Observations of atmospheric radionuclides from the Fukushima nuclear accident in Tsukuba, Japan

茨城県つくば市での福島事故由来の大気中人工放射性核種の観測

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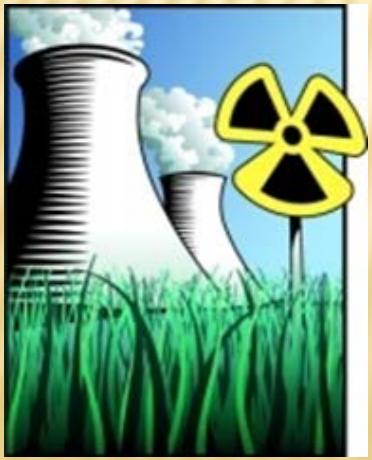
TALK OUTLINE

- Aim of the research
- Extent of the pollution by the Fukushima accident
- Observations and experimental at the MRI
- Gamma emitters in the atmosphere at the accident
- Plume transport reconstruction by the modeling
- Radio-Sr in the atmosphere at the accident
- Temporal changes in radio-Cs concentration in the atmosphere
- Long-term monitoring of radioactive fallout (^{90}Sr and ^{137}Cs); comparison with the Fukushima fallout
- Summaries

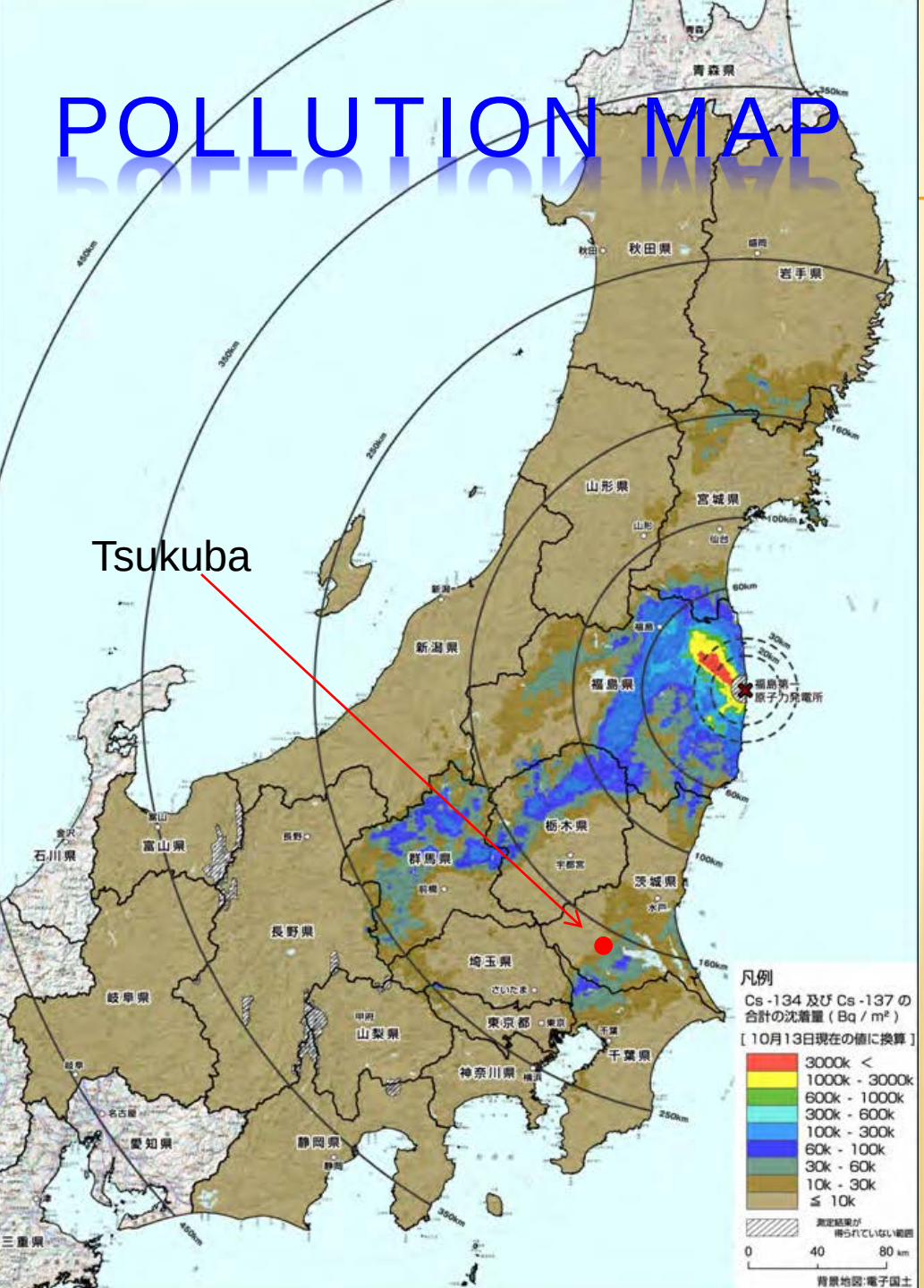
POINTS TO BE CLARIFIED ACCORDING TO OBSERVATION AND MODEL

Learn quantitative relations

- Transportation and diffusion
- Point source
- Emission inventory
- Temporal variation
- Dry deposition
- Wet deposition



POLLUTION MAP



Result of aircraft monitoring by the Ministry of Education, Culture, Sports, Science and Technology

(Total of cumulative Cs-134,137 pollution of the surface (kBq/m^2) in the range that the survey completed up to fall, 2011)

November 11, 2011 Announcement

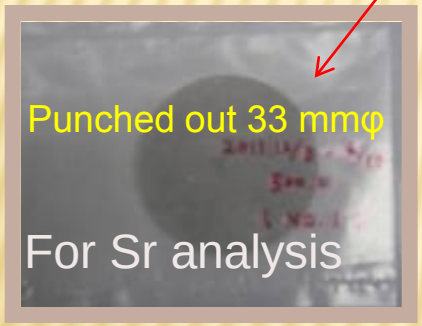
EMISSION ESTIMATE BY NUCLEAR AND INDUSTRIAL SAFETY AGENCY (OCT. 20, 2011)

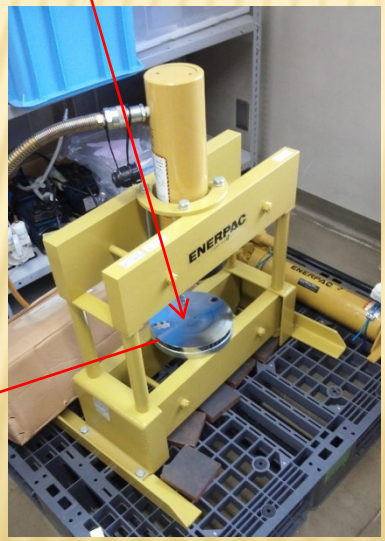
Nuclide	Half life	Emission (PBq)	Emission (PBq)
Kr-85	10.72y		33
Xe-133	5.25d	11000	6500
Te-129m	33.6d	3.3	240
Te-132	3.26d	88	~ 1150
I-131	8.04d	160	~ 1760
I-133	20.8h	42	910
Cs-134	2.06y	18	~ 47
Cs-136	13.1d		36
Cs-137	30.0y	15	85
Sr-89	50.5 d	2	~ 115
Sr-90	29.12y	0.14	~ 10
Pu-239	24065y	0.0000032	0.015
Pu-240	6537y	0.0000032	0.013

