International Environmental Modelling and Software Society (iEMSs)
2012 International Congress on Environmental Modelling and Software
Managing Resources of a Limited Planet, Sixth Biennial Meeting, Leipzig, Germany
R. Seppelt, A.A. Voinov, S. Lange, D. Bankamp (Eds.)
http://www.iemss.org/society/index.php/iemss-2012-proceedings

A new Lagrangian-probabilistic approach for the study of pollutants diffusion: analytical modeling and experimental validation using the Copenhagen data set

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Abstract: In this paper a Lagrangian-probabilistic model for the study of pollutants diffusion in the atmosphere is presented. As opposed to the models available in literature, where the transition Probability Density Function (PDF) of particles is Gaussian, in this new theoretical approach the transition PDF is bilateral exponential.

The model presented in this work, being based on an original formulation of the transition probability density, allows to represent and analytically calculate the mean concentration field produced by a time-varying point source. Comparing well results of the 1-D Laplace bilateral model (LBM) to those of the Gaussian approach, the three-dimensional model associated with PDF_{LB} has also been developed. The 3-D modeling allows the representation of more complex scenarios, remaining the PDF_{LB} function analytically flexible and easy to integrate. These peculiarities always lead to analytical solutions of the mean concentration field and allow to represent complex and time-varying scenarios, with low calculation requirements.

The model based on Laplace bilateral PDF has been then validated using a three dimensional approach and considering actual meteorological and emissions scenarios. The many data collected in experimental campaign of Copenhagen, conducted in years 1978-1979, have been compared with model results, highlighting a good agreement in almost all test conditions.

The good results obtained and presented in this study have opened new interesting perspectives for the approach proposed, such as the study of specific scenarios where pollutant emissions are characterized by instantaneous releases. These are typical situations of industrial equipment breakdown that can originate acute pollution scenarios, often characterized by a short transient phenomena. The possibility of using the model in these situations may make it suitable as an analytical tool of high value and flexibility for numerous engineering applications.

***Keywords**:* Air pollutant diffusion; Dispersion modelling; Lagrangian model; Gaussian plume.

1 INTRODUCTION

In recent decades, the growing attention that specialized literature has addressed to the study of pollutant diffusion in the atmosphere has accompanied the continuous and rapidly increasing evolution of environmental laws in the sector, as well as the growing public awareness towards environmental issues. In this context, the availability of effective simulation models is a valuable tool for the solution of complex problems in both design and control phases.

The wide and heterogeneous set of scenarios occurring in environmental impact assessments has pushed the development of several pollutant diffusion models, that require a rigorous classification of different theoretical approaches, as shown by legislation and related technical reports. The modeling in this field has therefore a fundamental role being an indispensable tool in design stage and also useful in check phase, if effectively employed to support measurement monitoring. Essential requirements of diffusion models include low calculation load, versatility in several application fields and adaptability to the largest number of possible scenarios.

The Laplace bilateral model (LBM), proposed by Cipollone and Foglia [2006], is a theoretical approach intermediate between the classic Gaussian model and the Lagrangian-probabilistic one, as discussed by Seinfeld [1985] and Zannetti [1994]. The need for further model development arises from the necessity of being able to represent, with sufficient accuracy but with the simplicity of the Gaussian model, relatively complex scenarios, without suffering the limitations arising from many simplification hypotheses and from high calculation loads associated to numerical solutions of the Lagrangian-probabilistic model, as shown by Indumati [2009] and Kesarkar [2007].

In this work a bilateral exponential, rather than a normal- or Gaussian-type, is adopted for the transition probability density function (PDF) of the Lagrangian-probabilistic approach. It allows the representation of a large number of scenarios in a relatively simple and effective manner, with the advantage of having an analytical expression for the mean concentration field of polluting species.

The proposed model has been validated against the Copenhagen data set; results show a good agreement in estimating both the position of maximum pollutant concentration and the values of mean concentration field.

