CHARACTERISTICS OF SPHERICAL Cs-BEARING PARTICLES COLLECTED DURING THE EARLY STAGE OF FDNPP ACCIDENT

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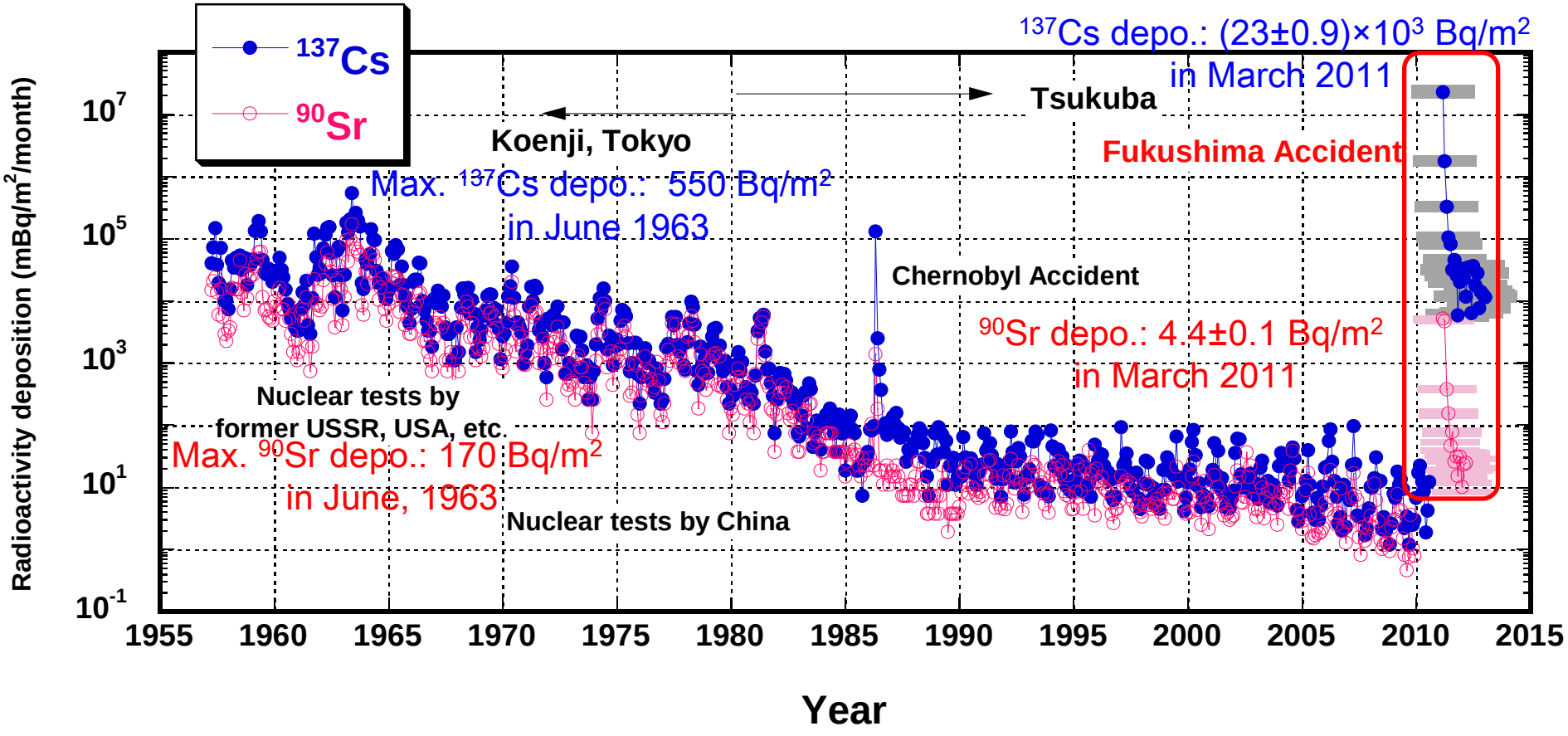
Talk outline

- Research backgrounds, aims of the present study and so forth
- Radioactive aerosol sampling at the Meteorological Research Institute (MRI), methodologies for analysis of Cs-bearing particles with state-of-the-art instruments including scanning electron microscope (SEM)
- Characteristics of spherical Cs-bearing particles including morphology, size and major components
- Modeling approach with state-of-the-art aerosol transport simulation considering the present spherical Cs-bearing particles
- Summary

Research Background: Temporal change in atmospheric deposition of radionuclides at MRI, Japan



Fukushima radioactivity emission inventory : Just a few kgs in weight !

Nuclear and Industrial Safety Agency Estimate

Nuclide	Half life	Emission/PBq
Kr-85	10.72y	
Pu-239	24,065y	0.0000032
Pu-240	6,537y	0.0000032

Conversion

Emission in mass

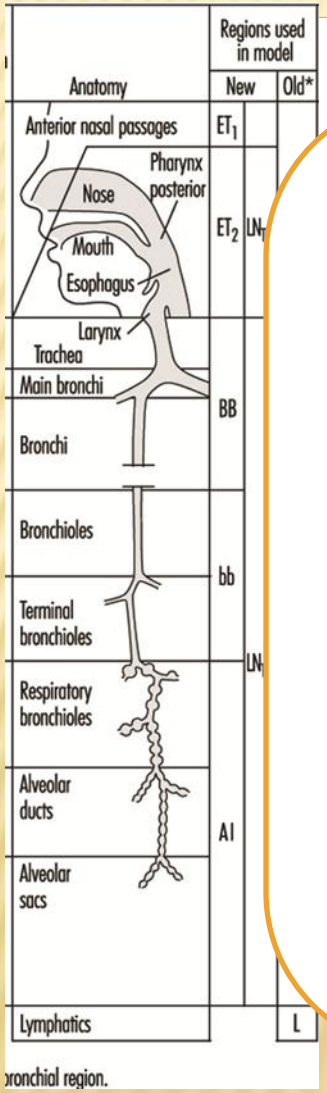
Nuclide	Half life	Emission/g
Xe-133	5.25 d	1590
Sr-90	29.12 y	28
Pu-239	24100 y	1.4
Pu-240	6540 y	0.4

Investigation is needed regarding mixed states with not only the radionuclide but also the general atmospheric aerosol.

∴ Most of radionuclides occurred in the aerosol form, which floated and transported in the atmosphere.

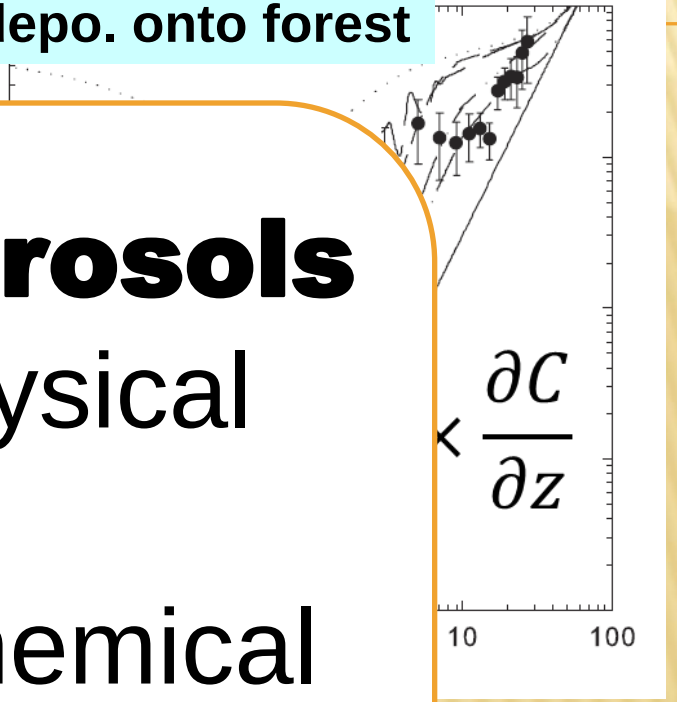
Cf. Daily SO_2 emission in Japan : 2×10^9 g (EAGrid2000)

Deposition of aerosols in environ. and humans



Inhalation

Dry depo. onto forest



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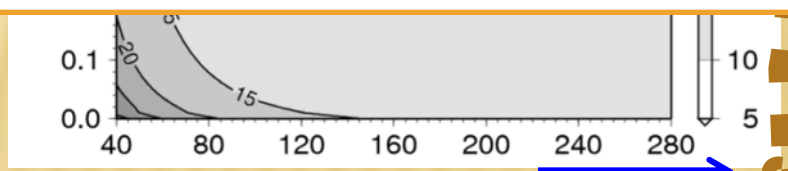
Jpn. Soc. Atmos. Environ.,
2014 (in press)

