The Anomaly in Atmospheric Radon Concentrations Prior to the 2011 Tohoku-Oki Earthquake in Japan

Yumi Yasuoka*, Hiroyuki Nagahama2, Jun Muto2 and Takahiro Mukai1

1Kobe Pharmaceutical University, 4-19-1, Motoyamakitamachi, Higashinada-ku, Kobe-shi, Hyogo Pref. 658-8558, Japan
2Tohoku University, 6-3, Aramaki Aza-Aoba, Aoba-ku, Sendai-shi, Miyagi Pref. 980-8578, Japan

Received 31 December 2017; revised 6 April 2018; accepted 1 May 2018

This review summarizes anomalous variations in radon concentration in Fukushima, Miyagi, and Tochigi Prefectures prior to the 2011 Tohoku-Oki Earthquake. Atmospheric radon concentrations in Fukushima, Hokkaido (Sapporo), and Wakayama Prefectures were analyzed based on at least five years of raw data, whereas the data periods obtained in at Miyagi (around approximately four years of data) and Tochigi (around approximately three years of data) Prefectures before the 2011 Tohoku-Oki Earthquake were shorter than five years. The data were fitted using sinusoidal regression to describe seasonal variations in atmospheric radon concentration. In 72% of prefectures, including the Miyagi and Tochigi Prefectures, the anomalous data extracted from the normal pattern of annual radon variation could be used to identify earthquake activity. We obtain anomalous results that the radon concentrations were simultaneously reduced in the Fukushima, Miyagi, and Tochigi Prefectures before the 2011 Tohoku-Oki Earthquake by analyzing the variations in radon concentration based on the normal seasonal variations in atmospheric radon concentration approximated by the sinusoidal regression curves.

Key words: Atmospheric radon, Tohoku-Oki Earthquake, Prediction, Anomalous

1. Introduction

This review summarizes the anomalous variations in the observed radon concentrations in Japan before the occurrence of the 2011 Tohoku-Oki Earthquake. Radon (222Rn), a radioactive gas with a half-life of 3.82 days, is released from soil, rocks, and water. In the 238U decay chain, radon is produced by the radioactive decay of 226Ra. Radon is released from the ground into the atmosphere. Thus, anomalous radon concentrations in groundwater, soil, and air have been reported prior to earthquakes.

Some reviews have been published on pre-seismic anomalies in radon concentrations in groundwater and soil at the earth’s surface1-5). However, compared to studies on radon concentrations in groundwater6-9) and soil10-13), few investigations have focused on the relationship between atmospheric radon and earthquakes5, 14-17).

Previously, we reported an anomalous increase in atmospheric radon concentration at Kobe Pharmaceutical University (N34.7°, E135.3°) before the Kobe earthquake (17 January 1995; Mw 6.9, depth = 16 km; N34.6°, E135.0°) based on over a decade of data (1984–1996)18). We observed that the dynamic evolution of radon concentration is sufficiently represented by the log-periodic accelerated peaks, which suggests a pattern in the manifestation of the precursory variation19). The obtained radon data were also compared with other
precursory data collected before the earthquake\textsuperscript{20}). In another study, we reported an anomalous increase in atmospheric radon concentration at Wakayama Medical University (N34.2°, E135.2°) before and after the 2011 northern Wakayama earthquake (July 5, 2011; Mw 5.0; depth = 7 km; N34.2°, E135.2°) based on several years of data (January 2000–June 2013)\textsuperscript{21}).

In this review, the atmospheric radon concentrations in Fukushima, Hokkaido (Sapporo), and Wakayama Prefectures were analyzed based on at least five years of raw data, whereas the radon concentrations in Miyagi and Tochigi Prefectures before the 2011 Tohoku-Oki Earthquake compelled to be analyzed based on approximately four years of data and approximately three years of data, respectively. The possibility of the fitting of a sinusoidal regression curve is explored for atmospheric radon seasonal variation in Japan. Hence, using normal seasonal variation for atmospheric radon concentration analyzed by a sinusoidal regression curve, this review summarizes whether the anomalous variation in radon concentration before the 2011 Tohoku-Oki Earthquake is capable of detecting, instead of using the five years’ raw data.

