Development of an Easy and Simple Method to Measure the Environmental Radioactivity in Trees with Efficient Personal Dosimeters

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We have developed a simple method for the long-term monitoring of the seasonal variation of the radio-caesium radioactivity in trees using commercially available integrating dosimeters. Experiments and Monte Carlo simulations were performed to assess whether the proposed dosimeter-based method could measure the seasonal variation. From the experimental results, the performance variation among individual dosimeters was found to be 1.6%, which is sufficiently small compared with the expected seasonal variation. Moreover, the simulation results indicated that the measured value of the dosimeter increased owing to the photons passing through a stem, and a correction factor was necessary to compensate the influence of photon absorption and scattering in stem. We suggested that the simple and easy dosimeter-based method could be useful to measure the seasonal variations of radioactivity in trees.

Key Words: radio-caesium, PHITS (Particle and Heavy Ion Transport code System), dosimeter, environment, Monte Carlo simulation

1. Introduction

Many radionuclides were released into the atmosphere owing to the Fukushima Dai-ichi Nuclear Power Plant accident.1–3 Among the released radionuclides, 137Cs contamination has become a serious concern because of its long half-life (30 years).4 Thus, it is important to understand and predict the movement of 137Cs in the environment to avoid unnecessary radiation exposure.

Seventy percent of the contaminated area is forested, but decontamination plans have only been implemented in the living areas and agricultural areas. Forests store more radio-caesium than other areas, and there is a possibility that the concentrated radio-caesium stored in the forests will migrate to rivers and lakes. Therefore, it is particularly important to understand the movement of radio-caesium within forested areas.5

To analyze the dynamics of radio-caesium circulation in a forest, the 137Cs dynamics in trees must be monitored at numerous sites over a long
period of time. Most monitoring methods are based on invasive or destructive techniques. Although such methods provide accurate data, considerable work is required to perform the measurement, and long-term monitoring cannot be performed with destructive monitoring methods. Therefore, most existing monitoring methods are unsuitable for long-term measurements. A nondestructive, simple, and easy method is required for measuring the dynamics of $^{137}$Cs in trees due to seasonal variations.

D-Shuttle, which is a new dosimetry system, was developed by Chiyoda Technol Corporation for the purposes of continuous long-term personal dose monitoring of residents in the area affected by the accident.\textsuperscript{6, 7) This commercially available integrating dosimeter is small, light, and low-priced and can automatically collect data. If this dosimeter can be applied to the monitoring of the radioactivity in trees, the radioasium dynamics in a forest can be measured with ease and at a low cost. However, the dosimeter will only measure the external exposure, and the displayed value is 1-cm dose equivalent. Thus, to determine the radioactivity within the tree from the displayed value of the dosimeter, it is necessary to consider the influence of photon scattering and absorption in the tree. Further, to measure the seasonal variation of radioasium radioactivity in the forest, multiple measurement sites are required, which would utilize many dosimeters. Therefore, the degree of variation among individual dosimeters should be determined.

In this study, measurements were performed using 18 dosimeters under 4 experimental conditions to investigate the performance variation among individual dosimeters. In addition, Monte Carlo simulations were performed to accurately measure the seasonal variation in a tree, and the influence of photon absorption and scattering within the stem was investigated. These results were used to reveal whether the proposed dosimeter-based method could accurately measure the seasonal variation of the radioactivity in trees.

2. Materials and methods

2.1 Evaluation of the performance variation among individual dosimeters

The D-Shuttle dosimeter consists of a small, lightweight silicon semiconductor detector (about $2.77 \times 2.77 \times 0.32 \text{ mm}^3$), and its sensitivity was calibrated with a $^{137}$Cs source.\textsuperscript{6, 7) The dosimeter integrates the number of detected photons with energies over about 60 keV and converts the integrated count to the dose rate. The battery of the dosimeter lasts about 1 yr, and the dosimeter automatically records the dose rate and the total integrated dose every hour.

