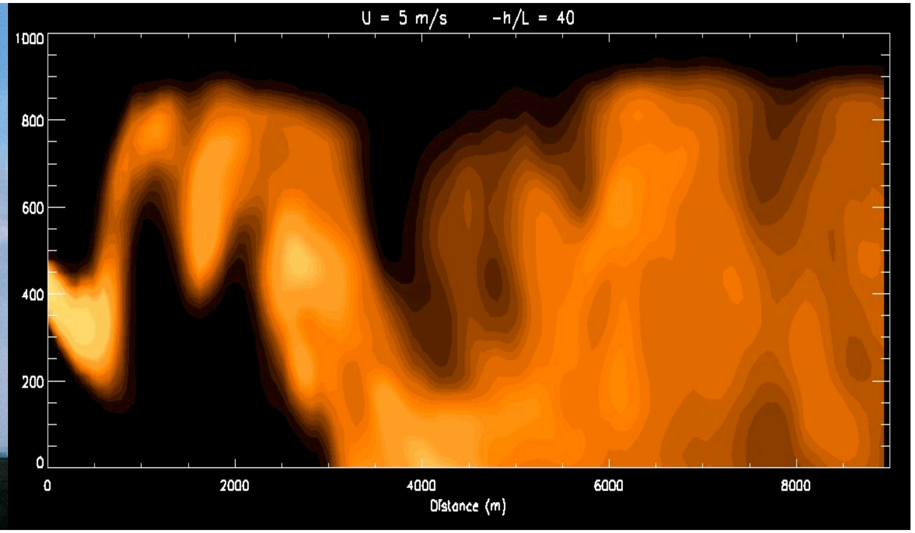
Turbulent dispersion and chemical transformation in the atmospheric boundary layer: Part I: Dry Convection



WAGENINGEN UNIVERSITY
METEOROLOGY AND AIR QUALITY

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Thanks: Alessandro Dosio
Stefano Galmarini

Laminar
Lasagna
(LES)

Turbulent
ratatouille (DNS)

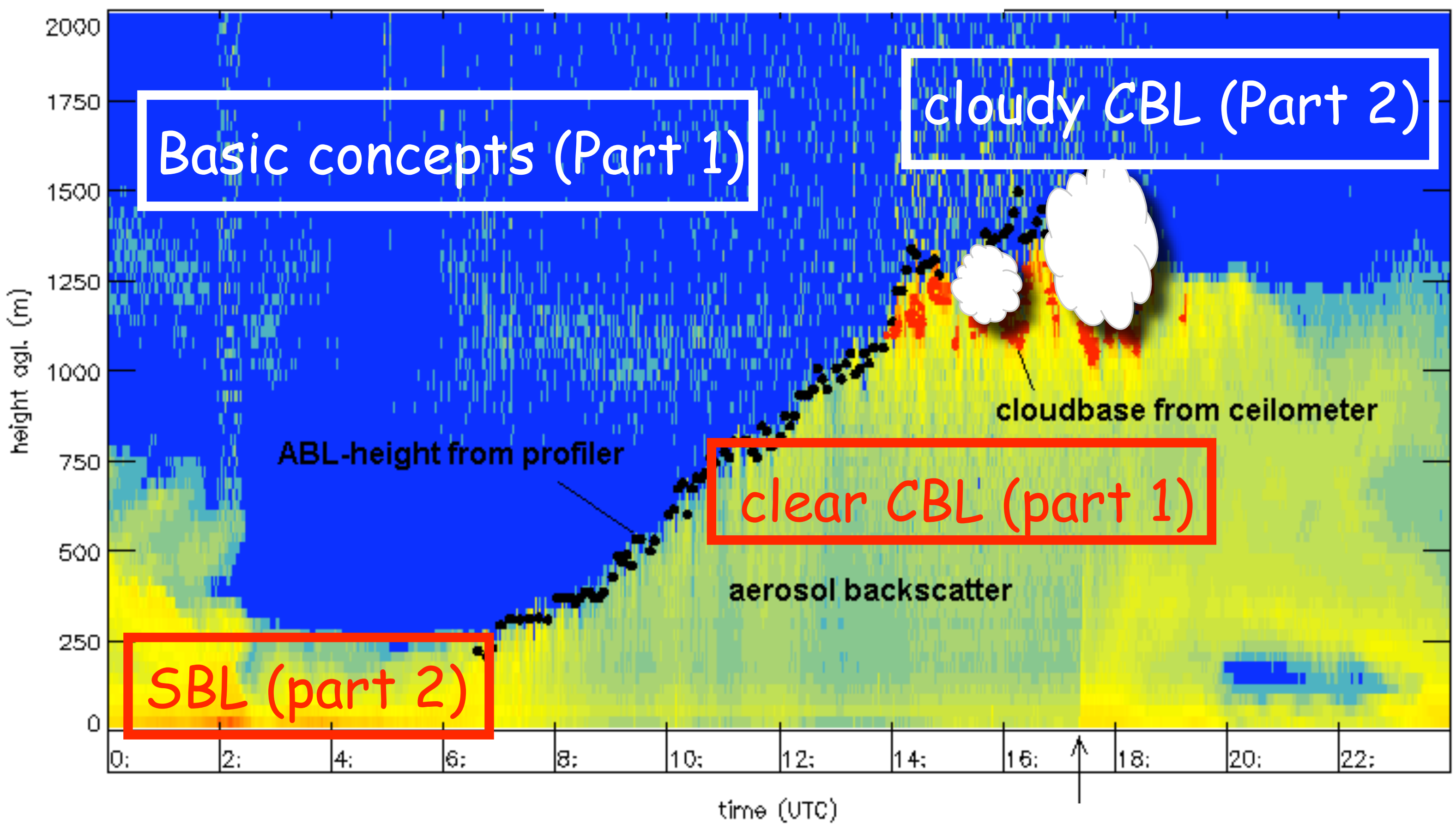
How do the Atmospheric Boundary Layer dynamics influence the transport, mixing, chemical transformation and removal of atmospheric (reactive) compounds?

Greenhouse gases (CO_2, N_2O, CH_4,...)
Reactive compounds(O_3, NO_x, VOC,...)

Scope of the 2 lectures

Discuss in detail how boundary layer structure determines the dispersion and transformation of atmospheric compounds

Organization of the 2 lectures



Basic concepts

Emphasis on connecting ABL dynamics (mainly atmospheric turbulence) to the dispersion and transformation of atmospheric compounds

<u>Dispersion</u>: Lagrangian approach

Taylor's diffusion equation

<u>Chemical transformation</u>: Eulerian approach

Conservation equation of reactants

Plume Dispersion is described by

Mean (1st moment) $\overline{C}(\vec{x},t)$

Spread (2nd moment) $\boxed{\sigma(\vec{x},t)}$

Symmetry (3rd moment) $S(\vec{x},t)$

Taylor's diffusion equation: Autocorrelation

Quantifying the "persistence" of the velocities fluctuations

$$R_j(\tau) = \frac{\overline{u_j(t)u_j(t+\tau)}}{\overline{u_j^2}}$$

$$\boxed{\begin{aligned} \tau = 0 &\rightarrow R(0) = 1 \\ \tau = \infty &\rightarrow R(\infty) = 0 \end{aligned}}$$

(Taylor, 1921)

Taylor's diffusion equation: Lagrangian time scale

Integral time scale that characterizes the energy containing eddies

$$T_j^L \equiv \int_0^{\infty} R_j^L(\tau)\,d\tau$$

Remember than

$$\boxed{T_j^L > T_j^E}$$

Taylor's diffusion equation: dispersion

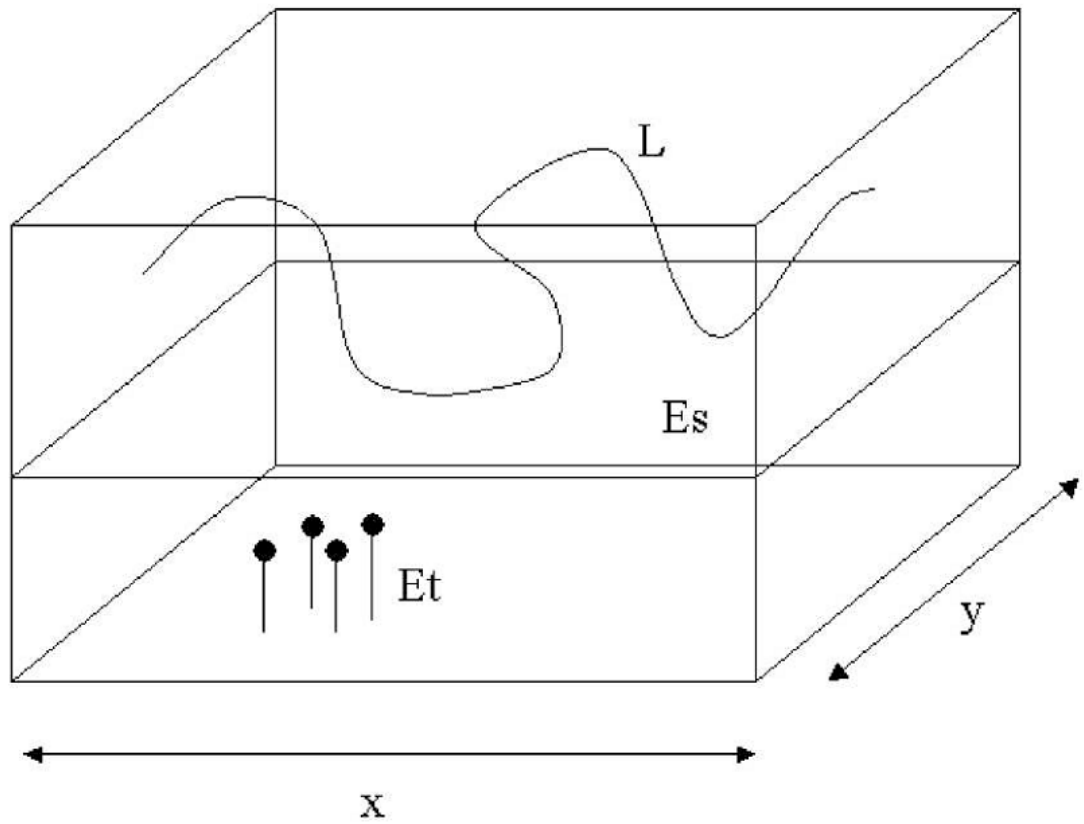
Connecting flow properties to dispersion properties (σ)

If turbulent field is homogenous:

$$\sigma_j^{\,2}(T) = 2\overline{u_j^{\,2}}\int_o^T dt \int_o^t R_j(\tau)d\tau$$

Through out the lecture, examples of $R(\tau)$ and T_L under different ABL conditions

Chemical transformations are easier to be treated in a fix system of coordinates (Eulerian)



Conservation eq. reactants: reaction term

Relevance of the chemical term

$$\frac{\partial C}{\partial t} + u_j \frac{\partial C}{\partial x_j} = \boxed{R(c_1, \ldots, c_n)}$$

Similar to: radiative flux divergences

phase changes

Conservation eq. reactants: <u>physical influences</u>

Turbulence and UV-radiation influence R

$$\frac{\partial \overline{C}}{\partial t} + \overline{U}_j \frac{\partial \overline{C}}{\partial x_j} + \frac{\partial \overline{u_j c}}{\partial x_j} = j\overline{A} - k(\overline{B}\,\overline{C} + \overline{bc})$$

(Averaged equation)

<u>Photolysis</u> j control by UV radiation

<u>Co-variance</u> quantifies how atmospheric turbulence mixes reactants

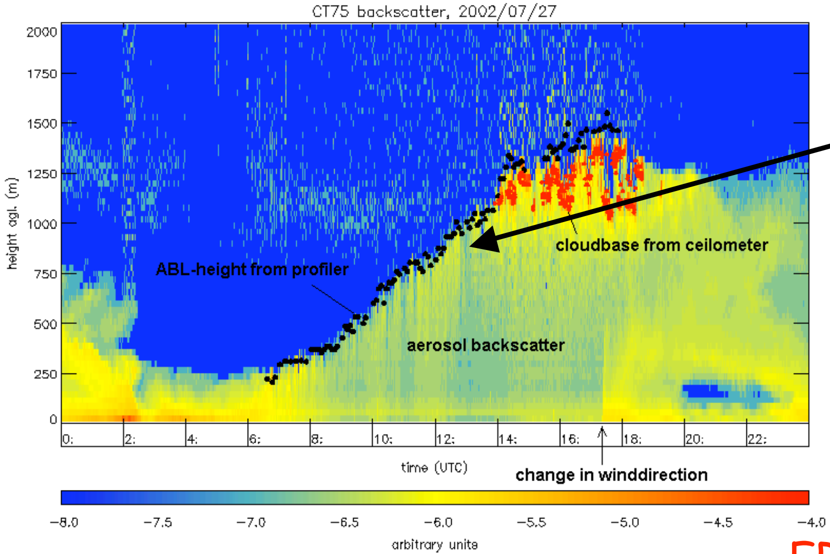
Outline

Refreshing the main characteristics of the convective boundary layer

Turbulent dispersion

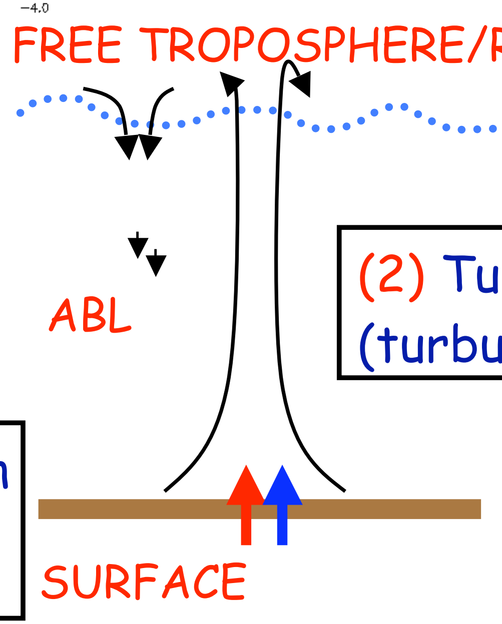
Reactivity

Convective Boundary Layer characteristics



Large diurnal variability

(3) Entrainment (exchange fluxes)



