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PRELIMINARY RESULTS FROM RADON OBSERVATION AT SYOWA STATION, ANTARCTICA, DURING 1996

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Abstract: The concentration of ^{222}Rn in surface air was measured for the first time at Syowa Station, Antarctica in the period September 1996–January 1997 by an electrostatic collection method.

The concentration of ^{222}Rn was higher in the first half than the latter half of the monitoring period. Daily mean ^{222}Rn concentration averaged 270 mBq/m^3 for the first half of the period and 150 mBq/m^3 for the latter half of the period. The maximum daily mean ^{222}Rn concentration was 630 mBq/m^3, and the minimum concentration was 70 mBq/m^3. The daily mean values were higher than those measurement in previous studies in Antarctica.

Remarkable ^{222}Rn concentration increases (radon storms), which accompanied cyclones, were observed twice during the austral spring at Syowa Station. The first radon storm occurred on 18–21 September and the second on 16–20 October 1996. The daily mean ^{222}Rn concentrations in the first and second radon storms attained 530 mBq/m^3 and 630 mBq/m^3 respectively. On the other hand, the hourly mean ^{222}Rn concentration attained 1200 mBq/m^3 in the second event. This implies that the ^{222}Rn concentrations vary on a short time scale. The main feature of the ^{222}Rn concentration record corresponds to intense mixing of air masses from mid-latitude continents to Antarctica. This implies that the variations of ^{222}Rn may be related to the wind direction. The concentrations of ^{222}Rn seem to increase with southerly wind and decrease with northerly or northeasterly wind.

1. Introduction

Antarctica has the cleanest atmosphere of any continent. The radioactive noble gas ^{222}Rn (radon) is chemically inert and disappears from the atmosphere only by radioactive decay with a 3.8 day half-life period. Since Antarctica has no regional sources of ^{222}Rn except for local outcrops, short-lived ^{222}Rn in the Antarctic atmosphere must be transported from outside. So,^{222}Rn can be useful as a tracer for continental air masses which contain various kinds of materials.

LOCKHART (1960), LOCKHART *et al.* (1966), WILKNISS *et al.* (1974), MAENHAUT *et al.* (1979), POLIAN *et al.* (1986), PEREIRA *et al.* (1988, 1992), PEREIRA (1990), HEIMANN *et al.* (1990), WYPUTTA (1997) have reported ^{222}Rn concentrations in Antarctica. Most ^{222}Rn concentrations are 0.01 to 0.03 Bq/m^3. But concentrations of 0.4 Bq/m^3 (PEREIRA

et al., 1988) and 0.08 Bq/m^3 (WYPUTTA, 1997) have been observed in the radon storm which is defined as an abrupt increase of ^{222}Rn with a cyclone.

Extremely high ^{222}Rn concentrations were first described by LAMBERT *et al.* (1970) as "radonic storms". Since then, increases of ^{222}Rn with increase of temperature and wind speed, and decrease of atmospheric pressure, have often been described. POLIAN *et al.* (1986) called this phenomenon "radonic storms", PEREIRA *et al.* (1988) "radon surges", and WYPUTTA (1997) "radon storms". The abrupt rise of ^{222}Rn concentration is interpreted as a transport of an air mass with high ^{222}Rn content of continental origin to Antarctica by means of a cyclone (LAMBERT *et al.*, 1970; WILKNISS *et al.*, 1974; PEREIRA *et al.*, 1988; WYPUTTA, 1997).

In contrast with the ^{222}Rn concentrations in Antarctica, YAMANISHI *et al.* (1991) observed outdoor ^{222}Rn concentrations in Japan of 3 to 12 Bq/m^3. The ^{222}Rn concentrations in the surface air of Antarctica are far less than those in Japan by a factor of 1/100 to 1/1000.

We used a sensitive radon detector, originally designed for low-level measurement, and estimated the ^{222}Rn concentrations for the first time at Syowa Station (69°00′S, 39°35′E) (referred to as SYO), Antarctica in 1996. We found radon storms at least two times in the austral spring of 1996.

In this paper, we focus on ^{222}Rn observations in the austral spring of 1996. First, we describe the radon detector and the observation system. Then we discuss the variations of ^{222}Rn and the possible relationship between radon storms and cyclones which passed over SYO.

2. Sensitive Radon Detector

The methods of radon detection used in this study are electrostatic collection of daughter nuclei of ^{222}Rn, and α-spectrometry using a PIN photodiode.

Figure 1 shows a schematic view of a sensitive atmospheric radon detector developed for continuous monitoring of low level radon concentration in the Superkamiokande experiment (TASAKA, 1996). It consists of a cylindrical stainless steel vessel with electrolytic polish, a PIN photodiode, a high voltage divider, an amplifier circuit, and a feedthrough. The negative high voltage is supplied to the P-layer of the photodiode, and the vessel is grounded. Then an electric field is produced in the vessel. The positively ionized daughter nuclei are collected on the surface of the photodiode and their α-activity is spectrometrically measured.

