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# MODELLING POLLUTANT REACTIONS DEPENDING ON INSTANTANEOUS CONCENTRATIONS: A DIRECT ESTIMATION OF THE SEGREGATION COEFFICIENT


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photo: Lyon (France, 2009) - Amicarelli

# Summary

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    1. Scalar reactions and concentration fluctuations
    2. Lagrangian micromixing modelling
2. Numerical model (Leuzzi et al.)
    1. Main features
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    3. Micromixing scheme (molecular diffusion)
    4. Conserved scalar theory (CST, scalar reactions) (new)
    5. Proposed variants for CST: NHRDL (and RDL-plume) and NFRDL (new)
3. 2D validation ($2^{nd}$ order kinetics) in grid turbulence (point source + backg)
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This study refers to Amicarelli et al. CAF (manuscript in revision), but Sec.4:
Stochastic Lagrangian micromixing modelling of reactive scalars in turbulent flows:
concentration fluctuations and improvements in the conserved scalar theory for non-homogeneous conditions

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# 1.1. Scalar reactions and concentration fluctuations

## Concentration turbulent fluctuations -> reactive scalar mean concentrations

- **Reynolds' decomposition** for concentration (turbulent regime):

$$C = \overline{C} + C' \qquad \text{E.1}$$

- **Reactive term** (**balance equation for $C_{mean}$** of a reactant –A or B-; **2-species 2nd order kinetics**): $T_m = -r * R_r$ (r: chemical reaction rate, $R_r$: reaction rate of Brown-Bilger 1996 JFM)

$$\overline{T} = \overline{-rC_A C_B} = -r\left(\overline{C_A} + C_A'\right)\left(\overline{C_B} + C_B'\right) = -r\left(\overline{C_A}\,\overline{C_B} + \overline{C_A' C_B'}\right) \qquad \text{E.2}$$

- **Segregation coefficient** (potential range: -1 to unlimited, ρ: correlation coefficient; $i_C$: intensity of fluctuations):

$$I_s \equiv \frac{\overline{C_A' C_B'}}{\overline{C_A}\,\overline{C_B}} = \frac{\overline{C_A C_B}}{\overline{C_A}\,\overline{C_B}} - 1 = \rho_{AB} i_{C,A} i_{C,B} \qquad \text{E.3}$$

- **Main motivation of the study:** modelling reactive pollutants with neither relevant systematic errors (due to no concentration fluctuations) nor ad-hoc (test case dependent) tuning.

# 1.1. Scalar reactions and concentration fluctuations

## Reactions -> Concentration fluctuations (e.g. concentration variance)

- Reactions alter the concentration mean gradients of the reactant and then the production term ($P_{\sigma C}$) in the balance equation of $\sigma_{CA}^2$ ($K_{T,i}$: turbulent dispersion coefficient):

$$P_{\sigma_C} = -2\overline{u_i' C'}\frac{\partial \overline{C}}{\partial x_i} \approx 2K_{T,i}\left(\frac{\partial \overline{C}}{\partial x_i}\right)^2 \qquad \text{E.4}$$

- Reactions activate a reactive term in balance equation of $\sigma_{CA}^2$:

$$\overline{R_2} = -2r\left(\overline{C_A}\,\overline{C_A' C_B'} + \sigma_{C_A}^2\,\overline{C_B} + \overline{\left(C_A'\right)^2 C_B'}\right) \qquad \text{E.5}$$

- State-of-the-art (scalar reactions via instantaneous concentrations)
  - ✓Lagrangian micromixing models: Sawford 2006 BLM, Cassiani 2013 BLM
  - ✓Pdf modelling (Garmory et al. 2006), heavy computational costs
  - ✓DNS: «very small» domains, «very heavy» computational costs
  - ✓Other models use tuned parameterizations for $I_s$

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# 1.2. Lagrangian micromixing models: main features
# Lagrangian turbulence + molecular diffusion + reactions

- Advantages:
  - ✓Unique system of equations for all the concentration moments+pdf
  - ✓Reactions depending on the instantaneous-like concentrations
  - ✓Direct estimation of the velocity auto-correlation function
  - ✓Mixing time depending on instantaneous-like parameters
  - ✓Low computational costs
  - ✓Neither convergence algorithm, nor computational mesh
- Shortcomings:
  - ✓Eulerian velocity statistics to be provided: (coupling with RANS model for the main flow or diagnostic fields from measures)
  - ✓Few studies available at the moment
- Potential application fields (–air/water quality-):
  - ✓**Micro-scale dispersion** in urban or aquatic environments (traffic sources, production or storage sites, ..)
  - ✓**Accidents:** acute emissions, explosions, inflammables, terrorism
  - ✓Impact from **odours** (e.g. energy from waste, production sites)
  - ✓**Pollutant transformations** (reactive scalars)
  - ✓Industrial processes: **combustion**, smoke treatment, desulfurization
  - ✓Energy from sewage (biological treatments)

