Inverse estimation of the emission of radioactive materials from Fukushima

Taichu Y. Tanaka, Takashi Maki, Mizuo Kajino, and Tsuyoshi T. Sekiyama

Meteorological Research Institute, Japan Meteorological Agency

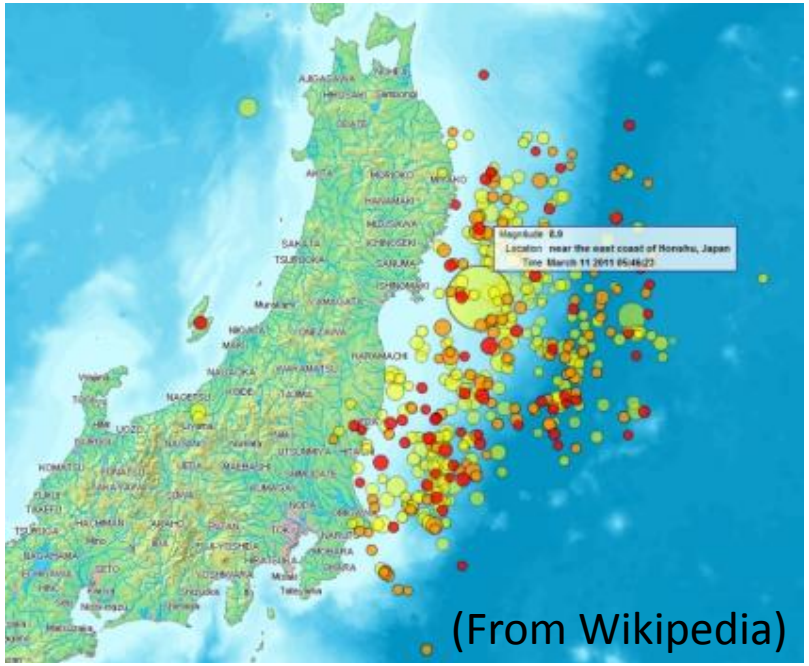
16 May 2012, ICAP workshop@ESA/ESRIN, Frascati

Outline

- Introduction
- Global simulation of radioactive materials from Fukushima
- Preliminary results of the inverse estimation of the emission flux of ^{137}Cs from the accident.

On 11 March 2011,

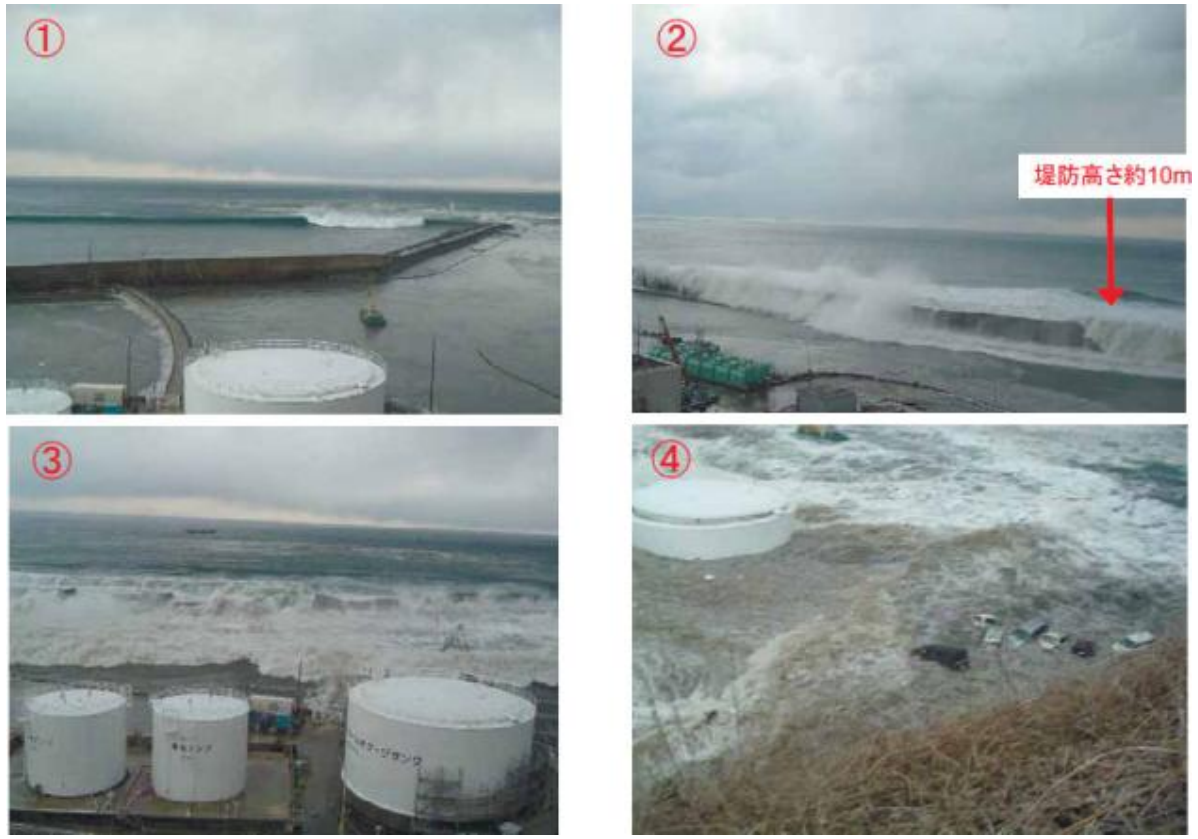
- 2011 Tohoku earthquake occurred:
  - A Huge earthquake with Mw9.0 occurred off the Sanriku, approximately 180km off the Pacific coast of Japan, and a large Tsunami attacked the Pacific coast of Tohoku (North eastern region) and Northern Kanto regions in Japan.
  - More than 19,000 people have been missing or passed away.



(From Wikipedia)

At Fukushima prefecture,

- The Tsunami flooded the Fukushima Dai-ichi nuclear power plant (Fukushima NPP1), and caused a total loss of electricity which leads to the meltdown of the reactors.
- Huge amount of radioactive materials are released to the environment.



(From TEPCO report)

Motivation of our study

- This disastrous accident of nuclear power plant should be investigated and documented as possible in detail for the future generation.
  - The amounts of released radioactive materials are essential data.
- We should improve the atmospheric tracer models and analysis technique by evaluating the emission, transport and deposition of this event.

Estimated amount of released ^{137}Cs

- Japan Atomic Energy Agency (Chino et al., 2011) 12.6 PBq (3/12-4/5)
- JAEA, revised 9.1 PBq (3/11-4/5)
- JNES report 15 PBq* (Initially, 6.1PBq)
- Stohl et al. (2012,ACP) 36.6 PBq(20.1–53.1PBq; 3/10-4/20)
- Winiarek et al. (2012,JGR) 10 - 19 PBq
- IRSN, France 10 PBq (-3/22)†
- ZAMG 66 PBq (the first four days)

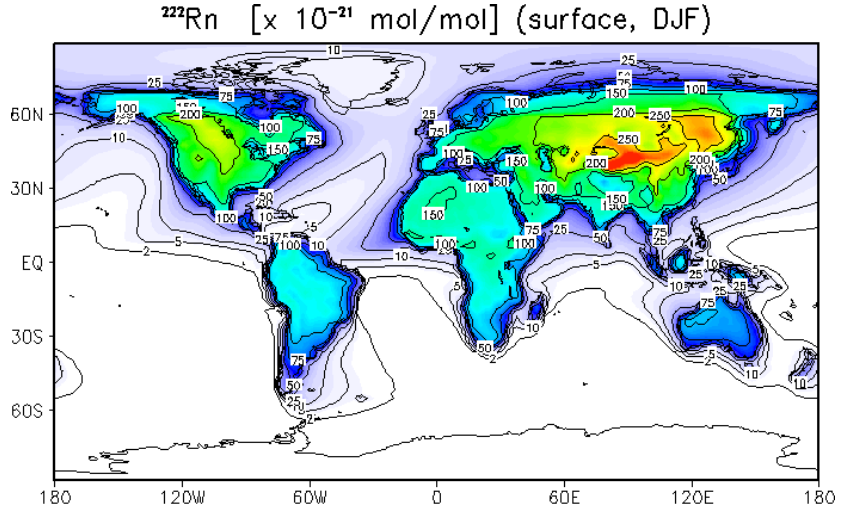
→ More than factor of 5 difference!

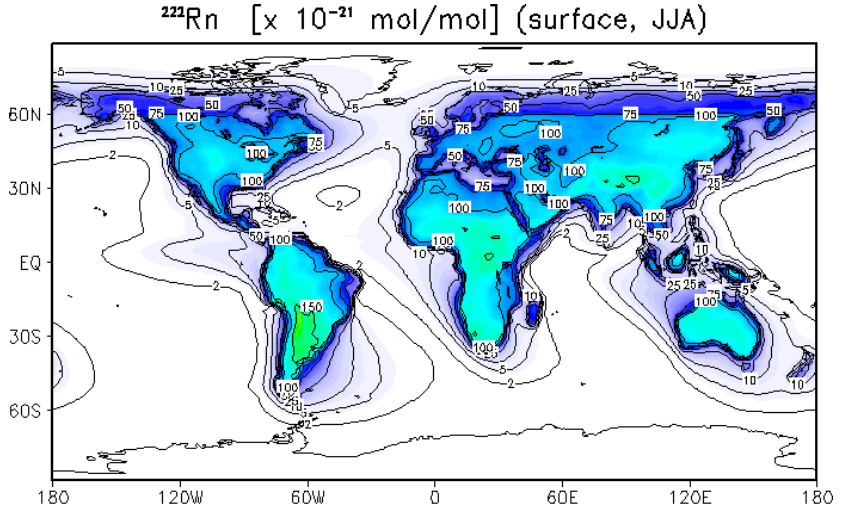
*Report of Japanese Government to the IAEA Ministerial Conference on Nuclear Safety - The Accident at TEPCO's Fukushima Nuclear Power Stations -, June, 2011

† http://www.irsn.fr/EN/news/Documents/IRSN_fukushima-radioactivity-released-assessment-EN.pdf

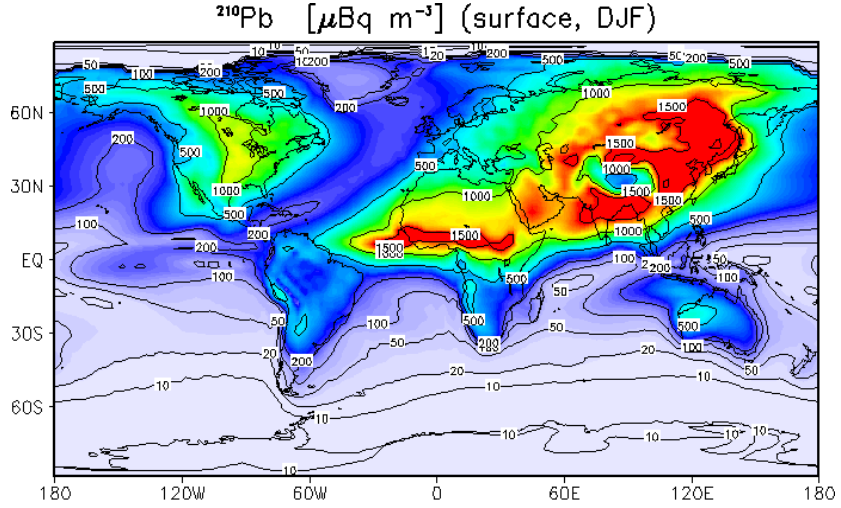
Simulation of ^{222}Rn and ^{210}Pb

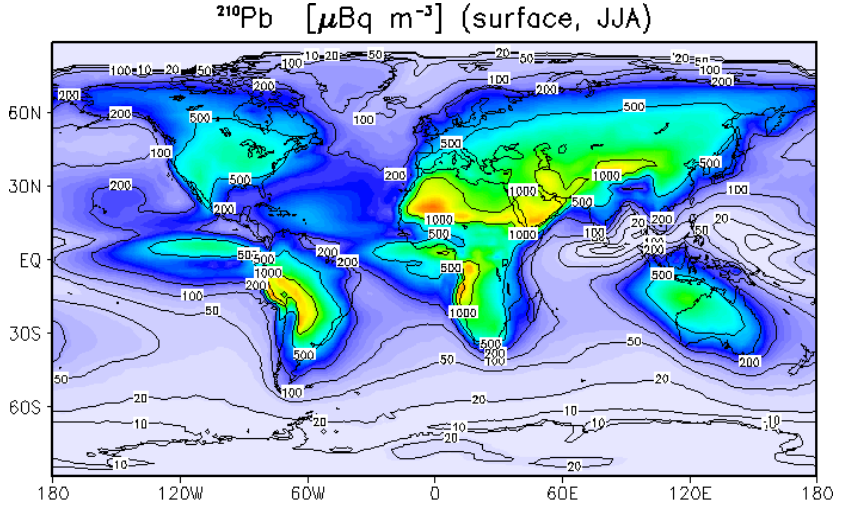
^{222}Rn





^{210}Pb



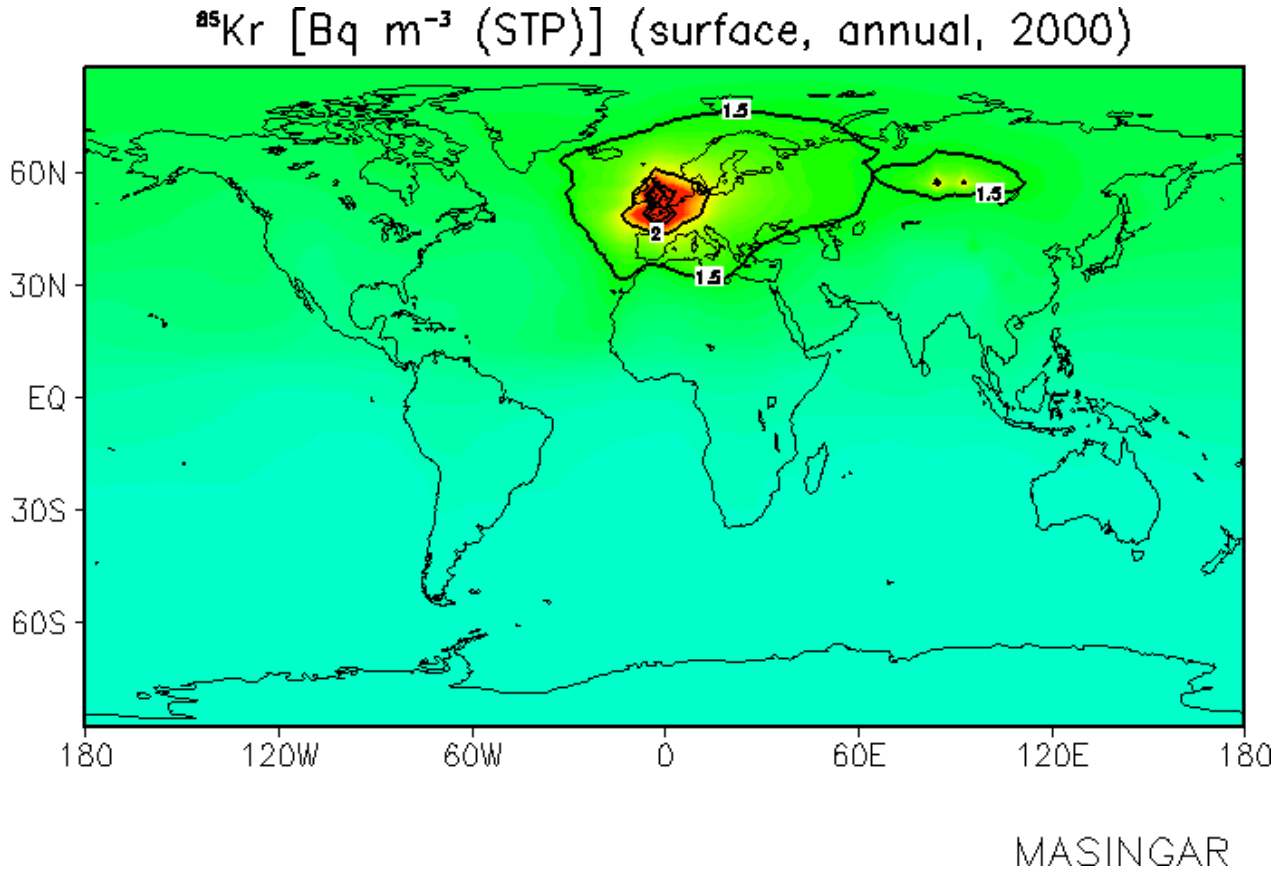


DJF (Winter in NH) MASINGAR

JJA (Summer in NH) MASINGAR

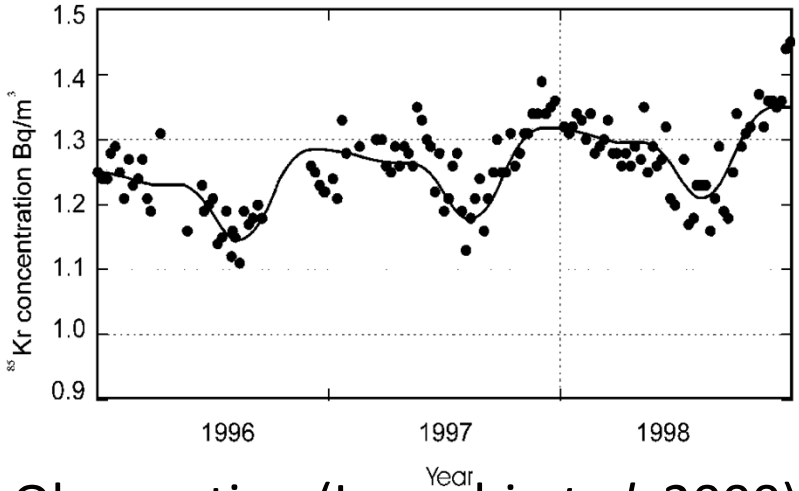
Above: ^{222}Rn, below: ^{210}Pb, surface concentration

Simulation of ^{85}Kr from nuclear reprocessing plant

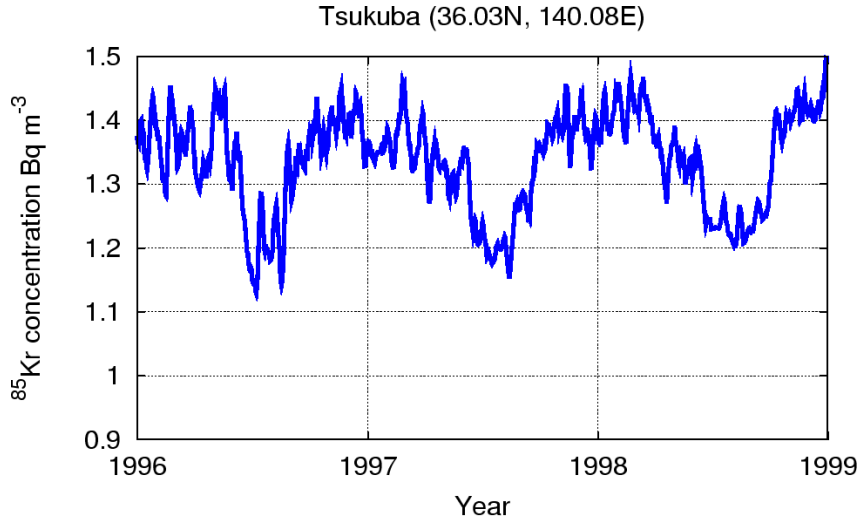


Simulated surface concentration of ^{85}Kr from 1955 to 2000.

