# Chapter 10
# Advanced Source Inversion Module of the JRODOS System


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**Abstract** The development of the source inversion algorithm is described which allows for estimation of the release rates of multiple nuclides and source height with the use of gamma dose rate (GDR) measurements. The method is applicable for the dispersion problems of different spatial scales: from $\sim$1 to $\sim$1000 km. The variational formulation of source inversion problem is used in which unknown release rates of different nuclides are adjusted to minimize the difference of calculated values and measurements. The sensitivities of calculated results with respect to release rates of different radionuclides are calculated with the aid of atmospheric transport model DIPCOT and the source receptor matrix (SRM) is thus constructed. The source inversion problem is regularized using prior (first guess) estimation of release rates. The method is proposed to account for the restrictions on the ratios of the release rates of different radionuclides in formulation of source inversion problem which allows for the assessment of the nuclide composition in radioactive release. The above restrictions are evaluated using the first guess source term. Parameterizations for the regularization parameters of source inversion problem which include root mean squared errors of measurents, first guess release rates, calculated values etc., are developed. The method was successfully tested using

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artificial measurements precalculated for the conditions of the ETEX experiment. Pilot implementation of the developed algorithm in the European nuclear emergency response system JRODOS is described.

**Keywords** Inverse problem · Source term estimation · Radioactive release RODOS

## 10.1 Introduction

After the accidents at nuclear power plants (NPPs) in Chernobyl and Fukushima a great effort has been spent for the development of algorithms capable of calculation of the source terms following the accident. These algorithms could be subdivided in two broad categories. The algorithms in the first category could be called 'bottom-up' (or process-based) because information concerning technical details of the accident and models of the physical processes in the reactor are used to assess the release rates. Excellent example of the source terms calculated in this way is the library of source terms described by Landman (2007). The difficulty in application of the 'bottom-up' algorithms arises from the very high uncertainty by a factor of 10–100 of their results even when exact information of technical details of the accident is available (US NRC 1990).

The algorithms in the second category could be classified as 'top-down' (or measurement-based) since release rates are evaluated by altering the solution of atmospheric transport model to measuremed values. These 'top-down' algorithms are frequently called also the 'source inversion' algorithms since the described problem is a particular example of the inverse problem occurring in many scientific and practical applications (Tarantola 2005). The drawback of the 'top-down' approach is that the inverse problem is frequently ill-conditioned, i.e., the solution is non-unique (or unstable) unless the problem is properly regularized with the aid of first guess estimations of the release rates which in turn are usually obtainted by using 'bottom-up' approaches.

In present work inverse modeling is employed and the source inversion is treated as a variational data assimilation problem with use of so-called source-receptor matrices (SRMs). The source receptor matrix characterizes sensitivity of calculated values (air concentrations, deposition, gamma dose rates) at points of measurements to values of nuclides emission rates. In present work SRM is calculated in a forward run of atmospheric dispersion model DIPCOT following the approach described by Tsiouri et al. (2012). The above approach was modified to take into account influence of deposition on gamma doses.

SRM concept can be applied both to scenarios where the source location is known or not. Scenarios where the source location is known (a nuclear facility) are considered. The estimated parameters are: (a) isotopic composition of the release,

(b) time-dependent emission rates and (c) effective height of the release. Generally, besides the source term, many other inputs to the dispersion models can be estimated. It is assumed that these inputs—including meteorological fields—are good enough to produce useful results.

This concept was successfully applied in the field of radiation protection (Davoine et al. 2007; Winiarek et al. 2011; Stohl et al. 2012; Saunier et al. 2013). However as a consequence of the described difficulties in achieving of robust solution of the inverse problems the source-inversion algorithms are usually not included in automated real-time decision support systems like JRODOS which are usually used in emergency phase of the accident. A strongly simplified form of source inversion method was developed for operational application in the context of the Comprehensive Nuclear Ban Treaty (Wotawa et al. 2003). In the work of Winiarek et al. (2011), automated algorithms were developed for nearly real-time evaluation of radioactive releases using concentration measurements. However GDRs measurements are much more widely available than concentrations and GDR measurements are routinely collected by every nuclear power plant. The problem of source term estimation based on GDR measurements is especially complex because the nuclide composition of release is poorly known in advance. In a work of Saunier et al. (2013) it was proposed to use information regarding possible ranges of ratios of released inventories of different nuclides to solve the problem of source inversion in case of multiple nuclides using GDR measurements and variational data assimilation method. In the work by Zhang et al. (2017) the Ensemble Kalman Filter was used for the same purpose. In frame of the EU FP7 PREPARE project (Raskob et al. 2016) the automated source inversion algorithm was developed in which GDR measurements collected at different distances from the source could be used to evaluate release rate, source height and nuclide composition of the release. The brief summary of the developed algorithm in frame of the PREPARE project was previously published in short communication by Kovalets et al. (2016). Recently the algorithm was integrated in the EU decision support system in case of nuclear emergencies JRODOS (Ievdin et al. 2010). The present work presents the algorithm and results of its testing in technical detail.

The novelty of present work consists in creation of the algorithm for the identification of the unknown time dependent release rate and nuclide composition following radiological accident using GDR measurements. In this algorithm all regularization parameters are constructed on the basis of the first guess source term provided by user with the aid of empirical parameterizations. The algorithm is implemented in automated Source Inversion Module of the JRODOS nuclear emergency response system and hopefully will become available to the users of the system.

## 10.2 Source Inversion Method

### *10.2.1 Source Term Estimation as an Inverse Problem*

Consider at first the most general statement of the inverse problem following Tarantola (2005). The observations are combined in a vector $\bar{y} \in R^{N_o}$, where $N_o$ is total number of measurements (in general different than the number of monitoring stations). The model parameters to be found in process of the solution of the inverse problem (such as time distributed source rate and other) are combined in a vector $\bar{x}$ (sometimes called "control vector"), while other parameters which are considered known are combined in vector $\bar{i}$. The model can be understood as—generally non-linear—operator $g(.)$ transforming sources to measurements: $\bar{y} \approx g(\bar{x})$ (approximate equality means that model is imperfect). Denote the conditional distribution function of the observations with given parameter vectors as $P(\bar{y}|\bar{x}, \bar{i})$. Prior information about vector $\bar{x}$ is described by the conditional distribution function $P(\bar{x}|\bar{i})$. Then the inverse problem could be posed as the problem of finding posterior distribution function $P(\bar{x}|\bar{y}, \bar{i})$ of the vector $\bar{x}$ with given vectors $\bar{y}$ and $\bar{i}$. The famous Bayes formula is used: $P(\bar{x}|\bar{y}, \bar{i}) \propto P(\bar{y}|\bar{x}, \bar{i})P(\bar{x}|\bar{i})$.

The important particular case is when distribution functions are Gaussian. Let also the covariance matrix of the distribution be diagonal. Then the distribution functions have the following form:

$$P(\bar{y}|\bar{x}, \bar{i}) \propto \exp\left[-\sum_l \frac{(y_l - g_l(\bar{x}))^2}{\sigma_{O,l}^2 + \sigma_{M,l}^2}\right] \tag{10.1}$$

$$P(\bar{x}|\bar{i}) \propto \exp\left[-\sum_l \frac{(x_l - x_l^B)^2}{\sigma_{B,l}^2}\right] \tag{10.2}$$

Here $\sigma_{B,l}$ is standard deviation of the prior (also called 'background' or 'first guess') estimate of the $l$th component of the vector $\bar{x}$; $\sigma_{M,l}$ is the rms of model error (provided that correct values of $\bar{x}$ are known) and $\sigma_{O,l}$ is the rms error of observations. Then posterior distribution function is of the form:

$$P(\bar{x}|\bar{y}, \bar{i}) \propto \exp\left[-\left\{\sum_l \frac{(y_l - g_l(\bar{x}))^2}{\sigma_{O,l}^2 + \sigma_{M,l}^2} + \sum_l \frac{(x_l - x_l^B)^2}{\sigma_{B,l}^2}\right\}\right] = \exp(-J). \tag{10.3}$$

Thus maximum of probability distribution function (10.3) coincides with the minimum of function $J$:

$$J(\bar{x}) = \sum_l \frac{(y_l - g_l(\bar{x}))^2}{\sigma_{O,l}^2 + \sigma_{M,l}^2} + \sum_l \frac{(x_l - x_l^B)^2}{\sigma_{B,l}^2} = (\bar{y} - g(\bar{x}))^{\mathrm{T}} \underline{\underline{R}}^{-1} (\bar{y} - g(\bar{x})) + (\bar{x} - \bar{x}^B)^{\mathrm{T}} \underline{\underline{B}}^{-1} (\bar{x} - \bar{x}^B). \tag{10.4}$$

Here $\underline{\underline{R}}$ and $\underline{\underline{B}}$ are error covariance matrices (diagonal in the present case) representing combined model and measurement errors and first guess errors, respectively: $r_{ll} = \sigma_R^2 = \sigma_{O,l}^2 + \sigma_{M,l}^2$, $b_{ll} = \sigma_{B,l}^2$.

Thus minimization of function (10.4) yields the solution of inverse problem $\bar{x}^S$:

$$\bar{x}^S = \arg\min_{\bar{x}}(J(\bar{x})). \tag{10.5}$$

Below control vector consists (unless noted otherwise) of time distributed emission rates: $\bar{x} = (q_1(t_1), q_2(t_1), \ldots, q_{N_{\mathrm{nu}}}(t_1), \ldots, q_{N_{\mathrm{nu}}}(t_{N_{\mathrm{s}}}))$, where $q_i(t_m)$ is release rate of $i$th nuclide at time step $m$, $N_{\mathrm{nu}}$ and $N_{\mathrm{s}}$ are number of nuclides and of time steps (onto which source function is split) respectively. In this case relationship $g(\bar{x})$ is linear and it is represented with matrix (sometimes called source-receptor matrix SRM): $g(\bar{x}) = \underline{\underline{G}}\bar{x}$, where the elements of SRM are then defined as $g_{ij} = \partial y_j / \partial x_i$.

The linear case is applicable to passive tracers and substances which do not undergo nonlinear chemical transformation and substances with a prescribed decay or growth rate, e.g. radionuclides (Seibert et al. 2002). In this case, the minimization problem (10.5) is a linear regression problem of minimizing the cost function

$$J(\bar{x}) = (\bar{y} - \underline{\underline{G}}\bar{x})^{\mathrm{T}} \underline{\underline{R}}^{-1} (\bar{y} - \underline{\underline{G}}\bar{x}) + (\bar{x} - \bar{x}^B)^{\mathrm{T}} \underline{\underline{B}}^{-1} (\bar{x} - \bar{x}^B) = J_1 + J_2 \tag{10.6}$$

Note that second term $J_2$ ensures strict convexity of the cost function (10.6) and thus uniqueness of the solution of minimization problem. Therefore, this term 'regularizes' the solution of (10.5). Without regularization the problem may be ill-conditioned, i.e. the solution is non-unique or unstable with respect to small changes in the parameters of the problem or with respect to numerical errors.

### 10.2.2 Nuclide Composition and Augmented Minimization Problem

The most common quantities used in the inversion of sources of radioactivity emitted into the atmosphere are the atmospheric activity concentration, the surface contamination (deposition) and the GDR from the cloud and deposited activity. Activity concentration and deposition measurements provide nuclide-specific data which can be used for estimation of isotopic composition of the release. In a few countries, concentration measurements of radioactivity are routinely available and

are used for source estimation (Winiarek et al. 2011). Many NPPs have the possibility to perform concentration measurements on an irregular basis, for instance, by using mobile laboratories in the case of a hypothetical release and measuring daily (or sub-daily) averaged concentrations of aerosols and nuclides of the iodine group. A significant obstacle in using these data in real-time applications is their poor time resolution, where sampling times are hours or even days long. Nevertheless, these data were successfully used for source inversion of the Fukushima disaster by Stohl et al. (2012), in tandem with the Lagrangian atmospheric transport model FLEXPART (Stohl et al. 2005).