2 LAPLACE BILATERAL MODEL

A Lagrangian-probabilistic model for the study of inert pollutants diffusion in the atmosphere is presented in this paper. As it is well known, the Lagrangian-probabilistic model focuses on the choice of the transition probability density function (PDF) of the pollutant particles. As opposed to the numerous models available in the literature, that take a Gaussian transition PDF, the model presented in this paper takes a transition PDF of bilateral exponential type, also known as Laplace bilateral (LBM).

The $\mathrm{PDF_{LB,x}}$ in a 1-D approach can be expressed as

$$\mathrm{PDF_{LB,x}} = \frac{\alpha_\mathrm{x}}{2} \mathrm{e}^{-\alpha_\mathrm{x}|\mathrm{x}-\bar{\mathrm{x}}(\mathrm{t}-\mathrm{t}')|} \tag{1}$$

where the variable x indicates the spatial position of the particle at time t, $\bar{x}$ the average trajectory of the particle in the time interval (t, t'), and α_x the shaped factor of the $PDF_{LB,x}$, closely related to the turbulent flow field in which the particle diffuses. In this way the bilateral function correctly represents the typical conditions of isotropic turbulence.

In a first theoretical approach the shaped factor α_x can be determined by imposing that the second order moment of PDF equals the variance of the Gaussian PDF

$$\sigma_{LB} = \sigma_{Gauss\,plume} \tag{2}$$

that can be evaluated through

$$\sigma_{LB}{}^{2} = \frac{2}{\alpha^{2}} \tag{3}$$

Extensive literature concerning the Gaussian model, as discussed by Seinfeld [1985], reports experimental correlations on how turbulent dispersions are influenced by the meteorological parameters and the macroscopic flow field.

The main advantage of the bilateral exponential PDF is the ease of integration, a peculiarity of this versatile function. As it is known, the average concentration field of a pollutant $\langle \mathrm{C}(\mathrm{x},\mathrm{t})\rangle$ is strictly dependent on the transition PDF and on the emission source distribution $\mathrm{S}(\mathrm{x},\mathrm{t}')$

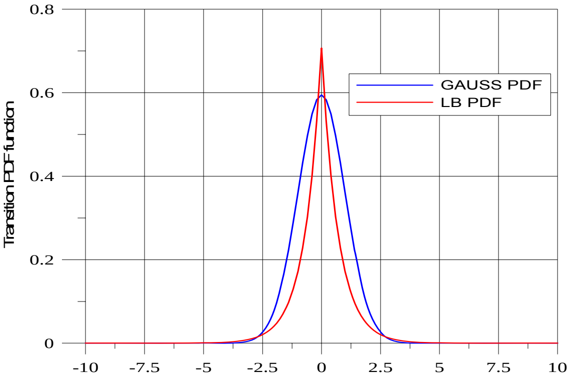

Figure 1. Comparison between the normal Gaussian PDF and the bilateral Laplace PDF.

$$\langle C(x,t)\rangle = \int_{\mathbb{R}} \int_{t} S(x,t')PDF_{LB,x}\,dx\,dt' \tag{4}$$

For a point source, the ease of integrating the transition $\mathrm{PDF_{LB}}$ leads to a concentration field $\langle \mathrm{C}(\mathrm{x},\mathrm{t})\rangle$ expressed in analytical form in most cases of practical interest. We can conclude, therefore, that MBL is of extreme engineering interest. Hence, LBM is considered a versatile and light tool for both design and control phases. A preliminary investigation has been carried out for estimating capabilities of the model, by comparing LBM and Gaussian plume models in a 1-D spatial domain. Results obtained by Cipollone and Foglia [2006] were good and pushed to extend the model the 3-D spatial domain, able to represent more effectively real scenarios.

