Integrating Radon Progeny Monitor

Y. Ishimori, K. Ito and S. Furuta
Ningyo-toge Environmental Engineering Center, Japan Nuclear Cycle Development Institute,
1550 Kamisaibara, Tomata-gun, Okayama 708-0698 Japan

INTRODUCTION

The Japan Nuclear Cycle Development Institute (JNC) carried out research and development projects related to uranium exploration in Japan from 1967 to 1987. Several mining facilities have been retained after the end of projects. Hence, the study on the behavior and the environmental impact of radon and its progeny from the mining facilities is being continued at the Ningyo-toge Environmental Engineering Center of the JNC (JNC Ningyo-toge)1)-3).

The limitations against radon are determined with equilibrium equivalent radon concentration (EECRn) in current Japanese laws, including the Japanese Mine Safety Law. The Law requires a lower exposure dose than 1mSv/year out of the site, in excess of the natural background level. 1mSv/year is equivalent to 9 Bq/m3 of EECRn*. It is therefore necessary to determine the long-term average concentration of radon progeny. Nevertheless, the environmental impact of radon is generally estimated from radon concentration by integrating radon monitors. This means that the radon progeny concentration monitors, which are suitable for long-term average concentration of radon progeny at outdoor fields where it is impossible to use an AC power source, have not appeared. Actually, long-term environmental surveillances of radon concentration have been carried out more frequently than that of radon progeny concentration.

Radon progeny monitors are categorized into active monitors and passive monitors. In general, the response of the active monitor is more stable than the passive monitor. The equilibrium factor (F value) by grab sampling method on and around the mining sites is shown in Figure 1. The variation of the F values is vary widely. Thus, active monitors are suitable for environmental monitoring. Several different techniques using the active method, including integrating monitors4)-11), have been used to determine radon progeny concentration. Since some of them require precision systems with AC power sources, they are big and expensive. Although others are simple and use batteries, almost all of them are developed for short-term monitoring; for example, personal dosimetry or area monitoring while working.

An integrating radon progeny monitor12) developed by the JNC Ningyo-toge collects radon progeny on a filter using a pump with a battery and detects alpha particles with a CR-39 detector. However, it differs from others in that it uses interval operation for sampling and it has a function to estimate the effect of components attached by diffusion during interval. This monitor makes possible the long-term measurements of radon progeny concentration for over a month without an AC power source.

In this paper, the character of the monitor and the results of experimental measurement at outdoor fields are reported.

MATERIALS AND METHOD

Composition of a monitor

The composition of the integrating radon progeny monitor developed by the JNC Ningyo-toge is shown in Figure 2. It is composed of two parts: ‘heads’ and ‘sampler’. The head is a holder of a track-etch detector (CR-39), an absorber and a filter. The sampler has a pump, a lead accumulator, a flow meter and a control system. It is 12cm(L)×18cm(W)×12cm(H) and weighs about 5kg.

The monitor has two heads, a ‘sampling head’ and a ‘BG head’. Each head has four channels to detect the target nuclides; CH1 is for RaA (Po-218), RaC’ (Po-214), ThC (Bi-212) and ThC’ (Po-212), CH2 is for RaC’ and ThC’, CH3 is for ThC’ and CH4 is for blank, that is, testing for abnormalities. The BG head is set to remove the counts of components attached to the monitor by diffusion. In this study, etch-pits of each channel of BG head are called BG etch-pits for the corresponded channel of the sampling head. The schematic diagram of a head and an absorber is shown in Figure 3.

Usually, the monitor actuates the pump for 5 or 10 minutes an hour and collects the radon progeny in the atmosphere on the filter. The sampling period can be changed from 1 minute to 60 minutes depending on the purpose: 60 minutes sampling period means continuous sampling. The sampling flow rate is constant at about 0.5 liter/minute. The monitor can work for over a month with a lead accumulator which has a capacity of 6V-10Ah when a pump is actuated for 10 minutes an hour. This means the long-term average concentration of radon progeny is measured without an AC power source.

* All current Japanese laws for radiation protection are according to ICRP Publication 26 yet. Now they are being revised according to ICRP Publication 60. The equivalent concentration of radon progeny for 1mSv/year will be changed soon.
A CR-39 detector is etched for 24 hours at 70 °C in 6N NaOH solution. The etch-pits are counted by an automatic system using the image processing method. Radon progeny concentration is obtained as EECRn by calculating from the result with sampling volume.

