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APPLICATION OF A NEW APPROACH IN AN ANALYTICAL MODEL TO SIMULATE THE DISPERSION OF A RADIOACTIVE POLLUTANT

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ABSTRACT

The contribution of this work is focused on presenting a new approach to dispersion models of radioactive pollutants, that is, pollutants that undergo chemical reactions. The model uses the two-dimensional advection-diffusion-reaction equation to represent the dispersion of the pollutant in the Atmospheric Boundary Layer and to denote the chemical reaction that the pollutant suffers is included a source term in the advection-diffusion equation. This study presents a new analytical solution for the transient two-dimensional advection-diffusion-reaction equation by the combination of the methods of separating variables with the Generalized Integral Laplace Transform Technique (GILTT). The validation of the new model is made using experiment data carried out close to the site of Angra dos Reis nuclear power plant, in Brazil. The results of the three-dimensional pollutant concentrations are compared with the ones obtained by the GILTT method with a Gaussian assumption in the y direction (called here as GILTTG). The numerical results show that the predicted concentrations of the proposed model are close to the GILTTG concentrations, with the gain of not having to perform a numerical inversion in the time variable, resulting in a fast time response, which is very important in the prevention of environmental impacts.

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INTRODUCTION

Prediction models of radioactive substances in the atmosphere is a matter of great importance for the prevention of environmental impacts in the event of an accident. Emergency plans are formulated on the basis of possible air concentration scenarios, and therefore they use mathematical models of contaminant dispersion in the atmosphere, which are able to relate causes (sources) to relative effects (pollutant concentration). This type of problem is represented by the classical advection-diffusion equation (Seinfeld and Pandis, 1998). The major analytical solutions in the literature for the advection-diffusion equation are for very specific cases, generally considering constant or simple turbulent diffusivity coefficients, for some references see the work of (Moreira *et al*., 2009). However, many advances were obtained using the GILTT method (Wortmann *et al*., 2005; Moreira *et al*., 2006, Buske *et al*., 2012a, 2012b Vilhena *et al*., 2012). For the solution of partial differential problems, this integral transformation technique combines an expansion in series with an integration. In the expansion a trigonometric, obtained from an auxiliary Sturm-Liouville problem, is used.

Integration is done over the whole range of the transformed variable, taking advantage of orthogonality property of the basis used in the expansion. This procedure results in an ordinary differential equations system, which, once solved, is easily inverted to obtain the result of the original equation. The transformed problem is solved analytically by the Laplace transform technique and diagonalization. The final concentration is obtained with a numerical inversion (Gauss quadrature) in the time variable. In this work, the analytical solution obtained by the GILTT method is improved, and for this the two-dimensional non-stationary advection-diffusion equation is solved, avoiding the numerical inversion made in the previous works that use the GILTT method. The solution is obtained through a combination of the methods of separating variables and GILTT.

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The great advantage is that, thus, the final concentration of pollutants is obtained much more quickly and efficiently because there are no numerical inversions in the new solution. The article is organized as follows: in section 2, we present the proposed solution to the advection-diffusion-reaction equation; in section 3, we find the turbulent parameterizations used in the simulations; Section 4 briefly describes the experiment used and shows the numerical simulations obtained by the proposed methodology. The results are compared with the experimental data and the GILTTG model (model that uses a numerical inversion to obtain the final solution). Finally, in section 5, an analysis of the proposed methodology is made.

Solution of the Advection-Diffusion-Reaction Equation

In order to describe the concentration field of a radioactive pollutant, the advection-diffusion-reaction equation is used. This equation is the basis for the majority dispersion models of pollutants in the atmosphere and allows to investigate the influence of the turbulent parameters in the concentration. In this work, we use the advection-diffusion-reaction equation in the transient two-dimensional form, shown below:

$$\frac{\partial C}{\partial t}+u\frac{\partial C}{\partial x} = \frac{\partial}{\partial z}\left(K_z\frac{\partial C}{\partial z}\right)+S \qquad \text{.....(1)}$$

where $C(x,z,t)$ is the mean pollutant concentration, u is the mean velocity component in the longitudinal direction, K_z is the turbulent diffusion coefficient in the vertical direction and S is the source term (representing the chemical reaction of the pollutant). In the proposed problem, it is considered that the pollutant undergoes a first order reaction, thus, the term is represented by:

$$S=-\lambda C(x,z,t) \qquad \text{.....(2)}$$

Being λ the decay constant of the pollutant under analysis in the model.