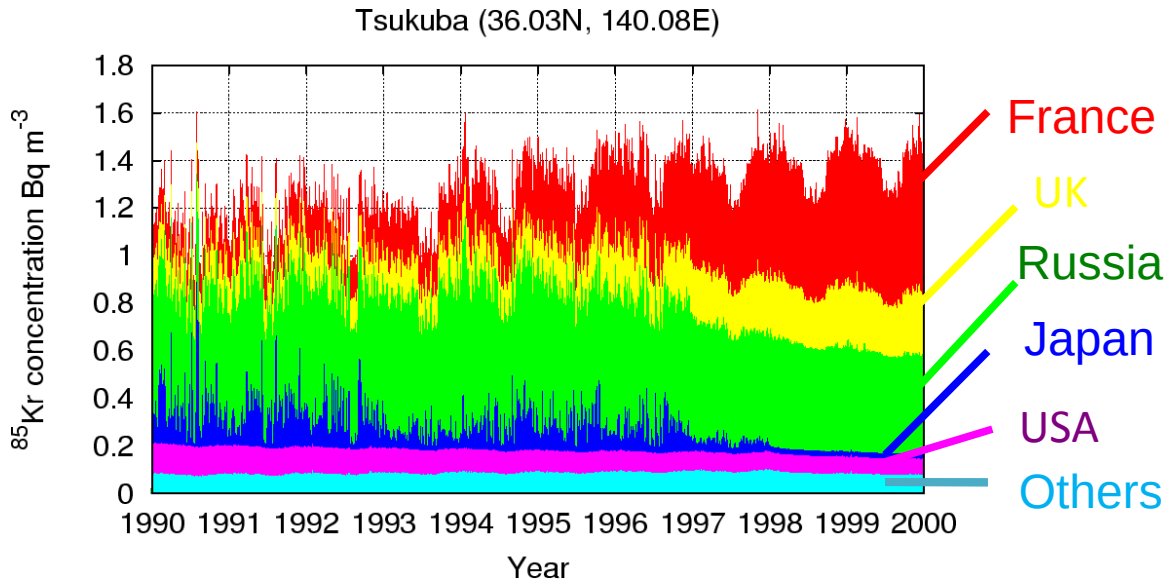
Comparison with observed concentration of ^{85}Kr at MRI/JMA, Tsukuba



Observation (Igarashi *et al.* 2000)



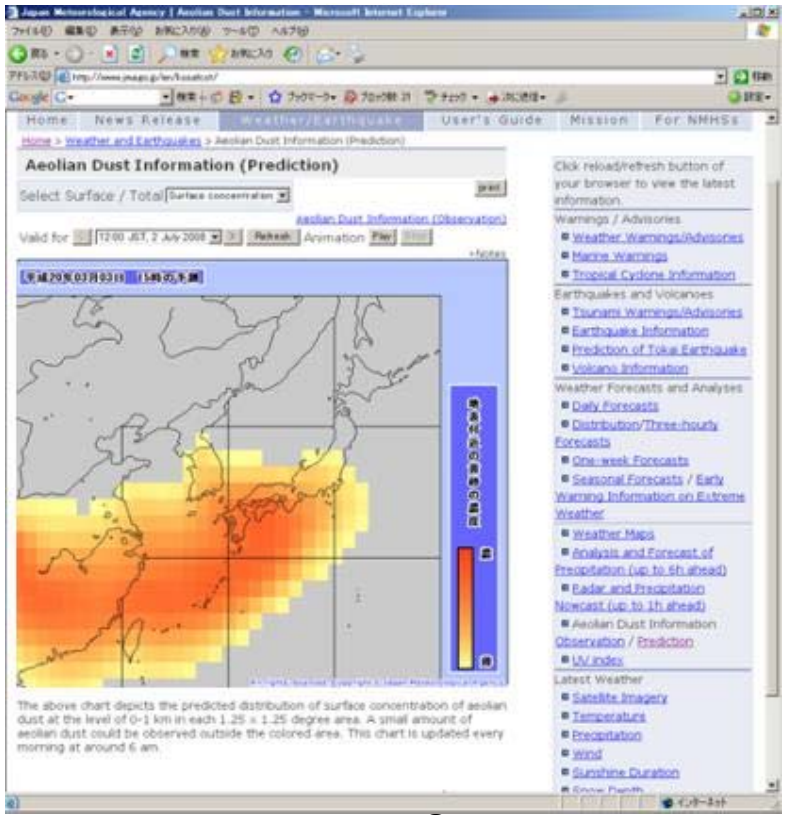
Simulated with MASINGAR



Simulation of the radioactive materials from the Fukushima accident

Brief description of the global model

- We used our global model called MASINGAR (Model of Aerosol Species in the Global Atmosphere), which calculates the distribution of aerosol species.
- MASINGAR has been used for the Asian dust information by JMA, and climate change experiments by Meteorological Research Institute.
- Here we will show the results of the developmental version of the model (MASINGAR mk-2).

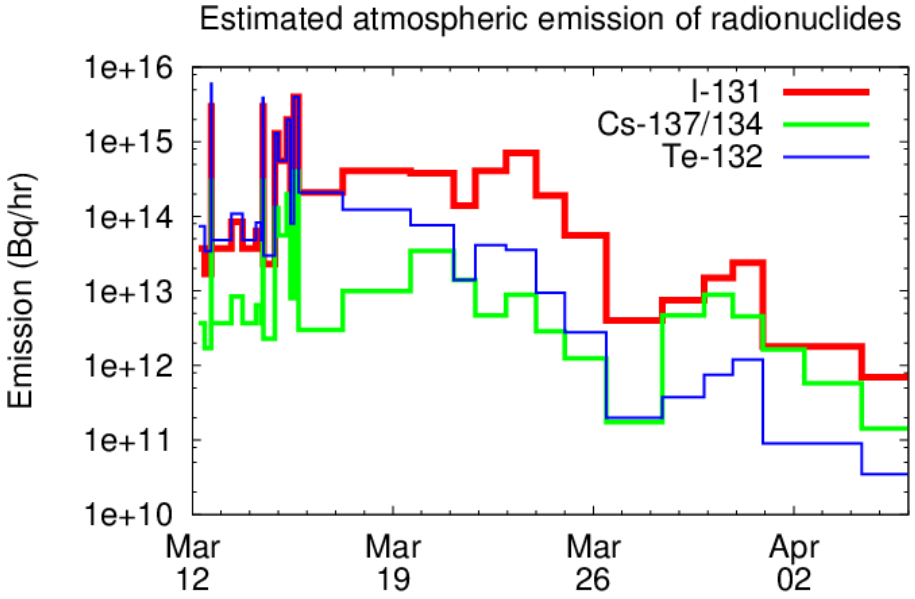


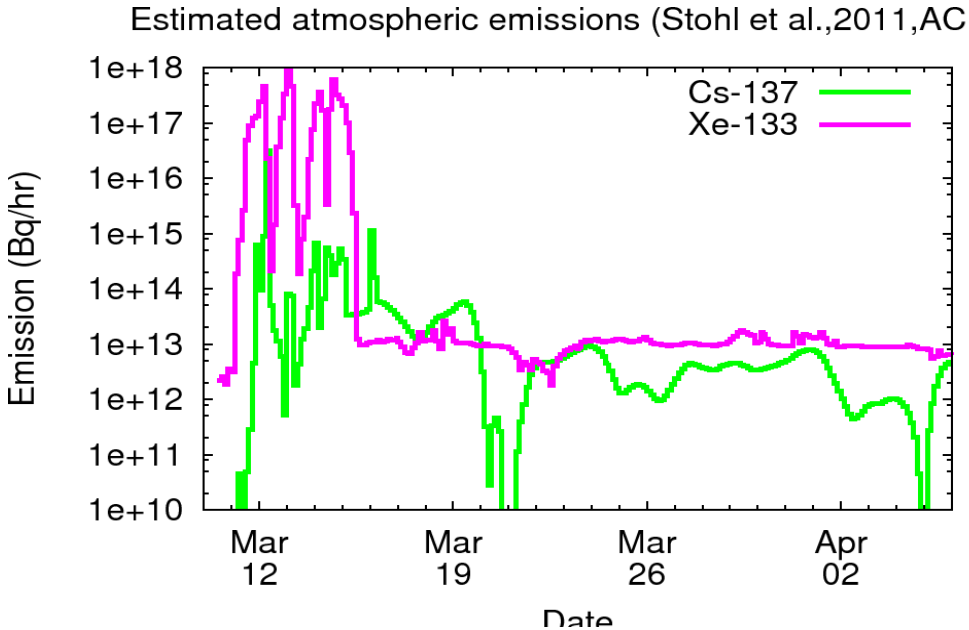
http://www.jma.go.jp/jp/kosa/

Experimental settings

- Included radionuclides: 6 species
  - I-131, Cs-137, Cs-134, Te-132, I-132, Xe-133
  - Xe-133 is treated as non-reacting gas with no dry and wet depositions.
  - Other species are assumed to be attached to aerosols (Lognormal size distribution with r_n=0.07μm, σ=2.0, and hydrophilic)
- Model resolution: Horizontal TL319 (0.56°, approx.60km), vertical 40 layers (ground– 0.4hPa)
- Atmospheric dynamical model: JMA/MRI unified general circulation model (MRI-AGCM3)
- Horizontal wind components are nudged using JMA global analysis (GANAL).

Estimated emissions from Fukushima Dai-ichi nuclear power plant

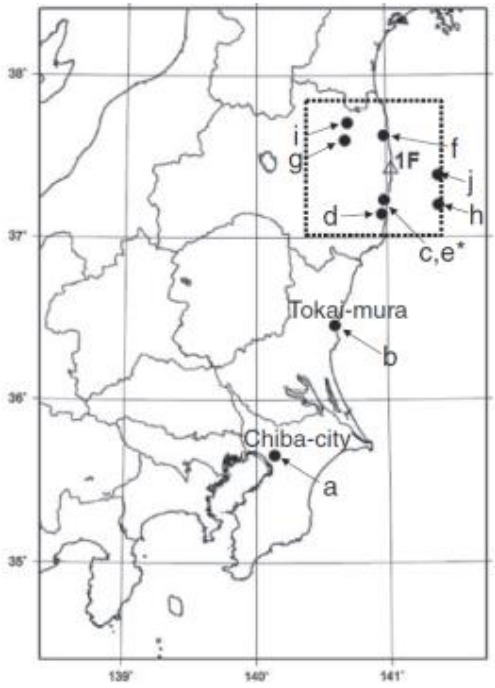




- Source:
  - ^{131}I, ^{137}Cs, ^{134}Cs, ^{132}Te : JAEA (Chino, personal communication)
  - ^{133}Xe: Stohl *et al.* (2011)
- For ^{137}Cs, estimated emission by Stohl et al. (2011) is used for a sensitivity study.

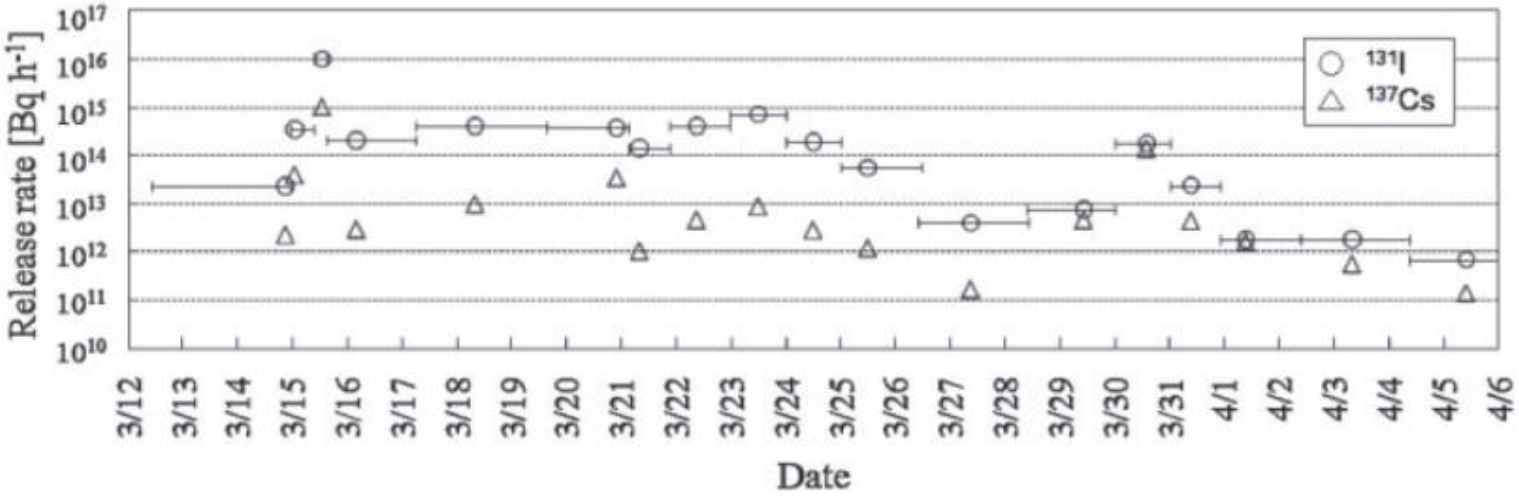
Prior studies (1)

Japan Atomic Energy Agency (JAEA) estimated emission amount of radio nuclei by comparing model simulation results and observation data (Chino et al., 2011)



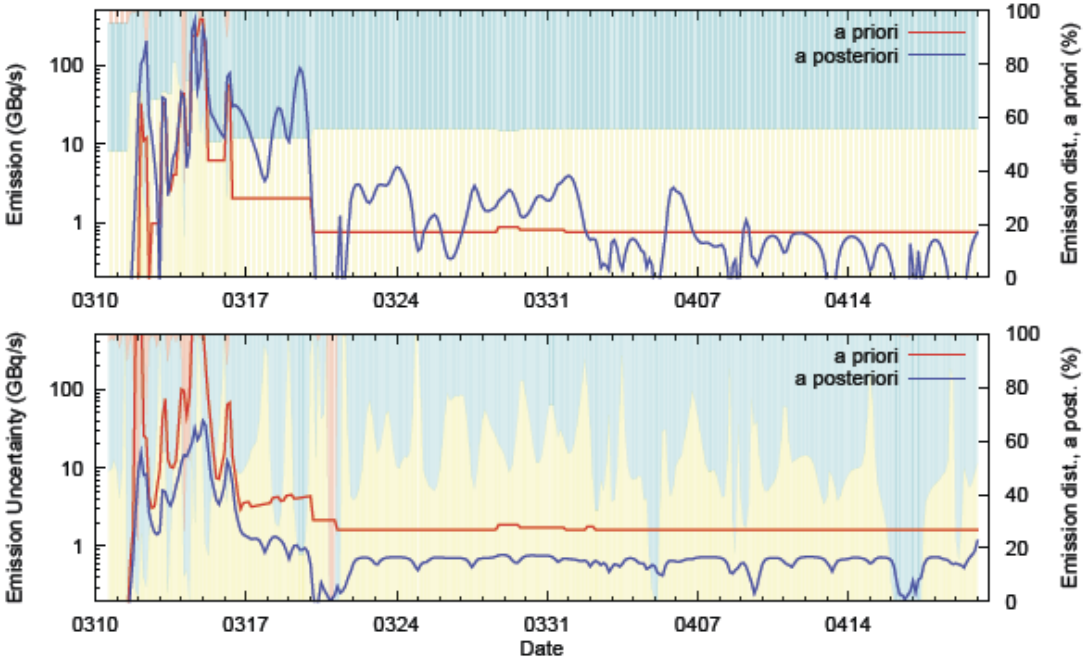
Data No.	Sampling date and time	Sampling point
1	2011/3/15 06:00-07:00	a
2	2011/3/15 06:55-07:15	b
3	2011/3/16 06:05-06:25	b
4	2011/3/18 10:50-11:18	c
5	2011/3/21 04:45-05:05	b
6	2011/3/21 10:50-11:08	d
7	2011/3/22 14:17-14:31	c
8	2011/3/23 13:15-15:10	e
9	2011/3/24 14:55-15:15	f
10	2011/3/25 15:05-15:22	g
11	2011/3/27 11:45-	h
12	2011/3/29 11:17-12:15	f
13	2011/3/30 15:11-15:32	g
14	2011/3/31 12:22-13:12	e
15	2011/4/1 12:00-12:20	i
16	2011/4/3 11:04-	j
17	2011/4/5 13:07-13:27	f

*Points c and e are almost overlapped.



Prior studies (2)

Norwegian Institute for Air Research(NILU) estimated emission amount of radio nuclei by their Lagrangian transport model and inverse model (Stohl et al., 2011).

