



Technical Memorandum 45

Atmospheric concentrations of radon and radon daughters in Jabiru East

RA Akber and JL Pfitzner

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Abstract

Akber RA and Pfitzner JL 1994. *Atmospheric concentrations of radon and radon daughters in Jabiru East*. Technical memorandum 45, Supervising Scientist for the Alligator Rivers Region, AGPS, Canberra.

The results of atmospheric concentration measurements of radon and PAEC (total and unattached) for a one year cycle in Jabiru East in the northern tropical region of Australia are reported. The average annual radon and PAEC total concentrations are $39 \pm 5 \text{ Bqm}^{-3}$ and $3.5 \pm 0.5 \text{ mWL}$ respectively. The high unattached radon daughter fraction, annual average of 0.21 ± 0.03 , is attributed to the low concentration of condensation nuclei, about $(1.3 \pm 0.3) \times 10^3 \text{ \# cm}^{-3}$. Seasonal and diurnal variations are evident in these parameters. The unattached fraction is lower than average during the Dry season and also in the early morning when peaks occur in the radon and radon daughter concentrations. The condensation nuclei concentration and the equilibrium factor are higher than average during the dry months of the year.

This report will appear in a similar form in the *Proceedings of the Workshop on the Health Effects of Induced Radionuclides: Implications for Radiation Protection in Mining* (in press), Jabiru 25 September 1992.

Atmospheric concentrations of radon and radon daughters in Jabiru East

Introduction

In March 1989, an automatic monitoring station was installed in a caravan which enabled simultaneous measurements of atmospheric concentrations of radon, radon daughters (total and unattached) and condensation nuclei. Wind speed and direction were also measured at the same time. The primary objective of these measurements was to estimate the contribution of the ERA Uranium Mine at Ranger to atmospheric radon and radon daughter concentrations at selected locations within a few kilometres from the mine site. During the first year this station was mostly located at ARRRI in Jabiru East, about 2.5 km NW of the mine (fig 1). This paper presents some of the results from this station for an operational period of one year, March 1989 to February 1990, with the exception of the months of June and September 1989 when the station was used elsewhere.

Locality

Jabiru East and Ranger are located in the northern tropical region of the Northern Territory in Australia within the boundary of a uranium mine lease that is surrounded by Kakadu National Park. The climate is characterised by distinct Wet (November–March) and Dry (May–September) seasons. These seasonal boundaries are, however, not precise. For an average year in Jabiru East about 67% of the annual rainfall occurs between January and March, while <2% occurs between May and September. Bush fire management regimes are followed by the Kakadu National Park and Ranger Mine managements. Most of the bush fires occur during the period May to August. About 25% of the average rainfall occurs during the months of November and December, but the showers are localised and less frequent. According to the information provided by the Bureau of Meteorology (1993) the average annual rainfall is 1437 mm. For most years evaporation exceeds rainfall. The area, like other parts of northern Australia, is subject to cyclones.

During the Dry season, wind speed and direction are determined by the ESE to SE trade winds. In all seasons there is a late afternoon–early evening wind from the general NW direction that may be associated with the sea breeze. The monsoonal winds from the NW during the Wet season are not as persistent as the SE winds of the Dry season. Wind speeds are generally higher during the day time (9am–3pm) and during the early months of the Dry season (Clark et al 1993).

Jabiru East is no longer a residential town, but laboratories and offices are attended by about 100 people during the day time. The nearby Aboriginal camps at Gulungul Creek and Magela 009 are inhabited by about 15–20 people each, including children. The township of Jabiru (1500 people including adults and children) is the major population centre near Ranger. The

