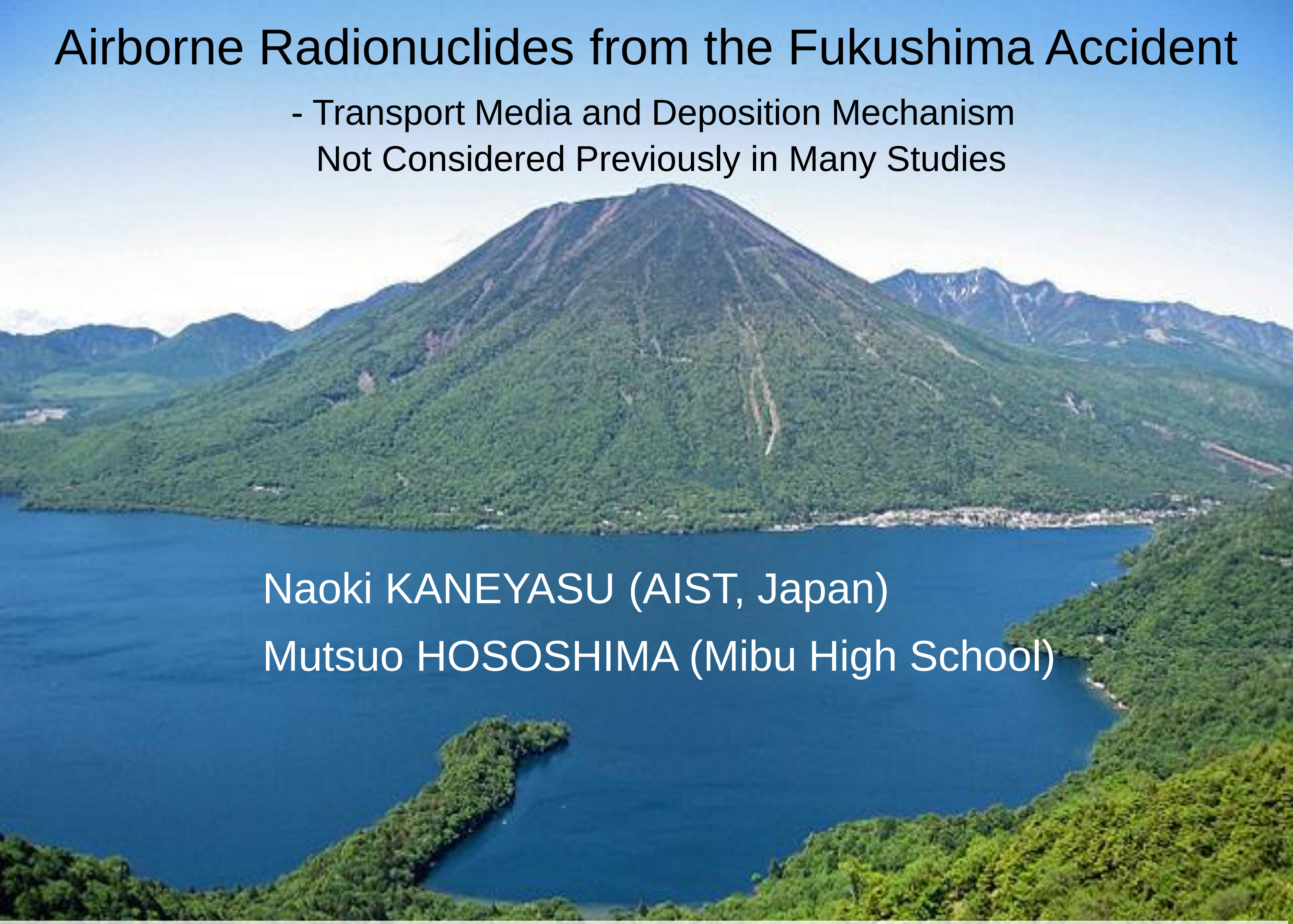
Airborne Radionuclides from the Fukushima Accident

- Transport Media and Deposition Mechanism Not Considered Previously in Many Studies



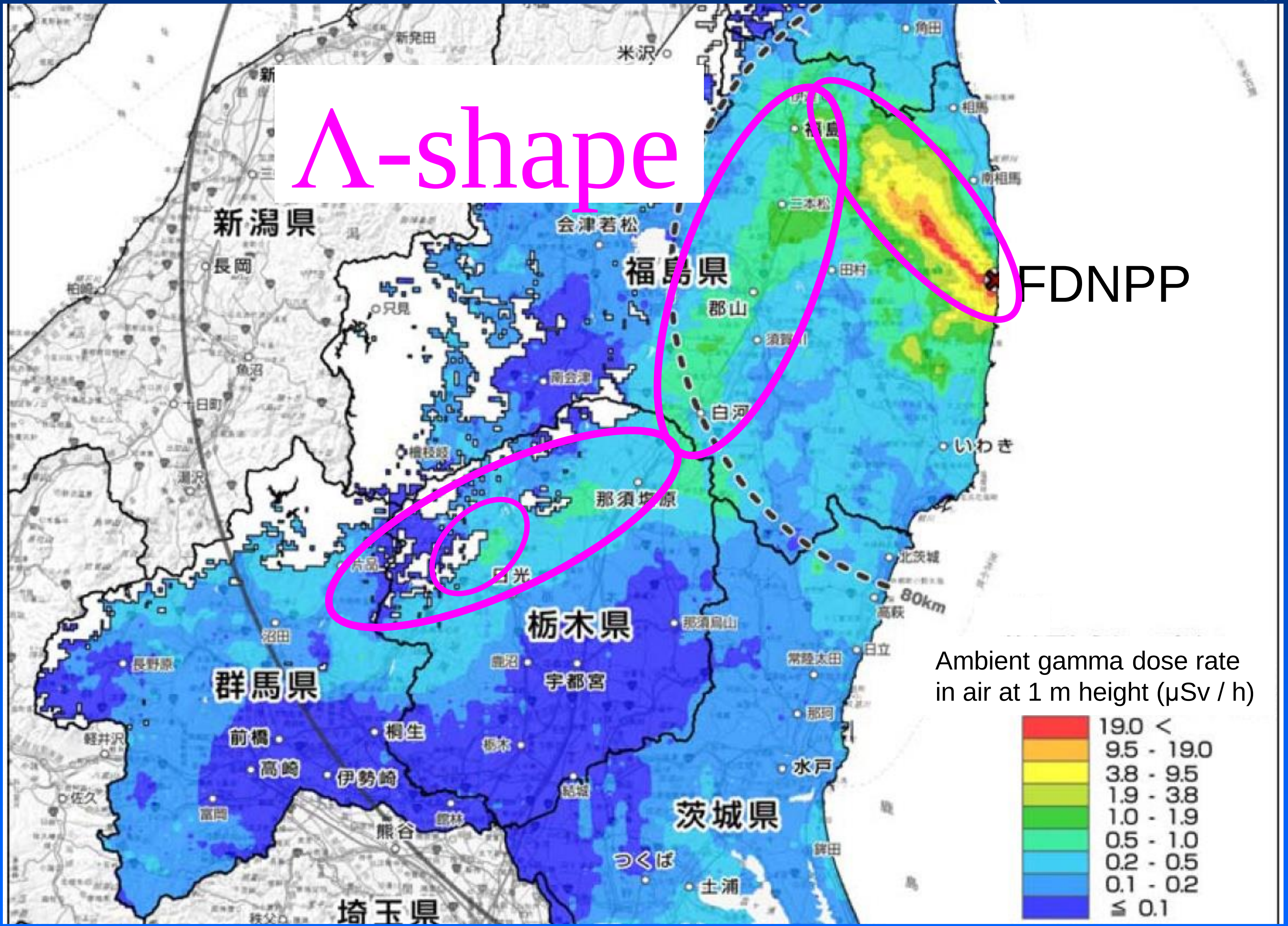
Naoki KANEYASU (AIST, Japan)

Mutsuo HOSOSHIMA (Mibu High School)

International Workshop on Dispersion and Deposition Modeling for Nuclear Accident Releases (2015.3.2)

Airborne monitoring of ambient γ dose rate in air

(as of June 28, 2012)



SW end of the contaminated area shifted toward mountain side

Car-borne survey system KURAMA Ⅱ

(Tanigaki, 2014)

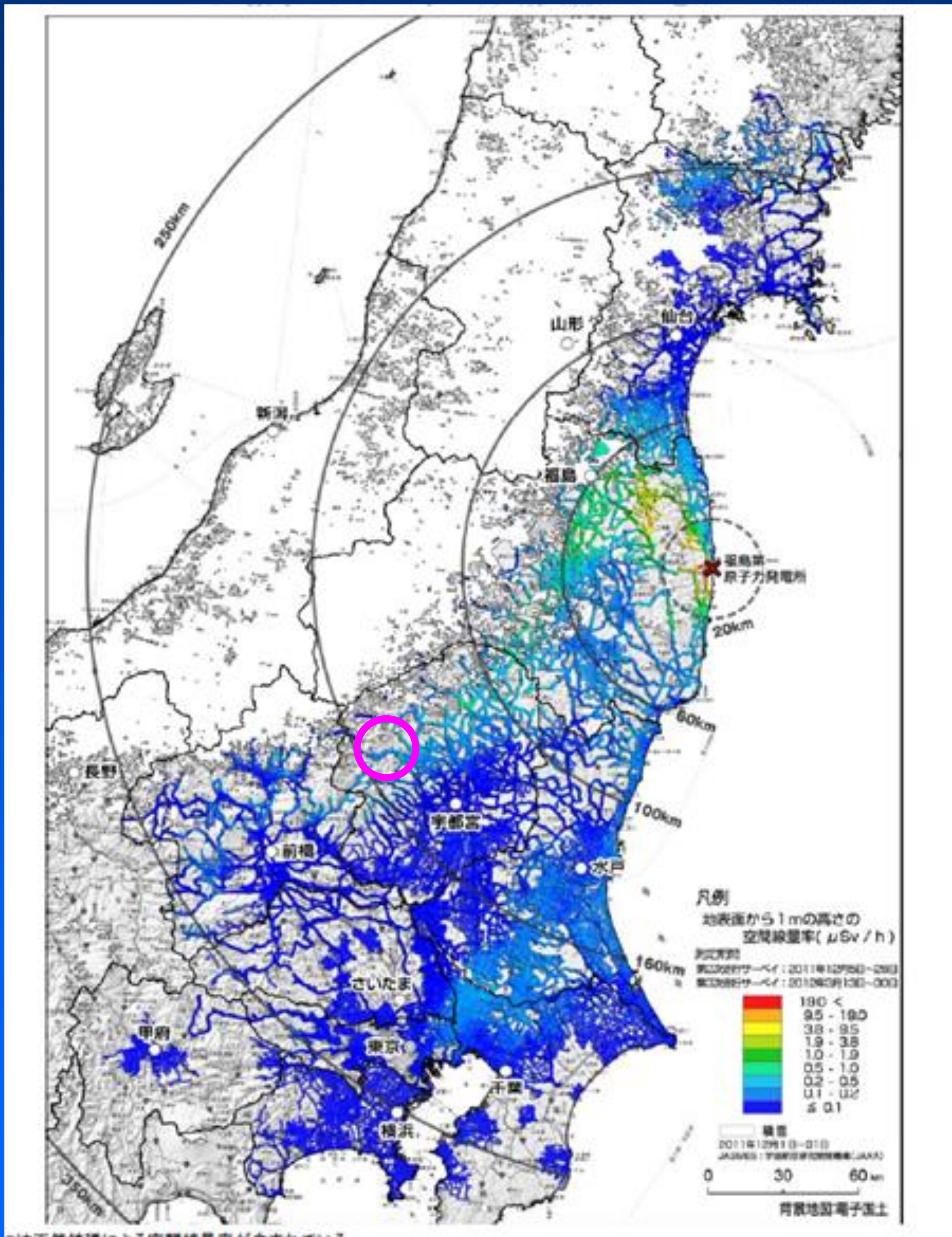


Fig. 7.5 In-vehicle unit of KURAMA-II. A CsI detector and a CompactRIO are compactly placed in a tool box 34.5×17.5×19.5 cm in size

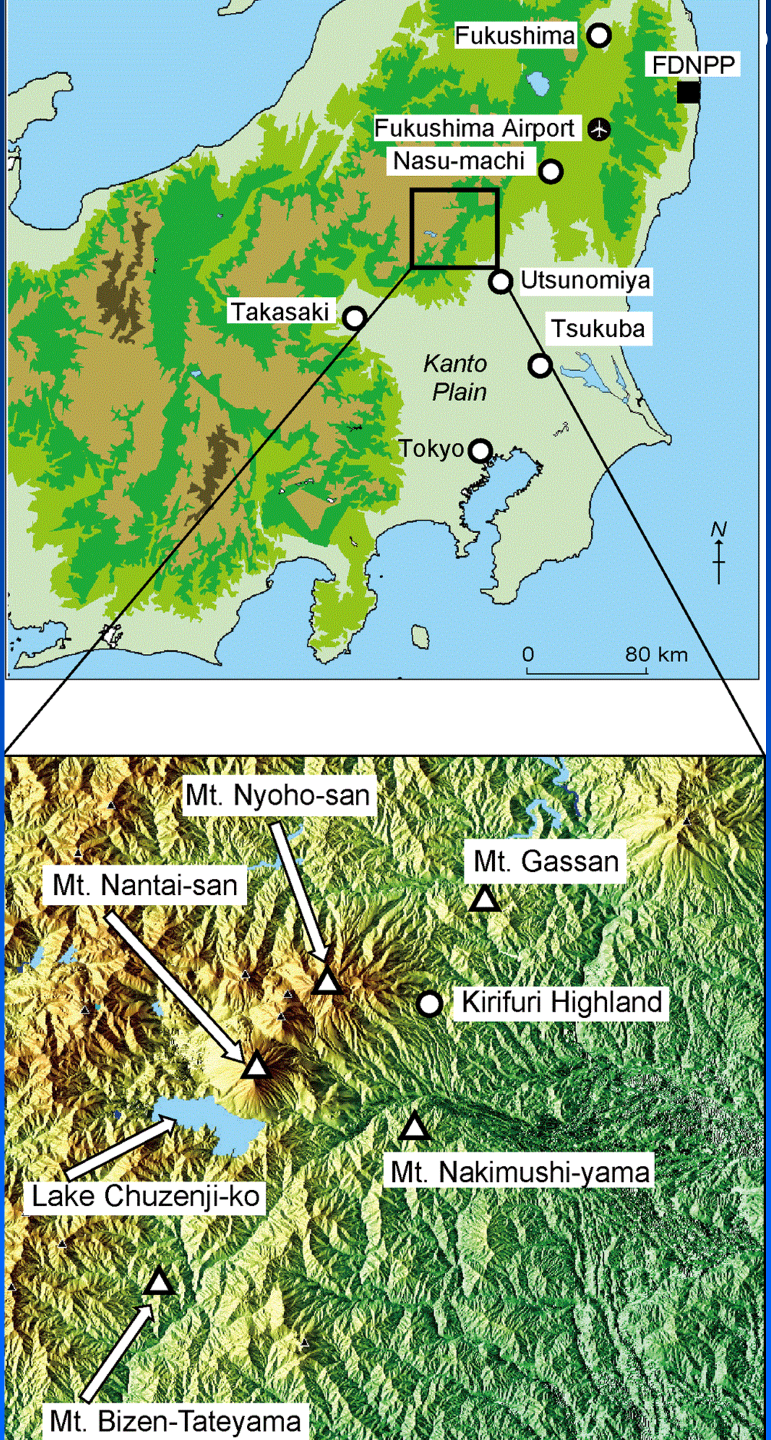
Radioactive contamination in a mountainous area north of Kanto Plain

