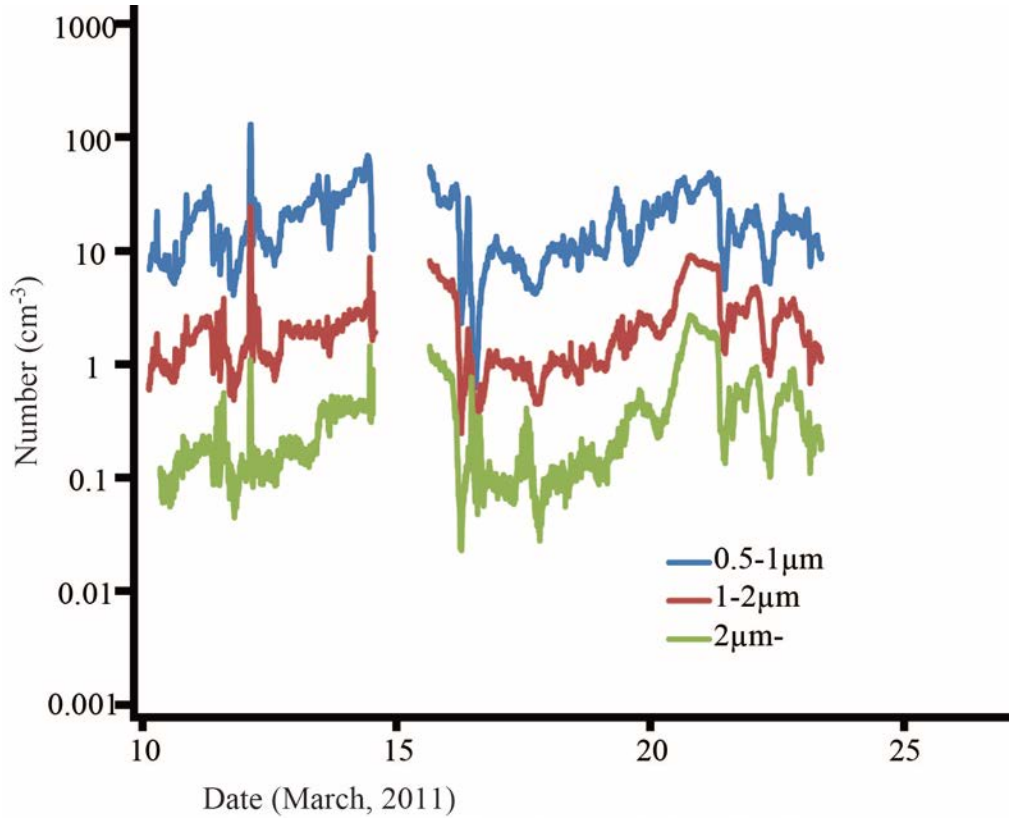
SUPPORTING INFORMATION FOR

Emission of spherical cesium-bearing particles from an early stage of the Fukushima nuclear accident

