1. Publication NO	2. Version	3. Date	5. Distribution		
INPE-3398-PRE/667	2nd*	January/85	☐ Internal ᠓ Externa		
4. Origin F	☐ Restricted				
DGA/DIG A					
6. Key words - selected  ANTARCTICA  RADON  AEROSOL	by the author	^(s)			
7. U.D.C.: 550.3:539.1	6(99)(047.1)				
8. Title	10. Nº of pages: 21				
ATMOSPHERIC RADON MEASU	11. Last page: 17				
PENINSULA: A PR	12. Revised by				
9. Authorship Enio B. D.J.R. M.B.A.	Nalin Babulal Trivedi  13. Authorized by				
Responsible author	Euro Ble_		Marco Antonio Raupp		

#### 14. Abstract/Notes

During the II Brazilian Scientific Expedition to Antarctica (summer 1983/1984), some preliminary measurements of the sea-level atmospheric radioactivity were performed. Radon, and radon daughter products were measured aboard the Brazilian Navy Ship "Barão de Teffé", along a cruise from King George Island (62°05'S, 58°23'W) in the Antarctic peninsula to the Brazilian coast (30°05'S, 49°42'W). An average of 2.1( $\pm$ 1.2) pCi/m³ was obtained for radon in the Antarctic peninsula region. The Drake Passage contributed with only 1.3( $\pm$ 0.7) pCi/m³. Large short term fluctuations superimposed on a slow but steady concentration decrease with time emphasized the need of additional long-term data for more thorough studies. The radon daughters collected on aerosol filters kept the same long-term concentration decrease with time but no short-term correlations were found. INAA performed on returned filters confirmed the expected preferential maritime origin of these aerosols with an average Cl/Na of 1.83.

\*Revised work in September 1985.

15. Remarks This work was partially supported by the FNDCT, under contract FINEP 537/CT and CIRM, subproject 9586. This work was present in the "Simpó sio Nacional do Programa Antártico", during Oct. 22-26, 1984, in São Paulo, and submitted for publication in the "Anais da Academia Bras. de Ciências".

# ATMOSPHERIC RADON MEASUREMENTS IN THE ANTARCTIC PENINSULA: A PRELIMINARY REPORT\*

E.B. Pereira and D.J.R. Nordemann

Instituto de Pesquisas Espaciais - INPE

Conselho Nacional de Desenvolvimento Científico e Tecnológico - CNPq

12200 São José dos Campos, SP, Brasil

M.B.A. Vasconcellos

Instituto de Pesquisas Energéticas Nucleares - IPEN

Cidade Universitária, C.P. 11049 (Pinheiros)

05508 - São Paulo, SP

<sup>\*</sup> This work was partially supported by a grant from CIRM - Programa Antartico Brasileiro, nº 9586.

- 1. ATMOSPHERIC RADON MEASUREMENTS IN THE ANTARCTIC PENINSULA:
  A PRELIMINARY REPORT
- Enio Bueno Pereira
   Daniel Jean Roger Nordemann
   Marina B.A. Vasconcellos (\*)
- Instituto de Pesquisas Espaciais INPE
   Conselho Nacional de Desenvolvimento Científico e Tecnológico CNPq
   C.P. 515, 12200 São José dos Campos, SP.
- (\*)Instituto de Pesquisas Energeticas Nucleares IPEN

  Cidade Universitária, C.P. 11049 (Pinheiros), 05508 São Paulo, SP.
- 4. Ministério da Marinha Comissão Interministerial para os Recursos do Mar - CIRM Programa Antártico Brasileiro Subprojeto 9586
- 5. Enio Bueno Pereira
  Instituto de Pesquisas Espaciais INPE
  C.P. 515
  12200 São José dos Campos, SP
- 6. ANTARCTICA, RADON, AEROSOL
- 7. ATMOSPHERIC RADON MEASUREMENTS

#### **ABSTRACT**

During the II Brazilian Scientific Expedition to Antarctica (summer 1983/1984), some preliminary measurements of the sea-level atmospheric radioactivity were performed. Radon, and radon daughter products were measured aboard the Brazilian Navy Ship "Barão de Teffé", along a cruise from King George Island (62° 05'S, 58° 23'W) in the Antarctic peninsula to the Brazilian coast (30° 05'S, 49° 42'W). An average of  $2.1(\pm~1.2)~\rm pCi/m^3~\rm was~obtained~for~radon~in~the~Antarctic$  peninsula region. The Drake Passage contributed with only  $1.3(\pm~0.7)\rm pCi/m^3$ . Large short term fluctuations superimposed on a slow but steady concentration decrease with time emphasized the need of additional long-term data for more thorough studies. The radon daughters collected on aerosol filters kept the same long-term concentration decrease with time but no short-term correlations were found. INAA performed on returned filters confirmed the expected preferential maritime origin of these aerosols with an average C/Na of 1.83.

