Algorithms for the inverse modelling of transport and transformation of atmospheric pollutants

A V Penenko[1,2]

[1] Institute of Computational Mathematics and Mathematical Geophysics SB RAS, ICM&MG SB RAS, Prospekt Akademika Lavrentieva 6, Novosibirsk, 630090, Russian Federation
[2] Novosibirsk State University, Pirogova st 1, Novosibirsk, 630090, Russian Federation

a.penenko@yandex.ru

Abstract. When studying air quality, a key parameter for assessment and forecast is information on emission sources. In applications, this information is not fully available and can be compensated by air quality monitoring data and inverse modelling algorithms. Because of the rapid development of satellite chemical monitoring systems, they are becoming more useful in air quality studies. Such systems provide measurements in the form of concentration field images. In this paper, we consider an inverse source problem and a corresponding data assimilation problem for a chemical transport model. The problem of assimilation of data given as images is considered as a sequence of linked inverse source problems. Each individual inverse problem solution is carried out by variational and Newton-Kantorovich type algorithms. In the numerical experiment presented, an emission source of a primary pollutant is reconstucted via the concetration field of a secondary pollutant. Both data assimilation and inverse problem solution algorithms are capable of approximating the unknown source.

1. Introduction

The development of algorithms for atmospheric chemical composition monitoring data assimilation is a strategic task in the context of re-industrialization and increased public interest in the environmental conditions. The chemical composition of the atmosphere is changing all the time and depends on many factors of natural and man-made origin. In turn, it has a significant impact on the entire biosphere and, in particular, on the well-being and life expectancy [1, 2].

Information on the concentration of chemicals can be obtained through various monitoring systems. However, it is still impossible to measure all values, including future pollutant distributions. Nevertheless, this information is necessary and important in planning and decision-making on a wide range of issues, from the design of new industrial facilities or environmental impact assessment of various technological events to the choice of personal transportation routes. In this regard, there is a need to obtain estimates of the distribution of chemicals in the atmosphere, as well as the forecast of their changes. The key problem in the modeling of the atmospheric chemical composition dynamics is the description of emission sources [3, 4]. In the near future, the state registration system should account for all major industrial, rural, and other economic objects that emit pollutants into the atmosphere. However, there are sources, such as small households and transport, that have random characteristics and cannot be strictly accounted. One of the possible ways is to use the data

assimilation methods. Overviews of modern data assimilation algorithms can be found in [5, 6]. With assimilation algorithms it is possible to estimate unknown parameters of the mathematical model, which can be used both to predict the composition of the atmosphere and to assess the current state. Currently the operational (and quasi-operational) systems use the variational approach for data assimilation (e.g. [7]). The disadvantage of the variational approach is that it can be parallelized only at the level of the solutions of direct and adjoint problems needed for obtaining the gradient of the cost functional. We consider the data assimilation problem as a series of linked inverse problems. Within the framework of this concept, we can try various algorithms for solving the individual inverse problems, not necessarily variational ones.

To fit the computational context, we are developing an alternative algorithm based on the use of sensitivity operators constructed of an ensemble of quasi-independent adjoint problem solutions [8, 9, 10]. The sensitivity operator allows transforming the inverse problem stated as a system of nonlinear PDEs to a family of operator equations depending on the given set of functions in the space of the measurement results. The resulting operator equations are treated by the relevant operator equation methods. This algorithm's parallelization potential is higher due to the independent solution of the adjoint problems of the ensemble.

The objective of the paper is to present the first results on the data assimilation of concentration field images in a 2D atmospheric chemistry transport model with the algorithm based on the sensitivity operator inversion and to compare them with the solution of the variational algorithm.