Comparison: Emission by the Chernobyl accident (Chernobyl forum)

SAMPLING, SAMPLE PREPARATIONS AND GAMMA MEASUREMENT

Total = Wet and dry deposition sampling





Hydraulic press machine

RADIOCHEMICAL SEPARATION OF RADIO-Sr

Major Removal

1
- Sample dissolution in nitric acid
- Adding stable Sr and Ca carriers

Cs

2
- Hydroxide precipitation (Bulk recovery of alkaline earths)
- Oxalate precipitation

Ca

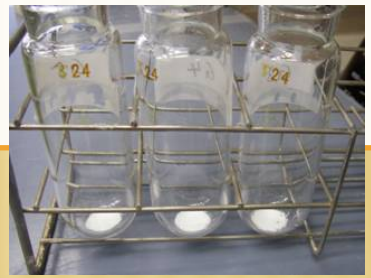
Ba, Pb, Ra

3
- Fuming nitric acid precipitation
- Barium chromate precipitation

^{90}Y

4
- Hydroxide precipitation (Recovery of Sr)
- Iron hydroxide precipitation

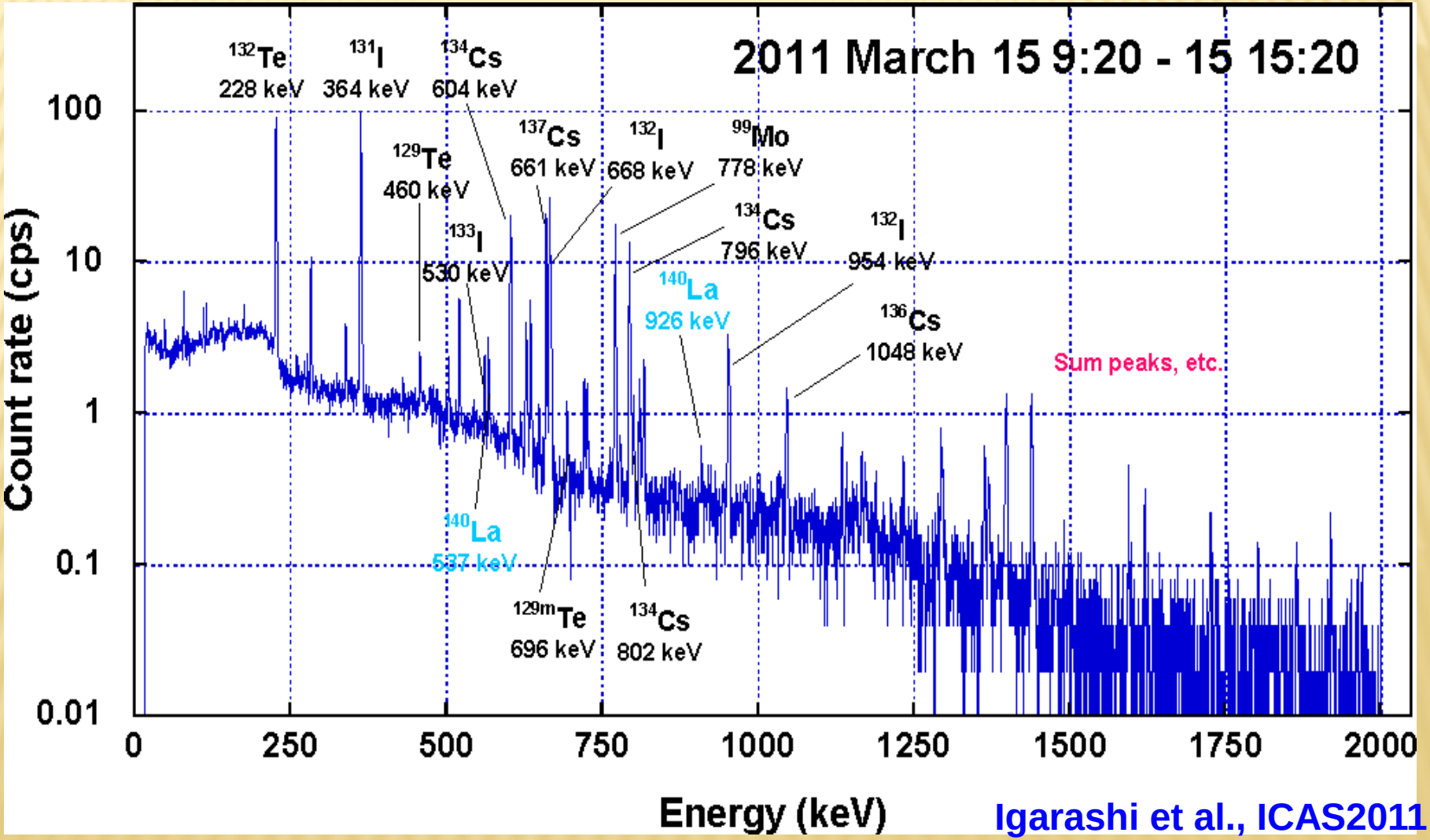
5
- Fixation as Sr carbonate
- Low BG β counting



Yield of Sr : about 60~90%

GAMMA-RAY SPECTRUM OF A HV FILTER SAMPLE

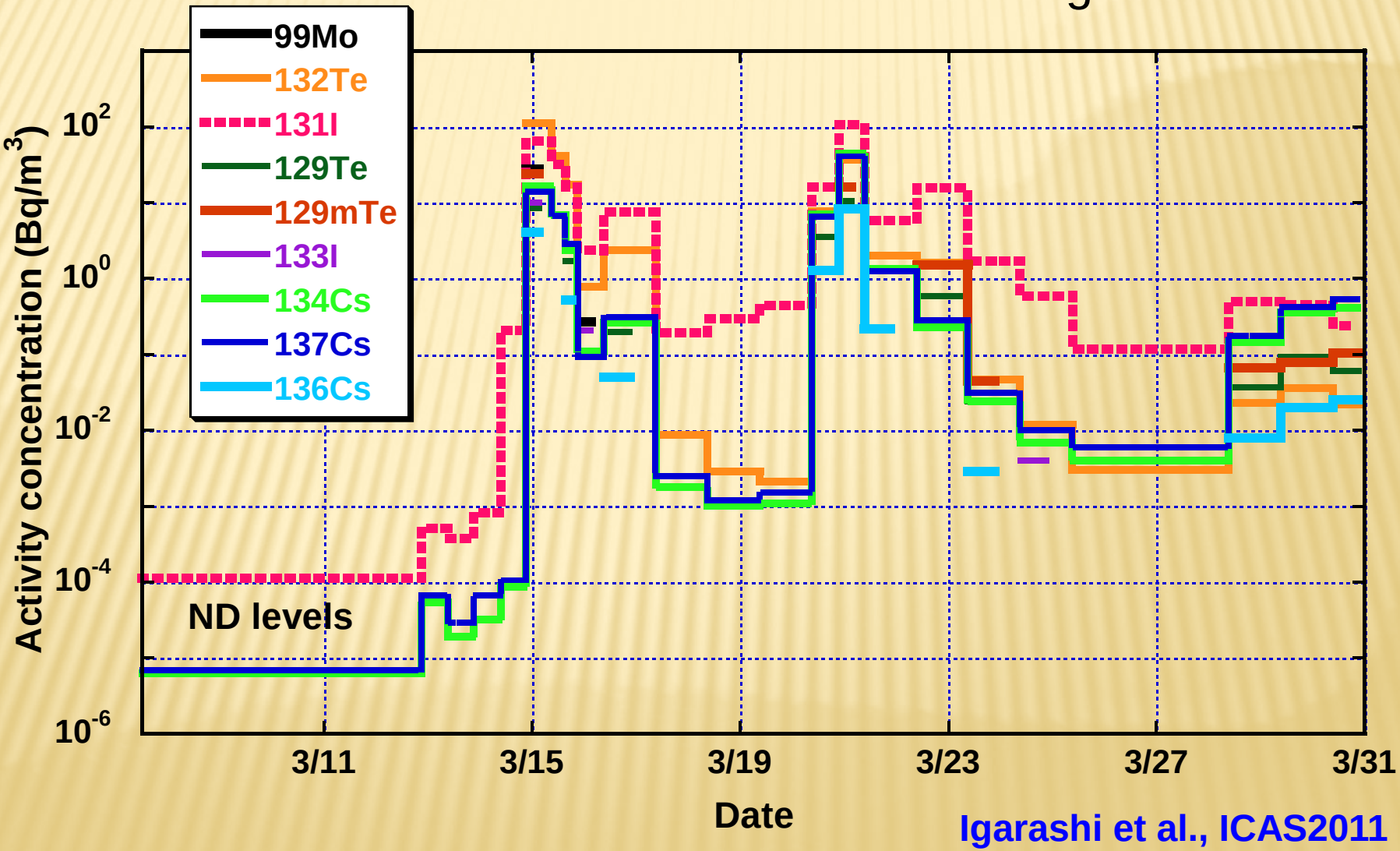
A sample collected at the observation field of MRI, Tsukuba