The radioactivity (Bq) of a target must be estimated from the recorded dosimeter value because the dosimeter is designed to measure the external exposure, and the recorded values represent the 1-cm dose equivalent ($\mu$Sv/h). Thus, a calibration experiment was performed using a radiation source with a known radioactivity. Fig. 1 shows a photograph and schematic of the experimental setup. A $^{137}$Cs source with a radioactivity of 18.5 MBq and 18 dosimeters were placed in a chamber, in which the internal light and temperature environment can be changed, and the distance between the $^{137}$Cs source and the dosimeters was 50 cm. Because the actual measurements of the seasonal variations of the radioactivity in a tree will be performed in the field, the measurement environment, namely the light and temperature, will vary. Therefore, measurements in this experiment were performed under four conditions: (1) with light at $22^\circ$C, (2) without light at $22^\circ$C, (3) with light at $30^\circ$C, and (4) without light at $30^\circ$C. Nine records were utilized for each condition. The measurement duration of each record was 1 h. In the analysis, the recorded values were converted to dose rates at $20^\circ$C using the temperature characteristic line, whose range is from $-20$ to $40^\circ$C, for the dosimeter pro-
vided by Chiyoda. Then, the converted dose rate was converted to counts per hour using the dose-rate–count-rate conversion factor provided by Chiyoda. The converted count rates of the four conditions were compared.

The performance variation among individual dosimeters was investigated by the following procedure. First, the mean values of the count rates of the nine records were calculated for each of the 18 dosimeters, and the mean of all obtained mean values, i.e., the mean of all measured values, was determined. Next, the statistical errors of the mean values of the nine measurements were calculated for each of the dosimeters, and the mean value of the statistical errors of the 18 dosimeters was determined. Then, the standard deviation of the measured values was determined using the mean values of the nine records for each of the 18 dosimeters. Finally, performance variation among the individual dosimeters under a given condition was calculated using the following equation:

\[ N_{\text{variation}} = \sqrt{N_{\text{deviation}}^2 - N_{\text{error}}^2} \]  

where \( N_{\text{variation}} \) is the performance variation among individual dosimeters under a given condition, \( N_{\text{deviation}} \) is the standard deviation of the measured values of the 18 dosimeters, and \( N_{\text{error}} \) is the mean value of the statistical errors of the 18 dosimeters.

2.2 Evaluation of the influence of photon absorption and scattering

The counting rates of the dosimeter for various distances between the \(^{137}\text{Cs}\) source and the dosimeter were calculated using a Monte Carlo simulation. In addition, the counting rates were calculated for the case in which the \(^{137}\text{Cs}\) source was positioned at the center of a stem of a tree and the dosimeter was placed on the side of the tree.

The Monte Carlo simulation was performed with the Particle and Heavy Ion Transport Code System (PHITS).\(^9\) To simulate the count rates in the case where photons from \(^{137}\text{Cs}\) were not absorbed and scattered by a stem, a point source emitting 662-keV photons and a rectangular parallelepiped (2.77×2.77×0.32 mm\(^3\)) of silicon imitating the dosimeter detector were placed in a simulation space filled by air (Fig. 2 (a)). The distance \( R \) between the point source and the rectangular parallelepiped was varied to 5–15 cm by 1 cm in different simulations. On the other hand, Fig. 2 (b) shows a schematic of the simulation setup in the case where photons were absorbed or scattered by the stem. A cylinder,
whose height was 10 cm, chemical formula was \( \text{C}_{5.29}\text{H}_{25.07}\text{O}_{11.41}\text{N}_{0.08}\text{S}_{0.004} \), and density was 0.81 g/cm\(^3\), was placed in the simulation space to imitate raw sugi wood (Japanese cedar). The radius of the cylinder \( R \) was varied to 5–15 cm by 1 cm in different simulations. The rectangular parallelepiped representing the dosimeter detector was placed on the side surface of the cylinder (wood). In both the simulations, \( 6.4 \times 10^9 \) photons were generated from each of the sources, and the energy distributions of the photons deposited in the rectangular parallelepiped were recorded for each distance.

In actuality, \(^{137}\text{Cs} \) is distributed in various parts of a tree, such as the bark, the heartwood, and sapwood.\(^9,10\) The radiation from the \(^{137}\text{Cs} \) in the bark is easy to detect by the dosimeter placed on the side of the tree because the distance between the radiocaesium and the dosimeter is short. On the other hand, the measurement of the \(^{137}\text{Cs} \) in the wood is more difficult because the radiation is attenuated by the stem. In addition, the increase of the distance between the radiocaesium and the dosimeter reduces the count rate. In the present simulations, the radiocaesium was placed at the center of the wood, which was the most difficult condition for the measurement of the radiocaesium in a tree. If this method was useful under the condition, it would also be useful under the other easier conditions.

