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ESTIMATION OF THE DEPOSITION RATES OF ATMOSPHERIC POLLUTANTS USING PARTICLE SWARM OPTIMIZATION

Roseane A.S. Albani

IPRJ/UERJ

roseanealves75@gmail.com

Vinicius V.L. Albani

Departamento de Matemática/UFSC

v.albani@ufsc.br

Alfredo Gamboa

IPRJ/UERJ

alfredo.soliz@iprj.uerj.br

Davidson Moreira

SENAI-CIMATEC

davidson.moreira@gmail.com

Antônio J. Silva Neto

IPRJ/UERJ

ajsneto@iprj.uerj.br

Abstract. Dry deposition removal process is a major environmental concern, especially because the material deposited over a surface may react with underlying substances producing potential harmful substances. Dry deposition rates can be obtained from observational data, though it may be a hard task. The computational modeling using atmospheric dispersion models can be an interesting alternative to the experimental investigation. In this work we propose a methodology based on inverse problems techniques to estimate the dry deposition rate of a pollutant from observed concentrations. More precisely, an objective function, that evaluates the discrepancy between observed and model concentrations, is minimized with respect to the deposition rates. We consider two analytical dispersion models to evaluate the concentrations that account for the deposition rates. The proposed methodology is tested with datasets from the Hanford field experiment and the results are compared with deposition rates obtained in the literature. As we shall see, the rates estimated using both models were close to the literature results and the model concentrations were adherent to the observed data.

Keywords: Dry deposition, Atmospheric Pollutant Transport, Inverse problems, Particle Swarm Optimization.

1. INTRODUCTION

The atmosphere continuously receives gases and particulate matter from different sources, being posteriorly removed mainly due to dry or wet deposition mechanisms. The dry deposition removal is a major environmental issue since the drop of gases and particulate matter over the earth's surface might react with other species, producing potentially harmful compounds which might threaten the underlying ecosystems in many ways. Investigating the geographical distribution and magnitude of the deposition of atmospheric pollutants is essential to decide which regions will be affected by their damaging effects, allowing the establishment of pollutant control measurements. The determination of the deposition rates can be performed using inverse problems modeling. This includes the definition of the forward problem, which is a mathematical description of the dispersion processes related, commonly represented by an advection-diffusion partial differential equation (PDE). Also, it is necessary to provide observational data and define a technique to solve the inverse problem. Deposition rates come up in one of the terms of the advection-diffusion equation or as a flux boundary condition. However, by the nonlinear nature of the estimation process, the adjoint advection-diffusion PDE is not suitable to model the forward problem implying a significant increase in the computational cost of the inverse problem solution. In this case, the forward problem needs to be solved for each iteration of the inversion procedure. To avoid such an issue, analytical solutions for the advection-diffusion equation or Gaussian plume-depletion models might be a reasonable alternative to

describe the forward problem since they are usually simpler and faster than numerical solutions.

Particle swarm optimization have been applied to identification parameters of atmospheric dispersion models, mostly, to source parameter estimation, see for reference (Albani *et al.*, 2020). Optimization techniques are based on the minimization of an objective function, which is composed by a data misfit term that evaluates the discrepancy between measured and observed concentrations. The set of estimated parameters consist of the minimizers of the objective function.

The problem of source identification has received much attention in last decades and a number of inversion techniques were proposed, many of them using deterministic models, as mentioned above, based on least-squares or Tikhonov-type regularization. Other authors proposed a series of stochastic techniques that are mainly based on Bayesian inference. See, for example Albani and Albani (2019); Albani *et al.* (2020); Albani and Albani (2020); Albani *et al.* (2021), and references therein.

