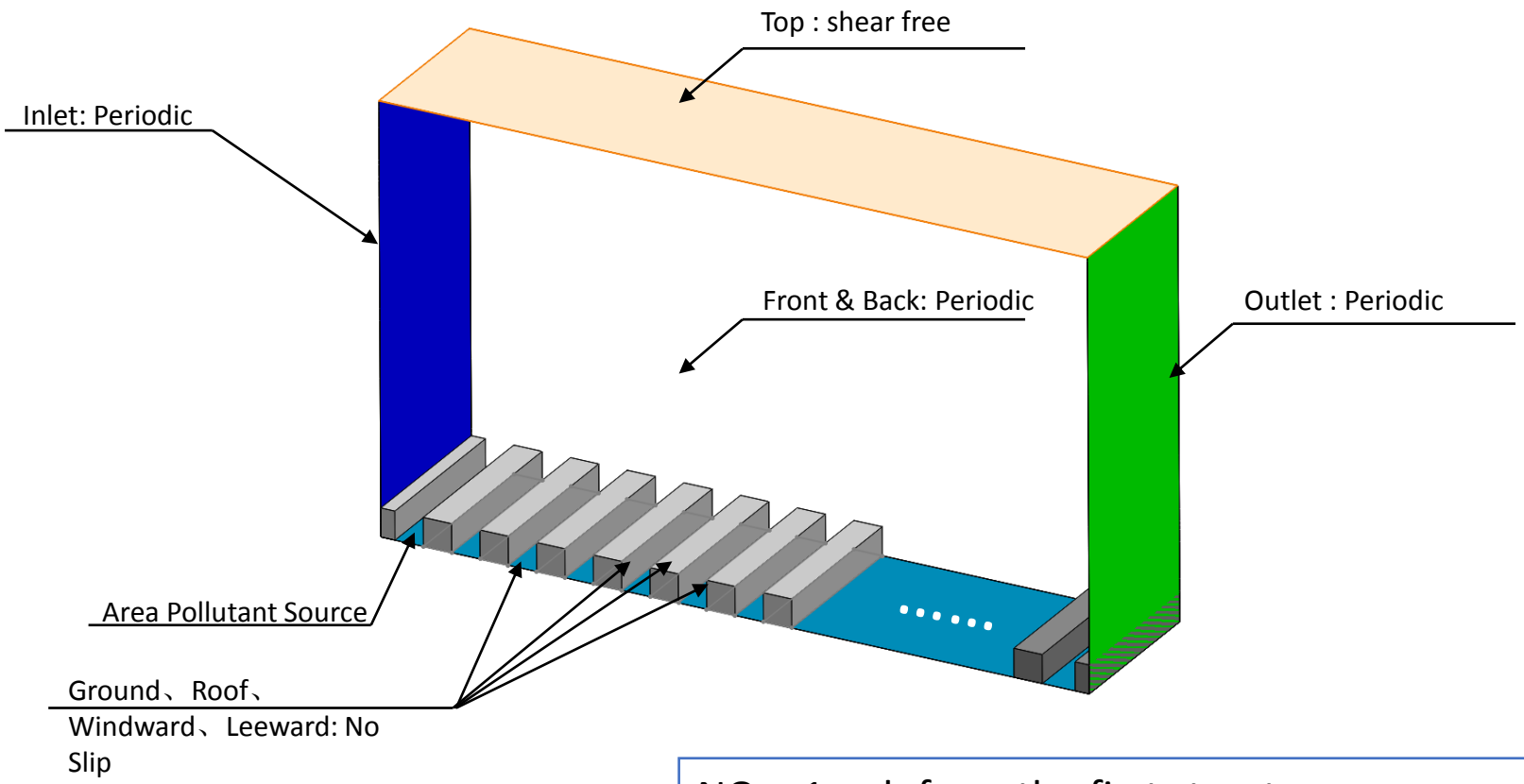


**Fig.6 Grid information in Street Canyon**

| Description | Parameters | Value |
|---|---|---|
| Grid Number | Nx<br>Ny<br>Nz | 864<br>96<br>84 |
| Grid Number in Street Canyon | Ncx<br>Ncz | 12<br>12 |
| Total Grid Number | N | 6,400,000 |