Receptors measuring GDR are the most widespread observation system. Unless devices for in situ spectrometry are used, these data provide only bulk measurements of gamma dose from cloud and deposition with no information on isotopic composition of the release. However, these data provide real-time information on plume passage with high temporal resolution and networks of these receptors are present around every nuclear installation or even country-wide and are relatively dense.

For source term estimation using GDR measurements with the present method, at least approximate knowledge of nuclide ratios is assumed. Following Hofman et al. (2015) the method based on minimization of the cost function (10.6) is modified in a way that it can be used for inversion of a source term with multiple nuclides using bulk GDR measurements. Nuclide ratios enter the problem in the form of additional linear equations, where the deviations from prescribed ratios are weighted by factors. The error variance of these deviations allows to control how strictly the prescribed ratios are fulfilled.

Firstly, let us illustrate the method with a simple case—a release of Xe-133, I-131 and Cs-137. If release rates for the respective radionuclides are denoted with $x_1$, $x_2$ and $x_3$, the nuclide ratios are formulated as follows (constants are assumed for the sake of the example here): $x_1 = 10x_2$, $x_1 = 2x_3$. This can be re-written as

$$\begin{aligned} x_1 - 10x_2 &= 0 \\ x_1 - 2x_3 &= 0. \end{aligned} \tag{10.7}$$

The above equations are used to augment our cost function. It then reads as follows:

$$J(\bar{x}) = (\tilde{y} - \underline{\underline{\tilde{G}}}\bar{x})^{\mathrm{T}} \underline{\underline{\tilde{R}}}^{-1} (\tilde{y} - \underline{\underline{\tilde{G}}}\bar{x}) + (\bar{x} - \bar{x}^B)^{\mathrm{T}} \underline{\underline{B}}^{-1} (\bar{x} - \bar{x}^B) = J_1 + J_2. \tag{10.8}$$

Here $\tilde{y}$ and $\underline{\underline{\tilde{G}}}\bar{x}$ are augmented in a way that Eqs. (10.7) added to $\tilde{y} - \underline{\underline{\tilde{G}}}\bar{x}$:

$$\begin{bmatrix} y_1 \\ \vdots \\ y_m \\ 0 \\ 0 \end{bmatrix} - \begin{bmatrix} g_{11} & g_{12} & g_{13} \\ \vdots & \vdots & \vdots \\ g_{1m} & g_{2m} & g_{3m} \\ 1 & -10 & \\ 1 & & -2 \end{bmatrix} \begin{bmatrix} x_1 \\ x_2 \\ x_3 \end{bmatrix}. \tag{10.9}$$

Correspondingly, the observation error covariance matrix $\underline{\underline{R}}^{-1}$ is augmented by inversions of variances of ratio conditions deviations:

$$\underline{\underline{\tilde{R}}}^{-1} = \begin{bmatrix} \sigma_{y_1}^{-2} & & & & \\ & \ddots & & & \\ & & \sigma_{y_m}^{-2} & & \\ & & & \sigma_I^{-2} & \\ & & & & \sigma_{Cs}^{-2} \end{bmatrix}. \tag{10.10}$$

Coefficients $\sigma_{\mathrm{I}}^2$, $\sigma_{\mathrm{Cs}}^2$ determine how large variances are allowed for errors in (10.7): $\sigma_{\mathrm{I}}^2 = E\left((x_1 - 10x_2)^2\right)$, $\sigma_{Cs}^2 = E\left((x_1 - 2x_3)^2\right)$, and symbol $E$ stands for the expected value. Thus besides the ratios of nuclide release rates the augmented algorithm requires the specification of their error variances as well. These inputs will be discussed below in Sect. 10.2.4.

Extension of the definition of the above augmented matrices to a general case when there is total number of $N_{\mathrm{nu}}$ nuclides and the source function is discretized with the total number of $N_{\mathrm{s}}$ time steps is straightforward. Obviously it is not necessary to use ratios of release rates of different nuclides to release rate of only the first nuclide. On the contrary, as it will be demonstrated below in Sect. 10.2.4, if the 1st nuclide is noble gas, the range of uncertainties for the ratios $a_{i,m} = x_{(m-1)N_{\mathrm{nu}}+i}/x_{(m-1)N_{\mathrm{nu}}+1}$ (here $m$ stands for time index) will be too large for all indices $i$ which do not correspond to noble gas. Therefore the following function is defined:

$$J(i) = \{j \in (1, \ldots, N_{\mathrm{nu}}), j \neq i\}. \tag{10.11}$$

Equation (10.11) sets correspondence between nuclide number $i$ and nuclide number $j$ with respect to which ratio of release rates $a_{i,m} = x_{(m-1)N_{\mathrm{nu}}+i}/x_{(m-1)N_{\mathrm{nu}}+J(i)}$ is calculated. Obviously, for each time step there could be set $N_{\mathrm{nu}} - 1$ nuclide ratios. The specific form of $J(i)$ implemented for usage in the JRODOS system will be described below in Sect. 10.2.4.

### *10.2.3 Source-Receptor Matrix*

In this work the aim is to estimate the release rate for each of multiple simultaneously released radionuclides significantly contributing to the GDRs measured at receptors. Although the source location is well known in our scenario, all possible release intervals $s: 1 \le s \le N_s$ of all nuclides $i: 1 \le i \le N_{nu}$ must be treated as separate sources in order to achieve this. Under this assumption, each elements $g_{lk}$ of the SRM matrix $\underline{\underline{G}}$ represents the contribution of a source $k = (s-1) \cdot N_{nu} + i$ releasing a particular nuclide $i$ at a given time interval $s$ to total GDR measurement $l$ sampled in a particular time-space point (interval). In the case when also the effective height of the release is to be estimated, sources index $i$ in $\bar{x}$ and $\underline{\underline{G}}$ will also have to iterate over vertical release intervals.

Here the algorithm of SRM calculation implemented in Lagrangian puff model DIPCOT of JRODOS is presented. In contrast to Lagrangian particle model, in Lagrangian puff models puffs are characterized by their finite size (as in Gaussian puff models) but, as in Lagrangian models, turbulent mixing is taken into account by adding in the right part of the equation of puff's movement the wind fluctuations in addition to Reynolds-averaged wind field. This approach leads to significant reduction in computational time in the Lagrangian puff model as compared to Lagrangian particle model. This approach had been studied theoretically by De Haan and Rotach (1998) and widely applied practically. In particular, the Lagrangian puff version of DIPCOT has been extensively validated over numerous wind tunnel and field experiments [see the results referenced in Andronopoulos et al. (2009, 2010) and Davakis et al. (2007)].

Although DIPCOT atmospheric dispersion model does not take into account such processes as resuspension of radionuclides, diffusion in soil, and wash-out, it is likely that those processes are more important in the long term (months and years after the accident), while in emergency phase and for time scales of the few days after the release which correspond to the spatial scale of the dispersion problem up to $\sim$1000 km the above processes are of minor importance.

In the next three sections elements of SRM are derived at first for the simplified case of single point source with the number of release intervals coinciding with the number of puffs $N_s = N_p$. Extension of the presented relationships for the case of multiple sources is straightforward. Extensions of SRM relationships for the case of release intervals of realistically large duration are discussed in Sect. 10.2.3.

#### 10.2.3.1 SRM Calculation in Case of Concentration Measurements

In all kinds of puff models concentration at a given point $(x, y, z)$ and time $t$ is calculated using the relationship (single nuclide is considered for simplicity):

$$C(x,y,z,t) = \frac{1}{(2\pi)^{3/2}} \sum_{p=1}^{N_p} \frac{M_p}{\sigma_{xp}\sigma_{yp}\sigma_{zp}} \exp\left[-\frac{1}{2}\frac{(x_p - x)^2}{\sigma_{xp}^2}\right] \exp\left[-\frac{1}{2}\frac{(y_p - y)^2}{\sigma_{yp}^2}\right]$$
$$\left\{\exp\left[-\frac{1}{2}\frac{(z_p - z)^2}{\sigma_{zp}^2}\right] + \exp\left[-\frac{1}{2}\frac{(z_p + z - 2z_g)^2}{\sigma_{zp}^2}\right]\right\},$$
$$M_p = q_p \tau \gamma(t,\tau,p) \times \Upsilon(t,\lambda,\ldots). \tag{10.12}$$

Here $\tau$ is time interval between appearance of successive puffs, $N_\mathrm{p}$ is the number of puffs, ($x_\mathrm{p}$, $y_\mathrm{p}$, $z_\mathrm{p}$)—coordinates of puff's centre, ($\sigma_{xp}$, $\sigma_{yp}$, $\sigma_{zp}$)—dispersions of matter distribution in a puff (calculated using different parameterizations), $z_\mathrm{g}$—height of ground surface above sea level, $M_\mathrm{p}$ inventory of a puff (total amount of radioactivity in Bq), $q_\mathrm{p}$—source rate corresponding to appearance of $p$th puff, function $\gamma(t,\tau,p) = \mathrm{sgn}(t - p\cdot\tau)$ excludes influence of puffs appeared after time $t$. Term $\Upsilon(t,\lambda,\ldots)$ in generalized form takes into account radioactive decay, dry and wet deposition. For instance in the case of noble gases, when deposition is absent, $\Upsilon(t,\lambda,\ldots) = \exp(-\lambda(t-(p-1)\tau))$, and $\lambda$ is the radioactive decay constant.

In Eq. (10.12) full reflection of the plume at the ground surface is assumed. Variations of Eq. (10.12) are possible to take into account reflection from the upper lid of the mixing layer, multiple nuclides with transformations described by matrix, etc. However in all cases it is possible to derive analytical relationships for the elements of source-receptor matrix.

Say, if relationship (10.12) is used, then for the case of instantaneous concentration at a spatial point $\bar{r}_j$ at particular time $t^k$, the elements of source-receptor matrix are calculated using the formula: $g^{(c)}_{l,(p-1)\cdot N_{nu}+i} = g^{(c)}\left(\bar{r}_j, t^k, p, i\right)$. Here index $l$ corresponds to the number of measurements (according to their storage in vector $\bar{y}$), $g^{(c)}(\bar{r}_j, t^k, p, i) = \partial C(\bar{r}_j, t^k)/\partial q_{p,i}$ obtained from formula (10.12) is the sensitivity of model-calculated concentration with respect to inventory of $i$th radionuclide in puff $p$, $g^{(c)}_{l,(p-1)\cdot N_\mathrm{nu}+i}$ is the corresponding element of matrix $\underline{\underline{G}}$.

#### 10.2.3.2 SRM Calculation in Case of Deposition Measurements

Dry deposition $C_\mathrm{d}$ of the nuclide $i$ in a given spatial point $j$ characterized by the position vector $\bar{r}_j$ is calculated using the equation:

$$\frac{\mathrm{d}C_{d,i}(t,\bar{r}_j)}{\mathrm{d}t} = F_\mathrm{d}(t,\bar{r}_j)C_a(t,\bar{r}_j) - \lambda_i C_\mathrm{d}(t,\bar{r}_j). \tag{10.13}$$

Here the first term in the r.h.s. described deposition flux, which is proportional to ground-level concentration $C_a$ in the point of interest; proportionality coefficient $F_\mathrm{d}$

depends on land use, friction velocity, etc. The second term describes radioactive decay of the deposited radioactivity.

