Radionuclide monitoring through atmospheric transport modeling

Matthew J. Krupcale John C. Lee

Nuclear Engineering and Radiological Sciences, University of Michigan
2355 Bonisteel Blvd, Ann Arbor, MI 48109-2104, USA

CVT Workshop 2018-11-01

Outline

Motivation

Physical transport model

- Drift-diffusion and advection-diffusion models
- FLEXPART and HYSPLIT Lagrangian models: Gaussian plume simulation

Radionuclide modeling

- ORIGEN API and RN transmutation coupling
- May 2010 RN measurements near Korean peninsula

Conclusions

Introduction

- Atmospheric transport modeling used for
  - Estimating radionuclide (RN) release magnitudes in accident scenarios such as Fukushima
  - Monitoring clandestine nuclear activities using simulated radionuclide detector responses and contributions
- Our studies have demonstrated many uncertainties
  - Physical model
  - Meteorological data
  - Measured IMS data
- Need to assess the implications of these various sources of error on simulation results

Outline

Motivation

Physical transport model

- Drift-diffusion and advection-diffusion models
- FLEXPART and HYSPLIT Lagrangian models: Gaussian plume simulation

Radionuclide modeling

- ORIGEN API and RN transmutation coupling
- May 2010 RN measurements near Korean peninsula

Conclusions

Atmospheric transport model

- Stochastic drift-diffusion equation for turbulent velocity: FLEXPART and HYSPLIT

$$\frac{\mathrm{d}u}{\mathrm{d}t} = \underbrace{-\frac{u}{T_\mathrm{L}}}_{\text{drift}} + \underbrace{\left(\frac{2\left\langle u'u'\right\rangle}{T_\mathrm{L}}\right)^{1/2}\xi(t)}_{\text{diffusion}} \tag{1}$$

  - T_L: Lagrangian time scale or turbulent velocity correlation time
  - $\langle u'u'\rangle$: Lagrangian velocity variance related to the Reynolds stress
  - $\xi(t)$: Gaussian white noise with mean $\langle\xi(t)\rangle = 0$ and covariance $\langle\xi(t_1)\xi(t_2)\rangle = \delta(t_2 - t_1)$

- Advection-diffusion equation: Gaussian plume model

$$\frac{\partial C}{\partial t} + \mathbf{u}\cdot\nabla C - \nabla\cdot(k\nabla C) = q(\mathbf{x}, t) \tag{2}$$

- Using the eddy viscosity $k = \langle u'u'\rangle T_\mathrm{L}$, we can relate Lagrangian parameters to Gaussian dispersion factors

$$\sigma^2 = 2kt = 2\left\langle u'u'\right\rangle T_\mathrm{L} t \tag{3}$$

Outline

Motivation

Physical transport model

- Drift-diffusion and advection-diffusion models
- FLEXPART and HYSPLIT Lagrangian models: Gaussian plume simulation

Radionuclide modeling

- ORIGEN API and RN transmutation coupling
- May 2010 RN measurements near Korean peninsula