Igarashi et al., ICAS2011

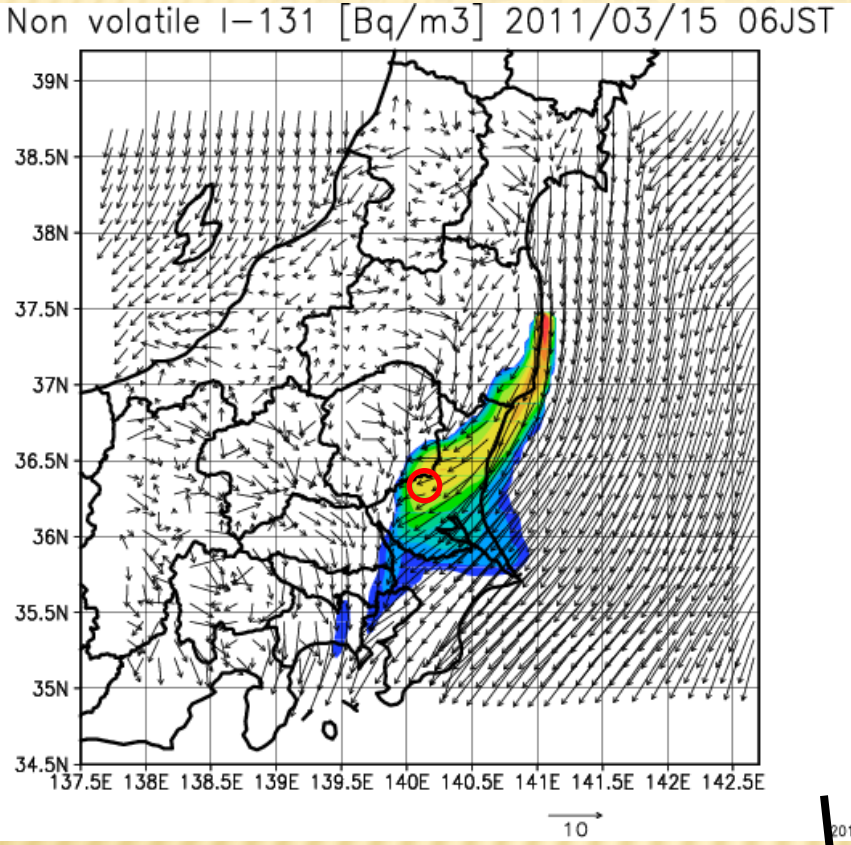
RADIOACTIVITY IN AEROSOL SAMPLES AT THE MRI IN MARCH 2011

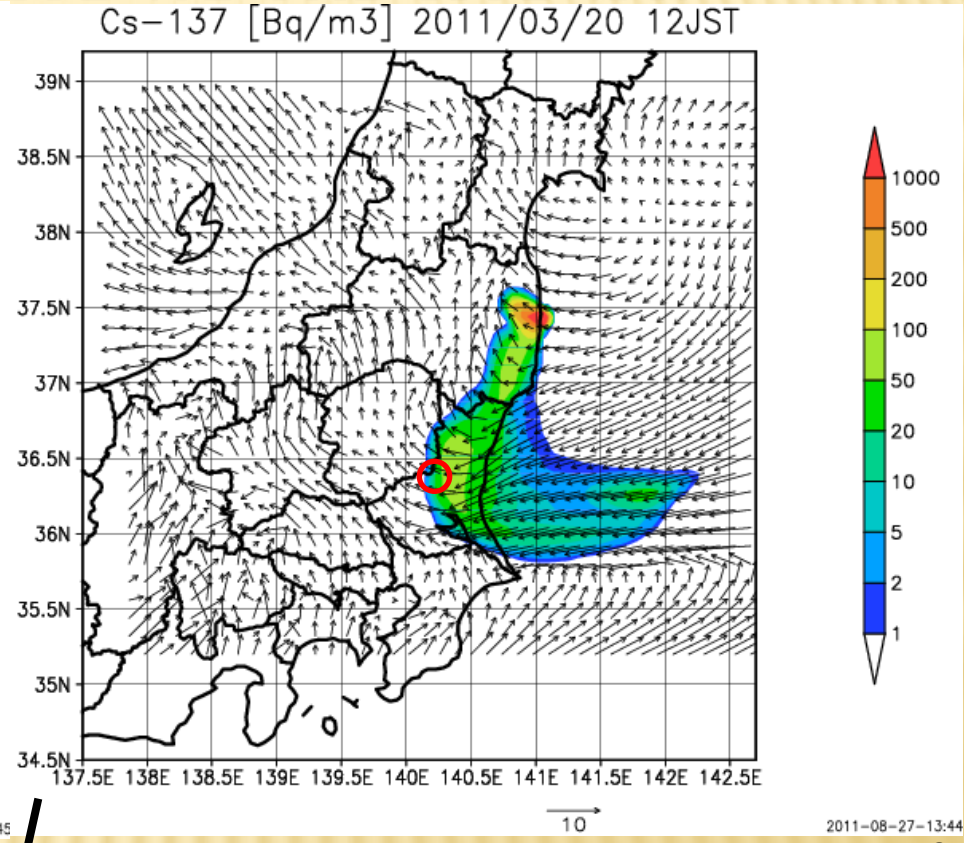
In logarithmic scale



Igarashi et al., ICAS2011

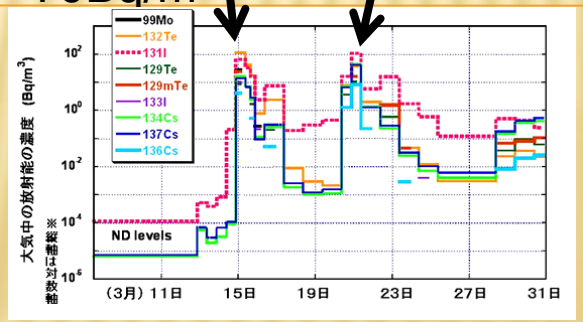
TRANSPORT OF RADIOACTIVE PLUME TO TSUKUBA (MAR./15 AND MAR./20)