Provided that initial deposition is zero the solution of the above equation depends on time in a following way:

$$C_{\mathrm{d},\mathrm{i}}(t,\bar{r}_j) = \int_0^t F_{\mathrm{d}}(\tau,\bar{r}_j)C_a(\tau,\bar{r}_j)\exp(-\lambda_i(t-\tau))\mathrm{d}\tau. \tag{10.14}$$

In model run the above integral is approximated by sum which is calculated in a time loop:

$$C^n_{\mathrm{d},i}(t^k,\bar{r}_j) = \sum_{n=0}^{k} F_{\mathrm{d}}(n\cdot\mathrm{d}\tau,\bar{r}_j)C^n_a(n\cdot\mathrm{d}\tau,\bar{r}_j)\exp(-\lambda_i(t^k-n\cdot\mathrm{d}\tau))\mathrm{d}\tau. \tag{10.15}$$

Calculation of sum in r.h.s. of the above equation is performed in a time loop (index $n$). Let $l$th element $y_l$ of measurement vector $\bar{y}$ is deposition at point $\bar{r}_j$ and at time $t_k$. Using the above formula the elements of the $l$th row of SRM corresponding to deposition measurements are calculated using the formula:

$$\begin{aligned} g^{(\mathrm{dep})}_{l,(p-1)\cdot N_{nu}+i} &= g^{(\mathrm{dep})}(\bar{r}_j,t^k,p,i) \\ &= \sum_{n=0}^{k} F_{\mathrm{d}}(n\cdot\mathrm{d}\tau,\bar{r}_j)g^c(\bar{r}_j,t^n,p,i)(n\cdot\mathrm{d}\tau,\bar{r}_j)\exp(-\lambda_i(t^k-n\cdot\mathrm{d}\tau))\mathrm{d}\tau. \end{aligned} \tag{10.16}$$

Here the same as above $g^{(\mathrm{dep})}(.)$ and $g^{(\mathrm{dep})}_{*,*}$ are sensitivity function of model-calculated deposition with respect to inventory of $i$th radionuclide in puff $p$ and the corresponding element of SRM. Thus in a time loop the elements of SRM corresponding to deposition measurements are updated using relationships for $g^{(c)}$ according to formula (10.16). It should be noted that change of deposition due to vertical diffusion in soil, wash-out, resuspension, etc. is not taken into account in DIPCOT and hence not accounted for in SRM calculation.

#### 10.2.3.3 SRM Calculation in Case of GDR Measurements

Ambient gamma radiation is created by airborne radionuclides and by deposition. GDR created by deposition is calculated in DIPCOT in a given point using the assumption of infinite homogeneous deposition. The resulting GDR value linearly depends on depositions of radionuclides at a given location.

There are different methods of calculating GDR from cloud using puff data. The methods range by the level of complexity from simplified method of infinite cloud

approximation to comprehensive 3D integration. The method of intermediate level of complexity recently implemented in DIPCOT (Andronopoulos and Bartzis 2010) closely approximates full 3D integral while retaining computational efficiency. However for all kind of methods of GDR calculation the resulting values are linear combination of concentration fields and hence puff inventories.

Therefore in simplest case of infinite cloud approximation elements $g^{(gdr)}$ of source-receptor matrix corresponding to GDR measurements are calculated as linear combination of $g^{(\mathrm{dep})}$ and $g^{(c)}$:

$$g^{(gdr)}_{l,(p-1)\cdot N_{nu}+i} = \beta_c \cdot g^{(c)}(\bar{r}_j, t^n, p, i) + \beta_d \cdot g^{(\mathrm{dep})}(\bar{r}_j, t^n, p, i). \tag{10.17}$$

In the case of more complex method of calculation of GDR from the cloud, the second term in the above equation is preserved, while the first term is calculated using the relationships presented by Tsiouri et al. (2012).

#### 10.2.3.4 Reduction of Control Vector

In the case of the Lagrangian puff model, the elements of source-receptor matrix are stochastic variables and this creates difficulties in direct usage of that matrix in the solution of the inverse problem. This difficulty can be overcome by the simple approach used by Tsiouri et al. (2012). Assume that during some interval $\Delta t$ the source rate could be considered constant. This is a common assumption in all existing accidental scenarios. For instance, in all of the scenarios existing in JRODOS (Landman 2007) the value of $\Delta t$ is in the range $10^3 - 10^4$ s. At the same time, the time interval between appearance of successive puffs in Lagrangian model is typically much shorter $\tau \approx 0.1-10$ s. Thus practically $\Delta t/\tau = \Pi \gg 1$.

Then the release period can be divided in $N_\mathrm{s}$ time intervals of size $\Delta t$ with $\Pi$ puffs in each time interval, so that total number of puffs is $N_\mathrm{P} = \Pi \cdot N_\mathrm{s}$. Even when $\Delta t = 10$ minutes, the value of $\Pi$ will be large (taking into account the above estimates for $\tau$). In each interval $m$ $(1 \le m \le N_\mathrm{s})$ the release rate can be considered equal to $q_m$; the values of $q_m$ constitute the reduced control vector $\bar{x}$ of size $N_\mathrm{s}$.

In the 'reduced' (and regularized) minimization problem the cost function is minimized with respect to the reduced control vector $\bar{x}$. Elements of source-receptor matrix $\underline{\underline{G}}$ corresponding to reduced minimization problem are obtained by summing elements of source receptor matrix (let's denote it here $g^{\mathrm{full}}$) corresponding to 'full' minimization problem (i.e. in which control vector consist of individual inventories of each puff):

$$g_{l,m} = \sum_{p=1}^{\Pi} g^{\mathrm{full}}_{l,((m-1)\Pi+p)}, \quad \forall l, m : 1 \le l \le N_o, 1 \le m \le N_\mathrm{s}. \tag{10.18}$$

According to Central Limit Theorem statistically stable values of $g_{l,m}$ are obtained when $\Pi$ are large enough.

The described regularization procedure has been previously implemented within the DIPCOT model (Tsiouri et al. 2012) and tested against field measurements of GDRs. The case of only one nuclide and GDR only from cloud has been considered in those works.

## *10.2.4 Parameters of the Minimization Problem*

### 10.2.4.1 First Guess Estimation of Source Term and Its Error Variances

As it was mentioned in previous sections, the usage of the prior information about the source term via the second term in the r.h.s. of Eq. (10.6) regularizes the solution of minimization problem. This form of regularization is useful when a first guess of emissions of particular nuclides exists (e.g. from accident-based analysis like MELCOR calculations (https://melcor.sandia.gov). The task of assignment of the first guess estimation of the source term is to be carried out by the user of the JRODOS system prior to running source inversion algorithm. The library of existing in JRODOS pre-defined source terms (Landman 2007) could assist in this task. The JRODOS system allows user extending this library by importing source term created by user into JRODOS database (Ievdin et al. 2015).

Once the first guess source term is provided, the error variances of this prior source term estimation are to be estimated. For the real-time applicability of the method robust simplifications are frequently applied, usually assuming error variance to be certain fraction of some norm of the source function, such as average or maximum value of the source function through the release interval (Tsiouri et al. 2012; Winiarek et al. 2011). In the present work, the same simplification is used: magnitude of error variance $\sigma_{B,i}$ of first guess source rate for the $i$th nuclide $\bar{x}_i^B$ is set equal to a certain fraction of its maximum value during the release period:

$$\sigma_{B,i} = \alpha_B \left\| \bar{x}_i^B \right\| = \alpha_B \max_m (x_{i,m}^B). \tag{10.19}$$

The particular value for $\alpha_B$ to be used by default in operational applications of the method could be assessed from the fact that the first guess estimations of the source term are usually characterized by very high uncertainty by a factor of 10–100 (US NRC 1990). If, for instance, there is a constant release of a single nuclide with the release rate estimated as $q_B$ and true release rate is expected to be found within the interval $(0.1q_B, 10q_B)$; this situation yields the estimate: $\sigma_B \approx 5q_B$. Thus the value of $\alpha_B = 5$ could be proposed to be used by default in emergency situations when first guess source term are characterized by high levels of uncertainty.

Note, that the example described above corresponds to the skewed probability distribution of the first guess estimation, while the Bayesian formulation (10.1–10.3) leading to cost function (10.6) assumed Gaussian distributions. Resolving of such controversy requires a change in the cost function as shown in work of Bocquet (2008), and could be subject for future research.

#### 10.2.4.2 Error Variances of Observed and Model-Calculated Values

Let's consider rms errors of observations $\bar{\sigma}_{\mathrm{O}}$ and of model $\bar{\sigma}_{\mathrm{M}}$. For implementation of source inversion algorithms in a real-time emergency response system like JRODOS some automatic procedure should exist for their assignment. Recall that observations and model errors enter the cost function (10.6) through the covariance matrix $\underline{\underline{R}}$ having diagonal elements $r_{ll} = \sigma_R^2 = \sigma_{O,l}^2 + \sigma_{M,l}^2$. Let's consider at first the error of observations. It consists of the instrumental error $\sigma_{\mathrm{d}}$ of the measurement device and an additional error which is introduced by the existence of a background field $\varphi$ of the measured quantity (e.g. background GDR). The user may wish to discard measurements which are close to the average value of the background field $\varphi_0$, therefore the error variance of observations is set to

$$\sigma_o = \alpha_\varphi \varphi_0 + \sigma_{\mathrm{d}}. \tag{10.20}$$

Here the value of $\alpha_\varphi$ depends on type of measurements and magnitude of natural perturbations of background field. For instance, in Ukraine the background GDR is about $\varphi_0 = 10^{-4}$ mSv/h, while the lowest GDR levels which can be used for alarm are considered between $1.5\varphi_0$ to $3\varphi_0$. Therefore the default value for $\alpha_\varphi = 2$ could be proposed for GDR measurements. In general the measurements which are close to background value contain also some useful information, especially in complex release and atmospheric transport patterns therefore by adjusting $\alpha_\varphi$ the user can control the degree to which such measurements are taken into account.

One possible choice of the value for observational error $\sigma_{\mathrm{d}}$ is to set it proportional to the measured value

$$\sigma_{d,i} = c_1 y_i + c_0, \tag{10.21}$$

where the constant of proportionality, $c_1$, is specified, e.g., by the manufacturer of a measuring device or by an expert estimate (Seibert et al. 2011; Tsiouri et al. 2011); $c_0$ can represent a "background value", a minimum measurable value. For the robust usage of source inversion method the information about the values of $c_1, c_0$ should ideally be present in JRODOS measurement database for each measurement station. However presently such data are absent in JRODOS database. Therefore the default value $c_0 = 2 \times 10^{-5}$ mSv/h is assumed for GDR measurements, typically characterizing such measurements used at Ukrainian NPPs. The value of $c_1$ is not taken into account in calculating $\sigma_{\mathrm{d}}$ since as far as for model error $\sigma_{\mathrm{M}}$ its

proportionality to measurement values is also assumed below; the value of $c_2$ could be implicitly taken into account in calculating $\sigma_M$.

The estimation of model error $\sigma_M$ is problematic. It can be, for example, be estimated using statistics of an ensemble of different dispersion models or one model with different inputs for a given source vector $\bar{x}$. Real-time application of such method is difficult. Therefore, in the present work the simple assumption is used:

$$\sigma_{m,i} = \alpha_m y_i. \tag{10.22}$$

The value of $\alpha_m$ could be estimated from the following considerations. Recall that the physical meaning of $\sigma_m$ is the error of model provided that exact values for the control vector (source term) are known. Therefore in $\sigma_m$ errors of different parameters that are known approximately but do not enter control vector (meteorology, parameters of turbulent mixing parameterizations, etc.) are implicitly taken into account.

In numerous validation studies of atmospheric dispersion models, a commonly used statistics for measuring model performance against measurements is the normalized mean squared error: $\mathrm{NMSE} = \left\langle (y_m - y_o)^2 \right\rangle / \langle y_m \rangle \langle y_o \rangle = \sigma_m^2 / \langle y_m \rangle \langle y_o \rangle$. The values of NMSE in different studies range from 0.5 to 10 and more. As it could be seen from the above relationship the following approximation for $\alpha_m$ could be proposed: $\alpha_m \approx \sqrt{\mathrm{NMSE}}$. The default value $\alpha_m = 1$ is proposed.

