Atmospheric Transport Modeling of Radionuclide Releases

Matthew J. Krupcale[1*], Paul W. Eslinger[2], Fariz A. Rahman[1]

[1]University of Michigan, [2]Pacific Northwest National Laboratory

John C. Lee, jcl@umich.edu, (734) 764-9379

Consortium for Verification Technology (CVT)

Introduction

Radionuclides (RN) in the atmosphere can be used to detect and monitor the activities of undeclared and inaccessible nuclear facilities. Determining the spatiotemporal characteristics of the radiological source assists in treaty compliance monitoring. Reconstructing the radiological sources from RN data involves inverse atmospheric transport modeling (ATM) and assimilating RN data to obtain an optimal source estimate. Backwards ATM can be performed by the HYSPLIT[1] or FLEXPART[2] code, and RN data can be assimilated through Bayesian or Kalman filtering methods. This work studies and validates ATM applications to verification technology.

Goals and objectives

- Support NNSA GNDD Technology Roadmap
  - Develop accurate RN ATM capability to monitor clandestine nuclear fuel cycle activities
  - Assess uncertainties in RN source estimation
- Compare and benchmark ATM codes with atmospheric transport data
- Benchmark ATM codes with available RN data from IMS stations
- Coordinate with seismic and infrasound data
- Reconstruct radiological source using inverse ATM with example RN data
- Explore Bayesian data assimilation methods
- Develop minimum variance algorithms for source reconstruction in nonlinear ATM applications

Source-receptor relationship

Forward	Adjoint/backward
• Source oriented	• Receptor oriented
$L\psi(\mathbf{r},t)=q(\mathbf{r},t)$	$L^*\psi^*(\mathbf{r},t)=h(\mathbf{r},t)$
• ψ calculated for each source q	• ψ^* calculated for each response h
• Useful when there are fewer sources than receptors	• Useful when there are fewer receptors than sources

ψ: scalar field (i.e. concentration), q: source field, h: response

Lagrangian and Eulerian atmospheric transport models

Lagrangian	Eulerian
• Local, moving frame of reference	• Global, fixed frame of reference
• Embarrassingly parallel for non-interacting air parcels such as inert gases	• Nonlinear advection term can cause numerical instability and smear scalar gradients due to nonphysical numerical diffusion
• Better representation of physics	
$L_{\mathrm{L}}=\partial_t-\nabla\cdot k\nabla+\alpha$	$L_{\mathrm{E}}=L_{\mathrm{L}}+\mathbf{u}\cdot\nabla$
$L_{\mathrm{L}}^*=-\partial_t-\nabla\cdot k\nabla+\alpha$	$L_{\mathrm{E}}^*=L_{\mathrm{L}}^*-\mathbf{u}\cdot\nabla$

k: diffusion coefficient, α: decay or buildup constant, $\mathbf{u}$: velocity

Adjoint formulation for estimated concentration

- The backward ATM calculation results in the adjoint concentration field $C^*(\mathbf{x},t|\mathbf{x}_j,t_j)$ at source $(\mathbf{x},t)$ due to a sample measurement at $(\mathbf{x}_j,t_j)$.
- The duality relationship $R=\langle C,h\rangle=\langle Q,C^*\rangle$ between the concentration and adjoint concentration fields allows us to compute the detector response using the adjoint concentration:

$$R(\mathbf{x}_j,t_j|\boldsymbol{\theta})\equiv\overline{C}_j(\boldsymbol{\theta})=\langle Q,C^*\rangle=\int_0^{t_j}\mathrm{d}t\int_\Omega Q(\mathbf{x},t|\boldsymbol{\theta})C^*(\mathbf{x},t|\mathbf{x}_j,t_j)\mathrm{d}\Omega$$