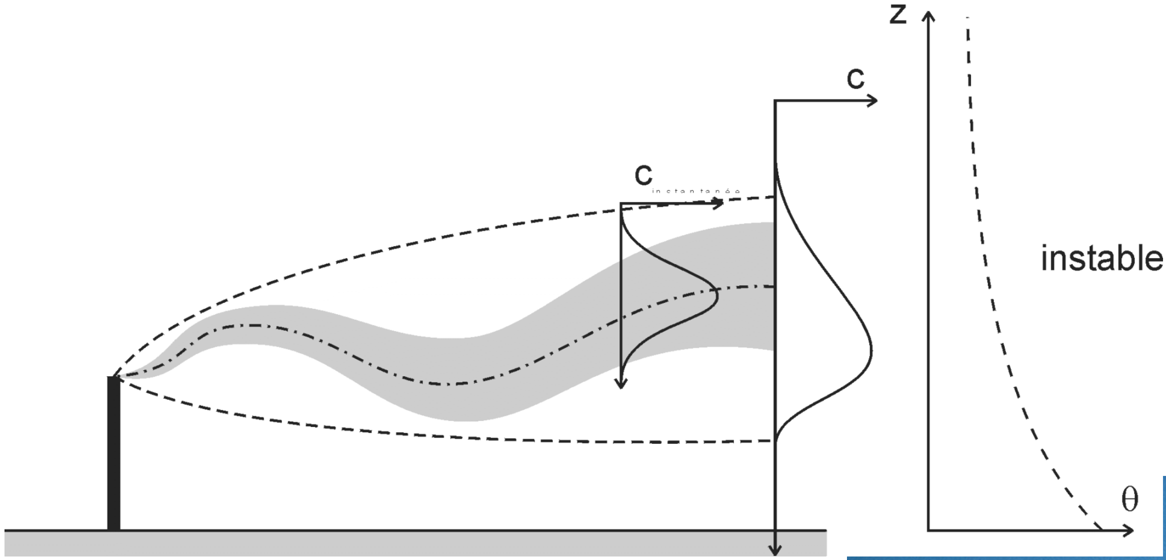
(2) Turbulent mixing (turbulent fluxes)

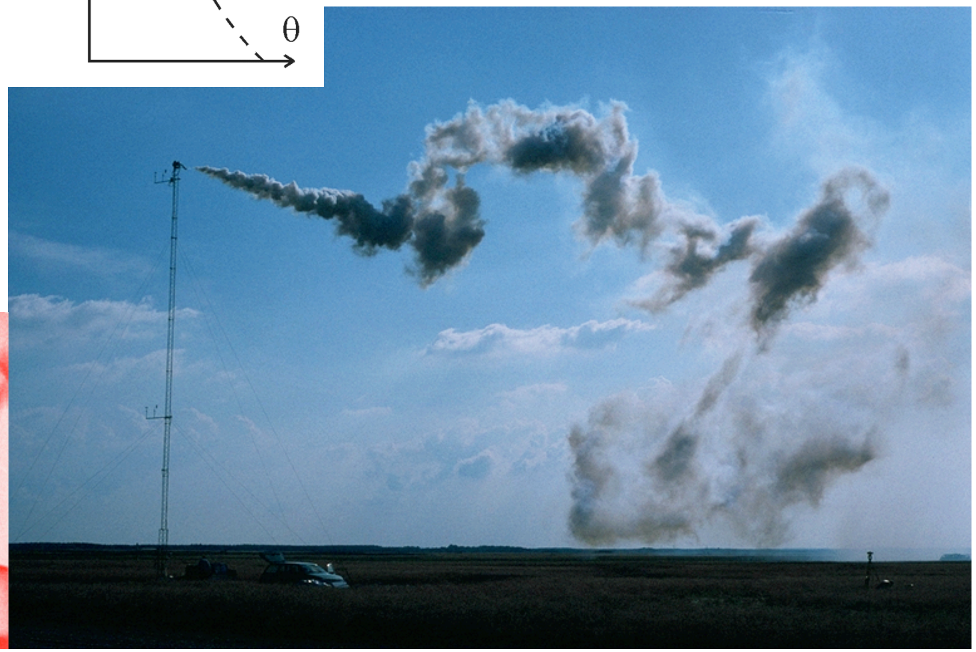
(1) Land-atmosphere interaction (including surface/emission variability)

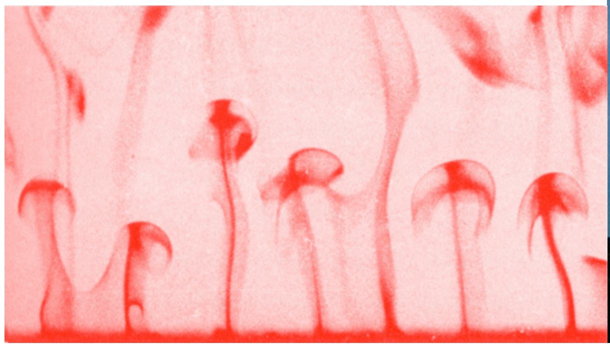
How does the CBL structure influence the dispersion and transformation of atmospheric compounds?

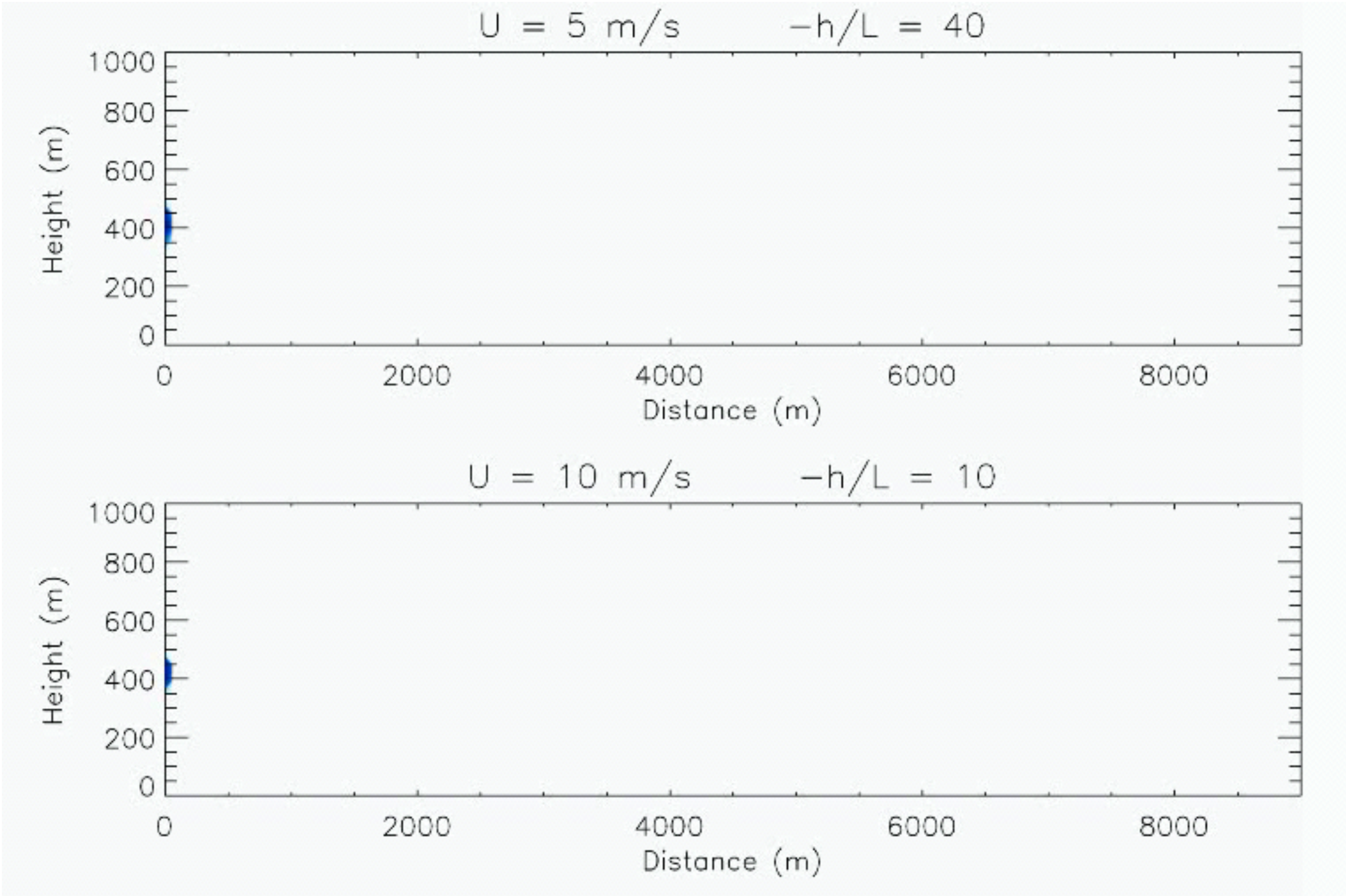
Turbulent dispersion and mixing driven by vigorous thermals and subsidence motions

Plume morphology








U = 5 m/s   −h/L = 40
Height (m)
Distance (m)
0
200
400
600
800
1000
0
2000
4000
6000
8000
U = 10 m/s   −h/L = 10
Height (m)
Distance (m)
0
200
400
600
800
1000
0
2000
4000
6000
8000

Using LES to understand and obtain the statistical properties of plume dispersion

Calculation Taylor's diffusion equation

$$\sigma_j^2(T) = \underbrace{2\overline{u_j^2}\int_o^T dt\int_o^t R_j(\tau)\,d\tau}_{\text{Each term calculated by LES}}$$

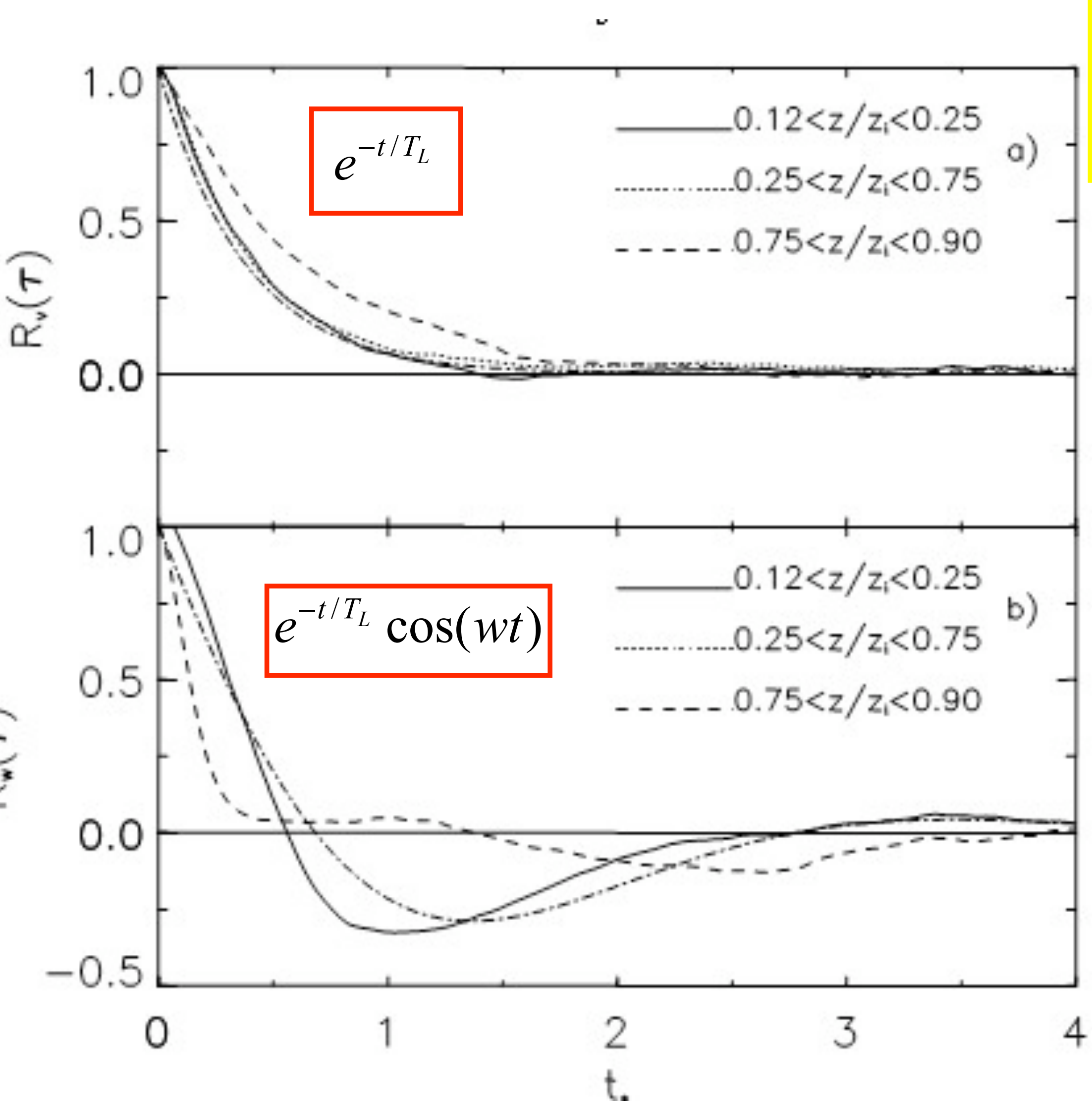
σ is calculated
explicitly by LES

Lagrangian autocorrelation velocities

$$\sigma_j^{\,2}(T) = 2\overline{u_j^{\,2}}\int_o^T dt \int_o^t \boxed{R_j(\tau)}\,d\tau$$

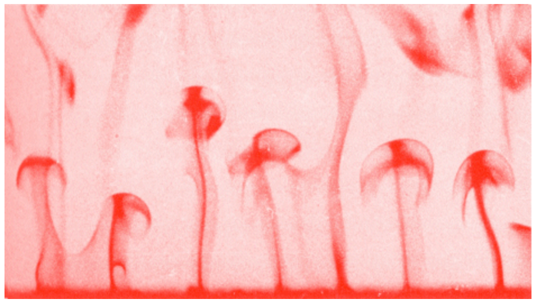
Horizontal

Vertical



Approximately, follows a Markov process (homogeneous and stationary)

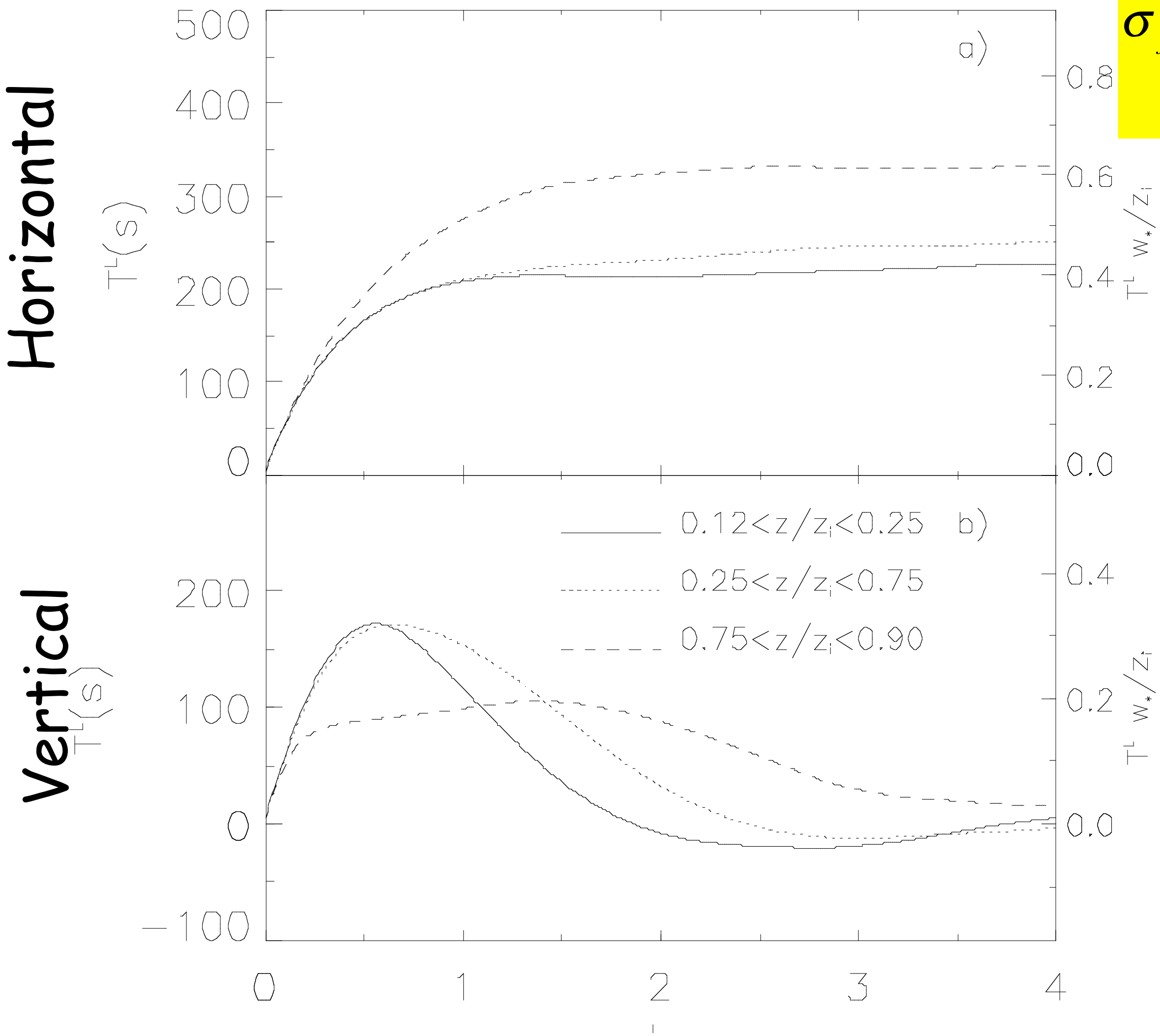
Influence by large-eddy structure.



