RADON CONCENTRATIONS PROFILES OVER THE BRAZILIAN AMAZON BASIN DURING WET SEASON

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Atmospheric radon measurements were performed airborne in the Brazilian Amazon Basin during the wet season ABLE-2B experiment. The vertical profiles of radon showed a small decrease of concentration with increasing altitude at a rate varying from 6.5 to 11 x 10^{-2} Bq m^{-3} km^{-1}. The calculation of the flux balance of radon in the troposphere above the Amazon Basin indicated a residual flux at the upper boundary of the measurement domain (6 km) of 0.14 atom cm^{-2} s^{-1}. This residue may be associated with the turbulent transport of radon due to cloud activity. A rough estimate of the cloud mass flux at 6 km was of the order of 0.4 x 10^{-2} kg m^{-2} s^{-1}. The horizontal survey of radon between Belém (1.5°S, 48.5°W) and Manaus (3°S, 60°W) performed at two different levels (300 m and 3000 m) indicated strong enhancement of radon near Santarém (2°S, 54.5°W) at the lower level, and a marked decrease at the higher level flight. It is suggested that mesoscale subsidence in this region inhibited the removal of mixed layer radon by the convective activity.

PERFIS DE CONCENTRAÇÃO DE RADÔNIO SOBRE A BACIA AMAZÔNICA DURANTE A ESTAÇÃO DAS CHUVAS – Medidas de radônio atmosférico foram realizadas na Bacia Amazônica brasileira durante a campanha GTE/ABLE 2B na estação úmida. Os perfis verticais, realizados a bordo da aeronave, mostraram um ligeiro decréscimo da concentração em função da altitude, com uma taxa de variação compreendida entre 6,5 e 11 x 10^{-2} Bq m^{-3} km^{-1}. O cálculo do balanço de radônio na troposfera acima da Bacia Amazônica indicou um fluxo residual no limite superior do domínio de medidas (6 km) de 0,14 átomos cm^{-2} s^{-1}. Este resíduo pode ser associado ao transporte turbulento de radônio devido à atividade das nuvens. Uma estimativa do fluxo de massa de nuvens a 6 km foi da ordem de 0,4 x 10^{-2} s^{-1}. O perfil horizontal de radônio entre Belém (1,5°S, 48,5°W) e Manaus (3°S, 60°W) realizado a dois níveis diferentes (300 m e 3000 m) indicou um forte aumento de radônio perto de Santarém (2°S, 54,5°W) na altitude inferior, e um decréscimo nítido na altitude superior do vôo. É sugerido que a subsidência de mesoescala nesta região inibiu o escape do radônio da camada misturada sob o efeito da atividade convectiva.

INTRODUCTION

The potential of radon as an atmospheric tracer has not been fully explored yet. This is particularly true when dealing with studies of vertical mixing in the free troposphere above the planetary boundary layer. This issue came back to discussion in a publication by Liu et al. (1984) on the basis of a compilation of several vertical concentration profiles in several places of the world. Earlier work by Turekian et al. (1977) and later by Lambert et al. (1982) presented very comprehensive studies on the global balance of radon and radon daughter products in the troposphere with direct implications on the mobilization and transport of trace-elements in the troposphere. Nevertheless, the available literature today reveals an unjustified lack of radon data for tropical regions of the world, particularly for the tropical rain forest areas, where important turbulent mixing associated with moist convection takes place in the lower troposphere.

The NASA's Amazon Boundary Layer Experiment (GTE/ABLE-2B) conducted broad studies on gas exchange and chemical budgets in the Brazilian Amazon rain forest. This paper describes for the first time, results of vertical concentration profiles and flux balance of radon in the troposphere above the Amazon Basin, obtained during the wet season.

INSTRUMENTS AND METHODS

Radon concentration profile measurements were performed in the Brazil Amazon rain forest during the wet season aboard the NASA-429 Electra aircraft.