2. Methods and algorithms

2.1. Data Assimilation Problem

Let us consider a two-dimensional spatial domain $\Omega = [0, X] \times 0, Y]$, a spatio-temporal domain $\Omega_T = [0, T] \times \Omega$, and an advection-diffusion-reaction model on the domains $\Omega_{[\underline{\theta}, \bar{\theta}]} = [\underline{\theta}, \bar{\theta}] \times \Omega \subset \Omega_T$.

$$\frac{\partial \varphi_l}{\partial t} + div(\vec{u}\varphi_l) - div(diag(\mu) grad \varphi_l) + P_l(t, \varphi)\varphi_l = \Pi_l(t, \varphi) + f_l + r_l, \quad (x, y, t) \in \Omega_{[\underline{\theta}, \bar{\theta}]}, \tag{1}$$

$$\varphi_l = g_l, (x, y, t) \in \partial\Omega_{[\underline{\theta}, \bar{\theta}]} / \{t = \underline{\theta}\}, \tag{2}$$

$$\varphi_l = \varphi_l^0, (x, y) \in \Omega, t = \underline{\theta}, \tag{3}$$

for $l = 1, \ldots, N_c$, where N_c is the number of considered substances, $\varphi_l(x, y, t)$ denotes the concentration of the l^{th} substance at a point $(x, y, t) \in \Omega_{[\underline{\theta}, \bar{\theta}]}$, and φ is the vector of φ_l for $l = 1, \ldots, N_c$. The function $\mu(x, y, t)$ corresponds to the diffusion coefficient, $\vec{u}(x, y, t)$ denotes the advection speed, f_l, g_l, φ_l^0 are *a priori* source function, boundary conditions, and initial conditions, correspondingly, r_l is a control function (uncertainty). Let $r \in F \subset L_2(\Omega_{[\underline{\theta}, \bar{\theta}]}; \mathrm{R}^{N_c})$, where F is a set of admissible sources. The destruction and production operator elements $P_l, \Pi_l : [0, T] \times \mathrm{R}_+^{N_c} \to \mathrm{R}_+$ are defined by the chemical kinetics system. We assume all the functions and model parameters to be smooth enough for the solutions to exist and the further transformations to make sense.

Direct problem: Given $\underline{\theta}$, f, g, φ^0, r, determine φ from (1)-(3). Let $\varphi[\underline{\theta}, \varphi^0, r]$ denote the solution of the direct problem.

Let us define a set of the measurement results in the interval and the operator $A^{[\underline{\theta}, \bar{\theta}]}$ that maps the uncertainty function (in our case this is r) into the measurement data in the assimilation window $[\underline{\theta}, \bar{\theta}]$:

$$U_{mes}^{[\underline{\theta},\bar{\theta}]} = \left\{ \left\{ \begin{cases} h_l, l \in L_{mes} \\ 0, l \notin L_{mes} \end{cases} \right\}_{l=1}^{N_c} | \, h_l \in L_2(\Omega_{[\underline{\theta},\bar{\theta}]}; \mathrm{R}^{N_c}) \right\}, \tag{4}$$

$$A^{[\underline{\theta},\bar{\theta}]} : \begin{cases} \mathrm{R}^{N_c} \times F \to U_{mes}^{[\underline{\theta},\bar{\theta}]} \\ \{\varphi^0, r\} \to \left\{ \begin{cases} \varphi_l[\underline{\theta}, \varphi^0, r], l \in L_{mes} \\ 0, l \notin L_{mes} \end{cases} \right\}_{l=1}^{N_c}. \end{cases} \tag{5}$$

Inverse problem: Given $\varphi(\underline{\theta})$ and $I^{[\underline{\theta},\bar{\theta}]}$ and some partial information on $\delta I^{[\underline{\theta},\bar{\theta}]}$, find $r^{[\underline{\theta},\bar{\theta}]}$ and $\varphi^{[\underline{\theta},\bar{\theta}]}$ from

$$A^{[\underline{\theta},\bar{\theta}]}(\varphi(\underline{\theta}), r^{[\underline{\theta},\bar{\theta}]}) = I^{[\underline{\theta},\bar{\theta}]} + \delta I^{[\underline{\theta},\bar{\theta}]}. \tag{6}$$

Let us consider a system of the intervals (assimilation windows) $\Theta = \left\{ [\underline{\theta}^k, \bar{\theta}^k] \right\}_{k=0}^{N_w - 1} \subset 0, T]$ such that $\bar{\theta}^k = \underline{\theta}^{k+1}$ and $\underline{\theta}^k < \bar{\theta}^k$, $\underline{\theta}^0 = 0$, $\bar{\theta}^{N_w - 1} = T$.