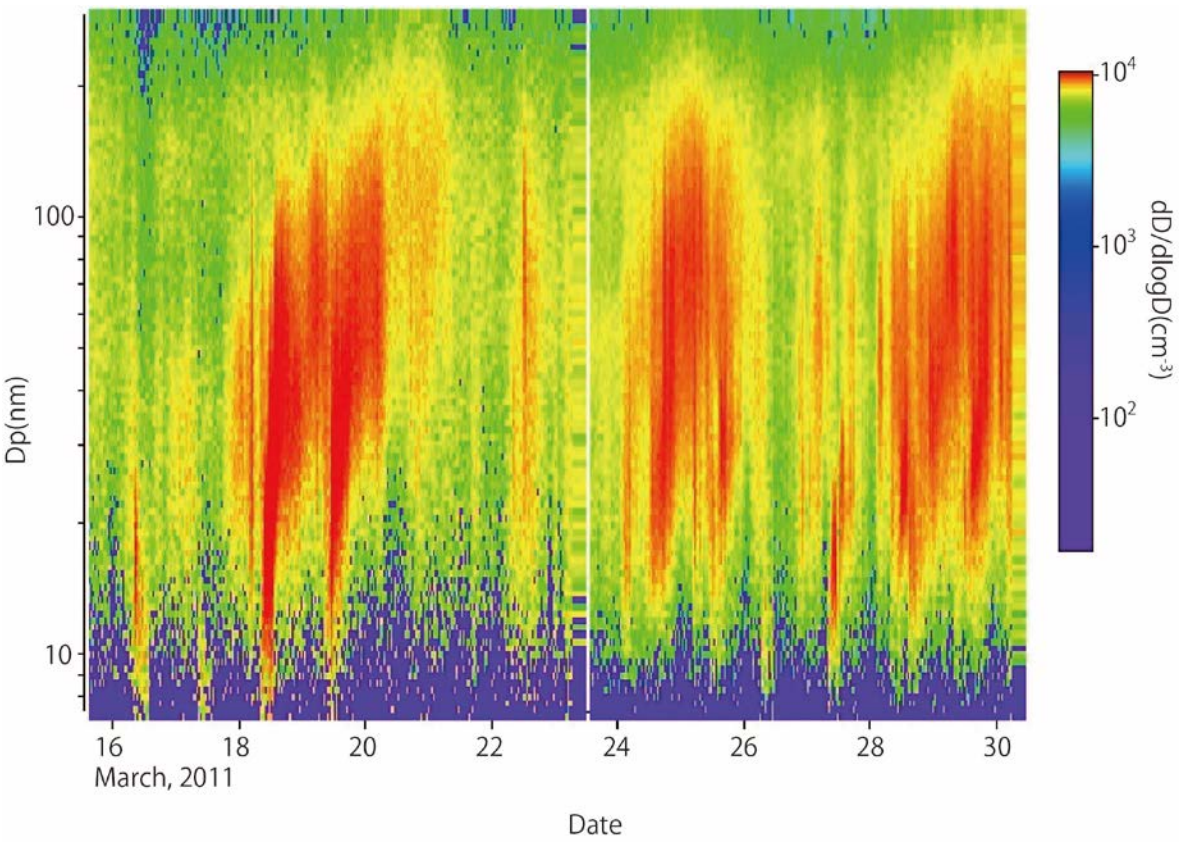
Kouji Adachi[1]*, Mizuo Kajino[1], Yuji Zaizen[1], and Yasuhito Igarashi[1]

1: Meteorological Research Institute, 1-1 Nagamine, Tsukuba, Ibaraki, Japan 305-0052

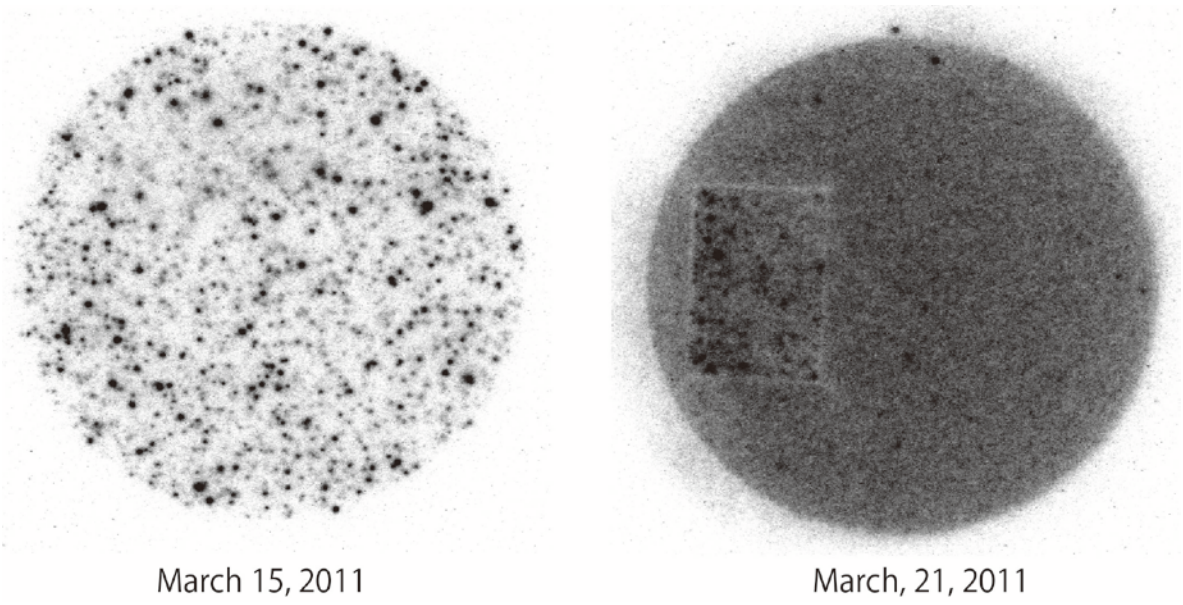
*Corresponding author: Kouji Adachi, Meteorological Research Institute, 1-1 Nagamine, Tsukuba, Ibaraki, Japan 305-0052, tel: +81-29-853-8570, adachik@mri-jma.go.jp