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Measurements were made with a real time radon-gas electrostatic precipitator described by Pereira et al. (1984, 1989). In this technique the outside air is continuously sampled through a membrane pump and introduced into a 0.042 m^3 hemispheric chamber. The radon daughters produced inside this chamber are mostly (> 80%) positive ions and precipitate through an electric field onto a collecting electrode consisting of a silicon alpha particle detector. Before entering the collection chamber however, the air is filtered in order to remove radon daughters produced in the outside air and attached to aerosol. Data acquisition was made by use of a single channel analyzer centered at the 6 MeV alpha peak of the ^{218}Po. The nuclear pulses within a preset energy range and time interval are recorded onto magnetic cassette tapes. Data is converted to radon concentration by a calibration procedure employing an international radium standard. Data is also corrected for cabin temperature and pressure, as well as for the specific humidity of air, and the results are given in units of local Bq m^{-3} (Bq = 1 decay per second). The efficiency or correspondence between count rates and radon activities of the air was 82.81 cps/(Bq m^{-3}) for typical parameters (pressure and humidity) and for the detector used (600 mm^2 silicon surface barrier detector, ruggedized type, made by EGG-ORTEC) in the conditions of this mission (Pereira & da Silva, 1989).

The response time of the instrument in the employed configuration is of the order of 10 minutes. Thus, the integration time was always adjusted above this limit, restricting the measurements to leveled flights with duration greater or equal to 15 minutes. The estimated noise of the instrument is 2.5 x 10^{-5} counts per second or 10^{-3} Bq m^{-3} at an altitude of about 550 m. With such a low noise and for the efficiency of detection of the instrument, the sensitivity depends mainly on the duration of the measurement, which has to be long enough to achieve high enough count rates for the results to be significant.

VERTICAL RADON CONCENTRATION PROFILES

Several vertical trace-element profiles were performed during the mission, although only in a few cases, acquired radon data were considered statistically significant due to the relatively large integration time of our radon meter when compared to the ascent-descent rate of the airplane.

Figure 1 shows the vertical radon concentration distribution for all flights. The data of Fig. 1 reflects a mean behavior of the vertical radon distribution over the Amazon Basin during the wet season.

The slope of this profile is fairly small, about 6.5 x 10^{-2} Bq m^{-3} km^{-1}, up to 3 km due to the strong turbulent transport by moist and dry turbulence. Above this level, there is an inversion of the slope reflected by a slight but significant (when considering error bars) increase of radon concentration with height. Above 3.5 to 4 km the normal decreasing profile is again established at a larger 11 x 10^{-2} Bq m^{-3} km^{-1} rate, which is consistent with less intense vertical mixing in the atmosphere.

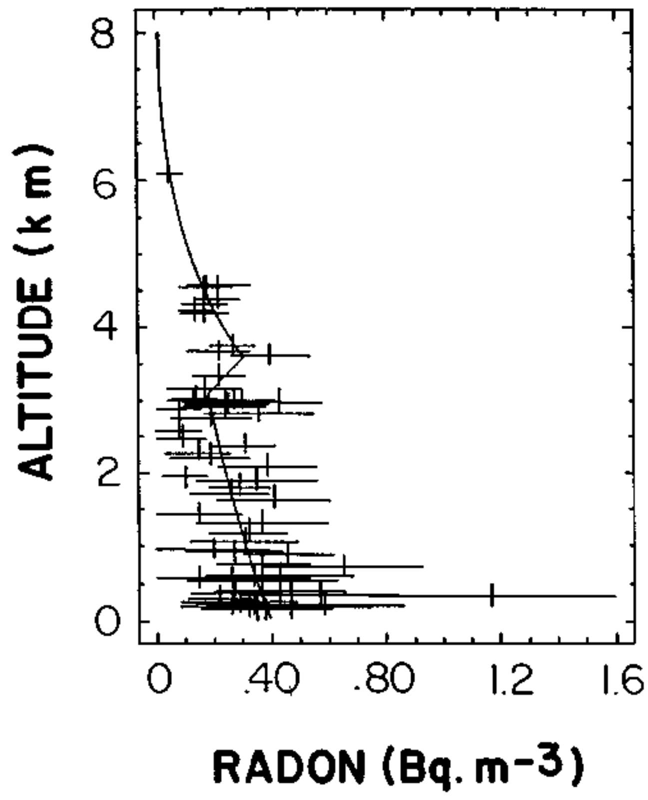

Figure 1. Radon versus geometric altitude for all leveled flights with Δt = 15 minutes.

