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Regression model analysis of the decreasing trend of cesium-137 concentration in the atmosphere since the Fukushima accident

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ABSTRACT

The decreasing trend of the atmospheric ^{137}Cs concentration in two cities in Fukushima prefecture was analyzed by a regression model to clarify the relation between the parameter of the decrease in the model and the trend and to compare the trend with that after the Chernobyl accident. The ^{137}Cs particle concentration measurements were conducted in urban Fukushima and rural Date sites from September 2012 to June 2015. The ^{137}Cs particle concentrations were separated in two groups: particles of more than 1.1 μm aerodynamic diameters (coarse particles) and particles with aerodynamic diameter lower than 1.1 μm (fine particles). The averages of the measured concentrations were 0.1 mBq m^{-3} in Fukushima and Date sites. The measured concentrations were applied in the regression model which decomposed them into two components: trend and seasonal variation. The trend concentration included the parameters for the constant and the exponential decrease. The parameter for the constant was slightly different between the Fukushima and Date sites. The parameter for the exponential decrease was similar for all the cases, and much higher than the value of the physical radioactive decay except for the concentration in the fine particles at the Date site. The annual decreasing rates of the ^{137}Cs concentration evaluated by the trend concentration ranged from 44 to 53% y^{-1} with average and standard deviation of 49 ± 8% y^{-1} for all the cases in 2013. In the other years, the decreasing rates also varied slightly for all cases. These indicated that the decreasing trend of the ^{137}Cs concentration was nearly unchanged for the location and ground contamination level in the three years after the accident. The ^{137}Cs activity per aerosol particle mass also decreased with the same trend as the ^{137}Cs concentration in the atmosphere. The results indicated that the decreasing trend of the atmospheric ^{137}Cs concentration was related with the reduction of the ^{137}Cs concentration in resuspended particles.

1. Introduction

The accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) on March 11, 2011 resulted in releases of large amounts of radionuclides to the atmosphere. The released radionuclides were deposited onto the ground causing significant contamination. The Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) and the U.S. Department of Energy (U.S. DOE) conducted airborne monitoring which showed that the area northwest from the FDNPP was highly contaminated (MEXT and U.S. DOE, 2011). Among the major released radionuclides, ^{137}Cs remains in the environment for a long time because of its relatively long 30 y radioactive half life (Lederer and Shirley, 1978). After been deposited onto the ground, it was considered to be resuspended into the atmosphere in a cycle of deposition and resuspension that affects the ^{137}Cs concentration trend expected from radioactive decay. The trend of the atmospheric ^{137}Cs concentration is related to public health by long-term internal exposure due to inhalation and it is desirable to predict the concentration trend due to resuspension from the contaminated ground. The decreasing trend of the atmospheric ^{137}Cs concentration in Fukushima city has been reported to be larger than that for the physical radioactive decay (Tsukada et al., 2012).

In the case of the Chernobyl accident, the decreasing trend and fluctuation of the ^{137}Cs concentration was analyzed by fitting the concentration to a model formula (Garger et al., 1994; Hatano et al., 1998; Ichige et al., 2015). All of these groups derived a decreasing relationship for the trend concentration which excluded fluctuation of the observed concentration and the behavior of the fluctuation by using long-term data. Because the seasonal variation and other

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sub-annual fluctuations are reduced by the smoothing, the trend concentration is more suitable for analyzing the decreasing trend.

In trend analyses, a regression model has been applied for particulate SO_4^{2-}, NO_3^- and NH_4^+, SO_2 and HNO_3. Sirois (1993, 1997) then Sirois and Barrie (Sirois and Barrie, 1999) applied a regression model including the terms of a polynomial trend and a seasonal cycle on the logarithmic concentration of gas and aerosols to reveal the features of the component in a time series. Seto et al. (2002, 2004) used a regression model composed of simple linear and seasonal terms for analyzing the trend of the lognormal ionic concentration in precipitation. Okuda et al. (2008) used a regression model for the log-normal metal concentration in aerosols to explain similar trends. For radionuclides, regression models are useful for understanding characteristics and variations of the trend because the concentration includes the decrease due to radioactive decay and the variations due to resuspension.

To determine the decreasing trend of the atmospheric ^{137}Cs concentration since the FDNPP accident, the ^{137}Cs concentration was measured in urban Fukushima and rural Date sites and applied in the regression model which decomposed them into the components of the trend and seasonal variation. The collected particles were separated into two groups using the 1.1 μm aerodynamic diameter as the separation point based on the dust particles dominant in the coarse mode (Seinfeld and Pandis, 2006) for focusing on the resuspended ^{137}Cs particles. The parameters related to the decreasing trend were compared to those of the Chernobyl accident reported in two previous studies (Hatano et al., 1998; Ichige et al., 2015). From the comparison, the characteristics of the decreasing trend in the atmospheric ^{137}Cs concentration were investigated considering similarities and differences between the two sampling locations, meteorological conditions and the ground contamination levels.

