

Review

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

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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 surface^{1–5}. However, compared to studies on radon concentrations in groundwater^{6–9} and soil^{10–13}, few investigations have focused on the relationship between atmospheric radon and earthquakes^{5,14–17}.

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

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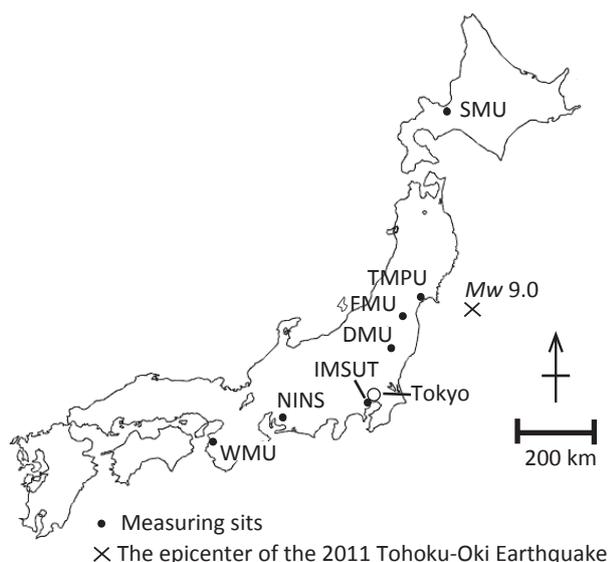


Fig. 1. Map of Japan showing the measuring sites and the epicenter of the 2011 Tohoku-Oki earthquake (M_w 9.0). The measuring sites were Sapporo Medical University (SMU) in Sapporo City, Hokkaido; Fukushima Medical University (FMU) in Fukushima City, Fukushima Prefecture; the Institute of Medical Science, 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 (WPMU) 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.

precursory data collected before the earthquake²⁰. In another study, we reported an anomalous increase in atmospheric radon concentration at Wakayama Medical University ($N34.2^\circ$, $E135.2^\circ$) before and after the 2011 northern Wakayama earthquake (July 5, 2011; M_w 5.0; depth = 7 km; $N34.2^\circ$, $E135.2^\circ$) based on several years of data (January 2000–June 2013)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

Monitor Name	DGM-101	PMT-TEL
Specifications		
Detection limit (Bq/m^3)	0.64	0.93
Detector	Gas-flow ionization chamber	ZnS(Ag) scintillator
Effective volume of chamber (l)	14	18
Gas-flow rate (l/min)	6.5	1.0
Data-collection interval (min)	1	10
Conditions of NIRS radon chamber		
Radon level (Bq/m^3)	1011 ± 72	2066 ± 206
Exposure period (h)	94	40

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; $N43.05^\circ$, $E141.33^\circ$) in Sapporo City, Hokkaido; Fukushima Medical University (FMU; $N37.69^\circ$, $E140.47^\circ$) in Fukushima City, Fukushima Prefecture; the Institute of Medical Science, the University of Tokyo (IMSUT; $N35.64^\circ$, $E139.72^\circ$) in Tokyo; the National Institutes of Natural Sciences, Okazaki Research Facilities, Centre for Radioisotope Facilities (NINS; $N34.95^\circ$, $E137.17^\circ$) in Okazaki City, Aichi Prefecture; Wakayama Medical University (WPMU; $N34.18^\circ$, $E135.18^\circ$) in Wakayama City, Wakayama Prefecture; the Tohoku Medical and Pharmaceutical University (TMPU; $N38.28^\circ$, $E140.89^\circ$) in Sendai City, Miyagi Prefecture; and Dokkyo Medical University (DMU; $N36.47^\circ$, $E139.82^\circ$) in Mibu Town, Tochigi Prefecture. The data at three sites (FMU, TMPU, and DMU) were analyzed with respect to the 2011 Tohoku-Oki Earthquake in Japan (M_w 9.0; depth = 24 km; $N38.10^\circ$, $E142.86^\circ$; black cross mark in Fig. 1).

3. Radon monitoring

3.1. Measurement of radon concentration with an exhaust monitor

We reported the atmospheric radon concentrations measured by an exhaust monitor²² originally used to detect any leakage of the radioisotope from the radiation facilities. A popular exhaust monitor in Japan is the DGM-101 instrument, which contains one gas-flow ionization chamber (Table 1).

