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Adjoint Approach to Estimate the Non-Steady Emission Rate of A Point Source

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Abstract
A method to estimate the non-steady emission rate of a point source by using pollutants concentration data is given. To solve this ill-posed inverse problem, a variational problem with constraints is proposed, where the objective functional for minimizing is the norm of the first derivative of the emission rate, and the integral constraints for the pollutants concentration contain full information, in a compact form, on the dispersion phenomenon in the atmosphere and include the anomaly of concentration data. A three-dimensional dispersion air pollution model with point sources is considered in a limited region to forecast the concentration of pollutants. The corresponding adjoint model and the duality principle are used to write the constraints in terms of influence (adjoint) functions. It is shown that both models are well posed in the sense of Hadamard. The corresponding discrete formulation, obtained from the variational problem, is an easily-solvable quadratic programming problem whose solution determines the emission rate of the source. Numerical results obtained for a simple box model and a one-dimensional diffusion problem demonstrate the method's ability.
Keywords: inverse problem, estimation of parameters, dispersion model, adjoint model.

I. Introduction

Dispersion models are routinely used to assess the impact of emission sources on air quality for different meteorological conditions. These models are also used at nuclear and industrial plants for emergency response and impact assessments for hazardous substances accidentally released into the atmosphere [1,2]. Apart from their direct application, such models are a fundamental instrument for solving such inverse problems as the assessment of the air pollution problem parameters [2,3,4].

The inverse problems involve the determination of the causes from the knowledge of the effects in a system. As the effects we consider observed or desired values of the state variables, and the causes are the initial conditions, the forcing or the parameter values that are capable of driving the system to the effects [3,5]. Frequently, the inverse problems are ill-posed, presenting different types of instabilities or the difficulties related with existence and uniqueness of solutions [5,6]. Therefore, the solution of inverse problems requires the use of such methods that allow us to suppress the instability and choose a solution congruent with the studied phenomenon. Such methods are known as the methods of regularization of inverse problems [5,6]. Among these methods, it should be noted those introduced by Tikhonov [7], which consist in minimizing a functional that combines the data errors and the norms of the control variable and some of its derivatives. The inverse problem considered here, consists in evaluating unknown sources of the atmospheric pollutants by using a given set of measured concentrations. The solution of such inverse problem can be used to estimate and verify emission inventories [8] of many (chemical or biological) toxic and radioactive species, as well as to detect unknown sources of atmospheric tracers. For example, the detection and location of nuclear testing or terrorism-related events can be achieved using observations of radionuclides in the atmosphere [2,9]. In particular, the solution of this inverse problem provides an opportunity to assess the intensity of undeclared emissions of dangerous contaminants [10].

In the study of such inverse problems, the intensity of the source is often considered to be stationary or defined as an impulse in time (explosion), and this feature is essentially used in all methods developed to estimate the intensity of the source. As a result, these methods do not cover all of the actual events of emissions and have a limited application [4,9,10]. Also, the developed methods frequently use analytical solutions of a simple dispersion model of pollutants (Gaussian plume model) that limits their application only to the cases of steady conditions of dispersion in the atmosphere [4,10,11].

In the present work, we suggest a method for assessing unknown non-steady emission rate of a new point source by using a linear dispersion model with time-variable parameters. It is assumed that an accident, or undeclared release took place at unknown time, but the location of the source is known (for example, it was discovered by a satellite). The inverse problem is ill-posed and a regularization method is necessary in order to find the solution [11,12,13]. It is

shown the usefulness of the Tikhonov regularization in the case when the functional to be minimized contains the derivative of the emission rate, and is subjected to integral constraints on the pollutant concentration which contains full information, in a compact form, on the dispersion phenomenon in the atmosphere and utilize the data on the anomalies of pollutant concentration. The adjoint solutions (or influence functions) used in the integral constraints exhibit the explicit relation between the emission rate and the data on the anomalies of pollutant concentration. The resulting variational problem is easily solved. In Section II, we formulate the general variational problem, while a dispersion model and its corresponding adjoint, which match the features of the general formulation, are presented in Section III. Finally, in Section IV the numerical approach of the variational problem is presented, and two examples of the solution of inverse problem are developed in detail using synthetic data.

II. Formulation of the problem

Let us suppose that a singular event, such as an explosion or release of a dangerous substance, happened completely during a time interval $(0,T)$ in a known place $\mathbf{r}_0$ located in a region $\mathbf{D}$, and the emission rate of new source $Q(t)$ is the control variable that must be determined. We also assume that the distribution $\varphi^0(\mathbf{r})$ of this pollutant in $\mathbf{D}$ at the initial moment $t=0$, and the emission rates $f(\mathbf{r},t)$ of the other sources located in $\mathbf{D}$ are well known, and hence, the actual state ϕ' in region $\mathbf{D}$ for $t\geq 0$ is known (for example, ϕ' can be determined with a model). Taking into account the new source, the dispersion model for calculating the pollutant concentration φ in time interval $(0,T)$ can be written as

$$\frac{\partial \varphi}{\partial t}+A\varphi = f(\mathbf{r},t)+Q(t)\delta(\mathbf{r}-\mathbf{r}_0) \text{ in } \mathbf{D}\times(0,T) \quad (1)$$

$$\varphi(\mathbf{r},0)=\varphi^0(\mathbf{r}) \text{ in } \mathbf{D} \quad (2)$$

where A is the linear operator that describes the advection of pollutants by winds or currents, turbulent diffusion, deposition and decay of pollutants due to chemical reactions, $\delta(\mathbf{r}-\mathbf{r}_0)$ is the Dirac delta, and $\mathbf{r}_0$ is the location of new source.

On the other hand, let us denote by ϕ' the solution of dispersion problem (1)-(2) only with forcing $f(\mathbf{r},t)$ ($Q(t)\equiv 0$). Thus, the anomaly of the pollutant concentration $\phi=\varphi-\phi'$ satisfies the dispersion problem

$$\frac{\partial \phi}{\partial t}+A\phi = Q(t)\delta(\mathbf{r}-\mathbf{r}_0) \text{ in } \mathbf{D}\times(0,T) \quad (3)$$

$$\phi(\mathbf{r},0)=0 \text{ in } \mathbf{D} \quad (4)$$

As it was noted before, the basic concentration of the pollutant ϕ' is supposed known as the solution of the dispersion model (3)-(4) with the initial distribution $\varphi^0(\mathbf{r})$ and forcing $f(\mathbf{r},t)$. Further, the concentration φ can be partially evaluated by means of its monitoring in some places of region $\mathbf{D}$. Therefore, the time series of the anomaly $\phi=\varphi-\phi'$ of the pollutant can be obtained at the monitoring sites.

It is reasonable to assume that the errors in the measurements of concentration φ are small. However, the errors in the basic function ϕ' are typically much larger than the errors of measurements, because this function is the result of solution of a computational model that does not describe exactly the processes of dispersion and transformations in the atmosphere. Therefore, we assume that the errors in the values of $\varphi-\phi'$, obtained in different monitoring sites, are of the same magnitude as the errors in function ϕ'. Such perturbations in the anomaly $\phi=\varphi-\phi'$ of concentration of pollutant generate the numerical instability in the process of inversion of data, and they are the fundamental reason for introducing one of the regularization methods. To this end, we first obtain integral equations that establish a direct relationship between the "cause" and the "effect" in the system.

In order to find an explicit relationship between the anomaly ϕ (state variable) and the emission rate $Q(t)$ (control variable), we introduce the influence function g as the solution of the following adjoint model:

$$-\frac{\partial g}{\partial t}+A^*g = p(\mathbf{r},t) \text{ in } \mathbf{D}\times(0,T) \quad (5)$$

$$g(\mathbf{r},T)=0 \text{ in } \mathbf{D} \quad (6)$$

The adjoint operator A^* is defined by means of the Lagrange identity [14]:

$$(A\phi,g)=(\phi,A^*g) \quad (7)$$

where $(h,g)=\int_{\mathbf{D}} hg\,d\mathbf{r}$ is the inner product of the Hilbert space $L_2(\mathbf{D})$.

