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POLLUTANT DISPERSION IN THE ATMOSPHERE WITH EDDY DIFFUSIVITY DEPENDING ON THE SOURCE DISTANCE

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Abstract. *The present study aims to solve the steady-state two-dimensional advection-diffusion equation with vertical eddy diffusivity dependent on the source distance in a vertically inhomogeneous planetary boundary layer. The Eulerian models with eddy diffusivity depending on the source distance capture the memory effect present in the adjacencies of the source, making the analytical models more reliable. For this, the GILTT (Generalized Integral Laplace Transform Technique) analytical method was applied in the resolution of the advection-diffusion equation. For the purposes of comparison and validation of the presented model, we used the experimental data from Copenhagen and Prairie Grass, comparing the results obtained by the solution with the coefficient Kz(x,z) with those obtained with the coefficient Kz(z). The obtained values suggest that the inclusion of the memory effect improves the description of the process of turbulent transport of contaminants in the atmosphere, only to sources close to the ground and under extremely unstable conditions*

Keywords: *GILTT; Pollutant Dispersion; Memory Effect; Advection-Diffusion Equation, Eulerian Models.*

1. INTRODUCTION

In the last century, industrial development, growing demand for energy, vehicular emissions and fires have generated a great impact on the environmen, especially as regards the quality of the air we breathe. Unfortunately, these changes have not been accompanied by adequate analysis and sanitation measures in order to assess their impact on the environment, the toxicity of their residues and their probable harm to health. Thus, the management and protection of air quality have become indispensable factors in ensuring socio-conomic development in a sustainable and enviromentally safe manner.

Air quality assessment is often carried out through monitoring networks, yet this monitoring and control instrument has limitations of use and high implementation costs. In this context, mathematical models are presented viable and low-cost alternatives that allow description, interpretation, management of accidental releases, evaluation of risk areas, identification of polluting sources, correction and control of the concentration pollutants in the atmosphere (Moreira & Tirabassi, 2004).

Currently, there are many mathematical models for this purpose, among them; we highlight computational simulations and analytical solutions of the diffusion-avection equation. Although they are widely used in simulation of atmospheric processes, computational simulations have limitations in computational cost, since they often require the use of supercomputers. On the other hand, the analytical or semi-analytic solutions may represent these processes with a high degree of accuracy at relatively low computational cost, in addition to being capable of represent the influence of the parameters related to this dispersion process of the pollutants in an explicit manner (Carvalho & Moreira, 2007). Moreover, the study and improvement of these solutions enable the development, validation, calibration and improvement of numerical models.

The process of dispersion of pollutants in the Planetary Boundary Layer (PBL) is, often, modeled by the diffusion-advection equation, which justifies the great interest of the community in the study of analytical solutions of this equation. In this sense there's a significant scientific advance in relation to obtaining analytic / semi-analytic solutions for the diffusion- advection equation , using different methodologies, as described in the works of (Rounds, 1955);(Yeh and Huang, 1975); (Moreira et al., 2005, 2009,2014); (Sharan and Modani, 2006); (Essa et al., 2007);(Tirabassi et al., 2008); (Pimentel et al., 2014).

Another important factor in the transport of pollutants can be found in the choice of the turbulent parameterization; after all, it describes the behavior of turbulence and nature around the source from the physical point of view. In his statistical theory of diffusion, Taylor (1922) showed that turbulent diffusion is different in regions that are close to and distant from a continuous source. In adjacent regions to the source, fluid particles keep the memory of its turbulent environment. For long travelling times, this memory is lost, and the particles only follow the properties of local turbulence. The diffusion coefficients that depend on the distance from the source enable the capture of the memory effect that is present in areas close to high and low source, turning analytical models more reliable as described in the Sharan et al. (1996), Degrazia et al. (2001) and Moreira et al. (2014).

In order to investigate how the coefficient of diffusion will influence the sensitivity of the proposed model, the diffusion-advection equation was solved analytically, applying the GILTT (Generalized Integral Laplace Transformation Technique). For more information on the GILTT method, consult (Wortmann et al. 2005); (Moreira et al., 2005); (Weymar, 2016); (Buske Daniela et al., 2017).