#### INTRODUCTION

Studies of radon and radon daughter concentrations in the atmosphere are of particular interest because of their radioactive (Figure 1) properties. The 3.82 day half-life of radon-222 associated with its chemical inertness may provide a time-scale for many diffusion and transport processes of trace gases in the atmosphere (Birot et al., 1970; Reiter, 1978; Druilhet et al., 1980; Larson and Bressan, 1980). Furthermore, radon daughters which are attached to the atmospheric aerosols furnish a major tool to trace the behavior and residence time of the atmospheric solid fraction (Turekian et al, 1977; 1981; Nevissi and Schell, 1980; Lambert et al., 1982; Schneider et al., 1983). Besides these and other fundamental applications of radon and radon daughters in the atmosphere, such studies may allow a better understanding of the very important problem of the anthropogenetic radioactive contamination of the atmosphere (Lambert, 1984).

The Antarctic continent has long been viewed as an oceanic area, as far as most atmospheric trace constituents are concerned. This is attributed to the almost negligible fraction of exposed continental surface as compared to total glaciated land area, and also to the reduced human activities in the region. Thus, the atmospheric radioactivity and the trace-element composition at the Antarctic latitudes are most probably an averaged result of several continental sources spread over the entire Earth. This simplifying "global" hypothesis has been used as a starting point for data interpretation by many geoscientists (Lockhart et al., 1966; Lambert et al., 1970; Aristarian et al., 1982; Pourchet et

al., 1983). In view of these characteristics, the Antarctic continent remains as one of the most recommended site for global studies of many atmospheric trace constituents, including the radioactive ones.

This paper reports the first of a series of concentration data for trace elements in the Antarctic atmosphere, that will follow the establishment of the first Brazilian permanent station for atmospheric studies in Antarctic by 1985. In this preliminary phase of the work, radon and radon daughters data were collected, along with supplemental wind, temperature, barometric pressure and major aerosol constituents.

#### **METHODS**

A novel, author patented, design of a mobile true-radon gas collector and measuring device (Pereira et al., 1984) was set up on board the Brazilian Navy oceanography ship Barão de Teffé. In this experimental set up the outside air is continuously sampled through a membrane pump at a constant flow of 0.3 m³ h $^{-1}$ . After passing through a filter stage (Millipore type MF, 0.45µm), the "particle free" air enters the precipitation chamber. This 42 liter hemispheric chamber is supplied with a strong radial electrical field in order to promptly collect the radon daughter ions produced by the decay of radon inside the chamber. These ions are collected upon the surface of a 400 mm² semiconductor detector (ORTEC) and have their emitted alpha particles counted by a suitable counting electronics. The counting rate can readily be converted into activity of radon per volume of sampled air (pCi/m³) by a simple multiplicative constant. The aerosol filters are collected

every 12 hours and the alpha particles due to the radon daughters attached to the atmospheric aerosols are counted as well. Next, these filters are carefully sealed inside plastic bags and stored for later Instrumental Neutron Activation Analysis (INAA) of Na,  $C\ell$  and  $A\ell$ . The INAA were performed at the Instituto de Pesquisas Energéticas e Nucleares (IPEN) in São Paulo.

Supplemental local temperature, wind and barometric pressure are also collected along with daily wheather maps furnished by the standard navigation equipment on the ship.

## EXPERIMENTAL RESULTS

The data were collected between February 22 and March 29, 1984; the whole cruise is illustrated in Figure 2. The first part of the measurements was made while the ship was in the neighborhood of King George Island  $(62^{\circ}\ 20^{\circ}\ S,\ 58^{\circ}\ 23^{\circ}\ W)$  and Elephant Island  $(61^{\circ}\ 20^{\circ}\ S,\ 54^{\circ}\ 55^{\circ}\ W)$ , from February 22 to March 9. The second part of the measurements corresponds to the Drake Passage from March 8 to 13. The last part of the data acquisition corresponds to the Atlantic measurements up to the Brazilian coast  $(30^{\circ}\ 05^{\circ}\ S,\ 49^{\circ}\ 42^{\circ}\ W)$ .