Indoor radon concentrations were simultaneously measured using the DGM-101 instrument and an atmospheric radon monitor (PMT-TEL, Pylon Electronics Inc., Canada; Table 1) to determine whether a measuring time of 1 h is sufficient. Both instruments were calibrated in a radon chamber at the National Institute

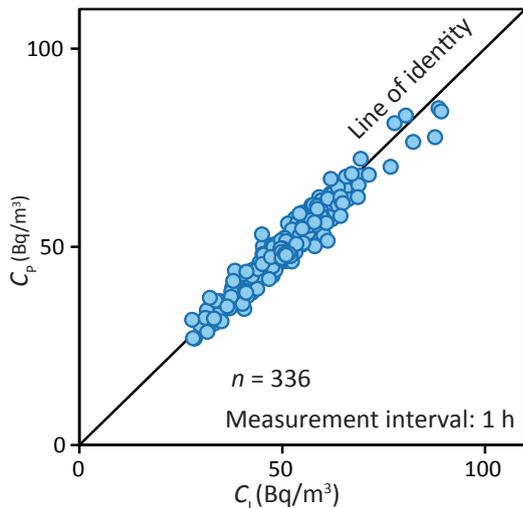


Fig. 2. Comparison of radon concentrations obtained by PMT-TEL (C_P Bq/m³) and DGM-101 (C_I Bq/m³) instruments. The figure has been modified from Ref. 22, which is available under the Creative Commons license from Oxford University Press.

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 Bq/m³) were compared to those measured by the PMT-TEL instrument (C_P Bq/m³), and the relative percent difference V_1 % was given by Eq.(1),

$$V_1 = \frac{100 (C_I - C_P)}{C_P}. \quad (1)$$

Using the simultaneously measured C_P Bq/m³ and C_I Bq/m³, the average value and standard deviation ($\bar{V}_1 + \sigma_1$) % were calculated. The 95% prediction interval (95% PI) was also calculated using Eqs. (2) and (3),

$$\bar{V}_1 - \gamma \sigma_1 \leq 95\% \text{PI} \leq \bar{V}_1 + \gamma \sigma_1, \quad (2)$$

$$\gamma = k \sqrt{\left(1 + \frac{1}{n}\right)}, \quad (3)$$

where the number of data points n was 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 γ 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/m³ with an

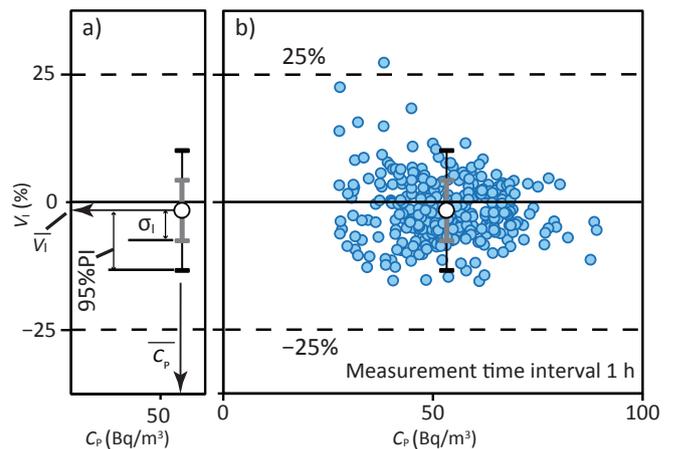


Fig. 3. Evaluation of 95% prediction interval (95% PI) of \bar{V}_1 based on integrated hourly data. Relationship between relative percent difference V_1 % and the reference radon concentration (C_P Bq/m³). (a) Evaluation of 95% PI; and (b) The 95% PI with V_1 . The figure has been modified from Ref. 22, which is available under the Creative Commons license from Oxford University Press.

average of 53 Bq/m³.