On-foot survey

Objective area

Nikko Mountain area

120 km north of Tokyo
160 km southwest of FDNPP



Portable-type γ-ray detectors used in on-foot survey

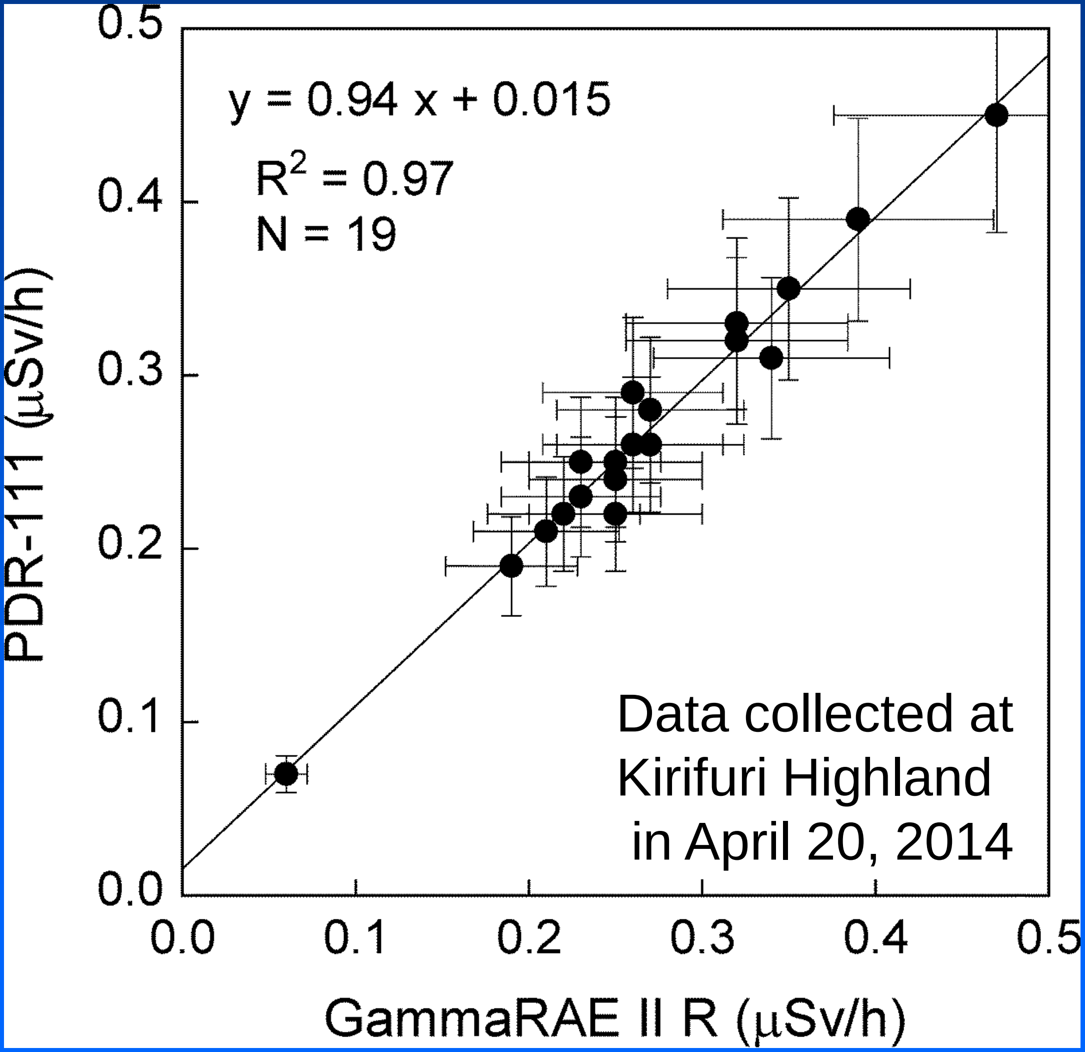
Gamma RAE II R (RAE systems)

…looks rugged and tough !

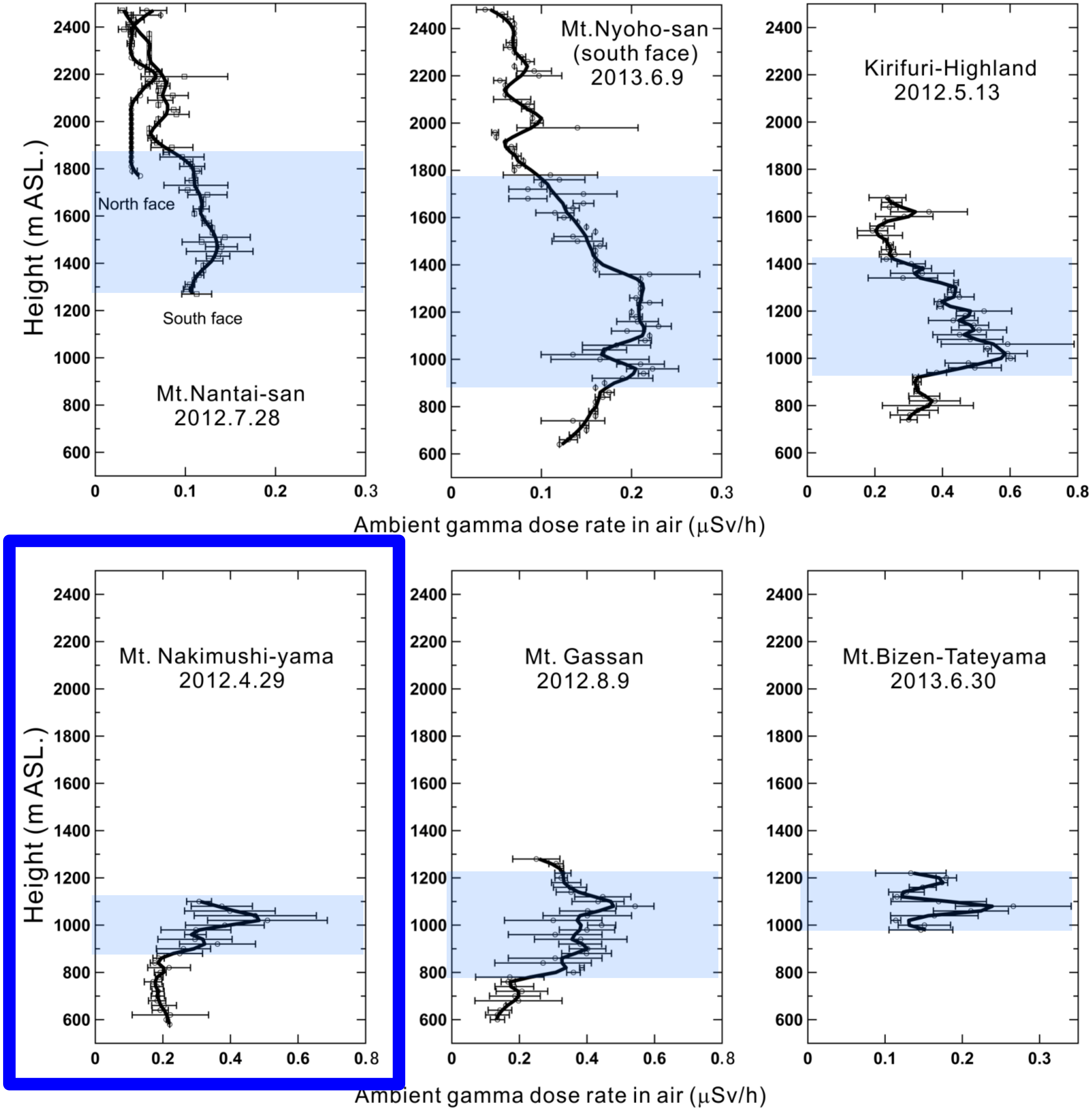
PDR-111 (Hitachi-Aloka Medical)

… looks fragile !

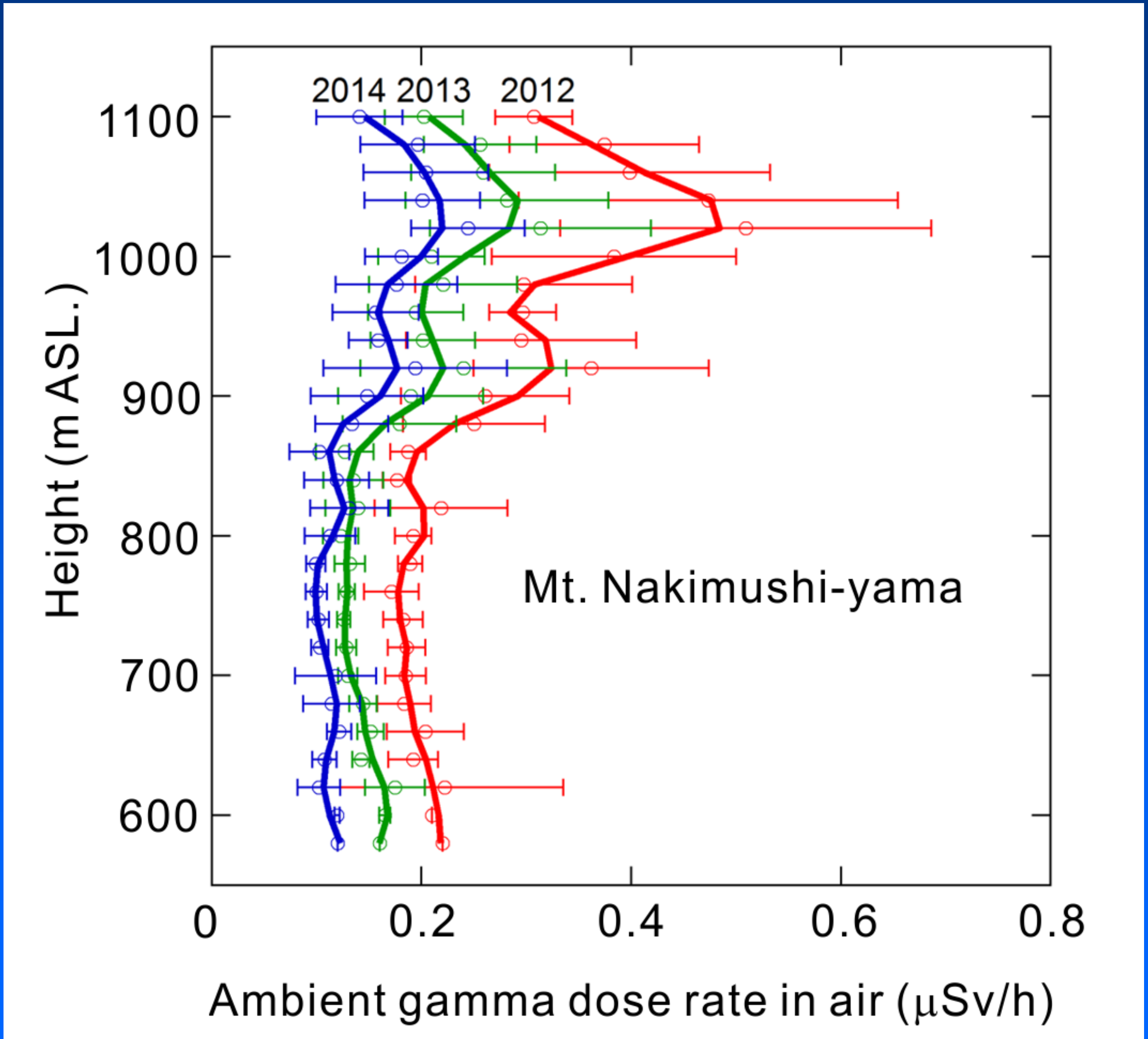
Comparison of the two instruments' readout



Altitudinal distribution of ambient γ dose rate in air in Nikko Mountain area


Mt.Nantai-san
2012.7.28
North face
South face
Mt.Nyoho-san
(south face)
2013.6.9
Kirifuri-Highland
2012.5.13
Mt. Nakimushi-yama
2012.4.29
Mt. Gassan
2012.8.9
Mt.Bizen-Tateyama
2013.6.30
Height (m ASL.)
Ambient gamma dose rate in air (μSv/h)

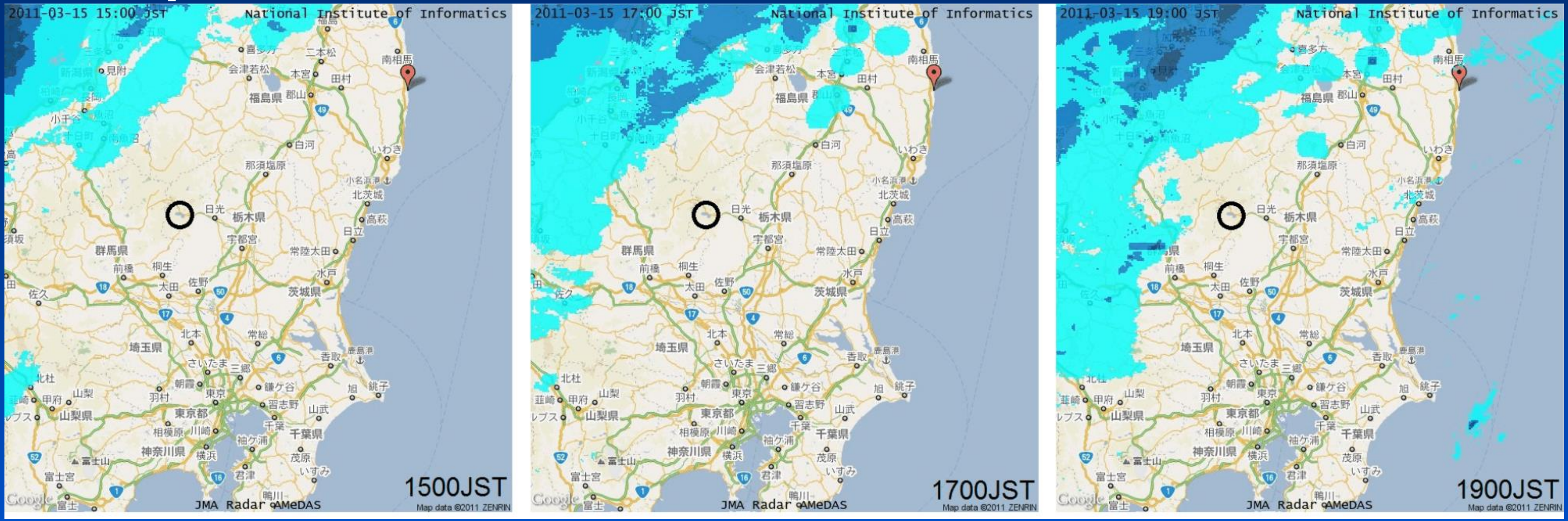
Decay of ambient γ dose rate in air in 3 years after the accident



If the distribution was formed by wet-deposition,

was it raining?

