AN ANALYTICAL AIR POLLUTION MODEL FOR BUOYANT PLUMES

Camila Pinto da Costa, camila@mecanica.ufrgs.br
Universidade Federal do Rio Grande do Sul - PROMEC - Porto Alegre, Brasil

Tiziano Tirabassi, t.tirabassi@isac.cnr.it
Institute ISAC of CNR , Bologna, Italy

Davidson Martins Moreira, davidson@mecanica.ufrgs.br
Universidade Federal de Pelotas, UNIPAMPA, Bagé, Brasil

Marco Tullio Vilhena, vilhena@mat.ufrgs.br
Universidade Federal do Rio Grande do Sul - PROMEC - Porto Alegre, Brasil

Abstract. *In this work we present the semi-analytical solution of the three-dimensional advection-diffusion equation using the GIADMT (Generalized Integral Advection Diffusion Multilayer Technique) method incorporating the plume rise effect (warm sources) in the model. A correct estimation of buoyant plume rise is one of the basic requirements for operative models applied for the determination of ground-level concentrations of airborne pollutant emitted by industrial stacks. To investigate the performances of the model, we will report numerical simulations of the ground-level concentrations compared with an experimental data set*

Keywords*: GIADMT, plume rise, advection-diffusion equation, analytical solution, pollutant dispersion*

1. INTRODUCTION

Eulerian approach for modelling the statistical properties of the concentrations of contaminants in a turbulent flow as the Planetary Boundary Layer (PBL) is widely used in the field of air pollution studies. Despite well known limits, the K-closure is largely used in several atmospheric conditions because it describes the diffusive transport in an Eulerian framework where almost all measurements are Eulerian in character, it produces results that agree with experimental data as well as any more complex model, and it is not computationally expensive as higher order closures are.

The advection-diffusion equation has been widely applied in operational atmospheric dispersion models to predict ground-level concentrations due to low and tall stacks emissions. In this work, we step forward presenting a solution for the three-dimensional advection-diffusion equation in order to simulate pollutant dispersion in atmosphere reporting a solution for the K-diffusion model assuming plume rise effect.

To accomplish this objective we solve the three-dimensional advection-diffusion equation by the GIADMT (Generalized Integral Advection-Diffusion Multilayer Technique) method. This method is a combination of the well known ADMM (Advection Diffusion Multilayer Methodl) and GILTT (Generalized Integral Transform Technique) methods. To more details about these approaches see the works of Vilhena et al. (1998), Moreira et al. (1999), Wortmann et al. (2005), Costa (2006) and Moreira et al. (2006).

The ADMM approach is based on the Laplace transform technique with numerical inversion considering the PBL as a multilayer system where in each layer the eddy diffusivity and wind are constants. The main feature of this method relies on the following steps: stepwise approximation of the eddy diffusivity and wind speed, the Laplace transform application to the advection-diffusion equation, semi-analytical solution of the set of linear ordinary equation resulting for the Laplace transform application and construction of the pollutant concentration by the Laplace transform inversion using the Gaussian quadrature scheme.

The GITT is a well-known hybrid method that had solved a wide class of direct and inverse problems mainly in the area of heat transfer and fluid mechanics (Cotta, 1993; Cotta and Mikhailov, 1997; Cheroto et al., 1999; Alves et al., 2002; Magno et al., 2002 and Cotta et al., 2003). The main steps of this method include the construction of the auxiliary Sturm-Liouville problem associated to the original problem, the determination of the integral transform technique in a series, using as basis the eigenfunction of the solved Sturm-Liouville problem, the replacement of this expansion in the original problem and taking moments. This procedure leads to a set of ordinary differential equation, which is classically solved by numerical methods.