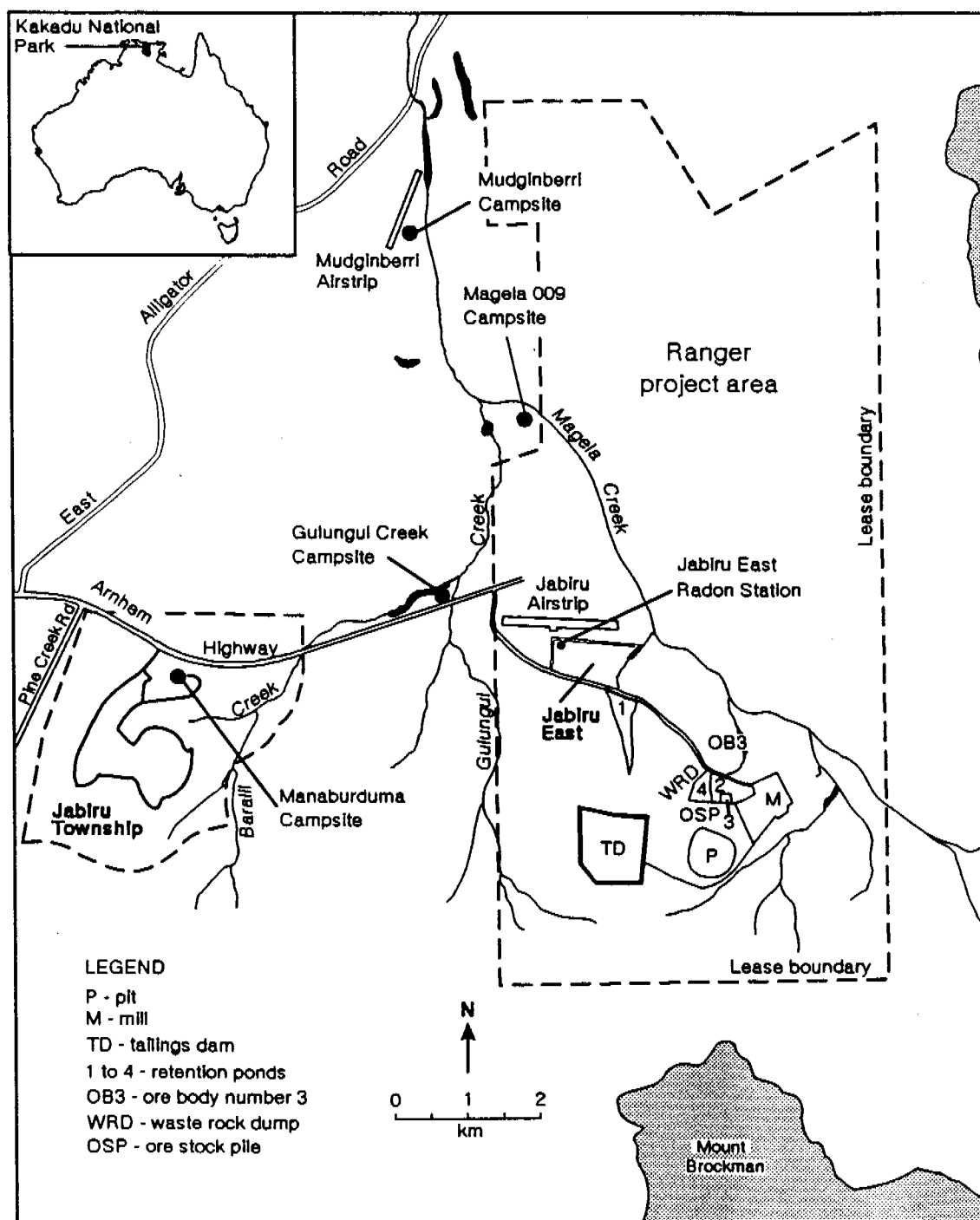


Figure 1 A locality map to show the population centres and the radon monitoring station in Jabiru East near ERA uranium mine at Ranger

permanent Aboriginal camp site at Manaburduma is occupied by twenty-five people (Wellings P, 1990 pers comm). Most visitors to Kakadu National Park come to Jabiru. The estimated number of Park visitors was about 220 000 in 1988. About 5% of the visitors came for a day trip only; the average stay for the remaining 95% was a little above four days (Preece 1989).

About 3000 tonnes of U_3O_8 was produced each year between 1981 and 1992 at Ranger but recently production has been reduced to about half this value. The average ore grade is about 0.3% U_3O_8 and the material less than 0.01% U_3O_8 is piled as waste rock. The ore stockpiles, waste rock dump, mining pit and tailings dam are the major radon sources. Their locations and the

location of the sampling point for the measurements presented in this paper are identified in fig 1. According to an estimate reported by Akber et al (1992a), the mine-related sources contributed about 20% of the total 38 Bqm^{-3} annual average atmospheric radon concentration in Jabiru East for the period July 1990–June 1991. The background and mine-related radon concentrations in Jabiru and Manaburduma camp area were somewhat lower perhaps due to the lower local soil radioactivity and larger distance from the mine site. The background radon concentrations at the other two Aboriginal campsites are not known, but model-based estimates show that the mine-related atmospheric radon concentrations are comparable with those in Jabiru East (Akber et al 1993). The data sets for this paper are limited to a sampling point in Jabiru East only, but the findings may be broadly applied to other centres of demographic interest in the expectation that the trends of radon and radon daughter behaviour in the outdoor atmosphere are likely to be similar within a range of a few kilometres.

Instruments

The radon detection system used in the present study is similar to that described by Whittlestone (1985). It consists of a primary filter and a dehumidifier, a thoron delay chamber, a particle generator for radon daughters attachment in a 200 L mixing chamber and a ZnS detector. The air is sampled at a rate of 10 L per minute. The mixing and attachment step in the chamber introduces an approximately 60 minute delay between the collection and the detection step. The output data are corrected for this delay prior to data processing. Under optimum operational conditions, the detection limit of this system is lower than 1 Bqm^{-3} .

Radon daughters are detected by measuring the count rate of alpha particles on a filter paper through which air is drawn at a rate of 1 L per minute. A modified alpha Nuclear 400 system is used for counting. The unattached daughter detection system is similar to the total detection system but the filter is replaced by a wire screen with a mesh size of 400 per inch. Following the mathematical treatment of Ramamurthi and Hopke (1989), the wire screen parameters lead to a 50% collection efficiency cut-off point (d_p –50%) at $\sim 7.1 \text{ nm}$. Front to total activity ratio (FT) of 0.7 is used for the calculations, which is in good agreement with the experimental findings of 0.67 ± 0.02 for $d_p > 99.9\%$ for an ultrafine (AMD 0–10 nm) activity size distribution mode (Solomon & Ren 1992). The detection efficiencies and the flow rates of radon and radon daughter detection systems are checked on a routine basis using standard thin alpha sources and flow rate meters. The calibrations are checked against the portable equipment calibrated at the Australian Radiation Laboratory.

The condensation nuclei concentration (CN) counter is a cloud chamber. The particle concentration is calculated from the difference in the optical transmission measured by a light emitting diode (LED) before and after the formation of a condensation cloud by the rapid expansion of the water saturated air sample. This instrument has been calibrated only once, at the time of installation in March 1989. The calibration can be traced back to a standard particle counter maintained by the CSIRO Division of Atmospheric Research.

