Investigation of chemical and physical properties of radioactive aerosols sampled from SPM tape filters using multiple synchrotron radiation X-ray analyses

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Large amounts of radioactive materials were released into the environment after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident caused by the Tohoku Earthquake and Tsunami on March 11th 2011. As one form of radioactive material emitted into the atmosphere, Cs-bearing radioactive spherical microparticle, known as Cs ball, has gathered attention in recent years. The Cs-ball was first found in aerosols collected at Tsukuba during March 14th and 15th 2011[1)] and identified as a water-insoluble glass material containing various heavy metals derived from fission products (FPs) of the nuclear fuel[2)]. While recent investigations suggest the widespread distribution of similar particulate radioactive materials in the environment such as soils sampled from the Fukushima prefecture[3)], the distribution of such radioactive particles in the Kanto region including the capital Tokyo just after the accident is still unclear. The present study thus focuses on suspended particulate matter (SPM) filter samples collected by operational air pollution monitoring stations[4)]. Radioactive aerosols were sampled from the filters collected at several stations during March 15th 2011 to investigate their chemical and physical properties in the nondestructive manner. The synchrotron radiation (SR)-X-ray microbeam of SPring-8 was used as an analytical probe for chemical composition analysis by X-ray fluorescence analysis (SR-μ-XRF), chemical state analysis by X-ray absorption near edge structure analysis (SR-μ-XANES) and crystal structure analysis by X-ray diffraction (SR-μ-XRD).

All particles sampled from the SPM filters in the present study have similar physical properties: a spherical shape and ~1 μm of diameter. Based on $^{134}Cs/^{137}Cs$ ratios (~1.0) of individual particles, it is expected these particles were emitted from the reactor No.2 or 3 of the FDNPP. These properties are similar to those of Cs-balls reported in previous studies[1,2)] except for their sizes (Cs-ball: ~2 μm in diameter). Various heavy elements (Rb, Mo, Sn, Sb, Te, Cs, Ba etc.) possibly derived from FPs were commonly detected from all these particles by the SR-μ-XRF. In addition, it was revealed that some particles contain trace amount of U. While we examined chemical states of four metal elements (Fe, Zn, Mo and Sn) contained in these particles by the SR-μ-XANES, all analytical results indicated that these elements exist as a glass state with high oxidation states in these particle. Results of the SR-μ-XRD also verified that these particles are amorphous materials. Because of these obvious similarities of chemical and physical properties between radioactive particles sampled from the SPM filters collected at the Kanto region and the Cs-balls, we thus concluded that particulate radioactive materials equated with the Cs-ball passed through the Kanto region on March 15th 2011. At the same time, our analytical results demonstrated the U possibly originated from the fuel certainly arrived to the Kanto region just after the accident.

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