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EMISSION RATE ESTIMATION THROUGH DATA ASSIMILATION OF GAMMA DOSE MEASUREMENTS IN A LAGRANGIAN ATMOSPHERIC DISPERSION MODEL

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This paper presents an efficient algorithm for estimating the unknown emission rate of radionuclides in the atmosphere following a nuclear accident. The algorithm is based on assimilation of gamma dose rate measured data in a Lagrangian atmospheric dispersion model. Such models are used in the frames of nuclear emergency response systems. It is shown that the algorithm is applicable in both deterministic and stochastic modes of operation of the dispersion model. The method is evaluated by computational simulations of a 3-days field experiment on atmospheric dispersion of ^{41}Ar emmited routinely from a research reactor. Available measurements of fluence rate (photons flux) in air are assimilated in the Lagrangian dispersion model DIPCOT and the ^{41}Ar emission rate is estimated. The statistical analysis shows that the model-calculated emission rates agree well with the real ones. In addition the model-predicted fluence rates at the locations of the sensors which were not used in the data assimilation procedure are are in better agrrement with the measurements. The above first evaluation results show that the method performs satisfactorily and therefore it is applicable in nuclear emergency response systems provided that more comprehensive validation studies will be performed.

1. INTRODUCTION

Real-time nuclear emergency response systems (ERSs) like RODOS [1], ARGOS [2], NARAC [3] calculate atmospheric dispersion and fallout following an accidental release of radioactivity using atmospheric dispersion models (ADMs). In emergency phase the uncertainty of the radionuclides emission rate in the atmosphere (usually called "source term" or "source function" in the frames of dispersion models) can be dominant among all other uncertainties (like uncertainties in meteorological information or model errors): the estimated source term can differ from the true one by a factor of 10 or more [4]. Therefore, correction of the radionuclides emission rate used in the dispersion model by utilizing all available sources of information is of primary importance especially in nuclear ERSs as well as in ERSs dealing with other kinds of hazardous materials [5]. A way to improve emission rate information is data assimilation (DA) of gamma dose measurements which are typically available around every nuclear power plant. In operational ERSs the Kalman Filtering (KF) approaches (standard, ensemble, and other) are most widely used for that purpose (e.g., [6], [7]). In several works the KF approach has been combined with different kinds of ADMs (i.e. with Gaussian plume models in [8] and with Lagrangian stochastic models as in [9]) for source term correction with assimilation of gamma dose measurements. Despite their definite advantages, like ability to deal with different sources of uncertainty, including model errors, robustness and relatively simple implementation, KF approaches are computationally quite expensive procedures, and that characteristic becomes even more important in case of large deviation of the first guess estimation of the control variable from the true one. On the other hand the variational approach to data assimilation, which was extensively used in case of source term correction with concentration measurements (e.g., [10], [11], [12], [12] etc) and which is very attractive from the viewpoint of computational effectiveness, has been rarely applied for source term (emission rate as a function of time) estimation with gamma dose rate measurements. Additional difficulty arises when variational approach is to be applied within a stochastic model such as the Lagrangian model DIPCOT [14], because the cost function gradient with respect to control variables becomes random function.

In previous work of the authors [15] a variational algorithm has been developed and implemented within the deterministic ('puff') atmospheric dispersion model for source term estimation using concentration measurements. The algorithm was validated through 'identical twin' tests. In the present work the variational method is extended for the case of assimilation of gamma dose rate measurements in both the deterministic and stochastic versions of Lagrangian ADM DIPCOT, which is used in the European nuclear ERS RODOS. The algorithm is validated through computational simulations of the 3-days field experiment on ^{41}Ar atmospheric dispersion in Mol, Belgium, [16]. Measurements of fluence rate in air (photons flux) are assimilated in the ADM DIPCOT in order to correct the first-guess estimation of the ^{41}Ar emission rate. The model-estimated emission rate agrees well with the true one. Also the model-predicted fluence rates at the locations of the sensors not used in the DA procedure are improved.

2. SHORT DESCRIPTION OF DIPCOT MODEL

2.1 Basics

DIPCOT [14] is a computational model, which simulates atmospheric dispersion estimating particles' (or puffs') trajectories. It has been comprehensively evaluated against numerous field and laboratory experiments on atmospheric dispersion (e.g., [17], [18], [19], [20]) and presently it is included in EU RODOS system. In ADM DIPCOT there are two modes of particles /puffs movement, the stochastic mode (SM) and the deterministic mode (DM). In deterministic mode puffs are transported by the mean wind velocity field and grow in size at a rate depending on atmospheric stability conditions. In stochastic mode puffs are transported, in addition to the mean wind, also by wind velocity fluctuations calculated by a Langevin equation. The latter is formulated for stationary homogeneous isotropic turbulence at the horizontal direction, and on inhomogeneous Gaussian turbulence in the vertical direction. Concentration of radionuclide *nu*, C_{nu}, at an arbitrary spatial point of the 3-dimensional domain (x, y, z) at time t is calculated as sum of contributions of all puffs:

$$C_{nu}(x,y,z,t) = \sum_{i=1}^{N_p} C_{i,nu} = \frac{1}{(2\pi)^{3/2}} \sum_{i=1}^{N_p} \frac{q_{i,nu}\tau\gamma(t,\tau,i)}{\sigma_{xi}\sigma_{yi}\sigma_{zi}} \times \exp\left(-\lambda_{nu}\left(t-(i-1)\tau\right)\right) \exp\left[-\frac{1}{2}\left(\frac{(x_i-x)^2}{\sigma_{xi}^2}+\frac{(y_i-y)^2}{\sigma_{yi}^2}\right)\right] \times \left\{\exp\left[-\frac{1}{2}\frac{(z_i-z)^2}{\sigma_{zi}^2}\right]+\exp\left[-\frac{1}{2}\frac{(z_i+z-2z_g)^2}{\sigma_{zi}^2}\right]\right\} \quad (1)$$

Here N_p is the total (maximum) number of puffs, $C_{i,nu}$ is the concentration of radionuclide *nu* in puff *i*, (x_i, y_i, z_i) are coordinates of i-th puff, z_g is ground height above sea level, τ is time interval between releases of puffs, $q_{i,nu}$ is the release rate of radionuclide *nu* corresponding to the time of the i-th puff's release, σ_{xi}, σ_{zi} are the parameters characterising the spatial distribution of concentration in the puff, the function $\gamma(t,\tau,i) = \mathrm{sgn}(t - i\cdot\tau)$ eliminates the influence of puffs not released by the time *t*, and λ_{nu} is the decay constant of radionuclide *nu*. Total reflection of the cloud from the underlying surface is assumed in (1). In deterministic mode the values of σ_{xi}, σ_{zi} are parameterized with the well-known Karlsruhe-Julich relationships [21], while in stochastic mode those values are defined following the approach from [22], which itself is based on Taylor's homogeneous diffusion theory.