Wind speed and direction are measured using standard meteorological equipment, calibrated in a wind tunnel at ANSTO. Meteorological measurements have been routinely carried out at the height of about 15 m above the ground, away from the interference of the arboreal canopy. Radon and radon daughter concentrations are also measured at the same height to improve the signal to noise ratio for estimates of the mine-related component of atmospheric radon and radon daughter concentrations using correlated wind direction measurements (Akber et al 1992a). Ambient aerosol densities are measured at a height of 2 m above the ground.

Further details of the instrumentation and operation have been described by Pfitzner and Whittlestone (1991).

Results and discussion

Continuous monitoring of radon and its daughters generates a time series of half-hourly concentrations in the atmosphere over prolonged periods of time. The average diurnal behaviour is illustrated here through a set of curves generated by averaging, on a half-hourly time basis, the total potential alpha energy concentration (PAEC total) data for the months of May, August and November 1989 and February 1990 (fig 2). The error bars correspond to the standard error in the mean values for each half-hour period. As the half life of radon daughters is comparable with the sampling time, an error is introduced due to their detection in the subsequent time intervals. The data are not corrected for this source of error but, as the response times of radon and radon daughter detection systems are similar, particularly during steady state conditions, this error is likely to cancel out in the calculation of the equilibrium factor. The inversion peaks in Jabiru East are generally broad, typically from 4 to 6 hours, shifting by up to a few hours on a day-to-day basis. Frequently more than one peak occurs. These characteristics of diurnal variations have led to broad peaks in the monthly averages, typically from 2 am to 12 noon. The diurnal trends of radon and PAEC unattached are similar to those for the PAEC total plotted in fig 2.

Mean daily concentrations of radon and radon daughters for each month (monthly averages) are generated by using half-hourly averages similar to those shown in fig 2. The values in fig 3 are the monthly average radon concentrations for the reporting period. The average rainfall data for Jabiru East (also shown in fig 3) illustrate the distinct Dry and Wet seasons. As expected, radon concentrations were highest during the Dry season, indicating higher emission rates from the drier soils. Also, the radon concentrations for the earlier months of the Dry season (May–August) are higher than those for the later months (October–November). Similarly, the values appear to increase in December and January – at the onset of the Wet season. This behaviour of atmospheric radon concentrations is consistent with the expected changes in radon emanation with the soil moisture content as reported by other authors. For example, laboratory based experiments of Strong and Levins (1982) showed that for uranium tailing materials, the moist pores or capillaries emanated more radon than dry or water saturated pores and capillaries. They observed the maximum emanation occurring at about 6% of water content by weight. Thamer et al (1981) also reported consistently lower emanation from the dry uranium ores when compared with the 5–20% water saturated samples.

The seasonal variations in the PAEC total and PAEC unattached are broadly similar to those of radon. In detail, though, the ratio of PAEC total relative to radon concentration is higher during the dry months than during the wet months. This trend is reflected by an increase in the equilibrium factor F which varies between 0.45 and 0.60 for July–November and between 0.25 to 0.45 for March–May and December–February periods (fig 4b). The trend for the unattached radon daughter fraction f_p (fig 4a) is opposite to that of the equilibrium factor F . This fraction, defined here as the ratio of PAEC unattached to PAEC total, is between 0.2–0.4 for the wet months of the year. It drops to a value around 0.1 for the dry months.

The behaviour of f_p and F can vary with changes in the ambient aerosol density. A recent review (NRC 1991) summarises the work of many authors but most of these studies have been conducted in mine environments or indoors in houses under European conditions. Porstendorfer (1987) reported that (for indoor air) a higher particle concentration at the time of formation of ^{218}Po increases the equilibrium factor value because the attachment is then faster than the plate out. The experimental observations showed a decreasing trend in the unattached fraction f_p as a function of the condensation nuclei concentration (CN) 10^3 – 10^6 # cm^{-3} range in five different rooms. Reineking and Porstendorfer's (1990) limited study in the ambient atmosphere at Gottingen is associated with CN 2 – 4×10^4 # cm^{-3} , f_p 0.01–0.02 and F 0.53–0.67; no correlation is evident from their results. Perhaps the observations by Whittlestone (1990) at Cape Grim are