Multiplying (3) by g and taking the integral over $\mathbf{D}\times(0,T)$ we get

$$\int_0^T\int_{\mathbf{D}} g\frac{\partial\phi}{\partial t}d\mathbf{r}dt + \int_0^T\int_{\mathbf{D}} gA\phi d\mathbf{r}dt = \int_0^T\int_{\mathbf{D}} gQ(t)\delta(\mathbf{r}-\mathbf{r}_0)d\mathbf{r}dt \quad (8)$$

The integration by parts of the first integral and the use of conditions (4) and (6) leads to

$$\int_0^T\int_{\mathbf{D}} g\frac{\partial\phi}{\partial t}d\mathbf{r}dt = \int_0^T\int_{\mathbf{D}} \phi(-\frac{\partial g}{\partial t})d\mathbf{r}dt$$

Using in (8) Lagrange identity (7) and the well-known property of Dirac delta one can get

$$\int_0^T\int_{\mathbf{D}} \phi\left\{-\frac{\partial g}{\partial t}+A^* g\right\}d\mathbf{r}dt = \int_0^T Q(t)g(\mathbf{r}_0,t)dt$$

Finally, taking into account equation (5) we obtain

$$\int_0^T\int_{\mathbf{D}} \phi\, p(\mathbf{r},t)d\mathbf{r}dt = \int_0^T Q(t)g(\mathbf{r}_0,t)dt$$

In particular, if the forcing $p(\mathbf{r},t)$ in (5) is defined as

$$p(\mathbf{r},t)=\delta(\mathbf{r}-\mathbf{r}_i)\delta(t-T)$$

then the last equation is reduced to

$$\phi(\mathbf{r}_i,T)=\int_0^T Q(t)g_i(\mathbf{r}_0,t)dt \quad (9)$$

Equation (9) is the desired explicit relationship between the emission rate $Q(t)$ and the anomaly of pollutant concentration at time $t=T$ and monitoring sites $\mathbf{r}_i$, $i=1,...,N$. It should be noted that the adjoint function g_i depends only on the dispersion conditions in the atmosphere and monitoring site $\mathbf{r}_i$, but it is independent of the control variable Q. Thus, in order to use equations (9), the N different adjoint functions must be calculated.

On the other hand, let $\mathbf{R}$ be a monitoring site and let $t_j \le T$ be a sampling time ($j=1,2,...,M$). Then the forcing $p(\mathbf{r},t)$ in (5) can be defined as

$$p(\mathbf{r},t)=\delta(\mathbf{r}-\mathbf{R})\delta(t-t_j),$$

and in addition to (9), one can obtain a set of alternative equations

$$\phi(\mathbf{R},t_j)=\int_0^T Q(t)g_j(\mathbf{r}_0,t)dt,\ j=1,2,...,M \quad (10)$$

where $\{\phi(\mathbf{R},t_j)\}_{j=1}^M$ is the time series (without errors) of the anomaly of pollutant concentration obtained at the monitoring site $\mathbf{R}$ and sampling time $t_j \le T$. Again, the adjoint function g_j depends only on the dispersion conditions in the atmosphere and the sampling moments t_j, and is independent of the control variable Q. Note that in order to use the formulas (10), it is necessary to calculate the *M* adjoint functions. Besides, according to (5)-(6),

$$g_j(\mathbf{r}_0,t)=0 \text{ in } (t_j,T)$$

and hence, equations (10) are reduced to

$$\phi(\mathbf{R},t_j)=\int_0^{t_j} Q(t)g_j(\mathbf{r}_0,t)dt,\ j=1,2,...,M \quad (11)$$

In the case of stationary conditions of atmospheric dispersion, the operator A is time independent, and

$$g_j(\mathbf{r}_0,t)=g_\alpha(\mathbf{r}_0,T-t_j+t) \text{ in } (0,t_j),\ j=1,2,...,M \quad (12)$$

where the adjoint function g_α, known as the basic kernel, is the solution of (5)-(6) with forcing $p(\mathbf{r},t)=\delta(\mathbf{r}-\mathbf{R})\delta(t-T)$. Thus, equations (11) accept the form

$$\phi(\mathbf{R},t_j)=\int_0^{t_j} Q(t)g_\alpha(\mathbf{r}_0,T-t_j+t)dt,\ j=1,2,...,M \quad (13)$$

and hence, in the stationary case, it is necessary to calculate only one adjoint function g_α to establish the explicit relationships between $Q(t)$ and the time series $\{\phi(\mathbf{R},t_j)\}_{j=1}^M$ of the anomaly of the pollutant concentration at the monitoring site $\mathbf{R}$. Thus, in the stationary case, the calculations are much simpler.

It should be emphasized that the use of equations (9) or (11) depends on the monitoring information available. Equation (9) is useful in the case when the pollutant concentrations are available simultaneously from different monitoring stations in $\mathbf{D}$, while equation (11) can be applied if there is a time series of pollutant concentrations obtained from a single monitoring station in the region. Hereafter, in the study of the inverse problem, only equations (11) will be used.

There are two particular cases when the source strength can be immediately identified from equations (11). First, when the mean value of the errors $\delta\phi_j$ in the anomaly of pollutant concentration

are equal to zero, and the intensity of source is constant: $Q(t)=Q_c$, $0<t<T$. Then, summing the relations (11) over j from 1 to M yields

$$Q_c=\left(\sum_{j=1}^{M}\phi_j\right)\left(\sum_{j=1}^{M}\int_0^{t_j}g_j(\mathbf{r}_0,t)dt\right)^{-1} \quad (14)$$

where ϕ_j is the anomaly of pollutant concentration registered at moment t_j and $\phi(\mathbf{R},t_j)=\phi_j-\delta\phi_j$, $j=1,...,M$. Second, when the mean value of errors $\delta\phi_j$ are equal to zero, and the source intensity is of the form of an impulse at time $t=t_e$: $Q(t)=Q_e\delta(t-t_e)$, $0<t_e<T$. Then, summing the relations (11) over j from 1 to M yields

$$Q_e=\left(\sum_{j=1}^{M}\phi_j\right)\left(\sum_{j=1}^{M}g_j(\mathbf{r}_0,t_e)\right)^{-1} \quad (15)$$

Suppose now that the emission rate of a point source is non-steady and a time series $\{\phi_j\}_{j=1}^{M}$ of the anomalies of pollutant concentration is available at the monitoring site $\mathbf{R}$. We now suggest a regularization method (Tikhonov regularization) for the inverse problem:

$$\text{minimize}\quad J(Q)=\frac{1}{2}\int_0^T\left(\frac{dQ}{dt}\right)^2dt \quad (16)$$

$$\text{subject to: } Q(t)\geq 0,\ 0<t<T,\ Q(0)=Q(T)=0, \quad (17)$$

$$\text{and } \varepsilon\leq\phi_j-\int_0^{t_j}Q(t)g_j(\mathbf{r}_0,t)dt\leq\varepsilon,\ j=1,..,M \quad (18)$$

where $\phi(\mathbf{R},t_j)=\phi_j-\delta\phi_j$ and $\delta\phi_j$ is the corresponding error in the anomaly of pollutant concentration at moments $t=t_j$, $j=1,...,M$.

The functional (16) is minimized in order to filter out perturbations and reconstruct the emission rate. Note that due to errors in the anomalies of pollutant concentration, it is impossible to directly use equation (11). That is why we consider the difference of terms in (18). In this constraint of variational problem (16)-(18), the positive parameter ε is introduced for extending the feasibility space in which there exists the solution of the inverse problem. As it will be explained in the examples below, this parameter influences the smoothness of the solution obtained, and its optimum value is a function of the maximum error in the anomaly.

Note that, both the objective function and the constraints directly depend of the control variable Q, and therefore, the discrete version of problem (16)-(18) will determine an optimization problem of many real variables (quadratic programming problem). Such a problem is posed and solved in Section IV with the *quadprog* routine of MATLAB [15]. In the next section, we describe the dispersion and adjoint models that complete the formulation of inverse problem.

