

## TEMPORAL VARIATION OF RADON PROGENY RATIO IN OUTDOOR AIR

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**Abstract**—This paper investigates the concentration ratio of radon progeny ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ ) in outdoor air. From 1988 to 1998, alpha spectroscopy was used intermittently to collect progeny data outside the author's office located in the northeastern part of Japan. Let the quantity  $R_{EEC}$  represent the ratio  $EEC(^{218}\text{Po})/[EEC(^{214}\text{Pb}) + EEC(^{214}\text{Bi})]$ , where, e.g.,  $EEC(^{218}\text{Po})$  denotes the equilibrium equivalent radon concentration contributed by  $^{218}\text{Po}$ . The concentration ratio  $R_{EEC}$  correlates with outdoor air stability. Statistical and time series analyses of  $R_{EEC}$  indicate (1) outdoor air is more stable in the morning than in the afternoon, (2) outdoor air is more stable in summer/autumn than in winter/spring, and (3) in spite of no significant correlation between  $R_{EEC}$  and wind speed, the power spectrum of  $R_{EEC}$  appears similar to that of wind speed. This is probably due to the fact that near-surface mixing is not very sensitive to wind speed.

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**Key words:** radon progeny; radioactivity, airborne; air sampling; statistics

### INTRODUCTION

THIS PAPER concerns an investigation of the concentration ratio of radon progeny ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ ) in outdoor air. Over a period of ten years (1988–1998), the author has used alpha spectroscopy to collect progeny data intermittently outside his office located in northeastern Japan. A preliminary report of this work has already been published (Kobayashi and Takaku 1997). Clarification of the behavior of radon progeny is an issue of wide importance to radiation protection, earthquake prediction, etc. Although there have been many studies referring to radon progeny in air (Budnitz 1974; George 1996), most of these represent progeny as potential alpha energy concentration,  $PAEC$ , or equilibrium equivalent concentration of radon,  $EEC_{Rn}$ . Only a few reports deal

with progeny concentration ratios, such as the ratio of the concentration of  $^{214}\text{Pb}$  to that of  $^{218}\text{Po}$ .

This paper investigates temporal variations of the radon progeny ratio in outdoor air. In most cases, progeny ratios are discussed in terms of concentration ratios,  $R_1 = C_B/C_A$ , and  $R_2 = C_C/C_A$ , where  $C_A$ ,  $C_B$ , and  $C_C$  denote the concentrations of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ , respectively. However, in reality,  $C_A$  varies very rapidly, while  $R_1$  and  $R_2$  often reveal values greater than unity. A less variable ratio is  $R_{EEC}$ . Let  $R_{EEC} = EEC(^{218}\text{Po})/[EEC(^{214}\text{Pb}) + EEC(^{214}\text{Bi})]$  where  $EEC(^{218}\text{Po})$  denotes the equilibrium equivalent radon concentration contributed by  $^{218}\text{Po}$ , etc. From ICRP 65 (ICRP 1993), the equivalent radon concentration is  $EEC_{Rn} = (0.104C_A + 0.515C_B + 0.382C_C)$  ( $\text{Bq m}^{-3}$ ). Thus if  $C_A$ ,  $C_B$ , and  $C_C$  are in units of  $\text{Bq m}^{-3}$ ,  $EEC(^{218}\text{Po}) = 0.104C_A$ , and similarly,  $EEC(^{214}\text{Pb}) = 0.515C_B$ , and  $EEC(^{214}\text{Bi}) = 0.382C_C$ .

The concentration ratio  $R_{EEC}$  correlates with the outdoor air stability. Under equilibrium conditions (i.e., the most stable air),  $C_A : C_B : C_C = 1 : 1 : 1$  and  $R_{EEC}$  has a value of 0.116. As the air becomes unstable (because of increased vertical mixing, etc.), e.g.,  $C_A : C_B : C_C = 1 : 0.6 : 0.4$ ,  $R_{EEC}$  takes on larger values (0.225 in this case). The aim of using  $R_{EEC}$  is (1) to make use of existing progeny monitoring products that measure  $R_{EEC}$ , and (2) to observe the variation of concentration ratios from the more stable criteria of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ .

