

High Radioactivity Particles in Japanese House Dusts

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Hypothesis: The Fukushima Dai-ichi accident released very high activity inhalable dust particles that travelled long distances.

Introduction: The 2011 earthquake and resulting tsunami in Northeastern Japan led to damages to four of the six nuclear power units at the Fukushima Dai-Ichi Station. Radiological materials escaped from the reactor units at the power plant site via air-borne plumes of contaminated gases, aerosols and particles, and by contaminated wastewaters.

Airborne dusts can transport radioactive materials as isolated individual particles containing high concentrations of radioisotopes. Alpha and beta emissions related to fission wastes and dispersed fuel particles are hazardous when inhaled or ingested. Radioactively-contaminated environmental dusts can accumulate in indoor spaces, potentially causing significant radiation exposures to humans via inhalation, dermal contact, and ingestion. These heterogeneously distributed hot particles can be difficult to detect and measure, making it likewise difficult to determine radiation dose to residents of contaminated areas.

Methods: Dust samples contaminated with Fukushima-related fission products were identified using gamma spectrometry. A set of high activity hot particles were isolated from the house dusts by autoradiography and physical separation of identified hot spots on air filter media. After isolation, hot particles were analyzed via scanning electron microscopy/energy-dispersive X-ray analysis. (SEM/EDS).

