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Bayesian Estimation of Source Term of Atmospheric Radiation Release with Interval Prior

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Abstract Detection of release of an atmospheric pollutant is a problem of interest in environmental sciences. We are concerned with estimation of unknown source term of the release. Formally, the problem is formulated as a linear model where measurements are explained using source-receptor-sensitivity matrix obtained from atmospheric transport model and source term vector which has to be estimated. Specifically, we estimate the release of radioactivity from measurements of gamma dose rate (GDR). The problem of isolation of activity of individual nuclides is poorly conditioned. We propose a probabilistic model with the prior information on intervals of nuclide ratios. The model parameters are estimated using the Variational Bayes method. The proposed algorithm is tested on simulated scenario with 16 nuclides and compared with state-of-the-art optimization approaches.

Keywords: Source term estimation; Variational Bayes inference; Gamma dose rate measurement; CVX optimization toolbox; Covariance matrix model

1 INTRODUCTION

The task of determination of a source term of an atmospheric pollutant is important in many situations such as radioactive release from nuclear power plants (Davoine and Bocquet [2007]), volcano eruptions (Kristiansen et al. [2010]), or emission of greenhouse gases (Stohl et al. [2009]). The source term is the vector of amounts of the pollutant released in regularly sample time. The location of the release is assumed to be known. Uncertainty in the source term is one of the largest source of errors in modeling and prediction of the pollutant dispersion in the atmosphere, see e.g. Stohl et al. [2012], hence, any improvement of the reliability of the source term estimation has significant impact.

The common approach for determination of the source term is to combine the data measured in the environment (e.g., radionuclide concentrations) with an atmospheric transport model. The quality of the estimated source term to a given measurements can be modeled and optimized using various approaches including the Bayesian approach (Bocquet [2008]), maximum entropy principle (Bocquet [2005]), or cost function optimization (Eckhardt et al. [2008]; Adam and Branda [2016]). Typically, the problem is formulated as a linear regression. The vector of measurements is assumed to be a product of a computed source-receptor-sensitivity (SRS) matrix determined using an atmospheric dispersion model and an unknown source term vector.

Commonly, the vector of measurements contains separated concentrations of assumed radionuclides in the case of radioactive releases. However, in this paper, we assumed the measurement in the form of gamma dose rate (GDR) measurements. This means that our measurement vector contains bulk GDR from a mixture of nuclides and not nuclide-specific concentration activity measurements. The advantage of this approach is easier measurement than measurement of activity concentration and much higher temporal resolution. The disadvantage of this approach is that the problem is ill posed and some further assumption has to be used.

One such approach is to reduce the SRS matrix by removing outlying measurements as in Martinez-Camara et al. [2014] which may be misleading if the removed measurement is informative. Another approach is to use some prior information. Particularly important are prior assumptions on nuclide ratios which can be available, e.g., from physical analysis, reactor inventory, or measurement of taken samples downwind of the release. Since exact nuclide ratios are hardly available, we aim to use intervals of ratios as a prior information in the same spirit as Saunier et al. [2013].

To our knowledge, two approaches are suitable for this kind of constraints: constraint optimization and Bayesian inference with restriction. For constraint optimization, we use CVX toolbox developed by Grant and Boyd [2008, 2014] where constraints on ranges of nuclide ratios can be formulated together with selected form of a regularization. As a regularization, we assume two common regularization terms: Tikhonov regularization (Golub et al. [1999]) and least absolute shrinkage and selection operator (LASSO) regularization (Tibshirani [1996]). For Bayesian inference, we developed generalization of approach by Tichý et al. [2016] where ranges of ratios are formulated as a restriction on covariance matrix of the source term. The aim of this contribution is study the developed Bayesian inference and to compare these approaches on data with simulated release of 16 nuclides measured using GDR measurement with prior knowledge of possible intervals of nuclide ratios.

2 LINEAR INVERSE PROBLEM IN SOURCE TERM ESTIMATION

We deal with following linear inverse model:

$$\mathbf{y} = M\mathbf{x} + \mathbf{e}, \tag{1}$$

where $\mathbf{y}$ is the measured vector with GDR measurement, M is known SRS matrix (Seibert and Frank [2004]) accumulating uncertainties from the atmospheric transportation model and the metheorological data, and $\mathbf{x}$ is the unknown source term vector which has to be estimated. We focus on the situation when ordinary least square solution fails since the matrix M is ill-conditioned and some additional information is necessary for suitable solution.