- We parameterize the source $Q(\mathbf{x},t|\boldsymbol{\theta})$ as a Dirac delta function in space and a uniform source in time
- Then the estimated concentration is calculated from the adjoint concentration field C^* as

$$\overline{C}_j(\boldsymbol{\theta})=\sum_{i=1}^{N_s}Q_i\int_{t_{\mathrm{b},i}}^{\min(t_j,t_{\mathrm{e},j})}C^*(\mathbf{x}_{\mathrm{s},i},t|\mathbf{x}_j,t_j)\mathrm{d}t$$

Data assimilation: parametric Bayesian approach

- N independent measurements of the activity concentration, d_j
- Assume Gaussian distribution, $d_j\sim\mathcal{N}(\overline{C}_j(\boldsymbol{\theta}),\sigma_j^2)$, so that the likelihood function for the data is

$$\mathcal{L}(\boldsymbol{\theta})\equiv p(\mathbf{d}|\boldsymbol{\theta},I)=\frac{1}{\prod_{j=1}^N\sqrt{2\pi}\sigma_j}\exp\left(-\frac{1}{2}\sum_{j=1}^N\left(\frac{d_j-\overline{C}_j(\boldsymbol{\theta})}{\sigma_j}\right)^2\right)$$

- A uniform *a priori* distribution for the independent source parameters is assumed, $\theta_k\sim\mathcal{U}([a_k,b_k])$ for each parameter k:

$$p(\boldsymbol{\theta}|I)=\prod_{i=1}^{N_s}\pi(Q_i)\pi(\mathbf{x}_{\mathrm{s},i})\pi(t_{\mathrm{b},i})\pi(t_{\mathrm{e},i})$$

- Then from Bayes' theorem, the updated or *a posterior* distribution for the source parameters given the data is

$$p(\boldsymbol{\theta}|\mathbf{d},I)=\frac{p(\mathbf{d}|\boldsymbol{\theta},I)p(\boldsymbol{\theta}|I)}{\int_{\boldsymbol{\Theta}}p(\mathbf{d}|\boldsymbol{\theta}',I)p(\boldsymbol{\theta}'|I)\mathrm{d}\boldsymbol{\theta}'}$$

- Due to the high dimensionality of $\boldsymbol{\theta}$, Markov Chain Monte Carlo (MCMC) algorithms are used to sample from the *a posterior* distribution
  - The Markov process equilibrium distribution is the target *a posterior* distribution
  - Samples $\boldsymbol{\theta}^{(k)}$ are taken in proportion to the target distribution so that less time is spent sampling regions of low probability

HYSPLIT and FLEXPART Lagrangian atmospheric transport codes

- Removal processes: deposition, radioactive decay
- Forward and backward modeling

HYSPLIT	FLEXPART
• Used in PNNL and others	• Used by CTBTO and others
• Graphical user interface	• Command-line interface
• Available through NOAA ARL	• Open source
• Supports many meteorological data sets with conversion	• Supports ECMWF and NCEP GFS meteorological data

HYSPLIT 2-source problem

Description

- Generated synthetic radionuclide sample data using 120-144 hours of forward ATM
  - Hypothetical medical isotope production facility radioxenon sources
    - 2012/02/28: Idaho, U.S.
    - 2012/03/01: Missouri, U.S.
    - 6.45×10^{15} mBq release over 1 hour period
  - Sample the plumes at noble gas IMS and additional hypothetical stations in North America
- Use backward/adjoint ATM in parametric Bayesian formulation to reconstruct source terms
  - For each sample collected, run backward ATM to generate adjoint concentration field over spatiotemporal region of interest up to 96 hours before initial detection
  - Use MCMC algorithm to sample from the *a posterior* distribution