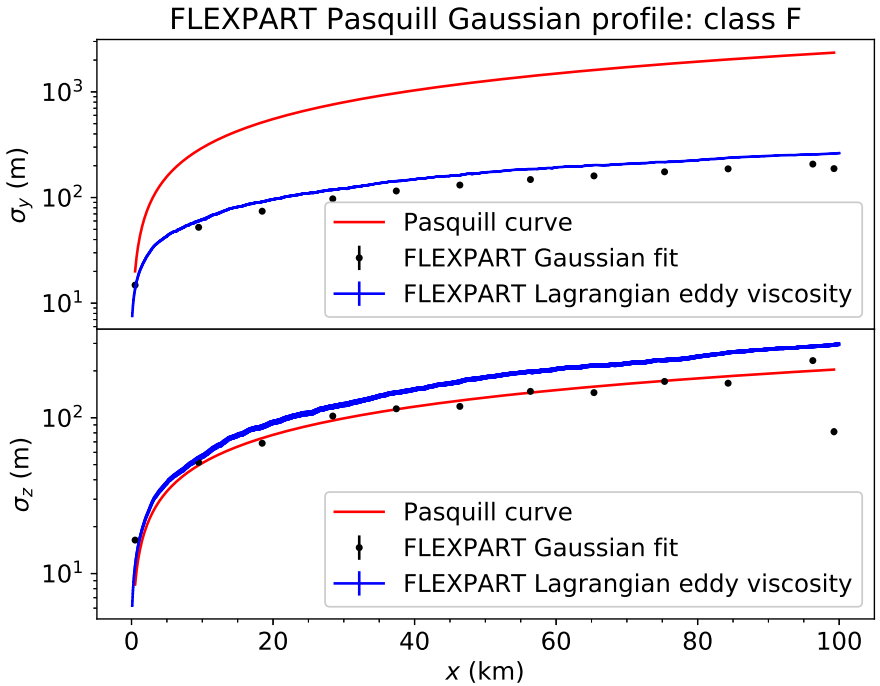
Conclusions

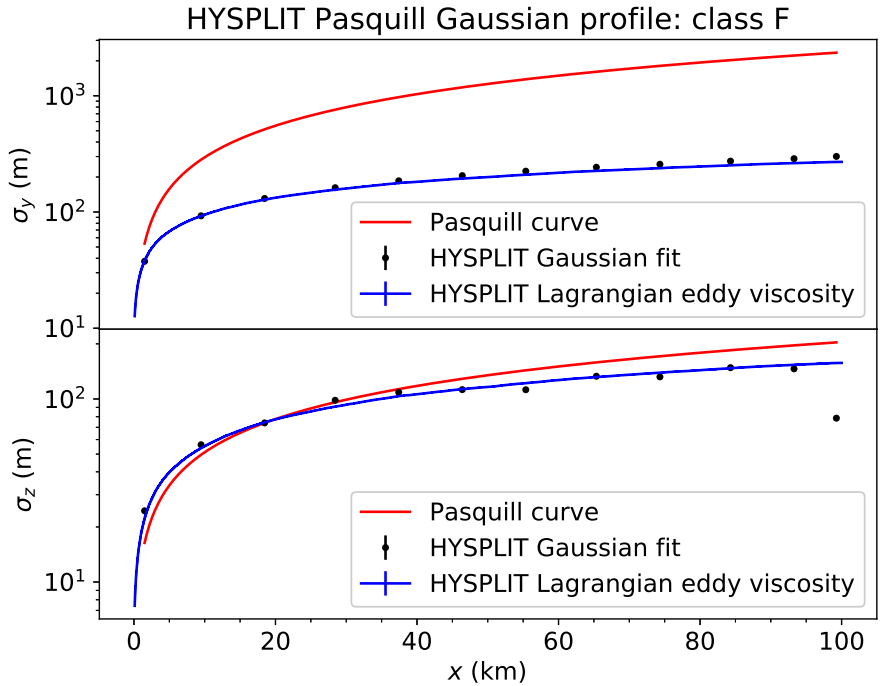
Gaussian plume model setup

- Simulated atmospheric dispersion under Pasquill F (stable) conditions
- Goal: compare the FLEXPART and HYSPLIT turbulence models for particle trajectory
- Method: implemented HYSPLIT turbulence model in FLEXPART

Dispersion factors

- Obtained the gaussian dispersion factors versus downwind distance x
  - Pasquill semi-empirical curves: $\sigma(x) = ax^b$
  - Gaussian curve fit for the dispersion parameters $\sigma(x)$
  - Lagrangian particle velocity statistics: $\sigma(t) = \sqrt{2\langle u'u'\rangle T_{\mathrm{L}} t}$
- Gaussian fit and Lagrangian dispersion factors in agreement
- FLEXPART (*left*) and HYSPLIT (*right*) dispersion factors quite similar for this case





Outline

Motivation

Physical transport model

- Drift-diffusion and advection-diffusion models
- FLEXPART and HYSPLIT Lagrangian models: Gaussian plume simulation

