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Radon-222 time series measurements in the Antarctic peninsula (1986–1987)

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ABSTRACT

Continuous measurements of radon-222 were conducted at Comandante Ferraz (62°S, 58°W), a Brazilian station in the Antarctic Peninsula, during the winters of 1986 and 1987. The average concentration of radon-222 was 0.026 ± 0.018 Bq m^{-3} in 1986 and 0.014 ± 0.008 Bq m^{-3} in 1987. The higher average of 1986 was attributed to the stronger winds in 1986 compared to those observed in 1987. Large surges of radon reaching up to 0.13 Bq m^{-3} were affected by local radon emissions due to soil defrosting. Bare soils and atmospheric stability also favored local contributions. Radon peaks attributed uniquely to air mass trajectories passing over the South American continent occurred during the 2 years of monitoring. Periodicities of 25 to 30 days in the time series of radon corroborate with previous results obtained by other authors in east Antarctica.

1. Introduction

Radon and its daughter products are used to study long-range transport processes in the troposphere with large uncertainties mainly due to the lack of sufficient number of world-wide well-distributed monitoring stations. Particularly in the Antarctica where the study of troposphere background trace-elements represents a major international goal, only a few isolated radon monitoring stations operate. Measurements of the injection of continental air into the antarctic troposphere employing radon as tracer are known only for the east antarctic and sub-antarctic areas (Polian, 1986). Such measurements were begun about two decades ago by Lambert et al. (1970) and revealed high radon episodes known as "radonic storms" which were attributed to rapid transport of continental air over the oceans. Furthermore, a 28-day periodicity was linked to these high radon events and later studied by Balkanski and Jacob (1988).

The Brazilian station Comandante Ferraz in the west Antarctica, (62°05′S, 58°23.5′W) is located on King George Island, in the South Shetland Islands of the Antarctic Peninsula (Fig. 1). Since 1986, radon-222 has been continuously measured by a novel design automatic radon monitoring system. This paper presents a preliminary comparative study of the first 2 years of operation.

2. Instruments and methods

The radon-222 in the atmosphere was measured by the electrostatic precipitation technique described in Pereira and da Silva (1989). This technique is based on the direct electrostatic deposition in a closed chamber of the locally produced radon decay products (polonium-218, polonium-214) onto a solid state alpha detector, followed by alpha spectrometry. This method does not depend on "secular" radioactive equilibrium assumptions and is free from source errors like aerosol scavenging and ion removal from the atmosphere. Background noise in this technique is very low, about 2.5×10^{-5} decays/s for a 5-day period measurement. Thus, this method allows a detection limit of 10^{-3} Bq m^{-3} at sea level with a

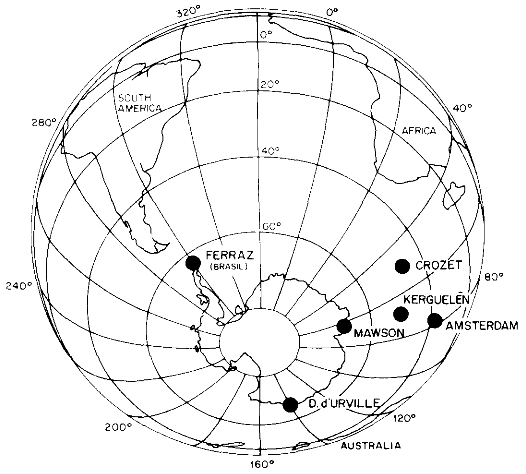

Fig. 1. Map of the southern hemisphere showing the position of some antarctic and subantarctic stations. Frei meteorological station and Ferraz Station are both located on King George Island.

350 lph air intake rate. This low background is instrumentally achieved by using a low-noise silicon alpha particle detector. Further noise reduction is possible by successively sampling and resetting the pulse counter at a relatively high rate (1/h) with respect to the data integration and acquisition rate (1/24 h). Data counts sampled in each counting cycle are tested against the neighbor data counts forward and backward. Values rising above the predicted statistical noise, and lasting less than the instrumental time response (100 min) are transient noise, and are hence deleted.

Instrument calibration was performed by using a standard radium source, and was verified every month by electronic procedures. The observed gain drifts were random and were always lower than the FWHM peak resolution for both the polonium-218 and polonium-214 alpha lines. Systematic errors were very unlikely.