**Calculation method**

Radon progeny concentration is calculated by the equation as follows when the counts of etch-pits on all channels are obtained with \( K \), \( S \), \( V \) and \( \eta \); in which \( K \) is the conversion constant which is \( 2.846 \times 10^{-5} \) in (Bq/m³)/(MeV/m³), \( S \) is the counting area in cm², \( V \) is the sampling volume in m³ and \( \eta \) is the geometrical efficiency.

\[
EECRn = K \times S(6D_1 + 1.69D_2)/V/\eta
\]

(1)

\( D_1 \) and \( D_2 \) are obtained by the equations as follows.

\[
D_1 = CH_1_{net} - 0.6406 \times CH_3_{net}
\]

(2)

\[
D_2 = CH_2_{net} - CH_3_{net}
\]

(3)

where \( CH_1_{net}, CH_2_{net} \) and \( CH_3_{net} \) means counts per unit area in cm² of the etch-pits on CH1, CH2 and CH3 from which corresponded BG etch-pits are removed, respectively. Therefore, \( D_1 \) and \( D_2 \) is counts per unit area of etch-pits by RaA+RaC’ and RaC’ only, respectively. Hence, \( (6D_1 + 1.69D_2) \) is proportional to the potential alpha energy concentration of radon progeny. It means that the counts by RaA only are not required. The equation (4) is the final equation calculated by substituting \( K \), \( S \) and \( \eta \) for the equation (1).

\[
EECRn = 1.161 \times 10^{-3}(6D_1 + 1.69D_2)/V
\]

(4)

Although radon progeny concentration is calculated theoretically in this investigation, the result was experimentally confirmed by several experiments (see Figure 4). The integrating monitor is comparable to commercialized monitors; which are the automatic monitor developed by the JNC, the WLM-200plus (Tracerlab Co. Ltd.; Germany) and the WLx (Pylon Co. Ltd.; Canada).

**Detectable limits**

Although the detectable limits in this method depend on the concentrations of RaA, RaB and RaC and the BG counts, they are estimated roughly.

\[
(CH_2_{net}/CH_1_{net})
\]

is estimated theoretically about 0.9 when \( ([RaB\ conc. + RaC\ conc.]/RaA\ conc.) > 0.5 \). BG counts are estimated experimentally at about 0.30 of counts in CH1 and about 0.34 of counts in CH2 although they have a wide range. Thus, the relative error, relative standard deviation, is calculated for each EECRn as Figure 5 when a measurement is carried out for 30 days with 5 minutes sampling an hour. The detectable range is from 0.8 to 1600 Bq/m³, when the triple of standard deviation is regarded as the detectable limit. In a similar way, the detectable range is from 0.3 to 800 Bq/m³ when a measurement is carried out for 30 days with 10 minutes sampling an hour. They are sufficient for environmental monitoring.

**MEASUREMENTS AND DISCUSSION**

Several experimental measurements were carried out at outdoor fields around uranium mining sites during the 3rd quarter of 1999. The location of the measurement points is shown in Figure 6. All of the points were chosen from the radon monitoring points of the JNC Ningyo-toge.

Two points, the JNC Ningyo-toge and the Katamo WR(B1) *, are located in sites where uranium waste rock piles exist. Three points, Katamo, Asabatake and Kawakami, are located in residential districts near the waste rock pile sites. Three points, Misasa (the Okayama univ. branch hospital), Kurayoshi (the JNC Kurayoshi dormitory) and Anagamo, are located in districts distant from the waste rock piles sites. These three points were set as the control. The monitor was used in the naturally ventilated case, the height of which is about from 1 to 1.5 meters (see Figure 7). The monitor worked for 5 minutes an hour through the sampling period. The results are shown in Table 1.

Table 2 shows the measurement results of radon concentration by electrostatic integrating passive radon monitors13) (Aloka Co. Ltd; Japan) over the same period. The radon monitor was used in the same case as the radon progeny monitor. Two cases, one for radon and one for radon progeny, were set adjacent to each other. The F value shown in Table 2 was calculated from the results of both radon progeny concentration and radon concentration.

