



# High Radioactivity Particles in Japanese House Dusts

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*(amended 5/19/2015 to correct mass of, and dose/risk from Nagoya hot particle)*

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Marco Kaltofen, Nuclear Science and Engineering

## **Presenter disclosure:**

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## Fuel fragmentation and dispersal



Above: Dust from a failed fuel rod, from US NRC NUREG 2121, *Fuel Fragmentation, Relocation, and Dispersal During the Loss-of-Coolant Accident*, 3/2012

The 2011 earthquake and tsunami in Japan damaged nuclear reactors in Fukushima, Japan. Radioactively-contaminated gases, aerosols and particulate matter were released.

These radioactively-contaminated dusts settled inside homes, where residents may be exposed to dusts by contact, inhalation or ingestion.

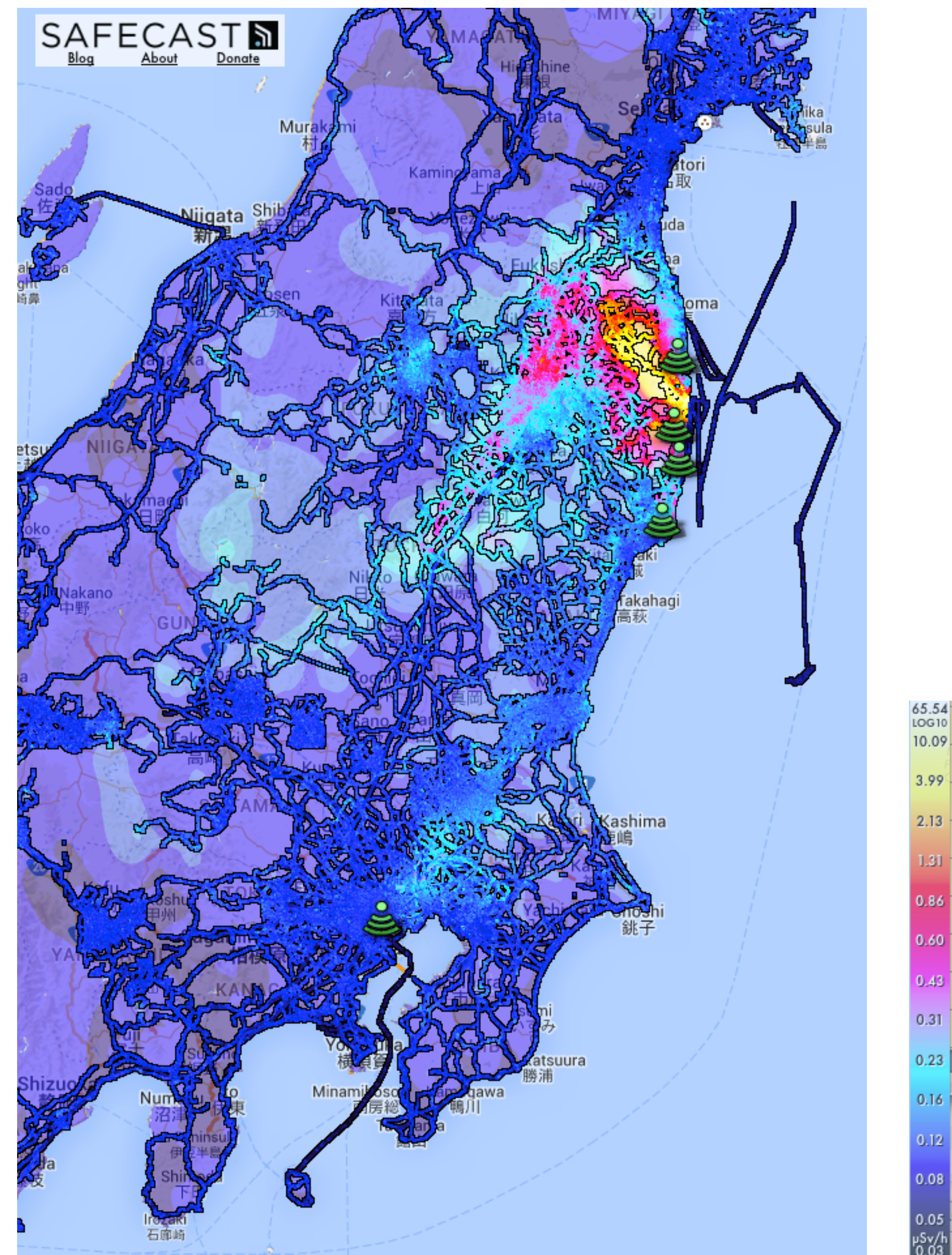


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Radioactively-hot particles on the respirable size range were routinely detected in a set of 84 household and street dust samples received from Japan.

Map by Safecast.org, used by permission.



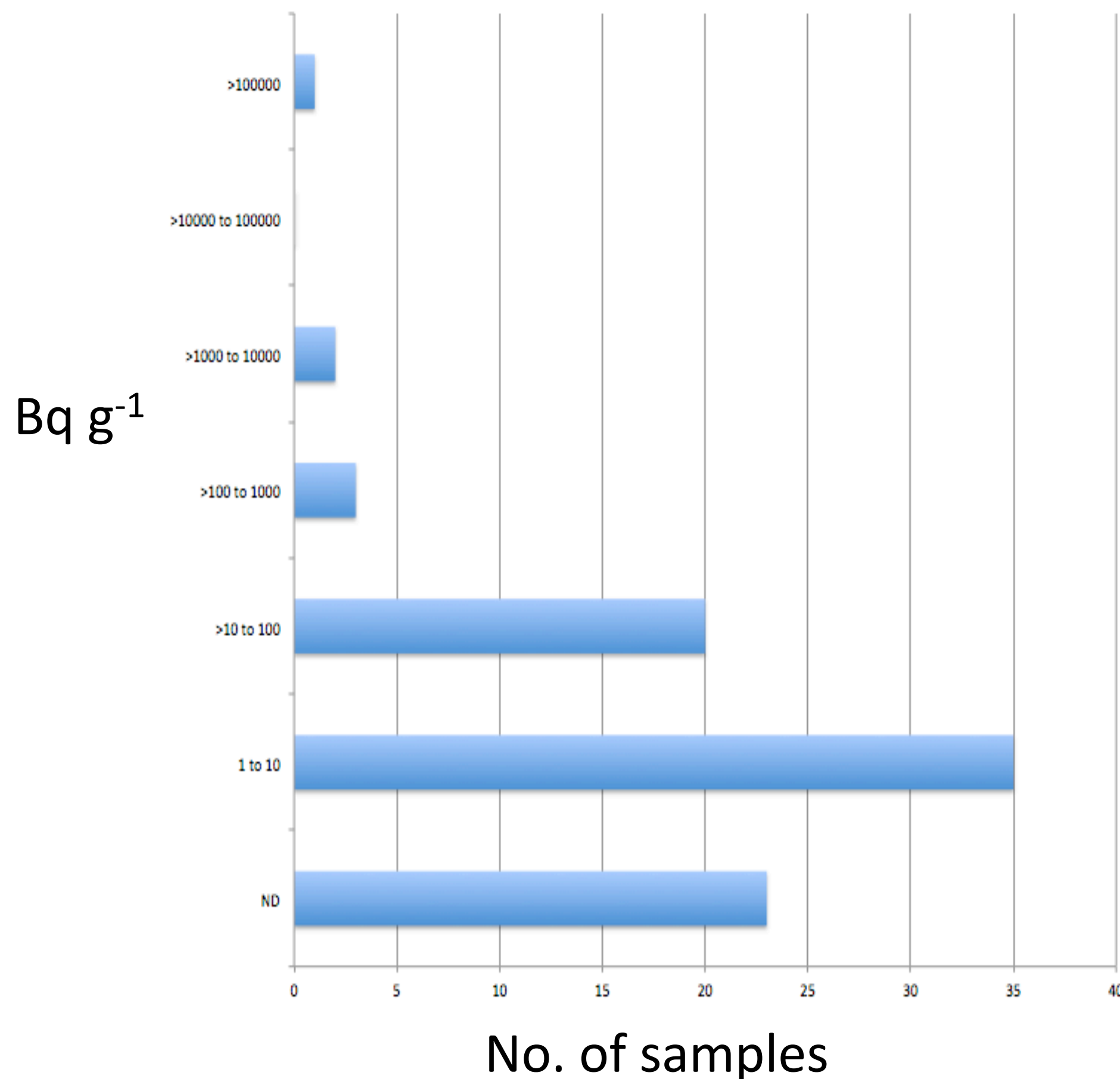




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## Japan dust data



Five of eighty-four dust samples had activities two or more orders above the median.