Figura 1. Radônio em função da altitude geométrica para todos os vôos nivelados para os quais Δt = 15 minutos.

A best fit line through data was adjusted by assuming two distinct sets of data points separate by the discontinuity at 3100-3600 m level.

The vertical temperature and moisture profiles over the Amazon region during ABLE-2B also indicate the presence of a slight temperature and moisture inversion at the 3-4 km level (Harris et al., 1990). This inversion is related to the subsidence which is established between convectively active periods. Except for the intense precipitating periods, shallow cumulus convection is present with cloud top level at about 3-4 km. The shallow cumulus play an important role in the vertical transport of heat and moisture from the mixed layer to the lower levels of the troposphere as discussed by Betts (1976). It is reasonable to assume that the shallow cumulus are also efficient in the transport of trace gases as discussed by Gidel (1983).

The effects of the strong vertical mixing below 4 km can be seen in two distinct vertical profiles for which the experimental conditions were optimized for the long integration time of the equipment. The flight of April, 29 (Fig. 2a, b) took place over "terra firme" or dry forest, about 80 km NE from the city of Manaus. During this flight a well mixed layer existed to about 2.5 km, topped by an inversion marked by a haze layer. The potential and equivalent potential temperature profiles indicated dry mixing to about 1 km and intense cloud induced mixing from 1 km to 2.5 km (Fig. 2b). These conditions are consistent with the

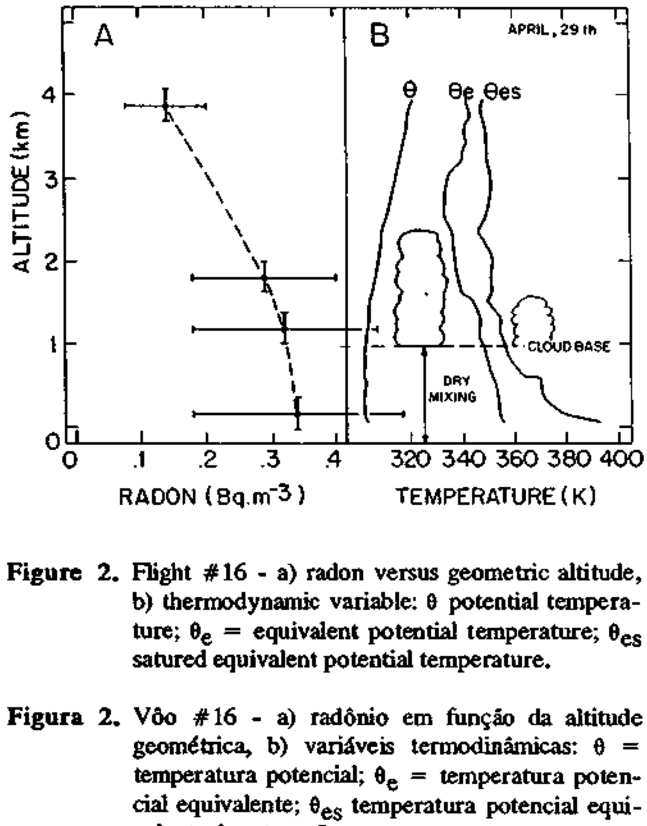

Figure 2. Flight #16 - a) radon versus geometric altitude, b) thermodynamic variable: θ potential temperature; θ_e = equivalent potential temperature; θ_{es} satured equivalent potential temperature.

Figura 2. Vôo #16 - a) radônio em função da altitude geométrica, b) variáveis termodinâmicas: θ = temperatura potencial; θ_e = temperatura potencial equivalente; θ_{es} temperatura potencial equivalente de saturação.

negligible slope of the radon vertical profile up to 2 km, and both radon and other meteorological data are conducive to an intense mixing up to this altitude.

