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Depth profiles of radioactive cesium in soil using a scraper plate over a wide area surrounding the Fukushima Dai-ichi Nuclear Power Plant, Japan

Norihiro Matsuda [a, *], Satoshi Mikami [a], Susumu Shimoura [b], Junko Takahashi [c], Masakazu Nakano [a], Kiyotaka Shimada [a], Kiichiro Uno [a], Shigetomo Hagiwara [a], Kimiaki Saito [a]

[a] Japan Atomic Energy Agency, 2-2-2 Uchisaiwai-cho, Chiyoda, Tokyo 100-8577, Japan
[b] Center for Nuclear Study, University of Tokyo, 7-3-1 Hongo, Bunkyo, Tokyo 113-0033, Japan
[c] Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan

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ABSTRACT

During the Fukushima Dai-ichi Nuclear Power Plant (NPP) accident, radioactive cesium was released in the environment and deposited on the soils. Depth profiles of radioactive cesium in contaminated soils provide useful information not only for radiation protection and decontamination operations but also for geoscience and radioecology studies. Soil samples were collected using a scraper plate three times between December 2011 and December 2012 at 84 or 85 locations within a 100-km radius of the Fukushima Dai-ichi NPP. In most of the obtained radioactive cesium depth profiles, it was possible to fit the concentration to a function of mass depth as either an exponential or hyperbolic secant function. By using those functions, following three parameters were estimated: (i) relaxation mass depth β (g cm^{-2}), (ii) effective relaxation mass depth β_{eff} (g cm^{-2}), which is defined for a hyperbolic secant function as the relaxation mass depth of an equivalent exponential function giving the same air kerma rate at 1 m above the ground as the inventory, and (iii) 1/10 depth $L_{1/10}$ (cm), at which the soil contains 90% of the inventory. The average β value (wet weight) including ones by hyperbolic secant function in December 2012, was 1.29 times higher than that in December 2011. In fact, it was observed that depth profiles at some study sites deviated from the typical exponential distributions over time. These results indicate the gradual downward migration of radioactive cesium in the soils. The $L_{1/10}$ values in December 2012 were summarized and presented on a map surrounding the Fukushima Dai-ichi NPP, and the average value of $L_{1/10}$ was 3.01 cm ($n = 82$) at this time. It was found that radioactive cesium remained within 5 cm of the ground surface at most study sites (71 sites). The sech function can also be used to estimate the downward migration rate V (kg m^{-2} y^{-1}). The V values in December 2012 ($n = 25$) were in good agreement with those found by a realistic approach using a diffusion and migration model. Almost all values ranged between 1.7 and 9.6 kg m^{-2} y^{-1} in this study.

1. Introduction

Many people living in northeastern Japan experienced the earthquake that occurred off the Pacific coast of Tohoku on March 11, 2011, and those living around the Fukushima Dai-ichi Nuclear Power Plant (NPP) were negatively affected by the subsequent accident at the plant. A large amount of radioactive nuclides, including approximately 1.5 × 10^{16} Bq of ^{137}Cs, was released in the environment because of this accident (NERHGJ, 2011). Currently, the Fukushima-derived radioactive cesium is the dominant radionuclide deposited on soils, and specific vertical distributions (depth profiles) have been observed in soils. Similarly, in those days after the Chernobyl accident, radioactive cesium, especially the ^{137}Cs that remains after the decay of ^{134}Cs (which has a physical half-life of 2.062 y), was of significant radiological importance (IAEA, 2006).

The Japanese Government acted quickly to implement continuing decontamination work to reduce the exposure dose to

* Corresponding author. Fukushima Environmental Safety Center, Japan Atomic Energy Agency, 2-2-2 Uchisaiwai-cho, Chiyoda, Tokyo 100-8577, Japan. Tel.: +81 3 3592 2703.
E-mail address: matsuda.norihiro@jaea.go.jp (N. Matsuda).

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local residents following the accident (NERH, 2011). On the basis of the Comprehensive Monitoring Plan drawn up at the Monitoring Coordination Meeting (DMCM, 2011) and the Enforced Plan on Environmental Monitoring from the Headquarters against Nuclear Disaster (HND, 2011), a project for large-scale environmental monitoring and mapping was initiated in June 2011 (Saito, 2014) with funding from the Ministry of Education, Culture, Sports, Science, and Technology (MEXT). As part of this project, a detailed survey was planned and conducted to clarify the depth profiles of radioactive cesium in soils using a scraper plate (Loughran et al., 2002) within a 100-km radius of the Fukushima Dai-ichi NPP.

It is important to understand the depth profiles of radioactive cesium not only for continuing radiation protection and environmental recovery but also for geoscience and radioecology studies. The depth profiles, especially their temporal changes, provide basic information about the vertical migration of radioactive cesium within the ground. According to previous studies, including those in the Chernobyl accident, the initial distribution of radioactive cesium in surface soils and their post-depositional redistribution within the soils are influenced by various physical, physicochemical, and biological processes (e.g., He and Walling, 1997; IAEA, 2006). Therefore, depth profiles represent fundamental data required for understanding the movement of radioactive cesium in the ground. Furthermore, this information can be used in evaluating the risks of downward migration in groundwater and root uptake by plants, which is an important terrestrial pathway in the food chain. In addition, varied depth distributions complicate the estimation of exposure dose even if the total inventory is the same, because upper soil layers can act as a shield against gamma rays (Anderson and Roed, 1994; ICRU, 1994). Knowledge of the current soil depth profile is important, as it will affect the costs involved in removing soil during decontamination as well as the amount of waste produced.

