# ENVIRONMENTAL IMPACT OF MINING

## Radiological Impacts of Mining

# Long-term study of groundwater dispersion of uranium at Ranger Mine

**M Iles, P Martin, B Ryan & C leGras**

## Background

Energy Resources of Australia (ERA) has monitored uranium (U) concentrations in water from numerous bores located within the Ranger mine lease since the 1980s. It has been observed that the monitoring data have shown increasing U concentrations in filtered water from several of the bores, particularly those close to the north and north-eastern walls of the tailings dam (ie OB13A and OB16) (fig 1). This project aims to use the $^{234}U/^{238}U$ isotope ratio to indicate the source of uranium and to identify mechanisms to account for the concentration changes. This was possible as uranium-234 is more mobile than $^{238}U$ due to the physical recoil of the atom following the alpha decay of $^{238}U$, and the displacement of the $^{234}U$ atom to a more chemically labile binding site. Consequently, the $^{234}U/^{238}U$ ratio is generally greater than unity in natural waters. In tailings dam water the ratio is close to unity following the chemical leaching process used in the milling of the ore (which extracts both isotopes with high efficiency).

## Results and discussion

Locations of the bores sampled are shown in figure 1. Initially, based upon the results of ERA's monitoring data, only bores to the north and east of the tailings dam were targeted for study. However, more recently, the extension of waste rock dumps into this area has made the interpretation of results for these bores difficult. Consequently, recent work has concentrated on other bores on the Ranger site. These data will provide a baseline against which any possible future changes in U concentrations in these bores can be investigated.

Most of the U isotope ratio measurements have been made on samples collected since 1996. Figure 2 shows the $^{234}U/^{238}U$ isotope ratio results obtained for filtered water samples from a number of bores at Ranger plotted against the reciprocal of the U concentration. On such a plot a sloping trend line can indicate mixing of waters with different isotope ratios and different concentrations (Osmond & Cowart 1992). The resulting mix would lie on a line between the two end members. With high U concentrations and a $^{234}U/^{238}U$ ratio close to 1.0, tailings dam water would lie close to the bottom left corner of these plots (x~0, y~1). Therefore, a line with a *y*-intercept of 1 can indicate mixing with tailings water. Results are described below according to whether or not there appears to be some change in the source or behaviour of U in the bores.

### No apparent source change

Since the beginning of the collection of bore monitoring data OB6A has consistently had elevated U concentrations compared to that measured at other Ranger observation bores. The $^{234}U/^{238}U$ ratios for OB6A have remained above 1.65, apart from May 1998 when a lower ratio (1.26 +/- 0.02) coincided with a higher than normal concentration.