In this contribution, we aim to use approximate information about ratios of nuclides from which the source term is consisted. Specifically, we assume the knowledge of the intervals in which lay ratios of selected nuclides and a chosen reference nuclide. The similar assumption was used by Saunier et al. [2013]. Assume that the interval with ratio for kth nuclide $\mathbf{x}_k$ is

$$a_k \leq \frac{\mathbf{x}_1}{\mathbf{x}_k} \leq b_k, \tag{2}$$

where $\mathbf{x}_1$ is the reference nuclide.

In following sections, we will formulate two approaches how to incorporate this information.

2.1 Optimization Approach

The problem (1) can be formalized as an optimization problem. As the first try, ordinary least square solution with positivity constraints and constraints (2) can be used:

$$\mathbf{x}^* = \arg\min_{\mathbf{x}\in\mathcal{X}} \left\{ ||\mathbf{y} - M\mathbf{x}||_2^2 \right\}, \text{ subject to } \mathbf{x} \geq 0,\ a_k \leq \frac{\mathbf{x}_1}{\mathbf{x}_k} \leq b_k,\ \forall k, \tag{3}$$

where $||\mathbf{x}||_2 = \mathbf{x}^T\mathbf{x}$ denotes quadratic norm of the vector $\mathbf{x}$. However, ill-conditioned matrix M implies pure quality and reliability of the solution. Thus, regularization term is typically employed as

$$\mathbf{x}^* = \arg\min_{\mathbf{x}\in\mathcal{X}} \left\{ ||\mathbf{y} - M\mathbf{x}||_2^2 + \alpha g(\mathbf{x}) \right\}, \text{ subject to } \mathbf{x} \geq 0,\ a_k \leq \frac{\mathbf{x}_1}{\mathbf{x}_k} \leq b_k,\ \forall k, \tag{4}$$

where $\alpha > 0$ is the weight of the regularization term $g(\mathbf{x})$. An examples of regularization terms are Tikhonov regularization, see Golub et al. [1999], or LASSO regularization, see Tibshirani [1996]:

$$g_{\text{Tikhonov}}(\mathbf{x}) = ||\mathbf{x}||_2^2, \tag{5}$$

$$g_{\text{LASSO}}(\mathbf{x}) = ||\mathbf{x}||_1, \tag{6}$$

where $||\mathbf{x}||_1 = \sum_i |x_i|$ denotes l_1-norm of the vector $\mathbf{x}$. Note that the solution strongly depends on the selection of the parameter α. This parameter will be manually selected to achieve the best solution in the forthcoming experiments.

We use the CVX toolbox from Grant and Boyd [2008, 2014] for convex optimization to obtain solution of the formulated optimization problem. Here, limitations of the nuclides ratios (2) can be directly taken into the account since they for a convex constraints.

2.2 Variational Bayes Inference

The Bayesian approach is based on calculation of the posterior distribution from the likelihood of the of the measurements and prior distribution of the unknown variable. The prior distribution serves as regularization of the problem Šmídl and Quinn [2006]. Evaluation of the posterior distribution is typically intractable, hence we seek for approximate solution in the form of conditionally independent distributions that minimize the Kullback-Leibler divergence to the true posterior which is known as the Variational Bayes method Miskin [2000]; Šmídl and Quinn [2006]. The Variational Bayes inference for the formulated problem (1)–(2) is similar to this by Tichý et al. [2016]; however, with further modeling of a covariance matrix of the vector $\mathbf{x}$.

The observation model of linear inverse problem (1) with isotropic Gaussian noise is

$$p(\mathbf{y}|\mathbf{x}, \omega) = \mathcal{N}_{\mathbf{y}}\left(M\mathbf{x}, \omega^{-1} I_p\right), \tag{7}$$

where symbol $\mathcal{N}$ denotes Gaussian probability distribution, I_p is identity matrix of the given size, and ω is unknown precision of noise with Gamma prior which needs to be also estimated from the data.