The dimensions of the stainless steel vessel are 50 cm in diameter and 35 cm in height, and the volume is 68.7 *l*. In order to keep the background at low level, the inside of the vessel is electrolytically polished. The PIN photodiode is electrically isolated from the stainless steel vessel by a ceramic feedthrough.

The PIN photodiode is a Hamamatsu Photonics S3204-06 with passivation finish. The glass cover of the diode is removed to measure the energy of α particles. The detection area, sensitive thickness, and capacity are 16 mm$\times$16 mm, 500 μm and 80 pF, respectively. A typical α-spectrum is shown in Fig. 2. In order to measure ^{222}Rn concentrations, only the ^{214}Po peak is used because that energy region has few noise events, no other overlapping α peaks, and higher collection efficiency of ^{214}Po than

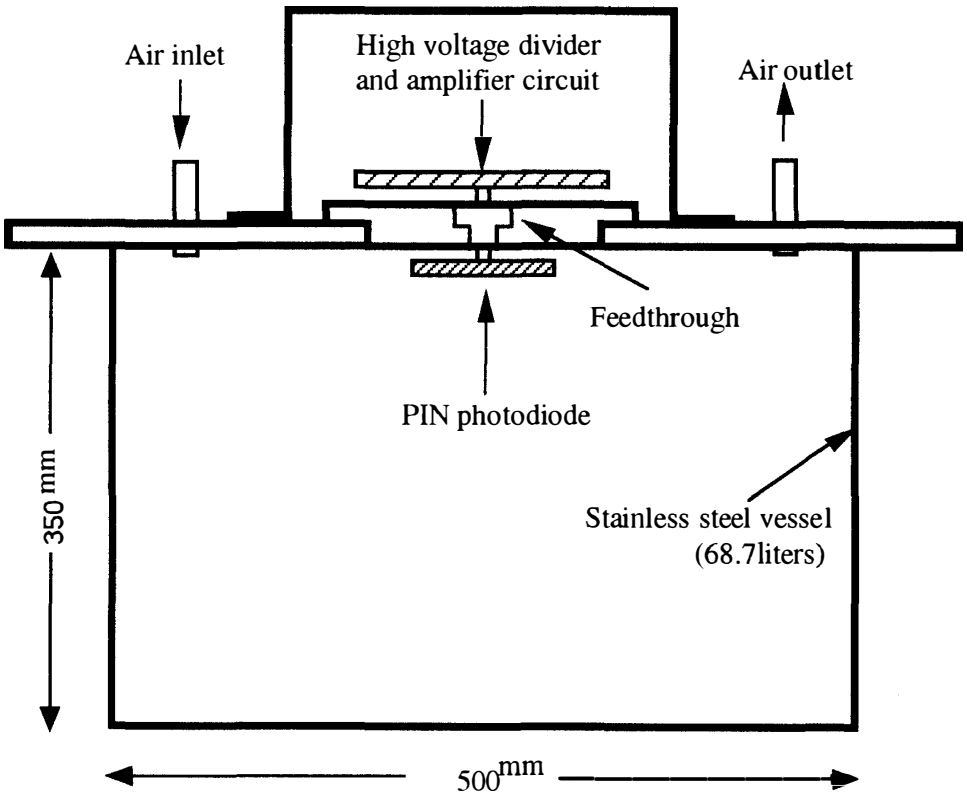

Fig. 1. A schematic view of a high sensitivity detector to measure the radon concentration in air.

^{218}Po on the diode. The signal region of ^{214}Po decay is from 125 to 156 ADC channel for the measurement of ^{222}Rn concentration at SYO (Fig. 2). It takes a few hours to reach radio-equilibrium for ^{214}Po.

Concerning ^{218}Po atoms (one of the daughter nuclei of ^{222}Rn), it is known that more than 90% of ^{218}Po atoms tend to become positively charged. The neutralization effect of ^{218}Po atoms has been pointed out in the electrostatic collection method (KOTRAPPA *et al.*, 1981; BUSIGIN *et al.*, 1981; CHU and HOPKE, 1988; HOWARD *et al.*, 1991). So, the humidity dependence of the calibration factor was carefully studied.

Figure 3 shows a schematic diagram of the calibration system for the atmospheric radon detector. The detector is connected to a radon source, an ionization chamber, a syringe, and a vapor trap. A small rock containing radium stored in a glass vessel of 3 *l* is used for the radon source. The ionization chamber is used as the reference device to measure radon concentrations in this system. We used a 1.5 *l* ionization chamber made by Okura Electric Co. Ltd. As international intercomparison of radon concentration measurements using this type of ionization chamber has been carried out at the Environmental Measurements Laboratory of USA, the device is known to be reliable. The absolute humidity in this system was controlled by a vapor trap consisting of a cooler and a vacuum bottle. A barometer, a dew-point meter, a flow meter, and a circulation pump were also included (Fig. 3). They were connected with 1/4 inch stainless steel tubes and rigid nylon tubes. Standard air of class A (Nihon Sanso Co. Ltd)