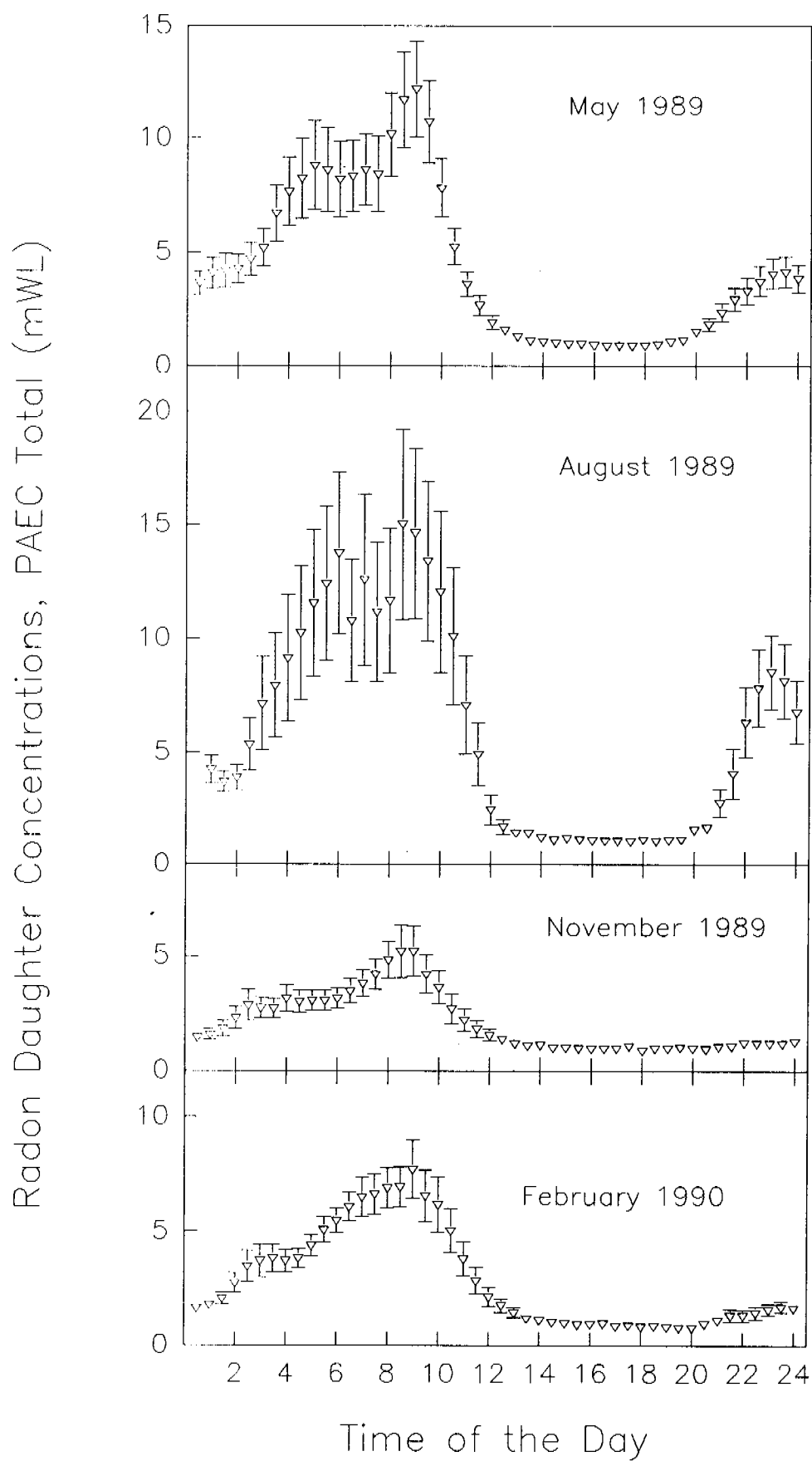


Figure 2 The average daily response curves of radon daughter concentrations for four different months in Jabiru East

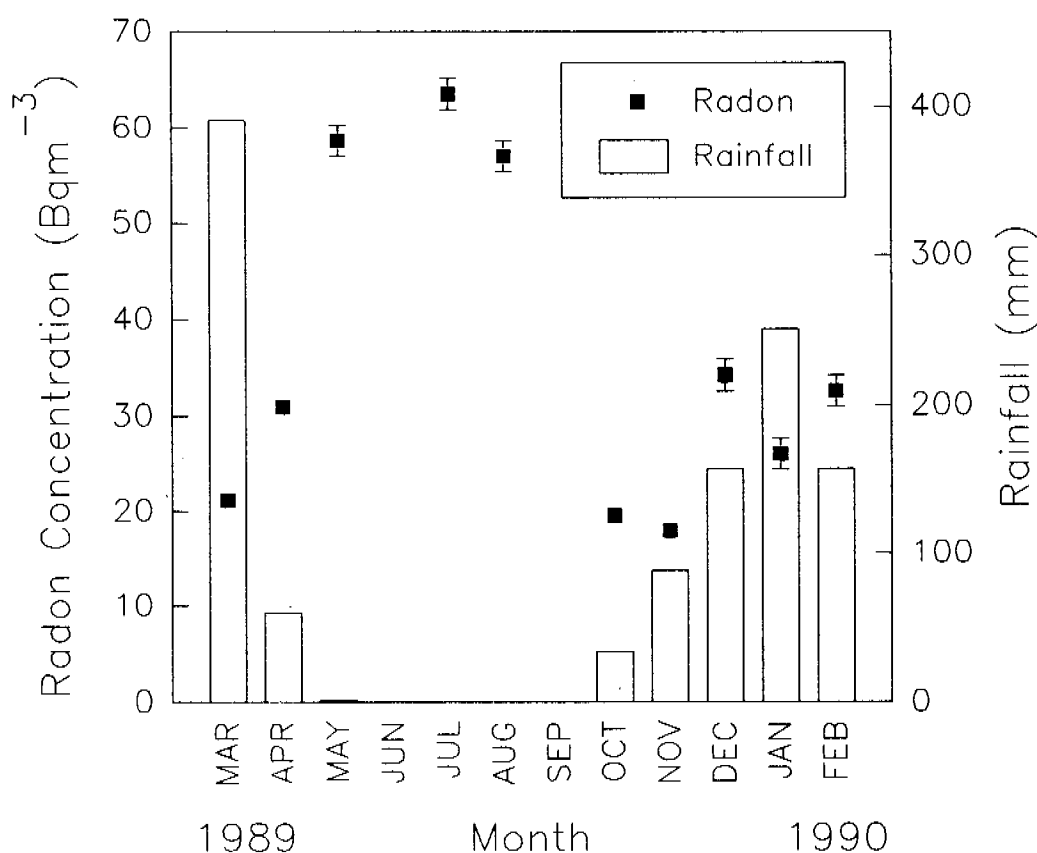


Figure 3 Monthly cumulative rainfall and average radon concentrations in Jabiru East during the period March 1989–February 1990