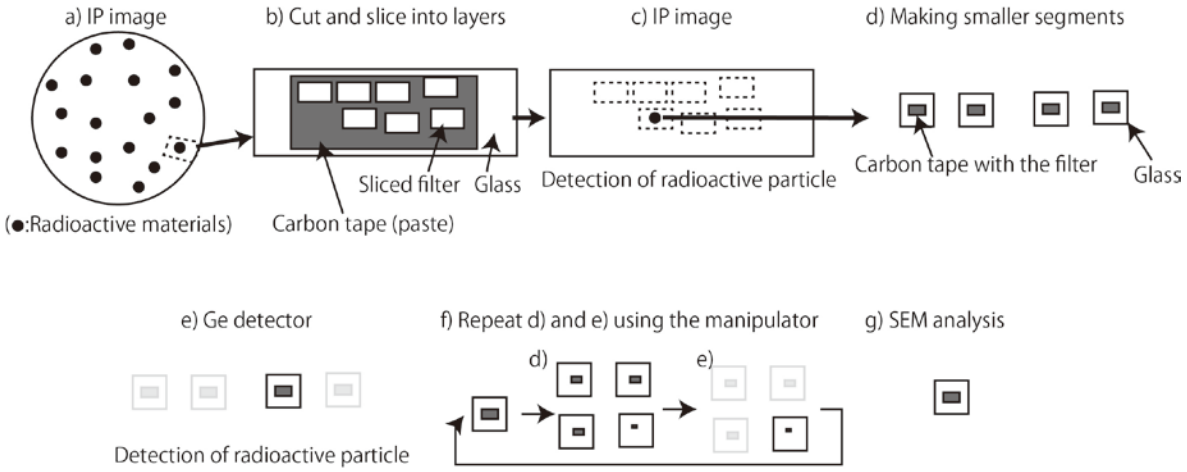
SI 1. Particle number concentrations for particles of 0.5-1.0, 1.0-2.0, and >2.0 μm. These data were measured using an aerosol particle sizer (APS). The data around March 15 were missing because of a regional unstable power supply caused by the earthquake. The APS was located on the rooftop site.



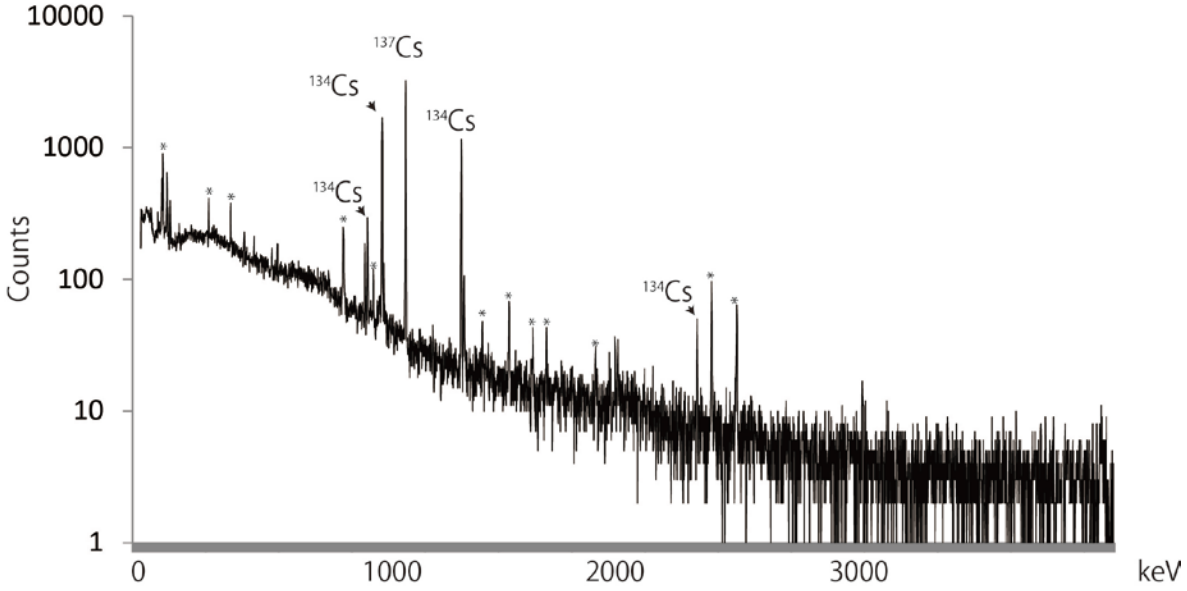
SI 2. The size distribution of aerosol particles between 7 and 289 nm using a scanning mobility particle sizer (SMPS). The averaged particle number concentration was 1.9×10^9 per m^3 during the second plume (March 20-21). The data prior to March 15, 17:18 (local time) were missing because of the emergency stop caused by the earthquake. The SMPS was located on the rooftop site.



