

A LAGRANGIAN DISPERSION MODEL WITH CHEMICAL REACTION

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INTRODUCTION

Lagrangian stochastic models are recognized to be powerful tools for pollutant dispersion at different scales in complex terrain and different stability conditions. The only shortcoming is the difficulty to include chemical reactions when NO₂ or Ozone concentrations have to be predicted in presence of NO_x emissions. This is mainly due to two reasons. As a matter of fact, from a theoretical point of view, the particle trajectory are usually considered independent and hence the concentration correlations and fluctuations cannot be calculated. This problem can be overcome by developing a two particle model (*Thomson 1990*) as done by *Crone et al. 1999* and *Van Dop 2001*. Unfortunately the theory of such models is limited to ideal turbulence assuming homogeneity and isotropy and hence this model cannot be applied in real conditions. The second problem is the limit of the computational resources which does not allow to reproduce the Ozone background concentrations.

In this work a Lagrangian stochastic (single particle) model is modified in order to account for simple chemical reactions and tested against measured wind tunnel data. These data refers to a reactive plume generated by a single point source of NO inside a turbulent grid flow doped with Ozone (*Brown and Bilger 1996*). The comparison between measured and simulated data has been made at different distance from the single point emission and has considered all the substances (NO, NO₂, O₃) involved in the chemical reactions and diffusion processes. In the model the cross correlation term between the concentration of the different chemistry compounds is neglected or parameterised. Despite of this simplification the results are rather satisfactory and seem to encourage the use of the model in actual practical applications in real atmosphere.

MODEL

SPRAY (*Tinarelli et al. 1994*) is a Lagrangian stochastic particle model designed to study the pollutants dispersion in complex terrain. It is based on the Langevin equation for the turbulent velocities (*Thomson 1997*), whose coefficients depend on a solution of the Fokker-Planck equation for a given Eulerian probability density function (PDF) of the turbulent velocity and on the inertial range turbulence theory respectively. In the two horizontal directions the PDF is assumed to be Gaussian. In the vertical direction the PDF is assumed to be non-Gaussian, so to deal with non-uniform turbulent conditions and/or convection. The vertical dispersion calculation is based on the generalized Langevin equation (*Thomson 1997*). The equations prescribing the evolution of the vertical velocity fluctuation w and the displacement z are the following:

$$dw = a(z, w)dt + \sqrt{C_0 e} dt dm$$

$$dz = w dt$$

where dm has zero mean and unit variance, C_0 is a constant and e is the dissipation rate of turbulent kinetic energy. $a(z, w)$ must be determined by solving the Fokker-Planck equation, obtaining:

$$a(z, w) = \frac{1}{P} \left(B_0 \frac{\partial P}{\partial w} + \Phi \right)$$

where $B_0 = \frac{1}{2} C_0 \mathbf{e}$ and, as suggested by Thomson 1997,

$$\Phi = - \frac{\int_{-\infty}^w w P(z, w) dw}{\int_{-\infty}^w P(z, w) dw}$$

$P(z, w)$ is the PDF that must be prescribed from the available measurements or parameterisations. In the present work, we used the Gram-Charlier PDF (Ferrero and Anfossi, 1998). The model makes use of the Gaussian assumption in the horizontal directions.

The dispersion model is coupled to the circulation model RAMS, which provides the mean flow. Following (Chock and Winkler, 1994a, 1994b) chemistry and dispersion are treated separately and sequentially, each particle (m) released by the source may bring mass of different substances. At each time step the particle position $X^{(m)}(t)$ is update using the stochastic model equation, then the concentrations of each substance is calculated in a fixed Eulerian grid and the chemistry is computed.

The basic equations for chemistry are

$$\frac{\partial c_A}{\partial t} = \frac{\partial c_B}{\partial t} = - \frac{\partial c_C}{\partial t} = -k c_A c_B$$

where $C_A(t)$, $C_B(t)$ e $C_C(t)$ are the concentration of the substances in the cells where the *well mixed condition* is supposed to be attained. The discretized form of the chemical equations are:

$$\begin{aligned} \langle c_A(\mathbf{x}_j, t_1) \rangle &= \langle c_A^*(\mathbf{x}_j, t_1) \rangle - k \Delta t \langle c_A^*(\mathbf{x}_j, t_1) \rangle \langle c_B^*(\mathbf{x}_j, t_1) \rangle \\ \langle c_C(\mathbf{x}_j, t_1) \rangle &= \langle c_C^*(\mathbf{x}_j, t_1) \rangle + k \Delta t \langle c_A^*(\mathbf{x}_j, t_1) \rangle \langle c_B^*(\mathbf{x}_j, t_1) \rangle \end{aligned}$$

where the asterisk indicates the old concentration in the cells obtained after the Lagrangian particles displacements but before the chemical reaction. Then the mass (M) of the different substances (α) are redistributed to each particle (n) in the cell through the following scheme:

$$\begin{aligned} M_a^{(n)}(t_1) &= \langle c_a(\mathbf{x}_j, t_1) \rangle \frac{V_j}{N_j} \quad \text{if} \quad \langle c_a^*(\mathbf{x}_j, t_1) \rangle = 0 \\ M_a^{(n)}(t_1) &= M_a^{(n)}(t_0) \frac{\langle c_a(\mathbf{x}_j, t_1) \rangle}{\langle c_a^*(\mathbf{x}_j, t_1) \rangle} \quad \text{otherwise;} \end{aligned}$$

where t_0 and t_1 indicates the time before and after the reaction, V_j and N_j are the volume and the number of particles inside the cell. In order to simulate the chemical reaction in a turbulent flows on a time scale less than the typical equilibrium scale the cross covariance term between the concentration fluctuation $\langle a'b' \rangle$ of the two compounds participating to the reaction should be accounted for. The presence of this term is often referred as “segregation” and

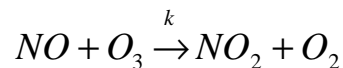
$$\mathbf{a} = \frac{\langle c_A' c_B' \rangle}{\langle c_A \rangle \cdot \langle c_B \rangle}$$

is the segregation coefficient.

RESULTS AND DISCUSSION

The test to verify the validity of the chemical algorithm introduced in SPRAY has been to reproduce the wind tunnel experiment performed by Bilger *et al.*, 1996 and already examined

by Van Dop, 2001. During the experiment a point source emission of NO has been set inside a flux of air doped with a uniform O₃ concentration, therefore the chemical reaction



will take place inside the plume. The NO, O₃ and NO₂ concentrations will be measured over planes perpendicular to the air flux at different distance from the source and along the plume centreline. The air with an O₃ concentration of 1 ppm enters the working section, which is 2.8 m in diameter and 8 m long, and crosses the turbulence generating grid made of square bars 65 mm x 65 mm and of pitch M= 320 mm. The nominal mean axial velocity of the flow (\bar{U}) is of 0.5 ms⁻¹ giving a Reynolds number $Re = \bar{U} M/\nu = 10700$ where ν is the cinematic viscosity. The point source has a diameter of 31.5 mm and is located at the centre of the working section at a distance of 3M (0.96 m) from the grid. Air with a velocity matched to \bar{U} and with a NO concentration of 515 ppm is released from the point source. The heat release at the concentrations used causes negligible rise in temperature. Turbulence and concentrations measurements have been performed at different distance from the point source ($x/M=7, 9, 12, 15, 17$) along arcs concentric to the plume centreline.

Constant background Ozone

The simulations of the chemical reactions were performed by considering a constant background ozone concentration. This approach allows to use particles only for simulating the inside plume concentrations. In this way a great amount of computer resources can be avoided. Nevertheless this particular condition cannot be considered completely realistic, because the ozone concentration should decrease inside the plume due to the reaction with NO. Hereafter we present the results of the simulation performed with a 0.01 m grid space in the cross wind direction for the concentration calculation. The scheme for the chemistry was applied every 1, 10, 20 and 40 time steps in the different simulations respectively.

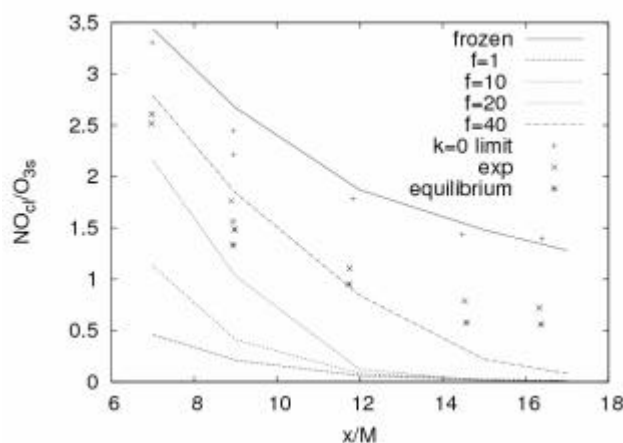


Figure 1; Centerline NO mean concentration normalized by O₃ source concentration.

Figure 1, shows the NO trend, normalized by the O₃ concentration at the source, as a function of the distance from the source, normalized by the wind tunnel turbulence grid size (M). The results of the “frozen case” (K=0), “equilibrium case” (K=8) and the cases with chemistry scheme activated at the different time steps frequency. For sake of comparison the measured wind tunnel data are also reported. It can be observed that, while the frozen case agrees with experimental data, the simulation performed with the chemistry scheme activated show a remarkable discrepancies which increase with the distance and with the frequency of the chemical reactions computation.