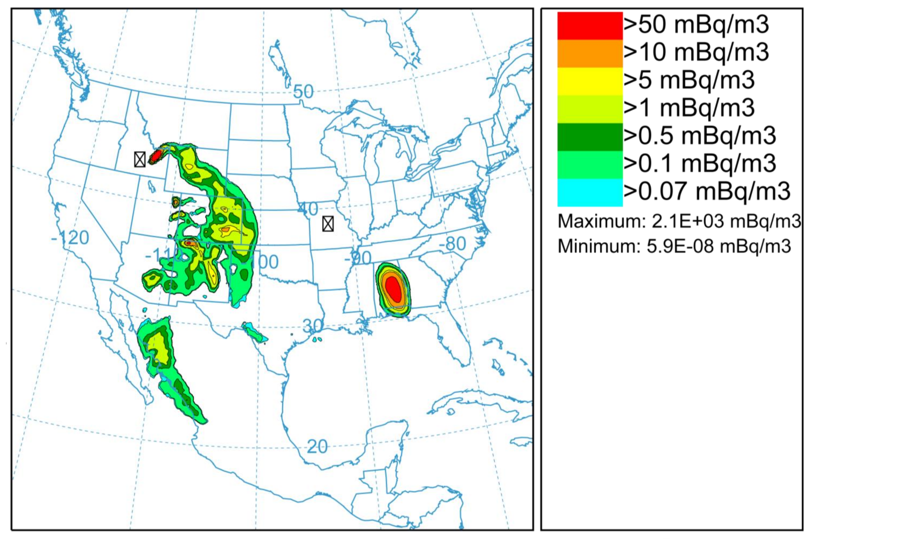

Fig. 1. Two-source emission of strength $q_0=6.45\times10^{15}$ mBq after 60 hours of forward transport simulation using HYSPLIT. There is no spatiotemporal overlap of the two plumes during the simulation.

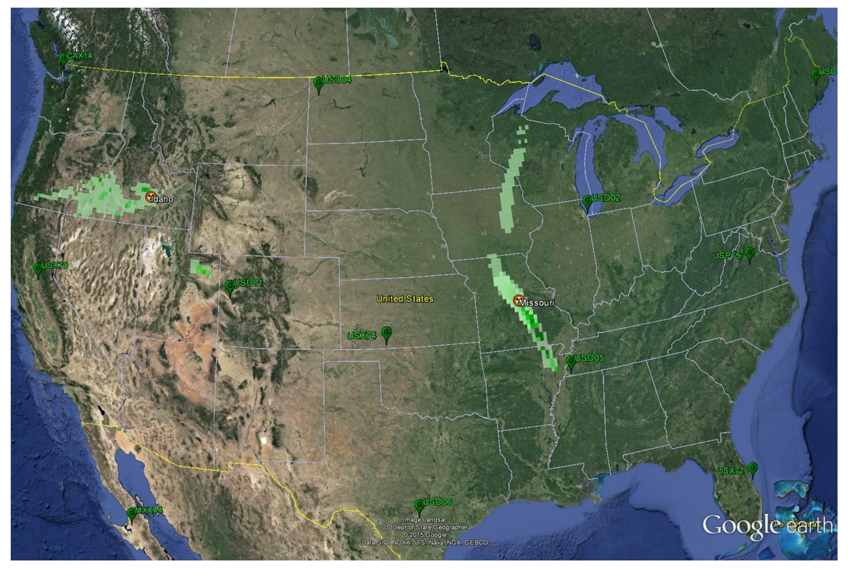

Fig. 2. Spatial marginal probability density functions for radiological sources obtained from Bayesian MCMC analysis