In a 3-D domain $\mathrm{PDF_{LB}}$ takes the form

$$PDF_{LB} = \prod_{i=x,y,z} PDF_{LB,i} = \prod_{i=x,y,z} \frac{\alpha_i}{2} e^{-\alpha_i|i-\bar{\imath}(t-t')|} \qquad with\ \ i = x,y,z \tag{5}$$

where $\mathrm{PDF_{LB,i}}$ components in the three spatial directions are mutually independent. In this case the analytical expression of the mean concentration field, space- and time-variant, if it is generated by a point source with constant mass flow rate, $\mathrm{S}(\mathrm{x},\mathrm{y},\mathrm{z},\mathrm{t}) = \mathrm{q}$, can be always obtained analytically from Eq. (4)

$$\langle C(x,y,z,t)\rangle = \begin{cases} q\dfrac{\alpha_y\alpha_z}{8\bar{u}} e^{-\alpha_y|y|} e^{-\alpha_z|z|} e^{-\alpha_x|x|}\left[e^{\alpha_x\bar{u}t} - 1.0\right], & t < \dfrac{x}{\bar{u}} \\ q\dfrac{\alpha_y\alpha_z}{8\bar{u}} e^{-\alpha_y|y|} e^{-\alpha_z|z|}\left[2.0 - e^{-\alpha_x x} - e^{\alpha_x(x-\bar{u}t)}\right], & t \geq \dfrac{x}{\bar{u}} \end{cases} \tag{6}$$

where $\bar{\mathrm{u}}$ is the mean speed of the macroscopic motion field and t is the instant in which the pollutant concentration field is assessed.

3 EXPERIMENTAL VALIDATION

3.1 Copenhagen data set

The experimental validation of the proposed model has been carried out on the basis of data given by Olesen and Lyck [2005], referring to the campaign of Copenhagen in years 1978-1979. This measurement campaign was conducted by releasing constant flow rates of sulphur hexafluoride (SF6) from a stack at 115 m.

The concentration field was measured at about 2-3 m from the ground, along three arcs (Arc1, Arc2, Arc3) distant about 2000, 4000 and 6000 m from the pollutant source, as shown in Figure 2.

For the calculation of the mean concentration field, three consecutive measures every twenty minutes were made. In the same spatial domain, measures were also conducted by Gryning [1981] on the more significant meteoclimatic parameters; these measured variables, considered as input data for the simulation of different scenarios with LBM model, are summarized in Table 1.

Table 1. Meteorological conditions and data during Copenhagen experiments.

$DATE$	$NCLD$ [%]	$u@10\ m$ [m/s]	T_{env} $[K]$	V_f [m/s]	Q [kg/h]	H_{inv}	Pasquill class	Data set
09/20/1978	70	2.1	285.5	2.5	11.5	1980	C	DS1
09/26/1978	60	4.9	288.7	2.5	11.5	1920	C	DS2
10/19/1978	50	2.3	285.6	2.5	11.5	1120	C	DS3
11/03/1978	40	2.5	284.8	2.5	8.3	390	C	DS4
11/09/1978	70	3.1	285.5	2.5	11.5	820	C	DS5
04/30/1979	50	7.2	280.1	2.5	11.2	1300	D	DS6
06/27/1979	60	4.1	292.2	2.5	8.6	1850	B	DS7
07/06/1979	50	4.2	293.8	2.5	10.8	810	B	DS8
07/19/1979	60	5.1	289.8	2.5	11.9	2090	D	DS9

NCLD: cloud cover; u @ 10 m: wind velocity measured at 10 m; T_{env}: environmental temperature; V_f : exit gas velocity; Q: mass flowrate of pollutant; H_{inv}: inversion height

The data collected in the experimental campaign of Copenhagen were used largely for the validation of pollutant diffusion in atmosphere, as demonstrated by numerous authors in workshops and conferences, including Mangia et al. [2004], Tirabassi and Rizza [1997], and Costa et al. [2006].

3.2 Results and discussion

The Copenhagen data set is used to assess the ability of LBM model to predict gaseous pollutant concentrations along arcs. First a statistical analysis has been carried out; in Table 2 values of some significant statistical variables - the fractional bias FB, the geometrical mean bias MG, the normalised mean square error NMSE and the factor of two Fa2 - are reported. They highlight a good agreement between the predicted model values and the experimental data.

Table 2. Statistical evaluation of model results in terms of downwind concentration Cmax and ratio C/Q.