One sample had no detectable excess radioactivity except for a hot particle which had uniquely high activity.

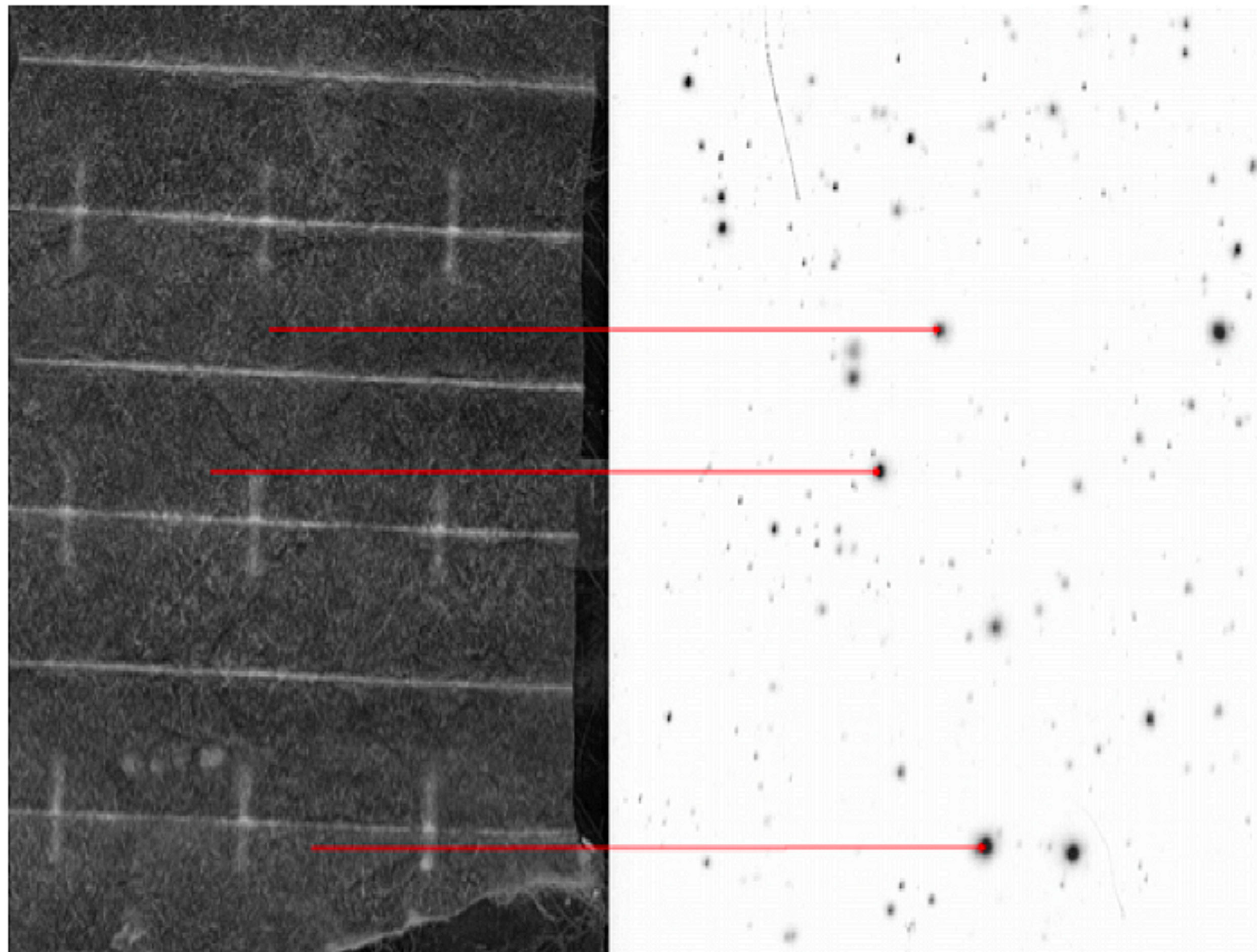
Median activity was 2.5 Bq g<sup>-1</sup>

Mean activity was 72 Bq g<sup>-1</sup>

RSD = 470 % including outliers



## Hot particle detection by autoradiography



Tokyo vehicle air filter image (Left) and 7 day exposure autoradiograph (Right).

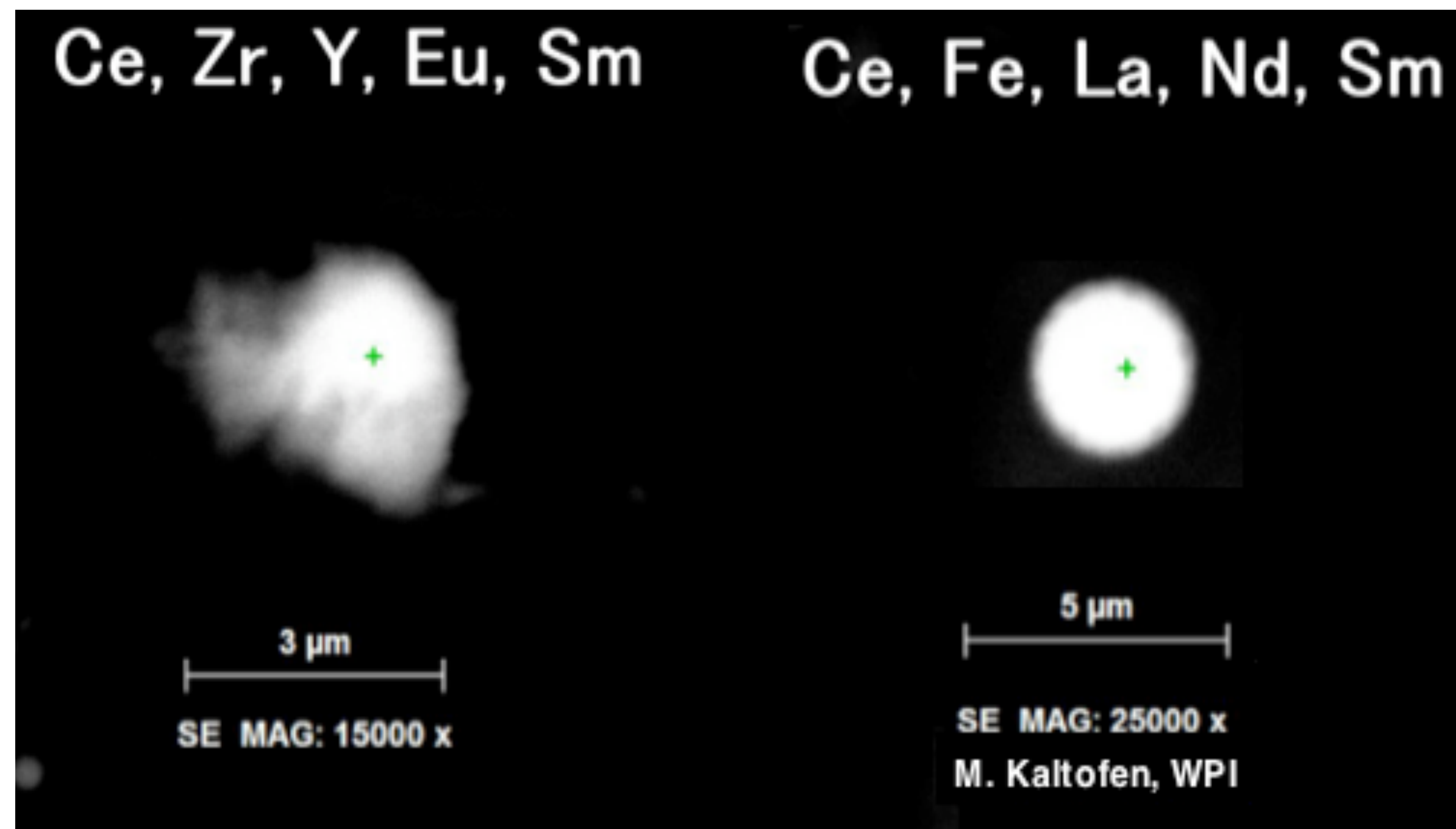
Corresponding points are connected by red lines.