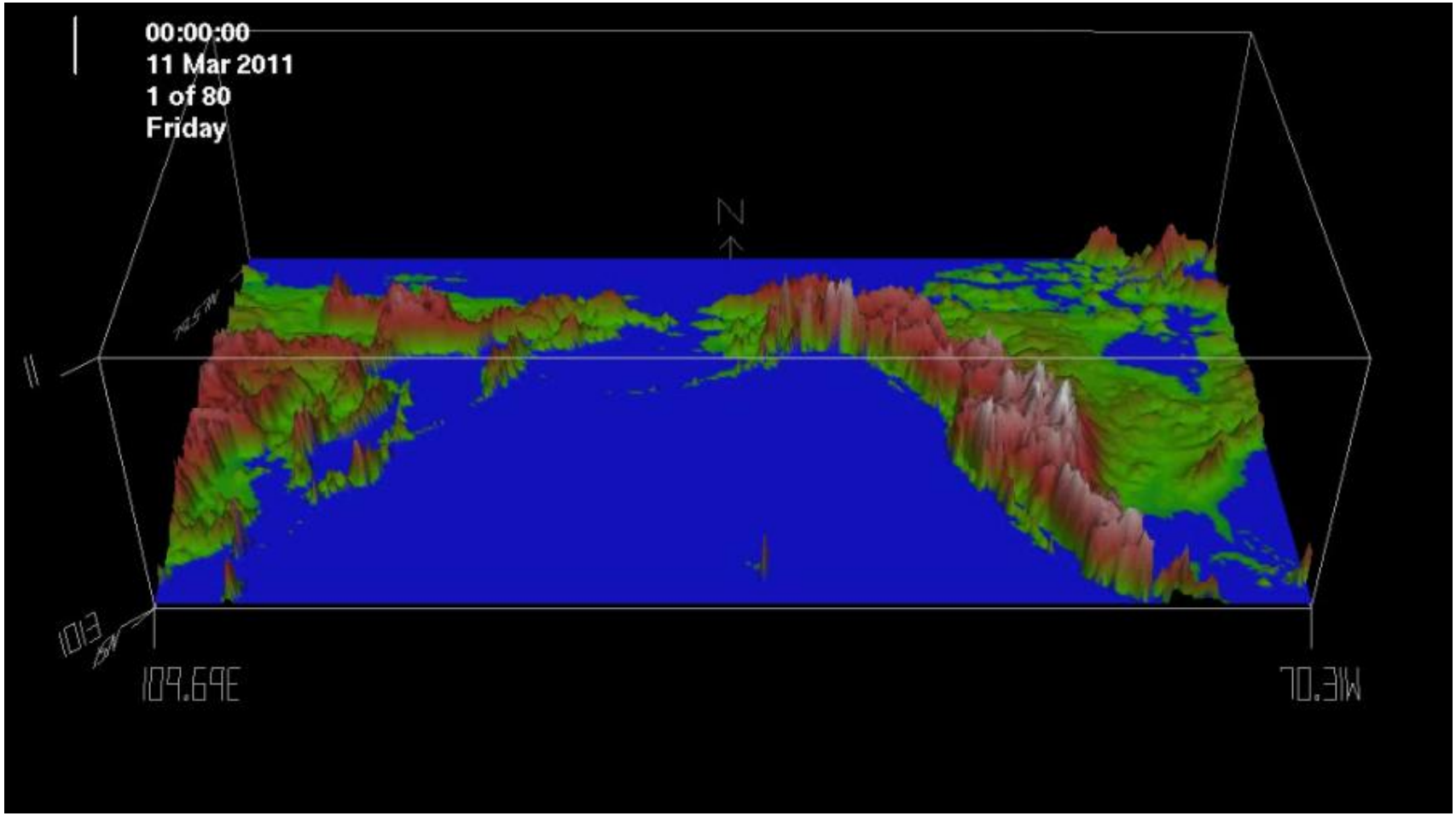


Emission amount (upper) and their uncertainty

Station name	Longitude	Latitude	Num	Data source
Okinawa	127.9	26.5	39	CTBTO
Takasaki	139.0	36.3	38	CTBTO
Wako	139.6	35.8	31	RIKEN
Tsukuba	140.1	36.0	24	NIES
Chiba	140.1	35.7	37	JCAC
Tokai-mura	140.6	36.4	69	JAEA S. Furuta (personal communication, 2011)
Guam	144.9	13.6	36	CTBTO
New Hanover	150.8	−2.6	36	CTBTO
Petropavlovsk	158.8	53.0	40	CTBTO
Wake Island	166.6	19.3	36	CTBTO
Midway Islan	−177.4	28.2	39	CTBTO
Sand Point	−160.5	55.3	37	CTBTO
Oahu	−158.0	21.5	39	CTBTO
Salchaket	−147.1	64.7	39	CTBTO
Vancouver	−123.2	49.2	39	CTBTO
Sacramento	−121.4	38.7	39	CTBTO
Yellowknife	−114.5	62.5	39	CTBTO
Ashland	−99.8	37.2	38	CTBTO
Resolute	−94.9	74.7	37	CTBTO
Melbourne	−80.6	28.2	39	CTBTO
Panama City	−79.5	9.0	39	CTBTO
Charlottesville	−78.4	38.0	39	CTBTO
Ottawa	−75.7	45.4	9	I. Hoffman (personal communication, 2011)
St.John's	−52.7	47.6	39	CTBTO
Iceland	−21.9	64.1	13	Ro5
Reykjavik	−21.8	64.1	38	CTBTO
Caceres	−6.3	39.5	16	Ro5
Orsay	2.2	48.7	19	Ro5
Sola	5.7	58.9	23	Ro5
Schauinsland	7.9	47.9	27	CTBTO
Braunschweig	10.5	53.3	19	Ro5
Osteras	10.6	59.9	22	Ro5
Spitsbergen	15.4	78.2	31	CTBTO
Longyearbyen	15.6	78.2	15	Ro5
Stockholm	17.6	59.2	39	CTBTO
Svanhovd	30.0	69.4	20	Ro5
Dubna	37.3	56.7	39	CTBTO
Kuwait City	47.9	29.3	39	CTBTO
Kirov	49.4	58.6	36	CTBTO
Zalesovo	84.8	53.9	39	CTBTO
Ulan-Bator	106.3	47.9	39	CTBTO
Quezon City	121.4	14.6	39	CTBTO
Ussuriysk	132.0	44.2	38	CTBTO

Available observation sites (43 sites in globe, 5 sites in Japan)

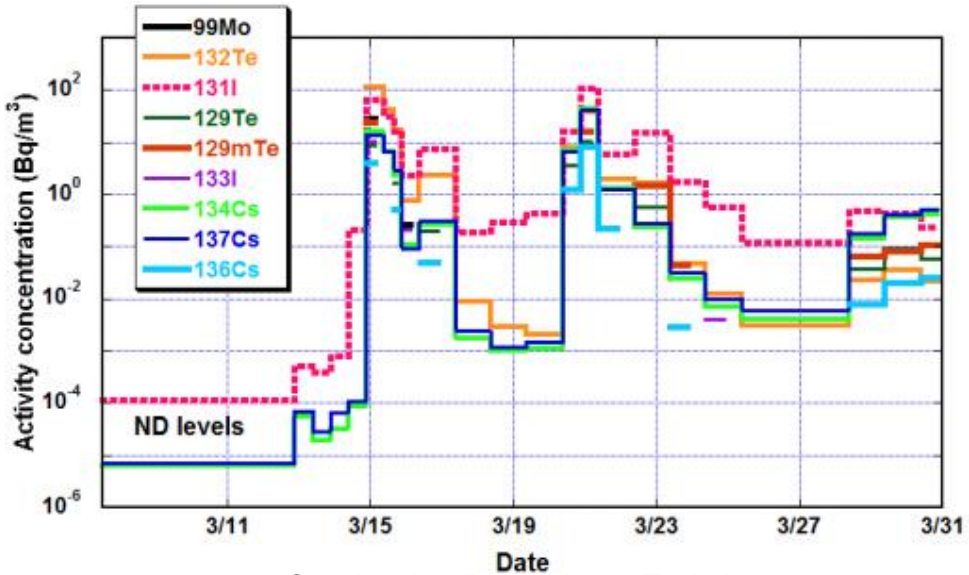
Simulated Cs-137



Comparison of simulated results with observations.

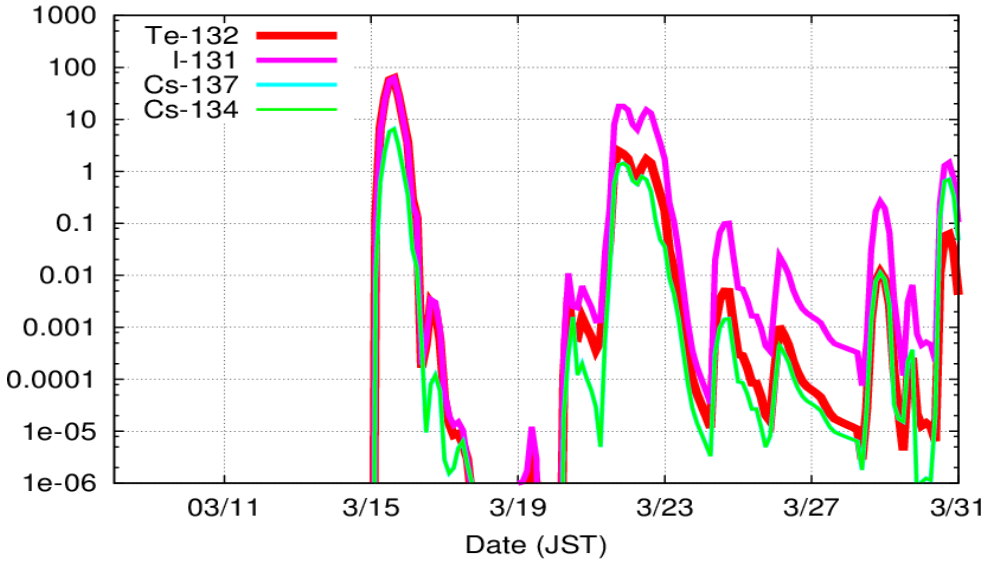
- Currently, we obtained the observed concentrations of radioactive materials from:
  - MRI, Tsukuba (Y. Igarashi)
  - The Comprehensive Nuclear Test-Ban-Treaty Organization (CTBTO)
  - Ring of 5 (Ro5, Masson et al. 2011)
  - Published references (Berkeley, Bowyer *et al.* 2011; Hoffman *et al.* 2011; Hsu *et al.* 2011)

Comparison of observed and simulated time series at Tsukuba, Japan



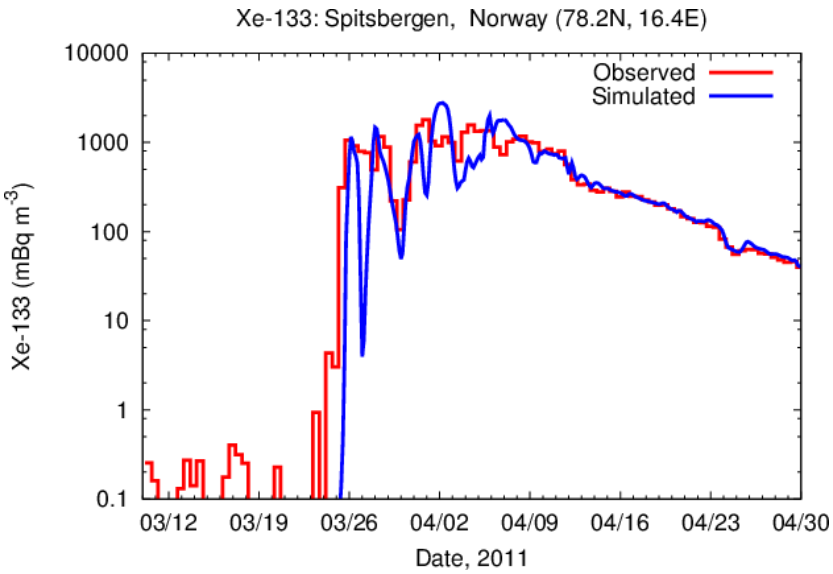
Time series of observed radionuclides on aerosols at Tsukuba, Japan (Igarashi et al., MRI)

Simulated radionuclides at Tsukuba

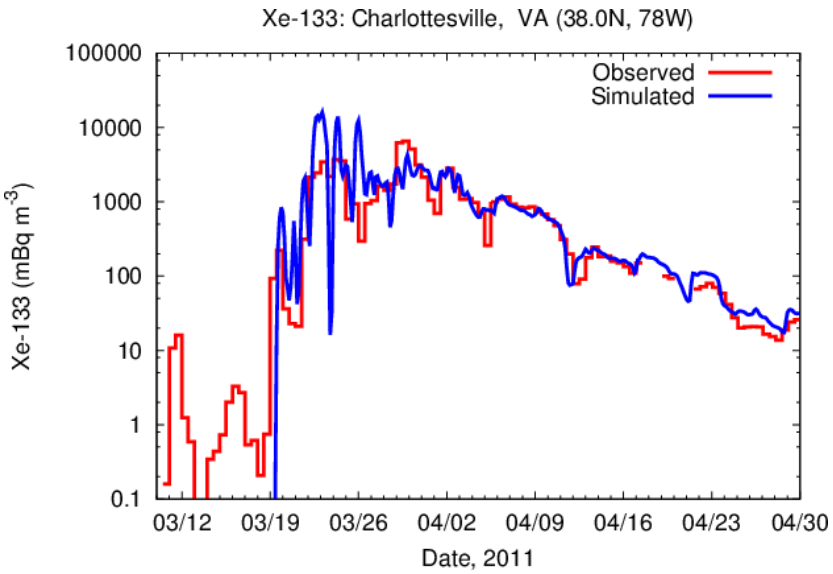


Simulated concentrations of I-131, Cs-137, Cs-134, and Te-132

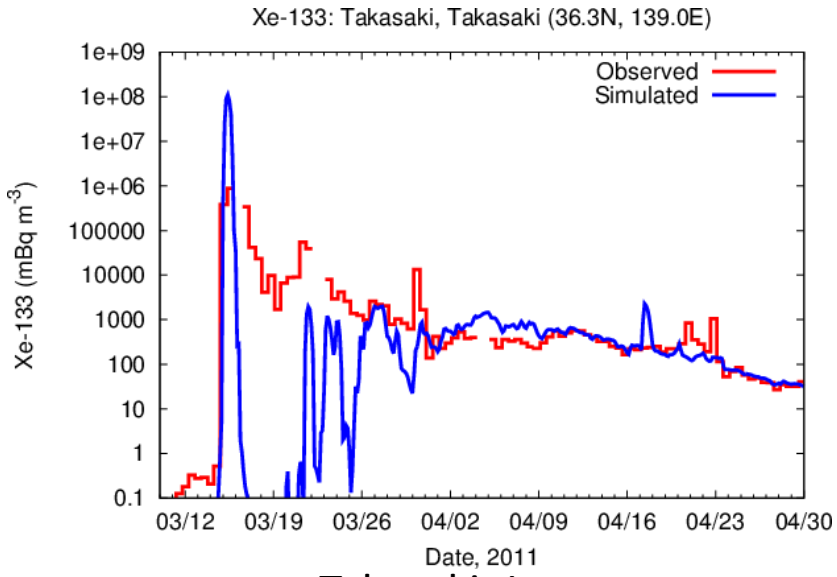
Comparison of observed and simulated ^{133}Xe



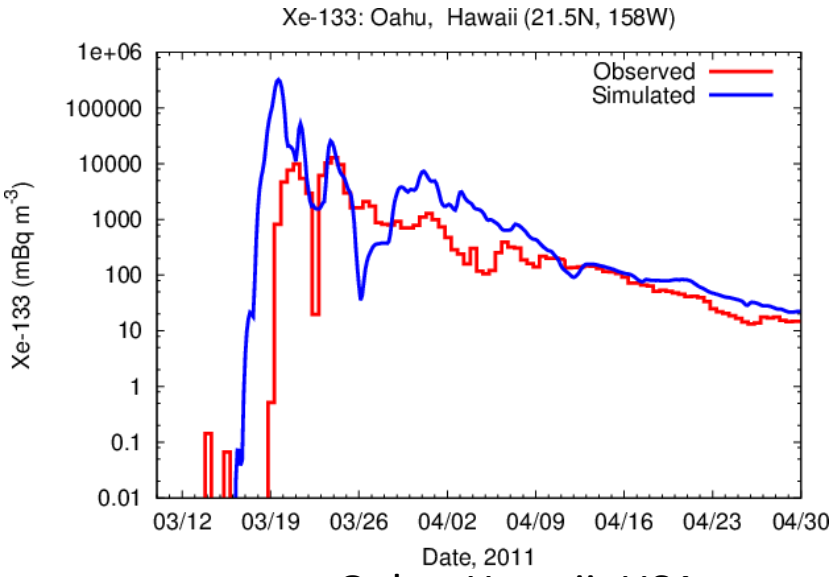
Spitsbergen, Norway



Charlottesville, USA

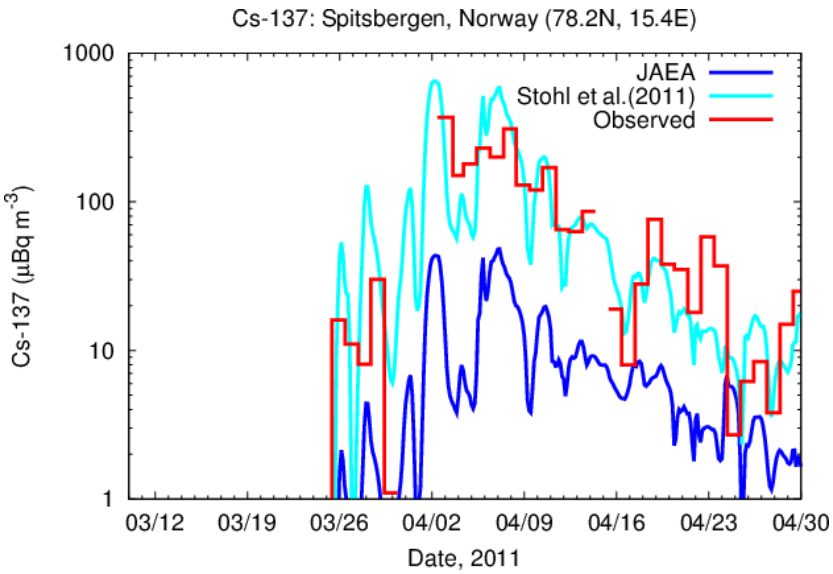


Takasaki, Japan

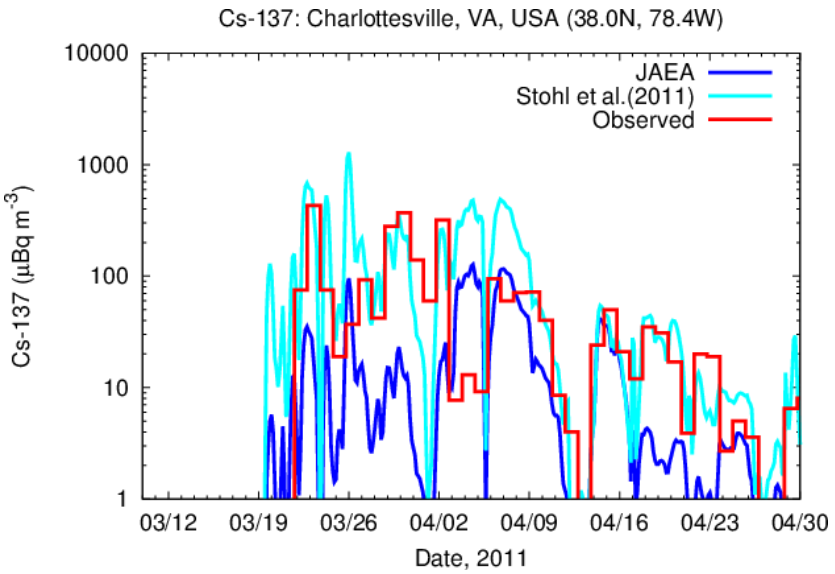


Oahu, Hawaii, USA

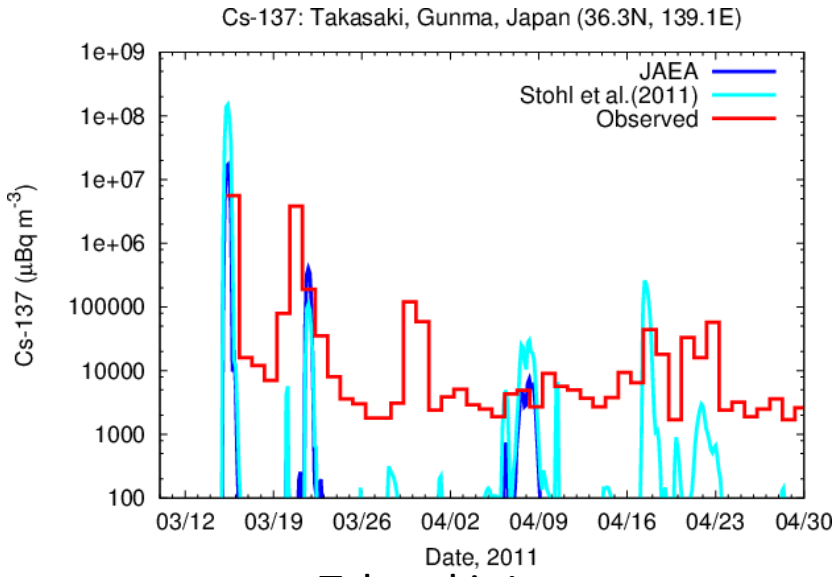
Comparison of observed and simulated ^{137}Cs