In this context, the proposed model was parameterized with the vertical diffusion coefficients proposed in the works by Mooney and Wilson (1993) and Moreira et al. (2002). The first one is applied to high moderate weather conditions convective, and the second, for low and strongly convective conditions, both exhibited as follows:

$$K_z(x,z) = f(x).g(z) \tag{1}$$

Where $g(z)$ is the functional form of the coefficient on the diffusion dependent only on the spatial z variable, and $f(x)$ is the correction of $g(z)$ points near the source.

To evaluate the performance of the model studied, data generated by the solutions were compared with experimental data from Copenhagen and Prairie Grass.

2. GILTT METHODOLOGY

The following are the main steps of the GILTT methodology applied to the diffusion- two-dimensional advection with diffusion coefficient that depend on the distance from the source, stationary, based on K theory, for a system of Cartesian coordinates whose source is in BPL, given by the following

$$U(z)\frac{\partial C}{\partial x} = \frac{\partial}{\partial z}\left(\left(K_z(x,z)\frac{\partial C}{\partial z}\right)\right) \tag{2}$$

with the following boundary conditions

$$-K_z(x,z)\frac{\partial C}{\partial z} = 0 \; em \; z = 0 \; e \; z = h \tag{3}$$

and the condition of source

$$C(0,z) = \frac{Q}{U(z)}\delta(z - H_s) \tag{4}$$

where Hs represents the height of the source, δ is the Dirac delta and h is PBL height.
To apply the GILTT technique it is necessary that the initial conditions or contour conditions are homogeneous, and that the problem is of finite dimension. In this context, GILTT will be applied to variable z.

The first step is to find the equation that represents the auxiliary problem of Sturm-Liouville. For this, Eq. (1) is substituted in Eq. (2), obtaining:

$$\frac{U(z)}{f(x)}\frac{\partial C}{\partial x} = \frac{\partial}{\partial z}\left[g(z)\frac{\partial C}{\partial z}\right] \tag{5}$$

In order to obtain the auxiliary equation, the substitution of the variable suggested in the Crank, (1968), as shown below:

$$dX = f(x)dx \tag{6}$$

Thus, Eq. (5) can be rewritten as

$$u(z)\frac{\partial C}{\partial X}=\frac{\partial}{\partial z}\left[g(z)\frac{\partial C}{\partial z}\right] \tag{7}$$

Using the chain rule to the diffusive term of Eq. (7) (Wortmann et al., 2005), and applying the necessary simplifications, the auxiliary problem equation is obtained as follows:

$$\phi(z)^{"}+\lambda^2\phi(z)=0 \tag{8}$$

and their respective boundary conditions

$$\phi^{'}(z)=0,\ \text{em}\ z\ =\ 0,h \tag{9}$$

This problem has the traditional solution given by:

$$\phi_n(z)=\cos\left(\frac{n\pi z}{h}\right)\text{com}\ n=1,2,3,.... \tag{10}$$

The next step is to expand the variable $C(X,z)$ in a series as displayed in the formula below:

$$c(X,z)=\sum_{n=0}^{\infty}A_n(X)\phi_n(z) \tag{11}$$

where $A_n(x)$, n = 0, 1, 2,..., are the unknown coefficients of the series. To determine them, replace Eq. (11) in Eq. (7), thus:

$$\sum_{n=0}^{\infty}\left(\int_0^h U(z)\phi_m(z)\phi_n(z)dz\right)\frac{dA_n(X)}{dX}=\sum_{n=0}^{\infty}\left[\int_0^h \phi_m(z)\left\{-g(z)\lambda^2\phi_n(z)+\frac{dg(z)}{dz}\frac{d\phi_n}{dz}\right\}dz\right]A_n(X) \tag{12}$$

For convenience, Eq. (12) will be rewritten in matrix form, resulting in:

$$A'(X)+FA(X)=0 \tag{16}$$

subject to the initial condition

$$Q\phi_m(h_s)=A_m(0).\frac{h}{2} \tag{17}$$

where $A(X)$ represents a vector , and matrix F is defined as $F=B^{-1}E$.