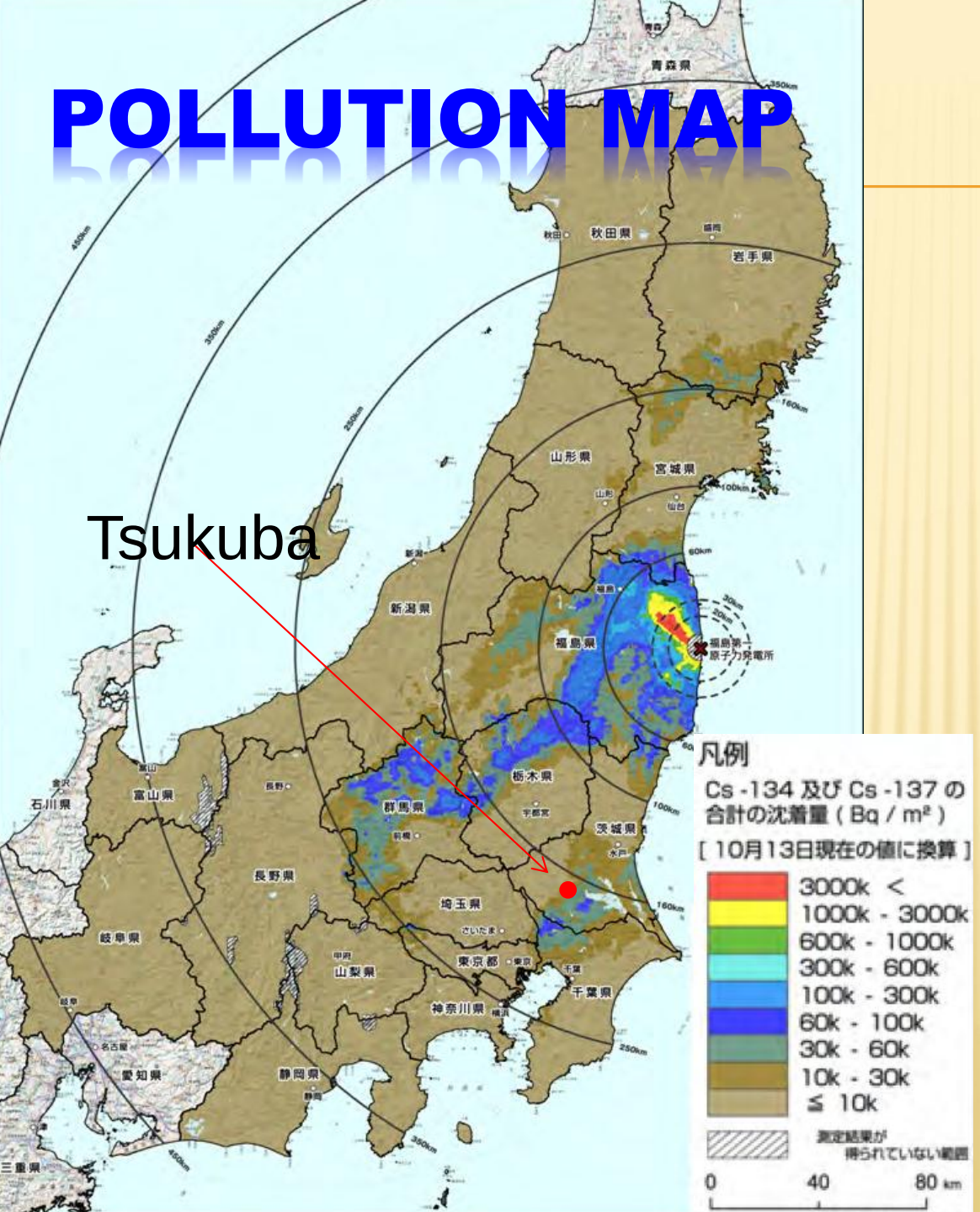


Particle size

Radioactive Aerosols

Aerosol size=physical
and
hygroscopicity=chemical
information
are very important!

POLLUTION MAP



Result of aircraft monitoring made by the Ministry of Education, Culture, Sports, Science and Technology (MEXT)

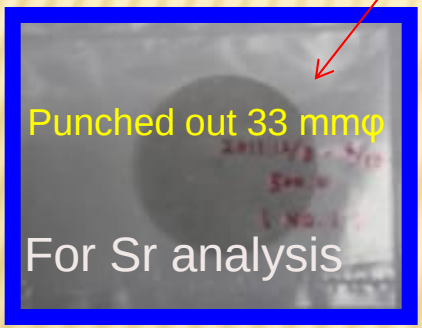
(Total of cumulative $^{134,137}Cs$ pollution of the surface (kBq/m^2) in the range that the survey completed up to fall, 2011)

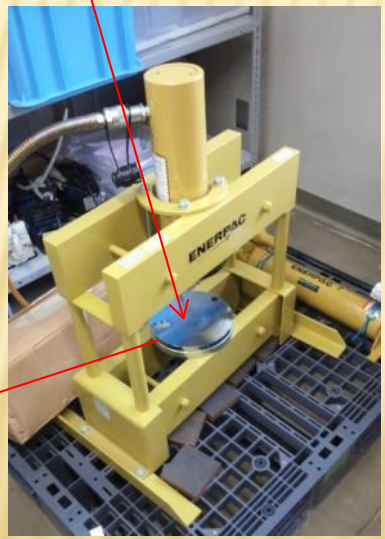
Nov. 11, 2011
Announcement

Sampling, sample preparations and radionuclides measurements

8" × 10“ Quartz fiber filter

Total = Wet and dry deposition sampling

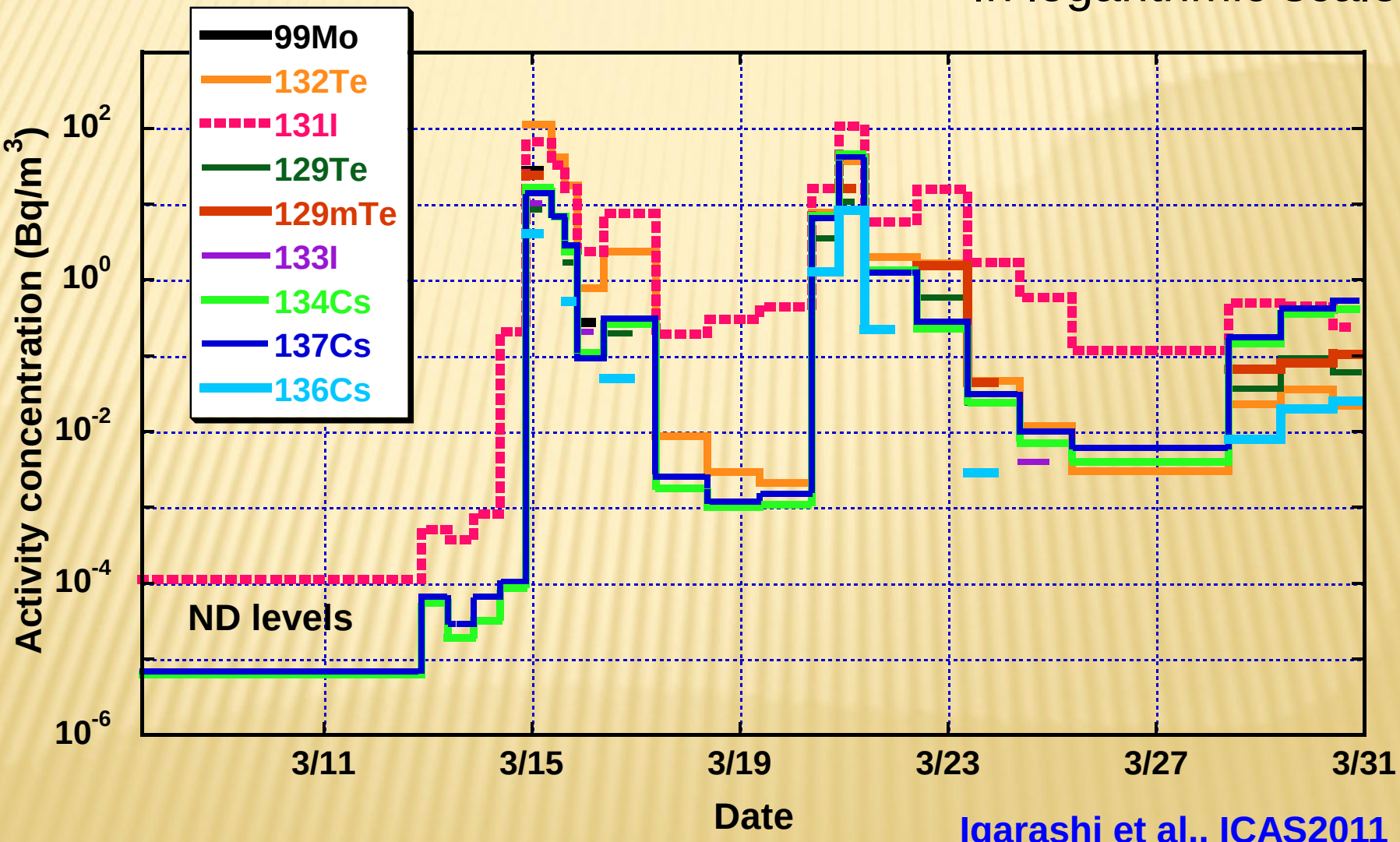




Hydraulic press machine

Radioactivity in aerosol samples at MRI in March 2011

In logarithmic scale



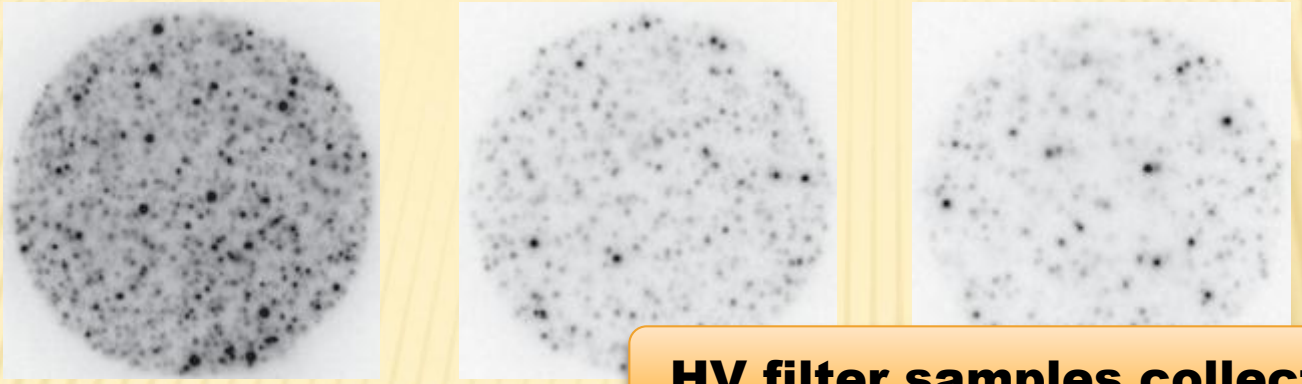
Igarashi et al., ICAS2011

Imaging plate picture of HV filter sample

Digital radiography with courtesy of Dr. Osada, Nagoya Univ.