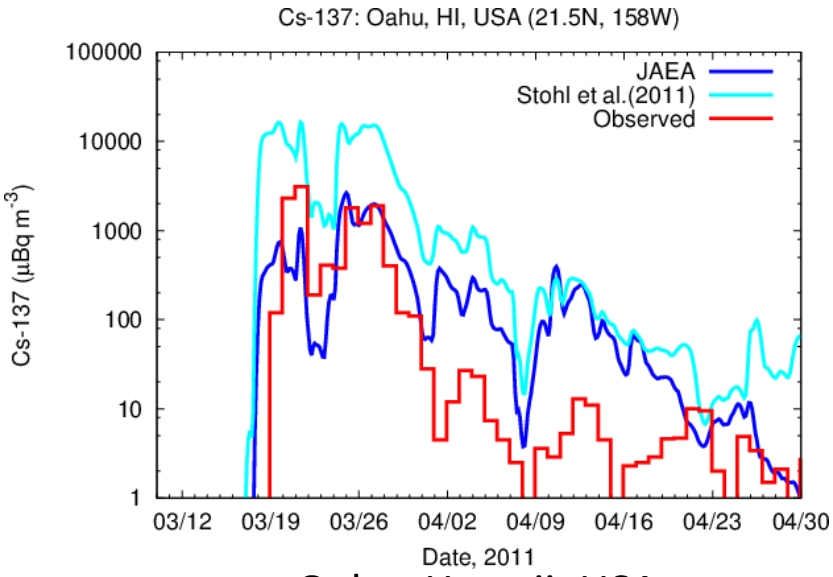
Spitsbergen, Norway



Charlottesville, USA



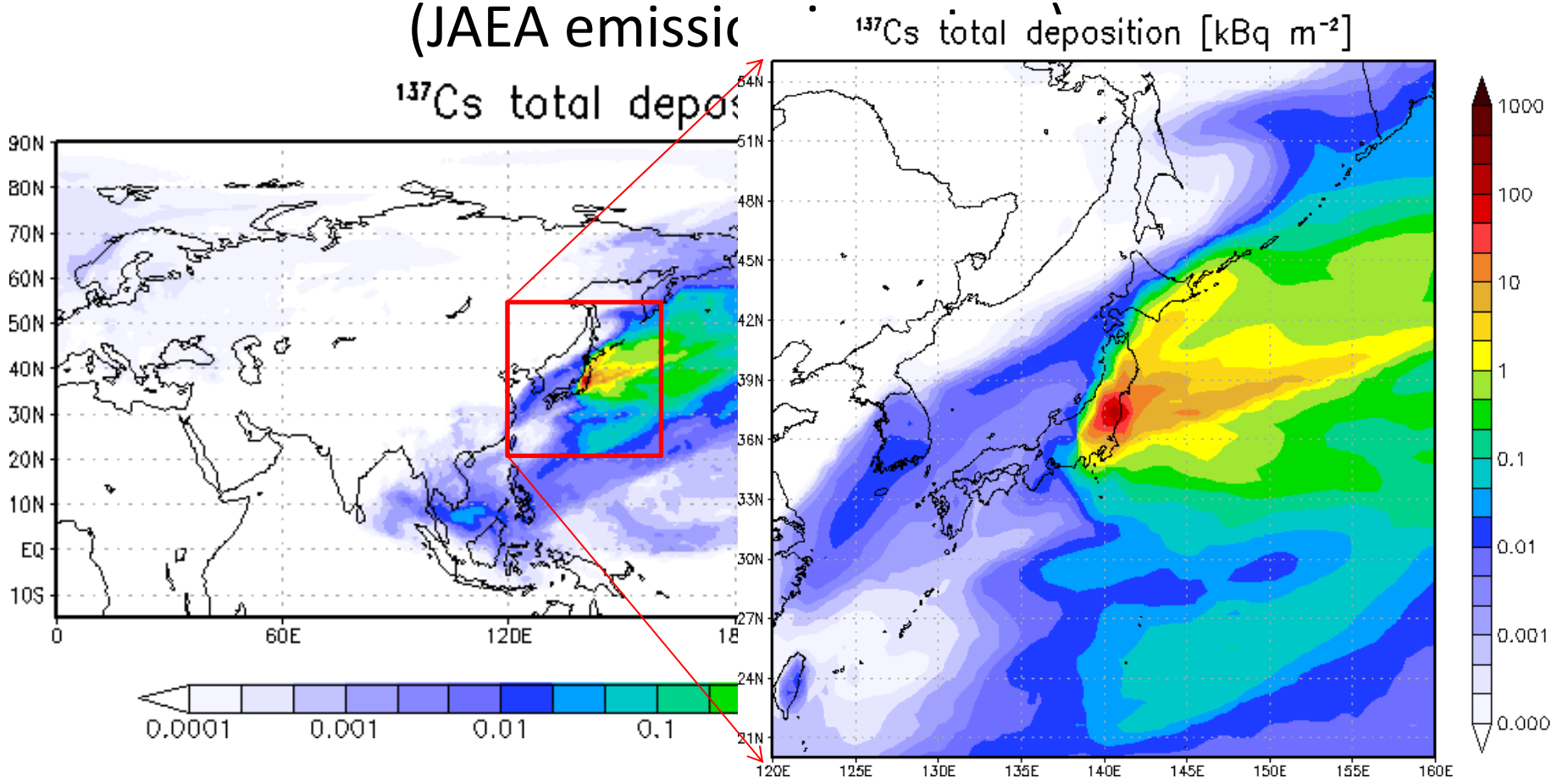
Takasaki, Japan



Oahu, Hawaii, USA

Total deposition of ^{137}Cs

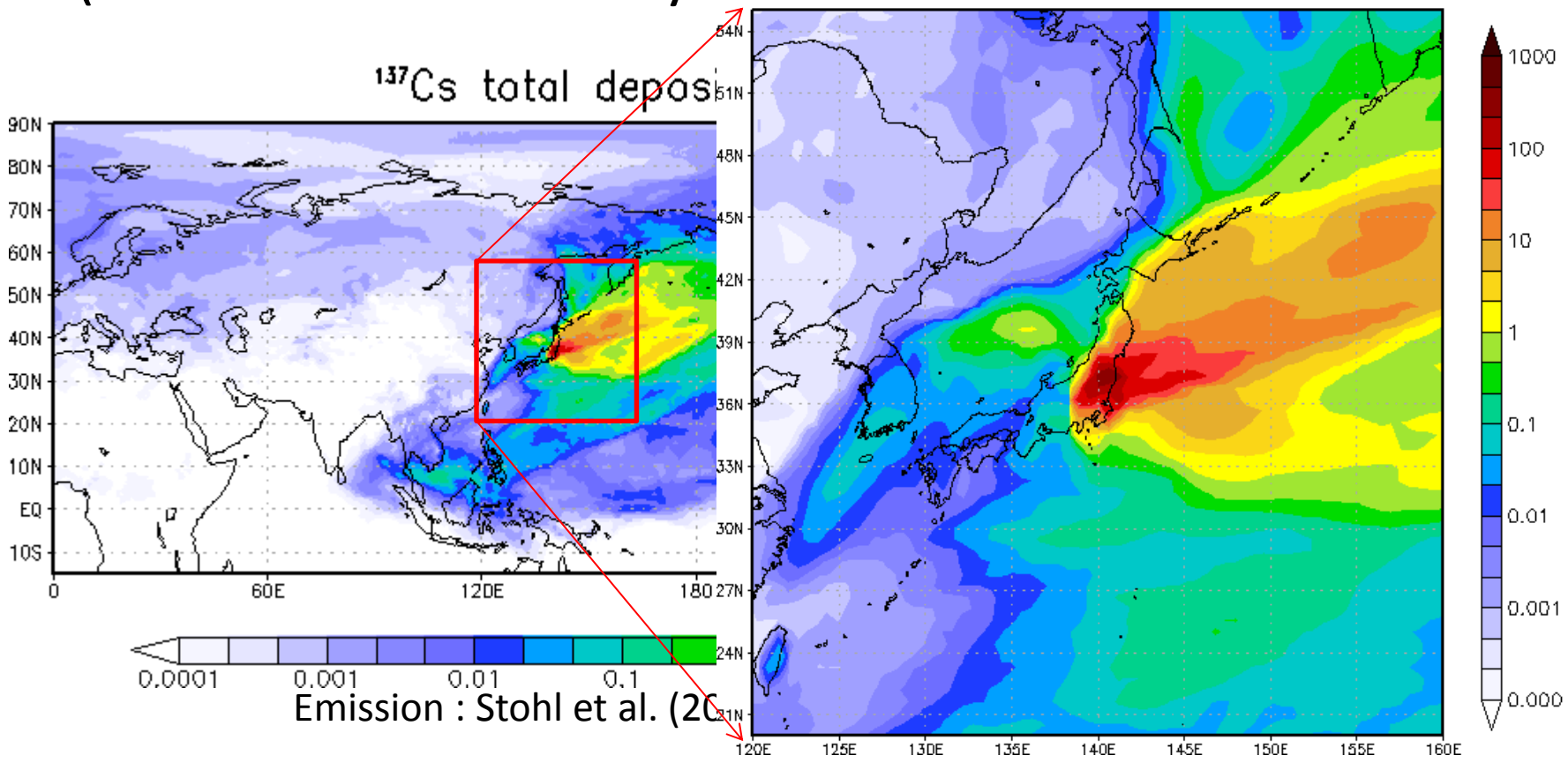
(JAEA emissi



- Deposited over land: 29.9% (2.0 PBq)
- Deposited over ocean: 70.1% (4.8 PBq)
  - ➢ Most of the ^{137}Cs is deposited over Northern Pacific

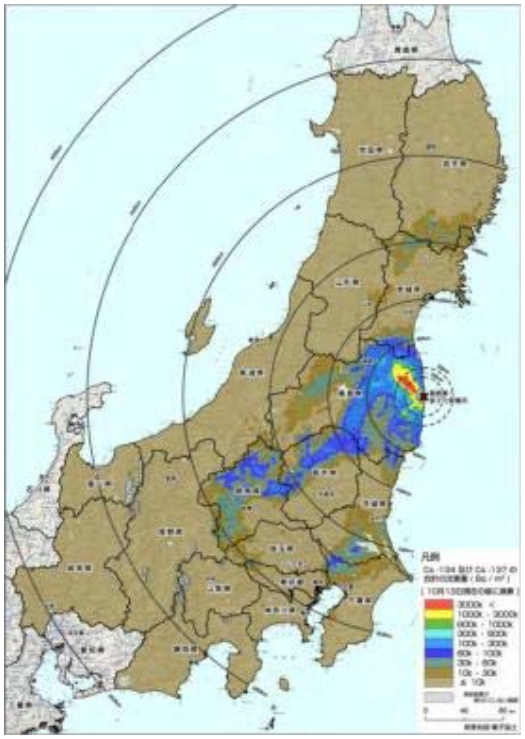
Total deposition of ^{137}Cs

(Emission estimated by S[illegible] 2011, ACPD)

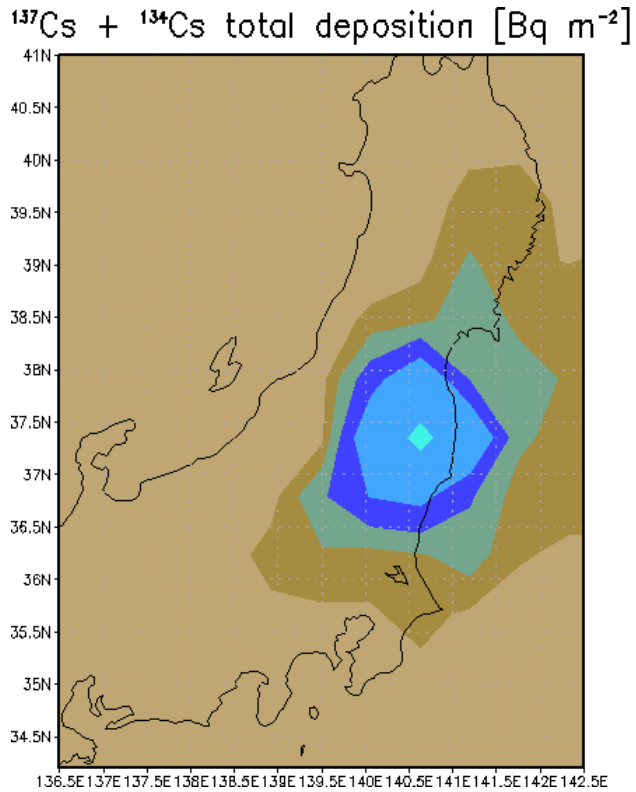


- Deposited over land: 17.6% (6.1 PBq)
- Deposited over ocean: 82.4% (28.8 PBq)

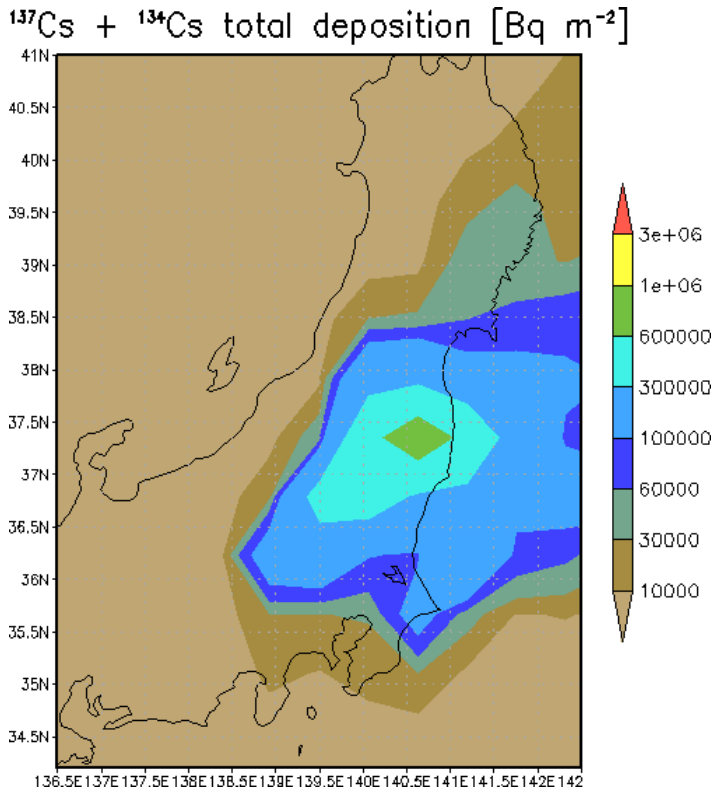
Comparison of ^{134}Cs and ^{137}Cs total deposition with aircraft observation



Observed ^{134}Cs+^{137}Cs deposition by aircraft measurement (MEXT, 11 Nov. 2011)



Simulated deposition with JAEA emission inventory



Simulated deposition with emission estimated by Stohl et al. (2011, ACPD)

The emission flux estimation system by inverse model

1. Concept of inverse model
2. Estimation of dust flux emission
3. Application to an accident of Fukushima-dai-ichi nuclear power plant

Principal Investigator: Takashi Maki

The concept of Inversion

Aim

To estimate initial nuclei emission flux by inverse model

Assumption

Observation ($\mathbf{y}$) is determined by linear combination of emission flux ($\mathbf{x}$) and Transport model processes ($\mathbf{A}$).

Features

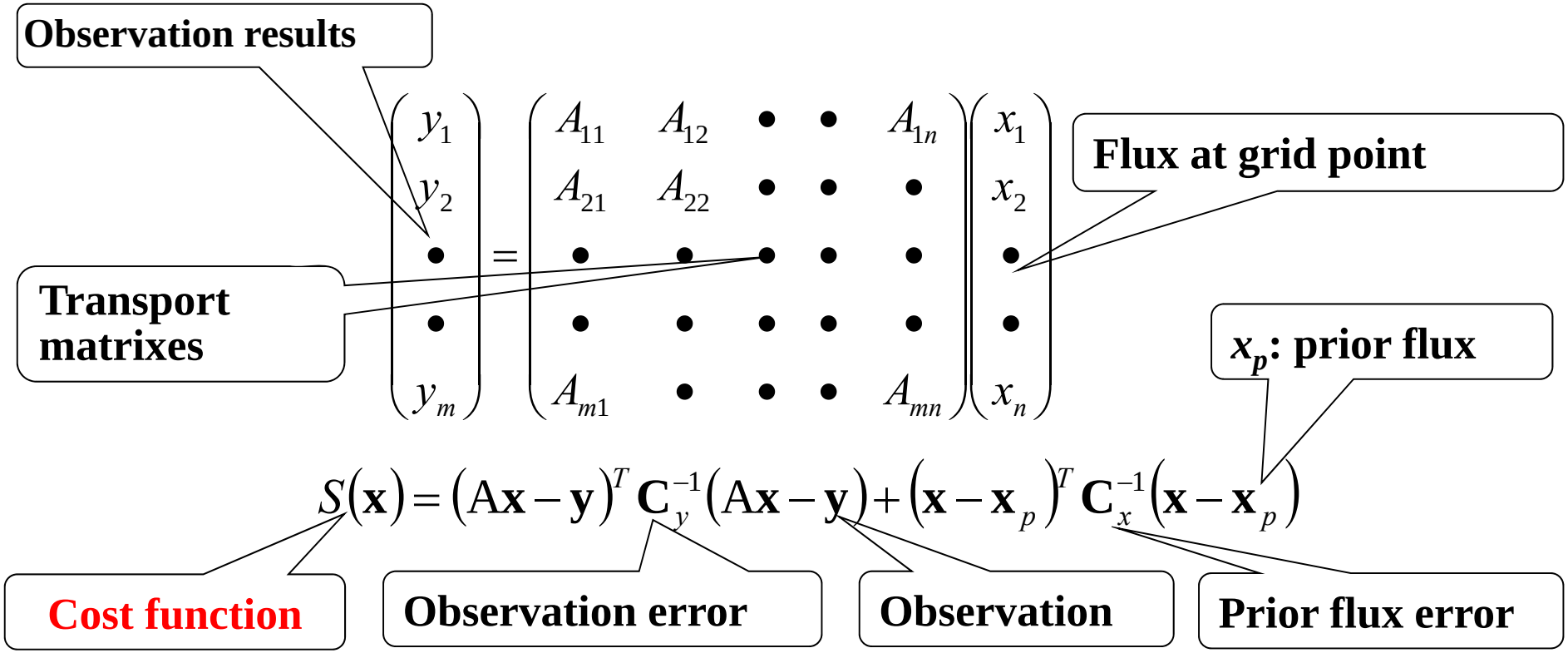
We obtain initial emission flux not only by observation ($\mathbf{y}$) and transport model ($\mathbf{A}$) but also prior information ($\mathbf{x}_p$).

The precision of the estimation depends on a quality of prior information.

$$S(\mathbf{x}) = (\mathbf{Ax} - \mathbf{y})^{\mathrm{T}} \mathbf{C}_y^{-1} (\mathbf{Ax} - \mathbf{y}) + (\mathbf{x} - \mathbf{x}_p)^{\mathrm{T}} \mathbf{C}_x^{-1} (\mathbf{x} - \mathbf{x}_p)$$

We obtain $\mathbf{X}$ which minimizes the cost function $S(\mathbf{x})$.