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 concentration²³. 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 Sm_i . Subsequently, the time series of radon variation Rn (or Rm) were determined from the detrended levels by subtracting S_i (or Sm_i) 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

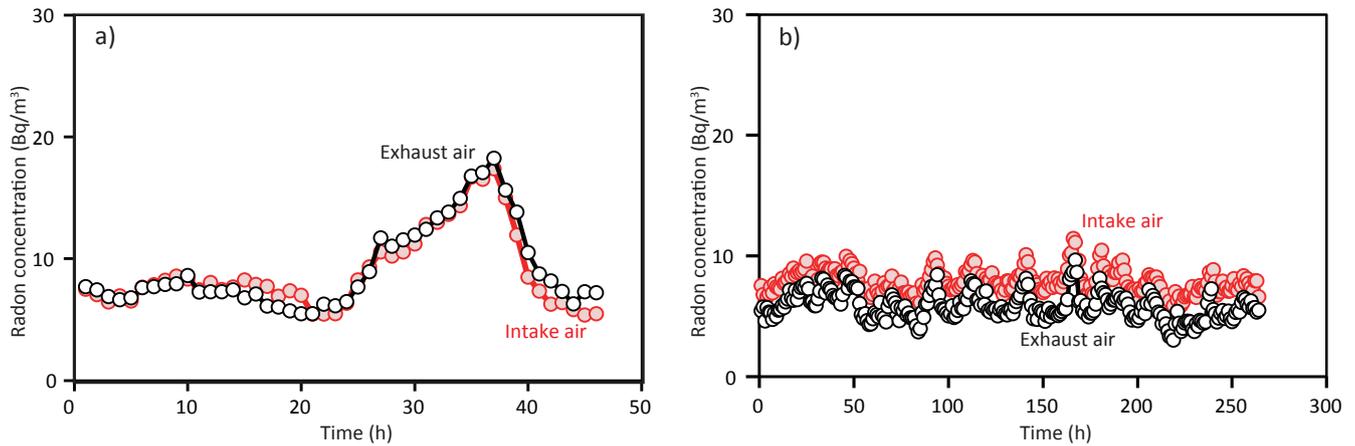


Fig. 4. Comparison of the directly measured radon concentration in outdoor air (solid red circles and red line) and the concentration in exhaust air (black open circles and black line) obtained using two gas-flow ionization chambers (DGM-101) at different rates of air flow. (a) 25.6 h^{-1} (very high flow rate); and (b) 4.9 h^{-1} . Adapted from Ref. 23 and reprinted with permission from Springer International Publishing AG.

Table 2. Locations of monitoring sites

Monitoring Site ^{a)}	Longitude, Latitude	Measurement Period (Normal Period)
SMU	N43.05°, E141.33°	2000–2009 (2005–2009)
TMPU	N38.28°, E140.89°	1 February 2007– 11 March 2011 (1 February 2007– 31 January 2010)
FMU	N37.69°, E140.47°	2003–11 March 2011 (2003–2007)
DKU	N36.47°, E139.82°	1 May 2008–11 March 2011 (1 May 2008–30 April 2010)
IMSUT	N37.69°, E139.72°	2005–2010
NINS	N35.63°, E137.17°	2003–2011
WMU	N34.18°, E135.18°	2000–2010

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 detected²⁴⁾.

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.*¹⁰⁾ 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^{5, 26)}.

The residual radon concentration (R_i) 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_i) 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

$$f(t) = a \sin \left\{ \frac{2\pi}{365} (t + \phi) \right\}, \quad (4)$$

where t day is the time elapsed after the start of observation ($t = 0$ corresponds to 1 January), a Bq/m³ is the amplitude, ϕ day is the phase shift, and 2π radians is equivalent to 365 day. For the S_{mi} predicted using Eq. (4), $a = 2.1 \text{ Bq/m}^3$, $\phi = 72$ days, and the coefficient of determination $R^2 = 0.88$. It was possible to apply a sinusoidal regression curve with $\phi \approx 70$ days (red dotted line in Fig. 5b) to the seasonal data variation.

