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Proceedings of the 5th International Conference on Inverse Problems in Engineering: Theory and Practice, Cambridge, UK, 11-15th July 2005

ESTIMATION OF EMISSION RATE FROM POLLUTANT SOURCES

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Abstract - Pollutant dispersion problem in the atmosphere is a hard issue, because the atmosphere presents several stability conditions, implying different parameterisations for the atmospheric turbulence. In some practical situations, such as accident, it is important to have available a tool capable of identifying the pollutant source emission strength on the basis of measured concentrations only. Therefore, the goal here is to identify the strength of pollutant sources knowing the ground level concentrations, where two different sources, a linear and an areal, are simultaneously emitting. A Langrangian particle model, based on the Langevin equation, is used to simulate the forward problem, while the inverse problem is formulated as an optimization problem. Two optimizers are employed to obtain inverse solutions: quasi-Newton method, and simulated annealing. They are deterministic and stochastic approaches, respectively. Under certain conditions, the deterministic approach is unable to find a good inverse solution.

1. INTRODUCTION

The pollutant dispersion in the atmosphere is a topic with high interest nowadays. However, inside of the Planetary Boundary Layer (PBL) the turbulence is a permanent feature [2, 11], making this problem in a very hard one. Models for air monitoring are not only important for describing the pollutant impact over urban or rural areas, or for a urban planning consideration (these are examples where the forward problem is important considering several scenarios), but other issues are also relevant, such as the pollutant source strength estimation, CO_2 diurnal cycle, and total ozone in the atmosphere. The last three issues are examples of inverse problems in atmospheric pollution. Inverse problems can be usually solved by an *implicit technique*, where the inverse problem is formulated as a constrained nonlinear optimization problem: the forward problem is iteratively solved for successive approximations of the unknown parameters. The associated forward problem is the solution of the dispersion model.

Two different approaches can be used to study the atmospheric dispersion: Eulerian and Lagrangian. The difference between them is related to the reference system. While in the Eulerian reference system is fixed, the Lagrangian reference system follows the average atmospheric motion. Both approaches are target of intense research today. However, each one has advantages and disadvantages to use. One main advantage considering Lagrangian models is good performance for simulating complex terrain or emission source of arbitrary size and geometry without limitation and numerical problems of the Eulerian models. However, these models have a greater computational cost than Eulerian models. Here, the Lagrangian particle model is used as the forward model.

The pollutant source estimation is carried out using an implicit strategy. Two procedures are employed for solving the optimization problem: quasi-Newton method, and simulated annealing: deterministic and stochastic schemes, respectively.

2. DIRECT MODEL

The Lagrangian particle model LAMBDA was developed to study the transport process and pollutants diffusion, starting from the Brownian random walk modeling [7–8]. In the LAMBDA code, full-uncoupled particle movements are assumed. Therefore, each particle trajectory can be described by the generalized three-dimensional form of the Langevin equation for velocity [14]:

$$du_i = a_i(\mathbf{x},\mathbf{u},t)dt + b_{ij}(\mathbf{x},\mathbf{u},t)dW_j(t) \tag{1a}$$

and

$$dx = (\mathbf{U}+\mathbf{u})dt\,, \tag{1b}$$

where $i, j = 1,2,3$, and x is the displacement vector, $\mathbf{U}$ is the mean wind velocity vector, $\mathbf{u}$ is the Lagrangian velocity vector, $a_i(\mathbf{x},\mathbf{u},t)dt$ is a deterministic term and $b_{ij}(\mathbf{x},\mathbf{u},t)dW_j(t)$ is a stochastic term and the quantity $dW_j(t)$ is the incremental Wiener process.

The deterministic (drift) coefficient $a_i(\mathbf{x},\mathbf{u},t)$ is computed using a particular solution of the Fokker-Planck equation associated to the Langevin equation. The diffusion coefficient $b_{ij}(\mathbf{x},\mathbf{u},t)$ is obtained from the Lagrangian structure function in the inertial subrange, $(t_K << \Delta t << t_L)$, where t_K is the Kolmogorov time scale and t_L is the Lagrangian decorrelation time scale.

