

Atmospheric Transport Modeling of Radionuclides

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Atmospheric transport modeling of radionuclides for Comprehensive Test Ban Treaty (CTBT) verification

- ▶ Support NNSA Ground-based Nuclear Detonation Detection (GNDD) Technology Roadmap
 - ▶ Develop accurate radionuclide (RN) atmospheric transport modeling (ATM) capability to monitor clandestine nuclear fuel cycle activities
 - ▶ Assess uncertainties in RN source estimation
 - ▶ Discriminate between ordinary civilian and clandestine nuclear activities
- ▶ Benchmark ATM codes with available RN data from IMS stations and aircraft volcanic ash data
- ▶ Coordinate with seismic and infrasound data



Atmospheric transport modeling using particle dispersion modelers I

- ▶ Solving the advection-diffusion equation for radionuclide concentration $C(\mathbf{x}, t)$ on a global scale can be challenging:

$$\hat{L}_E C(\mathbf{x}, t) = \frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C - \nabla \cdot (k \nabla C) - \alpha C = q(\mathbf{x}, t),$$

where

\mathbf{u} = Eulerian particle velocity

k = diffusion coefficient

α = scavenging/deposit, radioactive decay/buildup

$q(\mathbf{x}, t)$ = RN source



Atmospheric transport modeling using particle dispersion modelers II

- ▶ Turbulent motion is handled in a stochastic, Monte-Carlo method
- ▶ For non-interacting particles such as noble gases, this is an embarrassingly parallel procedure
- ▶ There are large amounts of meteorological data, I/O limitations and large numbers of simulation particles



ATM test case: Fukushima Daiichi nuclear accident

- ▶ Objectives
 - ▶ Estimated radiological release term
 - ▶ Compared radioxenon concentrations to IMS detector measurements
 - ▶ Parametrically studied sensitivity of results to source time dependence and magnitude
- ▶ Methods
 - ▶ FLEXPART model used with 1-hour global CFSR $0.5^\circ \times 0.5^\circ$ meteorological data
 - ▶ Time dependent release from March 12-16, 2011 in 24-hour uniform periods
 - ▶ Point source released at center of units 1-4
 - ▶ Noble gas and particulate species without deposition
 - ▶ Activity emissions estimated using ORIGEN2 reactor inventory estimates



Fukushima radioactivity releases

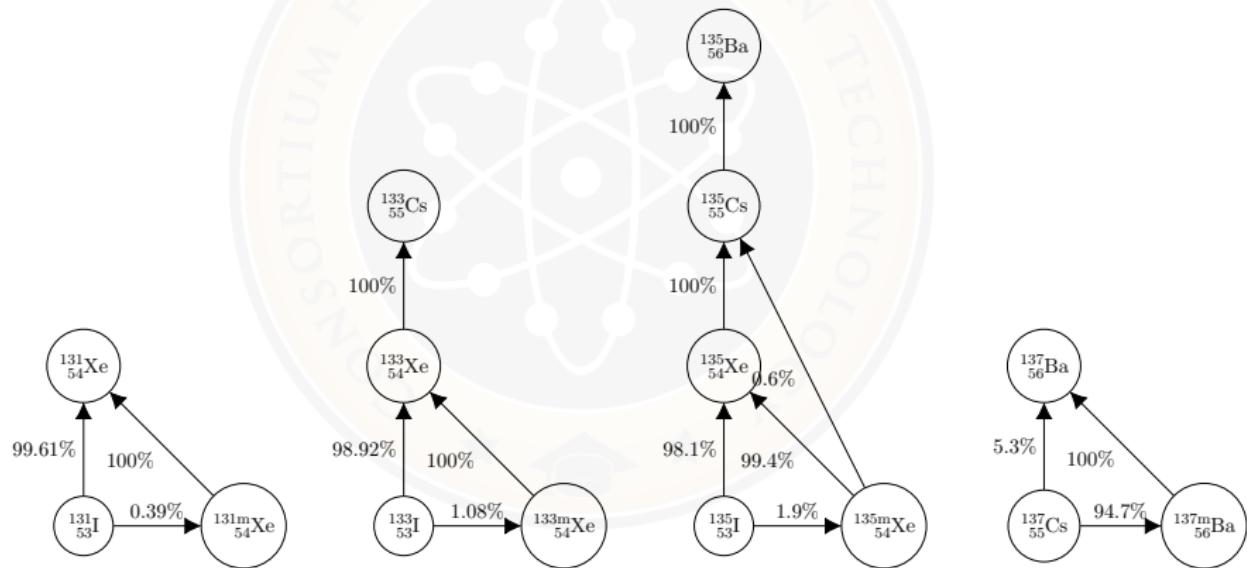
- ▶ Total Fukushima Daiichi radionuclide emissions and inventories (PBq) found in literature and in ORIGEN2 calculations
- ▶ 100% release for xenon gases; 10% for particulate
- ▶ Large uncertainties in the source intensity