Event 1

#5 March/14-15 (700 m^3) #6 March/15a (253 m^3) #7 March/15b (233 m^3)

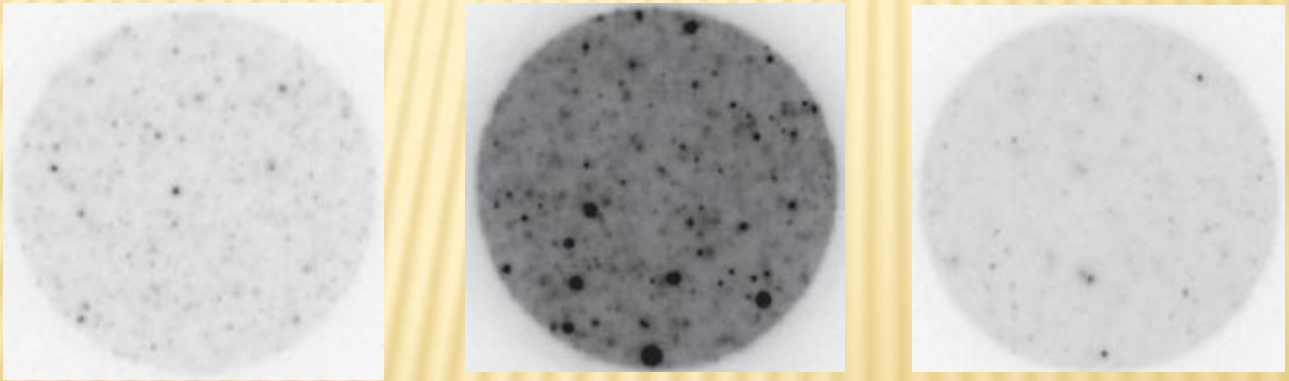


In April 2011 (more than a month later the accident) images were taken.

HV filter samples collected at MRI

Event 2

#8 March/15-16 (498 m^3) #14 March/20-21 (487 m^3) #15 March/21-22 (1011 m^3)

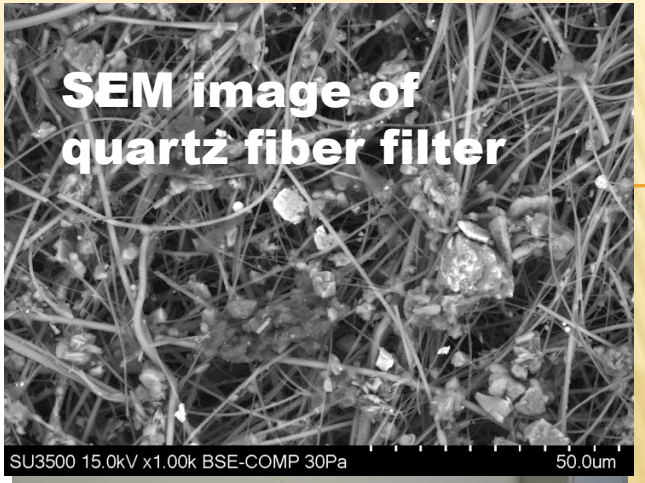


The BG image with homogeneous distribution vs Hot spots with irregular distributions like Japanese sesame senbei.

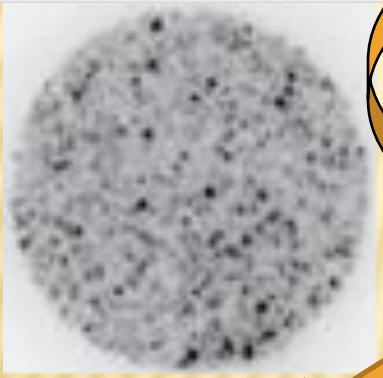
= Strong radioactive particle was seen.

However, the black spot in the image doesn't show the size of the particle directly.

