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A Bayesian Inference Model for the Estimation of Time-Dependent Pollutant Emissions

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Abstract. Source identification methodologies use inverse problems techniques combined with a dispersion model and observational data to estimate relevant source parameters. This work proposes a time-dependent model to estimate source parameters of multiple point releases. The forward problem or dispersion model accounts for the time variation of the wind field using a Fourier series that best fits the wind field time series of the experimental data. The source parameters are estimated by an adaptive Monte Carlo Markov Chain algorithm.

Key words. Source Estimation, Atmospheric Dispersion, Bayesian Inference, Monte Carlo Markov Chain Algorithms, Inverse Modeling.

1 Introduction

The source estimation of environmental releases is an interdisciplinary field of research involving applied mathematics, computation, and geophysics. It is usually employed to forecast sudden potentially harmful emissions in the atmosphere, rivers, lakes, and oceans at local or global scales. The models include the definition of a forward problem which is a mathematical description of the related physical processes, usually described by a partial differential equation (PDE). A net of observational data to feed the model is also necessary. In addition, it is essential to choose a technique to solve the inverse problem.

The main difficulties concerning source identification are the intrinsic ill-posedness of source identification, the correct description of the relevant physical processes involved, the accuracy of the methodologies to solve both the inverse and the forward models, and the noise in the observational data [6] are the main challenges. Several of these issues have been addressed in the literature. We recommend [4–6] for more information.

Including more data may partially address the ill-posedness of the inverse problem since it can reduce the signal to noise ratio. Some of our research [2, 4] were dedicated to the source identification of multiple atmospheric releases. However, we assumed the stationarity of measured quantities. An obvious drawback of this procedure is the loss of information.

In this work, by considering a dynamic model for the dispersion and the wind field, we allow the inversion model to account for more data. This also improves the physical description of the forward problem. The wind field is given by a truncated Fourier series fitted to observational wind time series.

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2 Source Estimation Methodology

In this section we describe all the techniques involved in the reconstruction of the source parameters considering point releases in the Atmospheric Boundary Layer (ABL). We start by presenting the calibration technique that is based on Bayesian inference, using an adaptive Monte Carlo Markov Chain (MCMC) algorithm. We then apply the inversion modeling to a dataset from the Fusion Field Trial 2007 (FFT07) experiment [13].

2.1 The Inversion Modeling Setting

Let C denote the set of concentrations evaluated numerically, using the dispersion model, at the sensors locations and measuring time instants. The set of experimental concentrations are denoted by C_{exp}. Denote by N the number of sensors and time instants where the concentrations are measured. The discrepancy between the evaluated and experimental concentrations is then modeled as

$$\varepsilon = \ln(C) - \ln(C_{\text{exp}}), \tag{1}$$

where ε is an N-dimensional Gaussian-distributed random variable, with mean zero and covariance matrix Σ. The $N \times N$-matrix Σ is equal to $\xi^{-1}I$, where ξ is a non-negative scalar so-called precision and I is the $N \times N$ identity matrix. Since we are assuming that concentration measurements are independent, the covariance matrix of ε has the present structure. The logarithm helps to turn the distribution of the concentration discrepancy close to symmetric.

We want to estimate the location and strength of M emission sources, more precisely, we must estimate the $4 \times M$-dimensional vector $\mathbf{v}_{\text{S}}$ containing the parameters of the sources, which is defined as follows

$$\mathbf{v}_{\text{S}} = [x_{\text{S},1}, y_{\text{S},1}, z_{\text{S},1}, Q_{\text{S},1}, \ldots, x_{\text{S},M}, y_{\text{S},M}, z_{\text{S},M}, Q_{\text{S},M}]^T, \tag{2}$$

where $(x_{\text{S},j}, y_{\text{S},j}, z_{\text{S},j})$ and $Q_{\text{S},j}$ are, respctively, the coordinate and the strength of the jth source, $j = 1, \ldots, M$.