The source term vector $\mathbf{x}$ is also model as Gaussian; however, truncated to positive values as an analogy to "subject to $\mathbf{x} \geq 0$" in (3), see Appendix A, with notation $t\mathcal{N}$ for truncated Gaussian distribution with given support:

$$p(\mathbf{x}|\Omega) = t\mathcal{N}_{\mathbf{x}}\left(\mathbf{0}, \Omega^{-1}, [0, +\infty]\right). \tag{8}$$

Here, Ω is unknown precision matrix of the vector $\mathbf{x}$. Since our aim is to model ratios between nuclides in vector $\mathbf{x}$, we use Ω in the specific form of modified Cholesky decomposition $\Omega = L\Upsilon L^T$ where Υ is diagonal matrix with Gamma prior on diagonal elements and L is lower triangular matrix with diagonal elements equal to one. The non-zero off-diagonal elements of L forming vectors $\mathbf{l}_k$ are then equal to (2) as $-\frac{\mathbf{x}_k}{\mathbf{x}_1}$ and the prior model for each modeled ratio between $\mathbf{x}_1$ and $\mathbf{x}_k$ is

$$p(\mathbf{l}_k|\psi_k) = t\mathcal{N}_{\mathbf{l}_k}\left(\mathbf{0}, \psi_k^{-1}, [a_k, b_k]\right), \tag{9}$$

where ψ_k is diagonal matrix with unknown precision parameters with Gamma priors which are also estimated from the data.

The minimization of the Kullback-Leibler divergence leads to a set of implicit equations which have to be solved iteratively and convergence to local minima is guaranteed. Note that in opposite to the optimization approaches and their α in (4), the proposed method has no direct tunning parameter. The algorithm is called the least square with the prior adaptive covariance with interval ratios restrictions (LS-APCi) algorithm.

3 EXPERIMENT

In this experiment, we consider a simulated release of a mixture of 16 nuclides: Cs-136, Cs-134, Cs-137, I-133, I-131, I-135, I-132, I-134, Kr-85m, Kr-88, Kr-87, Sr-90, Sr-89, Te-132, Xe-135, Xe-133.

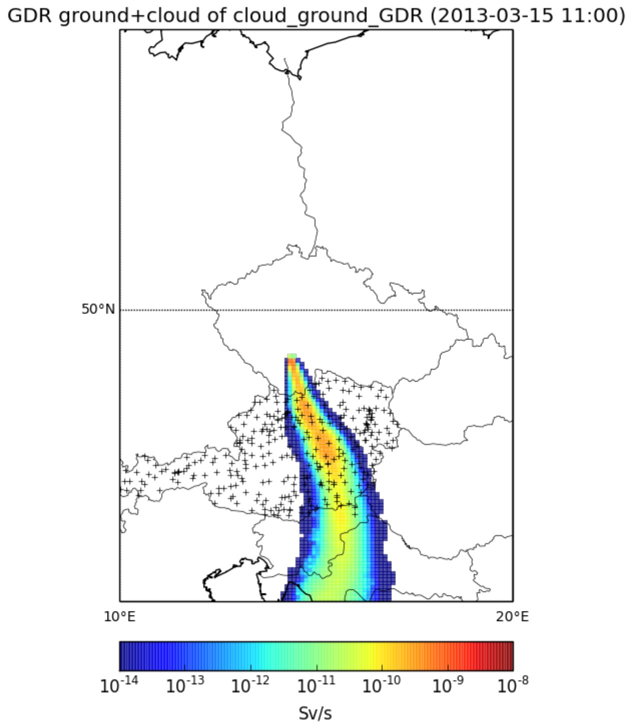

Figure 1. Gamma dose rate from the cloud shine and deposition 12 hours after start of the release.

We use the Lagrangian particle dispersion model FLEXPART (Seibert and Frank [2004]) with ECMWF Era-Interim meteorological fields with horizontal resolution 0.5 deg in the case of SRS matrix and 0.25 deg in the case of measurements. The spatial resolution of a model output is approximately 10×10 km. The Czech nuclear power plant Temelin is assumed to be the location of the release. The topology of the Austrian radiation monitoring network comprising of more than 300 receptors (Figure 1) is used in simulation of hypothetical GDR measurements with the temporal resolution one hour.

We emphasize that the measurements are in the form of the bulk of GDR so there is no distinction between contributions of single radionuclides. The effects such as different half-life or different atmospheric transportation properties reflected in the computed SRS matrix are extremely low and the problem of separation of the nuclides is highly ill-posed.

In this experiment, $\mathbf{y} \in R^{6720\times 1}$ and the total time of measurement is 14 hours, thus, the length of the vector $\mathbf{x}$ is 224 implying $M \in \mathbf{R}^{6720\times 224}$. The simulated release started 4 hours after start of the experiment and lasted for 6 hours followed by another 4 hours with no release. The original releases for each nuclide can be seen in Figures 2 and 3 using red dashed lines. The ranges of nuclide ratios are selected according to the expert opinion with true ratios laying inside the intervals.