Statistical and time series analyses of  $R_{EEC}$  indicate several interesting results. These are (1) outdoor air is more stable in the morning than in the afternoon, (2) outdoor air is more stable in summer/autumn than in winter/spring, and (3) in spite of no significant correlation between  $R_{EEC}$  and wind speed, the power spectrum of  $R_{EEC}$  appears similar to that of wind speed.

### MATERIALS AND METHODS

Intermittent measurements of  $^{222}\text{Rn}$  progeny employed membrane filters (Advantec A080A055C, pore size:  $0.8 \mu\text{m}$ ) and a self-contained alpha spectrometer (ORTEC 676) with a silicon surface-barrier semiconductor (active area:  $900 \text{ mm}^2$ , thickness of depletion layer:

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100  $\mu\text{m}$ ) (Martz et al. 1969; Kobayashi and Takaku 1997). The counting efficiency for alpha particles was 0.125. After 10-min sampling ( $20 \text{ L min}^{-1}$ ) of the progeny in air, 2 min were required to set the filter to the detector and evacuate the counting chamber, and the alpha activity of the filter was counted from 2 to 12 min and from 12 to 42 min after the end of sampling.

The alpha spectrum for progeny measurements consisted of three regions:  $^{218}\text{Po}$  region (4.6 to 6.2 MeV),  $^{214}\text{Po}$  region (6.2 to 7.9 MeV), and  $^{212}\text{Po}$  region (7.9 to 9.3 MeV). By the use of an evacuated chamber, interference between these three regions was negligible, especially because of very few counts in the  $^{212}\text{Po}$  region.  $t$  test for the affect of  $^{220}\text{Rn}$  progeny (Martz et al. 1990) indicated that the affect of  $^{212}\text{Bi}$  and  $^{212}\text{Po}$  was negligible. Filter decay data provided the concentrations of radon progeny,  $^{218}\text{Po}$  (RaA),  $^{214}\text{Pb}$  (RaB), and  $^{214}\text{Bi}$  (RaC).  $EEC(^{218}\text{Po})$ , etc., were calculated from these three concentrations. The lowest detection limit for overall  $EEC_{\text{Rn}}$  was  $0.07 \text{ Bq m}^{-3}$ .

The data consist of three Series 1–3 corresponding, respectively, to sampling times of 9:15 (1988–04–04 to 1993–07–09), 15:00 (1993–07–19 to 1997–04–30), and 9:00 (1997–05–01 to 1999–01–28). Measurements at identical times are indispensable for time series analysis. Series 1 and 2 compare morning and afternoon air. Series 3 was conducted with progeny data and detailed meteorological data. Progeny data in Series 3 indicated essentially the same results as Series 1. Measurements were performed every day except holidays. The outdoor air sampler was placed on the fourth floor of an emergency staircase of a five-story college building built in 1988.

## RESULTS AND DISCUSSION

### Basic statistics

Statistical analyses were made using SPSS10.0J (SPSS Inc. 1999). Table 1 shows basic statistics for Series 1 and Series 2. Table 1 also lists conventional concentration ratios,  $R_1 = C_B/C_A$ , and  $R_2 = C_C/C_A$ , where  $C_A$ ,  $C_B$ , and  $C_C$  denote the concentrations of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Bi}$ , respectively. Several data of  $C_A$ ,  $C_B$ , and  $C_C$  were set equal to zero because of too small value to

measure. Calculation of  $EEC_{\text{Rn}}$  is possible when there is at least one of non-zero value in  $C_A$ ,  $C_B$ , and  $C_C$ , whereas  $C_A = 0$  gives missing values in  $R_{EEC}$ . Similarly, the case where either  $C_A = 0$  or  $C_B = 0$  gives missing values in  $R_1$ , and either  $C_A = 0$  or  $C_C = 0$  gives missing values in  $R_2$ . In Series 1, there were 11 cases where  $C_A = 0$  resulting in 1,236  $R_{EEC}$  values, 26 cases of either  $C_A = 0$  or  $C_B = 0$  resulted in 1,221  $R_1$  values, and 23 cases of  $C_A = 0$  or  $C_C = 0$  resulted in 1,224  $R_2$  values. In the same manner, the different numbers of data points in Series 2 happened. In Table 1,  $R_1$  and  $R_2$  often show unreasonable values of greater than unity.

Upon comparing the results of Series 1 and Series 2, it was found that the mean value of  $R_{EEC}$  was lower in the morning, thus indicating that the outdoor air was more stable in the morning than in the afternoon.