Since the precision ξ is unknown, it is included in the set of the quantities that will be estimated. We define the likelihood function as follows,

$$P\left(C_{\text{exp}} \,|\xi, \mathbf{v}_{\text{S}}\right) \propto \xi^{\frac{N}{2}} \exp\left(-\frac{\xi}{2} \left\|\ln\left(C(\mathbf{v}_{\text{S}})\right) - \ln\left(C_{\text{exp}}\right)\right\|_{\ell_2}^2\right). \tag{3}$$

We also assume that ξ and $\mathbf{v}_{\text{S}}$ have independent prior distributions. The prior of ξ, as usual, is given by the distribution Gamma $\left(\frac{n_0}{2}, \frac{d_0}{2}\right)$. On the other hand, $\mathbf{v}_{\text{S}}$ has a uniform distribution in an appropriate set, that will be defined later.

The full conditionals for ξ and $\mathbf{v}_{\text{S}}$ are given as follows,

$$P\left(\xi \,|\mathbf{v}_{\text{S}}, C_{\text{exp}}\right) \propto P\left(C_{\text{exp}} \,|p, \mathbf{v}_{\text{S}}\right) P(\xi) \quad \text{and} \quad P\left(\mathbf{v}_{\text{S}} \,|p, C_{\text{exp}}\right) \propto P\left(C_{\text{exp}} \,|\xi, \mathbf{v}_{\text{S}}\right) P(\mathbf{v}_{\text{S}}). \tag{4}$$

The functions $P(\mathbf{v}_{\text{S}})$ and $P(\xi)$ denote, respectively, the prior probability densities of the quantities $\mathbf{v}_{\text{S}}$ and p. The full conditional for the precision ξ is proportional to a Gamma density, since the prior $P(\xi)$ is Gamma and the likelihood function is Gaussian. Thus, ξ given $\mathbf{v}_{\text{S}}$ and C_{exp} has the distribution Gamma $\left(\frac{\tilde{n}}{2}, \frac{\tilde{d}}{2}\right)$, with the parameters $\tilde{n} = n_0 + n$ and $\tilde{d} = d_0 + \| \ln(C_{\text{exp}}) - \ln(C(\mathbf{v}_{\text{S}}))\|_{\ell_2}^2$. The advantage of ξ be Gamma-distributed is the fact that this distribution can be easily sampled by a Gibbs sampler. To draw samples of ξ and $\mathbf{v}_{\text{S}}$, we use the Metropolis in Gibbs MCMC algorithm [6, 8, 10, 11], which is represented in Algorithm 1.

To make Algorithm 1 adaptive, the Gaussian random walk covariance matrix Z changes to increase or decrease the covariance accordingly to the discrepancy between concentrations evaluated with the proposed samples and the experimental data. For more details, see Ref. [6].

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Algorithm 1 Metropolis in Gibbs MCMC algorithm.

procedure MCMC ALGORITHM
  $j = 0$,
  $\xi^j = \xi, \mathbf{v}_{\mathrm{s}}^j = \mathbf{v}_{\mathrm{s}}$
  while $j \leq$ MaxIter **do**
    Draw $\mathbf{u} \sim N(\mathbf{v}_{\mathrm{s}}^j, Z)$
    Evaluate $\tilde{n}$ and $\tilde{d}$
    Draw $\xi^{j+1} \sim \mathrm{Gamma}\left(\frac{\tilde{n}}{2}, \frac{\tilde{d}}{2}\right)$
    Evaluate $P\left(\mathbf{v}_{\mathrm{s}}^j \middle| \xi^{j+1}, C_{\mathrm{exp}}\right)$ and $P\left(\mathbf{u} \middle| \xi^{j+1}, C_{\mathrm{exp}}\right)$
    Evaluate $\beta = \min\left(1, \frac{P\left(\mathbf{u} \middle| \xi^{j+1}, C_{\mathrm{exp}}\right)}{P\left(\mathbf{v}_{\mathrm{s}}^j \middle| \xi^{j+1}, C_{\mathrm{exp}}\right)}\right)$
    Draw $l \sim U[0, 1]$
    if $l < \beta$ **then**
      Accept: $\mathbf{v}_{\mathrm{s}}^{j+1} = \mathbf{u}$
    else
      Reject: $\mathbf{v}_{\mathrm{s}}^{j+1} = \mathbf{v}_{\mathrm{s}}^j$
    end if
    $j = j + 1$
  end while
end procedure