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**Hot particle sizes were  $< 10\text{ }\mu\text{m}$**



SEM analysis showed that the majority of the radioactively-hot particles detected were  $10\text{ }\mu\text{m}$  or less in size, meaning that they were potentially inhalable.

Nal gamma spectrometry detected  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$ , in all dust samples with activities significantly above background.

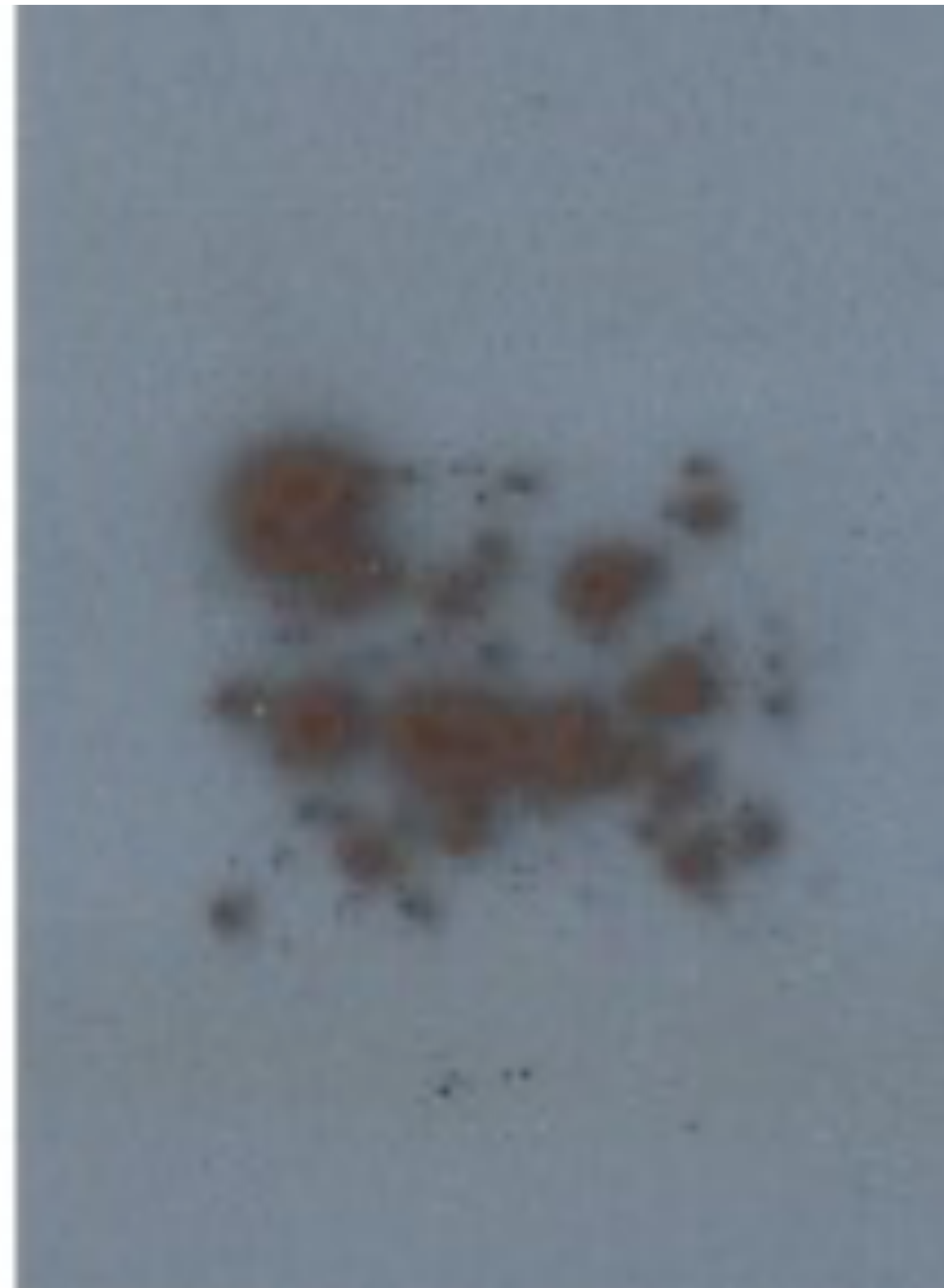
The isotopes  $^{60}\text{Co}$ ,  $^{226}\text{Ra}$ ,  $^{241}\text{Am}$ , and/or  $^{230}\text{Th}$  were detected in 5 of 84 samples using SEM/EDS.



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### Autoradiograph of black sand shows uniform activity.