In this work we step forward incorporating the plume rise effect (warm source) in the model using the approach proposed by Briggs (1975). A correct estimation of buoyant plume rise is one of the basic requirements for the determination of ground-level concentrations of airborne pollutant emitted by actual industrial stacks. This improvement turns out a more operative model. To investigate the influence of the plume rise effect, we report numerical simulations of the ground-level centerline concentrations compared with the observed concentrations measured during the Kinkaid experiment (Hanna and Paine, 1989).

To reach this goal, we outline the paper as follows: in section 2, we report the derivation of the GIADMT solution for the three-dimensional advection-diffusion equation. In section 3, the plume rise approach is presented. In section 4 the turbulent parameterisations assumed in this work are presented. The numerical results obtained by the GIADMT method are reported as well the comparison with experimental data are presented in section 5, and finally in section 6, the conclusions.

2. THE GIADMT SOLUTION

The advection-diffusion equation of air pollution in the atmosphere is essentially a statement of conservation of the suspended material, assuming that the pollutants are inert and have no additional sinks or sources downwind from the point source. The vertical (*w*) and lateral (*v*) components of the mean flow are assumed to be zero. The mean horizontal flow is incompressible and horizontally homogeneous. Then, for stationary conditions, we have:

$$u\frac{\partial c}{\partial x}=\frac{\partial}{\partial x}\left(K_x\frac{\partial c}{\partial x}\right)+\frac{\partial}{\partial y}\left(K_y\frac{\partial c}{\partial y}\right)+\frac{\partial}{\partial z}\left(K_z\frac{\partial c}{\partial z}\right) \tag{1}$$

for $0<z<z_i$, $0<y<L_y$ and $x>0$, where and z_i is the height of the PBL and L_y is faraway from the source, where c denotes the average concentration, K_x, K_y, K_z and u, v, w are the Cartesian components of eddy diffusivity and wind, respectively, and S is the source term. The *x*-axis of the Cartesian coordinate system is aligned in the direction of the actual wind, the *y*-axis is oriented in the horizontal crosswind direction, and the *z*-axis is chosen vertically upwards.

The mathematical description of the dispersion problem (1) is completed by boundary conditions. In the *z*-direction, the pollutants are subjected to the boundary conditions of zero flux at ground and PBL top:

$$K_z\frac{\partial c}{\partial z}=0 \qquad \text{at} \quad z=0, z_i \tag{2a}$$

In the *y*-direction, we have the conditions:

$$\frac{\partial c}{\partial y}=0 \qquad \text{at} \quad y=0, L_y \tag{2b}$$

and, for the source condition, a continuous point source of constant emission rate Q is assumed, with a fixed frame of reference with the *x*-axis coinciding with the plume (Arya, 2003):

$$uc(0,y,z)=Q\delta(z-H_s)\delta(y-y_o) \qquad \text{at} \quad x=0 \tag{2c}$$

where δ is the Dirac delta function and H_s is the source height.

To solve the advection-diffusion equation for inhomogeneous turbulence by the ADMM method, we must take into account the dependence on the eddy diffusivities and wind speed on the height variable (variable *z*). To reach this goal we discretize the height z_i of the PBL into *N* sub-intervals in such manner that inside each sub-region, $K(z)$ and $u(z)$ assume respectively the following average values:

$$K_n=\frac{1}{z_{n+1}-z_n}\int_{z_n}^{z_{n+1}}K_z(z)dz \tag{3}$$

$$u_n=\frac{1}{z_{n+1}-z_n}\int_{z_n}^{z_{n+1}}u(z)dz \tag{4}$$

for $n=1:N$.