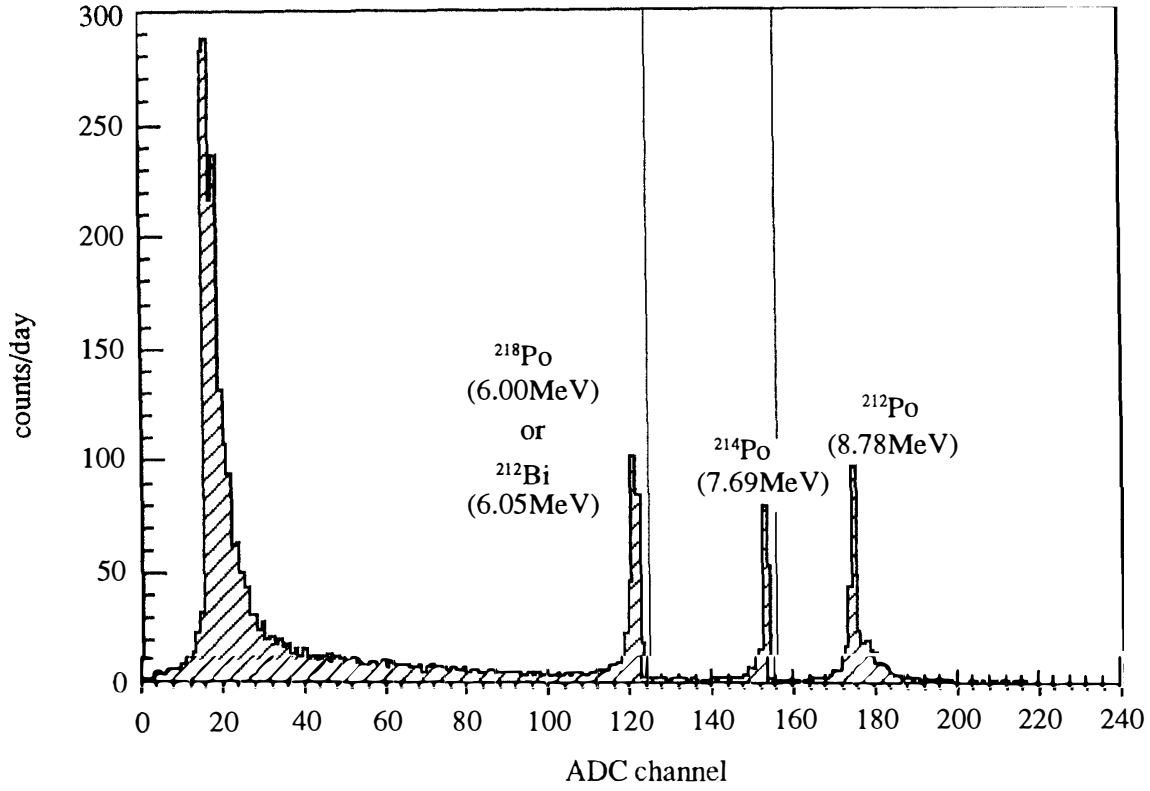

Fig. 2. A typical α-spectrum from ^{222}Rn and ^{220}Rn daughters. This spectrum was obtained at SYO in October 1996. The vertical lines indicate the range of the ADC 125–156 channel. Signals in this range were used as the energy of α particles from ^{214}Po decay. The signals around ADC channel 20 indicate electric noise.

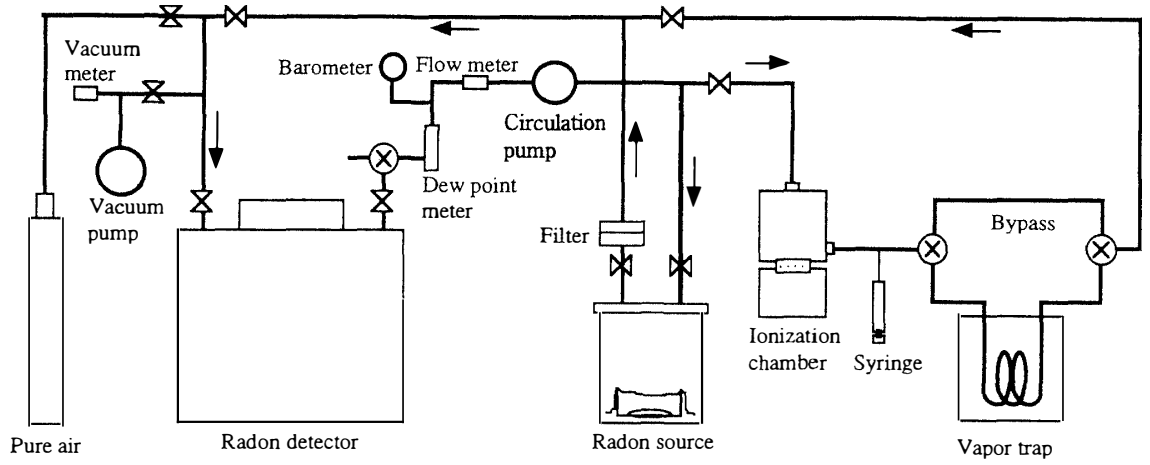

Fig. 3. A schematic diagram of the calibration system for high voltage dependence and humidity dependence measurement.

was used in the experiment with the circulation rate of 3 *l*/min. Before starting calibration, the leak rate was checked; it was less than 1×10^{-5} cm^3/s, which is negligible. The radon concentration was set to 8000 Bq/m^3 during the calibration.