2.2 Gamma dose calculation in DIPCOT

In this section a brief description is given of the gamma dose calculation methods used in DIPCOT. Detailed description is provided in [14] and [17].

The gamma radiation dose rate (Gy·s^{-1}) in air at a receptor point immersed in a radioactive cloud with arbitrary activity concentration distribution $C(x,y,z)$ (Bq·m^{-3}) emitting mono-energetic photons is

$$d_\gamma = \frac{\mu_\alpha E_\gamma}{\rho}\Phi = \frac{\mu_\alpha E_\gamma}{\rho}\frac{1}{4\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{Be^{-\mu r}C}{r^2}dxdydz \quad (2)$$

where μ_α is the total linear energy absorption coefficient for air (m^{-1}), E_γ is the photons' energy (J), ρ is the air density (kg·m^{-3}), Φ is the fluence rate in air (photons·m^{-2}·s^{-1}), μ is the total linear attenuation coefficient for air (m^{-1}), B is the "build-up" factor, that accounts for the flux of extra photons due to Compton

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scattering and r is the distance between the receptor point and the elementary source of volume $dxdydz$. μ_a, B and μ are functions of the photons energy.

For a semi-infinite radioactive cloud with homogeneous concentration c, the above volume integral can be calculated analytically and in that case the dose rate is:

$$d_{\gamma,\infty} = \frac{CE_\gamma}{2\rho} \quad (3)$$

In DIPCOT the radioactive plume is described as a number (N_p) of puffs. If N_{nucl} radionuclides are emitted in the atmosphere, each puff n (n = 1 to N_p) bears a certain activity load

$Q_{i,nu} = q_{i,nu}\tau \exp(-\lambda_{nu}(t-(i-1)\tau))$ (Bq) – see equation (1) – for nuclide nu (nu =1 to N_{nucl}). Furthermore, to take into account the fact that more than one photon with different energies are emitted per radioactive disintegration, the photons are divided in N_{gr} groups, according to their energies. The energies emitted per disintegration in each of the N_{gr} groups for the radionuclides of concern $E_{\gamma,nu,ig}$ are given as input data in DIPCOT. For each group ig (ig = 1 to N_{gr}) a central or "nominal" energy $E_{\gamma,\mathrm{nom},ig}$ is defined. It is assumed that a number of "equivalent photons" $f_{nu,ig}$ is emitted per disintegration of nuclide nu in each group ig with energy equal to the nominal energy of the group. The number of equivalent photons is calculated as follows:

$$f_{nu,ig} = \frac{E_{\gamma,nu,ig}}{E_{\gamma,\mathrm{nom},ig}} \quad (4)$$

Finally, the gamma radiation dose rate in air is calculated as the sum over all puffs, all radionuclides and the N_{gr} energy groups, as follows:

$$d_\gamma = \sum_{i=1}^{N_p}\sum_{nu=1}^{N_{nucl}}\sum_{ig=1}^{N_{gr}} f_{nu,ig}\frac{\mu_{\alpha,ig}E_{\gamma,\mathrm{nom},ig}}{\rho}\Phi_{i,nu,ig} = \quad (5)$$

$$\sum_{i=1}^{N_p}\sum_{nu=1}^{N_{nucl}}\sum_{ig=1}^{N_{gr}} f_{nu,ig}\frac{\mu_{\alpha,ig}E_{\gamma,\mathrm{nom},ig}}{\rho}\frac{1}{4\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{B_{ig}e^{-\mu_{ig}r}C_{i,nu}}{r^2}dxdydz$$

where $C_{i,nu}$ is given by (1).

For an infinite cloud of homogeneous activity concentration $C_{i,nu}$ for each nuclide, the dose rate becomes:

$$d_{\gamma,\infty} = \sum_{i=1}^{N_p}\sum_{nu=1}^{N_{nucl}}\sum_{ig=1}^{N_{gr}} f_{nu,ig}\frac{E_{\gamma,\mathrm{nom},ig}C_{i,nu}}{2\rho} \quad (6)$$

The assumptions of infinite cloud dimensions and homogeneous activity concentration can be applied in an atmospheric dispersion model at sufficiently large distances from the cloud source. However at small distances from the source and/or in complex terrain circumstances where the cloud is strongly inhomogeneous, the infinite cloud approximation results in significant over-estimation of the gamma radiation dose rate. In such cases the integral in equation (5), representing the fluence rate, has to be computed numerically. This is computationally very expensive, especially in the frame of an atmospheric dispersion model with N_p ~ 10^3 - 10^5 and calculating doses at a number of grid points in the order of 10^4. Therefore a method has been developed for the fast and accurate calculation of the integral in equation (5). The details of the method are given in [14] and [17]. The final relationship is the following:

$$d_\gamma = \sum_{i=1}^{N_p}\sum_{nu=1}^{N_{nucl}}\sum_{ig=1}^{N_{gr}} f_{nu,ig}\frac{\mu_{\alpha,ig}E_{\gamma,\mathrm{nom},ig}}{\rho}\times \quad (7)$$

$$\frac{1}{4\pi}\frac{Q_{i,nu}}{\sigma_{x',i}\sigma_{y',i}\sigma_{z',i}}\frac{1}{\mu_{ig}}\left(A_{1,i,ig}+\kappa A_{2,i,ig}\right)$$

where $\kappa = (\mu-\mu_\alpha)/\mu_\alpha$, $\sigma_{x',i}$, $\sigma_{y',i}$, $\sigma_{z',i}$ are the puff's dimensions in a new coordinates system with its origin at the puff's centre and the z-axis passing through the receptor point and $A_{1,i,ig}$, $A_{2,i,ig}$ are integrals, functions of the photons energy, the puff's dimensions and its distance from the receptor point. These integrals have been pre-calculated for a range of values of the above factors covering the range of values encountered in dispersion calculations for the different photons energies, puffs dimensions and distances between puffs and receptor (grid) points. The pre-calculated values of the integrals have been stored in a file in the form of 4-dimensional matrices to be read at the beginning of the DIPCOT run. Each time a dose rate has to be calculated by (7), a linear interpolation is performed in the stored values of A_1 and A_2.