Nuclide	$T_{1/2}$	Literature emissions	ORIGEN2 inventory	Estimated release
^{131m}Xe	11.84 d	N/A	6.70×10^1	6.70×10^1
^{133}Xe	5.2475 d	$(0.041 - 1.53) \times 10^4$	1.21×10^4	1.21×10^4
^{133m}Xe	2.198 d	N/A	3.58×10^2	3.58×10^2
^{135}Xe	9.14 h	5.6×10^0	4.28×10^3	4.28×10^3
^{135m}Xe	15.29 min	N/A	2.67×10^3	2.67×10^3
^{131}I	8.0252 d	$(1.5 - 1.9) \times 10^2$	6.02×10^3	6.02×10^2
^{133}I	20.83 h	3.02×10^2	1.26×10^4	1.26×10^3
^{135}I	6.58 h	N/A	1.20×10^4	1.20×10^3
^{137}Cs	30.08 y	$(1.2 - 3.7) \times 10^1$	6.98×10^2	6.98×10^0



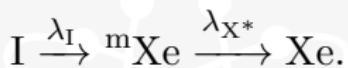
Post-processing of ATM simulation output: radioactive decay chain calculations for iodine, cesium and xenon

- ▶ Difficult to discriminate Xe-133m from Xe-133 in IMS data
- ▶ Short half lives (~ 1 d): ^{133m}Xe , ^{135}Xe , ^{135m}Xe , ^{133}I , ^{135}I



Post-processing of ATM simulation output: radioactive decay chain calculations for iodine, cesium and xenon I

- ▶ For Xe-131m and Xe-133, break the decay chains into two-step decay chains:

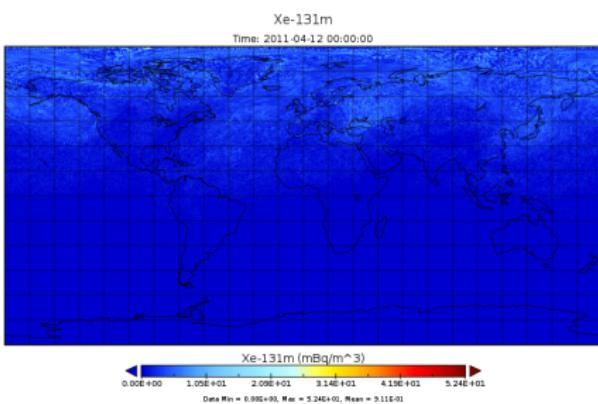
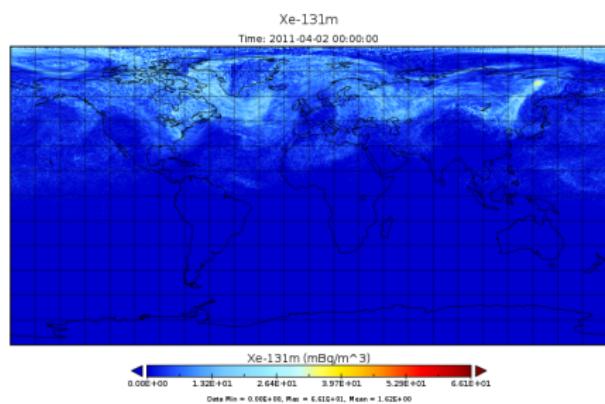
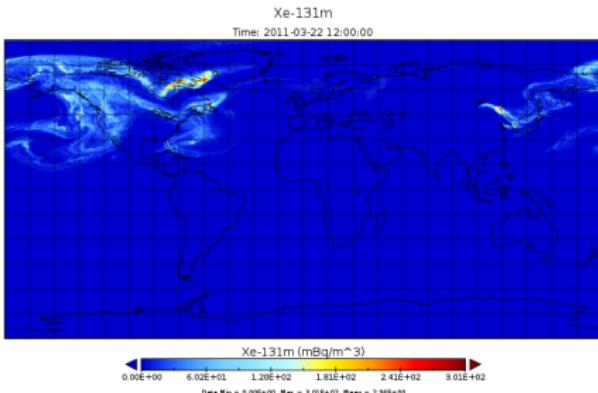
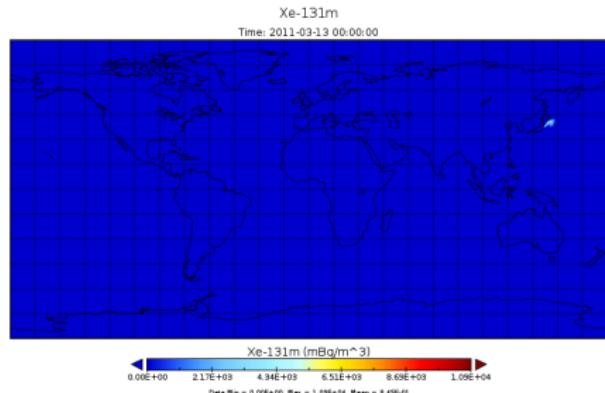


$$\frac{d}{dt} \begin{bmatrix} N_I(t) \\ N_{X^*}(t) \\ N_X(t) \end{bmatrix} = \begin{bmatrix} -\lambda_I & 0 & 0 \\ \lambda_I & -\lambda_{X^*} & 0 \\ 0 & \lambda_{X^*} & 0 \end{bmatrix} \begin{bmatrix} N_I(t) \\ N_{X^*}(t) \\ N_X(t) \end{bmatrix}.$$