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# 2.1. Numerical model: main features

- Model features (stationary model):
  - ✓ **Stage 1** (S1, **passive scalar means**) + **stage 2** (S2, **all other C statistics**)
  - ✓ **Macromixing scheme** (Lagrangian turbulence): estimation of trajectories of fictitious fluid particles representing a turbulent flow (Harris chain) (S1 S2)
  - ✓ **Micromixing scheme**: simulation of molecular diffusion processes (S2)
  - ✓ Scalar **reactions according to the conserved scalar theory** (CST): alternative approach to a reactive IECM scheme (S2)
  - ✓ Concentration Statistics on particle ("instantaneous") concentrations (S1 for $C_m$, S2)

- Model references of previous studies (also **3D + obstacles**):

Leuzzi et al. 2012 AtmosEnviron, Amicarelli et al. 2011b IJEP, Amicarelli et al. 2012 IJEP

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## 2.2. Macromixing scheme: Lagrangian turbulence

- Lagrangian Stochastic Equation (Markov process for velocity U):

$$\underline{X}(t+dt) = \underline{X}(t,\underline{x}) + \underline{U}(t)dt, \quad \underline{X}(t=0) = \underline{X}_0 \qquad \text{E.7}$$

$$dU_i = a_i dt + b_{ij} d\xi_i \qquad \text{E.8}$$

- Lagrangian structure function to determine b (Kolmogorov theory) ; $d\xi_i$ is a Gaussian Wiener process with mean zero and variance dt ($C_0$: Kolmogorov constant; $\delta_{ij}$: Kronecker's $\delta$):

$$\left[U_i(t+dt) - U_i(t)\right]\left[U_j(t+dt) - U_j(t)\right] = \delta_{ij} C_0 \varepsilon dt, \quad dt \to 0 \quad \Rightarrow \qquad \text{E.9}$$

$$\Rightarrow \quad b_{ij} = \delta_{ij}\sqrt{C_0\varepsilon} \qquad \text{E.10}$$

- "Well mixed condition" for independent Gaussian velocity probability density functions (pdfs) – Thomson 1987:

$$dU_i = \left\{ \begin{array}{l} -\dfrac{\delta_{ij} C_0 \varepsilon}{2} V_{jk}^{-1} U_k^{'} + \dfrac{1}{2}\dfrac{\partial V_{il}}{\partial x_l} + \overline{u}_l \dfrac{\partial \overline{u}_i}{\partial x_l} + \dfrac{\partial \overline{u}_i}{\partial t} + \\ + \left[ \dfrac{1}{2} V_{lj}^{-1} U_j^{'} \left( \dfrac{\partial V_{il}}{\partial t} + U_m \dfrac{\partial V_{il}}{\partial x_m} \right) \right] \end{array} \right\} dt + \sqrt{C_0\varepsilon}\, d\xi_i = \qquad \text{E.11}$$

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## 2.3. Micromixing scheme: molecular diffusion

- IECM (Interaction by the Exchange with the Conditional Mean) micromixing scheme (Pope 1998, Sawford 2004); $t_m$ is the mixing time scale, T the transformation term :

$$\frac{dC}{dt} = -\frac{C - \langle C|\underline{U}\rangle}{t_m} \qquad \text{E.12}$$

- Mixing time scale (Leuzzi et al. 2012 AtmosEnviron; $\sigma_{z0}$ is the source length scale, $t_f$ the fly time, $C_\varphi$ Richardson constant, ε: dissipation rate of turbulent kinetic energy q):

$$t_m = 0.8(3/2)^{-1/2}\left[(3/2)^{1/2}\frac{\sigma_{z0}^{2/3}}{\varepsilon^{1/3}} + \sqrt{2T_L t_f}\right] \le \frac{2q}{C_\varphi \varepsilon} \qquad \text{E.13}$$

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# 2.4. Conserved scalar theory (reactions)

(**a simplified reaction scheme in stationary regimes**: **2-stage model**; e.g. 2nd order kinetics with 2 species -A and B-; Sawford 2006 BLM)

- Definition of the mixture fraction $F_m$ ($F_{ms}$: stoichiometric $F_m$, $C_{A,1}$ and $C_{B,2}$: concentrations of unmixed species); Bilger et al.91:

$$F_m \equiv \frac{C_A - C_B + C_{B,2}}{C_{A,1} + C_{B,2}} \quad \Rightarrow \quad T = -rC_AC_B = rC_A\left[C_A + \left(F_{m,s} - F_m\right)\left(C_{A,1} + C_{B,2}\right)\right] \qquad \text{E.14}$$

