Detailed Evaluation of Natural Gamma-Radiation Field due to Uranium (238U) Series

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Peak counting rates of a Ge (Li) detector for γ-rays of interest and exposure rates at 1 m above the ground have been calculated using the soil and air concentration profiles of naturally occurring radionuclides at Tsukuba (Yatabe City, Tsukuba District, Ibaraki Prefecture). Decrease in the peak counting rate for 609 keV 214Bi γ-ray above the ground, which was due to diffusion loss of 222Rn under the ground, was evaluated to be about 44% of the peak counting rate calculated assuming uniform distribution of 222Rn under the ground, and decrease in the exposure rate was evaluated to be about 13% of the total exposure rate calculated assuming uniform distribution. Field measurement of γ-rays from sources in the soil and air was also made with the Ge (Li) detector at Tsukuba. The calculated peak counting rates where the diffusion loss of 222Rn was involved agreed with those observed in situ. During the observation the calculated peak counting rate for the 609 keV γ-ray from airborne 214Bi ranged 3~19% of that from uniformly distributed 214Bi source under ground, and the calculated exposure rate from the airborne 222Rn daughters ranged 1~9% of the total exposure rate.

KEYWORDS: peak counting rate, field gamma-spectrometry, laboratory gamma-spectrometry, natural radioactivity, gamma-radiation, exposure rates, radon-222, daughter products, soil, air, diffusion loss, spatial distribution, escape-to-production ratio

I. INTRODUCTION

Radionuclide concentrations in the soil and exposure rates have been obtained by field measurements of γ-rays (especially with a Ge (Li) detector) and compared with those by in-laboratory soil sample measurements(11)~(11). Most of these investigations have been carried out under the assumption that all the natural γ-emitters (uranium (238U) series, thorium (232Th) series and 40K) are uniformly distributed in the soil. Recently, the contribution of airborne 222Rn daughters to natural γ-radiation field has been calculated(11). The influence
of the diffusion loss of $^{222}$Rn under the ground, which is mainly dominated by $^{222}$Rn escape-to-production ratio, on the natural γ-radiation field has also been evaluated by calculation\textsuperscript{(13)}. These calculations meet the needs of the times\textsuperscript{(14)-(15)}. It is, therefore, highly desirable that these calculations are supported by experiments.

This paper describes experimental verification to the above-mentioned calculations. Measurements were made at Tsukuba (Yatabe City, Tsukuba District, Ibaraki Prefecture), the geology of which is Kanto loam\textsuperscript{(16)}, because it was considered that the $^{222}$Rn escape-to-production ratio of the volcanic ash soil was large\textsuperscript{(17)}.

II. SOIL AND AIR CONCENTRATION PROFILES OF NATURALLY OCCURRING RADIONUCLIDES

1. Soil Concentration Profiles of Naturally Occurring Radionuclides

Radium-226 ($^{226}$U), $^{233}$Th and $^{40}$K present in the soil are assumed to be distributed uniformly. The soil concentration profile of $^{222}$Rn can be calculated using the measured air ratio, $^{222}$Rn concentration in the soil air, $^{226}$Ra concentration and $^{222}$Rn escape-to-production ratio\textsuperscript{(13)}.

(1) Soil Phases

Soil is made up of solid, liquid and gaseous material. The sources present in the soil naturally, $^{226}$Ra ($^{238}$U), $^{233}$Th and $^{40}$K, are contained in soil particles (solid phase). Soil density, which affects the source activity per gram of soil, depends on soil water (liquid phase). For the uranium ($^{238}$U) series, most of the γ-rays are emitted by $^{214}$Pb and $^{214}$Bi which are daughters of $^{222}$Rn, a gaseous daughter of $^{226}$Ra. Radon-222 can emanate from soil particles, diffuse through the soil air (gaseous phase) to the surface and escape into the atmosphere, since it is a gas. Although a $^{232}$Th daughter, $^{220}$Rn, is also a gas, its extremely short half-life of 55.3 s reduces the effect of its migration in the soil to a negligible proportion.

The following terms about soil properties are defined in Ref. (18).

The water content by wet soil $w($%) is usually expressed on a percentage basis, i.e.

$$w = \frac{W_w}{W_T} \times 100,$$

where $W_w$ is the weight of water (g) and $W_T$ the total weight of a soil (g).