The estimated source term of the LS-APCi algorithm is displayed in Figure 2 using solid blue lines for each nuclide. Note that the estimated source term well corresponds to the true release and the main peaks in the estimate correspond to the peaks in true release. For comparison, we selected the best estimate from optimization approaches, CVX optimization with Tikhonov regularization, see Figure 3. The tuning parameter α was selected manually as the one providing best results. In spite of this, the estimated source term has a poor quality in comparison with the estimate from the LS-APCi algorithm since even peaks of the estimated source term do not agree with the true source term. The result from the CVX optimization with LASSO regularization is comparable with this in Figure 3 and the result from the CVX optimization with no regularization are significantly worse then this in Figure 3.

All results from the four methods are compared using computed mean absolute error (MAE) for each

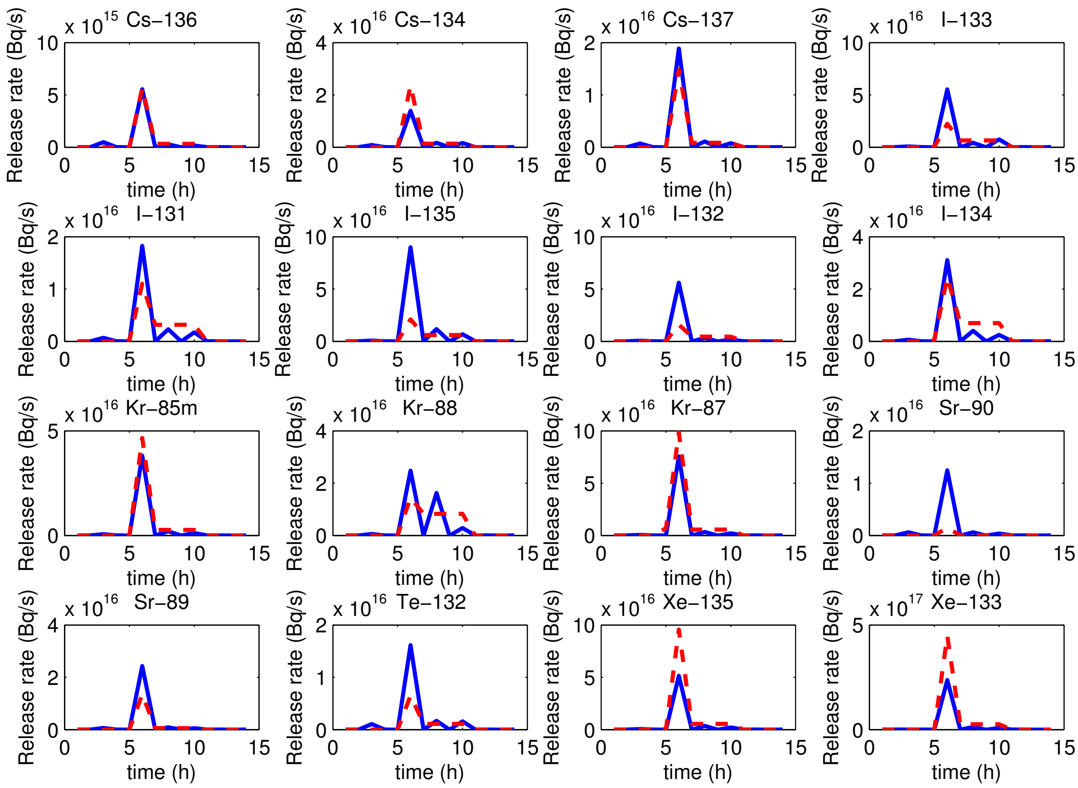

Figure 2. The results of the LS-APCi algorithm for the GDR data containing 16 nuclides. The dashed red lines are true source terms and the solid blue lines are estimated source term by the algorithm.

nuclide between the estimates and the true source term. The results are shown in Figure 4. It is shown that estimates of all nuclides provided by the LS-APCi algorithm are systematically better or comparable to optimization-based methods with one exception, Sr-90. It is also shown that the regularization in optimization problem is necessary for feasible solution since the result from the CVX optimization with no regularization provides results of poor quality. Notably, all methods were able to fit the measurement very well, see Figure 5. This agrees with the assumption that the matrix M are poorly conditioned and the measurement can be explained using many possible source terms, hence, some regularization and further prior information are crucial for source term determination from GDR measurement.