In order to simulate the turbulent diffusion employing the Langevin equation (1a) the coefficients $a_i(\mathbf{x},\mathbf{u},t)$ and $b_{ij}(\mathbf{x},\mathbf{u},t)$ can be written as functions of the turbulent velocity variances s_i^2 and decorrelation time scales (τ_{Li}). Accounting for the current knowledge of the PBL structure and characteristics, Degrazia *et al*. [4] have derived parameterizations for s_i^2 and t_{Li}. This parameterization is obtained from the Taylor statistical diffusion theory; observed spectral properties, where a linear combination of the two turbulent forcing mechanisms (shear + buoyancy) is assumed, and the value of the wavelength associated to the energy containing eddies. This parameterization give continuous values for the PBL at all elevations and all stability conditions from unstable to stable.

LAMBDA uses a large number of fictitious particles to simulate the atmospheric diffusion. Each particle can be marked for a mass. The spatial distribution of particles in the computational domain allows calculating the three-dimensional concentration field, $C(x, y, z, t)$, through the calculation of how many of them lie in a cell or imaginary volume centred at x, y, z, as follows:

$$C(x, y, z, t) = m_p \frac{N_v}{V_c} \tag{2}$$

where m_p is the mass of the each particle, N_v is the particle number in the cell and V_c is the cell volume. The mass of the each particle is determined from :

$$m_p = \frac{Q(t)\,\Delta t}{N_p}, \tag{3}$$

where $Q(t)$ is the emission rate, Δt is the time step and N_p is the particle number emitted per time step.

3. INVERSE MODEL

In order to set up the inverse analysis, it is assumed that the concentration obtained from the mathematical model is given by $C^{Mod}(\vec{\mathbf{r}}, \mathbf{Q})$, where $\boldsymbol{Q} = [Q_1(t),...,Q_n(t)]^T$ is the vector emission rate and $Q_n(t)$ represents the emission rate of the nth source and $C^{Exp}(\vec{\mathbf{r}})$ are data from concentration measurements. The solution of inverse problem is a function $\mathbf{Q}$ that minimizes the following objective function:

$$J(\mathbf{Q}) = \left\| C^{Exp}(\vec{\mathbf{r}}) - C^{Mod}(\vec{\mathbf{r}},\mathbf{Q}) \right\|_2^2 + a \left\| \mathbf{Q} \right\|_2^2 \tag{4}$$

where $\left\| \mathbf{Q} \right\|_2^2$ is the zeroth order Tikhonov regularization scheme, and a a positive parameter, called regularization parameter.

3.1 Optimization Algorithm

The optimization problem is iteratively solved by two different algorithms: the quasi-newtonian optimizer routine E04UCF, from the NAG Fortran Library [5] (deterministic method); and Simulated Annealing [11] (stochastic method).

Quasi Newtonian
The minimization of the objective function (4), subject to simple bounds on $\mathbf{Q}$, is solved using a first-order optimization algorithm from the NAG Fortran library. This routine is designed to minimize an arbitrary smooth function subject to constraints (simple bounds, linear and nonlinear constraints), using a sequential programming method. For the n^{th} iteration, the calculation proceeds as follows.

1. Solve the direct problem for $\mathbf{Q}^n$ and compute the objective function $J(\mathbf{Q})$.

2. Compute by finite differences the gradient $\nabla J(\mathbf{Q})$.

3. Compute a positive-definite quasi-Newton approximation to the Hessian $\mathbf{H}^n$:

$$\mathbf{H}^n = \mathbf{H}^{n-1} + \frac{b^n (b^n)^T}{(b^n)^T u^n} - \frac{\mathbf{H}^{n-1} u^n (u^n)^T \mathbf{H}^{n-1}}{(u^n)^T \mathbf{H}^{n-1} u^n}$$

where

$$b^n = \mathbf{Q}^n - \mathbf{Q}^{n-1}$$
$$u^n = \nabla J(\mathbf{Q}^n) - \nabla J(\mathbf{Q}^{n-1}).$$

4. Compute the search direction $\mathbf{d}^n$ as a solution of the following quadratic programming subproblem:
Minimize

$$\left[\nabla J(\mathbf{Q}^n)\right]^T \mathbf{d}^n + \frac{1}{2}(\mathbf{d}^n)^T (\mathbf{H}^n)\mathbf{d}^n,$$

subjected to: $l_q - p_q^n \le d_q \le u^n - p_q^n$.

5. Set, $\mathbf{Q}^{n+1} = \mathbf{Q}^n + b^n \mathbf{d}^n$ where the step length b^n minimizes $J(\mathbf{Q}^n + b^n \mathbf{d}^n)$.