The Equation (16) is then solved by applying the Laplace transform and diagonalization process (Segatto and Vilhena, 1999), providing the transformed solution:

$$\overline{A(s)}=W\left(s.I+D\right)^{-1}W^{-1}.A(0) \tag{18}$$

where D represents the diagonal matrix of eigen values, W the matrix of the autofunctions of F, its inverse, and I represents the identity matrix.

The elements of the matrix $sI+D$ have the form $s+d_i$, where d_i are the eigenvalues of matrix F or the elements of the diagonal matrix D. Since $sI+D$ is a diagonal matrix of algebra matrix, it is possible to say that its inverse is given by multiplicative inverse of the elements of the diagonal main elements, so the elements of matrix $\left(sI+D\right)^{-1}$ take the form $\frac{1}{s+d_i}$, whose inverse Laplace transform is e^{Xd_i}. Of Equation (6), one has to $X = \int_0^x f(x^*)dx^*$, meaning the diagonal matrix $L^{-1}\left\{(sI+D)^{-1}\right\}$, will have the elements $e^{d_i \int_0^x f(x^*)dx^*}$. Thus, the final solution of Eq. (18) is given by

$$A(X) = X.L^{-1}\left\{(s.I+D)^{-1}\right\}X^{-1}.A(0) \tag{19}$$

For more details, see Wortmann et al. (2005) and Moreira et al. (2005).

Thus, the solution of the problem expressed by Eq. (2) is finally obtained and given by:

$$c(X,z) = \sum_{n=0}^{\infty} A_n(X)\phi_n(z) \tag{20}$$

at where $\phi_n(z)$ represents the eigenvalues obtained from equation (10) (Sturm-Liouville problem) and vector $A_n(x)$ is the result of the solution of problem transformed (19). It should be noted that solution provided by (20) does not have any approximation, except for the truncation error incurred to the expansion of the series.

3. PARAMETERIZATION OF ATMOSPHERIC TURBULENCE

The parameterization of turbulence is the key factor in dispersion models. The reliability of each model depends on how turbulent parameters are calculated and is related to the understanding of BPL behavior in its current state (Mangia et al., 2002). Therefore, the atmospheric conditions are a major factor relevant in the parameterization process of the coefficient, since they describe several scenarios in the environment. In this way, expressions used to represent the parameterization of the turbulent diffusion coefficients are considered in accordance with the conditions of the atmosphere.

In this context, the model proposed in this paper was parameterized with two types of coefficients of vertical diffusion, the first one dependent on vertical variable z, and the second, dependent on the spatial variables x and z. It is intended, therefore, to investigate the influence that longitudinal distance source has in the process of diffusing the pollutant.

As for the vertical diffusion coefficient dependent on the spatial variable z, the formulations proposed by Degrazia et al. (1997) and Troen & amp; Mahrt (1986), both applied to atmospheric conditions were applied.

The diffusion coefficient proposed by Degrazia, Velho & Carvalho (1997) was deduced based on Taylor's statistical theory of and the spectrum of turbulent energy. It shall apply PBL and valid for long diffusion times obtained through the expression below:

$$K_z(z) = 0,22(\omega_* h)\left[\frac{z}{h}\left(1-\frac{z}{h}\right)\right]^{1/3}\left[1-\exp\left(-\frac{4z}{h}\right)-0,0003\exp\left(\frac{8z}{h}\right)\right] \tag{21}$$

where h represents the height of the convective layer, ω_* is the convective velocity and z represents the height above ground.

The diffusion coefficient proposed by Troen & Mahrt (1986), proposes an expression where turbulent diffusivities have a profile shape prescribed by z/h function and scale parameters from similarity arguments. Is applicable to all unstable PLC and calculated through of the following expression:

$$K_z(z) = k\omega_* z\left(1-\frac{z}{h}\right) \tag{22}$$

where $k \cong 0,4$ represents the constant of Von Kármam and h is the top of the BPL.

For the dependent vertical diffusion coefficient of the spatial variables x and y, the formulations proposed by Mooney and Wilson (1993) and Moreira et al. (2002) were.