# Boundary Conditions




NO： 1 ppb from the first street canyon
$NO_2$: set as zero
$O_3$: 1 ppb with the coming wind

**Fig.7 Boundary Condition**

# Velocity & NO Field

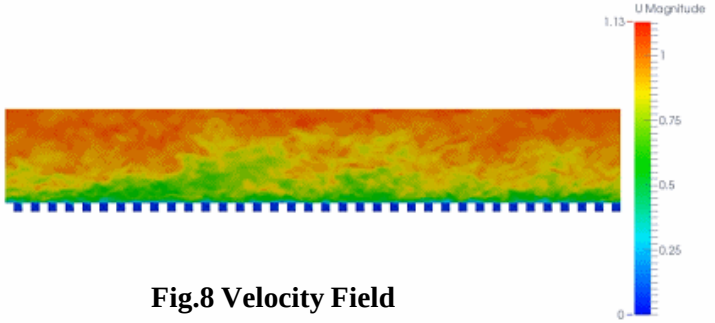



**Fig.8 Velocity Field**

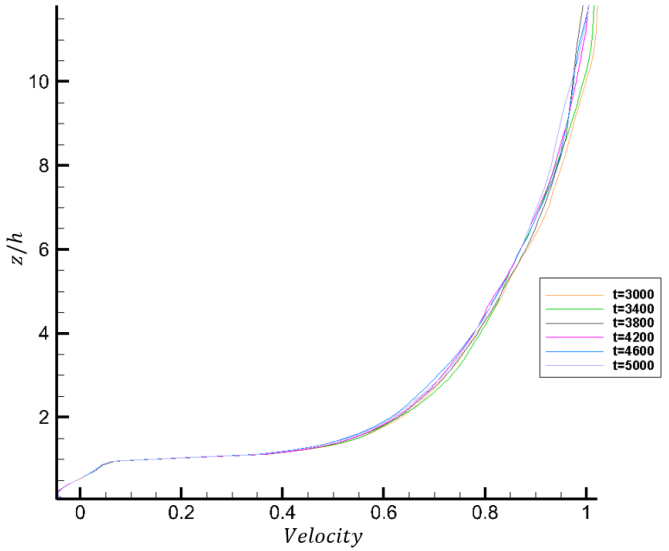



**Fig.10 Vertical Velocity Profile**

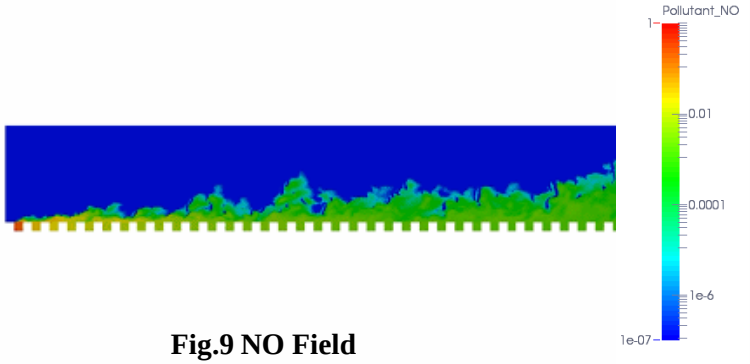



**Fig.9 NO Field**

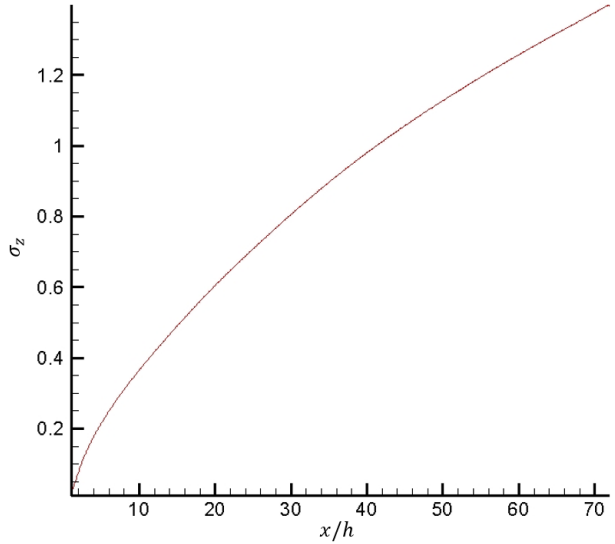



**Fig.11 Dispersion Coefficient $\sigma_z$**

# Reaction time scale

| | | |
|---|---|---|
| $NO_2 + h\nu \rightarrow NO + O$ | photolysis rate $j_1$ | In the presence of sunlight |
| $O + O_2 + M \rightarrow O_3 + M$ | rate constant $k_2$ | Independent of solar radiation |
| $O_3 + NO \rightarrow NO_2 + O_2$ | rate constant $k_3$ | |

- Ground-state oxygen atom O is highly reactive such that its formation is fast enough to counterbalance its depletion.

- Reaction time scale of NO: $\tau_{NO} = \frac{1}{k_3[O_3]_0}$

- Reaction time scale of O3: $\tau_{O_3} = \frac{1}{k_3[NO]_0}$

$k_3$ =4.43e-4$ppb^{-1}sec^{-1}$ → $\tau_{NO} = \tau_{o_3} = 2257$

# Diffusion Time Scale

- **Mass conservation of species：**