3. Experimental results

Radon concentrations observed in the raw data during 1986 clustered around a mode of 2.0×10^{-2} Bq m^{-3} with an average of $(2.6 \pm 1.8) \times 10^{-2}$ Bq m^{-3}. High radon episodes sometimes attaining 12.6×10^{-2} Bq m^{-3} and known as "radonic storms", occurred regularly during this period. In 1987, the mode was 0.8×10^{-2} Bq m^{-3}, with an average of $(1.4 \pm 0.8) \times 10^{-2}$ Bq m^{-3}, and smaller peaks of only 5.5×10^{-2} Bq m^{-3}.

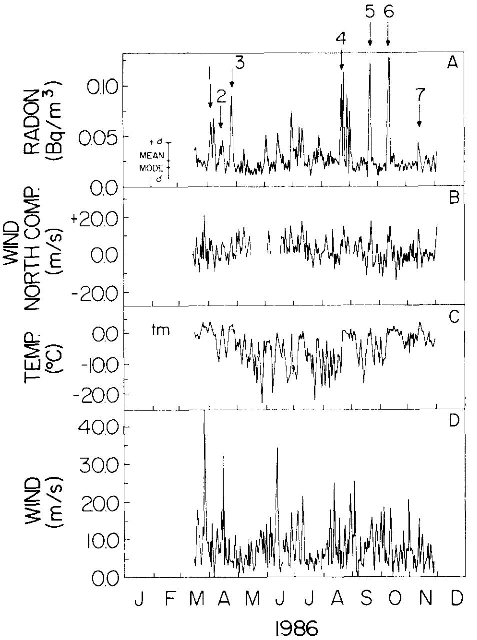

Fig. 2. Radon and meteorological parameters at Ferraz during 1986. (A) Daily radon concentrations, (B) north component of the 850 hPa wind velocities, (C) local air temperature, and (D) surface wind velocities. The dashed curve in (C) is the temperature (tm) for the onset of soil defrosting.

The daily record of radon is shown in Figs. 2A and 3A for the March–November period of 1986 and 1987, respectively. This period corresponds to the winter season at the antarctic station Ferraz. Both 1986 and 1987 daily records of radon showed occasional radon surges in association with increases in the local air temperature (Figs. 2C and 3C) and surface wind (Figs. 2D and 3D) in spite of the fluctuations in the data.

The dashed curves (tm) superimposed in Figs. 2C and 3C are the corresponding air temperature when soil defrost for the first 5 cm depth began to occur. This threshold temperature was derived from local measurements made during 1987. Fig. 4 depicts the soil temperature, showing the sudden change in slope at 0°C when the frozen

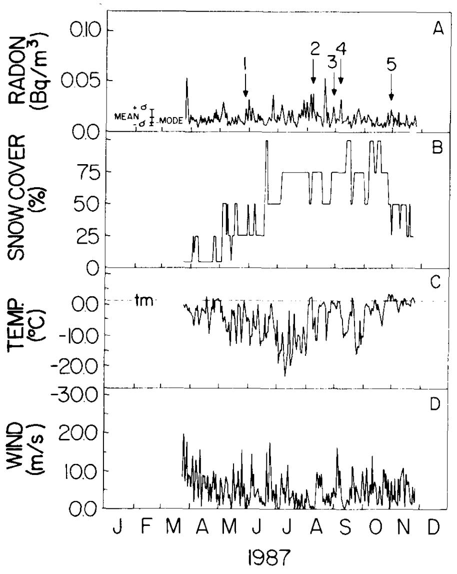

Fig. 3. Radon and meteorological parameters at Ferraz during 1987. (A) Daily radon concentrations, (B) fraction of snow cover in the soil, (C) local air temperature, (D) surface wind velocities.

soil melts. Below this, soil temperature and radon display a small unexplained inverse ratio. Fig. 5 is the plot of air temperature versus soil temperature with the estimated (tm) indicated by the arrow. Only in a few isolated cases, did the air temperature curve exceed (tm) during the winter season at Ferraz. During these events, the measured radon was strongly affected by local radon released from the soil. Local radon emissions hinder data interpretation and thus, radon was not monitored during the period from December to March when substantial snow and permafrost melt occurred.

Fig. 2B is the plot of the north component of 850 hPa winds computed from radiosonde data supplied by Frei meteorological facilities close to Ferraz on the same island. Unfortunately, this data set was not available for the 1987 winter.