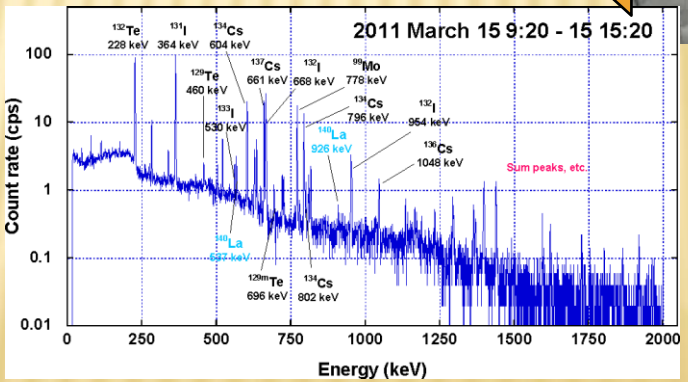
Exploit for strong radioactive particle

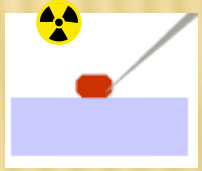


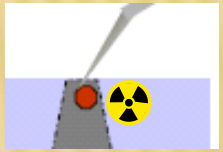
Providing position data



Low vacuum-low acceleration bias SEM·with EDS mapping







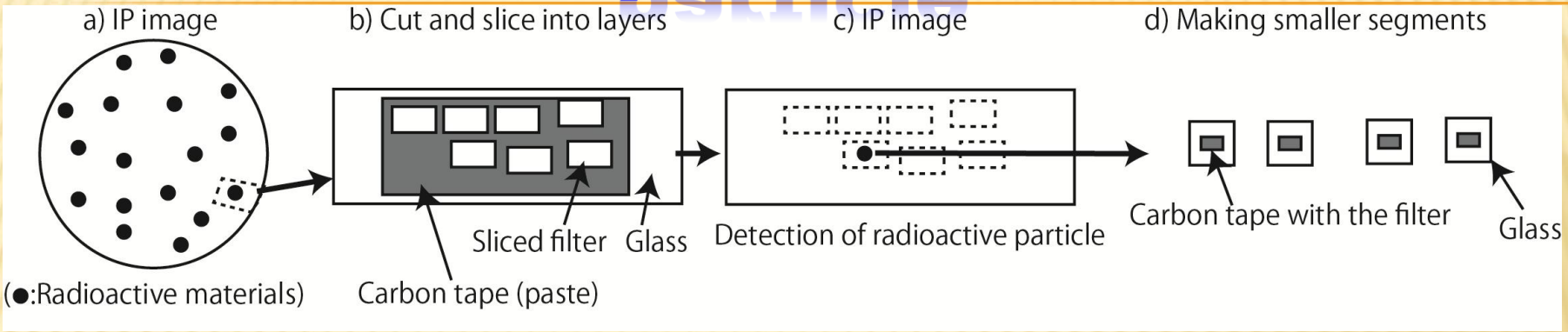
Pick up
Extraction of the particle

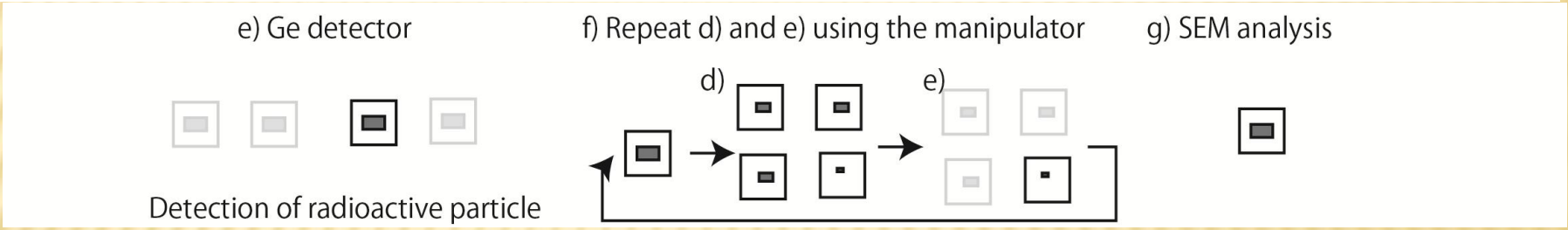
SPring-8

Nano-XRF

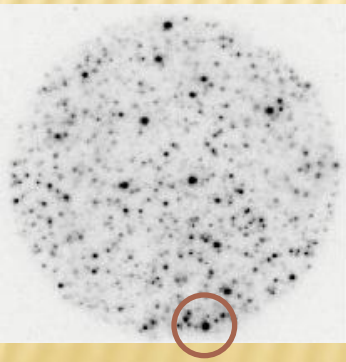
Activity measurement with Ge detector

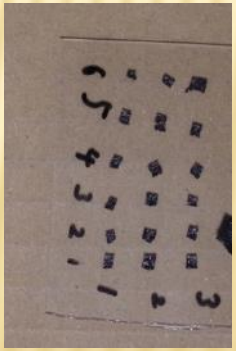
Exploit for strong radioactive particle

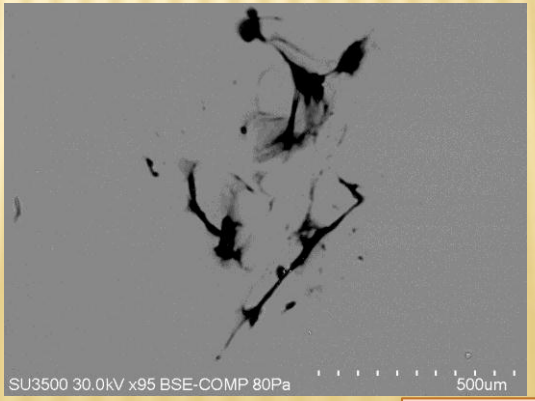


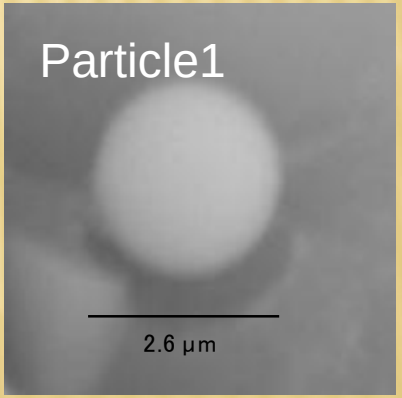


Adachi et al., Sci. Rep., 2013; doi: 10.1038/srep02554



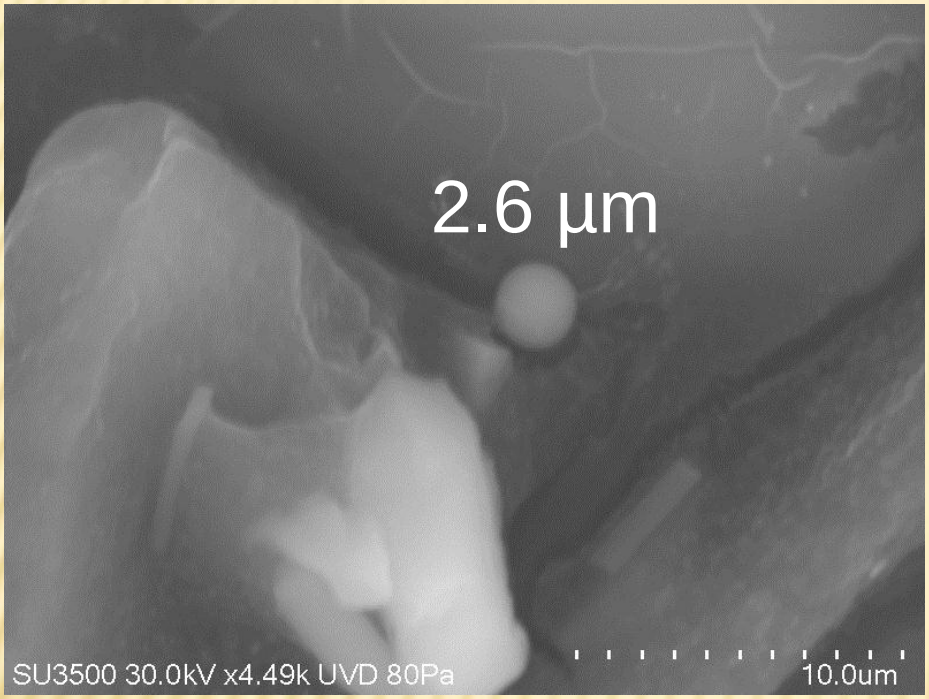


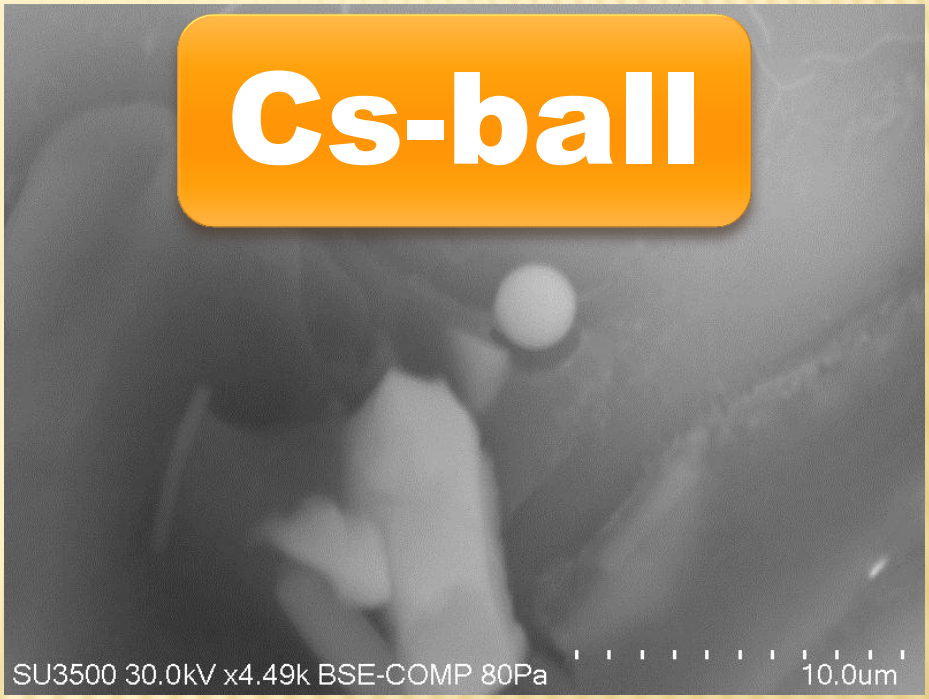




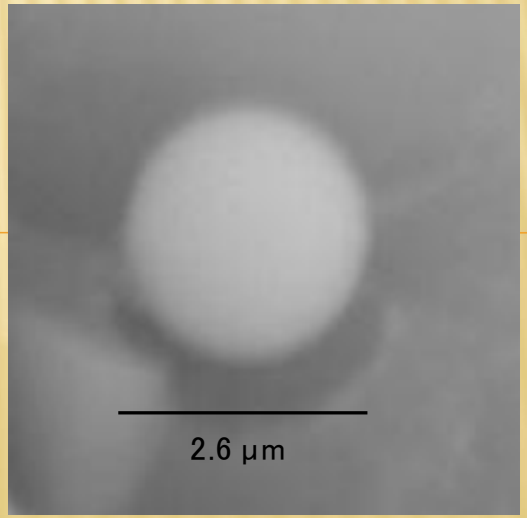
3 cm → 1 cm → 1 mm → 100 μm → **Downsize** → 2 μm