Precipitation in the afternoon of March 15



Radar / AMeDAS analysis chart

Nikko Mountain area

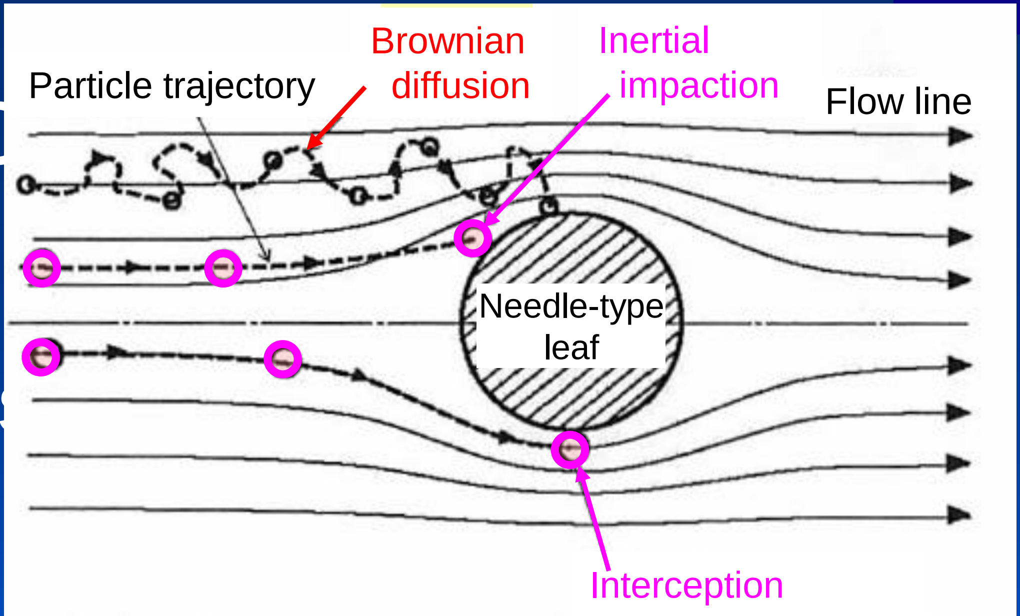
-0 0-1 1-2 2-4 4-8 (mm/h)

A hypothetical deposition mechanism

“Cloud / Fog / Oc

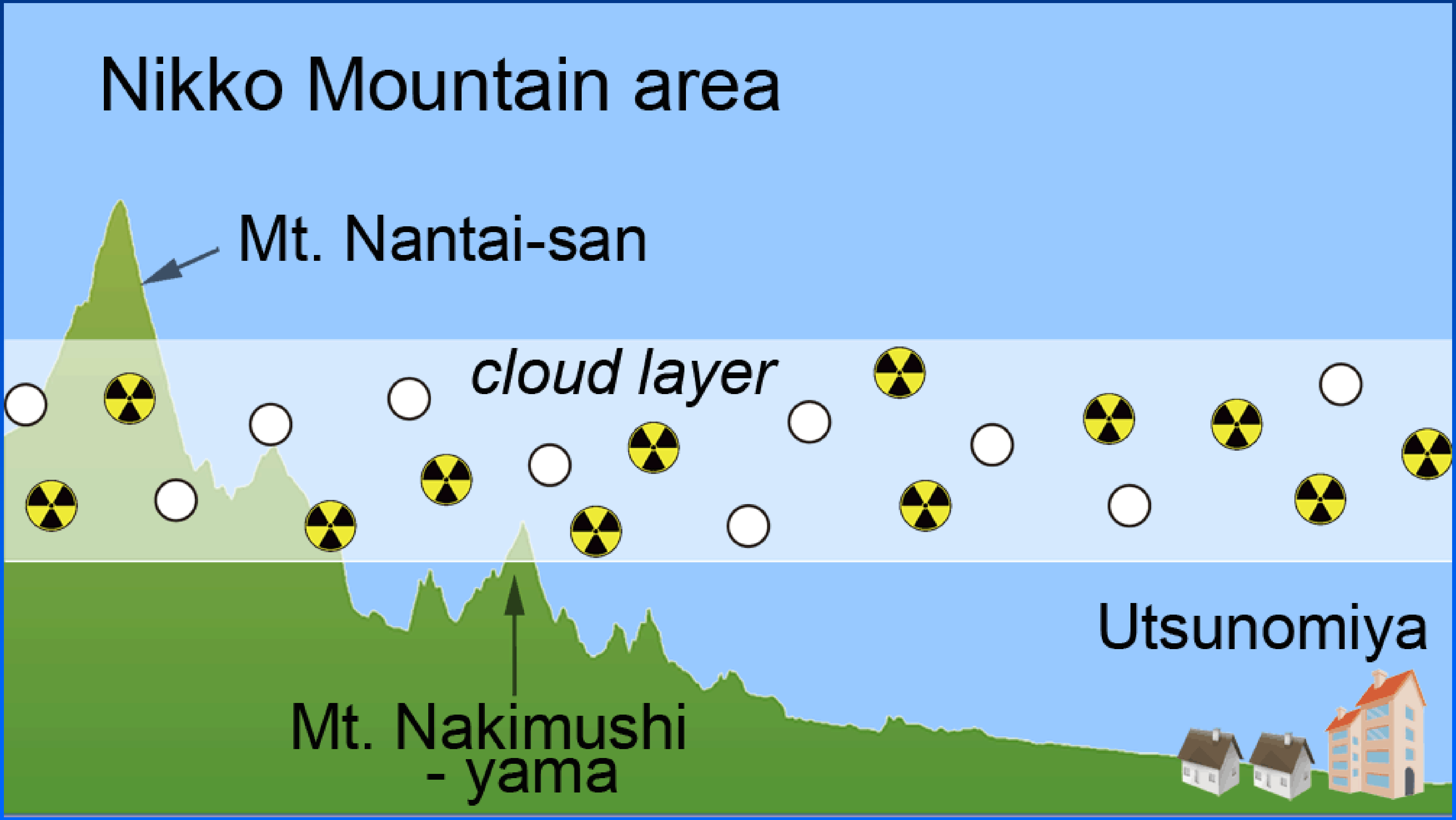
Water droplets with

several μm ～ s



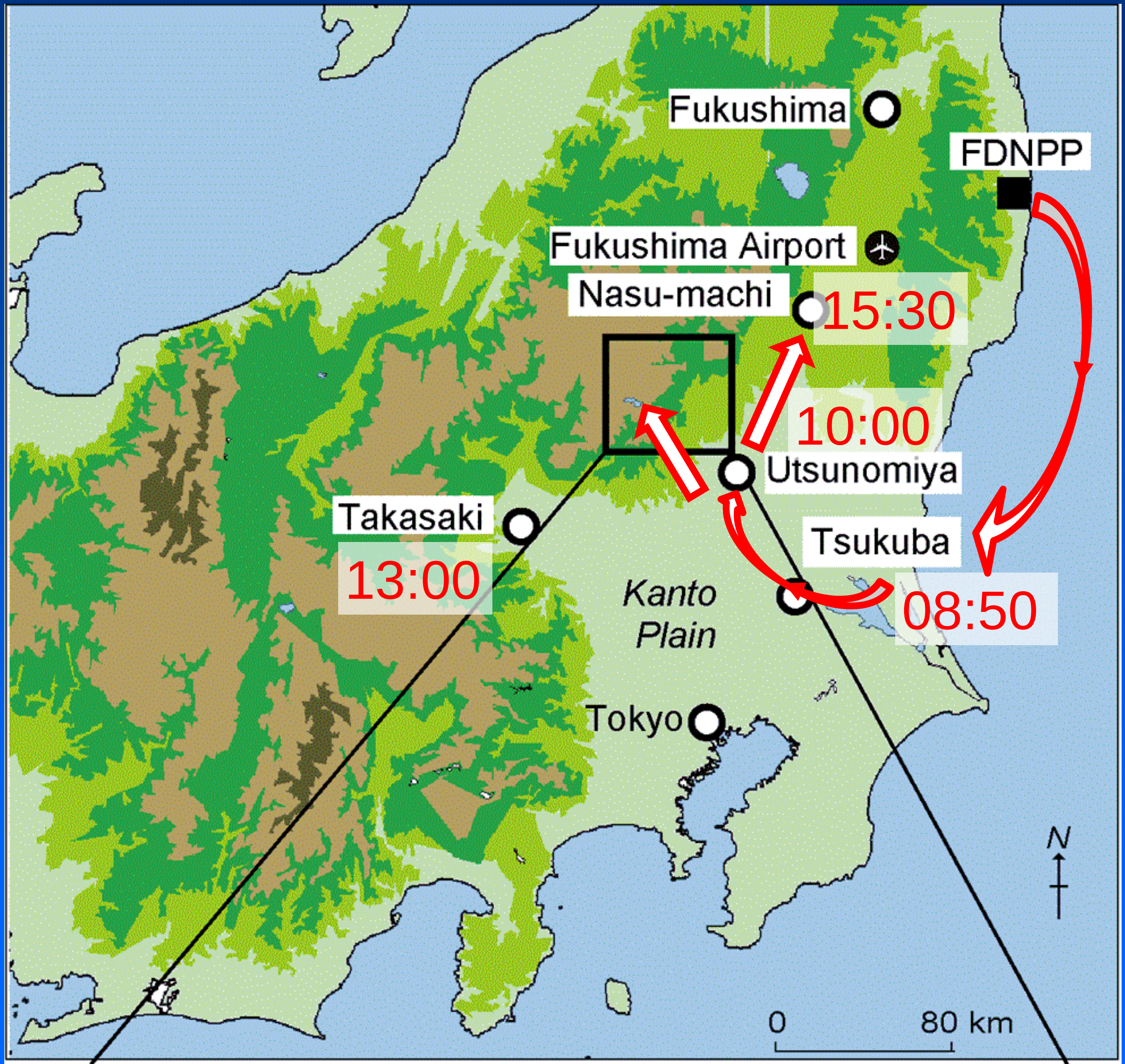
- Prevailing deposition mechanism
  - Accumulation mode aerosol (*e.g.*, sulfate)
    → Brownian diffusion
  - Cloud / Fog / Occult deposition
    → Inertial impaction, Interception, Gravitational settling

A hypothetical deposition mechanism of radionuclides in the area concerned



Arrival timing of radioactive plume at Nikko Mountain area

Transport path of radionuclides on Mar. 15



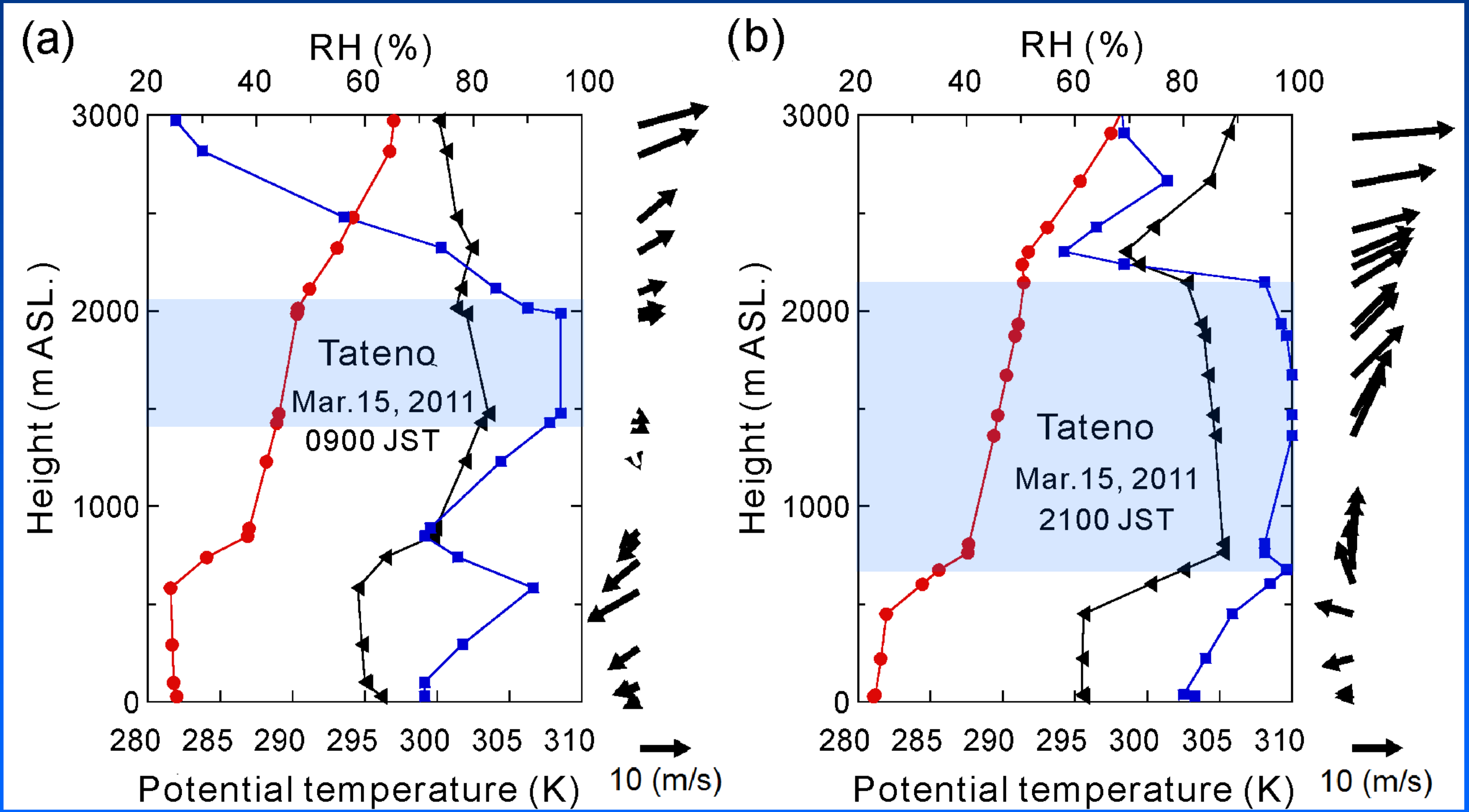
Expected arrival time

13:00 - 15:00?