4 CONCLUSION

In this paper, we study the problem of source term determination of atmospheric release from gamma dose rate (GDR) measurement where contribution from a mixture of radionuclides is observed. Since the problem is poorly conditioned, further regularization or prior information is necessary to obtain feasible solution. In this case, we employ approximate knowledge about nuclide ratios in the form of intervals. We formulate two approaches, optimization and Bayesian. For optimization approach, we employ CVX toolbox and formulate the problem as the constrained optimization problem with selected regularizations. For Bayesian approach, we formulate probabilistic model with restricted covariance matrix of the source term using truncated Gaussian distribution.

We compared these methods on data with simulated release composed of 16 radionuclides. We have shown that the Bayesian LS-APCi algorithm outperforms optimization methods in the sense of proximity of estimated source term to the true source term. Notably, no tunning parameters are necessary in the LS-APCi algorithm since all model parameters are estimated from data.

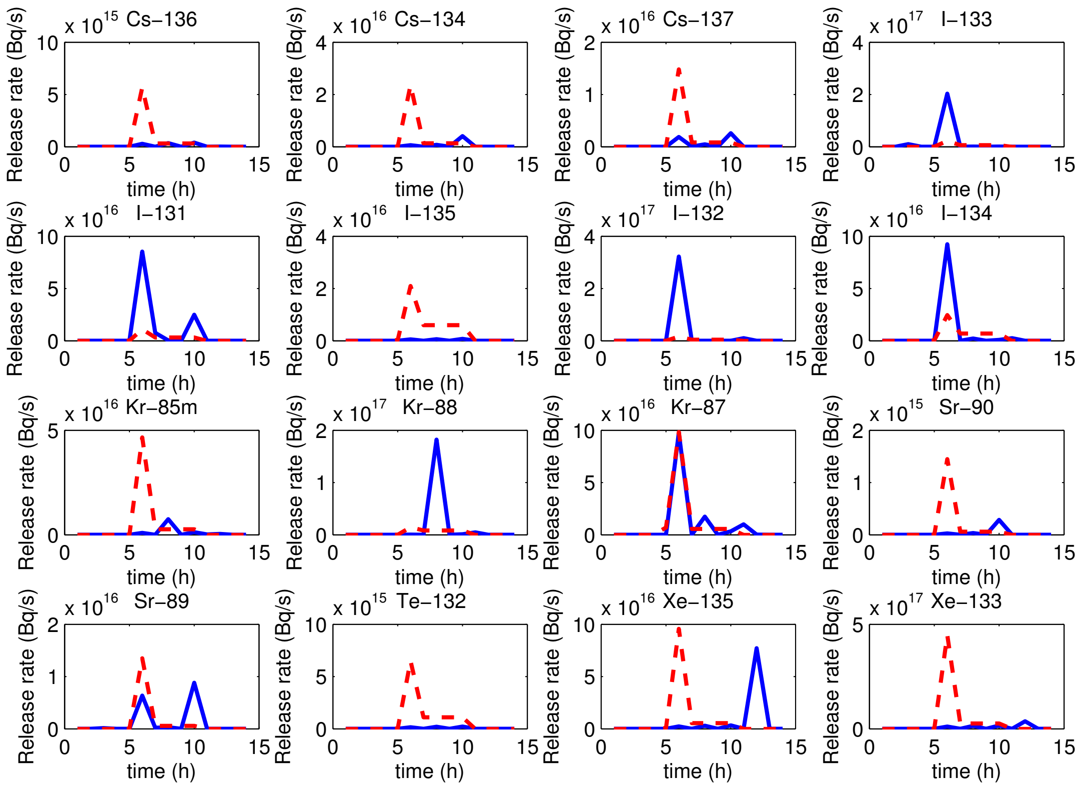

Figure 3. The results of the CVX optimization with Tikhonov regularization for the GDR data containing 16 nuclides. The dashed red lines are true source terms and the solid blue lines are estimated source term by the algorithm.