Figure 3 shows the radon concentration data averaged over 6 hours and filtered by a computer program in order to eliminate data noise due to electrical and mechanical interference from the ship.

The original raw data were acquired hourly (real time) to provide enough data points for the filtering procedure by the Chauvenet's criterium.

The error bars indicated are the one standard deviation ( $\pm \sigma$ ) for six averaged data points.

Figure 4 is of the total alpha counting for the aerosol filters. Due to the large counting time (12 hours), the filter measurements were very sensitive to the frequent power failures of the ship's laboratory. This problem did not interfere in the radon measurements due to the much shorter counting time employed (1 h). The local temperature, barometric pressure and true wind, were all read by the ship's standard equipment twice a day, at the time the filters were changed.

Table 1 shows the results for the INAA performed on the retrieved aerosol filters. Not all filters were analysed by this technique. Only those collected during the first part of the cruise were selected. To avoid contamination by the ship's engine discharge, we have also neglected those filters for which the relative wind direction could lead the discharge gases into the inlet of the sampling duct. This table also contains the total alpha counting data of the filters and the corresponding atmospheric radon content for comparative purposes

The volume of data colected in this cruise is far from being sufficient to derive any conclusive model for the circulation and balance of atmospheric radioactivity in Antarctica. We will therefore restrict ourselves to point out some of the most remarkable observed features that may deserve further investigation from a more complete set of data.

The first and most evident of these features is the steady trend of decrease of concentration as observed in Figures 3 and 4 for radon and radon daughters, during stages 1 and 2 of the cruise. This concentration decrease has a already been observed in the past by Lockhart et al. (1966). However, a clear explanation for this effect is still to be formulated. It is possible that this phenomenon is associated with the proximity of the southern winter season, when all summer exposed land areas are covered by winter snow. Superimposed on this long term concentration change, there are several short term changes that are probably connected to measoscale meteorological events. Thus, for example, the large radon concentration increase pointed out by the A in Figure 3 corresponds to a sharp increase of barometric pressure associated to a fast decrease in temperature. Other, but not so clear, correlations were also observed and are indicated by B and C. Immediately following the radon anomalies a depression followed by a second peak is allways observed. Surprisingly, wind direction seems to have no influence on the radon and daughters concentrations. On the other hand, the wind intensity shows a close positive correlation with these concentration changes. It is also interesting to observe that these short term radon changes do not keep any clear association with the total alpha counting of the aerosols filters in Figure 4. This can only be true with different processes (whichever they are) controlling the concentration of radon and of radon daughters in the atmosphere. The diurnal modulation, which is normally found in other continental areas, is not observed here because of the litle diurnal variation of the vertical mixing.

The very large peaks observed in Figures 3 and 4 after March 13 correspond to the increased influence of continental air masses from South America. The peak of March 15, for example, was observed when the ship passed at about 50 miles from the Malvinas (Falkland) Islands. A spectrometric analysis of the corresponding aerosol filters showed a very large fraction of radium-226, uranium-238, thorium-232 and some other radioisotopes not commonly found in soil dust. These filters are still under investigation.

On the average, the radon concentration obtained for the Antarctic Peninsula,  $2.1(\pm~1.2)pCi/m^3$ , is in close agreement with data obtained by other authors (0.1 to 4 pCi/m³ after Lambert et al., 1982). The average concentration obtained while crossing the Drake Passage was much lower,  $1.3~(\pm~0.7)~pCi/m³$  and no other references reporting atmospheric radon data for this region were found.

In the INAA of Table 1, the Na and Cl concentrations indicate a maritime origin for the aerosols, while the Al are indicative of a continental origin (soil dust) of the aerosols. From these data it is possible to conclude that the preferential source for the aerosols in the Antarctic Peninsula is maritime. Furthermore, the average Cl to Na ratio is 1.83, more than twice the ratio measured at the South Pole aerosols by Maenhaut and Zoller in 1977 (Cl/Na = 0.8). This ratio is very close to the ratio measured in the Antarctic Peninsula snows (Aristarain et al., 1982) and other coastal reas (Briat et al., 1974).