Calculated 3-day isobaric back trajectories (Fig. 4) showed 850 hPa air masses coming from the E while the 500 hPa comes from SW, from the central Brazil plateau where the "cerrado" or a "savana" type of environment predominates. However, the reliability on the back on trajectories is questionable in the presence of active convection since vertical mixing dilutes the air parcels. Also, the back trajectories are computed on isobaric levels, thus ignoring the effect of large-scale ascent or descent of the air parcels. Since the temperature field is very homogeneous on pressure surfaces in the tropical region, we believe that the isobaric trajectory assumption is not as serious as the neglect of the vertical mixing by moist convection.

The flight of May 2nd (Fig. 3a, b), on the other hand, was performed over the "varzea" or wetland, about 200 km E of Manaus. The moist convective activity during this flight, as indicated by the thermodynamic analysis (Fig. 3b) was highly suppressed above 930 hPa (700 m) where the top of

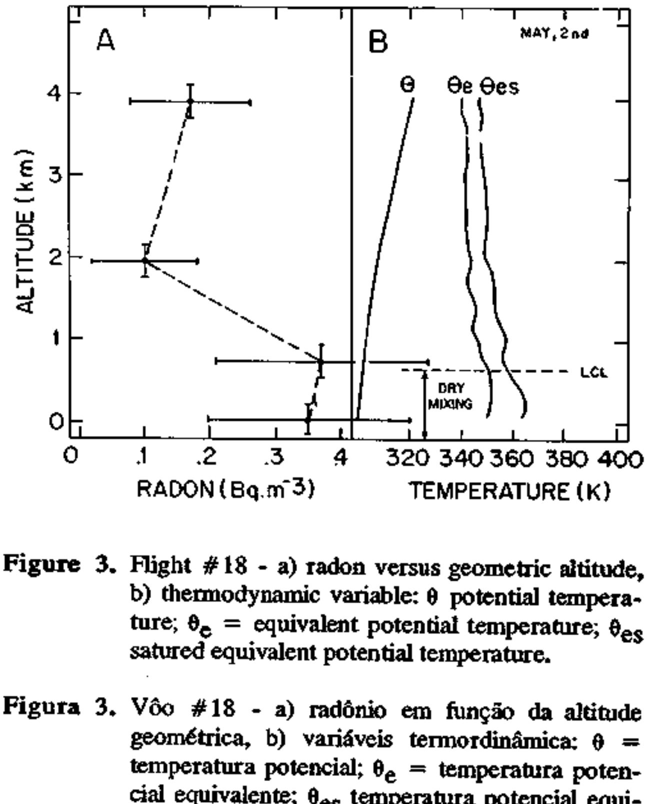

Figure 3. Flight #18 - a) radon versus geometric altitude, b) thermodynamic variable: θ potential temperature; θ_e = equivalent potential temperature; θ_{es} satured equivalent potential temperature.

Figura 3. Vôo #18 - a) radônio em função da altitude geométrica, b) variáveis termordinâmica: θ = temperatura potencial; θ_e = temperatura potencial equivalente; θ_{es} temperatura potencial equivalente de saturação.

the mixed layer was located. The isobaric back trajectories resembled that of the April 29 flight, except that the 500 hPa trajectory showed a more intense southerly component. Radon vertical concentration profiles for this flight indicates intense vertical mixing up to approximately 800 m (dry mixing) topped by a very stable profile up to 2 km.

The air at the 4 km level showed a radon concentration higher than below, with a distinct inverted radon profile. The increase of the radon concentration at higher levels is probably related to a different air mass, characteristic of central South America which is overlaying a stable layer. A low radon concentration in the stable layer is expected since the eddy transports from the mixed layer are inhibited. The radon concentration at the top layer (4 km) is the same for the April 29th and May 2nd profiles suggesting a unique source, possibly from the central Brazilian plateau as indicated by the isobaric trajectory analysis.