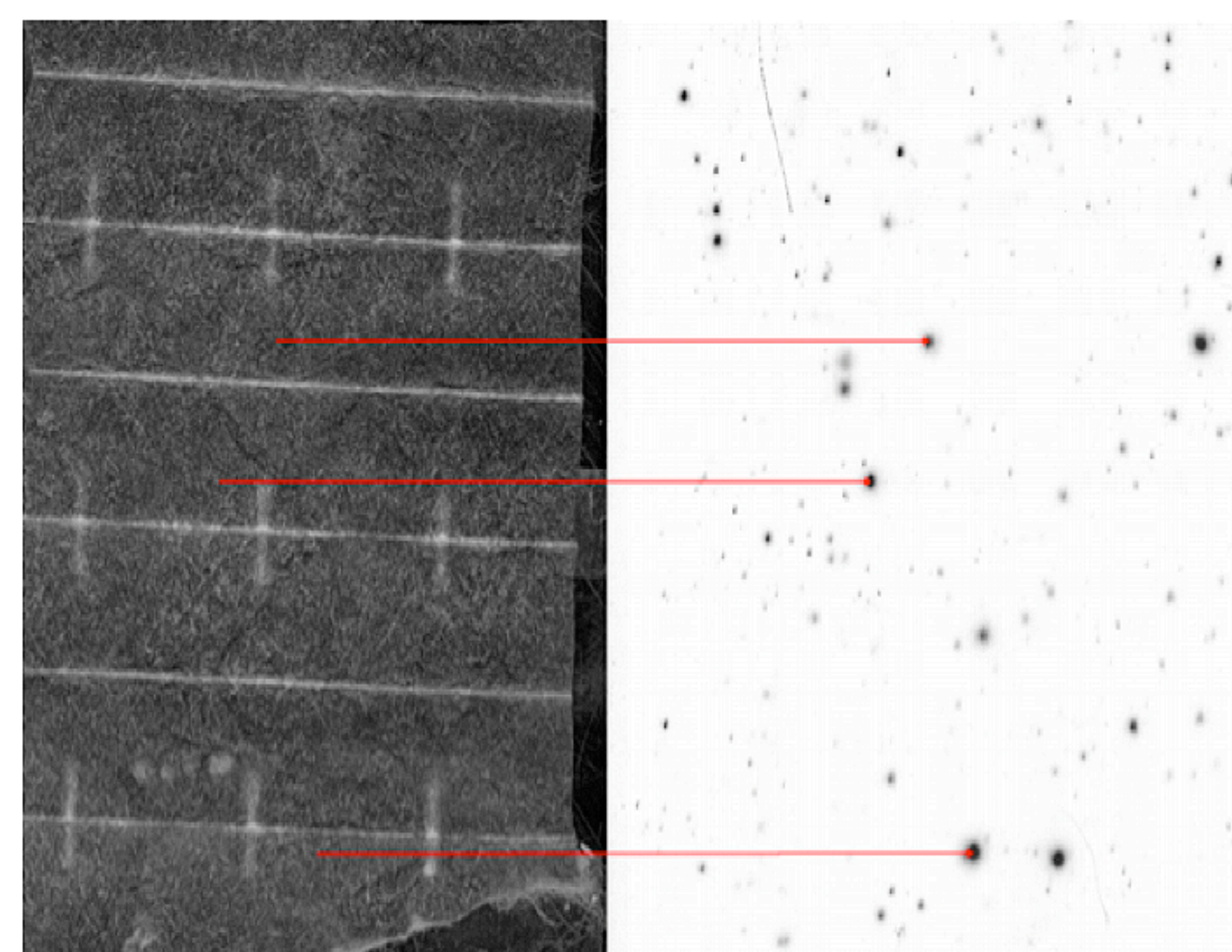
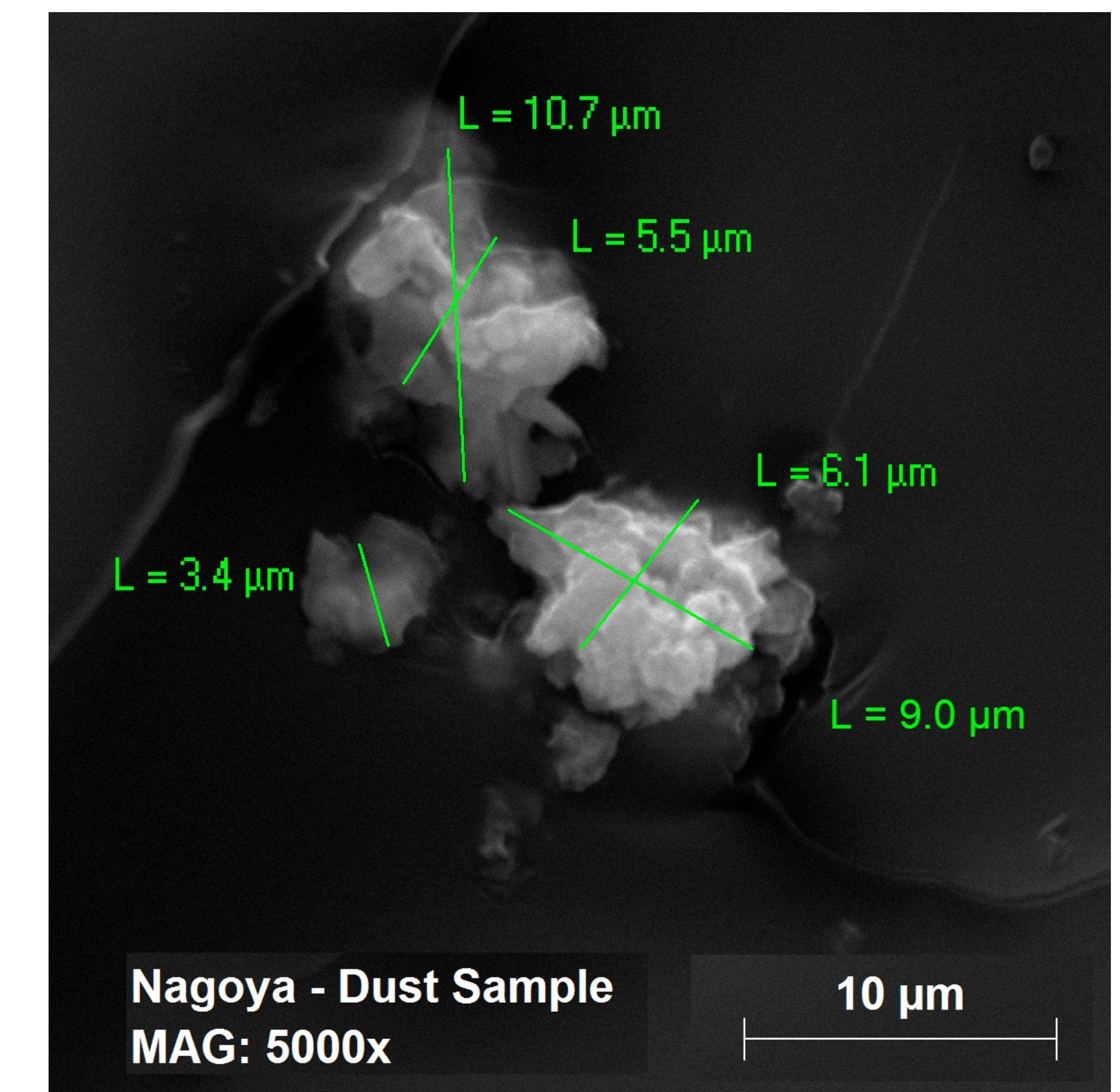
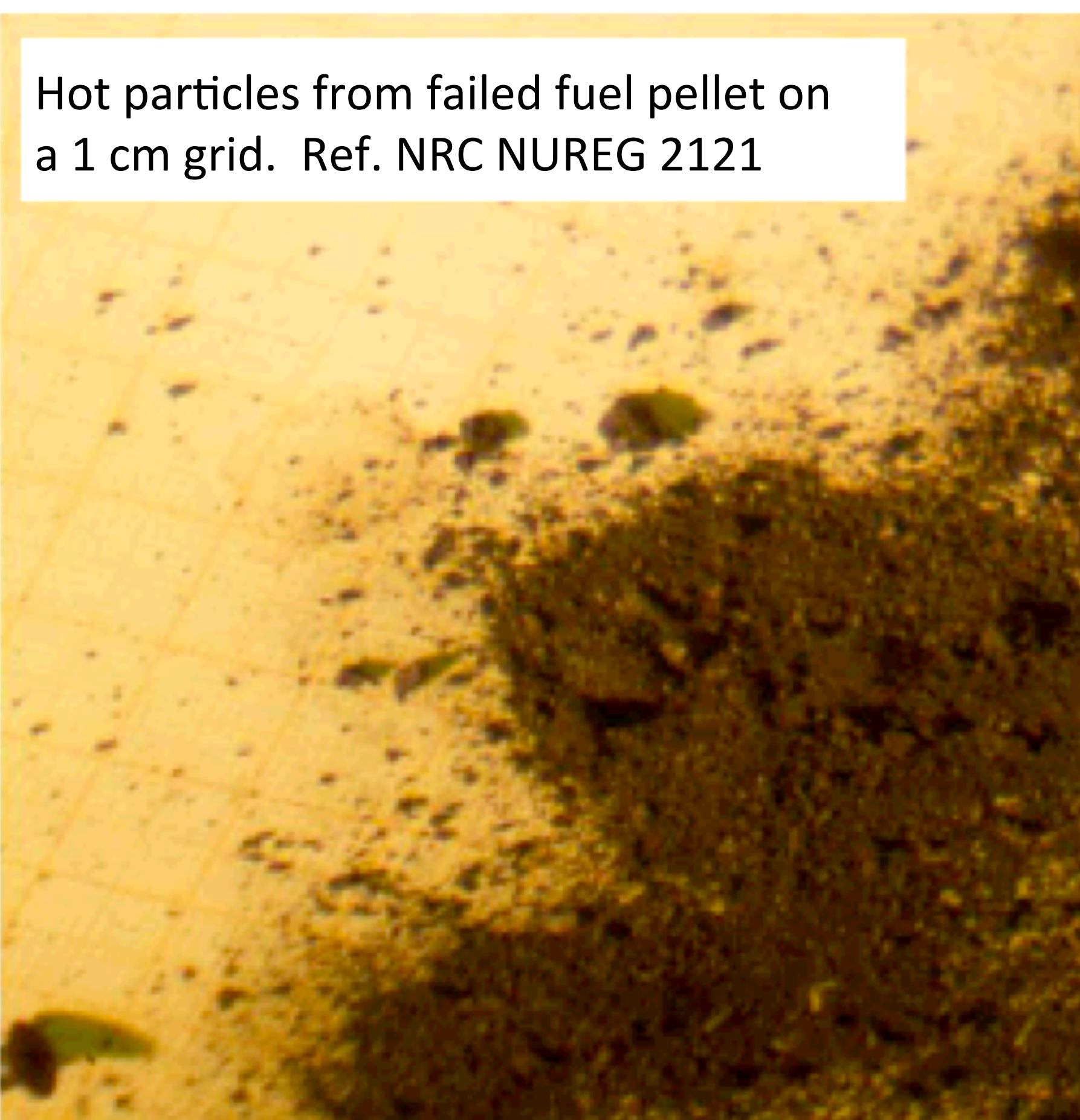
Results: The median total activity of eighty four Japanese bulk house dust samples studied was 2.5 KBq kg⁻¹. The mean total activity was 71.6 KBq kg⁻¹ with $\sigma = 339$ KBq kg⁻¹. Most of the activity detected was due to ¹³⁴Cs, ¹³⁷Cs, ⁶⁰Co and ²²⁶Ra. Short-lived ¹³¹I decayed away after gamma spectral measurements were made, but before hot particle analyses were completed. Cesium isotopes were in concentration ratios definitive for Fukushima discharges.