### 3. Results

#### 3.1 Evaluation of the performance variation among individual dosimeters

Table 1 summarizes the means of the measured values, \( N_{\text{error}} \), \( N_{\text{deviation}} \), and \( N_{\text{variation}} \), under each experimental condition.

| Experimental condition | Mean of measured value [cph] | \( N_{\text{error}} \) [cph] | \( N_{\text{deviation}} \) [cph] | \( N_{\text{variation}} \) [cph] (relative value [%]) |
|------------------------|-----------------------------|----------------|----------------|----------------------------------|
| 1                      | 545.70                      | 7.79           | 10.29          | 6.72 (1.26)                      |
| 2                      | 547.03                      | 7.80           | 10.82          | 7.50 (1.37)                      |
| 3                      | 545.82                      | 7.79           | 14.24          | 11.92 (2.18)                     |
| 4                      | 547.14                      | 7.82           | 11.78          | 8.81 (1.61)                      |

#### 3.2 Evaluation of the influence of photon absorption and scattering

Fig. 3 shows the simulation results obtained using the two simulation conditions. The horizontal axis shows the distance \( R \) between the \(^{137}\text{Cs} \) source and

![Fig. 2 Schematic of the simulation setups (a) without and (b) with the wood (Color online).](image-url)
the dosimeter, and the vertical axis shows the photon count in the detector per unit of generated particles. The points labeled 'Air' and 'Wood' indicate simulation results without and with the wood, respectively. The error bars in Fig. 3 indicate the statistical errors at each point. Here, the count is the integral value of the number of photon counts that deposit 60-keV or more energy to the detector because the dosimeter integrates the number of detected photons with energies over about 60 keV. In Fig. 3, the 'Wood' results are larger than the 'Air' results. Thus, the radioactivity of radiocaesium in a tree measured using this method may be larger than the actual radioactivity. To correctly measure the radioactivity, a correction factor to compensate the influence of photon absorption and scattering within the tree is required.

Fig. 4 shows the ratio of the simulation results of 'Air' and 'Wood'.

Fig. 4  Ratio of the simulation results of 'Air' and 'Wood'.

From the simulation results, the recorded value of the dosimeter increased when photons traveled through a stem. Fig. 5 shows the distributions of the energy depositions for 'Air' and 'Wood' calculated at R=10 cm. The yield for 'Wood' is larger than that for 'Air' below about 400 keV, likely a result of Compton scattering in the wood. Thus, in the case where the radiocaesium radioactivity is measured in a tree, the yield of radiation is higher because the influence of scattering in the stem is stronger than that of attenuation by absorption.

The background due to the ambient dose inhibits accurate radioactivity measurements in the field. It is impossible to distinguish between the signals of the $^{137}\text{Cs}$ in a tree and those of the high background radiation from the displayed value of the D-Shuttle because the dosimeter only displays a 1-cm dose equivalent as a measurement value. The radioactivity of the background is much higher than the radioactivity in the trees. Thus, in order to accurately observe the dynamics of radiocaesium in a tree with dosim-
eters, it is necessary to eliminate the influence of the background radiation from the measurement value of the dosimeters placed on the side of the tree. The influence of the background radiation can be eliminated by measuring only the background of a measurement point of radiocaesium in a tree with the other D-Shuttle and subtracting the background value from the measurement value (background radioactivity in a tree). In this case, however, a sufficient measurement time is necessary to reduce the statistical error of the result. Herein, the lower measurement limit of the radioactivity of radiocaesium in a wood was examined under an environmental condition. The average dose rate background during measurement is 1.25 µSv/h from the actual ambient dose in the Fukushima forest. The measured wood is a sugi (Japanese cedar) tree with a radius of 10 cm and a height of 30 cm, and the detection range of the dosimeter placed on the side of the wood is 15 cm above and below the dosimeter position. The dosimeter is also covered with a 3-cm lead shield to reduce the background radiation. In addition, the radioactivity of 137Cs at the center of the wood is about 2.6 kBq. This radioactivity was calculated from the dry weight of the wood and the 137Cs activity concentration data of a real sugi tree. The counts and statistical error of the measurement are described as follows:

**Measured value ± SE**

\[
\text{Measured value} = \frac{N_{\text{do}si} \cdot t}{\sqrt{N_{\text{do}si} \cdot t}} \quad (2)
\]

where \(N_{\text{do}si}\) is the counts per hour of the dosimeter attached to the side of the wood, SE is the statistical error, and \(t\) is the measurement time. The counts of the background can be expressed as follows:

**Background ± SE**

\[
\text{Background} = \frac{N_{\text{back}} \cdot t}{\sqrt{N_{\text{back}} \cdot t}} \quad (3)
\]

where \(N_{\text{back}}\) is the counts per hour of the background. Thus, the counts of radiocaesium in the wood can be calculated from Equations (2) and (3) as follows:

\[
\text{Measured value of Cs ± SE} = (N_{\text{do}si} - N_{\text{back}}) \cdot t \pm \sqrt{(N_{\text{do}si} + N_{\text{back}}) \cdot t} \\
= N_{\text{Cs}} \cdot t \pm \sqrt{(N_{\text{Cs}} + 2N_{\text{back}}) \cdot t} \quad (4)
\]

where \(N_{\text{Cs}}\) is the counts per hour of radiocaesium. Because the seasonal variation of the radiocaesium radioactivity in a tree is expected to be approximately 9–31%, a radioactivity measurement with a statistical error of 3% or less is required to observe this variation. From Equation (4), the required measurement time is described as follows:

\[
\frac{\sqrt{(N_{\text{Cs}} + 2N_{\text{back}}) \cdot t}}{N_{\text{Cs}} \cdot t} = 0.03 \quad (5)
\]

In this case, \(N_{\text{back}}\) without the lead shield is about 105.8 counts/h from the dose rate of the background (1.25 µSv/h). This value reduces to 31% (32.8 counts/h) by the shielding effect of the lead and the measured wood. \(N_{\text{Cs}}\) was estimated by Monte Carlo simulation. The simulation was carried out with a schematic, which is almost the same as the schematic shown in Fig. 2 (b); however, the height and \(R\) of the cylinder were respectively 30 cm and 10 cm. The count number of photons in the detector was calculated by the simulation. The measurement time was calculated from the count of generated photons in the simulation and the 137Cs radioactivity (2.6 kBq). In this case, the count number was 26.8 counts, and the measurement time was 12.6 h. Therefore, the counts per hour (cph) was approximately 2.13 counts/h. Furthermore, considering the calibration curve of \(\text{Ratio} = 0.50 + 0.020 \times R\) in Fig. 4, this value is 1.49 counts/h. Thus, from Equation (5), about 34,000 h of measurements are required to measure 2.6 kBq of radiocaesium in the wood with a statistical error of 3% using one dosimeter. Thus, the seasonal variation of the radioactivity in a tree cannot be measured with only one dosimeter. However, the measurement time can be reduced by increasing the number of detectors measuring the same position. In the above case, if fifteen dosimeters covered with the lead shield are
used, the time can be reduced to about 2,200 h, i.e., about 3 months. Therefore, this discussion suggests that there is a possibility that the proposed method using the commercially available D-Shuttle integrating dosimeter can accurately measure the seasonal variation of radioactivity in trees.

In Table 1, the discrepancy between the means of all measured values is within the statistical error range. This result shows that the measured value of the dosimeter is not affected by light, and the temperature correction of the measured value is effective. Thus, an accurate measurement can be obtained in the field by determining the temperature at the area where the dosimeter is placed. Here, we consider the influence of an error in the temperature measurement on the measured value of the dosimeter. We assumed that the use of a thermometer is simple and inexpensive, and the error of the temperature measurement is 1°C. When the temperature is shifted by 1°C, the measured value of the dosimeter has an error of 0.3%. This value is sufficiently small compared with the expected seasonal variation. In addition, the performance variation among individual dosimeters is 1.6%, which is also sufficiently small compared with the expected seasonal variation. Therefore, the error in the temperature and the performance variation have negligible influence on the dosimeter-based measurements.

5. Conclusion

We investigated whether the variation of the radiocaesium radioactivity in trees could be measured using personal D-Shuttle dosimeters using experiments and Monte Carlo simulations. It was suggested that the method could sufficiently observe the seasonal variation of radioactivity in trees.

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要旨

個人積算線量計を用いた樹体における環境放射能の簡易測定手法の開発

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樹体に含まれる放射性セシウムの季節変動を長期間モニタリングするため、市販の積算線量計を用いた簡易測定手法を開発した。開発した手法で季節変動が測定可能かどうか評価するため、実験及びモンテカルロシミュレーションを実施した。実験の結果、線量計指示値の個体間のばらつきは1.6%であり、期待される季節変動に比べて十分に小さいことがわかった。また、シミュレーションの結果、本手法で樹体に含まれる放射性セシウムを測定する場合、樹体による光子の吸収及び散乱の影響で測定値が増加するため、これを補正する必要があることがわかった。以上の結果から、本手法により、樹体に含まれる放射性セシウムの季節変動が測定可能であることが示唆された。