### Log-normality of the statistical distribution

Figs. 1 and 2 show statistical distributions for Series 1 (morning) and Series 2 (afternoon), respectively. A log-normal distribution is one of the well-known characteristics of radon progeny; Figs. 1 and 2 appear to indicate log-normality. To examine log-normality, the author performed the Lilliefors test (modified version of the Kolmogorov-Smirnov test) for the logarithm of the data for each fiscal year. Table 2 shows the results where “Year 1988” means data from April 1988 to March 1999 and encircled data passed the Lilliefors test (significance level exceeded 0.01).  $R_{EEC}$  passed the test 5 times for 9 years, while radon progeny passed 8 times.

### Seasonal variation

To observe the seasonal variation, one-way layout analysis was made for four sets of data, i.e., spring (March to May), summer (June to August), autumn (September to November), and winter (December to February). Table 3 and Fig. 3 show mean values of the four groups. In Fig. 3, two asterisks (\*\*) and one asterisk (\*) denote  $p$ -values of 0.01 and 0.05, respectively.  $R_{EEC}$  indicated a significantly higher level in spring and winter, i.e., the air was stable in summer and autumn.

**Table 1.** Basic statistics of radon progeny and ratios in outdoor air for Series 1 (progeny was sampled at 9:15) and Series 2 (progeny was sampled at 15:00). Arithmetic mean (AM), arithmetic standard deviation (ASD), geometric mean (GM), geometric standard deviation (GSD), minimum (Min.), maximum (Max.), and the number of data (#) are listed.

Series	Series 1							Series 2							
	1	AM	ASD	GM	GSD	Min.	Max.	#	2	AM	ASD	GM	GSD	Min.	Max.
$EEC_{\text{Rn}}$	3.35	2.20	2.78	1.85	0.37	14.88	1,247	$EEC_{\text{Rn}}$	2.16	0.98	1.95	1.61	.17	7.67	787
$R_1$	0.83	0.69	0.63	2.12	0.03	5.53	1,221	$R_1$	0.83	0.88	0.58	2.39	.01	7.64	748
$R_2$	0.81	0.64	0.65	2.00	0.02	7.07	1,224	$R_2$	0.81	0.73	0.61	2.16	.01	7.44	755
$R_{EEC}$	0.20	0.12	0.17	1.77	0.03	1.27	1,236	$R_{EEC}$	0.22	0.16	0.19	1.91	.02	1.95	777

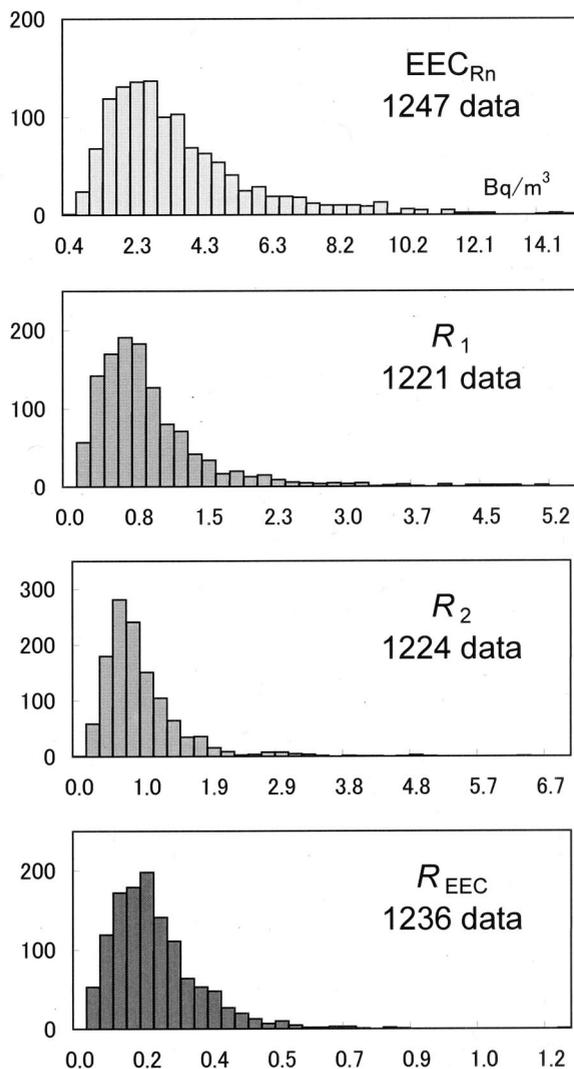


Fig. 1. Statistical distribution of radon progeny and ratios in outdoor air for Series 1 (progeny were sampled at 9:15).