Figure 4 shows the dependence of the calibration factor on high voltages for the radon detector. The absolute humidity was kept at 1.2 ± 0.2 g/m^3 in the experiment. The radon source was introduced into the circulation system to keep the radon concentration constant. We used -1500 V as the supplied high voltage in the calibration because the calibration factor was almost constant and electric noise became larger in the range over -1500 V.

Figure 5 shows the absolute humidity dependence of the calibration factor for the radon detector. Two methods were used to control the absolute humidity. A syringe was used in one method and a vapor trap in the other. The results agreed well (Fig. 5). The absolute humidity dependence on the calibration factors is clearly observed in the region below 1.6 g/m^3, and it becomes constant in the region larger than about 2.0 g/m^3 (Fig. 5). This kind of response of the detector is consistent with the study of CHU and HOPKE (1988). The calibration factors obtained in this measurement are 1.8 ± 0.1 and 0.75 ± 0.04 ((counts/day)/(mBq/m^3)) at observation humidities of 0.08 and 11 g/m^3 respectively.

The detection efficiency of this radon detector at 0.08 g/m^3 is estimated to be

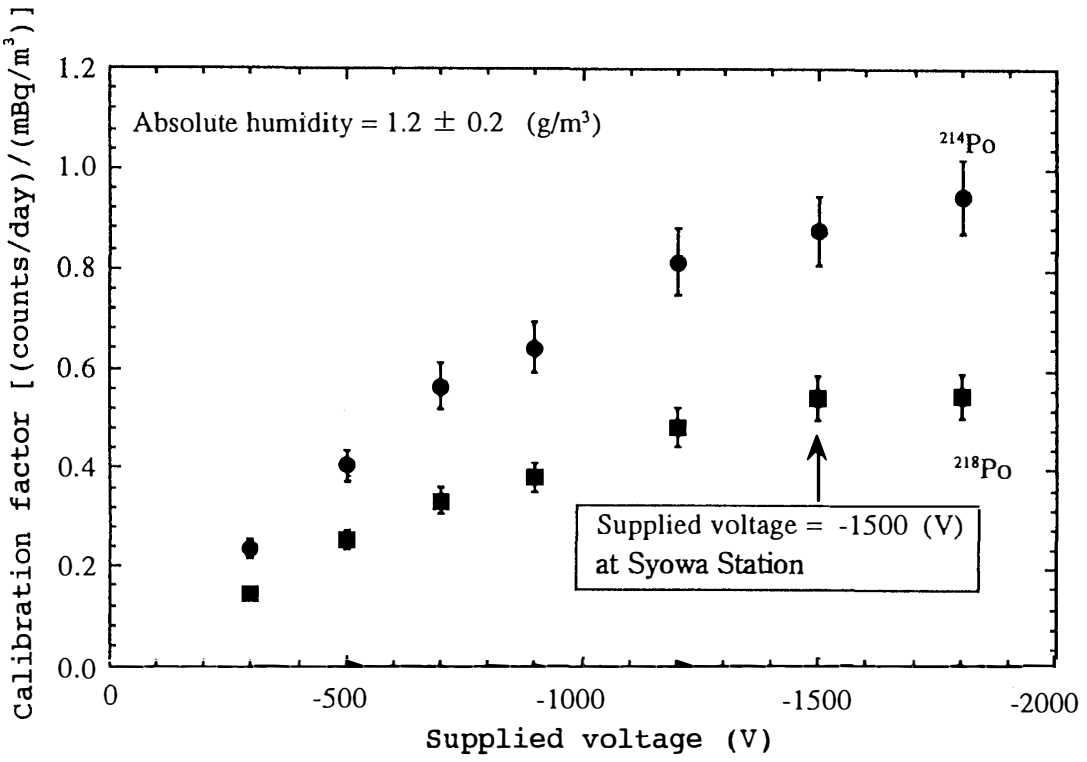

Fig. 4. High voltage dependence on the calibration factors for the high sensitive atmospheric radon detector at the absolute humidity of 1.2 ±0.2 (g/m^3). The supplied high voltage was −1500 V at SYO.