Now we are in position to solve the advection-diffusion equation by the Laplace transform technique for each sub-interval (neglecting the longitudinal diffusion):

$$u_n\frac{\partial c_n}{\partial x}=\frac{\partial}{\partial y}\left(K_{y_n}\frac{\partial c_n}{\partial y}\right)+\frac{\partial}{\partial z}\left(K_{z_n}\frac{\partial c_n}{\partial z}\right) \qquad z_n\le z\le z_{n+1} \tag{5}$$

for n = 1:N, where N denotes the number of sub-layers and c_n denotes the concentration at the n^{th} sub-interval. Besides which, two boundary conditions are imposed at $z = 0$ and z_i given by Eq. (2a) together with the continuity conditions for the concentration and flux of concentration at the interfaces. Namely:

$$c_n = c_{n+1} \qquad n = 1, 2,...(\mathrm{N}-1) \tag{6}$$

$$K_{zn}\left(\frac{\partial c_n}{\partial z}\right) = K_{zn+1}\left(\frac{\partial c_{n+1}}{\partial z}\right) \qquad n = 1, 2,...(\mathrm{N}-1) \tag{7}$$

must be considered, in order to be possible to uniquely determine the 2N arbitrary constants appearing in the solution of the set of problems (5).

Now, we are in position of applying the GITT method in the y-direction. Following the formalism of GITT, we begin expanding the variable $c_n(x, y, z)$ by the series:

$$c_n(x, y, z) = \sum_{i=0}^{\infty} \frac{\bar{c}_{ni}(x, z)\psi_i(y)}{\mathrm{N}_i^{1/2}} \tag{8}$$

Here $\psi_i(y) = \cos(\lambda_i y)$ are the eigenfunctions of the auxiliary Sturm-Liouville problem in the y variable and $\lambda_i = i\pi / L_y$ the corresponding eigenvalues. Now, likewise of the Moreira et al. (2005), we replace the Eq. (8) into Eq. (5) and we have:

$$u_n \sum_{i=0}^{\infty} \frac{\partial \bar{c}_{ni}(x, z)}{\partial x} \frac{\psi_i(y)}{\mathrm{N}_i^{1/2}} = K_y \sum_{i=0}^{\infty} \bar{c}_{ni}(x, z) \frac{\psi_i''(y)}{\mathrm{N}_i^{1/2}} + K_z \sum_{i=0}^{\infty} \frac{\partial^2 \bar{c}_{ni}(x, z)}{\partial z^2} \frac{\psi_i(y)}{\mathrm{N}_i^{1/2}} \tag{9}$$

Taking moments and solving the resulting transformed problem we come out with the result:

$$\widehat{c}_{ni}(s, z) = C_{1n} e^{(F_n+R_n)z} + C_{2n} e^{-(F_n-R_n)z} + \frac{Q}{2R_a}\left(e^{(F_n+R_n)(z-H_s)} - e^{(F_n-R_n)(z-H_s)}\right) \tag{10}$$

where

$$R_n = \sqrt{\frac{su_n + K_y\lambda_j^2}{K_z}} \quad \text{and} \quad R_a = \frac{N_i^{1/2}}{\psi_i(y_0)}\sqrt{K_z\left(su_n + K_y\lambda_i^2\right)} \tag{11}$$

The integration constants C_{1n} and C_{2n} are determined by solving the linear system resulting from of the application of the boundary and interfaces conditions.

The final concentration is finally obtained by inverting numerically the transformed concentration $\widehat{c}_{n\,i}$ expressed by Eq. (10) applying the Fixed Talbot (FT) algorithm (Valkó and Abate, 2004; Abate and Valkó, 2004). This procedure yields to the solution:

$$c_n(x, y, z) = \sum_{i=0}^{\infty} \frac{\psi_i(y)}{N_i^{1/2}} \left\{ \frac{r}{M^*} \left[\frac{1}{2} \hat{c}_{ni}(r, z) e^{rx} + \sum_{k=1}^{M^*-1} \mathrm{Re}\left[e^{xS(\theta_k)} \hat{c}_{ni}\left(s(\theta_k), z\right)\left(1 + i\tau(\theta_k)\right)\right]\right]\right\} \tag{12}$$

where

$$s(\theta_k) = r\theta(\cot\theta + i), \qquad -\pi < \theta < +\pi$$
$$\tau(\theta_k) = \theta_k + \left(\theta_k \cot\theta_k - 1\right)\cot\theta_k$$

$$\theta_k = \frac{k\,\pi}{M^*}$$

and r is a parameter based on numerical experiments. To control the round-off error in the computation of Eq. (12), we specify the precision requirement: number of precision decimal digits = M^*.