The large difference between the mean and median resulted from the contributions of two samples with activities above 1.0 MBq kg⁻¹ and a micron-scaled particle with activity greater than 1.0 PBq kg⁻¹. The particle was collected from a home in Nagoya, Japan. Nagoya is 460 km from the accident site. The particle's activity was 310 Bq as measured by gamma spectrometry. Its beta activity was 285 Bq. It contained both fission products and decay products of ²³⁸U at percent levels. X-ray microanalysis of this particle mapped varying concentrations of 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 %, as well as trace amounts of tin, lead, nickel, iron, and chromium. The very hot particle has a calculated volume of no more than 0.0012 mm³. Based on its composition as measured by SEM/EDS, its density is about 3.6 g cm⁻³.

NaI gamma and EDS spectrometry of hot particles found ²²⁶Ra, ¹³⁴Cs, and ¹³⁷Cs, ²⁴¹Am, and ²³⁰Th as the most commonly detected gamma photon-emitting isotopes. Autoradiographic, gamma spectral and SEM/EDS results demonstrated that qualitatively similar particles were present in about 25 % of dusts sampled. This quartile of the samples was contaminated with ¹³⁴Cs, an indicator contaminant for the reactor accident, and was autoradiographically positive for hot particles. SEM analysis showed that the majority of these hot particles were 10 um or less in size, meaning that they were potentially inhalable.

Conclusion: Radioactively-hot particles on the respirable size range were routinely detected, with one as far as 460 km from the release site.

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Tokyo vehicle air filter (L) autoradiograph (R)