$$\frac{\partial c_i}{\partial t} + \frac{\partial}{\partial x_j}(u_j, c_i) = D_i \frac{\partial^2 c_i}{\partial x_j \partial x_j} + R_i(c_1, \dots, c_N, T) + S_i(x, t)$$

$c_i$:species concentration of interest
$D_i$: molecular diffusion constant
$R_i$: rate of $c_i$ generated reaction
$S_i$: rate of $c_i$ generated by source

- There exists a closure approximation in introducing the concept of eddy diffusivity and its statistical approximations for the random concentration and velocity components from turbulent mixing.

$$\langle u_j^{"} c_i^{"} \rangle = -K \frac{\partial \overline{c_i}}{\partial x_j}$$

$K$ is the eddy diffusivity

- **For passive scalar** $\emptyset$

$$\langle w"\emptyset" \rangle = -K_\emptyset \frac{\partial \langle \overline{\emptyset} \rangle}{\partial z} \qquad K_\emptyset = -\frac{\langle w"\emptyset" \rangle}{\frac{\partial \langle \overline{\emptyset} \rangle}{\partial z}} \qquad \tau_d = \frac{\sigma_z^2}{4K_\emptyset}$$

- **For pollutant NO**

$$\langle w"NO" \rangle = -K_{NO} \frac{\partial \langle \overline{NO} \rangle}{\partial z} \qquad K_{NO} = -\frac{\langle w"NO" \rangle}{\frac{\partial \langle \overline{NO} \rangle}{\partial z}} \qquad \tau_{dNO} = \frac{\sigma_{zNO}^2}{4K_{NO}}$$