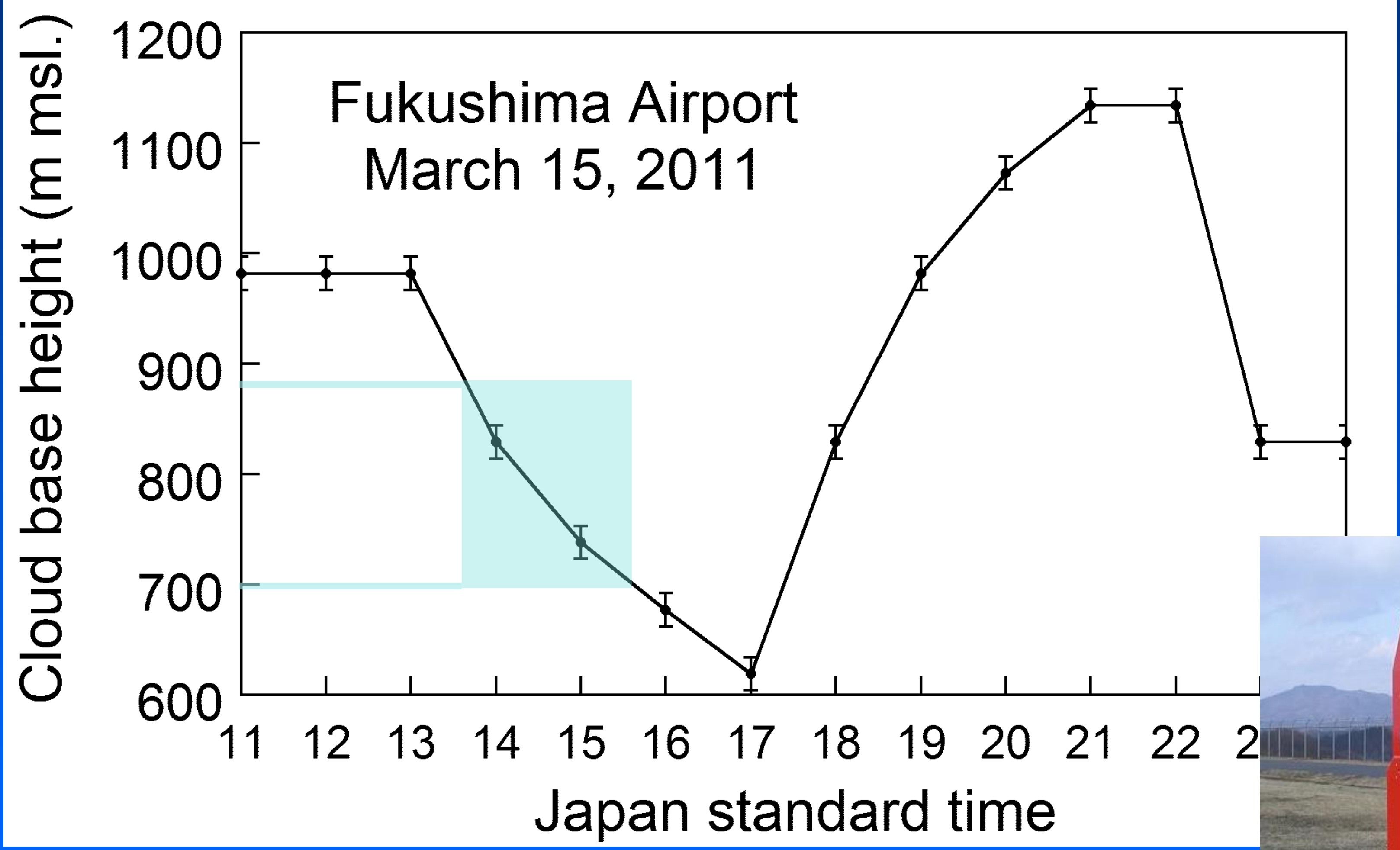
Was there clouds?

If so, which altitude ?

Upper sounding data at Tateno (Tsukuba) observatory



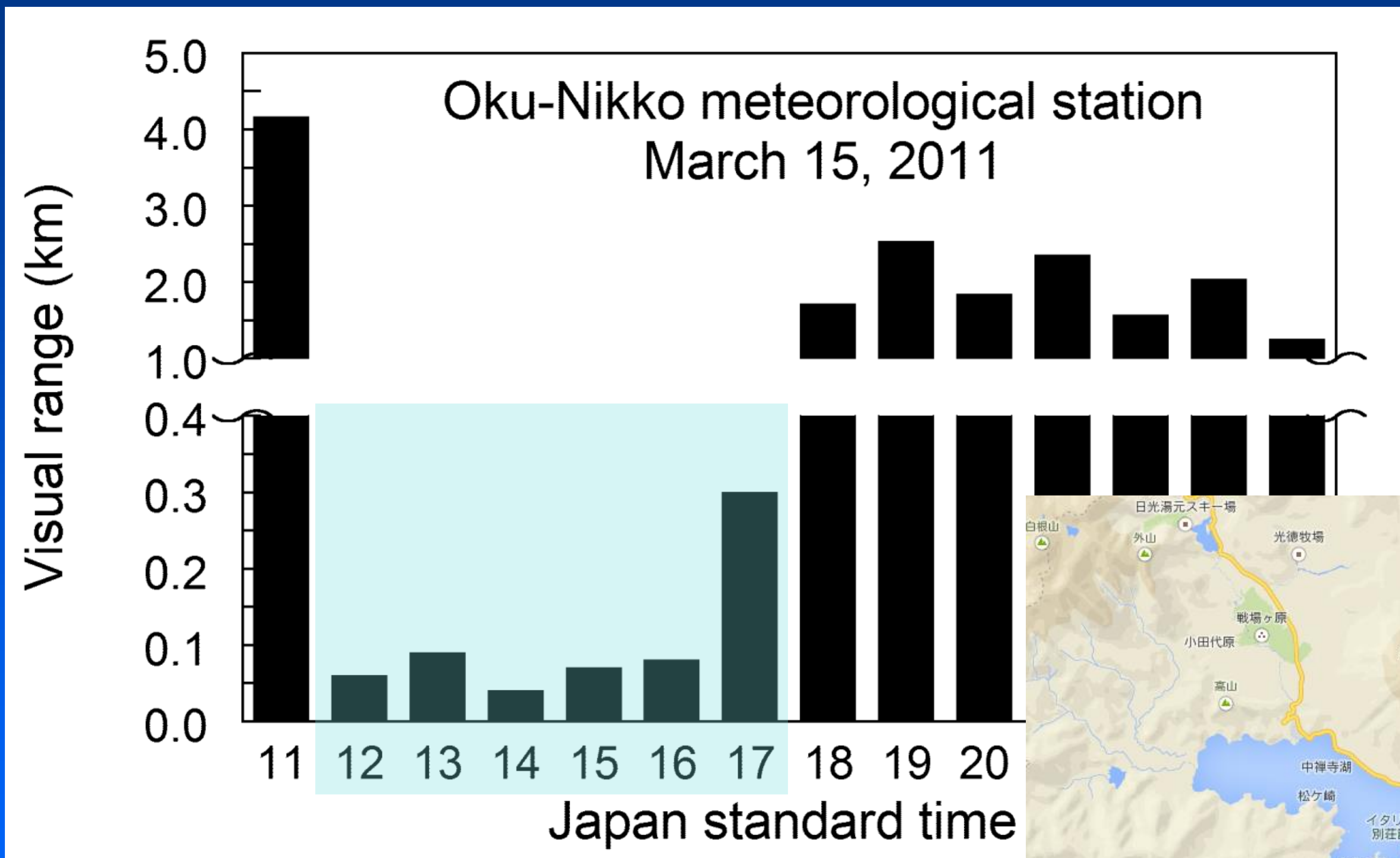
Cloud base height at Fukushima Airport



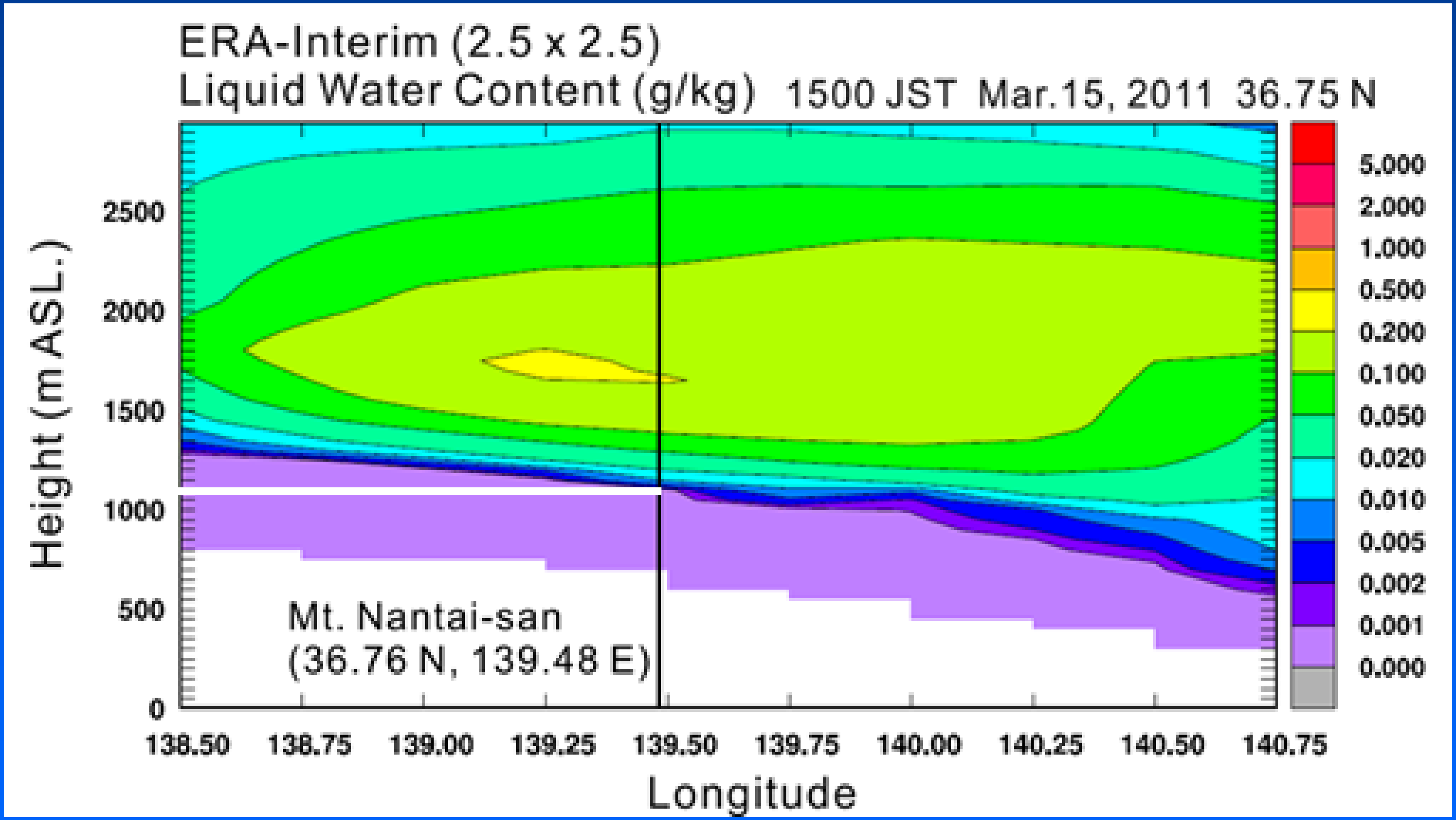
Ceilometer

Cloud base height in 14:00 –15:00 JST was approx. 700 – 900 m

Visual-range monitor at Oku-Nikko weather station (1292 m ASL.)

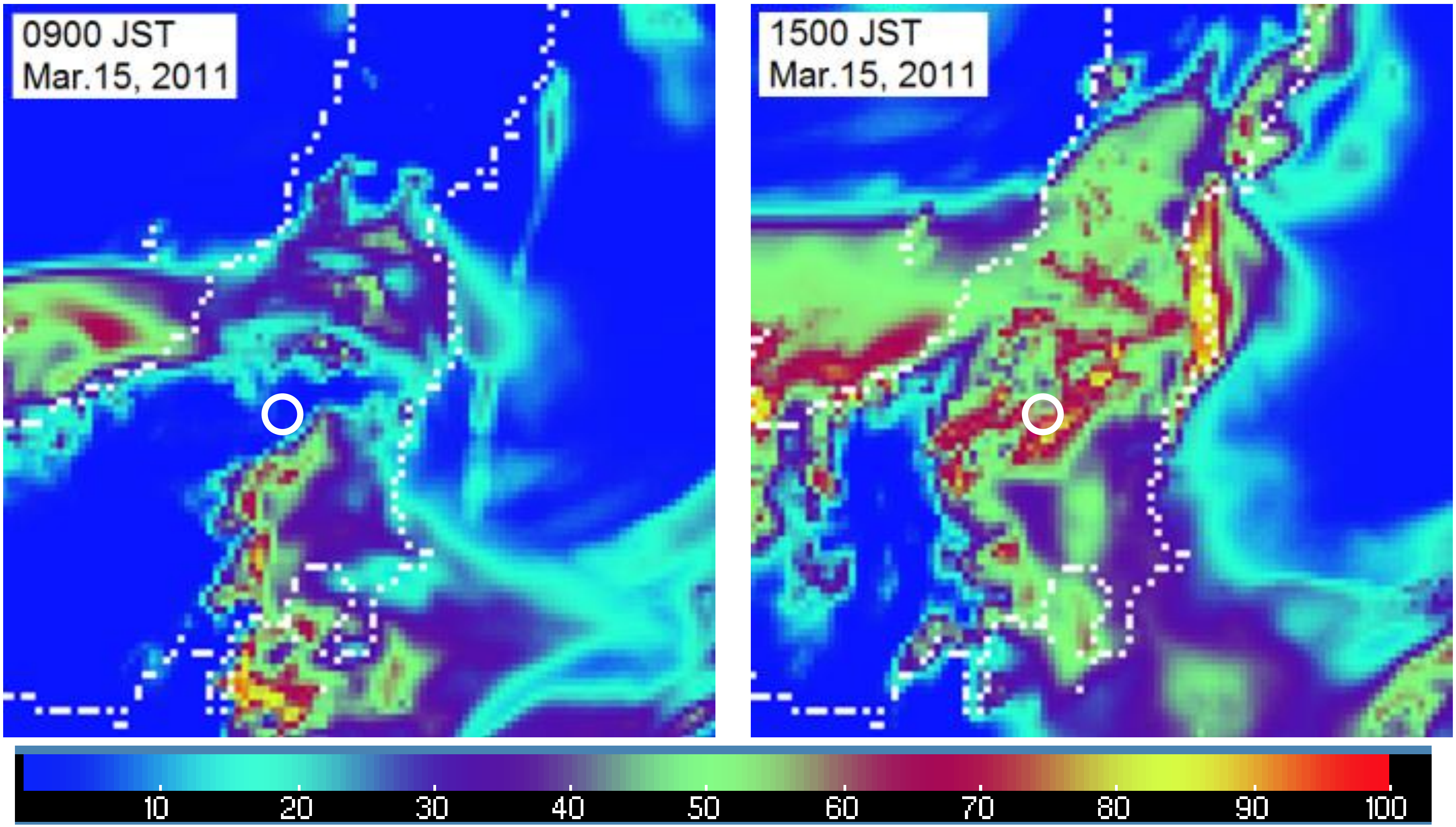


Liquid Water Content in air
reanalyzed in ECMWF ERA-Interim dataset



Cloud coverage of low stratus *in* the Grid Point Value data of JMA Mesoscale Model

(figure provided by Y. Takane, AIST)



Cloud coverage (%)

- Arrival timing of the radioactive plume

  → Early afternoon of Mar. 15, 2011

- Meteorological data

  → Cloud layer existed over the area from the early afternoon of Mar. 15

  → Cloud-base height : 700 - 900 m ASL.