The A $\ell$  measured as indicator of continental aerosols were in all, but a few cases, below the detection limits of the method employed. Nevertheless, it can be seen in Table 1 that whenever a measurements of A $\ell$  is obtained (March 2 and 8), a relative maximum in the total alpha counting is also observed. This is consistent with a continental origin for the alpha emitters since no correspondent radon maximum was observed.

#### CONCLUSIONS

This reconnaissance work suggests that more data collected over a much larger time interval are necessary to derive any well founded model on the circulation and balance of the atmospheric radiactivity in Antarctica. Nevertheless, a few preliminary conclusions can be darwn:

- 1 The concentrations of radon and radon daughters in the Antarctic Peninsula are not controlled by the same process.
- 2 Weather controlled fluctuations of radon prevail over the diurnal modulation normally observed in the non-glaciated continents.
- 3 The average radon concentration in the Antarctic Peninsula area was  $2.1(\pm 1.2) \text{ pCi/m}^3$  and in the Drake Passage was  $1.3(\pm 0.7) \text{ pCi/m}^3$ .
- 4 A preferential maritime origin was attributed to the aerosols collected in the Antarctic Peninsula.

5 - The measured Cl to Na ratio of 1.83 is very close to this ratio measured by several authors in the Antarctic snows. However, it is twice as large as the ratio measured in the South Pole aerosols.

# **ACKNOWLEDGEMENTS**

We thank Eng. Ailton M. Takashima and Mr. Sylvio L.M. Neto for their collaboration in the experimental setup and data acquisition.

# TABLES

Table 1 - Instrumental Neutron Activation Analysis of Aerosols of the Antarctic Peninsula. The activation time was 10 minutes at a neutron flux of 4 x  $10^{11}$  n/m<sup>2</sup>s. Decay time was 5 to 7 minutes.

## FIGURES

- Figure 1 Decay scheme of <sup>222</sup>Rn with some more important properties.
- Figure 2 Data acquisition route during the II Brazilian mission to Antarctica.
- Figure 3 Radon concentration data for the whole cruise: (1) Antarctic Peninsula, (2) Drake Passage and (3) South Atlantic.
- Figure 4 Total alpha counting performed on the aerosol filters along the cruise.

## REFERENCES

- Aristarain, A.J., Delmas, R.J. & Briat, M. (1982), Snow chemistry on James Ross island (Antarctic Peninsula). *J. Geophys. Res.*, <u>87</u>(13): 11004-11012.
- Birot, A., Adroguer, B. & Fontan, J. (1970), Vertical distribution of radon-222 in the atmosphere and its use for study of exchange in the lower troposphere. *J. Geophys. Res.*, 74(12):2373-2383.
- Briat, M., Boutron, C. & Lorius, C. (1974), Chlorine and sodium content of east Antarctic firm samples. J. Rech. Atmos., 8:895-901.
- Druilhet, A., Guedalia, D. & Fontan, J. (1980), Use of natural radioactive tracers for the determination of vertical excannge in the planetary boundary layer. In: Gesell, T.F. and Lowder, W.M. (eds.), Natural Radiation Environment III. Technial Information Center/U.S.

  Department of Energy, U.S.A., vol. 1, 226-241.
- Lambert, G., Polian, G. & Taupin, D. (1970), Existence of periodicity in radon concentration and in the large-scale circulation at lower altitudes between 40° and 70° south. *J. Geophys. Res.*, <u>75</u>(12):2341-2345.
- Lambert, G., Polian, G., Sanak, J., Ardouin, B., Buisson, A., Jegou, A.
  & Le Roulley, J.C. (1982), Cycle du radon et de ses descendants:
   application ā l'étude des échanges troposphère-stratosphère. Ann.
   Geophys., 38(4):497-537.

- Lambert, G. (1984), La radioactivité atmosphérique. *La Recherche*, <u>15</u> (157):938-948.
- Larson, R.E. & Bressan, D.J. (1980), Radon-222 as an indicator of
   continental air mass boundaries over ocean areas. In Gesell, T.R. &
   Lowder, W.M. (Eds.), Natural Radiation Environment III, Technical
   Information Center/U.S. Department of Energy, U.S.A., vol. 1,
   308-326.
- Lockhart, B.L. Jr., Patterson, L.R. Jr. & Saunders, A.W. Jr. (1966), Airborne radioactivity in Antarctica, *J. Geophys. Res.*, <u>71</u>(8): 1985-1991.
- Maenhaut, W. & Zoller, W.H. (1977), Determination of the chemical composition of the South Pole aerosol by Instrumental Neutron Activation Analysis. J. Radianal Chem., 37:637-650.
- Nevissi, A. & Schell, W.R. (1980), Use of lead-210 and polonium-210 as tracers of atmospheric processes. In: Gesell, T.F. & Lowder, W.M. (Eds.), Natural Radiation Environment III, Technical Information Center/U.S. Department of Energy, U.S.A., vol. 1, 405-414.
- Pereira, E.B., Nordemann, D.J.R., Takashima, A.M., Dutra, L.S.V. & Mantelli Neto, S.L. (1984), Um sistema para monitoração do radônio e seus produtos de decaimento na atmosfera. *Revista Brasileira de Geofisica*, 2(2):59-64.