The first is valid for points located near the source, whose wind direction coincides with that of the axis x, and to a moderately convective. Thus, it is used for kz, a modified form, the wording of which is given by the product of two independent diffusion coefficients, the first in terms of altitude and second in terms of the longitudinal distance from the source, given by the following:

$$K_z(x,z) = K_z(z)\left[1-\exp\left(-\frac{x}{L_1}\right)\right] \tag{23}$$

where $K_z(z)$ represents the functional expression of turbulent diffusivity depending only on the vertical variable z, $L_1 = U(H_s)\tau(H_s)$ is the scale along the wind through which the coefficient of diffusion reaches its asymptotic value and τ is the Lagrangian scale of time at the height of the source, given by $\tau(z) = K_z(z)/\sigma_w{}^2$, where σ_w represents the standard deviation of vertical velocity.

In the present study, Eq. (21) and (22) to the $K_z(z)$ da Eq. (23).

The turbulent diffusion coefficient proposed by Moreira et al. (2002), is only valid for a source at ground level under strongly convective conditions. Yaglom's similarity theory of (1972) was used to estimate these quantities, expressed by:

$$K_z(x,z) = \mu h \omega_* \left(\frac{x\omega_*}{\mu h}\right)^2 \left[\left(-\frac{L}{z}+3\right)\right] \tag{24}$$

where μ a constant whose optimal value found is 2.0 and L is the length of Obukhov.

4. WIND PROFILE

The equations adopted by the model to simulate the behavior of the average winds are those predicted by the theory of similarity (Panofsky and Dutton, 1984):

$$u = \frac{u_*}{k_*}\left[\ln\frac{z}{z_0} - \psi_m\left(\frac{z}{L}\right)\right] \tag{25}$$

where u_* is the scalar velocity relative to mechanical turbulence, k is von Karmanis constant, z_0 the roughness length and ψ_m the stability function expressed in the relations of Businger:

$$\psi_m\left(\frac{z}{L}\right) = -4,7\frac{z}{L} \text{ for } \frac{1}{L} \geq 0 \tag{26}$$

and

$$\Psi_m\left(\frac{z}{L}\right) = \ln\left(\frac{1+x^2}{2}\right) + \ln\left(\frac{1+x^2}{2}\right)^2 \text{-}2\arctan x + \frac{\pi}{2} \text{ for } \frac{1}{L} < 0 \tag{27}$$

where $x = \left(1 - 15\frac{z}{L}\right)^{1/4}$

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

$$\frac{u_2}{u_1} = \left(\frac{z_2}{z_1}\right)^n \tag{28}$$

in which u_1 and u_2 are the average speeds of the wind at points z_1 and z_2 measured in the direction of vertical z from the ground, and represents a number associated with the turbulence, roughness and difference between points taken as a reference.

5. RESULTS

To evaluate the accuracy of the model, the results obtained were compared with experimental data from Copenhagen and Prairie Grass.

The Copenhagen experiment, described by Gryning and Lyck (1984), was carried out north of in the municipality of Gladsaxe under neutral and unstable atmospheric conditions. The experiment consisted of the release without buoyancy of a chemical tracer, Sulfur Hexafluoride (SF6), from a 115 meter-tall tower. The samples were collected at ground level (z = 2m) by 60 collectors in positions located on three arcs concentric to the source and perpendicular to the direction of the average radial distances of 2, 4 and 6 km. It is worth noting that the region in which the experiment was performed was predominantly residential, with 0.6 m roughness length.

In contrast, the Prairie Grass experiment was carried out in O'Neill, Nebraska, under stable and unstable atmospheric conditions during the summer of 1956 (BARAD, 1958). It consists of a set of standards of observations (68 experiments) widely used in the validation to dispersal models. During the experimental trial, the sulfur dioxide (SO 2) was released from a source point at a height of 0.46 m (except for experiments 65 to 68) of the soil. Measurements concentrations were carried out at a height of 1.5 m along concentric arcs located in 50, 100, 200, 400 and 800 m intervals As this was a virtually flat terrain, the length of surface roughness is 0.6 cm. About half of the 68 experiments refers to thermal stratification (daytime) and the remainder was obtained in conditions (nighttime), in the presence of temperature inversions.