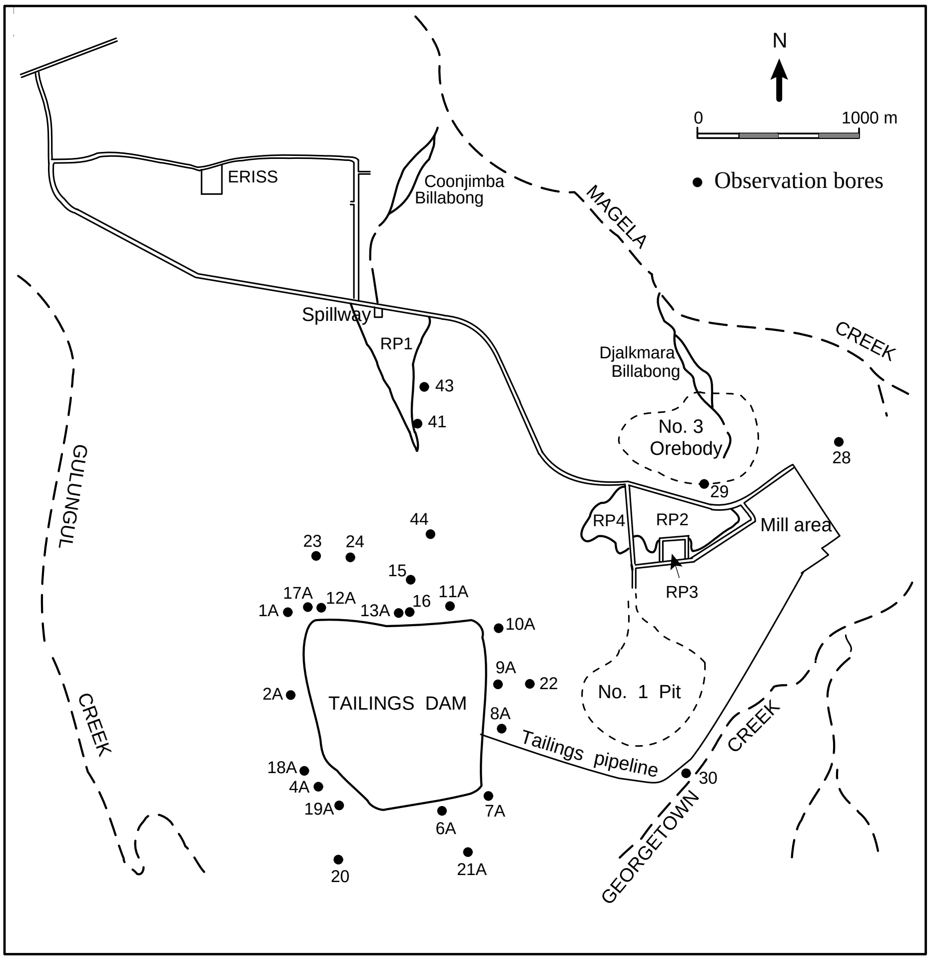



**Figure 1** Location of observation bores at Ranger

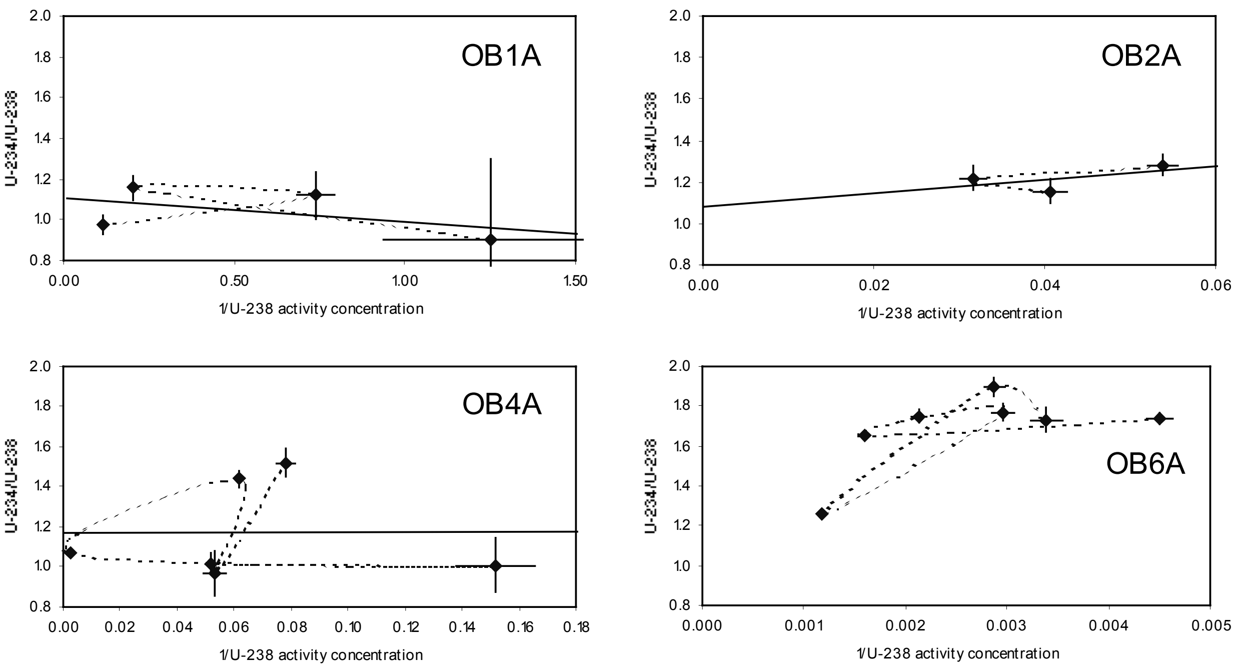
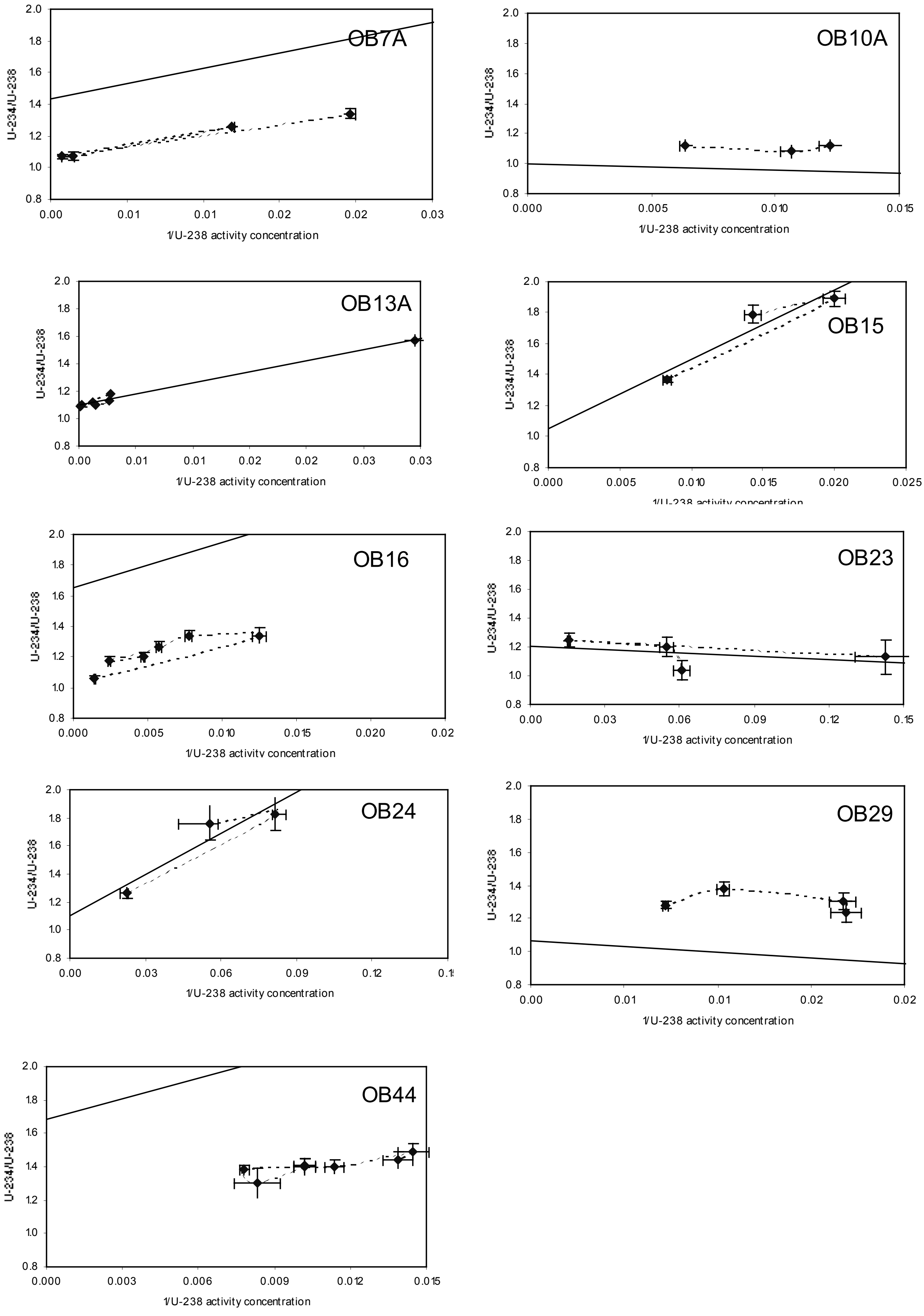



**Figure 2** $^{234}U/^{238}U$ ratio vs. the reciprocal of the $^{238}U$ concentration obtained for filtered water from a number of Ranger observation bores. Error bars reflect counting statistics alone, and correspond to one standard deviation. The mixing line is shown in bold, the data sequence is shown by the dashed line.

**Figure 2** continued



OB1A and OB4A show no correlation between U concentrations, which are variable in both bores and isotope ratios. The variability in U concentrations is most likely related to the high Fe concentrations for these bores, resulting in the formation of $Fe(OH)_3$ on sampling which removes the U from the filtrate fraction.

OB10A has relatively small un-correlated changes in both U concentration and isotope ratio values. OB23 and OB29 have fairly stable $^{234}U/^{238}U$ ratios un-correlated to U concentrations. OB2A and OB44 both appear to have fairly stable concentrations and isotope ratio values.

## Possible Source Change

In OB13A the U concentration increase since the 1980s has been by a factor of about 1000; since about 1992 concentrations have often been of a similar order of magnitude to, or even greater than, the concentrations in tailings dam surface water. There is a strong correlation between this large concentration increase and a large decrease in the $^{234}U/^{238}U$ ratio.

No isotope ratio values are available for OB16 prior to the time of marked U concentration increase in 1990. Where isotope ratios are available their decrease shows some correlation with the concentration increases, but the measured value remained greater than or equal to 1.18.

OB7A oscillates between high and low U concentrations, which strongly correlate with the isotope ratio values, which decrease at times to 1.07.

Although there is only a small dataset for OB15 and OB24 (no. of samples = 3), there is a strong correlation between U concentration and $^{234}U/^{238}U$ ratios. The U concentration in OB15 has almost halved while for OB24 it has approximately doubled. The change in concentration in these bores, though significant, is relatively small compared to some other bores.

In the following, three proposed mechanisms for increased U concentrations are examined for consistency with the observed data.

### Mechanism 1 — Transport of U from the tailings dam

Based upon time-series changes in U concentrations, and associated changes in isotope ratios, seepage could be affecting at least the following set of bores: OB7A, OB13A, OB15, OB16 and OB24. Figure 2 shows that the *y*-intercept values for each of these bores are greater than 1.0 implying that the U increases are not solely due to transport directly from the tailings dam.