III. Dispersion and adjoint models

The results described in the previous section are valid for any dispersion model (1)-(2), provided that the operator A is linear and its adjoint operator is defined by means of Lagrange identity. These models can differ by the number of physical and chemical processes taken into account and by the forms of parameterization of these processes. Hereinafter we briefly describe a model which can be used to predict the dispersion of a few substances emitted from various sources. The details can be found in [16].

3.1 Dispersion model

We now consider a general dispersion model for short-term forecast of various pollutants emitted from sources located in a region $\mathbf{D}$. Such a model can be used to describe the concentrations φ or ϕ as much as the anomaly ϕ of the released substance. Of course, the corresponding values of parameters, initial condition and forcing must be specified in each concrete case.

Let $\mathbf{D}=D\times(0,H)$ be a simply connected bounded domain in $\mathbf{R}^3$ whose boundary $\partial\mathbf{D}=S_0\cup S\cup S_H$ is the union of a cylindrical lateral surface S, the base S_0 at the bottom, and top cover S_H at $z=H$ (see Figure 1). In the domain $\mathbf{D}$, we consider the following dispersion model $\mathbf{M}$ for K quasi-passive pollutants:

$$\frac{\partial\phi_k}{\partial t}+\mathbf{U}\cdot\nabla\phi_k+\sigma_k\phi_k-\nabla\cdot(\mu\nabla\phi_k)-\frac{\partial}{\partial z}\mu_z\frac{\partial\phi_k}{\partial z}+$$
$$\nabla\cdot\boldsymbol{\phi}_k^s=f_k(\mathbf{r},t) \quad (19)$$

$$\boldsymbol{\phi}_k^s=-v_k^s\phi_k\mathbf{e}_3 \text{ in } \mathbf{D} \quad (20)$$

$$\phi_k(\mathbf{r},0)=\phi_k^0(\mathbf{r}) \text{ in } \mathbf{D} \quad (21)$$

$$\mu\nabla\phi_k\cdot\mathbf{n}-U_n\phi_k=0 \text{ on } S^- \quad (22)$$

$$\mu\nabla\phi_k\cdot\mathbf{n}=0 \text{ on } S^+ \quad (23)$$

$$\mu\nabla\phi_k\cdot\mathbf{n}=0 \text{ on } S_0 \quad (24)$$

$$\mu_z\frac{\partial\phi_k}{\partial z}-U_n\phi_k=-v_k^s\phi_k \text{ on } S_H^- \quad (25)$$

$$\mu_z\frac{\partial\phi_k}{\partial z}=-v_k^s\phi_k \text{ on } S_H^+ \quad (26)$$

$$\nabla \cdot \mathbf{U} = \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0 \text{ in } \mathbf{D} \tag{27}$$

For each k ($k = 1,...,K$), $\phi_k(\mathbf{r},t) \geq 0$ represents the concentration of k th primary pollutant, $\phi_k^0(\mathbf{r})$ is its initial distribution at $t = 0$, and $\sigma_k(\mathbf{r},t) \geq 0$ is the coefficient describing the decay of this element due to chemical transformations. Also, $\mu(\mathbf{r},t) > 0$ and $\mu(\mathbf{r},t) > 0$ are the turbulent diffusion tensors,

$$\mu = \begin{pmatrix} \mu_x(\mathbf{r},t) & 0 \\ 0 & \mu_y(\mathbf{r},t) \end{pmatrix}, \; \mu = \begin{pmatrix} \mu_x(\mathbf{r},t) & 0 & 0 \\ 0 & \mu_y(\mathbf{r},t) & 0 \\ 0 & 0 & \mu_z(\mathbf{r},t) \end{pmatrix}, \tag{28}$$

and the term $\nabla \cdot \boldsymbol{\phi}_k^s$ in (19), describes the change of concentration of particles per unit time because of sedimentation with constant velocity $v_k^s > 0$. It is assumed that the wind velocity $\mathbf{U}(\mathbf{r},t) = (u,v,w)$ is known and satisfies the continuity equation (27) in $\mathbf{D}$.

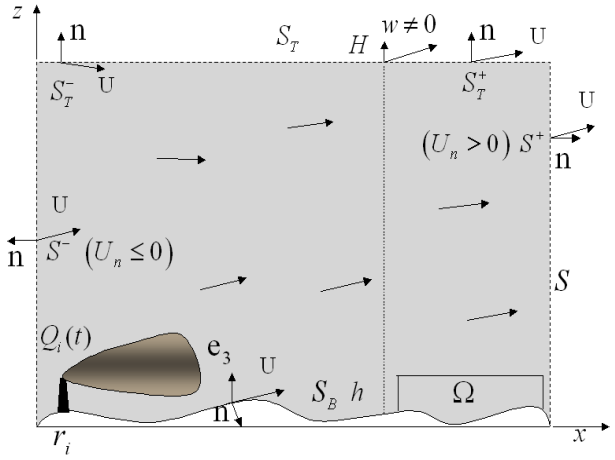

Fig. 1 Cross section of region $\mathbf{D}$.

Without loss of generality, we assume that the forcing

$$f_k(\mathbf{r},t) = \sum_{i=1}^{N} q_{ik}(t)\delta(\mathbf{r} - \mathbf{r}_i) \tag{29}$$

is formed by the point sources (for example, by various industries) located at the points $\mathbf{r}_i \in \mathbf{D}$, $i = 1,...,N$. Here $q_{ik}(t)$ is the emission rate of k th pollutant of the i th source. Thus,

$$q_i(t) = \sum_{k=1}^{K} q_{ik}(t) \tag{30}$$

is the emission rate of the i th source formed by the emission rates of K different pollutants, and $\delta(\mathbf{r} - \mathbf{r}_i)$ is the Dirac delta centered at the plant position $\mathbf{r}_i$, $i = 1,...,N$. Note that the line or area sources in region $\mathbf{D}$ can also be modeled as point sources [17], and therefore equation (29) is a rather general form of the forcing in equation (19).

The conditions on the open boundary $\partial\mathbf{D}$ of limited domain $\mathbf{D}$ lead to the well-posed problem in the sense of Hadamard [18]. We denote by $U_n = \mathbf{U} \cdot \mathbf{n}$ the projection of the velocity $\mathbf{U}$ on the outward unit normal $\mathbf{n}$ to the boundary S, which is divided into the outflow part S^+ where $U_n \geq 0$ (advective pollution flow is directed out of $\mathbf{D}$) and the inflow part S^- where $U_n < 0$ (advective pollution flow is directed into $\mathbf{D}$). It is assumed that region $\mathbf{D}$ is large enough and includes all important pollution sources. In other words, we suppose that there is no sources outside $\mathbf{D}$, and hence, the total flow of pollutants (the sum of the diffusive flow and advective flow) is zero on the inflow part S^- (condition (22)). The pollution flow is non-zero only on S^+, besides, according to (23), the diffusive flow on S^+ is assumed to be negligible as compared with the corresponding advective flow. The conditions (25) and (26) have similar meanings on S_H, where the sedimentation of the particles has been also taken into account. Equation (24) indicates that there is no flow of the substances through S_0, since $\mathbf{U} \cdot \mathbf{n}$ and v_k^s are both zero on the surface of irregular terrain (see Fig. 1). In general, (25) and (26) are necessary because $w = 0$ on S_0 and (27) lead to a non-zero vertical velocity component at S_H:

$$w(x,y,z,t) = -\int_0^z \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) dz \tag{31}$$