Radionuclide modeling

- ORIGEN API and RN transmutation coupling
- May 2010 RN measurements near Korean peninsula

Conclusions

Radionuclide transmutation: simple analytical solution

- For $^{131\mathrm{m}}\mathrm{Xe}$ and $^{133}\mathrm{Xe}$, we break the decay chains into two-step decay chains

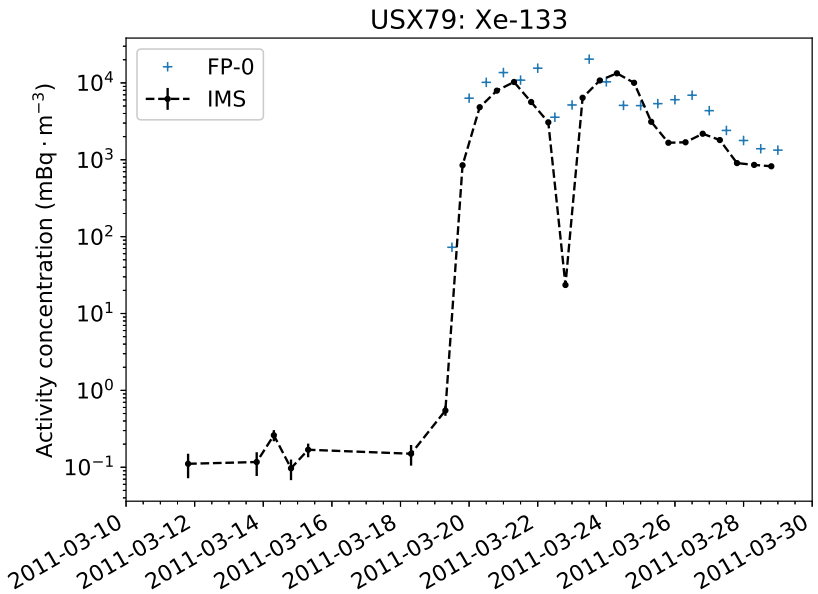
$$\mathrm{I} \xrightarrow{\lambda_{\mathrm{I}}} {}^{\mathrm{m}}\mathrm{Xe} \xrightarrow{\lambda_{\mathrm{X^m}}} \mathrm{Xe}.$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} N_{\mathrm{I}}(t) \\ N_{\mathrm{X^m}}(t) \\ N_{\mathrm{X}}(t) \end{bmatrix} = \begin{bmatrix} -\lambda_{\mathrm{I}} & 0 & 0 \\ \beta_{\mathrm{I}\to\mathrm{X^m}}\lambda_{\mathrm{I}} & -\lambda_{\mathrm{X^m}} & 0 \\ 0 & \beta_{\mathrm{X^m}\to\mathrm{X}}\lambda_{\mathrm{X^m}} & -\lambda_{\mathrm{X}} \end{bmatrix} \begin{bmatrix} N_{\mathrm{I}}(t) \\ N_{\mathrm{X^m}}(t) \\ N_{\mathrm{X}}(t) \end{bmatrix}.$$

- Isotopic depletion equations are integrated analytically
- Atmospheric concentrations are scaled by the source intensity and isotopic depletions

ORIGEN API implementation

- Used the ORIGEN libraries from the SCALE package to perform RN transmutation calculations
- Enables greater accuracy of simulated physics by coupling transmutation with transport and deposition
- Compared the time-averaged relative errors between FLEXPART simulated detector Xe-133 concentrations and IMS data



^{133}Xe	analytic correction	ORIGEN
JPX38	160%	75%
USX74	69%	97%
USX77	21%	25%
USX79	202%	209%

Outline

Motivation

Physical transport model

- Drift-diffusion and advection-diffusion models
- FLEXPART and HYSPLIT Lagrangian models: Gaussian plume simulation

Radionuclide modeling

- ORIGEN API and RN transmutation coupling
- May 2010 RN measurements near Korean peninsula