The bulk density of a soil $\rho_s$ (g·cm$^{-3}$) is represented by

$$\rho_s = \frac{W_T}{V_T} = \frac{W_w + W_s}{V_T},$$

where $W_s$ is the weight of soil solids (g) and $V_T$ the total volume of a soil (cm$^3$).

The specific gravity of soil solids $G_s$ is represented by

$$G_s = \frac{\gamma_s}{\gamma_w} = \frac{W_s}{V_s \gamma_w},$$

where $\gamma_w$ is the unit weight of water at 4°C, $\gamma_s$ the unit weight of the soil solids and $V_s$ the volume of soil solids (cm$^3$). This quantity is used to compute the porosity and the air ratio.

The porosity of a soil $n$ is represented by

$$n = \frac{V_v}{V_T} = 1 - \frac{W_s}{G_s \gamma_w V_T},$$

where $V_v$ is the volume of voids (cm$^3$).
The air ratio of a soil $n_a$ is represented by

$$n_a = \frac{V_a}{V_T} = 1 - \frac{1}{V_T \gamma \nu} \left( \frac{W_t}{G_s} + W_w \right),$$

(5)

where $V_a$ is the volume of air (cm$^3$).

(2) Soil Air Concentration Profile of $^{222}$Rn

After a 0.3-cm-i.d. brass tube was inserted into a bore hole, the drilling residue was repacked. Soil air samples were drawn from depths into evacuated 100-cm$^3$ flasks, using the tube sealed into the ground. The filter chamber of which the inner diameter was 6 cm and the inner height was 6 cm on the bottom of the tube was filled with glass wool to prevent soil particles from being drawn up into the sampling flask$^{(19)}$. Six tubes like the above were set at the depths of 15, 25, 35, 60, 80 and 100 cm at Tsukuba, respectively. The soil air sample was introduced into an ionization chamber after passing through calcium chloride in a glass chamber and a millipore filter (type PH) supported inside a holder. The ionization current due to $^{222}$Rn and its daughters was measured with a vibrating-reed electrometer 3 h after the introduction of the soil air sample, and automatically recorded. The $^{222}$Rn concentration in the soil air $C_s(z)$ (pCi·cm$^{-3}$ of soil air) is given by

$$C_s(z) = 5.15 \times 10^4 \times \frac{V}{R_x} P \exp (\lambda_{Rn} T),$$

(6)

where $V$ is the value recorded (mV), $R_x$ the input resistance of the vibrating-reed electrometer ($\Omega$), $P$ the correction factor for the dilution of soil air, $\lambda_{Rn}$ the decay constant of $^{222}$Rn (s$^{-1}$) and $T$ the time between sampling and measurement (s).

(3) Radionuclide Concentrations in Soil

The $^{226}$Ra, $^{232}$Th and $^{40}$K concentrations in sampled soil were determined by laboratory Ge(Li) $\gamma$-ray spectrometry. This was carried out with reference to the methods developed by Gustafson & Brar$^{(2)}$ and Kataoka & Ikebe$^{(20)}$.

(4) Radon-222 Escape-to-Production Ratio of Soil

The $^{222}$Rn emanating power of this soil (The $^{222}$Rn emanating power of soil E.P. is the activity of $^{222}$Rn that escapes into soil air per gram of dry soil at equilibrium state.) was also measured$^{(20)}$. Radon-222 escape-to-production ratio of the soil $\delta$ is given by

$$\delta = E.P./S_{Ra, dry},$$

(7)

where $S_{Ra, dry}$ is the $^{226}$Ra concentration in the soil (pCi·g$^{-1}$ of dry soil).

2. Air Concentration Profile of $^{222}$Rn Daughters

From Oct. 12~16, 1979, the $^{222}$Rn daughters concentrations in the air were measured at 1, 10 and 100 m above the ground on the Tsukuba Meteorology Tower. They were collected on a millipore filter (type RA) and were counted with a zinc-sulfide alpha scintillation counter. The sampling time was 20 min at about $12 \ell \cdot \min^{-1}$ and the counting time was 24 min at 1 and 10 m above the ground level. At 100 m, 30 min at about $18 \ell \cdot \min^{-1}$ and 25 min were chosen as sampling and counting times, respectively.