### Table 1. Specifications of the three measurement instruments and the conditions of the NIRS radon chamber

<table>
<thead>
<tr>
<th>Monitor Name</th>
<th>DGM-101</th>
<th>PMT-TEL</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Specifications</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Detection limit (Bq/m$^3$)</td>
<td>0.64</td>
<td>0.93</td>
</tr>
<tr>
<td>Detector</td>
<td>Gas-flow ionization chamber</td>
<td>ZnS(Ag) scintillator</td>
</tr>
<tr>
<td>Effective volume of chamber (l)</td>
<td>14</td>
<td>18</td>
</tr>
<tr>
<td>Gas-flow rate (l/min)</td>
<td>65</td>
<td>1.0</td>
</tr>
<tr>
<td>Data-collection interval (min)</td>
<td>1</td>
<td>10</td>
</tr>
<tr>
<td><strong>Conditions of NIRS radon chamber</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Radon level (Bq/m$^3$)</td>
<td>1011 ± 72</td>
<td>2066 ± 206</td>
</tr>
<tr>
<td>Exposure period (h)</td>
<td>94</td>
<td>40</td>
</tr>
</tbody>
</table>

## 2. Measuring sites

The radon concentrations were measured using DGM-101 (Hitachi, Ltd., Japan) exhaust monitors at the following sites (Fig. 1): Sapporo Medical University (SMU; N43.05°, E141.33°) in Sapporo City, Hokkaido; Fukushima Medical University (FMU; N37.69°, E140.47°) in Fukushima City, Fukushima Prefecture; the Institute of Medical Science, the University of Tokyo (IMSUT; N35.64°, E139.72°) in Tokyo; the National Institutes of Natural Sciences, Okazaki Research Facilities, Centre for Radioisotope Facilities (NINS) in Okazaki City, Aichi Prefecture; Wakayama Medical University (WMU) in Wakayama City, Wakayama Prefecture; the Tohoku Medical and Pharmaceutical University (TMPU) in Sendai City, Miyagi Prefecture; and Dokkyo Medical University (DMU) in Mibu Town, Tochigi Prefecture.

### Fig. 1. Map of Japan showing the measuring sites and the epicenter of the 2011 Tohoku-Oki Earthquake

The epicenter of the 2011 Tohoku-Oki earthquake (Mw 9.0). The measuring sites were Sapporo Medical University (SMU) in Sapporo City, Hokkaido; Fukushima Medical University (FMU) in Fukushima City, Fukushima Prefecture; the Institute of Medical Science, the University of Tokyo (IMSUT) in Tokyo; the National Institutes of Natural Sciences, Okazaki Research Facilities, Centre for Radioisotope Facilities (NINS) in Okazaki City, Aichi Prefecture; Wakayama Medical University (WMU) in Wakayama City, Wakayama Prefecture; the Tohoku Medical and Pharmaceutical University (TMPU) in Sendai City, Miyagi Prefecture; and Dokkyo Medical University (DMU) in Mibu Town, Tochigi Prefecture.
of Radiological Sciences. The radon level in the radon chamber was monitored by an AlphaGUARD which was calibrated by the German National Metrology Institute Physikalisch-Technische Bundesanstalt (PTB), and these values were in good agreement with the PTB values. The result to this correlation is shown in Figure 2. The radon concentrations measured by the DGM-101 instrument ($C_I \text{ Bq/m}^3$) were compared to those measured by the PMT-TEL instrument ($C_P \text{ Bq/m}^3$), and the relative percent difference $V_I$ % was given by Eq.(1),

