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Temporal variation of post-accident atmospheric ^{137}Cs in an evacuated area of Fukushima Prefecture: Size-dependent behaviors of ^{137}Cs-bearing particles

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ABSTRACT

The concentrations of ^{137}Cs in the air, which were divided into coarse (>1.1 μm ϕ) and fine (<1.1 μm ϕ) fractions of particulate matter (PM), were measured from October 2012 to December 2014 in an area evacuated after the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. Total atmospheric ^{137}Cs concentrations showed a clear seasonal variation, with high concentrations during summer and autumn related to the dominant easterly wind blowing from the highly radioactivity contaminated area. This seasonal peak was dominated by ^{137}Cs in the coarse PM fraction. The ^{137}Cs specific activity (massic ^{137}Cs concentration) in the coarse PM was also found to increase significantly in summer, whereas that in the fine PM showed no variability during the year. These results show that coarse and fine ^{137}Cs-bearing PM have different origins and behaviors in the resuspension process. The seasonal variation in atmospheric ^{137}Cs concentration was well correlated with the mean ^{137}Cs surface contamination (deposition density) around the observation site weighted by the frequency of wind direction, indicating that the atmospheric ^{137}Cs concentration in the observation site was explained by the distribution of the ^{137}Cs surface contamination and the frequency of different wind directions. We introduced a resuspension factor corrected for wind direction, consisting of the ratio of the atmospheric ^{137}Cs concentration to the weighted mean ^{137}Cs surface contamination, which evaluated the intensity of resuspension better than the conventional resuspension factor. This ratio ranged from 5.7×10^{-11} to 8.6×10^{-10} m^{-1} and gradually decreased during the study period.

1. Introduction

During the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in March 2011, large amounts of radiocesium (^{134}Cs and ^{137}Cs) were released and deposited in the surrounding environment (MEXT, 2011; Mikami et al., 2015; Saito et al., 2015). The transport of this radiocesium in the atmosphere has been investigated through field observations (e.g., Kanai, 2012; Tsuruta et al., 2014; Igarashi et al., 2015) and model simulations (e.g., Chino et al., 2011; Katata et al., 2012a, 2012b; Srinivas et al., 2012). After direct releases from the power plant became negligible, resuspension from the ground was the remaining major source of atmospheric radiocesium. Resuspension is an important process for evaluating human exposures to chemical pollutants, because it leads to inhalation and redeposition on agricultural plants. Therefore, it had been widely investigated to understand its mechanisms (e.g., Nicholson, 1988; Sehmel, 1980).

Studies of resuspension of radionuclides were conducted after the 1986 Chernobyl nuclear power plant accident (e.g., Garger, 1994; Garland and Pomoroy, 1994). These studies showed that the temporal and spatial changes in radionuclide resuspension are influenced not only by meteorological conditions but also by anthropogenic activities such as traffic and agricultural operations (Akimoto, 2015; Garland and Pomeroy, 1994; Wagenpfeil et al., 1999; Yamaguchi et al., 2012), and by natural events such as

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wildfire (Bondarkov et al., 2011). Studies of the Chernobyl accident aftermath also enabled researchers to describe long-term trends of resuspension using an exponential (Garland et al., 1992; Rosner and Winkler, 2001) or power function (Hötzl et al., 1992; Garger et al., 1997; Hatano and Hatano, 1997, 2003).

These research findings are useful for evaluating the radiocesium resuspension in the Fukushima area. However, contamination situations by the accidents (Steinhauser et al., 2014) and the land surface environments (e.g., climate, vegetation type, and land use), which are related to the characteristics and behavior of resuspended materials, are different between Chernobyl and Fukushima. For accurate predictions of resuspended atmospheric ^{137}Cs, it is necessary to clarify the controlling factors of resuspension in the Fukushima area.

We investigated FDNPP-derived ^{137}Cs in airborne particulate matter (PM) in Namie Town, an area highly contaminated by the FDNPP accident, to clarify the controlling factors on seasonal changes in resuspended atmospheric ^{137}Cs.

2. Samples and methods

Atmospheric PM was sampled at a site in the western part of Namie Town (Fig. 1), in the evacuation zone, from October 2012 to December 2014. The sampling site is located at the western edge of the highly contaminated zone. The initial surface contamination (deposition density) of ^{137}Cs (Bq m^{-2}) around the sampling site (10 km radius) was estimated to be 0.1–7 MBq m^{-2}, based on the third airborne survey conducted by the Ministry of Education, Culture, Sports, Science and Technology (MEXT, 2011). The sampling site is located in the valley of the Ukedo River draining from the Abukuma Mountains to the Pacific Ocean. Elevation is about 416 m based on the topographic map (Geospatial Information Authority of Japan). Land use around the sampling site (10 km radius) mainly consists of forest (81%) and farmland (17%) based on National Land Numerical Information (Ministry of Land, Infrastructure, Transport and Tourism, Japan) database.