III. FIELD $\gamma$-RAY SPECTROMETRY

1. Calculations of Peak Counting Rates of Ge(Li) Detector

The total flux density of uncollided $\gamma$-rays of energy $E_i$ at height $H$ above a flat air-ground interface (see Fig. 1) due to the sources in the soil is given by
\[ \Phi_{\text{prim},i} = 2\pi \int_{0}^{\pi/2} \int_{0}^{R} \frac{n_{i}A(z)}{4\pi r^{2}} r^{2} \sin \theta \]
\[
\cdot \exp \left\{ -\mu_{g} \rho_{g} \left( r - \frac{H}{\cos \theta} \right) \right\} \exp \left\{ -\mu_{a} \rho_{a} \frac{H}{\cos \theta} \right\} dr d\theta ,
\]

where \( \Phi_{\text{prim},i} \) is the uncollided \( \gamma \)-ray flux density \((\text{cm}^{-2} \cdot \text{s}^{-1})\), \( n_{i} \) the number of \( \gamma \)-rays of energy \( E_{i} \) per disintegration of \( ^{226}\text{Ra} \), \( ^{222}\text{Rn} \), \( ^{232}\text{Th} \) and \( ^{40}\text{K} \), \( z \) the distance from the air-ground interface (cm, \( z \) is negative below the ground), \( H \) the distance from the air-ground interface to the detector (cm), \( r \) the distance from each element of differential volume to the detector (cm), \( \theta \) the \( \gamma \)-ray angle of incidence with respect to the perpendicular to the air-ground interface (deg), \( A(z) \) the concentration of the source \( ^{226}\text{Ra} \), \( ^{222}\text{Rn} \), \( ^{232}\text{Th} \) or \( ^{40}\text{K} \) in the soil with depth \( z \) \((\text{s}^{-1} \cdot \text{cm}^{-3} \text{ of soil})\), \( \mu_{a} \) the mass attenuation coefficient of the air \((\text{cm}^2 \cdot \text{g}^{-1})\), \( \mu_{g} \) the mass attenuation coefficient of the ground \((\text{cm}^2 \cdot \text{g}^{-1})\), \( \rho_{a} \) the density of the air \((\text{g} \cdot \text{cm}^{-3})\) and \( \rho_{g} \) the soil bulk density \((\text{g} \cdot \text{cm}^{-3})\) \((21)(22)\).

For uniformly distributed sources such as the thorium \( ^{232}\text{Th} \) series and \( ^{40}\text{K} \) dependence of the uncollided \( \gamma \)-ray flux density on the \( \gamma \)-ray angle of incidence with respect to the perpendicular to the air-ground interface \( \theta \) is obtained by integration over \( r \), hence

\[ \Phi(\omega)_{\text{prim},i} = \frac{n_{i}A_{\text{Th,K}}}{2\mu_{a} \rho_{a}} \exp \left\{ -\frac{\mu_{a} \rho_{a} H}{\omega} \right\} , \]  

where \( \omega = \cos \theta \) and \( A(z) = A_{\text{Th,K}} \); \( A_{\text{Th,K}} \) is the \( ^{232}\text{Th} \) or \( ^{40}\text{K} \) concentration. \( \Phi(\omega)_{\text{prim},i} \) is represented by Eq. (9) for thorium \( ^{232}\text{Th} \) series and \( ^{40}\text{K} \), but for uranium \( ^{238}\text{U} \) series \( ^{226}\text{Ra} \) daughters it is as follows: Since

\[ A(z) = A_{\text{Ra}} \left\{ 1 - \delta \exp \left( \frac{\lambda_{\text{Ra}}}{D} z \right) \right\} , \]

is the \( ^{222}\text{Rn} \) concentration in the soil \((11)\), integration over \( r \) yields

\[ \Phi(\omega)_{\text{prim},i} = \frac{n_{i}A_{\text{Ra}}}{2\mu_{a} \rho_{a}} \left\{ \frac{1}{\mu_{a} \rho_{a}} - \frac{\delta}{\omega \sqrt{\lambda_{\text{Ra}}/D + \mu_{a} \rho_{a}}} \right\} \exp \left\{ -\frac{\mu_{a} \rho_{a} H}{\omega} \right\} , \]

where \( A_{\text{Ra}} \) is the \( ^{226}\text{Ra} \) concentration in the soil and \( D \) the diffusion coefficient for \( ^{222}\text{Rn} \) in the soil gas \((\text{cm}^2 \cdot \text{s}^{-1})\).