# Time scale of NO

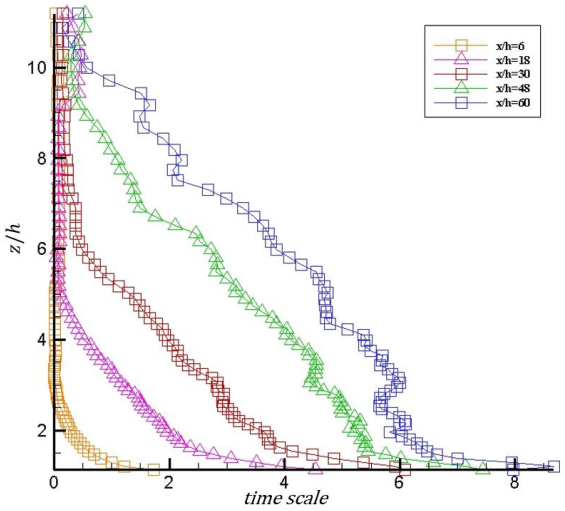



**Fig.12 Diffusion time scale of NO**

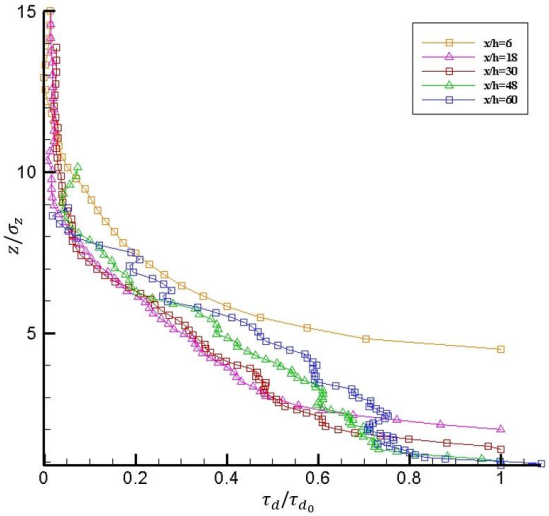



**Fig.13 Normalized Diffusion time scale of NO**

- The diffusion time scale $\tau_d$ of NO (normalized by the diffusion time scale near the roof level $\tau_{d0}$ at $z = 1.1h$)
- Near the first street canyon ($x = 6h$), $\sigma_z$ is relatively small and the plume is not yet fully developed.
- The pollutant concentration gradient is small near the domain top so the diffusion time scale remains small from $z = 3h$ and thereafter.
- 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 ($z/\sigma_z \geq 10$)