The following observations also imply that direct seepage from the tailings dam is not dominating U behaviour in this system:

- The very high U concentration observed in November 1996 for OB13A was several times higher than the U concentrations in tailings dam water.
- The U concentration observed in some bores appeared to fluctuate during the year with U concentration values being higher late in the Dry season (November) than early in the Dry season (May). No such seasonal effect is seen in ERA's monitoring data for sulphate which is sourced primarily from the tailings (leGras et al 1993).

### Mechanism 2 — Leaching of U from mine structure materials

Uranium concentration increases in OB13A and OB16, located close to the north wall of the tailings dam (fig 1), coincided with the raising of the tailings dam wall in 1990. Although not conclusive, the timing suggests that this was due to seepage from materials used in the dam wall construction, or altered groundwater flows due to the greater mass of the wall.

Waste rock dumps can contain unweathered, mineralised material which can be expected to degrade with exposure to rain and the atmosphere. As these waste rock dumps increase in height, the associated hydraulic head can be expected to increase and more material can be exposed to percolating rainwater which can dissolve U from the rocks. However, several observations do not support the rainwater percolation hypothesis, including: (1) the observed seasonal variations in U concentrations and ratios, (2) studies of the Ranger Land Application area have shown that U from the applied water is retained in the top few centimetres of soil (Akber & Marten 1992), and (3) U is comparatively immobile under the conditions expected in waste rock dumps, ie oxidising, near-neutral pH and relatively low conductivity.

#### Mechanism 3 — Mobilisation of U from aquifer rocks in the vicinity of the bore

The U isotopes should be equally labile when contained in carbonate minerals because these can be chemically leached with relative ease. Uranium mobilised from these minerals should therefore have a $^{234}U/^{238}U$ value near unity. Uranium-enriched carbonate facies are common in both Ranger ore and waste rock, and possibly also in aquifer material. If additional, highly labile $^{234}U$ was produced by recoil from a residual phase (such as silicate minerals), the isotope ratio may converge to near unity as carbonate leaching became the dominant source of U in borewater.

The available data do not support mobilisation of U from aquifer rocks as a dominant factor, ie the changes in U behaviour should be gradual and related primarily to changes in parameters such as the chemistry of the groundwater and the level of the water table. No simple relationship was observed between U concentrations, pH, conductivity and sulphate ion concentrations reported annually by ERA. However, the chemistry of U is very complex and a simple correspondence between these parameters and U concentrations cannot be expected. Also, displacement of either local or transported U from the aquifer walls by an ion exchange process would be preceded by displacement of a more exchangeable ion. No correlation was seen between measured sodium, potassium and U concentrations reported annually by ERA, although any ion-exchange effects would probably be masked by interactions with magnesium and calcium, which are present in much higher concentrations than U. It is possible that a relationship exists between U and ammonium ion concentrations but the latter has not been monitored.

## Conclusions

At this stage, none of the three postulated mechanisms can be unequivocally given as the major influence on U behaviour in this system. However, the fact that $y$-intercepts for U ratio plots are generally greater than 1.0 implies that the primary source of the observed U increases is not the tailings dam. Interpretation of the results for bores which have shown increasing $^{238}U$ concentrations is made more difficult by the lack of an extensive $^{234}U/^{238}U$ ratio database for the period prior to the concentration increases. Consequently, work on this project is presently targeted at providing such a baseline dataset for other bores on the Ranger site.

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# Development of a moving-grid dispersion model for radon and radon progeny

**P Martin**

## Introduction

Atmospheric dispersion of Rn and its progeny and subsequent inhalation can be one of the dominant pathways for public dose from uranium mining. Most of this dose arises from exposure to the progeny rather than the Rn parent. Three important parameters for dose estimation are the Rn progeny potential alpha energy concentration (PAEC)[1], the equilibrium factor ($E$)[2] , and the unattached fraction ($f_p$)[3]. Measurements of Rn progeny for Jabiru East by Akber and Pfitzner (1994) show that the unattached fraction is unusually high with an annual average of about 14%, suggesting that effective dose estimates should be revised upwards (Akber et al 1994b). However, their reported measurements were not able to distinguish between mine-origin and non mine-origin PAEC.

In this section, the development and validation of a model for the atmospheric dispersion of Rn and Rn progeny following exhalation of Rn from defined sources will be described. The majority of reported studies on behaviour of Rn progeny in the air relate to the indoor environment, while many outdoor studies deal with temperate and urban environments. This makes the choice of values for many of the parameters of the model problematic for a tropical, rural location such as Jabiru. Since the main purpose of the present work is to estimate whether or not the unattached fraction could make a significant difference to dose estimates in the ARR, a set of values for input parameters for the model has been derived from the literature which can be expected to be approximately correct. A sensitivity analysis has then been carried out to determine which of these parameters gives rise to the greatest uncertainties in model predictions (and hence which parameters would be the most important to address in future research on Rn and Rn progeny dispersion).

## Model description

### Description of the moving-grid dispersion model

The dispersion model uses a variation of the grid-cell method, referred to here as a *moving-grid* model. This uses a 2-dimensional grid of cells, and calculates the transfer of Rn and progeny successively from one grid to the next downwind of the source. A computer program, *RAPAD* (Radon And Progeny Atmospheric Dispersion model), has been written in the C programming language to carry out the model calculations (Martin 2000).

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1 PAEC is essentially a measure of the total amount of alpha energy which would be liberated upon the decay of all of the short-lived Rn progeny present in a unit volume of air.

2 $E$ is essentially the ratio of the actual PAEC to that which would be present if the progeny were in secular equilibrium with the Rn parent.

3 The unattached fraction, $f_p$, referred to here corresponds to the ratio of unattached PAEC to the total PAEC, where 'unattached' refers to PAEC associated with an aerosol with diameter less than approximately 4 nm.

**Calculation of dispersion**

The $x$ dimension (ie in the current wind direction) of each cell was 100 m, the $y$ (horizontal, perpendicular to $x$) dimension was 100 m and the $z$ (vertical) dimension was 10 m (ie the approximate height of the tree canopy). For calculation of the proportions to pass to the cells of the next grid, a Gaussian distribution in the $y$ and $z$ directions has been assumed. Increase in wind speed with height has been calculated from the power law:

$$U(z) = U(z_0)\left(\frac{z}{z_0}\right)^n$$

where $U(z)$ is the wind speed at height $z$ (m), $U(z_0)$ is the wind speed observed at reference height $z_0$, and $n$ is an empirical constant taken here to be 0.17 (Boeker & van Grondelle 1995). Predictions of mixing layer height $H_m(T)$ were based on the formulation suggested by Petersen et al (1992). Values ranged from 40 m to 1600 m, depending on time of day and wind speed.

**Calculation of ingrowth/decay/attachment/removal**

Figure 1 shows the model used for calculation of ingrowth, decay, attachment and removal of Rn and Rn progeny. $\lambda_1$ ... $\lambda_5$ represent the radioactive decay constants for $^{222}$Rn … $^{214}$Po, while $\lambda_A$ is the rate constant for attachment of Rn progeny. $\lambda_{RU}$ and $\lambda_{RA}$ are the rate constants for removal of Rn progeny by rain for unattached and attached progeny, respectively. $\lambda_{DU}$ and $\lambda_{DA}$ are the corresponding constants for dry deposition.