Data Assimilation Problem: Solve a sequence of N_w inverse problems in assimilation windows Θ. In the k-th problem $\underline{\theta} = 0$, $\bar{\theta} = \bar{\theta}^k$, and the available data are

$$\left\{ I^{[0,T]}(t), t \le \bar{\theta}^k \right\}, \quad k = 1, \ldots, N_w - 1,$$

Find $r(t)$ and $\varphi(t)$ for $t \in 0, T]$.

By analogy with 4DVAR, let us define the 4DIP algorithm solution of the data assimilation problem in the time moment $\bar{\theta}^k$:

$$\phi^{[\underline{\theta}^k, \bar{\theta}^k]}(t) = \begin{cases} \phi^{[\underline{\theta}^{k-1}, \bar{\theta}^{k-1}]}(t), t \le \underline{\theta}^k \\ \phi^{[\underline{\theta}^k, \bar{\theta}^k]}[\phi^{[\underline{\theta}^{k-1}, \bar{\theta}^{k-1}]}(\underline{\theta}^k), r^{[\underline{\theta}^k, \bar{\theta}^k]}](t), \underline{\theta}^k \le t \le \bar{\theta}^k, \\ \phi^{[\theta^{k+1}, T]}[\phi^{[\underline{\theta}^k, \bar{\theta}^k]}(\bar{\theta}^k), 0], \bar{\theta}^k \le t \end{cases}$$

$$r^{[\underline{\theta}^k, \bar{\theta}^k]}(t) = \begin{cases} r^{[\underline{\theta}^{k-1}, \bar{\theta}^{k-1}]}(t), t \le \underline{\theta}^k \\ r^{[\underline{\theta}^k, \bar{\theta}^k]}(t), \underline{\theta}^k \le t \le \bar{\theta}^k. \\ 0, \bar{\theta}^k \le t \end{cases}$$

where $r^{[\underline{\theta}^k, \bar{\theta}^k]}$ is the solution of the inverse problem (6) with

$$I^{[\underline{\theta}^k, \bar{\theta}^k]}(t) = I^{[0,T]}(t), t \in [\underline{\theta}^k, \bar{\theta}^k]. \tag{7}$$

If the inverse problems in 4DIP are solved with the minimization of the Tikhonov functional

$$J(r^{[\underline{\theta},\bar{\theta}]}) = \left\| r^{[\underline{\theta},\bar{\theta}]} \right\|_R^2 + \left\| A^{[\underline{\theta},\bar{\theta}]}(\phi^{[\underline{\theta}^{k-1}, \bar{\theta}^{k-1}]}(\underline{\theta}^k), r^{[\underline{\theta},\bar{\theta}]}) - I^{[\underline{\theta},\bar{\theta}]} \right\|_Q^2 \to \min,$$

then the method is a modification of 4DVAR with weak constraints (in [7] the initial state is fitted as well). Here R and Q denote the weights in the corresponding norms. In the numerical experiments we will use iterative regularization in the variational method instead of the Tikhonov one (i.e. $R = 0$).

In this paper, we consider both the 4DVAR and 4DNK methods, where NK instead of IP in the abbreviation 4DIP denotes that we use the Newton-Kantorovich type algorithm analogous to [9, 10] to solve the inverse problems within assimilation windows Θ.