C_{max}

FB	MG	NMSE	Fa2
0.33	1.64	1.01	1.27

C/Q

FB	MG	NMSE	Fa2
0.34	1.65	1.01	1.29

Considering all data available from the experimental campaign conducted by Gryning et al. [2002], Figures 3 and 4 show the scatter plot of measured and predicted concentration in absolute values and referred to emission flow-rate, being both terms significant for the validation of LBM model.

As shown in figures 5-10, the agreement between predicted and measured values is generally good; for some data set (DS1, DS6) the model has less ability to reproduce experimental data, while the agreement is very satisfactory for other data set (DS3, DS5).

For DS1 and DS6 data set, where the agreement between predicted and measured data is more slack, meteorological conditions tend to neutrality. In this case, downwind and crosswind profiles are represented in correspondence of two measurement arcs.

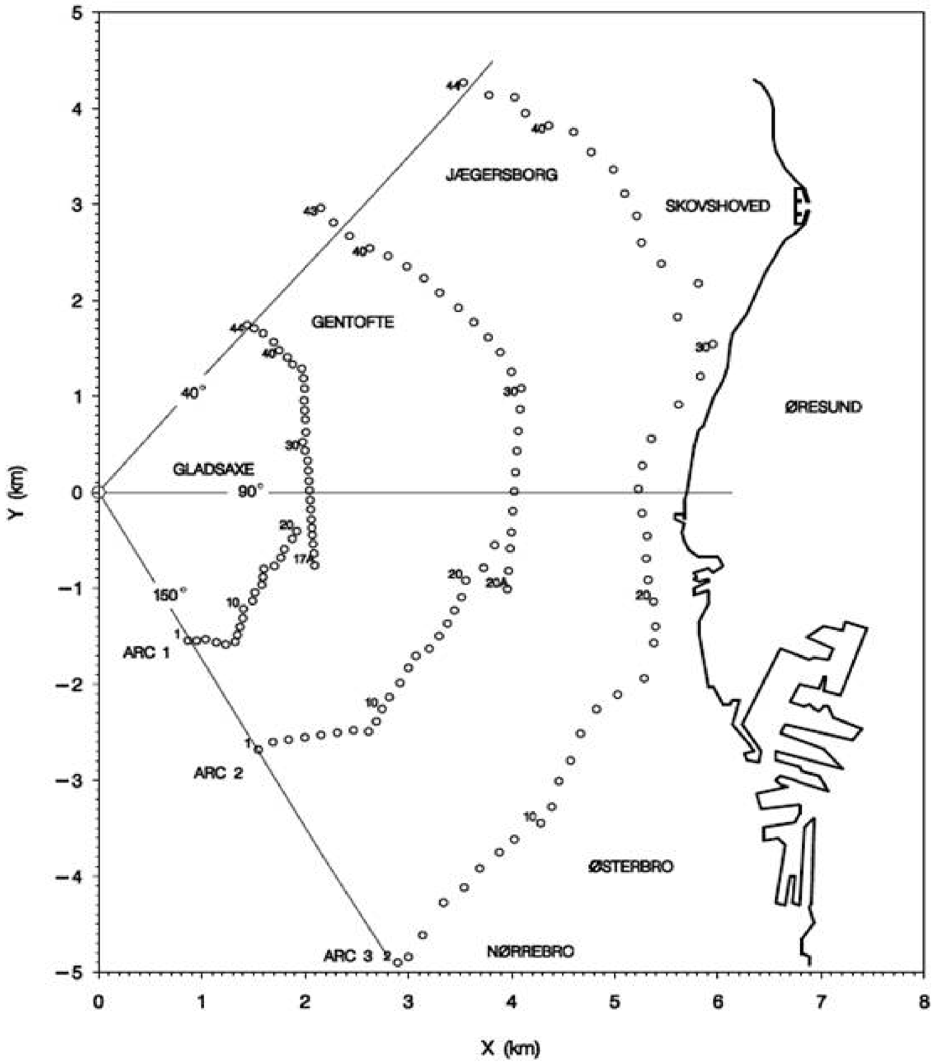

Figure 2. Measurement arc distribution of Copenhagen experiments, [12].