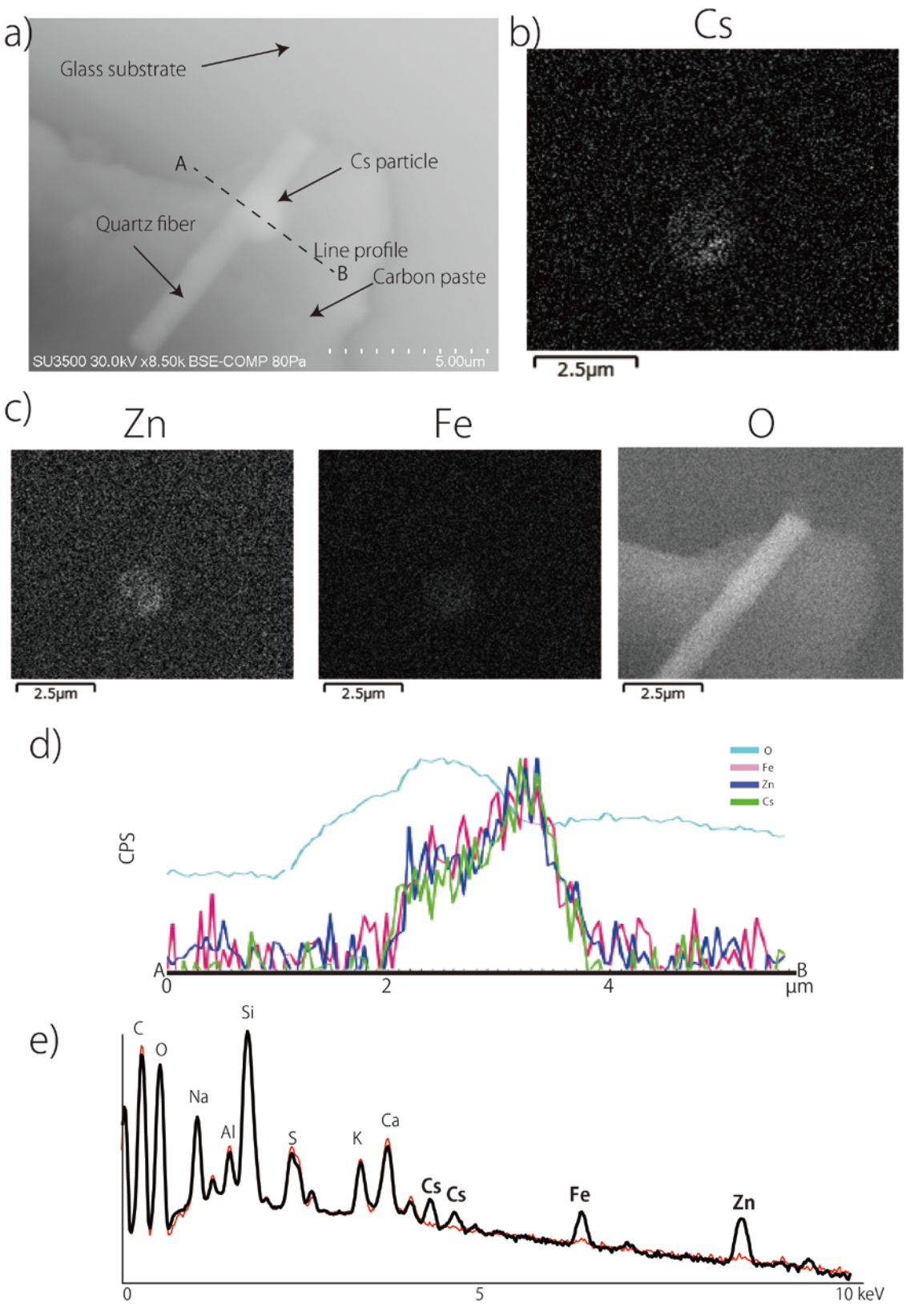
SI 3. The IP images of the PM2.5 filter samples collected on March 15 (left) and March 21 (right) on the rooftop site. The left filter includes many spots whereas the right filter appears dark all over. The features are similar to those in the Figure 2, which were collected at the ground level of the site. A part of each filter was cut for carbonaceous measurements and replaced (the trace is apparent in the right filter). A two hour exposure time was used for the IP analysis.



