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Measurements of aarnma radiation in the Fishermans Bend waste drum storage compound on 29/11/91

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1. <u>Summarv</u>

The purpose of this work was as follows:

(a) to explore whether calculations and measurements of dose rates from drums containing wastes from Fishermans Bend could indicate if the radioactive content of the drums was greater than the specific activity of 100 Bq per gram which is the legal definition of a "radioactive substance" in the Regulation of the NSW Radiation Control Act 1990 No. 13.

(b) to estimate dose rates at 10 m from an array of drums.

Estimates have been made of surface gamma dose rates from a 200 L drum containing (a) uranium ore @ 0.3% U₃O₈ from which 95 % of U had been extracted, and of dose rates 10 m distant from a stack of 15 pallets, stacked 3 tiers high containing 48 drums, (b) surface gamma dose rates from a 200 L drum containing monazite ore containing 9.5% thorium.

Measurements have been made of surface dose rates from a random selection of drums in the Fishermans Bend compound. Several surface dose rate measurements indicate that if the contents of the drums were homogeneous, several of the few drums monitored could contain a radioactive content in excess of 100 Bq per gram.

Measurements made of the gamma radiation dose rates at the perimeter fence in the vicinity of the Fishermans Bend drum stacks were indistinguishable from variations in the natural background radiation.

2. Lavout of Fishermans Bend Drum Storaae

Drums containing wastes from Fishermans Bend are located in three stacks in a fenced compound at LHRL

(Figure 1). Drums are stored on a square close spacing on pallets, i.e., 4 drums per pallet, with pallets stacked in three tiers. The pallets are generally close spaced with some access corridors through the stacks. Stage 1 storage consists of two stacks containing 1260 and 1824 drums, of approximate areal dimensions 10 m x 20 m and 32 m x 12 m, respectively. The stack

containing 1260 drums consists of two arrays, the first 15 pallets long x 4 pallets wide with a lengthwise corridor about one metre wide between the second array of 15 pallets long x 3 pallets wide. The stack containing 1824 drums consists of two arrays, each of19 pallets long x 4 pallets wide separated by a lengthwise one metre wide corridor . Stage 2 storage containing 6649 drums on an area of about 63 m x 20 m consists of ten arrays separated by nine corridors along the width axis. Six arrays each containing 720 drums are 15 pallets long x 4 pallets wide, one with 900 drums is 15 pallets long x 5 pallets wide, and three approximately 10 pallets long by 4 pallets wide, contain 480, 463, and 486 drums.

3. Basis of Dose Rate Calculations

Table 1 lists isotopes in the U-238 decay chain, together with their half-lives and activities per gram of U-238. Secular equilibrium was assumed in estimation of isotopic activity per kg of uranium ore containing 0.3% U_3O_8 , and in 1 kg of tailings resulting from extraction of 95% of U-238 and U-234. A 200 L drum was assumed to contain 340 kg of tailings at density 1.7 g cm⁻³.

Table 2 lists similar data for the Th-232 series. A 200 L drum was assumed to contain 492 kg of rnonazite ore at a density of 2.46 g cm⁻³ containing 9.5% Th-232 in secular equilibrium with products of the decay chain.

4. Dose Rate Calculations

Calculations detailed in Attachment A assumed the drums to be right circular cylindrical sources of dimensions 560 mm diameter and 875 mm tall, filled with a homogeneous ore or tailings of densities as above, with shielding characteristics similar to that of soil. The front array of a stack of 8 pallets with three tiers contained 48 drums.

Tabular data on shielding calculations was limited to beyond 7 cm from the outside face of a drum, together with a separate tabulation of the upper limit of the surface dose. Estimates are as follows:

(a) From one drum of uranium tailings at 7 cm:	3.5 µSv h-1 7.0 µSv h-1
(b) Upper limit at contact:	7.0 µSv h-1
(c) Dose rate at 10 m from a stack of drums:	0.4 µSv h ⁻1
(d) From one drum of monazite ore at 7 cm:	80 µŚv h-1
(e) Upper limit at contact:	100 µSv h-1
	-

5. Gamma radiation measurements

Measurements of gamma radiation flux were made on contact with a selection of drums on the outside of stacks and in access corridors in the three stacks.

Access to the sides of the majority of drums was limited to an arc of about 150 - 160 degrees per drum, with the exception of drums on the outer edges of pallets at the corners of stacks or corridors where an arc of about 300 - 320 degrees was possible. A digital readout instrument FAG FH 40F2 calibrated in μ Sv h⁻¹ was used. This instrument had a slow integration response; to expedite measurements, an Eberline ratemeter in direct readout mode was used to identify drums with greater potential gamma doserate. Measured dose rates are shown in Figure 2.

