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## Hourly atmospheric radionuclides after the Fukushima accident by analyzing filter-tapes of SPM monitoring sites

TSURUTA, Haruo<sup>1\*</sup>; OURA, Yasuji<sup>2</sup>; EBIHARA, Mitsuru<sup>2</sup>; OHARA, Toshimasa<sup>3</sup>; NAKAJIMA, Teruyuki<sup>1</sup>

<sup>1</sup>Atomsphere and Ocean Research Institute, the University of Tokyo, Japan, <sup>2</sup>Tokyo Metropolitan University, JAPAN, <sup>3</sup>National Institute for Environmental Studies, JAPAN

The current estimates for the internal radiation doses from inhalation by the Fukushima Daiichi Nuclear Power Station (FD1NPS) accident on March 11, 2011 have large uncertainty, because no observed data has been found of continuous monitoring of radioactive materials in the atmosphere in the Fukushima prefecture (FP) just after the accident. And most of the atmospheric transport models have simulated the surface deposition of radionuclides released by FD1NPS accident, to compare with many observed deposition datasets in eastern Japan. To retrieve the atmospheric transport of radioactive materials released from the FD1NPS, we collected the used filter tapes installed in Suspended Particulate Matter (SPM) monitors with beta-ray attenuation method operated by local governments in the air pollution monitoring network of eastern Japan. Then, we measured hourly Cs-134 and Cs-137 concentrations in SPM at 40 monitoring sites in the FP and Tokyo Metropolitan Area (TMA) located more than 170 km southwest of the FD1NPS, after more than one year. The period for measurements was during March 12-23, 2011, when atmospheric, aquatic, and terrestrial environments were seriously suffered in most of eastern Japan by a large amount of radioactive materials released from the FD1NPS. In this paper, a comprehensive study will be reported for the first time on a spatio-temporal variation of atmospheric Cs-137 concentrations in the FP and the TMA. Major results are as follows; (1) Nine major plumes with Cs-137 concentrations higher than 10 Bq m<sup>-3</sup> were found, of which 5 and 4 plumes were transported to the FP and TMA, respectively. (2) High radioactive materials from the FD1NPS were transported five times (March 12-13, 15, 18, 19, and 20-21) to the northern part of Hamadori located in the east coast of the FP, and which was little known until this study. Hence, the time-integrated atmospheric Cs-137 concentrations were highest among the FP and TMA during the period. (3) Two plumes were transported to Nakadori, located in the central part of the FP, on March 15, and March 20-21. The polluted air masses with high Cs-137 were observed in Nakadori for more than half a day from the evening of March 20 to the morning of March 21, and which was not recognized until now. (4) The radiation dose rate measured at some monitoring posts in Nakadori did not increase even when the plume (March 20) passed by. It was already too high to detect a new plume, due to the groundshine caused by the deposition of a large amount of radionuclides on the grounds by precipitation. (5) Two plumes transported to the TMA on March 16 and March 20 were newly found, in addition to the well-known two major plumes on March 15 and March 21, 2011. (6) A local area of relatively high Cs-137 deposition density in the TMA by precipitation on the morning of March 21, 2011, was consistent with an area where the time-integrated atmospheric Cs-137 concentrations were also high due to the transport of a plume on the morning of March 21, 2011. In the Fukushima prefecture, however, the correlation was not so clear. We thank to all the local governments in eastern Japan who kindly allowed us to measure radionuclides in the SPM collected on the used filter-tapes in the SPM monitors. We also thank Prof. S. Wakamatsu (Ehime University), and many other persons who cooperated to store the filter tapes. We sincerely acknowledge Y. Katsumura and M. Ishimoto in The University of Tokyo for supporting the measurements. Part of this study was financially supported by the Ministry of Education, Culture, Sports, Science and Technology, and the Ministry of Environment, Japan.

Keywords: Atmospheric Cs-137, FD1NPS, Spatio-temporal variation, SPM