- Modelling $F_m$ as a passive scalar and use of its instantaneous values to determine ($C_A$,$C_B$,$C_C$), according to alternative formulations («reaction limits»)
- **Reaction-Dominated limit** (**RDL**; **«State-of-the-art solution»**; instantaneous mixing at the inlet section with on-going passive FL concentrations as initial values; contact/reaction time = fly time):

$$C_A = C_{A,1}F_m \frac{\left(F_{m,s} - F_m\right)}{F_{m,s}\left(1 - F_m\right)e^{\left[\left(F_{m,s} - F_m\right)r\left(C_{A,1} + C_{B,2}\right)t_f\right]} - F_m\left(1 - F_{m,s}\right)}$$

$$C_C = F_m C_{A1} - C_A$$

E.15

$$C_B = C_A + \left(C_{A,1} + C_{B,2}\right)\left(F_{m,s} - F_m\right) = C_{B,2}\left[\frac{\left(F_{m,s} - F_m\right)}{F_{m,s}} + \frac{\left(1 - F_{m,s}\right)C_A}{F_{m,s}C_{A,1}}\right]$$

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## 2.5. Proposed variants for CST: Non-Homogeneous Reaction-Dominated Limit

- **Motivation:** RDL (state-of-the-art scheme) systematically overestimates reactions under non-homogeneous conditions
- **Non-Homogeneous Reaction-Dominated Limit (NHRDL); «New numerical solution»**

  Under non-homogeneous conditions, RDL reaction/contact time is computed as modified contact time, depending on the product of the reactant inst. concentrations along the particle trajectory :

$$t_{NHRDL,I}(t_f^*) = \frac{\int_0^{t_f^*} C_{A,FL}(t_f) C_{B,FL}(t_f) dt_f}{\int_0^{t_f^*} C_{A,FL}(t_f^*) C_{B,FL}(t_f^*) dt_f} t_f^* = \frac{\int_0^{t_f^*} C_{A,FL}(t_f) C_{B,FL}(t_f) dt_f}{C_{A,FL}(t_f^*) C_{B,FL}(t_f^*)} \quad \text{E.16}$$

- NHRDL: corrected contact time (discrete formulation):

$$t_{NHRDL}(t_f^*) = \frac{\sum_{l=0}^{n_{step}(t_f^*)-1} C_{A,FL}(t_f^* - l \cdot dt) \cdot C_{B,FL}(t_f^* - l \cdot dt) \cdot dt(t_f^* - l \cdot dt)}{C_{A,FL}(t_f^*) \cdot C_{B,FL}(t_f^*)} \quad \text{E.17}$$

- **No fluctuation RDL (NFRDL)**: RDL limit based on the mean values of $F_m$ and mean $t_f$(=X/U in this case)

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# 3.1. Test case (Brown-Bilger 96-98a) - numerical settings

## We keep working in 3D even if the case is 2D

- Domain size: Lx*Ly*Lz=17M*7M*7M (unbounded)
- $U_{ref}$ = 0.5m/s, dx=M/10, dt=0.001s
- M(grid spacing)=0.320m, $x_g$=-3M (grid position)
- $x_s$=(0;Ly/2;Lz/2) (source position)
- C(NO,source)=515ppm(D=31.5mm),C(O3,back.)=1.0ppm
- Kolmogorov constant ($C_0$)=2 (model first choice, close to 2.7: average chosen value for grid turbulence)
- Micromixing constant values: 0.45 (chosen according to the state-of-the-art procedure), 0.40 (Cassiani et al. 2013, BLM), 0.65 (most used value)
- Sim. particles: 7e6+7e6 (fast); 7e6+7e7(ref.); 2e7+2e8(long)
- 100 velocity classes for the conditional mean
- Only reaction: 2nd order kinetics (2 reactants, 1 product; r=0.37ppm/s) $NO + O_3 \rightarrow NO_2 + O_2$
- **Decaying grid turbulence** (main flow):

$$\sigma_v^2 = \sigma_w^2 = A * 0.041 * \bar{u}^2 \left( \frac{x - x_g}{M} \right)^{-c}, \qquad c = \begin{cases} 1.7, & \frac{x}{M} \le 10, \quad A = 2.68 \\ 1.27, & 10 \le \frac{x}{M} \le 17, \quad A = 1 \end{cases}$$

$$\varepsilon = -\frac{3}{2}\frac{\partial \sigma_w^2}{\partial t} = \frac{3}{2}\sigma_w^2 \frac{c\bar{u}}{(x - x_g)}$$