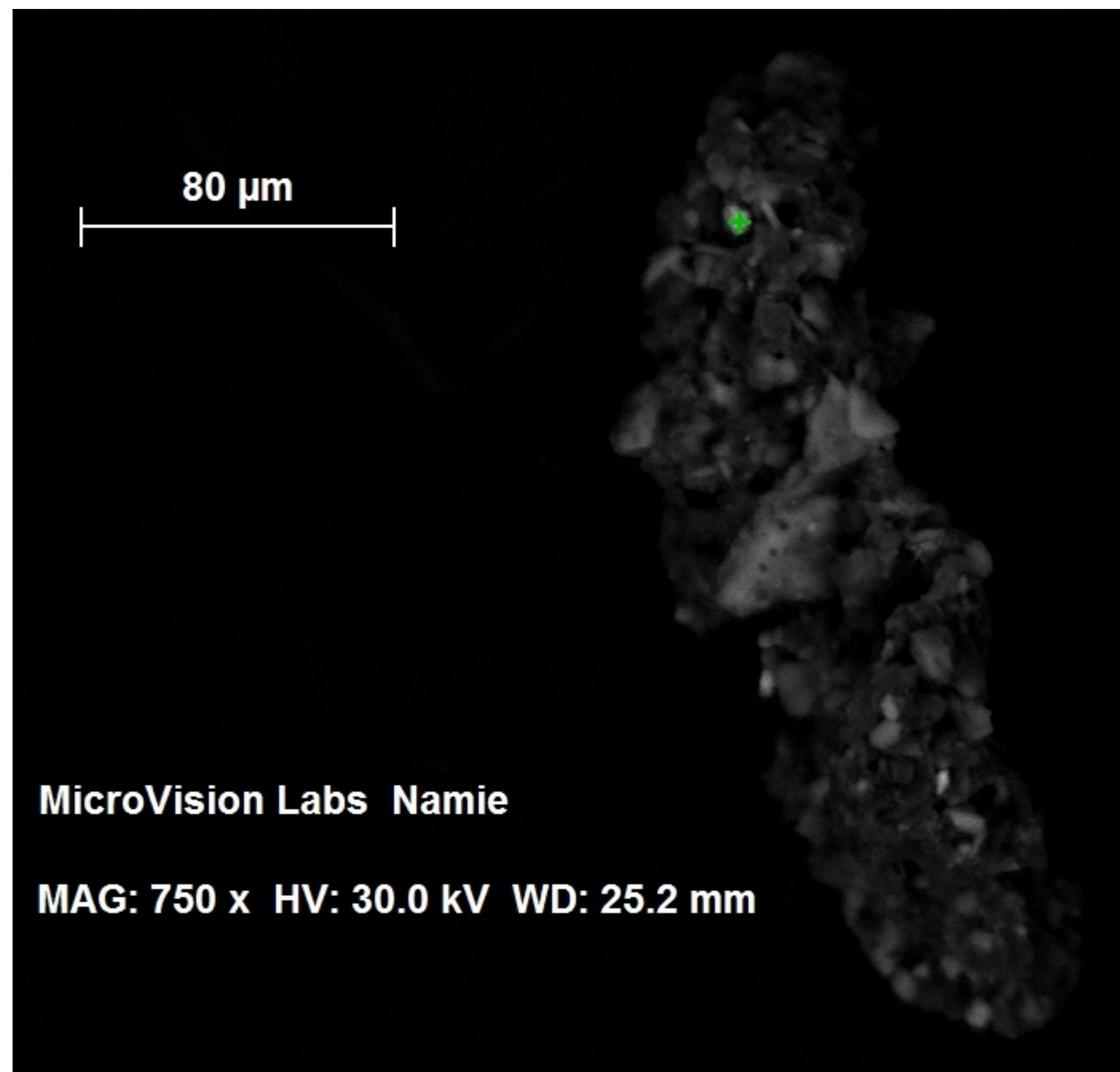
A sample of “Black Sand” was collected from Namie-machi, Futaba-gun, Fukushima Prefecture, alongside the exclusion zone fence.

It had a uniform activity of 1.5 MBq/kg of the fission products  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .

The sample also had 300 Bq/kg of  $^{60}\text{Co}$ . This isotope is a neutron activation product of iron.



## Black Sand sample from Namie, Japan

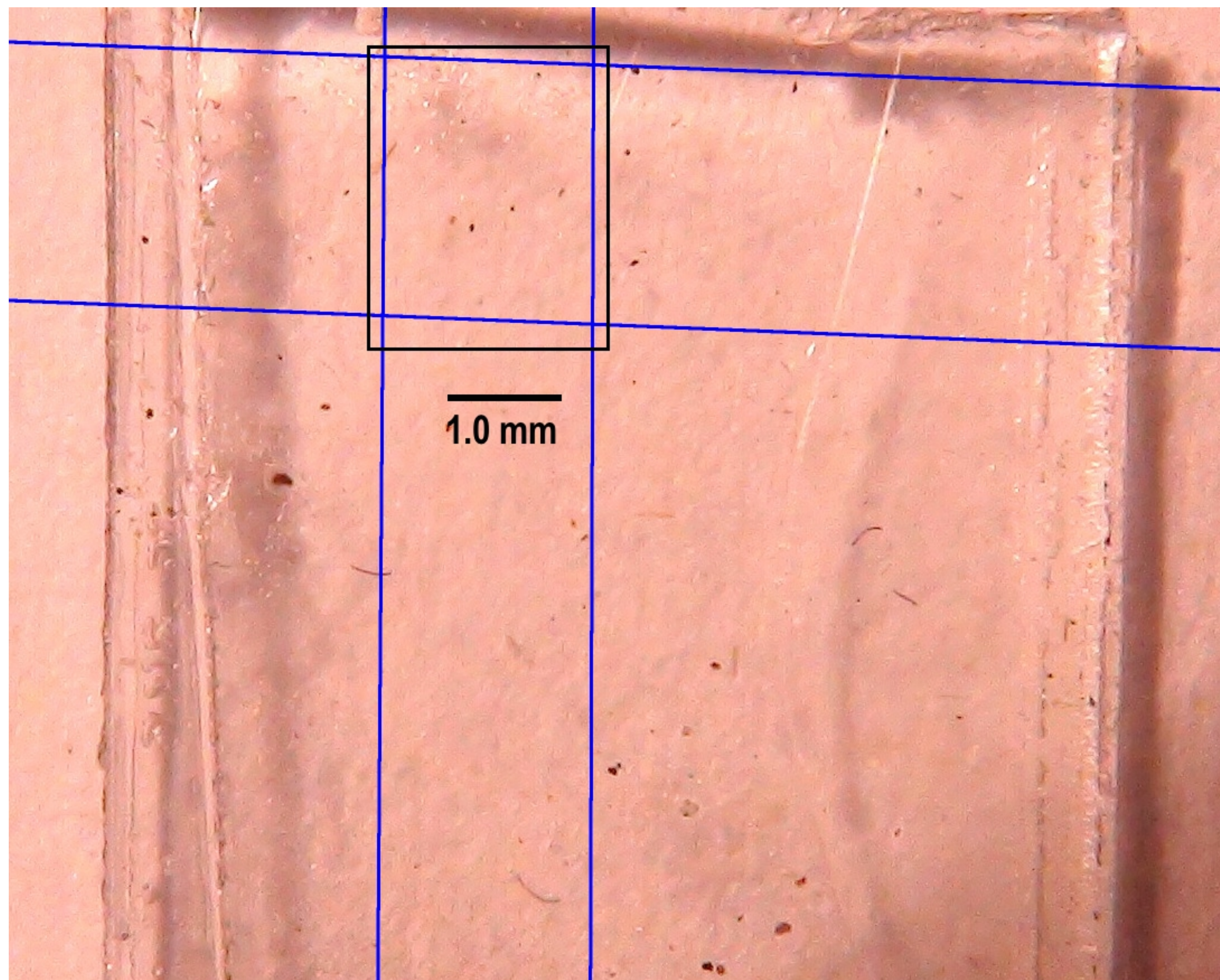


Contained thorium-cerium monazite crystals, 5 to 20  $\mu\text{m}$ , Y-Yb, and Hf-Pu-U particles, < 10  $\mu\text{m}$ .

Potentially nonradioactive artifacts included flakes of K-Br, 10 to 100  $\mu\text{m}$ , spherical W, < 10  $\mu\text{m}$ , Ga-As crystals, 10  $\mu\text{m}$ , and Cu-Sn flakes, < 30  $\mu\text{m}$ .