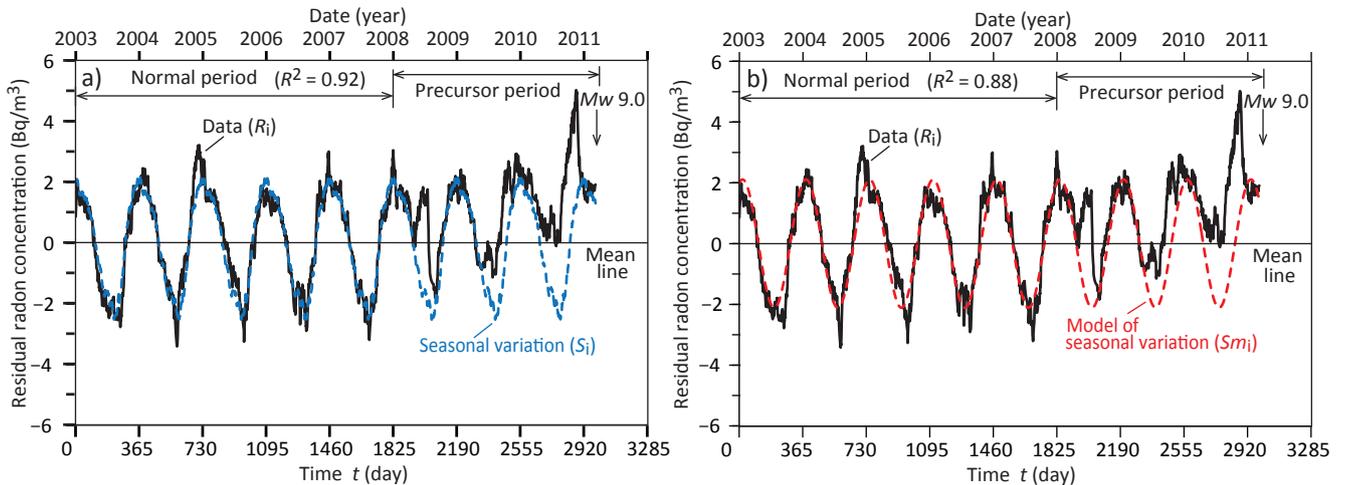


Fig. 5. Time series of residual radon concentration (R_i ; black line). The time at the start of observation (1 January 2003) was set as $t = 0$. (a) Comparison between R_i and seasonal variation (S_i ; blue dotted line); and (b) Comparison between R_i and the model of seasonal variation (S_{mi} ; red dotted line). R^2 indicates the coefficient of determination between R_i and S_i (or S_{mi}) during the normal period. Adapted from Ref. 24 and reprinted with permission from Elsevier B.V.

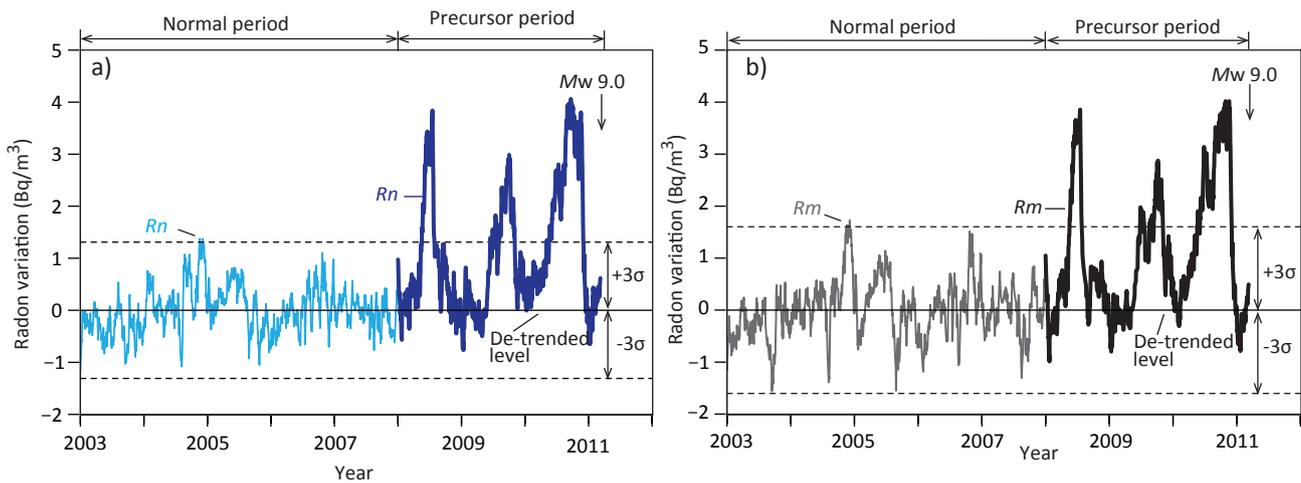


Fig. 6. Time series of radon concentrations from the de-trended levels. The downward arrow indicates the date of the 2011 Tohoku-Oki Earthquake in Japan. (a) R_n (normal period, light blue line; precursor period, dark blue line), which was obtained by subtracting S_i from R_i (Fig. 5a); and (b) R_m (normal period, gray line; precursor period, black line), which was obtained by subtracting S_{mi} from R_i (Fig. 5b). Adapted from Ref. 24 and reprinted with permission from Elsevier B.V.

