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A Charcoal Canister Survey of Radon Emanation at the Rehabilitated Uranium Mine Site at Nabarlek

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SUMMARY

This paper describes a recent survey of radon emanation measurements from the rehabilitated Nabarlek mine site. It was mined out in 1979, decommissioned in 1995 and provided a good test bed for assessment of rehabilitation, in terms of radon flux attenuation. Measurements have been made with charcoal canisters. Studies to measure the radon-220 flux by observing Tl-208 progeny of thoron, the effectiveness of trial covers and meteorological considerations will be reported.

1. THE NABARLEK MINE SITE

The Nabarlek uranium mine-site is located 150 km NE from Jabiru, near Cooper Creek, a tributary of the East Alligator River. Queensland Mines Ltd mined out the Nabarlek high-grade orebody in just over 4 months in 1979. The company stockpiled 600 000 tonnes of average 2% grade ore (1). The mineralisation extended to a depth of 72 metres, with a variable thickness of about 10 metres. Rehabilitation and decommissioning works at the site ended in 1995.

In the Environmental Impact Statement, Queensland Mines stated that rehabilitation would involve transfer of tailings to the pit, evaporation of surplus water in the pit during the dry season and then backfilling and covering with layers of low permeability clay, then sealing with waste rock. The plan was then to reapply the topsoil initially removed and revegetate the landscape. The resultant rehabilitation has included a "rip-rapped" surface to inhibit surface erosion and forming trenches about 1.5 metres deep. By 1999, these appeared in most places to have to have been filled in to a present depth of about 0.3 to 0.6 metres.

It is well established that inhalation of radon progeny from the radioactive decay of radon gas, is a potential health hazard, so that exposure limits (4 Working Level Months/yr) are recommended for the site by advisory and regulatory bodies (2). In a survey of the Nabarlek mine site, Leach and Lokan (3) measured the radon flux using brass canisters, filled with charcoal and set out as an array over the area of interest.

The aim of the present survey was to continue previous surveys of the site most recently that conducted by Martin (4) in the early 1990's. The grid locations of sites for the new survey were selected according to their average gamma dose rates measured in the earlier work. They included the former pit, plant runoff pond, ore stockpile area, ore stockpile run off pond, evaporation ponds and topsoil stockpile areas. The photograph in figure 1 which shows a control point on site with 9 charcoal canisters and 4 electrets in foreground.

2. MEASUREMENT DETAILS

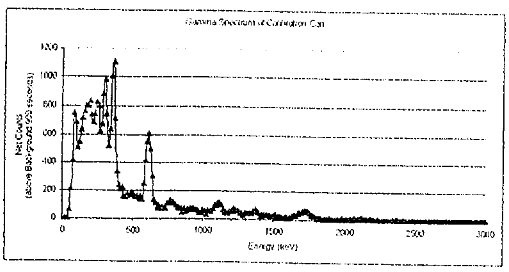
2.1 Detection by Charcoal Cups

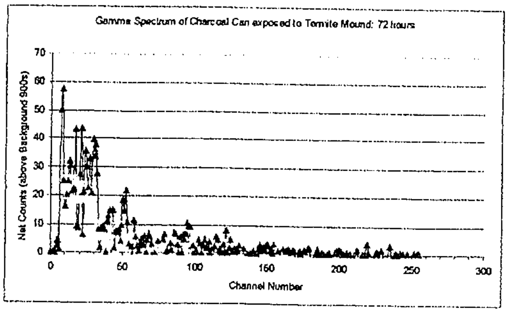
The charcoal cup method for determining radon emanation or concentration, uses the adsorption capacity of activated charcoal for radon-222 (radon) and radon-220 (thoron). One gram of activated charcoal typically adsorbs 2 to 6 Bq radon from the air with a radon activity concentration of 1 Bq l^{-1} (5). The minimum detectable activity for a 25 g charge of charcoal, based on background counts in our counting chamber is 2.2 Bq for the 100 litre air volume sampled or ~ 0.02 Bq l^{-1} (at 95% confidence).When used as a detector in a standard brass canister of area 0.002922 m^2, exposed to the surface for 3 days.

emanometer, an area sensitivity of 3 mBq m^{-2} s^{-1} may be inferred from the activity of 2.2 Bq measured

2.2 Spectra Measurements

Canisters used to measure emanation rates are embedded in the earth to a depth of ~1 cm, or mud is applied around the rim to prevent leakage of radon. In hard rock, the can may be fixed to and then sealed with a convenient putty such as *KneadIt*. The effect of temperature variation at the site was studied by a test site at ERISS.