The Bayesian Synthesis Inversion



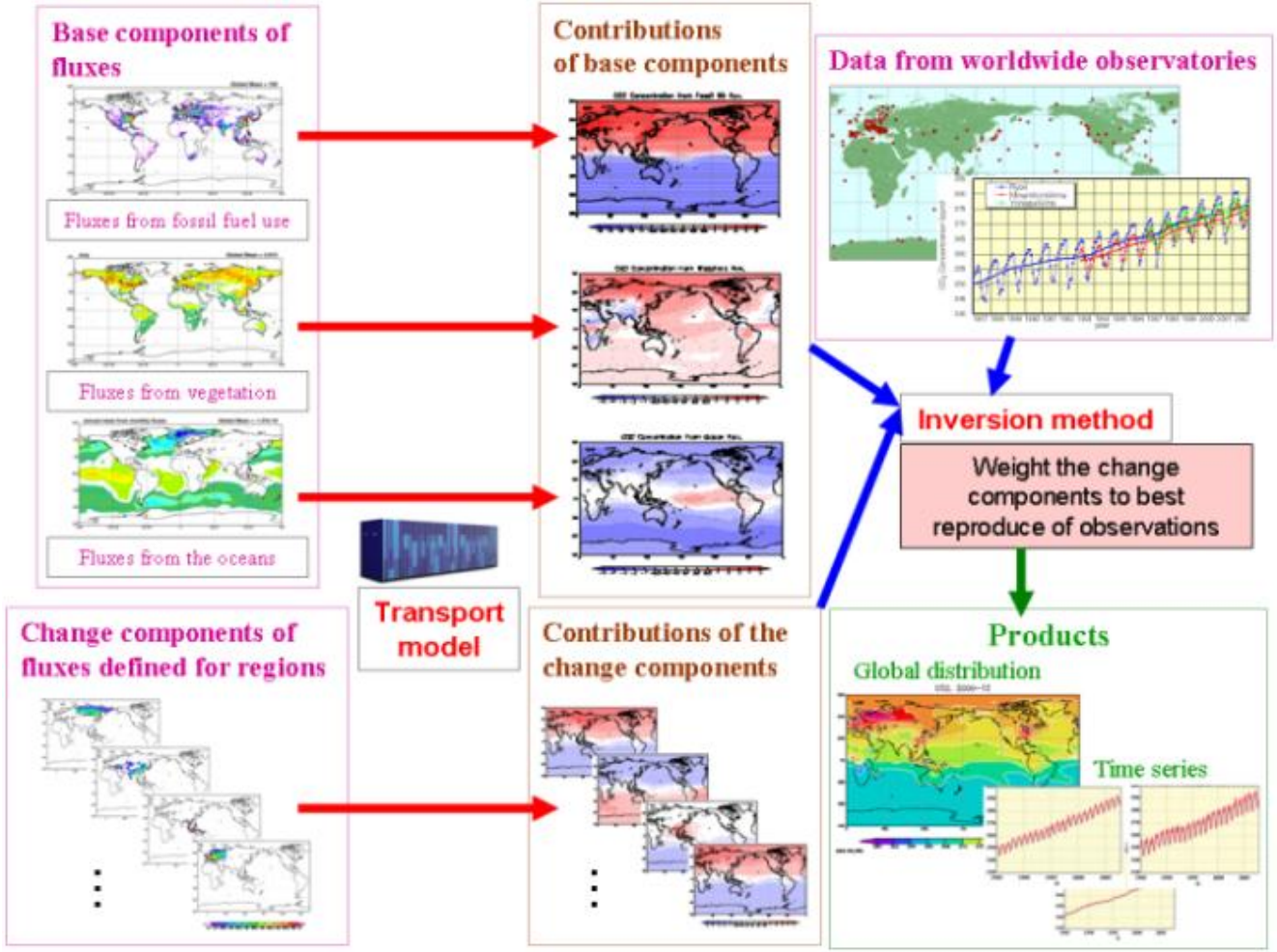
$(\mathbf{Ax}-\mathbf{y})^T \mathbf{C}_y^{-1}(\mathbf{Ax}-\mathbf{y})$ Shows the difference between model and observations

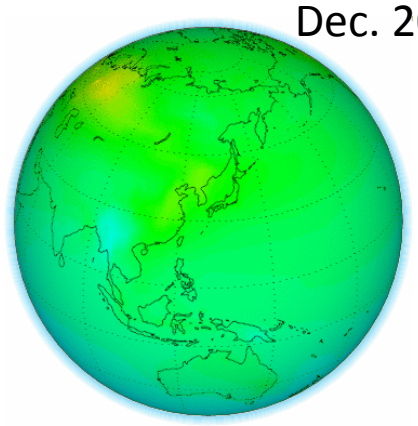
$(\mathbf{x}-\mathbf{x}_p)^T \mathbf{C}_x^{-1}(\mathbf{x}-\mathbf{x}_p)$ Shows the difference between results and prior information

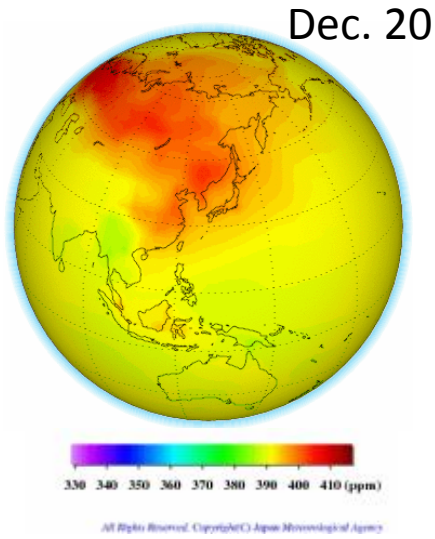
↓

We determine flux x which minimize cost function *S*(x).

CO_2 flux analysis by inversion



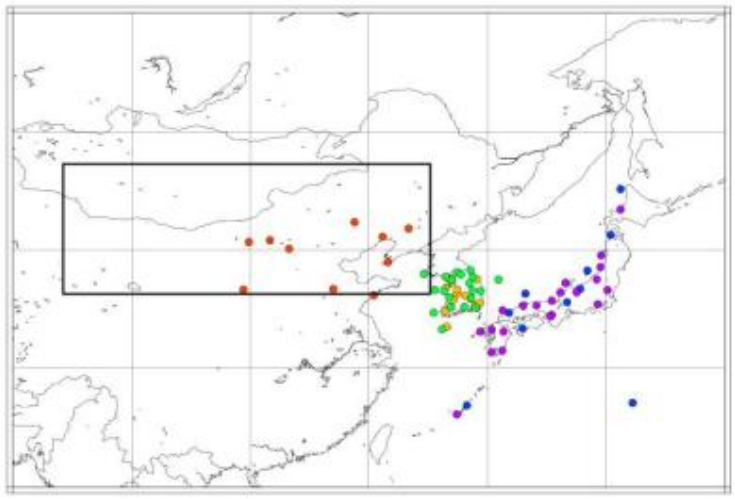




Greenhouse gas observations are made at about 150 stations worldwide. Global distributions of greenhouse gases are obtained from such limited observations using numerical models that simulate atmospheric transport and estimate CO_2 fluxes inversely from observations (Maki et al., 2010).

http://ds.data.jma.go.jp/ghg/kanshi/info_kanshi_e.html

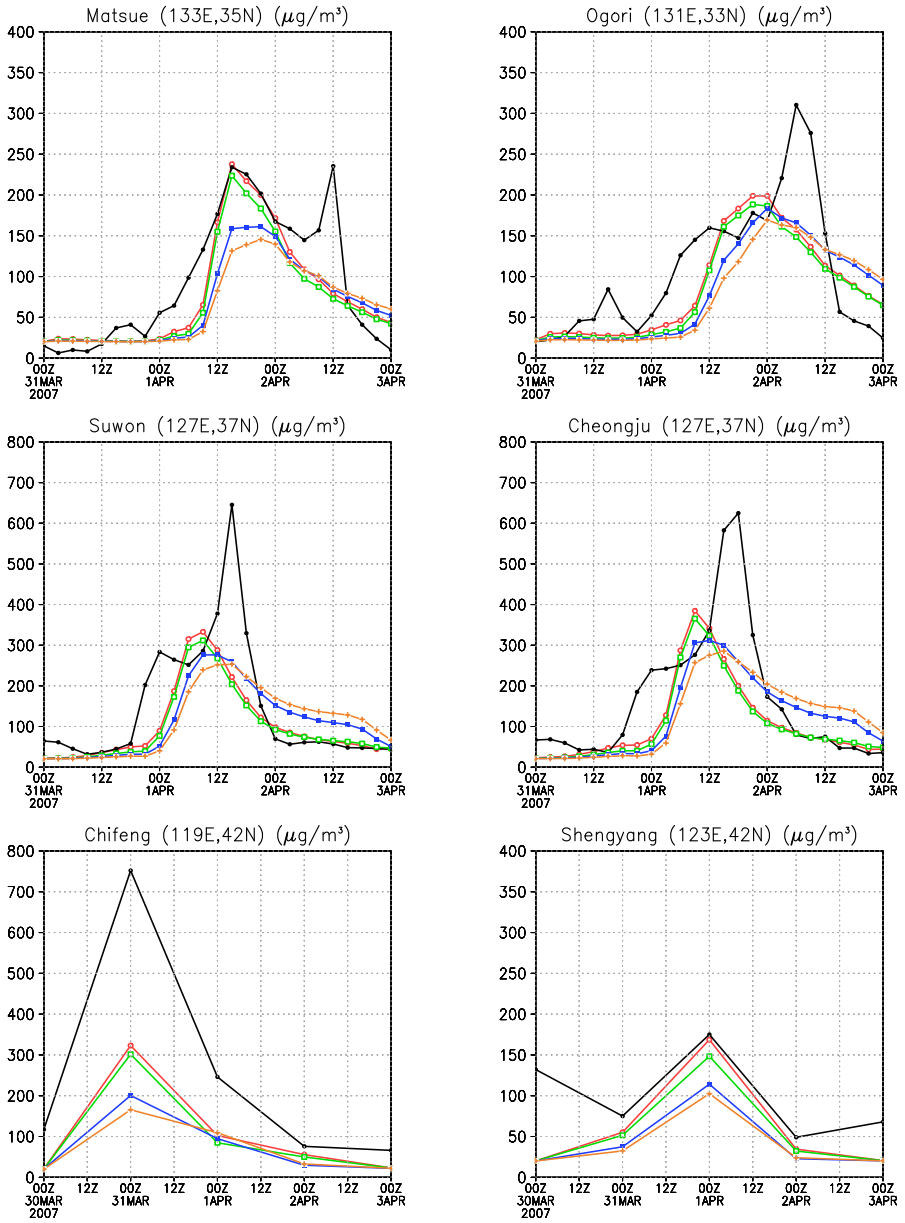
Dust emission analysis by inversion



Observation network of PM10

We could obtain more realistic dust concentration time series by inverse modeling.

We need more observation data near dust source area (Maki et al., 2011).



Dust Flux estimation results

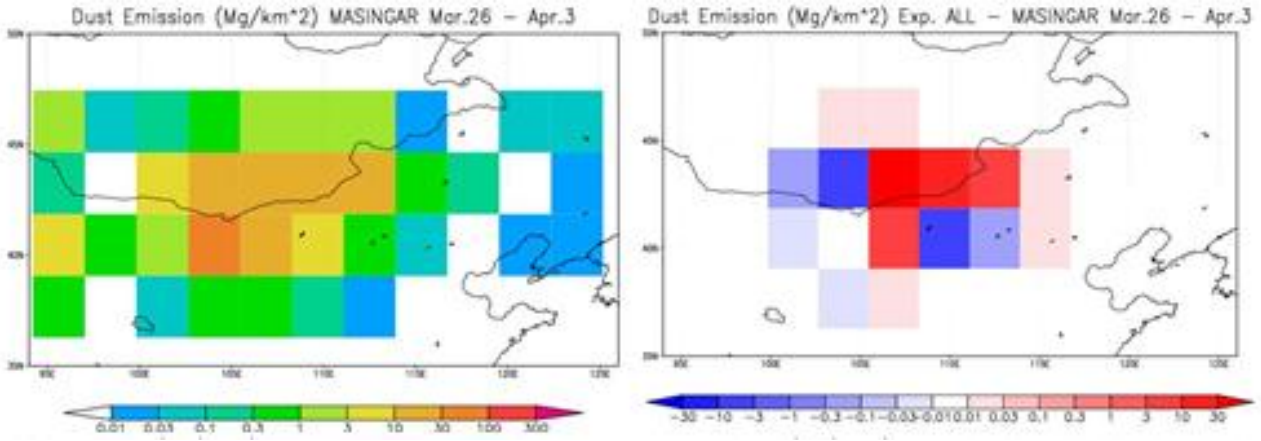


Table 2. Total estimated flux around the dust source area during the analysis period (26 Mar. 2007 – 3 Apr. 2007). Unit is Tg.					
Prior flux uncertainty	Exp. ADORC	Exp. KOREA	Exp. JK	Exp. ALL	**MASINGAR**
0.2 (20%)	20.4	23.6	23.8	23.4	**19.5**
0.5 (50%)	21.6	32.3	30.9	32.6	
1.0 (100%)	22.0	42.2	37.1	42.7	

*** We assume the dust source area to be a box region (Fig. 1) containing 44 grid cells in MASINGAR from 94 degrees east to 125 degrees east and from 36 degrees north to 47 degrees north. Exp. JK**

The estimated total dust flux amount are sensitive to observational data and their uncertainty.

We need more observation data near dust source area (Maki et al., 2011).

Application to Fukushima accident

- Huge amount of radio nuclei were emitted from Fukushima Dai-ichi nuclear power plant.
- Some institute forecasted spread of radio nuclei aerosol, but these information were not used to evacuation activity efficiently.

Fukushima power plant (TEPCO HP)

One of the main reason is that we could not know a amount, time-series of radio nuclei emission.

This hazardous accident has a feature that horizontal source area is known. We should use such information for emission estimation.

The feature of this research

	JAEA	NILU	MRI
Number of observation	17 (17)	43 (5)	49 (49)
Transport model	WSPEEDI	FLEXPART	MRI-PM/r MASINGAR mk-2
Model type	Lagrange	Lagrange	Euler
Meteorological Input	JMA GSM (0.25 x 0.2 deg)	ECMWF (0.18 deg) #GFS (0.5 deg)	MANAL (0.05 deg) JCDAS (1.25 deg)
Flux estimation	Peak comparison	Inversion	Inversion
Time resolution	variable	hours	6(3) hours output Daily

in second row shows observations in Japan mainland.

We consider it possible to analyze detailed estimation flux by using Japanese observation.

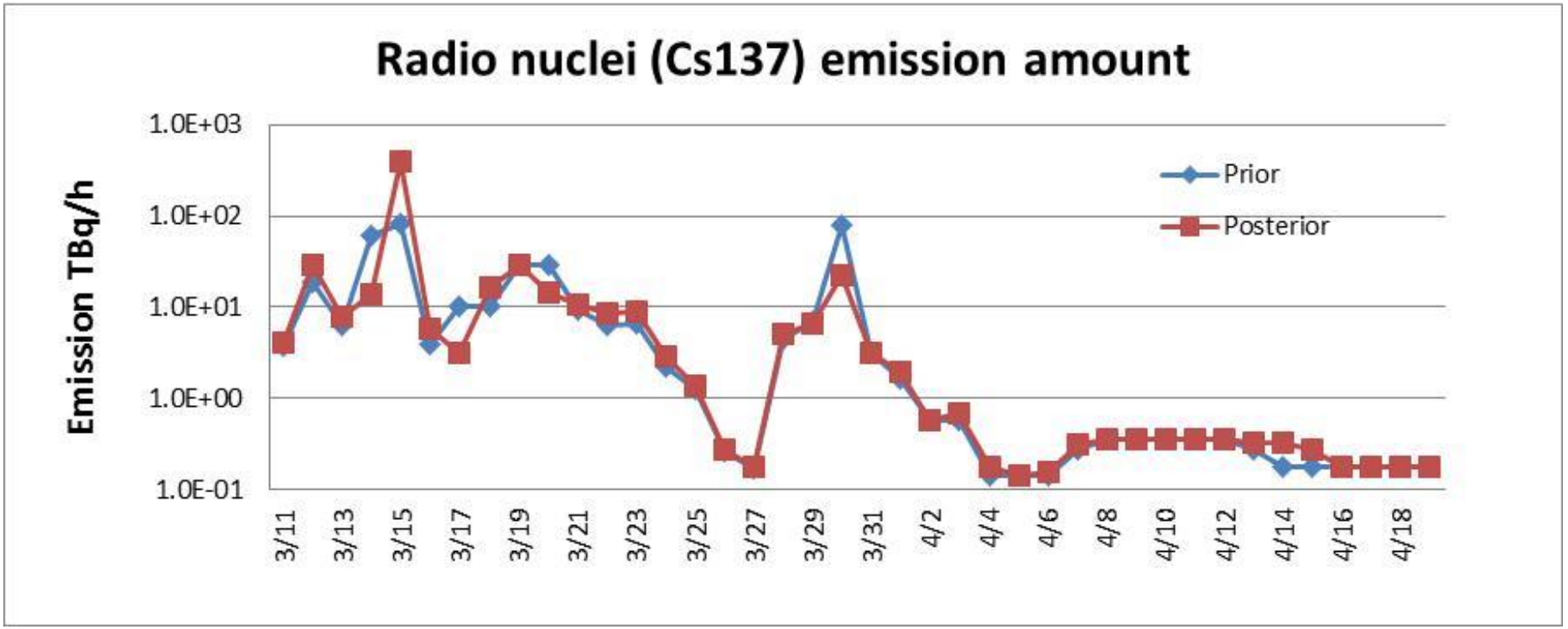
Preliminary result of our Inverse analysis with the global model

Observation: 50 sites observation data from CTBT, Ro5 and so on.

Model: MASINGAR-mk2 (Tanaka et al.)

Prior Information: Estimation flux by Chino et al. (JAEA)

Total Cs-137 flux: 9 PBq ➔ 13 PBq (↑increase)

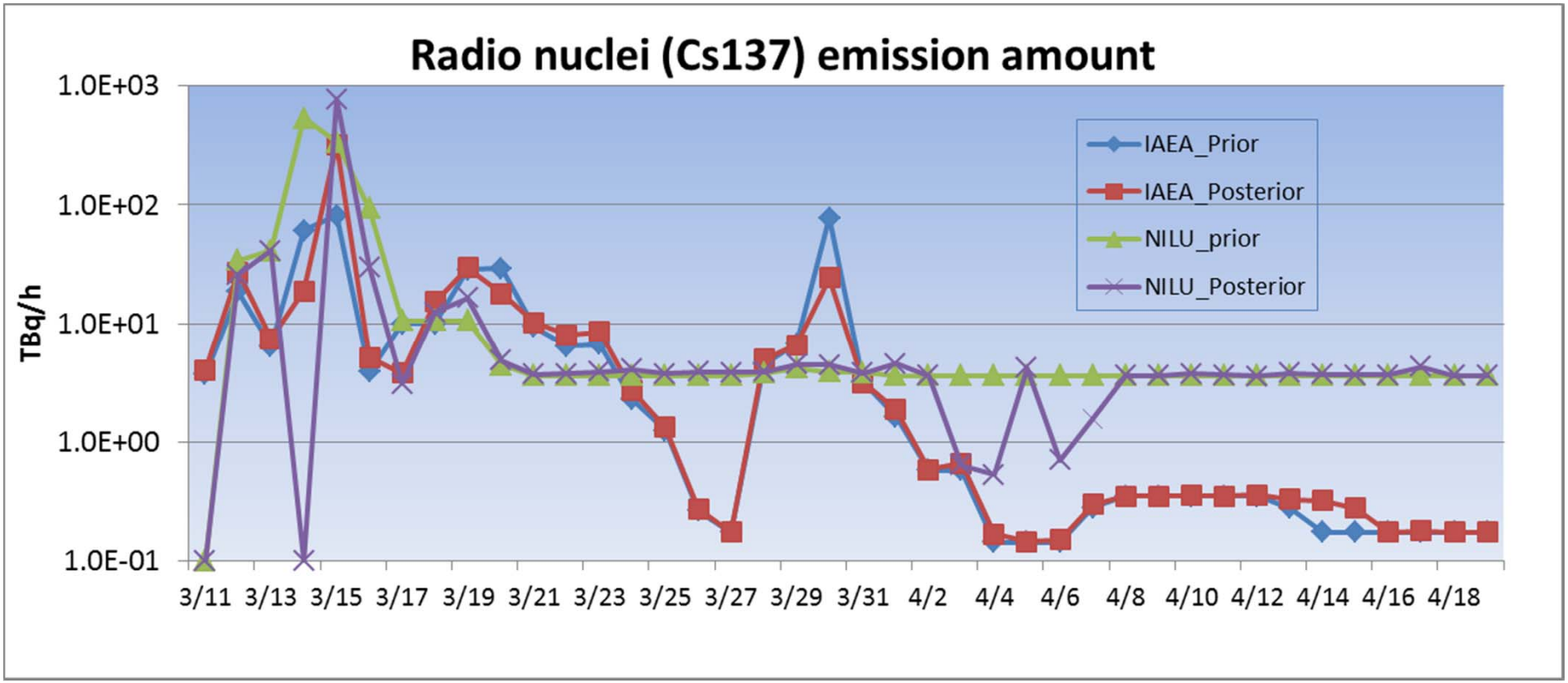


Preliminary estimated emission fluxes.