more relevant to this study as the data sets cover the particle concentration range of 30–3000 (# cm⁻³), similar to those observed by us in Jabiru East. Whittlestone reports a decrease in F value from 0.86 to 0.1 as the particle concentration falls from 10^3 to 10^1 (# cm⁻³). His data for unattached radon daughter concentrations are limited. One of the two data sets shows a downward trend in the f_p values from about 0.25 near 10^2 CN to about 0.05 near 2×10^3 CN. The other data set showed that f_p is not significantly different from zero in this particle density range. Earlier data of mine-related studies reviewed in NRC (1991) also show an inverse correlation between f_p and particle concentration values in the 10^3 – 10^6 region. The scatter in the data are large, typically up to an order of magnitude. Kojima and Abe (1988) observed diurnal variations in the aerosol density in a Japanese home. They noted that unattached radon daughter concentrations decreased when peaks occurred in the aerosol density.

Our observations of the condensation nuclei density (CN) are shown in fig 4c. Overall the CN values are low, averaging at 1.3×10^3 # cm⁻³. The values are higher during the months of little to no rain (April–November) than those of significant rain (December–March). A sudden drop in the CN value for the month of July 1989 is hard to explain. Bushfires, which are frequent during these months of the year, are generally managed on an area-to-area basis. This can cause fluctuations in the CN values and may be responsible for a significant upward trend for the month of August. The bar graph in fig 4c represents the average monthly mass loading obtained from high volume sampler data for the period 1983–88 at several sites in the vicinity of Jabiru East (Akber 1992). The mass loading and condensation nuclei concentration trends are broadly similar.

A comparison of the CN, f_p and F (fig 4) trends shows broad agreement with the results of other observers; the equilibrium factor F increases and the unattached radon daughter fraction f_p