In conclusion 2 methods for gamma radiation dose rate in air are available in DIPCOT: the infinite cloud approach (6) and detailed calculation (7).

3. DATA ASSIMILATION ALGORITHM

3.1 Statement of the problem

Consider the problem of modelling atmospheric dispersion on time interval $(0,T)$ with a point source releasing radionuclides during the time interval $(0,T_s)$, $T_s \le T$. It is assumed that during interval $(0,T)$ dose rate measurements are available from K stations located at spatial points $\bar{r}^k = \left(x^k, y^k, z^k\right)^T$, $1 \le k \le K$. Denote gamma dose rates, measured at time $t_n, 1 \le n \le N_O$ by the k-th station as $d^o(n,k)$. The available measurements during interval $(0,T)$ can be used to improve the source function information leading to improvement of the modelling results. The adjustable parameters in the assimilation procedure compose the control vector $\bar{\psi}$ which consists of source emission rates corresponding to times of releases of puffs: $\bar{\psi}^T = \left(q_1, ..., q_{N_p}\right) = \bar{q}^T$. The problem of data assimilation can be posed as an optimization problem of minimizing the following objective function with respect to control vector $\bar{\psi}$ [24]:

$$J = J_1 + J_2,$$
$$J_1 = \left(\bar{\psi} - \bar{\psi}^b\right)^T \underline{\underline{B}}^{-1} \left(\bar{\psi} - \bar{\psi}^b\right)$$
$$J_2 = \left(\bar{d}^o - \bar{d}^M\right)^T \underline{\underline{O}}^{-1} \left(\bar{d}^o - \bar{d}^M\right) \quad (2)$$

Here $\bar{\psi}^B$ is first guess estimation of the control vector, $\underline{\underline{O}}$, $\underline{\underline{B}}$ are covariance matrices of the errors of observations and background errors respectively; vector $\bar{d}^o \in R^{N_o K}$ consists of gamma dose rates $d^o(n,k)$, measured on each subinterval Δt_n by k-th station. The elements of d^O are ordered sequentially as follows: $d^o_{(n-1)K+k} = d^o(n,k)$. The corresponding vector that consists of the calculated dose rates at the K stations at the N_o time intervals is denoted by $\bar{d}^M = \underline{\underline{G}}\bar{\psi}$, where matrix $\underline{\underline{G}}$ of size $\left(N_o \cdot K \times N_p\right)$ is formally introduced relating $\bar{d}^M$ and $\bar{\psi}$. Details of its calculation will be clarified below. Thus the minimization of function (2) is a linear regression problem with constraint: $\bar{\psi} \ge 0$. In the present work covariance matrices are assumed to be diagonal with constant observation error σ_O^2 and background error σ_B^2. Those error parameters can reflect physical information concerning quality of measurements and of a priori information about background estimations of adjusted parameters. Errors σ_O^2 and σ_B^2 can be combined so that solution of minimization problem depend on only one parameter, namely $\sigma^2 = \sigma_O^2 / \sigma_B^2$. The latter can be estimated using different heuristics methods.

3.2 Calculation of the G-matrix

Now let us describe the details of computations of matrix $\underline{\underline{G}}$. As it immediately follows from (1) concentrations at given set of points are linearly related to vector $\bar{q}$: $\bar{c} = \underline{\underline{G}}^c \bar{q}$. This relationship has been used in previous work by the authors [15] and in other studies where measured concentration values are used for source function estimation. Since according to (3) and (6) in the case of infinite cloud approximation gamma dose rate is simply proportional to concentration at a given point, matrix $\underline{\underline{G}}$ can be computed simply by multiplication of the matrix $\underline{\underline{G}}^c$ by a constant:

$$\bar{d} = \underline{\underline{G}}\bar{q} = \frac{1}{2\rho}\left(\sum_{ig=1}^{Ngr} f_{ig} E_{\gamma,ig}\right)\underline{\underline{G}}^c \bar{q} \quad (3)$$

where N_{gr} is number of energy groups (typically $N_{gr} = 4$), $E_{\gamma,ig}$ are the emitted energies per disintegration for each of the energy groups. The constant value is evident from (3). Note, that since $E_{\gamma,ig}$ are nuclide dependent, the constant becomes dependent on nuclide type.

In case of the dose rate calculation method of [14] and [17], according to (7) computation of matrix $\underline{\underline{G}}$ is performed using the following relation (case of single nuclide is shown for simplicity):

$$g_{li} = \frac{1}{4\pi}\sum_{ig=1}^{Ngr} f_{ig} \frac{\mu_{\alpha,ig} E_{\gamma,ig}}{\rho \mu_{ig}} \times \frac{\left[\tau\gamma\left(t_l, \tau, i\right)\exp\left(-\lambda\left(t_l - (i-1)\tau\right)\right)\right]}{\sigma'_{xi}\sigma'_{yi}\sigma'_{zi}} \times \left(A_{1,i,ig,l} + \kappa_{ig} A_{2,i,ig,l}\right) \quad (4)$$

Here g_{il} is element matrix $\underline{\underline{G}}$, index i denotes puff number and index $1 \le l \le N_o K$ denotes a given observation at a given time. All terms entering right hand side of (4) have been explained above. Note that the dependence of A_1, A_2 on indices ig, i, l as shown explicitly by (4) reflects the fact that these coefficients depend on puff's size, distance to the receptor point and attenuation factor. The multiplier in square brackets is analogous to the same term in (1).

3.3 Control vector reduction

The number of puffs in a Lagrangian model as DIPCOT is in the order of 10^3 to 10^5 or even larger. Thus the dimension of the control vector of the optimization problem can become very large and this can lead to poor performance of data assimilation when the latter is performed in a straightforward way, as it will be shown in the next section. Another problem arises in case of stochastic mode of DIPCOT operation, when the puffs movement has a random component. In that case elements of matrix $\underline{\underline{G}}$ become random values (i.e, the same element in different runs will have different value), while traditional variational data assimilation approach deals with deterministic differential equations. Both the abovementioned problems can be solved with the simple 'control vector reduction' (CVR) procedure which was theoretically investigated in the context of variational data assimilation in [25].