Differences using other first guess emission

- Here, we also used estimation flux by Stohl et al. (2012) for the inverse analysis.

Total Cs-137 flux: 29 PBq ➔ 24 PBq (🡻 decrease)



Preliminary estimated emission fluxes.

In our inversion analysis,

- Estimated emission amount from Hydrogen explosion of the Unit 3 on 14 March was increased, with both first guesses.
- Both first guesses underestimated the release from the Unit 3 explosion?
- We should examine the height of the explosion.

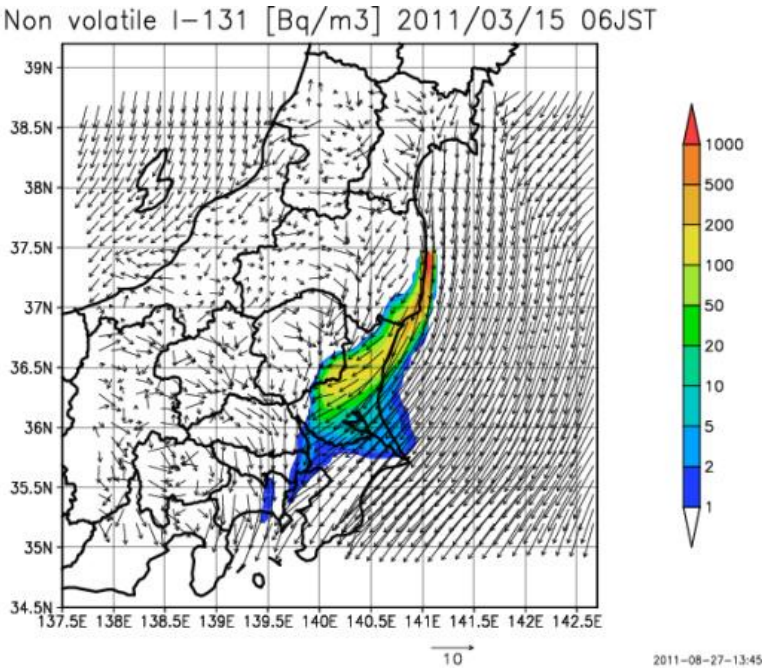
Explosion of the Unit 3 of Fukushima NPP1. (from TV news)

Summary of inversion analysis with our global aerosol model

- Preliminary results of our inversion analyses adjusted the emission flux of ^{137}Cs to 13–24 PBq, which are between JAEA (9.1 PBq) and NILU (36.6 PBq, Stohl et al. 2012).
- The estimated emission flux is highly sensitive to the selection of the first guess, because of the under-constraint of observations.

To reduce uncertainty,

- We should further investigate for the sensitivities in microphysical and deposition processes of aerosols, and use of observed deposition fluxes of ^{137}Cs.
  - Ex. Aerosol size distribution:
    - Stohl et al. (2012) : Lognormal: 0.4μm (aerodynamic mean), σ = 0.3
    - Takemura *et al.* (2011): 10μm



Simulation and estimation with a regional transport model

Questions

- In this model, we are trying to formulate the behavior of atmospheric radioactive materials in detail, and examine which processes are important.

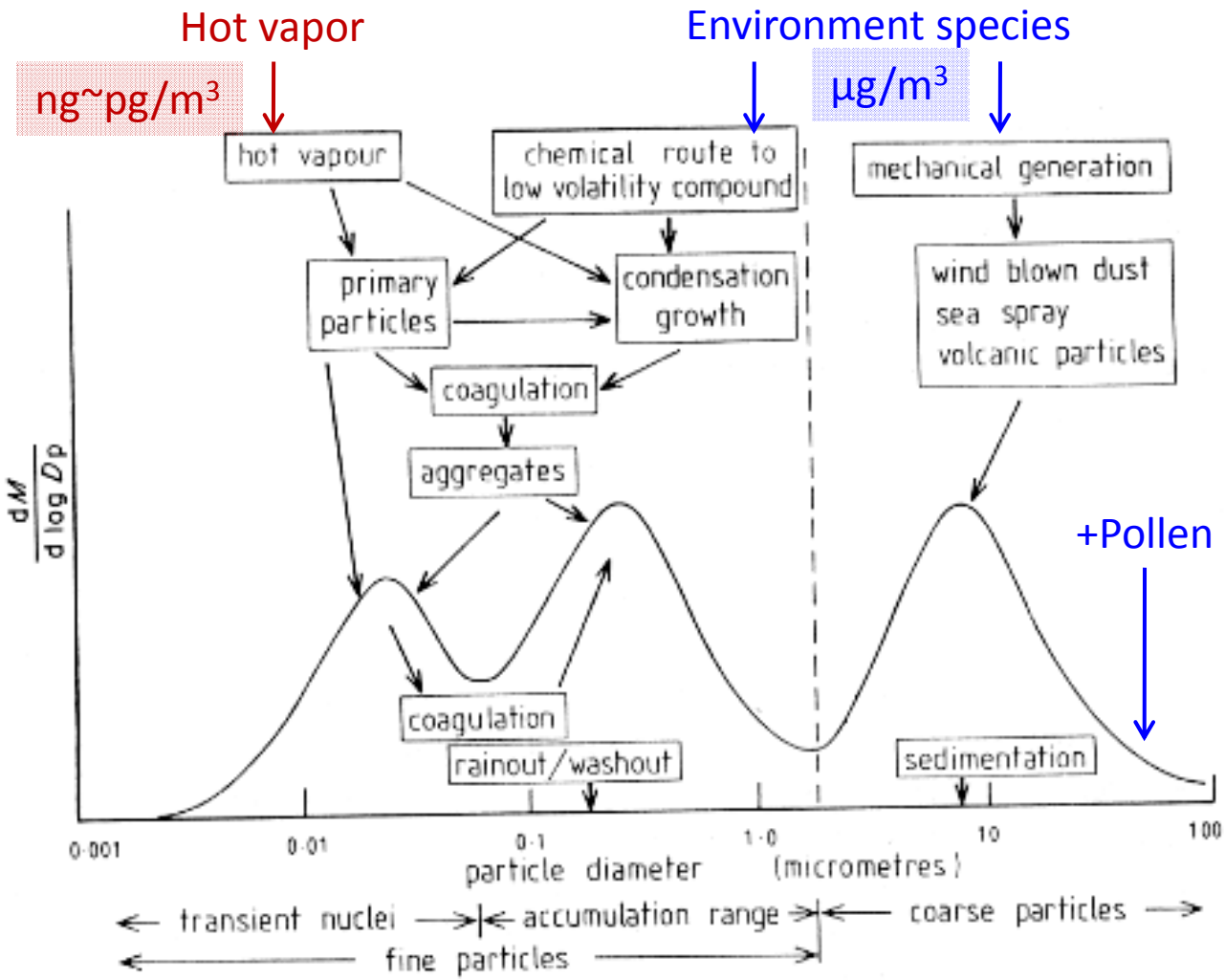
- To reproduce the deposition flux of radioactive materials, we have to reproduce the precipitation amount and other processes in
  - Dry deposition
  - Wet deposition by rain
  - Wet deposition by snow

- How do the chemical composition, size distribution, and surface conditions affect the simulated results?

Behavior of the radioactive materials in the atmosphere

^{131}I: 10^{15}Bq/h
↓
1g/h
($10^6 m^3$)
↓
1μg/m^3/h

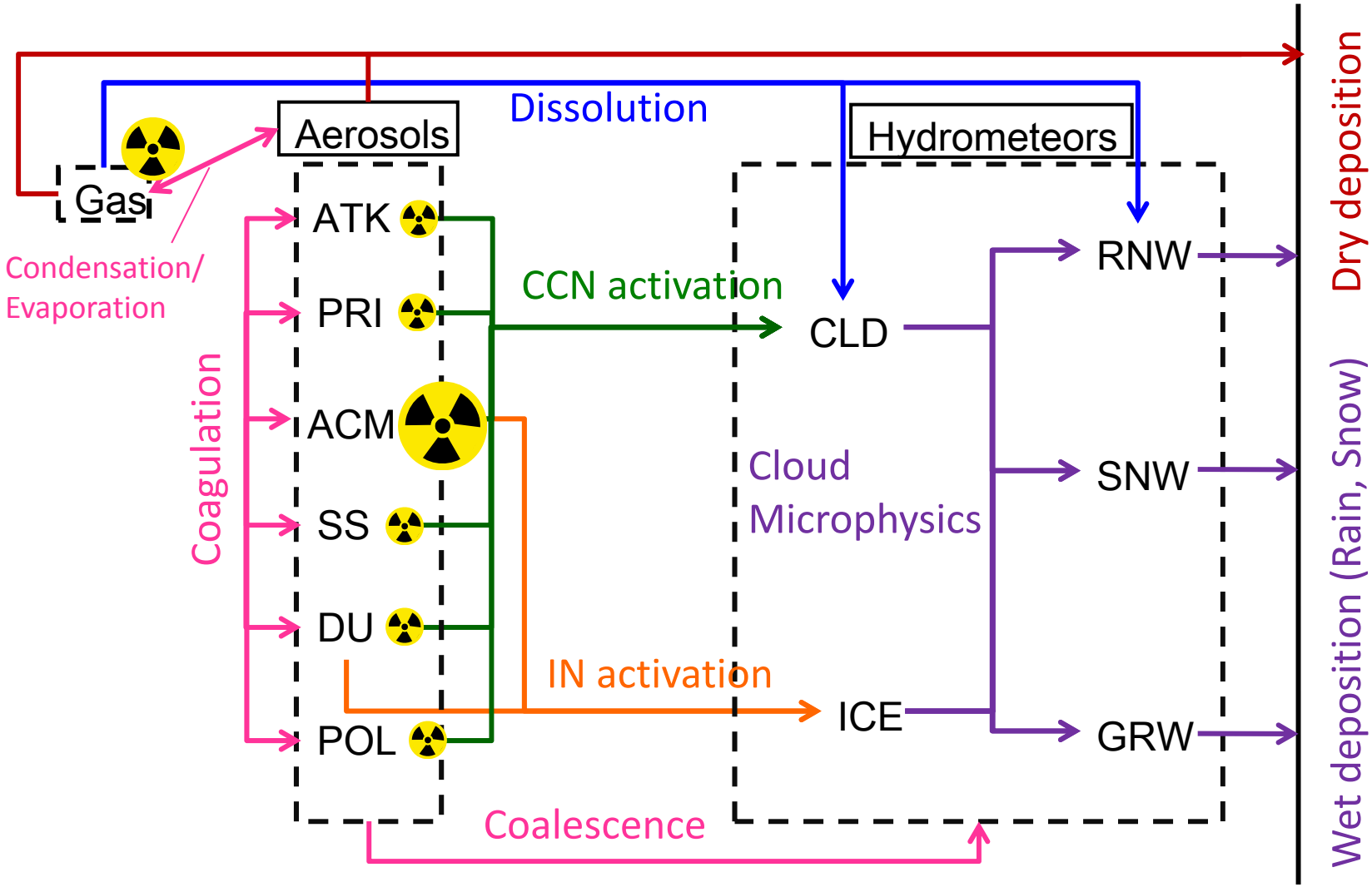
100 Bq/m^3
↓
1 pg/m^3
↓
(D=100nm)
↓
10^3particles /m^3



From SCOPE50, review paper of Chernobyl accident, Warner and Harrison (1993)

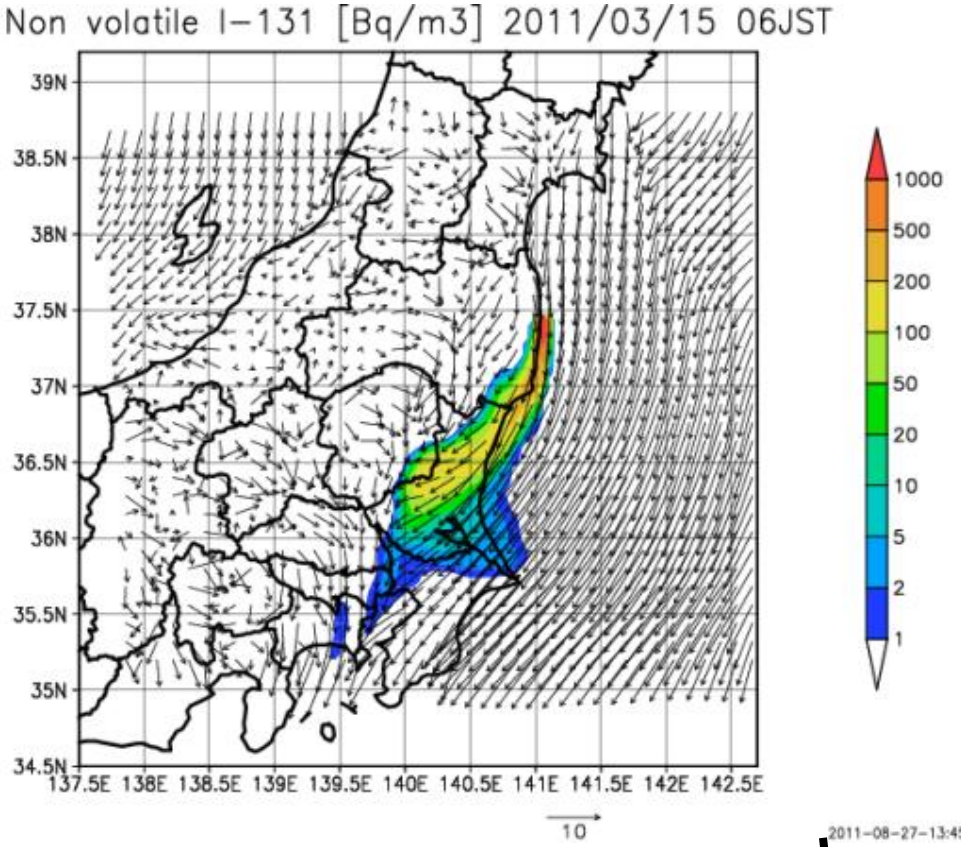
MANAL-NHM-PM/r (Δx=4km)

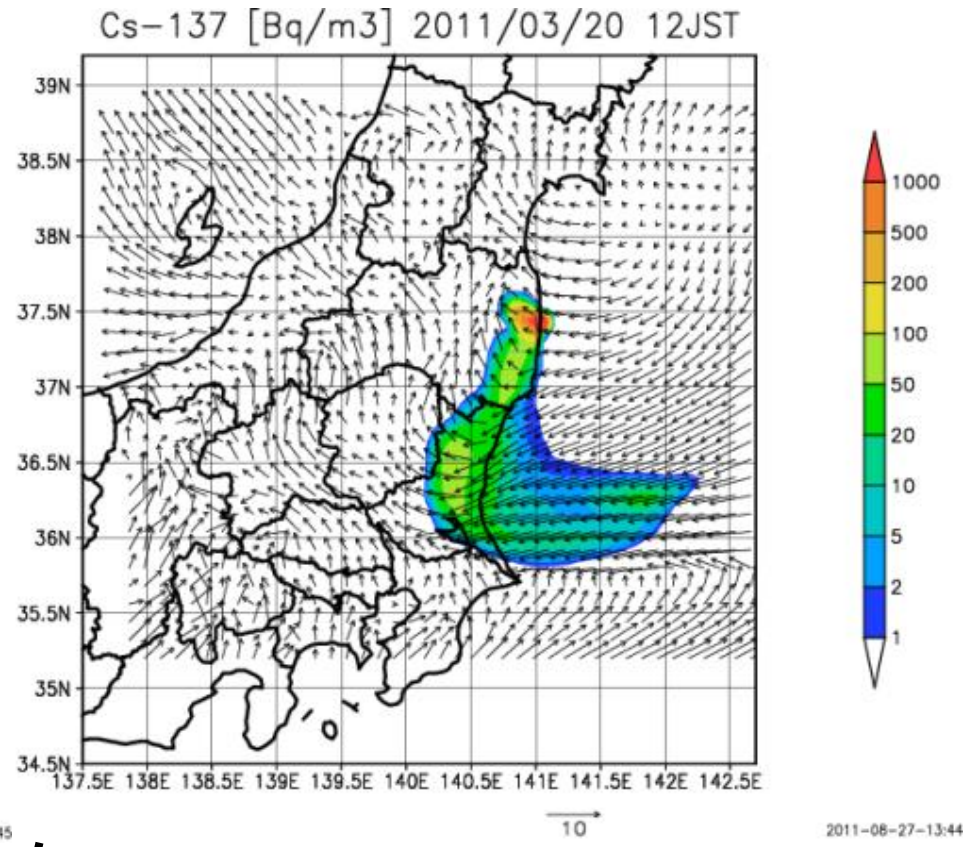
(Passive-tracers Model for radioactivity, off-line coupled with NHM)



Sub-micron (~0.1μm)	ATK (Aitken), PRI (primary nuclei), ACM (accumulation mode)
Super-micron (~1μm)	DU (soil particle), SS (Sea salt particle)
Super-large (~10μm)	POL (pollen)

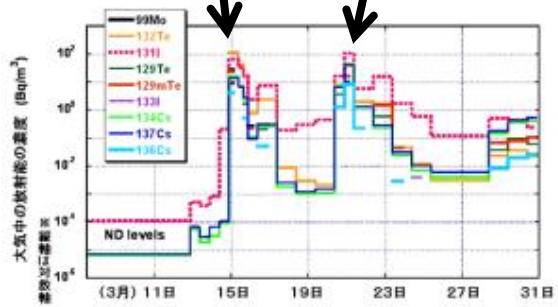
Simulated distributions of radioactive materials in aerosol for 15 and 20 March