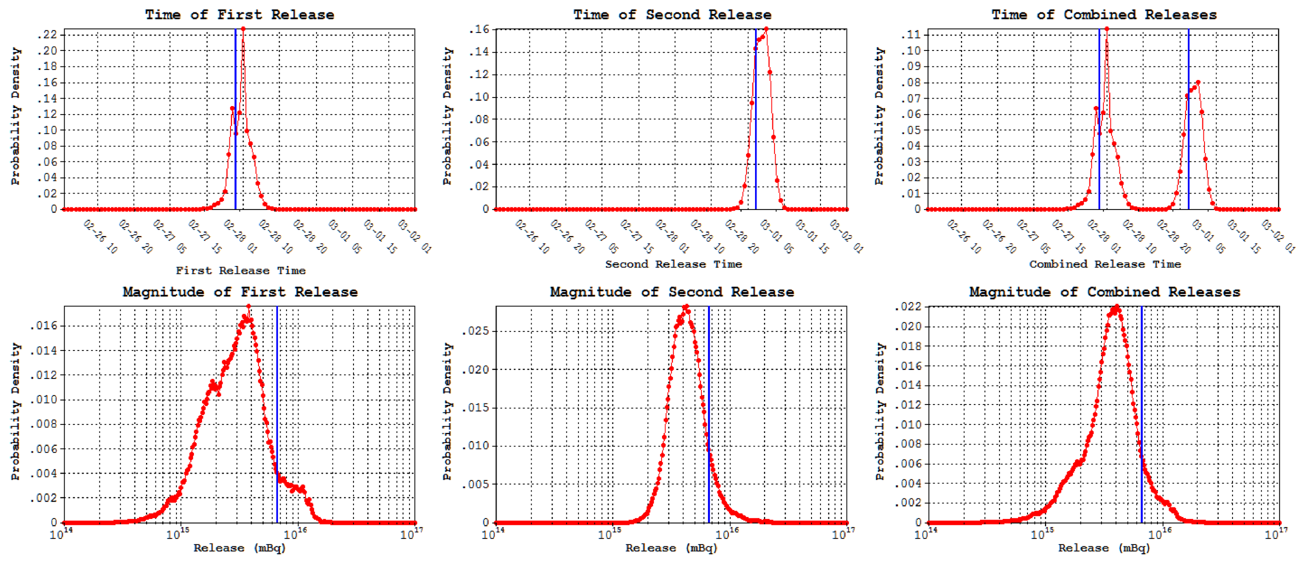

Fig. 3. Release time and magnitude marginal probability density functions for radiological sources obtained from Bayesian MCMC analysis. The blue lines show the actual release values.

FLEXPART and HYSPLIT volcano ash benchmark

Description

- Volcanic ash release in Iceland
- Line source emitting 6 types of ash over 48 hours
  - Start: 2012/05/17 1200 UTC
  - End: 2012/05/19 1200 UTC
  - 3 hour averaging period
- Concentrations from all ashes summed and averaged from 3 km to 20 km height above sea level
- Same meteorological data resolution used in FP/HS

Results

- FLEXPART shows south-west plume movement not seen in HYSPLIT
- Concentration magnitudes are on the same order
- More quantitative analysis to come