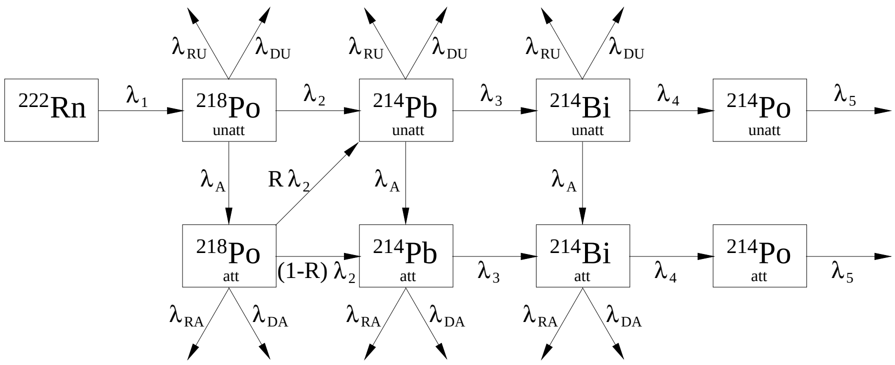


**Figure 1** Ingrowth/decay and attachment/detachment model for Rn and Rn progeny

$R$ is the probability of attached $^{218}$Po atoms becoming unattached following $\alpha$ decay due to recoil and has been taken as 0.8 (Porstendörfer 1994). The attachment rate constant ($\lambda_A$) is calculated from the product of $\beta$, the attachment coefficient of Rn progeny to an aerosol particle [$cm^3\ s^{-1}$], and $CN$, the condensation nuclei number concentration [$cm^{-3}$]. $\beta$ has been estimated from measurements for the open atmosphere reported by Porstendörfer and Mercer (1978).

Rate constants for removal of attached and unattached progeny by dry deposition were estimated using values published by Butterweck (1991). Rainfall scavenging rate coefficients have been estimated from the semi-empirical derivation of Seinfeld (1986).

# Input data

A 1-year meteorological dataset collected at Jabiru East between February 1989 and February 1990 was used. This dataset has hourly records for wind speed, direction and $\sigma_\theta$ (wind direction standard deviation) at 17 m height and for rainfall at ground level. Condensation nuclei concentrations were obtained from the study of Akber and Pfitzner (1994).

Akber et al (1993) reviewed the available data for Rn exhalation fluxes from the Ranger operation, and their recommended values have been used. These were: 300, 3760, 175, 1730 and 1090 kBq $s^{-1}$ for pit 1, the ore stockpile, waste rock dump, mill plant and tailings dam, respectively.

## Results and discussion

Table 1 shows a comparison of model predictions with experimental results (Akber et al 1994a) for the yearly average Ranger mine-derived signal for $^{222}$Rn and Rn progeny PAEC at 15 m height for Jabiru East and Jabiru Town. Unfortunately, experimental data for the mine-derived unattached fraction are not available for comparison with the predictions of the model.

Overall, the agreement between model and experimental results is reasonable (about a factor of two), and it would be advisable to obtain improved input parameters and to carry out testing at other sites before further refinement of the dispersion modelling approach is attempted.

**Table 1** Comparison of model predictions and experimental results (Akber et al 1993, 1994a) for the yearly average mine-derived signal at 15 m height for Jabiru East and Jabiru Town, March 1989 to February 1990

| | $^{222}$Rn (Bq $m^{-3}$) | | Progeny PAEC (nJ $m^{-3}$) | |
|---|---|---|---|---|
| | Model 10–20 m | Expt 15 m | Model 10–20 m | Expt 15 m |
| Jabiru East | 4.3 | 7 | 5.6 | 9.8 |
| Jabiru Town | 2.5 | 2 | 4.2 | 3.1 |

Table 2 shows the yearly average results from the model for the grid height 0–10 m at a number of human occupation sites (arranged in order of increasing distance from Ranger). As expected, $^{222}$Rn concentrations generally decrease with distance from Ranger. The low predicted concentrations for Magela 009 and Mudginberri are due to the fact that the predominant wind direction is from the east/south-east, whereas Magela 009 and Mudginberri lie towards the north of Ranger. The predicted equilibrium factors increase with distance from Ranger, while the unattached fractions decrease. This is due to the increasing time available for ingrowth and attachment of Rn progeny with greater travel distance.

**Table 2** Summary of model predictions for the yearly average mine-derived signal at 0–10 m height for Jabiru East, Magela 009, Jabiru Town and Mudginberri, Feb 1989 to Feb 1990

| Location | $^{222}$Rn (Bq $m^{-3}$) | Progeny PAEC (nJ $m^{-3}$) | Equilibrium Factor $E$ | Unattached Fraction $f_p$ |
|---|---|---|---|---|
| Jabiru East | 6.0 | 2.9 | 0.09 | 0.43 |
| Magela 009 | 1.2 | 1.0 | 0.16 | 0.27 |
| Jabiru Town | 3.1 | 2.6 | 0.16 | 0.29 |
| Mudginberri | 0.8 | 0.8 | 0.18 | 0.24 |

The results from the model runs imply that $f_p$ will be high for the Ranger mine-derived fraction of Rn progeny at the relevant living areas. This finding supports the conclusions of Akber and Pfitzner (1994), based on experimental data, that (total) $f_p$ values for the open air in the ARR are unusually high. The reason for this is that the area is remote from major industrial activity (apart from the Ranger mine itself), and so condensation nuclei concentrations are very low.

### Sensitivity to model parameters

Table 3 shows the results of a sensitivity analysis of the model to twelve of the main model parameters (the Wet season month of December 1989 was used in this analysis). The table shows the change in predicted values for mine-derived $^{222}$Rn concentration, equilibrium factor and unattached fraction at Jabiru Town (0–10 m height) when each parameter is independently increased by 10% from its base value.

**Table 3** Variation (%) in predictions for Jabiru Town with an increase of 10% in various model parameters, calculated for the meteorological dataset for December 1989. Calculation of the base values for $\sigma_y$, $\sigma_z$, $H_m(T)$ and $Q$ is complex and is detailed in Martin (2000).

| Parameter | Base value | $^{222}$Rn | Equilibrium factor $E$ | Unattached fraction $f_p$ |
|---|---|---|---|---|
| Attachment rate constant β ($cm^3\ s^{-1}$) | $6.0 \times 10^{-7}$ | 0 | 3.7 | -5.3 |
| Unatt. removal rate by rain $\lambda_{RU}$ ($s^{-1}$/(mm $hr^{-1}$)) | $1.4 \times 10^{-6}$ | 0 | $-1 \times 10^{-4}$ | $-4 \times 10^{-5}$ |
| Att. removal rate by rain $\lambda_{RA}$ ($s^{-1}$/(mm $hr^{-1}$)) | $8.0 \times 10^{-9}$ | 0 | $-7 \times 10^{-7}$ | $7 \times 10^{-7}$ |
| Unatt. deposition $\lambda_{DU}$ ($s^{-1}$) | $2.2 \times 10^{-2}$ | 0 | -1.9 | -2.2 |
| Att. deposition $\lambda_{DA}$ ($s^{-1}$) | $3.3 \times 10^{-4}$ | 0 | -1.7 | 1.7 |
| α-recoil detachment $R$ | 0.8 | 0 | -1.1 | 1.5 |
| Wind speed exponent $n$ | 0.17 | 4.5 | 1.5 | -1.6 |
| Horizontal dispersion $\sigma_y$ | | 0.41 | 0.27 | -0.16 |
| Vertical dispersion $\sigma_z$ | | -2.9 | 3.2 | 0.66 |
| Mixing layer height $H_m(T)$ | | -1.1 | -0.88 | 0.59 |
| Rn exhalation fluxes $Q$ | | 10.0 | 0 | 0 |

Predictions for $^{222}$Rn concentrations were found to be most sensitive to changes in the source Rn exhalation fluxes, the wind speed exponent ($n$) and vertical dispersion ($\sigma_z$ and $H_m(T)$). Equilibrium factor and unattached fraction predictions were most sensitive to changes in the attachment rate constant, deposition rate to the ground, and vertical dispersion ($\sigma_z$ and $H_m(T)$). As a result of this analysis, these factors will be targeted in future projects on atmospheric dispersion of radon and progeny in the region.