Data generated in this section were obtained from the data from the two previously mentioned experiments (Copenhagen and Prairie Grass) together with the turbulent parameterizations and wind profiles discussed in the previous sections.

Figure 1 shows the numerical convergence of parameterized solution with a coefficient of diffusion dependent on the distance from the source, Eq. (23), and the logarithmic wind profile, Eq. (25), and the micrometeorological data from Copenhagen's experiment 9. From this analysis, it is noted that for N > 80, the Concentrations reach numerical stability, that is, they suffer little variation, which shows that the method converges for values of N = 80.

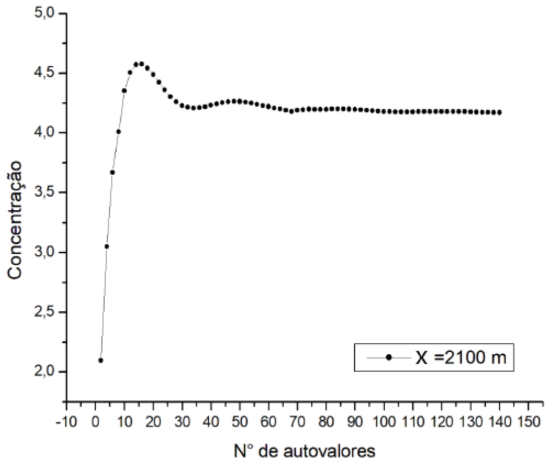

Figure 1. Numerical convergence of concentration of pollutants for Copenhagen's experiment 9 to x = 2100m.
Reference: Soledade et al. (2019)

In order to verify the contribution of the memory effect, the model was parameterized for relatively high sources, subject to moderately unstable atmospheric conditions, taking the micrometeorological data from Copenhagen's experiment.

In this context, the model was parameterized with coefficient proposed by Mooney & amp; Wilson (1993), Eq. (23), and compared with the parameterized solution with the coefficients proposed by Degrazia et al. (1997), Eq. (21), and Troen & Mahrt (1986), Eq. (22). Also, different wind profiles were adopted (power and logarithmic), in order to verify which of them provided the best accuracy in results.

Data obtained were statistically treated (Hanna, 1989), tabulated and organized in the Table 1. In cases I and II, the solution was parameterized with the vertical diffusion coefficient, given by Eq. (23), taking equations Eq. (21) and (22) for, respectively. In cases III and IV, the model was parameterized with the vertical diffusion coefficient, given by Eq. (21) and Eq. (22), respectively. The logarithmic wind profile was adopted for all cases (I, II, III and IV).

indicate good agreement between the data and the GILTT method. For a satisfactory analysis of the statistical indices, the optimal results are close to zero for the Nmse, Fb and Fs parameters and close to one for Color and Fa2 indicators (Hanna, 1989).

Table 1. Statistical model indices applied to the micrometeorological conditions of the experiment in Copenhagen.

	Coeficiente	Nmse	Cor	Fa2	Fb	Fs
I	$K_z(x,z)$	0.06	0.888	1.000	0.006	0.217
II	$K_z(x,z)$	0.06	0.891	1.000	0.009	0.182
III	$K_z(z)$	0.08	0,907	1,000	0.112	0.286
IV	$K_z(z)$	0.09	0.901	1.000	0.140	0.258

Reference: Soledade et al.; (2019)

Confronting the statistical indices of cases I and II with cases III and IV, one can observe that the values obtained in case I and II are relatively better than those provided in III and IV, with the statistical correlation index, whose value was slightly below 90%, but acceptable. However, it is not possible to ascertain the longitudinal distance of the source in the diffusion process, since the indices are relatively close.

Figures 2 and 3 show the scatter plots derived from Eq. (20), parameterized with diffusion coefficients given by Equations (22) and (23), respectively, for the wind profile logarithm, using the micrometeorological data from Copenhagen experiment. Where the straight line corresponding to the first bisector indicates the agreement between the estimated and observed results. Thus, the closer the points are to the bisector line, the greater the conformity between the observed and predicted values. As for the dashed straight lines indicate the confidence interval for the factor of two. Thus, all values obtained in graphs 2 and 3 are within a factor of two.