There are several methods to minimize the objective function, including the broad class of Metaheuristic techniques. Metaheuristic optimization methods such as the *Particle Swarm Optimization* (PSO), *Genetic Algorithms*(GA) , and *Simulated Annealing*, to refer to some popular techniques to the estimation of atmospheric dispersion parameters, are global search methods. Thus, algorithms based on these methods are less likely to fall on local minima points. Several optimization methodologies have been applied to the source estimation parameters of atmospheric emissions. PSO was applied to minimize a Tikhonov-type regularization functional in Ma *et al.* (2017) to identify source parameters. Ma *et al.* (2018) compares the performance of PSO and two other swarm intelligence optimization algorithms, the *Ant Colony Optimization Algorithm* and the *Firefly Algorithm* to retrieve the source strength and spatial coordinates. Wang *et al.* (2020) applied a hybrid strategy combining GA, PSO and SA to improve the accuracy and efficiency of the optimization process. (Albani *et al.*, 2020) applied a combination of two Metaheuristic optimization techniques, namely, the PSO and GA methods, with the gradient descent algorithm to estimate the source parameters of multiple atmospheric releases.

We propose a methodology based on analytical dispersion models associated to the Particle Swarm Optimization (PSO) technique to estimate the dry deposition rates of an atmospheric pollutant. We simulate a field experiment to evaluate the proposed methodology. The PSO calculates the minimizers of the corresponding Tikhonov-type functional.

2. MATHEMATICAL MODEL FOR THE DEPOSITION RATE ESTIMATION

In this work, we propose an inverse modeling to estimate the dry deposition rates using a combination of analytical models and the PSO technique. The estimation procedure consists on the minimization of a objective function using the PSO. The objective function accounts on the discrepancy between the observed or experimental data and the analytical model for the concentration distribution. Next, we describe briefly these methodologies.

2.1 Inverse Modeling Technique

The dry deposition rates must be estimated from the observed concentrations. To address this, a inverse modeling technique must be applied. Let \mathbb{C}_{exp} denote the vector containing the set of experimental concentrations observed at (x_i, z_i) , $i = 1, \dots, n$, $\text{bf} = [v_1, \dots, v_n]^T$ denote the set of dry deposition rates, and $C(\text{bf})$ denote the concentration given by some of the analytical models considered in this work. Thus, we must find \mathbf{v} such that

$$\mathbb{C}_{\text{exp}}(x_i, z_i) = C(x_i, z_i; v_i), \quad i = 1, \dots, n. \quad (1)$$

Eq. (1) is represents the so-called inverse problem (IP) of estimating the dry deposition rates from observed concentrations. The IP is solved by least-squares, i.e., by finding the minimizers of the functionals

$$\mathcal{F}_i(v_i) = (C(x_i, z_i; v_i) - \mathbb{C}_{\text{exp}}(x_i, z_i))^2, \quad i = 1, \dots, n. \quad (2)$$

The minimization of \mathcal{F}_i will be performed by PSO.

PSO belongs to the category of swarm intelligence methods. Its predecessors tried to reproduce the social behavior of flocks of birds and schools of fish considering some features, such as the nearest neighbor, velocity synchronization, optimal distance between individuals, and a sudden change in the direction of the group. The main objective of PSO is to provide information exchange between the group of individuals (or swarm) considering the aforementioned principles. PSO is considered a robust metaheuristic optimization algorithm with a simple computational implementation. The PSO accuracy is well documented, with applications in different problems arising in many areas in science and engineering. Moreover, they are simple to implement, with a vast number of pre-programmed codes available in different computing languages. For a precise description of these techniques the reader can look for the references (Echevarría *et al.*, 2019) and (Gendreau and Potvin, 2010).