The problem under study is subject to the following boundary and initial conditions, given below:

Boundary condition in z:

$$K_z\frac{\partial C}{\partial z}=0 \qquad \text{at } z=0\ ,\ h \qquad \text{.....(3.a)}$$

Source condition:

$$u\,C(t,0,z)=Q(t)\,\delta(z-H_s) \qquad \text{.....(3.b)}$$

Initial condition:

$$u\,C(0,x,z)=Q(0)\delta(x)\,\delta(z-H_s) \qquad \text{.....(3.c)}$$

where h is the height of the boundary layer, H_s is the height of the source and Q the emission pollutant rate.

In order to obtain the solution, a variable separation (Özisik, 1993) is applied, as $C(x,z,t)=X(x)Z(z)T(t)$. Replacing this function in equation (1) and separating the variables, we obtain the following system of ordinary differential equations (ODE):

$$\frac{1}{T}\frac{dT}{dt}=-\nu, \qquad \text{.....(4)}$$

$$u\frac{1}{X}\frac{dX}{dx}=\eta, \qquad \text{.....(5)}$$

$$\frac{1}{Z}\frac{d}{dz}\left(K_z\frac{dZ}{dz}\right)=-\lambda+\nu-\eta. \qquad \text{.....(6)}$$

By rewriting the eigenvalues (ν and η) in a more convenient way, and solving the ODE's (4) and (5), we have as solutions:

$$T_\nu(t)=e^{-\nu t}, \qquad \text{.....(7)}$$

$$X_{\alpha,\nu}=e^{\frac{\alpha+\nu}{u}x}=e^{\frac{\alpha}{u}x}e^{\frac{\nu}{u}x}=X_\alpha X_\nu, \qquad \text{.....(8)}$$

in which α = η - ν. The solution to equation (6) is obtained by the GILTT method, a well-known technique in the literature (Moreira *et al*., 2009).

Thus, one can rewrite the solution by grouping terms that have the same eigenvalues:

$$C_{\alpha,\nu}(x,z,t) = [T_\nu(t)X_\nu(x)][X_\alpha(x)Z_\alpha(z)]. \quad \text{(9)}$$

Note that each factor of equation (9), can be defined as:

$$\varphi_\nu(x,t) = T_\nu(t)X_\nu(x) = e^{-\nu t}e^{\frac{\nu}{u}x} = e^{-\frac{\nu}{u}(ut-x)}, \quad \text{(10)}$$

$$\phi_\alpha(x,z) = X_\alpha(x)Z_\alpha(z), \quad \text{(11)}$$

where $\phi_\alpha(x,z)$ is the solution obtained by the equation:

$$u\frac{\partial\phi_\alpha(x,z)}{\partial x} = \frac{\partial}{\partial z}\left(K_z\frac{\partial\phi_\alpha(x,z)}{\partial z}\right) - \lambda\phi_\alpha(x,z). \quad \text{(12)}$$

The concentration for each eigenvalue is:

$$C_{\alpha,\nu}(x,z,t) = \varphi_\nu(x,t)\phi_\alpha(x,z). \quad \text{(13)}$$

Since the domain at x is infinite, the eigenvalue ν is continuous, therefore the final concentration is given by:

$$C_{\alpha,\nu}(x,z,t) = \left(\int_0^\infty B(\nu)\varphi_\nu(x,t)d\nu\right)GILTT_\alpha(x,z), \quad \text{(14)}$$

Note that the eigenvalue ν (1/s) is considered continuous in [0,∞), so the final concentration is expressed as:

$$C(x,z,t) = \varphi(x,t)\,GILTT(x,z), \quad \text{(15)}$$

being:

$$\varphi(x,t) = \int_0^\infty B(\nu)\varphi_\nu(x,t)d\nu \quad \text{(16)}$$

$$GILTT(x,z) = \sum_{n=0}^{\infty} GILTT_{\alpha_n}(x,z)d\alpha_n \quad \text{(17)}$$

To determine the functions φ(x,t) and GILTT(x,z), conditions (3a) to (3c) are used.