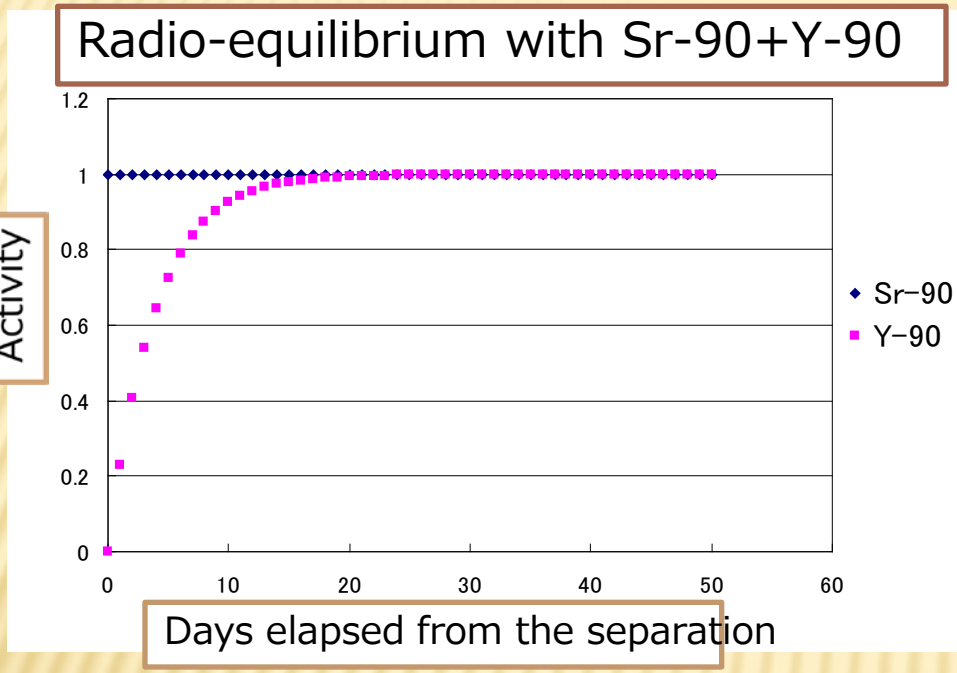
Observed I-131 at the MRI: 70Bq/m^3

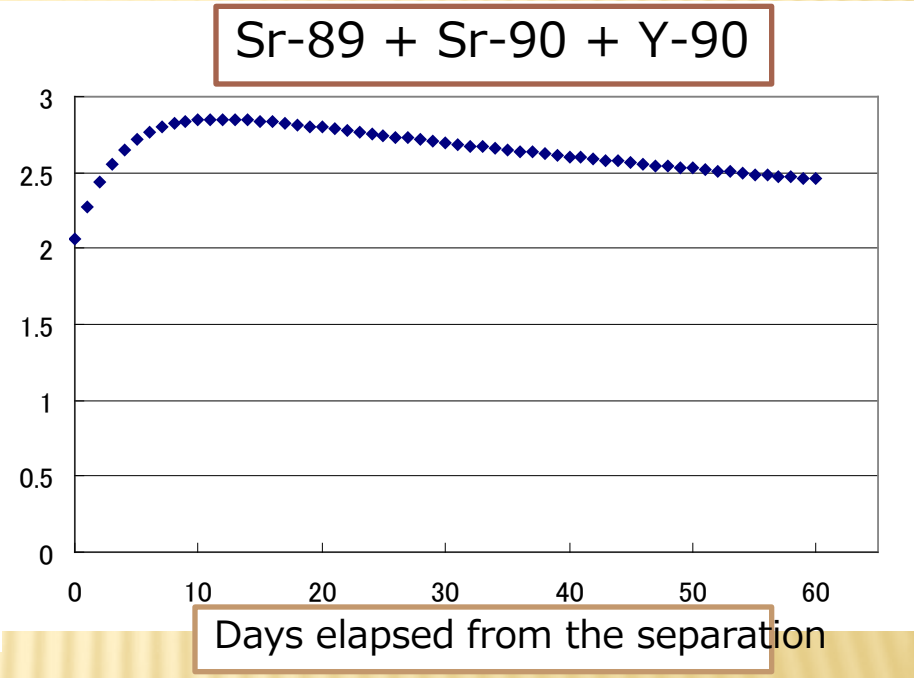
Observed Cs-137 at the MRI: 40Bq/m^3



Kajino et al., MSJ2011Fall

β-measurement of ^{89}Sr and ^{90}Sr(^{90}Y)