It is expected that the depth profiles of radioactive cesium will show an exponential distribution with depth in many cases (Walling and He, 1999). In some cases, temporal changes to the depth profiles have been observed after several years, in which the maximum concentration progressively moved from the surface layer to deeper ground layers (e.g., Bunzl et al., 1995; He and Walling, 1997; Hillmann et al., 1996; IAEA, 2006). To approximate the depth distribution, some fitting functions have been proposed and developed, such as the Gaussian (Genuchten and van Cleary, 1979; Konshin, 1992), double exponential (Zombori et al., 1992), Lorentz (Hillmann et al., 1996), and polynomial (van Siclen, 2011) functions. These functions are not satisfying in all cases, however, because the Gaussian function tends to underestimate the concentration of radioactive cesium in the deeper layers (Konshin, 1992), and the other functions incorporate independent distribution parameters and do not have common parameter with exponential functions, despite the expectation that the depth profiles will change from the initial distribution with time.

This study aims to determine the depth profiles of radioactive cesium in soils across a wide area around the Fukushima Dai-ichi NPP and to investigate the characteristics of these profiles, such as their temporal variation. For the depth profiles of radionuclides that show a peak at a certain depth, a new fitting function based on a hyperbolic secant function was applied as an alternative to the typical exponential function.

2. Materials and methods

2.1. Study sites

The semicircular region contained within a 100-km radius around the Fukushima Dai-ichi NPP site was divided into 5 km × 5 km square grids, each with a unique ID number, defined at the beginning of the project for large-scale environmental monitoring and mapping. Total of 84 or 85 square grids distributed evenly across this region were chosen for sampling, and the total area of these grids corresponds to approximately 1/5th of the area of square grids across the entire monitoring and mapping project in the semicircle. ID numbers are provided with reference to the Fukushima Dai-ichi NPP. For example, a grid with ID: 075N025 is located 75 km north and 25 km east of the NPP. One appropriate sampling location was selected within each grid unit, targeting locations with wide and flat landforms located at least 5 m away from buildings or trees. As the deposition of radioactive nuclides on the ground by rain or snow may be influenced by surrounding objects, sampling locations near buildings or trees may show uneven or anomalous concentrations. Sampling at locations with rocks and stones was also avoided, although the presence of a few pebbles was permitted. Furthermore, locations with visible disturbance by rodent activity were avoided due to potentially uneven mixing within the soil.

2.2. Soil sampling and radioactive cesium analysis

Soil sampling at each site was performed three times, once during each of the following periods: December 12–22, 2011 (81 sites) and April 17–19 (3 sites), 2012; August 21 to September 26 (85 sites), 2012; and November 26 to December 26, 2012 (85 sites). Three sites within the “Evacuation Zone” within 20 km of the Fukushima Dai-ichi NPP, were inaccessible during the main part of the first collection period, as the zone was out of bounds to the public at that time; so soil samplings at these sites were first conducted during April 17–19, 2012. All sampling dates were selected to consider the local rainy season from the beginning of June to mid-July and the typhoon season around September. The sampling locations in some of the sampling grids or some of the sampling grids had to be changed between collection periods due to disruptions by decontamination work.

The soil layer samples were collected using a scraper plate (Loughran et al., 2002) as per the standards of the International Atomic Energy Agency (IAEA) (Burkart, 2002). The soil sampling tools included a plate to scrape the soil and a stainless-steel guide frame to provide a fixed location and scraping area. The plate is equipped with numerous holes to allow attachment to a fixed bar, permitting a certain distance to be maintained between the guide frame and the edge of the plate. The inner dimensions of the guide frame are 150 mm × 300 mm; therefore, soils with a volume of 225 or 450 cm^3 were removed when sampling at 5- or 10-mm depth intervals, respectively. Vegetation cover over the scraping area was removed at the surface level, because it was not growing season of vegetation in which the radioactive nuclides were released from the accident. Gravel and pebbles on the ground surface were also removed. The soil was sampled at 5-mm intervals up to a depth of 20 mm, at 10-mm intervals between 20 and 50 mm, and at 30-mm intervals between a depth of 50 and 80 mm. The soil collected in each layer, including stones and pebbles, was placed in a resealable plastic sampling bag and labeled with the sampling date, ID number of the study site, and the depth. After the collection of soil was completed in a particular layer, the scraper plate was brushed and cleaned to avoid any cross-contamination between layers. This sampling process was repeated until the eighth layer of soil was collected.

The collected soil was carefully homogenized by hand in the plastic bag, and the wet weight of soil was measured using a portable scale. A constant volume of soil (excluding stones and pebbles) from the plastic bag was placed in a labeled φ56 mm (inner diameter: φ50 mm) × 68 mm plastic container. Then, the

plastic container was capped and sealed using a resealable plastic bag for contamination control.

Quantitative analyses of radioactive cesium in the samples in these plastic containers were conducted at the Institute for Environmental Sciences, the Japan Chemical Analysis Center, and the University of Tokyo using high-purity germanium detectors (GC3020-7500SL, Canberra, Meriden, USA, and other similar models). All concentration data of wet weight (Bq kg^{-1}) at each depth (cm) obtained in the present study will be available online from Japan Atomic Energy Agency (JAEA) in the near future (JAEA, 2014).