The obtained radon concentrations were about 10 – 20 Bq/m³ except one which was about 50 Bq/m³ at the Katamo WR(B1). Therefore, the radon concentration at the Katamo WR(B1) may be affected by radon released from waste rock piles. However, the radon progeny concentrations at the Katamo WR(B1) were not remarkably high. These results are agreed with our previous investigations11-12). The radon progeny concentrations at Misasa, Kurayoshi and Anagamo were higher than those in the sites or around the sites. The F value at control areas and residential districts around the sites shown in Figure 1, except Sugegawara, ranges from 0.031 to 0.70 (average: 0.36). The observed F values at control the areas and around

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* The authors usually use “R-5” as a point name of the Katamo WR(B1) for the radon concentration measurement. The point is located on the boundary of the Katamo –1,2 waste rock piles site.
the sites in this experiments were within the range. The F values at outdoor fields in Japan were reported by other researches. They are 0.6\(^6\) and 0.76\(^14\) at Nagoya and 0.69\(^15\) at Tokyo. Since Misasa, Kurayoshi and Anagamo are not urban but has moderate traffic flows for Japan according to their population, the aerosol concentration there is higher than at other points. Thus, it is natural that the F values there were near those of big cities rather than the average F value at the control areas and around the sites shown in Figure 1. The F value at the Katamo WR(B1) in this experiments was also in good agreement with Figure 1.

Table 3 shows radon concentrations at the Katamo WR(B1) by a continuous monitor (Alpha GUARD, Genitron Co. Ltd; German). It shows also radon progeny concentrations at the Katamo WR(B1) and at the JNC Ningyo-toge by automatic monitors developed by the JNC. These results also sustained the measurement results by the integrating radon progeny monitors in Table 1.

CONCLUSION

The integrating radon progeny monitor developed by the JNC Ningyo-toge collects radon progeny on a filter using a pump with a battery and detects alpha particles by a CR-39 detector. However, it differs from others in that it uses interval operation for sampling and that it has the function of estimating the effect of components attached by diffusion during the interval. The monitor makes possible the long-term measurements of radon progeny concentration over a month without an AC power source. The monitor is suitable for environmental monitoring. Several experimental measurements were carried out at laboratories and at outdoor fields. The measurement results by the integrating monitors have agreed with our previous investigations. The monitor was originally developed for determining the long-term average concentration of radon progeny at outdoor fields. It is also acceptable for determining the radon progeny concentration in houses because it is almost noiseless during sampling.

REFERENCES

9) C.S.Dudney and A.R.Hawthorne, Radon-222, \(^{222}\text{Rn}\) Progeny, and \(^{220}\text{Rn}\) Progeny Levels in 70 Houses. Health Phys., 58, 297-311(1990)
### Table 1. Measurement results by integrating radon progeny monitors during the 3rd quarter of 1999.

<table>
<thead>
<tr>
<th>Location</th>
<th>EECRn Bq/m³</th>
<th>Oct.</th>
<th>Nov.</th>
<th>Dec.</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td>in the site</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>the JNC Ningyo-toge</td>
<td>4.6 ± 0.8</td>
<td>3.6 ± 0.7</td>
<td>4.8 ± 0.7</td>
<td>4.4 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>the Katamo WR(B1)</td>
<td>5.3 ± 1.5</td>
<td>6.9 ± 1.3</td>
<td>9.3 ± 1.1</td>
<td>7.3 ± 0.8</td>
<td></td>
</tr>
<tr>
<td>Katamo</td>
<td>3.2 ± 0.9</td>
<td>6.6 ± 1.2</td>
<td>5.6 ± 0.9</td>
<td>5.1 ± 0.6</td>
<td></td>
</tr>
<tr>
<td>around the site</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Asabatake</td>
<td>6.4 ± 0.9</td>
<td>4.4 ± 0.8</td>
<td>0.8 ± 0.5</td>
<td>0.5 ± 0.6</td>
<td></td>
</tr>
<tr>
<td>Kawakami</td>
<td>1.6 ± 0.8</td>
<td>5.9 ± 0.9</td>
<td>5.4 ± 0.9</td>
<td>5.7 ± 0.6</td>
<td></td>
</tr>
<tr>
<td>Katamo WR(B1)</td>
<td>4.6 ± 0.8</td>
<td>3.6 ± 0.7</td>
<td>4.8 ± 0.7</td>
<td>4.4 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>Asabatake</td>
<td>6.4 ± 0.9</td>
<td>4.4 ± 0.8</td>
<td>0.8 ± 0.5</td>
<td>0.5 ± 0.6</td>
<td></td>
</tr>
<tr>
<td>Kawakami</td>
<td>1.6 ± 0.8</td>
<td>5.9 ± 0.9</td>
<td>5.4 ± 0.9</td>
<td>5.7 ± 0.6</td>
<td></td>
</tr>
</tbody>
</table>

* Only 5% of data was obtained during the period because the automatic monitor was used for another experiment.