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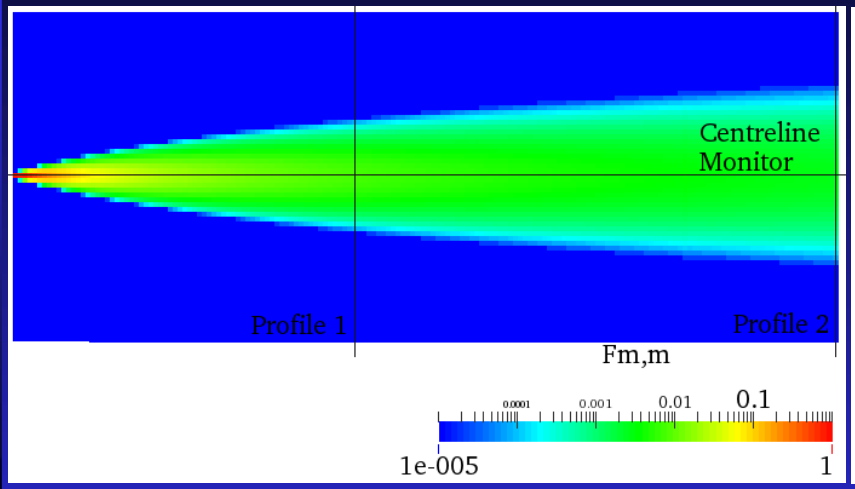
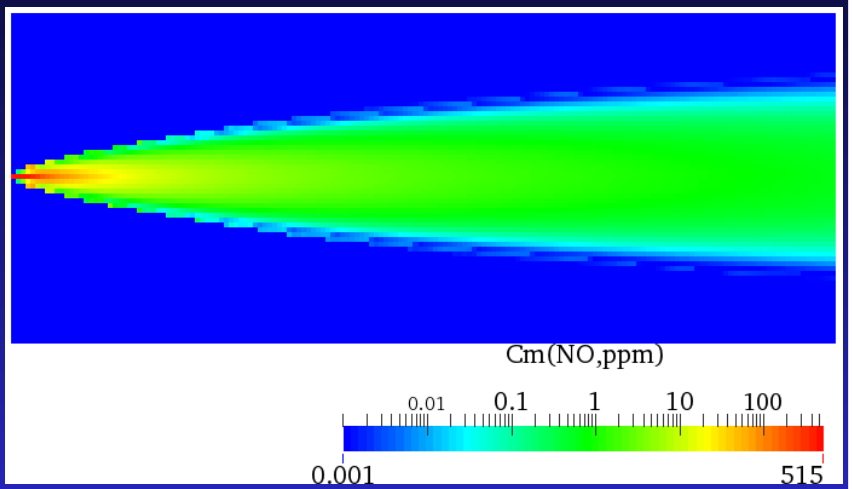
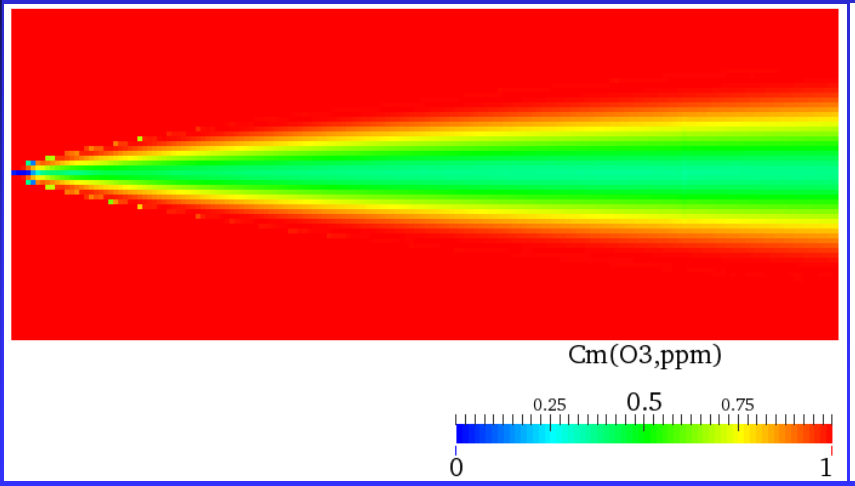
# 3.2. Mean concentrations (fields)
# Vertical slices containing the plume centreline