2.3. Fitting and estimation of distribution parameters for the depth profile

In the International Commission on Radiation Units and Measurements' report (ICRU, 1994), an exponential distribution function describing the concentration of radioactive cesium with a depth of mass per unit area, $A_m(\zeta)$, is defined as follows:

$$A_m(\zeta) = A_{m,0}\exp(-\zeta/\beta), \tag{1}$$

where $A_{m,0}$ is the concentration (wet weight) at the ground surface of the soil (Bq kg^{-1}) and β is the relaxation mass depth (g cm^{-2}), at which the concentration of radioactive cesium reduces to $1/e$ of the concentration at the ground surface. Mass depth ζ (g cm^{-2}) is given as follows:

$$\zeta = \int_0^z \rho(z')dz', \tag{2}$$

where z is the actual depth from the ground surface (cm) and ρ (g cm^{-3}) is the soil density. In these calculations, the mass depth ζ (g cm^{-2}) calculated from Eq. (2) is used, as opposed to the depth z (cm), because the shielding effect of the upper soil layer against gamma rays from radioactive cesium is related to ζ. The soil density ρ (g cm^{-3}) was determined based on the mass (wet weight) and the volume of each layer in the soil samples. The relaxation mass depth β as a distribution parameter was calculated from the obtained depth profile using a least-squares method. Furthermore, the inventory, $A_{inv.}$ (kBq m^{-2}), based on Eq. (1) is expressed as follows:

$$A_{inv.} = \{\beta \times (10,000/1000)\} \times A_{m,0}/1000. \tag{3}$$

To translate these calculations into useful information about the current depth profiles for decontamination work and other applications, an easily understood index is required. In this study, 1/10 depth $L_{1/10}$ (cm) at which the soil contains 90% of the inventory of radioactive cesium was defined based on inverse relaxation depth, $1/\alpha$ (cm), also denoted by β/ρ, because the concentration decreases exponentially with mass depth from the soil surface in most situations. In Eq. (1), ζ (g cm^{-2}) and β (g cm^{-2}) may be replaced by depth z (cm) and $1/\alpha$ (cm), respectively, assuming a constant soil density. The $L_{1/10}$ based on Eq. (1) can be expressed as follows:

$$L_{1/10} = (1/\alpha)/\log_{10}(\exp(1)). \tag{4}$$

In some cases, the maximal concentration of radioactive cesium in the soil shifted progressively to deeper layers with time, and the simple exponential function with only two parameters, i.e., $A_{m,0}$ and β, could not be applied to these cases. Therefore, to apply an approximate fitting to these depth profiles, an analytical function was defined on the basis of the hyperbolic secant (sech) and hyperbolic cosine (cosh) functions as follows:

$$A_m(\zeta) = (A_{m,0}/2)\cosh(\zeta_0/\beta)\mathrm{sech}(-(\zeta-\zeta_0)/\beta), \tag{5}$$

$$*\mathrm{sech}(x) = 1/\cosh(x) = 2/(\exp(x)+\exp(-x)),$$

where $A_{m,0}$ is the concentration at the ground surface (Bq kg^{-1}), ζ_0 is the mass depth from the ground surface where the concentration is at its maximum (g cm^{-2}), and β is the relaxation mass depth (g cm^{-2}). Furthermore, the maximum concentration $A_{m,\zeta 0}$ (Bq kg^{-1}) can be expressed as follows:

$$A_{m,\zeta 0} = (A_{m,0}/2)\cosh(\zeta_0/\beta). \tag{6}$$

This analytical function is considered optimal because it possesses the following properties: (i) it can simulate an asymmetric peak in a depth profile, (ii) it approaches an exponential function as the depth increases, and (iii) it has common parameters with the simple exponential distribution of Eq. (1). To determine $A_{m,0}$, β, and ζ_0 for an obtained depth profile, an iterative approach was applied using the solver function in Excel (Microsoft Office). For deeper layers, Eq. (5) can be approximated as follows:

$$A_m(\zeta) = A_{m,0}\cosh(\zeta_0/\beta)\exp\{-(\zeta-\zeta_0)/\beta\}, \tag{7}$$

in which the β in this equation conforms to the β in Eq. (1). Similarly, the inventory, $A_{inv.}$ (kBq m^{-2}), based on Eq. (5) is expressed as follows:

$$A_{inv.} = \{\beta \times (10,000/1000)\} \times A_{m,\zeta 0} \times [(\pi/2) - \arctan\{\sinh(\zeta_0/\beta)\}]/1000. \tag{8}$$

In cases where the exponential function (Eq. (1)) is applicable, the parameter β determines the relationship between the inventory and the air dose rate (Miller et al., 1990). However, in cases conforming to the sech function (Eq. (5)), the relationship is not determined by β alone, despite β being a common parameter in both equations. To estimate and analyze the air dose rate for a depth profile approximated by Eq. (5) using the simple relationship based on Eq. (1), the effective parameter, β_{eff}, was defined. The β_{eff} was determined as β in the exponential function to give the same air kerma rate at 1 m above the ground with the hyperbolic secant function and the same inventory. The air kerma rate was calculated based on the air kerma per unit source intensity with source depth (Saito and Jacob, 1995).