The boundary conditions are mathematically good, because the resulting problem (19)-(27) is well posed, that is its solution exists, is unique and continuously depends on the initial condition and forcing [19]. This follows from the fact that the problem operator

$$A\phi_k = \mathbf{U} \cdot \nabla \phi_k + \sigma_k \phi_k - \nabla \cdot (\mu \nabla \phi_k) - \frac{\partial}{\partial z} \mu_z \frac{\partial \phi_k}{\partial z} + \nabla \cdot \boldsymbol{\phi}_k^s \tag{32}$$

is nonnegative:

$$(A\phi_k, \phi_k) = \int_D \left\| \mu^{\frac{1}{2}} \nabla \phi_k \right\|_2^2 d\mathbf{r} + \frac{1}{2} \int_{S_0} v_k^s \phi_k^2 \left| \mathbf{e}_3 \cdot \mathbf{n} \right| dS +$$

$$\int_D \sigma_k \phi_k^2 d\mathbf{r} + \int_{S_H} \nu_k^s \phi_k^2 dS + \frac{1}{2}\int_{S\cup S_H} |U_n| \phi_k^2 dS \geq 0 \quad (33)$$

Here $(\phi,\eta) = \int_{\mathbf{D}} \phi\,\eta\, d\mathbf{r}$ is the inner product and $\|\varphi\|_2 = \left(\int_{\mathbf{D}} \varphi^2 d\mathbf{r}\right)^{1/2}$ is the norm. Then it can be shown [19] that

$$\|\phi_k\|_2 \leq T \cdot \max_{0\leq t\leq T} \|f_k(\mathbf{r},t)\|_2 + \|\phi_k^0\|_2 \quad (34)$$

The boundary conditions are also physically appropriate, since the integration of (19) over domain $\mathbf{D}$ leads to the mass balance equation

$$\frac{\partial}{\partial t}\int_{\mathbf{D}} \phi_k d\mathbf{r} = \sum_{i=1}^{N} q_{ik}(t) - \int_{S^+\cup S_H^+} U_n \phi_k dS - \int_{\mathbf{D}} \sigma_k \phi_k d\mathbf{r} - \int_{S_0} \nu_k^s \phi_k |\mathbf{e}_3 \cdot \mathbf{n}| dS \quad (35)$$

Thus, the total mass of the pollutants increases due to the nonzero emission sources $q_{ik}(t)$, and decreases because of advective outflow across $S^+ \cup S_H^+$, chemical transformations and sedimentation of pollutants on the ground.

Finally, in order to get the numerical solution of dispersion model (19)-(27) and its adjoint, the balanced and absolutely stable second-order finite-difference schemes based on the application of splitting method and Crank-Nicolson scheme are used [20,21].

3.2 Adjoint model

Clearly, the concentration of a pollutant ϕ_k in a point $(\mathbf{r},t)$, as well as the mean concentration

$$\mathbf{J}_{\Omega,\tau}^k = \frac{1}{|\Omega|\tau}\int_{T-\tau}^{T}\int_{\Omega} \phi_k(\mathbf{r},t) d\mathbf{r} dt \quad (36)$$

in a zone $\Omega \subset \mathbf{D}$ and time interval $(T-\tau,T)$ depend on the pollution emission rates of dispersion model (19)-(27). However, such relationships are implicit, whereas for the solution of some problems of control and decision-making is desirable to establish explicit relationships between the emission levels and the above-mentioned functional (concentration in a point, the average concentration, etc.). Such explicit relations can be obtained by means of solutions of the adjoint dispersion model [14,22] and Lagrange's identity (duality principle) [22,23]. The adjoint approach not only provides an effective and economical technique for the sensitivity study of the model solution with respect to variations in its parameters [24], but also permits to solve such important problems as optimal allocation of new industries [14], control of pollution emissions [16,25,26], detection of the industrial plants that violate prescribed emission rates [27], remediation of aquatic systems polluted by oil [28,29], etc.

To this end, we now consider in the domain $\mathbf{D}\times(0,T)$ the adjoint model associated with the original dispersion model by means of the Lagrange identity $(A\phi,g) = (\phi,A^*g)$:

$$-\frac{\partial g_k}{\partial t} - \mathbf{U}\cdot\nabla g_k + \sigma_k g_k - \nabla\cdot(\mu\nabla g_k) - \frac{\partial}{\partial z}\mu_z\frac{\partial g_k}{\partial z} - \nabla\cdot\mathbf{g}_k^s = p(\mathbf{r},t) \quad (37)$$

$$\mathbf{g}_k^s = -\nu_k^s g_k \mathbf{e}_3 \text{ in } \mathbf{D} \quad (38)$$

$$g_k(\mathbf{r},T) = 0 \text{ in } \mathbf{D} \quad (39)$$

$$\mu\nabla g_k\cdot\mathbf{n} + U_n g_k = 0 \text{ on } S^+ \quad (40)$$

$$\mu\nabla g_k\cdot\mathbf{n} = 0 \text{ on } S^- \quad (41)$$

$$\mu\nabla g_k\cdot\mathbf{n} + \mathbf{g}_k^s\cdot\mathbf{n} = 0 \text{ on } S_0 \quad (42)$$

$$\mu_z\frac{\partial g_k}{\partial z} + U_n g_k = 0 \text{ on } S_H^+ \quad (43)$$

$$\mu_z\frac{\partial g_k}{\partial z} = 0 \text{ on } S_H^- \quad (44)$$

We note that the adjoint model (37)-(44) being solved backward in time (from $t=T$ to $t=0$) also has a unique solution, which continuously depends on the forcing $p(\mathbf{r},t)$. This fact can be immediately shown by using the substitution $t' = T - t$ and comparing the resulting model with the original dispersion model [19].

The concrete form of forcing $p(\mathbf{r},t)$ in (37) depends on the functional under consideration. In particular, the forcing

$$p(\mathbf{r},t) = \begin{cases} \frac{1}{|\Omega|\,\tau}, & (\mathbf{r},t)\in\Omega\times(T-\tau,T) \\ 0, & (\mathbf{r},t)\notin\Omega\times(T-\tau,T) \end{cases}$$

allows us to evaluate the mean concentration (36) in the zone $\Omega\subset\mathbf{D}$ and time interval $(T-\tau,T)$. Indeed, if the last forcing is used then combining the solutions of dispersion model and its adjoint [14,22], one can obtain an alternative (dual to (36)) formula for the mean concentration

$$\mathbf{J}_{\Omega,\tau}^k = \sum_{i=1}^{N}\int_0^T g_k(\mathbf{r}_i,t) q_{ik}(t) dt + \int_{\mathbf{D}} g_k(\mathbf{r},0)\phi_k^0(\mathbf{r}) d\mathbf{r}, \quad k = 1,\mathrm{K}\,,K \quad (45)$$

in the domain $\Omega\times(T-\tau,T)$. The estimate (45) is the required formula that explicitly relates $\mathbf{J}_{\Omega,\tau}^k$ with

$\phi_k^0(\mathbf{r})$ and $q_{ik}(t)$. Although the solution g_k of adjoint model depends on the meteorological conditions and the parameters σ_k, v_k^s, τ and $|\Omega|$, it is independent of the emission rates $q_{ik}(t)$ and initial pollution distribution $\phi_k^0(\mathbf{r})$. This solution g_k is nonnegative and serves in (45) as the weight function for $\phi_k^0(\mathbf{r})$ and $q_{ik}(t)$. The last integral in (45) determines the contribution of ϕ_k^0 into $\mathbf{J}_{\Omega,\tau}^k$.

On the other hand, if the forcing $p(\mathbf{r},t)=\delta(\mathbf{r}-\mathbf{r}_s)\delta(t-t_j)$ is used in the adjoint model (37)-(44), then the resulting functional

$$\phi_k(\mathbf{r}_s,t_j)=\sum_{i=1}^{N}\int_0^T g_k(\mathbf{r}_i,t)q_{ik}(t)dt+\int_{\mathbf{D}} g_k(\mathbf{r},0)\phi_k^0(\mathbf{r})d\mathbf{r},\quad k=1,\mathrm{K},K \tag{46}$$

represents the explicit dependence of the concentration of pollutant ϕ_k in point $(\mathbf{r}_s,t_j)$ on the emission rates $q_{ik}(t)$ and initial pollution distribution ϕ_k^0. Note that equations (9), (10) and (11) are particular cases of result (46). Obviously, estimation (46) is the limiting case of equation (45) when τ tends to zero and zone Ω shrinks to a point.