Radio-eqlbrm. ^{90}Sr and ^{90}Y

Maximum β-energy

^{90}Sr(28.8y) 0.546MeV

^{90}Y(2.67d) 2.240MeV

^{89}Sr(50.52d) 1.497MeV

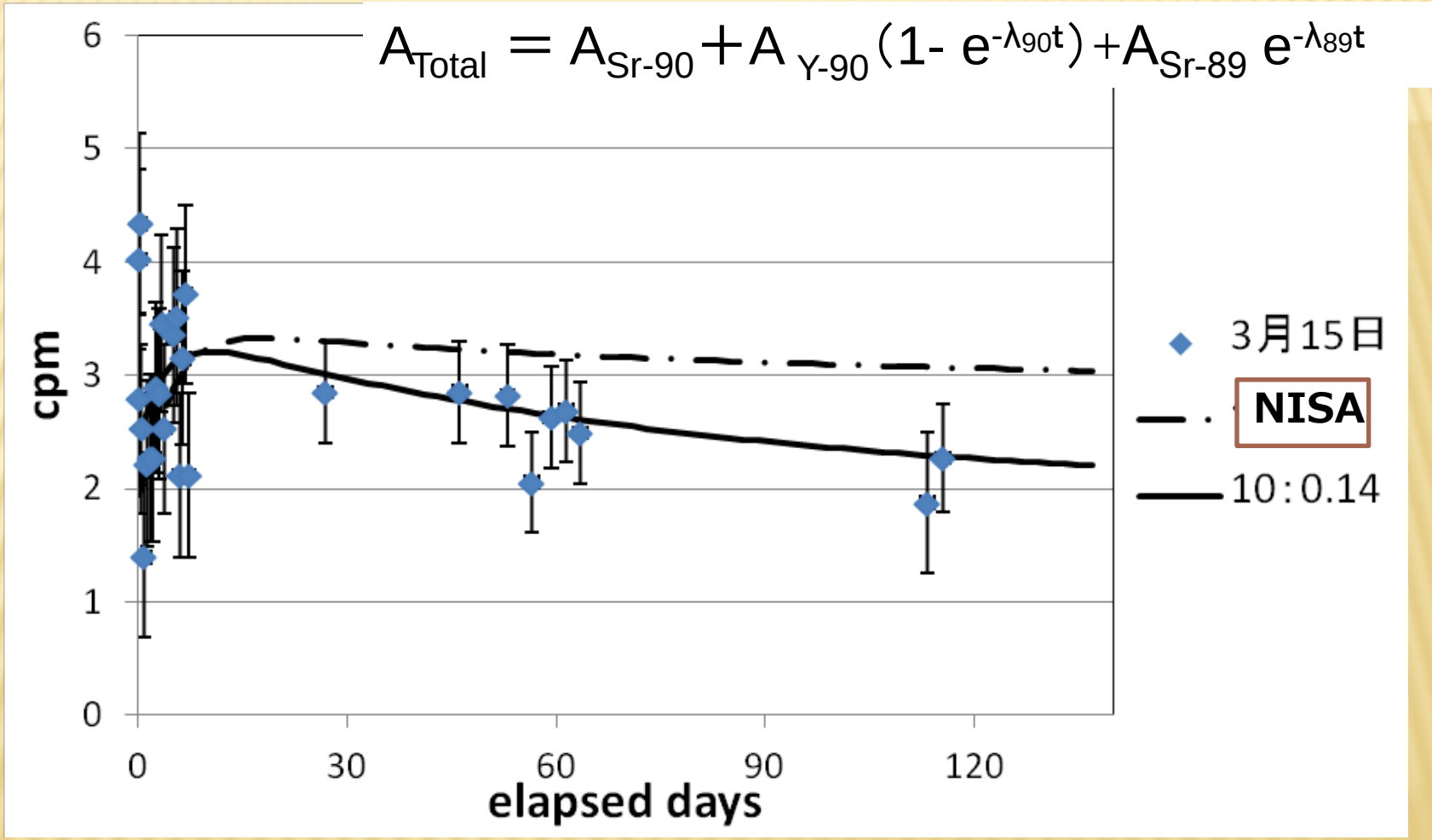
^{89}Sr : ^{90}Sr = 2 : 0.14

(based on NISA estimation)

Measured from 2011/Dec/7

Igarashi et al., 2012JpGU

DETERMINATION OF ^{89}Sr AND ^{90}Sr (MAR. 15, 2011 FILTER SAMPLE)

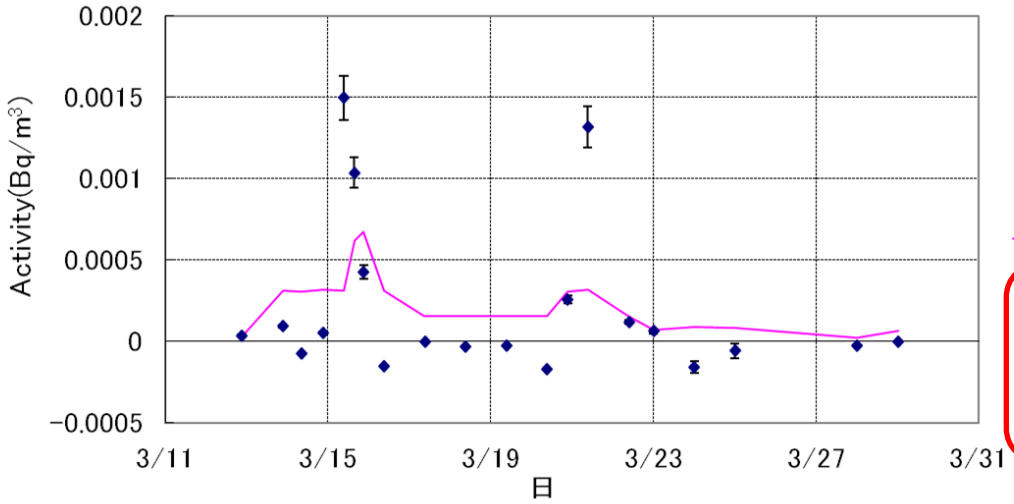