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



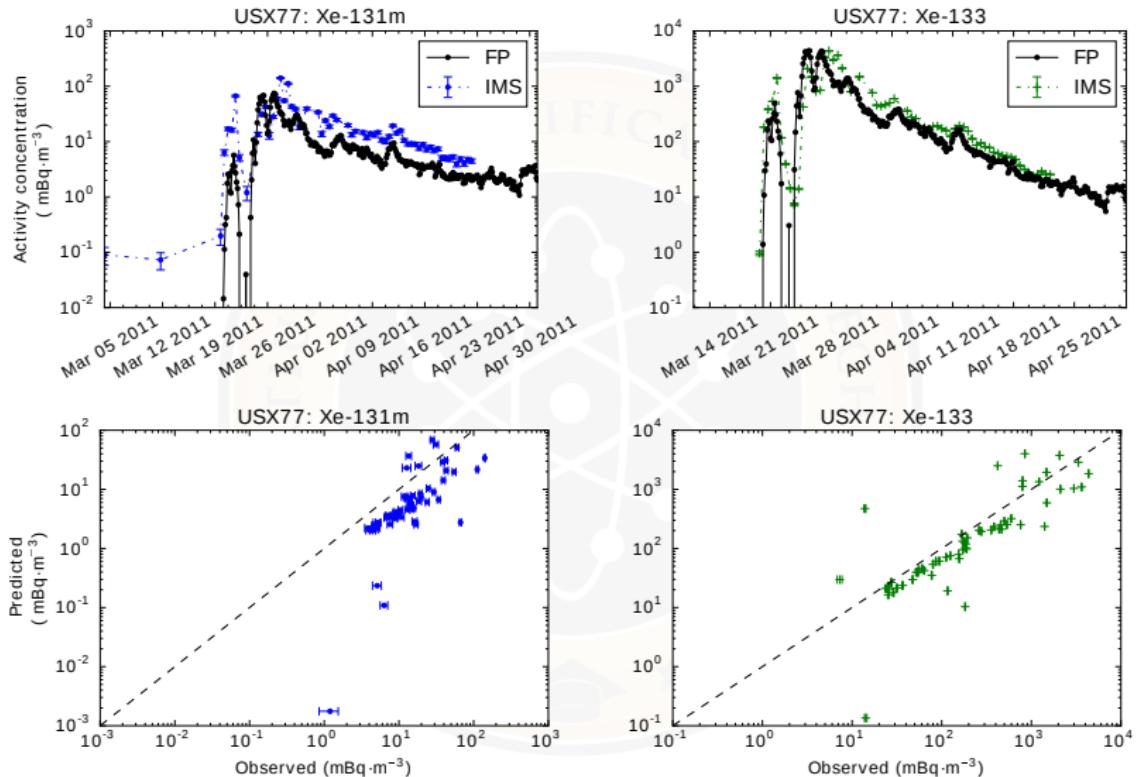
Fukushima ATM plume trajectory



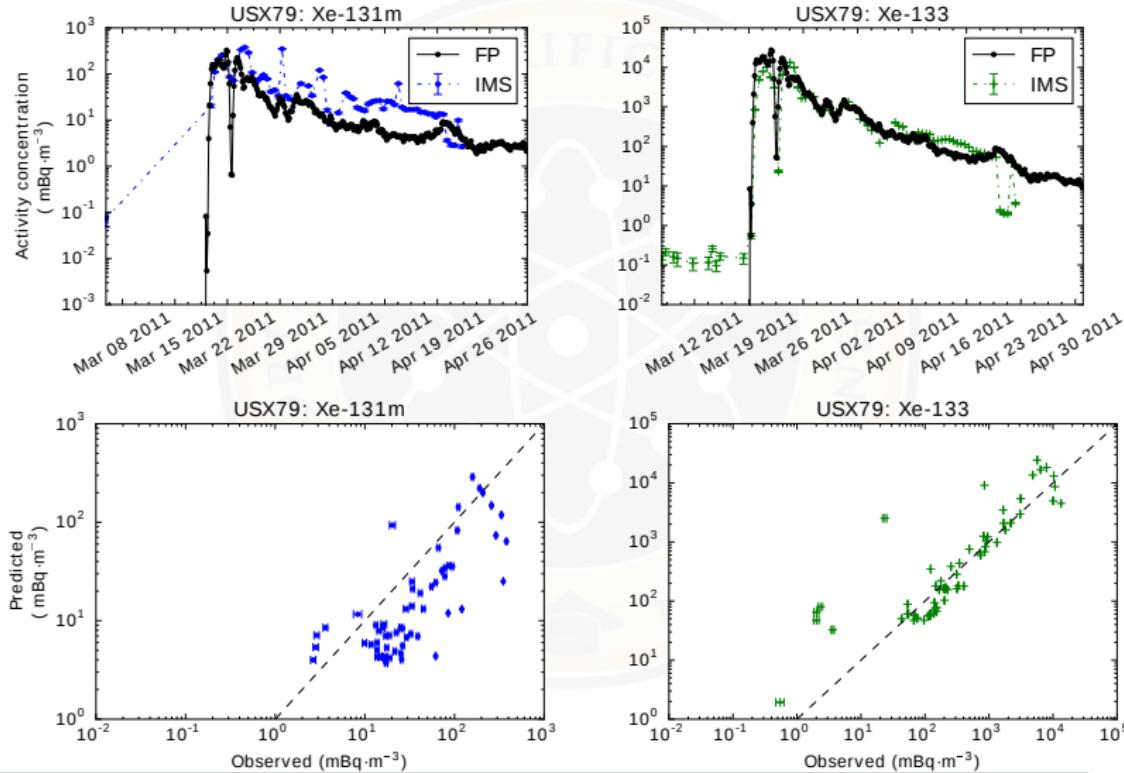
Consortium for Verification Technology