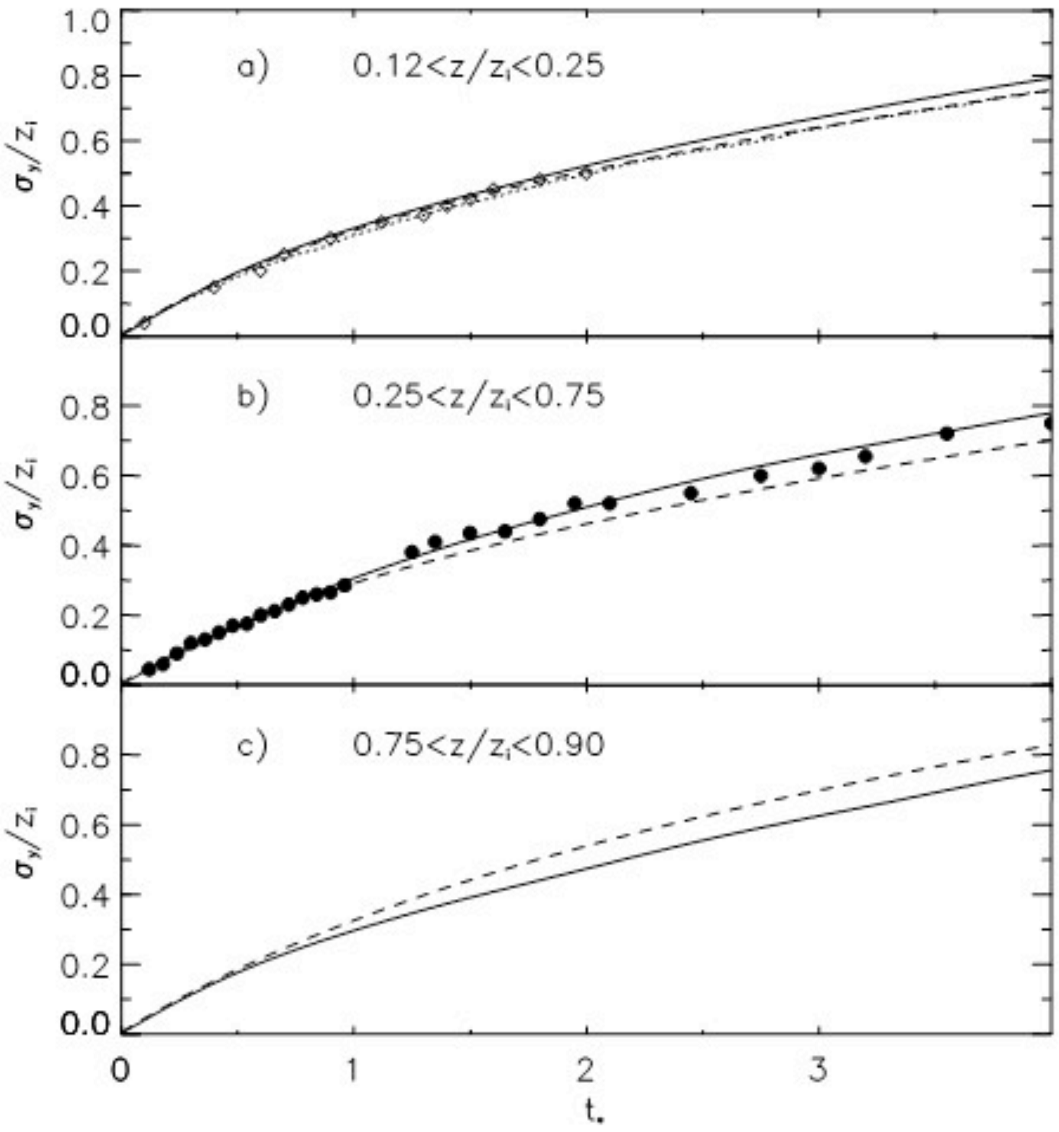
Lagrangian time scale

$$\sigma_j^{\,2}(T) = 2\overline{u_j^{\,2}}\int_o^T dt \boxed{\int_o^t R_j(\tau)d\tau}$$

$$\boxed{T_j^L \equiv \int_0^\infty R_j^L(\tau)d\tau}$$



Lateral dispersion (y-comp)



$$\sigma_j^{\,2}(T) = \boxed{2\overline{u_j^{\,2}} \int_o^T dt \int_o^t R_j(\tau)\, d\tau}$$

Continuous line $\overline{y'^2}(t)$

Dotted line σ_y

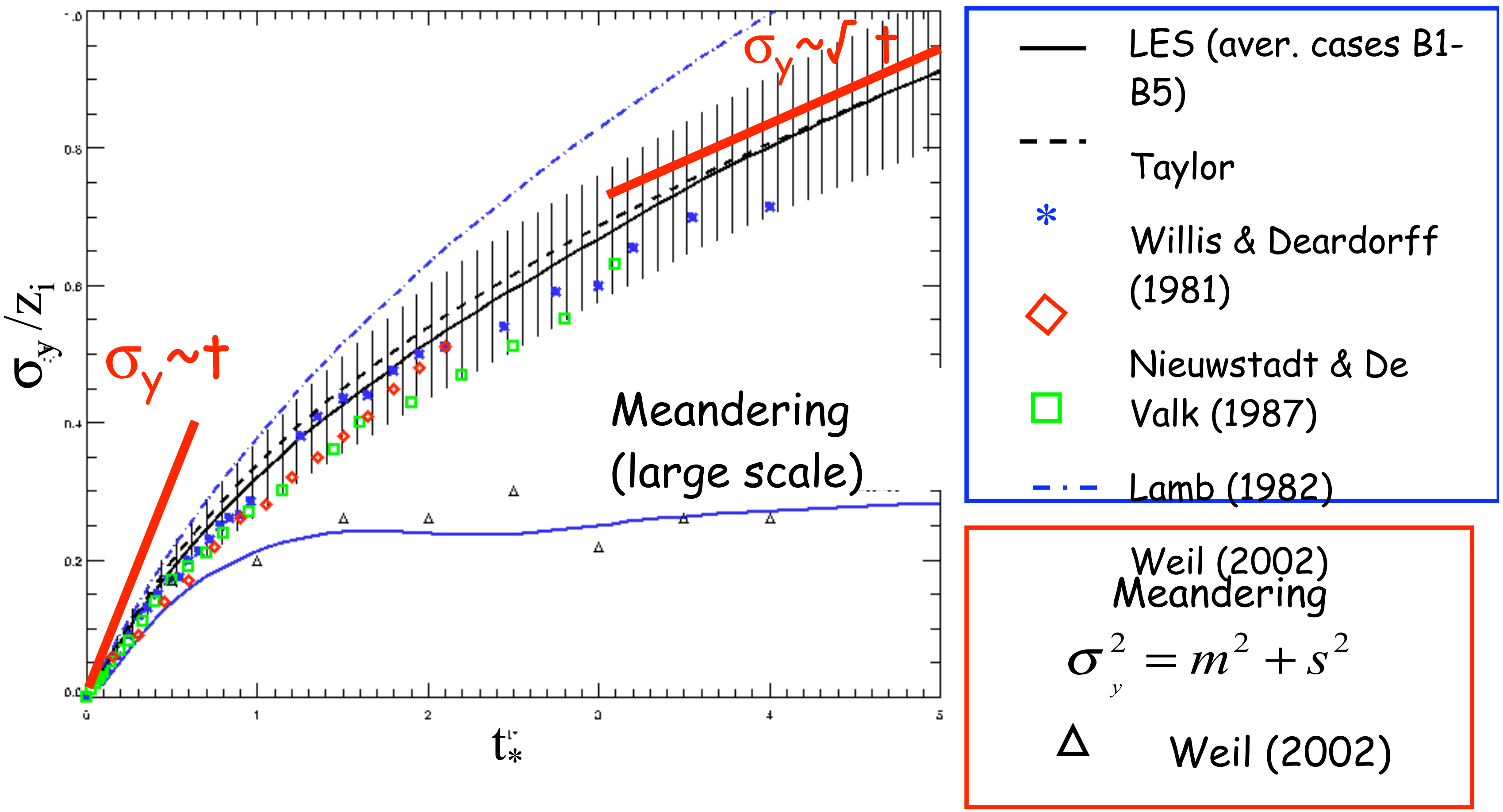
Dashed line Taylor eq.

● Water-tank data

$\overline{y'^2}(t)$ directly calculated from Lagrangian particles

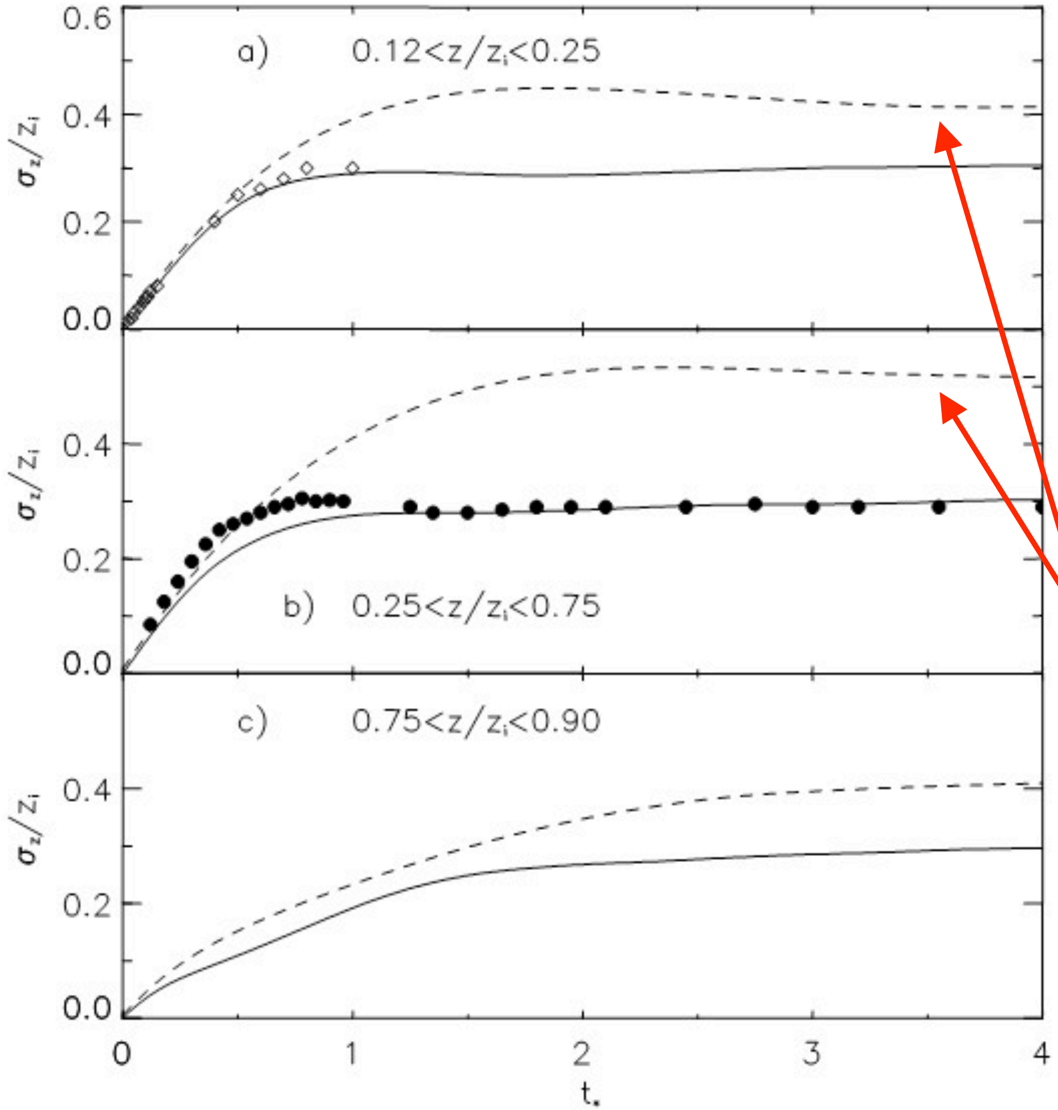
Lateral dispersion

LES results validated against other studies



Vertical dispersion (z-comp)

$$\sigma^2 = 2\overline{w^2}\int_o^T dt \int_o^\tau R(\tau)d\tau$$



Continuous line $\overline{z'^2}(t)$

Dashed line Taylor eq.