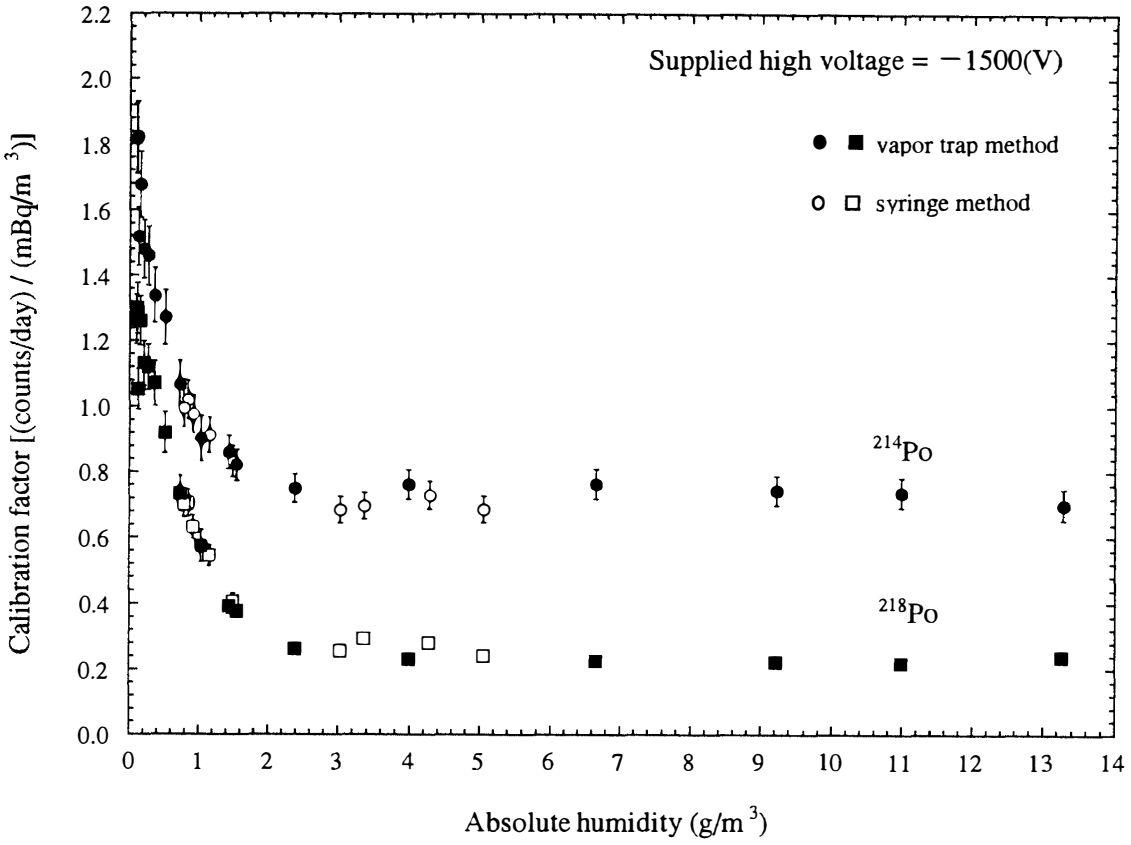

Fig. 5. Absolute humidity dependence on the calibration factors for the high sensitive atmospheric radon detector at supplied high voltage of −1500 V.

30%.

The background level of this radon detector was measured by using purified air for 1 week. The result of this background run is 13.4±2.6 (counts/day) giving detection limit at 1.4 g/m^3 of 27 mBq/m^3 when Currie's method (CURRIE, 1968) is applied.

3. Radon Observation System

Figure 6 schematically shows the radon observation system used at SYO. Surface outdoor air is taken into the system by a pump in the membrane dryer (Sunsep Dry) after passing through a filter (Borosilicate microfiber filters, GF75). The intake of outdoor air is placed at the roof of the laboratory, about 5 m above the ground. The intake tube on the roof is a stainless steel tube, 1 m in length and 1/2 inches in diameter, extending horizontally in the air. The intake tube is connected to a 1/2 inch synflex tube about 10 m in length, followed by a copper tube about 3 m in length and 1/4 inches in diameter. The copper tube is soaked in a water bath. Thus, the sampled air is warmed up to 20°C to decrease humidity, and also the moisture is removed by the membrane dryer. After that, the air is sent through the thermometer, the hygrometer, and the flow meter, and finally introduced into the detector. The whole length of the