During 1986, strong winds sometimes persisting for a few days with gusts up to 40 m s^{-1} occurred frequently. On the other hand, these events were less intense during 1987. Table 1 shows the results of average radon, wind, and temperature for the 2 winter periods at Ferraz. Radon, surface winds, snow cover, and temperature observations were made at the local station.

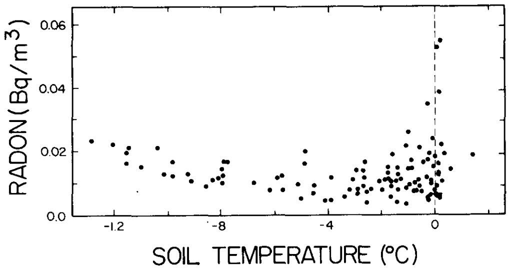

Fig. 4. Soil temperature versus radon during 1987.

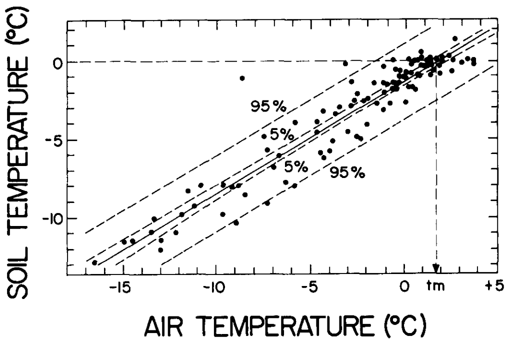

Fig. 5. Air temperature versus soil temperature during 1987. The dashed curve indicates the 5% and 95% confidence levels of the linear fitting. The arrow indicates the air temperature corresponding to the soil defrosting (tm).

The time series of radon concentrations at Ferraz were tested for periodicities by 2 independent methods, the first one applying the Fast Fourier Transform (FFT), the second one used the maximum entropy method of Burg (1967). Both methods produced similar results.

Table 1. *Average results for the two winter periods at Ferraz; radiosonde data for the 1987 winter period is not available*

	1986	1987
Radon (Bq m^{-3})	0.026 ± 0.018	0.014 ± 0.008
Surface winds (m s^{-1})	7.2 ± 6.0	5.0 ± 4.1
850 hPa winds[1] (m s^{-1})		
0° to 90°	9	*
90° to 180°	8	*
180° to 270°	11	*
270° to 360°	14	*
Air temperature (°C)	−5.0 ± 5.9	−4.7 ± 5.9

[1] Data from Frei meteorological station.

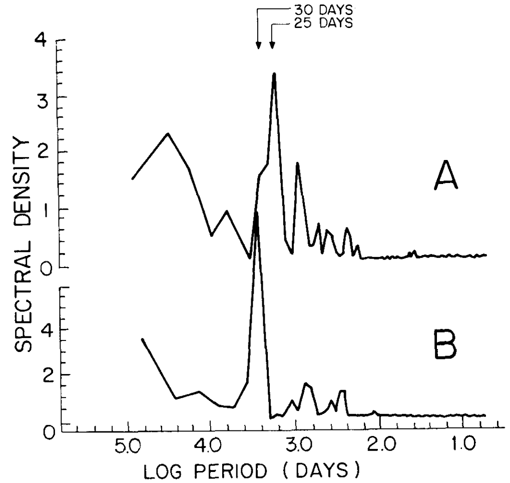

Fig. 6. Periodogram of radon-222 for winter 1986 (A), and 1987 (B), showing the major characteristic cycles. The x-axis is the natural logarithm of the period in days, and the y-axis is proportional to the squared amplitude of sinusoids of the FFT procedure (spectral density). The 2 major peaks correspond to a period of 25 days for 1986, and 30 days for 1987.

The FFT analysis is depicted in Fig. 6 and shows the normalized squared amplitude of the sinusoids (spectral density) versus the natural logarithm of the period (days). Due to the characteristic low count rate statistical fluctuation of the data, the radon time series has a large amount of very high frequency components which are higher than the Nyquist frequency. Here, aliasing can produce signals in the low range of the frequency spectrum, with which our study is most concerned. Smoothing the raw data before the Fourier analysis helps eliminate aliasing. A 6-point moving average smoothing was applied to raw data. The linear trend was also eliminated before the FFT.

The 1986 year was dominated by a 25-day period, while in 1987, the characteristic period was 30 days (arrows), with 95% confidence level of the Kolmogorov-Smirnov test for randomness. Nevertheless, the spectral density was not the same for the whole winter season. During 1986, the spectral density increased from nearly zero to a maximum in late winter (August-September). In 1987, the spectral density was higher than in the previous year, with only a small decreasing trend (Fig. 7).