2.2 Analytical models

This work use two analytical models to describe the concentration distribution, which accounts on the dry deposition process. The main advantage of using analytical models for the estimation of dry deposition rates is simplicity of the

solution that results in a lower computational time. A classical solution, evaluated with experimental data in (Doran and Horst, 1985) and the analytical solution for the atmospheric diffusion equation proposed by (Lin and Hildemann, 1997) are applied to be used as the forward model for the inverse problem of obtaining the dry deposition rates. The Doran and Horst (DH) model is the following:

$$C_{DH}(x, z, h) = \frac{Q}{\sqrt{2\pi}} \exp\left(-\frac{w(z-h)x}{u\sigma_z^2} - \frac{w^2x^2}{2u^2\sigma_z^2}\right) \times \left[\exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right)\right] \left(1 - 2\sqrt{2\pi} \frac{Vx}{u\sigma_z} \exp(\xi) \operatorname{erf}(\xi)\right) \quad (3)$$

where

$$V = v_d - \frac{w}{2} \quad \text{and} \quad \xi = \frac{z+h}{\sigma_z\sqrt{2}} + \frac{\sqrt{2}Vx}{u\sigma_z},$$

Q [g/s] is the pollutant emission rate, h [m] is the source height, w [m/s] is the settling velocity, σ_z [m] is the vertical dimension of the plume, which is a function of the downwind distance and the atmospheric stability, and v_d [m/s] is the dry deposition velocity, or dry deposition rate. Finally, u is the wind speed magnitude, given as (Panofsky and Dutton, 1983)

$$u = \frac{u_*}{\kappa} \left[\left(\ln \frac{z}{z_0} \right) + 5z/L \right] \quad (4)$$

where u_* [m/s] is the friction velocity, L [m] is the Monin-Obukhov length, z_0 [m] is the surface roughness length and κ is the Von Kármán constant.

The Lin and Hildemann (LH) model is the following:

$$C_{LH}(x, z) = G_z(x_s, z_s; x, z) - \frac{v_d}{b} \exp\left(\frac{v_d}{b} z_s\right) \int_{z_s}^{\infty} \left[\frac{2}{\sqrt{4\pi ab(x-x_s)}} \exp\left(\frac{a(z+\xi)^2}{4b(x-x_s)}\right) \right] \exp\left(-\frac{v_d}{b} \xi\right) d\xi, \quad (5)$$

where (x_s, z_s) are the source coordinates in the xz -plane, again, v_d is the dry deposition rate,

$$G_z(x_s, z_s; x, z) = \frac{1}{\sqrt{4\pi ab(x-x_s)}} \left[\exp\left(-\frac{a(z-z_s)^2}{4b(x-x_s)}\right) + \exp\left(-\frac{a(z+z_s)^2}{4b(x-x_s)}\right) \right]$$

is the Green's function of the Gaussian plume for the total reflection,

$$a = \frac{u(z_r)}{z_r^\alpha}, \quad \text{and} \quad b = \frac{K_z(z_r)}{z_r^\beta}. \quad (6)$$

where $u(z_r)$ [m/s] and $k(z_r)$ [m²/s²] are the wind magnitude and the vertical turbulent diffusion respectively, calculated in a reference height, α and β are constants that depend on the atmospheric stability conditions (Lin and Hildemann, 1997).

3. A CASE STUDY

In this work we use the dual-tracer datasets from the Hanford experiment to evaluate the proposed methodology to estimate dry deposition rates. During the Hanford tracer experiment, two tracer gases, one depositing, the Zinc Sulfide (ZnS), and another non-depositing, the Sulfur Hexafluoride (SF₆) were released simultaneously from a height of 2 m. A diagram representing the plume depletion experiment grid can be found in Fig. 1.

The concentrations of the two tracer gases were measured over sampling units arranged in five concentric arcs at $x = 100, 200, 800, 1600$ and 3200 m from the emission source, 1.5 m above the ground level. Figure 2 shows a diagram from the grid of the Hanford Experiment considering also the emission source located at the coordinate (0, 0) remarked as the black dot. A meteorological tower was located nearby 100 m to the north of the point of release to measure wind speed, wind direction and temperature. A detailed documentation of this experiment can be found in (Nickola, 1977). The ZnS crosswind-integrated concentration data, normalized by the release rates (Q), the meteorological data and the effective dry deposition rates are listed in Tab. 1. Because explicit measurements of the deposition flux was not available, since it was impractical, we use the effective dry deposition rates proposed in Doran and Horst (1985).