$$V_I = \frac{100 \left( C_I - C_P \right)}{C_P}.$$  

Using the simultaneously measured $C_P$ Bq/m$^3$ and $C_I$ Bq/m$^3$, the average value and standard deviation ($\bar{V}_I \pm \sigma_I$) % were calculated. The 95% prediction interval (95% PI) was also calculated using Eqs. (2) and (3),

$$\bar{V}_I - \gamma \sigma_I \leq 95\% \text{PI} \leq \bar{V}_I + \gamma \sigma_I,$$

$$\gamma = k \sqrt{\left(1 + \frac{1}{n}\right)},$$

where the number of data points $n = 336$, and $k$ indicates the student’s $t$ value at a significance of 0.05 for a two-tailed $t$-test determined based on the degree of freedom ($n - 1$). The $\gamma$ value in Eq. (3) was 1.97 in the case of $-25\% < 95\% \text{ PI} < 25\%$, and the individual percent difference was used to meet the efficiency criteria. We found that $C_I$ was consistent with $C_P$ because it successfully met the efficiency criteria (Figs. 3a and 3b). The PMT-TEL and DGM-101 instruments on hourly data were sufficient to measure the indoor radon concentration, which ranged from 28 to 89 Bq/m$^3$ with an average of 53 Bq/m$^3$.

3.2. Variation in atmospheric radon concentration measured by an exhaust monitor

Here, we examine whether the variation in DGM-101 data can be attributed to the variation in outdoor radon concentration. Two DGM-101 monitors were placed at the air intake and at the terminal exhaust duct of the Radioisotope Institute. The results showed that the radon concentration in the exhaust was the same as that in the air intake (Figs. 4a and 4b), suggesting that variations in outdoor radon concentration can be captured using an exhaust monitor.

The linear regression of mean for the radon concentration (mean line) is given by a linear function of the time series (Fig. 4 in Ref. 21). Furthermore, the time series of the residual radon concentration ($R_i$) was determined by subtracting the mean line from the original data. We divided $R_i$ into two periods: the normal and precursor periods, as depicted in Figure 5 and Table 2. Using the $R_i$ of composite year during the normal period, seasonal variation $S_i$ was determined. The sinusoidal regression curve of $S_i$ is referred to as the seasonal model variation $S_{mi}$. Subsequently, the time series of radon variation $Rn$ (or $Rm$) were determined from the detrended levels by subtracting $S_i$ (or $S_{mi}$) from $R_i$.

4. Normal seasonal variations in atmospheric radon concentration: a sinusoidal model

Anomalous atmospheric radon concentrations measured with exhaust monitors have been reported before earthquake activity. To identify anomalous variations in
a precursor period before an earthquake, it is necessary to take careful measurements of atmospheric radon concentration during a normal period. While considering the normal seasonal variations for atmospheric radon concentrations analyzed by the sinusoidal regression curve instead of the raw data obtained from the five years during the normal period, we investigated whether the anomalous variation in radon concentration before the 2011 Tohoku-Oki Earthquake was capable of being detected.

First, we used the hourly DGM-101 data obtained at FMU (Fig. 1 and Table 2) to analyze the daily minimum atmospheric radon concentration. It has been reported that minimum radon concentration (daily minimum value of radon concentration) is less affected by meteorological and geographical conditions. Papastefanou et al. reported that the difference in the daily minimum radon concentrations at both the locations (the distance is about 5 km) was observed to be small when the radon concentration within a river valley area was compared to that over a hillside area. Therefore the daily minimum radon concentration is affected by the average variation of radon released into air from the large area surrounding the monitoring station.

The residual radon concentration \( R_t \) is indicated by the black lines in Figure 5a and 5b. We obtained five years of “normal” radon concentration data (2003–2007) and compared them with data from the earthquake precursor period (2008 to 11 March 2011, when the 2011 Tohoku-Oki Earthquake occurred).