6. Test the convergence; stop or return to Step 1.

Simulated Annealing
This method is borrowed from materials science, where it is well known to produce a solid in a low energy state (such as a perfect crystal), you first heat the system to a high temperature, and then slowly cool it. At any given temperature, the probability that a change in the structure with energy change ΔE will occur is $\min\left(e^{-\Delta E / kT}\right)$, where k is Boltzmann's constant and T is the temperature of the system in degrees Kelvin. When the temperature is high, the system can change radically and many changes that do not lower the energy level are allowed. As the temperature decreases, fewer and fewer 'bad' changes are permitted, until finally a fairly optimal state is achieved. For a general optimization problem, an initial system is chosen and then iteratively optimized. During each pass, a generator of random changes the configuration of parameters $\mathbf{Q}$. Solve the direct problem for $\mathbf{Q}$. Compute the objective function $J(\mathbf{Q})$ and compute by finite differences the gradient $\nabla J(\mathbf{Q})$. The new system is accepted with probability:

$$P = \begin{cases} 1 & \text{if } \nabla J(\mathbf{Q}) < 0 \\ e^{-\Delta J(\mathbf{Q})/T} & \text{if } \nabla J(\mathbf{Q}) \ge 0 \end{cases}$$

where the value of T is gradually decreased according to an 'annealing schedule' as more optimizations are applied, thus causing the system to gradually accept fewer and fewer 'bad' changes.

4. NUMERICAL EXPERIMENT WITH TWO POLLUTANT SOURCES
For testing the inverse analysis for the pollutant emission rate estimation, a computational scenario is displayed: two pollutant source are present in the physical domain. The physical domain consists of a box with 5 Km × 5 Km of a flat terrain, with 1120 m of height. Inside this domain, an industrial area of size 500 m × 500 m is placed, releasing pollutant at 20 m height, additionally, a line source with aspect ratio of 40m × 5000 m in the y direction, representing a high way, emitting pollutant at level 20m. It is also considered a number of samplers

spread in the domain, measuring the pollutant concentration at ground level. Figure 1 represents our physical domain showing the samplers.

To simulate the concentration field, the mean meteorological data are used: $\overline{U}$, L, z_i (mean wind speed, Monin-Obukhov length, and Planetary boundary layer height) characteristic from Copenhagen experiment (19/10/1978 - (12:05h – 12:45h)) [9, 10] ($\overline{U}_{10m}$=2.6m/s, $\overline{U}_{120m}$=5.7m/s, L=-71m, z_i=1120m), and for the wind direction the angle q=180° was used. Wind standard deviations (s_i^2) and the Lagrangian decorrelation time scales (t_{L_i}) were calculated according to the turbulence parameterisation scheme suggested by Degrazia *et al*. [4].

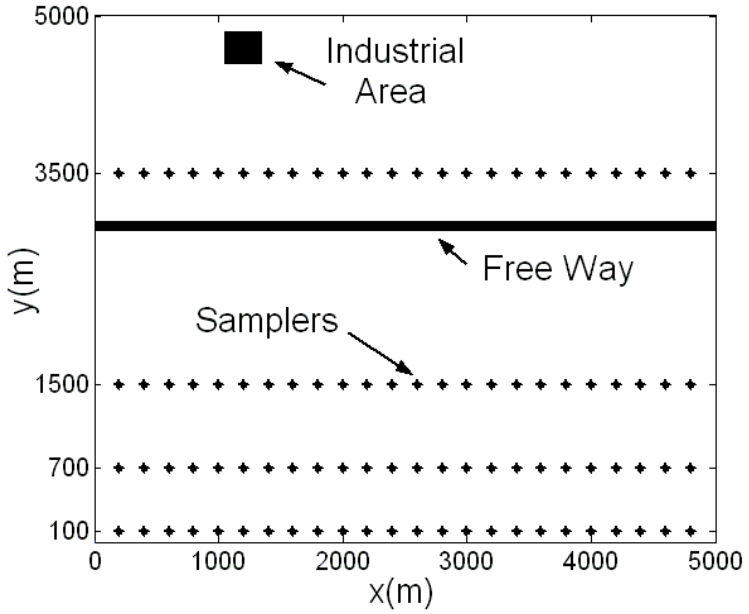

Figure 1: Fictitious experiment site.