When a Pb shield of thickness \( t \) is used, as shown in Fig. 1, Eq. (9) can be rewritten

\[ \Phi(\omega)_{\text{prim},i} = \frac{n_{i}A_{\text{Ra}}}{2\mu_{a} \rho_{a}} \exp \left\{ -\frac{\mu_{a} \rho_{a} (H-t)}{\omega} - \frac{\mu_{p} \rho \frac{1}{\omega}}{\omega} \right\} , \]

and Eq. (11) can be rewritten

\[ \Phi(\omega)_{\text{prim},i} = \frac{n_{i}A_{\text{Ra}}}{2\mu_{a} \rho_{a}} \left\{ \frac{1}{\mu_{a} \rho_{a}} - \frac{\delta}{\omega \sqrt{\lambda_{\text{Ra}}/D + \mu_{a} \rho_{a}}} \right\} \exp \left\{ -\frac{\mu_{a} \rho_{a} (H-t)}{\omega} - \frac{\mu_{p} \rho \frac{1}{\omega}}{\omega} \right\} . \]

If the \( ^{222}\text{Rn} \) daughters distribution in the air is uniform from \( z = H \) to \( z = H' \), \( \Phi(\omega)_{\text{prim},i} \) is given by
\[ \Phi(\omega)_{\text{prim},i} = \frac{n_i A_{\text{Ra},d}}{2} \cdot \frac{1}{\rho_a \alpha_a} \left\{ 1 - \exp \left( \frac{\mu_a R_a (H' - H)}{\omega} \right) \right\}, \]  

where \( A_{\text{Ra},d} \) is the \( ^{222}\text{Rn} \) daughters concentration in the air (s\(^{-1}\)·cm\(^{-3}\) of air).

The Ge(Li) detector (60 cm\(^3\)) was calibrated using a \( ^{226}\text{Ra} \) standard source. The angular response of the detector was measured at 10 positions from the detector axis, 0, 15, 30, 45, 60, 75, 90, 120, 150 and 180\(^\circ\), all at a source-detector distance of 1 m.

A peak counting rate \( N_{\text{prim},i} \) (cps) can be obtained using the following equation:

\[ N_{\text{prim},i} = \int_0^\pi R(\theta) \Phi(\omega)_{\text{prim},i} \, d\theta, \]

where \( R(\theta) \) is the angular response of the detector. The peak counting rates of Ge(Li) detector, therefore, can be calculated using the soil and air concentration profiles of naturally occurring radionuclides.

2. Field Measurements of \( \gamma \)-rays with Ge(Li) Detector

Field measurement of the \( \gamma \)-rays were made simultaneously with the observation of the \( ^{222}\text{Rn} \) daughters concentration in the air. The Ge(Li) detector was placed 1 m above the air-ground interface at the center of the experimental area of which the diameter was about 150 m. The arrangement is shown in Fig. 1. In measuring \( \gamma \)-rays from the airborne \( ^{222}\text{Rn} \) daughters, a Pb shield of 5 cm thickness was placed under the detector, as shown in the figure. This Pb shield was used to cut off the \( \gamma \)-rays coming from angles of 0 to about 80\(^\circ\). The sensitivity of the detector for measurement of the airborne \( ^{222}\text{Rn} \) daughters could be increased using this Pb shield. Counts were accumulated for live time of 8,192 s.

IV. RESULTS AND DISCUSSIONS

The soil properties measured here are shown in Table 1. The bulk density and water content by wet soil are considerably different from those chosen by Beck(23).

The \( ^{222}\text{Rn} \) concentration in the soil air was measured for each depth. The soil air concentration profiles of \( ^{222}\text{Rn} \) are shown in Fig. 2. The pipes fixed at the depths of 60, 80 and 100 cm were waterlogged in the soil on Oct. 12, 1979. Those of 80 and 100 cm were also waterlogged on Oct. 15. The ground water tables estimated from those observations are also given in Fig. 2. The observed \( ^{222}\text{Rn} \) concentrations in the soil air, except that at the depth of 60 cm, lie between those calculated using diffusion coefficients of 0.05 and 0.05/10 cm\(^2\)·s\(^{-1}\)(13). They are calculated under the assumption that no ground water exists there.