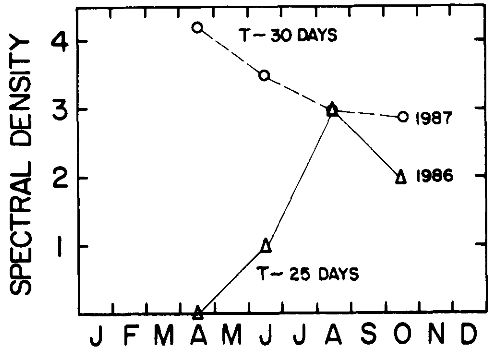

Fig. 7. Spectral density versus month in 1986 and 1987 for the characteristic periods of 25 days and 30 days, respectively.

4. Yearly mean values

In Table 2, the mean radon concentration at Ferraz for the observation period from March through November is given, along with the results from other antarctic and subantarctic stations for the same period, compiled from the literature. Results from antarctic stations Ferraz and Dumond d'Durville are alike, probably as a consequence of the influence further north of the large continental areas of South America and Australia, respectively. Results for the subantarctic islands in the Indian Ocean are comparatively larger than observed at Ferraz, and are

Table 2. *Comparative mean radon-222 concentrations in antarctic and subantarctic regions from March to December*

Station	^{222}Rn (Bq m^{-3})	Site
Ferraz (this work)	2.6×10^{-2} (1986) 1.4×10^{-2} (1987)	Antarctic Peninsula
Dumont d'Urville[1]	2.8×10^{-2}	Terre Adelie
Crozet[1]	5.3×10^{-2}	Indian Ocean
Kerguelen[1]	4.3×10^{-2}	Indian Ocean
Amsterdam[1]	4.1×10^{-2}	Indian Ocean
Mawson[1]	0.9×10^{-2}	Mawson Coast
Ocean lowest estimates[2,3,4]	4.0×10^{-2}	world-wide

[1] Polian et al. (1986).
[2] Schumann (1972).
[3] In: Lambert et al. (1982).
[4] Whittlestone (1985).

close to the ocean estimate. Mawson station presented the lowest result which is consistent with its relative isolation from other continental sources. Thus, the results of Table 2 clearly reflect the latitudinal effect on the mean radon concentration.

Data for the 1986 and 1987 winter periods at Ferraz are very distinct. During 1986, the mean radon concentration and wind intensity were higher by a factor of about two when compared to 1987. However, temperature had virtually the same average of −5°C. Surface winds decreased during 1987 to a relative minimum in mid-winter, with short duration events of higher winds.

5. Daily observations

Events of high radon concentration were generally associated with local increases in temperature and with surface winds. These conditions occurred during the entire winter of 1986. This can be seen by comparing Figs. 2A, 2C and 2D. With a few exceptions, major radon surges were always followed by a rise in air temperature and wind intensity at Ferraz. This was explained in a previous paper (Pereira et al., 1988) by the arrival of the warm sector of cyclonic systems that move eastward through the Drake Passage. These cyclonic wind circulations bring oceanic air masses from the south Pacific Ocean to the Antarctic Peninsula after a short transit over the tip of the South American cone. During the transit over the continent, the air masses are rapidly loaded with radon owing to the high emission bare soils of Patagonia, thus acquiring a full continental identity as far as the radon is concerned.

Results in Table 1 show that high-intensity winds arrived at King George Island, with a prevailing direction from the N–NW sector, during the 1986 winter. The computed north component of the 850 hPa winds plotted in Fig. 2B exhibits in all but a few cases the high-radon events linked to relative increases of this wind component.

The winter of 1987 showed less pronounced associations between radon, temperature and wind, owing to the poorer counting statistics/higher noise of the radon measurements.

Assuming that the average yearly emissions of radon from local sources were the same for the 2 years of data acquisition, it is possible to conclude that the higher average radon obtained for 1986 was not a consequence of a higher frequency of occurrence of favorable deep cyclonic systems during the 1986 winter, for 2 reasons. (1) The associated energy exchanges due to this transport of mass from lower latitudes produced no measurable difference in the local mean temperature as indicated by virtually the same yearly averages in Table 1 for the two winter periods. (2) The occurrence of these radon surges at Ferraz had a cyclic nature with comparable frequencies for both winters (see Fig. 6). A periodicity of 25 days was found for the 1986 winter and a 30-day periodicity for the winter of 1987. Thus, it is unlikely that the 2-fold difference in the mean radon measured at Ferraz could be supported by such a small difference in periodicity. It probably results from differences between the preferred routes of low pressure centers through the Drake Passage from one year to another. A thorough study of these routes based on weather satellite pictures would be of great help in an attempt to understand this difference.