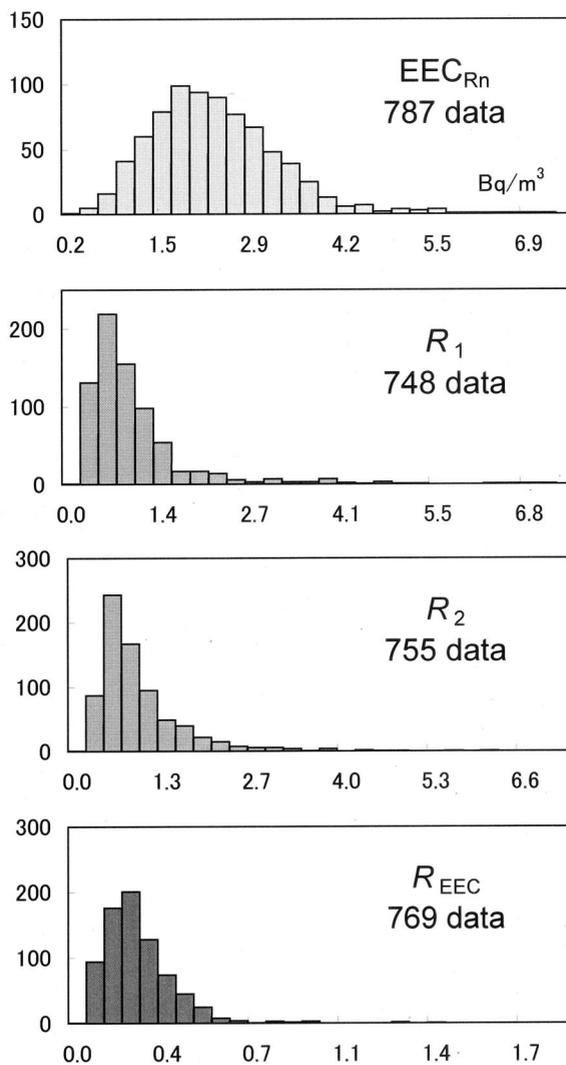


Fig. 2. Statistical distribution of radon progeny and ratios in outdoor air for Series 2 (progeny were sampled at 15:00).

**Correlation analysis**

Since Series 3 had perfect meteorological data, correlation analysis was made for this series. Table 4 shows Pearson product-moment correlation between ratios and meteorological data. In this table, the symbols + and -

denote significant positive and negative correlation, respectively. In the cases of + and -, every *p*-value was less than 0.01. Contrary to expectation, there was no correlation between  $R_{EEC}$  and wind speed. Table 5 shows correlation between ratios and progeny concentrations.  $R_{EEC}$  correlated

**Table 2.** Lilliefors test for the logarithm of the data for each fiscal year. Encircled data passed the test, i.e., the statistical distribution is log-normal.

Year	1988	1989	1990	1991	1992	1994	1995	1996	1997
Sampling time	9:15	9:15	9:15	9:15	9:15	15:00	15:00	15:00	9:00
#	209	237	225	235	240	268	186	175	209
$^{218}\text{Po}$									
$^{214}\text{Pb}$	○	○	○		○				
$^{214}\text{Bi}$			○		○				
$EEC_{Rn}$	○	○	○	○	○		○	○	○
$R_1$		○	○	○	○		○	○	○
$R_2$				○	○		○	○	
$R_{EEC}$		○		○	○	○	○		

**Table 3.** Seasonal variation of mean values of radon progeny ratio,  $R_{EEC}$ . Parenthesized are number of data used.  $R_{EEC}$  is high in spring and winter, i.e., the outdoor air is more stable in summer and autumn.

	Series 1 9:15	Series 2 15:00	Series 3 9:00
Spring	0.215 (336)	0.254 (193)	0.199 (71)
Summer	0.189 (334)	0.207 (171)	0.194 (103)
Autumn	0.191 (300)	0.195 (205)	0.174 (84)
Winter	0.211 (266)	0.247 (200)	0.262 (81)

**Table 4.** Correlation of radon progeny and ratios with meteorological data.  $R_{EEC}$  has no correlation with wind speed.