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# Radon exhalation rate from the rehabilitated Nabarlek surface

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## Introduction

One of the major data requirements for atmospheric dispersion models is detailed knowledge of the source term. In the case of radon dispersion in the environment, this is the radon exhalation rate from the ground surface. Ideally, variation both geographically and with time (ie season and time of day) should be known.

This paper describes research undertaken to obtain improved estimates of radon exhalation rates in the Alligator Rivers Region, particularly from the rehabilitated Nabarlek site. Once this work is completed, the results of dispersion models will be compared with measurements being made of radon concentrations in air at and near the site. Three separate sub-projects will be discussed here, relating to geographic variability over the Nabarlek site, and seasonal and diurnal variability.

In this paper, 'radon' refers specifically to the isotope $^{222}$Rn, while 'thoron' refers to $^{220}$Rn. $^{222}$Rn is a member of the uranium decay series and has a halflife of 3.8 days, while $^{220}$Rn is a member of the thorium decay series and has a halflife of 56 seconds.

## Geographic variability study at Nabarlek

Figure 1 shows the layout of the Nabarlek minesite during operations. No separate tailings dam was required since tailings could be placed directly in the mined-out pit. Following completion of milling operations in 1988, the tailings were covered by geotextile followed by a graded rock and leached sand layer of 1 to 3 metres. With final decommissioning of the mine in 1995, remaining contaminated material and unsaleable plant equipment were placed in the pit and covered with another layer of waste rock. Most of the other mine areas (with the exception of the topsoil stockpile and plant areas) were left covered with run-of-mine waste rock.

The work described here was undertaken to give information on the radon exhalation rate from this rehabilitated area, including information on the geographic variability over the site. The measurements were made in the late Dry season of 1999 (August/September) since the soil moisture content, and hence also the radon exhalation rate, should be reasonably stable at this time of year.

Measurements were carried out using the charcoal cup technique. Cups containing activated charcoal were left in place over a 3-day period before removal and counting on a $\gamma$-ray spectrometer. This method gives a measure of the exhalation rate of radon but not of thoron.

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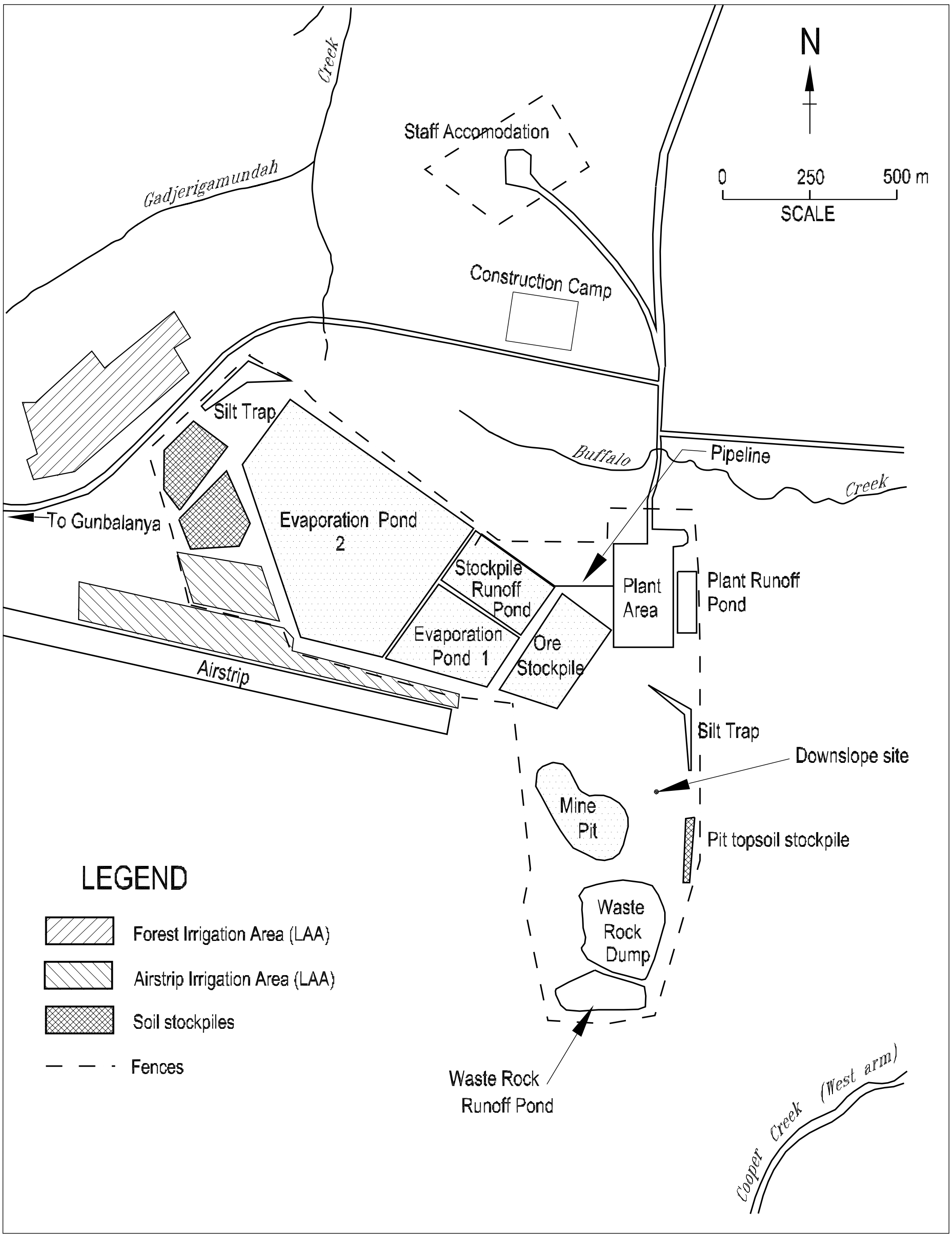


**Figure 1** Map of the Nabarlek site showing locations of major areas during operations, and the 'downslope' radon exhalation study site

Table 1 shows the results for radon exhalation rate for measurements made over the former locations of various structures on the Nabarlek fenced area. Also given is the total radon flux (in kBq $s^{-1}$) over each of these areas. Of these, the former pit gives rise to the greatest radon flux despite its small area (5 ha), due to its relatively high exhalation rate. It is unlikely that this higher exhalation rate is due primarily to radon sourced from the tailings, since these are approximately 13 metres below the final ground surface, and are below the ground water level (Waggitt & Woods 1998). A more likely reason is the substantially greater depth of waste rock over the pit, compared with the other areas.