## Atypically-hot particle from household dust

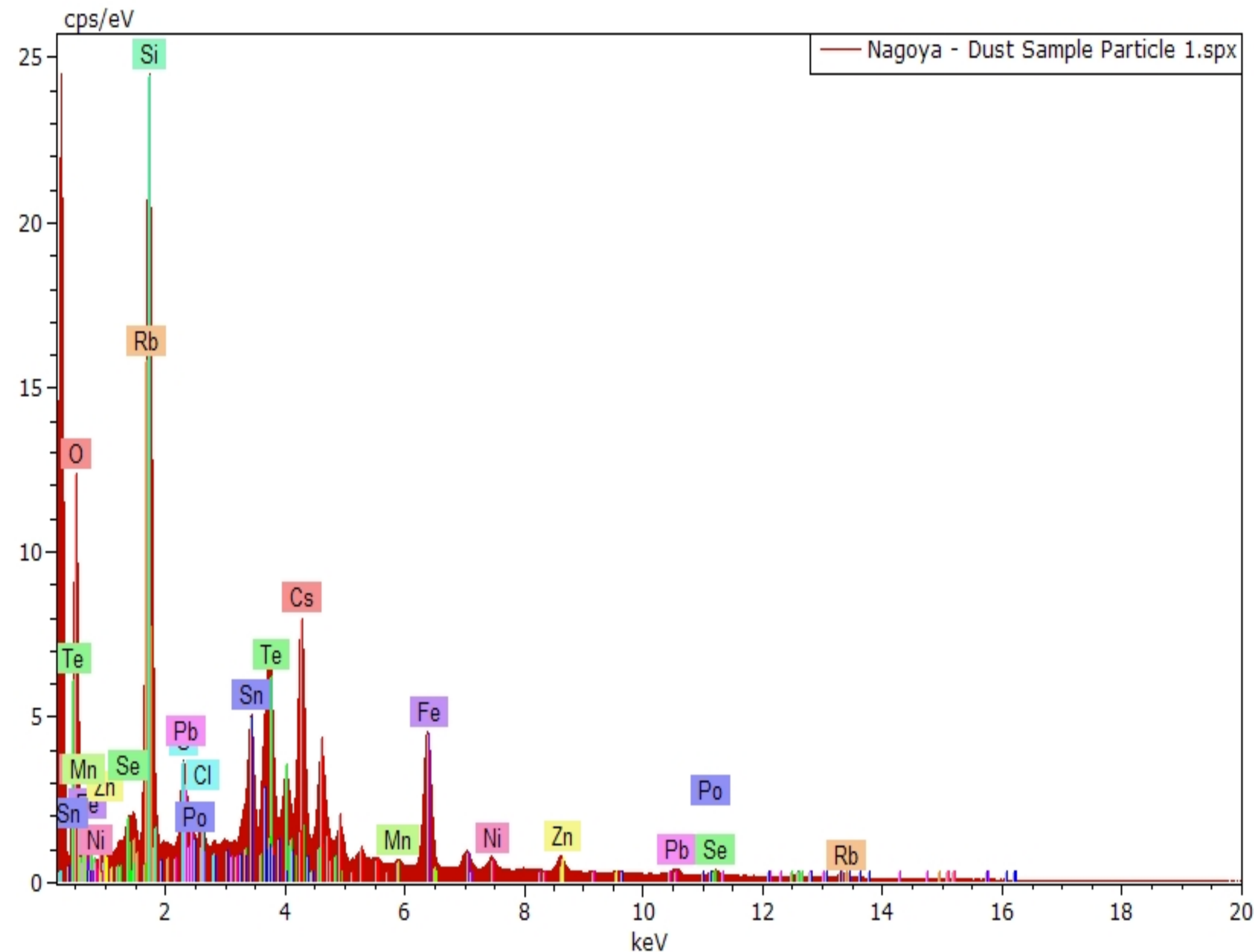


The excess radiation in a 0.3 kg vacuum cleaner bag dust sample from Nagoya, Japan came entirely from one particle.

The hot particle was isolated by sample division.



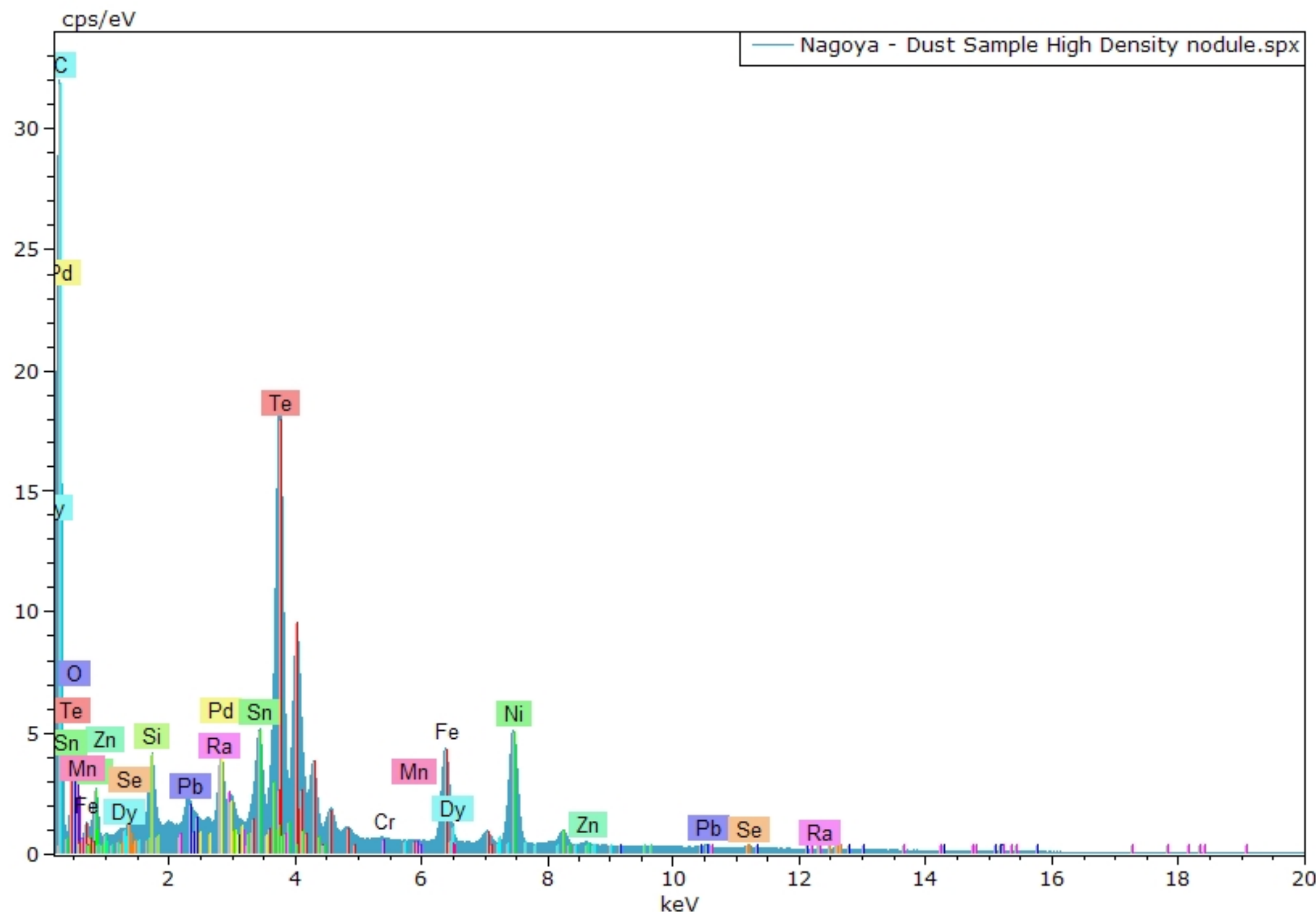
**High activity particle is an aggregate of smaller particles.**



Particle contained cesium oxides and other fission products.