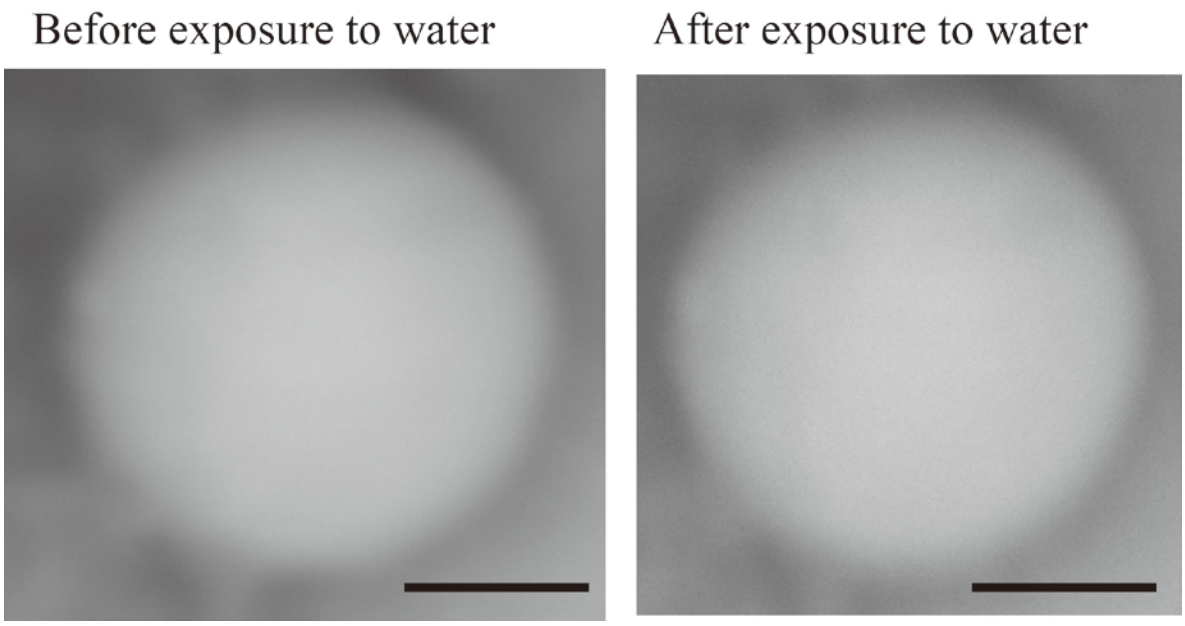
SI 4. The selection of radioactive particles for the SEM analysis. a) The IP image of the filter was captured, and a part of the filter that includes radioactive material was cut off (dotted line). b) The segment of the filter was separated into approximately 10 layers and put onto a carbon tape attached to a glass substrate. c) The IP image was taken to identify the position of the radioactive material. d) The carbon tape including the radioactive material was cut into small pieces. e) The radioactive material was detected using a Ge detector from the filter segments. f) The processes d) and e) were repeated to make the filter and the number of particles as small as possible. g) The radioactive particle embedded within carbon paste was analyzed using the SEM.