NISA estimate ･･･ ^{89}Sr : ^{90}Sr = 2 : 0.14

March 15th meas. ･･･ ^{89}Sr : ^{90}Sr = 10 : 0.14

Igarashi et al., 2012JpGU

ATMOSPHERIC CONCENTRATION OF ^{90}Sr AT THE MRI IN MARCH 2011



Max. on March 15
1.5 ±0.27mBq/m³

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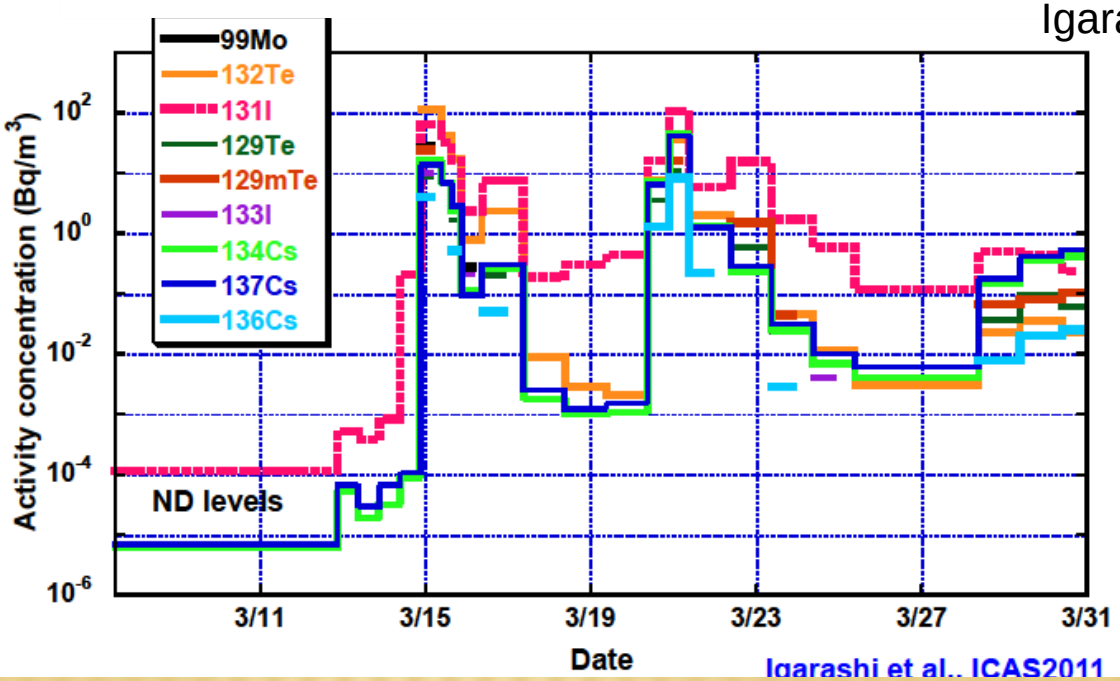
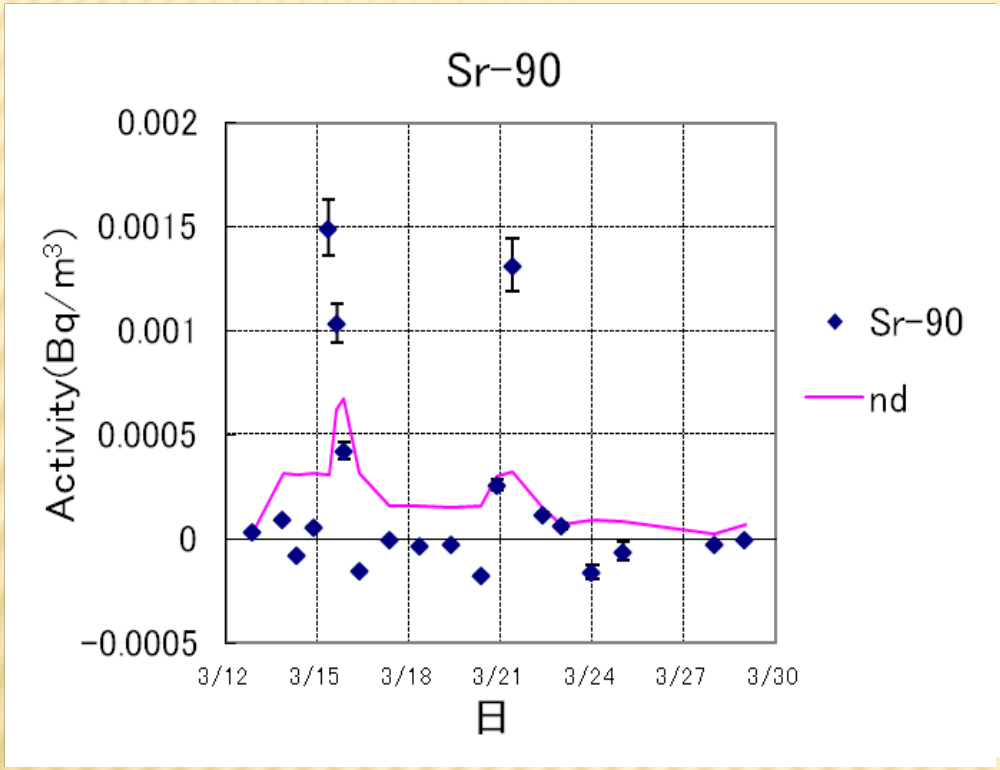
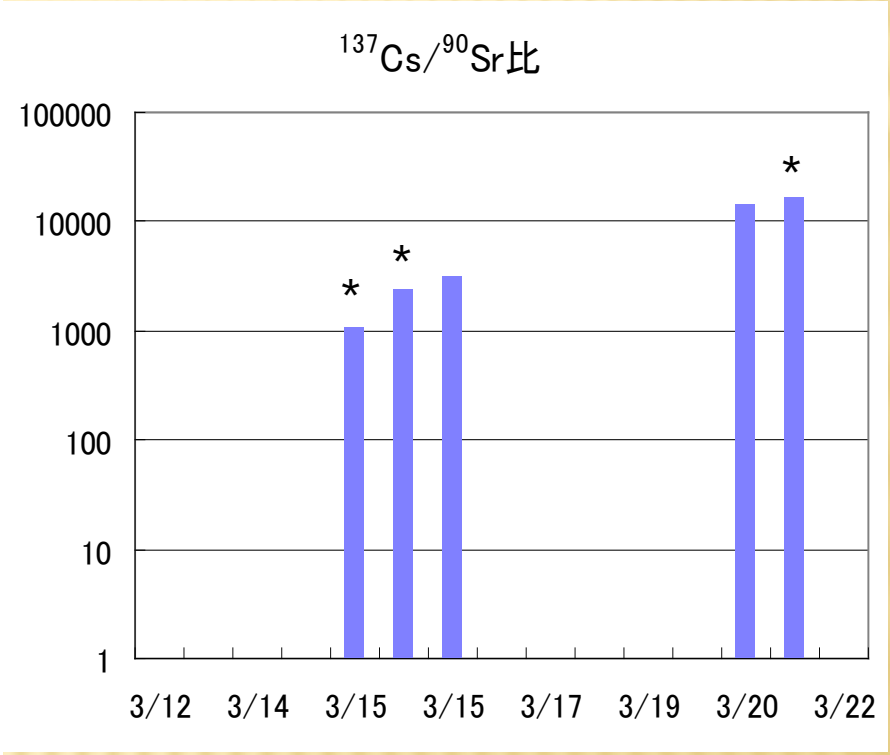