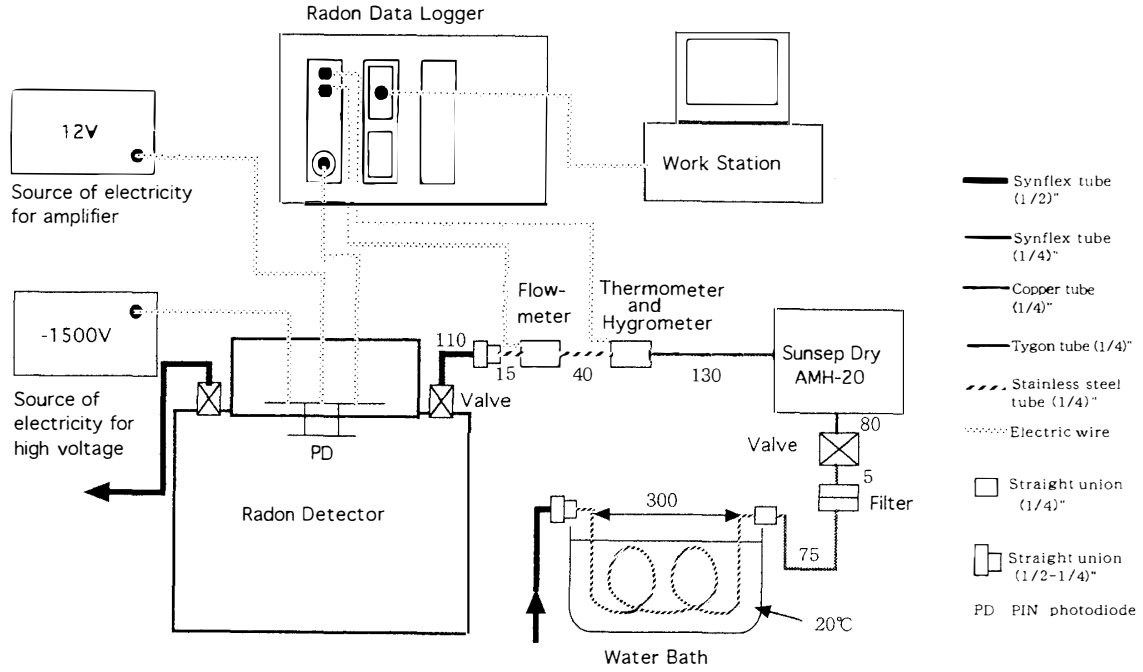

Fig. 6. A schematic diagram for the radon observation system at SYO. The total intake tube line is about 10 m long.

tube is about 18 m. This long intake tube line eliminates the short-lived radon isotopes, ^{220}Rn and ^{219}Rn (PEREIRA *et al.*, 1988). The signals from the detector are amplified and then converted to numerical data in the radon data logger. The digitized data are transmitted to a workstation once per 10 min and stored on a hard disk.

Although several cyclones (blizzards) passed over the station every month, radon observations were not obstructed by choking of intake tube with snow. The water bath, which controls temperature of intake air to be 20°C, is thought to prevent choking and clogging of the sampling line with snow and ice.

Outdoor surface air was continuously introduced into the radon detector at a flow rate of about 3.8 *l*/min. The ventilation rate was 3.4 times/hour. The absolute humidity in the observation system was (1.42±0.05) g/m^3 during the period July–October 1996 and larger than 1.6 g/m^3 during the period November 1996–January 1997. A preliminary calibration factor of 0.73 ((counts/day)/(mBq/m^3)) was applied in this study. The error due to the difference of the calibration factors was less than 7%.

4. Results and Discussion

4.1. Daily mean ^{222}Rn concentration

The measured ^{222}Rn concentrations are shown in Fig. 7. The histogram shows variations of daily mean of ^{222}Rn concentration from 1 September 1996 to 31 January 1997.

There are several data gaps. The largest one is the period 27 September–7 October 1996. This data gap is due to trouble in the system. Six data gaps are also shown in

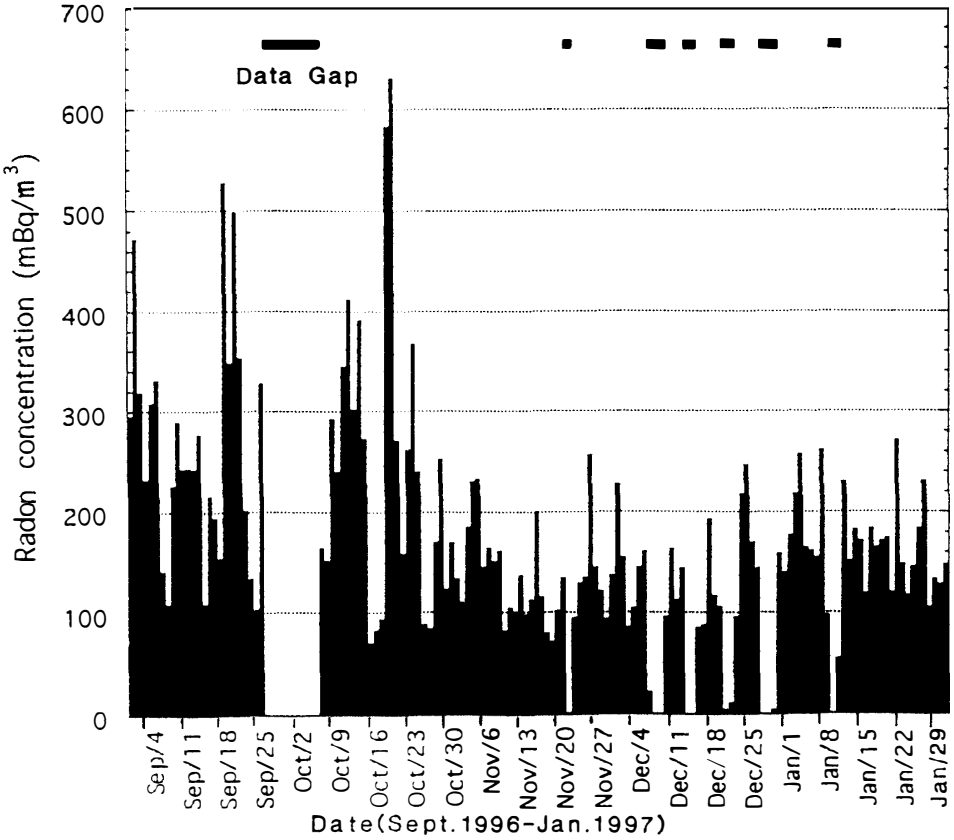

Fig. 7. The daily mean variations of ^{222}Rn concentrations during the period 1 September 1996–31 January 1997. Thick bars indicate data gaps during the monitoring period. Maximum daily mean concentration is about 600 mBq/m^3.