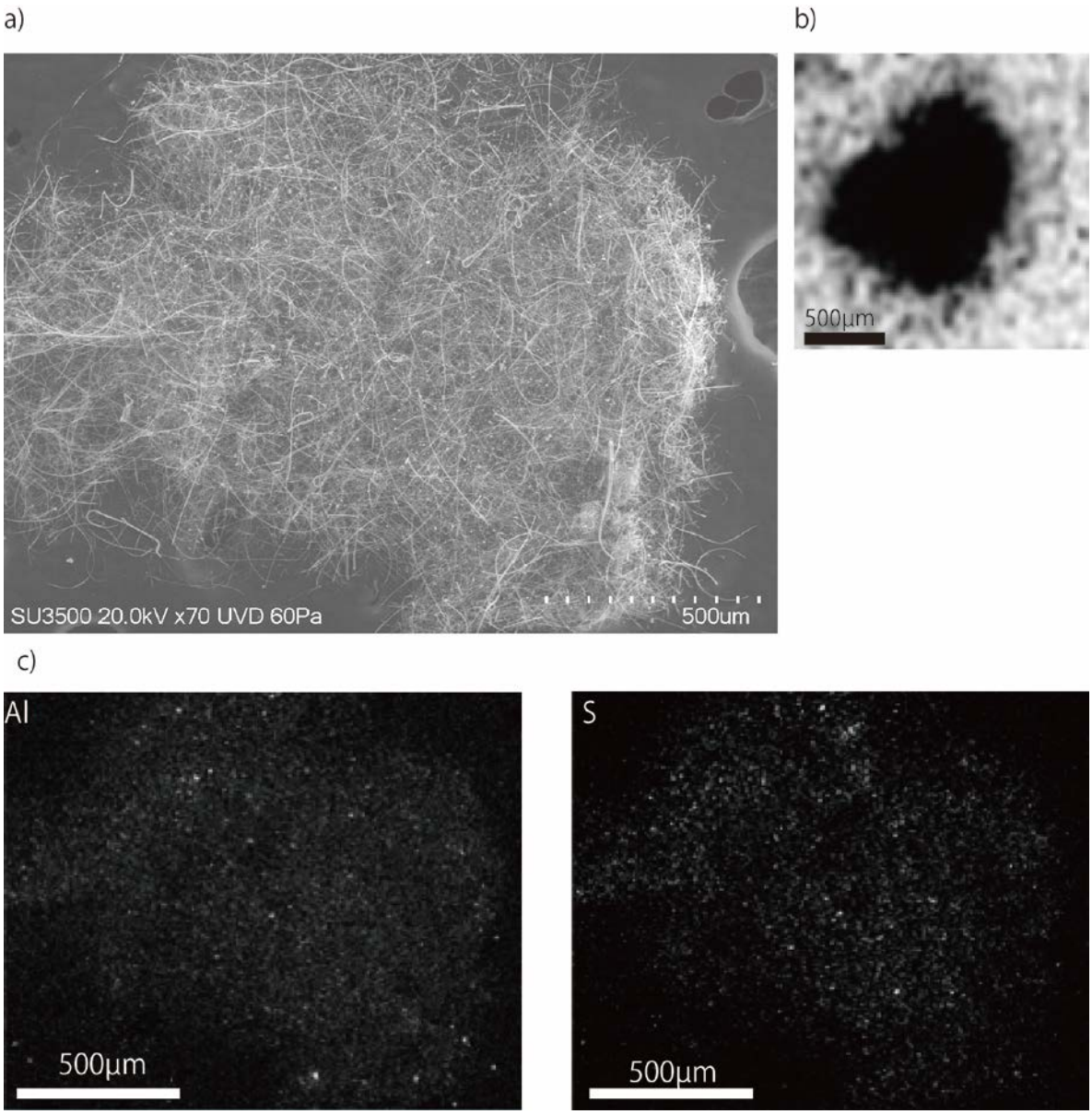
SI 5. The gamma spectrum of radioactive Cs particle 1. A 50,000 sec detection time was used. Peaks marked with * were from the background or the glass substrate. The sample was analyzed two years after the accident, and short-lived radionuclides would not have been detected even if they had been present when emitted.


a)
Glass substrate
A
Cs particle
Quartz fiber
Line profile
B
Carbon paste
SU3500 30.0kV x8.50k BSE-COMP 80Pa
5.00um
b)
Cs
2.5µm
c)
Zn
Fe
O
2.5µm
2.5µm
2.5µm
d)
O
Fe
Zn
Cs
CPS
A
B
0
2
4
µm
e)
C
O
Na
Al
Si
S
K
Ca
Cs
Cs
Fe
Zn
0
5
10 keV