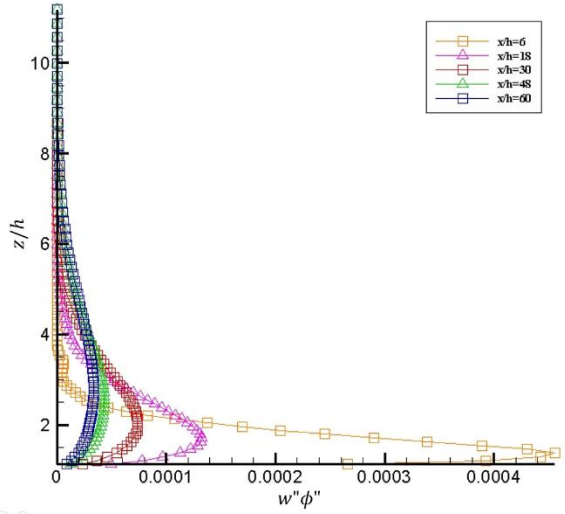



**Fig.14 Pollutant flux w”ϕ”**

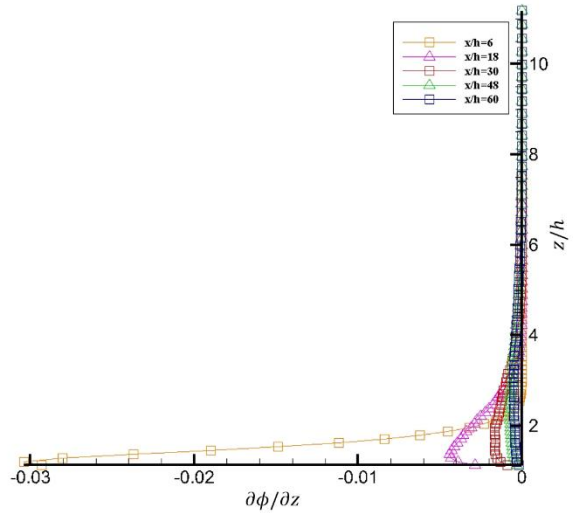



**Fig.15 Gradient of mean pollutant**

# Time scale of NO

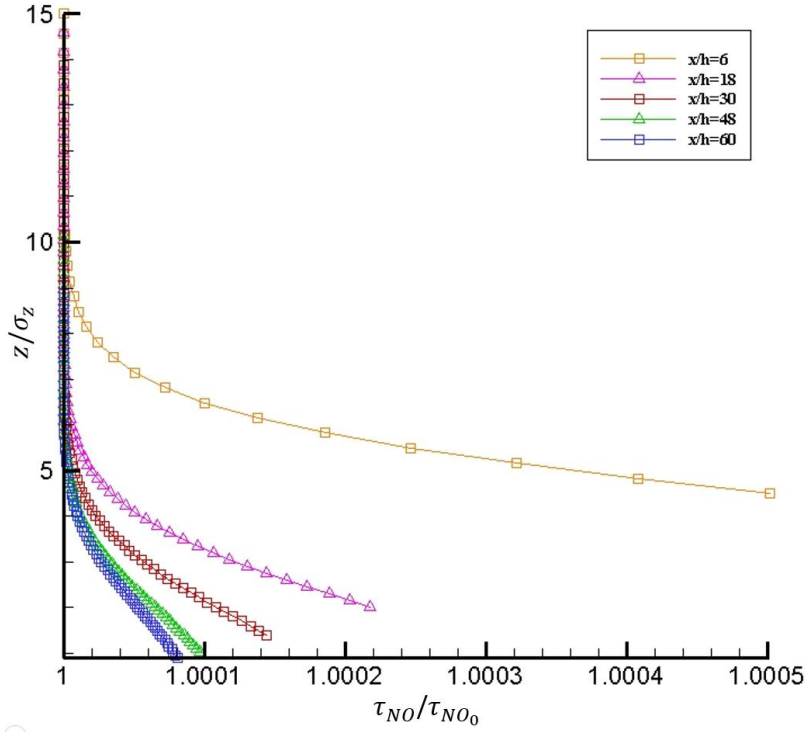



**Fig.16 Reaction time scale of NO**

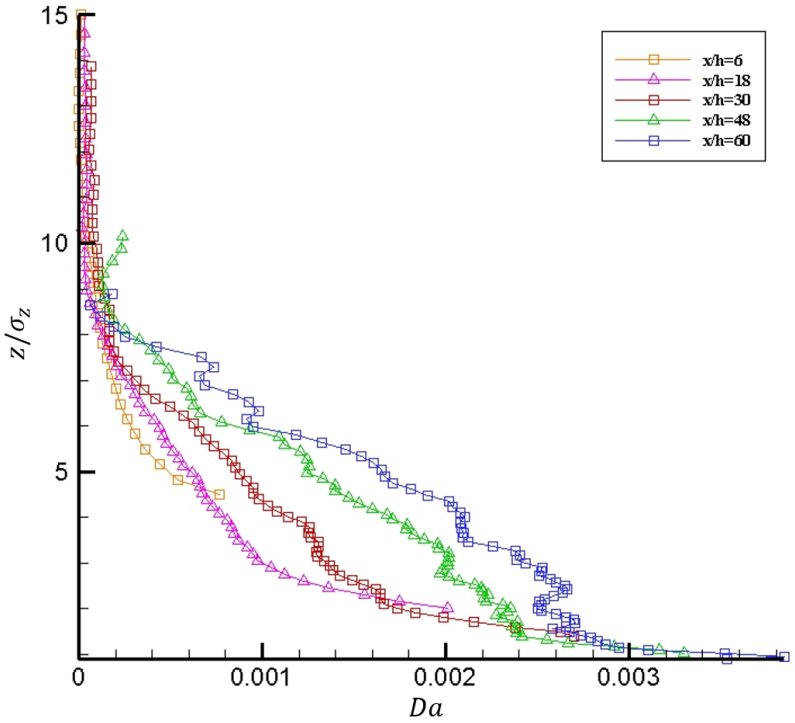



**Fig.17 Damköhler number**

- The $O_3$ concentration is lower than that in the upper domain
- 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

$$Da_{\mathrm{NO}} = \frac{\tau_d}{\tau_{\mathrm{NO}}} = \frac{\sigma_z^2}{4K_{\mathrm{NO}}} k_3[\mathrm{O_3}]$$

- Da>1: The pollutants are not mixed uniformly for complete reactions.
- Da<1: slow chemical reactions occur in the well-mixed condition.