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

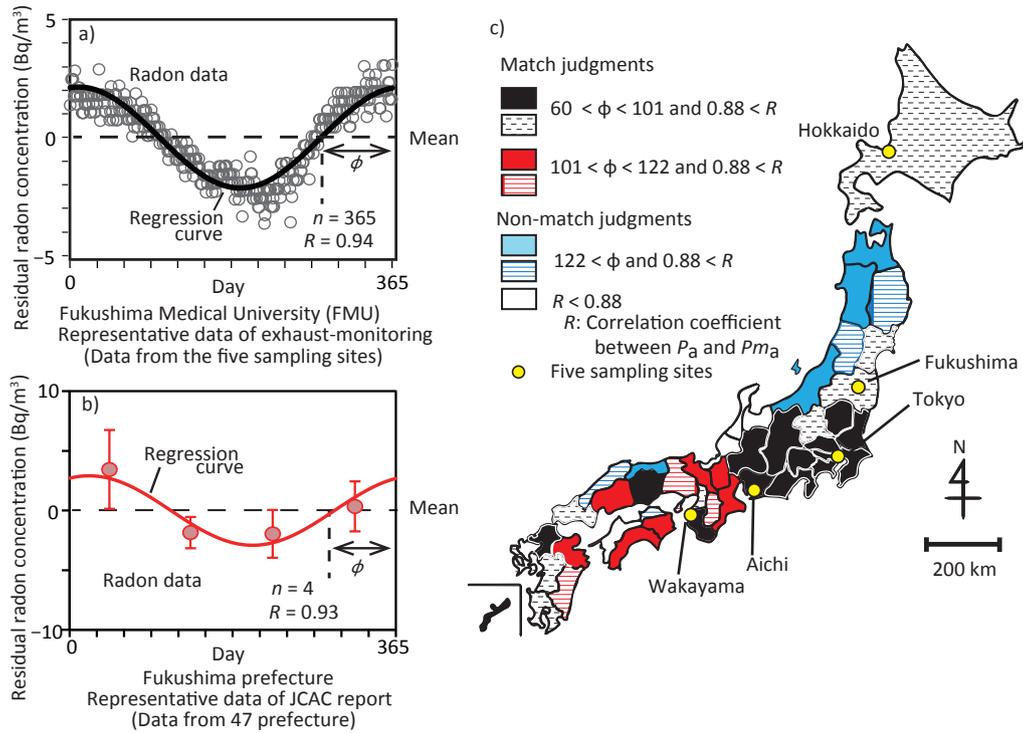


Fig. 7. Comparison of sinusoidal regression curves based on (a) exhaust data ($n = 365$ data/y) at FMU and (b) JACAC data ($n =$ four data/y) in Fukushima Prefecture. Error bars represent standard deviation. (c) Distribution of the phase shift (ϕ) values of the sinusoidal regression curve based on the JACAC data. Adapted from Ref. 25 and reprinted with permission from Elsevier B.V.

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 curve²⁵⁾. 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 Sm_i 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 (JACAC 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 JACAC data; this method was different from the method used to obtain the above data from the continuous exhaust-monitoring system.

In the case of $60 \text{ days} \leq \phi \leq 122 \text{ days}$ (see in Fig 7a), the variation in radon concentration is high in winter and low

in summer. As shown in Figure 7c, when $60 \text{ days} < \phi < 122 \text{ days}$ and $0.88 < R$ for the sinusoidal regression of the JACAC 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 JACAC 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