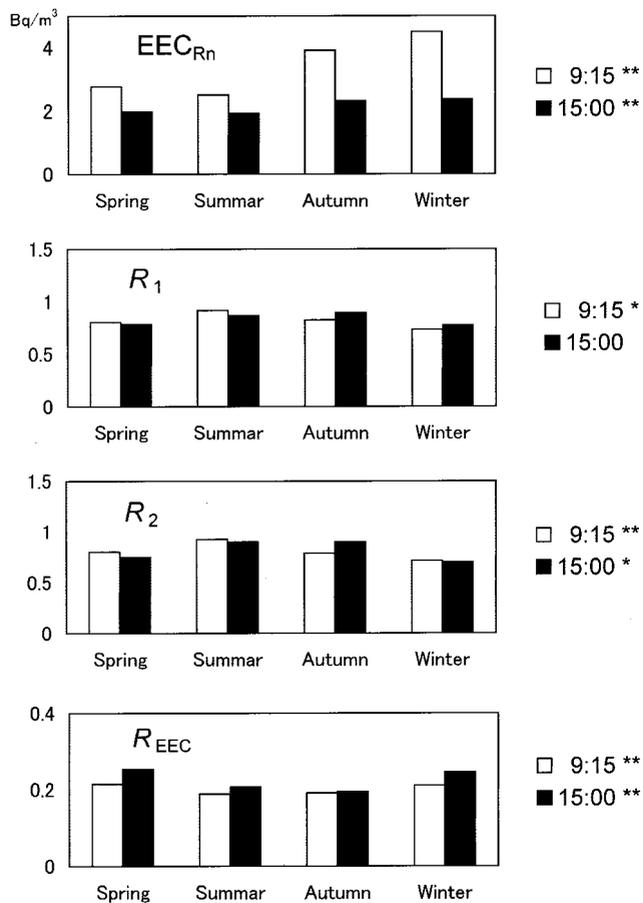
	Temperature	Humidity	Atm. Pressure	Wind speed
$EEC_{Rn}$	—		+	—
$R_1$		+		
$R_2$	+			
$R_{EEC}$	—			

**Table 5.** Correlation of ratios with progeny concentrations.

	$^{218}Po$	$^{214}Pb$	$^{214}Bi$	$EEC_{Rn}$
$R_{EEC}$	+	—	—	—
$R_1$	—	+		
$R_2$	—		+	

**Table 6.** Correlation of ratios with themselves.

	$R_{EEC}$	$R_1$	$R_2$
$R_{EEC}$	.	—	—
$R_1$	—	.	+
$R_2$	—	+	.

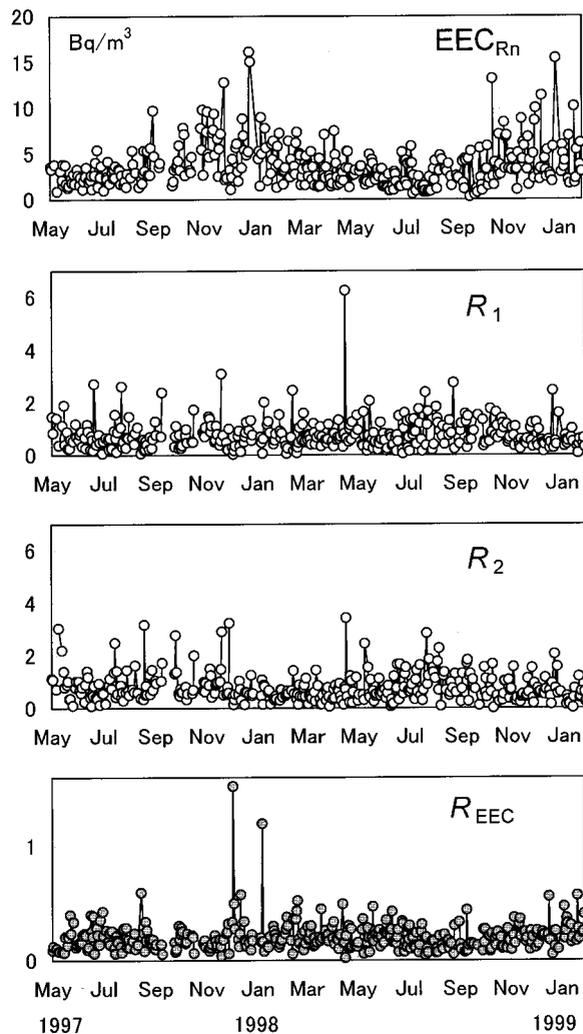


**Fig. 3.** Seasonal variation of radon progeny and ratios for Series 1 (progeny were sampled at 9:15) and Series 2 (progeny were sampled at 15:00).