# Branching point

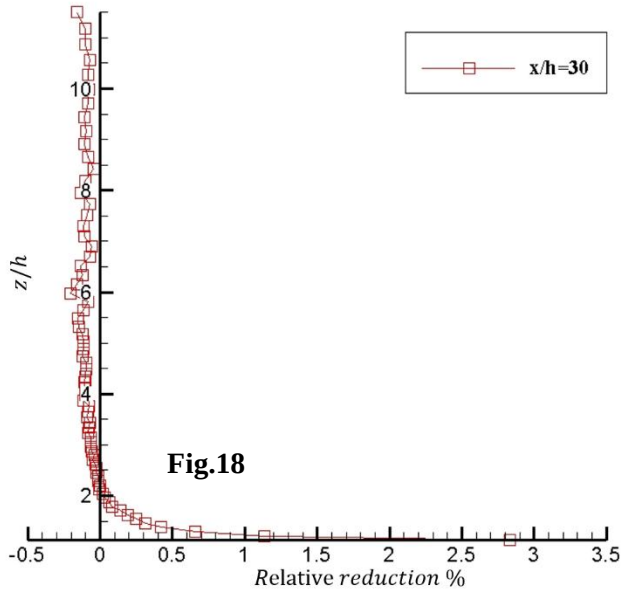



Fig.18

**Fig.18 Relative reduction in diffusion time scale at x/h=30**

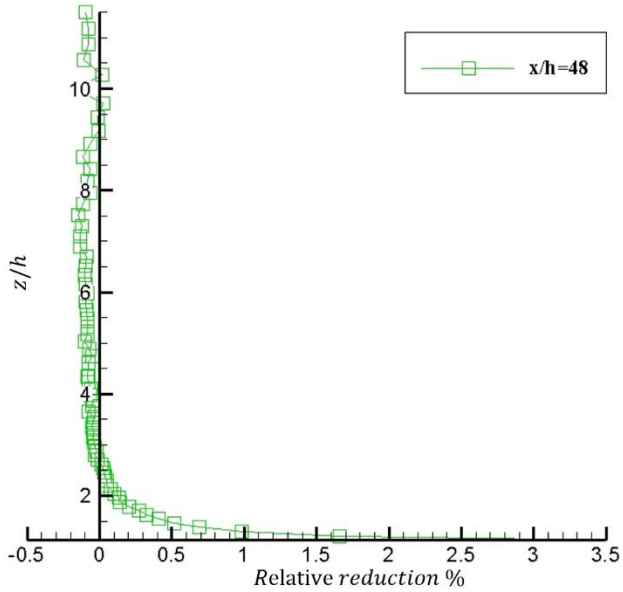



**Fig.19 Relative reduction in diffusion time scale at x/h=48**

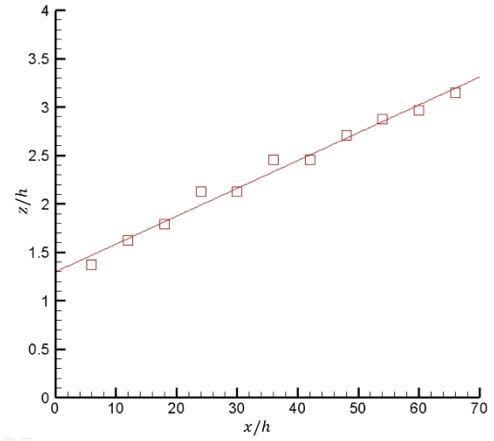



**Fig.20 Branching point along x/h**

$$\text{Relative Reduction } \% = \frac{\tau_{\text{passive scalar}} - \tau_{\text{NO}}}{\tau_{\text{passive scalar}}} \times 100\%$$

- A passive scalar is used the reference for reactive NO to investigate the effect of chemical reactions on the plume dispersion.
- Near the roof level, the chemical reaction tends to reduce diffusion time scale.
- There exists a level for this effect over which the diffusion time scale is increased by chemical reaction.
- The trajectory of branching point works as a function of streamwise distance $x$ in a linear relation

# Conclusion & Outlook

- The reaction time scale is much longer compared with the turbulent time scale when the initial concentration is small.(NO = $O_3$ = 1ppb)
- Two time scales are coupled with each other. In particular, pollutant chemistry could increase or decrease the diffusion time scale, i.e. dispersion coefficient.
- More different concentration cases are needed to analyze the relative contribution between chemistry and turbulent dilution to NO consumption along the plume dispersion.