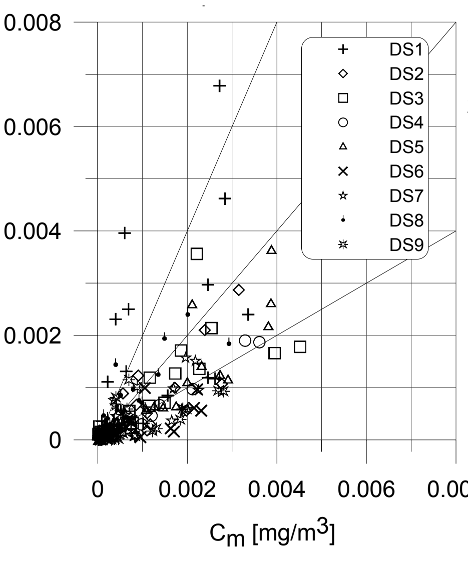

Figure 3. Scatter plot of measured and predicted concentration.

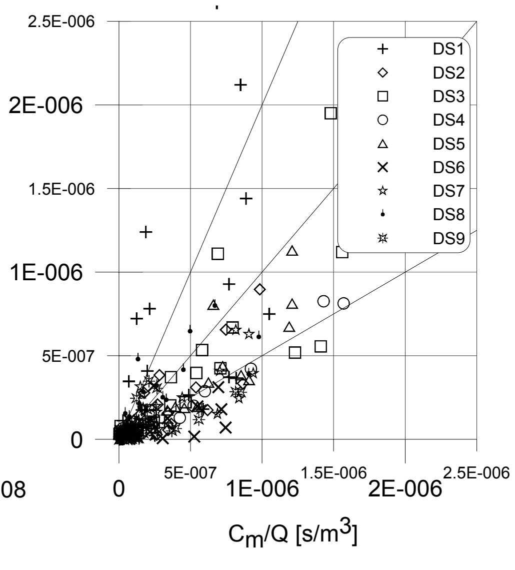

Figure 4. Scatter plot of concentration referred to emission flow-rate.

For DS1 data set, as shown in Figure 5, the model gives a good estimation of the position of maximum pollutant concentration on Arc1, the nearest to the source of pollution, being less satisfactory the correspondent estimated maximum value.

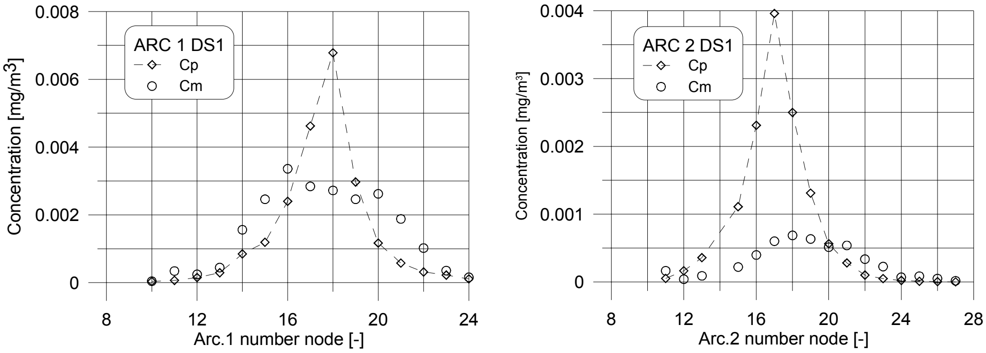

Figure 5. Concentration for DS1 data set on Arc1 and Arc2.

On Arc2, there are appreciable differences in estimating both the position of the maximum concentration and its value; in these cases LBM model overestimates the concentration field. For DS6 data set, as shown in Figure 6, the position of maximum pollutant concentration is estimated precisely in both the arcs, while the model gives a less accurate prediction of the concentration field along Arc1 underestimating the measured values.