with all progeny concentrations, whereas  $R_1$  and  $R_2$  indicated lack of correlation with two progeny concentrations. Table 6 shows correlation between the ratios themselves. Three ratios had significant correlation with each other.

**Time series analysis**

Time series analysis (spectrum analysis) was made for various variables using data from Series 3. To



**Fig. 4.** Time series for radon progeny and ratios in outdoor air using data from 1997-05-01 to 1998-01-09 in Series 3 (progeny were sampled at 9:00).

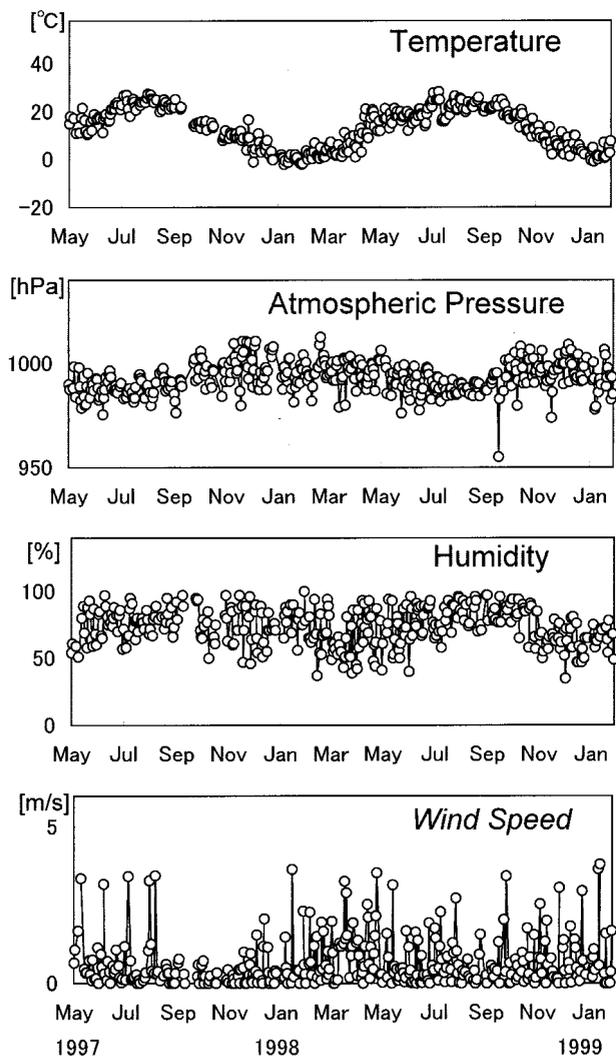


Fig. 5. Time series for meteorological data from 1997-05-01 to 1998-01-09 in Series 3 (progeny were sampled at 9:00).

estimate power spectra for time variations, the autoregressive model (Akaike 1969), equivalent to the maximum entropy method (Burg 1967), was adopted. BASIC programs were composed referring to Hino (1977) for time-series analysis. Figs. 4 and 5 show raw time-series data. Figs. 6 and 7 show the results of spectrum analysis for progeny/ratios and meteorological data, respectively. The power spectrum for  $R_{EEC}$  was very similar to that of wind speed. A 1-y period, that is always remarkable for radon progeny, was not significant for  $R_{EEC}$ . Three- to nine-day periods also appear for  $R_{EEC}$ , radon progeny, wind speed, and atmospheric pressure. These several-day periods are probably attributable to the passage of air masses. Twenty-day to forty-day peaks may be attributed to meteorological phenomena corresponding to the rotation period of the sun. Temperature indicated no significant periodicity except a large 1-y peak.