The seasonal variation \( S_t \) denoted by the blue dotted line in Figure 5a, which was calculated using the data obtained during the normal period, was influenced by the atmospheric turbulence and onshore-offshore pattern of the Asian monsoons. We established a model for seasonal variation by fitting a sinusoidal regression curve to the normal radon concentration data. In this model, the residual radon concentration \( f(t) \) is given by

\[
f(t) = a \sin \left( \frac{2\pi}{365} (t + \phi) \right),
\]

where \( t \) day is the time elapsed after the start of observation (\( t = 0 \) corresponds to 1 January), \( a \) Bq/m\(^3\) is the amplitude, \( \phi \) day is the phase shift, and \( 2\pi \) radians is equivalent to 365 day. For the \( S_m \) predicted using Eq. (4), \( a = 2.1 \) Bq/m\(^3\), \( \phi = 72 \) days, and the coefficient of determination \( R^2 = 0.88 \). It was possible to apply a sinusoidal regression curve with \( \phi = 70 \) days (red dotted line in Fig. 5b) to the seasonal data variation.
The radon variation ($R_n$) was obtained by subtracting $S_i$ (blue dotted line in Fig. 5a) from $R_i$ (black line in Fig. 5a), as depicted in Figure 6a. Furthermore, the radon variation ($R_m$), which is depicted in Figure 6b, was obtained by subtracting $S_{mi}$ (red dotted line in Fig. 5b) from $R_i$ (black line in Fig. 5b). The standard deviations of $R_n$ and $R_m$ for the normal period were calculated using $R_n$ and $R_m$ for the normal period, and a datum was determined to be anomalous if it exceeded three times the standard deviation ($\pm 3\sigma$). Figure 6 shows $R_n$ and $R_m \pm 3\sigma$ over the observation period. The curves of $R_n$ and $R_m$ are similar, and three anomalous peaks during the precursor period can be observed in each curve before the 2011 Tohoku-Oki Earthquake (indicated by downward-facing arrows in Fig. 6).

5. Annual variation in atmospheric radon concentration in Japan

The extraction of anomalous variations in radon concentration attributed to seismic activity would be greatly assisted by a complete understanding of the normal pattern of variation in radon concentrations. When $S_{mi}$ with $\phi \approx 70$ day (red dotted line in Fig. 5b) at FMU is close to the inverse correlation of the annual variation pattern of the surface temperature (the phase shift of the
sinusoidal regression curve of the surface temperature was approximately \((70 + 365/2)\) days (see Fig. 1 in Ref. 26)), it is reasonable to assume that the simple variation in the minimum radon concentration is matched by the atmospheric turbulence and the onshore-offshore pattern of Asian monsoons.

To describe seasonal variations in atmospheric radon concentration in Japan, we fit the data using a sinusoidal regression curve\(^{25}\). Using exhaust-monitoring hourly data from five sites (SMU, FMU, IMSUT, NINS, and WMU; Fig. 1 and Table 2) measured with DGM-101 instruments, the \(S_m\) were shown to be similar to this simple variation in Figure 7a, and the variations were considered to be mainly affected by the atmospheric turbulence and the onshore-offshore pattern of Asian monsoons. Moreover, we demonstrated that the data from the Japan Chemical Analysis Center (JCAC data) report can be used to estimate annual variations in radon concentration using sinusoidal regression (Fig. 7b)\(^{27}\). A solid-state nuclear track detector (three-month integrated values) was used to obtain the JCAC data; this method was different from the method used to obtain the above data from the continuous exhaust-monitoring system.

In the case of \(60 \leq \phi \leq 122\) days (see in Fig 7a), the variation in radon concentration is high in winter and low in summer. As shown in Figure 7c, when \(60 < \phi < 122\) days and \(0.88 < R\) for the sinusoidal regression of the JCAC data, the annual variation is primarily attributed to atmospheric turbulence and the onshore-offshore pattern of Asian monsoons. Importantly, the data for 72% of the Japanese prefectures included in the JCAC report (34 out of 47 prefectures) meet these requirements. However, we observe that the radon concentrations during winter do not increase by preventing the radon release using snow when the winter air mass reaches the north of the sites and ground is covered with snow; this occurs in 28% of the Japanese prefectures, which have north sides facing the sea. Thus, earthquake activity can be identified from anomalous radon concentration data in 72% of the prefectures, and these prefectures are suitable areas for obtaining earthquake-related radon variations.