Assume that during time interval Δt source release rate can be considered as constant with sufficient accuracy. In operational practice of ERSs Δt can be in the order of $10^3 - 10^4\, s$ (see e.g., [26]). At the same time DIPCOT uses significantly smaller time step τ between appearances of successive particles (0.1-100 s) so that $\Delta t / \tau = \Pi >> 1$. Then the source can be divided in P time intervals of the size Δt with Π puffs in each time interval, so $N_P = \Pi \cdot P$.. Clearly the value of P depends on the choice of the time interval Δt during which source rate could be considered as constant and thus it is a free variable that depends on the expert judgment of the user. Note that if P=1, then source rate is assumed to be constant during the whole release interval. In each interval j ($1 \le j \le P$) the release rate can be considered as constant and equal to $\tilde{q}_j$, here $\tilde{q}_j$ are the values characterizing source function of the $j-th$ group of puffs, which form the reduced control vector: $\underline{\tilde{q}}$ of the size P.

Instead of initial problem of minimization of (2) with respect to control vector $\overline{q}$ consisting of release rates of individual particles the 'reduced' minimization problem will be solved in which the same function will be minimized with respect to the reduced control vector $\underline{\tilde{q}}$. The $\underline{\underline{G}}^r$ matrix of the reduced minimization problem has size $\left(N_o K \times P\right)$. The formula for calculating the elements of the $\underline{\underline{G}}^r$ matrix from the elements of the $\underline{\underline{G}}$ matrix can be easily obtained:

$$g^r_{lj} = \sum_{m=1}^{\Pi} g_{l\left((j-1)\Pi+m\right)}, \forall l,\ 1 \le l \le N_o K, 1 \le j \le P\ .$$

(5)

Thus, when distributions of random variables g_{lp} satisfy conditions of Central Limit Theorem (which are in fact quite non-restrictive [27]) and with large enough Π, sums in (5) converge to stable values. As it will be shown below that circumstance allows for using of the variational data assimilation method with CVR also in case of stochastic Lagrangian particle models. Another important advantage of using CVR is increase in computational efficiency and accuracy because of reducing the size of the control vector by a factor of Π.

The described above DA procedure has been implemented within the model DIPCOT. Matrix $\underline{\underline{G}}$ is calculated during forward run mode and cost function (2) with constraint of positive control vector values is minimized using the International Mathematics and Statistics Library (IMSL ® package) [31].

4. RESULTS

An atmospheric dispersion experiment, described by [16] (see also [28]), was carried out at the BR1 research reactor of the Belgium Nuclear Research Centre (SCK-CEN) in Mol, during first week of October 2001. In that experiment atmospheric dispersion of ^{41}Ar has been studied. The available measurements include: a) measured ^{41}Ar emission rate from the stack of the BR1 research reactor; b) meteorological data by a weather mast; c) monitoring of the gamma radiation field (fluence rate) due to the decay of ^{41}Ar. In previous work [17], the DIPCOT model in forward run operation mode and its gamma dose calculation method were evaluated against the measured data from the Mol experiment. In the present work data assimilation runs have been performed for the cases of Wednesday 3^{d} October 2001, afternoon

("Day 1") and Thursday 4th of October 2001 ("Day 2") experiments.

The raw wind velocity data of 1 min sampling at heights 69 m and 78 m have been averaged on 10 min intervals to drive the dispersion model, together with the Pasquill-Gifford stability categories given in 10 min intervals in the experimental data base. The meteorological data were pre-processed by the meteorological pre-processor FILMAKER of the RODOS system [29] to prepare input meteorological fields for the DIPCOT model.

Atmospheric air was led through the reactor at the rate of 9.4 m^3/s and was emitted from the 60-m stack, giving rise to a routine ^{41}Ar emission rate of ≈1.5 x 10^{11} Bq/h. The measurements of the release rate were available each 1 minute. The radiation field was monitored by an array of four NaI(Tl) detectors belonging to SCK-CEN (SCK-NaI), four NaI(Tl) detectors belonging to Danish Emergency Management Agency (DK-NaI), one Germanium detector belonging to Technical University of Denmark (DTU-HPGe) and one Germanium detector belonging to SCK-CEN (SCK-HPGe). A map of sensors locations is presented in Figure 1. During Day 1 experiment SCK sensors were collocated with DK-NaI and therefore they are not shown.

Simulations with DIPCOT have been performed with the following set of parameters. The puffs were released at a time interval $\tau \approx 3\,s$. The method described by equation (7) has been used for calculations of the fluence rates at the positions of sensors. Two sets of simulations have been performed: one set in deterministic and one set in stochastic mode of DIPCOT operation. The first guess source emission rate was set by a factor of 10 greater and also by a factor of 10 less than the true rate. A value of $\sigma = 10^{-9}$ has been used. That value corresponds to the measurement error $\sigma_o \sim 10\,m^2/s$ [16] and assumed error in estimation of the first guess emission rate by the factor of 100: $\sigma_B \sim 10^{10}\,Bq/s$.

Different number of source time intervals (parameter P of CVR procedure) has been used in different runs. For Day 1 experiment the values of P=1, 9, 27, 111, 3500 have been used corresponding to the following values of time intervals during which source rate is assumed to be constant: $\Delta t \approx 175, 20, 6.5, 1.5, 0.05\,\text{min}$ respectively.

The last value of P represents the maximum possible value in this run: $P_{max} = N_P = 3510$. In Day 2 experiment the tested values of P were: 1, 4, and 16 corresponding to the values of $\Delta t \approx 485, 120, 30\,\text{min}$ respectively. Thus for both days of experiment values of Δt most typical in operational use of ERSs [26] equal to 20 and 30 minutes (P=9 and 16) had been tested. Other values of Δt (and hence of the parameter P) had been tested in order to clarify sensitivity of the results with respect to that parameter.

In assimilation runs for Day 1, experimental data of the four DK-NaI sensors have been used in data assimilation procedure while data of the DTU-HPGe sensor have been used for validation of the calculations. Since on Day 1 experiment SCK sensors were collocated with the DK-NaI sensors, the former were used neither in assimilation runs nor in validation process. In assimilation runs for Day 2 experiments, data of the four DK-NaI sensors have been used in data assimilation procedure while other available measurements from the other sensors SCK NaI 2-4, DTU-HPGe and SCK-HPGe (Figure 1) have been used in validation process.