#### 10.2.4.3 Ratios of Release Rates for Different Nuclides and Corresponding Error Variances

The modified formulation of the minimization problem was described in Sect. 10.2.2, which is used to take into account information regarding ratios of source rates of different nuclides consisting of release composition. Nuclides are subdivided into groups, and release ratios are calculated for different nuclides within each group with respect to the 'reference' nuclide representing each group. Additionally, ratios of release rates between 'reference' nuclides are calculated (Fig. 10.1). By default 3 groups of nuclides are used: Noble gases, Iodine and Aerosols.

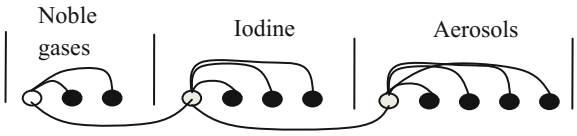



**Fig. 10.1** Scheme of calculating ratios between nuclides: vertical lines denote groups of nuclides; empty circle—representative nuclide within group; curved lines connect nuclides for which ratios of release rates are calculated

Now let's consider the issue of how ratios of source rates of different nuclides are calculated. Such ratios are calculated using first guess estimation of source term. Using the notation introduced in Sect. 10.2.2, relationships for ratios (which are generally time dependent) could be rewritten as: $a_{i,m} = x^B_{(m-1)N_{\text{nu}}+i} / x^B_{(m-1)N_{\text{nu}}+J(i)}$. The relationship *J(i)* was introduced in (10.11).

In reality release ratios are time dependent. For instance, in many source term scenarios noble gases are emitted at first, while aerosols are released later. The corresponding time dependence of release ratios could be theoretically evaluated from first guess source term. However in practical situations when exact start time of emissions is not known it is not reasonable to use time dependent prior release ratios assessed from first guess source term since time dependence of true release ratios depends on start time of emission. Therefore at first *time integrated* ratios of the release rates of different nuclides are calculated from the first guess source term:

$$a_i = \sum_{m=1}^{N_s} \left( x^B_{(m-1)N_{\text{nu}}+i} / x^B_{(m-1)N_{\text{nu}}+J(i)} \right). \tag{10.23}$$

For those time steps at which the denominator in (10.23) is zero, the corresponding ratio is set to a large value ($a_{\text{inf}} = 10^5$). However, if in the first-guess source term nuclides are present for which the total released inventory through the whole release period is zero, such nuclides are excluded from the minimization problem and the source term for such nuclides is not analyzed. Elements of the solution vector for such nuclides remain zero. Therefore, the denominator in (10.23) is always greater than zero.

Then, as described above, the matrix $\underline{\underline{G}}$ and the observation vector $\bar{y}$ are augmented with elements corresponding to relationships:

$$x_{(m-1)N_{\text{nu}}+i} - a_i x_{(m-1)N_{\text{nu}}+J(i)} = 0. \tag{10.24}$$

Of course, the solution of augmented minimization problem (10.8) satisfies relationships (10.24) only approximately and corresponding error variances

$$\tilde{\sigma}^2_{R,i} = \mathrm{E}\left( \left( x_{(m-1)N_{\text{nu}}+i} - a_i x_{(m-1)N_{\text{nu}}+J(i)} \right)^2 \right), \tag{10.25}$$

are to be estimated and used in the augmented matrix $\underline{\underline{\tilde{R}}}$ which was introduced above. In the minimization process, $\tilde{\sigma}_R$ are used as measures of allowable deviations of the r.h.s. of Eq. (10.24) from zero.

Let's consider the issue of $\tilde{\sigma}_R$ estimation. Consider for simplicity the case of two nuclides with corresponding estimated (analysed) release rates $q_1$, $q_2$ considered as random variables and ratio of release rates $a$: $q_2 - a \cdot q_1 = 0$. Let the random variable $a = a_0 + \delta a$ have the expected value $E(a) = a_0$ and variance $E\left((a-a_0)^2\right) = \sigma_a$. Then consider the random variable $\xi$: $\xi = q_2 - a_0 q_1 = q_2 - (a - \delta a) q_1 = \delta a \cdot q_1$.

It is clear that the expected value of $\xi$: $E(\xi) = 0$, while variance of $\xi$: $E(\xi^2) = \sigma_a \cdot \sigma_{q_1}$. The well-known theorem about the expected value and variance of the product of uncorrelated random variables is used in the above considerations. Of course, the variance of the analysed source rate $\sigma_{q_1}$ is not known, but it can be approximated with the corresponding variance of first guess estimation: $\sigma_{q_1} \approx \sigma_{B,1}$. The above considerations are easily extended yielding the relationship:

$$\tilde{\sigma}_{R,i} = \sigma_{a,i} \cdot \sigma_{B,J(i)}, \tag{10.26}$$

where $\sigma_{B,J(i)}$ are estimated using (10.19).

The values of $\sigma_{a,i}$ could be pre-calculated in advance on the basis of several source terms generated for a given accident scenario using process-based tools for source term estimation such as MELCOR (https://melcor.sandia.gov) and/or using expert estimations. However, if such pre-calculated values of $\sigma_{a,i}$ are not available, JRODOS is to be able to provide automatic assessment of those parameters. For this purpose, expert estimations for the bounds of possible ratios of inventories released for different nuclides for the case of the Fukushima accident presented by Saunier et al. (2013) as:

$$0.6 \le \frac{x_{132\mathrm{Te}}(t)}{x_{134\mathrm{Cs}}(t)} \le 16; \quad 2 \le \frac{x_{131\mathrm{I}}(t)}{x_{134\mathrm{Cs}}(t)} \le 100; \quad 0.1 \le \frac{x_{133\mathrm{Xe}}(t)}{x_{134\mathrm{Cs}}(t)} \le 10^4, \tag{10.27}$$

were compared to bounds for release ratios obtained on the basis of literature review by Kovalets et al. (2014):

$$5 \le \frac{x_{132\mathrm{Te}}(t)}{x_{134\mathrm{Cs}}(t)} \le 150; \quad 6 \le \frac{x_{131\mathrm{I}}(t)}{x_{134\mathrm{Cs}}(t)} \le 130; \quad 0 \le \frac{x_{133\mathrm{Xe}}(t)}{x_{134\mathrm{Cs}}(t)} \le 5 \times 10^4. \tag{10.28}$$

From the above data it could be concluded that in all cases the ratio between upper $(a_U)$ and bottom $(a_B)$ bounds of release ratios: $\omega = a_U/a_B$ varies from $\omega = 20$ to $\omega = 50$ between non-noble gas nuclides and $\omega \ge 10^5$ for the ratios of release rates of noble gas/to non-noble gas nuclide.

Let us assume that for the case of non-noble gas nuclides $a$ is Gaussian-distributed with the expected value $a_0$ and hence $a_U \approx a_0 + 2\sigma_a$, $a_B \approx a_0 - 2\sigma_a$ (known property of Gaussian distribution is that with 95% probability the value of a is found in the interval $(a_0 - 2\sigma_a, a_0 + 2\sigma_a)$). Let us assume $\sigma_a = \alpha_a \cdot a_0$ and substitute this into the above definition of $\omega$: $\omega = a_U/a_B = (a_0 + 2\alpha_a \cdot a_0)/(a_0 - 2\alpha_a \cdot a_0)$. Then the value of $\alpha_a = 0.5$ fits well the above range of $\omega = 20{-}50$.

In case when $a$ is the ratio of release rates of noble gas to non-noble gas nuclides, it spans a too large range of values and it is most likely that its probability distribution is skewed (non-Gaussian). Analogous situation was considered above for the case of error variance of first guess estimation and in the same way the value of $\alpha_a = 5$ could be reasonably used to define $\sigma_a = \alpha_a \cdot a_0$ in such a case.

Since first guess source terms are usually time dependent, instead of using the above parameterizations for $\sigma_a$, it can be defined alternatively from the first guess source term:

$$\sigma_{a,i}^{(1)} = \sqrt{\frac{1}{N_s}\sum_{m=1}^{N_s}\left(\left(x^B_{(m-1)N_{nu}+i}/x^B_{(m-1)N_{nu}+J(i)}\right) - a_i\right)^2}, \tag{10.29}$$

where $a_i$ was defined by (10.23).

Both methods are combined for calculating $\sigma_a$ by choosing the maximum out of the two values obtained using each method:

$$\begin{aligned} \sigma_{a,i} &= \max\left(\sigma_{a,i}^{(1)}, \sigma_{a,i}^{(2)}\right) \\ \sigma_{a,i}^{(2)} &= \alpha_a \cdot a_i, \end{aligned} \tag{10.30}$$

where in (10.30) $\alpha_a = 0.5$ is the default value used for ratios between release rates of non-noble gas nuclides (aerosols, iodine) or between noble gas nuclides (e.g. ratios of release rates of Xe-133 to Kr-85), while $\alpha_a = 5$ is the default value used for ratios of release rates of non-noble gas nuclide to noble-gas nuclide (e.g. Cs-137 to Xe-133). Note that according to Fig. 10.1 the ratio of the last type appears only once out of total number of $N_{nu} - 1$ ratios which are used if release consists of $N_{nu}$ nuclides.

### *10.2.5 Release Height Estimation*

A possibility to establish release height which was used in many works is to consider a vertically distributed release e.g. Talerko (2005), Stohl et al. (2012). In this case, the control vector $\bar{x}$ again consists only of source rates of different puffs. But contrary to the point release considered above, in the case of a vertically distributed release, puffs are released at different heights. Then the minimization problem is again linear regression and the source-receptor matrix could be established during a single forward run of the model. The release height could be defined as the centre mass of the plume:

$$z_R = (1/H_{max}) \int_0^{H_{max}} q(z)dz, \tag{10.31}$$

where $q(z)$ is the source rate vertically distributed from the ground level to maximum height $H_{max}$.

Further simplification of the source inversion problem with respect to release height estimation is achieved by subdividing the vertical interval of possible release height into subintervals such that in each subinterval the release is distributed

uniformly. This approach has been utilized by Stohl et al. (2012) in which the release following the Fukushima accident has been distributed over three layers: the lower 50 m, between 50 and 300 m and between 300 and 1000 m. In Saunier et al. (2013), also for the Fukushima accident, the release rate has been uniformly distributed over the lowest 160 m.

The calculation of the source-receptor function corresponding to a vertically distributed release has been implemented in the standalone DIPCOT code and the solution of the corresponding minimization problem is presented in the next section. However, in operational practice of the real-time system this approach may be too expensive in terms of computational requirements, since it requires dealing with much more particles in a single run as compared to one with a source at single fixed height.

Therefore, in the JRODOS system, a simplified approach of a release at a fixed height is implemented; the user can do separate inversions for each possible release height, and compare the associated cost function value and/or other obtained statistics of model-measurements comparisons, such as *NMSE*. The result with the lowest value of the selected criteria (e.g. cost function value) would correspond to the best height among the assumptions.

### *10.2.6 Minimization Method*

Minimization of the cost function (10.8) could be performed by using a direct method that solves the system of linear equations resulting from equating the derivative of the cost function $J$ with respect to $\bar{x}$ to zero e.g. Tarantola (2005). This method was applied at first stages of model testing (Sect. 10.4.2), however, its drawback is that it can potentially yield in unphysical negative values.