2. Material and methods

The two studied sites were located in Fukushima City and Date City (Fig. 1). The Fukushima site is in an urban area 62 km from the FDNPP and the Date site is in a rural area 55 km from the FDNPP. Aerosol samplings were conducted on the roof top of a building at 7 m and 11 m above ground surface in Fukushima and Date sites, respectively, by using high-volume air samplers (HV-1000R, SHIBATA) at a flow rate of 566 L min^{-1}. Collection was done using polytetrafluoroethylene filters with an Andersen air sampler (AH-600, Tokyo Dylec) which separated the airborne particles for the aerodynamic diameter of 1.1 μm at the 50% cut-off collection efficiency. The sampling was conducted at about half month intervals from September 2012 to June 2015. The detailed sampling intervals are shown in the supplementary file.

For analyzing the radioactivity of radiocesium in the collected filter samples, half of a filter was cut into small pieces that were packed into a 100 mL U8 plastic bottle. The radioactivity of ^{137}Cs in the samples was measured from the γ-ray spectrum by a Ge detector for 10,800–86,400 s. For quantifying the radioactivity, the γ-ray spectrum at 661 keV was used for ^{137}Cs. For low concentration samples, the measuring time was extended to keep the counting error within 10%. The concentration data is shown in the supplementary file.

3. Regression model

The regression model for the atmospheric radiocesium concentrations is based on the relation derived by Hatano and Hatano (1997, 2003). They showed the concentration decreases as

$$C_{trend} = Ae^{-\Lambda t}t^{-4/3} \quad (1)$$

where C_{trend} is the trend component of the concentration originally defined as the mean concentration (Garger et al., 1994; Hatano and Hatano, 1997) which is free of the fluctuating concentration including seasonal variation, t is days from the accident (in their case Chernobyl), A and Λ are parameters for the constant related to an initial state and exponential reduction, respectively. They also reported that the fluctuating concentration was proportional to the one-third power of the averaging period. In this study, the relation in Equation (1) was applied for the ^{137}Cs concentration in the case

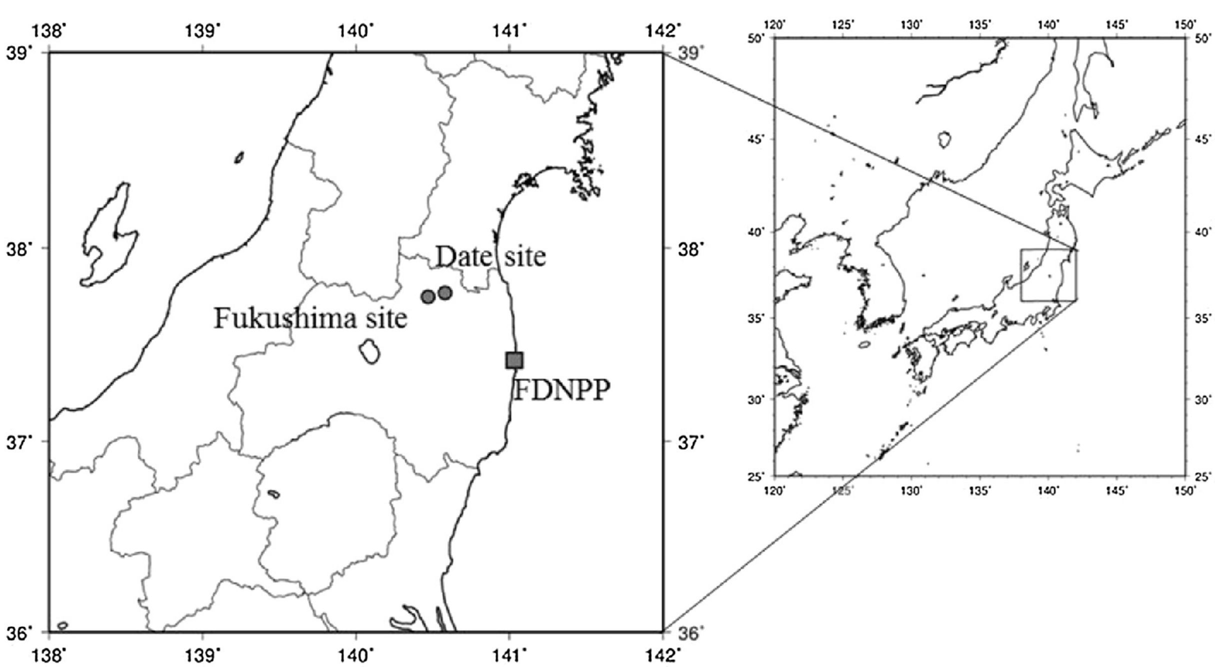

Fig. 1. The locations of the sampling sites and FDNPP.