Figure 2-a) presents examples of source emission rate estimations as result of assimilation of fluence rate data for the case of Day 1 experiment in case of stochastic version of DIPCOT. Results with the different number of groups P in control vector reduction procedure are presented. Figure 2-b) presents the source emission rate estimations for the case of Day 2 experiment with stochastic version of DIPCOT. In this figure the DA method performance setting the first guess source function by a factor of 10 less than the true one is presented. As it is evident from Figure 2-a) in the case of $P = N_P = 3510$ the source function is very poorly adjusted by the DA procedure. However results substantially improve with decreasing P. Note, that in all presented cases, except P=1, at the beginning and at the end of release the source function is not adjusted successfully. This happens because the measurements of the DK-NaI sensors were not available in the beginning and last period of the release. In case of P=1 source function is assumed to be uniform and thus covers the whole time period. However as it is evident from Figure 2 the adjusted source functions in all cases except the case with P=3510 are much better than the first guess source function.

This qualitative result is confirmed with the results of mean relative absolute error (MAE) and mean relative bias (MRB) presented in Table 1 ($MAE = \left\langle \left| q^a - q^t \right| \right\rangle / \left\langle q^t \right\rangle$, $MB = \left\langle q^a - q^t \right\rangle / \left\langle q^t \right\rangle$, where q is source function, $\langle\ \rangle$ means averaging, superscripts 'a' and 't' denote analyzed and true source function respectively). The results obtained by both stochastic and deterministic versions of DIPCOT and for both Day 1 and Day 2 experiments and with the different values of the CVR parameter P are presented in Table 1. The level of improvement is about the same for both days experiments. Generally as follows from these results in all cases source function analyzed in assimilation runs is much better than the first guess function.

In all cases presented in Table 1 the analyzed source function is underestimated by about 30-40%. This

systematic underestimation of the source function could be due to some error introduced by gamma dose calculation method as presented above and by other model errors. For comparison, assimilation runs with the 'infinite cloud approximation' (equation (6)) have been performed, which, according to [17], is less accurate than the method given by equation (7). Assimilation runs with the 'infinite cloud approximation' resulted in significantly worse values of MRB ($-0.8 \le MRB \le -0.5$) as compared to results presented in Table 1. Thus in present formulation model errors cannot be improved with the data assimilation procedure and the 'weak constraint' formulation is required in order to do that [30].

As it also follows from the results in Table 1, an important and good feature of the data assimilation algorithm is that the quality of the results is satisfactory for a wide range of values of the CVR parameter P. The time lengths Δt of source intervals corresponding to the values of P presented in Table 1 vary from several minutes to several hours and thus, as it was discussed above, completely fall within the range of Δt typically used in operational ERSs.

Another, indirect but important indicators of the level of improvement achieved with the use of data assimilation algorithm are the results of comparisons of the calculated fluence rates against the measurements that were not used in data assimilation procedure. Such results are presented in Figure 3 for Day 1 and in Figure 4 for Day 2 experiments. Results of the SCK-HPGe sensor are not shown because the measurements by that sensor didn't cover major part of the simulation period. As it is evident from Figures 3 and 4 the results achieved with data assimilation and with both deterministic and stochastic modes of DIPCOT operation are very good. However for Day 2 experiment the stochastic mode evidently better captures the maximum values of fluence rates.

The normalized mean squared error (NMSE) and fractional bias (FB) of the fluence rates calculated in first guess and different assimilation runs are presented in Table 2. As follows from Table 2 the levels of errors for all cases are good and are not too far from the errors of the forward runs achieved with the true source function as reported in [17]. Also from Table 2 it is seen that the results of assimilation runs obtained with the stochastic version of DIPCOT are generally better than the results obtained with the deterministic version.

CONCLUSIONS

An efficient algorithm is developed which allows for uknown source emission rate estimation with data assimilation of gamma dose measurements in the framework of both stochastic and deterministic versions of Lagrangian puff atmospheric dispersion model DIPCOT. The algorithm is implemented for two different methods of gamma dose rate calculation – the semi-infinite cloud approximation and a more detailed numerical method. The proposed "control vector reduction" procedure allows for substantial improvement in numerical efficiency and accuracy of the data assimilation method and makes it suitable for the use with stochastic version of DIPCOT.

The developed method is evaluated against the measurements in field experiment on atmospheric dispersion of ^{41}Ar performed in Mol [8]. In DA runs the first guess source emission rate has been set by a factor of 10 greater and also less than the true one. In all cases of DA runs the statistical indicators of errors of the estimated source emission rate as compared to the measured one were greatly reduced. The errors of the calculated fluence rates as compared to the set of independent measurements, which were not used in DA procedure were also substantially reduced.

The presented methodology is developed and evaluated for a single radionuclide, if there are different radionuclides then the first guess estimation of source term consists of information about amounts of separate nuclides. The easiest but coarse way to account for different nuclides in the present algorithm is to fix their proportion to the value specified in first guess estimation. The more complex and more correct way is to solve adjoint equations to the system of equations governing radionuclides decay, which should be subject to further work.

Despite the fact that there are not a lot available data sets with gamma dose measurements, the data set that we used covers atmospheric stability categories ranging from slightly unstable to very stable. Therefore, the presented results demonstrate potential of the developed algorithm for application in operational nuclear emergency response systems. However operational implementation of the proposed algorithm will require additional validation studies..

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FIGURE CAPTIONS

Figure 1. Map of sensors locations for conditions of Mol experiment; '×' – location of the source of release; '■' – locations of the DK-NAI sensors (A-D) on Wednesday; '●' – location of the DTU-HPGe sensor on Wednesday; '□' – locations of the DK-NAI sensors (A-D) on Thursday; 'Δ' – locations of the SCK-NAI sensors (1-4) on Thursday; '○','*' – locations of the DTU-HPGe and SCK-HPGe sensors on Thursday. Coordinates are shown in UTM coordinate system. The zone is 31U.

Figure 2. Release rate estimations as result of fluence rate assimilation performed for conditions of Mol experiment.. Results obtained with stochastic mode of DIPCOT operation are presented. a) - Day 1 experiment; b) - Day 2 experiment. Thick black line – true release rate; square symbols – first guess estimation of release rate; dashed line – results of DA run with P=1; thin solid line – results of DA run with P=9 (a) and with P=4 (b); dotted line – results of DA run with P=27 (a) and with P=16 (b); circles – results of DA run with P=3510.