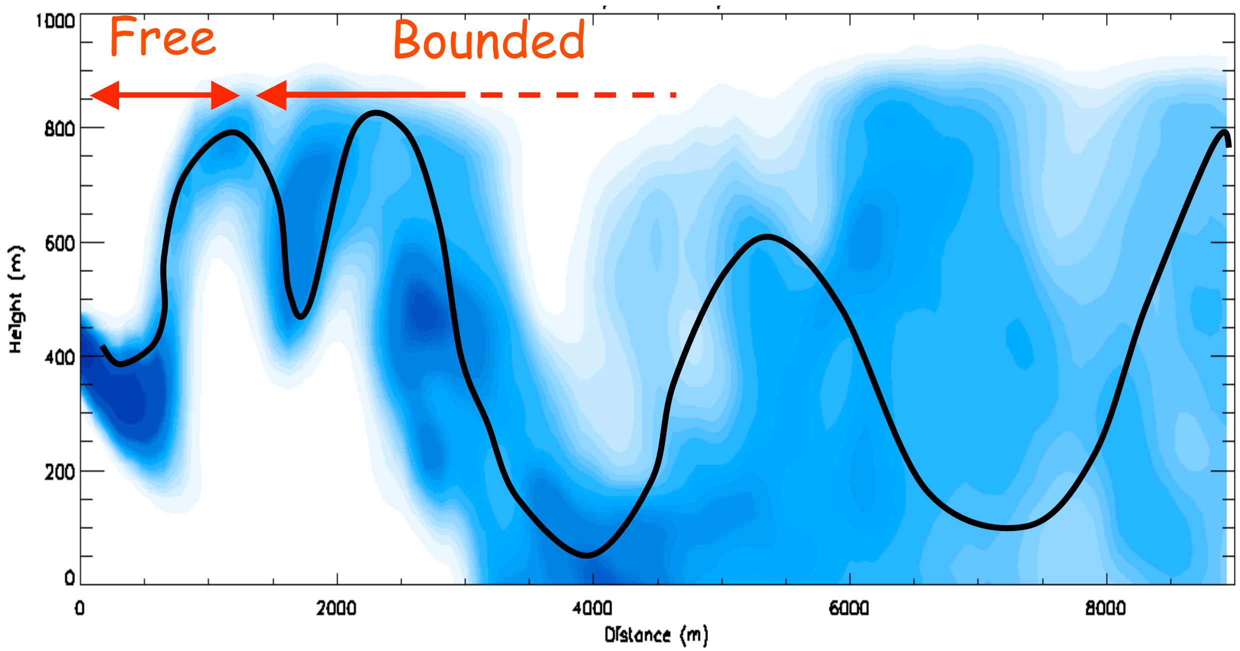
• Water-tank data

Overestimation of Taylor's theory

$\overline{z'^2}(t)$ directly calculated from Lagrangian particles

Redefining the Lagrangian time scale

To account for "free" and "bounded" motion



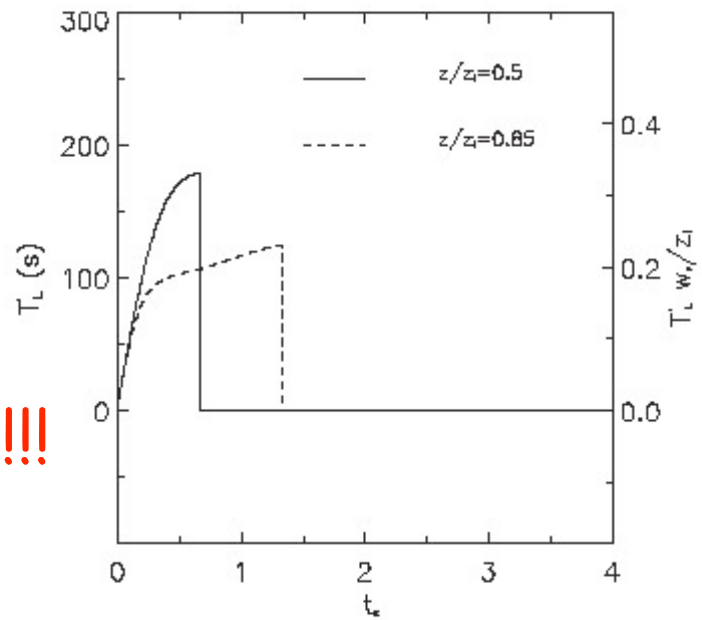
Free movement ($\tau<t_o$) (before reaching the boundary)

$$T_L^{'}(t) = T_w^L(t) = \int_0^t R_w^L(\tau)d\tau$$

$$\sigma^2 = 2\overline{w^2}\int_o^T dt \boxed{\int_o^\tau R(\tau)d\tau}$$

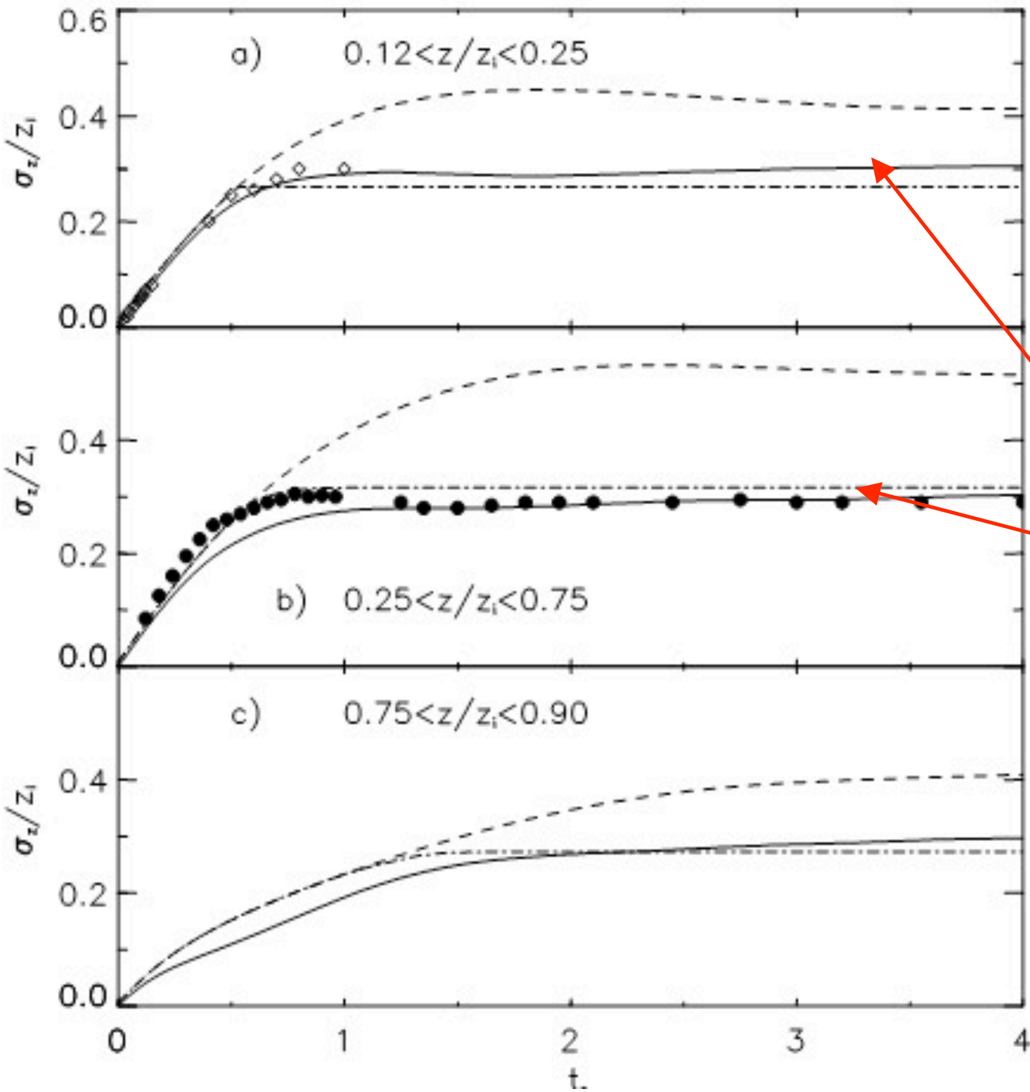
Bounded movement ($\tau>t_o$) (after reaching the boundary)

$$T_L^{'}(t) = 0$$



Not a theoretical explanation!!!

Vertical dispersion (z-comp)



$$\sigma^2 = 2\overline{w^2}\int_o^T dt \int_o^\tau R(\tau)d\tau$$

Continuous line $\overline{z'^2}(t)$

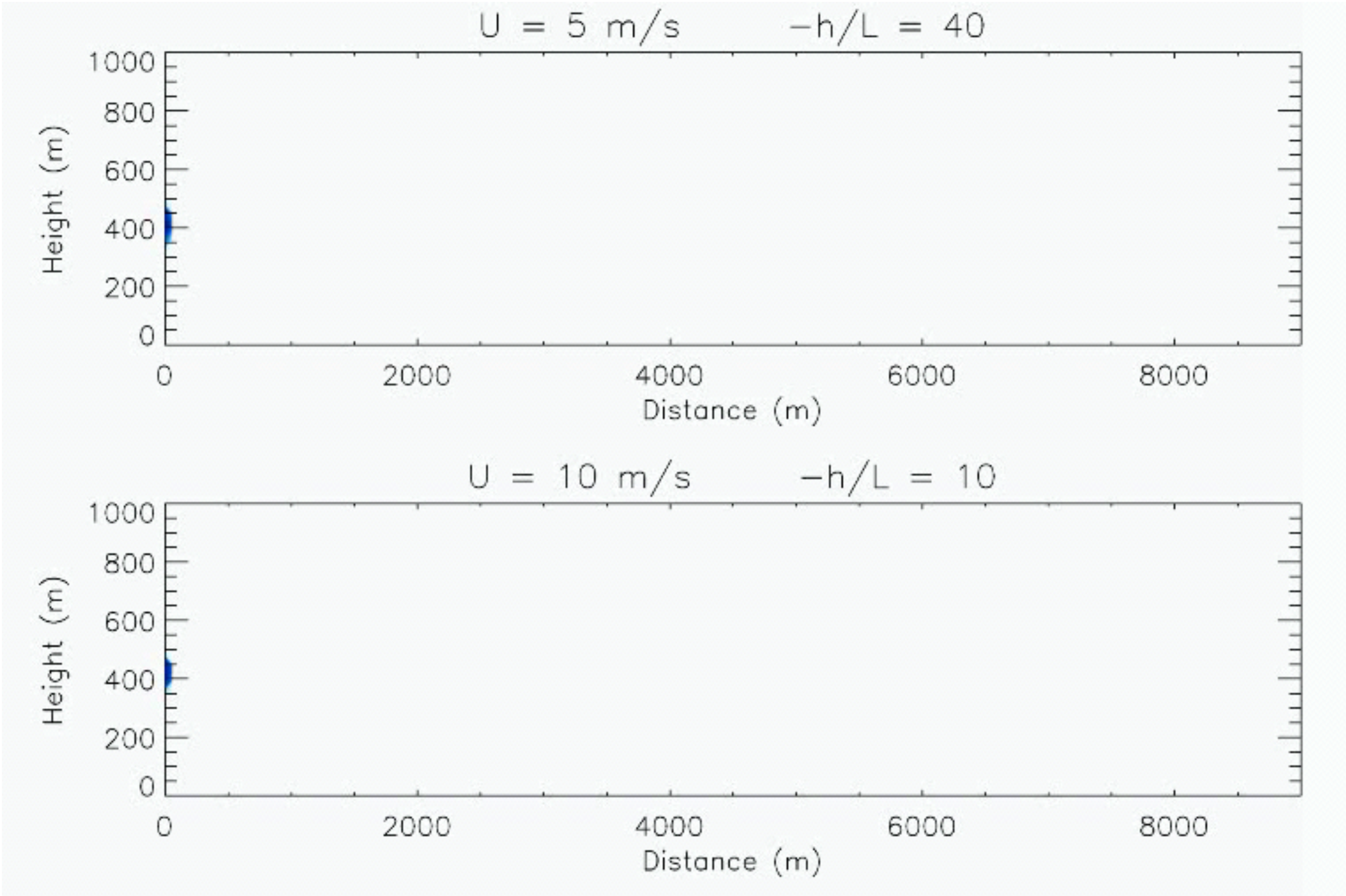
Dashed line Taylor eq.

• Water-tank data

Dashed-dotted new T_L

Previous simulation were done in free convective conditions.

What is the role of wind and shear in dispersion under convective conditions?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Height (m)
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200
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1000
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2000
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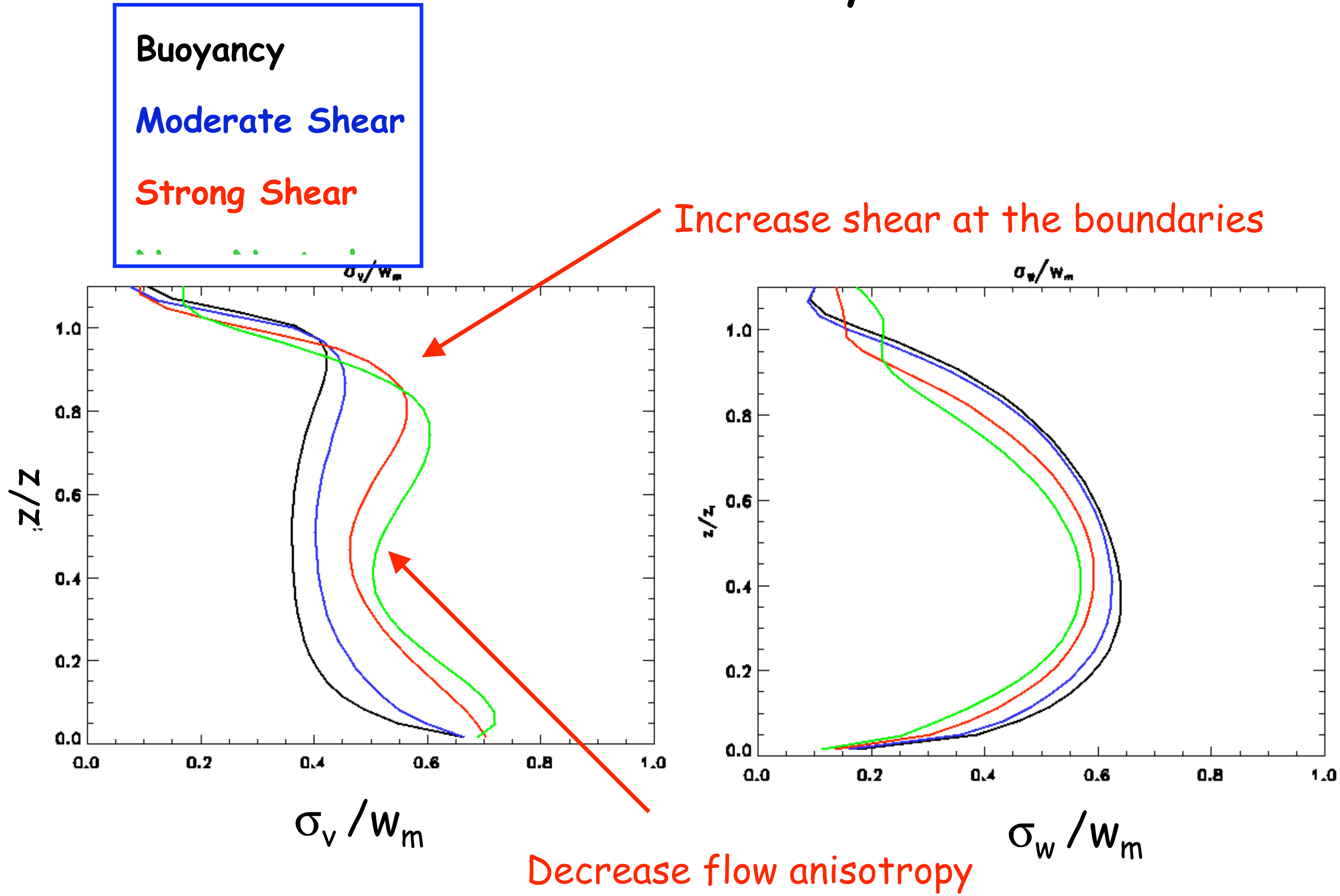
Profiles velocity variances