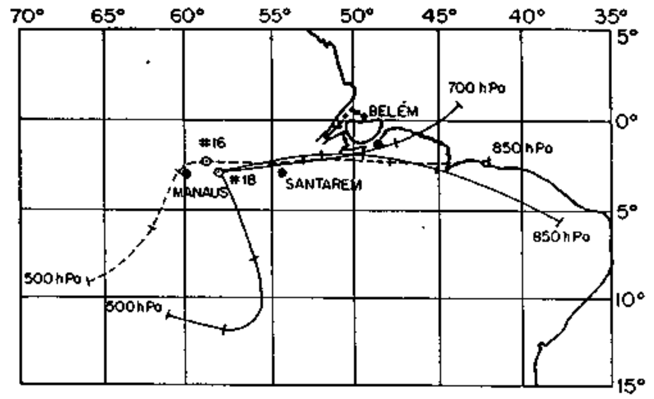

Figure 4. 3 day isobaric back trajectories for flight #16 and #18.

Figura 4. Retrotrajetórias isobáricas de três dias para os vôos #16 e #18.

FLUX BALANCE

The vertical profile of radon concentration in the Amazon Basin during the wet season presented marked discrepancies when compared to the profile data compiled by Liu et al. (1984) for several temperate and cold regions of the world. The Amazon region profile is much less steeper and starts with an initial concentration which is twice as low as in Liu's compilation. These two major discrepancies have important implications to the understanding of the dynamics of the lower troposphere forest regions, as far as trace-elements transport is concerned.

In order to study this subject, an estimate of the flux balance of radon is necessary. The flux balance can be done by measuring the ground flux, calculating the integral rate of decay, the horizontal divergence of radon up to a certain level in the troposphere where the radon concentration is negligible. The residual of the balance as determined from this technique is associated with the turbulent flux of radon at the top level of the radon balance domain. The integral form of the radon balance is:

$$\frac{\partial}{\partial n}\int_{P_s}^{P_t}\bar{n}\,dp + \nabla .\int_{P_s}^{P_t}\overline{V n} = -\overline{\omega' n'}P_t + F_s - \lambda\int_{P_s}^{P_t}\bar{n}\,dp$$

Here, λ is the decay constant for radon, $\bar{n}$ is the number density of radon represented in the Fig. 1 by the best fit line, P_t is the pressure of the level above which radon concentration is assumed negligible, $\overline{\omega' n'}$ is the turbulent flux at the top level where the primes denote deviations from the average vertical velocity in pressure coordinate (ω) and radon concentration, the (–) indicates an average over the area, $\overline{V}$ is the wind velocity from the wind field data, and F_s is the surface flux of radon. The time transients associated with the horizontal radon divergence are also neglected as a first working assumption. The top level P_t is estimated to be located at about 350 hPa (approximately 8 km), based on a best fit extrapolation from the data collected during the experiment (Fig. 1). The integration of the radon concentration up to this level gives a net rate of decay (R_d) of 0.14 $cm^{-2}\ s^{-1}$.

The ground flux of radon, F_s was measured by Trunbore & Wofsy (personal communication) during the mission in the Ducke reserve and averaged 0.33 $cm^{-2}\ s^{-1}$. The ground flux of radon is the time average from a single point in the dryland. Aside from the problems of source heterogeneities due to uneven distribution of radium in the soil, the problem of flux suppression due to flooded lands during the wet season was not taken into account. Niero et al. (1984) have applied satellite remote sensing techniques to a wetland area near the city of Manaus and found that 32% of the studied area was covered with water during the peak of the rainy season, while this coverage was reduced to about 19% in the dry season.

Thus, since the ground flux data was obtained in an unflooded area during the wet season, a first order correction factor for the surface emission of radon, based on Niero's findings, was applied to the data. Such correction reduce the ground flux to 0.25 $cm^{-2}\ s^{-1}$. This value is, however, substantially lower than the commonly accepted 0.75 $cm^{-2}\ s^{-1}$ continental average (Turekian et al., 1977) which suggests that the directly measured 0.33 $cm^{-2}\ s^{-1}$ is probably a best estimate.