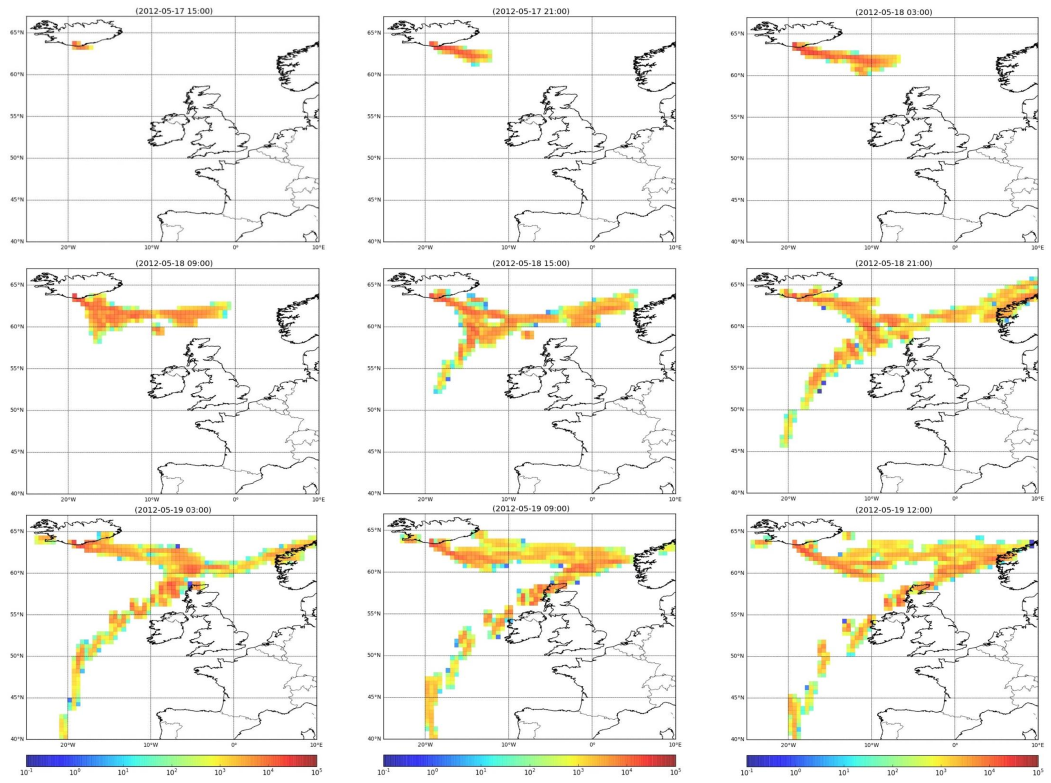

Fig. 4. FLEXPART volcanic ash forward ATM in 6 hour intervals. Units are in ng/m³.

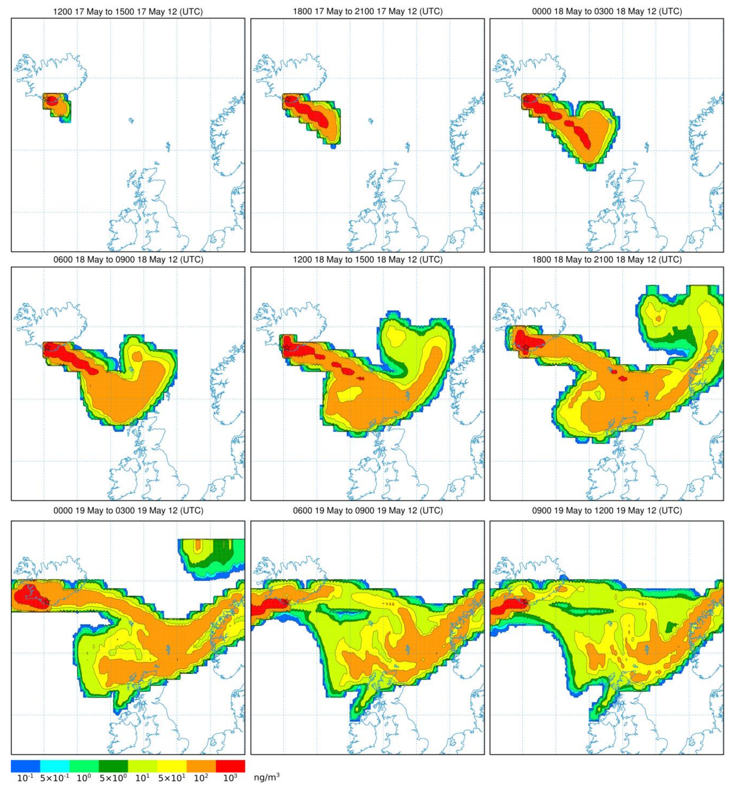

Fig. 5. HYSPLIT volcanic ash forward ATM in 6 hour intervals.

Conclusions and future work

- Reconstructed synthetic 2-source release using HYSPLIT backward ATM with MCMC sampling of *a posterior* source parameter distribution
- Benchmarking of FLEXPART and HYSPLIT with volcanic ash study
- Perform quantitative comparison between FLEXPART and HYSPLIT to help characterize ATM uncertainties
- Utilize IMS radionuclide data to simulate and reconstruct radiological release
- Explore optimal source estimation methods

This work was funded in-part by the Consortium for Verification Technology under Department of Energy National Nuclear Security Administration award number DE-NA0002534

National Nuclear Security Administration