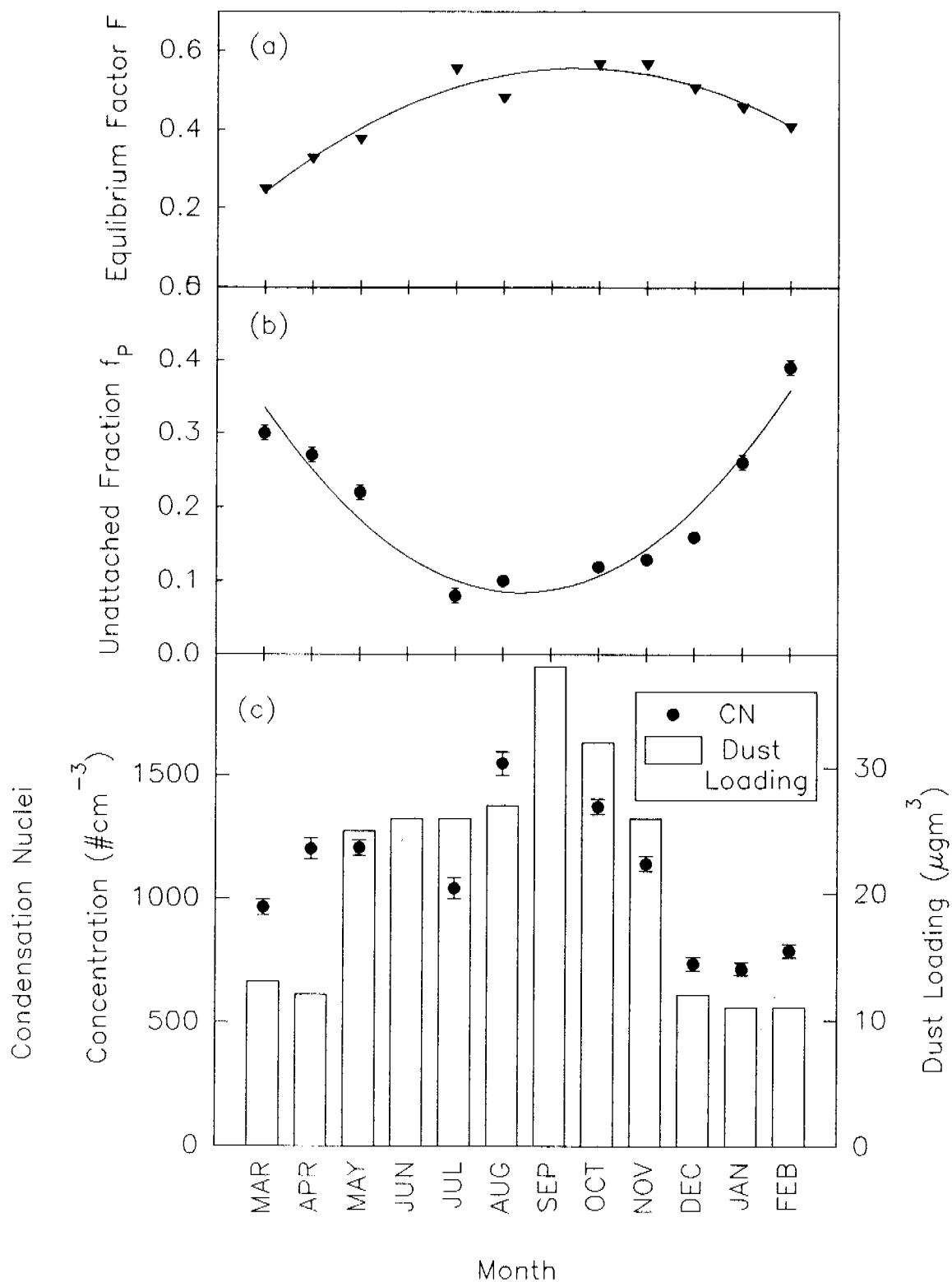


Figure 4 Monthly average values of (a) unattached fraction, (b) equilibrium factor and (c) condensation nuclei concentration and dust loading. With the exception of the dust loading values, the data are for Jabiru East during the period March 1989–February 1990. The dust loading data are the average values for the period 1983–1988 for several sites in the vicinity of Jabiru East

decreases with increases in the concentration of condensation nuclei. Our data set is, however, applicable to the outdoor non-urban conditions found in northern tropical Australia. The low CN values also explain the high unattached radon daughter concentrations compared with most other reported values which correspond to higher aerosol density environments.

Diurnal trends were observed in the unattached radon daughter fraction and the equilibrium factor. These trends show that, in addition to correlation with the concentration of condensation nuclei, the early morning calm conditions also appear to affect the attachment behaviour of radon daughters. This is illustrated in fig 5 (a-d) showing diurnal variations in f_p , F, CN and wind speed for an average day in the months of November 1989 and February 1990. The wind speed is generally low during the late night to early morning period. The CN values for this period are also low; they rise between 8-10 am and broadly follow the wind speed trend of the day. The downward trend of CN between 4-8 pm during the month of February is probably due to wash-out during the frequent afternoon rainfall.

Despite the lower CN values, both f_p and F show a downward trend during the early morning hours. This behaviour can probably be explained, at least qualitatively, through examining the production and fate of radon daughter atoms in the atmosphere. Lower F values occur when radon daughters have not achieved equilibrium with the parent radon in the atmosphere. This situation may be true for the early morning low wind speed periods of limited radon transport. The radon source under these conditions is expected to be local and the radon is detected before it reaches equilibrium with its daughters. During the daytime, radon is mixed more regionally and there is a longer period for daughter ingrowth and an increase in the F values.

A study by Woods (1989) shows diurnal variations in the equilibrium factor measured at the tailings repository of the Nabarlek uranium mine. Three sets of data were reported in that study covering periods of several days in October 1985, September 1987 and October 1988. The times of the equilibrium factor peaks vary in these data sets, with the September 1985 behaviour being similar to that observed in the present work for Jabiru East. Woods (1989) does not comment on the reported observations.

Early morning calm conditions also appear to favour the attachment of radon daughter atoms to the ambient aerosol, thereby leading to lower f_p values. Investigations of the exact cause of this behaviour are beyond the scope of this paper. The ambient aerosol characteristics, other than the density, such as the average particle size and the size distribution, can substantially alter the attachment rate of radon daughters (NCRP 1988, NRC 1991, Reineking & Postendorfer 1990). Little is presently known about the diurnal or seasonal variations in these parameters in Jabiru East. Hence the cause of low f_p values during the calm to low wind speed periods of the day should be further explored.

The average wind speed is low during the late months of the Dry season and may, in addition to the increase in the average CN values, be responsible for the low unattached radon daughter fractions during these months.

Table 1 summarises the annual average values of variables measured during this study. The estimates of PAEC (total) effective dose in Jabiru East (table 2) were obtained using a dose conversion factor of 8 mSv/WLM for adults and 10 mSv/WLM for juveniles. These conversion factors were based upon the value recommended by ICRP for the mine workers (ICRP 1985), corrected for different breathing rates for adults and juveniles (ICRP 1979) and the different organ size factor for juveniles (IAEA 1982). The ICRP dose conversion factor for inhalation of radon daughters by workers corresponds to values of the unattached radon daughter fraction in the approximate range 0-0.05 (ICRP 1981). The estimates of dose in Jabiru East need to be corrected upwards in the light of the information on the unattached radon daughter fraction reported in this paper. Using an average of the Jacobi-Eisfeld and James-Birchall models corresponding to 0.1 μm AMD (NEA 1983) a correction by a factor of 1.7 is proposed, corresponding to the annual average unattached fraction of 0.14 in Jabiru East. A more detailed