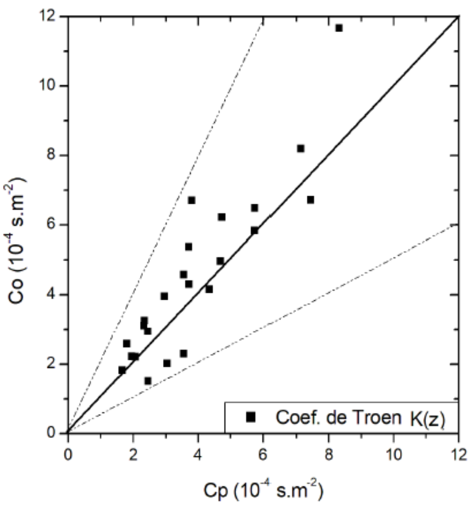

Figure 3. Data from observed concentrations (C_o) in the experiment compared to Data from maximal concentration predicted (C_p) in case II scatter plot. The first bisector indicates the agreement between the estimated and observed results.
Reference: Soledade et al. (2019)

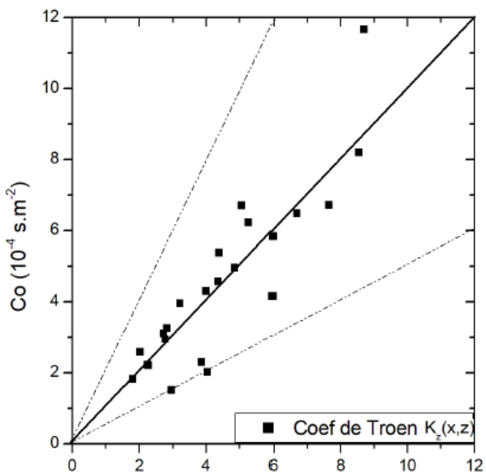

Figure 2. Data from Observed concentrations (C_o) in the experiment compared to Data from maximal concentration predicted (C_p) in case IV scatter plot. The first bisector indicates the agreement between the estimated and observed results.
Reference: Soledade et al. (2019)

The analysis of dispersion plots suggests a greater agreement between the observed and predicted values for case II (Figure 3) when compared to Case IV (Figure 2), since there is a greater concentration of points on the 1st bisector of the scatter plot. However, this analysis is not corroborated when comparing the statistical indices.

In the second part of the research, the effect of memory for the light sources, subject to to intensely convective atmospheric conditions was investigated, taking into account micrometeorological data from the Prairie Grass experiment. In this scenario, the solution was parameterized with the coefficient proposed by Moreira et al. (2002), Eq. (24), and compared with the parameterized models with the vertical diffusion coefficient , given by Eq. (23), with Eq. (21) for and vertical diffusion coefficient given by Eq. (21).

The statistical indices obtained in Table 2 indicate reasonable agreement between this study's experimental data and the GILTT method only for the case I. Analyzing the statistical indices (Hanna, 1989) we notice that this model satisfactorily simulates the observed concentrations in relation to values of Fb and Fs close to zero and relatively close to 1.

Confronting the statistical indices of Table 2, it is observed that all indices obtained in case I are better than cases II and III, thus revealing, the importance of a satisfactory parameterization of the solution, considering that the diffusion coefficient proposed by Moreira et al. (2002) better describes the conditions of strong convection present in the Prairie Grass experiment, while the coefficient proposed by Mooney & amp; Wilson (1993) (cases II) was formulated into an moderately convective atmosphere.

Table 2. Statistical indices of the model applied the micrometeorological conditions of the Prairie Grass experiment.

	Coeficiente	Nmse	Cor	Fa2	Fb	Fs
I	$K_z(x,z)$	0.52	0.814	0.830	0.165	0.006
II	$K_z(x,z)$	0.84	0.727	0.620	0.320	0.269
III	$K_z(z)$	0.90	0.780	0.630	0.420	0.480

Reference: Soledade (2019)

The results also highlight the insertion of the x variable into the vertical diffusion coefficient. It is of fundamental importance in capturing the memory effect, a fact that can be verified by comparing the statistical indices of cases I and II with case III.