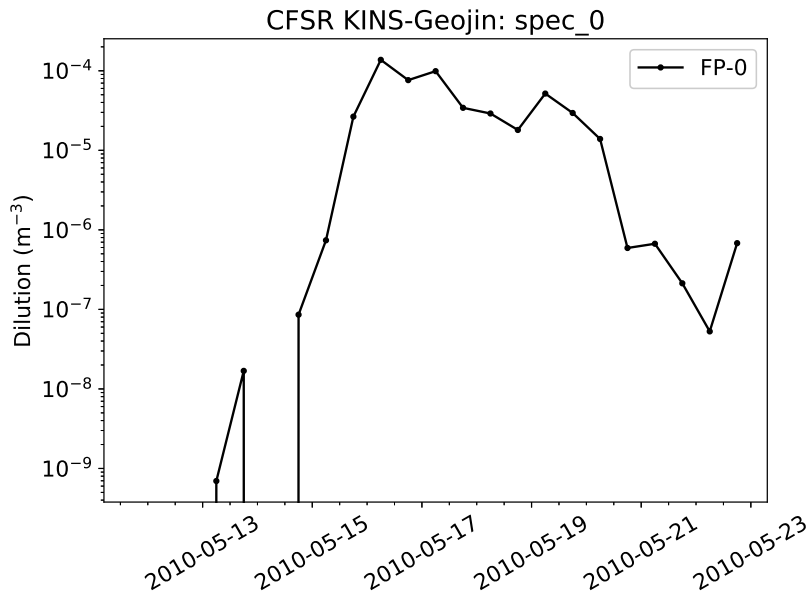
Conclusions

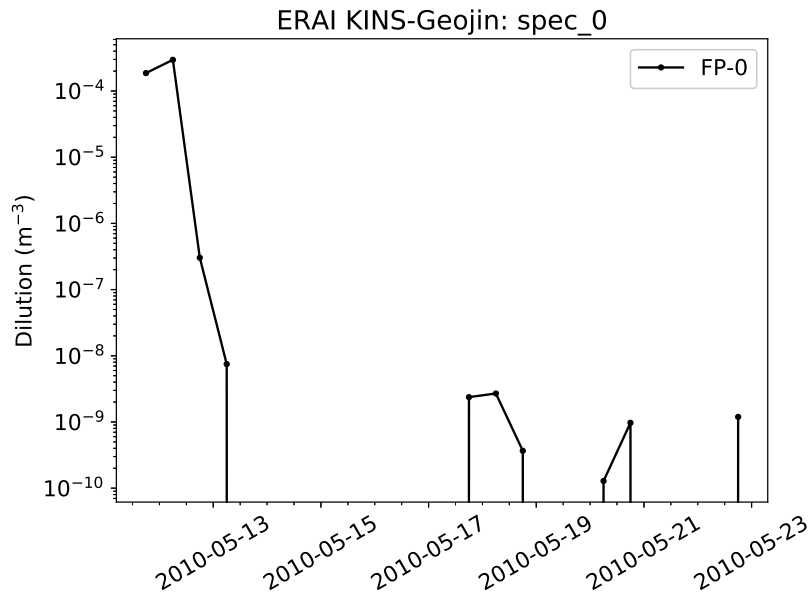
May 2010 RN measurements near Korean peninsula

- Articles[De 12, Wri13] published in 2012 and 2013 analyzing unusual RN measurements near Korean peninsula in May 2010
- Results and claims
  - $^{140}\mathrm{La}/^{140}\mathrm{Ba}$ ratio detected at Okinawa used to estimate emission time
  - Most likely candidate origin for measurements at Okinawa and Geojin was North Korea based on ATM
  - Isotopic ratios were consistent with a nuclear explosion or nuclear reactor near start-up
  - May have been a low-yield, decoupled nuclear test by North Korea on 11 May 2010
- Issues
  - Not supported by seismic evidence[Sch12, Kim16]
  - Some isotopes not detected
  - Cannot be certain nuclides came from nuclear explosion or from NKTS based on ATM