2.2 The Prior Density for the Source Parameters

Since the number of source parameters is large, we incorporate some prior information to simplify the search for the solutions. Based on an isopleth, i.e., the distribution of the concentrations through the computational domain in the xy-plane, as well as the wind direction, it is possible to find a region where the sources must be located. Moreover, also using the isopleth, it is possible to identify if the sources are located far apart from each other or close to each other. Furthermore, based on the concentration distribution in the domain, we assume that the emissions have similar strengths and are close to each other. If we order the sources, the parameters of the first emission must vary inside the set Λ, which is defined below:

$$\Lambda = [x_{\min}, x_{\max}] \times [y_{\min}, y_{\max}] \times [z_{\min}, z_{\max}] \times [Q_{\min}, Q_{\max}]. \tag{5}$$

Then, we define the parameters of the other sources in terms of the parameters of the first emission, i.e.,

$$[x_j, y_j, z_j, Q_j]^T = [x_1 + l_{x,j}, y_1 + l_{y,j}, z_1 + l_{z,j}, Q_1 + l_{Q,j}]^T, \quad j = 1, \ldots, M, \tag{6}$$

where the vector $[x_1, y_1, z_1, Q_1]^T$ contains the parameters of the first source, and the non-negative scalars $l_{x,j}$, $l_{y,j}$, $l_{z,j}$, and $l_{Q,j}$, $j = 1, \ldots, M$, represent the distance between the parameters of the jth source to the first emission. These distances can vary in preset intervals of the form $[l_{x,\min}, l_{x,\max}]$, $j = 1, \ldots, M$.

Based on such definitions implied by the prior information, we assume that the prior density of $\mathbf{v}_{\mathrm{S}}$ is the uniform density in the set $\Lambda \times L^{M-1}$, with

$$L = [l_{x,\min}, l_{x,\max}] \times [l_{y,\min}, l_{y,\max}] \times [l_{z,\min}, l_{z,\max}] \times [l_{Q,\min}, l_{Q,\max}]. \tag{7}$$

The power $M - 1$ in the definition of the prior density represents the Cartesian product of the set L with itself $M - 1$ times. We use truncation to make the MCMC algorithm proposals to stay inside the set $\Lambda \times L^{M-1}$.

2.3 The Forward Problem

The dispersion model is described by the following advection-diffusion Partial Differential Equation (PDE)

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$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C - \nabla \cdot (\mathbf{K} \nabla C) = \sum_{j=1}^{M} S_j \text{ in } \quad \Omega \times [0, T] \tag{8}$$

where $C = C(x, y, z, t)$ $[g/m^3]$ is the time averaged concentration. The computational domain is represented by Ω and it is within the Atmospheric Boundary Layer (ABL). The wind field is represented by $\mathbf{u}$, and the the tensor $\mathbf{K}$ accounts for the ABL turbulence state, and it is a diagonal matrix with the non-zero components K_x, K_y, and K_z which represent the turbulent diffusion in the x, y and z directions, respectively. The right-hand side of the equation corresponds to the contribution of the set of point source emissions, where M represents the total number of sources, and S_j is the jth source, defined as

$$S_j(x, y, z, t) = Q_{\mathrm{S},j} \delta(x - x_{\mathrm{S},j}) \delta(y - y_{\mathrm{S},j}) \delta(z - z_{\mathrm{S},j}) \mathcal{I}_{[t_j^{\mathrm{S}} - \beta_j, t_j^{\mathrm{S}} + \beta_j]}(t),$$

wherein $[x_{\mathrm{S},j}, y_{\mathrm{S},j}, z_{\mathrm{S},j}]^T$ are the source locations, $Q_{\mathrm{S},j}$ is the emission rate, $j = 1...M$ and δ is the Dirac delta distribution. Finally, $\mathcal{I}_{[t_j^{\mathrm{S}} - \beta_j, t_j^{\mathrm{S}} + \beta_j]}(t)$ represents the indicator (characteristic) function of the jth emission time interval $[t_j^{\mathrm{S}} - \beta_j; t_j^{\mathrm{S}} + \beta_j]$, with $\beta_j > 0$.