**Table 1** Original area, and measured post-rehabilitation radon exhalation rates and radon flux, for major areas of the Nabarlek site

| Location | Area (ha) | $^{222}Rn$ exhalation rate ($mBq\ m^{-2}\ s^{-1}$) | | | $^{222}Rn$ flux ($kBq\ s^{-1}$) |
|---|---|---|---|---|---|
| | | Mean | Standard deviation | *n* | |
| Pit | 5 | 971 | 739 | 42 | 49 |
| Plant Runoff Pond | 1.1 | 278 | 203 | 24 | 3 |
| Ore Stockpile | 6 | 77 | 59 | 21 | 5 |
| Stockpile Runoff Pond | 3 | 137 | 120 | 18 | 4 |
| Evaporation Pond 1 | 5 | 169 | 86 | 12 | 8 |
| Evaporation Pond 2 | 25 | 103 | 102 | 83 | 26 |
| Topsoil Stockpiles | 7 | 31 | 28 | 17 | 2 |

## Seasonal variability study

Measurements of radon in air in the Alligator Rivers Region show that concentrations are lower in the Wet season than in the Dry season by about a factor of two to three (Akber & Pfitzner 1994). It was expected that at least part of this variability is due to variation in radon exhalation rates due to changes in soil moisture content. This sub-project aims to obtain quantitative data on this variability by measuring radon exhalation rates from a limited number of sites over the course of a yearly cycle.

The main part of this work is planned to be carried out in 2002 and 2003, however, some preliminary data have been collected from one site to give an indication of the variability which could be expected. The site is a forest area east of Baralil Creek near Jabiru Town. Measurements have been made at approximately monthly intervals between September 2000 and February 2001. The instrument used gives the exhalation rates (over a measurement period of 30 minutes) of both radon and thoron.

Figure 2 shows the results obtained for radon and thoron exhalation rates, as well as soil moisture content measured from a 5 cm depth sample taken close to the measurement site (ensuring that the exhalation site itself is not disturbed). The strong influence of soil moisture on the radon exhalation rate is apparent, with the rate falling from about 35–45 $mBq/m^2/s$ in September/October, through about 30 $mBq/m^2/s$ in November/December, to $0 \pm 1$ $mBq/m^2/s$ for a 15% moisture content on the February measurement.

The thoron exhalation rate also fell markedly in February. Most of the thoron can be expected to originate from the top 1 to 2 centimetres owing to its very short halflife; the change in February was therefore consistent with a hypothesis that the high moisture content was very effective at retaining radon and thoron within even the top layer of the soil.

These results demonstrate that seasonal variability in radon and thoron exhalation rates can be dramatic, giving strong evidence that this is an important factor in variation of concentrations in air. However, exhalation rate variability can be expected to vary from location to location, depending on such factors as the local drainage pattern and soil porosity. These will be particularly important when considering rates from minesite areas such as waste rock dumps.

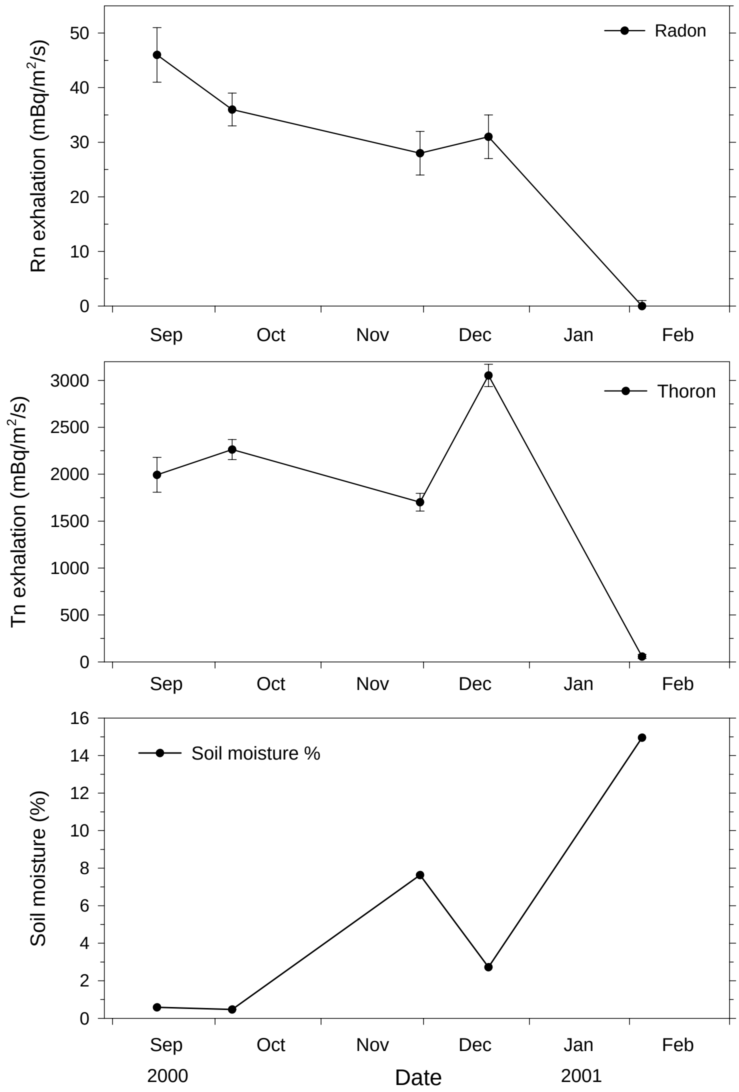


**Figure 2** Seasonal variability in radon and thoron exhalation rate, and surface soil moisture, at the Baralil Creek forest site. Error bars reflect counting statistics alone, and correspond to one standard deviation.

## Diurnal variability study

Few data are available in the literature on diurnal variability in radon exhalation rates. Todd et al (1998) measured radon and thoron exhalation rates over the course of a 24-hour period at a site at Jabiru East. Their results were equivocal, mainly due to the relatively low radon exhalation rate from this site (giving rise to large counting statistical errors), but indicated that if a diurnal cycle occurred for radon it was no greater than about 20% of the mean exhalation rate at this site.

A separate series of measurements were made between the 16$^{th}$ and 17$^{th}$ June 1998 at the 'downslope site' at Nabarlek (see fig 1). This site was chosen for study due to its relatively high radon exhalation rate. The instrument used was the same one used in the above seasonal variability measurements. Figure 3 shows that there was no apparent cycling for either radon or thoron exhalation rate over the daily cycle. A two-tailed t-test ($\alpha$ = 0.05) did not show a significant difference between daytime and nighttime exhalation rates for either radon or thoron.