Thermoluminescent dosimeters (TLD's) were positioned for a period of 984 hours (five weeks) on the perimeter fence west of the compound containing the Fishermans Bend waste drums as shown in Figure 1. Monthly cumulative doses are listed in Table 3.

6. Discussion

6.1 Background dose rate

Monthly cumulative doses measured on the perimeter fence west of the Fishermans Bend drum stacks recorded in Table 3 were in the range 0.09 - 0.19 mSv, with a mean value of 0.12 mSvand a standard deviation of 0.04 mSv. Prior to receipt of the waste drums from Fishermans Bend, other dosimeters at the south west of the LHRL site distanced from B.59 (where thorium concentrates are stored) recorded monthly doses in the range 0.07 - 0.29 mSv (Wright, P., Private communication) with an average monthly dose of 0.11 mSv and a standard deviation of about +/- 0.05 mSv. These latter indicate that any contribution to dose rate at the perimeter fence from the Fishermans Bend drums stacks was indistinguishable from variations in the natural background.

6.2 Dose rate measurements on contact with Drums

Tables 1 and 2 indicate that a drum containing a homogeneous 100 Bq g^{-1} of either the U-238 or the Th-232 decay chain in secular equilibrium could have an upper dose rate limit of about 2.2 - 2.6 μ Sv h⁻¹ on contact. Figure 2 shows that a substantial proportion of the drums monitored were below this value. Lowest readings of 0.2 - 0.3 μ Sv h⁻¹ were recorded at the west and east faces of the Stage 2 stack and in the recess within the Stage 2 stack. These are within the range of background radiation above, suggesting that drums in this area contain little radioactive content. The dose rate at the east face of this stack sets an upper limit of 0.2 μ Sv h⁻¹ to the contribution from B.59 to the background in this vicinity.

Dose rates within corridors inside stacks were variable, with rates from 0.6 to $14 \,\mu$ Sv h⁻¹measured along one corridor. The highest dose rate detected, $14 \,\mu$ Sv h⁻¹, was measured cn a corridor along with other rates as low as 0.6 μ Sv h⁻¹. A dose rate of 12 μ Sv h⁻¹ was measured at the east face of one of the Stage 1 drum stacks. The lack of access to the drums prevented measurements which might indicate the degree of homogeneity in the drums, and some or all of the higher dose values may be due to local 'hot spots' within the drums. On the limited evidence, however, if the material in the drums is homogeneously distributed, then several of those monitored could contain . considerably in excess of 100 Bg g⁻¹of radioactive material.

The proportion of drums possible to monitor in-situ is confined to those drums on the outside of stacks and along corridor walls, i.e., a maximum of about 3108 drums out of 9733, or 31.9 per cent of drums. In the random exercise, the third tier of drums was out of reach, and not all corridors of drums were monitored. If the random exercise monitored about 10% of the total possible, then about one percent of the drums monitored had surface dose rates greater than would be expected from a radioactive content of 100 Bq per gram.

6.3 Dose rates at a distance from an array of drums

A dose rate of 0.4 uSv h⁻¹ (0.29 mSv per month) was estimated at a distance of 10 m from a stack of drums of uranium ore tailings containing 319 Bq g⁻¹ with 48 drums in the front face of a stack 8 pallets wide and three tiers tall. Dose rates of this order were observed on contact with some drums on the exterior of stacks, suggesting a low average activity content. Radiation dose rates 50 m distant from the above stack would be about 0.01 mSv per month . By comparison, dose rates measured by TLD's at the site fence west of the Fishermans Bend compound were within observed variations in the background dose rate.

7. <u>Conclusions</u>

7.1 Assuming homogeneity of material contained in each drum, measurements on a small proportion of the drums of radioactive wastes from Fishermans Bend at LHRL indicate' that some drums (possibly one percent of those monitored) could contain a radioactive content in excess of 100 Bq per gram.

7.2 A legal viewpoint is required on whether the meaning of .'radioactive substance' in the NSW act could apply to a group of packages (the Fisherman's wastes as a whole) or to an individual package, i.e., a drum.

7.3 Measurements of gamma radiation dose rate at the perimeter fence around the Fishermans Bend drum stacks is within observed variations in the natural background radiation.

Table 1. Drums containing tailings from ore of the U-238 series.