- Ambient γ dose rate in air

  → Maxima at approx. 900 - 2000 m ASL.

⇩

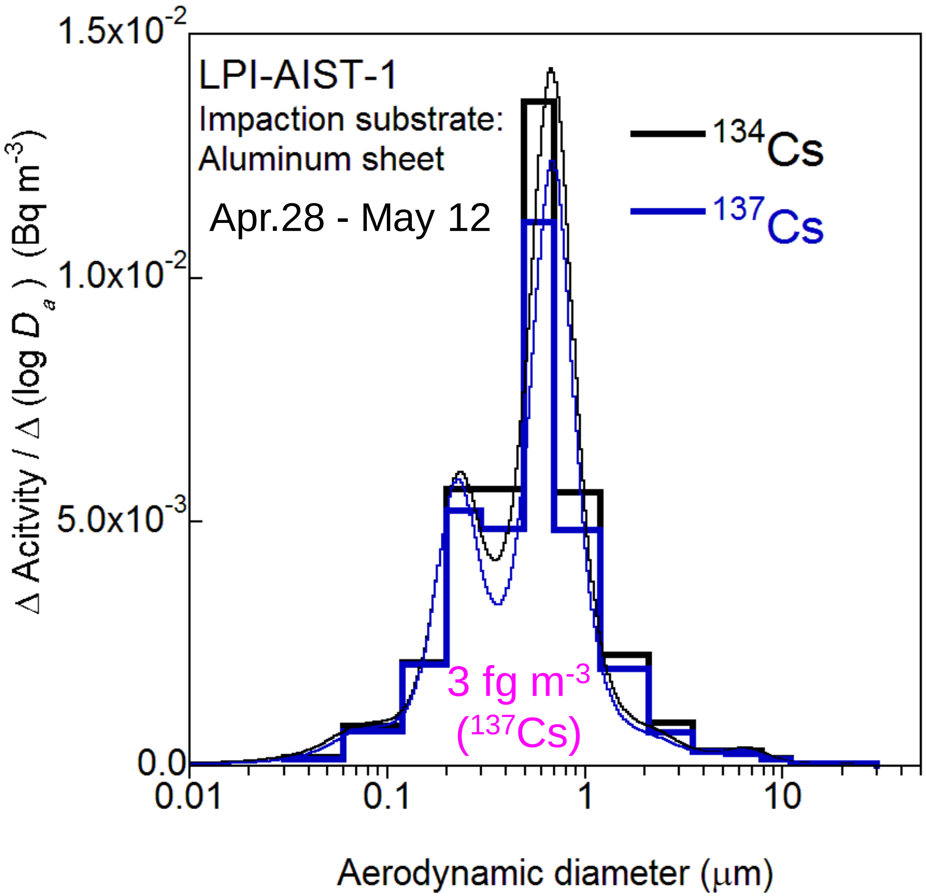
Radioactive contamination in Nikko Mountain area is caused by cloud / fog/ occult deposition

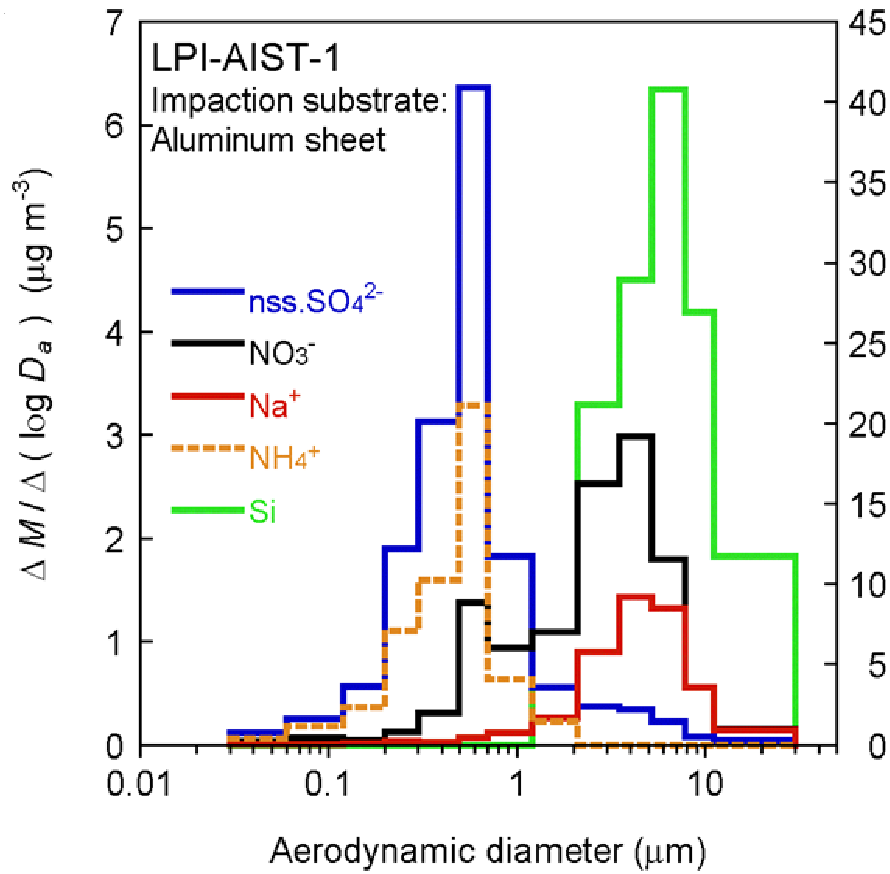
How were radionuclides

included in could droplets?

- *Physico-chemical* property -

Activity size dist. of $^{134,137}Cs$, and Mass size dist. of major aerosol components





- **Double peak** in the **Accumulation Mode** size range
- ^{137}Cs mass conc. 3 fg m^{-3} cannot form this size distribution

Airborne radiocesium in late April to May, 2011

- The overlapping size distributions
- 100% water-soluble
- Double-peak stricture in activity size dist.

suggesting…

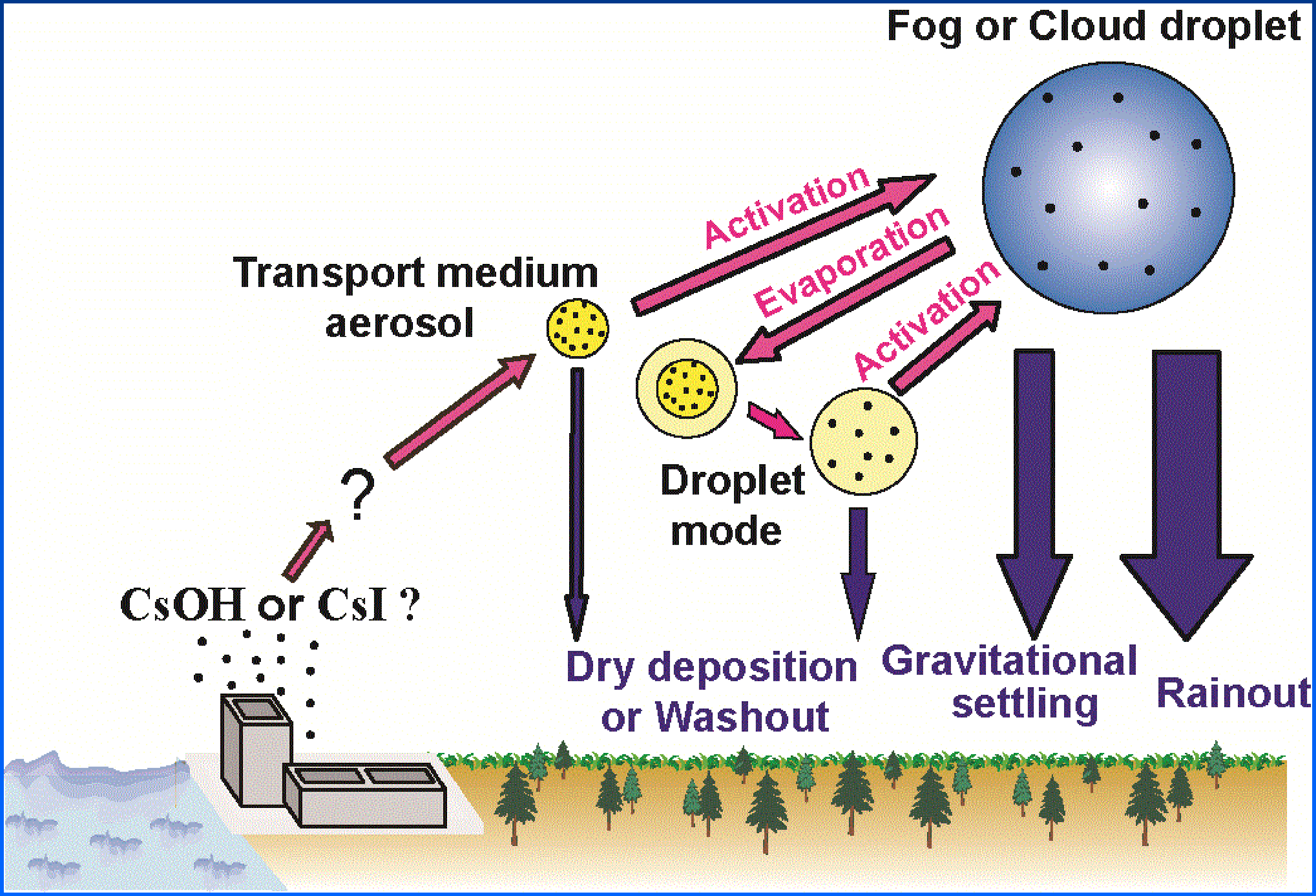
Transport medium...”carrier”

was

sulfate aerosol

Proposed behavior of radiocesium discharged from FDNPP in late April to May, 2011

Kaneyasu *et al*. (2012)

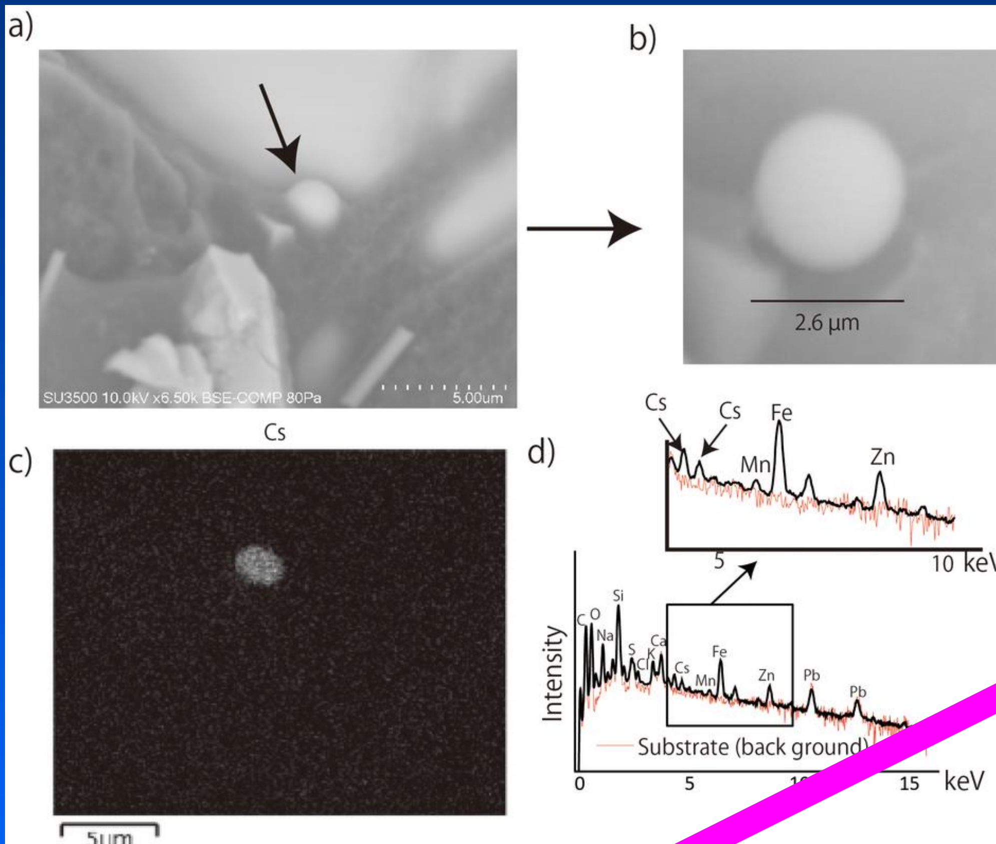


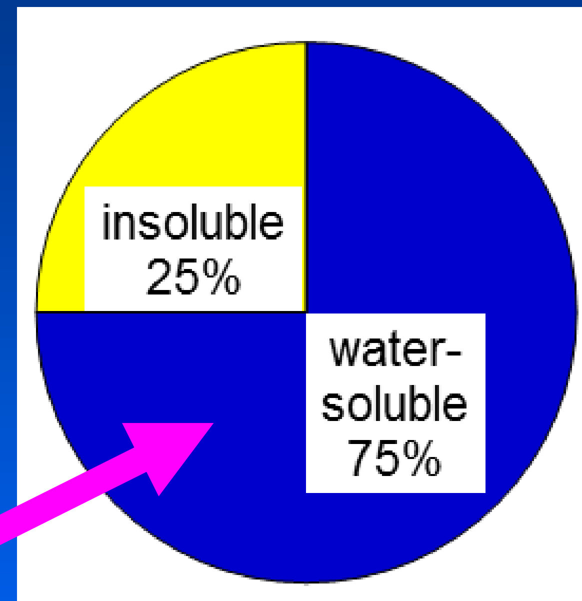
Another type of

radioactive particles

to be considered

Cs bearing insoluble particles in a filter sample collected on Mar.15 at Tsukuba (Adachi *et al.* 2013)