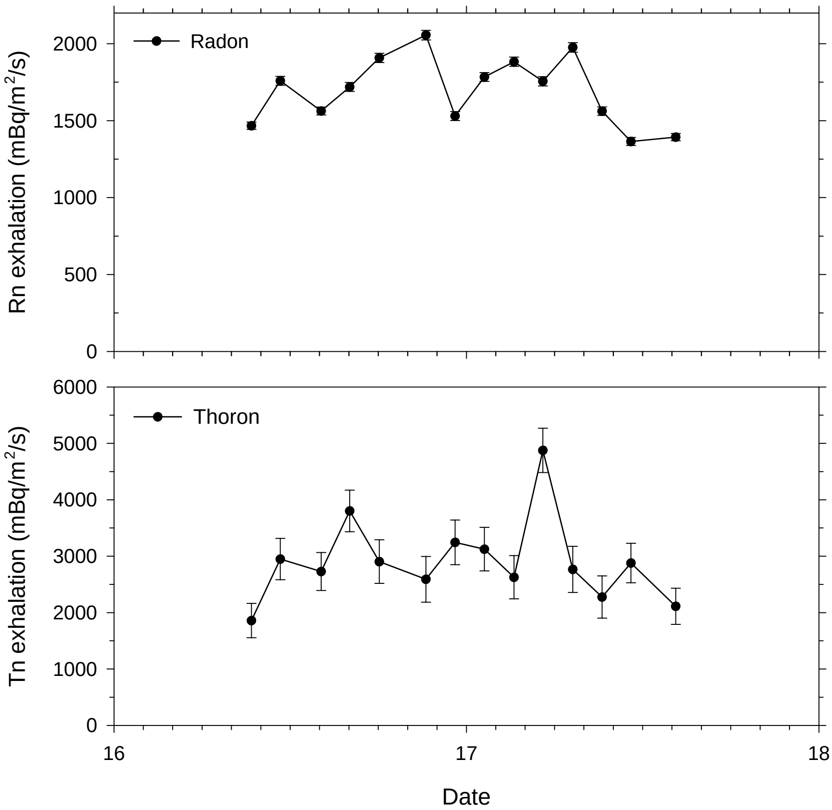


**Figure 3** Test of diurnal variability in radon and thoron exhalation rates at the Nabarlek 'downslope site'. Error bars reflect counting statistics alone, and correspond to one standard deviation.

# Conclusions

Data have been obtained for the radon exhalation rate from major areas of the rehabilitated Nabarlek minesite for the August/September period. The majority of the radon flux from the measured areas is sourced from above the former pit.

Preliminary data show that the difference in radon exhalation rate between Dry and Wet seasons can be large. However, this variability is expected to depend upon site-specific factors and so further, detailed studies are planned. The available data on diurnal variability indicate that it is not large, at least during the Dry season.

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# Identification of traditional Aboriginal foods for radiological assessment

**B Ryan & P Martin**

## Introduction

Previous studies conducted by ***eriss*** of radionuclides in Aboriginal bushfoods have focussed on bioaccumulation by aquatic animal and plant species (Pettersson et al 1993, Martin et al 1998). This was due to the importance of the aquatic transport pathway, particularly during the operational phase of uranium mining operations. Despite the work which has been carried out to date, there remain several significant areas of uncertainty, including the following:

- Knowledge of the bushfood consumption by the relevant Aboriginal groups is poor. This includes knowledge of both the range and quantities of foods eaten, as well as of food preparation methods (eg cooking methods).
- There is little available information on radionuclide uptake by terrestrial animals and plants. This pathway becomes particularly important once rehabilitation of minesites occurs.

This project aims to address these issues. Due to the large range of bushfoods eaten, a staged approach has been taken. Initially, work concentrated on consultation with Aboriginal people. This was followed by general observations of bushfood consumption. In addition, some studies of bioaccumulation by edible plants have been undertaken, primarily because of the importance of these in terms of rehabilitation.

## Rehabilitation issues

The Commonwealth Government and the NT Government have agreed upon conceptual plans for rehabilitation of the Ranger Uranium Mine (RUM) site when mining has ceased. The major objective relevant to revegetation is:

> To revegetate the disturbed sites of the Ranger Project Area with local native plant species similar in density and abundance to that existing in adjacent areas of Kakadu National Park, in order to form an ecosystem the long-term viability of which would not require a maintenance regime significantly different from that appropriate to adjacent areas of the Park.

After the rehabilitation of the RUM site there will be radiological protection issues associated with the land use expectations by local Aboriginal people. Local people may want to use the abandoned mine areas for hunting and gathering, if there is food accessible and available. Of the nineteen species that have currently been flagged as rehabilitation plants (RUM Rehabilitation #26 2001) seventeen have direct uses for local Aboriginal people. These uses range from using the wood for digging sticks and didgeridoos to the harvesting of fruit from fruit trees. Therefore the planting of some of these species will encourage the occupation and use of the rehabilitated contaminated sites by local Aboriginal people for various periods of time.

In Momega (western Arnhem Land), Altman (1984) identified over 80 floral species used as bush foods, and in total 170 species of flora and fauna were observed being consumed. The foods that were chosen for initial study after discussion with local Aboriginal people in Kakadu and people who dealt with Aboriginal people on a daily basis were a number of common fruits and yams, for which no radiological data existed. These fruits and yams can play a significant role in the diet of an Aboriginal person at different times of the year.

The results in table 1 obtained for four species of common fruit will be discussed here; these are *Buchanania obovata*, *Persoonia falcata*, *Vitex accuminata* and *Syzygium eucalyptoides*.

**Table 1** Fruits analysed in the Alligator Rivers Region

| Scientific Name | Common Name | Gundjehmi name |
|---|---|---|
| *Buchanania obovata* | Green Plum | *Andudjmi* |
| *Persoonia falcata* | Geebung | *Andaak* |
| *Vitex accuminata* | Black Plum | *Anbalindja* |
| *Syzygium eucalyptoides* | White Apple | *Anbongbong* |

## Sample description, collection and preparation

After extensive consultation with and advice from local organisations it was arranged that an ***eriss*** researcher would accompany Aboriginal people from the Kakadu Family Resources Centre on food gathering trips. These expeditions began in 1997 and continued through to 2000. When on these trips, the researcher would act as a driver and an observer only, gleaning information from Aboriginal people about collection techniques, cataloguing food collected and observing methods of preparation. Information on the amount of bush food consumed, where it was gathered and the time of year was also recorded.

*Buchanania obovata* or *andudjmi* was collected from two sites on the Ranger Uranium Mine lease area. These fruit ripen during the pre-monsoon storm season (late Oct-Nov-Dec) or *Gunumeleng*. The first site was situated approximately 200 metres west of the tailings dam western wall and the second site in the former Jabiru East town area near the Gagadju workshop. A tarpaulin was put under the tree and then the tree was shaken, with fruit falling from the branches. The ripe fruit were then collected and bagged. These fruit are often eaten raw when collected or sometimes they are taken and pulped into a paste and then eaten.

*Persoonia falcata* or *andaak* were collected from the old Jabiru East town site from several different trees in the area. These fruits are available during *Gunumeleng* and *Gudjeuk* (monsoon season Jan–March). The sample collection method was identical to *Buchanania obovata*. The fruit are dried and eaten after soaking.

*Vitex accuminata* or *anbalindja* were collected from the East Alligator area, within the boundaries of the park ranger station and are also available during *Gunumeleng* and *Gudjeuk*. The same method was used for collection as for previous fruits. These small black fruit are eaten raw after collection and are sweet tasting.