To complete the forward problem definition, (8) can be associated to the following boundary conditions:

$$\mathbf{n} \cdot \nabla C = 0, \tag{9}$$

in which the vector $\mathbf{n}$ represents the outward normal on the boundaries of Ω at $z = z_0$ and $z = H$, and

$$C = 0 \quad \text{elsewhere}, \tag{10}$$

and the following initial condition

$$C = C_0 \quad \text{in} \quad \Omega \quad \text{for } t = 0, \tag{11}$$

wherein z_0 represents the surface roughness length and H denotes the upper boundary of the computational domain Ω. To avoid solving Equation (8) for all the iterations of Algorithm 1, we take advantage of the linearity of Eq. (8) to establish a relationship between the sources and the sensors through the so-called adjoint state PDE. This procedure considerably reduces the computational cost of the simulations, since the direct problem is solved only once for all the iterations. Representing the observed concentration at the kth sensor, at the time instant T by $C_{\exp}(x_k, y_k, z_k, T)$, it follows that

$$C_{\exp}(x_k, y_k, z_k, T) = \sum_{j=1}^{M} \int_0^T \int_\Omega C_k^* S_j d\Omega dt = \sum_{j=1}^{M} \langle C_k^*, S_j \rangle, \tag{12}$$

where C_k^* is the solution of the following adjoint-state PDE [9]:

$$-\frac{\partial C^*}{\partial t} - \mathbf{u} \cdot \nabla C^* - \nabla \cdot (\mathbf{K} \nabla C^*) = \mathcal{S}_k, \tag{13}$$

wherein $\mathcal{S}_k(x, y, z, t) = \delta(x - x_k) \delta(y - y_k) \delta(z - z_k) \mathcal{I}_{[t_{\min}, t_{\max}]}(t)$, with (x_k, y_k, z_k), the spatial coordinates of the kth sensor for N sensors as well as a sampling time interval $[t_{\min}, t_{\max}]$. The terminal and boundary conditions associated to Eq. (13) are the following:

$$C^* = 0 \quad \text{on} \quad \Omega \quad \text{for } t = t_{\max}, \tag{14}$$

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$$\mathbf{n} \cdot \nabla C^* = 0 \quad \text{at } z = z_0 \text{ and } z = H, \quad \text{and } C^* = 0 \quad \text{elsewhere.} \tag{15}$$

Again, $\mathbf{n}$ denotes the outward normal on the boundary of Ω. In the adjoint-state PDE, the sensors work as sources. Thus, the concentration at each sensor will be obtained by the scalar product between the source term and the solution of the PDE (13) according to Eq. (12).

In this work the Finite Element formulation [7] is applied to solve Eq. (12). Here this formulation is not presented since it is performed in the author's previous works such as in Refs.[4, 5].

2.4 A Case study

We evaluate the proposed methodology to estimate the source parameters, namely, the source position and emission rates using the Fusion Field Trial (FFT07) tracer experiment [13]. We briefly describe the experiments here. More detailed description can be found in [1, 2].

The FFT07 were performed several times, considering different atmospheric conditions and different number of emissions. In this work, we consider the datasets from Trial 55, in which the tracer gas propylene (C_3H_6) was released during 15 minutes at a constant rate from four point sources 2 meters above the ground level.

During FFT07 trial 55, the tracer gas propylene (C_3H_6) was released during 15 minutes at a constant rate from four point sources, located two meters above the ground surface level. After released, the (C_3H_6) was sampled over a total of 100 tracer sampling units, arranged over a rectangular grid sampling of 475 m $\times$ 450 m. The sensors were displayed keeping the distance of 50 m from each other.

The FFT07 provides datasets regarding meteorological and concentration measurements to allow the evaluation of evaluate short-range source identification techniques. The meteorological variables were recorded during about 50 minutes over different positions at 2, 4, 8, 16 and 32 meters above the ground level. The high frequency datasets of wind and temperature were employed to calculate the turbulent fluxes of sensible heat and momentum to further compute the Monin-Obukhov length (L), the friction velocity (u_*) and the surface roughness length (z_0). Those are input parameters used to obtain the vertical turbulent diffusion profile.

Details concerning the datasets applied to feed the forward model can be found in Ref.[4]. In order to avoid to solve the Navier-Stokes equations over the computational domain, we derive the wind field from the measured wind time series and obtain its best fit to a Fourier series. The resulting wind profile was applied to perform the simulations.

3 Results and Discussion

Next, we present the numerical evaluation of the Algorithm 1 using the FFT07 datasets. The numerical evaluation of the forward problem in Eqs. (13)–(15) is omitted here and we refer the reader to Ref. [1].