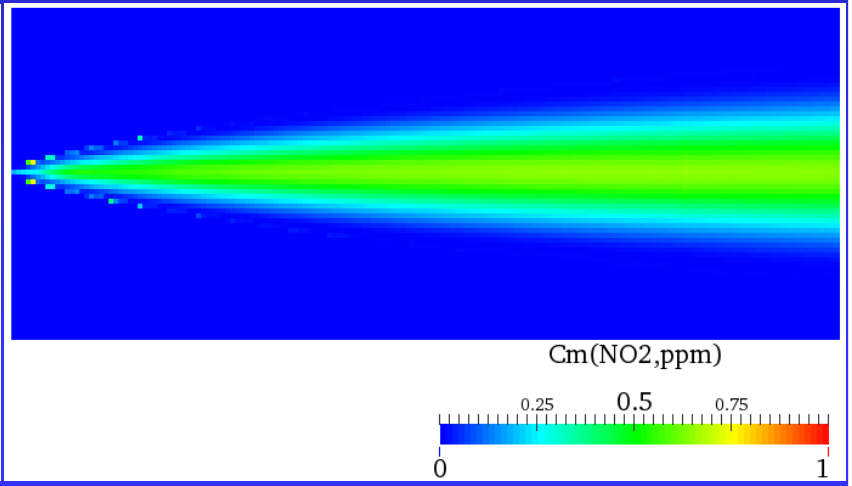

$F_{m,m}$ (y=0)




$C_m$ (NO) (y=0)

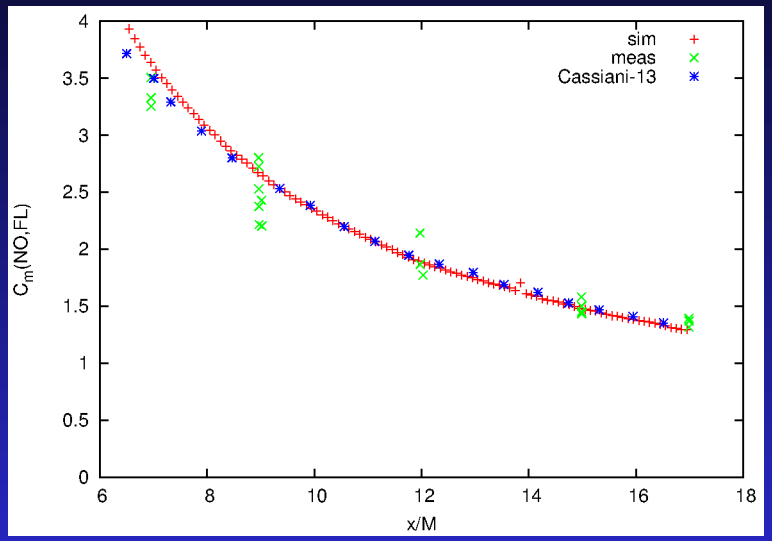



$C_m$ ($O_3$) (y=0)

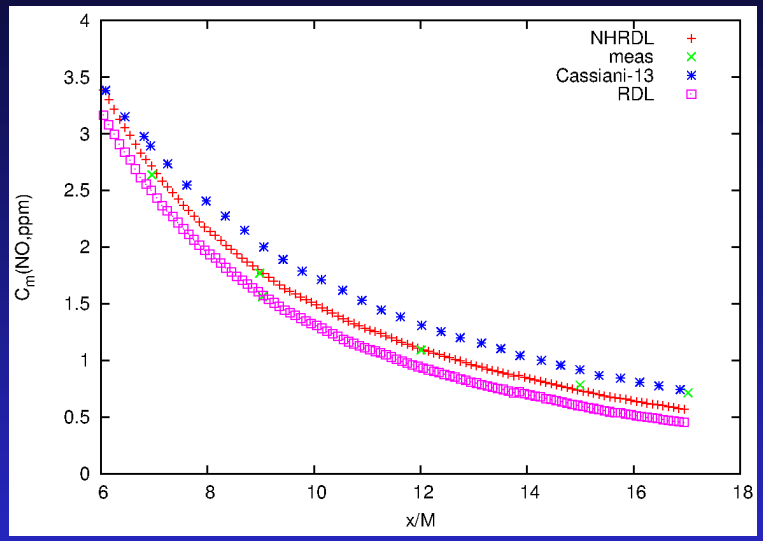



$C_m$($NO_2$) (y=0)