SEM image of particle 1



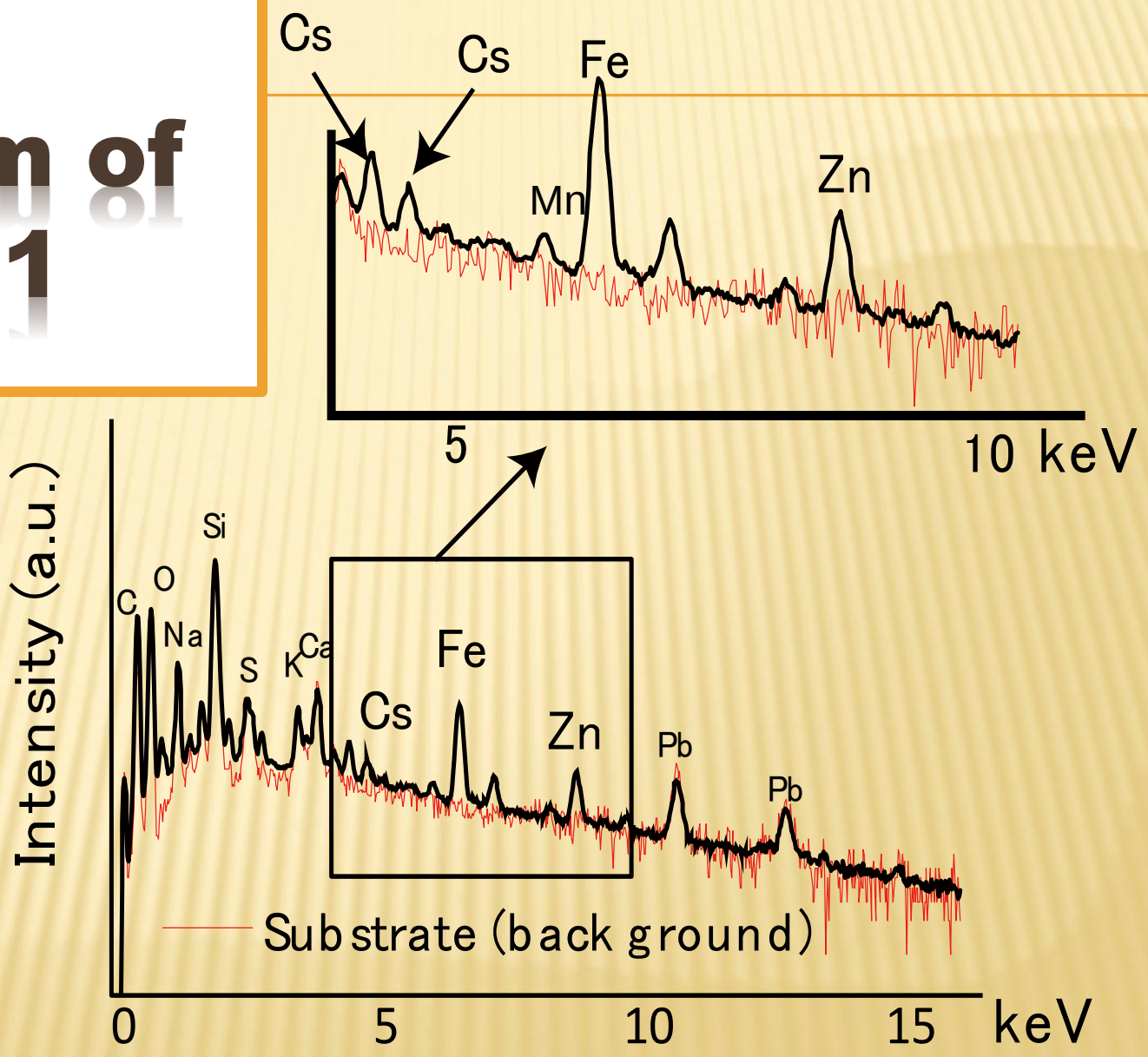


Detected elements by EDS:
O, Na, Si, Cl, Fe, Mn, Zn, Cs



Adachi et al., Sci. Rep., 2013; doi: 10.1038/srep02554

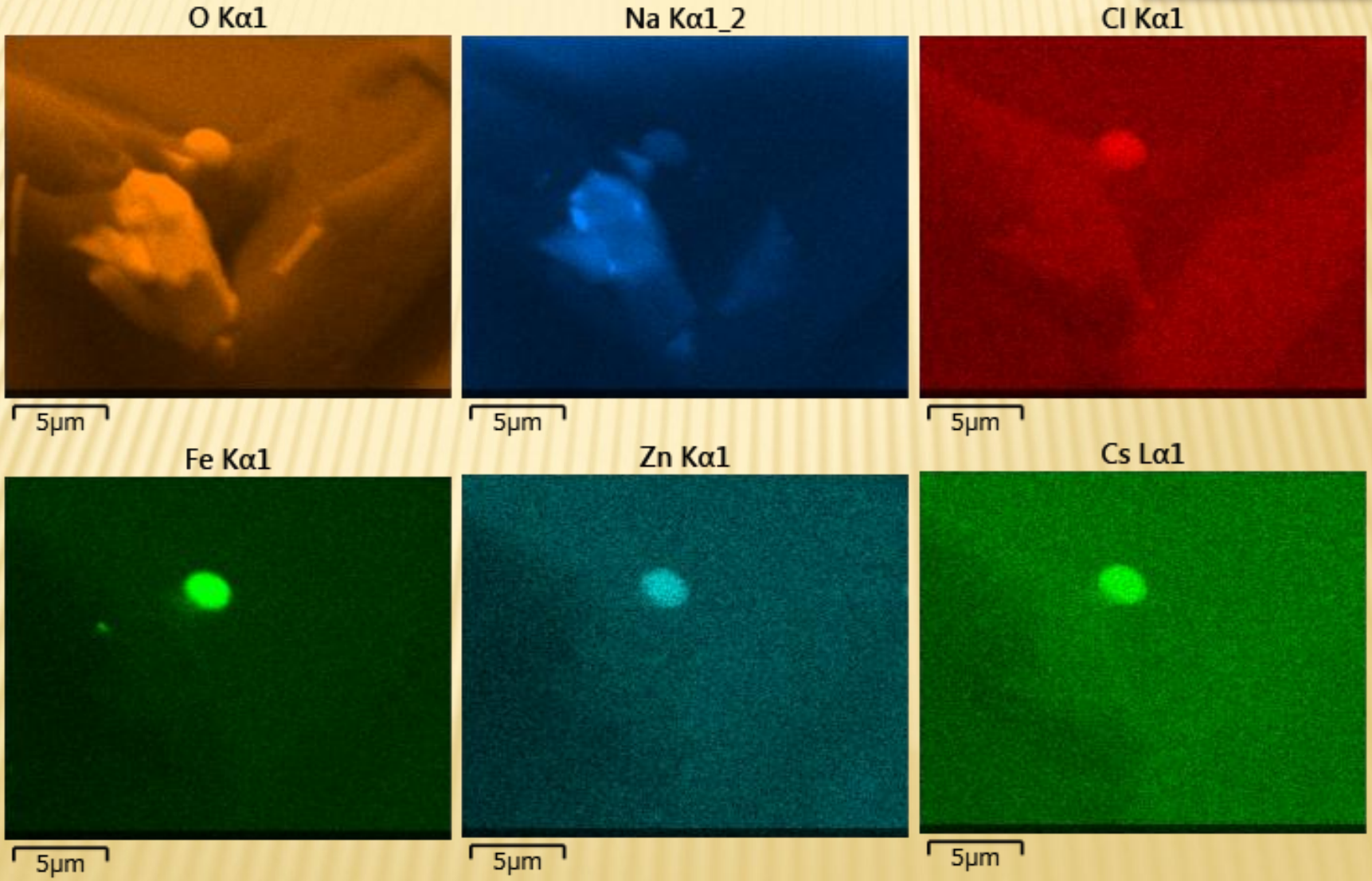
EDS spectrum of Particle 1



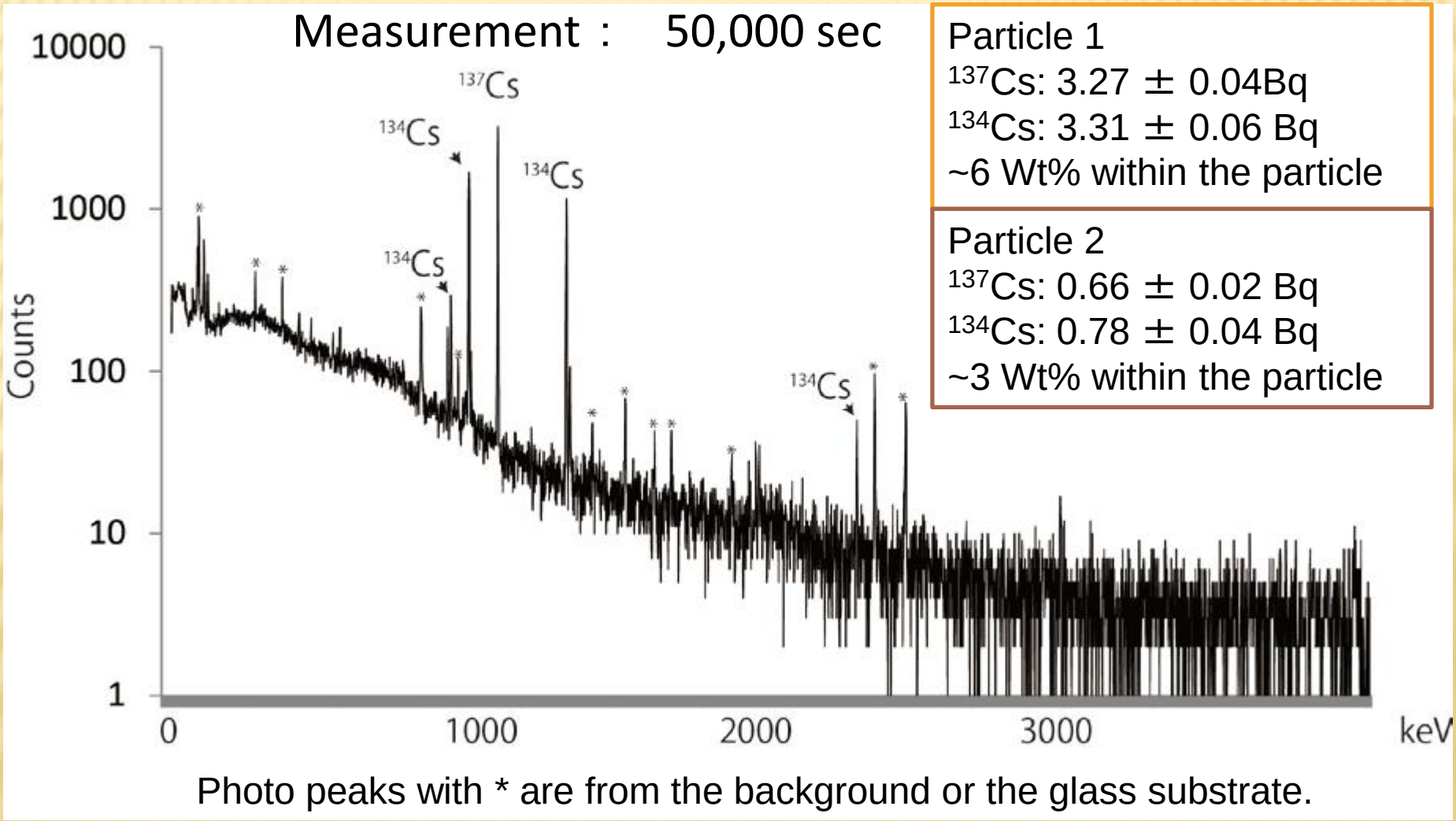
Adachi et al., Sci. Rep., 2013; doi: 10.1038/srep02554

Elemental mapping by EDS analysis

Particle 1

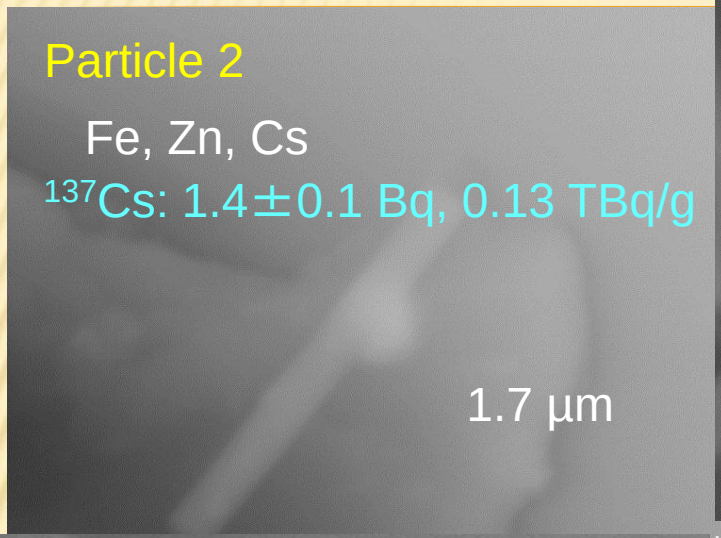


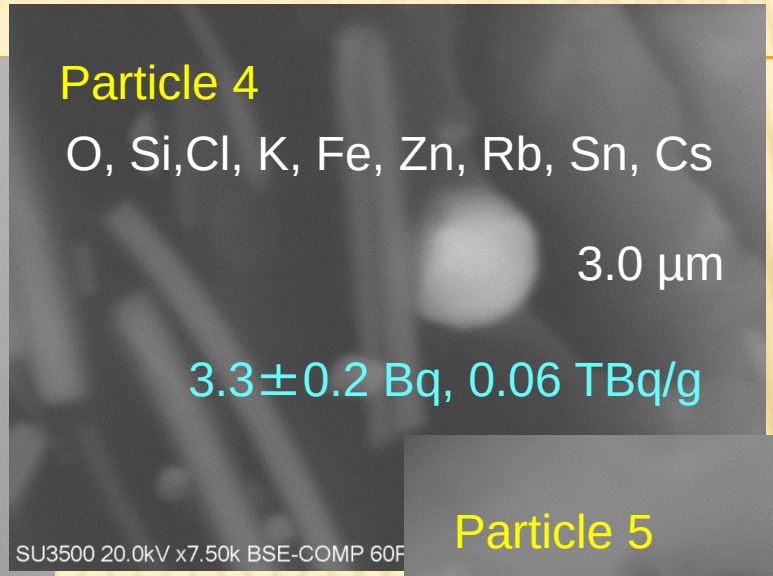
Gamma-spectrum of Cs-ball from Mar.14-15 HV filter



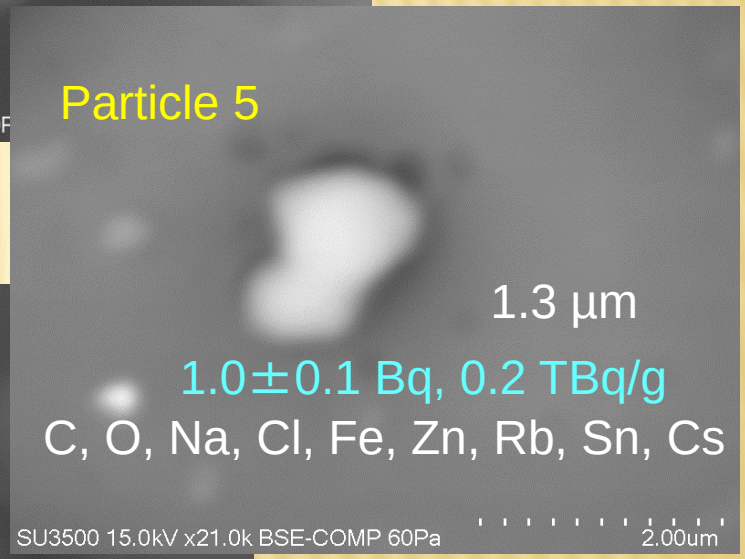
Adachi et al., Sci. Rep., 2013; doi: 10.1038/srep02554