Therefore another method was chosen to be used by default: nonnegative least squares (Lawson and Hanson 1974). Implementation of this method (NNLS subroutine) was extracted from freeware Netlib library (http://www.netlib.org). Before applying the linear regression solver, the cost function (10.8) to be minimized was brought to the standard form which is used by linear regression solvers:

$$J = J_1 + J_2 = \left\| \underline{\underline{\hat{G}}} \bar{x} - \hat{y} \right\|_2. \tag{10.32}$$

This is possible since all covariance matrices entering the cost function (10.8) are assumed to be diagonal and therefore matrix $\underline{\underline{\hat{G}}}$ and vector $\hat{y}$ in (10.32) can be calculated using the previous definitions as:

$$\underline{\underline{\hat{G}}} = \begin{pmatrix} \underline{\underline{\tilde{R}}}^{-1/2} \cdot \underline{\underline{\tilde{G}}} \\ \underline{\underline{B}}^{-1/2} \end{pmatrix}, \quad \hat{y} = \begin{pmatrix} \bar{y} \\ \underline{\underline{B}}^{-1/2} \cdot \bar{x}^B \end{pmatrix}. \tag{10.33}$$

It was checked that the results of the NNLS method were identical to the results of the direct method when the last didn't yield negative results.

## 10.3 Integration of the Source Inversion Module in the JRODOS System

### *10.3.1 Implementation of the Source Inversion Module (SIM)*

#### 10.3.1.1 SIM Structure

The source inversion module was initially developed and implemented in Fortran90 within the standalone version of Lagrangian dispersion model DIPCOT. After this initial implementation adaptation of the developed code for the work within the JRODOS system was performed as described above and specific details of this integration are described in this section. First the important details of SIM implementation are described.

The implemented source inversion module could be logically subdivided into four main submodules:

- Steering submodule which initializes input data, allocates memory for necessary arrays. In the standalone version data are initialized by reading files. In the integrated version, this submodule communicates with the JRODOS system and receives input data from databases. Then it consequently calls other submodules and gets output data from them;
- Submodule of SRM calculation based on a modification of the DIPCOT atmospheric dispersion model (Andronopoulos et al. 2009, 2010);
- The task of the data processing submodule is to pre-calculate all parameters of minimization problem, described in Sect. 10.2.4, such as error variances, ratios of release rates of different nuclides, initialize corresponding matrices and vectors;
- The minimization submodule which prepares matrices and vectors in a form suitable for further usage in minimization procedure. In particular, the 'design' matrix (which is SRM complemented to include first guess estimation of source term) is prepared and passed to the linear regression solver.

The key variables used in each submodule are stored in Fortran modules of the implemented SIM code.

### 10.3.1.2 Workflow of Main Computational Blocks

The preparation of the source term estimation consists of two main stages: (1) computation of the source-receptor matrix and (2) preparation of the regularization parameters and minimization. The flowchart of the source-receptor matrix calculation is presented in Fig. 10.2.

The workflow is very similar to the forward model run with the following two main differences: (a) gradients of deposition with respect to puff's inventories are

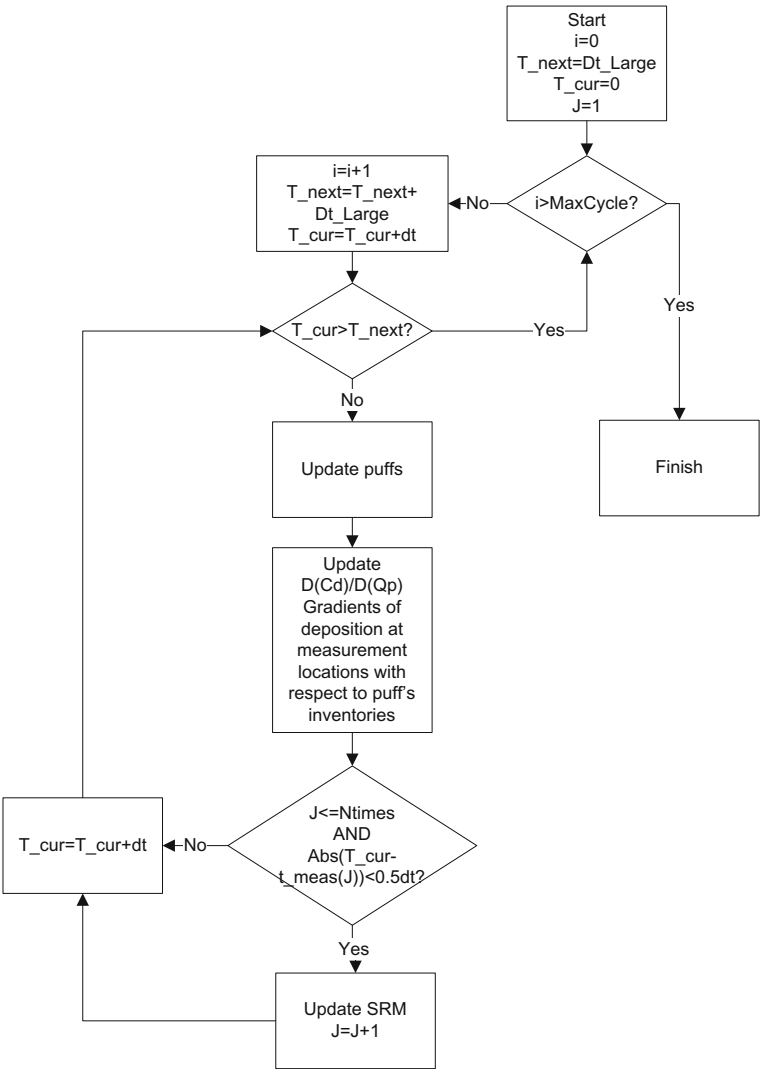



**Fig. 10.2** Flowchart of SRM calculation

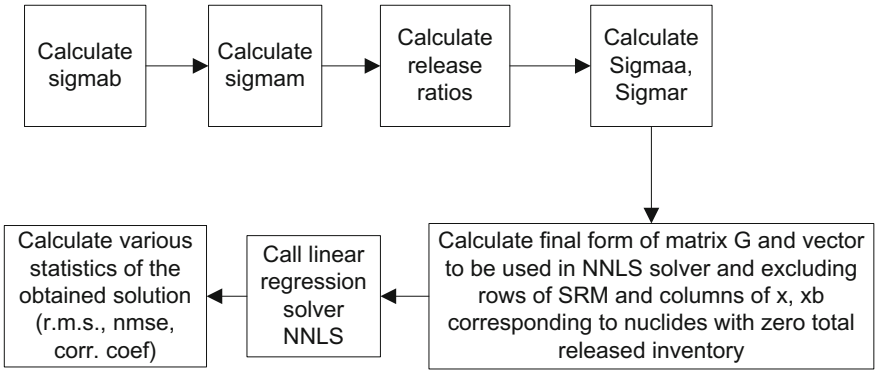



**Fig. 10.3** Flowchart of data preprocessing and minimization procedure

updated at every time step by updating the sum according to (10.16); (b) when model time fits next measurement time to be processed, rows of the source receptor matrix corresponding to measurements at a given time are calculated. As it is clear from this description measurements should be sorted by time.

After the SRM has been calculated and before the minimization procedure is called, various regularization parameters entering the minimization problem (10.8) are calculated, including ratios of nuclide rates and their variances, etc. (Fig. 10.3). Then according to (10.33), matrices and vectors are brought into the form suitable for usage in the linear regression solver, and at this stage those rows of SRM and of solution and first guess vectors which correspond to nuclides with zero total released inventory in the first guess estimation are excluded. The linear regression solver (cf. Sect. 10.2.6) is called afterwards and when the solution has been obtained, various statistical measures related to the difference of simulated results against measurements are calculated, including the final value of the cost function, r.m.s. deviation, and NMSE (Fig. 10.3).

## *10.3.2 SIM Integration in JRODOS*

### 10.3.2.1 SIM Operation Within the JRODOS

The flowchart of full SIM operation within the JRODOS is shown in Fig. 10.4. The workflow of SIM calculation was described above while other steps of SIM operation within the JRODOS include initialization with user-defined parameters and data from system databases and saving output results in project database. Most of the computational time during a full cycle of source term estimation is spent on SRM calculation; therefore it is reasonable to give users the possibility after completion of the first SIM run to repeat the minimization part with changed values of regularization parameters when other parameters of the problem were not

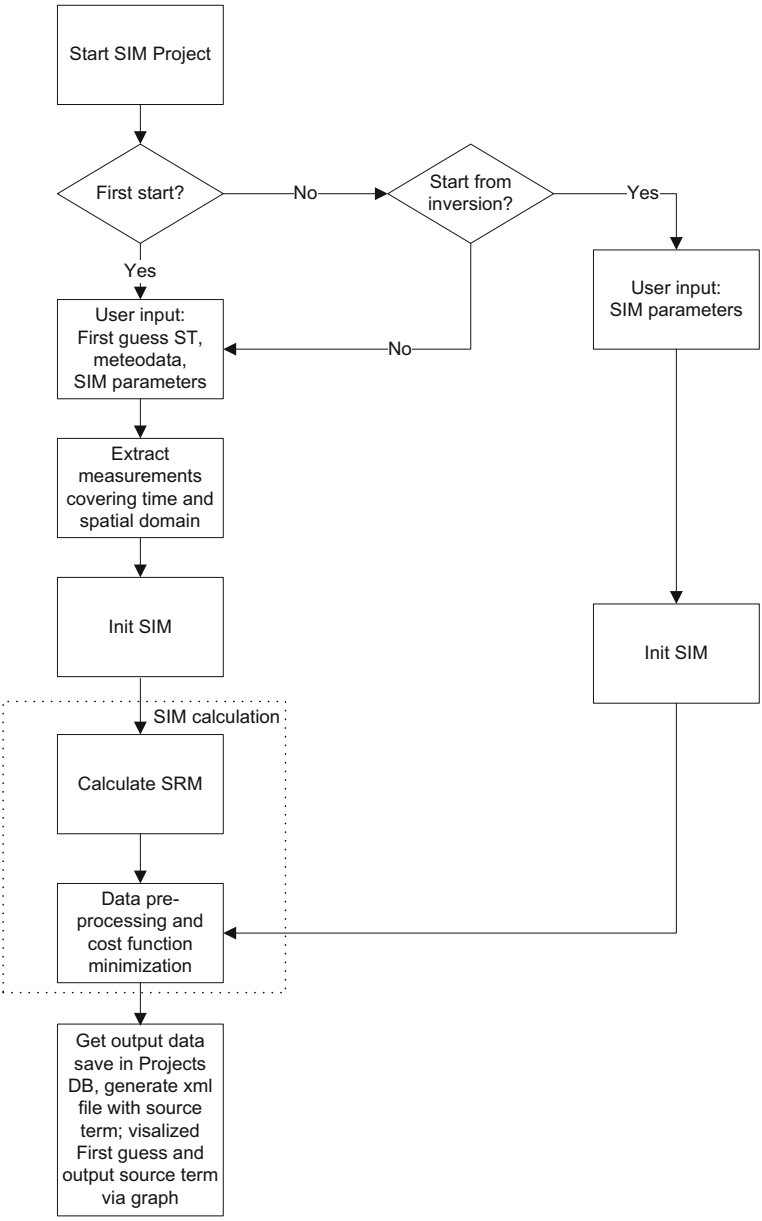



**Fig. 10.4** Flowchart of SIM operation within the JRODOS system

changed (i.e. no new measurements, no change in start and end times of first guess source term, same meteorology, etc.). This part of workflow is also shown in Fig. 10.4.

The input data include:

- Steering parameter (start_from_inversion)
- Set of variables describing measurement data, with the measurement values sorted by increasing time of measurements
- Set of variables describing first guess source term, including start and end time of release, released inventories per time step
- Time resolution of analyzed source term
- End time of simulation
- Set of regularization parameters
- Other parameters: the same as for normal LSMC run: meteorology, etc.

Output data include:

- Analyzed source term, presented also in form suitable for usage in normal LSMC run (xml file)
- Statistical indicators of quality of solution in tabular form
- SRM and corresponding measurement data and first guess, available for export to binary files for the users who may wish to try their own minimization algorithms.