Applying the source condition (3b) in the final solution (15), yields:

$$u\,C(t,0,z) = Q(t)\,\delta(z-H_s) = u\varphi(0,t)GILTT(0,z). \quad \text{(18)}$$

In equation (18), it can be established the following relations:

$$\varphi(0,t) = Q(t), \quad \text{(19)}$$

$$GILTT(0,z) = \frac{\delta(z-H_s)}{u}. \quad \text{(20)}$$

To determine the constant *B(ν)*, is used:

$$\varphi(x,t) = \int_0^\infty B(\nu)\varphi_\nu(x,t)d\nu = \int_0^\infty B(\nu)e^{-\frac{\nu}{u}(ut-x)}d\nu, \quad \text{(21)}$$

and by the source condition equation (19), follows:

$$Q(t) = \int_0^\infty B(\nu)e^{-\nu t}d\nu = L\{B(\nu);\nu\to t\}. \quad \text{(22)}$$

To obtain *B(ν)* the inverse Laplace transform is applied in equation (22):

$$L^{-1}\{Q(t);t\to\nu\} = B(\nu) \quad \text{(23)}$$

The constant *B(ν)* in equation (21) is replaced, resulting in:

$$\varphi(x,t) = \int_0^\infty L^{-1}\{Q(t)\}e^{\frac{\nu}{u}x}e^{-\nu t}d\nu = Q\left(t-\frac{x}{u}\right) \quad \text{(24)}$$

Therefore, the final solution is:

$$C(x,z,t) = Q\left(t - \frac{x}{u}\right)\sum_{n=0}^{\infty}\phi_{\alpha_n}(x,z), \quad \text{.....(25)}$$

Where the function $\phi_{\alpha_n}(x,z)$ is obtained applying the GILTT method (Moreira *et al*., 2009). Thus, the final solution is well determined.

Application of the source in the problem

To obtain the solution of the proposed problem, the Green solution for equation (1) will be presented. With the known Green solution, it is possible to find solutions for different forms of sources depending on the time (Q(t)), and the principle of superposition guarantees solutions for different types of source, since the problems are linear operators.

Initially, to determine the Green solution, it is considered a source with Dirac delta distribution, that is, $Q(t) = Q_c\delta(t)$. Using this type of source in equation (25), we have:

$$C(x,z,t) = Q_c\delta\left(t - \frac{x}{u}\right)\sum_{n=0}^{\infty}\phi_{\alpha_n}(x,z). \quad \text{.....(26)}$$

A highly concentrated function is considered:

$$\delta(t_1) = \lim_{b\to 0}\frac{1}{\sqrt{\pi b}}e^{-\frac{t_1^2}{b}}, \quad \text{.....(27)}$$

applying $t_1 = t - \frac{x}{u}$, one obtains:

$$\delta\left(t - \frac{x}{u}\right) = \lim_{b\to 0}\frac{1}{\sqrt{\pi b}}e^{-\frac{\left(t-\frac{x}{u}\right)^2}{b}} = \lim_{b\to 0}\frac{1}{\sqrt{\pi b}}e^{-\frac{(ut-x)^2}{bu^2}}. \quad \text{.....(28)}$$

Assuming that $b = \frac{4K_x t}{u^2}$ and doing $K_x \to 0$, one can rewrite equation (28) as follows:

$$\delta\left(t - \frac{x}{u}\right) = \lim_{K_x\to 0}\frac{u}{\sqrt{4\pi K_x t}}e^{-\frac{(ut-x)^2}{4K_x t}}. \quad \text{.....(29)}$$

Therefore, the final solution is:

$$C(x,z,t) = \lim_{K_x\to 0}\frac{Q_c u}{\sqrt{4\pi K_x t}}e^{-\frac{(ut-x)^2}{4K_x t}}\sum_{n=0}^{\infty}\phi_{\alpha_n}(x,z). \quad \text{.....(30)}$$

In order to find solutions for different types of time-dependent sources, is considered $\delta(t-\tau)$ in equation (29):

$$\delta\left(t - \tau - \frac{x}{u}\right) = \lim_{K_x\to 0}\frac{u}{\sqrt{4\pi K_x t}}e^{-\frac{(u(t-\tau)-x)^2}{4K_x t}}. \quad \text{.....(31)}$$

Ready, Green's solution is:

$$C(x,z,t,\tau) = \lim_{K_x\to 0}\frac{Q_c u}{\sqrt{4\pi K_x t}}e^{-\frac{(u(t-\tau)-x)^2}{4K_x t}}\sum_{n=0}^{\infty}\phi_{\alpha_n}(x,z). \quad \text{.....(32)}$$