2.2. Newton-Kantorovich Type Algorithm

In this section we consider the interval $[0,T]$, but without loss of generality the same can be carried out for the interval $\left[\underline{\theta},\overline{\theta}\right]$. By combining the results obtained in [9, 10] for the diffusion-reaction models, the following relation can be derived. For any $h \in U_{mes}$, $r^{(2)}, r^{(1)} \in F$,

$$\left\langle A(r^{(2)}) - A(r^{(1)}), h \right\rangle_{L_2(\Omega_T;\mathrm{R}_\rho^{N_c})} = \left\langle \Psi\left[r^{(2)}, r^{(1)}; h\right], r^{(2)} - r^{(1)} \right\rangle_{L_2(\Omega_T;\mathrm{R}_\rho^{N_c})}, \tag{8}$$

where $A(.) = A^{[0,T]}(\varphi(0),.)$, $\Psi\left[r^{(2)}, r^{(1)}; h\right]$ is the solution of the adjoint problem

$$-\frac{\partial \Psi_l}{\partial t} - \vec{u}grad(\Psi_l) - div(diag(\mu)grad\Psi_l) + (G(t,\varphi\left[r^{(2)}\right],\varphi\left[r^{(1)}\right])\Psi)_l = h_l,$$

$$(x,y,t) \in \Omega_T, \tag{9}$$

$$G(t,\varphi^{(2)},\varphi^{(1)}) = diag\left(P(t,\varphi^{(2)})\right) + \bar{\nabla}P(t,\varphi^{(2)},\varphi^{(1)})^* diag(\varphi^{(1)}) - \bar{\nabla}\Pi(t,\varphi^{(2)},\varphi^{(1)})^*, \tag{10}$$

$$\Psi = 0, (x,y,t) \in \partial\Omega_T / \{t = T\}, \tag{11}$$

$$\Psi = 0, (x,y) \in \Omega, t = T. \tag{12}$$

In (10), $.^*$ denotes the ajoint matrix with respect to the weighted Euclidean scalar product on $\mathrm{R}_\rho^{N_c}$, the symbol $\bar{\nabla}$ denotes the divided difference operator that maps the vector-function $S:[0,T]\times\mathrm{R}^{N_c} \to \mathrm{R}^{N_c}$ to the vector function $\bar{\nabla}S:[0,T]\times\mathrm{R}^{N_c}\times\mathrm{R}^{N_c} \to \mathrm{R}^{N_c\times N_c}$, such that for any $t \in [0,T]$ and $\varphi, \delta\varphi \in \mathrm{R}^{N_c}$,

$$S(t,\varphi+\delta\varphi) - S(t,\varphi) = \bar{\nabla}S(t,\varphi+\delta\varphi,\varphi)\delta\varphi. \tag{13}$$

The following relation holds for any r:

$$I_U - A_U(r) = M_U[r,r]\left(r^{(*)} - r\right) + w, \tag{14}$$

$$w = \left(M_U[r^{(*)},r] - M_U[r,r]\right)\left(r^{(*)} - r\right) + \delta I_U, \tag{15}$$

where $r^{(*)}$ is the exact solution of the inverse problem,

$$M_U[r^{(2)},r^{(1)}]:\begin{cases} F \to \mathrm{R}^{\Xi} \\ r \mapsto \sum\limits_{\xi=1}^{\Xi}\left\langle \Psi\left[r^{(2)},r^{(1)};u_\xi\right], r\right\rangle_{L_2(\Omega_T;\mathrm{R}_\rho^{N_c})} e_\xi \end{cases}, \tag{16}$$

$$A_U:\begin{cases} F \to \mathrm{R}^{\Xi} \\ r \mapsto \sum\limits_{\xi=1}^{\Xi}\left\langle A(r), u_\xi\right\rangle_{L_2(\Omega_T;\mathrm{R}_\rho^{N_c})} e_\xi \end{cases}, \quad I_U = \sum_{\xi=1}^{\Xi}\left\langle I, u_\xi\right\rangle_{L_2(\Omega_T;\mathrm{R}_\rho^{N_c})} e_\xi, \tag{17}$$

and

$$A_U(r+\delta r)-A_U(r)=M_U[r+\delta r,r]\delta r. \tag{18}$$

As the projection basis U, we consider the trigonometric basis:

$$U_\Theta=\left\{e_{\eta\theta_t\theta_x\theta_y}\mid 0\le\theta_x\le\Theta_x,0\le\theta_y\le\Theta_y,0\le\theta_t\le\Theta_t,\eta\in L_{mes}\right\}, \tag{19}$$

$$\left(e_{\eta\theta_t\theta_x\theta_y}\right)_{ij}^m=\left\{\begin{cases}\dfrac{1}{\sqrt{\rho_\eta}}C(T,\theta_t,t^m)C(X,\theta_x,x_i)C(Y,\theta_y,y_j), l=\eta\\ 0,\ l\ne\eta\end{cases}\right\}_{l=1}^{N_c}, \tag{20}$$

$$m=1,\ldots,N_t,i=1,\ldots,N_x,\ j=1,\ldots,N_y$$

$$C(T,\theta,t)=\frac{1}{\sqrt{T}}\begin{cases}\sqrt{2}\cos\left(\dfrac{\pi\theta t}{T}\right),\theta>0\\ 1,\theta=0\end{cases}. \tag{21}$$

For the solution of (14), we apply the Newton-Kantorovich type algorithm based on the truncated SVD with the use of the right inverse matrices [11, 12, 9]:

1. Define the initial approximation $r^{(0)}$, the step $\Delta\Sigma>0$, and the maximum allowed conditional number $cond_{\max}$.

2. Let us run iterations by the conditional number of the sensitivity operator matrix, starting with $\Sigma=1$:

(a) We compute the matrix $m^{(k)}$ of the sensitivity operator $M_U[r^{(k)},r^{(k)}]$ and the singular value decomposition of $m^{(k)}\left(m^{(k)}\right)^T$. Let σ_l be the singular values of $m^{(k)}$. If $\Sigma>cond_{\max}$, the algorithm stops.

(b) If not, we define the increment

$$\delta r^{(k)}=\Pr_{src}\left(m^{(k)}\right)^T\left[m^{(k)}\left(m^{(k)}\right)^T\right]_p^+\left(I_U-A_U(r^{(k)})\right),$$

where

$$\Pr_{src}\ z=\left\{\begin{cases}z_l,l\in L_{src}\end{cases}\right\}_{l=1}^{N_c}, \tag{22}$$

$$\left[m^{(k)}\left(m^{(k)}\right)^T\right]_\Sigma^+=\sum_{l=1}^{p}\frac{U_l}{\sigma_l^2}\langle .,U_l\rangle_{\mathrm{R}^\Xi},\quad \sigma_1/\sigma_p\le\Sigma<\sigma_1/\sigma_{p+1}, \tag{23}$$

$\langle .,.\rangle_{\mathrm{R}^\Xi}$ is the Euclidean scalar product in R^Ξ, $\{U_l\}_{l=1}^{rank(C)}$ is the orthonormal system of left singular vectors of $m^{(k)}$.

(c) We choose $r^{(k+1)}=r^{(k)}+\gamma^{(k)}\delta r^{(k)}$ to satisfy the condition (dividing $\gamma^{(k)}$ several times by 2, if necessary)

$$\left\|I_U-A_U(r^{(k+1)})\right\|_{\mathrm{R}^\Xi}<\left\|I_U-A_U(r^{(k)})\right\|_{\mathrm{R}^\Xi},\ \left\|I-A(r^{(k+1)})\right\|<\left\|I-A(r^{(k)})\right\|.$$

(d) The algorithm continues to work until the iterations are stabilized. After stabilization, the number of singular values and vectors under consideration increases: $\Sigma:=\Sigma\Delta\Sigma$.

For numerical solution of the multi-dimensional problem, a splitting scheme with respect to the spatial dimensions and physical processes is applied. Discrete-analytical approach with locally adjoint problems is used to obtain consistent numerical schemes for the individual splitting stages [13].