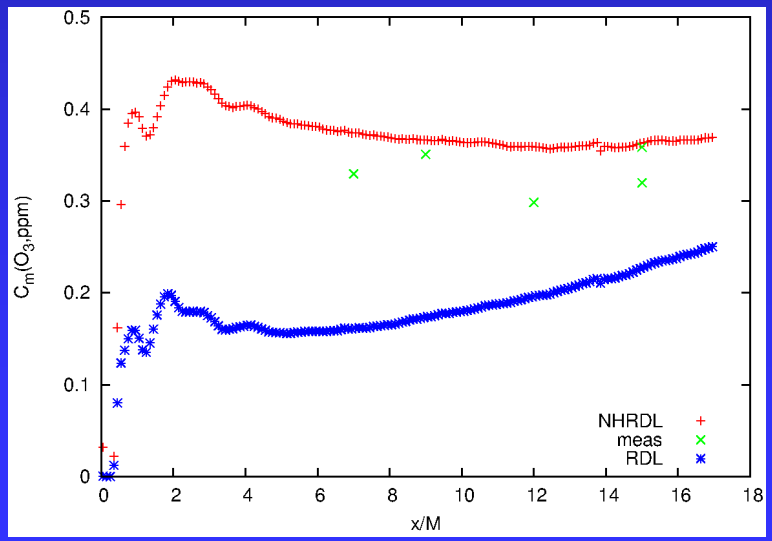
# 3.2. Mean concentrations (validation and inter-comparisons, centreline)



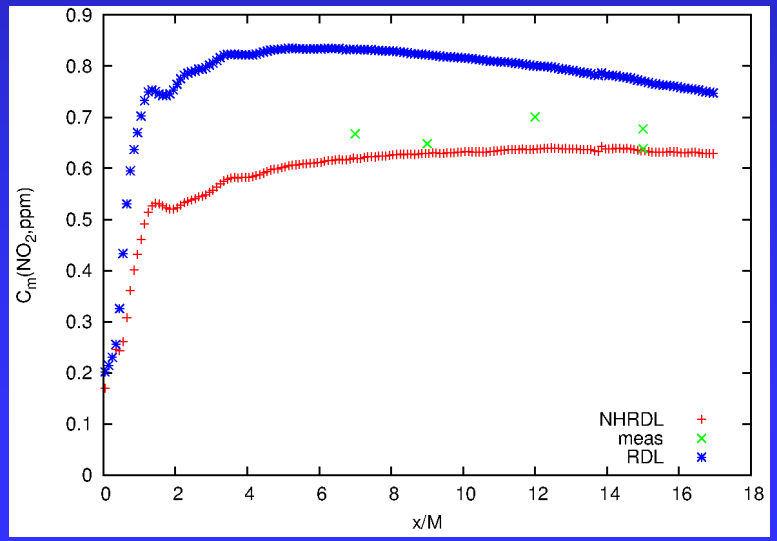

$F_{m,m}$ (x,y=0,z=0)




$C_m(NO)$ (x,y=0,z=0)

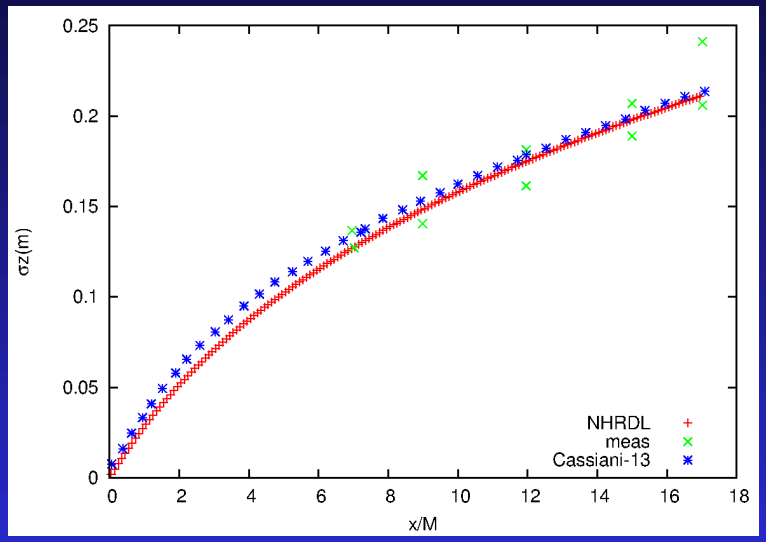



$C_m(O_3)$ (x,y=0,z=0)

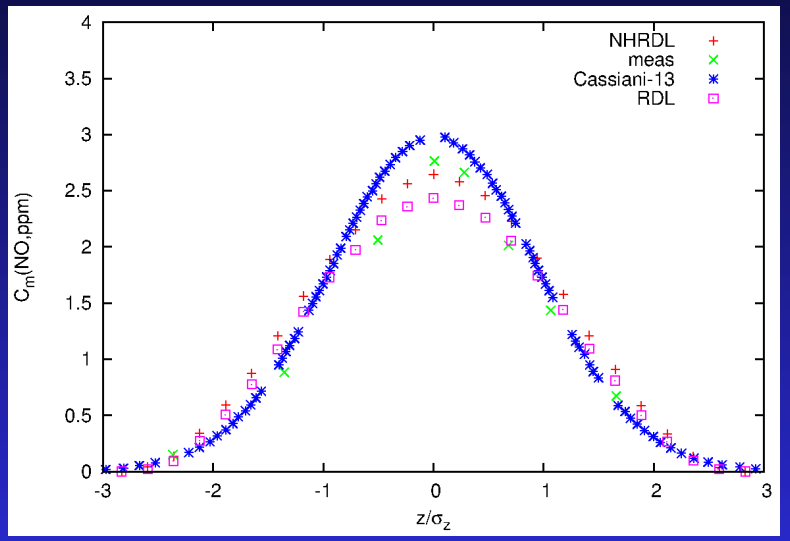



$C_m(NO_2)$ (x,y=0,z=0)