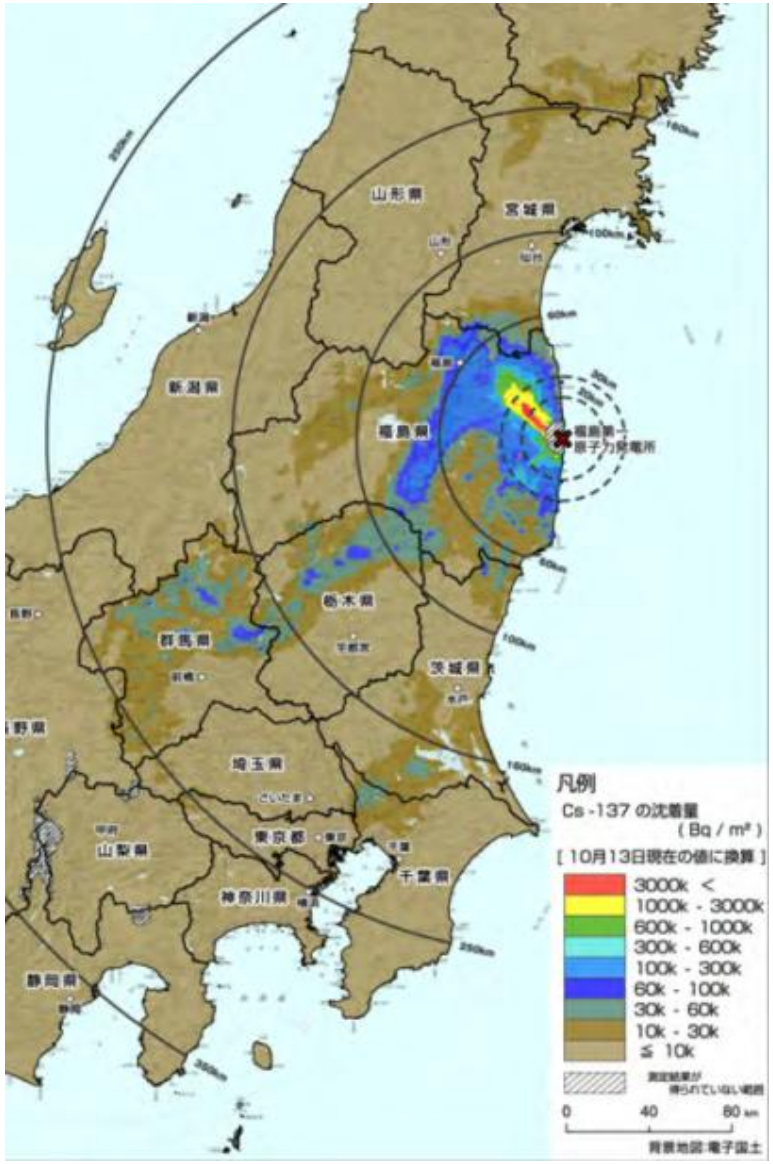


Observed I-131 in Tsukuba: 70Bq/m^3

Observed Cs-137 in Tsukuba: 40Bq/m^3

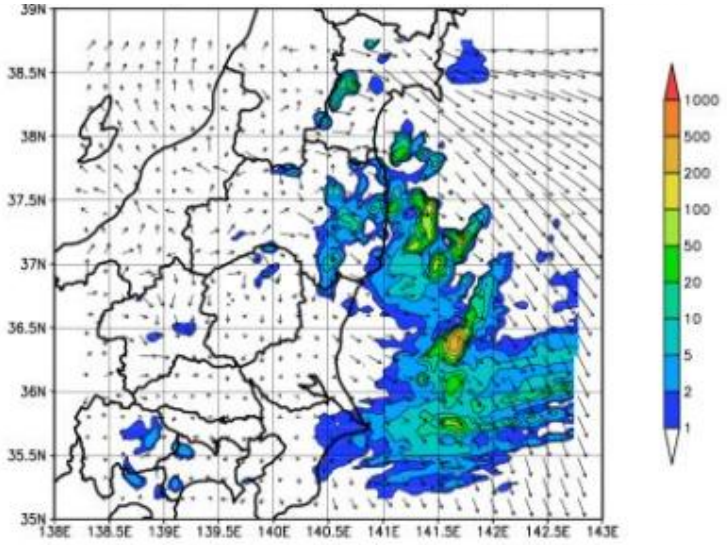


Uncertainties in wet removal processes

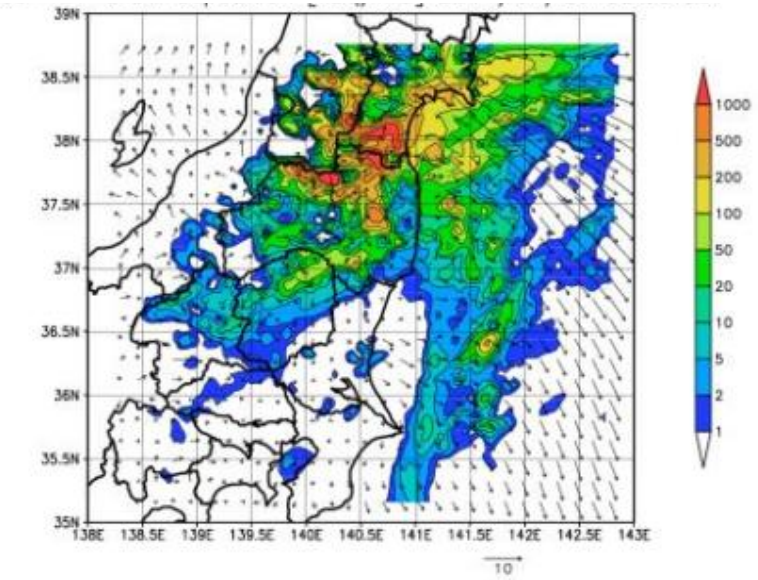


Aircraft monitoring of deposited Cs-137 (MEXT)

Accumulated deposition of Cs-137 by rain (kBq/m^2

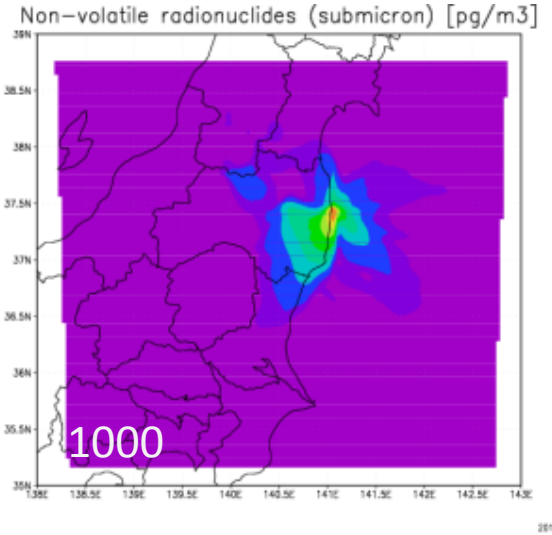


Accumulated deposition of Cs-137 by snow(kBq/m^2)

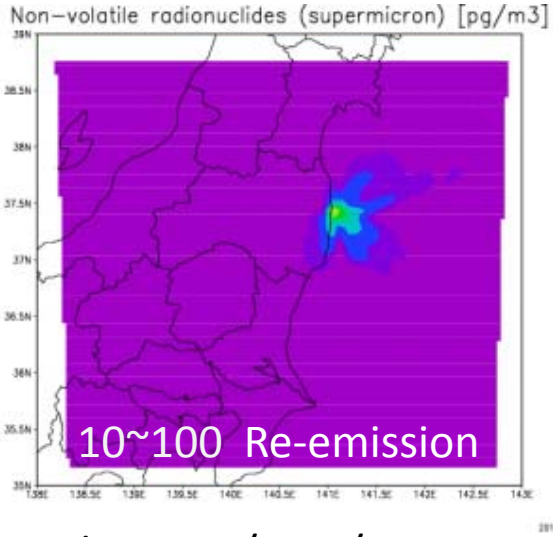


Size distribution and dry deposition velocity

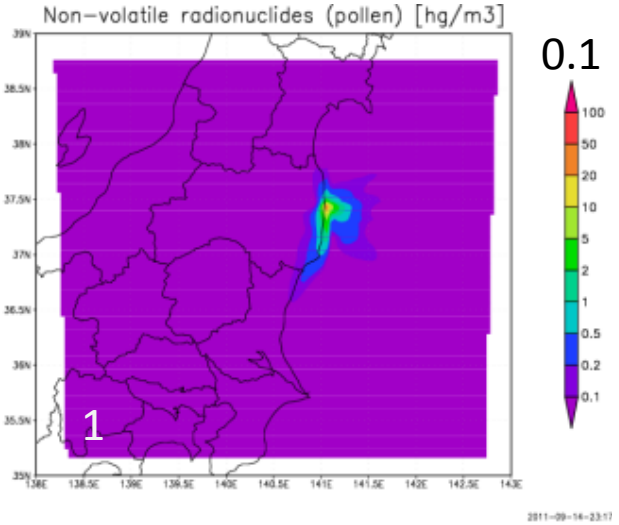
Dg < 1μm (ATK+PRI+ACM)



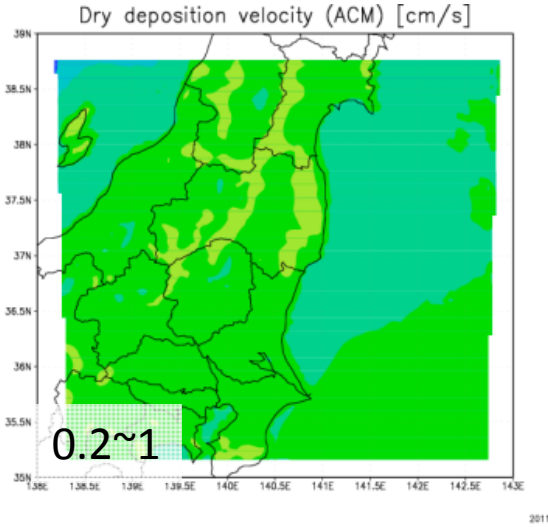
Dg > 1μm (DU+SS)

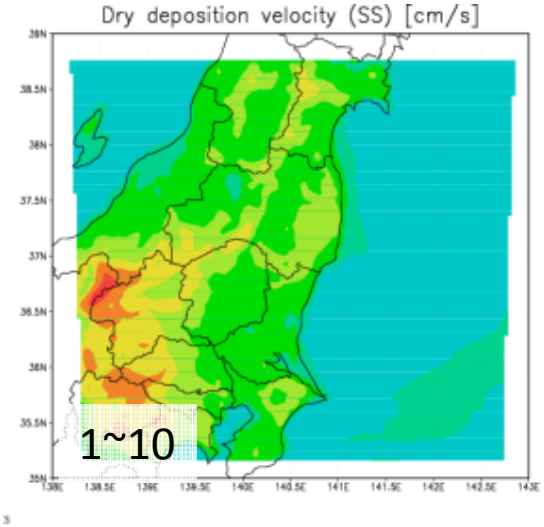


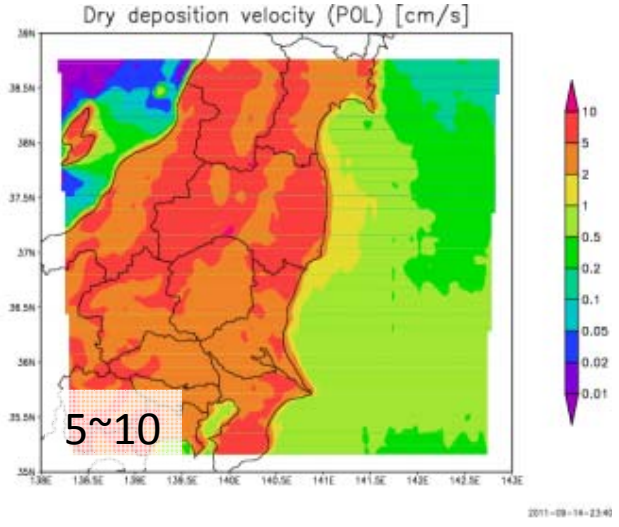
Dg > 10μm (POL)



Averaged dry deposition velocity during 3/11-3/22 : Function of wind speed and roughness



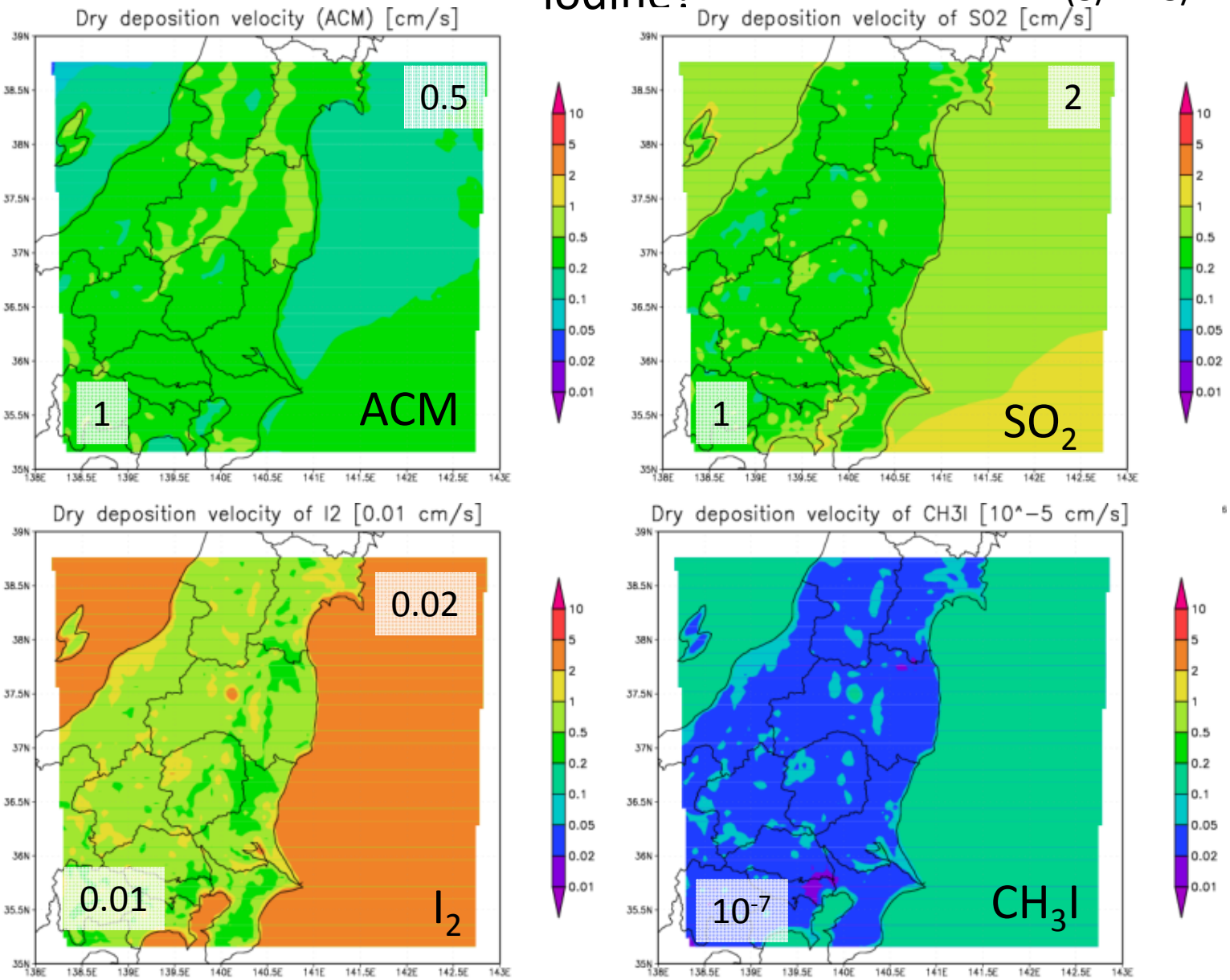




Ground level concentration: fine particles dominate CsI, coarse particles are one order smaller, pollens are two order smaller.
Dry deposition velocity is one order larger over land than over ocean, coarse particle are similar or one order larger than fine particles, pollens are one order larger.

How do the dry deposition velocity of gas-phase and aerosol-phase iodine?

(3/11-3/22 average)

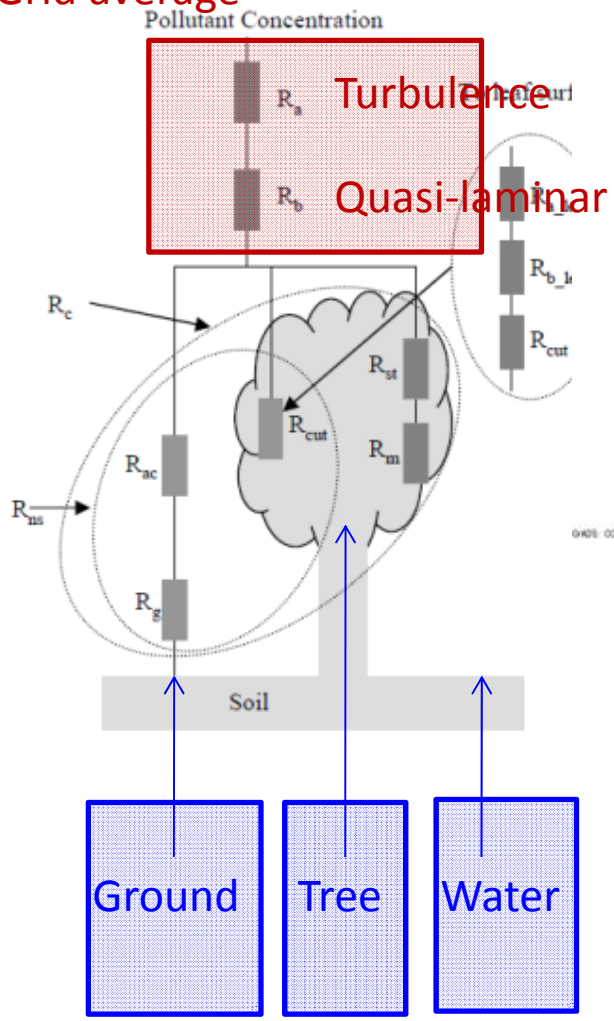


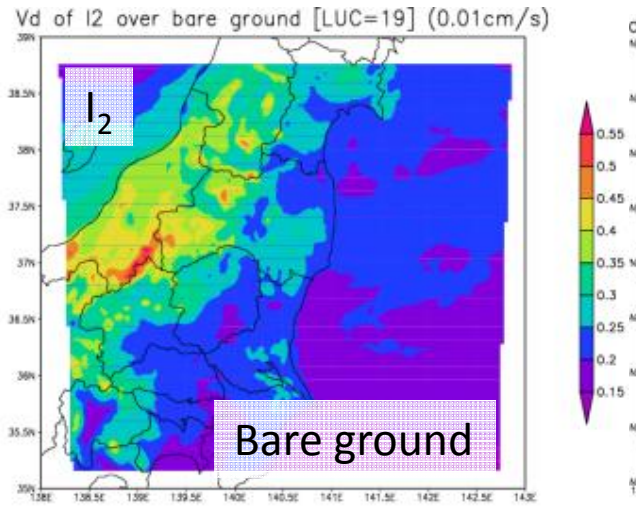
Dry deposition velocity of I_2 and CH_3I are 2 order smaller than those of SO2 and submicron particles.