3. Results

As an example, we consider the modified atmospheric chemistry transformation mechanism from [14]. The reaction rates depend on time. The chemical transformation system can be rewritten in the form of a system of ordinary differential equations of the production-destruction type forming the reaction term of (1).

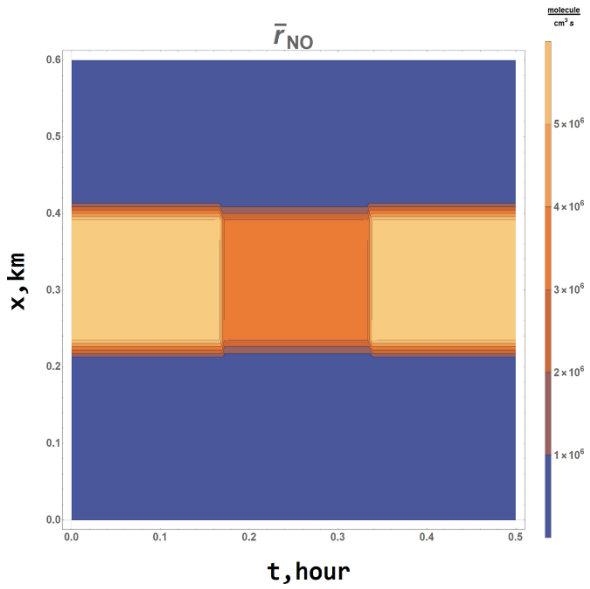


Figure 1. Exact source function cross section at $y = 300m$.

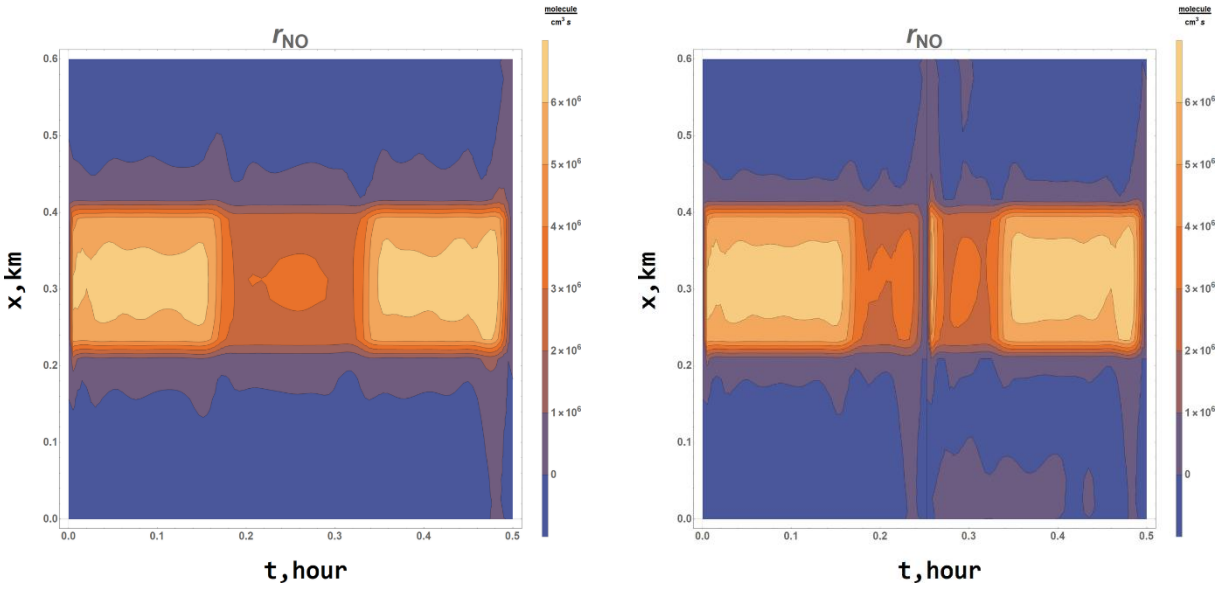
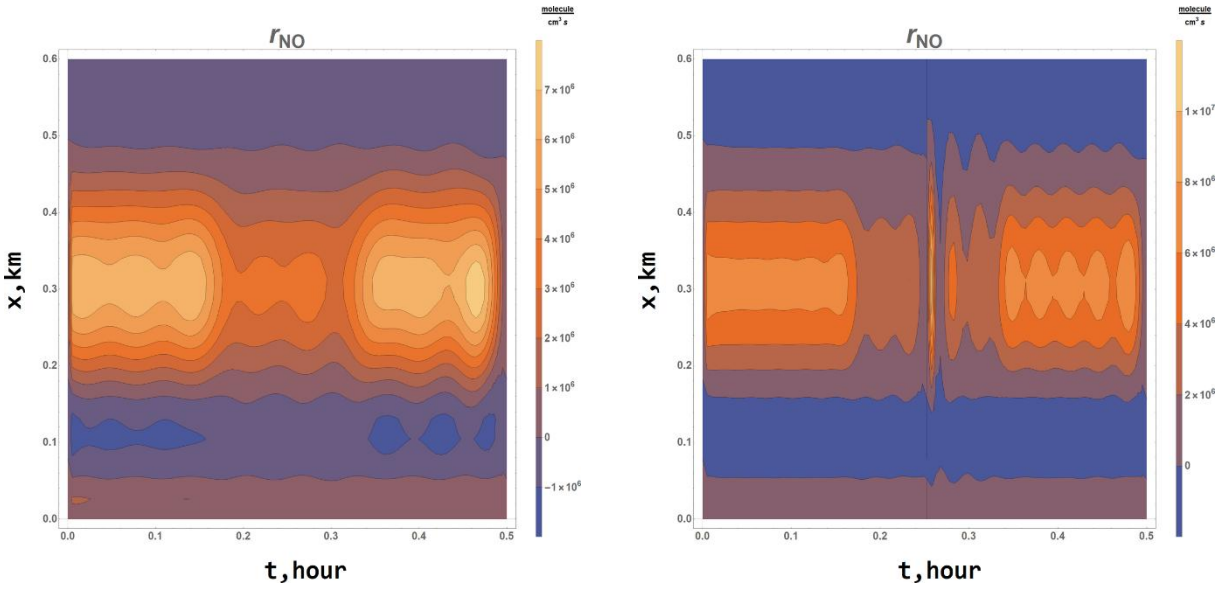


