Meteorological parameters contributing to variability in ^{222}Rn activity concentrations in soil gas at a site in Sapporo, Japan

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Continuous ^{222}Rn monitoring in soil gas since November 22, 2004 has revealed variability in activity concentration with time in the semi-natural woods on the campus of Hokkaido University in Sapporo, Japan. Among various factors affecting soil radon levels and variability, temperature was found to be dominant during three seasons when activity concentrations of ^{222}Rn showed a diurnal high and nocturnal low with a boundary around 10 o'clock in the morning. This pattern was disturbed by low pressure fronts with occasional rain. The activity gradually decreased as soil temperatures decreased from late November to mid-December. After the ground surface was completely covered with snow, soil radon levels became low with a small fluctuation. There were several peaks of ^{222}Rn on the time-series chart in winter. Those peaks appearing in early winter and early spring may be interpreted by considering meteorological parameters. In a few cases, the radon activity suddenly increased with increasing pressure in the soil at a depth of 10cm, which may be associated with subsurface events such as seismic activity in the area.

Key words: ^{222}Rn, soil gas, continuous monitoring, meteorological parameters, seismic effects

Introduction

There are many factors affecting radon activity concentration in soil gas, and meteorological parameters such as temperature, pressure and precipitation are known to be important contributions.

Seasonal variation of soil radon has been discussed with controversial observations. Winkler et al (2001) compared the variability resulting from different methods, spatial heterogeneity and seasonal fluctuations at a test site located at Neuherberg in Germany. Among several notable results, they observed a winter-high in radon levels due to frozen top soil. An extreme case was reported by Sundal et al (2004) who measured soil radon together with geochemical analyses of bedrock, groundwater and sediment at an ice-marginal deposit in Western Norway. They ascribed anomalously high seasonal changes in soil (and also indoor) radon concentration to subterranean airflows caused by temperature differences between soil air and atmospheric air. Iskandar et al (2004) investigated the dependence of radon emanation power on soil temperature using radium rich soil samples collected in Japan. They obtained a temperature-dependent equation to calculate emanation power at various temperatures from -20°C to 45°C. Over more than one year, Kitto (2005) measured radon flux from soil, along with meteorological and indoor radon measurements, finding that radon flux from soil has a slight seasonal pattern with the greatest exhalation occurring during the late summer months due to the lower moisture content and cracks in the clay soil in summer. The measured flux ranged from 0 to 140 mBq/m^2s with a mean of 37 ± 22 mBq/m^2s. The low flux in winter was caused by a combination of frozen ground and periodic snow melt, whereas low flux in spring most likely resulted from increased precipitation.

Changes in soil radon are thought to be a possible precursor of earthquakes. Among many studies concerning radon and earthquakes, Zmazek et al (2005) reported a statistical technique to identify soil radon anomalies caused by earthquakes in Slovenia by monitoring soil radon concentration, barometric pressure and soil temperature using a barasol probe (MC-450, ALGADE, France), along with other meteorological data like air temperature and precipitation, and also seismic data. Changes in radon concentration that deviated significantly from the mean value were related to seismic activity.

Walia et al (2003) also investigated relationship between radon anomalies and seismic parameters in the northwest Himalayas in India. They compared their results of soil radon monitoring from 1992 to 1999 with seismic data supplied by the Indian Meteorological Department and developed an empirical relationship between earthquake magnitude, epicentral distance and amplitude of radon anomaly. The proposed relationship was a linear relationship between log of the magnitude and log of the product of radon anomaly amplitude and epicentral distance. They concluded that there is no universal empirical relationship that relates radon data with all earthquakes occurring around the world. They further investigated spatial variations in radon and helium concentrations in soil gas across the Shan-Chiao fault in northern Taiwan (Walia et al., 2005). They confirmed that evaluation of both radon and helium was a powerful tool for the detection and mapping of active fault zones.

In our previous study, soil radon was measured temporally with a scintillation Lucas cell at a site on the campus of Hokkaido University, Sapporo, Japan. Soil radon level was varied to a large extent every day even after the probe was permanently emplaced at the point and sampling time was set constant as far as possible. The result together with those by a laboratory experiment suggested that the changing air-filled porosity due to changing soil humidity may be an important parameter controlling soil radon variability (Fujiyoshi et al., 2005). The current study has further investigated factors affecting soil radon levels by continuous monitoring with a Barasol probe since November 2004 at the same location on the campus of Hokkaido University. The obtained data extends these observations to evaluate the influence of meteorological parameters on soil radon levels and variability.

Monitoring Site and Methods

Details of the monitoring site and characteristics of the soil were described previously (Fujiyoshi et al., 2005). A continuous monitoring probe for soil radon (Barasol, Algade, France) was buried in the soil at a depth of 10 or 30 cm. This instrument has a battery powered solid state silicon detector and monitors temperature and barometric pressure with a data logger. It detects alpha-particle emissions of radon in soil gas hourly. The detector sensitivity is 0.02 pulses/h for 1 Bq/m^3 and the saturation volumetric activity is 3 MBq/m^3. Barometric pressure was also measured hourly just above the ground surface (KADEC-U21, Kona System, Japan) and the data set was collected at the end of each month. Humidity of the soil at depths of 20 and 50 cm was monitored hourly with a probe which could store data obtained for about 6 months (Profile Probe, Delta-T Devices Ltd., UK). Instruments were kept in the snow during the winter months from November 2004 to late March 2005, and also from December 2005 to March 2006.

Results and Discussion

Soil radon has been measured temporally by an active technique utilizing a scintillation Lucas cell (Pylon, Canada) at a point in the woods on the Hokkaido University campus since June 2002. In an earlier study, changing air-filled porosity caused by meteorological phenomena was found to affect soil radon levels (Fujiyoshi et al., 2005). During the observation period, there was a sudden increase in soil radon, which coincided with a large local earthquake on September 26, 2003 with the epicenter located offshore near Tokachi in Hokkaido, Japan. Because of the perceived link between earthquakes and changes in soil radon, the present study was undertaken to obtain detailed data on other factors that may contribute to changes in soil radon levels. The current data was obtained from continuous monitoring with a Barasol probe at the same location as the previous study.

Figure 1 shows time-series plots of soil radon levels together with hourly mean values of soil temperature measured at a depth of 10 cm from September 24-30, 2005. This is representative of data obtained at other times. Soil radon levels have a diurnal high and a nocturnal low. Such variability is strongly associated with atmospheric (and thus soil) temperature as shown in the figure. Consequently, there is a good correlation between daily ^{222}Rn concentration and temperature in soil at a depth of 10 cm ($0.78<r<0.94$; Fig. 2). The data indicates that an increase in soil temperature by 1 °C may result in an increase in soil radon level as 740 Bq/m^3 under similar meteorological conditions to those in the observation period of Sep. 24-30 2005.

However, this correlation is reduced by a low pressure front with occasional rain from October 1 to 2, 2005 as shown in Fig. 3. It should be noted that ^{222}Rn activity decreased sharply at 8pm on October 2 and the level remained low during the following day. This may correspond to the drop in temperature monitored at 10cm depth from the ground surface. The typical daily pattern in soil radon levels appeared again on October 4 as seen in Fig. 4, and similar variability in radon concentration was observed during most of the year except in winter.

Figure 5 shows the time-series plots of ^{222}Rn activity concentration and soil temperature from November 22 2004 to March 17 2005 and from December 1 2005 to March 2 2006. The data obtained in 2004-2005 winter can be roughly divided into three parts: i) from November 22 to December 31 when gradual decreases in soil radon levels were observed with decreasing soil temperature; ii) from January 1 to 19 when radon levels and soil temperature were low with little fluctuation; and iii) from January 20 to March 17 when ^{222}Rn activity concentration in soil gas was low, but with occasional high values during nearly constant soil temperatures (0°C). The low soil radon during winter can be related to the snow on the ground. The date when Sapporo city was completely covered with snow (lingering snow) was December 5, 2004 as reported by the Sapporo District Meteorological Observatory (http://www.data.kisyou.go.jp). From that time, snow depth increased to approximately 34cm by the end of the year. In contrast, soil temperature did not correspond to atmospheric temperature which was occasionally lower than -10°C. The discrepancy between temperatures in the soil and atmosphere may contribute to a soil layer of high humidity due to melting snow.