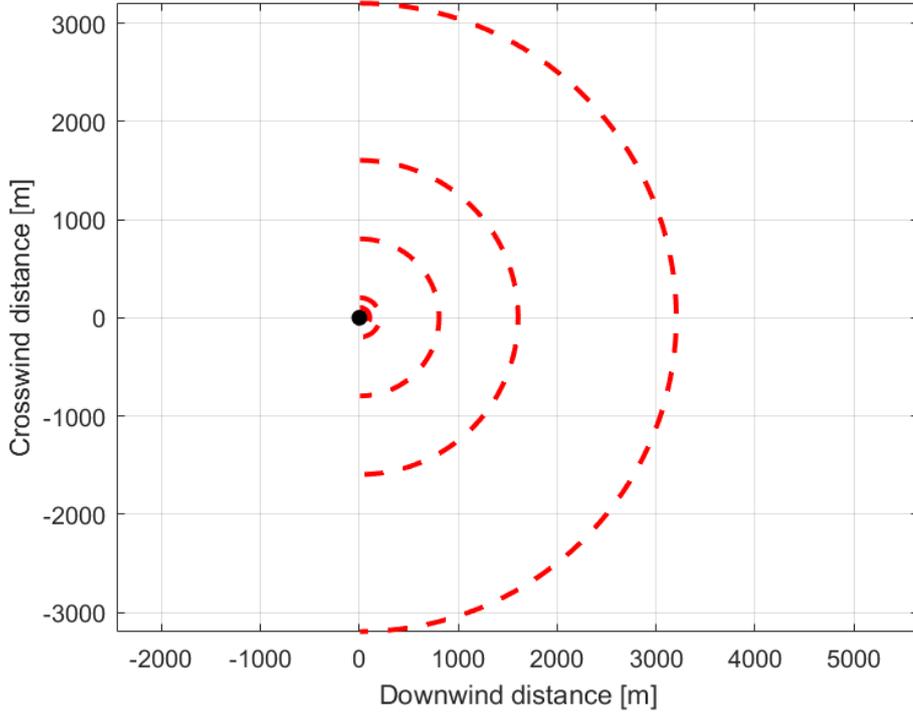


Figure 1: Diagram representing the plume depletion experiment grid. Adapted from (Doran and Horst, 1985).

4. RESULTS

As we mentioned before, the inverse problem is structured as the minimization of the functional in Eq. (2) by the PSO technique. The analytical concentration models in Eq. (3) and Eq. (5) are considered in the estimation jointly with the ZnS crosswind-integrated concentrations from Tab. 1. In other words, \mathbb{C}_{exp} in Eq. (2) represents the ZnS crosswind-integrated concentrations.

The estimation of the dry deposition rates using the DH model in Eq (3) is performed directly, as all the parameters in analytical formula for the concentrations are known. However, the estimation considering the LH model is more complex, as the parameters α and β in the definition of a and b in Eq. (6) are not known *a priori*. Thus, to achieve better performance, we added a and b to the set of unknown parameters that must be estimated. Thus, for the LH model, we shall estimate v_d , a and b , v_d depends on the downwind distances and a and b are constant with respect to x (downwind distance) and z (height). As the number of ZnS crosswind-integrated concentrations is small (3 for each set of experiments) and the number of unknowns is larger than the dataset, we perform the estimation in two steps:

1. Assume that v_d is constant for all the downwind distances and estimate v_d , a , and b by minimizing, using PSO, the Tikhonov functional (Tikhonov and Arsenin, 1998):

$$\mathcal{F}(a, b, v_d) = \sum_{j=1}^3 (C(x_j, z; a, b, v_d) - \mathbb{C}_{\text{exp}}(x_j, z))^2 + 10^{-6} (a^2 + b^2 + v_d^2),$$

with (a, b, v_d) varying in the set $[10^{-3}, 15] \times [10^{-3}, 15] \times [10^{-3}, 1.0]$ and $C(x_j, z; a, b, v_d)$ is the concentration given in Eq (5) at (x_j, z) , using the parameters a , b and v_d . x_j , with $j = 1, 2, 3$ represents the downwind distances from the source and assumes the values 800, 1600 and 3200m.