3. PLUME RISE

A correct estimation of buoyant plume rise is one of the basic requirements for the determination of ground level concentrations of airborne pollutant emitted by industrial stacks. In fact, maximum ground level concentration is roughly inversely proportional to the square of the final height h_e. For this reason, in many simple dispersion models, stack gases are assumed to be emitted from a virtual source located at height h_e along the vertical above the stack. The effective plume height h_e (elevation of plume centerline relative to ground level) results from the sum of stack height H_s and plume rise Δh:

$$h_e = H_s + \Delta h \tag{13}$$

Some formulas provide the plume rise as a function of the distance, but most of them provide a constant value (final plume rise) that the plume reaches at a large downwind distance. These formulas contain height depending atmospheric variables normally specified at the stack outlet height.

Several studies and review works have provided semi-empirical formula for evaluating Δh (e.g., Briggs, 1975; Stern, 1976; Hanna et al., 1982; and many others). Others researchers have provided more complex and comprehensive descriptions of several physical interactions between the plume and the ambient air (e.g., Golay, 1982; Netterville, 1990). Relevant and exhaustive review papers on the plume rise subject can be found in the literature, for instance, Briggs (1975) and Weil (1988). In this work, we are utilizing the formulas of Briggs (1975) applied by Moreira (2000).

Briggs (1975) made a distinction between neutral and unstable conditions accounting for the effects of ambient turbulence on the plume rise. While self-generated turbulence affects the entrainment process near the source, ambient turbulence (with both small and large scale eddies) becomes important further downwind. Small scale eddies, are responsible for the increase of the plume growth rate beyond that given by self-induced turbulence. The breakup model (Briggs, 1975; Weil, 1988) assumes that plume rise finishes when ambient turbulence "breaks up" the self-generated structure of the plume, causing a vigorous mixing, and, consequently, gradually loses buoyancy and momentum and eventually level off. Thus, this process leads to an asymptotic rise. According to Briggs, the plume breakup occurs when the ambient rate of dissipation of turbulent kinetic energy, ε_a, exceeds the one of the plume ε. Large scale eddies (updrafts and downdrafts in the convective boundary layer (CBL)) may transport plume segments up and down, thereby dispersing the plume by vertical meandering and pushing some of them to the surface. When this happens, the time averaged ground level concentration is more dependent on how many times, during the averaging period, the plume touches the ground than on the height of the asymptotic rise. As a consequence, in the CBL case, the leading parameter is assumed to be the surface sensible heat flux, which plays the major role in the development of updrafts and downdrafts.

In strong convection ($z_i/|L| > 10$) the model "breaks up" has a final behavior given for:

$$\Delta h = 4.3\left(\frac{F}{U w_*^2}\right)^{3/5} z_i^{2/5} \tag{14}$$

where the rate of ambient dissipation is assumed to be $0.1\,\frac{w_*^3}{z_i}$. The buoyancy parameter F is given for:

$$F = g V_i r_i^2 \frac{(T_i - T_a)}{T_i} \tag{15}$$

where V_i and T_i are the vertical velocity and temperature, respectively, in the exit of the chimney, T_a is the ambient temperature, g the acceleration of the gravity and r_i is the radius of the source. The model defines a "touchdown" for moderate convective conditions predicts the behavior of the plume for:

$$\Delta h = 1.0\left(\frac{F}{Uw_d^2}\right)\left(1+\frac{2H_s}{\Delta h}\right)^2 \tag{16}$$

where w_d is the medium speed of the downdrafts, considered as $w_d = 0.4 w_*$. The resulting equation is iteratively solved for Δh. In neutral stability, the "breaks up" model predicts the following behavior:

$$\Delta h = 1.3\frac{F}{Uu_*^2}\left(1+\frac{H_s}{\Delta h}\right)^{2/3} \tag{17}$$