Simulated RN measurements at Geojin detector

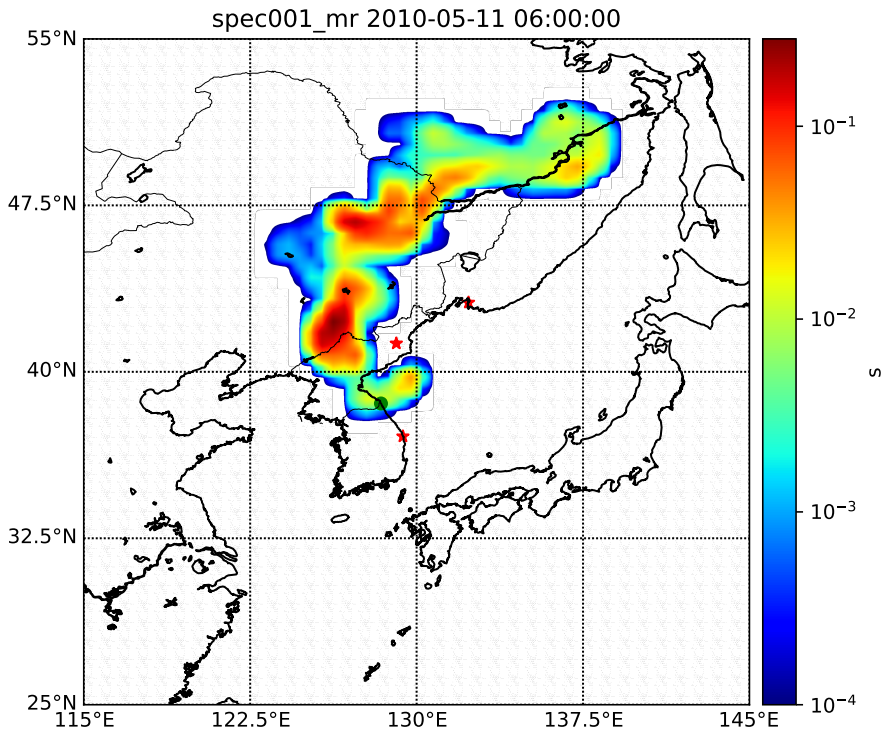
- Simulated RN measurements at Geojin South Korea due to 1-hour emission source from NKTS at 2010-05-11 06:00 UTC when using CFSR (*left*) and ERA-Interim (*right*) meteorological data sets
- Measurement depends strongly on the meteorological data
- Geojin Xe-133 and Xe-135 detection in the second half of May 13 may not be supported by ATM





Emission source contributions to Geojin detector I

- Adjoint/backward trajectory calculated for emission source contributions to detector at Geojin
- Contributions again depend strongly on the meteorological data
- There are large regions which may account for emissions in the postulated window 2010-05-10 to 2010-05-12 UTC



Emission source contributions to Geojin detector II

Table: 2°×2° spatial average of regional contribution (in seconds) from 0 to 5 meters above ground level comparing meterological data sets and emission times

Emission time (UTC)	Met data	Location		
		Bolshoy Kamen, RU	NKTS	Uljin, ROK
2010-05-10 00:00	CFSR	8.3e-3	4.5e-2	2.5e-4
	ERAI	5.7e-3	1.0e-3	3.6e-5
2010-05-11 06:00	CFSR	4.6e-7	4.7e-3	2.7e-4
	ERAI	9.5e-7	0	0
2010-05-12 00:00	CFSR	0	4.6e-3	4.4e-3
	ERAI	4.5e-5	5.0e-5	0

- Different meteorological data sets show similar source contributions on May 10 from Bolshoy Kamen when compared to NKTS
- Uljin shows similar source contribution on May 12 when compared to NKTS when using CFSR
- Need to study IMS and correct for RN measurements to compare with data