#### 10.3.2.2 User Interfaces

To run the source inversion module, the user has to start the project of the corresponding type (Fig. 10.5, Project tree). Entering of basic data necessary to run SIM is very similar to input data of the JRODOS Local-Scale Model Chain (LSMC), therefore the existing user interface of LSMC called RoLite and described in Ievdin et al. (2015) was modified accordingly (Fig. 10.6).

As in the case of LSMC, the first guess source term could be input in different forms: time dependent release (in Bq) for different nuclides, or release for groups of nuclides (Bq), or released fractions of reactor inventory. Existing JRODOS software components convert the input source term data in whatever form into release rates for different nuclides, and this is used as first guess source term in SIM. Therefore there were no changes in UI input window for setting the source term (except that the corresponding tab in RoLite was renamed to 'First guess source term').

Meteorological data for running SIM can be input by the user or obtained from NWP models. The parameter timestep in Fig. 10.6 is used as time resolution of source term analysis when running SIM. The duration of the analysis is to be selected by the user, and only measurements falling into the selected time interval will be extracted for usage in source inversion procedure. Note that only measurements of those providers which were registered for a given site using standard tools existing in JRODOS will be used in the source inversion procedure.

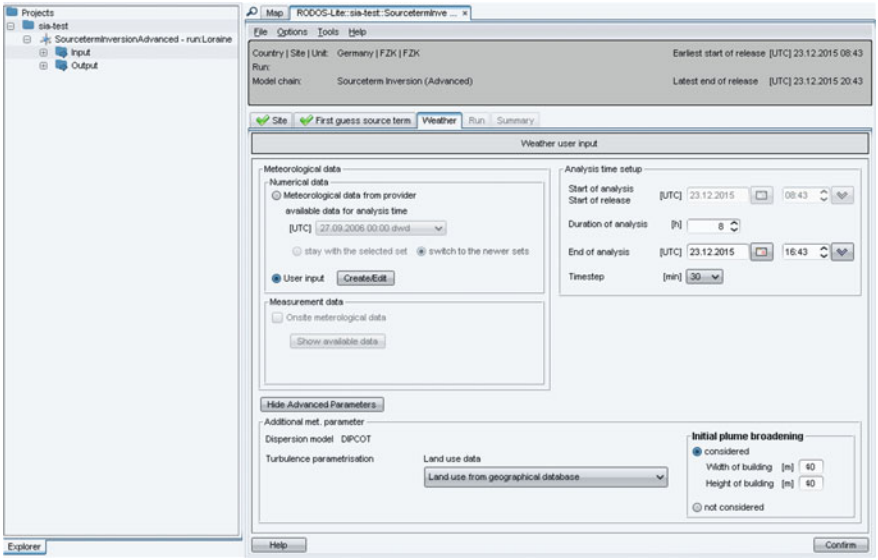
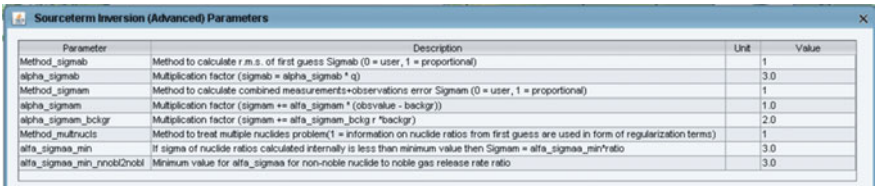



**Fig. 10.5** Setting SIM calculation through the modified RoLite interface of the JRODOS system




| Parameter | Description | Unit | Value |
|---|---|---|---|
| Method_sigmab | Method to calculate r.m.s. of first guess Sigmab (0 = user, 1 = proportional) | | 1 |
| alpha_sigmab | Multiplication factor (sigmab = alpha_sigmab * q) | | 3.0 |
| Method_sigmam | Method to calculate combined measurements+observations error Sigmam (0 = user, 1 = proportional) | | 1 |
| alpha_sigmam | Multiplication factor (sigmam += alfa_sigmam * (obsvalue - backgr)) | | 1.0 |
| alpha_sigmam_bckgr | Multiplication factor (sigmam += alfa_sigmam_bckg r *backgr) | | 2.0 |
| Method_multnucls | Method to treat multiple nuclides problem(1 = information on nuclide ratios from first guess are used in form of regularization terms) | | 1 |
| alfa_sigmaa_min | If sigma of nuclide ratios calculated internally is less than minimum value then Sigmam = alfa_sigmaa_min*ratio | | 3.0 |
| alfa_sigmaa_min_nnobl2nobl | Minimum value for alfa_sigmaa for non-noble nuclide to noble gas release rate ratio | | 3.0 |

**Fig. 10.6** Selection of SIM parameters for calculation of regularization parameters through menu 'Model parameters' of the JRODOS system

Figure 10.6 shows menu 'Model parameters' for parameters $\alpha_B, \alpha_m, \alpha_r, \alpha_a$ which are described in Sect. 10.2.4 above. The parameters Method_* also shown in model parameters window are set to 1 and presently cannot be changed. This means that error variances described above are calculated with the entered values for $\alpha_B, \alpha_m, \alpha_r, \alpha_a$ using relationships described in Sect. 10.2.4. Other options (methods) for calculation of error variances could be implemented in the future.

Figures 10.7 and 10.8 show output results of the source inversion module indicated in Project Tree. The output results include the source term prepared in the xml file which could be then imported to standard a LSMC Project (Fig. 10.7). Statistical indicators of quality of the achieved solution are presented in the form of a table (Fig. 10.8) and include the norm of residual $\|\bar{r}\|_2 = \|Gx - y\|_2$ before and after minimization and the final achieved value of the cost function. The norm of the measurement vector $\bar{y}$ is also displayed since the relative value of $\|\bar{r}\|_2 / \|\bar{y}\|_2$ may be of interest.

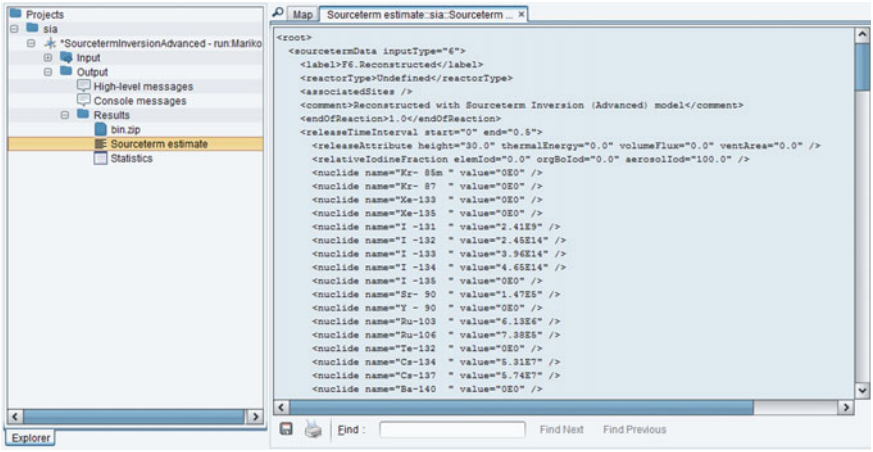



**Fig. 10.7** Output results of SIM in Project Tree (left) and estimated source term in xml format ready for export to LSMC (right)

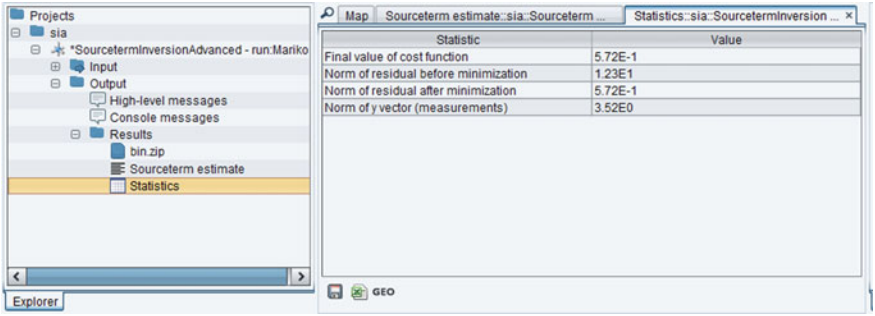



**Fig. 10.8** Statistical indicators of quality of solution of source inversion problem

## 10.4 Results of Calculations

### *10.4.1 Description of the Test Cases*

The developed source inversion method was applied for the meteorological and geographical conditions of the European Tracer Experiment ETEX (Gryning et al. 1998). The computational domain, topography etc. is shown in Fig. 10.9. Artificially generated measurements were used for testing the source inversion method, i.e. values of GDR calculated by DIPCOT using the true source term in the points of locations of sensors (Fig. 10.9) with 10 min time resolution.

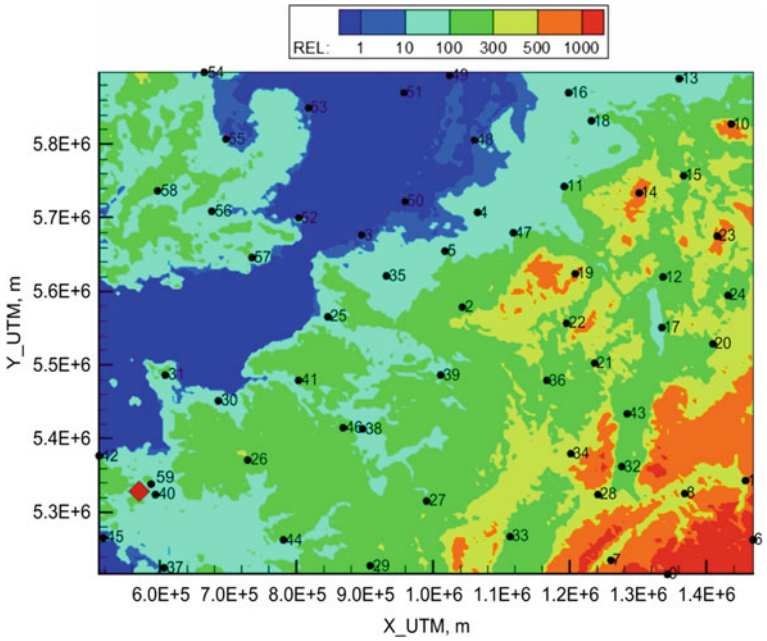



**Fig. 10.9** Topography and locations of sensors and source in the computational domain

Two source terms were used for testing:

1. Source term 1. Simplified source term with constant release rate during 12 h of three radionuclides with total released activities 1.0E+18 Bq of Xe-133, 1.0E +17 Bq of I-131 and 5.0E+17 Bq of Cs-137, source height is 8 m.
2. Source term 2. Hypothetical release of 21 nuclides (Rb-88, Sr-89, Sr-90, Sr-91, Ru-103, Ru-105, Ru-106, Te-131 m, Te-132, I-131, I-132, I-133, I-134, I-135, Cs-134, Cs-136, Cs-137, Kr-87, Kr-88, Xe-133, Xe-135). There were two phases in the release period, having durations of 0.5 and 4 h (source term Muehleberg-1 from flexRISK study, http://flexrisk.boku.ac.at/). Release rates varied from 0 to 1E+12 Bq/s. During the first phase of release 100% of the inventory of noble gases and 90% of aerosols and iodine was emitted. During next phase of release emission rate was constant (Fig. 10.10). It was a vertically distributed release, described by 10 vertical levels within the layer 0–100 m with a Gaussian profile of release rate (height of maximum release rate at 50 m).