Figure 2. Reconstructed source at the domain Ω_T cross section $y = 300m$ with variational algorithm: (a) as inverse problem solution and (b) as data assimilation problem solution with two windows.



(a) (b)

Figure 3. Reconstructed source at the domain Ω_T cross section $y = 300m$ by the Newton-Kantorovich type algorithm as (a) inverse problem solution and as (b) data assimilation problem solution with two assimilation windows.

Assume that the distributed source emits NO only (i.e. $L_{src} = \{NO\}$) with the intensity presented in Figure 1. Concentration field dynamics images of O_3 across the whole domain are available as the measurement data (i.e. $L_{mes} = \{O_3\}$). Projections of these images to U are used as I_U. In the data assimilation scenario, the time interval was split into two equal non-intersecting intervals (assimilation windows). In the inverse problem scenario there is only one assimilation window $[0,T]$.

The initial guess to the sources is $r^{(0)} = 0$. The model parameters are $\mu = 10m^2s^{-1}$, $\vec{u} = (1,1)ms^{-1}$. The grid parameters are $T = 1800s$, $X = Y = 600m$, the number of points in time $N_t = 100$, the number of points in space $N_x = N_y = 24$, and the number of substances $N_c = 22$. The numbers of projection functions for the Newton-Kantorovich algorithm were $\theta_x = \theta_y = 5$, $\theta_t = 16$.

The results of the source reconstruction with the help of the variational algorithms are presented in Figure 2 and with the Newton-Kantorovich type algorithm, in Figure 3. Both calculations were made on a workstation with 8 parallel computation cores.