The equilibrium state for a radon-air-water system with respect to the diffusion process is described by the temperature-dependent Ostwald coefficient. The coefficient for radon is 0.5 at 0°C and is 0.26 at 20°C. Diffusion of ^{222}Rn gas originating from deeper portions of the soil to the atmosphere is hampered by thick snow cover and is dissolved in the aqueous phase present in the upper portion of the soil. This may contribute to low ^{222}Rn concentration in soil gas during the winter after lingering snow at our site. The influence of water on soil radon concentration was evaluated by Oufni (2003). He determined the diffusion coefficient and exhalation rate of radon in Moroccan quaternary samples using the SSNTD technique, finding that increasing porosity makes water fill the empty pores and the diffusion of radon reduces.

As shown in Fig. 5, a similar result was obtained during the next winter from December 1, 2005 to March 2, 2006 when the probe was set in the soil at a depth of 30cm. It should be noted that an extremely high peak was observed on January 1, 2006. Inspection of the data shows that relatively high values of ^{222}Rn concentration appeared from December 28, which subsequently disappeared by January 5. As shown in Table 1, an earthquake with intensity of 2 occurred in Sapporo on December 13, 2005. The epicenter was located offshore near Tokachi in Hokkaido (M = 5.5). It is not clear at this moment if the radon anomalies observed from December 28 to January 5 would directly relate to this earthquake.

Previous studies have shown increasing soil radon concentrations in winter due to the frozen top soil (Winkler et al., 2001, Kitto, 2005). In contrast, the current study demonstrates low soil radon concentrations during winter, but the different soil temperatures are likely a major consideration since the top soil at our Sapporo site was never frozen, but always wet by melting snow after lingering snow. This also contributes to relatively constant soil temperature (0°C) in the mixture of snow and melting water during winter months (Fig. 5).

With respect to the soil ^{222}Rn peaks appearing in Fig. 5, some may be explained by considering changes in barometric pressure in soil (P_s) which accompanied changes in soil temperature (T_s). Figure 6 shows a typical sequence in which ^{222}Rn concentrations increased gradually on January 29 and decreased on the next day. During that time, a low pressure front arrived on January 29-30 that resulted in a small increase in soil temperature and a corresponding increase in radon level from 500 to 2500 Bq/m^3.

Similar events were observed in early spring from March 5 to 9, 2005. As shown in Fig. 7, barometric pressure in soil (P_s) began to decrease sharply from midnight of March 7 for about 24hrs, and peaked at approximately 10am on March 8. During this time, soil temperature (T_s) increased continuously from 0.1 to 0.6 °C even under a thick snow cover (~1m), which would affect the radon levels as shown in Fig. 8. It is interesting to note that the soil temperature increased gradually on a daily basis, until the typical diurnal and nocturnal changes in temperature appeared again on March 16, 2005.

There are additional peaks that should be noted on December 7 and December 23 in 2004 and March 18, 2005 in which they were not influenced by soil temperature. Time-series data on ^{222}Rn activity concentrations in the above three cases was plotted together in Fig. 9 (December 7-9 2004, December 22-25 2004 and March 18-20 2005). Soil radon increased considerably from 2000 to 6000 Bq/m^3 from midnight of March 18 until approximately 3 pm the next day, after which it decreased sharply to about 2500 Bq/m^3 at the end of the day. For the following whole day the concentration decreased slowly to a normal winter level of 500 Bq/m^3. As shown in the figure, a similar pattern was observed from December 22 to 24, 2004 when the soil radon levels began to increase around 5am on December 22 and decreased again on the next day to near 1500 Bq/m^3. These events seemed to correspond with changing barometric pressure as measured at a depth of 10cm (Fig. 10).

With respect to barometric pressure, the difference between atmospheric pressure (P_a) and soil pressure (P_s) is plotted on time-series chart of Fig. 11. It is clear from the figure that P_a-P_s values were kept constant (about 15 hPa) with time from March 18 to 20, 2005. In contrast, they were varied to a large extent in two other cases (Dec. 7-9 and Dec. 22-25). It may be due to a thick snow cover (1 m) on the soil surface in March, 2005 compared with that (20-40cm) in December, 2004.

Figure 12 shows a relationship between ^{222}Rn activity concentration and the difference in barometric pressure (P_a-P_s) on two radon anomalies observed on December 7-8 and December 23-24, in which only limited time span corresponding to each radon peak showed linearity from 10 am to 8 pm and from 10 am to 4 pm on December 7 and December 8, respectively. In any case, good correlation ($0.80<R<0.97$) exists between them.

Collectively, the data indicate the existence of some internal activity which forced gaseous constituents, including radon, out to the upper portion of the soil still covered with snow in winter. Seismic activity is a likely candidate for such an out-gassing process.

According to seismic data reported by Japan Meteorological Agency, there were several earthquakes with intensity higher than 1 measured in Sapporo during our observation period of 2004-2006. Table 1 lists the earthquakes that occurred during our observation period. Comparing data shown in Table 1 with the radon anomalies observed in this study, the peak on March 18 2004 may not be regarded as a direct precursor of any of the earthquakes listed in the table, since there was no earthquake reported for more than 6 months since January 31, 2005. However, two other anomalies observed on December 7-8 and December 23 may be related either after-effects and/or precursory phenomena of the earthquakes that occurred on December 6 and 14, respectively.

Zmazek et al (2005) compared three techniques to distinguish the anomalies caused by environmental parameters from those resulting solely from seismic activity. They found a simulation model (regression tree) to be the most applicable and the model was used to analyze data on soil radon measured during the non-seismically active periods. According to this technique, a discrepancy between measured values of radon concentration and values predicted by the model indicates a measure of earthquake prediction.

Another approach for earthquake prediction was reported by Planinic et al (2004). They measured radon concentration in soil gas continuously using the LR-115 nuclear track detectors for analysis with seismic activities, barometric pressure, precipitation and air temperature for four years at three sites in Croatia. By investigating linear and exponential multiple regression, they developed an algorithm to predict radon concentration as functions of barometric pressure, precipitation and atmospheric temperature.

The present authors could not find a quantitative relationship with which one could quantitatively evaluate the influence of meteorological parameters on temporal radon variations. However, it was relatively simple to detect anomalies from time-series radon data in winter when the ground is completely covered with snow, and soil temperature, the most important meteorological parameter influencing soil radon variability, is kept constant around 0°C. Possible anomalies may then be found by plotting ^{222}Rn activity concentration against the difference of barometric pressure in the atmosphere (P_a) and in soil (P_s). Additional data is being collected in this study to further elucidate quantitative factors controlling soil radon level together with meteorological and seismological data.

Conclusion

Soil temperature was found to be the most important factor influencing soil radon level and variability. The activity concentration was low in winter when the ground surface was covered with snow. Long-term and continuous monitoring of radon in soil gas, together with meteorological and seismological data, is important for evaluating relationships with seismic activity.

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Table 1. Earthquakes with intensity greater than 1 from November 2004 to March 2006*

Date	Time	Altitude (° ‘ N)	Longitude (° ‘ E)	Magnitude	Depth (km)	Intensity in Sapporo
Nov. 27 2004	07 42	42 19.5	143 04.8	5.6	51	1
Nov. 29 2004	03 32	42 56.7	145 16.5	7.1	48	3
Dec. 06 2004	23 15	42 50.8	145 20.5	6.9	46	2
Dec. 14 2004	14 56	44 04.6	141 41.9	6.1	9	2
Jan. 18 2005	23 09	42 52.5	145 00.4	6.4	50	1
Jan. 31 2005	18 39	41 45.8	143 49.0	5.4	48	1
Aug. 06 2005	11 46	38 08.9	142 16.6	7.2	42	1
Nov. 15 2005	06 38	38 01.8	144 53.3	7.1	83	1
Dec. 13 2005	06 01	43 12.5	139 24.8	5.5	29	2

* Earthquake data were reported the Japan Meteorological Agency (http://www.data.kisyou.go.jp).

Figure Legends

Figure 1 Hourly change in ^{222}Rn activity concentration in soil gas and mean soil temperature measured at a depth of 10cm from September 24 to 30, 2005.

Figure 2 Relationship between ^{222}Rn activity concentration and soil temperature from September 24 to 30, 2005.

Figure 3 Time-series plots of ^{222}Rn activity concentration, soil temperature and barometric pressure in soil at a depth of 10cm from September 30 to October 3, 2005. Values of soil temperature in the figure were 100 times larger than the original data.

Figure 4 Hourly change in ^{222}Rn activity concentration in soil gas from October 4 to 7, 2005.

Figure 5 Monitoring ^{222}Rn activity concentration and soil temperature during winter months from November 22, 2004 to March 17, 2005 and from December 1, 2005 to March 2, 2006.

Figure 6 Time-series plots of ^{222}Rn activity concentration, soil temperature and barometric pressure in soil gas at a depth of 10 cm from January 28 to 30, 2005. Data on soil temperature in the figure were 1000 times higher than the original ones.