IV. Numerical approach and examples

4.1 Formulation of the quadratic programming problem

In order to calculate numerical solutions of variational problem (16)-(18), and estimate the unknown emission rate $Q(t)$ of new source, we introduce here a quadratic programming problem. To this end, we define a finite set of basic functions (linear splines) as follows:

$$\gamma_l(t)=\begin{cases}1+(t-t_l)/\Delta t, & t_{l-1}\le t\le t_l\\ 1-(t-t_l)/\Delta t, & t_l\le t\le t_{l+1}\\ 0, \text{ otherwise}\end{cases} \tag{47}$$

where $t_l=l\cdot\Delta t$ are the nodes of a regular mesh in interval $[0,T]$, ($l=0,1,...,L$, $\Delta t\cdot L=T$), and the functions γ_0 and γ_L are equal to zero outside of this interval. The basic functions (47) have the following useful properties:

$$\gamma_l(t_j)=\begin{cases}1, & l=j\\ 0, & l\ne j\end{cases},\quad l,j=0,1,...,L \tag{48}$$

$$\int_0^T\left(\frac{d\gamma_0}{dt}\right)^2dt=\int_0^T\left(\frac{d\gamma_L}{dt}\right)^2dt=\frac{1}{\Delta t},\ \int_0^T\left(\frac{d\gamma_l}{dt}\right)^2dt=\frac{2}{\Delta t},\quad l=1,...,L-1 \tag{49}$$

and

$$\int_0^T\left(\frac{d\gamma_{l-1}}{dt}\right)\left(\frac{d\gamma_l}{dt}\right)dt=-\frac{1}{\Delta t},\quad l=1,...,L \tag{50}$$

Besides,

$$\int_0^T\left(\frac{d\gamma_j}{dt}\right)\left(\frac{d\gamma_l}{dt}\right)dt=0,\quad |j-l|>1 \tag{51}$$

We now propose the emission rate $Q(t)$ in the following form:

$$Q(t)=\sum_{l=1}^{L-1}Q_l\gamma_l(t) \tag{52}$$

where, due to property (48), $Q_0=Q(0)=0$, $Q_L=Q(T)=0$ and $Q_j=Q(t_j)$, $j=1,...,L-1$. Substituting equation (52) into functional (16), and taking into account the properties (49)-(51), we get

$$J(\mathbf{Q})=\frac{1}{2}\mathbf{Q}^tH\mathbf{Q}$$

where $\mathbf{Q}=(Q_1,Q_2,...,Q_{L-1})^t$ is a vector in the space $\mathbf{R}^{L-1}$, and H is a tridiagonal, symmetric and positive-definite matrix of order $L-1$, whose entries are defined by the inner products $H_{jl}=\int_0^T\gamma_j'\gamma_l'dt$, that is,

$$H=\frac{1}{\Delta t}\begin{pmatrix}2&-1&0&0&\mathrm{L}&0\\-1&2&-1&\mathrm{O}&&0\\0&-1&2&\mathrm{O}&0&\mathrm{M}\\0&\mathrm{O}&\mathrm{O}&\mathrm{O}&-1&0\\\mathrm{M}&&0&-1&2&-1\\0&0&\mathrm{L}&0&-1&2\end{pmatrix}$$

Without loss of generality, we assume that the sampling time moments coincide with the mesh nodes in the interval $[0,T]$ and therefore $M=L$. Thus, substituting equation (52) into integral equation (11), we get

$$\phi(\mathbf{R},t_j)=\sum_{l=1}^{j}a_{jl}Q_l,\quad j=1,..,L \tag{53}$$

where

$$a_{jl}=\int_{t_{l-1}}^{t_{l+1}}\gamma_l(t)g_j(\mathbf{r}_0,t)dt \tag{54}$$

Taking into account these approximations, the corresponding quadratic programming problem for the variational problem (16)-(18) can be posed as follows:

$$\text{minimize} \quad J(\mathbf{Q}) = \frac{1}{2}\mathbf{Q}^t H \mathbf{Q} \tag{55}$$

$$\text{subject to: } Q_l \geq 0, \quad l = 1,...,L-1, \; Q_0 = Q_L = 0 \tag{56}$$

$$\text{and} \quad -\varepsilon \leq \phi_j - \sum_{l=1}^{j} a_{jl} Q_l, \leq \varepsilon, \quad j = 1,..,L \tag{57}$$

where coefficients a_{jl} are given by (54). Note that, in order to simplify the constraints (57), we can calculate the coefficients a_{jl} by using second order formulas (like trapezoidal rule):

$$a_{jj} = \frac{\Delta t}{2} g_j(\mathbf{r}_0, t_j), \; j = 1,...,L\text{-}1, \quad \text{and} \quad a_{jl} = \Delta t \cdot g_j(\mathbf{r}_0, t_l), \quad l = 1,...,L-1, \; l < j \leq L \tag{58}$$

Note that the quadratic programming problem (55)-(57) have a solution because the feasibility space given by (56)-(57) is a nonempty compact set in the space $\mathbf{R}^{L-1}$, and the objective function $J(\mathbf{Q})$ is continuous. Besides, since H is a positive-definite matrix then $J(\mathbf{Q})$ is a strictly convex function, and hence, the solution of (55)-(57) is unique in the convex feasibility space (56)-(57) [30]. Such a solution can be calculated using the *quadprog* routine of MATLAB [15], as soon as the adjoint functions have been determined.

As for the *quadprog* routine, we can note that in the case when the problem, we are going to solve with *quadprog*, has only upper and lower bounds, i.e., no linear inequalities or equalities are specified, the default algorithm is the large-scale method. Moreover, if such a problem has only linear equalities, i.e., no upper and lower bounds or linear inequalities are specified, the default algorithm of *quadprog* is also the large-scale method. This method is a subspace trust-region method based on the interior-reflective Newton method described in [31]. Each iteration involves the approximate solution of a large linear system using the method of preconditioned conjugate gradients (PCG). Otherwise, medium-scale optimization is used, and *quadprog* uses an active set method, which is also a projection method, similar to that described in [32]. This method determines an initial feasible solution by solving a linear programming problem. Due to the structure of quadratic programming problem (55)-(57), the second method of *quadprog* routine is applied in the examples.

4.2 Numerical examples

In order to show the performance of quadratic programming problem (55)-(57) in the approximation of the solution of inverse problem, some numerical examples are presented in this section. The examples take into account the errors in the data, since the anomaly of concentration $\phi = \varphi - \hat{\varphi}$ is the difference between sample values of the concentration and solution values of the dispersion model, and both types of data have errors. Due to unavoidable errors in real data, as well as by the fact that the inverse problem is ill-posed, the determination of intensity $Q(t)$ of new source is not an easy task, and the formulations (16)-(18) and (55)-(57) are necessary as we shall see. In the synthetic examples of this section, the errors are uniformly distributed.

4.2.1 Example 1

We begin with a simple dispersion model of dimension zero (box model) for the pollutant concentration anomaly [17],

$$\frac{d\phi}{dt} + (\sigma + \frac{1}{V} u a^2)\phi = \frac{1}{V} Q(t), \quad 0 < t < T \tag{59}$$

$$\phi(0) = 0 \tag{60}$$

It is assumed here that at any moment, the pollutant forms an homogeneous mixture with the air in the region $\mathbf{D} = [0,a] \times [0,a] \times [0,a]$, which is a box of length a and volume $V = a^3$. The steady velocity u of the wind is horizontal, and σ denotes the chemical transformation coefficient for the pollutant. Note that problem (59)-(60) is a particular case of the general mass balance equation (35) for a single pollutant, without taking into account the sedimentation ($v_k^s = 0$), and considering just one point source in region $\mathbf{D}$ with emission rate $Q(t)$.