The LAMBDA domain was 5 Km × 5 Km in the horizontal, and the vertical domain was set equal to z_i. The time step was maintained constant ($\Delta t = 1s$). The PDF Gram-Charlier truncated to the third order was chosen [8]. One hundred particles were released at each time step during 2400 time steps for each source. The emission rate was 10 g/s industrial area and 50 g/s for free way.

To obtain the concentration field a gaussian noise (level at 10%) was added in the measured meteorological data, for each parameter: z_i, L, U and V. After the concentration computation, a 10% of gaussian noise was also added. This is used noise to simulate a real atmospheric turbulence. Besides, the meteorological sensors and concentration monitoring devices present an intrinsic measurement error. For a better estimation of the ground level concentration (GLC) in this non stationary case, the GLC is computed 10 times (each 2 min) during each 20 min period and then is taken an averaged for the GLC value. The mean concentration and the variance at each sampler are presented in Figure 2.

Figure 2 shows that the LAMBDA code represents a “well-possed problem”: continuous dependence of the solution on the data. However, the inverse solution is an example of a solution which varies considerably for a small variation of the data, in which, according to Hadamard – see reference [1] – is not really a solution in the physical sense.

To test the inverse problem methodology, looking for a minimum for the functional given by eqn. (4), two data set are used as experimental data:

Experiment-1: The mean concentration present in Figure 2.

Experiment-2: The solution that present major desagreement with the results of the direct model (LAMBDA simulation without noise in meteorological data).

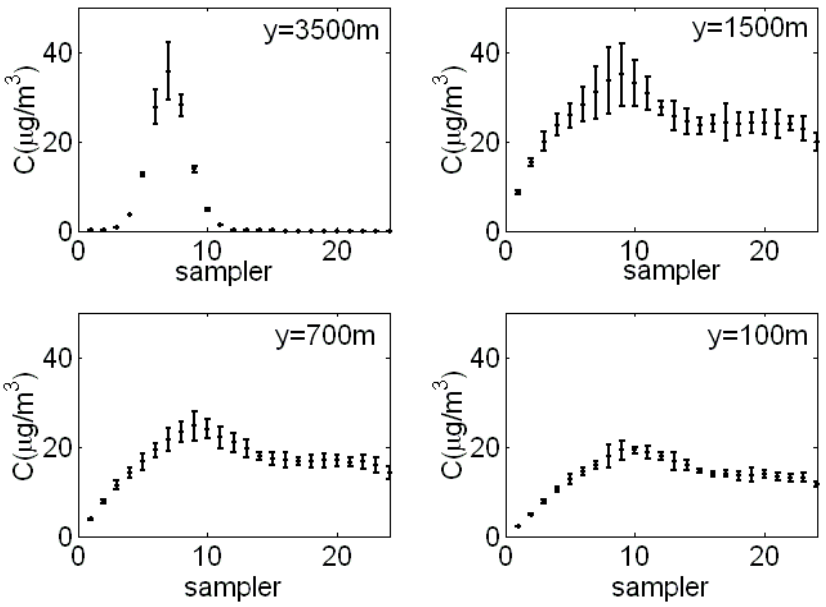

Figure 2: Mean experimental concentration with the variance for the fictitious experiment in each line of samplers.

5. RESULTS WITH THE DIRECT MODEL

A simulation using the meteorological data without noise was used as direct model. The model performance is avaliable by Fractional Standard Deviation (*FS*):

$$FS = 2\frac{s^{Exp} - s^{Mod}}{s^{Exp} + s^{Mod}}$$

where s is concentration standard deviations.

5.1 Comparison with Experiment-1

Figure 3 shows the scatter diagram between experimental and simulated concentration for the Experiment 1. Observing Figure 3 a very good agreement between experimental data and modeled data is noted, this is also shown by the Fractional Standard Deviation, *FS*=-0.011.

A LAMBDA simulation with the Degrazia's turbulence parameterization was tested with Copenhagen Experiment giving good results, and, in that case, the *FS* was *FS*=-0.056 [3]. This comparison shows that our fictitious experiment (Experiment 1) probably does not have a real correspondence with a nature phenomenom.