*Syzygium eucalyptoides* or *anbongbong* were also collected at the East Alligator Ranger Station and are available during *Gunumeleng* and *Gudjeuk*. The ripe fruits were picked directly off the tree and are pink in colour; they are eaten raw.

All fruit samples were weighed and then oven dried at 60°C, then crushed with a mortar and pestle to a fine powder. The fruit samples are more commonly unwashed when consumed, so they were not washed when prepared for analysis. Artificial tracer isotopes of known concentration were added to the samples and the samples were then digested completely using concentrated $HNO_3$ and HCl. Analysis was performed by $\alpha$-spectrometry. Sample sizes ranged from 2 to 10 grams, whilst detection limits were 0.1 mBq per sample.

Soil samples were taken from the base of the trees that the fruits were collected from. The samples were weighed and oven dried at 60°C, then ground to a fine powder using a ring mill prior to analysis. The sample was then cast into a resin disc and counted using low level, high-resolution $\gamma$-spectrometry as described by Murray et al (1987).

## Results

Radionuclide analysis results are shown here as dry fruit concentration (Bq/kg) and as a concentration factor (CF).

The CF for a nuclide in an organism is defined as the activity of the nuclide per unit fresh weight (or wet weight) of the organism, divided by the activity of the same nuclide per unit weight of substrate (the physical medium from which the organism is assumed to obtain the radionuclide). In the present case the substrate is the soil the fruit tree grows in.

Concentration factors enable a prediction of the radionuclide concentrations which will be present in an edible food for a given substrate concentration. The concentration factor method is discussed in detail in ICRP (1978) and IAEA (1982).

In the case that site- or species-specific data are not available, default concentration factors for generic food types are often used for dose predictions. The International Atomic Energy Agency (IAEA) publishes such default values. Those for crops are based upon common agricultural crops such as above ground vegetables, leafy vegetables, root vegetables and grains and are intended to represent the edible parts at crop maturity. Default values are given in the following tables for comparison purposes, and are from IAEA (1982) Table XVII, page 64.

### Radium

The $^{226}Ra$ concentrations for both soil and fruit were an order of magnitude higher for the *Buchanania obovata* than for any of the other fruits. This may be partly explained by the *Buchanania obovata* fruit tree location, which is situated on the Ranger Uranium mine lease area. The *Persoonia falcata* is also on the mine lease area but it is not near an access road and is surrounded by moderately thick bushland, thus protected from the dust thrown up by traffic on the unsealed road. The CFs in table 2 for $^{226}Ra$ for all four of the fruits are similar to the default values.

**Table 2** Derived and default IAEA concentration factors for $^{226}Ra$

| Species | Dry Fruit Conc $^{226}Ra$ (Bq/kg) | Conc. Factor (CF) | Default IAEA CF |
|---|---|---|---|
| **Buchanania obovata* | 17.1 | 0.022 | 0.04 |
| *Persoonia falcata* | 0.7 | 0.013 | 0.04 |
| *Vitex accuminata* | 2 | 0.037 | 0.04 |
| *Syzygium eucalyptoides* | 1.3 | 0.012 | 0.04 |

*Average of the tailings dam site and Gagadju workshop site

### Polonium

For calculation of CFs for $^{210}Po$, the soil concentrations have been assumed to be the same as those for $^{210}Pb$. Derived CFs in table 3 for $^{210}Po$ in the *Buchanania obovata*, *Vitex accuminata* and *Syzygium eucalyptoides* are one order of magnitude higher than the default values. The derived CF for the *Persoonia falcata* is two orders of magnitude higher than the default CF.

**Table 3** Derived and default IAEA concentration factors for $^{210}Po$

| Species | Dry Fruit Conc. $^{210}Po$ (Bq/kg) | Conc. Factor (CF) | Default IAEA CF |
|---|---|---|---|
| **Buchanania obovata* | 3.7 | 0.007 | 0.0002 |
| *Persoonia falcata* | 3.7 | 0.046 | 0.0002 |
| *Vitex accuminata* | 0.5 | 0.004 | 0.0002 |
| *Syzygium eucalyptoides* | 1.6 | 0.007 | 0.0002 |

* Average of the tailings dam site and Gagadju workshop site

## Uranium

Measured CFs in table 4 for $^{238}U$ in the *Buchanania obovata*, *Persoonia falcata* and *Syzygium eucalyptoides* compare favourably with the default values. The derived CF for *Vitex accuminata* was about five times higher than the default CF.

**Table 4** Derived and default IAEA concentration factors for $^{238}U$

| Species | Dry Fruit Conc $^{238}U$ (Bq/kg) | Conc. Factor (CF) | Default IAEA CF |
|---|---|---|---|
| **Buchanania obovata* | 0.6 | 0.001 | 0.002 |
| *Persoonia falcata* | 0.4 | 0.004 | 0.002 |
| *Vitex accuminata* | 0.4 | 0.011 | 0.002 |
| *Syzygium eucalyptoides* | 0.1 | 0.001 | 0.002 |

* Average of the tailings dam site and Gagadju workshop site

## Order of importance

Table 5 shows, for each of the fruits studied, the relative order of importance of the radionuclides $^{210}Po$, $^{226}Ra$, $^{234}U$ and $^{238}U$, in terms of radiological dose for a mine rehabilitation situation. This was calculated by multiplying the concentration factor by the dose conversion factor for ingestion of the relevant radionuclide, and then expressing the result as a percentage of the same calculation for $^{226}Ra$. This calculation assumes secular equilibrium for the uranium series radionuclides in the soil, which is reasonable for mine waste rock covering a rehabilitated site.

**Table 5** Radionuclide relative order of radiological importance

| Species | $^{210}Po$ | $^{226}Ra$ | $^{234}U$ | $^{238}U$ |
|---|---|---|---|---|
| *Buchanania obovata* | 143 | 100 | 0.9 | 0.8 |
| *Persoonia falcata* | 1571 | 100 | 5.3 | 4.8 |
| *Vitex accuminata* | 43 | 100 | 5.3 | 4.8 |
| *Syzygium eucalyptoides* | 257 | 100 | 1.1 | 1.0 |

The table shows that the general order of importance was: $^{210}Po \geq {}^{226}Ra \gg [{}^{234}U \approx {}^{238}U]$. Consequently, it will be more useful to target $^{210}Po$ and $^{226}Ra$, rather than the uranium isotopes, in any future research studies and/or monitoring regimes on this topic.

## Conclusions

Wild fruit and vegetables play an important part in a traditional Aboriginal diet, and radionuclide uptake by these foods will be particularly important for the post-rehabilitation situation for uranium mines in the Alligator Rivers Region. Unfortunately, in comparison with situation for aquatic flora and fauna, there is relatively little information available for radionuclide concentrations in local fruit and vegetables.

In this paper, data have been presented for several fruits. In terms of radiological dose for a mine rehabilitation situation, $^{210}$Po and $^{226}$Ra were found to be of greater importance than the uranium isotopes. The local values for concentration factors are sometimes up to two orders of magnitude different from the IAEA temperate environment default values. These results highlight the need to use local values wherever possible.

Other important factors that have emerged include food preparation and consumption habits of Aboriginal people, as they are quite different from European techniques and could potentially affect radionuclide intake estimates. This is difficult information to obtain because of the intrusive nature of such studies, and must be undertaken over a number of years.

At present, work on this project is concentrating on analysis of yam samples.

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