We will try to assess $Q(t)$ directly from (59)-(60). To this end, two well known finite central difference formulas of second order of approximation are used in (59) to get

$$\frac{\phi_{j+1} - \phi_{j-1}}{2\Delta t} + r \frac{\phi_{j+1} + \phi_{j-1}}{2} = \frac{1}{V} Q_j$$

where $r = \sigma + \frac{1}{V} u a^2$, Q_j approximates $Q(t_j)$ at the moments $t_j = j\Delta t$ ($j = 0,1,...,L$), $\Delta t = T/L$ is the time period between sample data, and $\{\phi_j\}_{j=0}^{L}$ is the time series of the anomaly of pollutant concentration. Grouping the terms in the last equation, we get

$$Q_j = \frac{V}{2}\left\{\left(r + \frac{1}{\Delta t}\right)\phi_{j+1} + \left(r - \frac{1}{\Delta t}\right)\phi_{j-1}\right\},$$

$$j = 1,...,L-1 \tag{61}$$

In order to test equation (61) we consider the input signal (emission rate)

$$Q(t)=\begin{cases}0, & 0\le t<2\\ q, & 2\le t\le 4\\ 0, & 4<t\le 10\end{cases} \tag{62}$$

and the output signal (pollutant concentration)

$$\phi(t)=\begin{cases}0, 0\le t<2\\ \frac{q}{rV}\left(1-\exp(-r(t-2))\right), 2\le t\le 4\\ \frac{q}{rV}\left(\exp(-r(t-4))-\exp(-r(t-2))\right), 4<t\le 10\end{cases} \tag{63}$$

for dispersion model (59)-(60). Figure 2 shows both signals for the following parameters: $a=1\,km$, $u=0.5\,kmh^{-1}$, $\sigma=0.0001\,h^{-1}$, $T=10\,h$ and $q=100\,kgh^{-1}$.

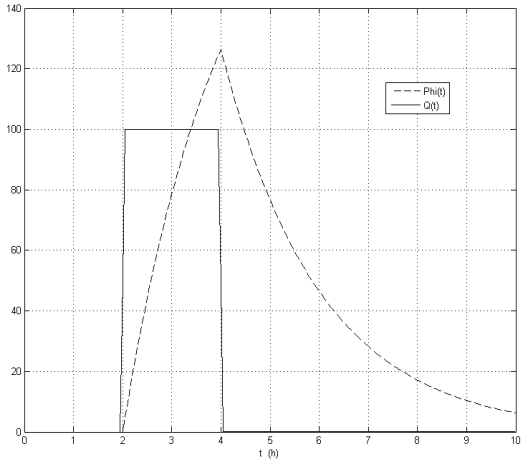

Fig. 2 Emission rate $Q(t)$ and anomaly of pollutant concentration ϕ given by (62) and (63).

Taking into account equation (63), the synthetic time series is given as

$$\phi_j=\phi(t_j)+\delta\phi_j,\ \ \Delta t=T/L,\ t_j=j\Delta t,\ \ j=0,1,...,L \tag{64}$$

where the perturbations (errors) $\left\{\delta\phi_j\right\}_{j=0}^{L}$ are uniformly distributed. Figure 3 shows an example of the time series calculated with (64) for $\Delta t=0.05\,h$, where the values $\delta\phi_j$ were randomly chosen from a uniform distribution in the interval $(-0.5,0.5)$. In this example, the amplitude of errors was normalized in order to be up to about 15% of the maximum output signal ϕ.

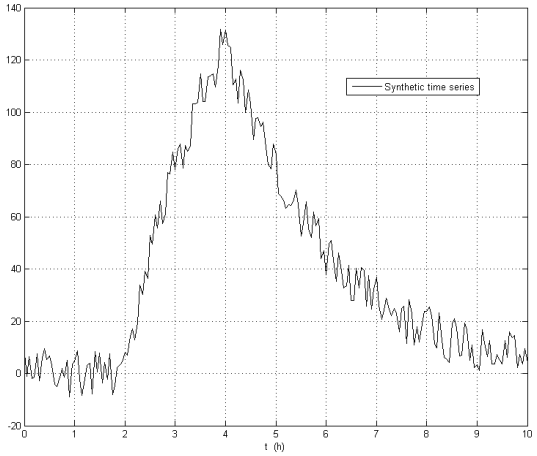

Fig. 3 Synthetic time series $\left\{\phi_j\right\}_{j=0}^{L}$ for the anomaly of pollutant concentration.

The result of applying the scheme (61) with the synthetic time series shown on Figure 3 is plotted in Figure 4. It is clearly seen that the amplitude of errors has grown up to 175% (since the inverse problem is ill-posed), and hence, the emission rate $Q(t)$ has been lost. This result can be explained as follows. Substituting (64) into equation (61), we get

$$\left|Q(t_j)-Q_j\right|\le V\delta\phi\left(r+\frac{1}{\Delta t}\right)+o\left((\Delta t)^2\right)$$

where $\delta\phi=\max_j\left\{\left|\delta\phi_j\right|\right\}$. Thus, the error of amplitude increases with volume V and sample frequency $1/\Delta t$. In particular, if the number of observations in the interval $(0,T)$ increases, then $\Delta t\to 0$ and therefore the estimate of $Q(t)$ using the scheme (61) becomes worse. Other methods based on the finite differences are performed similarly to scheme (61) [5].

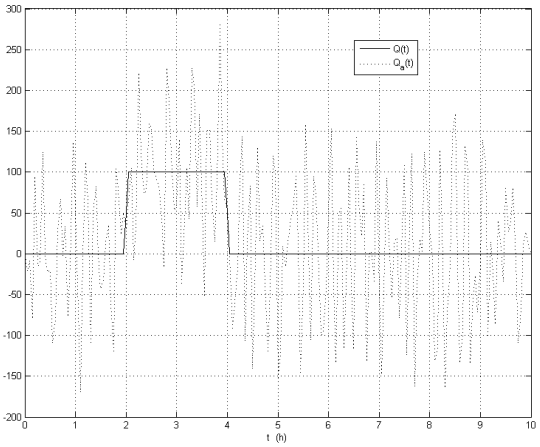

Fig. 4 Emission rate $Q_a(t)$ obtained from scheme (61) and emission rate $Q(t)$ given by (62).

We now consider the quadratic programming problem (55)-(57) plus (58) to solve this particular inverse problem. For the dispersion model (59)-(60), the explicit relationship between the unknown emission rate $Q(t)$ and the pollutant concentration anomaly ϕ can be established through the solution of equation (59) by means of Laplace transform [33]. Such a solution takes the following integral form

$$\phi(t)=\frac{1}{V}e^{-rt}\int_0^t Q(\xi)e^{r\xi}d\xi,\ \ 0<t<T \tag{65}$$

where $r=\sigma+\frac{1}{V}ua^2$. Then the corresponding equations (11) can be written as

$$\phi(t_j)=\int_0^{t_j}Q(t)g_j(t)dt,\ \ t_j=j\Delta t,\ \ j=1,...,L$$

where the kernel of integral equation is $g_j(t)=\frac{1}{V}e^{-r(t_j-t)}$, $0<t<t_j$. Besides, due to the steady dispersion conditions considered in this example, the basic kernel is $g_\alpha(t)=\frac{1}{V}e^{-r(T-t)}$, $0<t<T$, which possesses the property $g_\alpha(T-t_j+t)=g_j(t)$ for $t\le t_j$ (see equation (12)). The Figure 5 shows the input signal (62) and solution of quadratic programming problem (55)-(57) with (58). Such a solution was calculated by using the *quadprog* routine of MATLAB [15]. In this example, the time series $\{\phi_j\}_{j=0}^{L}$ is generated by means of (64) and it is plotted in Figure 3. It is clear that the numerical solution of regularization method (16)-(18) has improved the estimation of the emission rate found through the simple scheme (61).