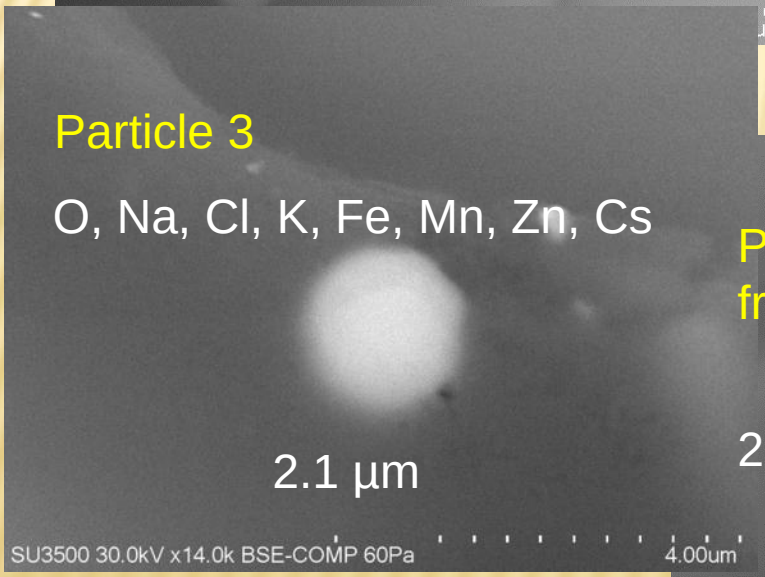
Collection of Cs-ball

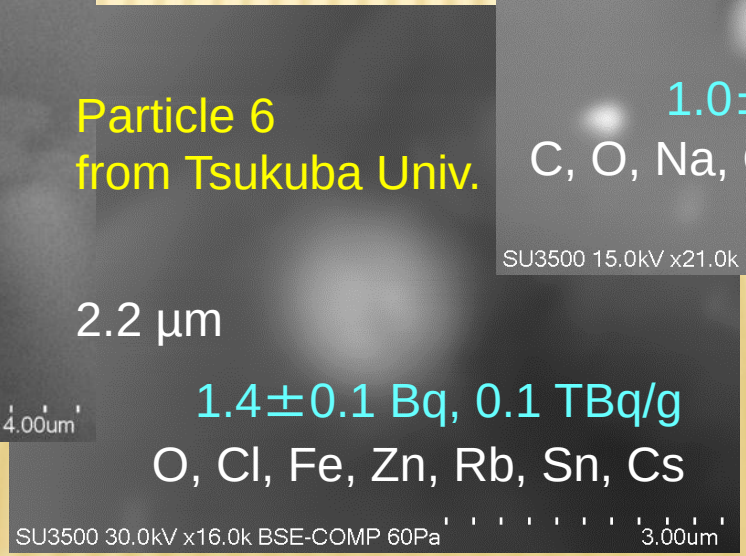




Specific activity calc. assume density of 2 g/cm^3

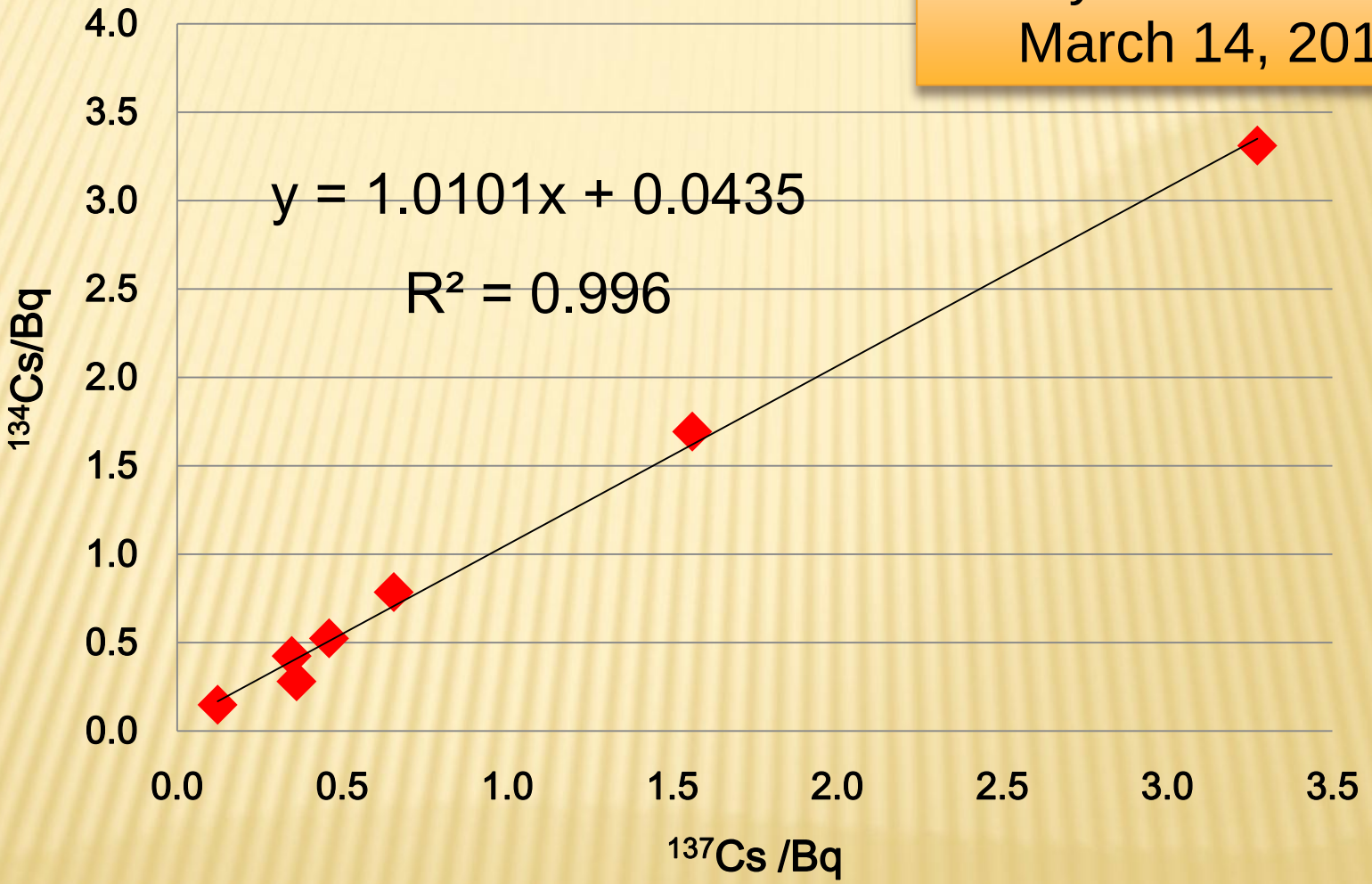






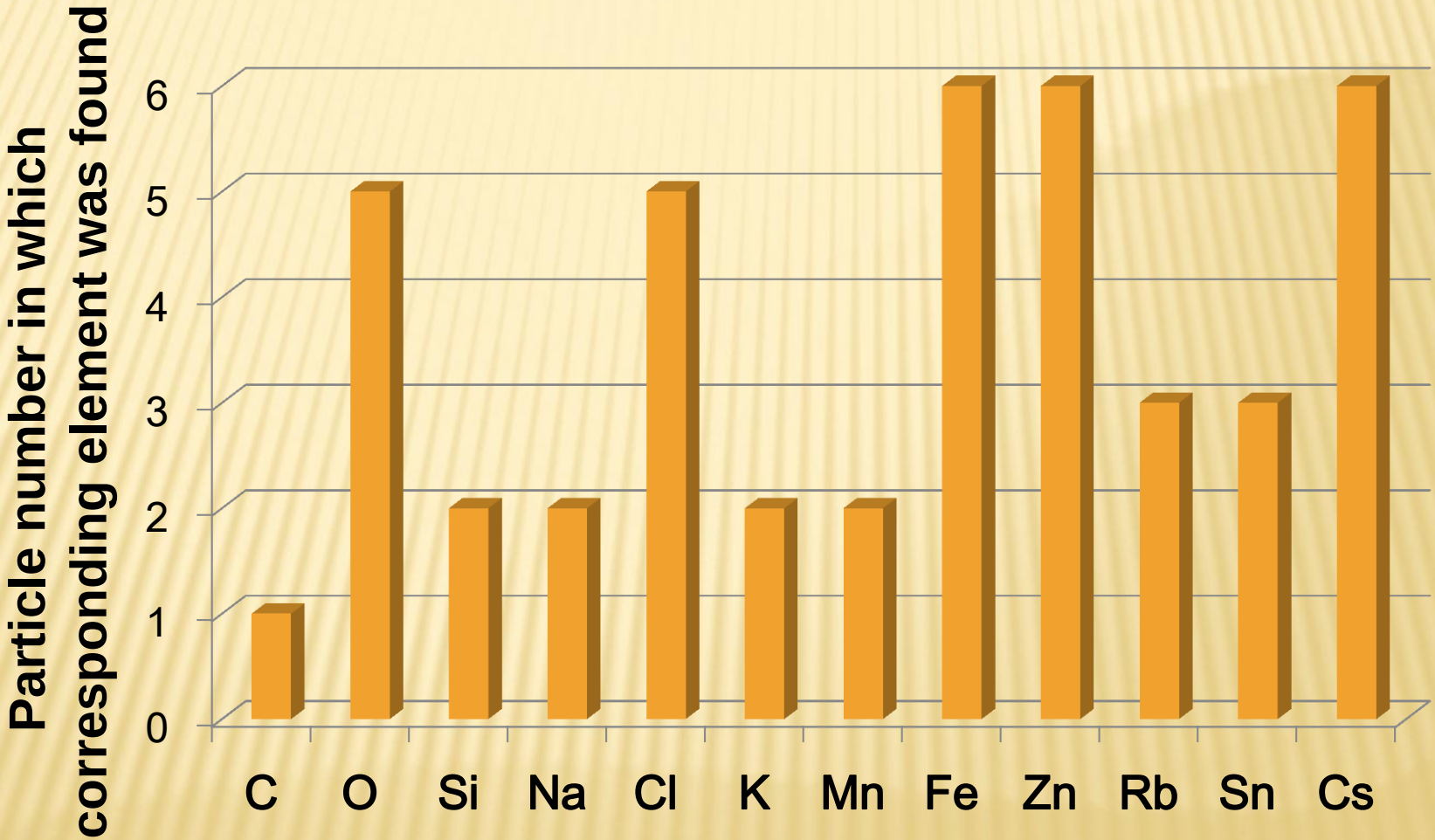
Cesium isotopic composition of Cs-ball (In total 7 particles)

