Report Rapport

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A FEASIBILITY STUDY - IN VIVO MEASUREMENT OF LEAD 210 IN NEWFOUNDLAND FLUORSPAR MINERS

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A research report prepared for the
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Research report

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A report prepared by M.W. Davis, Monserco Limited
Under contract to the Atomic Energy Control Board

ABSTRACT

A feasibility study was attempted to determine if skeletal burdens of Pb210 in Newfoundland fluorspar miners could be measured by in-vivo techniques using phoswich detectors inside a shadow shield. The detection system comprised two 12.7 cm diameter phoswich detectors with 3 mm thick front crystals of NaI (Tl activated) and 5 cm thick back crystals of CsI (Tl activated). Calibration of the system was carried out using a head phantom impregnated with Pb210 and a minimum detection limit of 0.20 nCi in the skull was calculated. Pb210 burdens in the skull and knee were measured in each of two ex-miners who had received radon daughter exposures estimated at 1766 and 1235 Working Level Months (WLM). The last exposures had been 25 and 19 years ago respectively and the Pb210 burdens had decreased to the point where they were undetectable using this technique. The estimated exposures are not inconsistent with the upper limits of exposure calculated using Eisenbud's model and assuming 0.2 nCi skull burdens. Among thirty other potential candidates for a full scale study most had exposures less than 3500 WLM and based on the limited data obtained from this work, results from a full scale study would have significant statistical uncertainties. Unfortunately during the course of this work a negative attitude developed among the candidates and the research had to be stopped prematurely.

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RÉSUMÉ

Une étude de faisabilité a été tentée afin de déterminer si les charges osseuses de plomb 210 chez les travailleurs des mines de spath fluor de Terre-Neuve pouvaient être mesurées par des techniques in vivo au moyen de scintillateurs-sandwiches à l'intérieur d'un écran partiel. Le système de détection se composait de deux scintillateurs-sandwiches de 12,7 cm de diamètre renfermant des cristaux avant d'iodure de sodium (Tl activé) d'une épaisseur de 5 cm. L'étalonnage du système a été effectué à l'aide d'une tête fantôme imprégnée de plomb 210; on a calculé une limite minimale de détection de 0,20 nCi dans le crâne. Des charges de plomb 210 au crâne et au genou ont été mesurées chez deux anciens mineurs qui ont reçu des doses de produits de filiation du radon évaluées à 1766 et 1235 unités alpha-mois (WLM). Les dernières radioexpositions remontaient à 25 et 19 ans respectivement et les charges de plomb 210 avaient diminué au point où cette technique ne permettait plus de les déceler. Les expositions estimatives ne sont pas incompatibles avec les limites maximales d'exposition qui ont été calculées grâce au modèle d'Eisenbud et en supposant une charge crânienne de 0,2 nCi. Parmi 30 autres candidats possibles pour une étude exhaustive, la plupart avaient subi des expositions inférieures à 3500 WLM et, d'après les données limitées tirées de la présente étude, les résultats d'une étude exhaustive comporteraient d'importantes incertitudes statistiques. Malheureusement, les candidats ont commencé à avoir une attitude négative durant l'étude et il a fallu interrompre prématurément la recherche.

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1.0 INTRODUCTION AND SUMMARY

This report describes the results of a study to determine the feasibility of measuring Pb210 in ex-fluorspar miners in St Lawrence, Newfoundland. The fluorspar miners are unique in so far as, in the course of their work, they have been exposed solely to short-lived radon daughters without simultaneous exposure to long-lived radioactive dust. If it is present in the body in sufficient quantity Pb210, a radon daughter product, may be detected using in-vivo counting techniques. Furthermore, knowing the years over which the miners incurred their exposures and the rate at which Pb210 is eliminated from the body, the measured burdens can be used to estimate the magnitude of the exposures to radon daughters and the resulting lung doses.

The objective of the study was to determine the feasibility of measuring Pb210 in the skull and knee bones of the miners using two phoswich detectors and a mobile shadow shield. Where possible the Pb210 burdens were to be related to radon daughter exposure estimates.

Using radon daughter exposure estimates obtained from Health and Welfare Canada, 30 potential candidates were selected for a future full scale study and five were selected for feasibility measurements to be made in Mississauga. During a meeting with potential candidates in St. Lawrence it was apparent that only two were willing to travel to Mississauga. However, all but one were willing to participate if the measurements took place in St. Lawrence.