In this work, the penetration of the plume is not considered due to the boundary conditions of the K-model. Then, if the plume is completely preyed, Weil (1979) suggests that the restriction geometric limit for Δh is:

$$\Delta h = 0.62(z_i - H_s) \tag{18}$$

In certain cases, Briggs (1975) recommends to use the formulas that provides the minimum plume rise; this result is "the most conservative", since it gives rise to the maximum values of concentration expected at the ground, thus limiting the risk of a possible underestimation. Then, the formulas can be summarized as it proceeds:

$$\Delta h = min(\text{Eqs. } 14, 16, 17, 18) \tag{19}$$

4. BOUNDARY LAYER PARAMETERIZATION

In order to illustrate the suitability of the discussed formulation to simulate contaminant dispersion in the PBL, we evaluate the performance of the solution (12) against experimental ground-level concentration. To do this we have to introduce a boundary layer parameterization. The literature reports many, greatly varied formulas, for the calculation of the vertical turbulent diffusion coefficient (Seinfeld and Pandis, 1997). As examples of applications of our new solution we tested the following vertical diffusion parameterization suggested by Pleim e Chang (1992) for convective conditions:

$$K_z = k\, w_* z\left(1-\frac{z}{z_i}\right) \tag{20}$$

and the following lateral diffusion parameterization suggested by Degrazia et al. 1997:

$$K_y = \frac{\sqrt{\pi}\,\sigma_v}{16(f_m)_v q_v} \tag{21}$$

with:

$$\sigma_v^2 = \frac{0.98 c_v}{(f_m)_v^{2/3}}\left(\frac{\psi_\varepsilon}{q_v}\right)^{2/3}\left(\frac{z}{z_i}\right)^{2/3} w_*^2 \tag{22}$$

$$q_v = 4.16\frac{z}{z_i} \tag{23}$$

$$\psi_\varepsilon^{1/3} = \left[\left(1-\frac{z}{z_i}\right)^2\left(-\frac{z}{L}\right)^{-2/3} + 0.75\right]^{1/2} \tag{24}$$

$$(f_m)_v = 0.16 \tag{25}$$

where k is the von Karman constant ($k = 0.4$), w_* is the convective velocity scale, σ_v is the Eulerian turbulent lateral velocity variance, q_v is the stability function, ψ_ε is the non-dimensional molecular dissipation rate function and $(f_m)_v$ is the lateral peak wavelength.

The wind speed profile was described by a power law expressed as follows (Panofsky and Dutton, 1988):

$$\frac{\overline{u}_z}{\overline{u}_1} = \left(\frac{z}{z_1}\right)^p \tag{26}$$

where $\overline{u}_z$ and $\overline{u}_1$ are the mean wind velocity at the heights z and z_1, while p is an exponent that is related to the intensity of turbulence (Irwin, 1979).

5. NUMERICAL RESULTS

In order to show an example of application of the solution and to evaluate the performance of the proposed PBL parameterization we have applied the Kinkaid dataset (Illinois - USA). The Kincaid field campaign (Bowne and Londergan, 1981) concerns an elevated release in a flat farmland with some lakes. For the experiment a SF6 tracer was released from a 187 tall stack and data were recorded on a network consisting of roughly 200 samplers positioned in different arcs in an area of 50 km downwind of the source. The data set includes the meteorological parameters as friction velocity, Obukhov-Monin length and height of boundary layer. The plume rise effect (warm source) was evaluated using the approach proposed by Briggs (1975). The measured concentration level is frequently irregular with high and low concentrations occurring intermittently along the same arc, moreover there are frequently gaps in the monitoring arcs. For the above reasons a variable has been assigned as a quality factor in order to indicate the degree of readability of data (Olesen, 1995). The quality indicator (from 0 to 3) has been assigned. Here, only the arc maximum concentration data with quality factor 3 relatively to convective conditions were considered.