ISOTOPE	T 1/2		Bq g-1 U-238	1 kg of U-ore (0.3% U3O8) Bq	1 kg tailings (95% U extn) Bq	1 x 200L drum (340 kg of tailings) Bq
			(secular equil.)			
U-238	4.50E+09	у	12321	31344	1567	5.33E+05
Th-234	2.41E+01	у	12321	31344	1567	5.33E+05
Pa-234	1.18E+00	m	12321	31344	1567	5.33E+05
U-234	2.50E+05	У	12321	31344	1567	5.33E+05
Th-230	7.60E+04	y	12321	31344	31344	1.07E+07
Ra-226	1.62E+03	y	12321	31344	31344	1.07E+07
Rn-222	3.83E+00	d	12321	31344	31344	1.07E+07
Po-218	3.05E+00	m	12321	31344	31344	1.07E+07
Pb-214	2.68E+01	m	12321	31344	31344	1.07E+07
Bi-214	1.97E+01	m	12321	31344	31344	1.07E+07
Po-214	2.70E-06	m	12321	31344	31344	1.07E+07
Pb-210	2.20E+01	у	12321	31344	31344	1.07E+07
Bi-210	5.00E+00	d	12321	31344	31344	1.07E+07
Po-210	1.38E+02	d	12321	31344	31344	1.07E+07
Pb-206	stable		0	0		
				Bq kg-l ore B	8q kg-1 tailings	3
TOTAL			172494	438815	319708	1.09E+08

1 x 200 L drum holds 340 kg of ore. Density of ore: 1.7 g cm-3

Specific activity of tailings = 319 Bq g-I

Calculated gamma radiation dose rate = 7 µSv h-1

Dose rate equivalent to 100 Bq.g-1 = 7 x 1001319 = 22 μ Sv h-l

			1	kg of monazite	e 1 x 200 L drum
				ore	monazite ore
ISOTOPE	T 1/2		Bq g-1 Th-232	Bq	Bq
Th 000			(secular equil.)	000700	4 075 00
Th-232	1.91 E+00	Y	4070	339783	1.67E+08
Ra-228	5.75E+00	Y	4070	339783	1.67E+08
Ac-228	6.13E+00	h	4070	339783	1.67E+08
Ra-224	3.66E+00	d	4070	339783	1.67E+08
Rn-220	5.50E+01	S	4070	339783	1.67E+08
Po-216	1.50E-01	S	4070	339783	1.67E+08
Pb-212	1.06E+01	h	4070	339783	1,67E+08
Bi-212	6.06E+01	min	4070	339783	1.67E+08
Po-212 (64%)	3.04E+02	ns	4070	217461	1.07E+08
Tl-208 (36%)	3.05E+00	m	4070	122322	6.02E+07
Pb-208	stable	m			
				Bq kg-l ore	Bq per drum
TOTAL			32560	3.06E+06	1.50E+09
					1
Monazite contair	ns 9.5% Tho	rium, a	and 60% rare eart	h oxides	
Specific activity	of Th-232 =	9 x I	I E+6 g Ci-l	9.00E+06	
			(0.11	u Cig-1)	
			· ·) Bg g-1)	
Sp Gr Th02 = 9.	69; Sp Gr so	oil = 1.7	· ·	,	
Density of mon	•	2.46	g cm-3		
			492 kg of monaz	ite ore	
Specific	c activity of M	lonazite	e ore ≂ 3060 Bq g-	1	
Calcu	lated gamma	radiati	on dose rate $= 80$	µSvh-l	
			∎,		

Table 2. Drums containing Monazite ore (Th-232 series).