In order to measure a total of five subjects to complete the feasibility study, the mobile counting equipment was transported to Newfoundland. However, between the time of the candidates meeting and the arrival of the equipment in St. Lawrence, almost all had turned against participation in the project. Only two ex-miners agreed to be measured in addition to three controls.

The in vivo measurements of Pb210 were made using a shadow shield and two phoswich detectors. The shadow shield comprised a steel lined chamber with up to 7.5 cm of lead in each wall to reduce background radiation. The phoswich detectors were 12.7 cm in diameter with thin front crystals of sodium iodide (NaI) and 5 cm thick back crystals of cesium iodide (CsI). The system was calibrated by using a head phantom painted with Pb210 on loan from the Radiation Protection Bureau. Throughout the series of measurements on the ex-miners and controls, fluctuations in background were monitored by repeated measurements of a water phantom.

In the gamma spectra of the ex-miners' skulls and knees no 47 keV photopeak from Pb210 could be identified by visual inspection. This was verified by the statistical analysis of each spectrum. Also, using the shape of the spectra from control subjects, it was possible to show that the 47 keV region of the spectra from ex-miners was not different than gamma spectra from control subjects with respect to Pb210 burdens.

Estimates of the minimum detection limits for Pb210 in the skull were based on the measurements of the head phantom. It was concluded that the two ex-miners (estimated exposures of 1776 and 1235 Working Level Months (WLM), ending 25 and 19 years ago respectively) have bone burdens of <0.20 nCi Pb210.

In view of the negative attitude that had developed on the part of the remaining ex-miners , it was decided that the study be carried no further.

2.0 BACKGROUND

In 1933 fluorspar mines were opened up in St Lawrence, Newfoundland and employed local men. Operations expanded and continued through the 1950s when it was observed that an apparently high incidence of lung cancer affected only the male population. In 1956 at the request of the Newfoundland Department of Health, the Occupational Health Division of the Department of National Health and Welfare began studying the extent of the dust hazard in the mine. In 1958 the scope of the project was expanded to include radiation measurements, epidemiological and clinical investigations. The outstanding environmental finding in the fluorspar mines was the discovery of concentrations of radon and daughter products in the air well in excess of suggested maximum permissible concentrations (Ref. 1) The origin of the radon gas was determined to be water flowing into the mine from adjacent rock formations containing radium and uranium. The fluorspar mines themselves had no unusual levels of radon precursors.

In the early 1960s improved ventilation and flood control systems were installed in the mines and the airborne concentrations of radon and daughter products were significantly reduced below the maximum allowed levels. Consequently, radon daughter exposures to the miners were greatly reduced from this period to the close of the mines in the late 1970s. The exposure of the miners to radon daughters resulted in the buildup of a radioisotope of lead (Pb210) in their bodies. Radon daughters in the lungs decay to Pb210 (see Figure 1) which is transported by the blood to the bone. Pb210 has a radiological half-life of 21 years and its effective half-life in bone has been variously reported as 9.2 years by Eisenbud (Ref 3), 15 years by Holtzman (Ref 4) and 3.6 years by Blanchard (Ref 5).

The radioactive decay of Pb210 is accompanied by the emission of a 47 keV gamma ray which can penetrate thin layers of tissue and be detected by scintillation counters outside the body. Because there is a minimum of tissue over the skull bone, and head size is one of the least variable of all body dimensions, the skull is the region of choice for in vivo measurement of Pb210.

3.0 MEASUREMENT METHOD

The in vivo measurements of Pb210 in the ex-miners were made by detecting the 47 keV gamma rays using the Compton suppression characteristics of phoswich detectors shielded from natural background radiation by a shadow shield.

The phoswich detectors were 12.7 cm in diameter with 3 mm thick front crystals of NaI (Tl activated) and 5 cm thick back crystals of CsI (Tl activated). By using phoswich detectors rather than single scintillator detectors the signal to noise ratio can be improved by a factor of four to six. Compton scattering interactions were distinguished from true low energy events in the front crystal by electronically rejecting events that occurred in both crystals. The faces of the two detectors were positioned perpendicular to one another and located in contact with the head or knee of the subject as shown in Figure 2.

The shadow shield is a large steel lined chamber with lead walls of various thicknessess up to 7.6 cm. The shield is large enough to accomodate a 2 metre tall person lying at full length. Openings at both ends allow visual and verbal contact with the subject throughout the measurement.