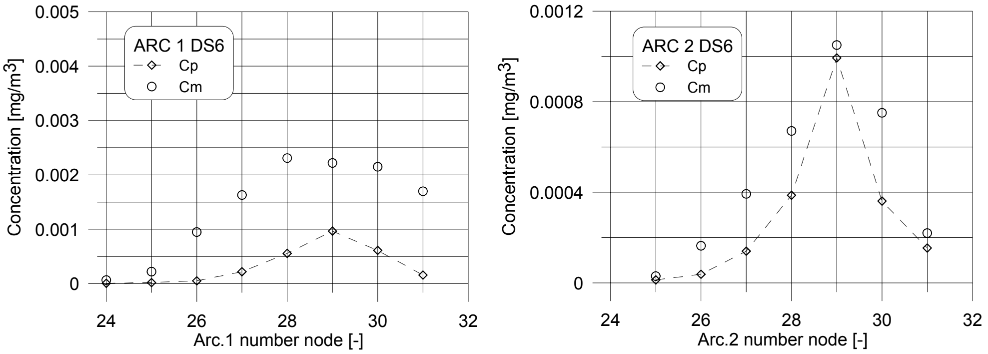

Figure 6. Concentration for DS6 data set on Arc1 and Arc2.

It is noteworthy to observe that, as shown in Figure 7, inaccuracies are also reflected in downwind profiles. For both data set, the LBM model anticipates the position of maximum pollutant concentration and underestimates the concentration field in neutrality meteorological conditions.

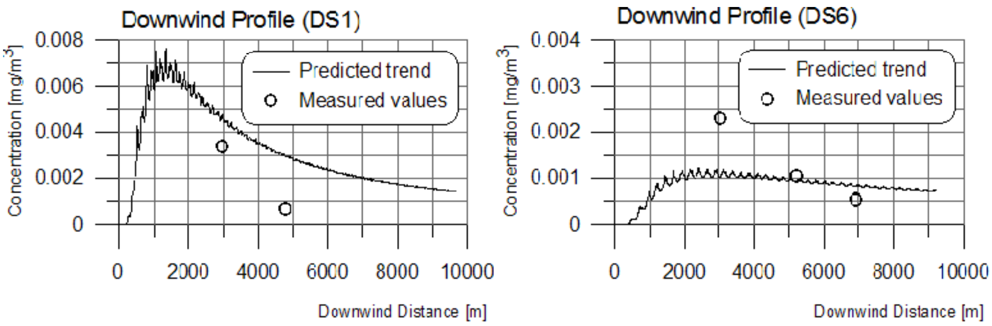

Figure 7. Downwind concentration for DS1 and DS6 data set.

The proposed LBM model gives more accurate estimations for DS3 and DS5 data set; in fact, as shown from crosswind profiles in Figures 8 and 9, model results match well both the maximum value and the mean concentration field. In Figure 10, downwind profiles are reported for these two data set, showing that predicted values approximate close the experimental data.

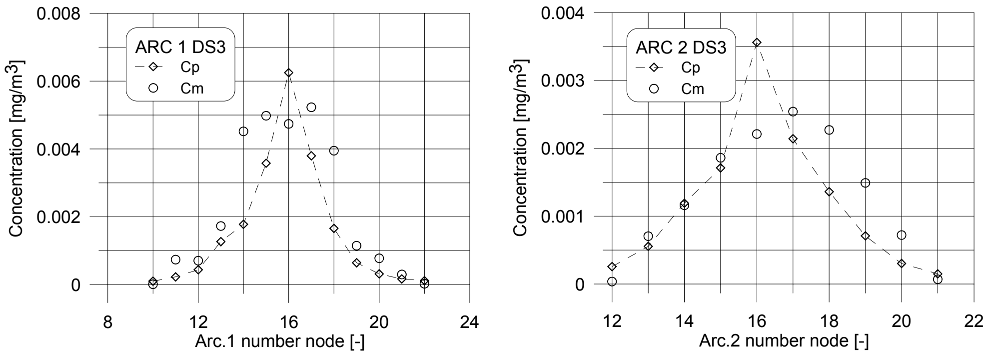

Figure 8. Concentration for DS3 data set on Arc1 and Arc2.

