Atmospheric Chemical Reactions in Turbulent Flows: Application to an Urban Environment

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Introduction

- Turbulent mixing controls the segregation and hence the rate of chemical reaction of tracers
- Chemical-turbulence interaction is a subgrid process, and is often neglected by large-scale chemical transport models, leading to miscalculation of tracer production
- Subgrid chemical-turbulence interaction is investigated in a fundamental representation, in planetary boundary layer, and in an urban setting in this project

Theory

- The effect of turbulent motions on chemical reaction can be estimated from the ratio between the turbulent and chemical timescales, or the **Damköhler number**

$$Da = \frac{\tau_{turb}}{\tau_{chem}}$$

- For the reactions with ***Da* << 1**, the tracers are efficiently mixed and react with **chemical timescale**. For the fast chemical reactions with ***Da* >> 1**, the tracers are not well mixed by turbulence before they react, and therefore react with the **turbulent timescale**
- The interaction between chemistry and turbulence can be represented by the **Reynolds averaged continuity equation**[1]:

$$\frac{\partial \bar{C}_i}{\partial t} + \frac{\partial \overline{w'C_i'}}{\partial z} = \nu_c \frac{\partial^2 \bar{C}_i}{\partial z^2} + \sum_{j=1}^{N} k_{ij}\bar{C}_j + \sum_{j=1}^{N}\sum_{k=1}^{j} k_{ijk}\left(\bar{C}_j\bar{C}_k + \overline{C_j'C_k'}\right) + Q_i$$

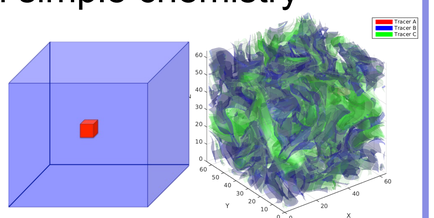
, where $(C_i^{'}, w_i^{'})$ are the turbulent components of concentration C_i and velocity field w_i, ν_c is the molecular diffusivity, k_{ij} and k_{ijk} are the uni-molecular and bi-molecular reaction coefficient

- The covariance terms involve in **closure problems**[2], such as $\overline{C_i'C_j'} = \left(\frac{\overline{C_iC_j}}{\bar{C}_i\bar{C}_j} - 1\right)\bar{C}_i\bar{C}_j$ **,** and are often **parametrized** but these single-valued schemes are often over-simplified and may lead to miscalculation of tracer production

Subgrid chemical-turbulence interaction: A fundamental representation

Simulation settings:

- 3D tracer advecting in turbulent field with simple chemistry (**A** + **B** → **C**)
- Constantly emitted sources of **Tracer A** at centre and **Tracer B** at background



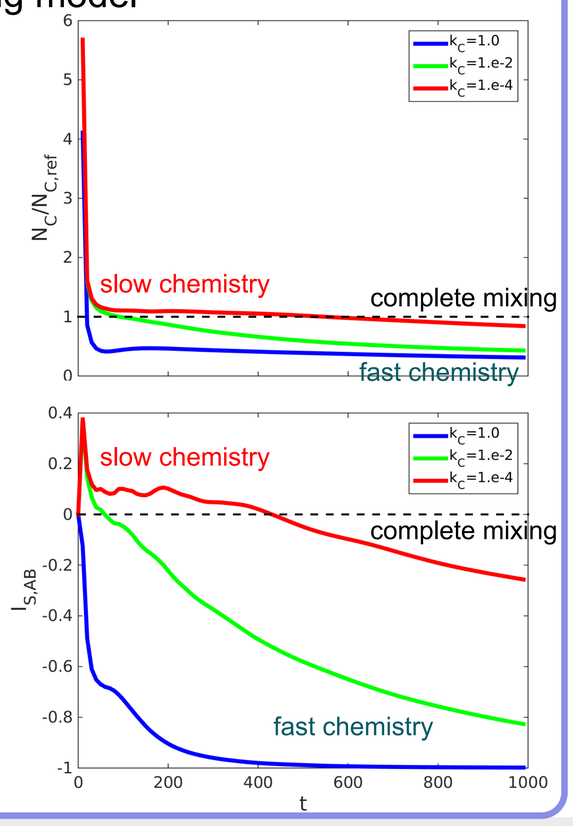
Initial condition At t = 500

Evaluation parameters:

1. Intensity of segregation between Tracer A & B $\mathbf{I_{S,AB}} = \frac{C_A'C_B'}{\bar{C}_A\bar{C}_B}$
2. Normalised total amount of Tracer C produced $\boldsymbol{N_C/N_{C,ref}}$, where $N_{C,ref}$ is from the complete-mixing model

Results:

- With a smaller chemical reaction rate k_c (slow chemistry),
  - ➢ the corresponding Damköhler number Da is smaller
  - ➢ tracers are less segregated
  - ➢ production approaches to the value in complete-mixing model
- **N_C is over-estimated** in complete-mixing model except for very slow chemistry



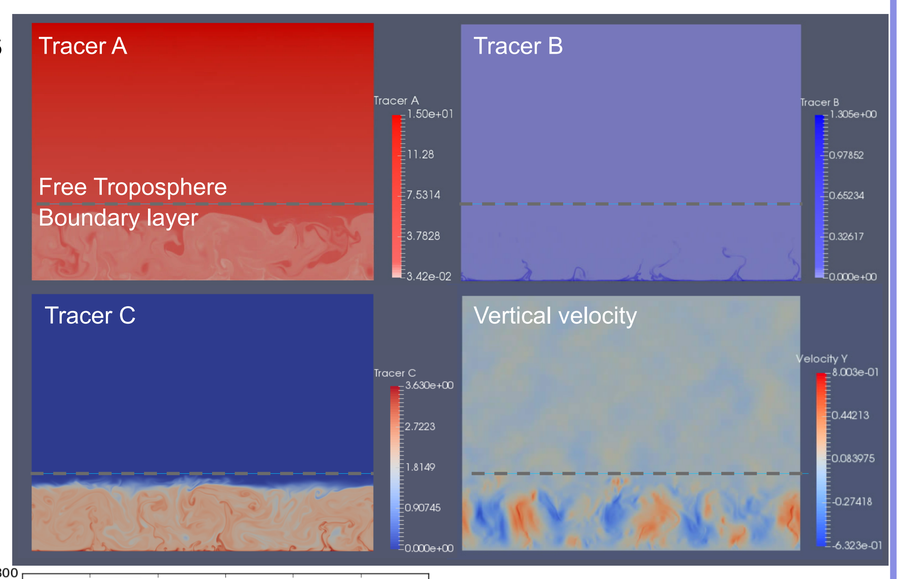
Application in planetary boundary layer

Simulation settings:

- Direct numerical simulation[3] with simple chemistry (**A** + **B** → **C**)
- Entrainment of **Tracer A** from free troposphere and **Tracer B** from the surface

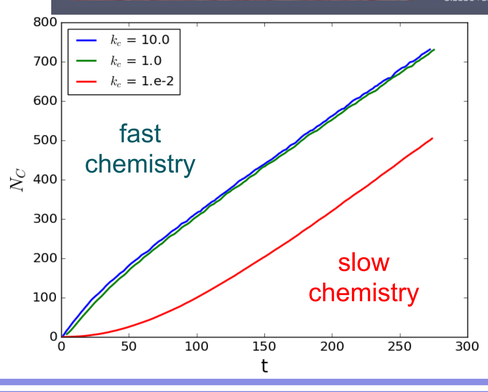
Preliminary results:

- Inhomogeneous production of Tracer C that relates to the regions of updrafts and downdrafts



Density plots of the tracer concentrations and the vertical component of velocity field with k_c = 1.0

- Turbulence-dependent production of Tracer C for fast chemistry



Total amount of Tracer C produced N_C in DNS simulations with varied chemical reaction rate k_c

Application to the region of Hong Kong

- **Complex topography**: Hilly terrains, the largest numbers of skyscrapers in the world, coastal location → **Turbulence**
- **Substantial supply of pollutants**: Intense urbanization, the "World's Factory" in the adjacent Pearl River Delta → **Chemistry**
- **Inhomogeneous emission**: Complex landuse, greenlands between urban areas, heavy transport network
- **High-resolution chemical transport modeling** with resolution up to 100 m and surface topography will be employed to study the chemical-turbulence interaction in the city
- The results from this study can then be compared with the **local observational data** and other related studies done in other scales

Aerial photo of Hong Kong: the photo shows the south-west region of the New Territories and part of the Kowloon Peninsula and the Hong Kong Island. The topography of Hong Kong is complicated with hilly terrains, an extent of high-rise buildings and its surrounding South-China Sea, making the region a highly turbulent urban area.

Next steps…

- DNS simulations with NO_X-O_3 chemistry scheme
- LES simulations with urban-rural emission inhomogeneity
- Simulations with WRF-Chem coupled to LES in Hong Kong with complex urban and terrain topography

References

1. Verver, G. H. L., H. Van Dop, and A. A. M. Holtslag. "Turbulent mixing of reactive gases in the convective boundary layer." *Boundary-Layer Meteorology* 85.2 (1997): 197-222.
2. Stull, Roland B. *An introduction to boundary layer meteorology*. Vol. 13. Springer Science & Business Media, 1988.
3. Garcia, Jade Rachele, and Juan Pedro Mellado. "The two-layer structure of the entrainment zone in the convective boundary layer." *Journal of the Atmospheric Sciences* 71.6 (2014): 1935-1955.

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