2. Use the estimated values for the parameters a and b in the formula given in Eq (5) and estimate the values of v_d , considering the ZnS crosswind-integrated concentrations for the arcs in $x = 800, 1600$ and 3200 m from the emission source. In this step, the functional in Eq. (2) is minimized.

This strategy of minimization is inspired by the splitting strategy in the estimation of multiple parameters and used in Albani *et al.* (2020); Albani and Zubelli (2020). We do not use in the estimation the ZnS crosswind-integrated concentrations given in the arcs in $x = 100$ and 200 m.

The PSO function used in this work is from the global optimization toolbox of MATLAB, with the settings below:

- The PSO algorithm is initialized considering the values for the particles in the range 10^{-3} to 1.

Experiment	Meteorological Data	Downwind Distance (Arc) [m]	C/Q [s/m ²]	v_d [m/s]
Exp 1	$L = 166$ m $u_* = 0.4$ m/s	100	0.0132	-
		200	0.00647	-
		800	0.00224	0.0421
		1600	0.000982	0.0405
		3200	0.000586	0.0365
Exp 2	$L = 44$ m $u_* = 0.26$ m/s	100	0.0389	-
		200	0.0224	-
		800	0.00747	0.0193
		1600	0.00325	0.0180
		3200	0.00231	0.0174
Exp 3	$L = 77$ m $u_* = 0.27$ m/s	100	0.0217	-
		200	0.0116	-
		800	0.00306	0.0314
		1600	0.00132	0.0302
		3200	0.000662	0.0284
Exp 4	$L = 34$ m $u_* = 0.2$ m/s	100	0.0409	-
		200	0.0272	-
		800	0.00804	0.0175
		1600	0.00426	0.0162
		3200	0.00314	0.0131
Exp 5	$L = 59$ m $u_* = 0.26$ m/s	100	0.0539	-
		200	0.0310	-
		800	0.00525	0.0156
		1600	0.00338	0.0147
		3200	0.00292	0.0114
Exp 6	$L = 71$ m $u_* = 0.3$ m/s	100	0.0539	-
		200	0.0310	-
		800	0.00525	0.0156
		1600	0.00338	0.0147
		3200	0.00292	0.0114

Table 1: Meteorological, Depositing tracer concentration and the effective dry deposition rates (v_d) obtained from (Doran and Horst, 1985). Table adapted from (Doran and Horst, 1985).

- Hybrid function was set as *fmincon*.
- Inertia range was set as [0.8; 1.2].
- Function Tolerance was set as 10^{-30} .
- The inertia term in the algorithm must be chosen such that its importance decreases during the algorithm execution. We set the inertia interval as [0.8,1.2], so it starts with values larger than 1, and assumes smaller values as the algorithm evolves.

Notice that, there is a hybridization in the minimization, and the *fmincon* function uses a gradient-descent technique to perform a local search and refine the results given by the PSO.

As we can observe in Fig 2 and in Tabs. 1–2, the estimated dry deposition rates using the LH model were, in general, more accurate than the ones obtained using the DH model. One of the main reasons for that, can be the larger number of estimated parameters in the LH model, that allowed the calibrated model be more adherent to the data than the DH. Such better adherence to the concentration data, as Fig. 3 and Tabs. 1–2 show, can also be due to the following, by considering the parameters a and b as additional unknowns in the model, in some sense, we are including the uncertainty in the observed meteorological data into the model estimation. In other words, by adjusting a and b , we are letting the