The \( ^{226}\text{Ra}, ^{232}\text{Th} \) and \( ^{40}\text{K} \) concentrations of the soil are given in Table 2. They are the average of six samples. Using the \( ^{226}\text{Ra} \) concentration in this table and the \( ^{222}\text{Rn} \) emanating power, which amounted to 0.448±0.032 pCi·g\(^{-1}\) of dry soil, \( \delta \) was calculated and found to be 47.6±4.8%. This value is normal as volcanic ash soil but is higher than those

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Soil properties at Tsukuba</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water content by wet soil</td>
<td>49.5%</td>
</tr>
<tr>
<td>Porosity</td>
<td>78.4%</td>
</tr>
<tr>
<td>Air ratio</td>
<td>22.0%</td>
</tr>
<tr>
<td>Bulk density</td>
<td>1.14 g·cm(^{-3})</td>
</tr>
<tr>
<td>Specific gravity</td>
<td>2.67</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Table 2</th>
<th>( ^{226}\text{Ra}, ^{232}\text{Th} ) and ( ^{40}\text{K} ) concentrations in Tsukuba soil</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nuclide</td>
<td>Concentration (pCi·g(^{-1}) of dry soil)</td>
</tr>
<tr>
<td>----------</td>
<td>-----------------------------------------------</td>
</tr>
<tr>
<td>( ^{226}\text{Ra} )</td>
<td>0.940±0.068</td>
</tr>
<tr>
<td>( ^{232}\text{Th} )</td>
<td>0.828±0.032</td>
</tr>
<tr>
<td>( ^{40}\text{K} )</td>
<td>7.44 ± 0.20</td>
</tr>
</tbody>
</table>
of other soils except calcareous soil [17].

Using the above mentioned quantities, the soil concentration profile of $^{222}$Rn was calculated by the Eq. (10). The result are given in Fig. 3. From this figure, it is found that $^{222}$Rn diffuses through the soil air and escapes considerably into the atmosphere.

Some examples of the air concentration profiles of the $^{222}$Rn daughters are shown in Fig. 4. The concentration observed at each level above the air-ground interface during this period varied between 10 and 200 pCi·m$^{-3}$.

The peak counting rates for the $\gamma$-rays of interest at 1 m above the ground were calculated using Eq. (15). The calculated and observed peak counting rates are given in Table 3(a) and (b). Cases (a) and (b) in the table show examples without Pb shield and with Pb shield, respectively. The errors (standard deviations) of the calculated counting rates of $\gamma$-rays from the ground are ascribed
to the concentrations of the radionuclides in the soil and to the response of the Ge(Li) detector. The $^{222}\text{Rn}$ daughters concentration used in calculating the contribution from the airborne nuclides is the average of three levels. The height $H$ and $H'$ are taken to be 1 and 1,000 m, respectively. For the 609 keV $^{214}$Bi peak in (a) and (b), the sum of both the calculated counting rate due to ground and air origin agrees with the counting rate of in-situ measurement. They almost agree for the 1,461 keV $^{40}$K peak. They do not agree for the 583 keV $^{208}$Tl peak in (b), although they agree in (a). The $^{220}$Ru exhalation rate observed by Megumi & Mamuro$^{(24)}$ and Ikebe & Shimo$^{(25)}$ ranged $10^{-15}$~$10^{-14}$ Ci·cm$^{-2}$·s$^{-1}$. The peak counting rate for the 583 keV $^{208}$Tl γ-rays estimated using these values and the air concentration profiles of $^{212}$Bi$^{(26)}$ was evaluated to be $2.4\times10^{-5}$~$1.8\times10^{-3}$ cps. The difference between the calculated and observed peak counting rates exceeds this range.

At Tsukuba, the decrease in the peak counting rate for the 609 keV γ-rays due to the diffusion loss of $^{222}$Rn under the ground was estimated from the difference between the calculations. The result showed that the decrease amounted to 44% of what was expected for the uniformly distributed $^{214}$Bi source under the ground. During the observation the peak counting rate due to the airborne $^{214}$Bi source was 3~19%.