# 3.2. Mean concentrations (Plume spread and profiles)

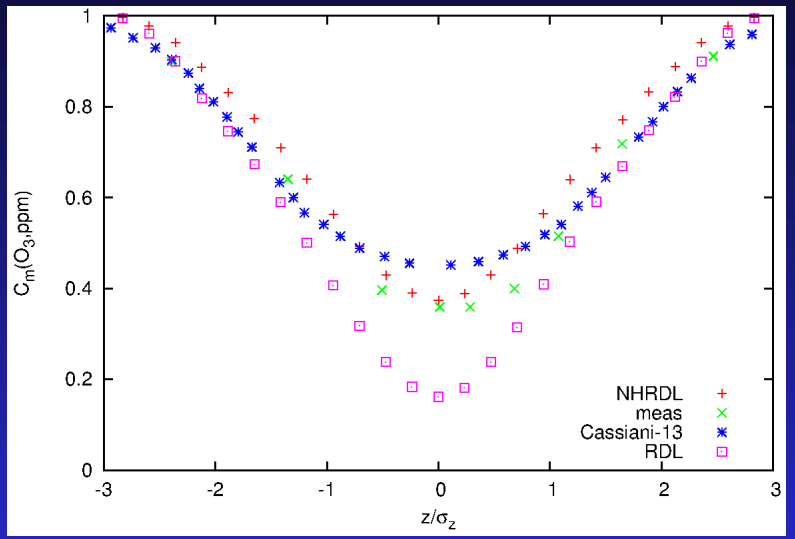



Plume spread $\sigma_Z$ (x)

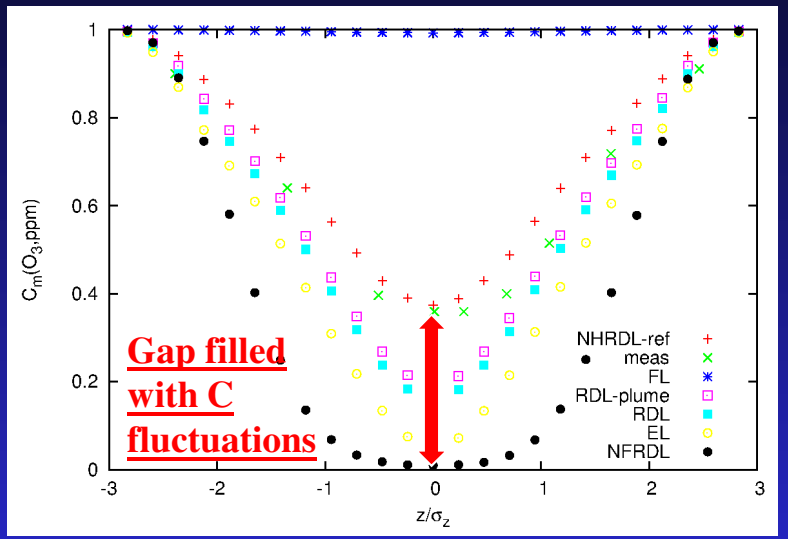



$C_m$(NO) (x=7M,y=0,z)

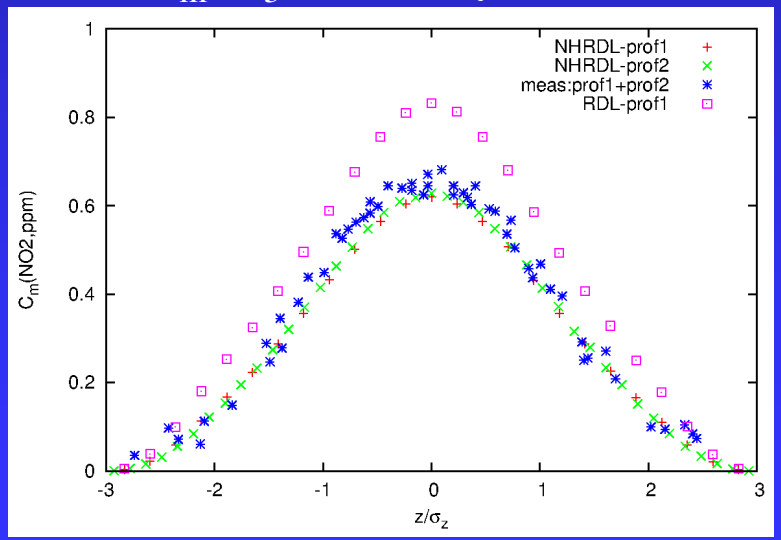
# 3.2. Mean concentrations (validation and inter-comparisons, profiles)