Figure 3. Measured by DTU-HPGe sensor (symbols) and calculated fluence rates with data assimilation for the case of Day 1 experiment. Bold and dotted lines - stochastic version of DIPCOT, P=27 and P=1 respectively; dashed and thin solid lines – deterministic DIPCOT, P=27 and P=1 respectively.

Figure 4. Measured (symbols) and calculated fluence rates with data assimilation for the case of Day 2 experiment, P=16. a) - SKC-NaI-2, b) - SKC-NaI-3, c) - SKC-NaI-4, d) - DTU-HPGe sensors. Thick line – deterministic run; dotted line – stochastic run.

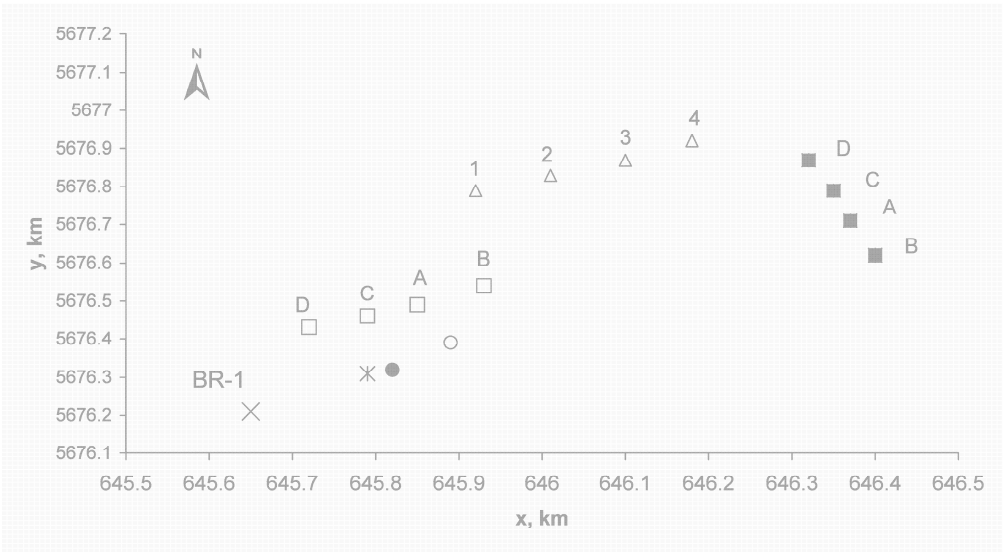
Tables

Table 1. Mean absolute relative error (MAE) and mean relative biases (MRB) of calculated source function as compared to measured source function. Errors of the first guess source function as well as the errors of source functions corrected in assimilation runs with different values of CVR parameter P and with puff and Lagrangian models for 03/10/2001 and 04/10/2001 Mol, Belgium experiments are shown.

Experiment (date)	**P**	**Stochastic /Deterministic (S/D)**	**MAE**	**MRB**
03/10/2001	First guess	S,D	9.0	9.0
03/10/2001	111	D	0.55	-0.37
03/10/2001	27	D	0.51	-0.38
03/10/2001	9	D	0.44	-0.38
03/10/2001	1	D	0.50	-0.50
03/10/2001	111	S	0.71	-0.27
03/10/2001	27	S	0.59	-0.28
03/10/2001	9	S	0.45	-0.35
03/10/2001	1	S	0.39	-0.39
04/10/2001	First guess	S,D	0.9	-0.9
04/10/2001	16	D	0.40	-0.36
04/10/2001	4	D	0.42	-0.38
04/10/2001	1	D	0.62	-0.52
04/10/2001	16	S	0.42	-0.37
04/10/2001	4	S	0.44	-0.38
04/10/2001	1	S	0.58	-0.46

Table 2. Normalized mean square errors and fractional biases calculated on the basis of sensors which were not used in DA. Results of forward runs with first guess source function, as well the results of assimilation runs with different values of CVR parameter P and with puff and Lagrangian models for 03/10/2001 and 04/10/2001 Mol, Belgium experiments are shown.

Experiment (date)	**P**	**Stochastic /Deterministic (S/D)**	**NMSE**	**FB**
03/10/2001	First guess	D	8.94	0.83
03/10/2001	111	D	0.61	-0.19
03/10/2001	27	D	0.51	-0.17
03/10/2001	9	D	0.57	-0.19
03/10/2001	1	D	0.48	-0.3
03/10/2001	First guess	S	9.7	0.83
03/10/2001	111	S	0.83	-0.27
03/10/2001	27	S	0.58	-0.218
03/10/2001	9	S	0.61	-0.26
03/10/2001	1	S	0.23	-0.21
04/10/2001	First guess	D	13.94	0.84
04/10/2001	16	D	0.63	-0.059
04/10/2001	4	D	0.85	-0.12
04/10/2001	1	D	1.71	-0.33
04/10/2001	First guess	S	17.32	0.88
04/10/2001	16	S	0.58	0.039
04/10/2001	4	S	0.73	-0.013
04/10/2001	1	S	0.94	-0.17



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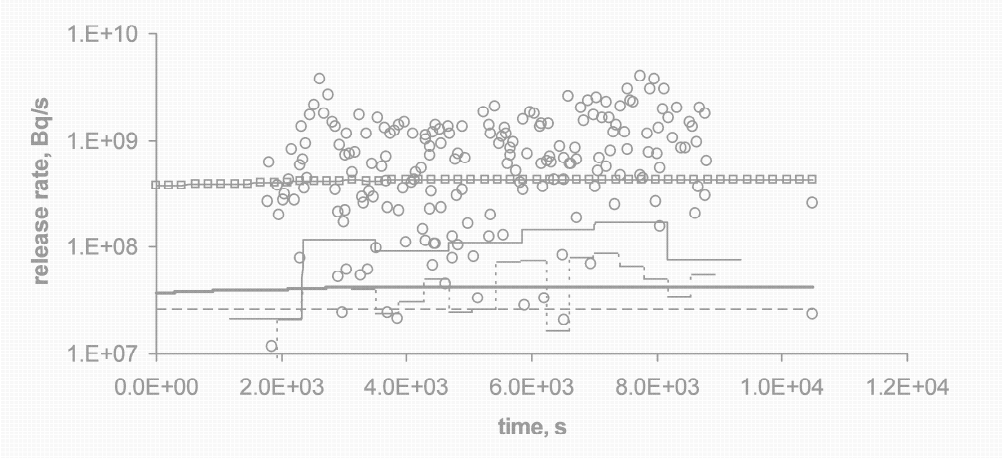