### Table 2. Radon concentration by electrostatic integrating passive monitors and calculated F value during the 3rd quarter of 1999.

<table>
<thead>
<tr>
<th>Location</th>
<th>Rn concentration Bq/m³</th>
<th>F value</th>
</tr>
</thead>
<tbody>
<tr>
<td>in the site</td>
<td></td>
<td></td>
</tr>
<tr>
<td>the JNC Ningyo-toge</td>
<td>9.9 ± 1.0</td>
<td>0.44</td>
</tr>
<tr>
<td>the Katamo WR(B1)</td>
<td>49.9 ± 4.8</td>
<td>0.15</td>
</tr>
<tr>
<td>Katamo</td>
<td>14.2 ± 1.4</td>
<td>0.36</td>
</tr>
<tr>
<td>Asabatake</td>
<td>10.8 ± 1.1</td>
<td>0.50</td>
</tr>
<tr>
<td>Kawakami</td>
<td>8.8 ± 0.9</td>
<td>0.64</td>
</tr>
<tr>
<td>around the site</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Misasa</td>
<td>19.2 ± 1.9</td>
<td>0.58</td>
</tr>
<tr>
<td>(the Okayama Univ. Branch Hospital)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kurayoshi</td>
<td>12.6 ± 1.3</td>
<td>0.78</td>
</tr>
<tr>
<td>(the JNC Kurayoshi Dormitory)</td>
<td>15.1 ± 1.5</td>
<td>0.59</td>
</tr>
<tr>
<td>Anagamo</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Table 3. Radon and radon progeny concentrations by continuous or automatic monitors during the 3rd quarter of 1999.

<table>
<thead>
<tr>
<th>Location</th>
<th>Radon concentration and EECRn Bq/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>in the site</td>
<td></td>
</tr>
<tr>
<td>the JNC Ningyo-toge</td>
<td>(EECRn) 4.1 4.7 5.6* 4.8</td>
</tr>
<tr>
<td>the Katamo WR(B1)</td>
<td>(Radon Conc.) 59.0 50.7 41.6 50.0</td>
</tr>
<tr>
<td>the Katamo WR(B1)</td>
<td>(EECRn) 7.8 6.6 5.1 6.5</td>
</tr>
</tbody>
</table>

* Only 5% of data was obtained during the period because the automatic monitor was used for another experiment.
Figure 1. Variation of F value.
Note: Asabatake-1, Asabatake-2, Asabatake-2B, Asabatake-3, Katamo-1,2, Katamo-S2, Katamo-3, Ayumidani, Kan’no kura-1, Kan’no kura-2, Nakatsugo and Choja are names of the waste rock piles sites. (B), (B1) and (B2) are the boundary of the sites. (M) is the closed mouth of a level. Sugegawara is the name of a control area. Control (Others) shows all data of control areas and residential districts around the sites except Sugegawara.

Figure 2. Composition of the integrating radon progeny monitor.
Figure 3. Schematic diagram of a head and an absorber. The head is composed of 10 parts: ① a lid of the detector holder, ② a detector (CR-39), ③ an absorber, ④ body of the head, ⑤ a Teflon ring, ⑥ a membrane filter, ⑦ a back-up filter, ⑧ a packing, ⑨ a Teflon ring and ⑩ a filter holder. ⑩ also contains the parts for setting the sampler on. The absorber is divided into 4 channels to detect target nuclides; CH1 is for RaA (Po-218), RaC" (Po-214), ThC (Bi-212) and ThC' (Po-212), CH2 is for RaC and ThC', CH3 is for ThC' and CH4 is for blank.
Figure 4. Comparison with commercialized monitors: In this graph, axes are set on different position from the usual because more than two commercialized monitors were used at the same time in some experiments.

Figure 5. Calculation of Relative Error.
Figure 6. Location of the Measurement Points.

Figure 7. Naturally ventilated case.