For the experiment addressed in this work, the emission source is continuous and represented by the following function:

$$Q(t) = \begin{cases} 0, & if \quad t = 0 \\ Q_c, & if \quad t > 0 \end{cases} \quad \text{.....(33)}$$

being Q_c the constant emission rate. Applying the source (33) in the Green solution (32), one comes to:

$$C(x,z,t) = \frac{Q_c}{2}\left[erf\left(\frac{1}{\sqrt{4K_x t}}(ut - ut_0 - x)\right) - erf\left(\frac{1}{\sqrt{4K_x t}}(ut - ut_1 - x)\right)\right]\sum_{n=0}^{\infty}\phi_{\alpha_n}(x,z). \quad \text{.....(34)}$$

Turbulent Parameterizations

In this work, to calculate the three-dimensional concentration, it is assumed that in the lateral direction the concentration is dispersed in the form of a Gaussian distribution. Thus, the three-dimensional concentration must take into account the dispersion parameter σ_y. Therefore, the concentration at the ground level (z = 0) is calculated by the following expression:

$$C(x,0,0,t) = \frac{C(x,0,t)}{\sqrt{2\pi}\sigma_y} \qquad \text{.....(35)}$$

where the concentration *C(x,0,t)* is calculated by equation (34) and for the lateral dispersion parameter σ_y, for small variations in the wind direction in relation to the position of the receivers, the expression presented by (Degrazia, 1998) is used:

$$\frac{\sigma_y^2}{h^2} = \frac{0.21}{\pi}\int_0^\infty \sin^2\left(2.26\psi^{1/3}X_{n'}\right)\frac{dn'}{(1+n')^{5/3}n'^2}, \qquad \text{.....(36)}$$

where $X = \frac{xw^*}{uh}$ is the nondimensional distance and $\psi^{1/3} = 0.97$ the dissipation function. For the case of great variation of the wind direction is considered the following expression for σ_y:

$$\frac{\sigma_y}{x} = \sigma_\theta\left(1+0.031x^{0.46}\right)^{-1}, \qquad \text{.....(37)}$$

where $\sigma_\theta = 15$ for unstable conditions (Blackadar, 1997).

In order to compare the approach presented in this work with the classical GILTTG technique, the same parameters of the turbulent diffusion coefficients and wind profile of Weymar (2012) will be used. By this way, the parameterizations of the vertical and longitudinal diffusion coefficients (Degrazia *et al*., 1997) are represented by the following expressions:

$$\frac{K_z}{w_* h} = 0.22\left(\frac{z}{h}\right)^{1/3}\left(1-\frac{z}{h}\right)^{1/3}\left[1-\mathrm{e}^{\left(-\frac{4z}{h}\right)}-0.0003\,\mathrm{e}^{\left(\frac{8z}{h}\right)}\right] \qquad \text{.....(38)}$$

$$K_x = \frac{0.583\mathrm{h}\;\mathrm{c}_i fi_{13}^2 w_* X\left(\frac{z}{h}\right)^{4/3}\left(1.03\sqrt{c_i}\,fi_{13}fm_*^{2/3}X+0.55\left(\frac{z}{h}\right)^{2/3}\right)}{\left(2.06\sqrt{c_i}\,fi_{13}fm_*X+0.55fm_*^{1/3}\left(\frac{z}{h}\right)^{2/3}\right)^2} \qquad \text{.....(39)}$$

where fi_{13} is the reduced maximum frequency, fm_* is the normalized frequency of the spectral peak and $c_i = 0.3$.
For the wind field the power profile is used (Panofsky and Dutton, 1984):

$$u_z = u_1\left(\frac{z}{z_1}\right)^n \qquad \text{.....(40)}$$

in which u_z and u_1 are the mean horizontal velocities of the wind at z and z_1 heights, and α is an exponent that is related to the turbulence intensity (Irwin, 1979).