Figure 7 Time-series plots of barometric pressure and soil temperature at a depth of 10 cm from March 5 to 8, 2005.

Figure 8 Plots of ^{222}Rn activity concentration in soil gas versus temperature in soil measured at a depth of 10 cm from March 5 to 9, 2005.

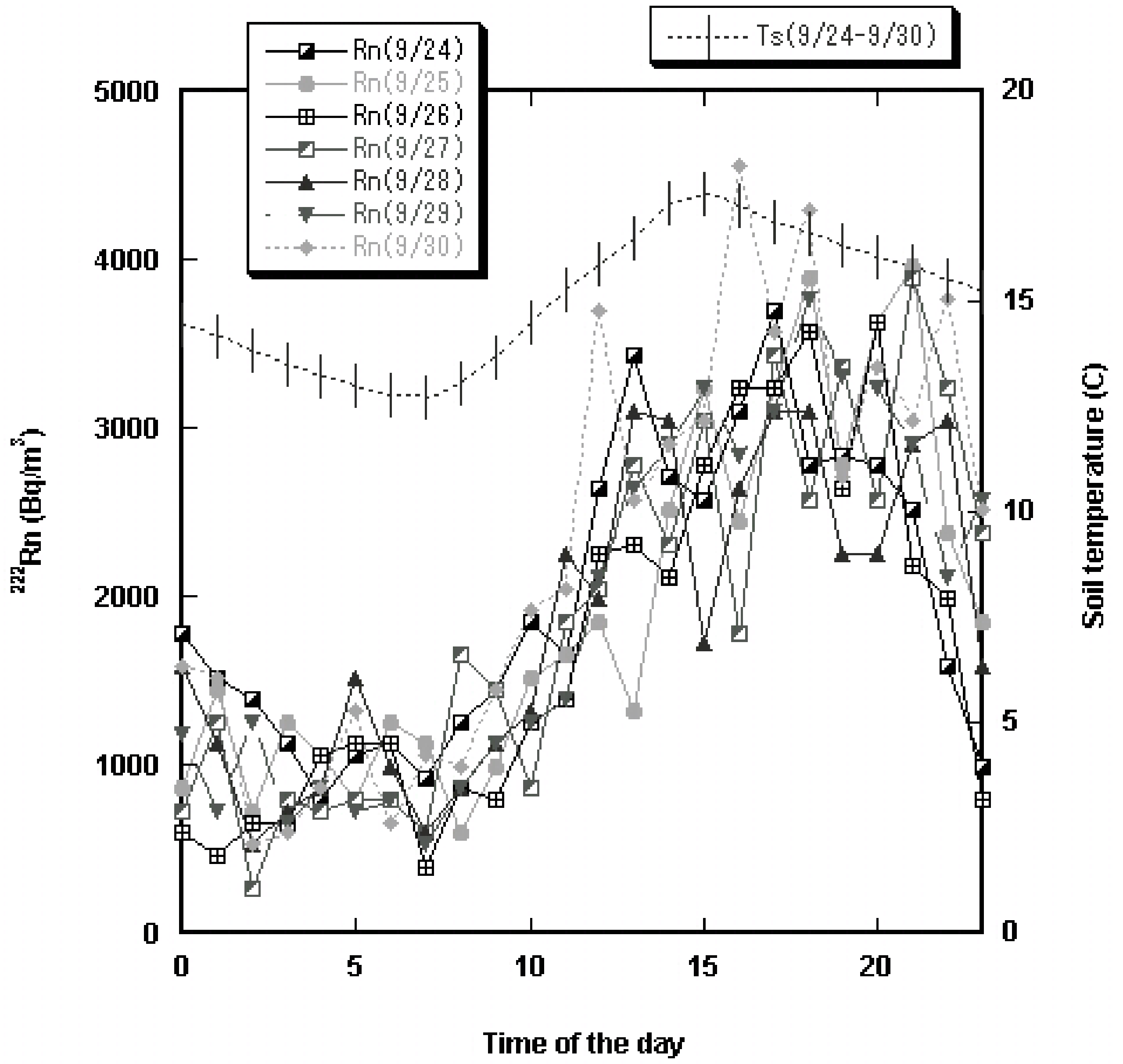
Figure 9 Time-series plots of ^{222}Rn activity concentration from December 7-9 2004, December 22-25, 2004 and from March 18-20, 2005.

Figure 10 Relationship between ^{222}Rn activity concentration in soil gas and barometric pressure in soil measured at a depth of 10 cm on December 7, 8, 23 and 24 in 2004 and March 18 and 19 in 2005. Data on December 7 and 8 are plotted only for a limited time span corresponding to each radon anomaly for 10am-8pm and for 10am-4pm, respectively. Correlation coefficient (R) of each line is shown in the figure.

Figure 11 Time-series plots of the difference in barometric pressure in the atmosphere (P_a) and in soil at a depth of 10cm (P_s) from December 7 to 9, December 22 to 25, 2004 and from March 18 to 20, 2005.

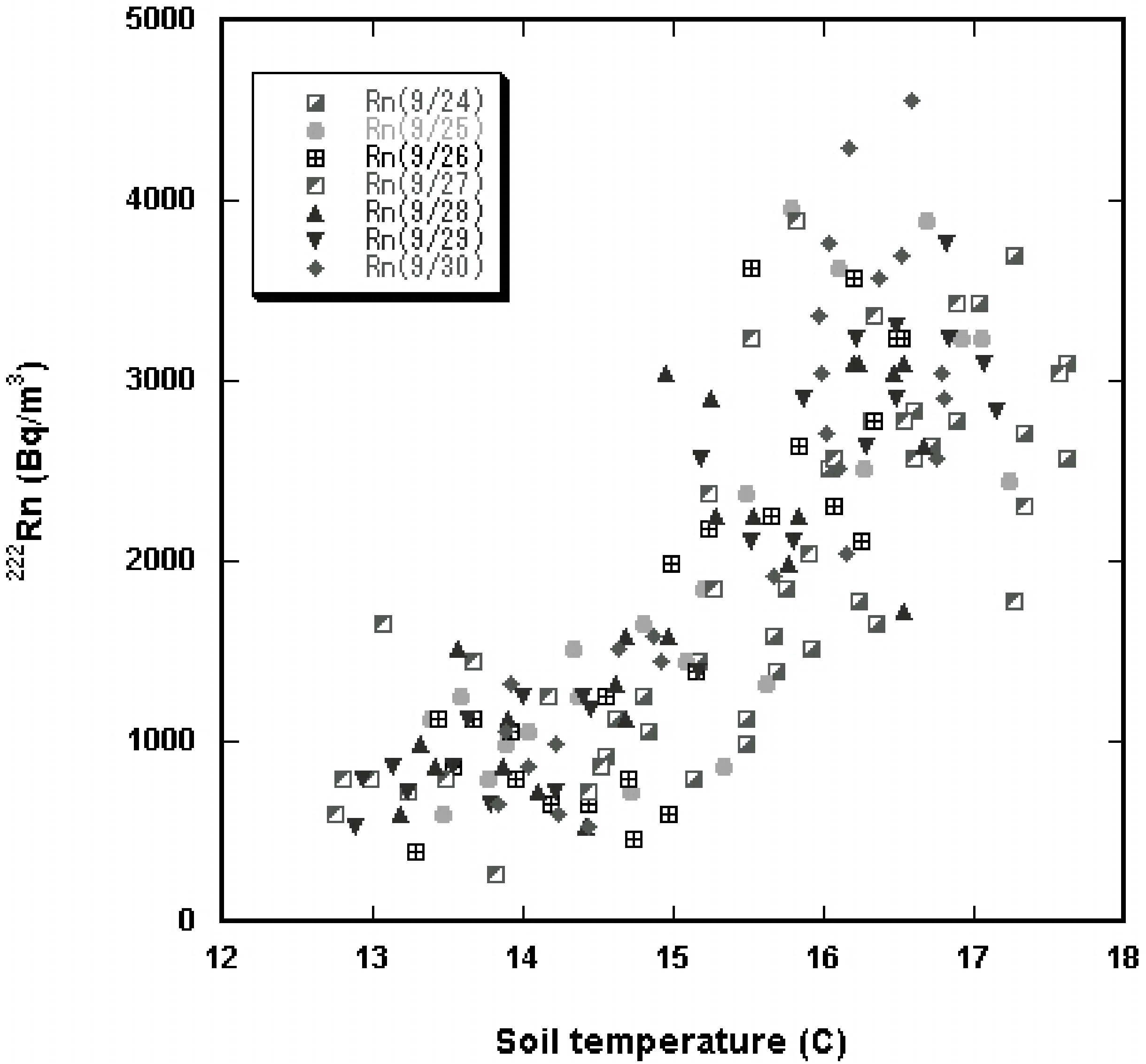
Figure 12 Relationship between ^{222}Rn activity concentration and pressure difference in the atmosphere (P_a) and in soil at a depth of 10 cm (P_s) on December 7 (10am-8pm), December 8 (10am-4pm), December 23 and 24, 2004. Correlation coefficient (R) of each line is shown in the figure.