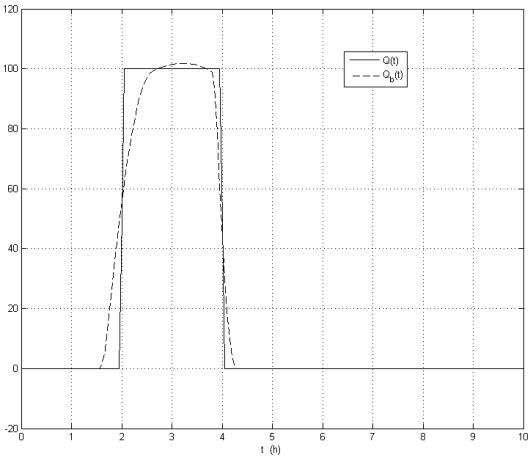

Fig. 5 Emission rate $Q_b(t)$ obtained from (55)-(57) and emission rate $Q(t)$ given by (62).

On the other hand, the numerical experiments show that if the parameter $\varepsilon\to+\infty$ then the solution of (55)-(57) is smoothed and tends to zero, since the feasibility space expands so that it contains the global minimum of the objective function J. At the other extreme, when $\varepsilon\to\delta\phi^+$ where $\delta\phi=\max_j\{|\delta\phi_j|\}>0$, the regularization leads to the best approximation of the emission rate. The Figure 6 shows the solution of (55)-(57) for the synthetic data presented in Figure 3, where $\varepsilon=e\cdot\delta\phi$, $e=1.8,\ 1.4$ y 1.0. The relative errors of the corresponding approximations are $0.32,\ 0.28$ and 0.19. Finally, when $\varepsilon<\delta\phi$, the solution of inverse problem is unstable (the oscillations are shown in Figure 4). Since for real data the value of $\delta\phi$ is unknown, such a change in the behavior of the solution indicates the point where the optimum value of $\delta\phi$ has been reached.

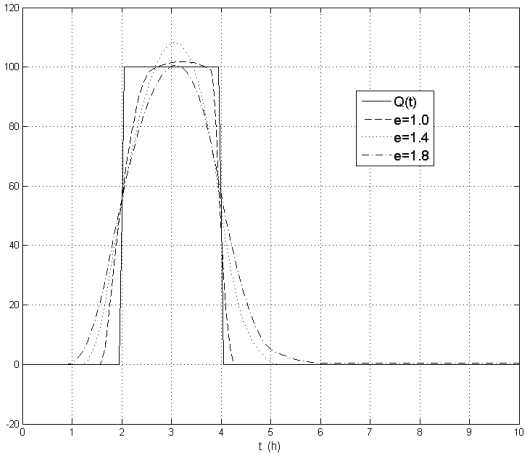

Fig. 6 Behavior of the solution of quadratic programming problem (55)-(57) when $\varepsilon\to\delta\phi^+$.

4.2.2 Example 2

We now consider a one-dimensional model of diffusion for the pollutant concentration anomaly $\phi=\phi(x,t)$:

$$\frac{\partial\phi}{\partial t}-\mu\frac{\partial^2\phi}{\partial x^2}=Q(t)\delta(x-x_0),\ \ 0<x<1,\ \ 0<t<T \tag{66}$$

$$\mu\frac{\partial\phi}{\partial x}(0,t)=0,\ \ t>0 \tag{67}$$

$$\mu\frac{\partial\phi}{\partial x}(1,t)=-\varsigma\phi(1,t),\ \ t>0 \tag{68}$$

$$\phi(x,0)=0,\ \ 0<x<1 \tag{69}$$

where $\mu>0$ is the diffusion coefficient and x_0 is the location of a point source in domain $\mathbf{D}=(0,1)$ with

emission rate $Q(t)$. The condition (67) means that there is no pollutant flow at $x=0$ (the closed boundary), and condition (68) means that at $x=1$ (the open boundary) the flow of pollutant is proportional to ϕ with $\varsigma>0$. As a result, the mass of pollutant in $\mathbf{D}$ increases due to the emission rate Q and decreases because of the outflow at the boundary point $x=1$ according to the following mass balance equation:

$$\frac{\partial}{\partial t}\int_0^1 \phi(x,t)dx = Q(t) - \varsigma\phi(1,t)$$

Similarly to the general dispersion model (19)-(27), the diffusion model (66)-(69) is well posed, since its solution exists, is unique and continuously depends on the initial condition and forcing. This follows from the fact that the problem operator A defined as

$$A\phi = -\mu\frac{\partial^2\phi}{\partial x^2}$$

is nonnegative:

$$(A\phi,\phi) = \int_0^1 \mu\left(\frac{\partial\phi}{\partial x}\right)^2 dx + \varsigma\phi^2(1,t)$$

By solving the Sturm-Liouville problem for the diffusion model (66)-(69), its non-negative solution can be expressed by Fourier series [34] as:

$$\phi(x,t) = \sum_{k=1}^{\infty}\left(\int_0^t d_k Q(\xi)e^{-\lambda_k^2\mu(t-\xi)}d\xi\right)\cos(\lambda_k x) \quad (70)$$

where

$$d_k = 2\cos(\lambda_k x_0)[1+\frac{\mu}{\varsigma}\sin^2(\lambda_k)]^{-1},\ \ k=1,2,\mathrm{K}\ ,$$

$\{\cos(\lambda_k x)\}_{k=1}^{\infty}$ is an orthogonal system of functions (eigenfunctions), and the corresponding frequencies λ_k (eigenvalues) are the roots of equation

$$\varsigma\cos(\lambda) - \mu\lambda\sin(\lambda) = 0\,.$$

Such frequencies can be efficiently calculated by Newton's method [35].

Note that for the numerical experiments of this section we used the Fejér series [36,37], calculated from the Fourier series (70), in order to have the pointwise convergence to $\phi(x,t)$. That is, in practice we consider in (70) the Fejér coefficients:

$$d_k = \left(1-\frac{(k-1)}{K}\right)d_k,\ \ k=1,2,\mathrm{K}\ K\ ,$$

instead of d_k, where K is the truncation number of the series.

On the other hand, the application of Lagrange identity $(A\phi,g)=(\phi,A^*g)$ to the diffusion model (66)-(69) leads to the following adjoint model

$$-\frac{\partial g}{\partial t} - \mu\frac{\partial^2 g}{\partial x^2} = p(x,t),\ \ 0<x<1,\ \ 0<t<T \quad (71)$$

$$\mu\frac{\partial g}{\partial x}(0,t) = 0,\ \ t>0 \quad (72)$$

$$\mu\frac{\partial g}{\partial x}(1,t) = -\varsigma g(1,t),\ \ t>0 \quad (73)$$

$$g(x,T) = 0,\ \ 0<x<1 \quad (74)$$

where the forcing is defined as $p(x,t)=\delta(x-\mathbf{R})\delta(t-t_j)$, and $\mathbf{R}$ is the monitoring point in domain $\mathbf{D}$.

As it was shown in Section II, the use of solution of adjoint problem (71)-(74) leads to the formula

$$\phi(\mathbf{R},t_j) = \int_0^{t_j} Q(t)g_j(x_0,t)dt$$

that corresponds to equation (11). Here, the adjoint function g_j can be expressed as

$$g_j(x_0,t) = \sum_{k=1}^{\infty} c_k e^{-\lambda_k^2\mu(t_j-t)}\cos(\lambda_k x_0),\ \ t\le t_j \quad (75)$$

where

$$c_k = 2\cos(\lambda_k\mathbf{R})[1+\frac{\mu}{\varsigma}\sin^2(\lambda_k)]^{-1},\ \ k=1,2,\mathrm{K}$$

are Fourier coefficients of series (75).

In the numerical experiments, as it was mentioned before, we consider in the series (75) the Fejér coefficients

$$c_k = \left(1-\frac{(k-1)}{K}\right)c_k,\ \ k=1,2,\mathrm{K}\ K\ ,$$

instead of c_k, where K is the truncation number of the series.

In this example, due to the steady dispersion conditions, the basic kernel

$$g_\alpha(x_0,t)=\sum_{k=1}^{\infty}c_k e^{-\lambda_k^2\mu(T-t)}\cos(\lambda_k x_0),\quad 0<t<T \tag{76}$$

possesses the property $g_\alpha(x_0,T-t_j+t)=g_j(x_0,t)$ for $t\le t_j$ (see equation (12)).