In Eq. (5), ζ (g cm^{-2}), ζ_0 (g cm^{-2}), and β (g cm^{-2}) can be replaced by depth z (cm), depth z_0 where the concentration is at a maximum (cm), and $1/\alpha$ (cm), respectively. The $L_{1/10}$ based on Eq. (5) is expressed as follows:

$$L_{1/10} = (1/\alpha)\times\sinh^{-1}\Big(\tan\Big(\big(9\times(\pi/2) + \tan^{-1}(\sinh(-z_0/(1/\alpha)))\big)\Big/10\Big)\Big) + z_0. \tag{9}$$

In contrast with this analytical function, a more realistic approach involving a diffusion and migration model (DM model) has been proposed and discussed (e.g., Pegoyev and Fridman, 1978; Reynolds et al., 1982; He and Walling, 1997; Porto et al., 2001, 2003; Krstic et al., 2004). For an undisturbed soil, the post-depositional redistribution of radioactive cesium within the soil over time is assumed to be governed by various physical, physicochemical, and biological processes operating in the soil system itself (He and Walling, 1997). This redistribution can be represented by simple diffusion and convection equations, characterized by an effective diffusion coefficient D (kg^2 m^{-4} y^{-1}) and a downward migration rate V (kg m^{-2} y^{-1}), respectively. A solution from Szerbin et al.

(1999), usually known as the Fokker–Planck equation (Risken, 1984), is

$$A_m(\zeta,t) = A_{inv.}\exp(-\lambda t)\times 1/2[\pi D t]^{1/2}\times\exp\left\{-[\zeta - Vt]^2\Big/4Dt\right\}, \quad (10)$$

where ζ is the mass depth (kg m^{-2}), t is the time since the deposition of radionuclides from the Fukushima Dai-ichi NPP accident (y), $A_m(\zeta,t)$ is the concentration of radioactive cesium with mass depth and at the time of t (Bq kg^{-1}), $A_{inv.}$ is the initial surface concentration (equal to the inventory) (Bq m^{-2}), and λ is the decay constant of radioactive cesium. In contrast, the downward migration rate V based on the ζ_0 values by the sech function in Eq. (5) will be approximated using the following equation (cf. Walling et al., 2002) modified for the Fukushima accident:

$$V \fallingdotseq \zeta_0/t. \quad (11)$$

3. Results and discussion

3.1. Typical depth profiles of ^{134}Cs and ^{137}Cs with an exponential distribution

The typical depth profiles of radioactive cesium, such as those in Fig. 1a and b, can be fitted with an exponential function, indicating that the cesium concentration exponentially decreases with mass depth. For example, from the depth distributions from the first collection period at ID: 000N000, the inventory of ^{134}Cs and ^{137}Cs within a depth of 80 mm was 346 and 530 kBq m^{-2}, respectively, on the measurement date. The ^{134}Cs/^{137}Cs ratio of these values was 0.65, which is in agreement with the theoretical value (0.70) predicted according to physical decay using an initial ratio of 1.0 on March 11, 2011 (MEXT, 2011). In addition, the concentrations of ^{134}Cs and ^{137}Cs at the ground surface, $A_{m,0}$, were 27.1 and 40.7 kBq kg^{-1}, respectively, and the relaxation depth β for ^{134}Cs and ^{137}Cs were 1.36 and 1.38 g cm^{-2}, respectively. The values of β for ^{134}Cs and ^{137}Cs were not significantly different, which is expected due to their identical chemical properties. Thus, the depth profiles were similar for ^{134}Cs and ^{137}Cs. There was a good agreement between the inventory of ^{137}Cs provided by measurement up to 80 mm depth (530 kBq m^{-2}) and that calculated using Eq. (3) (560 kBq m^{-2}).

Typical exponential distributions of radioactive cesium were observed at 72 sites in the first collection period. However, this decreased to 62 sites and 53 sites in the second and third collection periods, respectively. For example, the depth profiles at ID: 015S045 showed an exponential distribution in the first and second collection periods but this has changed by the third collection period (Fig. 1). This indicates that the downward migration of radioactive cesium has occurred in these soils or, in some cases, soil mixing may have occurred due to soil disturbance, cultivation, or decontamination work.

3.2. Temporal changes in the depth profiles of ^{134}Cs and ^{137}Cs

If the exponential function in Eq. (1) is applied to a depth profile with a peak at a certain depths, such as Fig. 1c, then the fitted line will be different from the actual depth distribution, and the value of β, corresponding to the slope of the fitted line, will tend to be an overestimation (gently) compared with the actual exponential decrease in the deeper layers. Therefore, to approximate these temporal changes in the depth profiles of radioactive cesium, the sech function (Eq. (5)) was applied. For example, in the depth distribution from the third collection period at ID: 005S025, the inventory of ^{134}Cs and ^{137}Cs within a depth of 80 mm was 38 and 67 kBq m^{-2}, respectively, on the measurement date. The ^{134}Cs/^{137}Cs ratio of these values was 0.57, which is in agreement with the theoretical value (0.58) predicted from physical decay using an initial ratio of 1.0 on March 11, 2011. In addition, the ratios (^{134}Cs/^{137}Cs) of concentrations at the ground surface $A_{m,0}$ and the maximal concentrations in the ground A_{m,ζ_0} were 0.55 (=0.6 kBq kg^{-1}/1.1 kBq kg^{-1}) and 0.56 (=1.0 kBq kg^{-1}/1.8 kBq kg^{-1}), respectively. In addition, the values of β and ζ_0 for ^{134}Cs and ^{137}Cs were 1.52 and 1.56 g cm^{-2}, and 1.70 and 1.69 g cm^{-2}, respectively. The values of all the above parameters were not significantly different between ^{134}Cs and ^{137}Cs, despite the depth profiles changing over time; so we decided to focus solely on ^{137}Cs in this study. The calculated value of β for ^{137}Cs was considerably-improved from 3.25 g cm^{-2} using the exponential function to 1.56 g cm^{-2} using the sech function. Further measured inventory of ^{137}Cs up to 80 mm depth (67 kBq m^{-2}) agreed well with the value calculated (68 kBq m^{-2}) using Eq. (8).

