PLPM (PHOTOCHEMICAL LAGRANGIAN PARTICLE MODEL): FORMULATION AND PRELIMINARY VALIDATION.

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INTRODUCTION

PLPM (Photochemical Lagrangian Particle Model) is a Lagrangian particle model developed in last years by ENEA and ENVIROWARE Srl. In this presentation, we discuss the physical formulation of PLPM, the main features of the kernel density reconstruction methods and some preliminary tests carried out to evaluate the model and the density reconstruction methods. In particular, the Kincaid data set (Bowne e Loondergan, 1983), along with the Model Validation Kit (Olesen, 1998), has been used to test the model on the dispersion of non–reactive compounds. The results obtained so far suggest bringing them to the attention of the air quality modelling community, together with the questions that are still open and would benefit of the efforts of other researchers.

MODEL DESCRIPTION

General features

PLPM is a three-dimensional dispersion model interfaced to the diagnostic meteorological model CALMET (Scire et al., 1999). Within the model a fixed number of particles is released by each source at each time step. Each released particle can be assumed to be composed by several pollutants (i.e. each particle contains information regarding different substances). In the complete version of the model, the pollutants in each particle will vary qualitatively and quantitatively with time since the chemical reactions may result in the production or loss of some species. These portions of atmospheric pollutants are macroscopic since include a large number of molecules, but they are small enough to be considered as points.
Particles motion is the resulting effect of the mean wind field and the turbulent diffusion. The particles' random walk induced by turbulence is Markovian. Given the position of a particle at time t, its position at the time t+Δt is be given by:

$$x_i(t+\Delta t) = x_i(t) + \Delta t\left(u_i + u_i'\right) \tag{1}$$

where i=1,2,3 indicates respectively the x, y and z direction, u_i is the mean wind component along the i-th direction and u_i' represents the turbulent velocity fluctuation along the same i-th direction. The time evolution of the velocity fluctuation is described in the most general terms by the non-linear Langevin equation introduced by Thomson (1987):

$$du_i' = a_i\left(x, \underline{u}', t\right)dt + b_{ij}\left(x, \underline{u}', t\right)d\xi_j(t) \tag{2}$$

Where a_i and b_{ij} are functions of space, velocity and time, and $d\xi_j(t)$ is a random increment of a Wiener process with independent components.

The fluctuating turbulent term at time *t* is correlated to the one at time $t+\Delta t$: turbulence reminds its previous state for a certain period, and a Lagrangian time scale can be defined as the value at which the autocorrelation coefficient is equal to *1/e*. The Lagrangian time varies for different turbulent regimes. Under convective conditions two models can be alternatively used to describe the turbulent vertical motion: the homogeneous skewed model of Hurley and Physick (1993) and the quasi-homogeneous model of Bianconi et al. (1999), both based on Thomson (1987) model.

The concentration field

The most common method of computing the concentration field is the *box counting*. It simply consists in defining a grid, summing the particles' masses in each cell and dividing by the grid cell volume to obtain the concentration value. This method has been shown to suffer of a major disadvantage: the concentration values depend on the cell volume and on the number of particles used. For large volumes the concentration field is too smoothed, while for small volumes the concentration field is perturbed by a high numerical noise, unless a very high number of particle is used

A different numeric technique for computing concentrations in a Lagrangian particle model is the *kernel density estimator,* which permits to reduce the number of particles and to compute a completely grid-free and continuous concentration field in each point of the domain.

The kernel estimator implemented in PLPM has the following form

$$c(x,y,z,t) = \sum_{i=1}^{n} \frac{m_i}{\lambda_x \lambda_y \lambda_z} d\left(\left|\frac{x_i - x}{\lambda_x}\right|\right) d\left(\left|\frac{y_i - y}{\lambda_y}\right|\right) d\left(\left|\frac{z_i - z}{\lambda_z}\right|\right) \tag{3}$$

where $c(x,y,z,t)$ is the concentration in (x,y,z) at time *t*; *n* is the total number of particles in the domain, (x_i, y_i, z_i) and m_i are the position and the mass of *i*-th particle and $d(u)$ is the Epanechnikov function:

$$d(u) = \begin{cases} \frac{3}{4}(1-u^2) & |u| \le 1 \\ 0 & |u| > 1 \end{cases} \tag{4}$$

The λ_i parameters are the *bandwidths* (one for each space direction). They states the amplitude of the volume in which the mass of the particle is spread in the domain. Bandwidths are the critical parameters for the good estimation of the concentration field: too small bandwidths can lead to a largely irregular concentration field (large variance) whereas bandwidths overestimation can result in a large bias of the reconstructed field. Thus, an optimal method for bandwidth calculation is needed, aimed to minimize both variance and bias.