Vertical velocity horizontal cross-sections

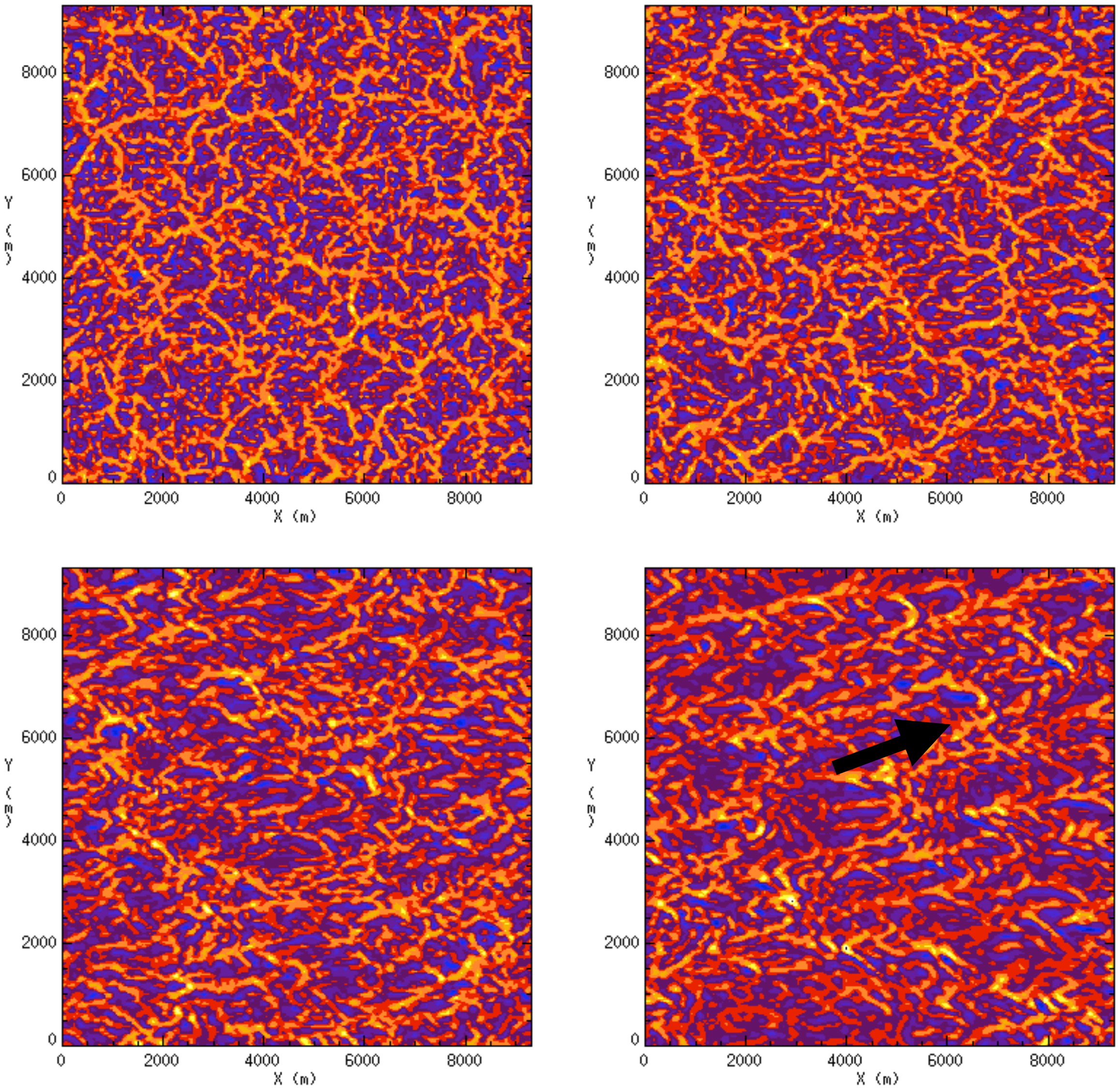
(z/z_i=0.175)

Buoyancy

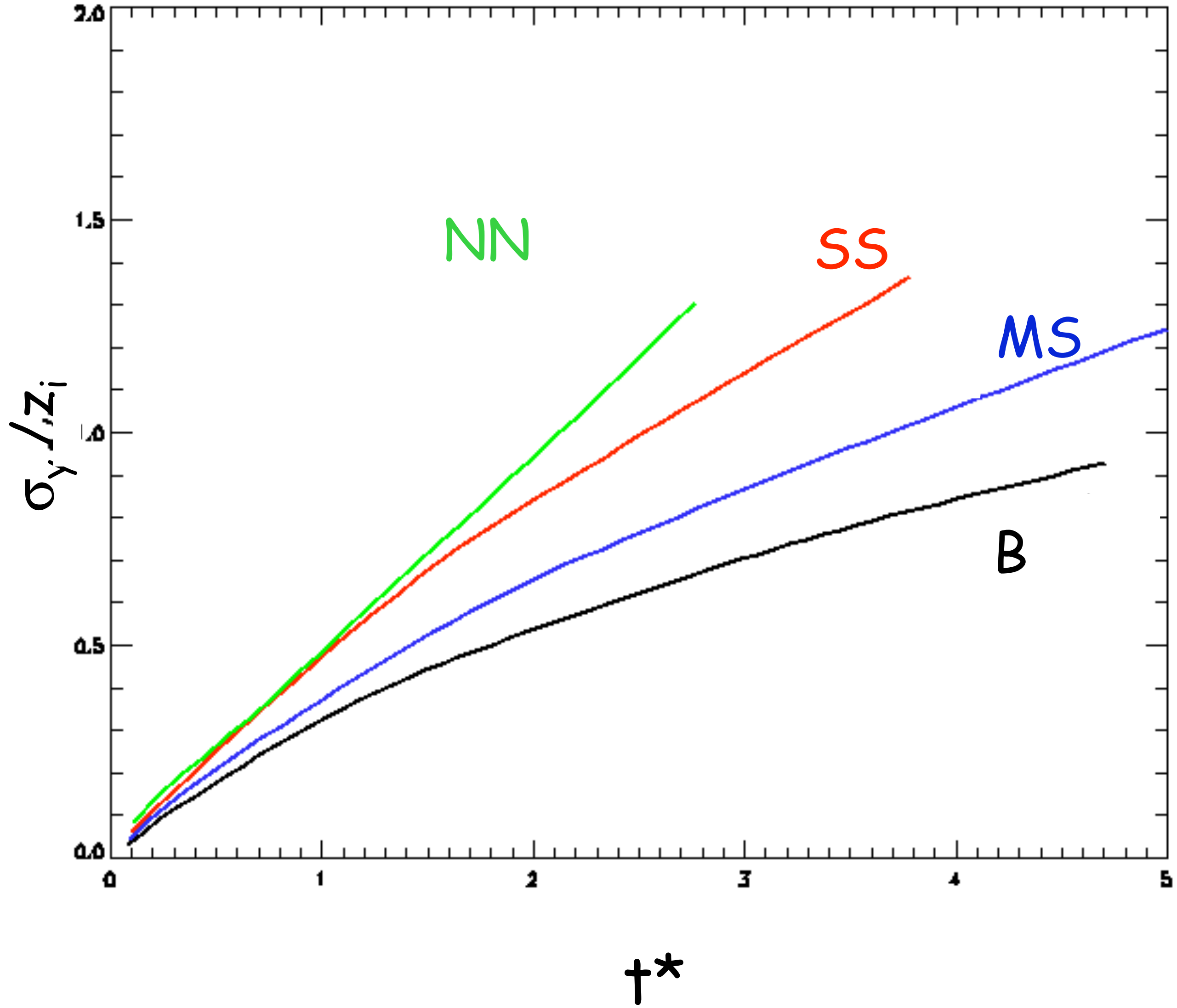
Moderate shear

Strong shear

Near neutral



Lateral dispersion (σ)



and now we move to

chemistry

Do we need to treat chemical species differently that inert atmospheric compounds?

or

What's the importance of the reactive term in the

Conservation eq. reactants: physical influences

Turbulence and UV-radiation influence R

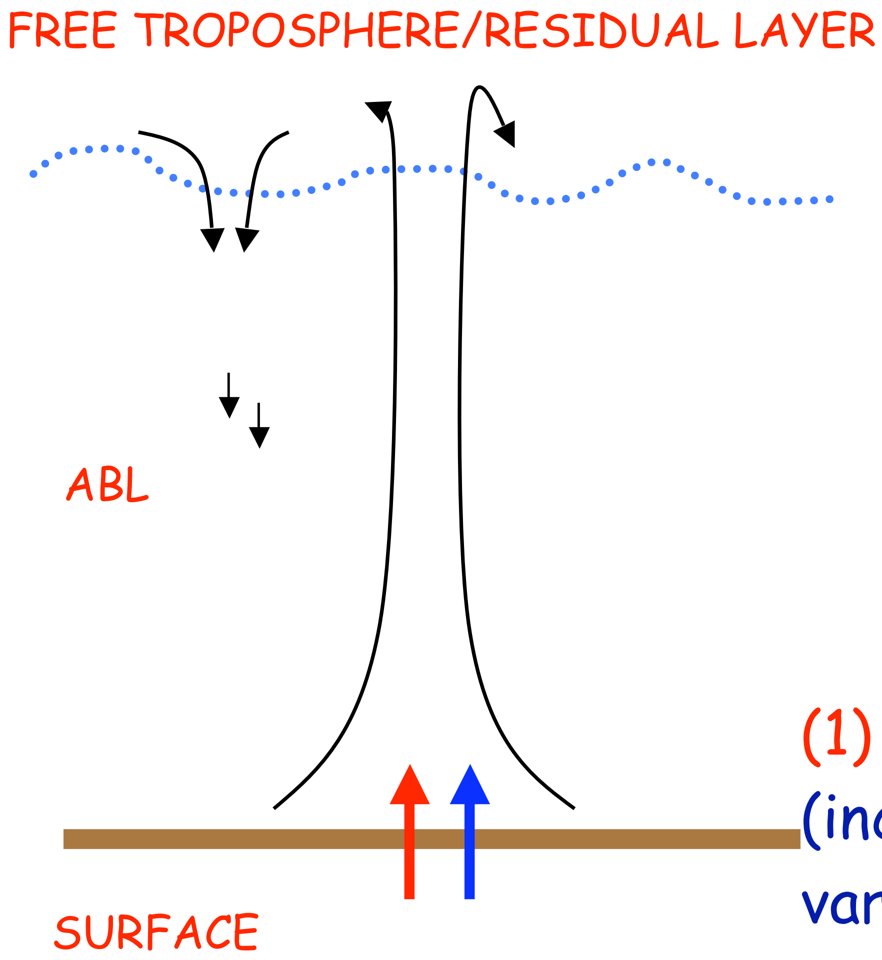
$$\frac{\partial \overline{C}}{\partial t} + \overline{U}_j \frac{\partial \overline{C}}{\partial x_j} + \frac{\partial \overline{u_j c}}{\partial x_j} = j\overline{A} - k(\overline{B}\,\overline{C} + \overline{bc})$$

(Averaged equation)

Photolysis j control by UV radiation

Co-variance quantifies how atmospheric turbulence mixes reactants

Essential processes to be represented in the ABL



(3) Entrainment
(exchange fluxes)

(2) Turbulent mixing
(turbulent fluxes)

(1) Land-atmosphere interaction
(including surface/emission variability)

(1) Land/Atmosphere exchange

Is the flux of
chemically reactive species
constant with height in the
Atmospheric Surface Layer?

In the atmospheric surface layer flux is (almost) constant with height

$$\boxed{\frac{\partial \overline{w'\varphi'}}{\partial z} \approx 0}$$

$$\varphi = \theta, U, V, q, CO_2, \ldots$$

But for chemically active species

$$-\frac{\partial \overline{w'\varphi'}}{\partial z} \pm \boxed{R_{\varphi}} \approx 0$$

Chemical reaction term (production/destruction)

$$\varphi = O_3, NO, NO_2, RH, NH_3, CO, \ldots$$

Chemical/aerosols transformations lead to a flux divergence

$$\boxed{\begin{array}{l} A + B \rightarrow C \\ \quad C \rightarrow A + B \end{array}}$$

$$\frac{\partial \overline{w'c'}}{\partial z} = -j\bar{c} + k\left[\bar{a}\bar{b} + \overline{a'b'}\right]$$

As a result, the flux of chemically active species can vary with height in the ASL

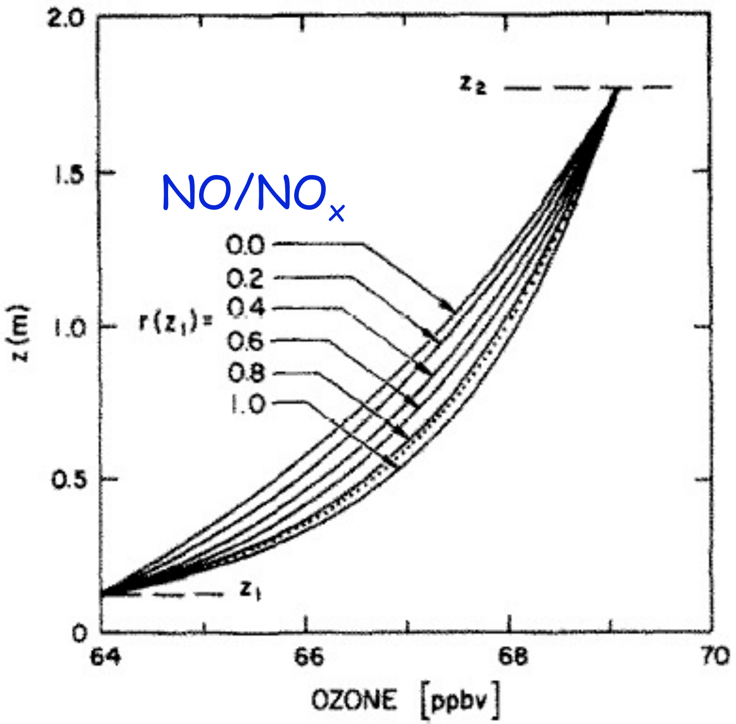
Flux divergence of the system NO-O_3-NO_2