3.3. Disturbed depth profile

In some cases, a disturbed depth profile of radioactive cesium was observed, despite the profiles obtained on other occasions showing a normal exponential profile. For example, profiles from the three different collection periods at the same study site, ID: 025S015, are shown in Fig. 2. In this, an almost homogeneous and unnatural depth profile was observed during the second collection period (Fig. 2b), however, the inventory up to a depth of 80 mm remained relatively constant between collection periods. The sampling location, including the scraping area, may have been disturbed. The activities of earthworms or small animals, such as

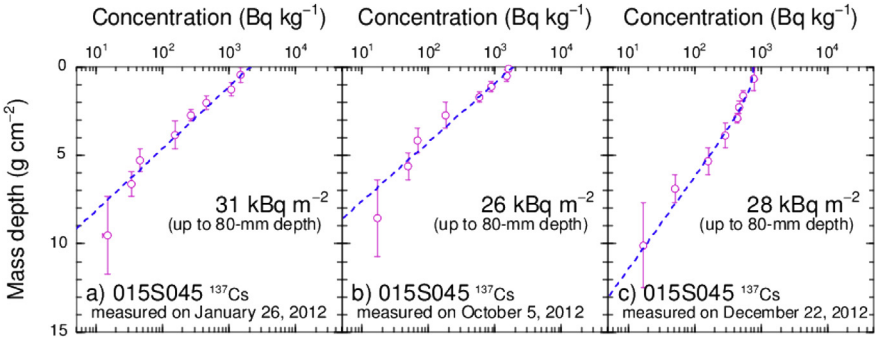

Fig. 1. Depth profiles for ^{137}Cs concentration with mass depth (wet weight) and approximate curves of best fit sampled in: a) December 2011, b) August 2012, and c) November 2012 at ID 015S045.

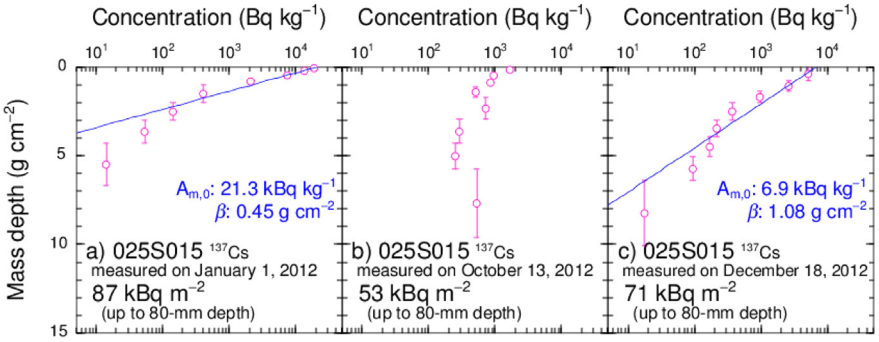

Fig. 2. Depth profiles for ^{137}Cs concentration with mass depth (wet weight) and approximate curves of best fit sampled in: a) December 2011, b) August 2012, and c) November 2012 at ID 025S015. An almost homogeneous and unnatural depth profile due to soil mixing was observed in b), however, the inventory up to 80 mm remains roughly the same.

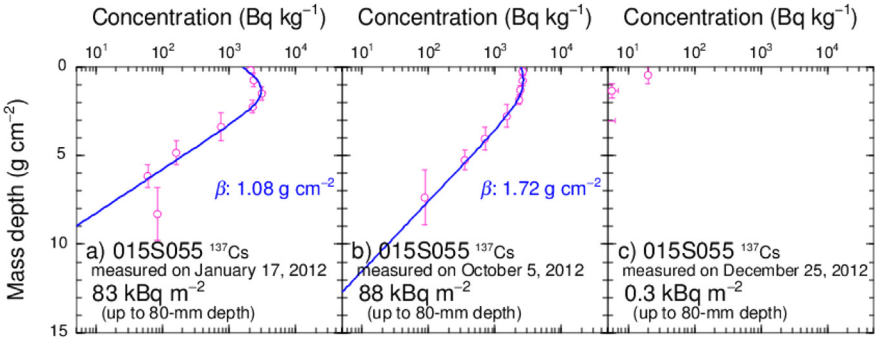

Fig. 3. Depth profiles for ^{137}Cs concentration with mass depth (wet weight) and approximate curves of best fit sampled in: a) December 2011, b) August 2012, and c) December 2012 at ID 015S055. The significant decrease in the inventory in c) was assumed to be due to decontamination work.

rodents, can also alter the depth profile. This type of homogeneous vertical distribution of ^{137}Cs concentration was also observed in sandy soil collected in coastal areas in June 2013. In another example, temporal changes in the depth profiles at ID: 015S055 are shown in Fig. 3. The significant decrease in the inventory seen here (Fig. 3c) is assumed to be due to decontamination work. If it was known in advance that a sampling location was decontaminated, then that site was abandoned in favor of another appropriate location in the same square grid. At locations where any type of disturbance in the depth profiles were considered to have occurred, exponential or hyperbolic secant fitting was not performed, and the data were not used for analysis.