Experimental data and Numerical Results

For model validation was used a controlled release of radioactive material performed in 1985 at the Itaorna Beach, close to the nuclear reactor site Angra dos Reis in the Rio de Janeiro state, Brazil. Details of the dispersion experiment is described by Biagio *et al*. (1985). The experiment consisted in a controlled releases of radioactive tritium loaded water vapor from a meteorological tower of 100m height during five days (November 28 to December 4, 1984). During the whole experiment, four meteorological towers collected the relevant meteorological data. Wind speed and direction were measured at three levels (10m, 60m, and 100m) together with the temperature gradients between 10m and 100m. Some additional data of relative humidity were available in some of the sampling sites, and were used to calculate the concentration of radioactive tritium loaded water in the air (after measuring the radioactivity of the collected samples). All relevant details, as well as the synoptic meteorological conditions during the dispersion campaign are described in (Biagio *et al*., 1985). The data from the 5 experiments were used to obtain the numerical results and are presented in Table 1. The total time of emission was of 90min for each experiment, being all the cases around noon. Water vapor collection was performed on aluminum plates in numbered locations (according to Figure 1) in three subsequent periods of 20min each, 30min after the start of the release. The same meteorological data were used in the work of Weymar (2012). In Table 1, u_{10} is the reference velocity at 10 meters in height (m/s), u_* represents the friction velocity (m/s), w_* is the vertical convective velocity scale (m/s), h is the boundary layer height (m) and Q is the emission rate of the source (MBq/s). Figure 2 shows the scatter plots of predicted concentrations, for the GILTTG model (with numeric inversion) and the proposed approach (without numeric inversion), with the observed concentrations for the 5 experiments.

Table 1. Meteorological parameters and emission rate for the Angra dos Reis experiment (Biagio *et al*., 1985)

Exp.	Period	$u(10)$ (m/s)	u_* (m/s)	w_* (m/s)	h (m)	Q (MBq/s)
1	1	1.83	.32	.46	965.09	20.46
	2	2.43	.42	.60	1259.98	20.46
	3	2.76	.48	.69	1447.64	20.46
2	1	2.59	.44	.63	1321.64	25.34
	2	2.21	.38	.55	1152.75	25.34
	3	2.18	.38	.54	1133.98	25.34
3	1	2.21	.38	.55	1152.75	20.46
	2	1.97	.34	.49	1026.75	20.46
	3	2.61	.46	.66	1367.21	20.46
4	1	1.23	.21	.31	643.40	24.34
	2	1.01	.18	.25	525.44	24.34
	3	1.05	.18	.26	544.21	24.34
5	1	1.95	.34	.49	1018.71	31.32
	2	1.54	.27	.39	804.24	31.32
	3	2.61	.45	.65	1356.49	31.32