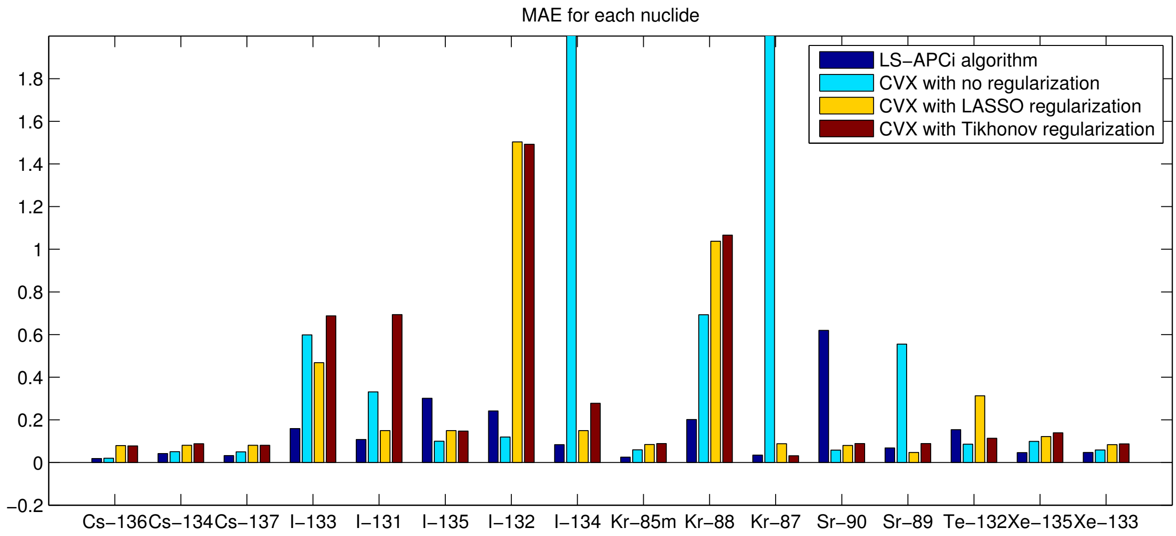

Figure 4. Computed mean absolute error between true source term and the estimated source term for each nuclide for all tested methods.

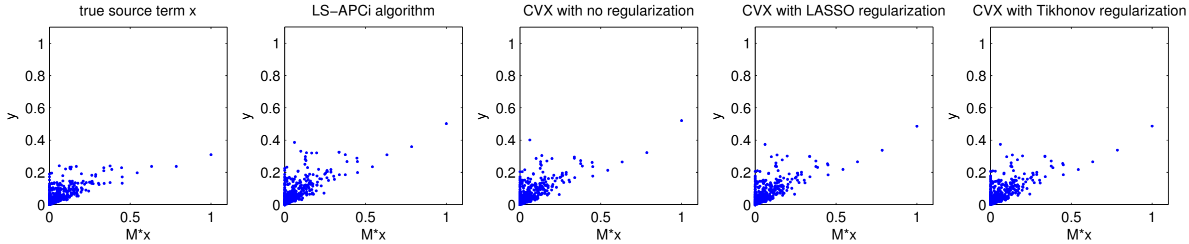

Figure 5. Fit of measurement vs. model with estimated source term **x** for all tested methods.

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A TRUNCATED GAUSSIAN DISTRIBUTION

Truncated Gaussian distribution, denoted as $t\mathcal{N}$, of a scalar variable x on interval $[a; b]$ is defined as

$$t\mathcal{N}_x(\mu, \sigma, [a, b]) = \frac{\sqrt{2}\exp\left(\frac{1}{2\sigma}(x-\mu)^2\right)}{\sqrt{\pi\sigma}(erf(\beta) - erf(\alpha))}\chi_{[a,b]}(x), \tag{10}$$

where $\alpha = \frac{a-\mu}{\sqrt{2\sigma}}$, $\beta = \frac{b-\mu}{\sqrt{2\sigma}}$, function $\chi_{[a,b]}(x)$ is a characteristic function of interval $[a, b]$ defined as $\chi_{[a,b]}(x) = 1$ if $x \in [a, b]$ and $\chi_{[a,b]}(x) = 0$ otherwise. $\mathrm{erf}()$ is the error function defined as $\mathrm{erf}(t) = \frac{2}{\sqrt{\pi}}\int_0^t e^{-u^2}\mathrm{d}u$.

The moments of truncated Gaussian distribution are

$$\langle x \rangle = \mu - \sqrt{\sigma}\frac{\sqrt{2}[\exp(-\beta^2) - \exp(-\alpha^2)]}{\sqrt{\pi}(\mathrm{erf}(\beta) - \mathrm{erf}(\alpha))}, \tag{11}$$

$$\langle x^2 \rangle = \sigma + \mu\widehat{x} - \sqrt{\sigma}\frac{\sqrt{2}[b\exp(-\beta^2) - a\exp(-\alpha^2)]}{\sqrt{\pi}(\mathrm{erf}(\beta) - \mathrm{erf}(\alpha))}. \tag{12}$$

For multivariate case, see Šmídl and Tichý [2013].

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