Figure 1 shows the observed and predicted scatter diagram of ground-level centerline concentrations using the above parameterizations, for wind and eddy diffusivities profiles, and the GIADMT model for the Kinkaid dataset. The Fig. 1 point out that a good agreement is obtained between experimental data and the model considering the centerline concentrations.

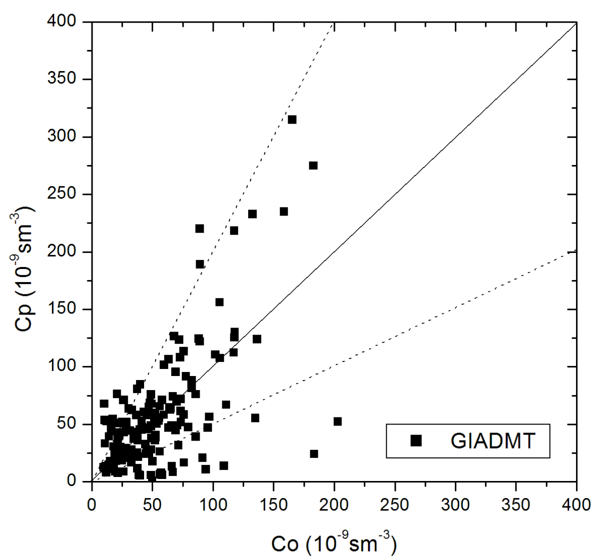

Figure 1. Observed (Co) and predicted (Cp) crosswind ground-level integrated concentration scatter diagram for the GIADMT model. Dotted lines indicate a factor of two.

In Table 1 some performance measurements are presented. We used the statistical evaluation procedure described by Hanna (1989) and defined in the following way:

NMSE (normalised mean square error) = $\overline{(C_o - C_p)^2} / \overline{C_o}\,\overline{C_p}$

COR (correlation coefficient) = $\overline{(C_o - \overline{C_o})(C_p - \overline{C_p})} / \sigma_o \sigma_p$

FA2 (factor of 2) = $C_p / C_o \in [0.5,2]$

FB (fractional bias) = $\left(\overline{C_o} - \overline{C_p}\right) / \left(0.5\left(\overline{C_o} + \overline{C_p}\right)\right)$

FS (fractional standard deviations) = $(\sigma_o - \sigma_p) / 0.5(\sigma_o + \sigma_p)$

where subscripts o and p refer to observed and predicted quantities, respectively, σ the standard deviation and an overbar indicates an average.

Table 1. Statistical evaluation of performances for Kinkaid data set

NMSE	COR	FA2	FB	FS
0.50	0.63	0.73	-0.04	-0.32

The analysis of the above results shows a reasonable agreement between the computed values against the experimental data.

6. CONCLUSIONS

We have presented a semi-analytical solution of the three-dimensional steady state advection diffusion equation incorporating the plume rise effect, which could be applied in operative models for describing turbulent dispersion of many scalar quantities, such as air pollution, radioactive material, heat and so on. In order to show the performances of the solution in actual scenarios, we introduced some parameterizations of the PBL and compared the values predicted by the solutions with data collected during the well-known Kinkaid experiment. The analysis of the results shows a reasonably agreement between the computed values against the experimental ones.

When using models, while they are rather sophisticated instruments that ultimately reflect the current state of knowledge on turbulent transport in the atmosphere, the results they provide are subject to a considerable margin of error. This is due to various factors, including in particular the uncertainty of the intrinsic variability of the atmosphere. Models, in fact, provide values expressed as an average, i.e. a mean value obtained by the repeated performance of many experiments, while the measured concentrations are a single value of the sample to which the ensemble average provided by models refer. This is a general characteristic of the theory of atmospheric turbulence and is a consequence of the statistical approach used in attempting to parameterize the chaotic character of the measured data.

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