Decay corrected as of March 14, 2011

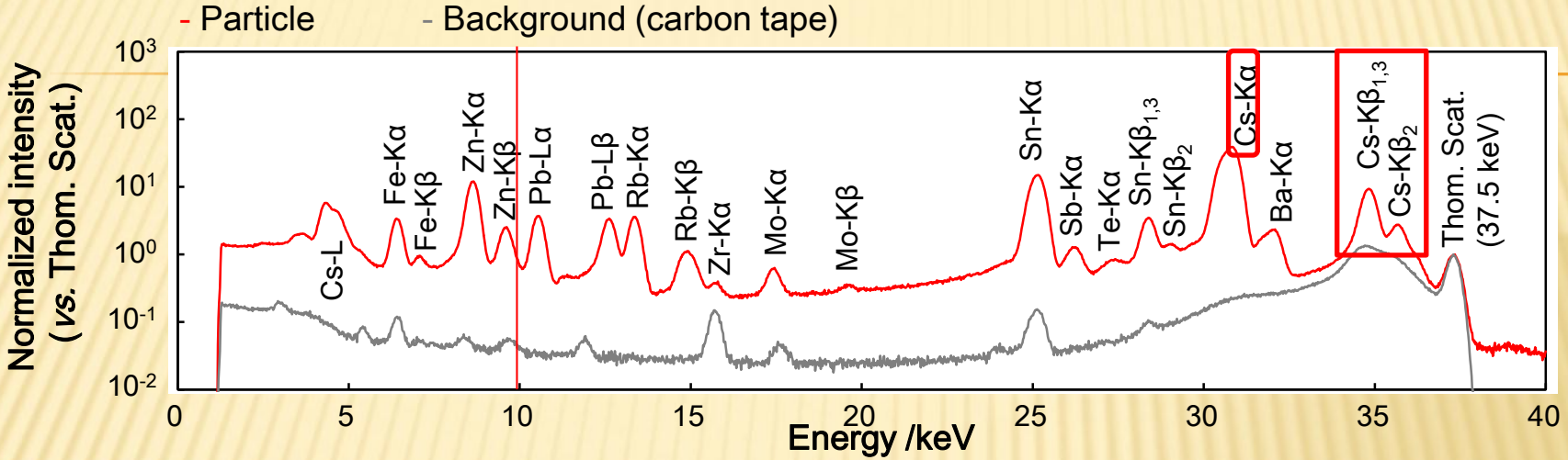


*One data was not yet subjected to SEM analysis.

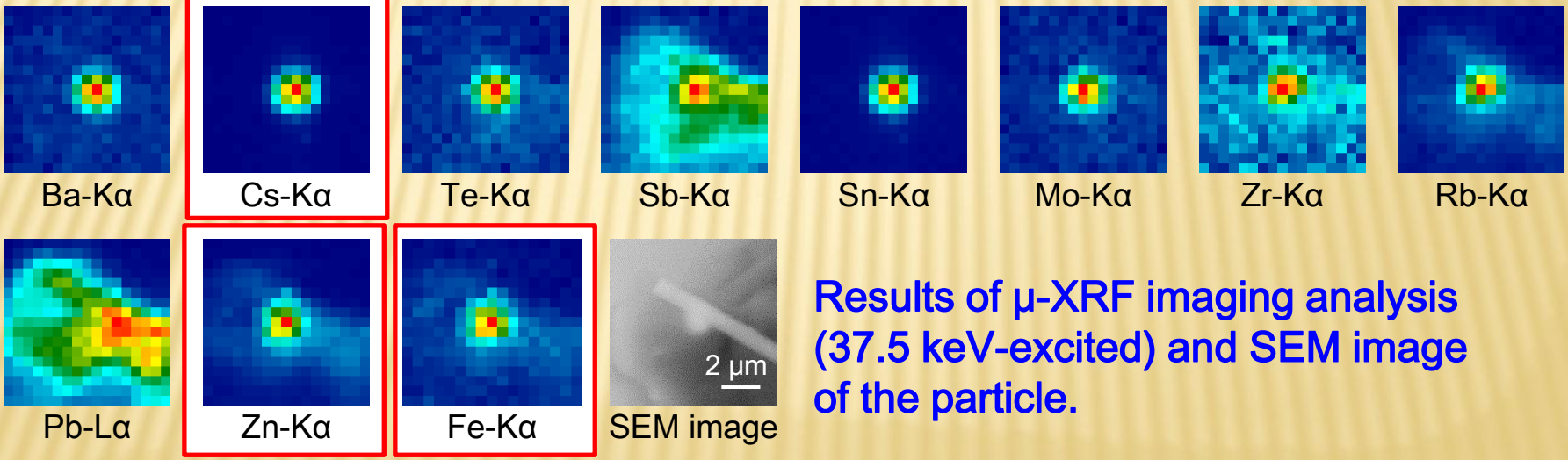
Elements identified in Cs-ball by SEM-EDS (In total 6 particles)



Results of synchrotron radiation-μ-XRF analyses



Comparison of XRF spectra of the particle and background (37.5 keV-excited).



Results of μ-XRF imaging analysis (37.5 keV-excited) and SEM image of the particle.

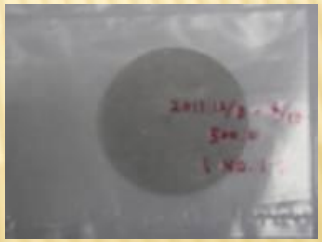
Elements in particle: **Ba, Cs, Te, Sb, Sn, Mo, Zr, Rb, Zn, Fe**

(Pb : derived from an attached fragment of the filter)

Unpublished data: Courtesy of Prof. Nakai and Dr. Abe, Tokyo Univ. Sci.

Extraction efficiency of ^{137}Cs from HV filter samples

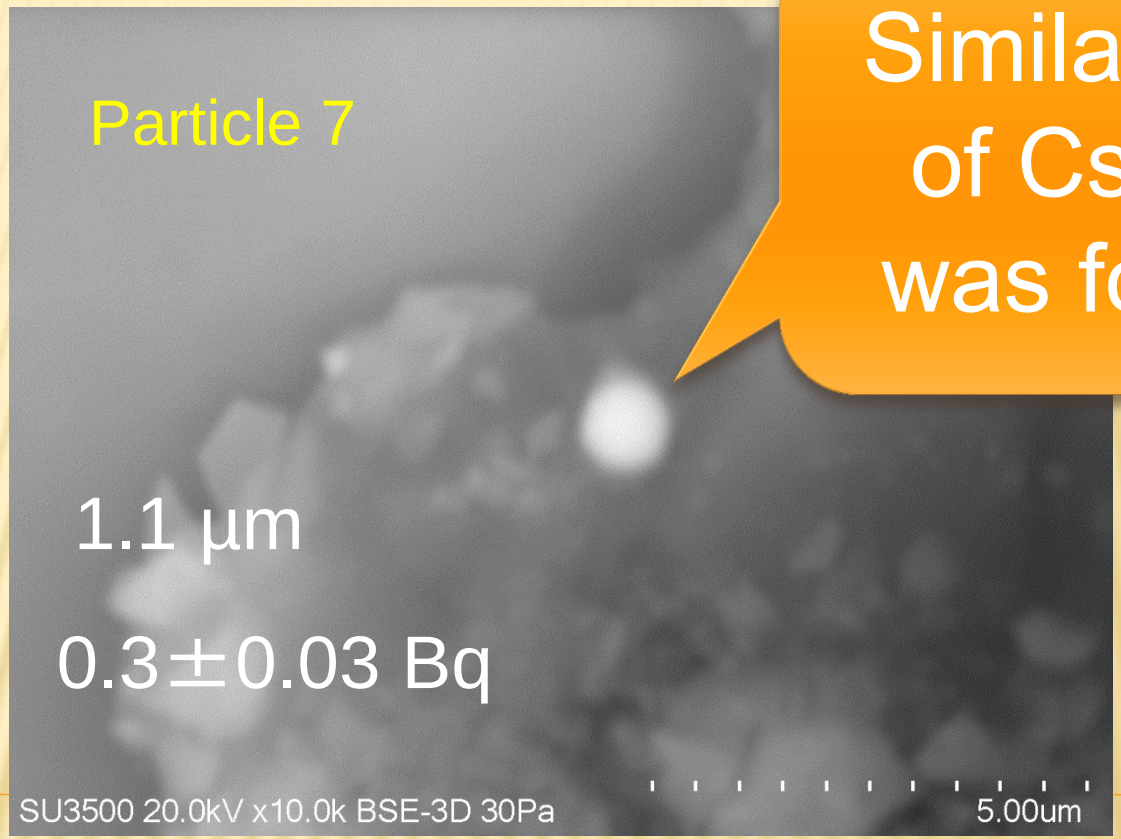
Sample no.	Before ext. ^{137}Cs (cps)		After ext. ^{137}Cs (cps)		Water extraction (%)	Nitric acid extraction (%)
Mar. 14 21JST – 15 9JST	28.4	±0.120	20.9	±0.105	26.5%	70.4%
Mar. 15 9:JST-15 15JST	7.75	±0.028	7.36	±0.014	5.0%	82.0%
Mar. 15 15JST-15 21JST	2.17	±0.015	2.28	±0.008	-5.1%	69.5%
Mar. 15 21 JST-16 9JST	1.74	±0.013	0.63	±0.004	63.8%	97.3%
Mar. 16 9JST-17 9JST	0.92	±0.010	0.60	±0.004	34.8%	94.3%
Mar. 20 9JST-21JST	7.06	±0.027	3.28	±0.009	53.5%	99.8%
Mar. 20 21JST-21 9JST	84.0	±0.22	33.5	±0.13	60.1%	99.7%
Mar. 21 9JST-22 9JST	4.74	±0.022	2.23	±0.008	53.0%	99.5%



Mar. 14-15 sample contained insoluble materials not only in water but hot nitric acid !
Characteristics of Mar.20 plume and particles seem to be different from those of 14-15.

SEM image of a remained particle on HV filter after nitric acid extraction

Mar. 15 sample
IP exposed for 24h.