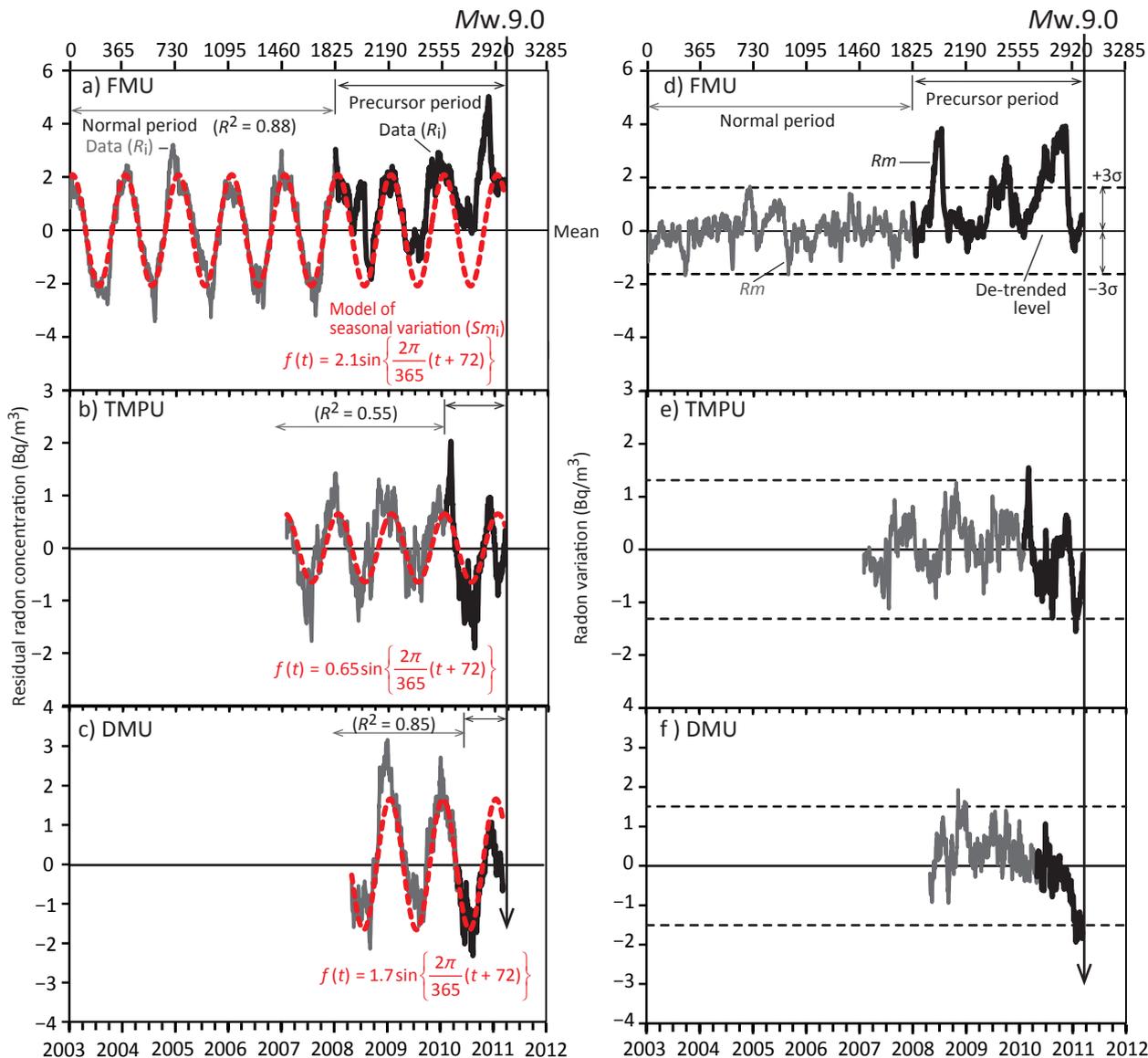


Fig. 8. Time series of residual radon concentrations R_i (normal period, gray line; precursor period, black line), model of seasonal variation (S_{m_i} ; red dotted line), and variation in radon concentration R_m (normal period, gray line; precursor period, black line). (a) R_i and S_{m_i} at FMU; (b) R_i and S_{m_i} at TMPU; (c) R_i and S_{m_i} at DMU; (d) R_m at FMU; (e) R_m at TMPU; and (f) R_m 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.

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 (M_w 9.0)²⁹. 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 S_{m_i} 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, which phase shift was obtained in FMU, it was possible to use data that was obtained. The R_m 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 R_m during the normal period at each site.

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.

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