The horizontal radon divergence requires several assumptions since the available data does not permit an adequate description of the horizontal variability. However, the wind measurements taken from the large-scale radiosonde network allow an estimate of the time averaged horizontal mass divergence over the Amazon Basin. We have assumed that the time averaged radon concentration over the adjacent oceanic regions is negligible and that the basin concentration is homogeneous. In order to compute the horizontal divergence term of the radon budget, an objective analysis scheme was applied to the radon concentration in order to estimate grid point values in an equally spaced horizontal grid used for the estimation of the horizontal mass divergence (Barnes, 1964). Figure 5 is a schematic view of the radon balance over the Amazon Basin. The vertical fluxes in parenthesis represents the estimate taking into account the suppresion of ground fluxes due to flooded areas.

On the assumption that this flux unbalance is real, it is possible to choose an explanation between two processes that may be able to counterbalance the ground flux surplus. The decoupling between the forest environment and the atmospheric boundary layer is one of such processes. It has been pointed out in recent publication (Wofsy et al., 1988) that the vertical exchange of CO_2 between the forest atmospheric

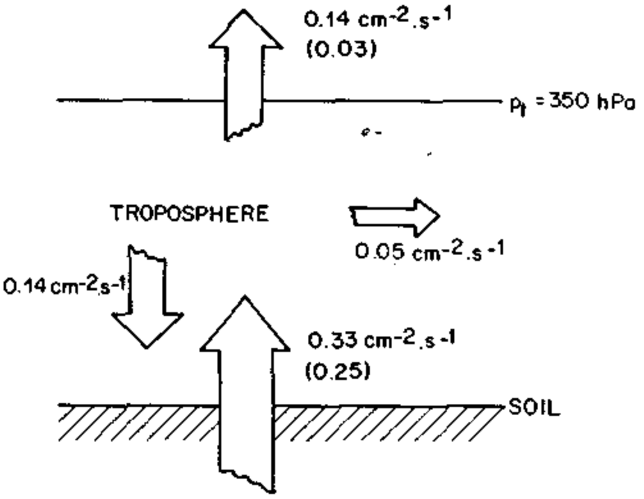

Figure 5. Flux balance of radon over the Amazon Basin.

Figura 5. Balanço de fluxo de radônio sobre a Bacia Amazônica.

environment and the troposphere is reduced during daytime and more active at night. Assuming that the radon flux from the forest canopy to the boundary layer is completely suppressed during daytime, the net flux of radon averaged over several day cycles would be reduced by a factor of 2. This factor is of the right order of magnitude to explain the observed flux unbalance of 2.36 to 1.79 (F_s/R_d).

Another possibility called to explain the flux unbalance is turbulent vertical transport of radon associated with the cloud activity. High concentration values of radon are excepted within clouds because of the higher values observed below cloud base. The cloud induced subsidence in the environment among clouds brings low radon air down. Thus, a net upward transport of radon is expected in convectively active regions such as the Amazon Basin.

A rough estimate of the cloud mass flux (M_c) at the top level of the box (P_t) can be estimated using Yanai et al. (1971) method adapted to the radon budget. The turbulent flux associated with the cumulus activity at P_t can be parameterized as:

$$\overline{\omega' n'} = M_c \, (n_c - \bar{n}) \, / \, g$$

where g is the gravity acceleration, n_c is a typical cloud radon concentration and $\bar{n}$ is the environmental value of radon outside clouds at level P_t (350 hPa). Assuming that the cloud radon at P_t is approximately equal to the mixed layer value (i.e., no entrainment and negligible decay between cloud base and P_t) we estimate M_c to be of the orfer of 10^{-2} kg m^{-2} s^{-1} for the upper value of surface flux and 0.4 x 10^{-2} kg m^{-2} s^{-1} for the lower value. The upper bound is equivalent to about 90 hPa/day in terms of cloud vertical velocity and approximately 35 hPa/day in the lower limit. The upper bound is quite reasonable when compared with