Figures 4 and 5 show the dispersion plots derived from Eq. (20), parameterized with the diffusion coefficients given by Equations (21) and (24), respectively, for the logarithmic wind profile, using the micrometeorological data of the Prairie Grass experiment.

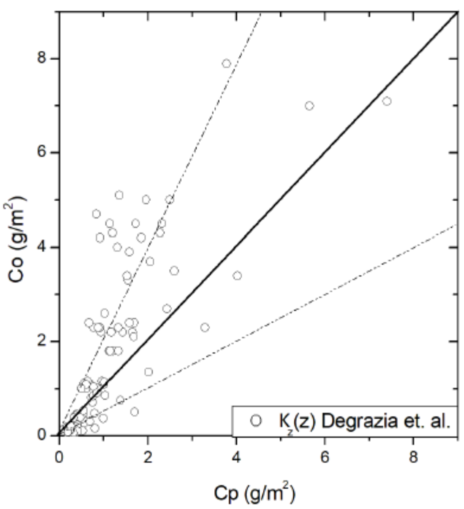

Figure 4. Data from observed concentrations (Co) in the experiment compared to Data from maximal concentration predicted (Cp) in case II scatter plot in case III . The first bisector indicates the agreement between the estimated and observed results.
Reference: Soledade et al. (2019)

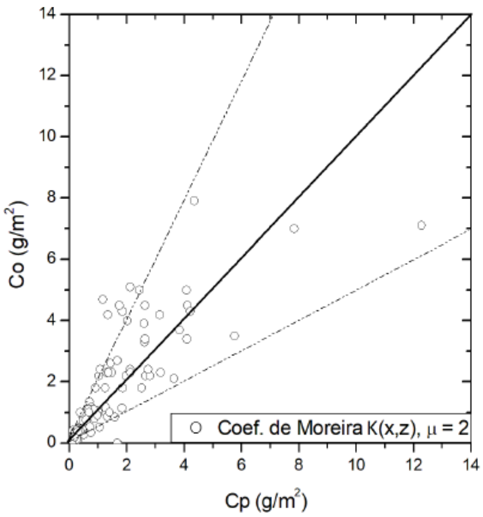

Figure 5. Data from observed concentrations (Co) in the experiment compared to Data from maximal concentration predicted (Cp) in case II scatter plot in case I. The first bisector indicates the agreement between the estimated and observed results.
Reference: Soledade et al. (2019)

The analysis of the scatter plots corroborates the results obtained by the statistical indices. Comparing Figures 4 and 5, it was observed that in case I (Figure 5) there is a greater number of points within a factor of two than in case III (Figure 4), which shows the influence of the memory effect on the solution.

6. CONCLUSIONS

For the Copenhagen experiment (relatively high source and conditions moderately convective atmospheric), the concentrations provided by the parameterized model with coefficient $K_z(x,z)$ indicated satisfactory results. This analysis is validated by statistical indices provided in Table 1 (Case I and II). Despite the good performance, it was not possible to confirm the influence of the memory effect, because when confronting the parameterized models with the coefficient $K_z(x,z)$ (cases I and II) with that parameterized with the coefficient $K_z(z)$ (cases III and IV), it can be seen that their statistical indices are very close.

With respect to the Prairie Grass experiment, it was possible to verify the influence of the memory. The concentration values obtained for the parameterized models with coefficient $K_z(x,z)$ proposed for low sources, under strongly convective conditions (MOREIRA et al. al., 2002), presented the best results when compared to the parameterized model with coefficient $K_z(z)$, as evidenced by the indices Table 2. Therefore, the concentrations obtained with the diffusion dependent on distance of source coefficients are better than the results achieved with coefficient $K_z(z)$, which suggests that the inclusion of memory effect, as predicted by the theory of Taylor, improves the description of the process involving the turbulent transport of a passive pollutant abandoned from a low, punctual and continuous source.

Thus, the analysis of the results reveals that the solution of the diffusion- advection methodology suggested in this paper, together with parameterizations that describe the physics of vertical non-homogeneous turbulence and dependent on the distance from the source, yielded good accuracy for the concentrations obtained in the experiments from Copenhagen and Prairie Grass, but the memory effect was only identified in low sources under strongly convective atmospheric conditions.

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