Figure 1



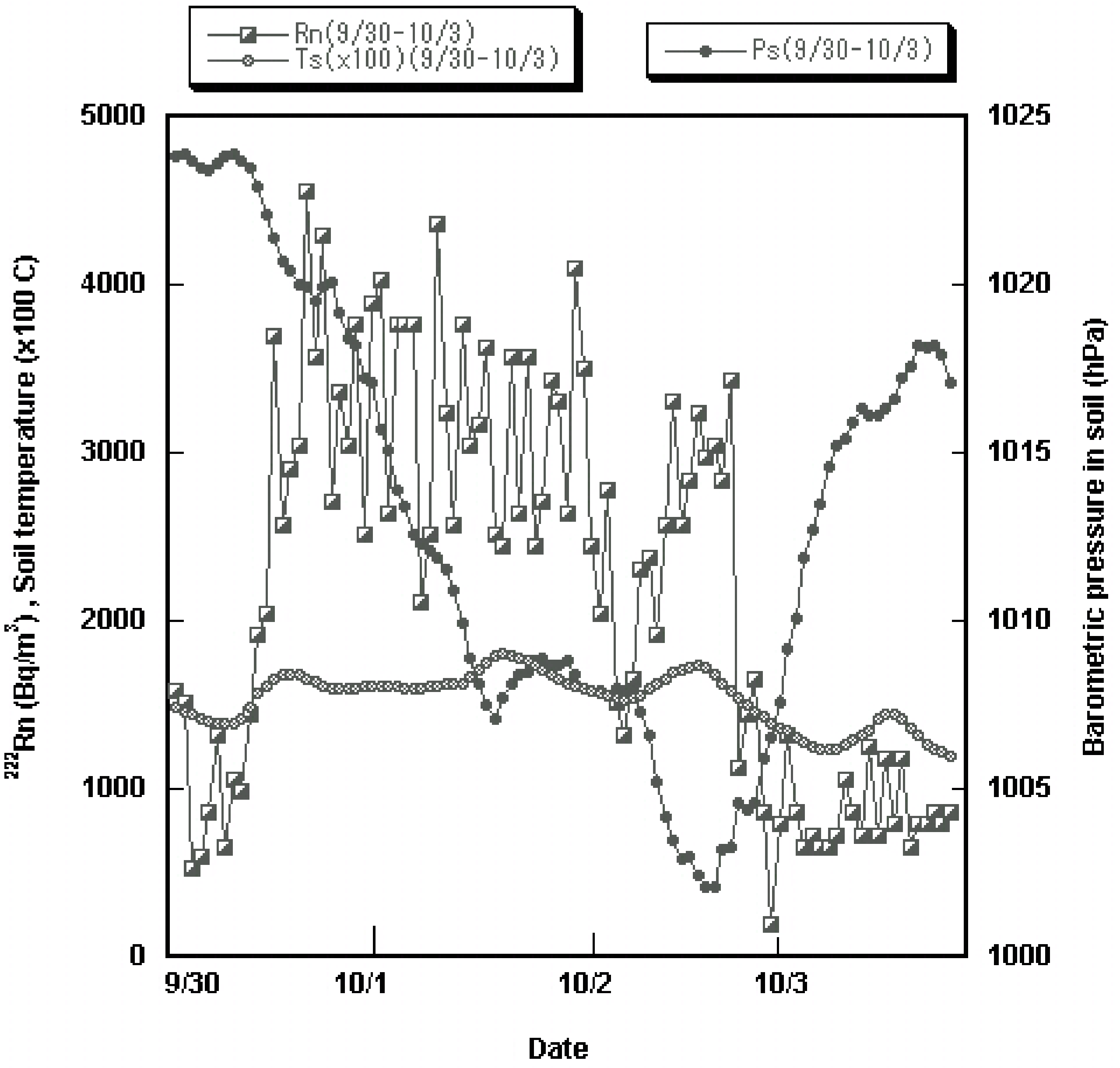
Rn(9/24)
Rn(9/25)
Rn(9/26)
Rn(9/27)
Rn(9/28)
Rn(9/29)
Rn(9/30)
Ts(9/24-9/30)
^{222}Rn (Bq/m^3)
Soil temperature (C)
Time of the day
0
1000
2000
3000
4000
5000
0
5
10
15
20

Figure 2



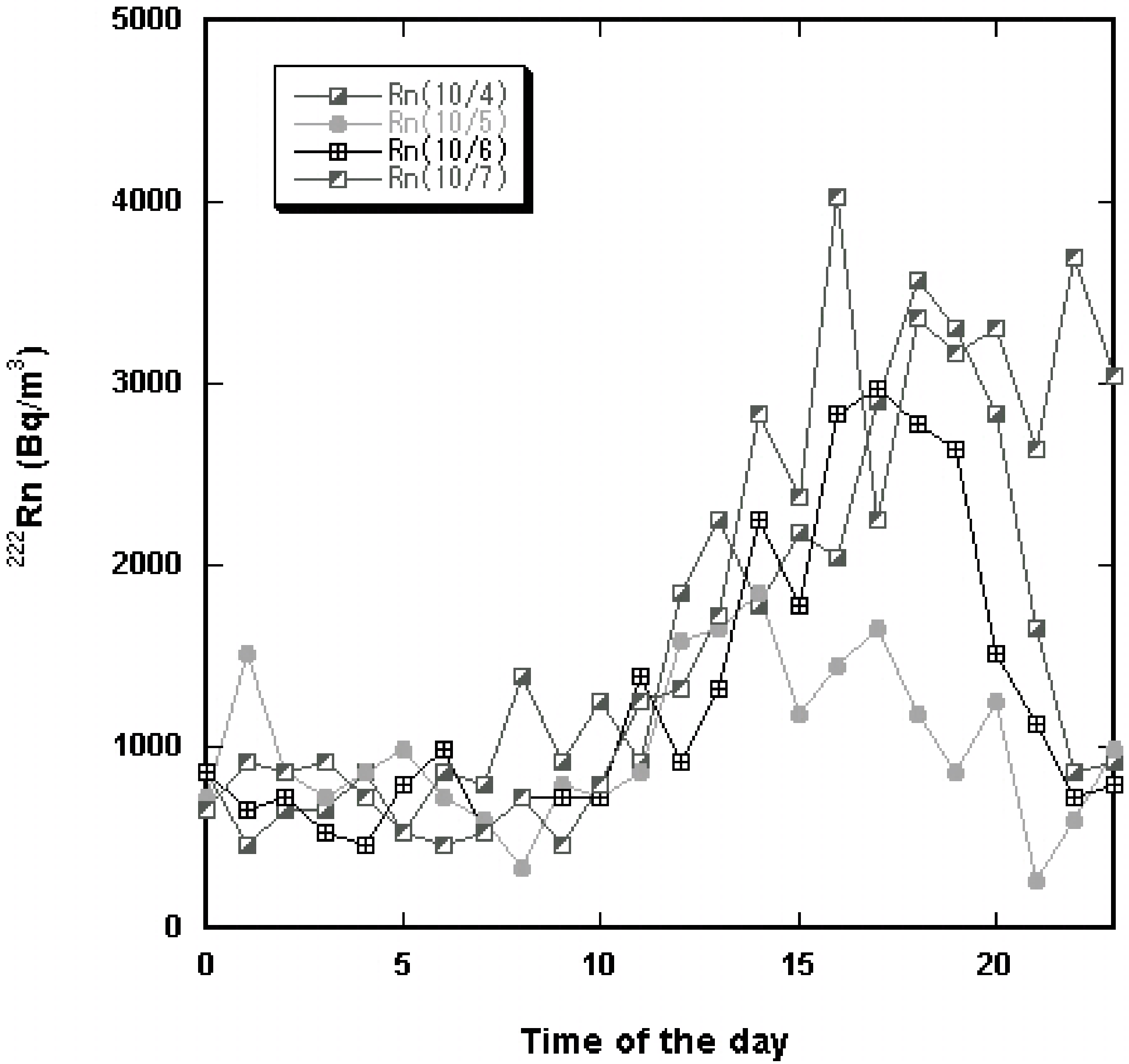
Rn(9/24)
Rn(9/25)
Rn(9/26)
Rn(9/27)
Rn(9/28)
Rn(9/29)
Rn(9/30)
^{222}Rn (Bq/m^3)
5000
4000
3000
2000
1000
0
12
13
14
15
16
17
18
Soil temperature (C)