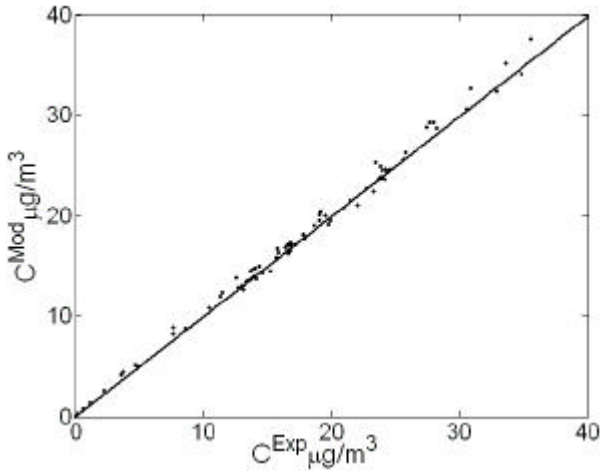

Figure 3. Scatter diagram between experimental and simulated samplers ground-level concentrations for Experiment 1.

5.2 Comparison with Experiment-2

The numerical experiment was carried out with the data from Experiment-2. Figure 4 shows the scatter diagram between experimental and simulated concentration. Clearly, the simulation produces an overestimation of the concentration, in this case the *FS*=-0.072. This *FS* value is greater than that presented by Carvalho et al. [3]. The conclusion is: this second data set emulated better the usually measurements.

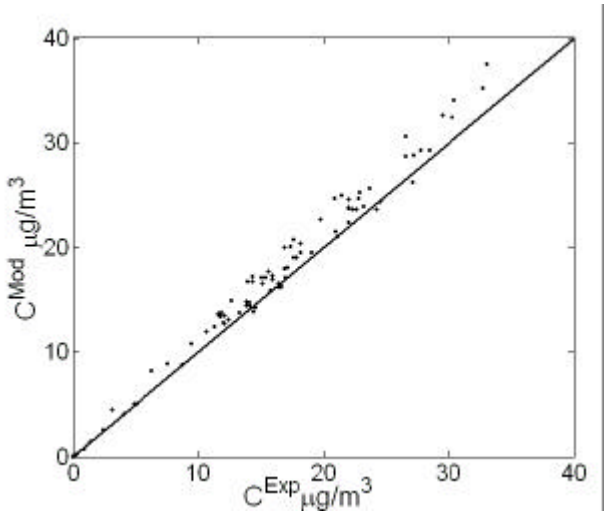

Figure 4. Scatter diagram between experimental and simulated samplers ground-level concentrations for Experiment-2.

6. RESULTS FOR THE EMISSION RATES ESTIMATION

For estimation of the emission rates two differents optimizators are used: Simulated Annealing, and a quasi–Newtonian. Tables 1 and 2 present the emission rates estimated by an inverse problem metodology for Experiment-1 and Experiment-2, respectively. For all estimation, the initial emission rate was assumed Q=0.1 g/s.

Inversion solution using Simulated Annealing produced betters, for both experiments results (see Tables 1 and 2), and the quasi-Newtonian optimizer is unable to find an aceptable answer for Experiment-2. Table 3 presents the number of interations employed for each optimizator. As pointed out by Roberti *et al*. [12], Simulated Annealing is more expensive from computational point of view than the Quasi-Newtonian scheme. However, the latter technique did not procuce good inverse solutions in the presence of higher noise level.

Table 1. Emission rate estimation for the Experiment-1.

	True Value	Quasi - Newton	Simulated Annealing
	Q (g/s)	Q (g/s)	Q (g/s)
Industrial area	10	8.7	9.4
Free way	50	48.7	50.7

Table 2. Emission rate estimation for the Experiment-2.

	True Value	Quasi-Newton	Simulated Annealing
	Q (g/s)	Q (g/s)	Q (g/s)
Industrial area	10	17.5	9.2
Free way	50	17.5	47.1

Table 3. Number of interations spent for the optimizators.

	Quasi-Newton	Simulated Annealing
Experiment 1	65	1575
Experiment 2	31	391

7. CONCLUSION

The implicit methodology was effective for estimating the emission rate from the pollutant sources. A Lagrangian model was used for computing the pollutant concentration field. Two different strategies for solving the optimization problem were considered: a deterministic approach (quasi-Newtonian), and a stochastic one (Simulated Annealing). Although the Simulated Annealing is more expensive than quasi-Newton method - in terms of CPU time - it presents better results for the emission rate estimation.

Ackowledgements

The authors would like to thank to the Brazilian and Italian financial support agencies. The first author acknowledges CNPq-Brasil (Proc. 20.0046/04-7) for the 6 months period of the doctorate research in the ISAC-CNR (Turin, Italy) under supervision of the Dr. D. Anfossi.

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