The particle also contained radium and polonium, which are decay products of the original nuclear fuels.

## High activity particle X-ray microanalysis results



Tellurium up to 48.0 %,  
Cesium up to 15.6 %,  
Rubidium up to 1.22 %,  
Polonium up to 1.19 %,  
Dysprosium up to 0.18 %

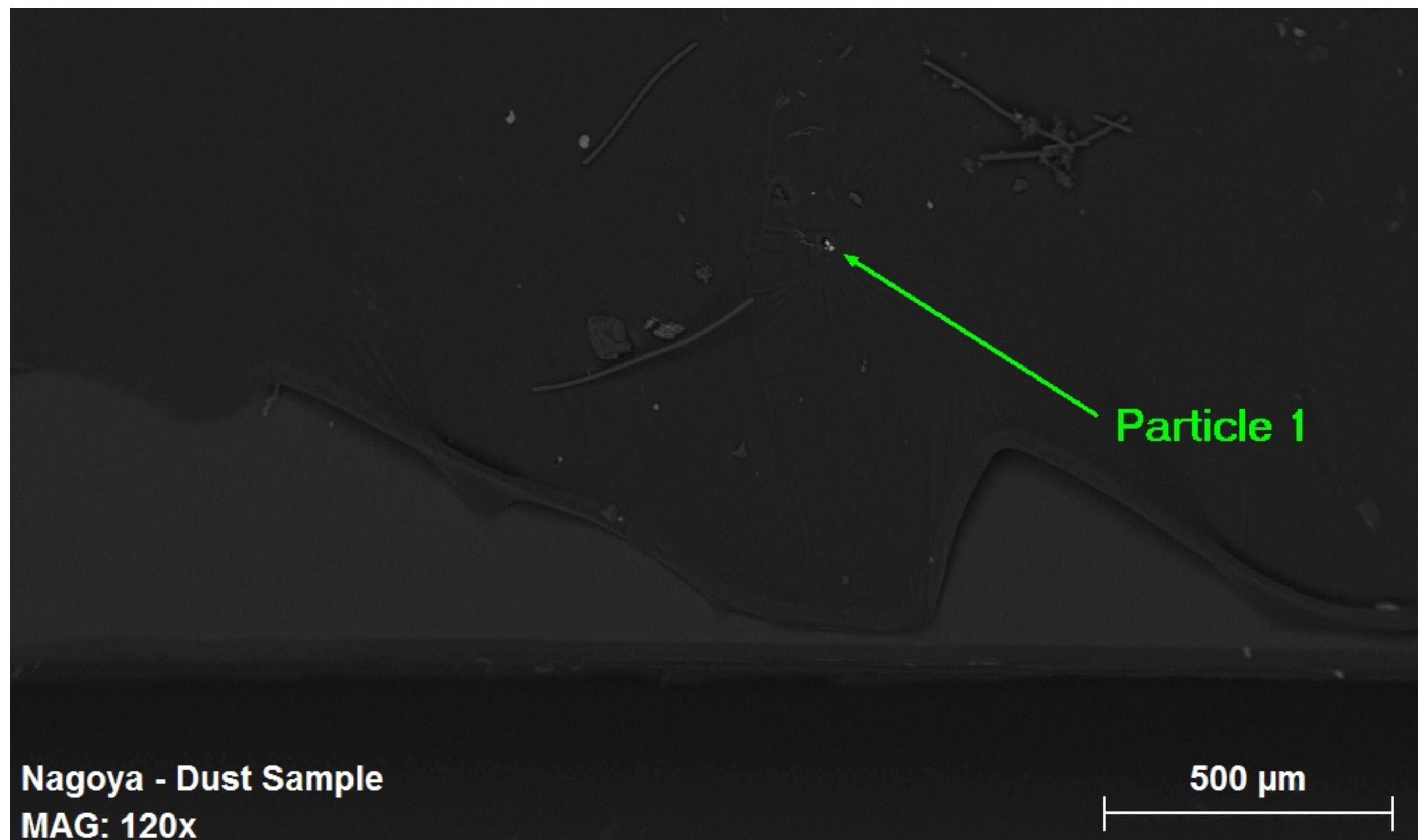
Individual subparticles also  
contained tin, radium, lead,  
nickel, iron, and chromium.



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## 120X Scanning electron microscope view of hot particle.

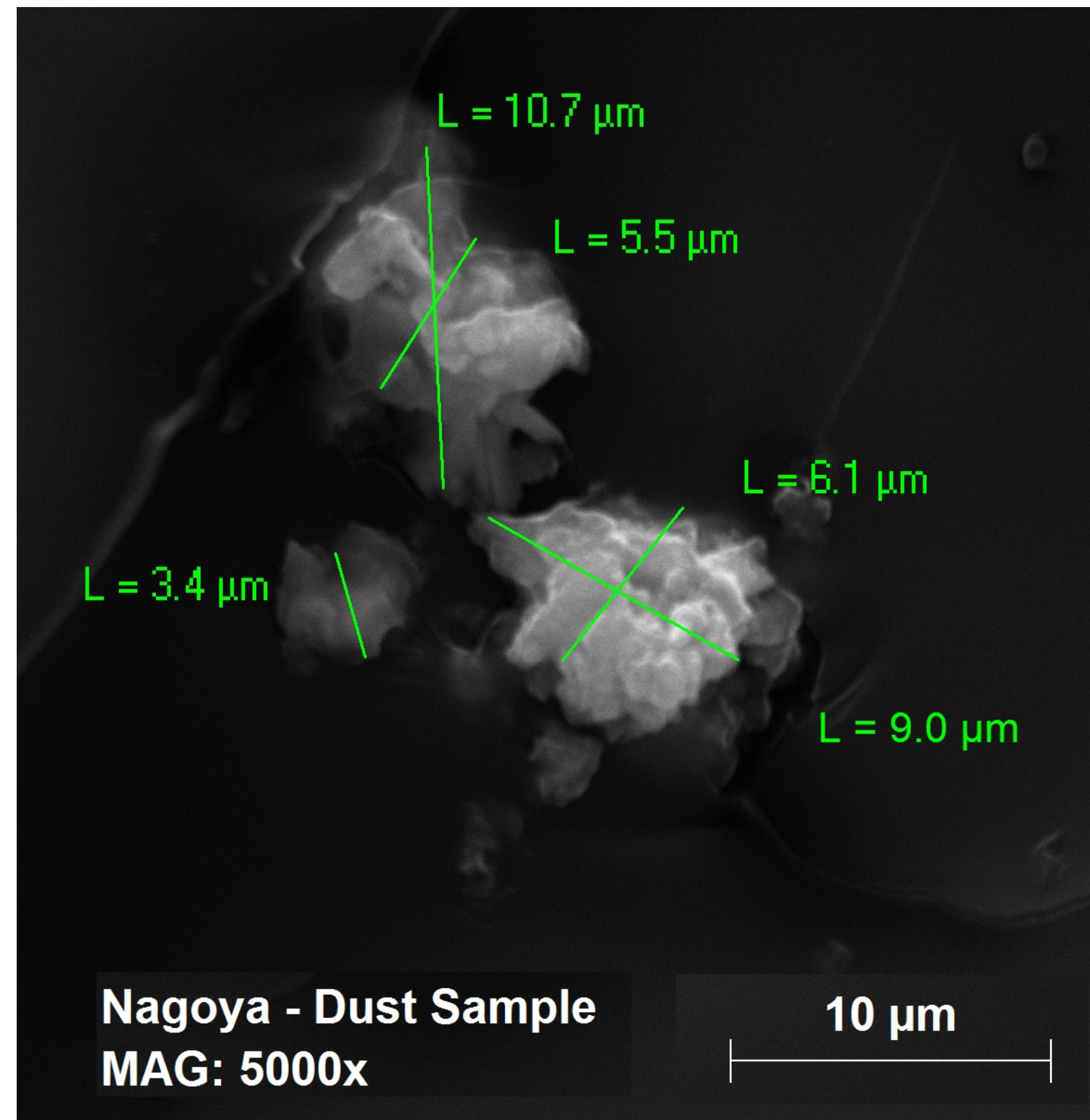


Activity standardized  
against  $^{137}\text{Cs}$  is:  
 $2.03 \pm 0.05 \text{ kBq}$ .