IMS data issues

- Large fluctuations in IMS RN concentrations observed
- Difficulty discriminating between $^{133}\mathrm{Xe}$ and $^{133\mathrm{m}}\mathrm{Xe}$
- Background levels of $^{135}\mathrm{Xe}$ almost always present
- Rarely have all four isotopes above $\mathrm{MDC} \sim 0.1\,\mathrm{mBq\,m^{-3}}$
- Unidentified $^{133}\mathrm{Xe}$ observations before and after DPRK weapons tests: January 2016 and September 2017
- $^{140}\mathrm{Ba}$ and $^{140}\mathrm{La}$ measured by particulate detectors in May 2010 and after Fukushima Daiichi accident

Case study results summary

- Gaussian plume study
  - For Pasquill class F stable atmosphere, the FLEXPART and HYSPLIT turbulence models produce similar dispersion despite different treatment of Lagrangian time scale
- Fukushima Daiichi nuclear accident study
  - Good agreement between ORIGEN API and analytic calculation for noble gas simulation
  - Integration of ORIGEN API enables more accurate modeling of coupled transport, deposition, and transmutation
- May 2010 RN measurements
  - Atmospheric RN measurements and source emission contributions depend strongly on the meteorological data used in the atmospheric transport model
  - Cannot rule out possibility of other potential sources besides NKTS

Future work

- Compare HYSPLIT turbulence model under unstable and neutral atmospheric conditions
- Simulate particulate deposition coupled with ORIGEN decay
- Study IMS RN issues
  - Ubiquitous $^{135}\mathrm{Xe}$ levels
  - Spurious measurements of $^{133}\mathrm{Xe}$
  - Significance of IMS rarely detecting $^{140}\mathrm{Ba}$ and $^{140}\mathrm{La}$
- Use adjoint methods for source estimation methods using Bayesian maximum a posteriori or Kalman filtering algorithms

CVT Impact

- CVT Workshops: 3 presentations and 1 poster
- National laboratory internships
  - PNNL Summer 2015: Dr. Paul Eslinger and Dr. Ted Bowyer
  - LLNL Summer 2017: Dr. Matthew Simpson
- CTBTO uses FLEXPART ATM for their analyses
- Source estimation and sensitivity/UQ analyses useful for nuclear accident scenarios and monitoring clandestine nuclear activities
- Publications

[Kru] M. J. Krupcale, M. D. Simpson, and J. C. Lee, "Uncertainty quantification in FLEXPART atmospheric transport modeling," *Atmos. Chem. Phys.* To be submitted.

[Kru17] M. J. Krupcale, J. C. Lee, and T. W. Bowyer, "Radiological source term estimates of the Fukushima Daiichi nuclear accident," *Trans. Am. Nucl. Soc.* **116**, 741 (2017).

[Kru18] M. J. Krupcale, M. E. Harkema, and J. C. Lee, "Derivation and comparison of FLEXPART and Gaussian plume models," *Submitted for presentation at Am. Nucl. Soc. Meeting* (2018).

Acknowledgements

The Consortium for Verification Technology (CVT) would like to thank the NNSA and DOE for the continued support of these research activities.

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

References

[De 12] L.-E. De Geer, "Radionuclide Evidence for Low-Yield Nuclear Testing in North Korea in April/May 2010," *Science & Global Security* **20**, 1 (2012).

[Kim16] W.-Y. Kim, P. G. Richards, D. P. Schaff, and K. Koch, "Evaluation of a seismic event, 12 May 2010, in North Korea," *Bulletin of the Seismological Society of America* **107**, 1 (2016).

[Sch12] D. P. Schaff, W.-Y. Kim, and P. G. Richards, "Seismological constraints on proposed low-yield nuclear testing in particular regions and time periods in the past, with comments on "Radionuclide Evidence for Low-Yield Nuclear Testing in North Korea in April/May 2010" by Lars-Erik De Geer," *Science & Global Security* **20**, 155 (2012).

[Wri13] C. M. Wright, "Low-Yield Nuclear Testing by North Korea in May 2010: Assessing the Evidence with Atmospheric Transport Models and Xenon Activity Calculations," *Science & Global Security* **21**, 3 (2013).