To measure Pb210 in the skull, each subject assumed a prone position in the shadow shield, the detectors were positioned as shown in Figure 2 and a 67 minute measurement was made. For the knee measurement, also 67 minutes duration, the subject lay supine and the right knee joint was located in contact with the detector face.

The height, weight, age, head circumference (taken just above the eyes) and knee circumference were also measured and recorded for each subject.

The efficiency of the system was checked periodically by positioning the head phantom painted with Pb210 in the same location as the subject's head shown in Figure 2. The phantom, on loan from the Radiation Protection Bureau (RPB), is constructed of a styrofoam head mold, the outer surface of which has been coated with Pb210 paint. A layer of plaster was then applied over the outer surface to simulate the thin layer of tissue over a human skull. The RPB phantom has been cross calibrated against two standard skulls prepared by N. Cohen at New York University Medical Center. It was reported that the effective Pb210 activity on the RPB phantom is 64 nCi.

Natural background radiation from radon gas and cosmic sources fluctuates throughout the day. The shadow shield does not shield background completely. Therefore, variations within the shield were monitored by repeated measurements of a water phantom which contained no radioactivity. A plastic bottle of water (circumference 49 cm) was positioned in contact with the face of the detectors in the same location as the subject's head shown in Figure 2 and background radiation scattered from the water phantom was measured.

4.0 RESULTS

Throughout the series of ex-miner and control measurements the efficiency of the two detector phoswich system was measured four times using the head phantom painted with Pb210. The average value of the calibration factor was 0.120 counts per second per nCi. (cps/nCi) and the observed standard deviation among the results was 4.3%. This average was in good agreement with previous calibration measurements of the system.

The energy resolution of the detectors was checked using the 60 keV photopeak from an Am241 source. The full width at half maximum was 7.5 keV, also consistent with past measurements.

As Indicated above gamma spectra were measured for the head and right knee of each of the two ex-miners and three controls. The counting time for each was 4000 seconds. Segments of the gamma spectra are shown in Figures 3a to 3e. On the left are plots of the raw data and on the right are plots of smoothed data obtained by 5 point averaging technique. A 47 keV photopeak is not evident in any of the raw data plots and only a very small peak appears in the smoothed spectrum of the knee of ex-miner 1. As shown below that peak is not statistically significant and is an artifact of the smoothing and/or chance.

The total number of counts in each of two energy intervals 38-55 keV and 79-111 keV and the ratios of these counts are shown in Table 1. For the three controls the average ratio (head) is 0.331 with a standard deviation of 0.017. The average ratio (knee) is 0.314 with a standard deviation of 0.016.

The 38-55 keV interval includes 47 keV gamma rays from any Pb210 present in the head/knee measured. It is expected that when measurable Pb210 is present the 79-111 keV interval should be unaffected. Therefore, the ratio of the two regions would be significantly greater than 0.331 for the head and 0.314 for the knee. The interval ratios have been plotted for controls and the two ex-miners, in Figure 4. There are no statistically significant differences among them.

A second method for subtracting the background from beneath a photopeak in a spectrum was also used. The region of each spectrum in the vicinity of 47 keV was divided into 3 equal energy intervals of 6 keV, 39-45, 45-51 and 51-57 keV. The middle interval includes any 47 keV peak from Pb210 and the upper and lower intervals were used to calculate the background in the middle interval assuming a straight line background model. When the intervals have equal energy intervals the predicted background, B, in the middle interval is calculated by

$$B = (A1 + A3) / 2$$

where A1 = the integral counts in the low energy interval
A3 = the integral counts in the high energy interval

The net counts, N, in any 47 keV peak are given by

$$N = A2 - B$$

where A2 = the integral counts in the middle energy interval

The standard deviation for N is given by

$$\sigma_N = \{ \sigma_2^2 + (\sigma_1^2 + \sigma_3^2) / 4]^{1/2}$$

where $\sigma_i = (A_i)^{1/2}$

The net counts in each A2 interval were calculated using this method and the results are shown in Table 3. In all cases the net counts are less than two standard deviations of uncertainty and therefore not significantly above the background at the 95 % confidence level.

The minimum detection limit when defined as three standard deviations above background is calculated to be

$$(3 \times 32) / (0.12 \times 4000) = 0.20 \text{ nCi}$$

where the standard deviation of the background in the A2 interval is 32 counts and 0.12 cps/nCi was the calibration factor measured using the Pb210 head phantom.