This yields a potential  
internal dose of  
 $1.15 \times 10^{-3} \text{ J/yr}$ .  
*(amended 5/19/2015)*



## Calculating activity and potential dose



The target organ is presumed to be the lung, which has a typical mass of 0.88 kg at adulthood, NIH 2014 data.

This gives a lung dose of 1.15 mGy annually, or a committed lung dose of 0.03 Sv, adjusted for decay.

Particle density =  $4.2 \text{ g/cm}^3$

Particle volume =  $933 \text{ } \mu\text{m}^3$

Specific activity =  $5.2 \times 10^{15} \text{ Bq/kg}$

a.k.a. 5.2 PBq/kg.

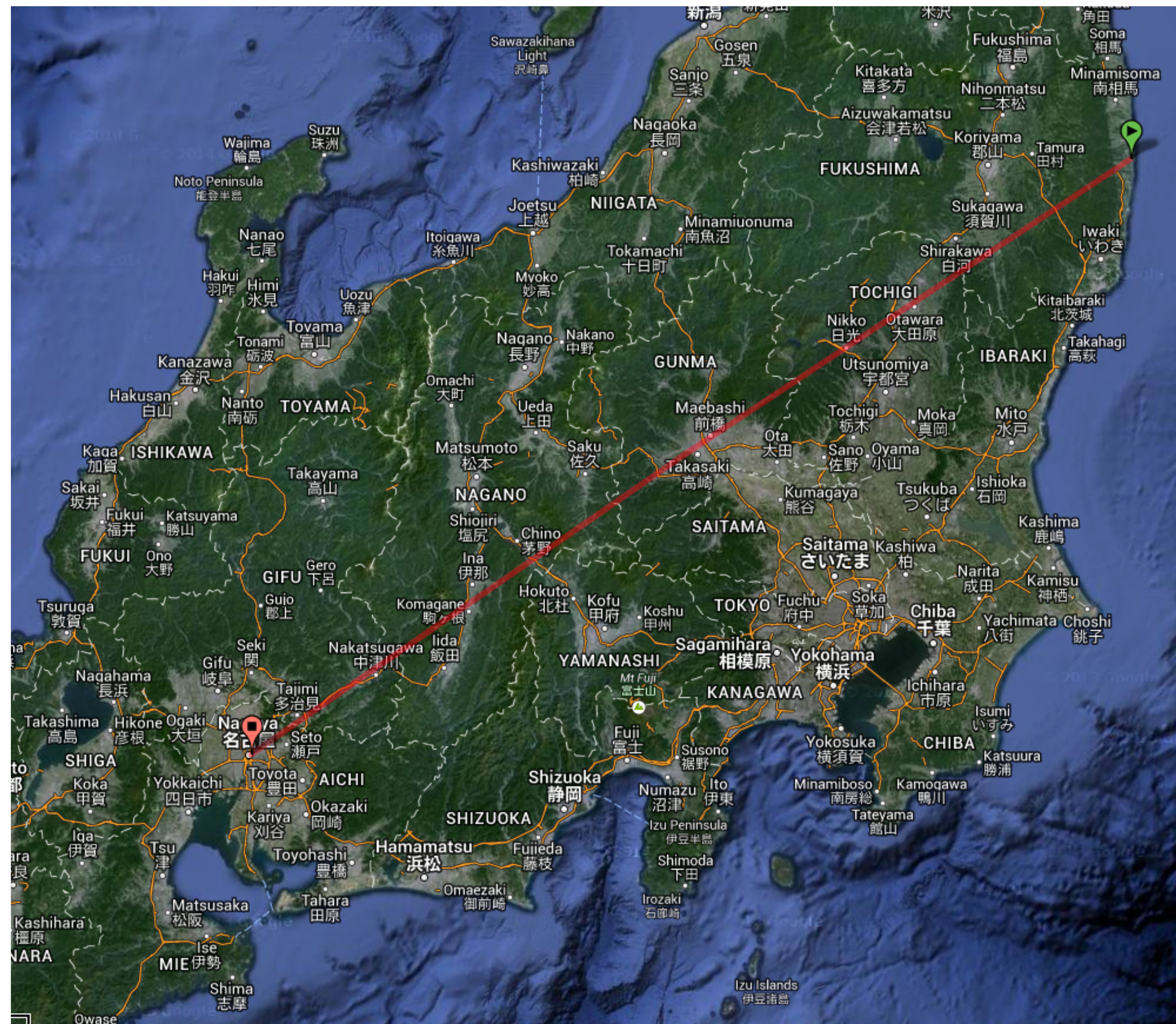
*(amended 5/19/2015)*



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## How far did this particle travel?



The distance between the hot particle sample site and Fukushima Daiichi is 450 km.

The particle is small enough to have travelled by air, but it also can be accidentally transported on clothing or vehicles.





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## Calculating excess cancer risk



Using RADRAT, the National Cancer Institute Radiation Risk Assessment Tool. For a male child, age 2 years when initially exposed in 2011, with a lung exposure of 1.15 cGy per year, the mean excess lifetime cancer risk is: 0.5 per 1000.

*(amended 5/19/2015)*





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## US/Canada Sample Data

### *Airborne dust samples:*

Positive autoradiographic results for Seattle, WA dusts

2X increase in total beta activity of air filters in March/April 2011

No detections of hot particles with Cs, Am, or Pu

### *Soil samples:*

Max. result:  $0.26 \pm 0.04$  Bq g<sup>-1</sup> for combined <sup>134</sup>Cs, and <sup>137</sup>Cs

Confirmed by USGS data (See: Wetherbee et al, 2012)

### *Seaweed samples:*

1/45 samples positive for combined <sup>134</sup>Cs, and <sup>137</sup>Cs,

Positive sample collected outside Bremerton Naval Base in WA.