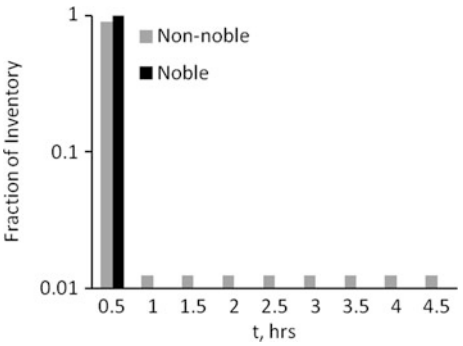



**Fig. 10.10** Emitted fraction of inventory at different phases of release: noble gases and aerosols + iodine (denoted as 'non-noble')

### *10.4.2 Testing of Source-Receptor Matrix*

Figure 10.11 demonstrates the correctness of the implementation of the source receptor matrix. Results calculated in forward run excellently agree with the results obtained by multiplication of the SRM with the vector of true source rates. Note that as it is evident from the Fig. 10.11, the influence of deposition on GDR is correctly accounted for in SRM calculation: GDR does not diminish to zero after plume passage but remains nearly constant due to radiation created by deposition of long-lived racionuclides.

Table 10.1 presents a quantitative comparison of measurements generated in forward run and of respective values calculated using the SRM. More specifically, the norm of residuals $\|Gx - y\|_2$ and the norm of the 'measurement' vector $\|y\|_2$ calculated using SRM and 'measurements' computed in the same model run (Run 0) and with 'measurements' computed in a different run (Run 1) are presented. Excellent results for Run 0 are evident in case of both source terms, while results of SRM as compared to results of independent forward run are not so good but still acceptable. This difference is caused by the stochastic nature of the Lagrangian

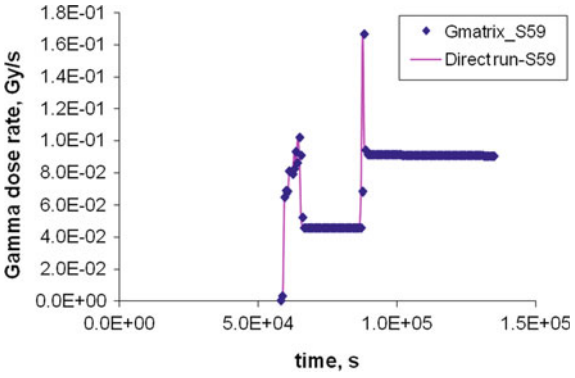



**Fig. 10.11** Time series of GDR at sensor No. 59 (most close to point of release) calculated with forward model (line) and reconstructed using the source-receptor matrix (signs) for the case of Source term 1

**Table 10.1** Norm of residual $\|Gx - y\|_2$ and norm of 'measurement' vector $\|y\|_2$ calculated using SRM and 'measurements' computed in the same model run (Run 0) and with 'measurements' computed in different run (Run 1)

| Source term | Run | Norm of residual (normr) | Norm of $y$ (normy) | Normr/normy |
|---|---|---|---|---|
| 1 | 0 | 2.8E−06 | 0.36 | 7.8E−06 |
| 1 | 1 | 0.012 | 0.36 | 3.3E−02 |
| 2 | 0 | 5.1E−07 | 3.9E−02 | 1.3E−05 |
| 2 | 1 | 2.0E−03 | 3.9E−02 | 5.2E−02 |

model and thus even after application of the regularization procedure (control vector reduction) described in Sect. 10.2.3, the SRM is not identical to independently calculated results of the forward model.

## *10.4.3 Results of Source Inversion with Adjusted Regularization Parameters*

In this section the results obtained with predefined and/or adjusted values of error variances are presented.

### 10.4.3.1 Source Term 1

In the first experiment, it is assumed as prior knowledge that the release is constant both in time and vertically. The SRM and the vector of samples $y$ were calculated in different runs of DIPCOT. Because DIPCOT is stochastic model, the results obtained in different runs are somewhat different. The release can be fully described by three numbers (dim $\boldsymbol{x} = 3$), representing releases of the three nuclides, and the augmented cost function is identical to that in Sect. 10.2.2. The matrix $\underline{\underline{G}}_1 \in R^{N_o \times 3}$ was calculated from $\underline{\underline{G}} \in R^{N_o \times 12}$ by aggregating corresponding columns.

The comparison of the true and estimated emissions using different methods is presented in Table 10.2:

**Table 10.2** True and estimated release rates

| | True release, Bq/s | Estimated release rates (Bq/s) | | |
|---|---|---|---|---|
| | | A1 | A2 | A3 |
| Xe-133 | 2.32E+13 | −5.24E+11 | 1.80E+13 | 2.28E+13 |
| I-131 | 2.32E+12 | 8.53E+12 | 4.44E+12 | 2.28E+12 |
| Cs-137 | 1.16E+13 | 6.67E+12 | 1.03E+13 | 1.14E+13 |

- (A1) pure linear regression, no weighting, no regularization ($J_2 = 0$ in Eq. (10.6); at this stage direct solver of linear regression problem was used which could yield negative values
- (A2) minimization of cost function (10.6) without nuclide ratios, and
- (A3) minimization of (10.8). Release ratios used in A3 are the precise nuclide ratios of the true source term.

In the second experiment, the assumption of the homogeneity of the release is dropped and released activities in all 12 time slots are estimated. This means that we estimate release rates in 12 independent phases for three nuclides—dim **x** = 36 (it might be interesting to examine influences of prescribing some continuity between the ratios in neighbouring phases). Analogously to case A1 presented above, in case of solving linear regression without any weighting or regularization the result is quite poor. When the same as in case A2 presented above the inversion method is applied based on the cost function (10.6) without any conditions on nuclide ratios, a more reasonable result is obtained. But estimated emissions again do not resemble the true release as shown in the upper Fig. 10.12.

For taking into account nuclide ratios the matrices $\bar{y}, \underline{\underline{G}}$ must be augmented by 24 rows and $\underline{\underline{R}}$—with 24 rows and columns (12 for I-131 ratios and 12 for Cs-137 ratios). In this setup, the influence of variances of nuclide ratios on the performance of the estimations is examined. If the first guess ratios for I-131 and Cs-137 are distorted by factors 100 and 0.01, respectively, as compared to the true ratios, and if higher augmented variances are used ($\sigma_R = 1E15$), the result is shown in bottom Fig. 10.12. Release rates of Xe-133 and Cs-137 were estimated more or less precisely, rates of I-131 fluctuate around the true value.

#### 10.4.3.2 Source Term 2

A time interval of 0.5 h was selected for calculations of SRM. Vertically, the release is distributed over 10 point sources uniformly distributed from 0 to 100 m. In the vertical, the profile is not homogeneous, emitted activity has a Gaussian distribution with the mode at 50 m. SRM is computed in a way that the actual release is preceded and followed by a 2-h period with zero emissions. This practically means that the release is splitted on 4 phases with a total number of 17 time intervals. The dimension of the source vector is 21 nuclides × 10 levels × 17 time intervals = 3570. There are 12,744 10-min GDR measurements from 59 receptors. As before, there are two runs of DIPCOT, one for $\underline{\underline{G}}_1 \in R^{12,744 \times 3570}$ and one (independent run)—for $\bar{y}$. As it was shown in Table 10.1, the results between different runs of the stochastic model (mentioned as Run 0 and Run 1 in Table 10.1) are not much different, but it is not an identical twin experiment.

Since there are zero emissions of noble gases in the second phase, a minimum emission $x_{\min} = 1.0\text{E} + 3\,\text{Bq/s}$ is defined in order to prevent numerical problems.

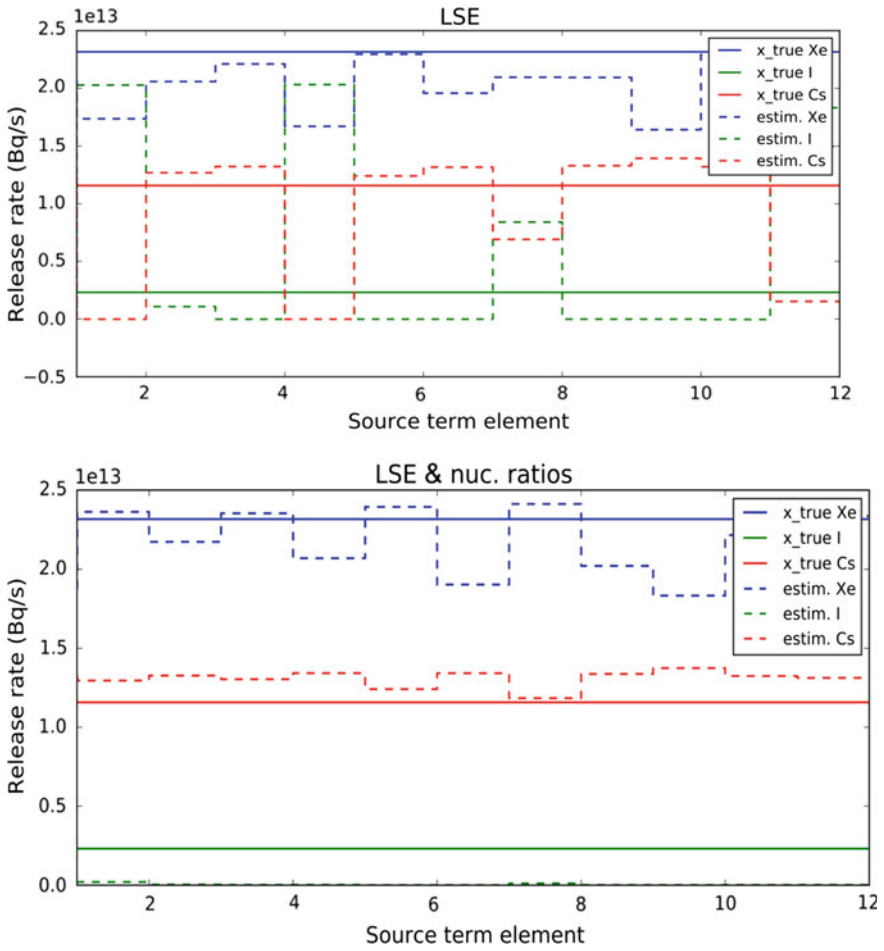



**Fig. 10.12** Results of solving the source inversion problem in case of source term No. 1, (upper)—without additional conditions for nuclide ratios; (bottom)—with using conditions for nuclide ratios as described in text

This minimum value is used for calculation of ratios etc. In all cases, the observation error is set to $\bar{\sigma}_{\mathrm{m}} = 0.1 \cdot \bar{y} + 10^{-6}$.

At this stage each time slot is not treated separately, but columns of SRM are aggregated in a way that emissions of each nuclide at each vertical source for the whole phase are estimated. Thus, the dimension of the source vector is 21 nuclides $\times$ 4 phases $\times$ 10 levels = 840. As first guess, all values are assumed to be zero $x_B = 0$. Its standard deviation is set to be 1.0E12 for the releases in the second phase (the first release phase) and 1.0E9 otherwise.