Detection of the radon is measured by the radioactive emanations of its progeny and gamma ray spectroscopy is a method of doing this. Usually, a single channel analyser is used to record the gamma ray emanations from the Bi-214 radon daughter, which emits gamma photons at an energy of 609 keV. The authors previously used this method (6) and estimated emanation rates from a system with a counting efficiency of 0.7 %. (0.007 counts.s^{-1} Bq^{-1})

Counting is performed at least three hours after the canisters are collected, to allow secular equilibrium to be established for the progeny Pb-214 (half-life 27 minutes) and Bi-214 (20 minutes), which are relatively short-lived compared with their parent radon (3.82 days). In the current survey, gamma spectrometry furnished a method for counting peaks due to several progeny of radon and also determining possible contamination due to other radioactive species (such as thoron). A portable 3" × 3" NaI(Tl) spectrometer counted gamma ray emanations in a lead castle in 256 channels over a range from 12 keV to 3 MeV, in windows of width 12 keV. This meant that as well as Bi-214 (609 keV), counts from Pb-214 (295 and 395 keV) and Bi-214 (1120 and 1740 keV) could be used to improve counting efficiency. The remaining lines from Pb-214 contributed to a counting efficiency of about 3 % (0.03 counts s^{-1} Bq^{-1}).

The portable spectrometer uses a 15 kBq Cs-137 source as a reference and for internal spectrum stabilisation. However it had to be removed so that it would not interfere with the Bi-214 (609 keV) line. No internal stabilisation was employed. Instead, the regions of interest for the Pb214 (295, 353 keV) and Bi-214 (609 keV) were defined in every survey, by comparing the strongest spectrum from the charcoal cans to that of the calibration can, normalised to the Pb-214 and Bi-214 peaks of interest. Drift in the counts in channels within each Region of Interest (ROI), was checked and the variation over the ten minute counting interval per canister, was found to be indistinguishable from Poisson statistics.

The radon emanation rate J in Bq m^{-2} s^{-1} is then calculated using the formula (7):

$$J = \frac{N.\lambda^2.e^{(\lambda.td)}}{\varepsilon.A.(1 - e^{(-\lambda.te)}).\ (1 - e^{(-\lambda.tc)})}$$

where N is the net counts in the regions of interest,
λ is the decay constant for radon,
t_d is the delay period from the end of exposure to the beginning of the counting interval
ε is the counting efficiency of the system in the regions of interest
A (m^2) is the area of the canister
t_e is the period of exposure of the charcoal in the canister
t_c is the counting interval of the charcoal.

The radon activity concentration in the air in Bq m^{-3} is calculated using the formula (8):

$$[Rn] = \frac{N.e^{(\lambda.(te/2 + td)}}{\varepsilon\ CF\ t_e}$$

where CF (m^3 s^{-1}) is the calibration factor, or volume sampling rate at a specific temperature and humidity relating the radon concentration to activity in the can.

2.3 Thoron Contamination

It has been remarked that the photopeak due to the 609 keV gamma rays from Bi-214 must be determined after the thoron progeny Tl-208 has decayed, so that the photopeak of the latter, at 580 keV, does not pile onto that of the former (9). Indeed, the consequences of the photopeak at 2,600 keV from Tl-208 could also result in a contribution to the counts for Pb-214 (295 and 353 keV) as well as the Bi-214, due to Compton scattering.

To investigate this, an exposed charcoal canister was corrected for background and then subject to spectrum stripping, by subtracting from it the spectrum of the cali-bration can which is solely due to radon-222 progeny. The results are presented in Figure 2 and by inspection, it appears that from 25 g of charcoal, there is no contribution from thoron progeny; certainly no obvious photopeaks due to Tl-208 at 2,600, 510, 580 or 860 keV.

This would be supported by an estimation of the amount of radon-220 adsorbed relative to the radon-222. Given that the flux ratio of radon to thoron at Nabarlek is expected to be 1:40, while the decay constants are 6,000:1, it is expected there will be about 140 times the number of radon atoms adsorbed onto the charcoal.

3. METHODOLOGY

Since the rip-rapped surface at Nabarlek follows an undulating pattern across the landscape, it was important not to introduce bias in the location of survey points. A regular grid may have located points on the crests or troughs of the landscape, rather than an equal selection of both. Moreover, revegetation has produced heavily grassed areas and patches with significant leaf litter. It was decided therefore to randomise the selection of survey points, to ensure independence.

A grid was mapped onto each location, each square 50x50 m^2 and co-ordinates for the survey point in each grid were computed from randomly generated numbers. Given the limitations of time, the number of points was typically limited to about 10. When more than about a dozen grid squares covered a given location, the restriction of time demanded that certain grid squares be selected and a random number generator was again used to choose which these would be. In total, 84 survey points were established on and off site; including nine environmental sites selected at arbitrary points that ranged from 50 to 2 000 metres from the perimeter fence. The x and y coordinates in each selected grid square were transformed to r,θ coordinates (range and bearing) and then a prismatic compass and tape measure used to approximately locate these points on site. The exact co-ordinates were then established using the Aircheck Digital Global Positioning System (DGPS), as eastings and northings in accord with WGS84. Additionally, three of the survey points were repeatedly measured over the 40 days of the survey, to gauge possible variation due to seasonally dependent parameters over that time. These control points included one point 500 metres off site, at the turnoff to the Myra Camp.

