Widespread distribution of radiocesium-bearing microparticles from the Fukushima nuclear accident

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011 emitted a considerable amount of radioactive materials. As a form of radioactive materials emitted from the FDNPP at the early stage of the accident, radiocesium-bearing microparticles (CsMPs) have been investigated by numerous researchers in recent years [1,2]. They are solid particles consisting primarily of silicate glass and contain trace amounts of various heavy elements associated with nuclear fuel and fission products (FPs). Because they do not easily dissolve in water, there is concern that they may have a long-term impact on the environment and human body.

A careful investigation of suspended particulate matter (SPM) collected hourly on filter tape at automated air pollution monitoring stations across Eastern Japan revealed that radioactive materials emitted from the FDNPP were transported over Eastern Japan via several major plumes during 12–23 March 2011 [3]. Among them, the second plume (P2) spread into the Kanto Plain, including the Tokyo metropolitan area, on 15 March. Hypothesizing that CsMPs were the major carrier of radioactive Cs in P2, we aim to verify the widespread distribution of CsMP in P2 over the greater Kanto Region (in and around the Kanto Region), focusing on how far CsMPs were transported southwest from the FDNPP. We examined radioactive aerosol particles from the SPM filter tape samples collected hourly on 15 March at several stations in the greater Kanto Region to compare their physical/chemical characteristics with those of previously reported CsMPs isolated from various environmental samples.

To nondestructively investigate the chemical characteristics of CsMPs isolated from the SPM filter tape samples in detail, multiple X-ray analyses using a synchrotron radiation microbeam (SR-μ) X-ray were conducted at SPring-8 **BL37XU** [4]. X-ray fluorescence (XRF) analysis using a high-energy SR-μ-X-ray was applied to carry out the nondestructive identification and qualitative detection of trace amounts of heavy elements within individual microparticles. The chemical state analysis of some metal elements in the particles was carried out by X-ray absorption near-edge structure (XANES) analysis. X-ray powder diffraction (XRD) analysis was also conducted to reveal the crystal structures of the particles. Brilliant X-rays from an advanced SR light source at SPring-8 enabled the use of a combination of these X-ray analytical techniques in a nondestructive manner.

Eight CsMPs were successfully isolated from SPM filter tape samples collected at seven monitoring stations (A–G) in the greater Kanto Region, including the Tokyo metropolitan area, on 15 March 2011 (see Fig. 1). The particles had a spherical shape with diameters of ~1 μm. Gamma-ray spectra of the eight particles indicated both ^{134}Cs and ^{137}Cs in each particle with activity ratios of ~1.0 (decay corrected as of March 2011), suggesting that these particles were emitted from either reactor No. 2 or 3 of FDNPP. As a result of the SR-μ-XRF analyses of individual particles, they were found to have qualitatively similar chemical compositions. The SR-μ-XRF spectra of four representative particles (A1, B1, C1, and E1) are shown in Fig. 2. The XRF analysis using a monochromatic SR-μ-X-ray with high energy (37.5 keV) for excitation can detect trace amounts of heavy elements within individual microparticles, although lighter elements (such as Si) that are major components of the particle could not be detected in the spectrum. In addition to sharp K-line peaks of Cs that had been identified by gamma-ray spectroscopy, the following eight heavy elements were detected in all particles: Fe, Zn, Rb, Mo, Sn, Sb, Te, and Ba. Several trace heavy elements (e.g., Zr, Ag, Cd, and U) specific to certain particles were also found. All elements identified in the CsMPs by SR-μ-XRF analyses can be associated with materials in the FDNPP: the nuclear fuel, FPs, and components of the reactors. The SR-μ-XANES analysis of the chemical states of four metal elements (Fe, Zn, Mo, and Sn) indicated that these elements exist as cations in silicate glass with high oxidation numbers, as

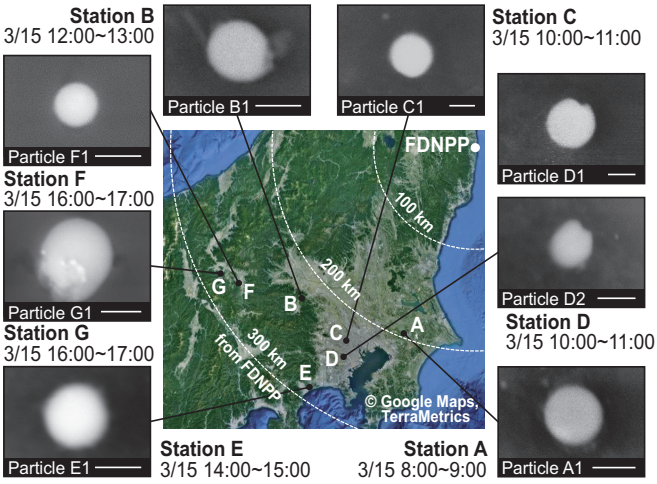

Fig. 1. Locations of monitoring stations A–G in the greater Kanto Region and eight radioactive particles isolated in this study. Collection times of the filters are shown for each station. The scale bars under the SEM images show a length of 1 μm.