Bandwidth calculation

In the present version of PLPM four different methods are available for bandwidth calculation:

Kernel RL3 **(Receptors-based, Locally-defined, three-dimensionally ordered)**

- Bandwidths are *associated to the receptors*, i.e. the points where the concentration has to be estimated.
- For each receptor *all particles are ordered* by their distance from the receptor.

- A group of nearest neighbours of the receptor is defined as containing the closest particles carrying one eightieth of the total particles mass (for each chemical species)
- The bandwidth associated to the receptor, in each direction, is the maximum projection of the distance of the nearest neighbours in that direction.

Kernel PG **(Particles-based** and **Globally-defined)**

- Bandwidths are *associated to the particles*. The same set of bandwidth, one for each space direction, is associated to each particle.
- Bandwidths are calculated, in each direction as (De Haan,1999):

$$\lambda_i = \alpha \cdot A(K) \cdot n^{-\frac{1}{d+4}} \cdot \min\left(\sigma_i, \frac{R_i}{1.34}\right) \quad \text{with } i=1, \ldots, d$$

where σ_i and R_i are the standard deviation and the interquartile range of the particles distribution in each space direction; n is the total number of particles, d the number of space dimensions ;α is a tuning parameter (set equal to 0.85 by DeHaan) and $A(K)$ is a parameter depending on the particle distribution and the kernel function.

Kernel PL1 **(Particles -based**, **Locally-defined**, **one-dimensionally ordered)**.

- Bandwidths are *associated to the particles* and each particle has a different set of bandwidths.
- The neighbourhood for each particle is defined again as containing the closest particles carrying one eightieth of the total particles mass (for each chemical species) but *three different neighbourhoods are defined for each space direction,* based on the projection of particles distances on the three axis.
- For each direction the bandwidth is set as the projected distance of the first particle excluded from each of the neighbourhoods.

Kernel PL3 **(Particles-based**, **Locally-defined, three-dimensionally ordered)**

- Bandwidths are *associated to the particles* and each particle has a different set of bandwidths.
- A *single neighbourhood* for each particle is defined again as containing the closest particles carrying one eightieth of the total particles mass (for each chemical species). *Three-dimensional distances are considered in the ordering process.*
- In each direction the bandwidth is set as the maximum projection of the distances of the nearest neighbours in that direction.

PRELIMINARY VALIDATION

The performances of the inert version of PLPM have been compared with the data set of the well known Kincaid release experiment. Meteorological fields have been calculated by means of CALMET, starting from the data of the National Weather Service included in the kit[1]. The only source of the experiment emits a plume of buoyant SF_6 inert gas. The present version of PLPM does not include a gradual plume rise algorithm, thus the source height has been corrected to an effective height $h_{eff}=h_s+\Delta h$. Δh has been calculated by means of the Briggs formulae as reported in Seinfeld and Pandis (1998), pag.932. Hourly emissions have been divided in 120 puffs

[1] Due to the simple orographic and meteorological features of the Kincaid domain, using CALMET is not strictly necessary and good results could be achieved also through simpler meteorological preprocessors. Anyway, the strict interdependence of PLPM and CALMET suggested to proceed with the complete model suite.

released every 30 seconds, each containing 30 particles, for a total emission rate of 3600 particles/hour.

Performance indexes

In Table 1 the usual performance indexes for model validation are shown. Selected data consist of the Kincaid subset with quality index Q ≥ 2. The four kernel based approaches described in previous section are analyzed together with the box counting method for density reconstruction.

Table 1. Performance indexes for different concentration calculation approaches on the data set with Q ≥ 2.