- Geometric diameter : 2.6 µm …. fairly large!
- 69.5～82.0 % of ^{137}Cs is extractable by nitric acid solution
- 2 particles out of 100 hot-spots in imaging plate photo

Physico-chemical properties of radionuclides,

such as size and water-solublity

… may affect the spatial distribution of contaminated area,

via deposition process

To fully reproduce it in numerical models,

- Formation of cloud droplets by the activation of CCN, which comprises a curtain fraction of aerosols

and

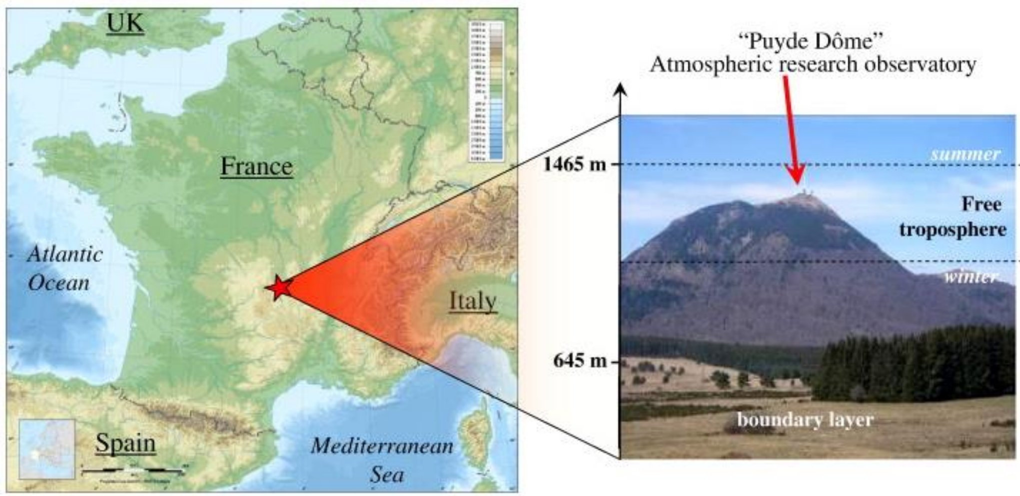
- Subsequent formation of precipitation and rain-out of radionuclides, via
  - ✓ "cold-rain" (Bergeron-Findeisen) process
  - ✓ riming by water or mixed-phase cloud

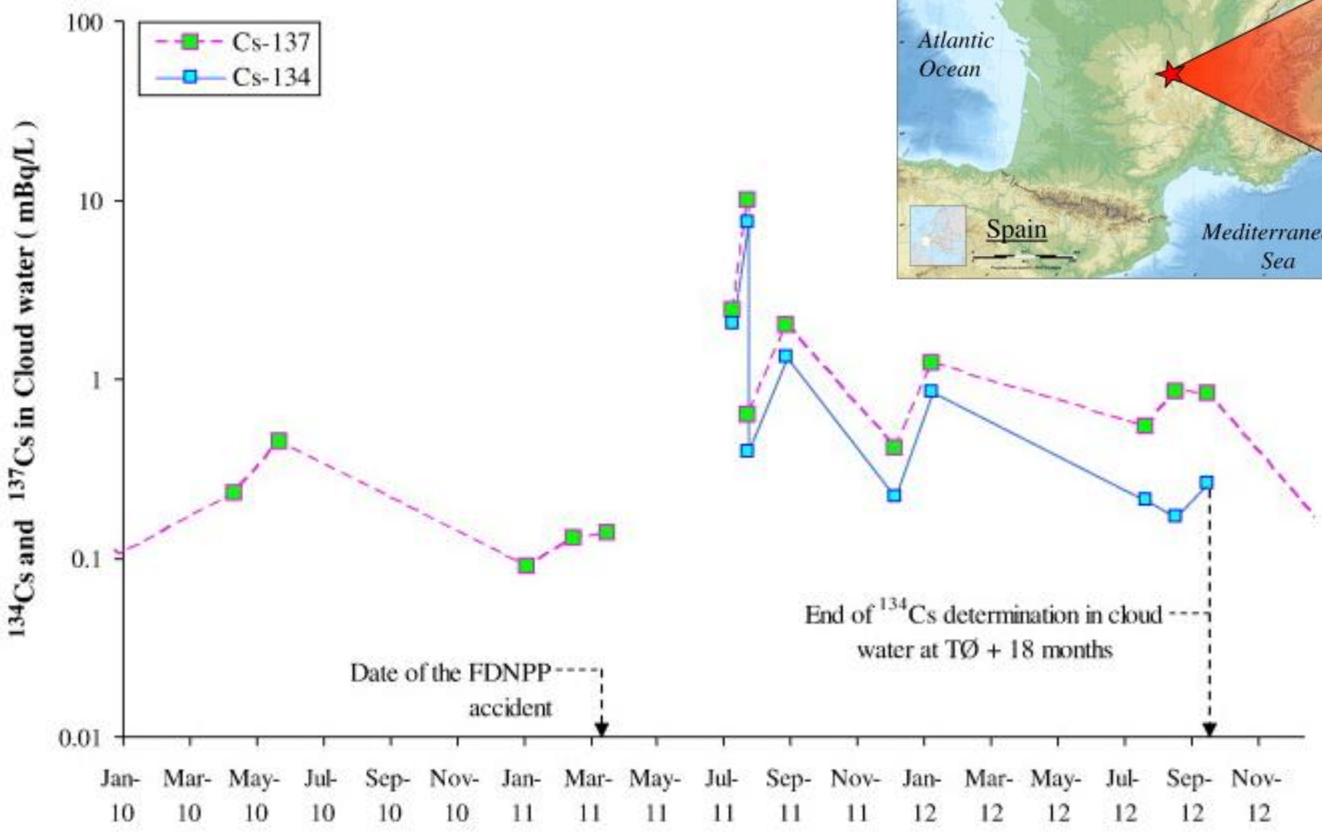
have to be considered…

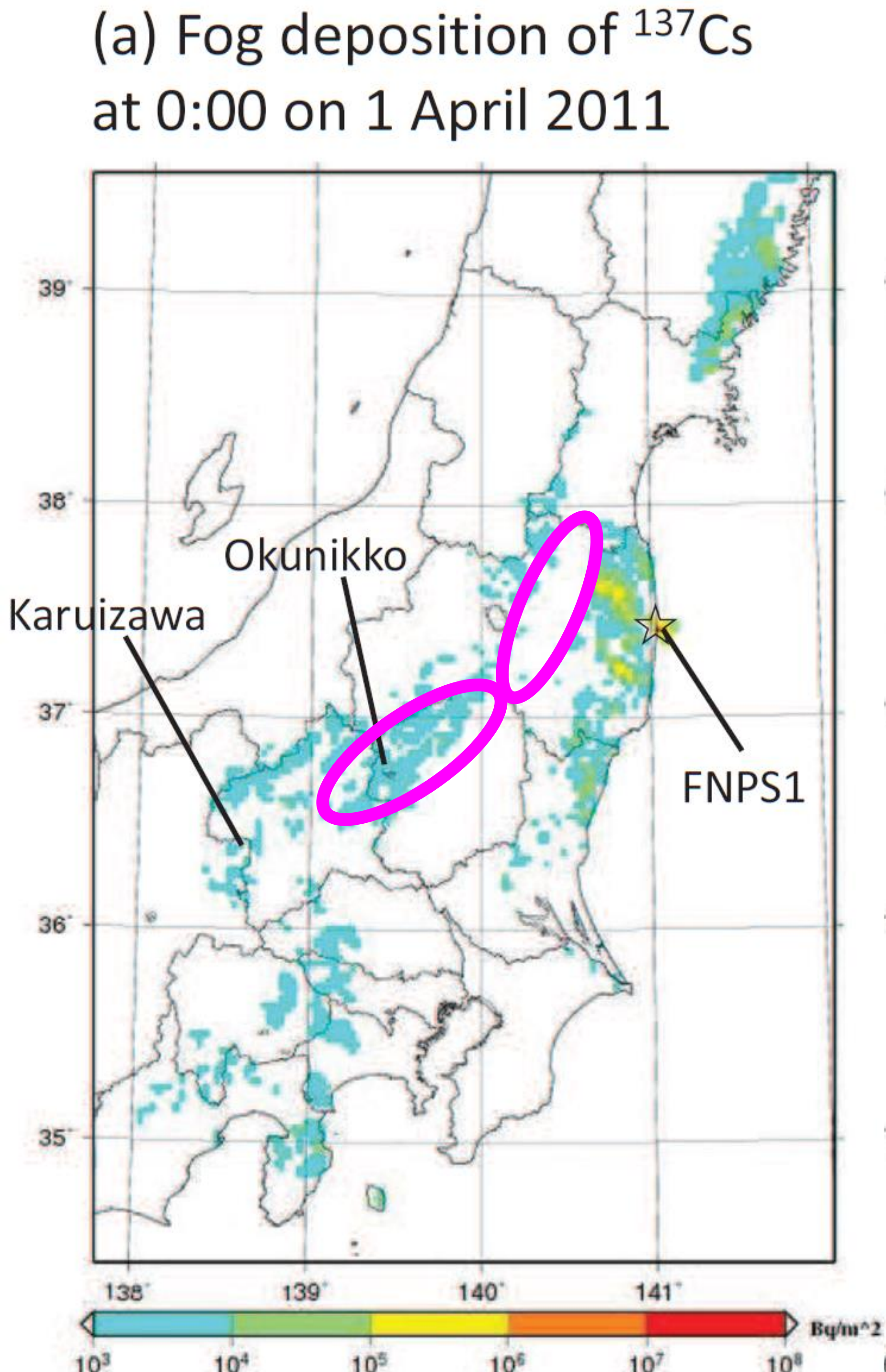
Related studies

Radiocesium in cloud water collected at Mt. Puyde Dôme, France

(Masson et al., 2014)







Katata et al. (2014)

Simplified fog deposition scheme was introduced in their WSPEEDI- II model

→ qualitatively reproduced the deposition pattern in mountain areas concerned

Thank you for your attention

Environmental Science & Technology

Article

Previous Art

Altitude-dependent Distribution of Ambient Gamma Dose Rates in a Mountainous Area of Japan caused by the Fukushima Nuclear Accident

Mutsuo Hososhima and Naoki Kaneyasu

Environ. Sci. Technol., Just Accepted Manuscript
DOI: 10.1021/es504838w
Publication Date (Web): February 23, 2015

CAS Section: Air Pollution and Industrial Hygiene

Abstract

Large amounts of airborne radionuclides were deposited over a wide area in eastern Japan, including mountainous regions, during the devastating Fukushima Dai-ichi nuclear power plant accident. Altitudinal distributions of ambient gamma dose rate in air were measured in a mountainous area at the northern rim of the Kanto Plain, Japan, using a portable instrument carried along the mountain trails. In the Nikko Mountain area, located 120 km north of Tokyo, the altitudinal distribution exhibited maxima at about 900–2,000 m above sea level (ASL). This area was not affected by precipitation until 2300 Japan Standard Time (JST) on March 15, 2011. By that time, a substantial amount of radionuclides had been transported from the damaged reactor, according to the numerical simulations using transport models. Meteorological sounding data indicated that the corresponding altitudes were within the cloud layer. A visual-range monitor deployed in an unmanned weather station at 1,292 m ASL also recorded low visibility on the afternoon of March 15. From these findings, it was deduced that the altitude-dependent radioactive contamination was caused by the cloud/fog deposition process of the radionuclides contained in aerosols acting as cloud condensation nuclei.

This work has just been published online in *ES&T* as "just accepted manuscript" (Feb. 23, 2015)

Another important issue

related to the cloud/fog/occult deposition

“edge effect”

Sellafield accident (1957)

(Z Ould-Dada et al., 2002)

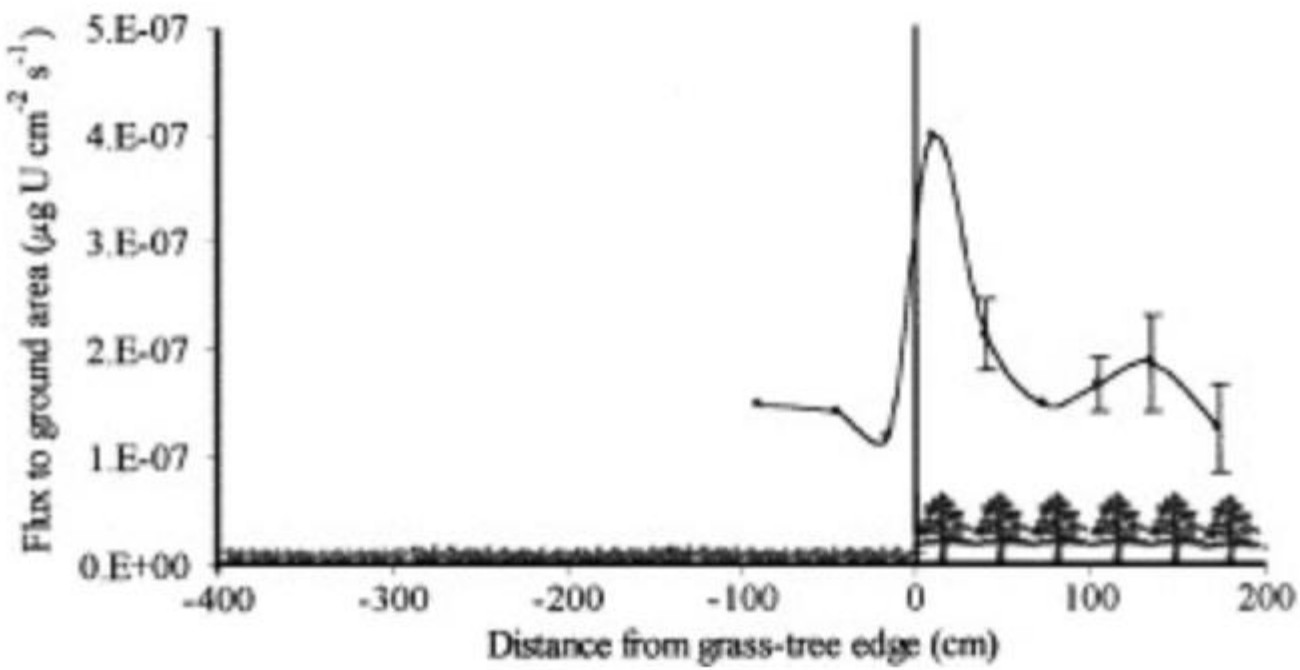


Fig. 5.

Variation of uranium aerosol flux to grass and spruce with distance from grass–tree edge.