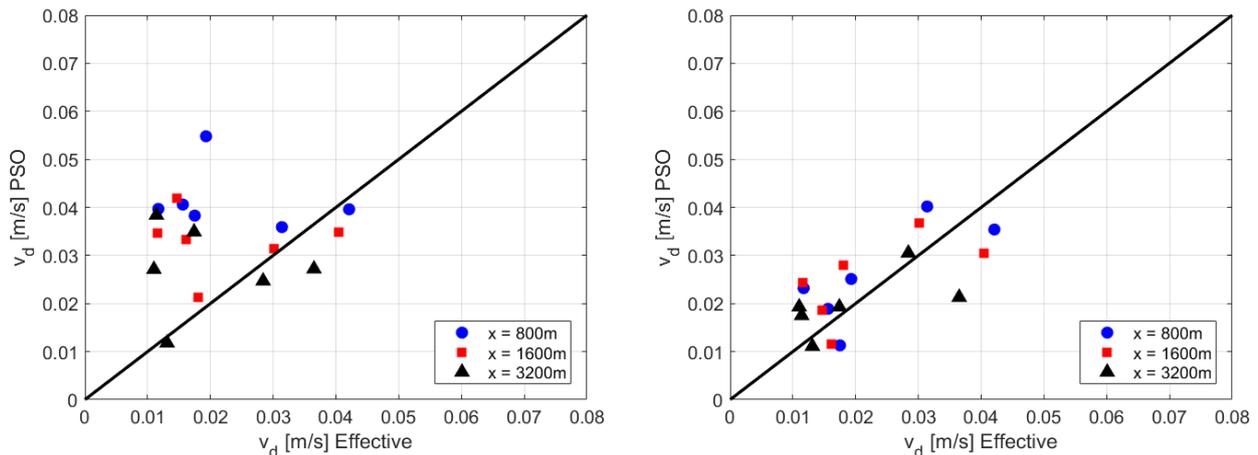


Figure 2: Effective \times predicted (PSO) dry deposition velocities using the DH and LH methodologies given, respectively, in Eq (3) (Doran and Horst, 1985)(left) and in Eq. (5) (Lin and Hildemann, 1997)(right).

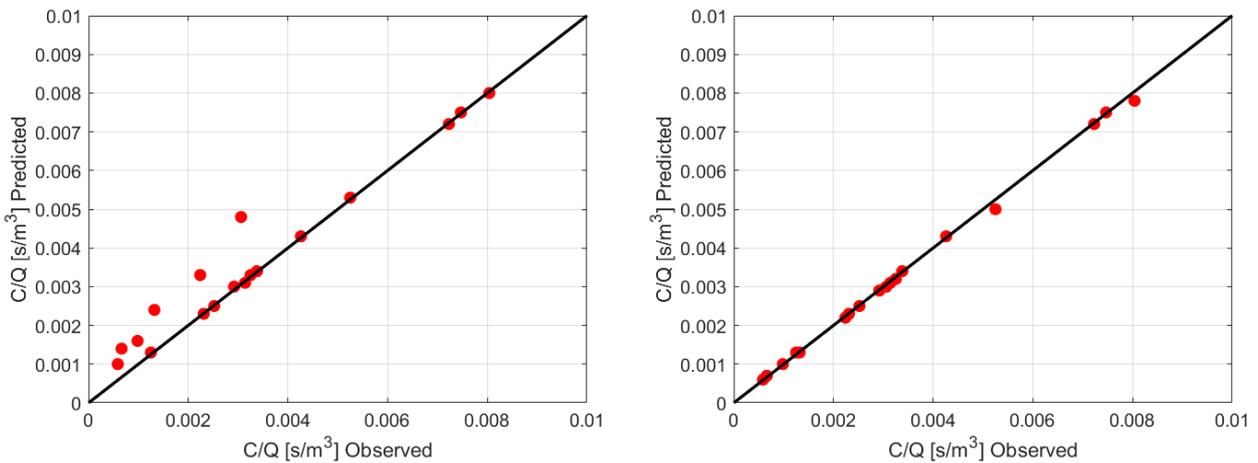


Figure 3: Observed \times predicted concentrations of dry deposition velocities with PSO using two different analytical methodologies given by (Doran and Horst, 1985)(left) and (Lin and Hildemann, 1997)(right).

concentration data indicate what are the more appropriate values for the parameters given by meteorological information. Overall, both models, the DH and the LH, performed well in the calibration presenting estimated dry deposition rates close to the values found in the literature for the same experimental dataset. The comparison between predicted and observed ZnS concentrations in Fig. 3 and the statistical indices in Tab. 3 corroborate the previous arguments, since a close agreement with observed ZnS concentration data is verified.