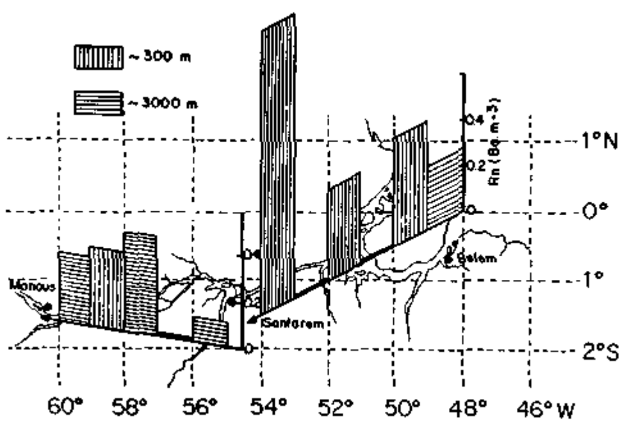

Figure 6. Radon concentration at two levels during the trans-basin flights #12 and #13 (April, 24).

Figura 6. Concentração de radônio a dois níveis durante os vôos transamazônicos #12 e #13 (abril, 24).

typical values obtained in other convectively active tropical regions (Yanai et al., 1971, 1976).

TRANS-BASIN FLIGHT

An horizontal survey of radon was made between the cities of Belém (1° 30'S, 48° 30'W) and Manaus (2° 56'S, 59° 58'W) on April 24th. This flight was flown successively at 300 m and 3000 m levels in equal length legs of about 15 minutes each and the results are shown in Fig. 6.

The general behavior of the radon horizontal survey is that it does not directly display the influence of maritime dilution towards the coastal zones (Belém). The concentration of radon near Belém at the two levels showed a rather continental, well mixed, characteristic. Moving inland towards Santarém, the upper level concentrations decreased to values below the detection limit of the instrument between 50°W and 54°W. The 300 m level radon, on the other hand, presented nearly constant values until around 54°W, near Santarém, where a strong 4-fold concentration increase occurred. This was the largest airborne radon measurement obtained during the whole mission (1.25 Bq m^{-3}). From this point to Manaus, radon at the 3000 m level showed a concentration increase to a relative maximum of 0.42 Bq m^{-3} near 58°W, while the 300 m level showed a relative minimum near 57°W.

Due to the proximity to the source, the 300 m radon data probably reflects: (a) the inhomogeneity in the soil fluxes, (b) meteorological influences such as the decrease of the vertical turbulent mixing due to the suppression of moist convection and (c) different air mass trajectories. The 4-fold radon increase near Santarém, was obtained when the airplane was flying above dryland, with slighly higher topography and reduced forest coverage. It is also a region of lower mean precipitation when compared to all other regions flown (Ratisbona, 1976). These conditions favor an enhanced ground flux of radon. The infrared satellite images shown in Fig. 7 (from April 24, 00 UTC to April 25, 00 UTC every 6 hours) suggests subsidence over the Santarém area when the measurements where taken between 12 and 18 UTC on April 24.

The vertical temperature and moisture profiles obtained by the aircraft also suggest enhanced subsidence in this area; there is a relatively strong temperature and moisture inversion at 0.9 km, as shown in Fig. 8 at approximately 52.5°W in correspondence with the high/lower radon concentration at the lower/higher altitude of the

Figure 7. Infrared satellite images corresponding to flights #12 and #13. A) 6:00 UTC; B) 12:00 UTC; C) 18:00 UTC; D) 0:00 UTC

Figura 7. Imagens infravermelhas de satélite relativas aos vôos #12 e #13. A) 6:00 UTC; B) 12:00 UTC, C) 18:00 UTC; D) 0:00 UTC