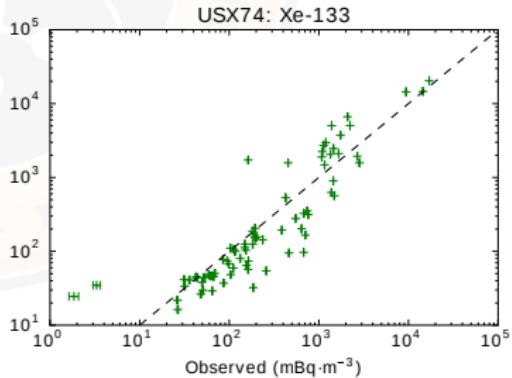
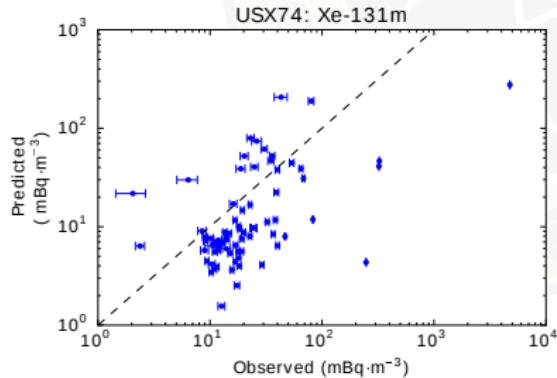
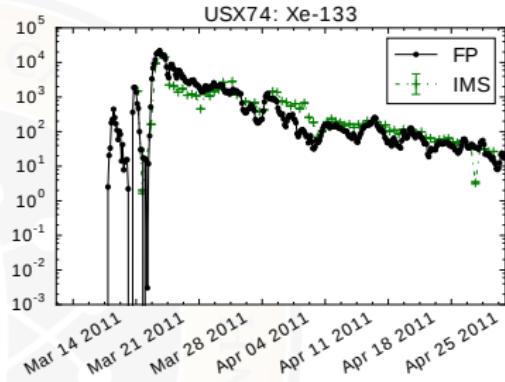
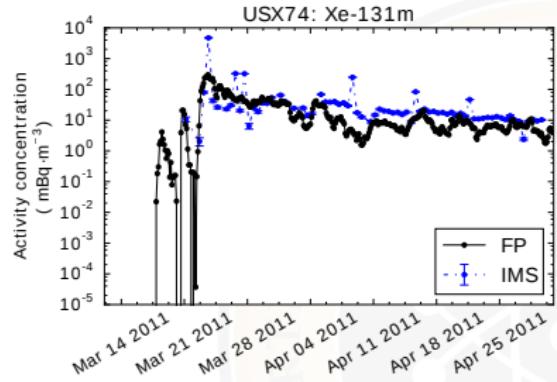
Fukushima ATM US IMS data: Wake Island, US