4. Discussion

The numerical experiment demonstrates the difference between the inverse problem and the data assimilation problem. In the inverse problem, all measurement data are available at the beginning of the solution process, while the portions of measurements in the data assimilation case arrive in the course of calculations. In the data assimilation scenario, the windows do not intersect. As a result, in Figures 2 (b) and 3 (b) we can see an additional discontinuity at the window border, in contrast to Figures 2 (a) and 3 (a), correspondingly, which show the inverse problem solution. The lack of computational resources did not allow us to take higher resolution (defined by $\theta_x, \theta_y, \theta_t$) for the Newton-Kantorovich type algorithm. Nevertheless, both algorithms were able to estimate the unknown source in all the presented cases. It seems that the edges are more sharp in Figure 3 (b)

compared to Figure 3 (a), because the time interval of the reconstruction was two times less. Hence, the same number of data $\theta_x \cdot \theta_y \cdot \theta_t$ is used to reconstruct less unknown variables.

5. Conclusions

Two types of inverse modeling problems were considered for a transport and transformation model of atmospheric pollutants: an inverse source reconstruction problem and a data assimilation problem. The measurement data were available in the form of concentration field images. The data assimilation problem was considered as a sequence of linked inverse problems. As inverse problem solution algorithms, variational and Newton-Kantorovich type algorithms were compared. In the numerical experiment, an emission source of a primary pollutant was reconstucted via the concentration field of a secondary pollutant. In the above-considered scenario, both algorithms were capable to estimate the unknown source.

Acknowledgments

This work was supported by the Russian Scientific Foundation under project 17-71-10184 (Development of Algorithms based on Sensitivity Operator) and by ICMMG SB RAS order no. 0315-2016-0004 (Development of Variational Algorithms). The vectorization and optimization of computer programs were carried out with support of the Ministry of Education and Science of the Russian Federation (NSU-NNC SB RAS joint laboratories 4.1.3).

References

[1] Gurjar B, Molina L and Ojha C (eds) 2010 *Air Pollution: Health and Environmental Impacts* (CRC Press) ISBN 1439809623
[2] Landrigan P J 2017 *The Lancet Public Health* **2** e4–e5
[3] Holnicki P and Nahorski Z 2015 *Environmental Modeling & Assessment* **20** 583–597
[4] Markakis K, Valari M, Perrussel O, Sanchez O and Honore C 2015 *Atmospheric Chemistry and Physics* **15** 7703–23
[5] Bocquet M *et al* 2014 *Atmospheric Chemistry and Physics Discussions* **14** 32233–32323
[6] Dimet F X L, Souopgui I, Titaud O, Shutyaev V and Hussaini M Y 2015 *Nonlinear Processes in Geophysics* **22** 15–32 URL http://www.nonlin-processes-geophys.net/22/15/2015/npg-22-15-2015.pdf
[7] Elbern H, Strunk A, Schmidt H and Talagrand O 2007 *Atmospheric Chemistry and Physics Discussions* **7** 1725–1783
[8] Marchuk G 1964 *Reports of the USSR Academy of Sciences / Ed. Science* **156** 503–506 (In Russian)
[9] Penenko A, Nikolaev S, Golushko S, Romashenko A and Kirilova I 2016 *Math. Biol. Bioinf.* **11** 426–444 (In Russian) URL http://www.matbio.org/article.php?lang=eng id=296
[10] Penenko A V 2018 *Numerical Analysis and Applications* **11** 73–88
[11] Cheverda V A and Kostin V I 1995 *Journal of Inverse and Ill-Posed Problems* **3** 131–48
[12] Kaltenbacher B, Neubauer A and Scherzer O 2008 *Iterative Regularization Methods for Nonlinear Ill-Posed Problems* (Berlin. Germany: De Gruyter) ISBN 3110204207
[13] Penenko V, Tsvetova E and Penenko A 2014 *Computers & Mathematics with Applications* **67** 2240–56
[14] Stockwell W and Goliff W 2002 *Journal of Geophysical Research* **107** 4643–50