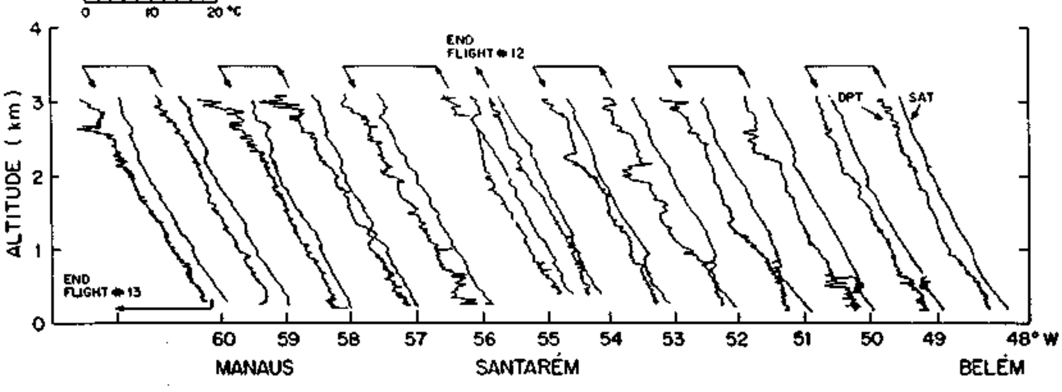

Figure 8. Dew point temperature (DPT) and static air temperature (SAT) during the trans-basin flights #12 and #13.

Figura 8. Temperatura de ponto de orvalho (DPT) e temperatura estática do ar (SAT) durante os vôos transamazônicos #12 e #13.

transbasin flight shown in Fig. 6. The thermodynamic inversion inhibits the vertical mixing above the mixed layer thus restricting radon and other trace elements to the mid-troposphere. The trajectory analysis over the region discards the possibility of different air masses at lower levels since the wind was predominantely from the east through the whole flight.

CONCLUSIONS

The atmospheric radon concentrations measured in the Amazon Basin during the ABLE-2B period (April 13 to May 13, 1987) displayed low values in regard to current values found worlwide. The vertical profiles of radon showed a small decrease in concentration (6.5×10^{-2} Bq m^{-3} km^{-1}) up to approximately 3 km. Above this level there was a marked shallow inversion followed by a sharper decrease with height at a rate of the order of 11×10^{-2} Bq m^{-3} km^{-1}.

The flux balance of radon in the troposphere above the Amazon Basn was evaluated by using data obtained from the vertical concentration profiles, an estimate for the ground flux and horizontal radon flux led to a residual value of 0.14 cm^{-2} s^{-1}. A possible explanation for this unbalance is related to the contribution of the turbulent vertical transport of radon associated with the vigorous moist convective activity observed in the Amazon region. A rough estimate from the radon data of the cloud mass flux at the upper boundary of the sampling domain (6 km) was of the order of 10^{-2} kg m^{-2} s^{-1} which is equivalent to a cloud vertical velocity of about 90 hPa per day. The estimated cloud mass flux was of the expected order of magnitude when compared to estimates based on the static energy and water balance in other convectively active tropical regions (Yanai et al., 1976).

The two detailed radon vertical profiles of April 29th and May 2nd showed distinct radon profiles above the 0.7 km level. The April 29th flight displayed a well mixed profile up to about 2 km revealed by the nearly constant radon concentration and by the thermodynamic variables. On the other hand, the flight of May 2nd exhibited a rapid decrease of radon above 0.7 km as a consequence of the suppression of moist convection as indicated by the thermodynamical analysis.

The horizontal survey of radon between Belém and Manaus did not display the influence of maritime dilution towards the coastal zones. A strong 4-fold enhancement of radon was found near the city of Santarém (1.25 Bq m^{-3}). Between Belém and Santarém a radon depleted region was found in association with an atmospheric subsidence.

The preliminary analysis presented in this paper emphasizes the importance of the vertical transport of radon by the convective activity in the tropical wet forest. However, the measurements are still rather incomplete in order to fully support the cloud mechanism. The measurements of radon are limited by the time response of the instrument and the horizontal and vertical extent of the sampled domain. It remains as a suggestion for future work in this area to measure radon at the outflow level of the deep convective systems in order to identify the role of dilution of the mixed layer radon which is fed into the clouds. Simultaneous radon measurements over a wider domain in addition to the energy and water balance would allow a more precise determination of the components of the radon budget as well as a better understanding of the cloud induced eddy transports.

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