Fukushima ATM US IMS detector radioxenon data comparisons: Oahu, HI, US

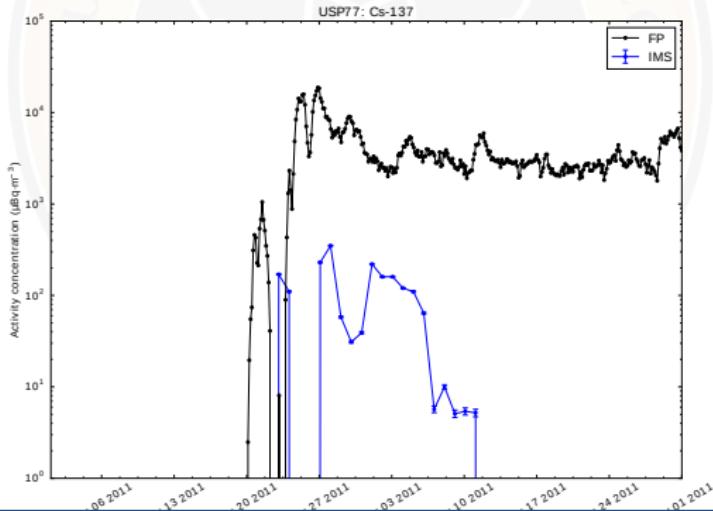


Fukushima ATM US IMS detector radioxenon data comparisons: Ashland, KS, US



Fukushima ATM US IMS detector particulate data comparisons: Cs-137

- ▶ Long Cs-137 half-life focuses on atmospheric transport alone
- ▶ Over-predicted concentration without deposition and 1% inventory emission
- ▶ Time profile leads data but follows similar profile with time condensed



Fukushima ATM US IMS detector data comparisons

- ▶ Simulation under-predicted concentrations at JPX38 (Takasaki, Japan)
- ▶ Focus on Xe-131m and Xe-133
 - ▶ Background levels of Xe-135 not accounted for in simulation
 - ▶ Discriminating between Xe-133m and Xe-133 is unreliable during most of Fukushima accident
 - ▶ Using two radioxenon concentrations generally not reliable for isotopic ratio discrimination of explosions
- ▶ FLEXPART model without deposition overpredicts Cs-137 concentrations; with deposition severely underpredicts



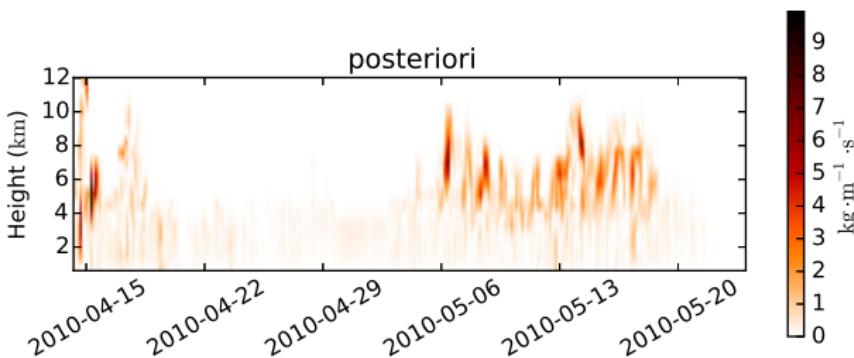
ATM test case: 2010 Eyjafjallajökull volcanic eruption I

- ▶ Objectives
 - ▶ Compare volcanic ash simulation results to observational data
 - ▶ Parametrically study sensitivity of results to different deposition schemes
 - ▶ Compare capabilities of different ATM codes (HYSPLIT and FLEXPART)
- ▶ Methods
 - ▶ FLEXPART model used with 1-hour global CFSR $0.5^\circ \times 0.5^\circ$ meteorological data



ATM test case: 2010 Eyjafjallajökull volcanic eruption II

- ▶ Maximum *a posteriori* estimated time-dependent line source at 19 height levels and 319 3-hour time periods from April 14 to May 24, 2010.
- ▶ Best estimate of eruption time and height ash mass emission profile. Courtesy of Stohl *et al.*