6. Anomalous variations in atmospheric radon concentration prior to the 2011 Tohoku-Oki Earthquake in Japan

Ozawa et al.\(^{28}\) reported spatial patterns of post-seismic crustal deformation caused by earthquakes of the Pacific Ocean off the coast of Fukushima and Ibaraki Prefectures in 2008 along with an earthquake off the coast of
Fukushima Prefecture in 2010 (Fig. 2 in Ref. 28).

Using the daily minimum radon concentration data collected at FMU, TMPU, and DMU (Fig. 4 and Table 2), the $R_i$ curves of FMU (Fig. 8a is the same as Fig. 5b), TMPU (Fig. 8b) and DMU (Fig. 8c) were obtained prior to the 2011 Tohoku-Oki Earthquake ($Mw.9.0$). The normal periods of atmospheric radon concentration at TMPU and DMU were shorter than five years (Table 2). When using normal seasonal variation for atmospheric radon concentration estimated by a sinusoidal regression curve (even if the data spanned less than five years), we examined whether anomalous variations in radon concentration occurred in Miyagi and Tochigi Prefectures before the 2011 Tohoku-Oki Earthquake. When the $Rm$ variations of FMU (Fig. 8a is the same as Fig. 5b), TMPU (Fig. 8b) and DMU (Fig. 8c) could be determined using the sinusoidal regression curve with a phase shift of 72 days, it was possible to use data that was obtained. The $Rm$ curves of FMU (Fig. 8d is the same as Fig. 6b), TMPU (Fig. 8e), and DMU (Fig. 8f) revealed outliers and sharp simultaneous reducing over time in radon concentration greater than $\pm 3\sigma$ before the 2011 Tohoku-Oki Earthquake in comparison to the variation in $Rm$ during the normal period at each site.

Fig. 8. Time series of residual radon concentrations $R_i$ (normal period, gray line; precursor period, black line), model of seasonal variation ($Sm_i$: red dotted line), and variation in radon concentration $Rm$ (normal period, gray line; precursor period, black line). (a) $R_i$ and $Sm_i$ at FMU; (b) $R_i$ and $Sm_i$ at TMPU; (c) $R_i$ and $Sm_i$ at DMU; (d) $Rm$ at FMU; (e) $Rm$ at TMPU; and (f) $Rm$ at DMU. The vertical black line indicates the date of the 2011 Tohoku-Oki Earthquake occurred in Japan. Modified from Ref. 29 and reprinted with permission from the authors, who retain the copyright.
7. Conclusion

When atmospheric radon concentration was analyzed, we used at least five years of raw data. However, the data periods obtained in at Miyagi (around approximately four years of data) and Tochigi (around approximately three years of data) Prefectures before the 2011 Tohoku-Oki Earthquake were shorter than five years. Fitting with sinusoidal regression curves was explored to describe the seasonal variations in atmospheric radon concentration in Japan. A normal pattern of annual radon variation with a phase shift of approximately 70 days was found in 72% of prefectures. Sinusoidal regression was applied to the data from Fukushima, Miyagi, and Tochigi Prefectures, and we constructed three models of seasonal variation in atmospheric radon concentration. Finally, the time series of radon variations were determined from the detrended levels by subtracting the seasonal variations from the raw data. We found the simultaneous reducing over time in radon concentration before the 2011 Tohoku-Oki Earthquake was capable of being detected. The results highlight the possible link between anomalous changes in radon concentration and seismic deformation. The analysis of data obtained from exhaust monitors at radioisotope institutes throughout Japan is currently under way.

Conflict of Interest Disclosure

The authors declare that they have no conflict of interest.

References