3.4. Temporal changes in β and β_{eff} values

The values of β and related parameters of ^{137}Cs calculated by either the exponential or sech functions are summarized in Table S1 in supplementary materials, together with the geological location of the study site measured using global positioning system. Averaged β values including ones by hyperbolic secant function and standard deviations from the three collection periods (December 12–22, 2011 and April 17–19, 2012; August 21 to September 26; and November 26 to December 26) were 1.14 ± 0.56 (n = 83), 1.32 ± 0.77 (n = 82), and 1.47 ± 0.95 (n = 82) g cm^{-2}, respectively. The minimum and maximum values of β in each sampling period were also 0.34, 0.33, and 0.46 g cm^{-2}, and 4.42, 4.58, and 5.31 g cm^{-2}, respectively. Note that the exact number of days since the accident is not considered to be important because the depth profile is assumed to change discontinuously in response to weather conditions. The change in the relative frequency distribution of β between collection periods is shown in Fig. 4, which also shows that the obtained β values were lower than 3.0 g cm^{-2} at most study sites. These results suggest that radioactive cesium has downwardly moved in these soils. The averaged β values increased by a factor of 1.29 (=1.47 g cm^{-2}/1.14 g cm^{-2}) between the first and third collection periods, which represents approximately one year.

Average β_{eff} values for each successive collection period were 1.30 (n = 83), 1.57 (n = 82), and 1.91 (n = 82) g cm^{-2}, respectively. Here, the average was calculated using β_{eff} values for sech functions and β values for exponential functions together. According to the

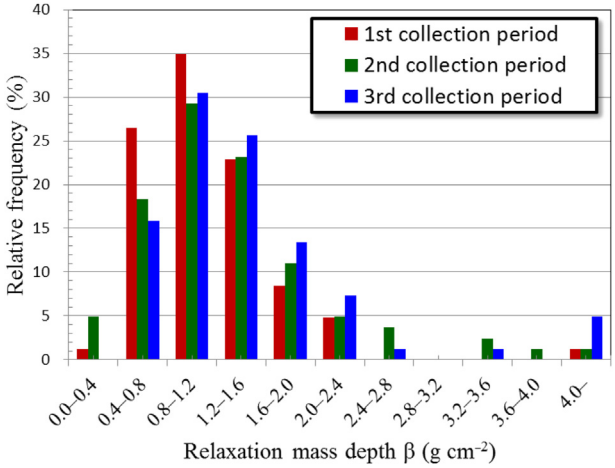

Fig. 4. Relative frequency distribution of the obtained relaxation depth β (g cm^{-2}) for ^{137}Cs (wet weight) in each collection period.

International Commission on Radiation Units and Measurements (ICRU), β values are expected to range from 0.1 to 3 g cm^{-2} within one year, 1–7 g cm^{-2} after several years, and 2–20 g cm^{-2} after more than 10 years (ICRU, 1994). The obtained averaged β and β_{eff} values around the Fukushima Dai-ichi NPP were both consistent with the ICRU data. This consistency implies that the migration of radioactive cesium into the depth direction is within normal ranges, though the ICRU data was summarized only about exponential depth profiles.

3.5. Regional variation in 1/10 depth

The 1/10 depths recorded during the third collection period around the Fukushima Dai-ichi NPP were plotted on a map (Fig. 5). The average value of $L_{1/10}$ was 3.01 cm ($n = 82$), and it was found that the majority of radioactive cesium remained within 5 cm of the ground surface in most study sites (71 sites).

According to the 1970s soil map of Japan (MLIT, 1971), most soil types within a 100-km radius of the Fukushima Dai-ichi NPP are