Figure 2-a)
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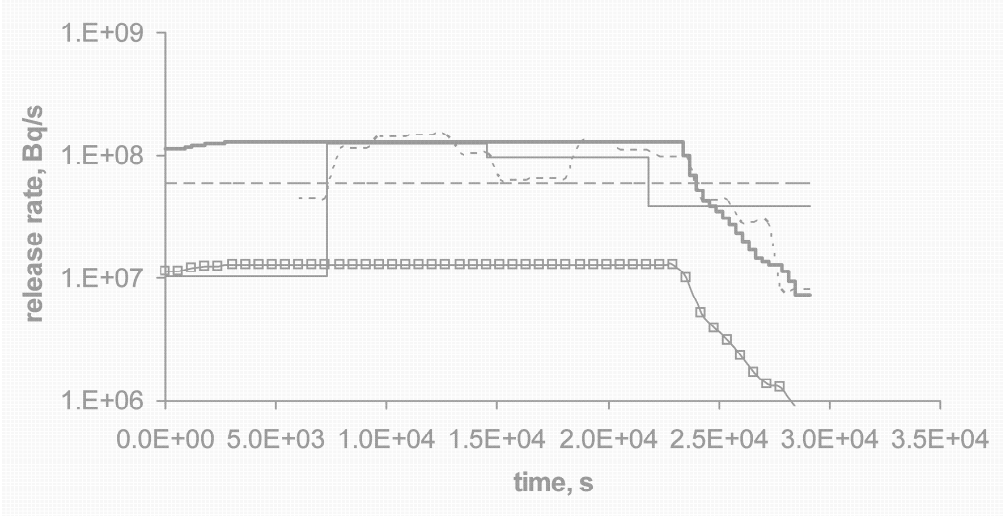

Figure 2-b)

Figure 2. Release rate estimations as result of fluence rate assimilation performed for conditions of Mol experiment.. Results obtained with stochastic mode of DIPCOT operation are presented. a) - Day 1 experiment; b) - Day 2 experiment. Thick black line – true release rate; square symbols – first guess estimation of release rate; dashed line – results of DA run with P=1; thin solid line – results of DA run with P=9 (a) and with P=4 (b); dotted line – results of DA run with P=27 (a) and with P=16 (b); circles – results of DA run with P=3510.
1787x945mm (87 x 85 DPI)

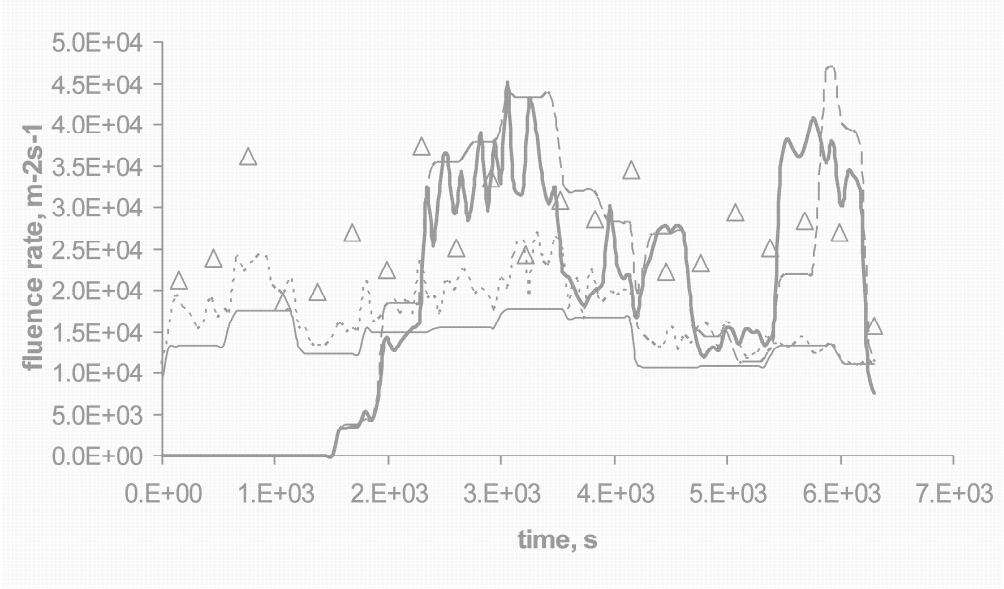

Figure 3. Measured by DTU-HPGe sensor (symbols) and calculated fluence rates with data assimilation for the case of Day 1 experiment. Bold and dotted lines - stochastic version of DIPCOT, P=27 and P=1 respectively; dashed and thin solid lines – deterministic DIPCOT, P=27 and P=1 respectively.
1795x1081mm (87 x 85 DPI)