The exposure rates calculated using the soil and air concentration profiles of naturally occurring radionuclides are given in Table 4. The calculation was carried out

### Table 3 Comparison of results for $^{214}$Bi, $^{208}$Tl and $^{40}$K by field γ-spectrometry, air sampling followed by α-counting and soil sampling followed by laboratory γ-spectrometry

(a) Without Pb shield

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Energy (keV)</th>
<th>Calculation (cps)</th>
<th>In-situ measurement$^1$ (cps)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Ground</td>
<td>Air</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>609</td>
<td>$(5.38\pm0.65)\times10^{-2}$</td>
<td>$0.99\times10^{-2}$</td>
</tr>
<tr>
<td>$^{208}$Tl</td>
<td>583</td>
<td>$(6.04\pm0.62)\times10^{-2}$</td>
<td>—</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>1,461</td>
<td>$(1.29\pm0.11)\times10^{-1}$</td>
<td>—</td>
</tr>
</tbody>
</table>

(b) With Pb shield of 5 cm thickness

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Energy (keV)</th>
<th>Calculation (cps)</th>
<th>In-situ measurement$^1$ (cps)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Ground</td>
<td>Air</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>609</td>
<td>$(0.55\pm0.07)\times10^{-2}$</td>
<td>$1.86\times10^{-2}$</td>
</tr>
<tr>
<td>$^{208}$Tl</td>
<td>583</td>
<td>$(0.66\pm0.07)\times10^{-2}$</td>
<td>—</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>1,461</td>
<td>$(1.75\pm0.15)\times10^{-2}$</td>
<td>—</td>
</tr>
</tbody>
</table>

$^1$ Oct. 13, 1979, 4: 10~6: 27, $^{17}$ Oct. 16, 1979, 4: 25~6: 42, $^{17}$ Calculated assuming uniform distribution.

### Table 4 Calculated exposure rates at 1 m above the ground, Tsukuba

<table>
<thead>
<tr>
<th>Sources</th>
<th>Exposure rate ($\mu$R·h$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ground</td>
</tr>
<tr>
<td>$^{238}$U series</td>
<td>$0.557\pm0.063$</td>
</tr>
<tr>
<td></td>
<td>min. 0.034 (9)</td>
</tr>
<tr>
<td>$^{232}$Th series</td>
<td>$0.913\pm0.103$</td>
</tr>
<tr>
<td></td>
<td>(34)</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>$1.11\pm0.10$</td>
</tr>
<tr>
<td></td>
<td>(41)</td>
</tr>
<tr>
<td></td>
<td>$0.669\pm0.044$</td>
</tr>
<tr>
<td></td>
<td>(25)</td>
</tr>
<tr>
<td></td>
<td>$2.34\pm0.13$</td>
</tr>
<tr>
<td></td>
<td>min. 0.034 (9)</td>
</tr>
<tr>
<td>Total</td>
<td>$2.69\pm0.15$</td>
</tr>
<tr>
<td></td>
<td>(100)</td>
</tr>
</tbody>
</table>

$^1$ Calculated under the assumption that $^{222}$Rn and its daughters are uniformly distributed in the soil.
using the Monte Carlo technique. A detailed description of the Monte Carlo $\gamma$ transport code system used here is given by Minato(27). The decrease in the exposure rate due to the diffusion loss of $^{222}$Rn under the ground amounts to 13% of the total exposure rate calculated assuming the uniform distribution of $^{222}$Rn in the soil. The exposure rate from the airborne $^{222}$Rn daughters amounts to 1~9%. The exposure rate from the naturally occurring radionuclides is, therefore, evaluated to be 88~96% of the total exposure rate calculated assuming uniform distribution of $^{222}$Rn in the soil.

During this observation the airborne $^{222}$Rn daughters concentration correlated closely with the peak counting rate for the 609 keV $^{214}$Bi $\gamma$-rays. A detailed discussion about the influence of airborne $^{222}$Rn daughters on the natural $\gamma$-radiation field will be presented in another paper in succession.

V. CONCLUSION

The soil and air concentration profiles of the naturally occurring radionuclides have been observed. The peak counting rates for the $\gamma$-rays of interest at 1 m above the ground were calculated using these profiles where the diffusion loss of $^{222}$Rn was involved. The calculated peak counting rates agreed well with those measured in situ. This fact justifies that the natural $\gamma$-radiation field is not a little affected by the diffusion loss of $^{222}$Rn under the ground and airborne sources at this observation site. In order to determine the concentration of the radionuclides in the soil by the field $\gamma$-spectrometry and/or the natural $\gamma$-radiation field by in-laboratory soil sample measurement exactly, it is useful to know above-mentioned points. The influence of the diffusion loss of $^{222}$Rn on the exposure rate have been studied at several selected sites, and will be presented in another paper.

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REFERENCES