The sets Λ and L that compose the set $\Lambda \times L^3$, see Eqs. (5)–(7), where the prior density of the vector of source parameters is defined, are the following,

$$\Lambda = [-100, 100] \times [50, 600] \times [0.013, 50] \times [0, 30] \quad \text{and} \quad L = [-25, 25] \times [-250, 250] \times [-1.5, 1.5] \times [-5, 5],$$

where the unit for the spatial parameters is meter and the unit for the source strengths is g/s. These sets are defined based on the wind direction and on the isopleth in Figure 1 in Ref. [3]. It is worth mentioning that the true source locations are inside the resulting searching region.

To initialize Algorithm 1, we draw an uniformly-distributed sample for the source parameters inside the search region $\Lambda \times L^3$, and the initial step for p is generated by the prior distribution Gamma($\frac{n_0}{2}, \frac{d_0}{2}$) with $n_0 = 10^{-3}$ and $d_0 = 10^{-3}$. The resulting Markov Chain has a total of 50

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thousand states. The first 10 thousand states are defined as the burn-in set. In the remaining states, 800 states were selected, since we used a step-size of 50 states. The summary statistics of the resulting chain can be found in Table 1.

Parameter	True	Min.	Q1	Median	Q3	Max.	GR Test
$x_{S,1}$ [m]	33.0	-51.1	-24.52	-18.94	-12.95	9.3	1.00
$y_{S,1}$ [m]	171	50.0	76.9	100	120	208	1.02
$z_{S,1}$ [m]	2.00	3.80	9.89	11.3	12.6	19.0	1.00
$Q_{S,1}$ [g/s]	11.4	10.1	13.8	14.6	15.5	18.6	1.00
$x_{S,2}$ [m]	33.8	-26.6	-5.7	-0.3	5.2	27.3	1.00
$y_{S,2}$ [m]	241	213	286	300	315	381	1.02
$z_{S,2}$ [m]	2.00	2.37	8.55	9.91	11.3	17.8	1.00
$Q_{S,2}$ [g/s]	11.4	5.70	9.2	10.0	10.8	14.3	1.00
$x_{S,3}$ [m]	30.0	-12.3	14.3	20.01	25.81	50.97	1.00
$y_{S,3}$ [m]	313	266	346	366	386	466	1.03
$z_{S,3}$ [m]	2.00	2.32	8.72	10.1	11.5	17.9	1.00
$Q_{S,3}$ [g/s]	4.65	2.20	5.70	6.57	7.50	10.8	1.00
$x_{S,4}$ [m]	26.0	-11.9	9.9	15.6	21.4	41.0	1.00
$y_{S,4}$ [m]	384	375	488	507	527	593	1.01
$z_{S,4}$ [m]	2.00	1.82	7.84	9.28	10.6	17.1	1.00
$Q_{S,4}$ [g/s]	11.4	10.0	13.7	14.5	15.4	19.1	1.00
p	-	0.01	0.02	0.02	0.02	2.61	1.00

Table 1: Comparison between the true values (True) and the summary statistics of the chains of the unknown parameters. The summary is given by the minimum value (Min.), the first quartile (Q1), the median value (Median), the third quartile (Q3), and the maximum value (Max.). We also include the value of the Gelman-Rubin convergence test (GR Test) for each variable, whose optimal value is 1.00.

As Tab. 1 shows, the values provided by the Gelman-Rubin test [12] are close or equal to 1.00, the ideal value. It indicates the convergence of the chains. Moreover, the major part of the true source parameters are inside the sample regions. On the other hand, if the source parameters are outside the sample regions, they are placed in the neighborhood of the sample regions. If we compare the present results with those from Ref. [3], we observe that they are more accurate. This is probably due to the improvements made in the modelling of the forward operator, that considers the dynamic change in the wind direction. It is worth mentioning that the present methodology must be refined so that, the regions defined by the Markov chain states include all the true source parameters.

4 Concluding Remarks

We used a time-dependent dispersion model that allowed to consider the evolution of the wind direction in the forward problem and to use time series of measured concentrations in the source estimation. Such changes considerably improved estimates if we compare with those from Ref. [3]. However, further improvements are still needed in order to find more accurate results.

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