“edge effect”

Kyshtym (1957) and Chernobyl (1986) accidents

(Tikhomirov and Shcheglov, 1994)

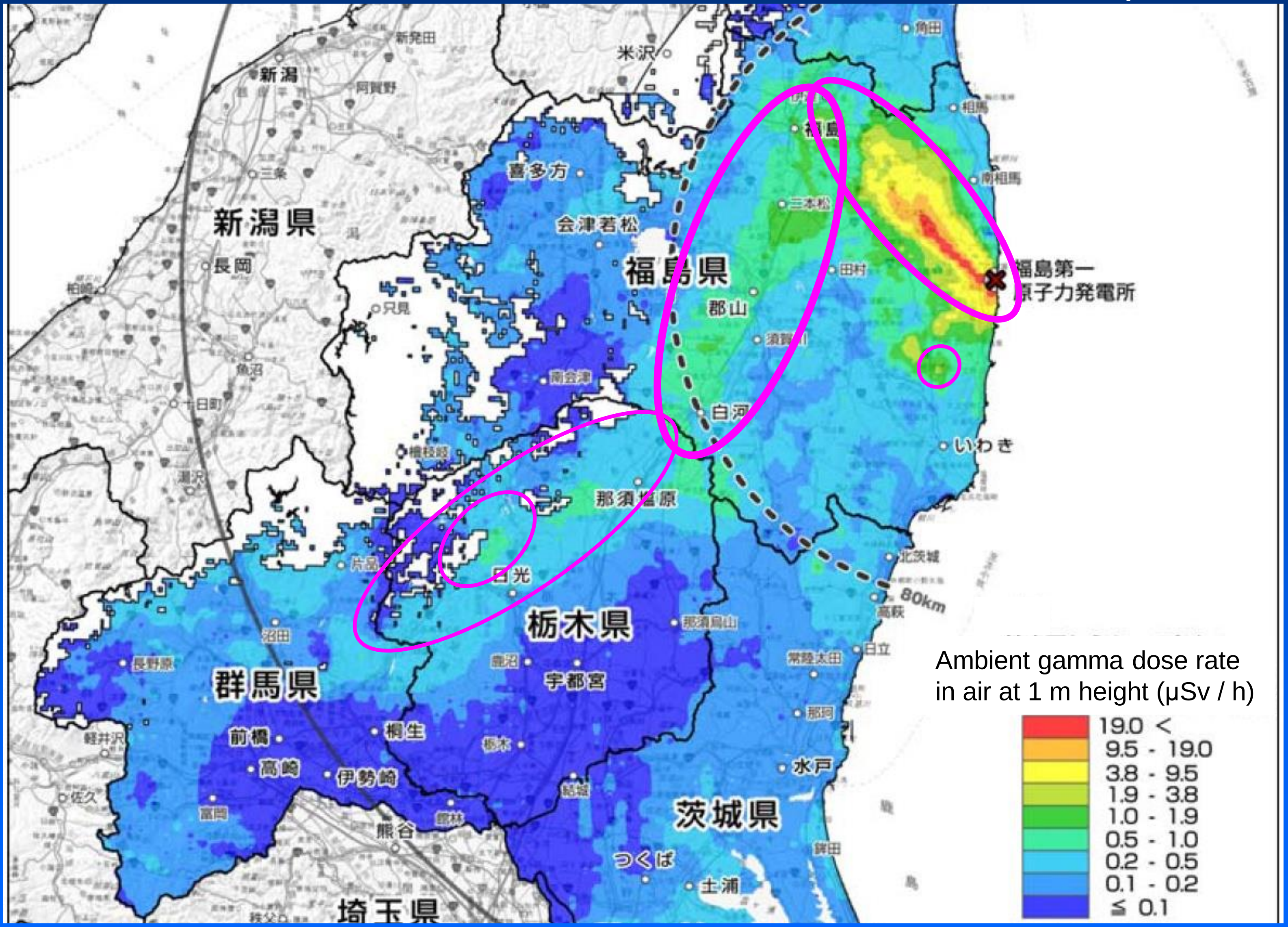
“A two- to fivefold elevated radionuclide precipitation is observed at the forest edges facing the ejection source and as far as 20-50 m deep into the forest …”

A possible case of

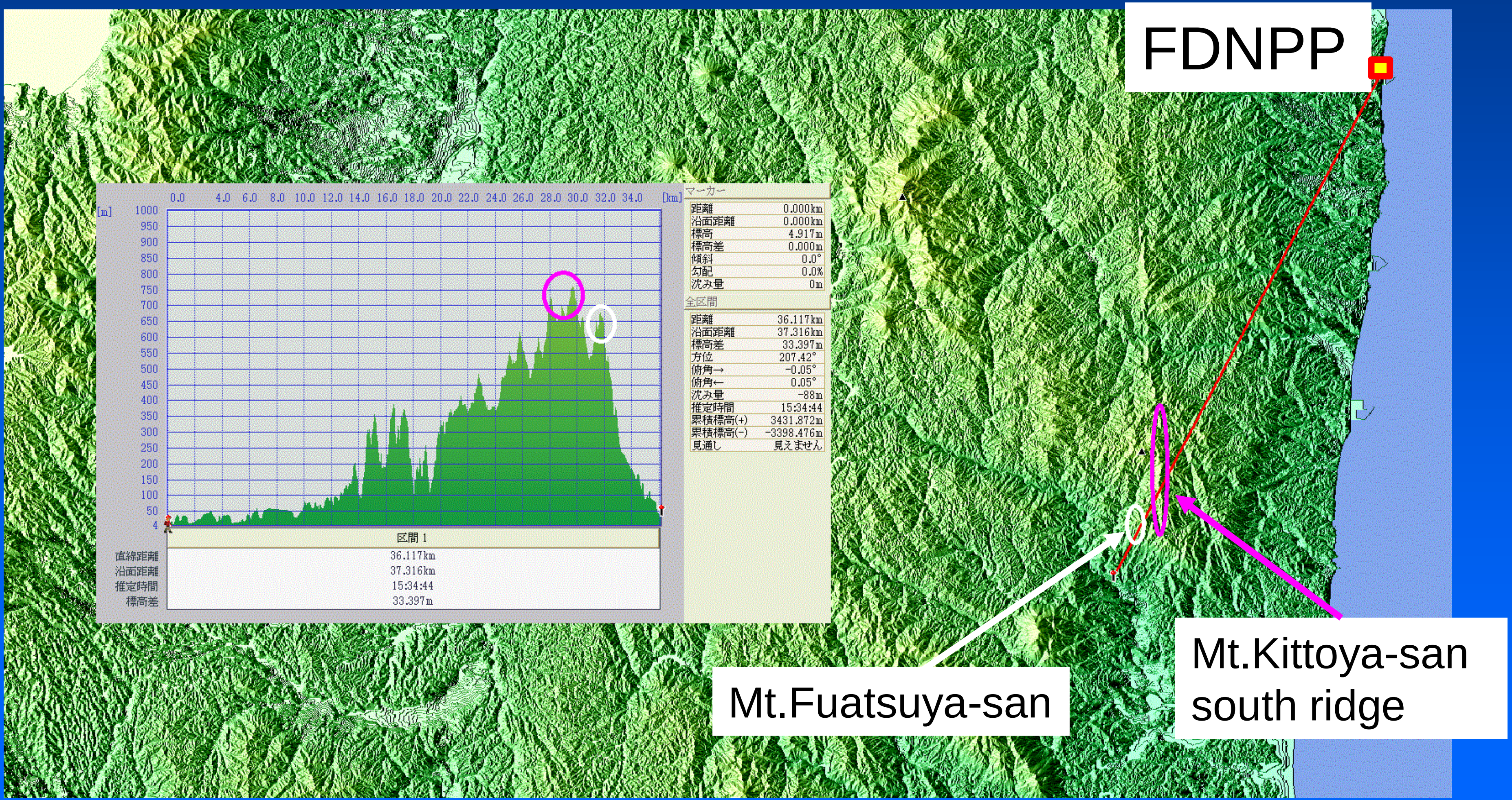
topographically formed cloud

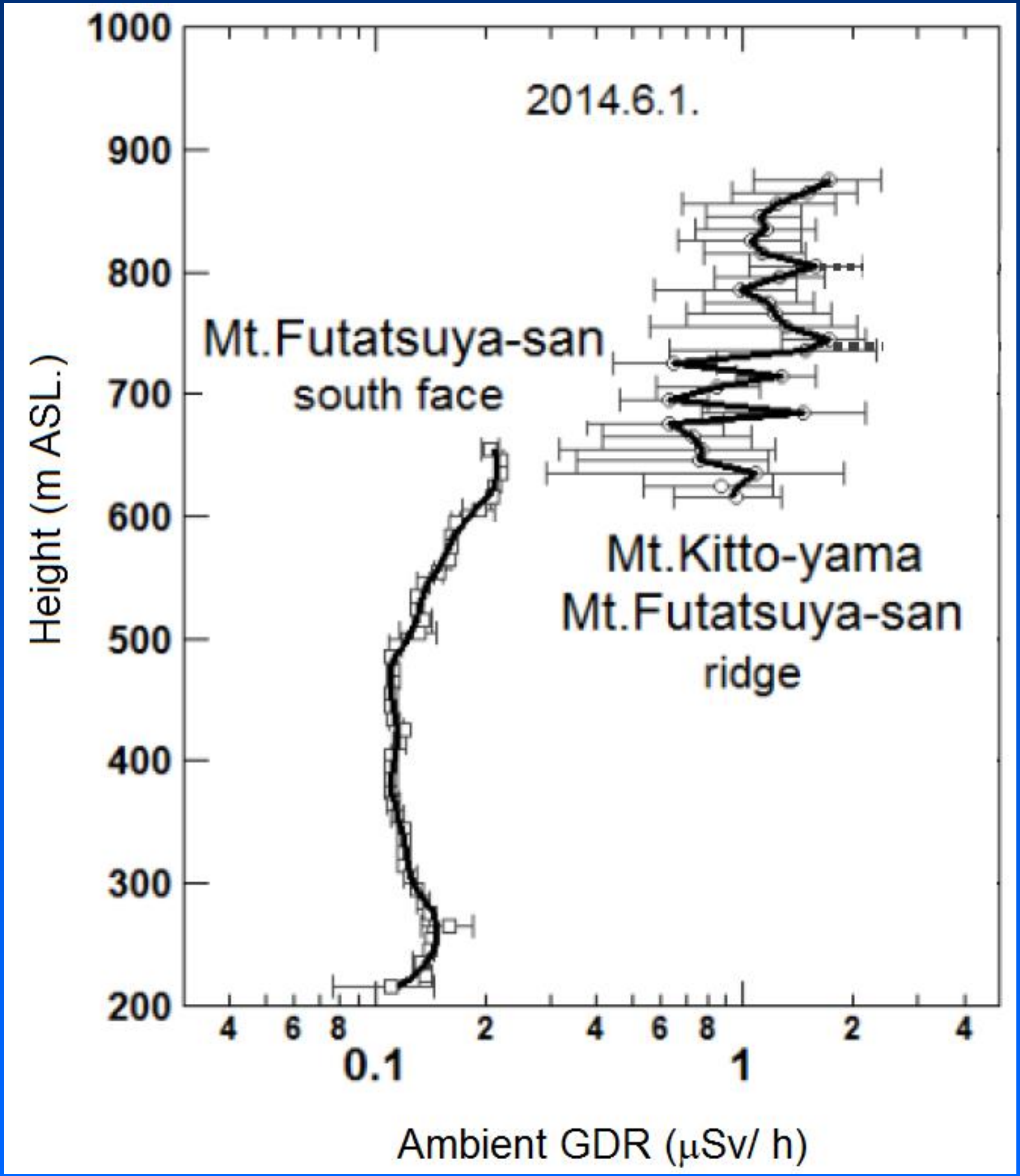
Airborne monitoring of ambient gamma dose rate

(June 28, 2012)

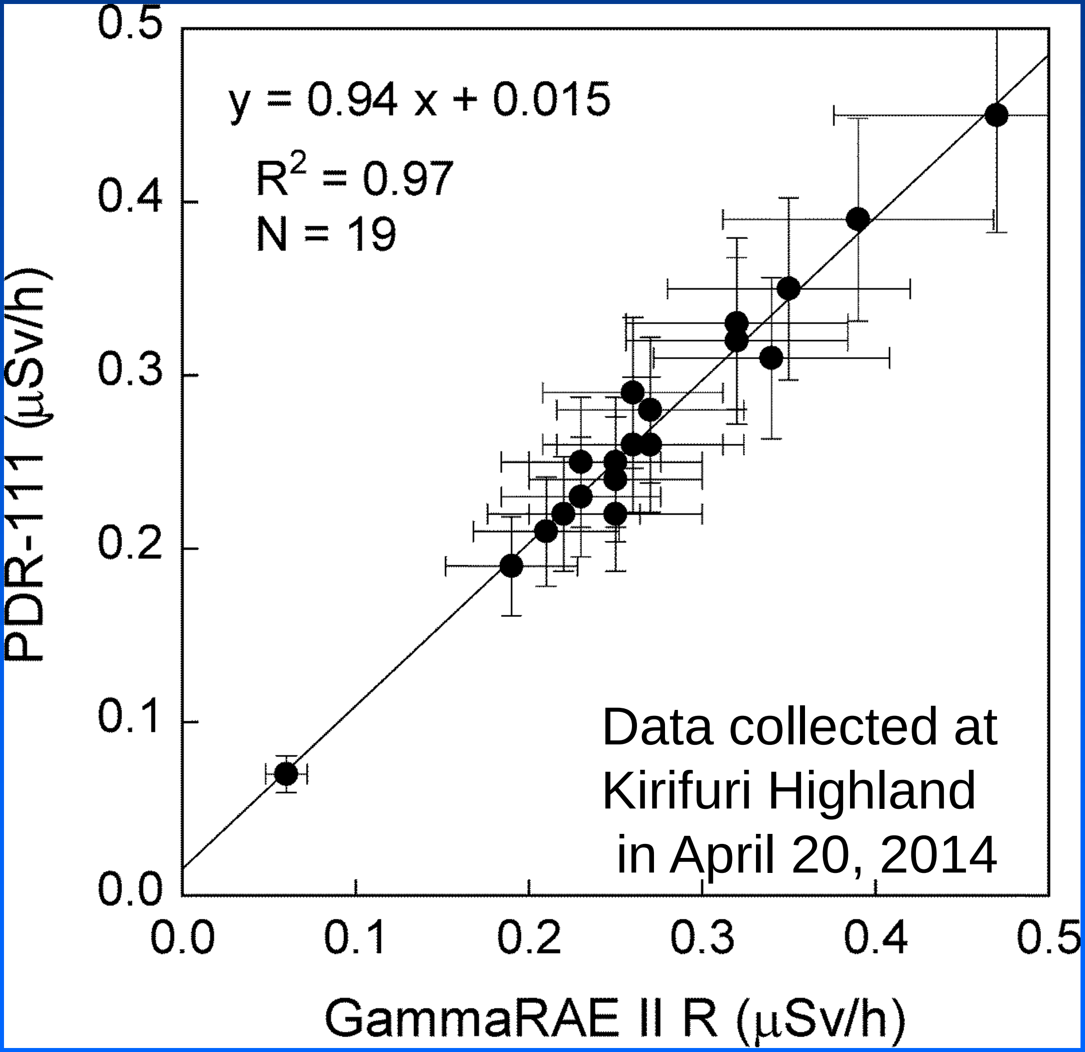


Mountains 25 km SSW of FDNPP




2014.6.1.
Mt.Futatsuya-san
south face
Mt.Kitto-yama
Mt.Futatsuya-san
ridge
Height (m ASL.)
1000
900
800
700
600
500
400
300
200
4
6
8
0.1
2
4
6
8
1
2
4
Ambient GDR (μSv/ h)