$^{137}Cs/^{90}Sr$ RADIOACTIVITY RATIO IN MARCH 2011

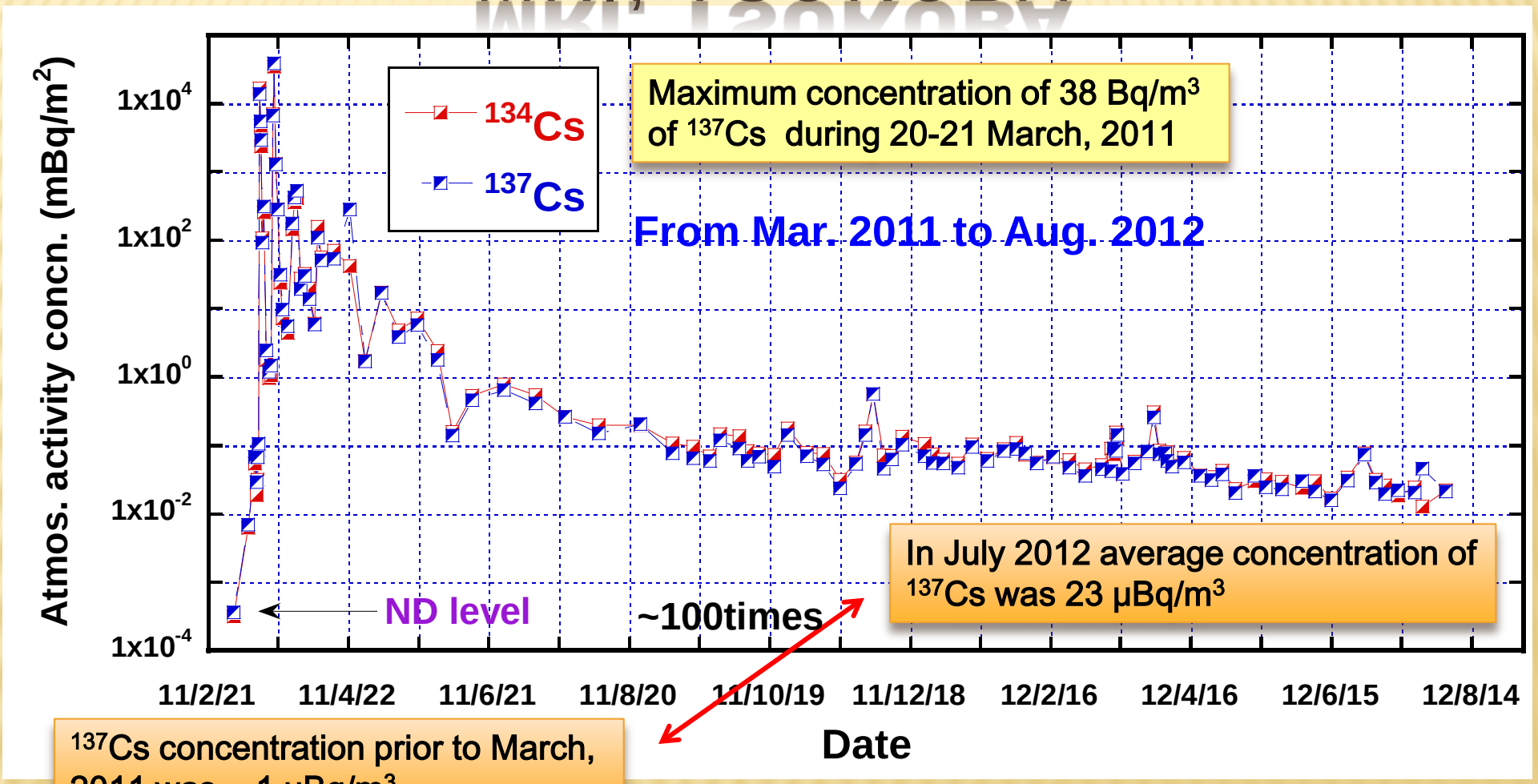




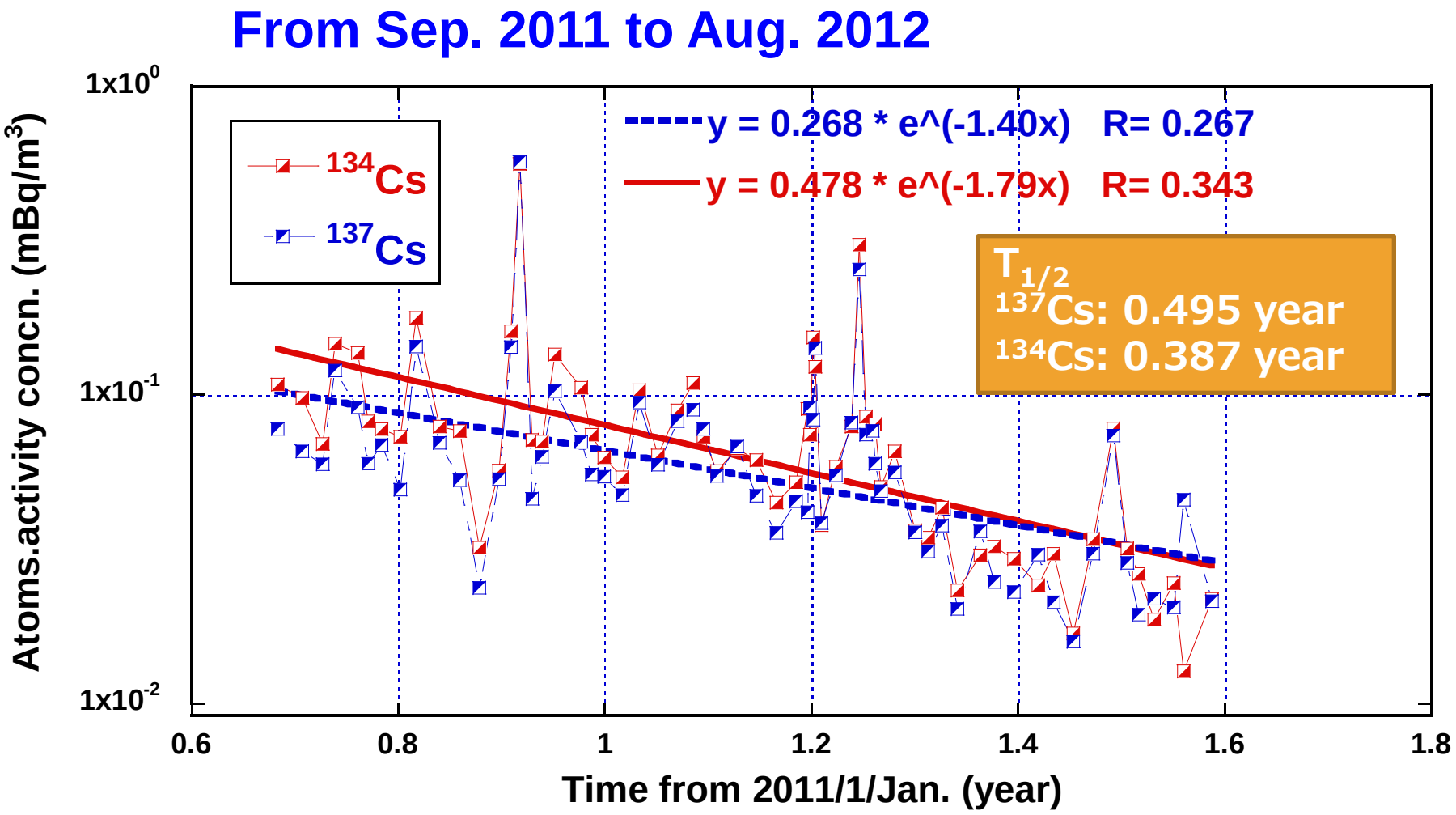
- $^{137}Cs/^{90}Sr$ ratio in the weapon fallout during the1960-70
  ⇒ About 1.6 (Krey et al., 1970)
- Chernobyl accident in 1986 ⇒ 95.7 (Aoyama et al., 1991)