The PM samples were collected for 1-month periods at a position 1.8 m above the ground using a high-volume aerosol sampler coupled with a single-stage impactor (HV-1000F, Tokyo Dylec, Japan). Particles coarser and finer than 1.1 μm median aerodynamic diameter were collected on separate quartz fiber filters at an air flow rate of 566 L min^{-1}. Flow rate was automatically controlled at a constant value. Lowering of flow rate by filter clogging did not occur even in a high PM concentration period, suggesting that the influence of long-term sampling on collection efficiency was negligible. Sample collection was unsuccessful in March and April 2014 and in part of February 2014.

The filter samples were dried in a desiccator, then weighed and pressed into disks 3.5 cm in diameter for radioactivity measurement. Weights of PM were obtained from the difference between the weights of filters before and after sampling.

The activity concentrations of ^{134}Cs ($T_{1/2} = 2.06$ y) and ^{137}Cs ($T_{1/2} = 30.17$ y) were determined by low-energy photon spectrometers (EGM3800-20, Eurisys Mesures, France; EGMP 80-20-R, Canberra Industries, USA). The counting efficiency of the detector was calibrated with several radioactive standards (IAEA-SOIL6, IAEA-375, IAEA-315, International Atomic Energy Agency, Austria; MP650, AEA Technology, UK; MX033U8PP, Japan Radioisotope Association, Japan). Wind speed and direction at a position 2 m above the ground were recorded every 10 min by a meteorological instrument (Hobo Weather Station, Onset Computing Corp.) at the observation site.

The relationship between atmospheric ^{137}Cs concentration, wind direction and the geographic distribution of radiocesium surface contamination was analyzed by using QGIS 2.2.0 software and geographic data from the Fundamental Geospatial Data (Geospatial Information Authority of Japan) and National Land Numerical Information (Ministry of Land, Infrastructure, Transport and Tourism, Japan) databases. The ^{137}Cs deposition density data were based on the third airborne survey of 2 July 2011 (MEXT, 2011). Data

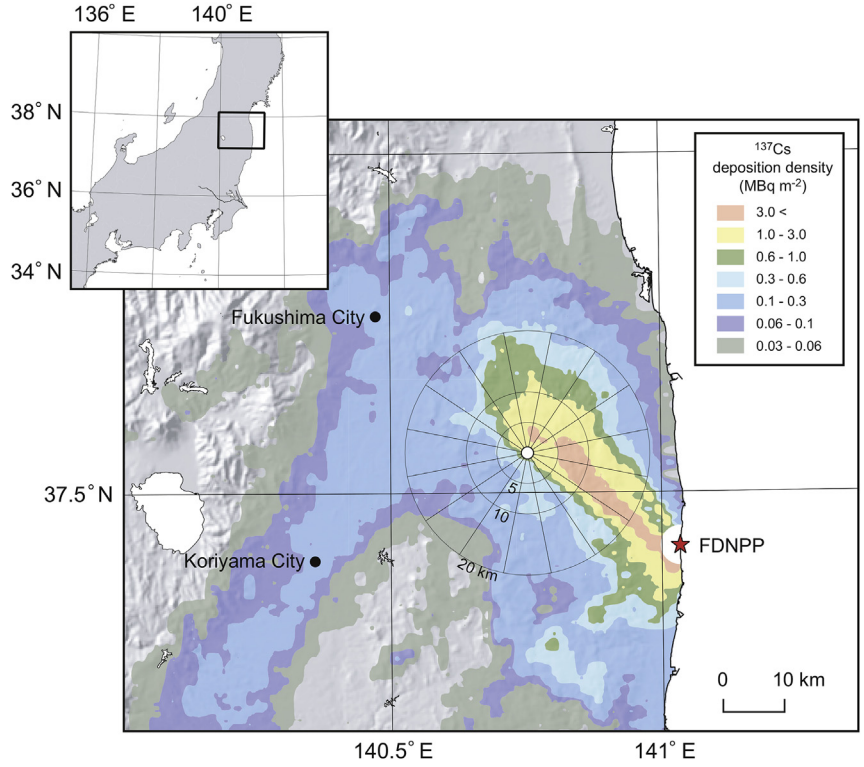

Fig. 1. Map and location of the sampling point in Namie Town, Japan. Distribution of the initial ^{137}Cs deposition density from MEXT (2011, 2012).