6. Local emissions versus continental radon

The high radon events or "radonic storms" that occurred at air temperatures higher than tm are to some degree affected by local emissions of radon due to soil defrost. These events are indicated by numbered arrows in Figs. 2A and 3A. An example of a "radonic storm", produced almost entirely by local radon emissions in the event no. 3, is shown in Fig. 2A. It occurred when winds were very low, below 3 m s^{-1}, and temperatures were well above tm. Nevertheless, these conditions alone cannot explain such large excursions in the radon record, because the concentrations decrease to baseline levels short after the peak, although the temperature may remain above tm for many days. This is the case in the event no. 6 in Fig. 2A. Very stable atmospheric conditions near ground and bare soils can also enhance high radon events as is illustrated by the example of the peak just after event no. 1 in Fig. 2A. Here, surface wind velocities decreased to zero after a long period of high temperatures ($t > \mathrm{tm}$) resulting in the growth of radon due to the absence of vertical mixing and horizontal mass divergency.

The soil melt contributions to high radon episodes are most probably a very transient phenomenon during which the soil gas radon trapped by the frozen soil increases to equilibrium or near equilibrium with the radium content of the soil. When this frozen soil cap melts, it releases a surge of radon that may exceed the normal steady-state soil flux by orders of magnitude. Thus, soils subjected to temperatures above tm for long periods will not affect the measurements because they have already been relieved of their major radon burden. Fig. 4 illustrates this phenomenon. Above the discontinuity at 0°C in Fig. 4, radon decreases to values found symmetrically below this temperature.

Owing to this effect, the radon records of 1986 and 1987 show no signs of seasonal trend. Radon measurements are also not affected by the snow cover as seen in Fig. 3B. All this corroborates with the hypothesis that most of the radon measured at Ferraz cannot be accounted for by the steady-state local soil emission. Instead, it is imported from elsewhere.

Individual high radon episodes, however, are in many cases affected by local surges from soil defrosting and should not be taken into account in a tracer study without proper corrections. These corrections can be made by simultaneous measurements of the other short-lived radon isotopes which cannot be transported for large distances from their production sites. A new system incorporating these simultaneous measurements is scheduled to begin operation at Ferraz during the 1990 summer season.

7. Conclusions

Radon concentrations measured at the Brazilian Ferraz station in the Antarctic Peninsula averaged $(2.6 \pm 1.8) \times 10^{-2}$ Bq m^{-3} during the winter period of 1986 and $(1.4 \pm 0.8) \times 10^{-2}$ Bq m^{-3} for the winter period of 1987. Results for the 2 consecutive years were very distinct. Mean radon concentration was higher for the 1986 winter period when the mean wind intensity was also high. Mean temperatures did not present a significant difference for these 2 periods. The comparative analysis of the time series, however, showed radon surges correlating well with air temperature rises and with high local winds during 1986, while 1987 showed a more chaotic behavior. Episodes of high radon concentrations at Ferraz were mostly due to the long-range (2000 km) transport of continental air masses from South America by deep cyclonic systems passing through the Drake Passage. In some cases, however, these events could be explained entirely by local emissions of radon from soil defrosting. The radon time series showed a major periodicity of 25 days for 1986 and 30 days for 1987. The global characteristic of the periodicity of atmospheric radon in antarctic and subantarctic regions has long been observed but yet poorly studied. The 28-day periodicity was first observed by Lambert et al. in 1970 in Terre Adelie. Lambert's result is in close agreement with the 25–30 day periodicity of Ferraz. Balkanski and Jacob (1988) have also simulated radon surges in connection with a 28-day periodicity. Periodic mass-energy transport in high latitudes was also observed in meteorological parameters by Kidson (1966) and Webster and Keller (1974). Nevertheless, there is a great need of a co-ordinated effort to perform simultaneous measurements of radon and other trace elements in several widely distributed stations in Antarctica in order to understand the nature of these periodicities in the Southern Hemisphere.

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