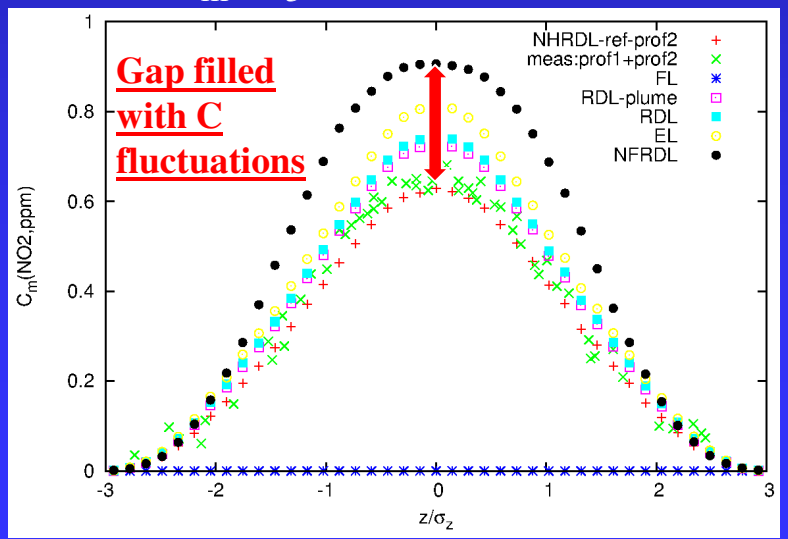

$C_m(O_3)$ (x=7M,y=0,z)




$C_m(O_3)$ (x=7M,y=0,z)




$C_m(NO_2)$ (x=7M-17M,y=0,z)




$C_m(NO_2)$ (x=17M,y=0,z)

# Summary

1. Introduction
   1. Scalar reactions and concentration fluctuations
   2. Lagrangian micromixing modelling
2. Numerical model (Leuzzi et al.)
   1. Main features
   2. Macromixing scheme (Lagrangian turbulence)
   3. Micromixing scheme (molecular diffusion)
   4. Conserved scalar theory (CST, scalar reactions) (new)
   5. Proposed variants for CST: NHRDL (and RDL-plume) and NFRDL (new)
3. 2D validation ($2^{nd}$ order kinetics) in grid turbulence (point source + backg)
   1. Test case and numerical settings
   2. Mean concentrations and mean plume
   3. Standard deviations of concentration
   4. Intensities of fluctuations
   5. Segregation coefficients and reaction rates
   6. Concentration skewnesses
   7. Concentration kurtosis
   8. Sensitivity to the micromixing constant
   9. Convergence
   10. Simplified modelling of the product species
4. 2D validation ($2^{nd}$ order kinetics) in grid turbulence (line sources)
5. Conclusions

**Results available via email (manuscript under revision)**
**No time here to discuss them**

# 4. Conclusions

1. **Implementation of a state-of-the-art CST schemes (Reaction-Dominated Limit RDL)** into a 3D Lagrangian micromixing model for reactions
2. **Definition and implementation of a modified formulation under Non-Homogeneous conditions (NH-RDL limit)** for CST
3. 2D **validation** (best available) on grid turbulence with 2nd order kinetics (2 reactants, 1 product): **$C_m$, $\sigma_C$, $S_{k,C}$, $K_{u,C}$, $I_s$, $i_C$, $R_r$ for $F_m$, NO, $O_3$, $NO_2$.**
4. **Inter-comparisons** with Cassiani 2013 BLM
5. **Fluctuations cannot be neglected in modelling $C_m$ (reactive pollutants)**
   Only passive scalar C means don't need concentration fluctuations (Pope98)
   For several years HARMO hosted a dedicated topic on C fluctuations.
6. **Noticeable advantages using NH-RDL limit** to correct the reaction overestimations of RDL under non-homogeneous concentration fields
7. **Lagrangian micromixing models are much faster than DNS and pdf models** (both for means and higher order statistics); these all represent a small minority among the air quality codes. **Other models normally do not treat reactions depending on concentration fluctuations (ad-hoc tuning for $I_s$).**

**Thanks for your attention**

**Any correction and suggestion is highly appreciated**