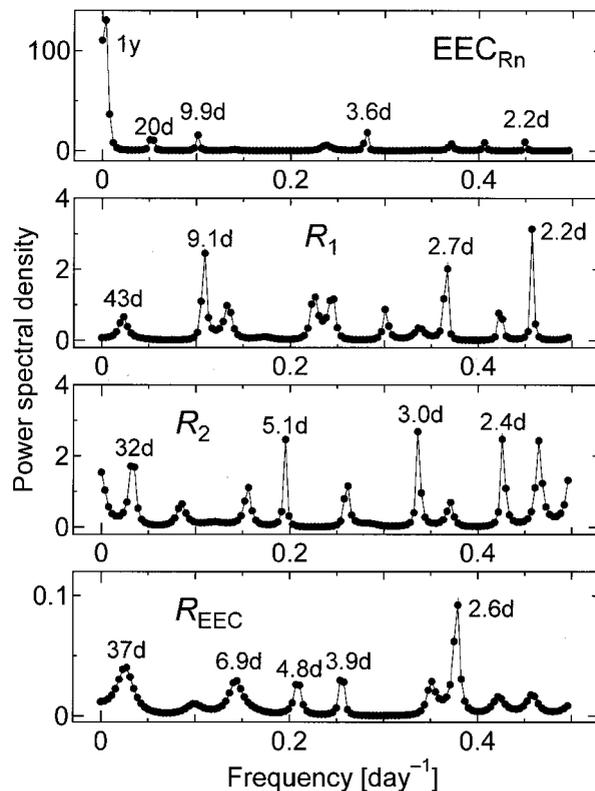


Fig. 6. Power spectra for radon progeny and ratios in outdoor air using data from 1997-05-01 to 1998-01-09 in Series 3 (progeny were sampled at 9:00).

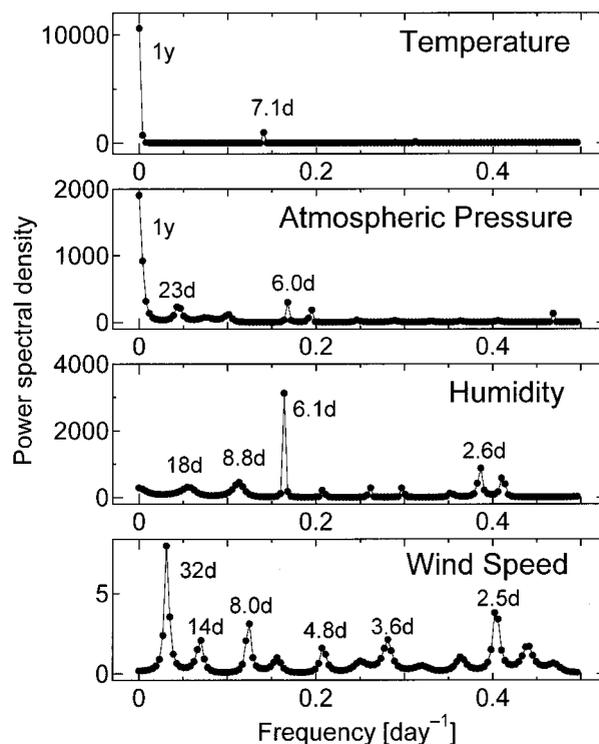


Fig. 7. Power spectra for meteorological data from 1997-05-01 to 1998-01-09 in Series 3 (progeny were sampled at 9:00).

Wind speed is well known to affect radon progeny concentration in air. Also in this study, Pearson correlation analysis indicated strong negative correlation between  $EEC_{Rn}$  and wind speed (Table 4). On the one hand, wind speed evidently reduced progeny concentration. On the other hand, as mentioned previously, the ratio  $R_{EEC}$  had no correlation with wind speed. In spite of no significant correlation between  $R_{EEC}$  and wind speed, the power spectrum of  $R_{EEC}$  was very similar to that of wind speed. The lack of correlation with wind speed may reflect that near-surface mixing is not very sensitive to wind speed.

### CONCLUSION

The concentration ratio of radon progeny in outdoor air was investigated using data obtained intermittently. Statistical analysis on  $R_{EEC}$  indicated that outdoor air was more stable in the morning than in the afternoon, and more stable in summer/autumn than in winter/spring. In spite of no significant correlation between  $R_{EEC}$  and wind speed, the power spectrum of  $R_{EEC}$  was similar to that of wind speed probably because near-surface mixing is not very sensitive to wind speed.

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