Monin-Obukhov similarity theory applied to reactive species

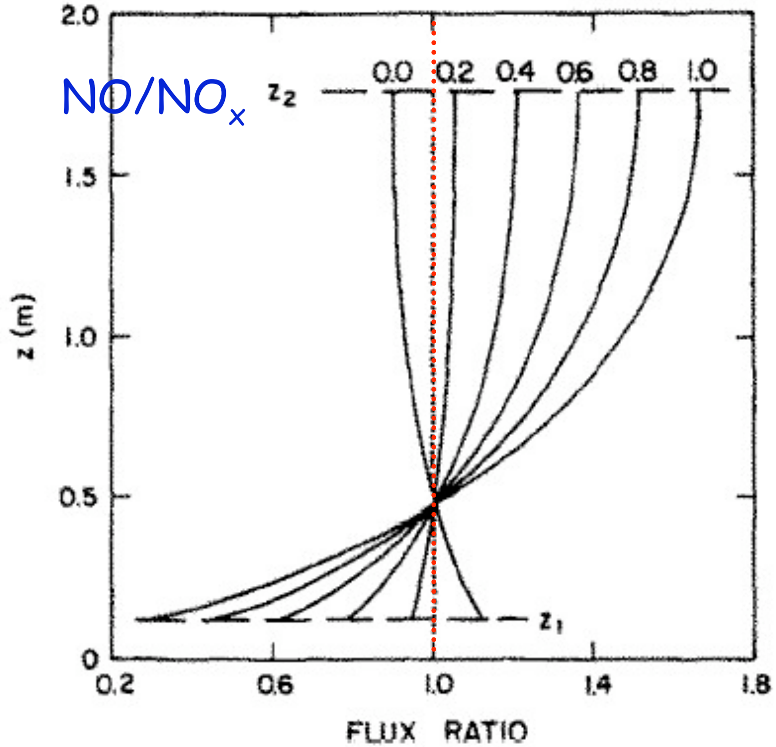
$NO + O_3 \rightarrow NO_2$

$NO_2 \rightarrow NO + O_3$

Departure log profile



Departure constant flux

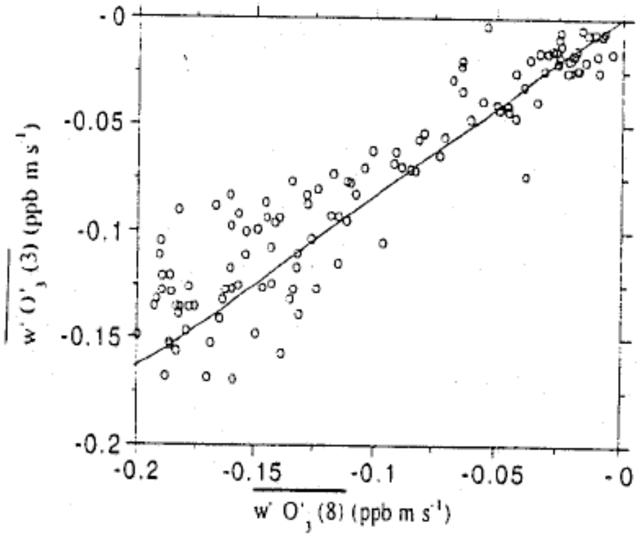


(Fitzjarrald and Lenschow, 1983)

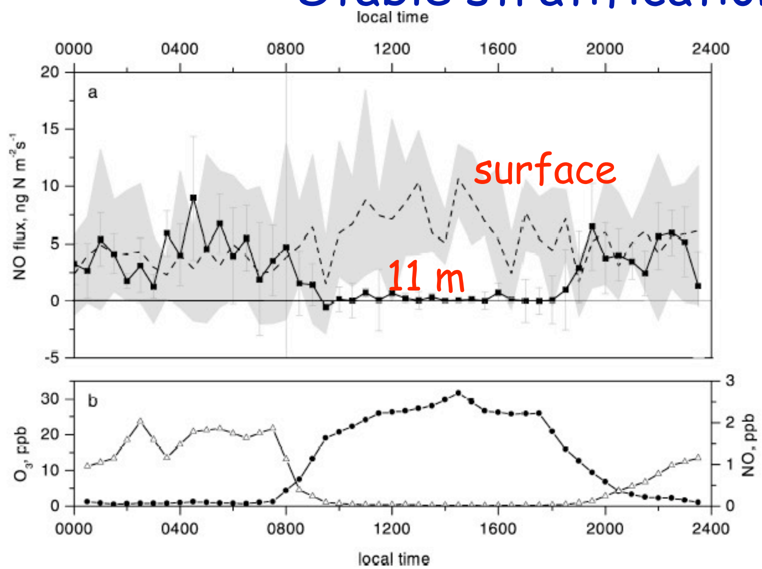
Do we have experimental evidence?

Not so much…but interesting

Grass
Day time
Unstable stratification



Inside canopy
Day time
Stable stratification



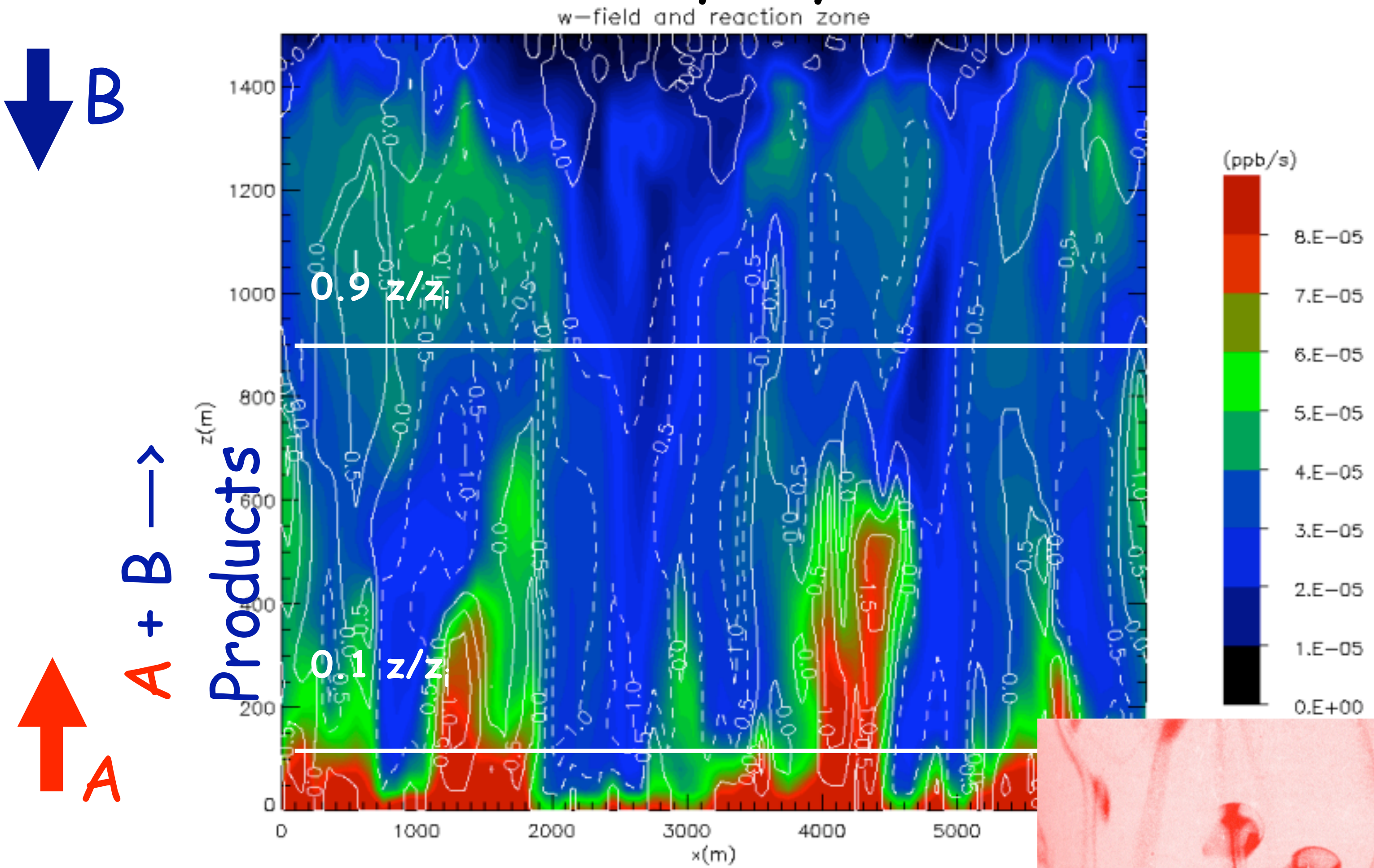
(Rummel et al. 2002)

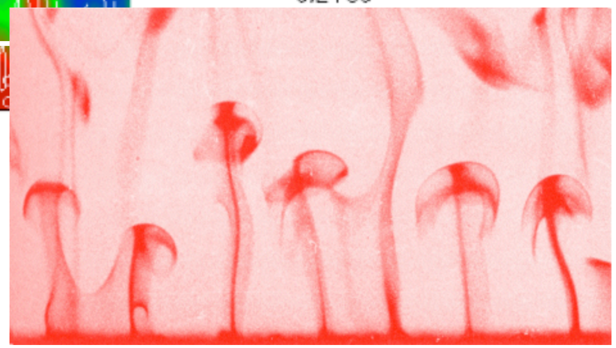
Practical implications of the flux divergence of chemically active species

(2) Turbulent mixing

Does atmospheric turbulence control the chemically reactivity?

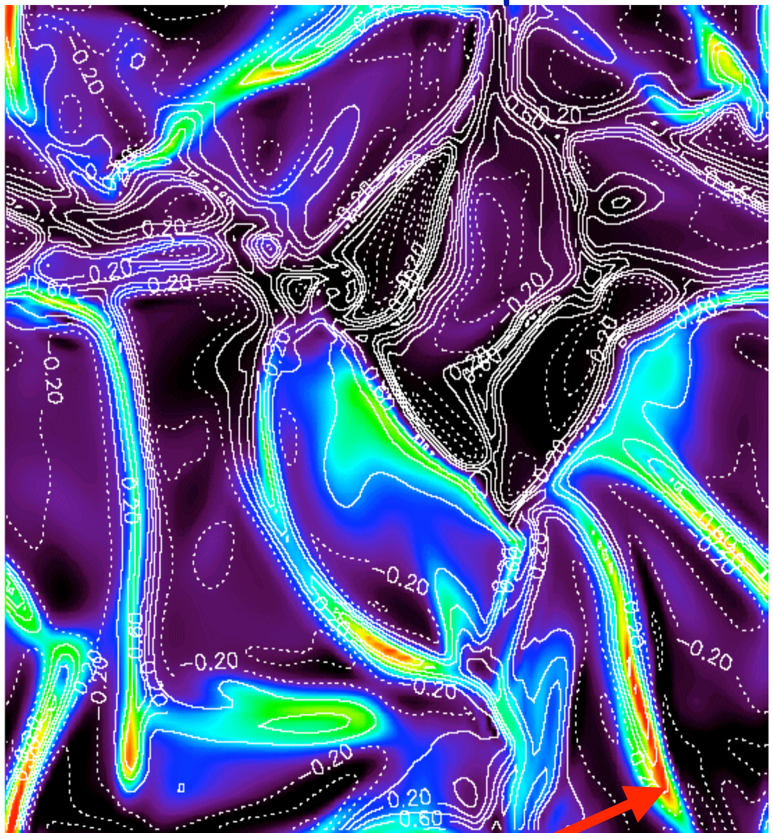
Segregation of chemical species due to the boundary layer structure



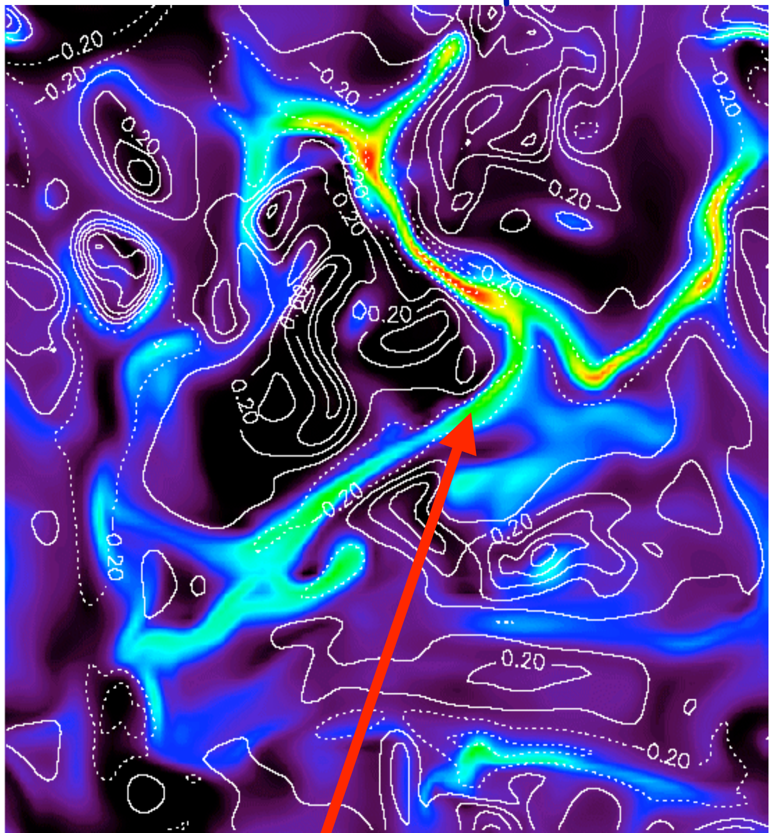


Horizontal cross section vertical velocity and reaction zones

$0.1\ z/z_i$



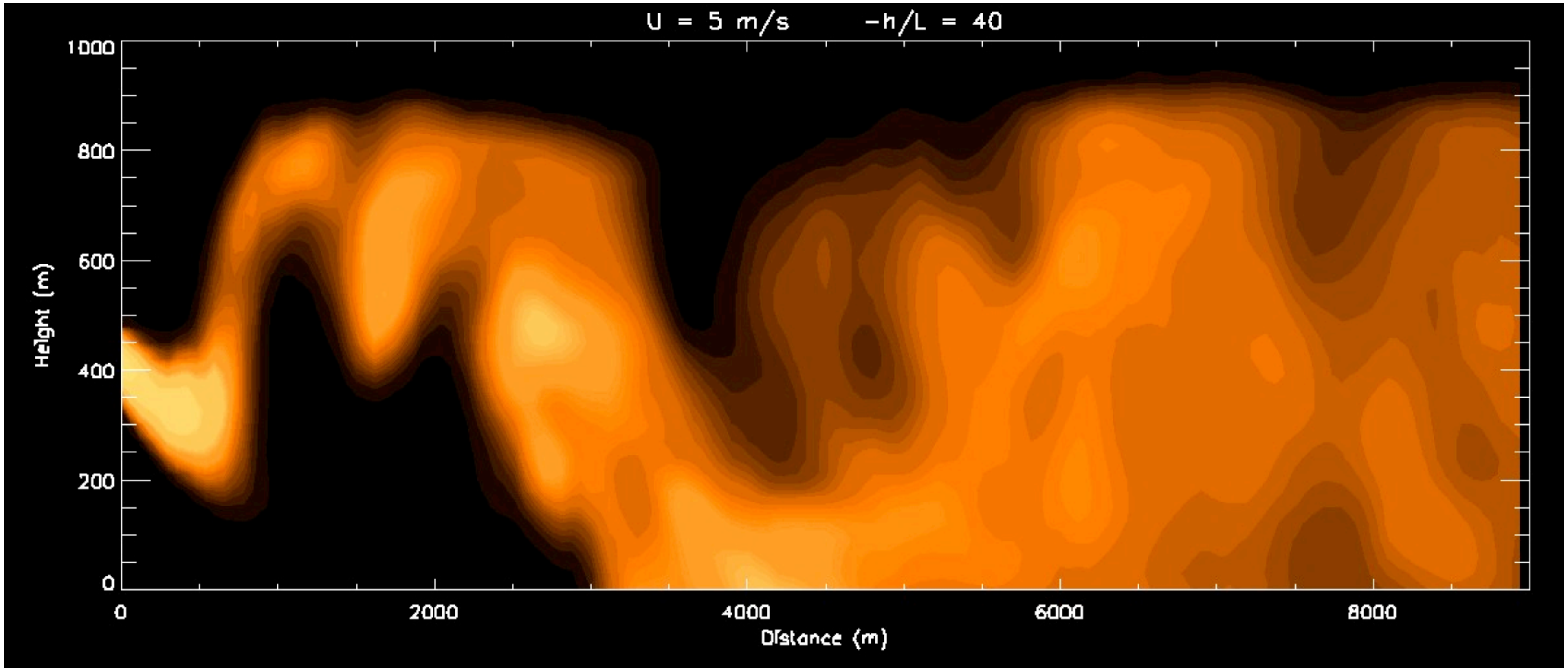
$0.9\ z/z_i$



Reaction zone in the updraft

Reaction zone in the downdraft

A similar turbulent control can occur in chemically reactive plumes



Chemical species can be segregated and then reactivity is controlled by turbulent dispersion!!

