Time Scale Analysis of Chemically Reactive Pollutants over Urban Roughness in the Atmospheric Boundary Layer

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• According to Hong Kong Environmental Protection Department (EPD), the total emission of NO\textsubscript{x} in Hong Kong in 2013 is about 113,220 tonnes.
• The emission sources of NO\textsubscript{x} in Hong Kong include power electricity generation, road transport, navigation, civil aviation, and other fuel combustion sources.
• Road transport is one of the major sources of NO\textsubscript{x}.

• While most practical dispersion models assume inert pollutants, emissions from traffic exhaust are chemically reactive.
• NO can be oxidized by ozone (O\textsubscript{3}) in the atmosphere.
• With sunlight, NO\textsubscript{2} can also decompose into NO and O\textsubscript{2}.
• Chemical reaction in the atmosphere is much more complicated than that.

Data Source: http://www.epd.gov.hk/epd/english/environmentinhk/air/data/emission_inve.html
Elevated pollutant concentrations are commonly observed in urban areas, such as street canyons, threatening human health. Dynamics are complicated by atmospheric turbulence, geometry/orientation of buildings, thermal stratification and chemical kinetics, etc. The oxidation rate of NO is affected by both physical process and chemical process. The physical process is mixing of the plume with the ambient air. The chemical processes are the molecular reactions of NO with species in the surrounding air. There exists a wide range of turbulent eddies which act on dispersing plume. Dispersion and mixing of material, which constitute the plume, are caused by eddies of all sizes in the atmospheric boundary layer.

- Large eddies (larger than the cross-section of plume, in the order of tens to several hundreds of meters): cause meandering of the plume
- Middle eddies (about the size of cross-section of plume or smaller): cause broadening and internal mixing of plume.
- Small eddies (in the order of millimeters): important for chemical reaction
Objectives

• Develop a CFD model for simple NO$_x$-O$_3$ chemistry

• Analyze the plume dispersion characteristics of passive scalar and chemically reactive pollutant.

• Analyze the plume characteristics in different cases with different O$_3$ concentration.

• Compare the time scales of diffusion and chemical reaction
Methodology

• Model
  Large-eddy simulation with one-equation SGS model

• Governing equations (filtered)
  • continuity equation
    \[ \frac{\partial \bar{u}_i}{\partial x_i} = 0 \]
  • momentum conservation
    \[ \frac{\partial \bar{u}_i}{\partial t} + \frac{\partial}{\partial x_j} \bar{u}_i \bar{u}_j = -\Delta \bar{P} \delta_{ij} - \frac{\partial \bar{p}}{\partial x_i} + (\nu + \nu_{SGS}) \frac{\partial^2 \bar{u}_i}{\partial x_j \partial x_j} \]
  • Transport equation for pollutant
    \[ \frac{\partial \phi}{\partial t} + \bar{u}_j \frac{\partial \phi}{\partial x_j} = \frac{\partial}{\partial x_j} \left( D \frac{\partial \phi}{\partial x_j} \right) + S(\phi) \]

• First step is to handle irreversible chemical reaction
  \[ \text{NO} + \text{O}_3 \xrightarrow{k_3} \text{NO}_2 + \text{O}_2 \]

• Source term for NO, NO\textsubscript{2} and O\textsubscript{3}
  \[ \frac{d[\text{NO}]}{dt} = -k_3[\text{O}_3][\text{NO}] \quad \frac{d[\text{O}_3]}{dt} = -k_3[\text{O}_3][\text{NO}] \]
  \[ \frac{d[\text{NO}_2]}{dt} = k_3[\text{O}_3][\text{NO}] \]
Geometry and Boundary Conditions

Computational Domain

Boundary Condition

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<thead>
<tr>
<th>Description</th>
<th>Parameter</th>
<th>Value</th>
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<tbody>
<tr>
<td>Domain Length</td>
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<tr>
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<td>12h</td>
</tr>
<tr>
<td>Domain Height</td>
<td>Z</td>
<td>12h</td>
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<tr>
<td>Building Height</td>
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<td>1h</td>
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<tr>
<td>Street Width</td>
<td>b</td>
<td>1h</td>
</tr>
<tr>
<td>Number of Street Canyons</td>
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</table>

<table>
<thead>
<tr>
<th>Case NO.</th>
<th>NO Concentration /ppb</th>
<th>O3 Concentration /ppb</th>
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<tbody>
<tr>
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<tr>
<td>Case 2</td>
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<td>10</td>
</tr>
<tr>
<td>Case 3</td>
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<td>Case 4</td>
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<tr>
<td>Case 6</td>
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Velocity scale: friction velocity  \( u_* = \sqrt{\tau_w / \rho} \)  \( \tau_w \) is the wall shear stress.