At first, different values of the uncertainty $\tilde{\sigma}_R$ are specified in the augmentations of $\underline{\underline{R}}$ for each phase. The following results shown in Figs. 10.13 and 10.14 were

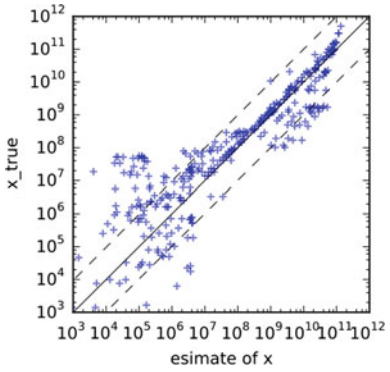



**Fig. 10.13** Estimated versus true release rates (Bq/s) obtained with assumption of four phases and variable variances of nuclide ratios for each phase (see text)

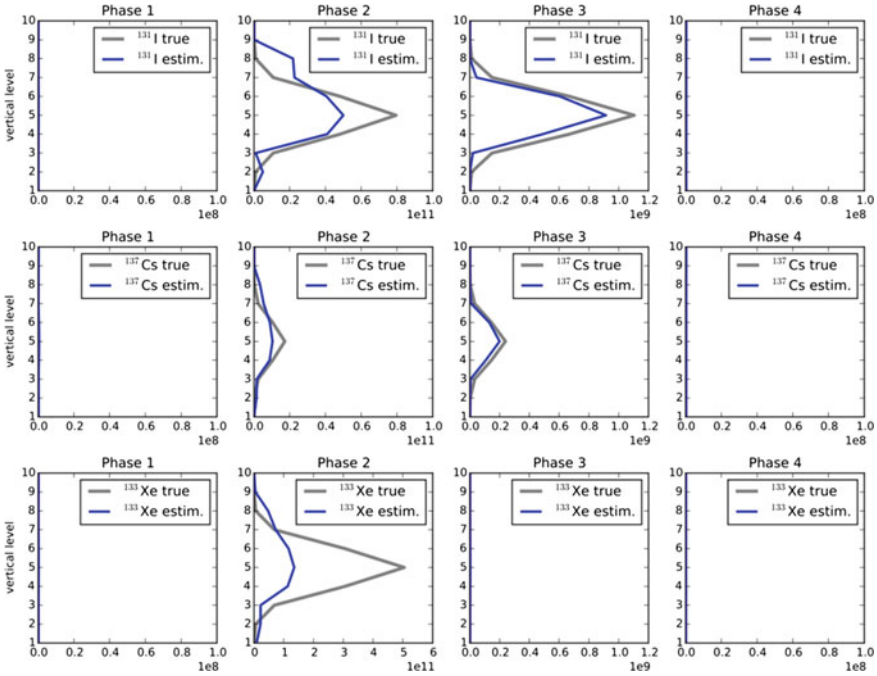



**Fig. 10.14** Estimated and true profiles of release rate (Bq/s) at different vertical levels for selected nuclides with assumption of four phases and variable variances of nuclide ratios for each phase (see text)

obtained with variances of nuclide ratios equal to 1.0E1, 1.0E10, 1.0E8, 1.0E1, respectively, for the different phases. The low values for the first and last phases mean that nuclide ratios in these 'no-release' phases are imposed strictly. The results obtained without any additional information about timing of the release will be presented in the next section.

The higher emissions were estimated correctly. Underestimation of lower emissions is caused by big differences in emission of respective vertical sources even in one phase. Such big differences cannot be parameterized by a single number neither in first guess error nor in nuclide ratio variance. However, the results obtained are still acceptable and resolve the most important features of the release (timing and magnitude of higher emissions).

### *10.4.4 Results with Automatically Calculated Error Variances*

In the previous section, results of testing the source inversion algorithm with arbitrarily adjusted values of the regularization parameters were presented. In operational practice of SIM usage within the JRODOS, regularization parameters are to be pre-calculated using the approach presented in Sect. 10.2.4. Therefore, here results of testing with automatically calculated values of regularization parameters are presented.

The same source term 2 as described in Sect. 10.4.1 was used but a point release was assumed with the release height equal to 50 m. In the tests presented, two different first guess source functions were used. Both source functions were created by shifting true source function backward in time by 2 h and then: increasing (Case A) and decreasing (Case B) true source function values by the factor of 10. Thus, the first guess release started immediately with the beginning of the simulations while true release started 2 h later. The source term was estimated with the time resolution equal to 0.5 h. The first guess source term was provided with the same time resolution.

The results of source inversion are presented in Fig. 10.15. Results are shown for selected nuclides Kr-87, Cs-137 and I-131, representative of each of the three groups (noble gases, iodine and aerosols). As it is seen from the results presented in those figures, the time of maximum emission is accurately evaluated by the algorithm. If the first guess source function is too low (Case B), the reconstructed source term is strongly underestimated for noble gas Kr-87. This is a consequence of lower values of root mean squared errors of the first guess release rates which according to (10.19) are proportional to the prior estimations of the release rates. In contrast the Case B yields overestimation of release rate of Kr-87. Emission rates of Cs-137 obtained in both cases A and B are almost identical and underestimated by the factor of about 2. Emission rate of I-131 is excellently identified in Case A while in Case B it is underestimated by the factor of 3.

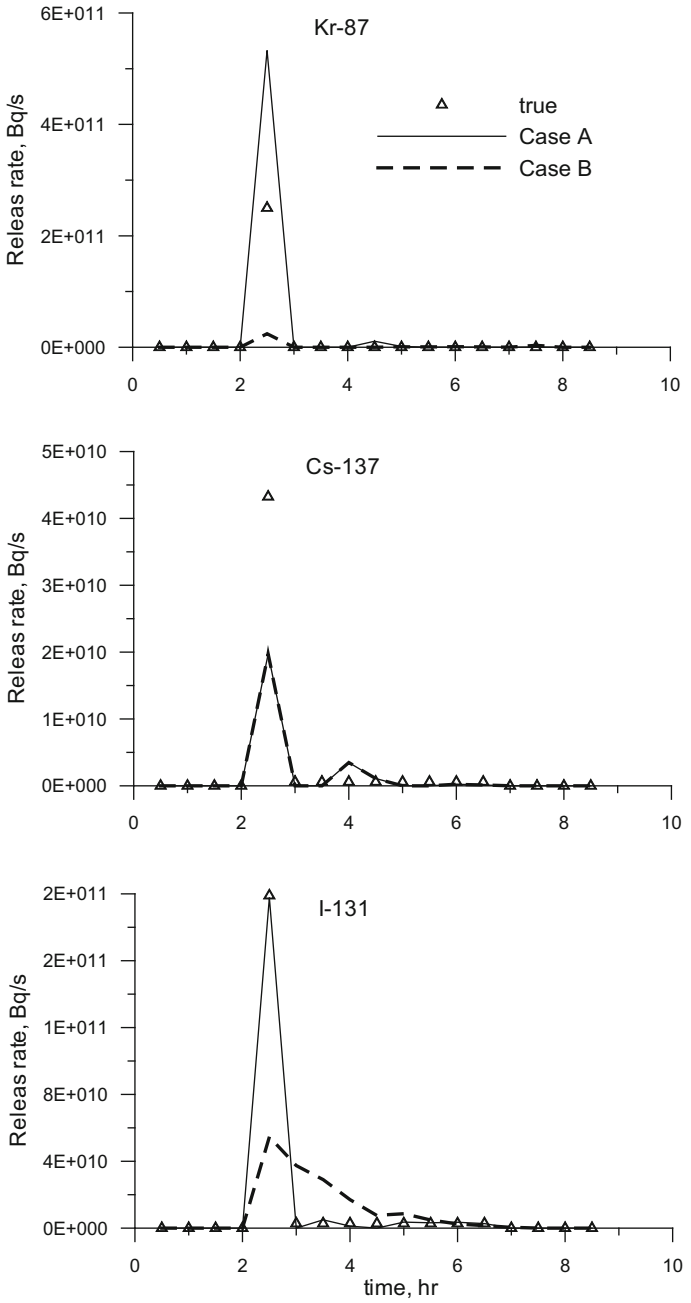



**Fig. 10.15** True (triangles) and calculated release rates for Kr-87, Cs-137, and I-131 in the cases A (solid line) and B (dashed line) as described in text

**Table 10.3** Mean absolute error and mean relative bias of the first guess and estimated emission rates

| Case | MAE | MRB |
|---|---|---|
| First guess-A | 10.8 | 9.0 |
| First guess-B | 1.1 | −0.9 |
| Source inversion-A | 0.49 | −0.24 |
| Source inversion-B | 0.96 | −0.76 |

Table 10.3 presents statistical indicators of the accuracy of the evaluated source rates calculated over all 21 nuclides and over all time periods in cases A and B. The mean relative absolute error MAE $= \langle |x_{sol} - x_{true}| \rangle / \langle x_{true} \rangle$ and mean relative bias MRB $= \langle x_{sol} - x_{true} \rangle / \langle x_{true} \rangle$ are presented. According to the definition of MAE when first guess release rate is by the order of magnitude greater than true release rate this leads to large values of MAE (in Case A presented in Table 10.3 MAE of first guess equals 9). When first guess release rate is much less than the true value this leads to the value of MAE asymptotically reaching MAE = 1 when first guess release rate reaches zero. This explains why for the Case B MAE of first guess equals 1.1.

In Case A errors of the first guess source rate are reduced very significantly in result of source inversion procedure. It is interesting to note that errors in estimated source term (MAE = 0.49, MRB = −0.24) are similar to errors obtained by Tsiouri et al. (2012) for the case of source inversion of release rate using GDR measurements in case of a single nuclide. In results of Tsiouri et al. (2012) MAE varied from 0.39 to 0.71 and MRB from −0.27 to −0.9. Since present case of release of multiple nuclides is much more complex than the case of a single nuclide it could be concluded that the level of improvement achieved in Case A is very good.

In Case B the level of improvement is essentially worse as compared to Case A and it is marginal as compared to initial errors of first guess source term. This is due to mentioned above effect of reduced variances of first guess source term estimation which according to Eq. (10.19) are proportional to first guess source rate itself. In result the solution is too strongly forced to first guess estimation. Therefore in setting first guess source term for usage in JRODOS SIM it is advised to use conservative estimates.

Table 10.4 presents results of source height estimation. The source height in the run with the minimum obtained value of the cost function is considered to be the result of source inversion. The Case A of first guess source term is used. As in Table 10.1, two runs were performed: with using measurements calculated in the same run in which SRM was calculated (Run 0); and with using measurements calculated independently (Run 1). In both cases, the algorithm is able to estimate the source height, but in the case of Run 1 the cost function minimum is not as distinct as in the case of Run 0. If the model error would increase even more, this minimum will be further smoothed and might even disappear. Therefore, further testing of this algorithm will require use of real measurements.

**Table 10.4** Dependence of the resultant cost function value on the release height for the case of using measurements from the same run in which SRM was calculated (Run0) and from an independent model run (Run1)

| Release height, m | $Jmin^{1/2}$ (Run 1) | $Jmin^{1/2}$ (Run 0) |
|---|---|---|
| 10 | 0.0066 | 0.0060 |
| 25 | 0.0061 | 0.0065 |
| **50** | **0.0020** | **0.0004** |
| 75 | 0.0030 | 0.0020 |
| 100 | 0.0033 | 0.0070 |
| 150 | 0.0120 | 0.0120 |
| 200 | 0.0120 | 0.0160 |

Figures in bold denote the minimum value achieved and the corresponding estimated release height. The first guess source term correspond to the Case A

## 10.5 Conclusions

In this work the development of a source inversion algorithm is described which allows to evaluate time-dependent release rates of different radionuclides and source height with the aid of GDR measurements collected at different distances from the source: from $\sim 1$ to $\sim 1000$ km. The automated version of the algorithm was developed which could be used in real-time. The algorithm was integrated in the Source Inversion Module of the European nuclear decision support system JRODOS.

A variational approach for the problem of source inversion was used. The source-receptor matrix is calculated with the aid of the atmospheric transport model DIPCOT.

To estimate release rates of different radionuclides from GDR measurements, in formulation of source inversion problem the SRM and measurement vector are augmented with additional linear relationships describing ratios of release rates of different radionuclides. The possible ranges for the respective ratios are calculated from the first guess source term provided by user.

Parameterizations for the regularization parameters of source inversion problem which include root mean squared errors of measurents, first guess release rates, calculated values etc., are developed.

Testing of the proposed method using the artificial measurements precalculated for the conditions of ETEX experiment had shown that in all cases the error of the evaluated release rates are less than the errors of the respective first guess. The level of improvement was better when first guess release rates were greater than the true values. Therefore in setting first guess source term for usage in JRODOS SIM it is advised to use conservative estimates.

It is also recommended to use the developed SIM together with the meteorological data assimilation tools previously implemented in the JRODOS system

(Kovalets et al. 2004; Davakis et al. 2007; Andronopoulos et al. 2016). Further testing of the algorithm is planned to be performed using real data from field experiments and/or real-case accident scenarios.

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