Fig. 7 after 22 November 1996. These are due to temporary suspension for other reasons.

The daily mean of ^{222}Rn concentration has the maximum value of over 600 mBq/m^3 on 20 October 1996. Although there is a large data gap in Fig. 7, there seems to be a difference between the period 1 September 1996–25 October 1996 and the period 26 October–31 January 1997.

Notwithstanding some spike-like records, the mean ^{222}Rn concentration before an abrupt increase (before 25 October, 1996) is about 270 mBq/m^3. And the mean ^{222}Rn concentration after an abrupt increase is about 150 mBq/m^3. The mean ^{222}Rn concentration at SYO is higher than the values of previous studies (*e.g.*, 74–115 mBq/m^3 at Little America V (LOCKHART, 1960); 17–74 mBq/m^3 at Terra Adelie (LAMBERT *et al.*, 1970); 11–15 mBq/m^3 at the geographical South Pole (MAENHAUT *et al.*, 1979); 28 mBq/m^3 at Dumont d'Urville (HEIMANN *et al.*, 1990); and 26 mBq/m^3 at Georg-von-Neumayer (WYPUTTA, 1997)).

Let us now consider the relation between synoptic weather and ^{222}Rn concentration in the following. Extremely low values of ^{222}Rn concentration in September 1996 are considered to be related to cyclones which passed over SYO. These values were

recorded on 7, 8, 15, 18, 24, and 25 September 1996 (Fig. 7). After middle in October, cyclones passed over SYO frequently, and the same relation was found between ^{222}Rn concentrations and cyclones. The variations of ^{222}Rn concentration before middle in October were larger than after. The reason for this differences left as a topic for future research.

4.2. Case study of radon storm at SYO

Abrupt increases of ^{222}Rn concentration apparently accompanying cyclones are recognized in two events in Fig. 7.

The first event occurred in middle in September, 1996. The strong cyclone arrived at SYO on 18 September and departed on 21 September. Extremely high concentrations of ^{222}Rn, reaching 500 mBq/m^3 or more, were recorded during this event. Mean wind speeds per day were 10.8, 22.8, 23.1, and 6.5 m/s, on 18, 19, 20, and 21 September, respectively (Table 1). The north-easterly or east-northeasterly wind was usually dominant in cyclones at SYO. After a cyclone, the southerly wind would bring extremely high ^{222}Rn concentration. The minimum atmospheric pressure and maximum temperature were 982.8 hPa and −6.0°C respectively (Table 1).

The second event occurred about one month later, during 16–20 October 1996. The maximum ^{222}Rn concentration attained 600 mBq/m^3 or more. Mean wind speeds per day were 18.7, 11.8, 20.7, 9.5, and 1.3 m/s on 16, 17, 18, 19, and 20 October 1996, respectively (Table 1). The northeasterly or east-northeasterly wind changed to a southeasterly wind at 1700 LT on 19 October 1996. The ^{222}Rn concentration increased abruptly at the same time and decreased after the wind direction changed to the north-northeast at 0300 LT on 20 October 1996. The minimum atmospheric pressure and the maximum temperature were 958.5 hPa and −8.1°C, respectively (Table 1).

Figure 8 shows variations of hourly mean ^{222}Rn concentration during the period 7–30 October 1996. WYPUTTA (1997) noted that the main feature of the concentration record was the existence of rapid and sharp ^{222}Rn episodes. The same feature is found in our measurements (Fig. 8). The maximum ^{222}Rn concentration reached 1200

Table 1. Average daily weather in the periods 18–21 September and 15–20 October 1996.

Date	Pressure hPa	Temperature °C	Relative humidity %	Wind direction	Wind speed m/s	Weather
9/18	991.3	−13.1	60	NE	10.8	Blizzard
9/19	984.9	−6.8	93	ENE	22.8	Blizzard
9/20	982.9	−6.0	94	ENE	23.1	Blizzard
9/21	982.8	−7.0	87	NE	6.5	Cloudy
10/15	996.3	−15.2	60	NE	3.6	Snowy
10/16	975.4	−9.1	87	NE	18.7	Blizzard
10/17	983.0	−8.1	83	NE	11.8	Snowy
10/18	966.0	−8.3	88	ENE	20.7	Blizzard
10/19	958.5	−9.7	87	NE	9.5	Fine
10/20	961.0	−17.7	67	SE	1.3	Very fine