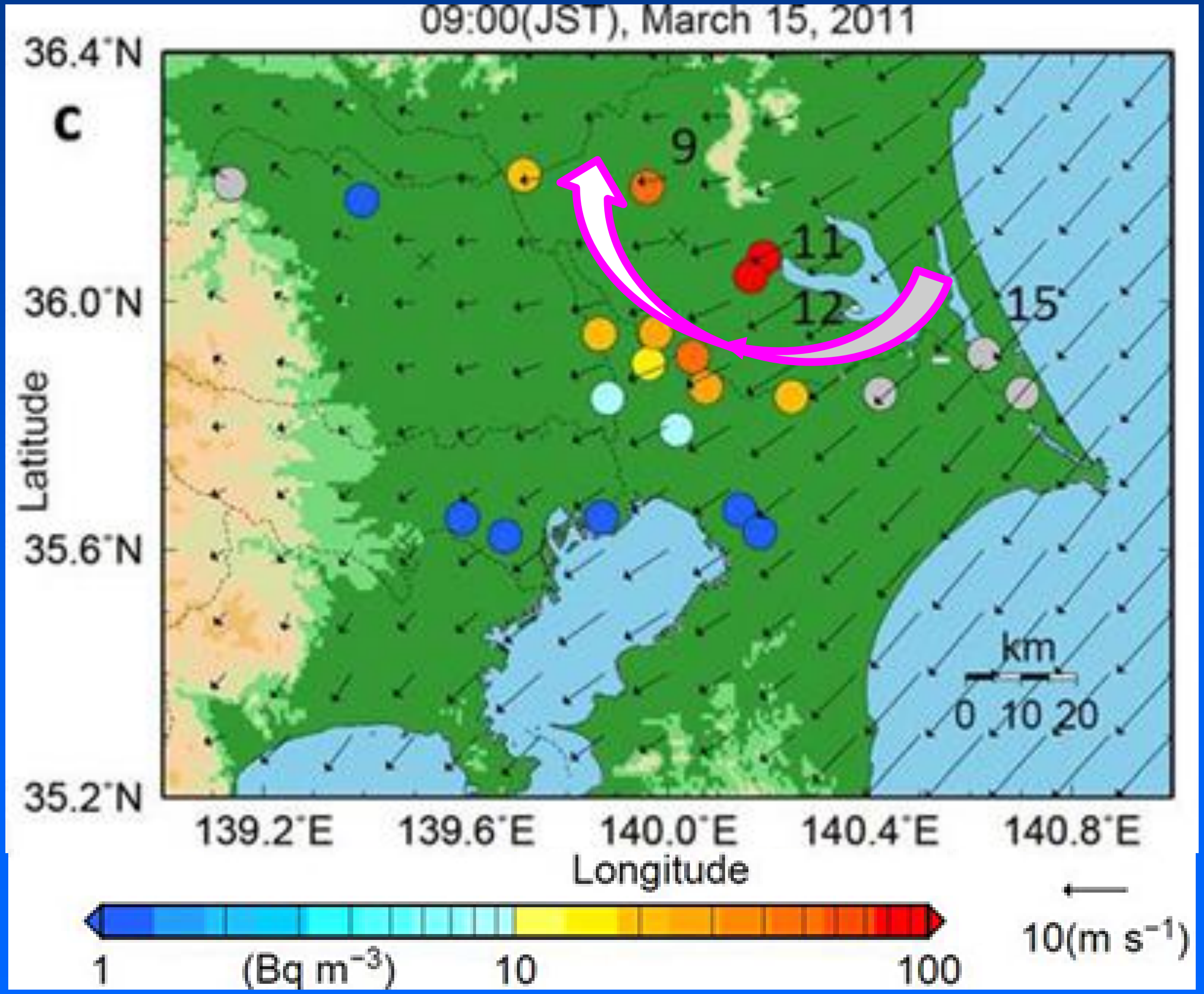
Comparison of the two instruments' readout



Kirifuri Highland on April 20, 2014

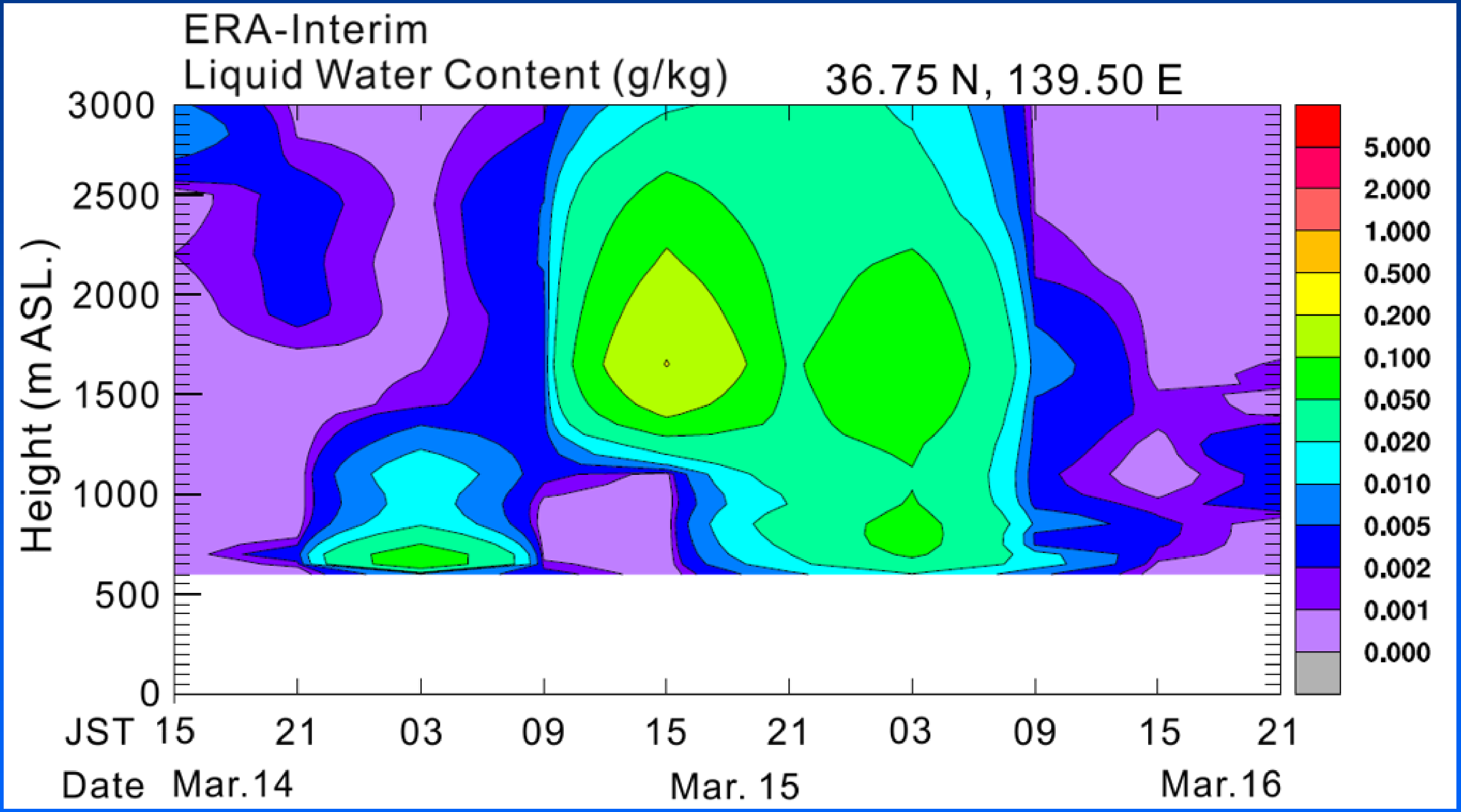
^{137}Cs activity con. measured from tape-filters of automated particulate matter monitors

(Tsuruta et al., 2014)



Liquid Water Content in air

reanalyzed in ECMWF ERA-Interim dataset



Solubility of Chernobyl-derived $^{134,137}Cs$ in aerosols collected at Tenenessee in 1986

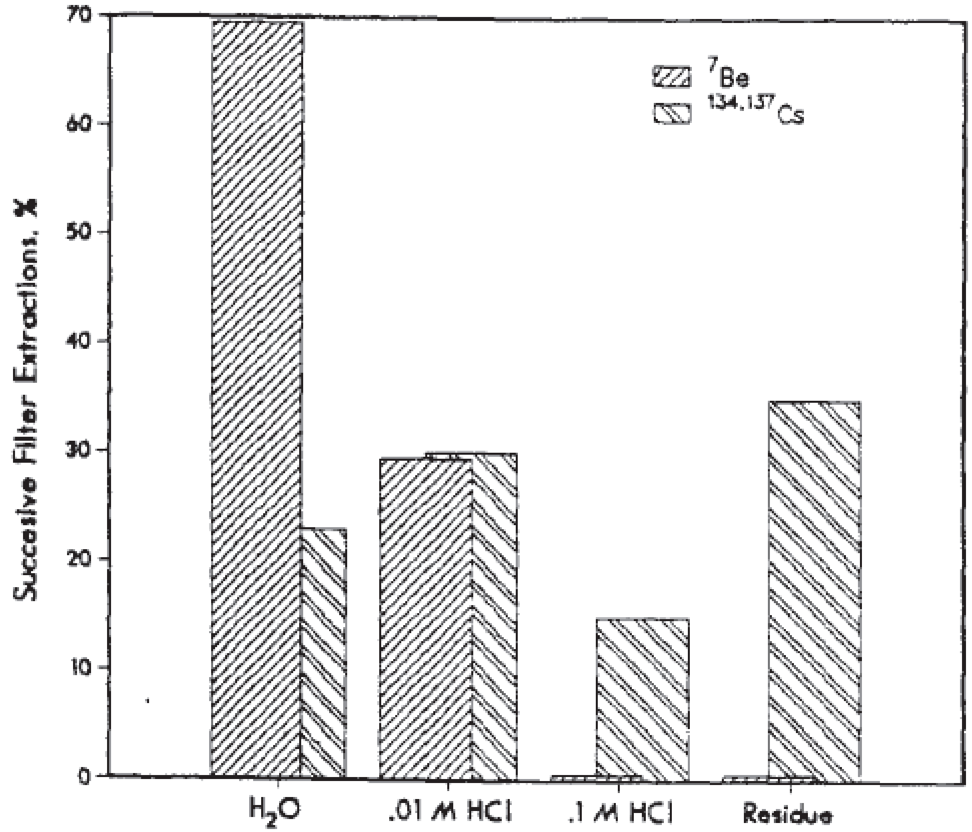


Fig. 6. The effect of progressively stronger acids on the extraction of natural ^{7}Be and Chernobyl $^{134,137}Cs$ from <0·41 μm aerosols sampled in Tennessee from 20 to 23 May 1986.

- Solubility to H_2O is less than 23 %

Artificial forest-fire experiment conducted in Lithuania 4-year after the Chernobyl accident

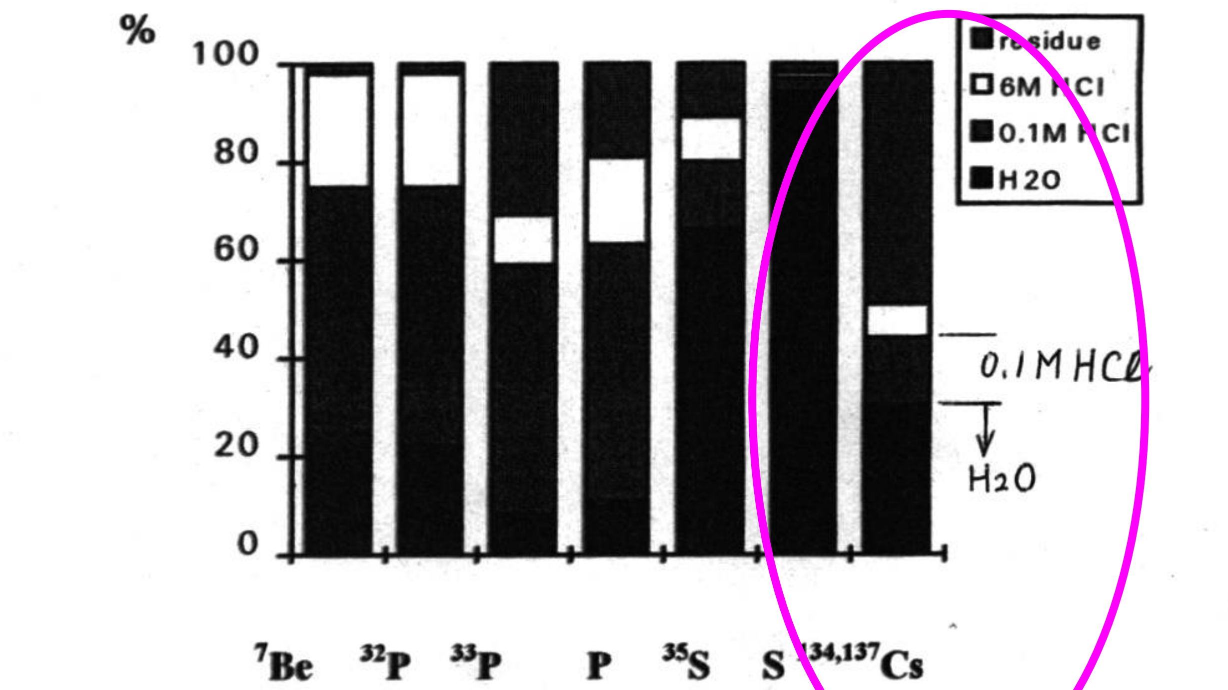


Fig. 1. Solubility of radionuclide carriers, stable phosphorus and sulphur in different solvents, % (aerosols sampled in Vilnius, June 18–21, 1990).

An Investigation of Changes in Radionuclide Carrier Properties

G. Lujanienė,[a] B. I. Ogorodnikov,[b] A. K. Budyka,[b] V. I. Skitovich[b] & V. Lujanas[a]