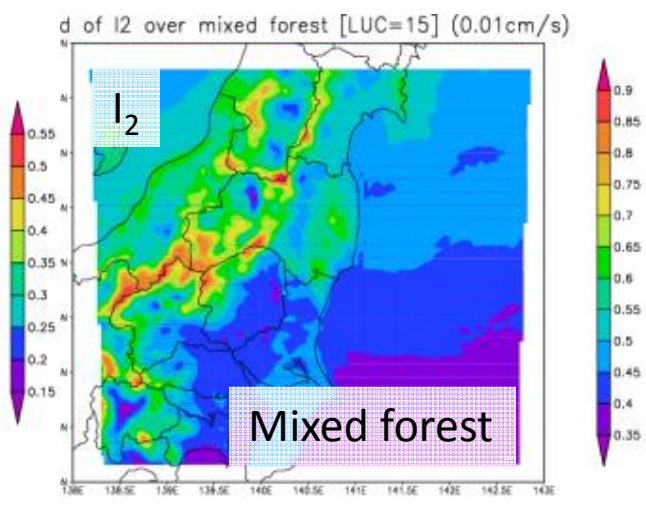
Effects of land surface types on the deposition velocity

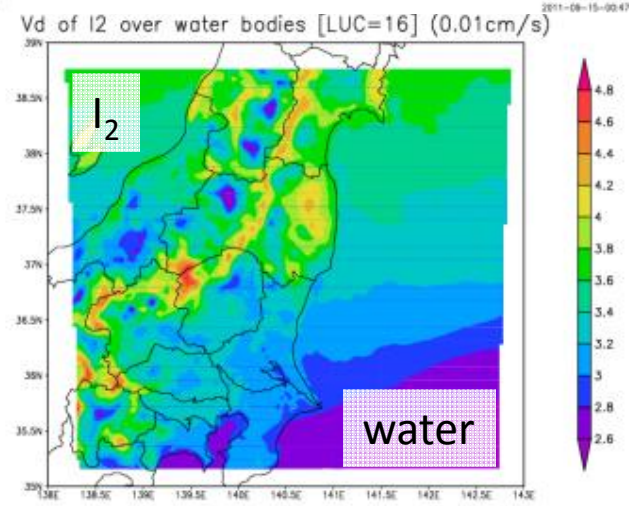
(3/11-3/22)

Grid average









Deposition velocity of Iodine gas on water surface are 5 times faster than mixed forest, and 10 times faster than bare ground.

For submicron particles, deposition velocities of mixed forest and water surface are about 2 times faster than that of bare ground.

Inverse analysis with the regional model

Observation

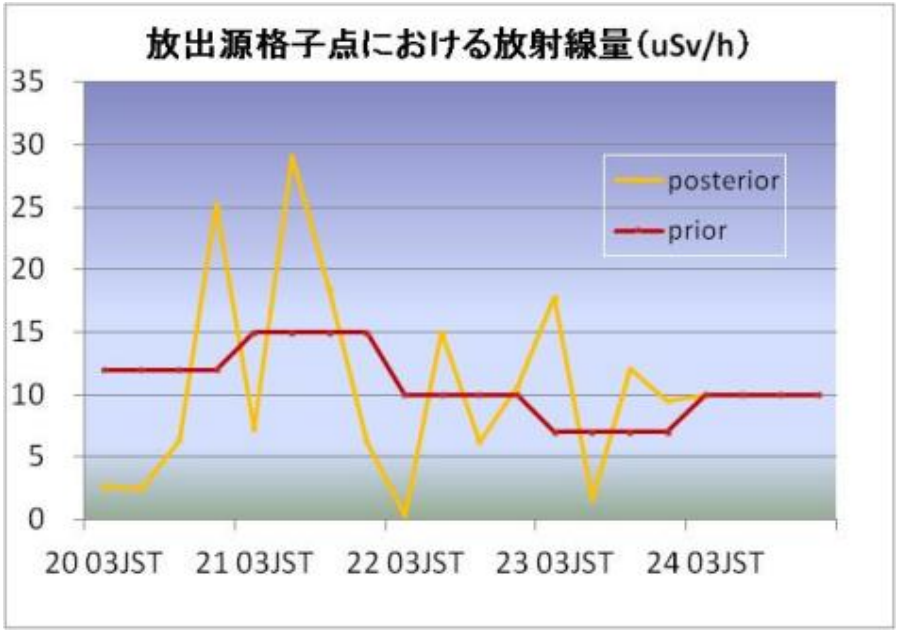
We collected 49 sites observation data in Japan with simple quality control system.

Model

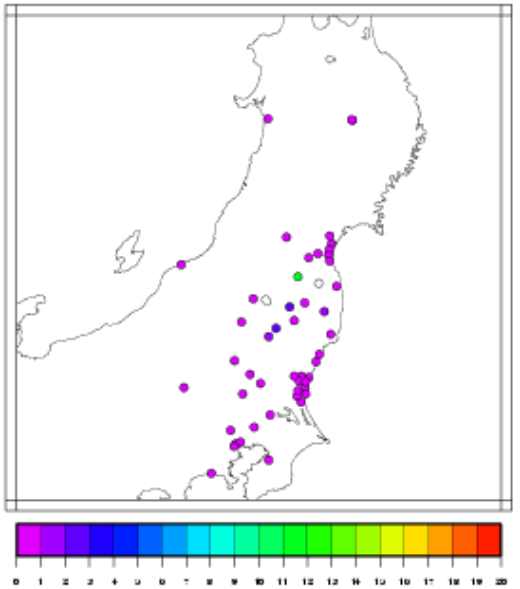
MRI-PM by Kajino et al.,

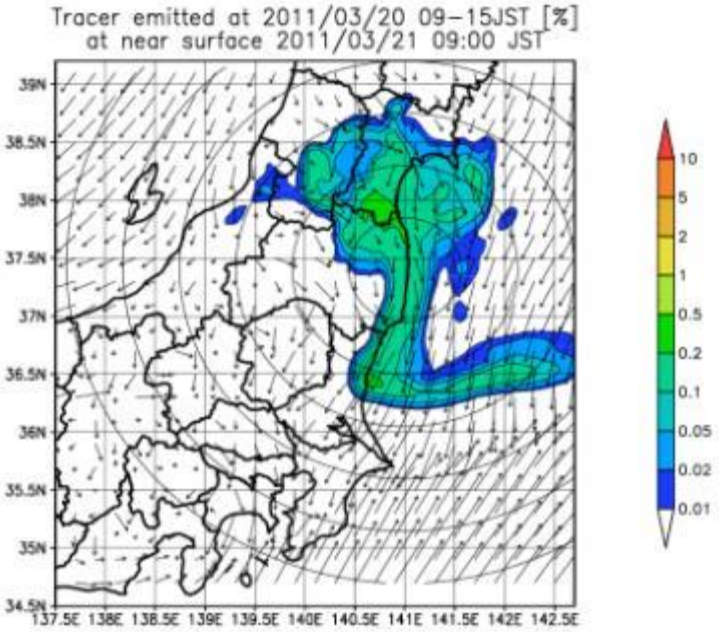
Prior Information

We used estimation flux by Chino et al.



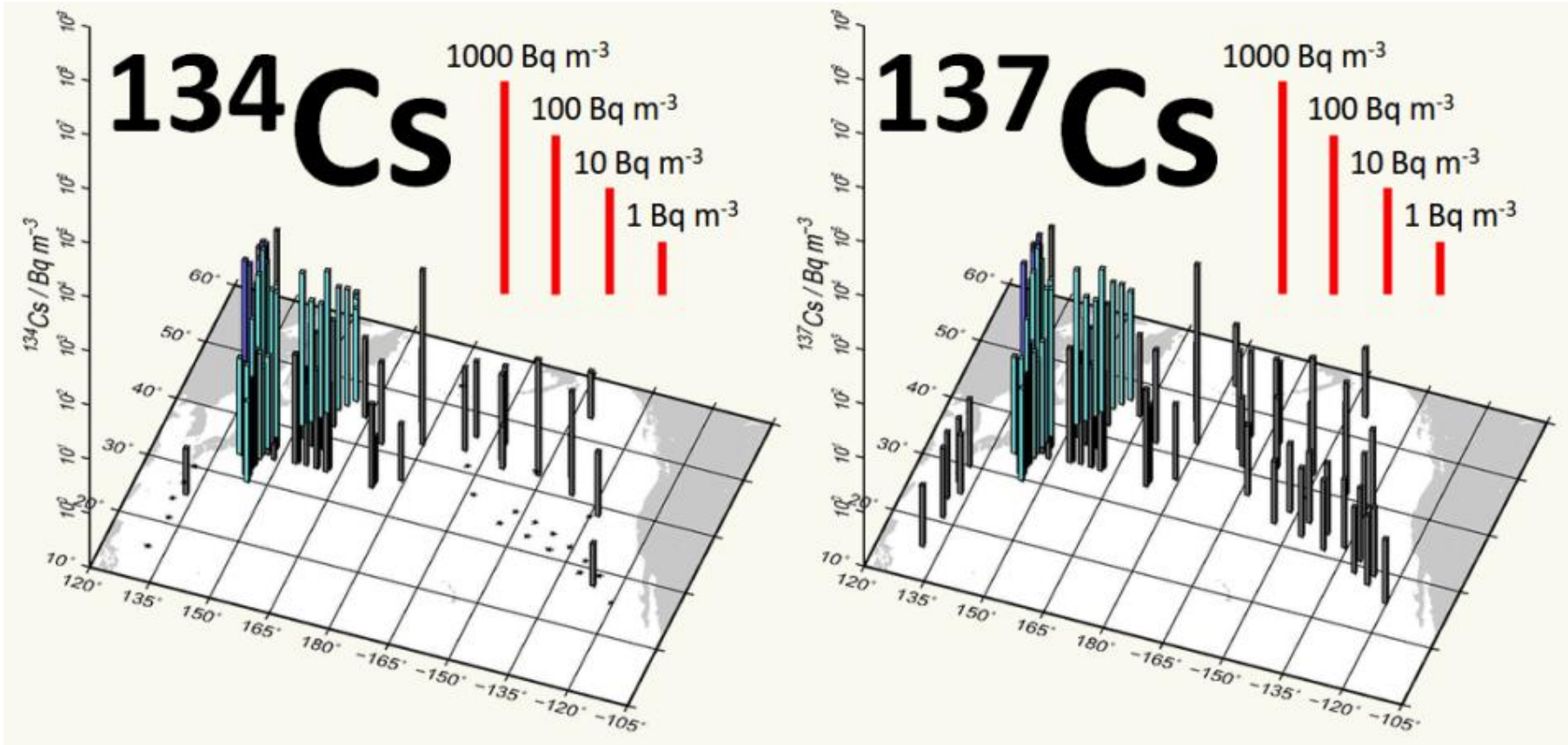
Preliminary estimated emission fluxes.



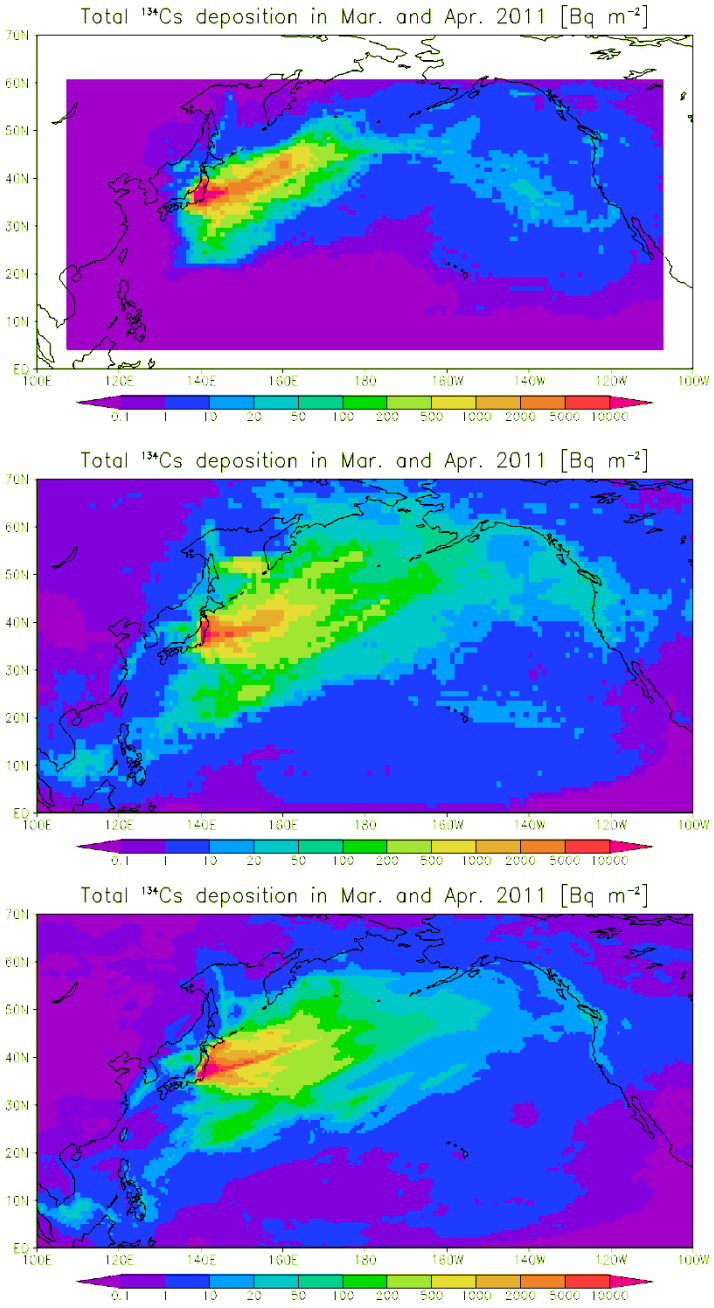


Estimation from the ocean deposition

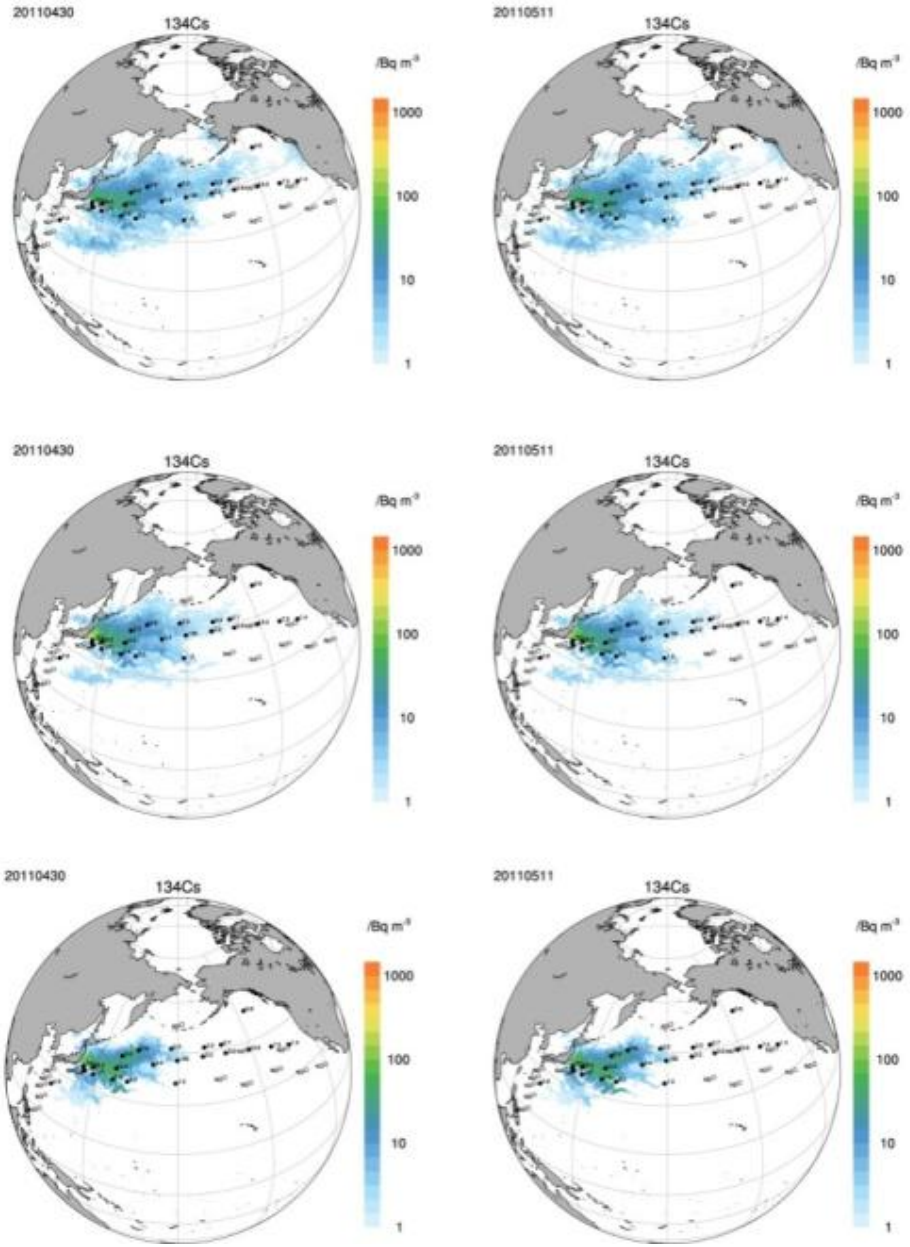
Observed Cesium in the Pacific surface water



(M. Aoyama, MRI/JMA)



Multi model calculation: Atmospheric transport models calculation for deposition behaviour over the North Pacific Ocean



Ocean transport model calculation for radiocaesium activity in the ocean

Estimated Cs-137 in the Ocean Environment

- Estimated atmospheric input to Pacific
  - 13.8 - 16.1 PBq
- Direct discharge form the power plant
  - 3.5 ± 0.7 PBq
- Cs-137 in Pacific Ocean before the accident, mainly due to nuclear weapon test
  - 69 PBq

Estimated total ^{137}Cs emission flux

	Total Flux	
This study	13 – 24 PBq	(3/10-4/20)
JAEA (Chino et al., revised 2011)	9.1 PBq	(3/10-3/31)
Stohl et al. (2012)	36.6 PBq (20.1 – 53.1)	(3/10-4/20)
Winiarek et al. (2012)	10 – 19 PBq	
MELCOR analysis (Gauntt et al.)	16.4 PBq	From Stohl et al. (2012)
IRSN	10 PBq	
ZAMG	66.6 PBq	From Stohl et al. (2012)
Aoyama et al. (ms. in preparation)	15.2 – 20.4 PBq	From obs. and numerical model analysis

Summary and Future Plans

- We are constructing an inverse modeling system to estimate emission flux by Fukushima power plant.
- Preliminary emission flux was obtained. We have a plan to estimate emission flux in higher time resolution and longer period.
- We have a plan to provide our estimated flux to global/regional model to understand transport and deposition processes.
- It is recommended that we need inversion inter-comparison to avoid estimation error by using only one model.

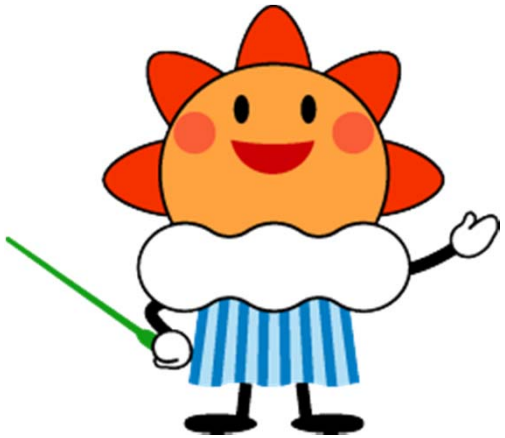
Future plans

- Data assimilation of radioactive materials using Ensemble Kalman Filter
  - Data assimilation with radiation dose rate

This is the end of the presentation.

Thank you very much!

Grazie mille!



Comparison of the time series of the emission of Cesium 137