Igarashi et al., 2012JpGU

TEMPORAL CHANGE IN ATMOS. RADIO-Cs CONCENTRATION AT THE MRI, TSUKUBA

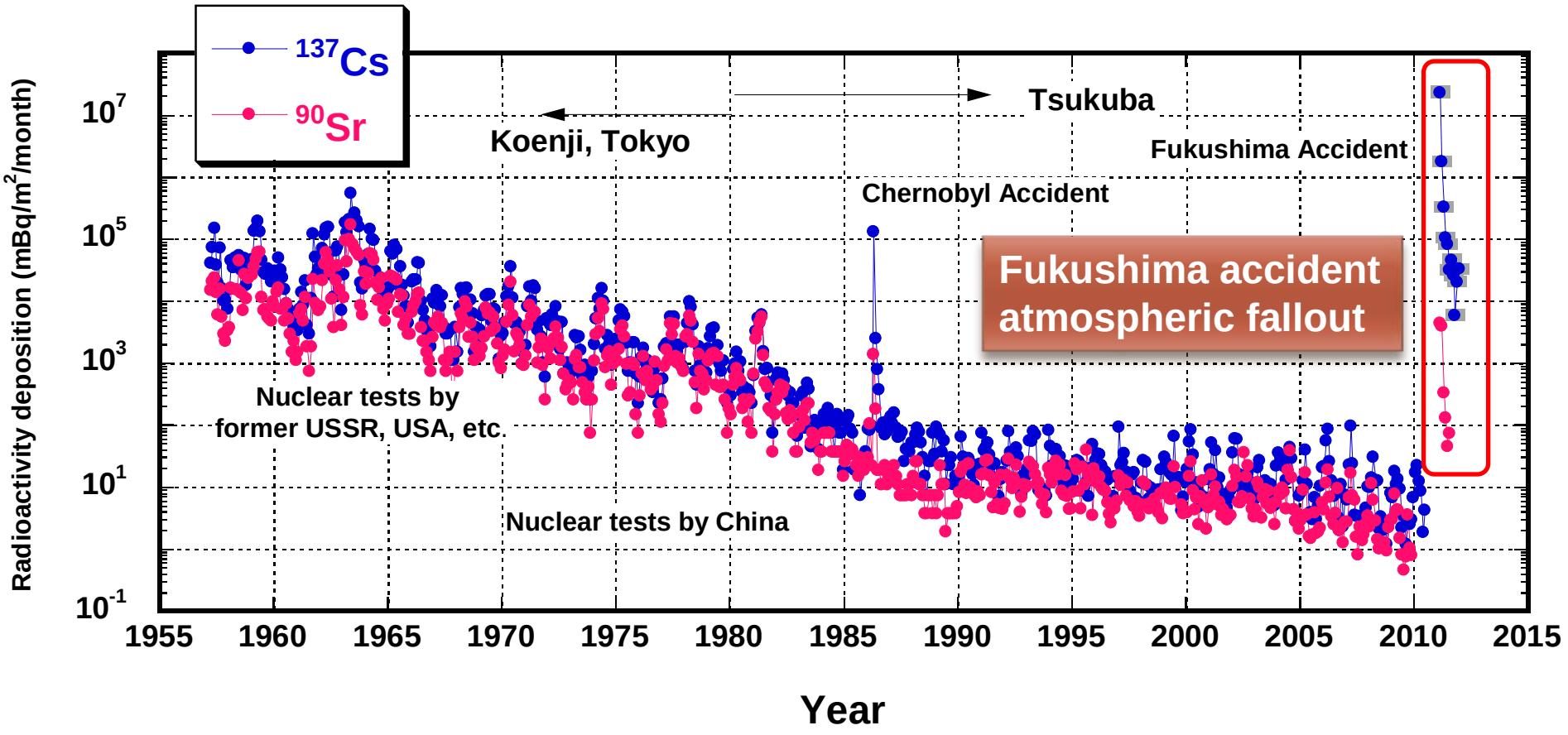


DECREASE IN ATMOSPHERIC RADIO-CS CONCENTRATIONS



LONG-TERM DEPOSITION RECORD AT THE MRI, TSUKUBA WITH THE FUKUSHIMA FALLOUT

Analysis of the total deposition sample collected monthly



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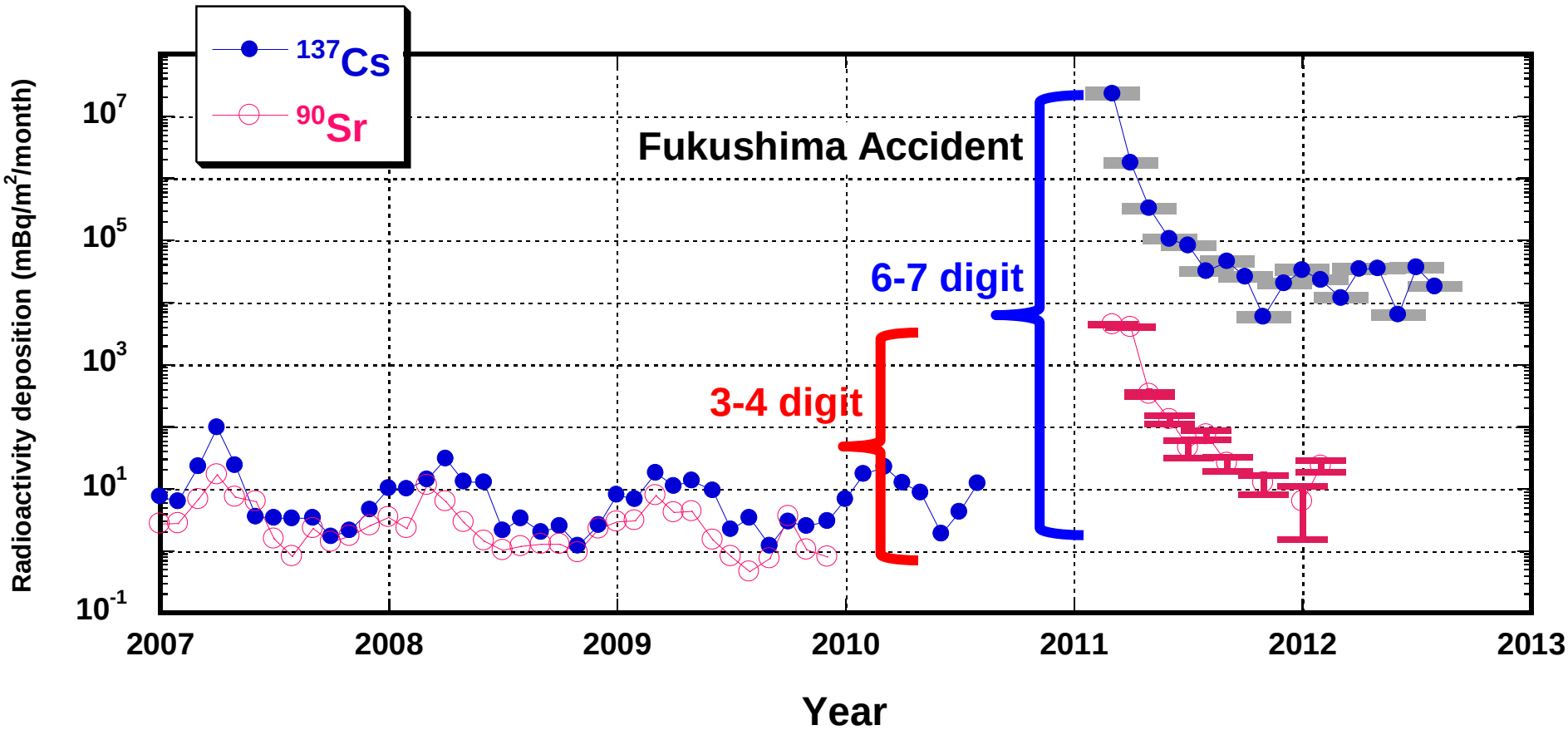
THE MONTHLY DEPOSITIONS BEFORE AND AFTER THE FUKUSHIMA ACCIDENT



ATMOSPHERIC DEPOSITION AFTER THE FUKUSHIMA ACCIDENT AT THE MRI, TSUKUBA

Unit: Bq/m^2 /month

Month	^{90}Sr	Erros	^{137}Cs	Errors	Ratio
Mar/2011	4.36	±0.09	2.308E+04	±924	5292
Apr/2011	4.00	±0.07	1.776E+03	±1.3	444
May/2011	0.33	±0.03	330	±0.3	989
Jun/2011	0.13	±0.02	104	±0.1	804
Jul/2011	0.05	±0.01	82.0	±0.1	1808
Aug/2011	0.07	±0.01	31.9	±0.1	435
Sep/2011			45.9	±0.1	
Oct/2011			25.8	±0.1	
Nov/2011			5.9	±0.0	
Dec/2011			20.3	±0.1	
Jan/2012			32.6	±0.1	

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SUMMARY 1

- We have continued observations of atmospheric concentrations of radioactive Sr and Cs, etc. and their depositions at the MRI, Tsukuba before and after the Fukushima nuclear accident.
- The plume transport to the MRI from the Fukushima accident was captured and reconstructed by the transport modeling.
- By tracking the temporal change of total β-activity of Sr, which was resolved into ^{89}Sr and ^{90}Sr.
- The relative ratio of 70 is estimated for ^{89}Sr and ^{90}Sr in March 2011, which is about 5 times larger than the NISA estimate.
- The $^{137}Cs/^{90}Sr$ ratio was in the range of 400 ~ 20000 in filter samples, suggesting the Cs enrichment during the transport.
- The ^{137}Cs monthly deposition at the MRI was $(23\pm0.9)\times10^3$ Bq/m^2 in March 2011, which is 6 to 7 orders of magnitude larger than pre-accident level.
- Equal amounts of ^{134}Cs and ^{137}Cs deposited, giving rise to the surface pollution of approximately 50 kBq/m^2 in Tsukuba, matched nearly with that by the MEXT mapping.

SUMMARY 2

- ^{90}Sr depositions of 4.36±0.09 Bq/m^2 in March 2011, which is less than 0.02% of the total ^{137}Cs fallout in the month.
- Level of ^{90}Sr deposition was 3-4 orders greater than pre-accident level, which did not reach one by nuclear tests during the 1960s; impact by ^{90}Sr will not be so large as radioactive Cs.
- At the end of 2011, the radioactive fallout lowered 3-4 orders from that on the accident, yet the several-Bq/m^2-deposition is continuing. This corresponds to the level in the early 1980s when China carried out the last atmospheric nuclear test.
- At the end of 2011, ^{137}Cs concentration was at the level of tens μBq/m^3. Since the re-suspension continues over a long period of time, it is necessary to watch its future trends.
- Apparent decrease in the atmos. Cs concentration occurs with a half-life of ca. 6 months since Sep. 2011, suggesting that the removal occurs relatively quickly at the 2ndary emission sources.
- Identification of the major source of 2ndary emission to the atmosphere (re-suspension) is required.

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