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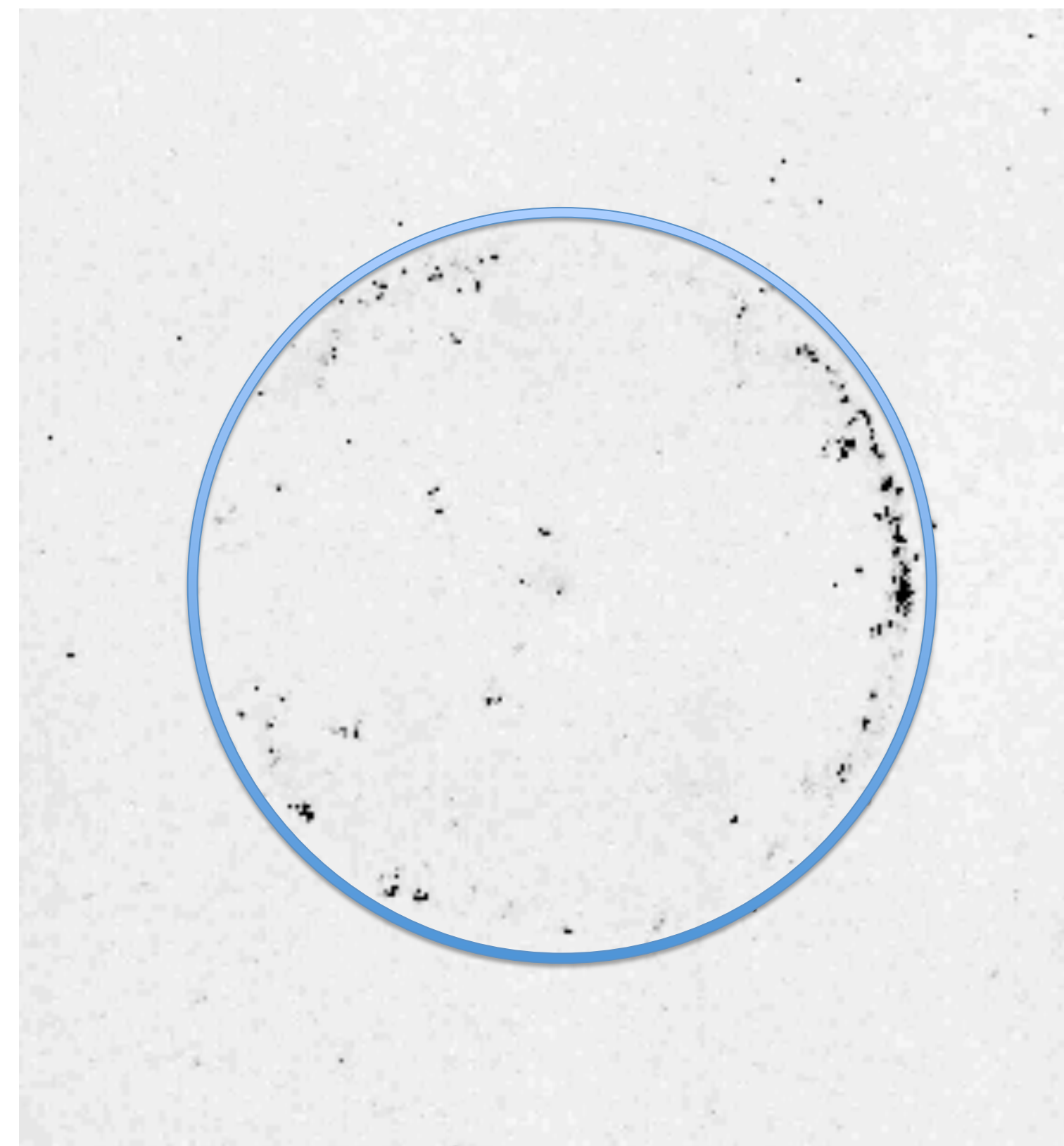
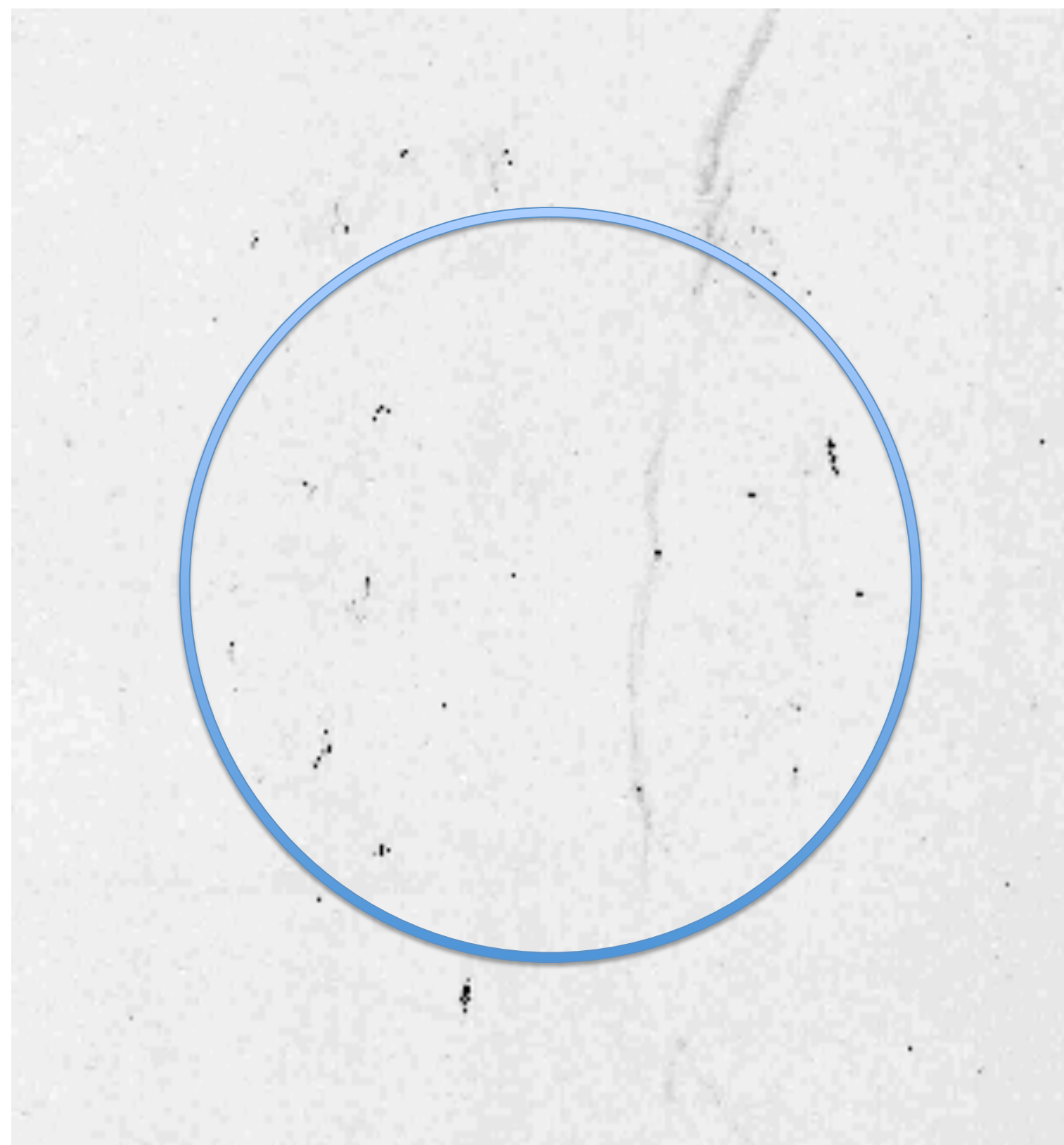
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**Seattle, WA data: 1x to 2x background for total beta in air.**

Autoradiographs of quantitative 35 mm air filters

Left: March 28 to April 1, 2011

Right: April 11 to April 14, 2011







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## What can you do with this research?

You can measure the actual or probable radiation dose to an individual, and calculate their potential excess cancer risk.

The drawback is that you have to measure the actual intake or calculate the probability of exposure to hot particles.

In this sample set, where there is some skewness, the mean activity is significantly higher than the median activity.

In practical terms, this means that a few individuals experience a higher dose than average data would suggest.





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## Conclusions

Radioactively-hot particles on the respirable size range were routinely detected, with one atypically hot particle sampled 450 km from the release site.

The dust sample set mean activity was significantly higher than the median activity due to these atypically hot dust samples or individual particles.

Some individuals may actually receive a dose that is skewed higher than the mean dose calculated from average environmental data.