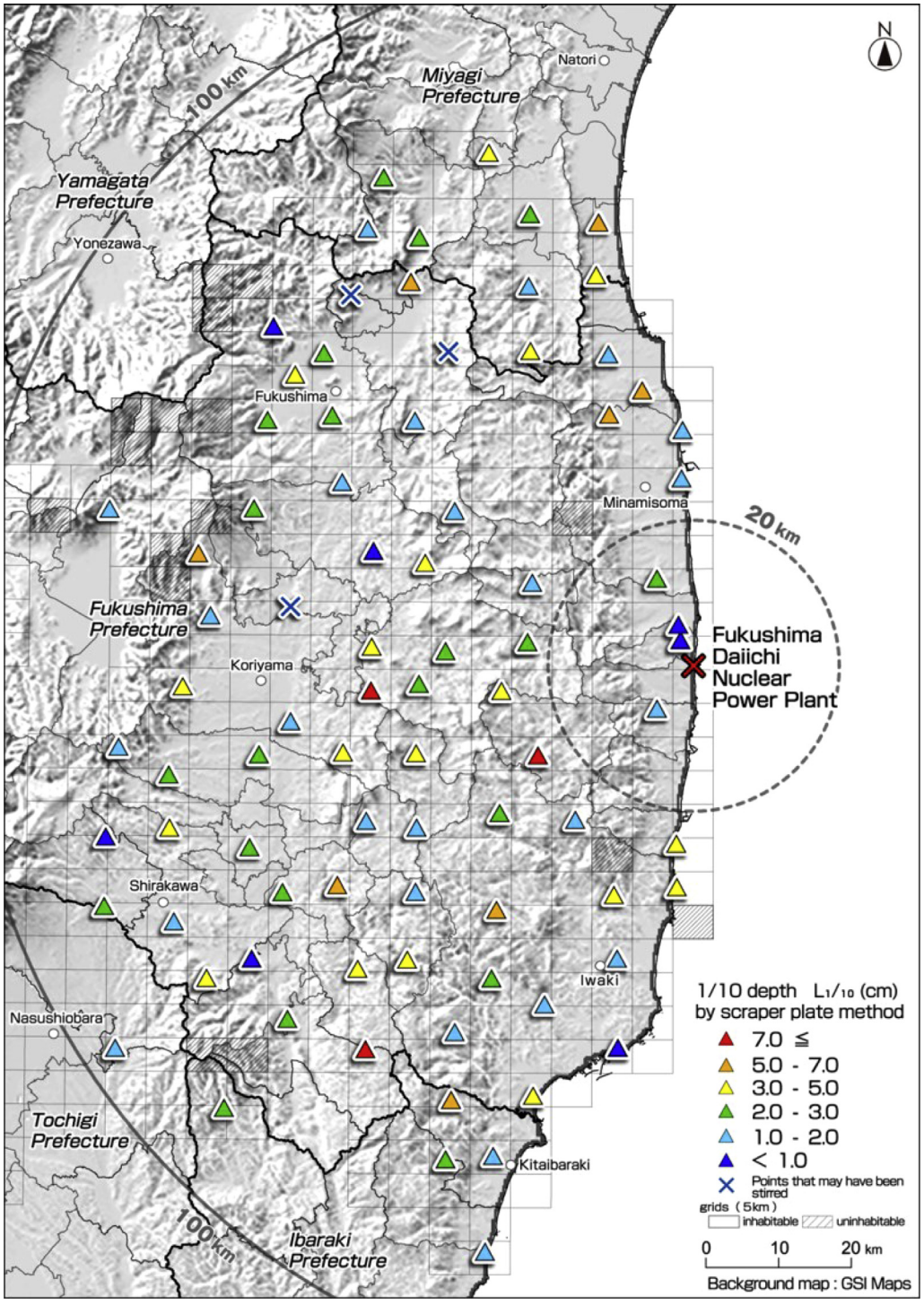

Fig. 5. Regional distribution of 1/10 depth $L_{1/10}$ (cm), at which 90% of the inventory of radioactive cesium is contained, in December 2012 around the Fukushima Dai-ichi NPP.

Brown Forest soils, which are either Cambisols or Stagnosols when translated in the World Reference Base for Soil Resources 2006 classifications (FAO et al., 2006) with the help of a bulletin by Obara et al. (2011). However, most study sites were located around public facilities within developed areas, such as community centers, Shinto shrines, Buddhist temples, and parks. In such cases, a clear relationship between the characteristics of the depth profile and soil type may not be apparent. Photographs taken at the study sites where the $L_{1/10}$ depth was found to be larger than 5 cm were carefully rechecked. It was found that most of these sites were characterized by many bumps of various sizes on the ground surface, but no evidence of human passage. Thus, these study sites were located on farmed land outside the urban areas. This may suggest that ground roughness induced by small animals, such as rodents or earthworms, which would be more prevalent in such agricultural land, play an important role in determining the initial distribution and redistribution of radioactive cesium. In particular, ID: 015S020, having the highest observed value of $L_{1/10}$ of 17 cm in the third collection period, showed an almost homogeneous depth profile. The sech function in Eq. (5) could be also fitted to these depth profiles and showed good agreement in the measured values.

3.6. Comparison of V values between the DM model and the sech function

The downward migration rate V obtained from the DM model (Eq. (10)) was compared with that based on ζ_0 values of the sech function in Eq. (11), illustrated in Fig. 6 for temporal changes of the depth profiles in the third collection period ($n = 25$). The V values were in good agreement with each other, except for two data points (i.e., ID: 060S040 and 065N010). The V values are determined by the mass depth, at which the concentration of radioactive cesium is at its maximum; therefore, the good agreement suggests that the mass depths were properly identified by both the DM model and the sech function. V values for ^{137}Cs are normally in the range of 0.2–1.0 kg m^{-2} y^{-1} (He and Walling, 1997); however, the obtained V values in this case were higher than the normal range. In addition, the DM model systematically underestimated the measured concentration of radioactive cesium as it exponentially decreased with increasing mass depth, as shown in Fig. 7. These findings may suggest that the concentration peak shift over time in Fukushima was influenced not only by diffusion and migration processes but also other factors, such as the mixing of topsoil by frosts that appear on and in the ground during the winter and the beginning of spring. Although, the collection period of this study were chosen after the consideration of the seasonal precipitation characteristics in Japan, further work should also consider the effects of snowfall or frost during the winter season. Furthermore, at those study sites with large differences between the V values of the DM model and the sech function, i.e., ID: 060S040 (Fig. 7b) and 065N010, the concentration of radioactive cesium is seen to gently decrease with increasing mass depth. The large discrepancy in the deep layers is assumed to hinder the determination of the concentration peak's mass depth in the DM model, causing the V values to be significantly different in such cases.