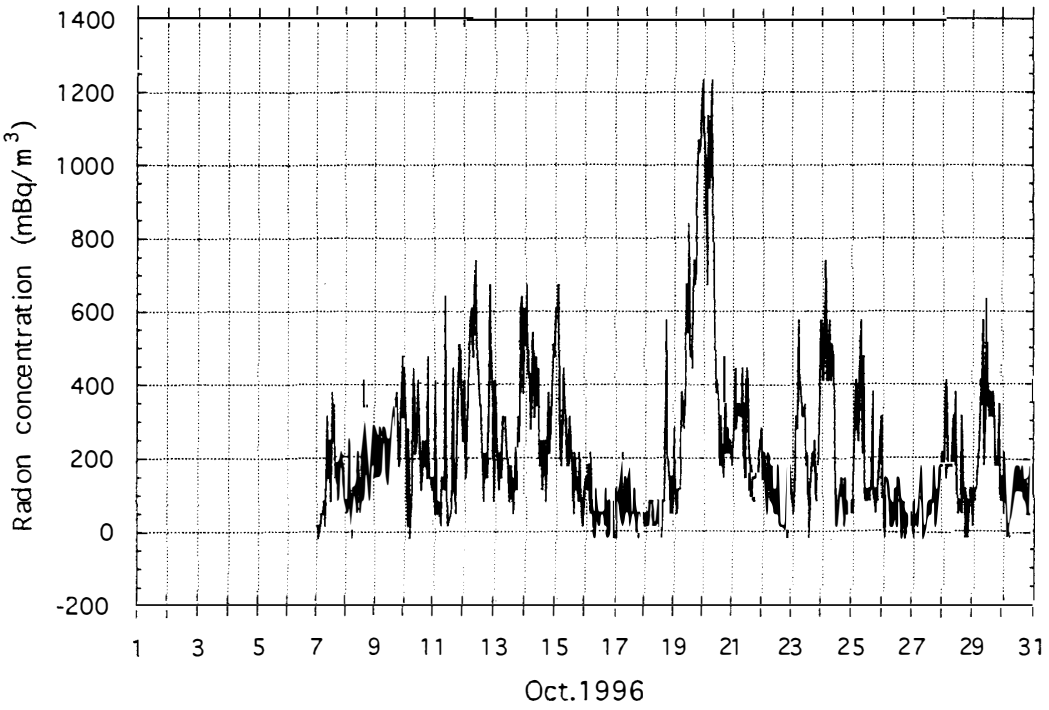

Fig. 8. The hourly mean variations of ^{222}Rn concentrations during October 1996. A data gap is shown from 1 to 6 October 1996. Maximum hourly mean concentration was about 1200 mBq/m^3 on 20 October 1996. Remarkable spike-like episodes of the ^{222}Rn concentration are recognized. The negative values indicated ^{222}Rn concentrations below the detection limit of 0.5083 counts/hour.

mBq/m^3, which was double the daily mean value on the same day (Figs. 7 and 8). This implies that the ^{222}Rn concentrations vary on a short time scale. The variation of ^{222}Rn appears to be related to transport of continental air masses and oceanic matter to Antarctica. Local meteorological features during the period 15–21 October were as follows (Table 1). A cyclone arrived at SYO at 1600 LT on 15 October, the wind direction changed from the south to the north. The ^{222}Rn concentration gradually decreased. The maximum wind speed attained 13.6 m/s at 2356 LT on 15 October. The daily mean wind speeds during the cyclone were 18.7, 11.8, and 20.7 m/s on 16, 17, and 18 October, respectively (Table 1). In this period, the ^{222}Rn concentration was about 100 mBq/m^3 which, about half the value before 14 October 1996. The north-easterly wind changed to the west-southwesterly wind at 1700 LT on 19 October 1996. The wind speed decreased to 2.3 m/s at the same time. The extremely high ^{222}Rn concentration, about 1200 mBq/m^3, coincided with this change of wind direction and decrease of wind speed.

4.3. Variation of ^{222}Rn concentration other than during the two radon storms

The concentration of ^{222}Rn decreases with change of wind direction from the south to the north. Although it is necessary to study the relationship between ^{222}Rn variations and meteorological conditions in detail, the spike-like moderately high ^{222}Rn variations shown in Fig. 8 seem to correspond to the local southerly wind. There are many

outcrops not covered with snow south of SYO. Rocks containing radioactive materials, *e.g.* ^{238}U, such as gneiss, granite, and pegmatite, are exposed there (YANAI *et al.*, 1974a, b). The moderately high ^{222}Rn variations are assumed to be caused by the local southerly wind transporting radon.

The cases other than the two radon storms also show moderately high ^{222}Rn concentration after relatively weak cyclones with wind speed over 10 m/s in many cases (Fig. 7). In those cases, it is considered that the ^{222}Rn concentrations decrease with approach of a cyclone and increase with departure of the cyclone from SYO. It is difficult to explain why spike like high ^{222}Rn variations exist. Further study should be needed to explain this phenomenon.

5. Conclusion

It is found that high ^{222}Rn concentrations are coupled with cyclonic weather events such as the first and the second events during the periods 18–20 September and 16–20 October 1996. It is concluded that radon storms occurred at least two times at SYO in the austral spring of 1996. The great variations of ^{222}Rn can be interpreted as the result of transport of continental air masses to Antarctica by cyclones. Continuing observations of ^{222}Rn at Antarctica will be needed to study polar atmospheric physics and chemistry combined with other physical and chemical observations in the future.

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