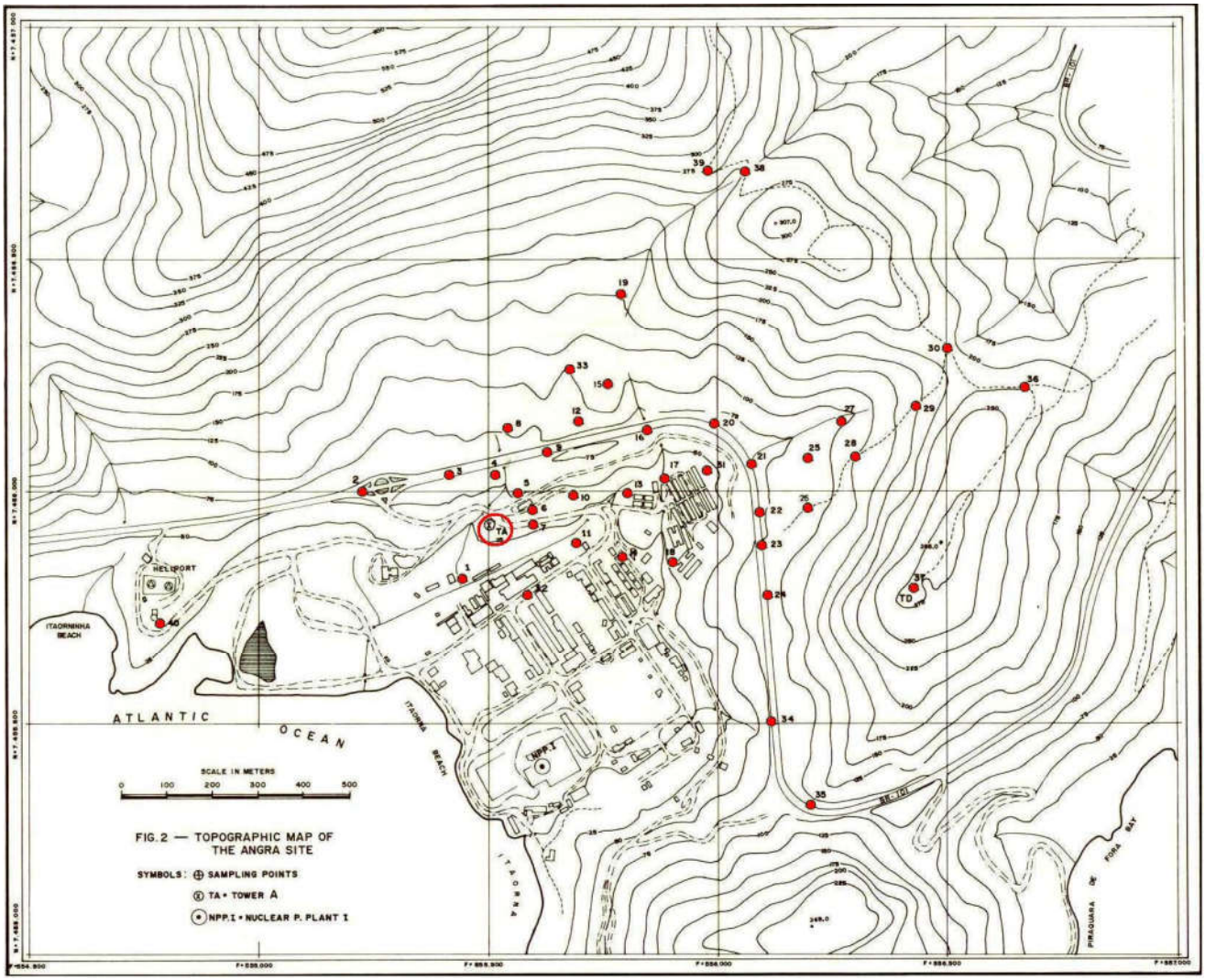

Figure 1. Topographic map of the Angra dos Reis experiment (Biagio *et al*., 1985)

The ideal for the model is that the generated concentration is the same as the measurement experimentally. Thus, the points of this graph would be on the straight identity. It is noticed in the scatter plots that the concentrations generated by the proposed model are very close to the concentrations generated by the GILTTG model, in some cases they have the same values. The present model (without numerical inversion) can reproduce results very close to the GILTTG model (with numerical inversion) and in a much faster way. In order to verify the proximity of the predicted concentrations of the models with and without numerical inversion, the graphs of the absolute and relative errors for each of the concentrations are presented in Figure 3. From the graphs it is noticed that in some cases the absolute and relative errors are equal to zero, showing that the concentrations are the same, in general the errors made are considered acceptable. With the results presented in the graphs, we obtain an average absolute error of approximately 4% and an average relative error of 10%, showing a compatibility between the models. To make a statistical comparison of the models with the experimental data, the following statistical indices are used: normalized mean square error (NMSE); correlation

coefficient (COR); factor of two (FAT2); factor of five (FAT5); fractional bias (FB) and standard fractional deviation (FS), described in Hanna (1989). Table 2 shows the results of the statistical indices for the two models in experiments 1-2; 3; 4; 5 and all experiments.