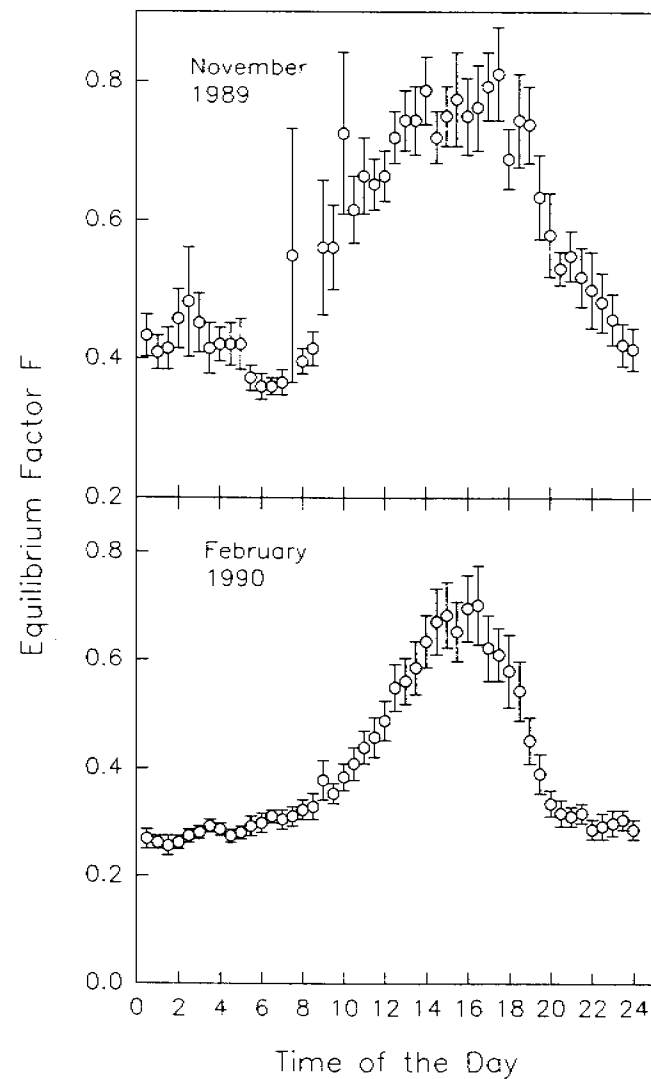


Figure 5a Average daily response curves of the equilibrium factor (F) for the months of November 1989 and February 1990 in Jabiru East

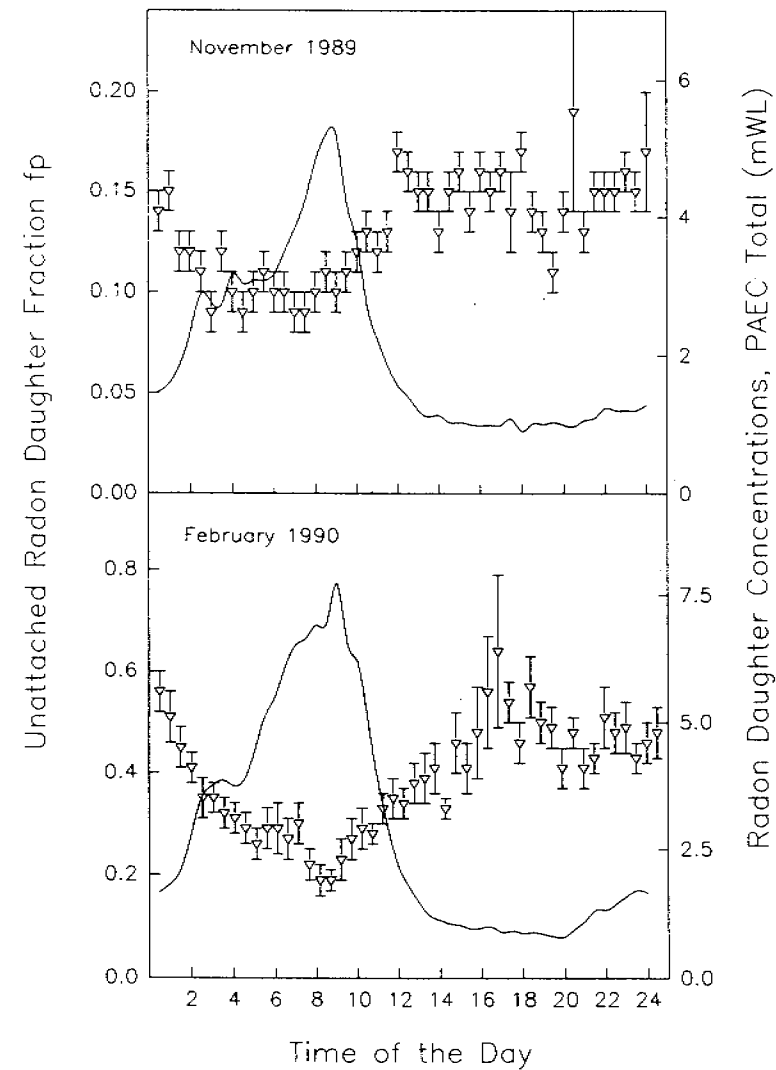


Figure 5b Average daily response curves of the unattached radon daughter fraction (f_p). The corresponding PAEC total values (line) are drawn to demonstrate the difference in diurnal trends of f_p and PAEC total