1. The vertical velocity profile fits well with that of the wind tunnel measurement.
2. The turbulence in LES is smaller than that of wind tunnel measurement for \( z \leq 0.6h \). It is because the turbulence in the wind tunnel is not only generated from roughness but also from upstream.
1. $w''$ is important in the momentum transport and pollutant transport in the vertical direction.

2. $w''w''$ in LES fits well with the wind tunnel data especially in the inertial sublayer.
1. The background O$_3$ mixed with NO from the area source in a molecular scalar.
2. When background [O$_3$] increases, the plume of NO is compressed.
3. In case [O$_3$] = 500 ppb and [O$_3$] = 1000 ppb, almost all the NO is consumed by the background O$_3$
1. The concentration of pollutant is normalized by the pollutant concentration at the roof level.
2. Near the roof level, the characteristics of concentrations of NO and passive scalar are very different.
3. Perhaps it is because O₃ entrains below the roof level. Near the roof level, the chemical reaction is quite fast that consumes more NO.
\( \sigma_z \) is the vertical dispersion coefficient. In the current study, it is calculated by the following equation.

\[
\sigma_z = \sqrt{\iiint (z - z_c)^2 \phi \, dy \, dt \, dz} / \iiint \phi \, dy \, dt \, dz
\]

\( z_c \) is the plume center height (the location of maximum concentration is used as the plume center height).
Diffusion time scale

Molecular/turbulent transport and mixing processes are involved in the plume dispersion in the atmosphere. The time scale of dispersion reflects the effects of different processes taking place on a wide spectrum of scales.

- Diffusion time scale

\[ \tau_d = \frac{\sigma_z^2}{4K} \]

- For passive scalar

\[ \sigma_z = \left( \frac{2Kx}{U} \right)^{1/2} \]

- So we can derive the diffusion time scale

\[ \tau_d = \frac{2Kx}{4KU} = \frac{x}{2U} \]

Reaction time scale

Reaction time scale is the time for the chemical reaction to take place.

\[ \tau_{NO} = -\frac{1}{k_3 \langle [O_3] \rangle} \]
In the cases $[O_3]_0 = 1000$ ppb and $[O_3]_0 = 500$ ppb, the minimum reaction time scale is 2.26 second and 4.53 second, which is much smaller than the diffusion time scale away from the pollutant source. The fast chemistry implies that most of NO is consumed by $O_3$ titration.

In the cases $[O_3]_0 = 1$ ppb and $[O_3]_0 = 10$ ppb, the reaction time scale is much longer than the diffusion time scale. Physical dispersion thus dominates the pollutant removal.
Time scale

- In the cases $[O_3]_0 = 50$ ppb and $[O_3]_0 = 100$ ppb, the reaction time scale and the diffusion time scale are comparable.

- In the case $[O_3]_0 = 100$ ppb, in the near field next to the pollutant source, diffusion time scale is smaller and diffusion dominates the pollutant removal. In the far field, the reaction time scale is smaller, thus chemical reaction dominates the pollutant removal.
1. The behavior of the passive scalar and NO is different. It shows that the chemical reaction has major effects on the dispersion behavior of pollutant.

2. The current LES model is validated with the wind tunnel experiments. The velocity data in LES fits well with that of the wind tunnel measurement.

3. The vertical concentration profile of reactive pollutant is different from that of passive scalar. The chemical reaction enhances the plume height and reduces the dispersion coefficient compared with that of the passive scalar. The dispersion coefficient of chemically reactive pollutant is weaker than its inert counterpart so pollution chemistry reduces plume width and coverage.

4. At a low level of $[O_3]_0 (= 1 \text{ ppb})$, $\tau_{NO}$ is longer than $\tau_d$ so physical dispersion dominates pollutant removal. For $[O_3]_0$ at 100 ppb, the physical and chemistry timescales are comparable with each other.
Thank you!