The Figure 7 shows the input signal $Q(t)$ given by (62) and the corresponding output signal $\phi(\mathbf{R},t)$ calculated according to (70) for the following parameters: $x_0=0.3\ km$, $\mathbf{R}=0.8\ km$, $\varsigma=1.0\ km$, $\mu=0.5\ km^2$, $T=10\ h$, $q=100\ kgh^{-1}$ and $K=10000$.

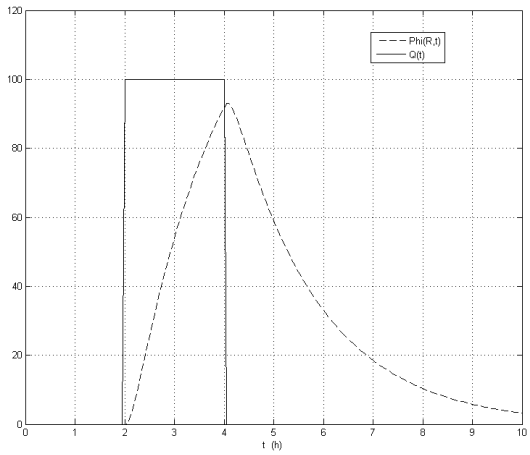

Fig. 7 Emission rate $Q(t)$ given by (62) and concentration ϕ at $x=\mathbf{R}$ calculated with (70).

In the Figure 8 is shown the basic kernel $g_\alpha(x_0,t)$ calculated according to (76) and two examples of adjoint functions $g_j(x_0,t)$ (see (75)). As it can see, each adjoint function is a displacement in time of the basic kernel (equation (12)).

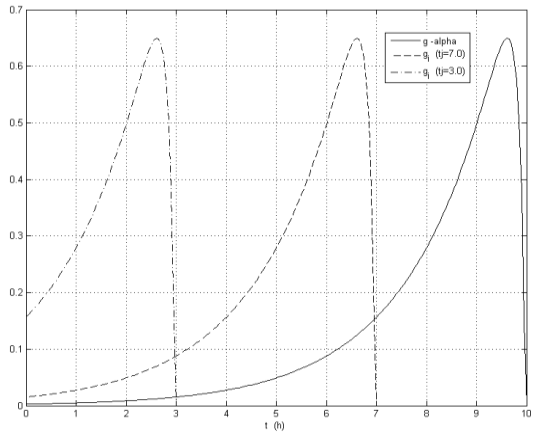

Fig. 8 Basic kernel $g_\alpha(x_0,t)$ and adjoint functions $g_j(x_0,t)$ for $t_j=3.0$ and $t_j=7.0$.

Once the adjoint functions have been obtained by means of the basic kernel we can consider the quadratic programming problem (55)-(57) with (58) to solve the inverse problem. As in example 1, the synthetic time series $\{\phi_j\}_{j=0}^{L}$ for the anomaly of pollutant concentration is generated by formula (64), where the errors $\{\delta\phi_j\}_{j=0}^{L}$ are uniformly distributed:

$$\phi_j=\phi(\mathbf{R},t_j)+\delta\phi_j,\quad \Delta t=T/L,$$
$$t_j=j\Delta t,\quad j=0,1,...,L.$$

The Figure 9 shows the time series calculated for $\Delta t=0.05\ h$, where the values $\delta\phi_j$ were randomly chosen from a uniform distribution in the interval $(-0.5,0.5)$. In this example, the amplitude of errors was normalized so as to be up to about 15% of the maximum output signal $\phi(\mathbf{R},t)$. The Figure 10 shows the input signal (62) and the behavior of the solutions of quadratic programming problem (55)-(57) with (58) when $\varepsilon\to\delta\phi^+$, where $\varepsilon=e\cdot\delta\phi$, $\delta\phi=\max_j\{|\delta\phi_j|\}>0$ and $e=8.0,\ 4.0,\ 2.0,\ 1.0$. These solutions were calculated using the *quadprog* routine of MATLAB [15]. The behavior of solutions with respect to parameter ε is similar to that described in example 1. Indeed, for $\varepsilon=\delta\phi$ we get the best approximation of the emission rate $Q(t)$ that the method can provide.

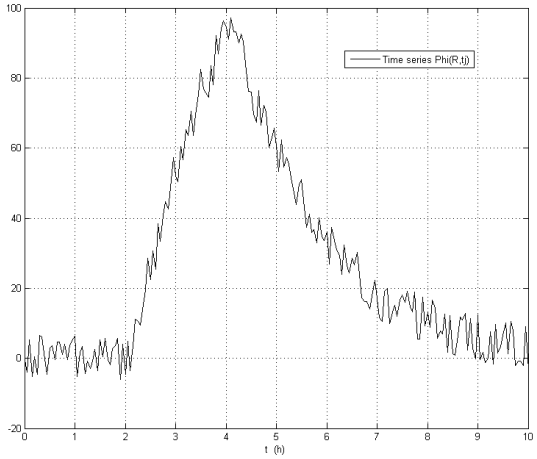

Fig. 9 Synthetic time series $\{\phi_j\}_{j=0}^{L}$ for the anomaly of pollutant concentration at $x=\mathbf{R}$.

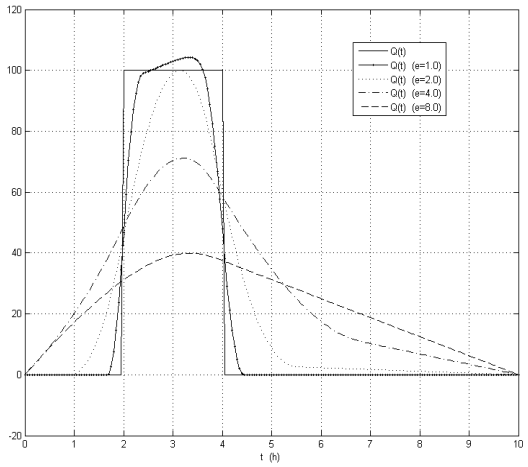

Fig. 10 Behavior of the solution of quadratic programming problem (55)-(57) when $\varepsilon \to \delta\phi^{+}$.

This example is interesting because a parcel of the pollutant released at $x_0 = 0.3$ and moment t, is propagated by diffusion in both directions (to the left and to the right). The part of this parcel that has been dispersed to the right, is registered at the monitoring point $\mathbf{R} = 0.8$ at moment $t_1 > t$, while the other part of the parcel is registered at moment $t_2 > t_1$ like a feedback process (reflection). This dynamical phenomenon is the consequence of the presence of closed boundary at $x = 0$. The problem is that such a feedback can be erroneously interpreted as the presence of one more source located to the left from the location of the true source, and that this false source can generate an additional impulse in $Q(t)$ outside of the time interval $(2,4)$. However, thanks to information provided by the adjoint solutions, this false interpretation is recognized by the regularization process, and finally the only source is correctly determined (see Figure 10).

V. Conclusions

A variational problem to find the unknown non-stationary emission rate $Q(t)$ of a point source is formulated. This method requires at least one time series of the anomaly of pollutant concentration in a monitoring site. It is shown that the use of adjoint functions is fundamental to establish integral constraints on variational problem, which explicitly relate such anomalies and $Q(t)$. Analytical expressions obtained from these relations allow estimating the emission rate in two particular cases, namely when $Q(t)$ is constant and when $Q(t)$ is an impulse in time (as in case of explosion). In the general case, the minimization of the L2-norm of first derivative of $Q(t)$ is a useful regularization for unstable inverse problems, since such regularization filters the errors in the data and permits to obtain a smooth approximation of $Q(t)$. This approximation of $Q(t)$ is the solution of a discrete problem associated with the variational problem. Besides, such a discrete problem represents a simple quadratic programming problem which is solved efficiently by *quadprog* routine of MATLAB. Numerical experiments show that for determining the emission rate, the optimal value of parameter ε is the maximum value of the error in the data of the anomaly of pollutant concentration.

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