# Thank you!

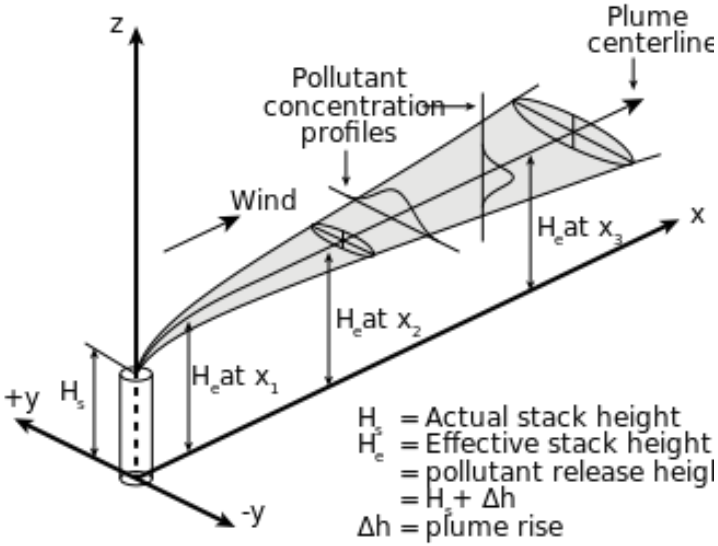
# Q&A

**Acknowledgment**

The first author (Z Wu) gratefully acknowledges the Hong Kong Research Grants Council (RGC) for financially supporting his study through the Hong Kong PhD fellowship (HKPF) scheme. This research is conducted using the HKU Information Technology Services research computing facilities that are supported in part by the Hong Kong UGC Special Equipment Grant (SEG HKU09). This project is partly supported by the RGC GRF HKU 714913E.


Zhangquan WU
wzqmec@gmail.com
The University of Hong Kong

# Gaussian Plume Model




**Point source**

$$C(x,y,z) = \frac{Q}{2\pi u \sigma_y \sigma_z} exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left\{exp\left[-\frac{(z-H)^2}{2\sigma_z^2}\right] + exp\left[-\frac{(z+H)^2}{2\sigma_z^2}\right]\right\}$$

**Line source**

$$C(x,y,z) = \int_{y1}^{y2} \frac{Q}{2\pi u \sigma_y(s)\sigma_z(s)} exp\left(\frac{-z^2}{2\sigma_z^2(s)} - \frac{(y-s)^2}{2\sigma_y^2(s)}\right) ds$$

σy: standard deviation of the concentration in the spanwise direction

σz: standard deviation of the concentration in the vertical direction

- The larger σ means the broader distribution. Further from the source the plume becomes broader and so the value of σ is greater.
- The Gaussian plume pollutant dispersion model can gives a fairly good estimate of the pollutant concentration when using accurate parameters.
- σy and σz are related to atmospheric turbulence, surface roughness and distance from the pollutant source.

# σz: Dispersion Coefficient

$$\sigma_z^2 = \frac{\iiint (z - z_c)^2 \emptyset dydtdz}{\iiint \emptyset dydtdz}$$

time and spanwise average

$$\sigma_z^2 = \frac{\int (z - z_c)^2 \emptyset dz}{\int \emptyset dz}$$

$$z_c = \frac{\int z \emptyset dz}{\int \emptyset dz}$$

$$\sigma_z^2 = \frac{\sum_{i=1}^{N} (z_i - z_c)^2 \emptyset_i \Delta z_i}{\sum_{i=1}^{n} \emptyset_i \Delta z_i}$$

$$z_c = \frac{\sum_{i=1}^{N} z_i \emptyset_i \Delta z_i}{\sum_{i=1}^{n} \emptyset_i \Delta z_i}$$

$\emptyset_i$:concentration at a certain position
$z_c$: plume center height

Basically, $\sigma_z$ is the length scale of the pollutant plume width