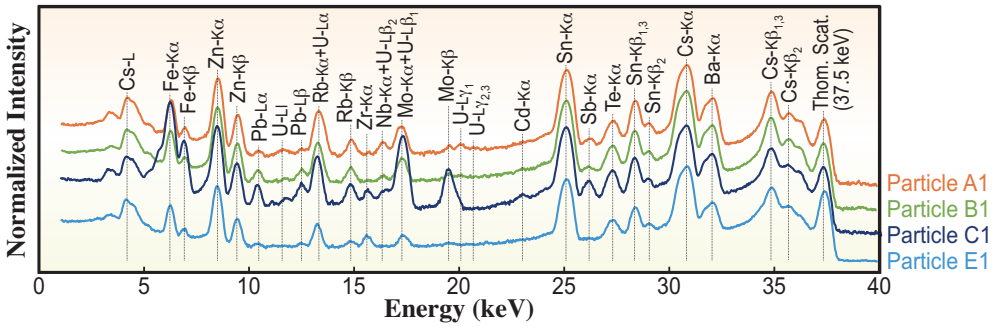

Fig. 2. SR-μ-XRF spectra of four representative CsMPs (A1, B1, C1, and E1). The intensities of each spectrum were normalized by the Thomson scattering peak and displayed on a logarithmic scale.

shown in Fig. 3. The SR-μ-XRD analysis of individual particles showed no diffraction peaks of a crystal structure for any of the particles, confirming that these particles have amorphous bodies.

Previously reported CsMPs can be categorized into two major types, type A and type B, based on the differences in physical/chemical characteristics [2]. All the analytical results on the eight CsMPs investigated in this study show that these particles are essentially the same as type A CsMPs. Type A CsMPs were first discovered from aerosols collected from the evening of 14 March to the morning of 15 March 2011 in Tsukuba, 170 km south-southwest of the FDNPP [1]. Since the discovery, a number of type A CsMPs have been identified from various environmental samples, mainly soils collected in Fukushima Prefecture. This is the first direct evidence demonstrating that type A CsMPs emitted from the FDNPP were widespread over the greater Kanto Region, farther than 250 km from the FDNPP, during the daytime of 15 March with a temporal resolution of one hour.

To estimate the emission time of type A CsMPs and identify their transport pathway(s) from the FDNPP to the greater Kanto Region, the trajectory analysis of air parcels that passed over the seven monitoring stations A–G was conducted. As a result, it was concluded that polluted air parcels containing type A CsMPs that passed over the greater Kanto Region on 15 March were emitted from the FDNPP between the evening of 14 March and the early morning of 15 March. Consistent with these estimated emission times, the pressure inside the reactor pressure vessel (RPV) in reactor No.2 decreased after the usage of a safety relief valve at around 19:03 JST on 14 March, followed by three sharp RPV pressure peaks at around 21:00 and 23:00 JST 14 March and 01:00 JST 15 March. At around 03:00 JST on 15 March, the pressure inside the primary containment vessel of reactor No.2 exceeded its designed value. In contrast, no incident was reported for reactor No.3 during the same period, except for a hydrogen explosion at 11:01 JST 14 March. It is therefore hypothesized that the incidents in reactor No. 2 were the most likely source of the type A CsMPs, rather than the hydrogen explosion in reactor No. 3.

Further investigation is necessary to estimate the environmental and health impacts of the CsMPs that travelled into the metropolitan area. Information regarding the widespread distribution of CsMPs can be useful toward calculating the inhalation dose of radionuclides during the early stage of the accident.

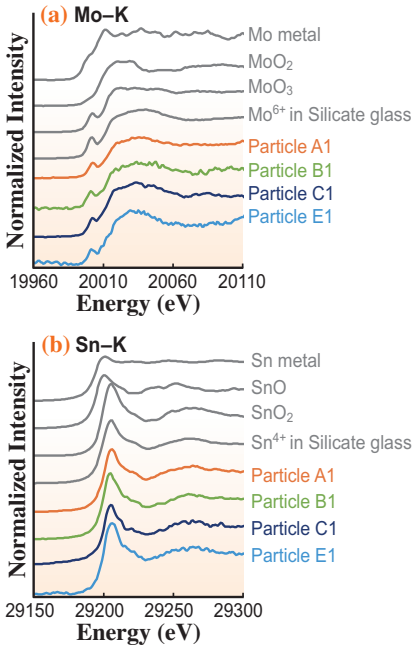

Fig. 3. SR-μ-XANES spectra of four representative CsMPs (A1, B1, C1, and E1) isolated from the SPM filter and reference materials. **(a)** Mo-*K* edge and **(b)** Sn-*K* edge.

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References

[1] K. Adachi *et al.*: Sci. Rep. **3** (2013) 2554.
[2] Y. Igarashi *et al.*: J. Environ. Radioact. **205-206** (2019) 101.
[3] H. Tsuruta *et al.*: Sci. Rep. **4** (2014) 6717.
[4] *Y. Abe, S. Onozaki, I. Nakai, K. Adachi, Y. Igarashi, Y. Oura, M. Ebihara, T. Miyasaka, H. Nakamura, K. Sueki, H. Tsuruta, Y. Moriguchi: Prog. Earth Planet. Sci. **8** (2021) 13.*