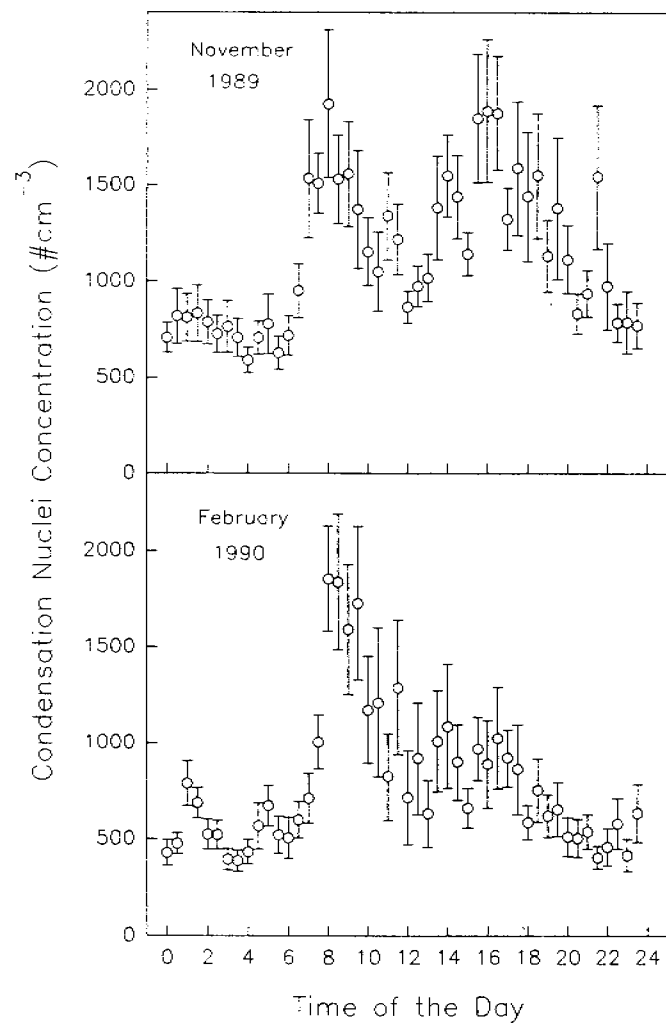


Figure 5c Average daily response curves of condensation nuclei concentrations

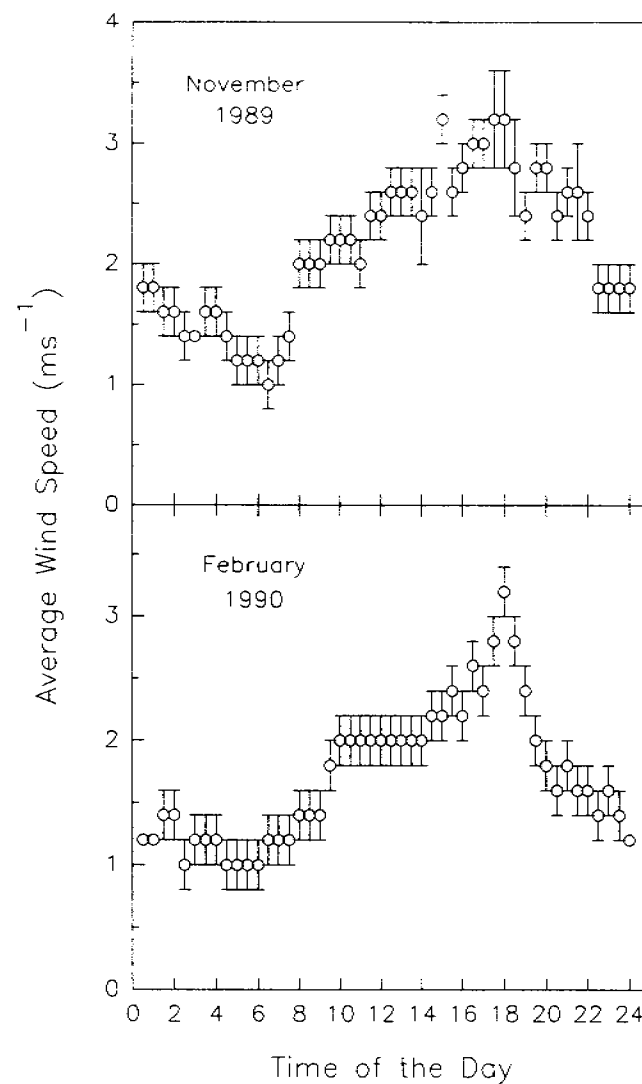


Figure 5d Half-hourly average wind speeds for the months of November 1989 and February 1990

Table 1 Average annual values of various parameters observed during this study in Jabiru East for March 1989–February 1990 period

Parameter	Value
Radon (Bqm ⁻³)	39 ± 5
PAEC total (mWL)	3.3 ± 0.5
PAEC unattached (mWL)	0.46 ± 0.07
CN (#cm ⁻³)	(1.3 ± 0.3) 10 ³
F	0.45 ± 0.03
f _p	0.21 ± 0.03 ^a
	0.14 ± 0.03 ^b

a based on half-hourly PAEC (unattached) to PAEC (total) ratios

b based on the annual average PAEC (unattached) to PAEC (total) ratio

Table 2 Annual effective dose for Jabiru East

Source	Period	Effective dose (mSvy ⁻¹)			
		Total		Mine-related	
		Juvenile	Adult	Juvenile	Adult
Akber et al (1992a) ^a	July 1989–June 1990	1.6	1.3	0.21	0.16
Akber et al (1992b)	July 1990–June 1991	1.5	1.2	0.22	0.17
Akber et al (1993)	March 1989–Feb 1990	–	–	0.30 ^b , 0.26 ^c	0.24 ^b , 0.21 ^c
This study	March 1989–Feb 1990	1.8	1.4		
Average		1.7	1.3	0.26	0.21

a Note an overlapping period of observations with other reported values. Data not included in average.

b Model

c Experimental

correction process could be followed but it will be complex because both the diurnal and the seasonal variations in radon daughter concentrations and values of the unattached fraction need to be taken into account in making an estimate of the annual effective dose. Any upward revision of the mine-related component of the effective dose is expected to be smaller than that for the background component because the radon transport from the Ranger Uranium Mine to centres of population is most significant during the dry months of the year and during the early hours of the morning when values of the unattached radon daughter fraction are lower than average. ICRP is currently in the process of revising the dose conversion factors for radon inhalation. Further adjustments in dose estimates may, therefore, be required when the outcome of this revision is known.

Acknowledgment

Dr A Johnston's contribution at the early stage of this study is acknowledged.

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