- Pourchet, M., Pinglot, F. & Lorius, C. (1983), Some meteorological applications of radioactive fallout measurements in Antarctic snows.

  J. Geophys. Res., 88(10):6013-6020.
- Reiter, E.R. (1978), Atmospheric Transport Processes, Part 4:

  Radioactive Tracers. Technical Information Center/U.S. Department of Energy, U.S.A.
- Schneider, J.K., Gagosian, R.B., Cochran, J.K. & Trull, T.W. (1983), Particle size distribution of n-alkanes and  $^{210}$ Pb in aerosols of the coast of Peru. *Nature*, 304(4):429-432.
- Turekian, K.K., Nozaki, K. & Benninger, L.K. (1977), Geochemistry of atmospheric radon and radon products. *Ann. Rev. Earth Planet. Sci.* 5:227-255.
- Turekian, K.K. & Cochran, J.K. (1981), <sup>210</sup>Pv in surface air at Enewetak and The Asian dust flux to the Pacific. *Nature*, 292(6):522-524.

TABLE 1

	Element (pg/m³)			Total alpha	Rn
Date	Na	Cl	Al	(1/12h)	(pCi/m³)
2/26 I	1642	3283	trace	20	2.7
2/27 I	1195	1594	_	18	2.3
2/27 II	764	1176	-	21	1.6
2/28 I	797	<u>-</u>	trace	25	2.0
3/1 II	1669	2346	8.57	29	1.8
3/3 II	694	1272	trace	11	1.4
3/07 I	315	571	-	<b>1</b> 1	1.4
3/07 II	316	884	trace	10	1.4
3/08 I	357	909	trace	11	1.4
3/08 II	728	873	22.8	13	1.4

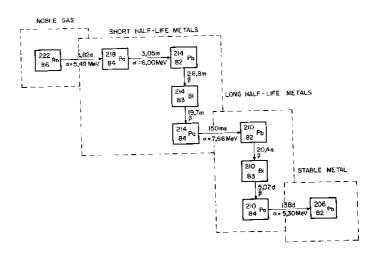


Fig. 1

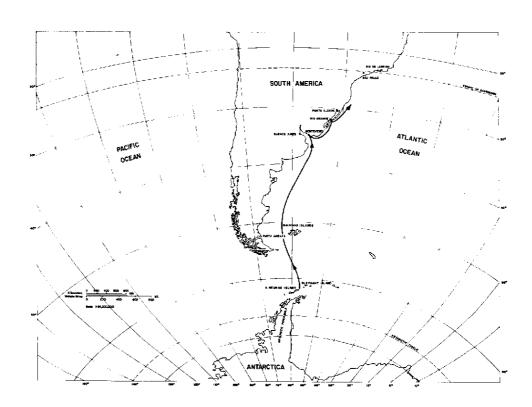


Fig. 2

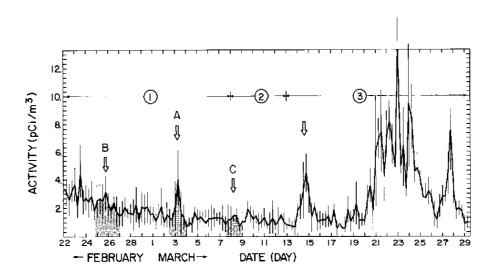


Fig. 3

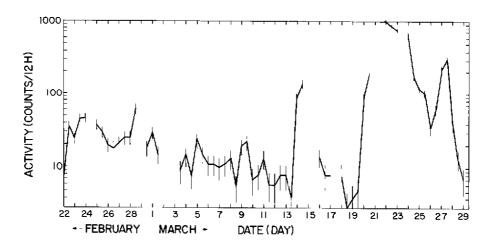


Fig. 4