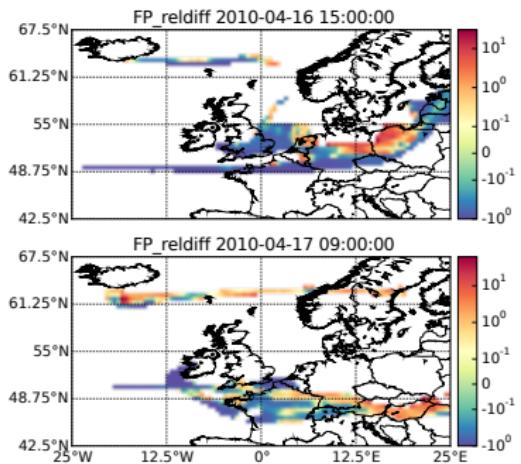
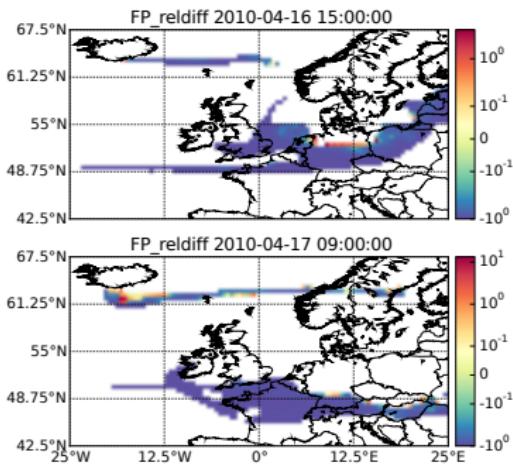


- ▶ Ash particle size distribution discretized using bimodal lognormal diameter distribution



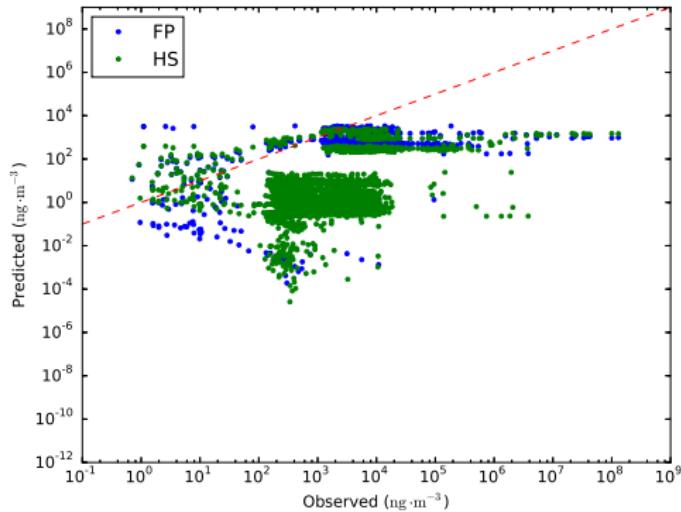
Volcanic ash ATM literature comparisons

- ▶ Compared ash concentrations to literature at two time steps with and without deposition
- ▶ Typically within 100% relative difference except around plume edges; some parts within 10%
- ▶ With dry deposition
- ▶ Without deposition



Volcanic ash ATM aircraft sampling data

- ▶ Ash concentrations measured by aircraft compared to FLEXPART (blue) and HYSPLIT (green) concentrations along the trajectory.
- ▶ Ash concentration mostly underpredicted



Conclusions and future work

- ▶ Improvements in ability discriminate between Xe-133m and Xe-133 would be helpful
- ▶ Incorporating estimates of background radioxenon concentrations may be useful, in particular Xe-135
- ▶ Dry and wet deposition models in FLEXPART probably require improvement
- ▶ Compare different meteorological datasets to characterize uncertainties
- ▶ IMS stations have not registered meaningful radionuclide data from recent DPRK weapons tests
- ▶ Adjoint source estimation procedures using Kalman filtering and Bayesian maximum *a posteriori* estimation