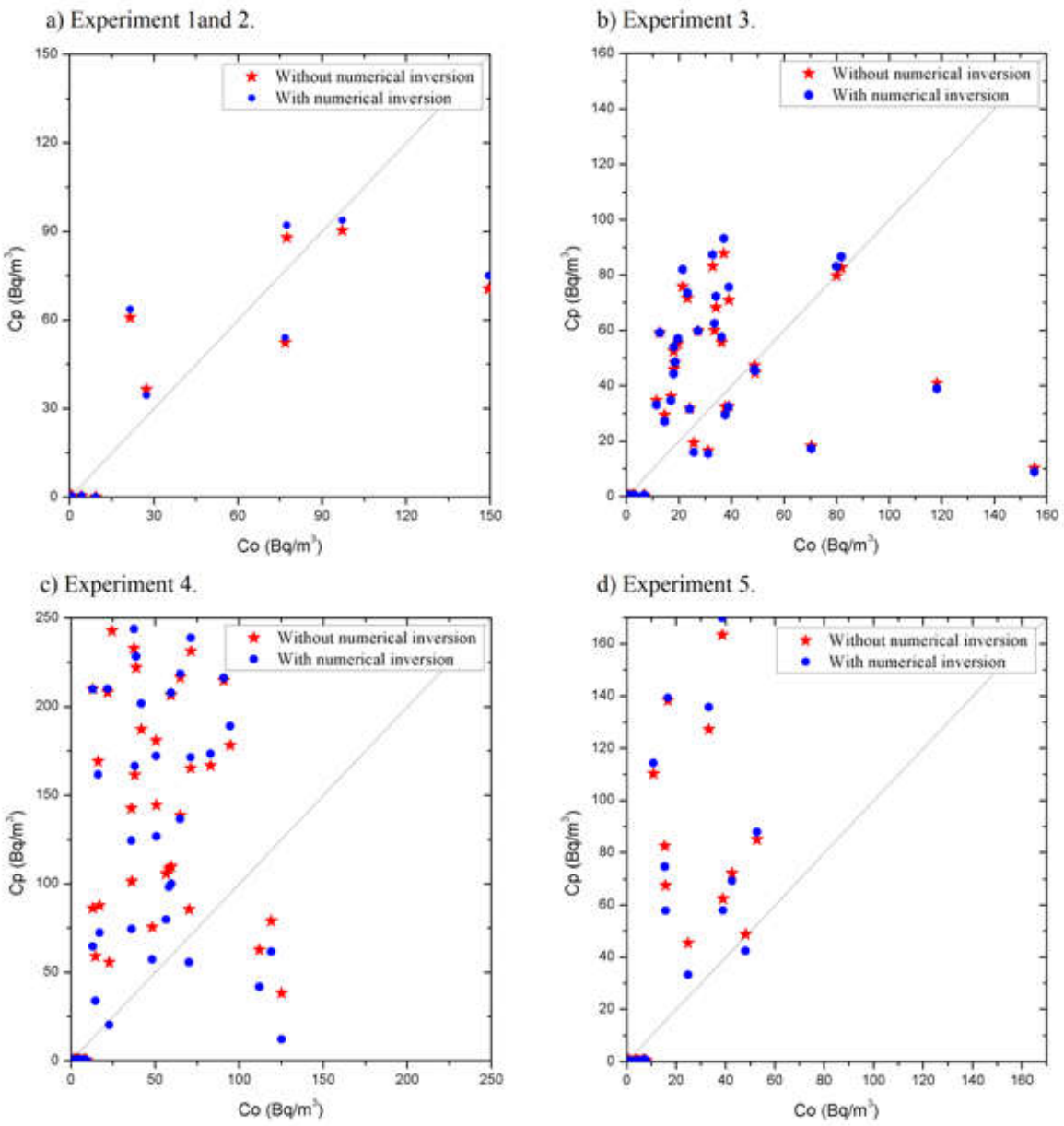

Figure 2. Scatter plot of the methods without (present) and with numerical inversion (GILTTG). In the graphs the predicted concentrations generated by the methods are compared with the observed concentration for the five experiments

Table 2. Results of statistical indexes of models with numerical inversion (GILTTG) and without numerical inversion (present).

Exp.	model	*NMSE*	*COR*	*FAT2*	*FAT5*	*FB*	*FS*
2-Jan	Present	0.96	0.87	0.62	0.9	0.15	0.25
	GILTTG	0.88	0.88	0.67	0.86	0.12	0.21
3	Present	1.25	0.35	0.29	0.76	-0.18	0.09
	GILTTG	1.31	0.33	0.31	0.76	-0.2	0.04
4	Present	2.34	0.42	0.2	0.68	-0.9	-0.83
	GILTTG	2.53	0.37	0.2	0.66	-0.86	-0.88
5	Present	4.45	0.7	0.26	0.42	-0.84	-1.04
	GILTTG	4.69	0.68	0.26	0.45	-0.83	-1.05
All	Present	2.66	0.49	0.3	0.68	-0.65	-0.67
	GILTTG	2.82	0.46	0.32	0.67	-0.62	-0.69

Again, the agreement between GILTTG (with numerical inversion) and the proposed model (without numerical inversion). The results of the statistical indexes of the models are very close to each other, both in the cases of individual experiments and in the case considering all experiments. It is also noticed that in some cases the indexes become equal. It is worth mentioning once again that the proposed model is able to reproduce the predicted concentrations of the GILTTG model (this model uses a numerical inversion to obtain the final solution) with the great advantage of not having to do a numerical inversion, by this way, making that the proposed model has a much faster response, which is of great value in mitigating actions in the event of an accident.