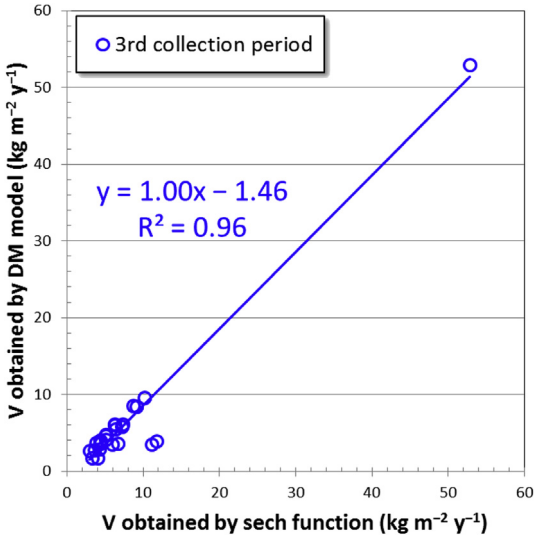

Fig. 6. Comparison of the downward migration rates V (kg m^{-2} y^{-1}) obtained by the DM model and those based on the sech function in December 2012.

4. Conclusions

Soil samples were collected using a scraper plate at 84 or 85 square grids within a 100-km radius of the Fukushima Dai-ichi NPP to obtain the depth profiles of radioactive cesium in the soil. Note that, some of the sampling sites were changed because of decontamination work. Considering the rainy season in Japan, three collection periods were determined, i.e., December 12–22, 2011 (81 sites) and April 17–19, 2012 (3 sites); August 21 to September 26, 2012 (85 sites); and November 26 to December 26 2012 (85 sites). Depth profiles for ^{134}Cs and ^{137}Cs were obtained from the quantitative analysis of the soil samples using high-purity germanium detectors. Parameters characterizing the vertical distributions were analyzed by fitting functions using either a traditional exponential equation or a newly defined hyperbolic secant equation. The hyperbolic secant equation was used for those depth profiles that changed with time, in which a concentration peak of radioactive cesium existed at a certain depth in the soil. The obtained relaxation depth β (g cm^{-2}) of ^{137}Cs is a useful parameter for quantifying distribution, and its characteristics were investigated in connection with several affecting factors. The average values and the standard deviations of β (wet weight) obtained in each of the three collection periods were 1.14 ± 0.56 (n = 83), 1.32 ± 0.77 (n = 82), and 1.47 ± 0.95 (n = 82) g cm^{-2}, respectively. These results suggest that radioactive cesium has downwardly moved in these soils. The value of β (wet weight) was found to increase by an average of 30% over approximately one year. Moreover, the effective parameter β_{eff} may be used for dose estimation and other applications. It is based on the exponential function in Eq. (1) and was calculated using the conversion factor of Saito and Jacob (1995) for those depth profiles that were well approximated by the sech functions. Average β_{eff}

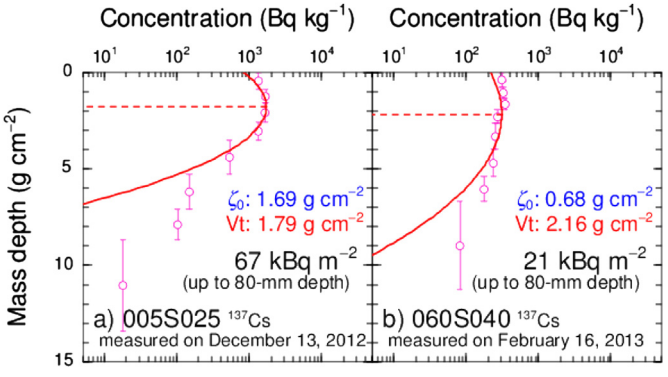

Fig. 7. Depth profiles for ^{137}Cs concentration with mass depth (wet weight) and approximate curves of best fit fitted using the DM model in Eq. (10) sampled in: a) November 2012 at ID 005S025, and b) December 2012 at ID 060S040. The obtained ^{137}Cs concentration gently decreases with increasing mass depth (wet weight) in b).

values for each of the three collection periods were 1.30 ($n = 83$), 1.57 ($n = 82$), and 1.91 ($n = 82$) g cm^{-2}, respectively; these were reasonably consistent with the ICRU data, which suggest that values should range from 0.1 to 3 g cm^{-2}. Next, the 1/10 depth $L_{1/10}$ (cm), at which 90% of the inventory of radioactive cesium is found, was plotted on a map around the Fukushima Dai-ichi NPP for use as a part of the decontamination strategy and for other applications. The average value of $L_{1/10}$ was 3.01 cm ($n = 82$) in December 2012, and it was found that radioactive cesium remained within 5 cm of the ground surface at most study sites (71 sites). Finally, the downward migration rate V (kg m^{-2} y^{-1}) in December 2012 ($n = 25$) was estimated by both the DM model and the sech function. The obtained V values were in good agreement with each other, with the exception of two study sites; however, the values of both were far higher than the normal range, i.e., 0.2–1.0 kg m^{-2} y^{-1}, as proposed by He and Walling (1997).

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Appendix A. Supplementary material

Supplementary data related to this article can be found online at http://dx.doi.org/10.1016/j.jenvrad.2014.10.001.

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