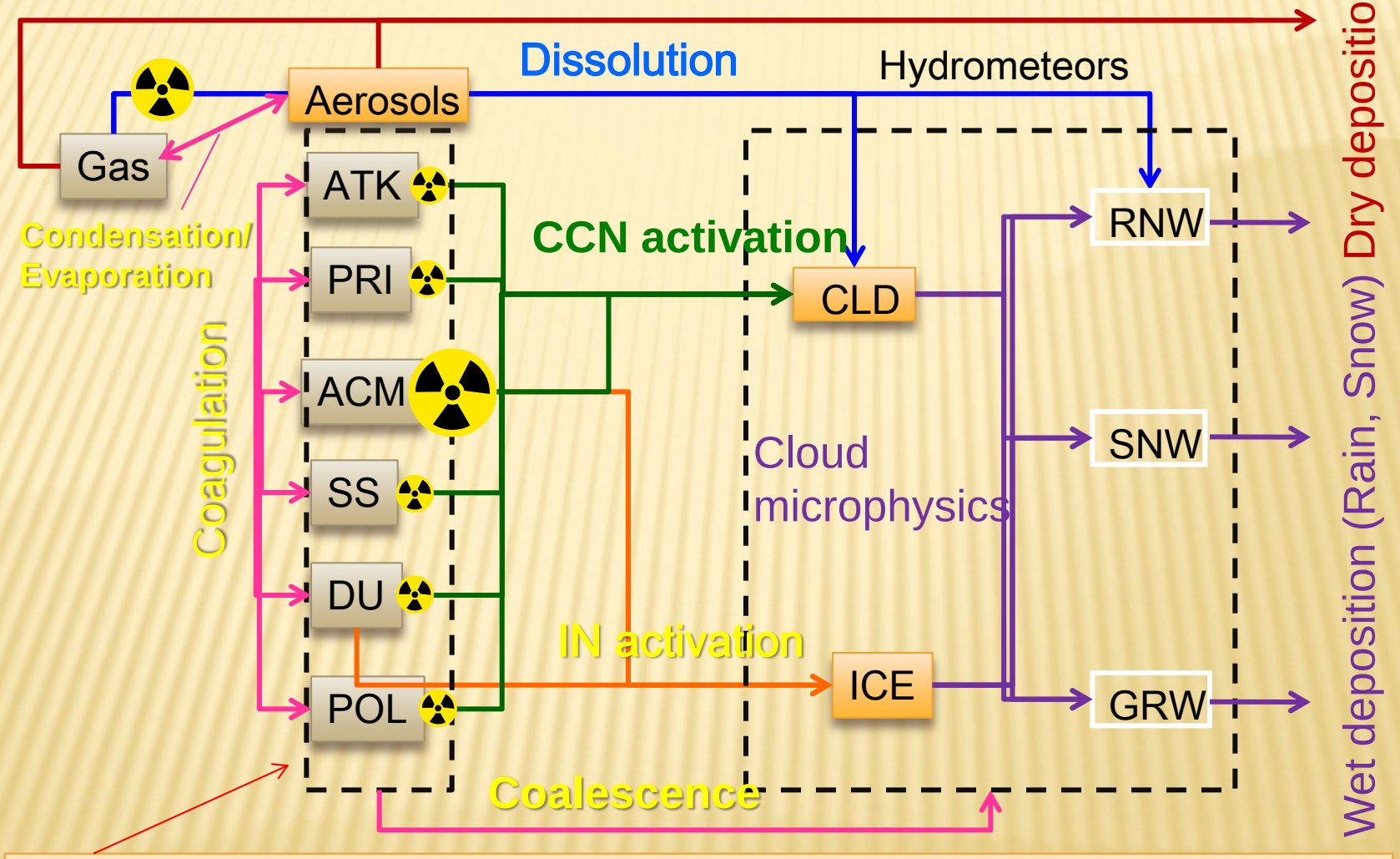


Similar type of Cs-ball was found!

Detected elements by EDS:
O, Si, Na, Ca, Mg, Fe, K, Al, Cl, Zn, Cs

MANAL-NHM-PM/r (Δx=4km)

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

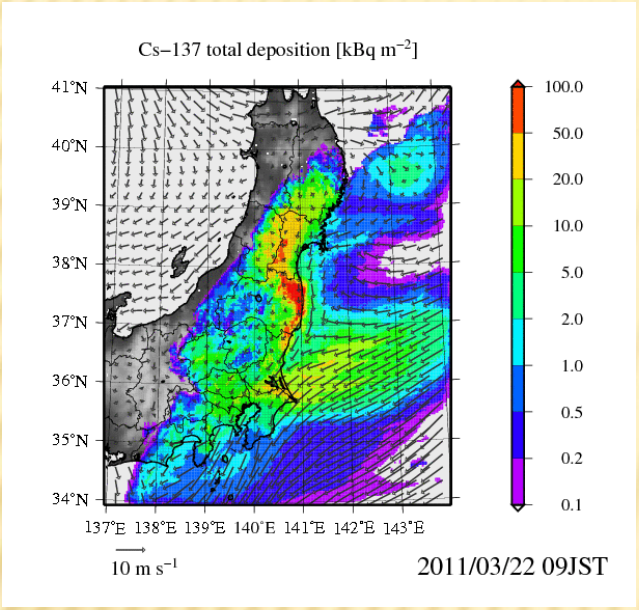


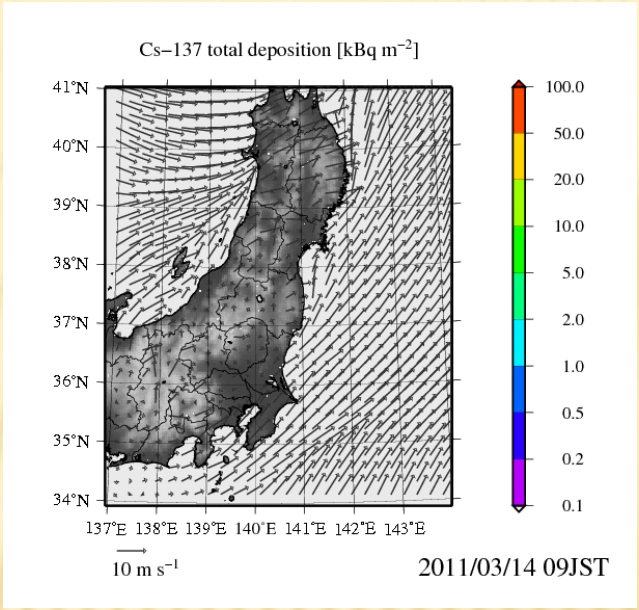
ATK (Aitken mode), PRI (primary radionuclides), ACM (accumulation mode), DU (dust), SS (sea salt), POL (pollen)

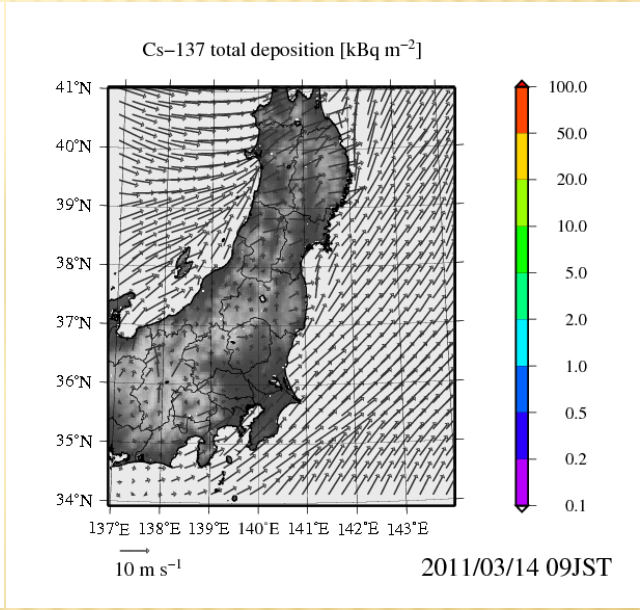
Difference in simulated depo. due to difference in aerosol characteristics

March 11 to 22, 2011
Total depo.（kBq m^{-2}）

Cs depo. emitted on March 14 only（kBq m^{-2}）



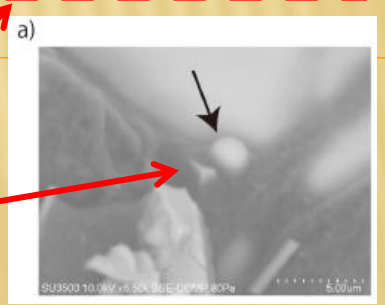




Submicron hygroscopic (CMD = 102 nm, κ=0.4)

CMD= 2μm, κ=0

Dry depo. velocity becomes about 4 times. No CCN activity. Removal by only rain drops encounter (scavenging).



SUMMARY 1

- We have found spherical Cs-bearing particles (Cs-ball) from HV filter samples collected during Mar. 14-15, 2011, when the first radioactive plume arrived from the FDNPP accident at the MRI as well as Univ. Tsukuba, Japan
- So far seven Cs-balls were found from any given black dot on IP image of the filter sample.
- They have basically spherical morphology, characterized composition of Fe, Zn, Mn, O, etc. with an appreciable amount of elemental Cs.
- They are a few microns in diameter (corresponding to PM2.5) with a few Bq activity but as high specific activity as sub-terra Bq/g.
- They are insoluble; even refractory to conc. nitric acid. Also, the extraction experiments showed that the Cs-ball occupied major part in the Mar. 14-15 Fukushima plume.
- They would persist for a long time in the environment as well as in living organisms.

SUMMARY 2

- While the investigation on radioactive aerosol during the second plume arrival gave no such Cs-ball.
- Aerosol model simulation gave the results that non-hygroscopic super-micron Cs-ball could be removed more by the dry deposition and below-cloud scavenging than conventional sulfate-hosted particles.
- A few Cs-balls have been subjected to the analysis of synchorotron X-ray analyses by using Spring-8 (Prof. Nakai and Dr. Abe, Tokyo Univ. Sci.).
- The finding should be a key to understand the processes of the FDNPP accident, to accurately evaluate the health and environmental impacts and to improve efficiency of the decontamination of the polluted area.

- Further studies are recommended for the present Cs-ball in more detail !

ACKNOWLEDGMENT

- Y. Igarashi acknowledge the following people for their support and collaboration;
  - Prof. I. Nakai, Dr. Y. Abe and Mr. Iizawa (Tokyo Univ. Sci.) for their advanced analysis by using Spring-8.
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