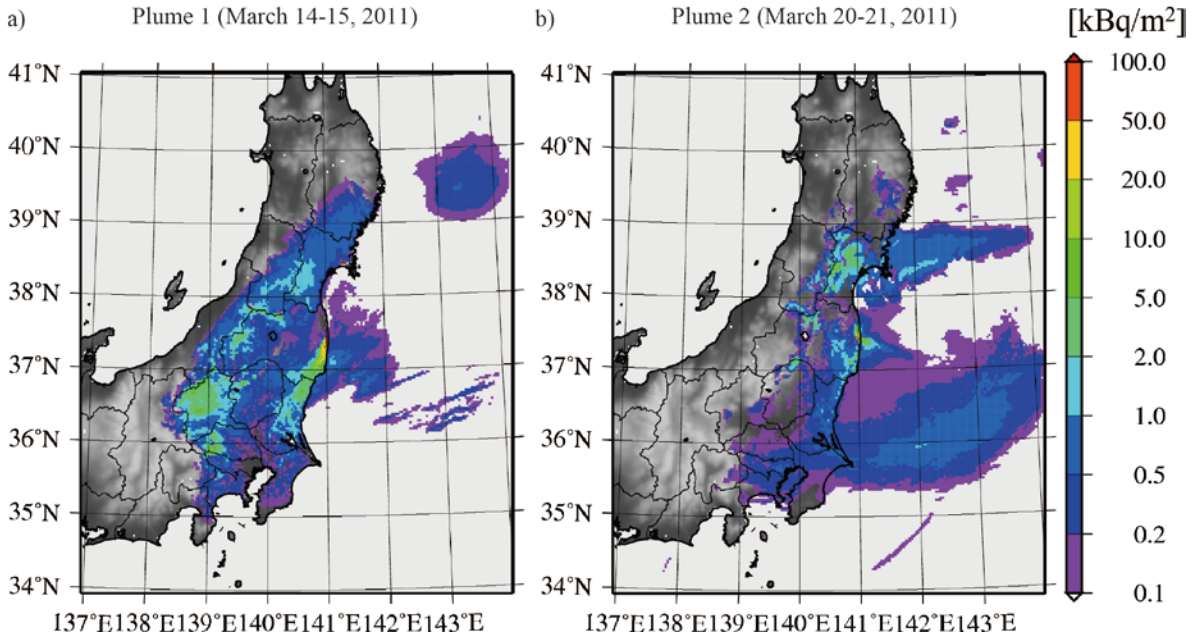
SI 6. The SEM analyses of radioactive Cs particle 2 from the March 14-15 sample. 1) An SEM image of the Cs-bearing particle. The particle was embedded within a carbon paste and was partially covered with a quartz fiber. b) The elemental mapping of Cs in the particle. c) The elemental mapping for Zn, Fe, and O in the particle. Unlike in particle 1, oxygen is not apparent. d) The line profile of the selected elements over the particle. The analyzed line is shown in a). e) The EDS spectra of the particle (black line) and glass substrate (red line).



SI 7. SEM images of the Cs particle 1 before and after exposure to water. There is no change between the images, suggesting that the particle is largely insoluble to water. Scale bars: 1 µm.



SI 8. The SEM analyses of the filter sample from March 20-21. a) An SEM image of a filter segment containing a diffused spot (Fig. 2). The filter was divided into several layers (SI 4-b), and this image corresponds to one of those layers. b) An IP image of the filter. Radioactive materials are distributed all over the filter. c) Elemental mapping images for Al and S, which represent aluminosilicate minerals and sulfate particles, respectively. These particles are distributed all over the filter.



SI 9. The total (dry + wet) deposition of ^{137}Cs released from the FNPP between (a) March 14, 17:00 and March 15, 02:00 (JST) and (b) March 19, 20:00 and March 20, 07:00. The Cs carriers are assumed to be water-soluble submicron particles, such as sulfates, in both plume. The Regional Air Quality Model 2 (RAQM2) was used for the model calculation. We used the Generic Mapping Tools (GMT) developed at University of Hawaii to draw the figure. The model elevation in the figures was generated based on a 1km resolution Global 30 Arc-Second Elevation (GTOPO30) of U.S. Geological Survey (USGS).