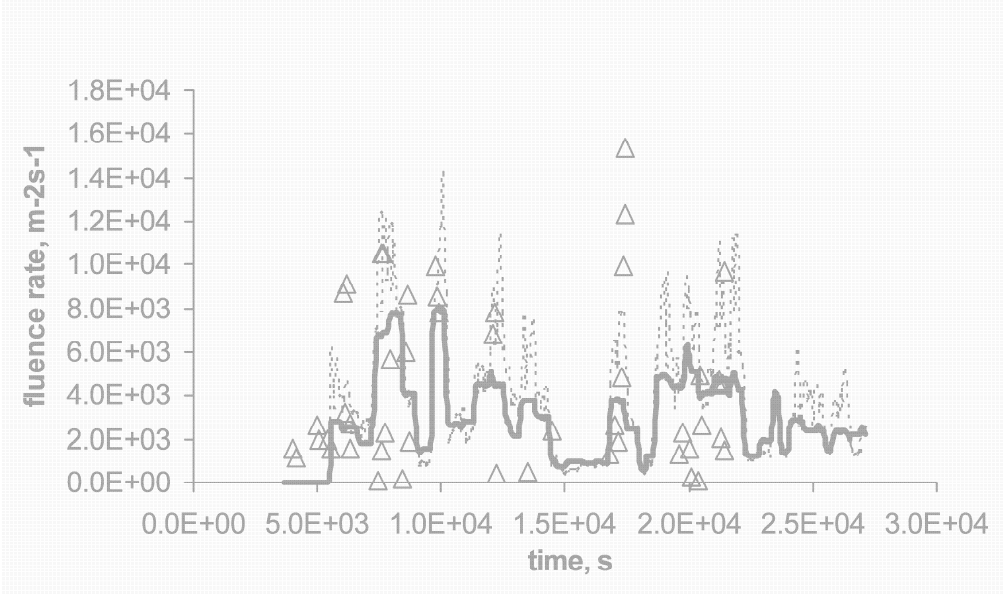

Figure 4-a)
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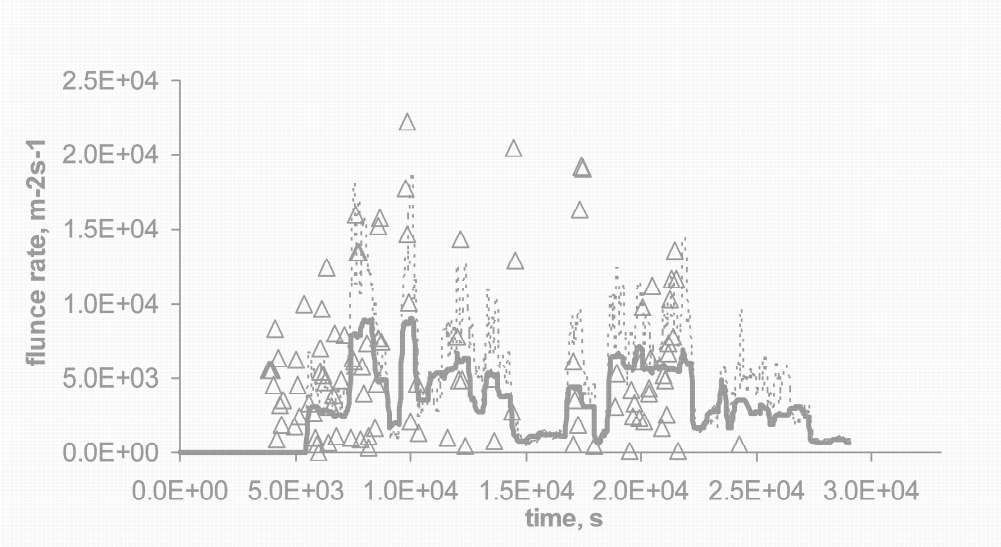

Figure 4-b)
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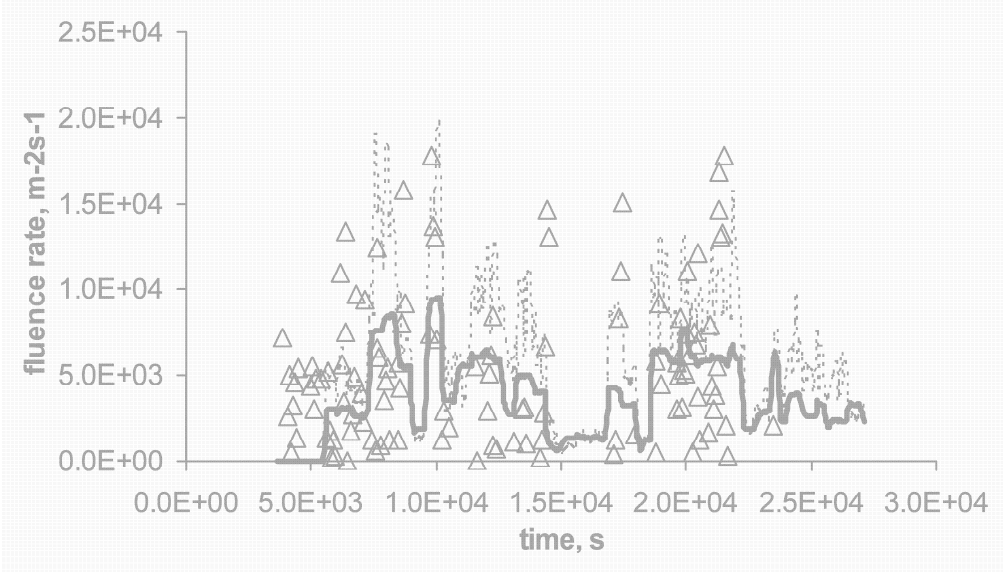

Figure 4-b)
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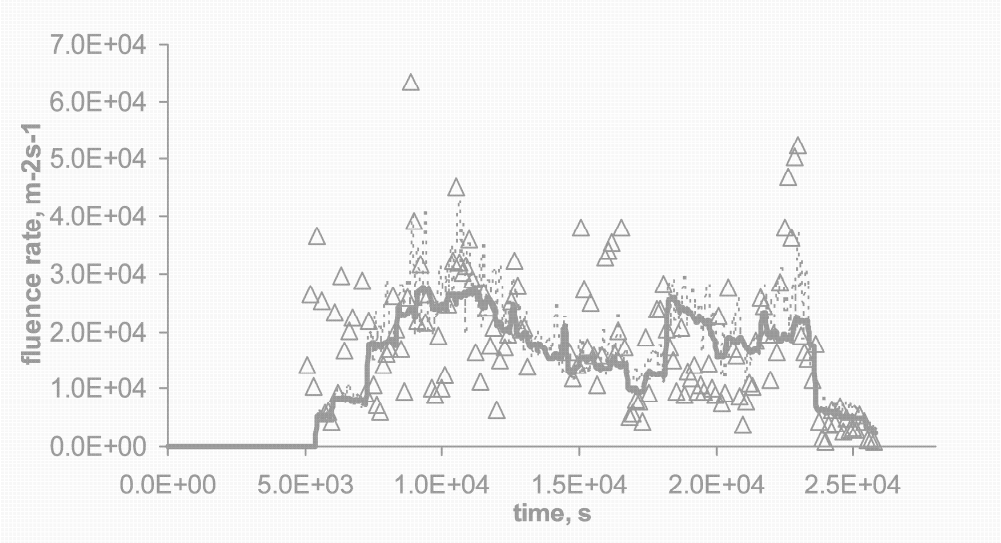

Figure 4-d)

Figure 4. Measured (symbols) and calculated fluence rates with data assimilation for the case of Day 2 experiment, P=16. a) - SKC-NaI-2, b) - SKC-NaI-3, c) - SKC-NaI-4, d) - DTU-HPGe sensors. Thick line – deterministic run; dotted line – stochastic run.
1755x980mm (87 x 85 DPI)