How efficient is turbulence in mixing the reactants?

When does turbulence control the reactivity?

In particular, for reactions with a similar chemical time scale to the turbulent mixing time scale

Definition Damköhler number:

$$\boxed{Da = \frac{\tau_t}{\tau_c}}$$

Classification regimes (rough):

Da << 1 Chemistry is slow compared with turbulence => SPECIES ARE WELL MIXED

Da = O(1) Chemistry and turbulence similar time scales => CONTROL Ability of turbulence to efficiently mix reactants

Da >> 1 Chemistry is faster than turbulence => NOT DEPENDING ON THE TURBULENCE

How can we quantify the effect of segregation/heterogeneous mixing?

$$\frac{\partial c}{\partial t} = -\frac{\partial \overline{w'c'}}{\partial z} - j\bar{c} + k\left[\bar{a}\bar{b} + \overline{a'b'}\right]$$

Coefficient Intensity segregation

$$Is = \frac{\overline{a'b'}}{\overline{A}\,\overline{B}}$$

Co-variance between reactants

A + B -> Products

Classification regimes:

Da << 1 SPECIES ARE WELL MIXED then

Is =0

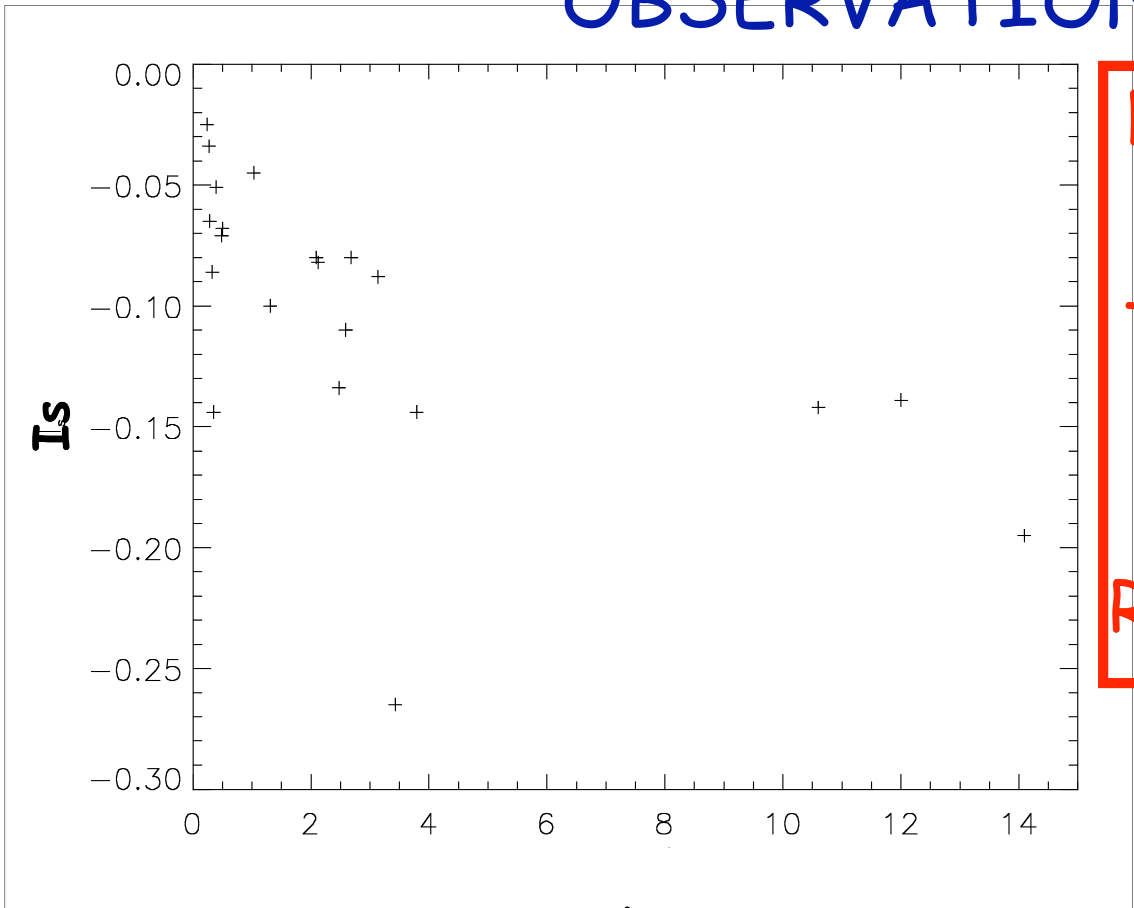
Da => O(1) CONTROL by TURBULENCE then

(non-premixed) -1 < Is < ∞ (premixed)

Depends on the way species are introduced

NO-plume released in the atmospheric surface layer and reacting with ozone

OBSERVATIONS



NO/O_3

Negative value =>
Slow down of
the reaction rate

Is= -.15 =>
Reaction rate 15%
slower

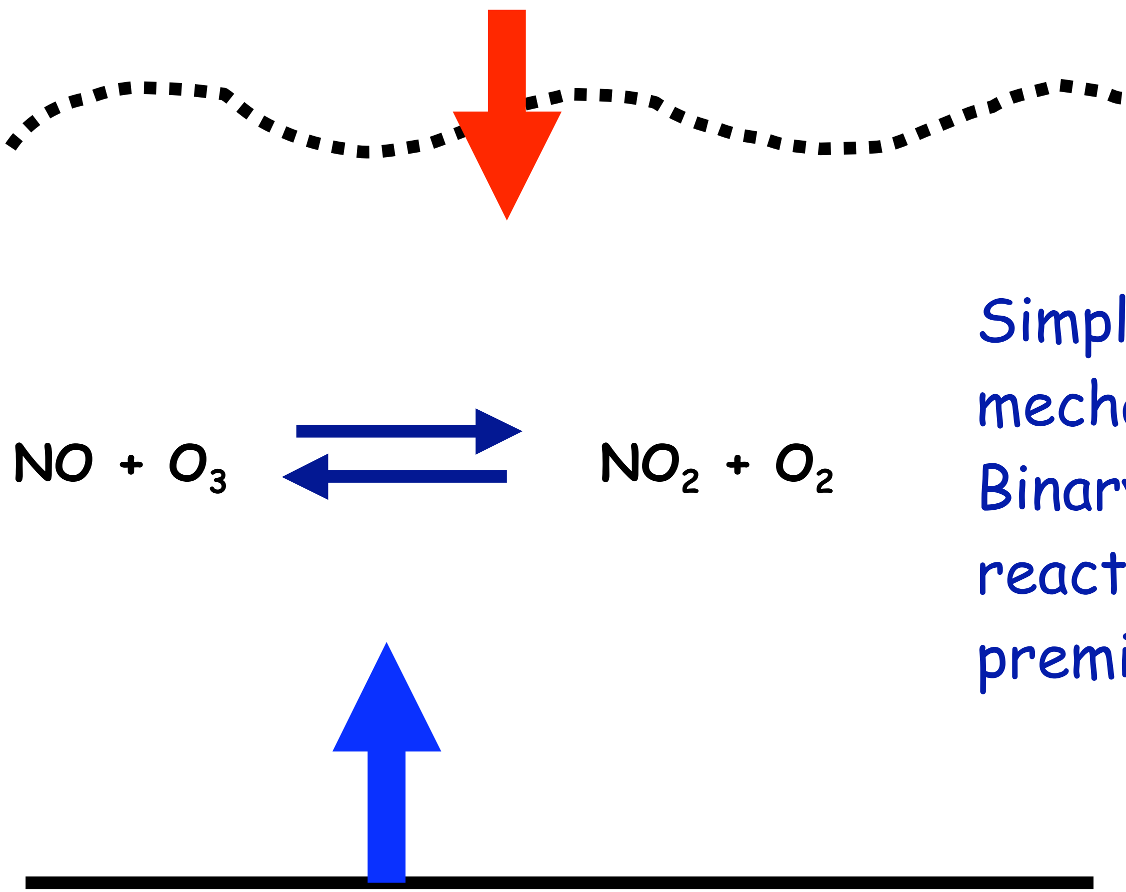
(Komori et al., 1991)

Intensity of segregation depends on:

I: Ability of turbulence to mix chemical species

II: Horizontal/vertical variability emissions

I: Ability of turbulence to mix chemical species

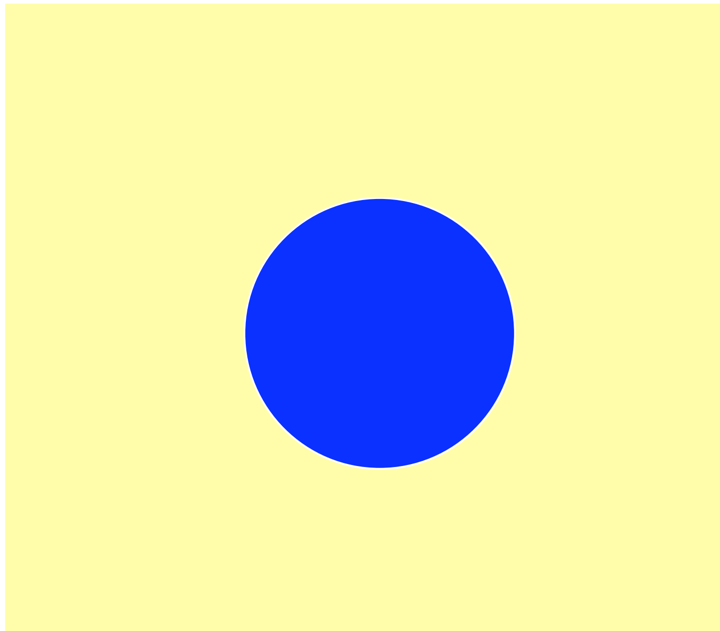


Simple chemistry mechanism.
Binary irreversible reaction/Non-premixed

Setting up numerical experiments using LES to investigate the combined effect of turbulence and emission heterogeneity

Uniform/Homogeneous

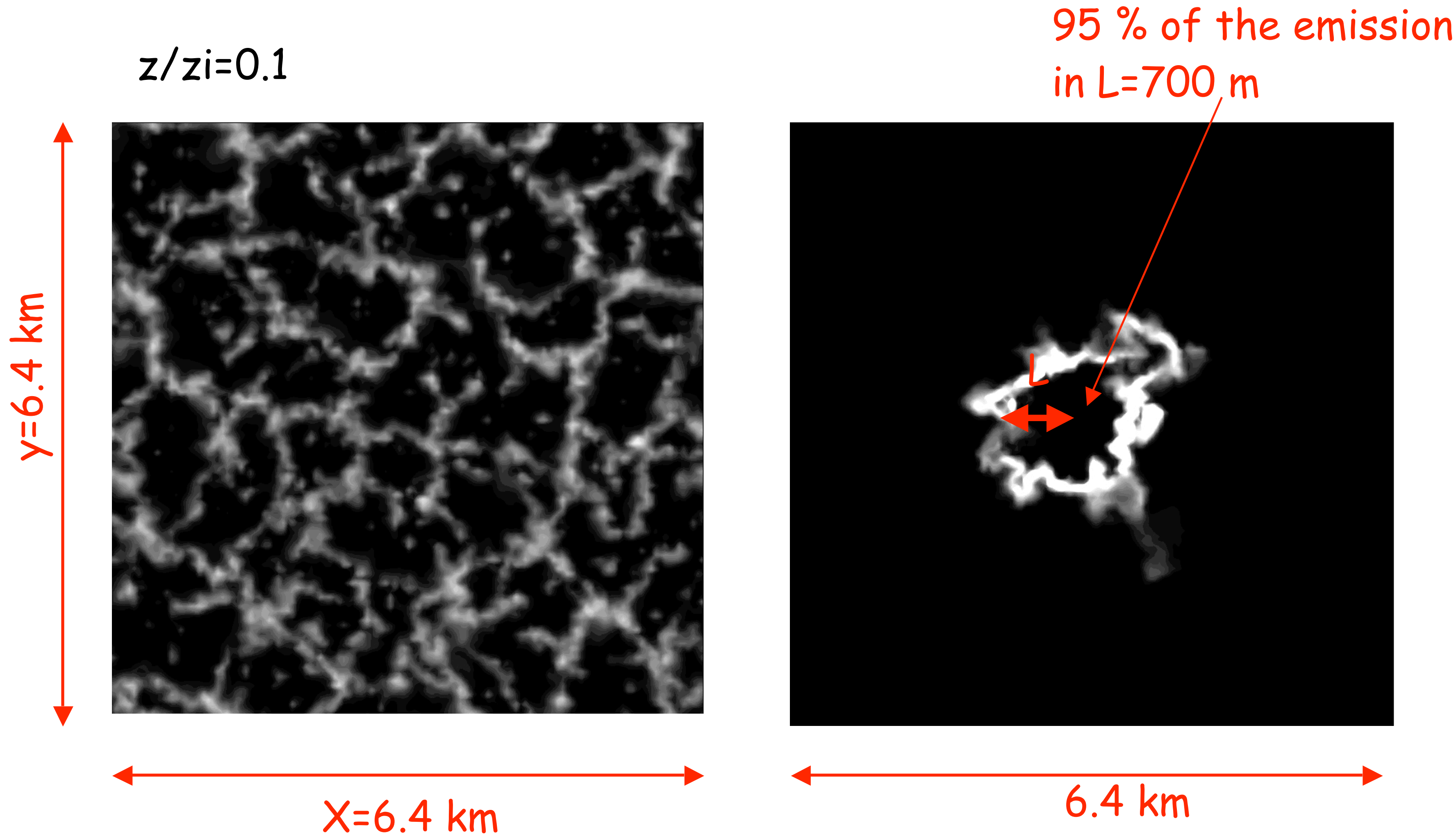
Non-uniform/Heterogeneous



Same amount of species is emitted!!!