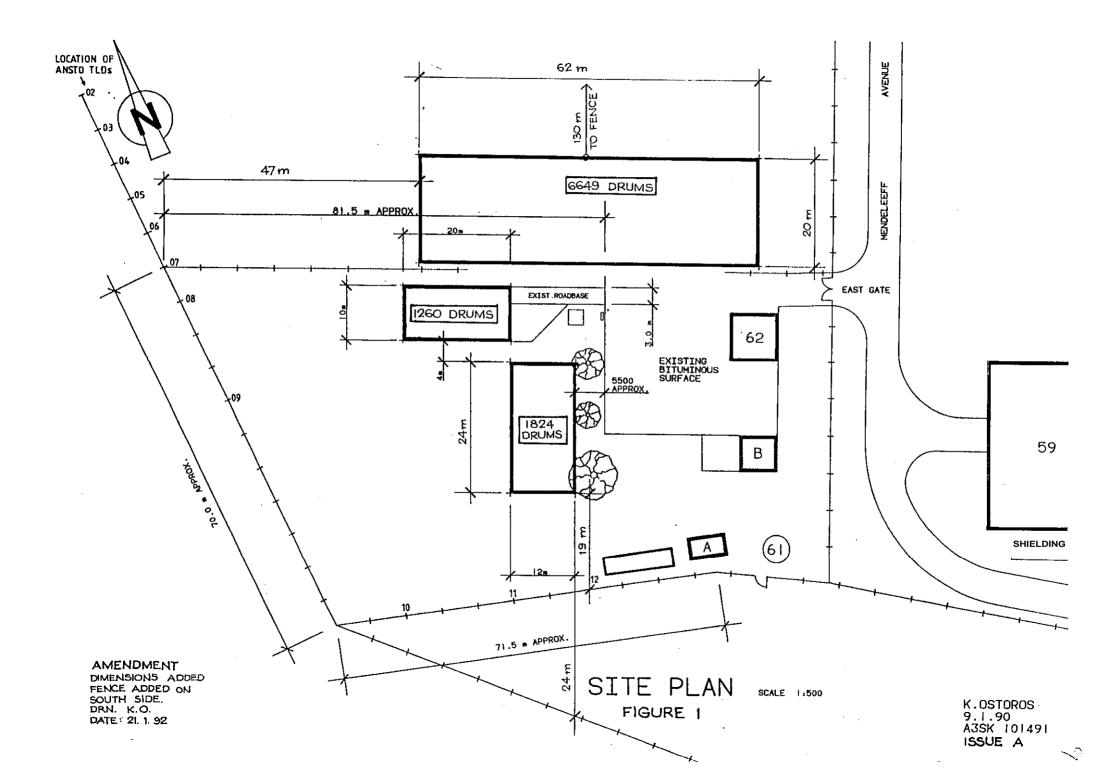
Dose rate equivalent to 100 Bq g-1 = 80 x 100/3060 = 2.6 µSv h-1

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Table.	З.	Radiation	Monitorina	at th	<u>e perimeter</u>	fence of	the
					Bend Drums		

Identification No.	Monthly Gamma Dose mSv month ⁻¹
2	0.185
3	0.096
4	0.163
5	0.089
6	0.089
7	0.096
8	0.111
9	0.037
10	0.103
11	0.163
12	0.140



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ATTACHMENT A

GAMMARAY DOSE FROM DRUMS OF TAILINGS U238 SERIES

Assuming a failing density of 1.7 g cm⁻³ and a disintegration rate of 31344 Bq/Kq of tailings for each of the gamma sources. The important gamma source isotope is Bi-214 with 1.17 gammas per disintegration at energies above 0.60 MeV., the secondary source is Pb-214 with lower gamma energies. A drum is 88 cm high with a radius of 28 cm. The source intensity S is $31.3 \times 1.7 = 53$ disintegrations cm-3s-1..

The estimated dose at 7 cm from the side of one drum is 3.5 μSvh^{-1} . The Shielding Compendium Tables do not give a value on the surface. They give an upper limit at the surface of flux = $S/2\mu$ which gives a dose of 7.2 μSvh^{-1} .

, The dose at 10 m is estimated as 0.39 μ Svh⁻¹ from 69 drums. The'stage 1 stack has 48 drums in the first layer. The total stack area = 12 x 2.92 = 35 m² while the area for 48 drums = 24.5 m². To allow for drums visible behind the first layer the number of stacks is assumed to be 48 x 35/24.5 = 69.

At a high energy of 1.8 MeV the first layer of drums is 4.4 mfp thick and the attenuation is $B(4.4)e^{-4.4} = .07$. The effect of layers beyond the first, is less than a factor of 1.07.

The dose at 10 m is corrected to 0.42 μ Syh⁻¹. This dose is about 4 times a yearly background dose of 1 mSv.

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Brian McGregor Nuclear Technology Program.

GAMMA-RAY DOSE FROM DRUMS-OF TAILINGS U238 SERIES

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E (MeV)	yield	FTD	μ	28µ	G(7cm)	Sx28xFTDxYxG	G(280)	Dose at 10 m
		$\mu Svh^{-1}/\gamma cm^{-2}S^{-1}$	cm-1			π		
0.20	.11	.0045	.204	5.7	.138	.03	.0028	.003
0.25	.17	.005	.189	5.3	.146	.06	.0030	.006
0.30	.19	.0055	.177	5.0	.152	.07	.0032	.008
0.35	.37	.006	.165	4.6	.167	.18	.0036	.020
0.61	.52	.013	.133	3.7	.208	.66	.0049	.083
1.12	.32	.022	.099	2.8	.275	.92	.0056	.100
1.77	.23	.027	.079	2.2	.350	1.03	.0070	.110
2.20	,10	.032	.071	2.0	.377	.57	.0074	.060
								n.389

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GAMMA-RAY DOSE FROM DRUMS OF TAILINGS - THORIUM SERIES

It is assumed that monazite contains 9.5% Thorium, the activity is 4070 Bq g^{-1} of Thorium and the ore density = 2.46.