Figure 3



Rn(9/30-10/3)
Ts(x100)(9/30-10/3)
Ps(9/30-10/3)
5000
4000
3000
2000
1000
0
1025
1020
1015
1010
1005
1000
^{222}Rn (Bq/m^3) , Soil temperature (x100 C)
Barometric pressure in soil (hPa)
9/30
10/1
10/2
10/3
Date

Figure 4


Rn(10/4)
Rn(10/5)
Rn(10/6)
Rn(10/7)
^{222}Rn (Bq/m^3)
Time of the day
0
1000
2000
3000
4000
5000
0
5
10
15
20

Time series plots of Rn222

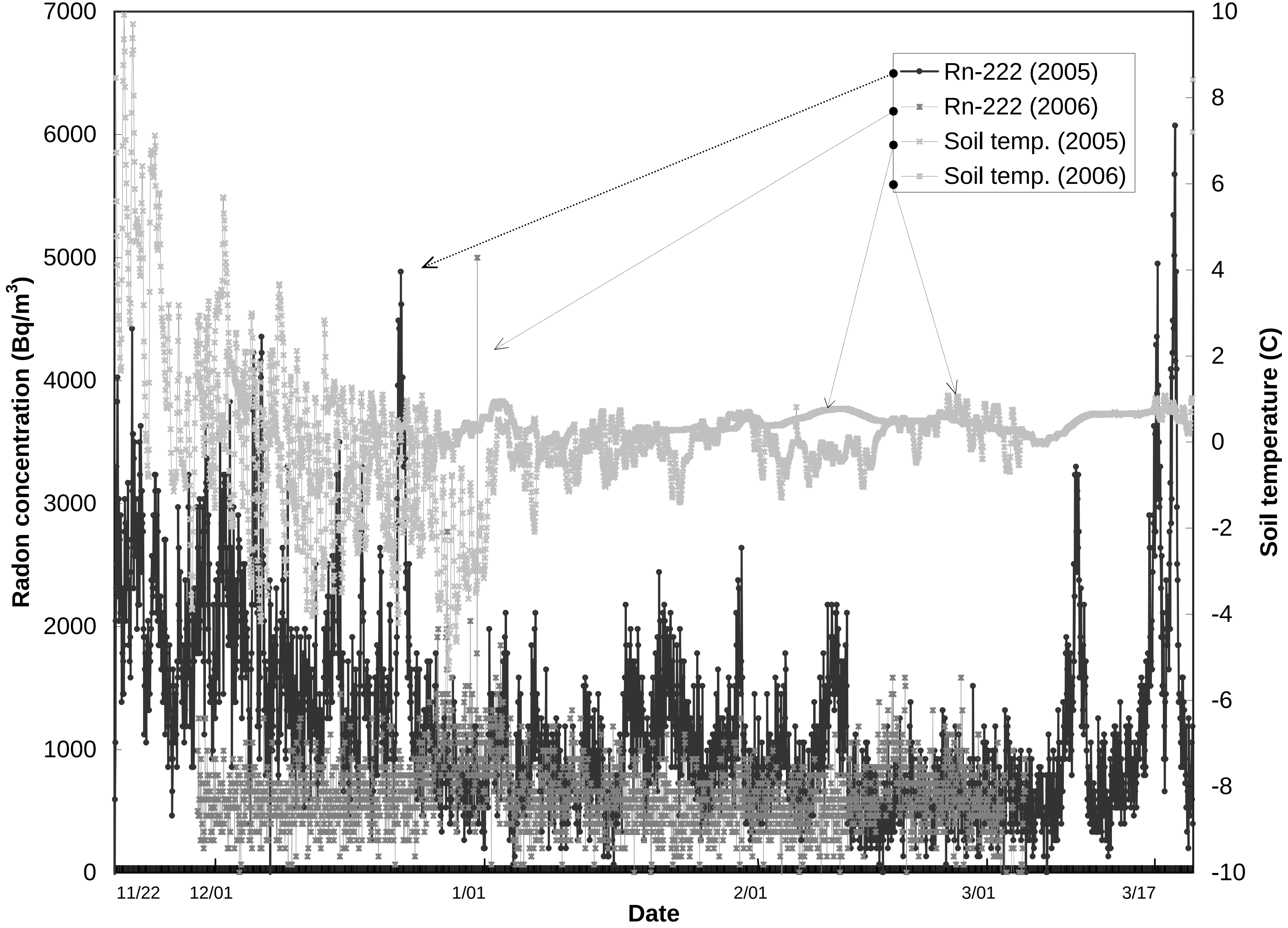


Figure 6

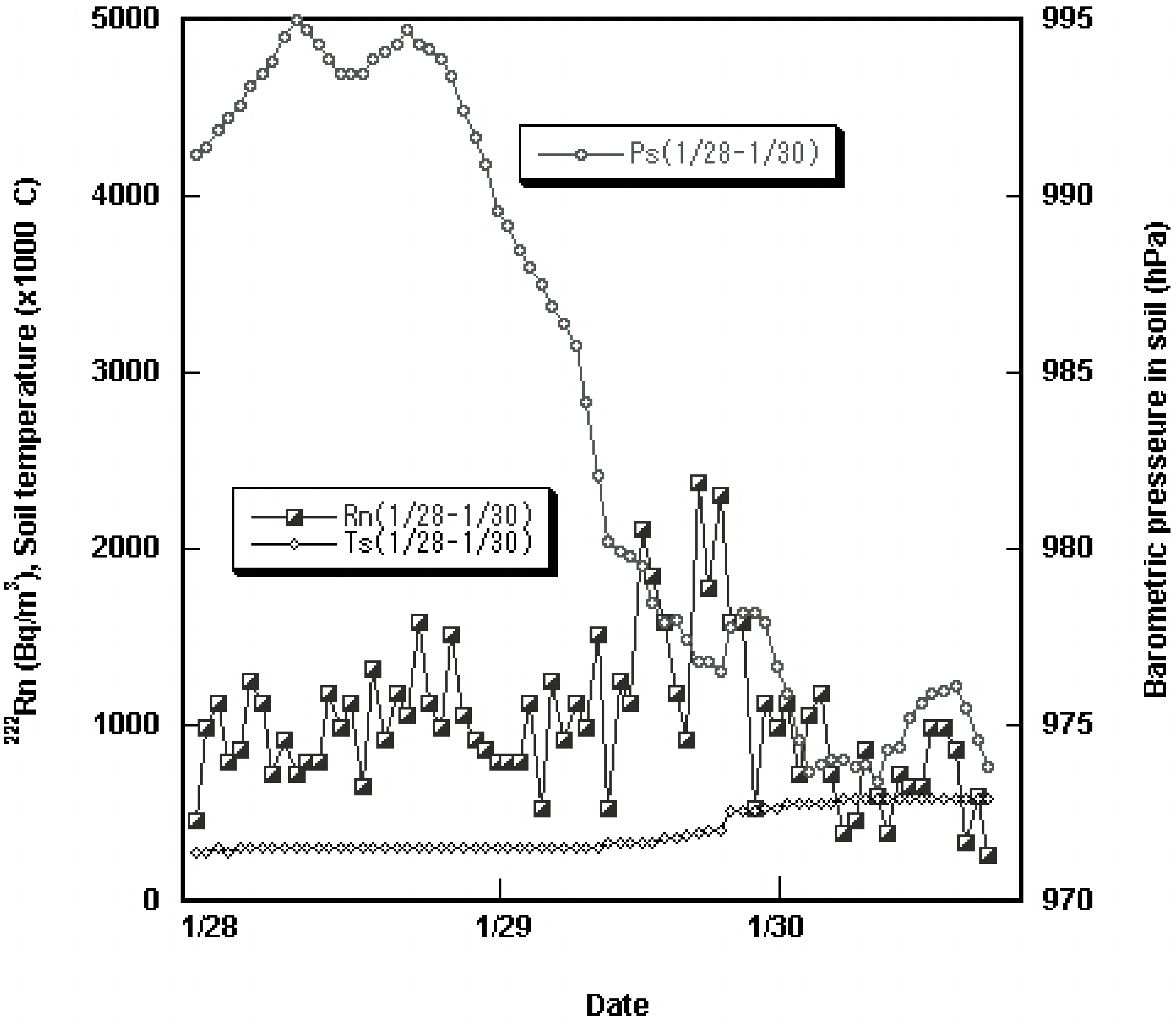

5000
4000
3000
2000
1000
0
995
990
985
980
975
970
Ps(1/28-1/30)
Rn(1/28-1/30)
Ts(1/28-1/30)
^{222}Rn (Bq/m^3), Soil temperature (x1000 C)
Barometric presseure in soil (hPa)
1/28
1/29
1/30
Date