5. CONCLUSIONS

The Particle Swarm Optimization (PSO) technique combined with two analytical dispersion models were applied to estimate dry deposition rates of a tracer gas in the atmosphere from observed concentrations. As a case study, we used the datasets from the Hanford tracer experiment (Nickola, 1977) and crosswind- integrated concentrations. The effective dry deposition rates obtained in (Doran and Horst, 1985) were used for comparison and to verify the accuracy of the estimations. The analytical models used in the inversion methodology were defined in Eqs. (3)–(5), and proposed, respectively, by (Doran and Horst, 1985) and (Lin and Hildemann, 1997).

The estimation process for the LH model Eq. (5) was more complex than the calibration of the DH in Eq. (3), since it has additional unknown parameters. In this case, the estimation was divided in two steps as the number of unknowns was larger than the dataset. One of such steps was based on Tikhonov regularization Tikhonov and Arsenin (1998). The estimated dry deposition rates for the LH model were closer to the ones obtained in (Doran and Horst, 1985), considering all the six experiments. This result was probably due to the larger number of unknowns and the more complex calibration process. Moreover, the estimated concentrations for the LH model were also more adherent to the data than the concentrations obtained by the DH model. Overall, both models performed well and provided accurate solutions, with the LH model presenting a better performance.

Experiment	Arc [m]	Doran & Horst Model		Lin & Hindelmann Model			
		v_d	C/Q [s/m ²]	a	b	v_d	C/Q [s/m ²]
Exp 1	800	0.0396	0.0033	0.0838	0.1211	0.0354	0.0022
	1600	0.0348	0.0016			0.0304	0.0010
	3200	0.0272	0.0010			0.0213	0.0006
Exp 2	800	0.0548	0.0075	0.9136	0.9858	0.0251	0.0075
	1600	0.0212	0.0033			0.0280	0.0033
	3200	0.0349	0.0023			0.0193	0.0023
Exp 3	800	0.0359	0.0048	0.2474	0.4469	0.0402	0.0031
	1600	0.0314	0.0024			0.0368	0.0013
	3200	0.0247	0.0014			0.0305	0.0007
Exp 4	800	0.0383	0.0080	0.0010	1.1893	0.0113	0.0080
	1600	0.0334	0.0043			0.0116	0.0043
	3200	0.0118	0.0031			0.0111	0.0032
Exp 5	800	0.0406	0.0053	0.0010	1.9807	0.0189	0.0053
	1600	0.0420	0.0034			0.0187	0.0034
	3200	0.0384	0.0030			0.0175	0.0029
Exp 6	800	0.0397	0.0072	0.2976	0.0891	0.0232	0.0072
	1600	0.0347	0.0025			0.0243	0.0026
	3200	0.0271	0.0013			0.0193	0.0012

Table 2: Estimated values for the dry deposition rate (v_d) and concentration at different downwind distances (arcs) using the Doran & Horst and Lin & Hindelmann models in Eqs. (3)–(5), respectively. For the Lin & Hindelmann model, the parameters a and b were also estimated, with the resulting values presented above.

Index	DH	LH	Ideal
NMSE	0.02	0	0
RMSE	0	0	0
R	0.92	0.94	1
f_b	-0.09	0	0
f_s	0.07	0	0
FAC2	0.94	1	1
MAE	0	0	0

Table 3: Statistical indices to evaluate the agreement between observed and predicted ZnS concentrations using v_d estimated by PSO technique considering the analytical models by (Doran and Horst, 1985)(DH) and (Lin and Hildemann, 1997)(LH).

6. ACKNOWLEDGEMENTS

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