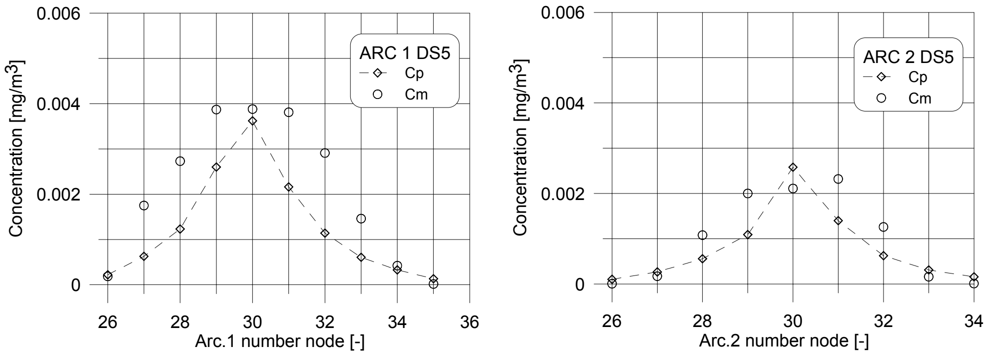

Figure 9. Concentration for DS5 data set on Arc1 and Arc2.

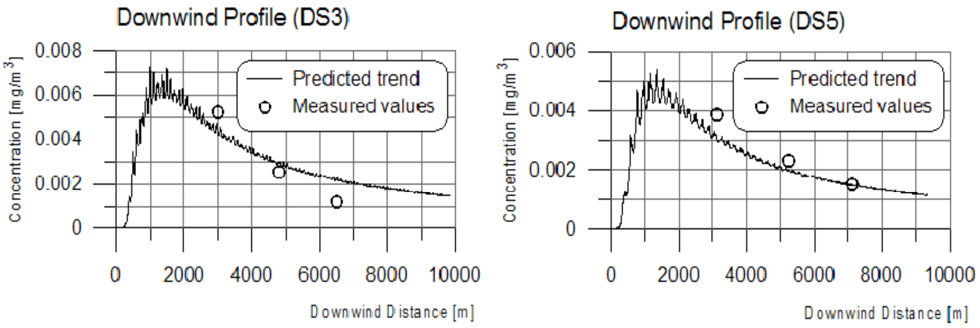

Figure 10. Downwind concentration for DS3 and DS5 data set.

4 CONCLUSIONS

In this work the development of an original one- and three-dimensional theoretical approach of pollutant diffusion in atmosphere has been presented. The model has been validated experimentally using the Copenhagen data set, a measurement campaign conducted in years 1978-1979.

The results obtained from the model are generally in a good agreement with measured values. Model validation has shown that the proposed approach is able to characterize scenarios more complex than those simulated in this paper; it can be also used for studying specific scenarios where pollutant emissions are characterized by instantaneous releases. However, it is necessary to identify experimental correlations that couple well with the structure of Laplace bilateral when it is used in meteo-climatic scenarios that tend to the neutrality or to the strong stability.

The use of Laplace bilateral approach as transition PDF in the Lagrangian-probabilistic model allows for limiting calculation requirements and expressing the concentration field through an analytical formulation, as a function of space and time variables. This characteristic make particularly interesting this theoretical approach. It allows a simple characterization also for quite complex scenarios, differently from the classical approach that usually requires a higher calculation load for the numerical solution of the concentration field.

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NOMENCLATURE

$\langle C(x,t)\rangle$	Concentration field	σ	PDF variance
H_{inv}	Inversion height	ΔH	Plume rise
q, Q	Flow rate	Fa2	Factor of two
$S(x,t')$	Emission source distribution	FB	Fractional Bias
t	Time variable	LBM	Laplace bilateral model
$\bar{u}$	Mean wind velocity	MG	Geometrical mean bias
x	Spatial position	NCLD	Cloud cover
$\bar{x}$	Average trajectory	NMSE	Normalized mean square error
T_{env}	Environmental temperature	PDF	The transition probability density function
V_f	Exit gas velocity		
α_x	Shape factor	SF6	Sulphur hexafluoride