Figure 7

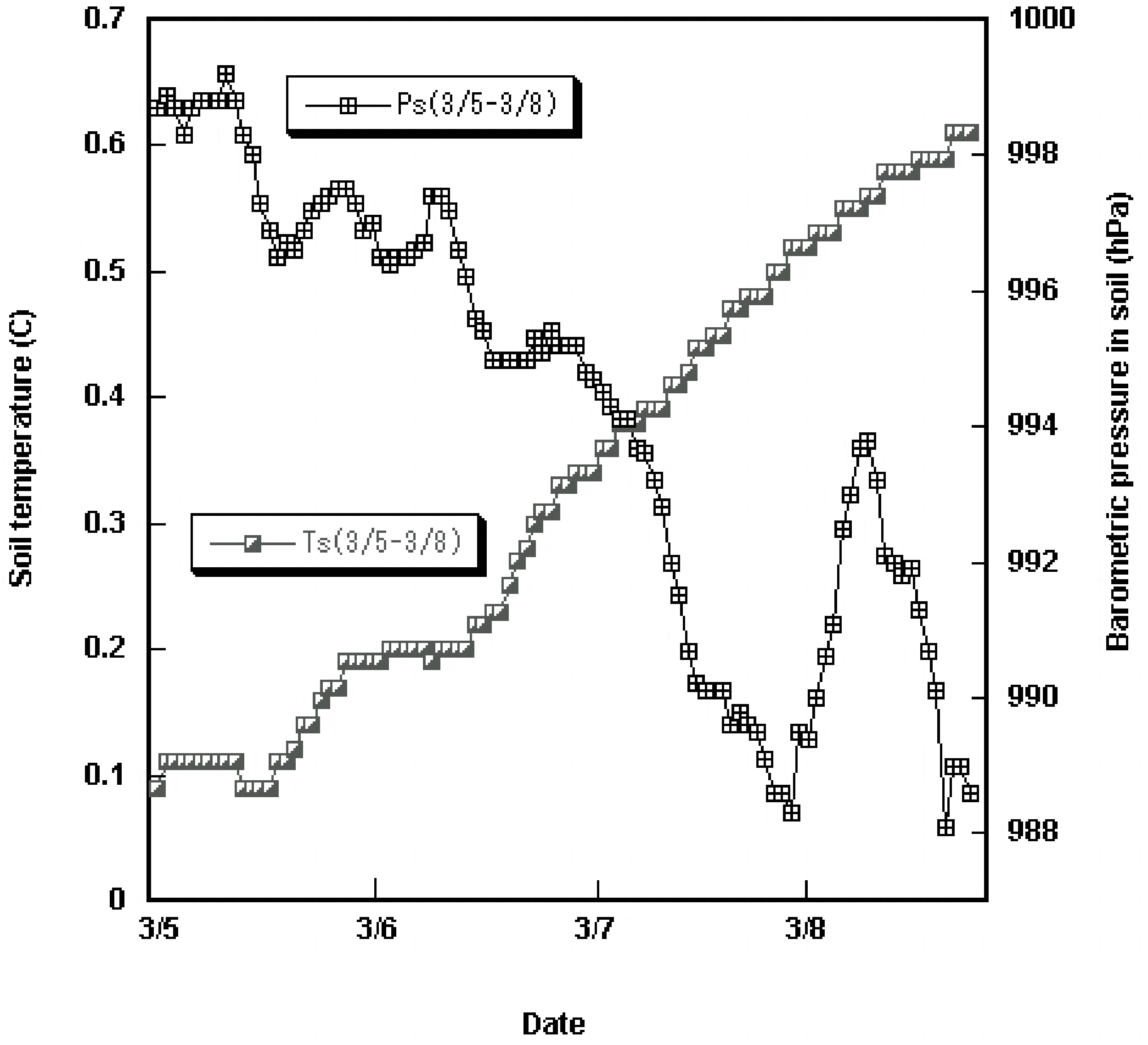
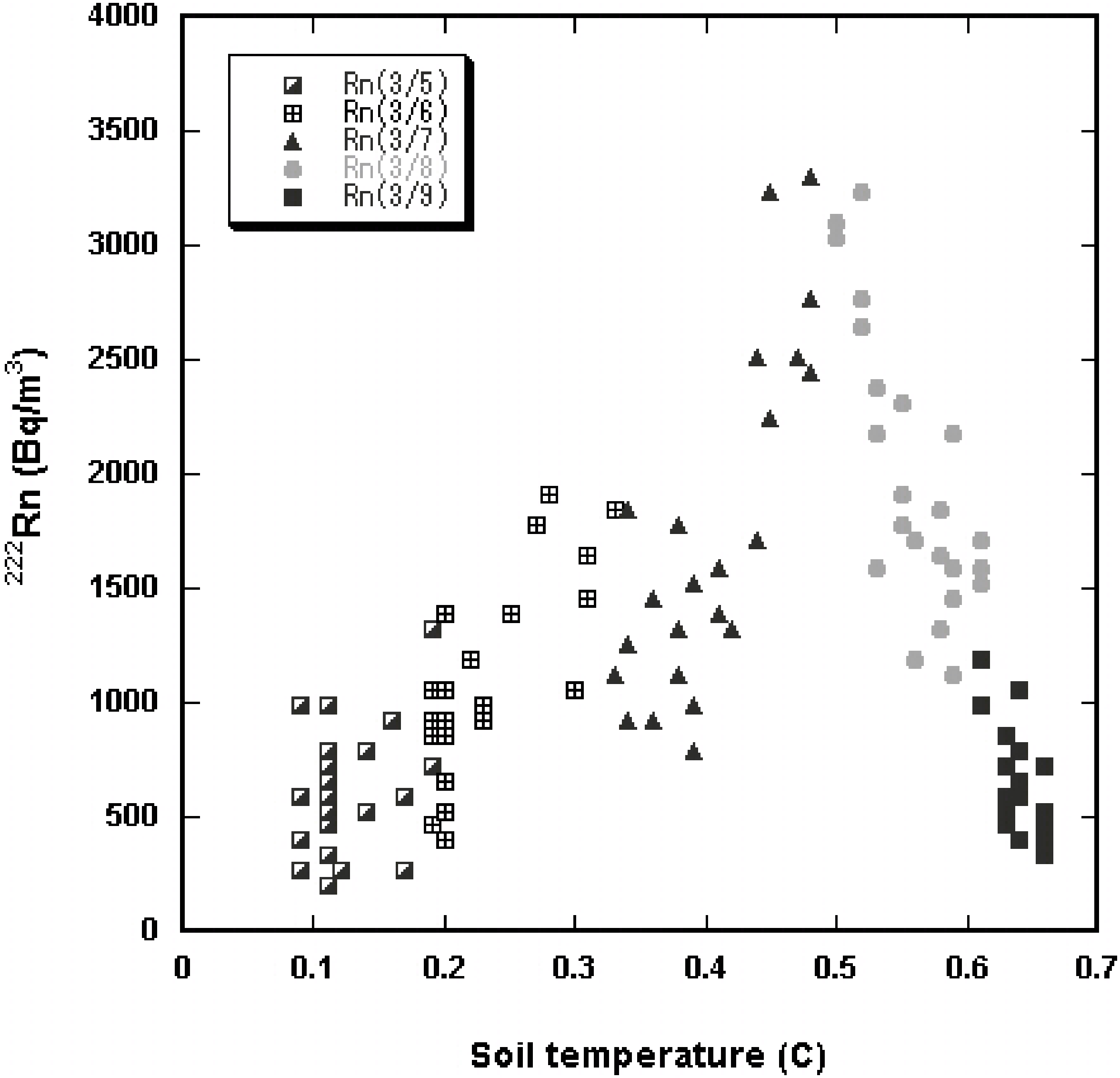
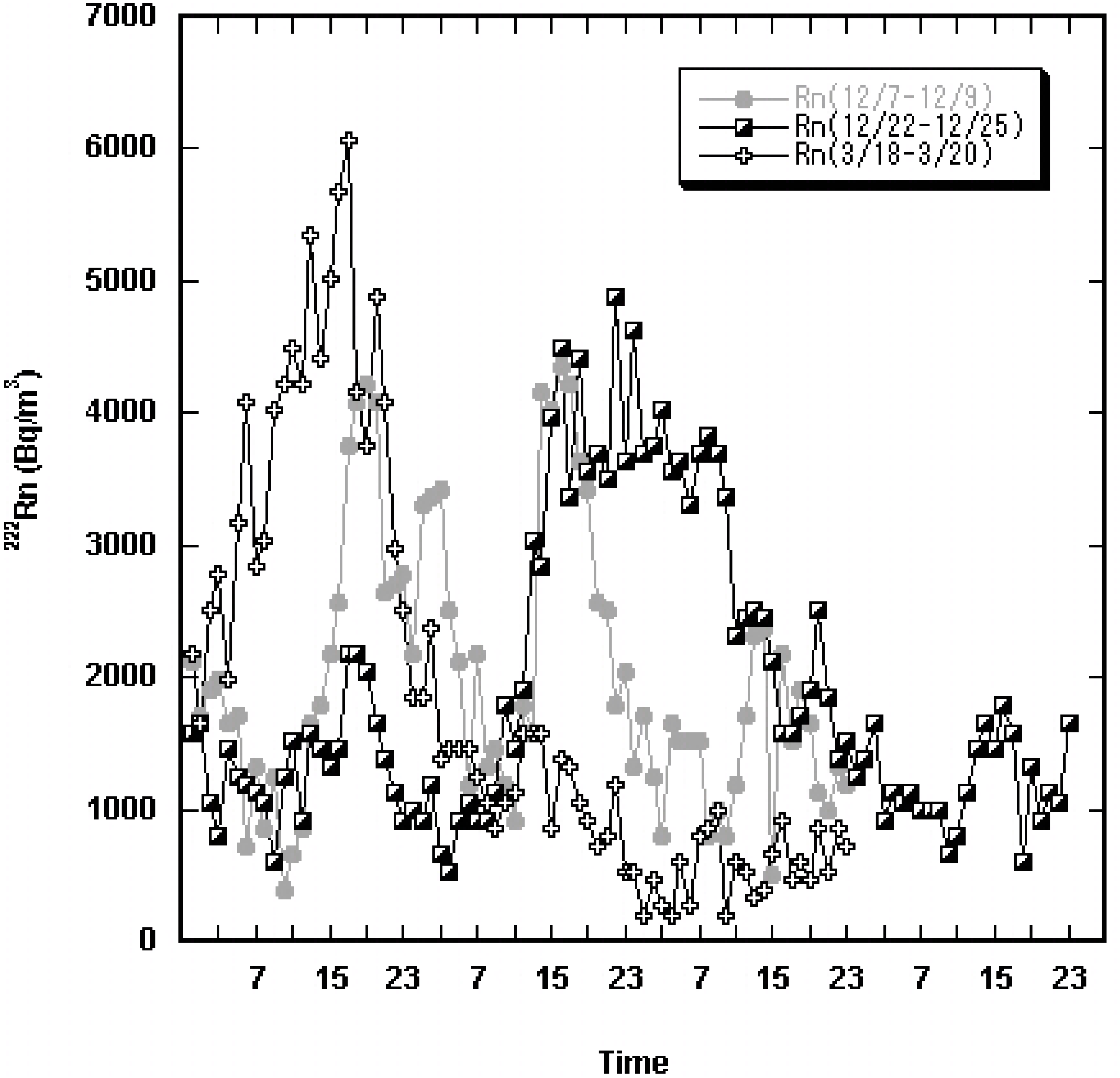