Q=2,3 (N=586)	Average [ng/m^3/(g/s)]	Sigma [ng/m^3/(g/s)]	Bias [ng/m^3/(g/s)]	NMSE	R	FAC2	FB	FS
Measures	40.96	39.26	0.00	0.00	1.000	1.000	0.000	0.000
Box Counting	128.35	119.41	-87.39	4.31	0.083	0.065	-1.032	-1.010
Kernel RL3	19.47	19.99	21.49	3.07	-0.029	0.301	0.711	0.651
Kernel PG	18.09	19.09	22.87	3.00	0.137	0.300	0.775	0.691
Kernel PL1	26.17	35.42	14.79	2.60	0.082	0.309	0.441	0.103
Kernel PL3	21.88	21.26	19.08	2.56	0.040	0.334	0.607	0.595

In Table 2 similar results are shown for the Kincaid data with Q = 3.

Table 2 . Performance indexes for different concentration calculation approaches on the data set with Q = 3.

Q=3 (N=338)	Media [ng/m^3/(g/s)]	Sigma [ng/m^3/(g/s)]	Bias [ng/m^3/(g/s)]	NMSE	R	FAC2	FB	FS
Measures	54.34	40.25	0.00	0.00	1.000	1.000	0.000	0.000
Box Counting	141.17	115.97	-86.84	2.99	-0.037	0.092	-0.888	-0.969
Kernel RL3	18.27	18.14	36.07	3.32	-0.029	0.269	0.993	0.758
Kernel PG	18.45	16.51	35.88	3.02	0.119	0.260	0.986	0.836
Kernel PL1	25.30	26.50	29.04	2.24	0.044	0.272	0.729	0.412
Kernel PL3	20.96	19.12	33.37	2.64	0.060	0.311	0.886	0.712

Performance indexes clearly show that the kernel based method is largely preferable to the box counting. All the kernel methods lead to a positive bias, i.e. a general underestimation of the concentration data. As far as the four bandwidths calculation methods are concerned, particles-based methods (PG, PL1 and PL3) perform better than the RL3 receptor-based. Also local approaches (PL1 and PL3) seem preferable to the PG global approach. Performances of PLPM are comparable to performances of other regulatory models as AERMOD or ISCST3 on the same data set (CERC, 2001).

Residual analysis

A residual analysis[2] has been performed to evaluate model performances for different values of some significant parameters (as emission temperature or speed, stability class, wind speed and so on). Five percentiles (5th, 25th, 50th, 75th and 95th) of the residual distributions for different values of the emission temperature and for the PL3 kernel approach are shown in figure 1. Models perform better as much the 50th percentile of the residual distribution is close to the unity and the distribution is stiff around the unity value.

Figure 1 shows as model performance is better for low temperature values, when the effect of the buoyant plume rise is smaller. All the other reconstruction methods (not reported here) show a similar trend. Higher emission temperatures lead to a clear underestimation of the predicted values confirming the need for a detailed treatment of gradual plume to be implemented in the next version of PLPM.

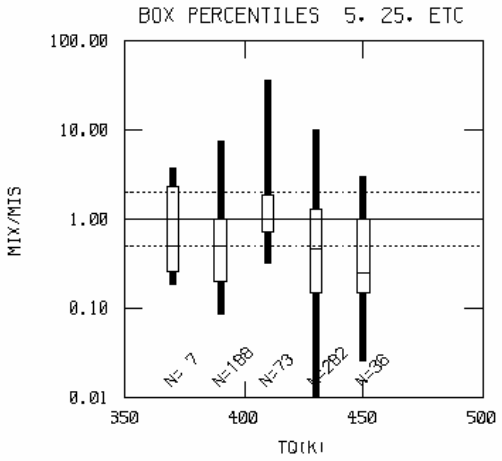

Figure 1. Residual analysis for PL3 concentration reconstruction approach and varying emission temperature on the data set with $Q \geq 2$.

CONCLUSIONS

A first prototype of a fully Lagrangian photochemical particle model, PLPM, has been presented and discussed. Peculiar features of this model are the high resolution, being linked to the CALMET meteorological model and the grid independence, obtained by using the kernel density estimator.

The main results of the model validation show as PLPM achieves the performances level of some well known regulatory models suggested by the EPA. Also this validation exercise, has shown as density reconstruction methods based on the kernel approach are preferable to the usual box counting method. A residual analysis has suggested next steps in the model development, as the implementation of complete description of the gradual plume rise.

[2] Residuals are defined as the ratios C_p/C_o of predicted and observed values.

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