The source term $S = 340 \text{ Bg g}^{-1}$ of ore = 884 Bq cm⁻³ of ore. An estimate of the surface flux = $\frac{0.8 \times S \times y}{2\mu}$

GAMMA-RAY DOSES

Isotope	E (MeV)	Yield	FTD	μ	Dose (µSvh ⁻¹)
Pb212	0.26	.62	.005	.27	4
- Ac228	0.93	.49	.019	.185	18
T1208	0.58	.46	.013	.195	11
	2.61	.38	.035	.106	44
		••			77
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The dose at the surface of a drum filled with thorium ore is estimated as 77 μ Svh⁻¹

RADON SURVEY OF FISHERMANS BEND DRUM STACKS AT ANSTQ

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Herewith an explanation of attached tables from Stewart W

Lettering of "Blocksⁿ refer to the stacks of drums under tarpaulins as shown in Figure 2 of the Report "Measurement of gamma radiation in the Fishermans Bend waste drum storage compound on 29/11/91" revised 22/1/92. Each block has a contiguous airspace.

Radon concentrations Bq m⁻³inside each block refers to the Rn-222 resulting from the drum emission (and from the concrete base of the block). The numerals 1-4 refer to sampling locations within the block, and a mean value is given.

The Ambient Air concentration refers to measurements in the open air outside the blocks.

The Source concentration data 1-3 refers to concentrations resulting from introduction of a Rn-222 source emitting 740 Bq Rn-222 min⁻¹into block H. Four measurements were made at different locations in block H **.** A source contribution was estimated from the mean of each measurement (e.g., 147 - 32 = 115 Bq Rn-222 min⁻¹ and mixing rate = 740/115 = 6.43 m³ min⁻¹.

The mixing rate was then used to determine the Rn-222 leakage rate from drums in the block (56 x 6.43 = 360 Bq min⁻¹).

Note that Rn-222 daughter WL's measured in the Fishermans Bend compound were practically identical with those measured at the meterological station, i.e., the drum stacks do not contribute a measurable amount to Rn levels in air at LHRL. Rn in the steel store were 50 % greater than those in the open.

Mike Costello. 24/1/92 Radon daughter concentrations expressed as milli-Working Levels measured simulataneously in the centre of the Fishermens Bend waste stacks (FB), at the meteorological tower near B44 (MT) and in the former steel store (SS). The measurement period was 1600 on Friday 17 to 1000 on Wednesday 22 January 1992.

LOCATION	MI	FB	SS
Conditions			
wind speed <1 m s ⁻¹ direction: 0-90 90-180 180-270 270-360	• 4 5 • 35 • 47 • 47	.41 .39 .36 .34	1.67 1.90 1.91 1.20
wind speed >1 m s ⁻¹ direction: 0-90 90-180 180-270 270-360	.37 .36 .41 .64	.38 .34 .40 .66	.81 1.07 1.78 1.53
All data: mean	.47	- 46	1.13
Estimated radon concentra Rq m ⁺ , error on mean	ation, base 6 .02	ed on an eo 6 .02	quilibrium factor of 0.3. 14 0.05

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		BLOCK E	BLOCK F	BLOCK G	BLOCK H	BLOCK I	BLOCK J	BLOCK K	BLOCK M
DATE		16.1.92	16.1.92	17.1.92	17.1.92	18.1.92	20.1.92	21.1.92	22.1.92
ADON	1	92	121	88	45	23	32	226	342
ONC.	2	30	173	47	38	95	34	1112	267
Bg/m3	3	6	56	99	12	51	33	79	34
	4	*							215
l Mea		43	117	78	32	56	33	472	214
mbient.	Airi	2.9	29	4.5	4.5	23	7	22	23
on <u>e agv</u> Souke e	,				<u>Neo</u>	233	42	209	94
Conc.	2				Source	116	137	136	66
Boy/m3	3				made and	4 91	49	166	83
					in Black	4			
Mean	≁]	. =	147	76	170	81
SOURCE CONTRIBU	710-					115	44	138	49
MIKING m3/m	RAT					6.43	16.8	54	15-1
Radon h Lalé Bgi	0.6.	se 469	* 1275	\$ 956 *	34-9*	360	554	2549	3231