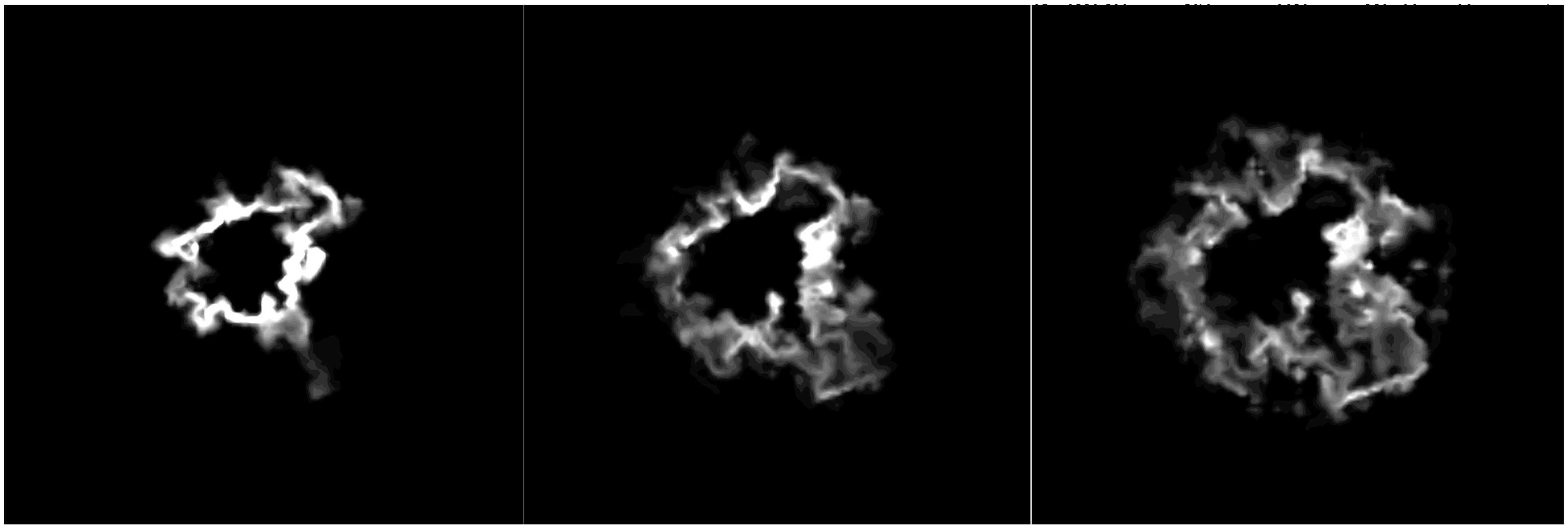
Location of the reaction rate:

Horizontal cross section

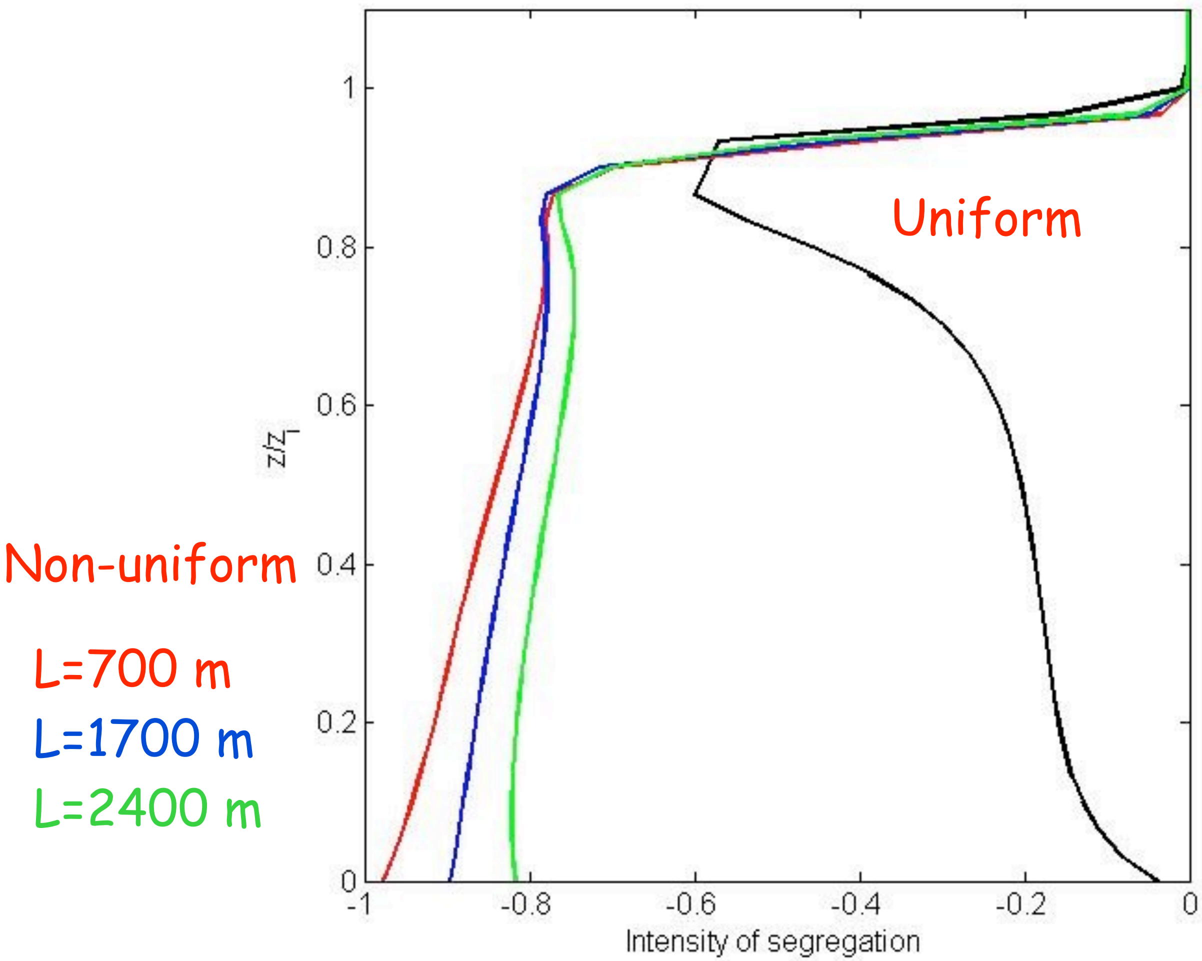


Different length scale of the surface emission

L=700 m　　L=1700 m　　L=2400 m

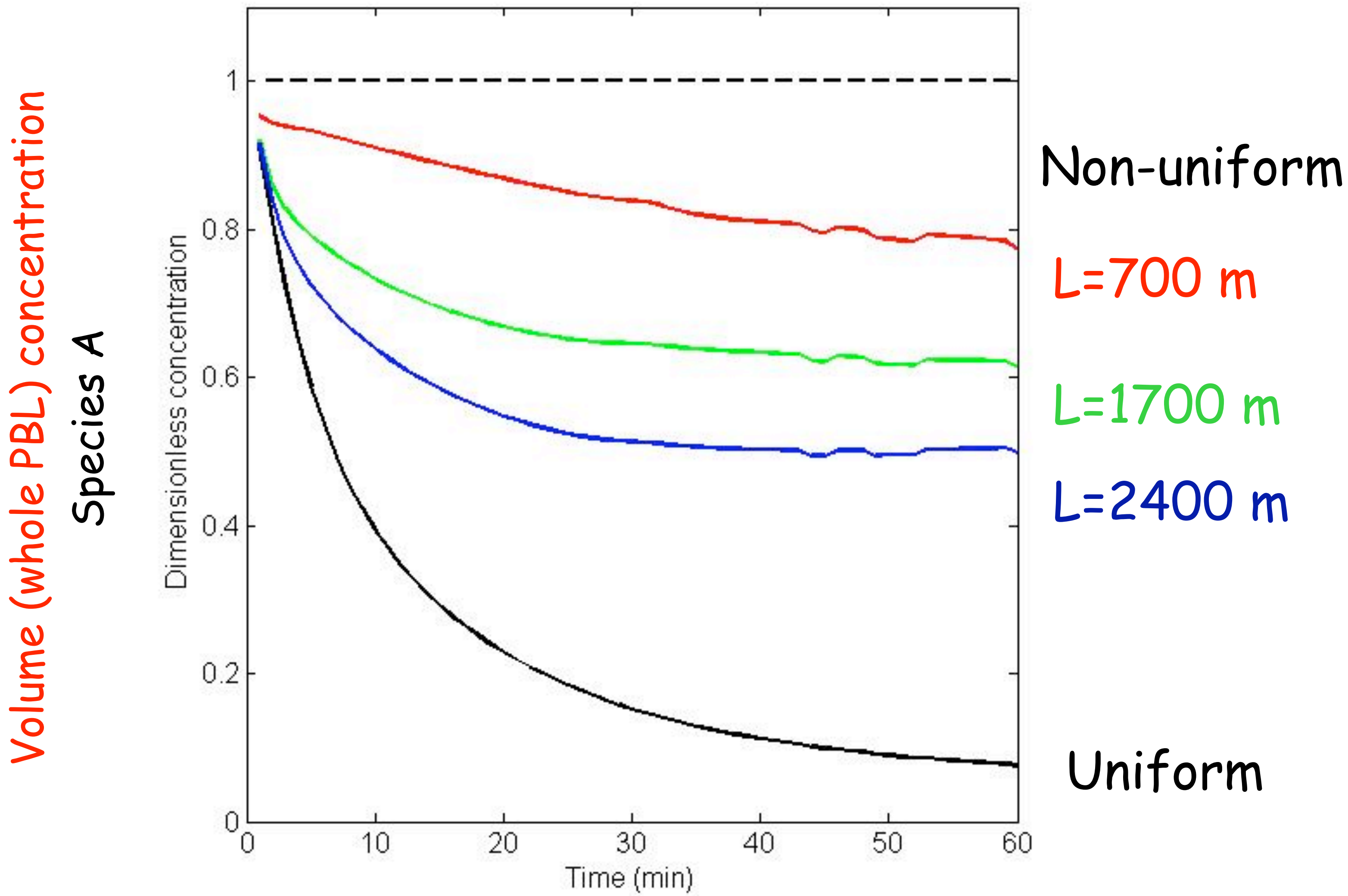


Quantifying the effect of the emission heterogeneity by the intensity of segregation

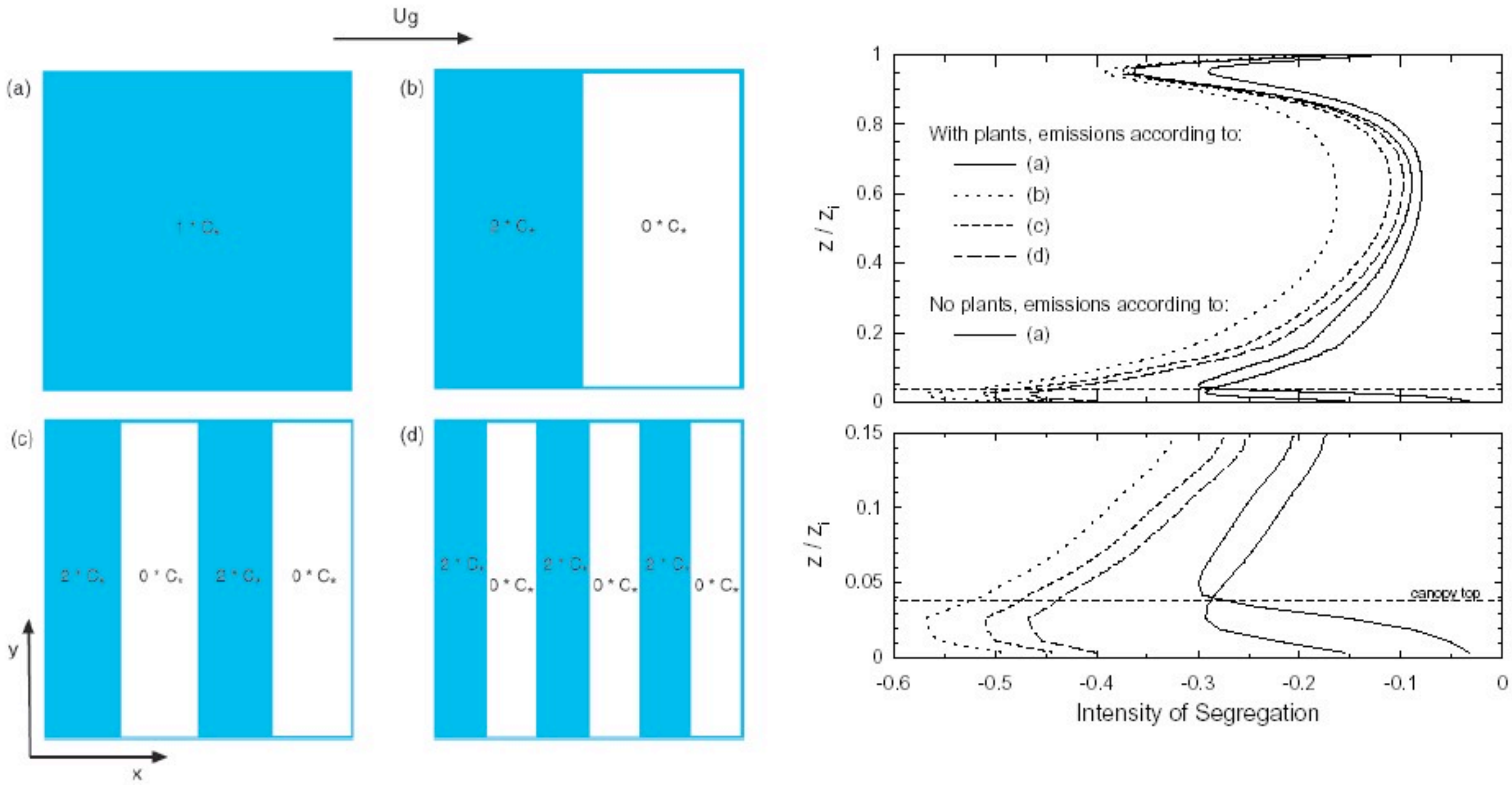


The heterogeneity of emission can control the reactivity of concentration

A + B -> P



Horizontal variability emissions and canopy effects



(Patton et al., 2000)

(3) Entrainment/Exchange

What is the role of entrainment on the exchange of reactants?

To be studied during the tutorial 7 (optional)!!!

Boundary layer dynamics control dispersion and reactivity of atmospheric compounds

Challenges ahead (I)

To integrate the dispersion chemical transformation in all the relevant spatial/temporal scales

Micrometeorology

Canopy

Boundary layer dynamics

Orography

Mesoscale

Stratification effects

Stratification and rotation effects

Challenges ahead (II)

To account for the interaction/feedbacks of physical/chemical processes

Atmospheric dispersion near the surface and in stable stratified conditions

Clouds

Deposition/Sedimentation heavy particles (pollen)

Heterogeneous chemistry

.....