Ps(3/5-3/8)
Ts(3/5-3/8)
Soil temperature (C)
Barometric pressure in soil (hPa)
Date
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
988
990
992
994
996
998
1000
3/5
3/6
3/7
3/8

Figure 8



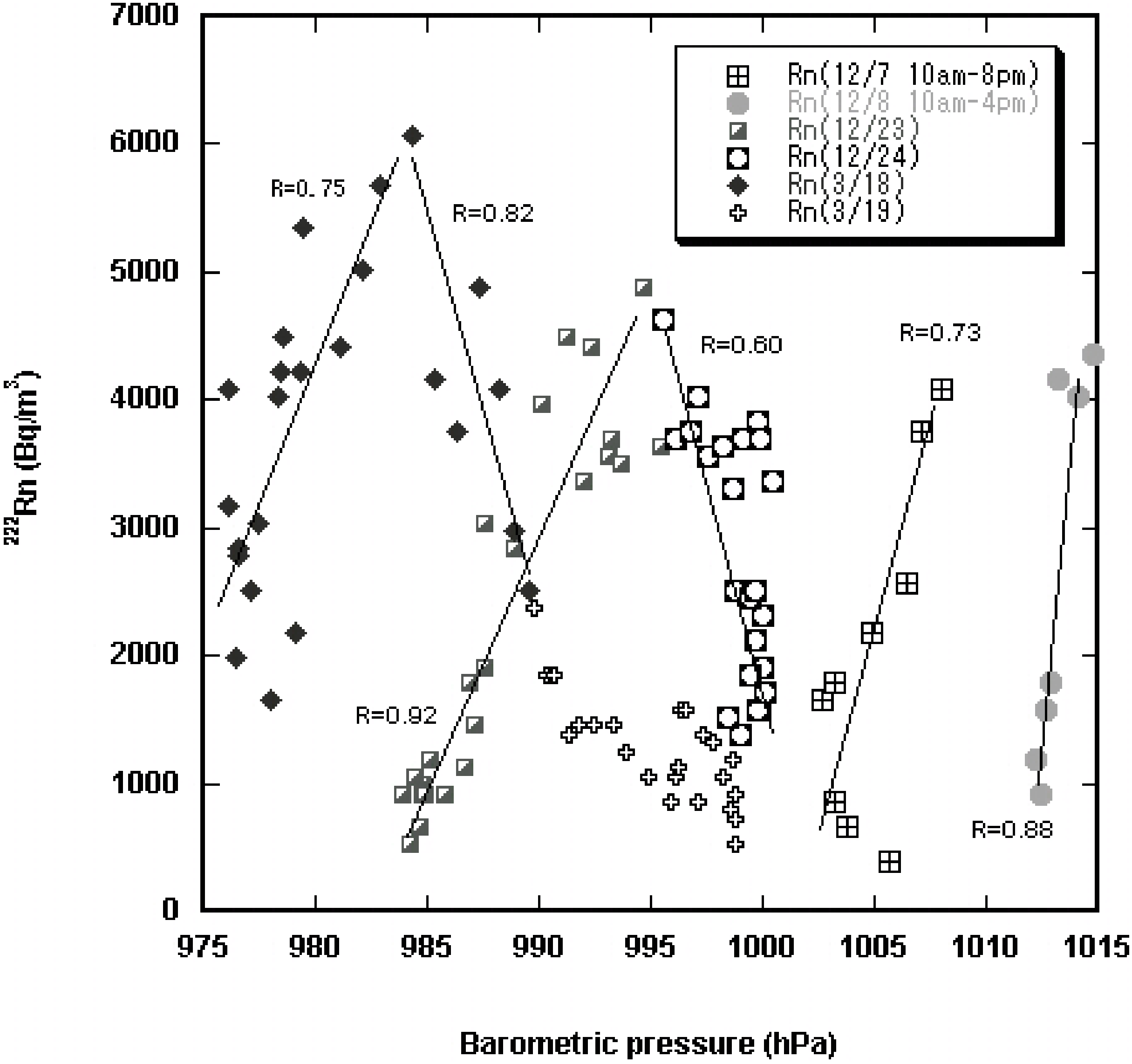
Rn(3/5)
Rn(3/6)
Rn(3/7)
Rn(3/8)
Rn(3/9)
4000
3500
3000
2500
2000
1500
1000
500
0
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
^{222}Rn (Bq/m^3)
Soil temperature (C)