4. RESULTS

Table 1 includes the radon flux and radon activity concentration measured at the Control Sites. It is concluded that the radon flux has not been measurably affected by temperature variations of the canisters under field conditions. Similarly, the deployment of canisters in the morning or evening appears to be of little consequence and the use of insulation marginally reduces the standard deviation in the flux measured from the ground.

Table 2 shows the radon flux from selected locations on the Nabarlek Site. These results compare with the survey by Kvasnicka 3 years ago, when he estimated 4710 $mBq.m^{-2}\ s^{-1}$ for the former pit and 840 $mBq.m^{-2}s^{-1}$ for the Evaporation Pond 2. From the photograph of the site, it is clear that embedded rock forms a significant portion of the emanating surface, and their inclusion reduces the flux from measurements exclusively based on soft, porous material of the surrounding earth. These results must be normalised to their radium-226 content and, with radon in-growth over the next month, the samples will be assayed for this content.

By way of intercomparison, electrets were deployed with the charcoal canisters used to measure radon concentration and both were deployed alongside the radon station.

In 1993, the design for rehabilitating the pit area was unchanged; mill arisings of 680 000 tonnes compacted by a bulldozer, a top layer of stockpile pad and waste rock, then surface preparation for seeding. A 23 m cover at the centre and 14 m at the edge was planned, with a 1:20 slope. There was no anticipated problem from gully erosion at these slopes and slope lengths. The estimated erosion rate of 25 to 100 mm per 1,000 years in 1985, was revised to 22 mm per 1 000 years, with a calculated 635 000 years before exposure of the highest point of the tailings. It was further calculated that 5% of its present radioactivity would remain after that time.

The half value layer for compacted clay and screened (35mm) waste rock were both determined in 1986 to be approximately 80 mm for gamma radiation, which they assumed adequately attenuated, by the minimum 14 m thick cover. The radon flux from the bare tailings was calculated at 3.63 $Bq\ m^{-2}\ s^{-1}$ and the attenuation factor of the cover to be 2.00×10^{22}, following the Code of Practice and assuming 2.39×10^{12} Bq radium in secular equilibrium with the uranium, in the 680 000 tonnes of deposited tailings.

The calculations were similar to those performed in 1985, where the radon flux was determined to be $1.81 \times 10^{-22}\ Bq\ m^{-2}\ s^{-1}$ and the clay layer was removed from the design, since it was considered to have minimal effect.

We suggest that the source of radon contributing to an average flux from the pit of $1080 \pm 860\ mBq\ m^{-2}\ s^{-1}$ is the waste rock.

ACKNOWLEDGMENTS

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TABLES

Table 1 includes the radon flux and radon activity concentration measured at the Control Sites.

Table 1: Summary of the Flux and Concentration at the Control Sites

site:								
	MYRA	**month**	Aug	Aug	Aug-Sep	Sep		
		date	13-16	20-23	30-2	10-13		
	J	**(mBq.m⁻².s⁻¹)**	**18**	**17**	**21**	**17**		
	[Rn]	**(Bq.m⁻³)**	**47**	**-11**	**32**			
	RADON STATION	**Month**	Aug	Aug	Aug	Aug-Sep	Sep	Sep
		Date	13-16	20-23	24-27	30-2	4-7	10-13
	J	**(mBq.m⁻².s⁻¹)**	**90**	**95**	**95**	**107**	**93**	**97**
	[Rn]	**(Bq.m⁻³)**	**98**	**139**	**201**		**133**	
	PLINTH D	**month**	Aug	Aug	Aug	Aug-Sep	Sep	Sep
		date	13-16	20-23	24-27	30-2	4-7	10-13
	J	**(mBq.m⁻².s⁻¹)**		**1125**		**857**		**866**

The average values for the radon-222 flux from each location on the Nabarlek site are presented in Table 2 below:

Table 2: Radon Flux from selected locations on the Nabarlek Site

SITE	J	+/- s.d.
	(mBq/m2 /s)	(mBq/m2/s)
PIT	1082	865
PROP	273	205
ORE STOCKPILE	77	60
ORE STOCKPILE RUNOFF POND	152	131
EVAPORATION POND 1 / TAILINGS DAM	172	86
EVAPORATION POND 2	105	101
TOPSOIL STOCKPILE	25	21
ENVIRONMENTAL SITES	90	137