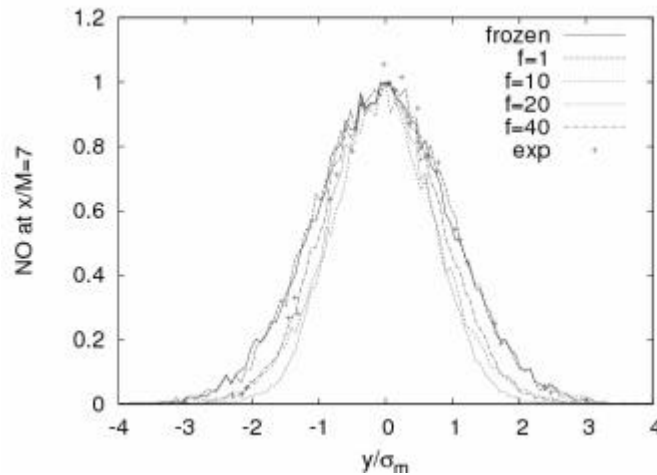


Figure 2; Mean NO concentrations at $x/M=7$ normalized by the centerline value.

Figure 2 shows the results corresponding to the transversal sections at $x/M=7$ for NO normalized with the centerline value, as a function of the cross wind direction y , normalized with the plume half width (σ_m). Although the agreement between predicted and observed values at the nearer sections seems to be rather good, thanks to the normalization with the centerline values, the results become worse as the distance increases (farthest distances are not shown here) due to the great number of zero NO concentration particles.

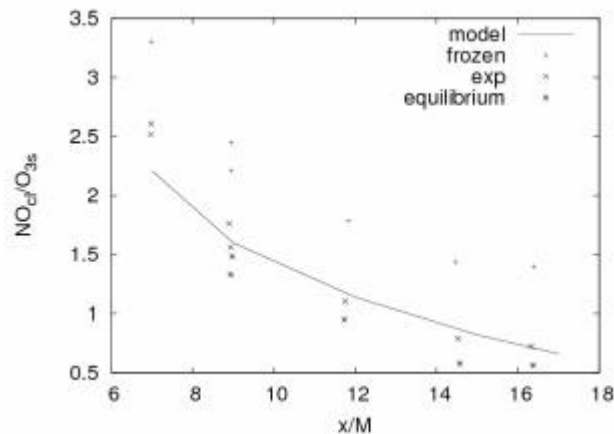


Figure 3; As in Figure 1 but with the modified background O_3 concentration and parameterized segregation coefficient

Modified background Ozone concentration

In order to perform a more realistic simulation, we considered a non uniform distribution of ozone accounting for a the lower concentrations inside the plume. In this simulation the cross-wind resolution was reduced to 0.02 m. Furthermore, a parameterization of the segregation coefficient was tested:

$$\langle C_A C_B \rangle = \langle C_A \rangle \cdot \langle C_B \rangle - C_{A0} C_{B0} G(y, z)$$

where C_{A0} e C_{B0} are the initial concentrations and $G(y,z)$ is a Gaussian function along the cross wind directions, obtained by experimental data interpolation. The NO normalized “center line” trend is shown in figure 3; it can be observed a better agreement with experimental data which demonstrates as the parameterization of the segregation coefficient together with the lower O_3 concentrations inside the plume act successfully also at greater distances. As a matter of fact, due to the segregation the reaction proceeds slower maintaining sufficiently high the NO concentrations.

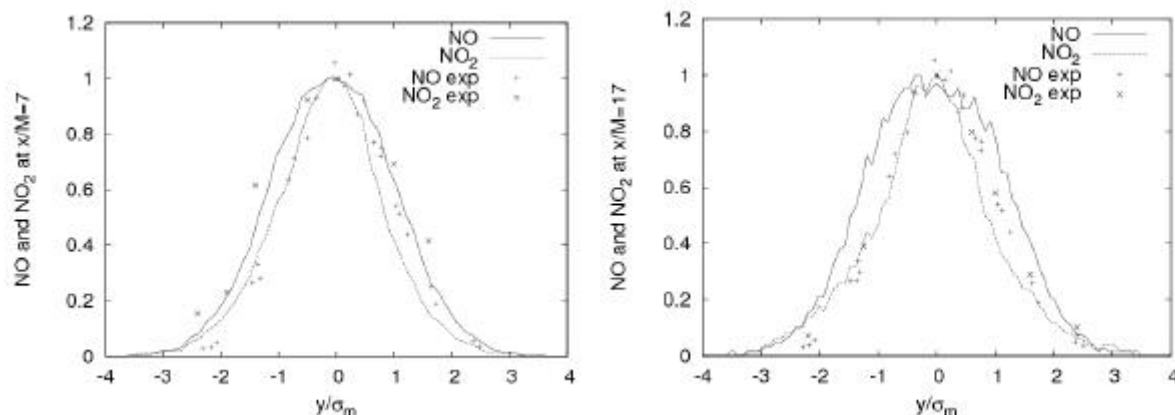


Figure 4 a,b; Mean NO and NO₂ concentrations at $x/M=7$ (a: left) and $x/M=17$ (b:right) normalized by the centerline values for the same case as Figure 3.

The NO and NO₂ concentration distributions in ppm at $x/M=7$ and 17 as a function of the normalized cross-wind directions, compared with measurements are respectively shown in Figure 4 a and b. Although the O₃ profiles inside the plume is not usually known in practical applications the results are encouraging and suggest the possibility to use this general approach in real cases.

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