Figure 9



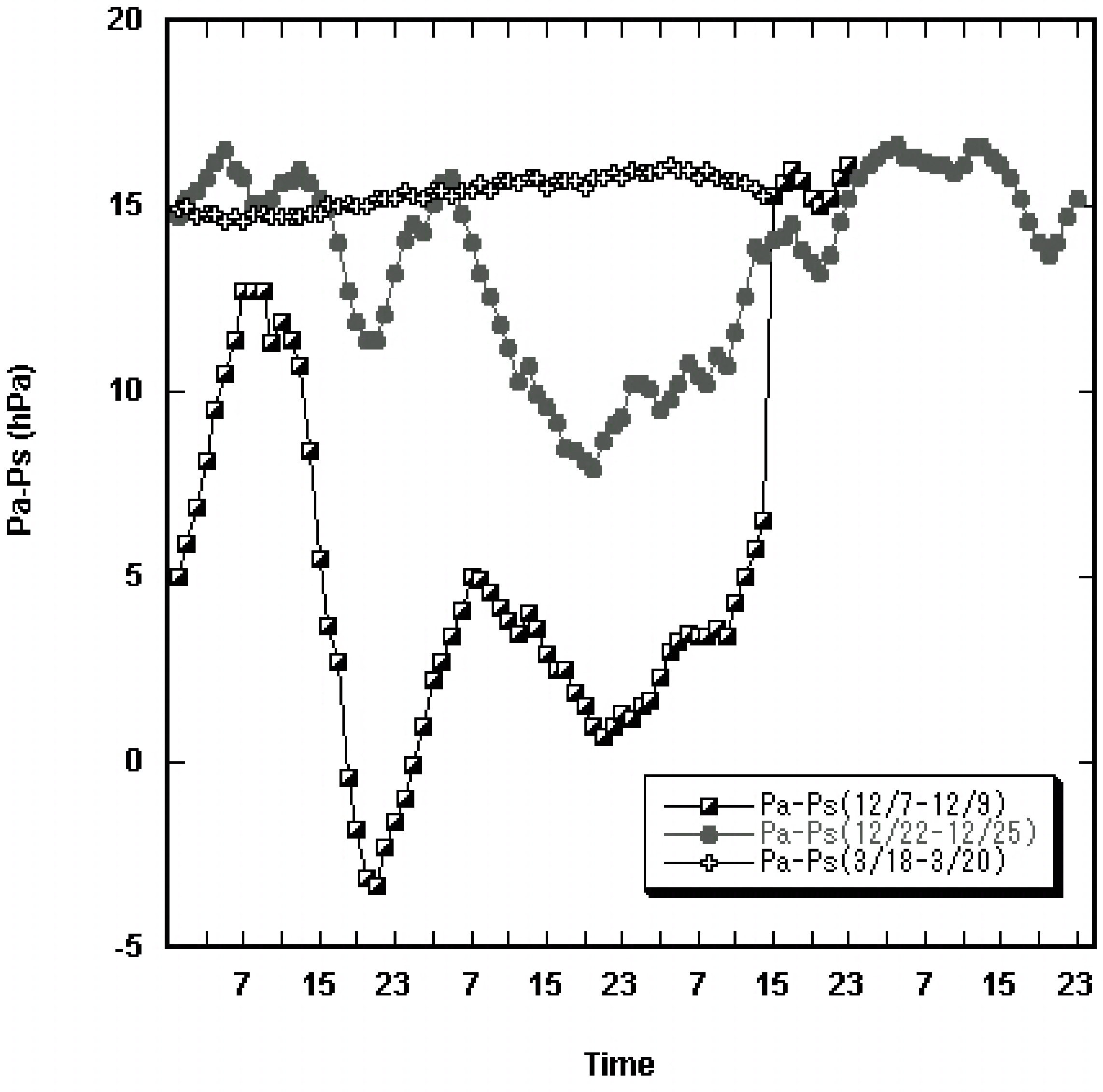
Rn(12/7-12/9)
Rn(12/22-12/25)
Rn(3/18-3/20)
^{222}Rn (Bq/m^3)
0
1000
2000
3000
4000
5000
6000
7000
7
15
23
7
15
23
7
15
23
7
15
23
Time

Figure 10



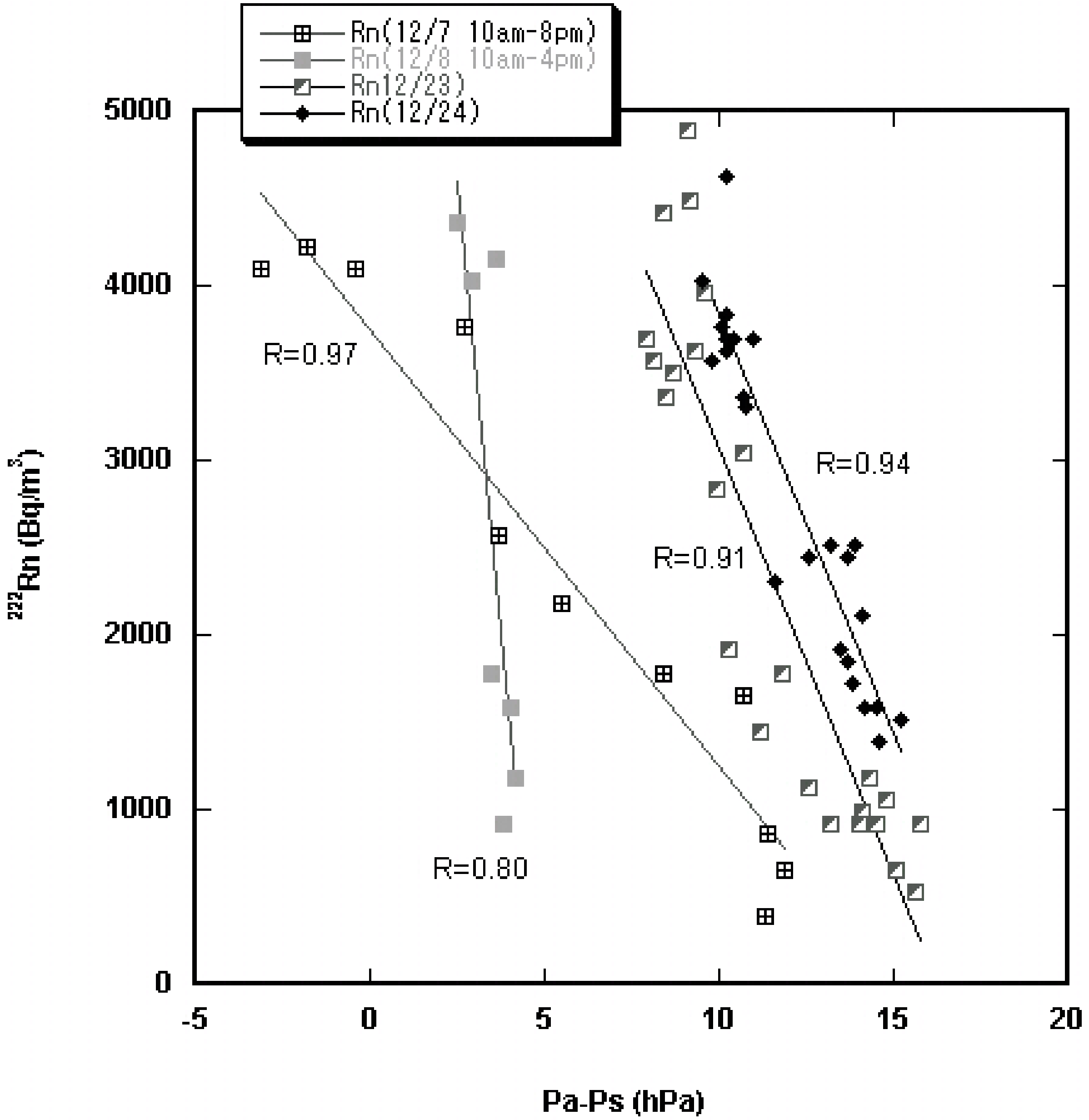
Rn(12/7 10am-8pm)
Rn(12/8 10am-4pm)
Rn(12/23)
Rn(12/24)
Rn(3/18)
Rn(3/19)
R=0.75
R=0.82
R=0.60
R=0.73
R=0.92
R=0.88
^{222}Rn (Bq/m^3)
Barometric pressure (hPa)
0
1000
2000
3000
4000
5000
6000
7000
975
980
985
990
995
1000
1005
1010
1015

Figure 11



20
15
10
5
0
-5
Pa-Ps (hPa)
7
15
23
7
15
23
7
15
23
7
15
23
Time
Pa-Ps(12/7-12/9)
Pa-Ps(12/22-12/25)
Pa-Ps(3/18-3/20)

Figure 12



Rn(12/7 10am-8pm)
Rn(12/8 10am-4pm)
Rn12/23)
Rn(12/24)
R=0.97
R=0.80
R=0.91
R=0.94
5000
4000
3000
2000
1000
0
-5
0
5
10
15
20
^{222}Rn (Bq/m^3)
Pa-Ps (hPa)