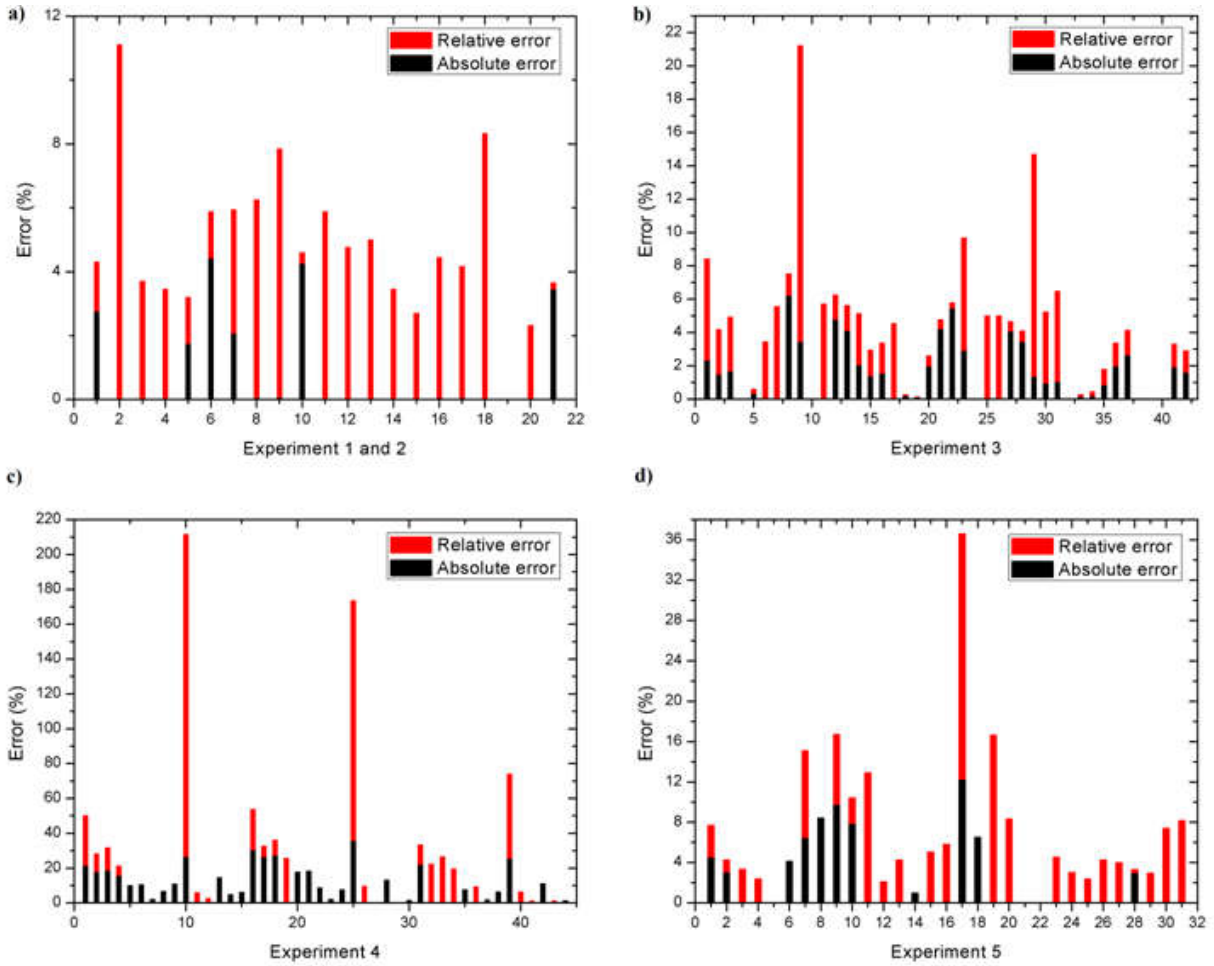

Figure 3. Comparison between the methods without (present) and with (GILTTG) numerical inversion. The graphs show the absolute and relative errors for the five experiments

Conclusion

With this work, it can be said that the proposed model is an evolution of the analytical models that use the GILTT method, because the presented methodology uses the GILTT technique and improves its performance. In addition, the results presented by the proposed model represent, in a very similar way, the results of the GILTTG model. It should be noted that the final solution obtained in this work does not have to make a numerical inversion in the time variable, different from the previous works.
The methodology developed in this work shows that the presented technique can also be applied in the advection-diffusion-reaction equation, that is, it extends its application to problems involving first-order chemical reactions.

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