Upper limits of the WLM exposures of the two ex-miners were made using equation 3 from Eisenbud (Ref 3) and making the following assumptions:

- the actual skull burdens were at the minimum detection limit of 0.2 nCi

- the radon daughter exposure rates were constant throughout the exposure period

- the deposition to the skeleton was 8.4 pCi Pb210/WLM.

- effective half life of Pb210 is 9.2 years.

The upper limits calculated by the above method were 1573 and 1207 WLM for miners M1 and M2 respectively.

5.0 DISCUSSION

The calibration factor for the phoswich detector system was stable throughout the series of measurements with a standard deviation of 4.3 % and it was in good agreement with previous calibrations.

There were no obvious Pb210 photopeaks in the gamma spectra of the skulls and knees of the controls and ex-miners. When the shapes of the spectra from controls were compared to the spectra from two ex-miners - there were no statistically significant differences. The 47 keV region of the spectra taken from ex-miners was tested using a straight line background model and adjacent energy intervals. The 47 keV region was not statistically different than the background and the ex-miners skull burdens were calculated to be <0.20 nCi Pb210.

The estimated exposures of the two ex-miners were 1766 and 1235 WLM based upon measurements of airborne radon daughter concentrations and their last exposures were 25 and 19 years ago respectively. Based upon their work history and assuming that their skull burdens were 0.2 nCi, the minimum detectable using the technique discussed in section 4, the upper limits of their exposures were calculated to be

1573 and 1207 WLM using the Eisenbud model. These values are consistent with the exposures and uncertainties estimated from measurements of airborne radon daughter concentrations. The remaining potential candidates have exposures that are generally less than twice 1766 WLM and all are less than three times. Based on the limited data here, it appears that the results of Pb210 measurements on the other candidates would have significant statistical uncertainties attached.

6.0 REFERENCES

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3. In Vivo Measurement of Pb210 as an Indicator of Cumulative Radon Daughter Exposure in Uranium Miners, Eisenbud, M et al, Health Physics 1969, Vol 16, p637

4. Sources of Pb210 in Uranium Mines, Holtzman, R.B. Health Physics 1970, Vol 18, 105

5. Blood and Skeletal Levels of Pb210-Po210 As a Measure of Exposure to Inhaled Radon Daughter Products, Blanchard, R.L., Archer, V.E., Saccomanno, G. Health Physics 1969, Vol 16, p585

FIGURE 1

RADON 222 AND RADON DAUGHTERS

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{226}_{88}Ra$	Radium	1602y	4.60 (6%) 4.78 (95%)	---	0.186 (4%)
$^{222}_{86}Rn$	Emanation Radon (Rn)	3.823d	5.49 (100%)	---	0.510 (0.07%)
$^{218}_{84}Po$ 99.98% 0.02%	Radium A	3.05m	6.00 (~100%)	0.33 (~0.019%)	---
$^{214}_{82}Pb$	Radium B	26.8m	---	0.65 (50%) 0.71 (40%) 0.98 (6%)	0.295 (19%) 0.352 (36%)
$^{218}_{85}At$	Astatine	~2s	6.65 (6%) 6.70 (94%)	? (~0.1%)	---
$^{214}_{83}Bi$ 99.98% 0.02%	Radium C	19.7m	5.45 (0.012%) 5.51 (0.008%)	1.0 (23%) 1.51 (40%) 3.26 (19%)	0.609 (47%) 1.120 (17%) 1.764 (17%)
$^{214}_{84}Po$	Radium C'	164μs	7.69 (100%)	---	0.799 (0.014%)
$^{210}_{81}Tl$	Radium C"	1.3m	---	1.3 (25%) 1.9 (56%) 2.3 (19%)	0.296 (80%) 0.795 (100%) 1.31 (21%)
$^{210}_{82}Pb$	Radium D	21y	3.72 (.000002%)	0.016 (85%) 0.061 (15%)	0.047 (4%)
$^{210}_{83}Bi$ ~100% .00013%	Radium E	5.01d	4.65 (.00007%) 4.69 (.00005%)	1.161 (~100%)	---
$^{210}_{84}Po$	Radium F	138.4d	5.305 (100%)	---	0.803 (0.0011%)
$^{206}_{81}Tl$	Radium E"	4.19m	---	1.571 (100%)	---
$^{206}_{82}Pb$	Radium G	Stable	---	---	---

FIGURE 2

PHOSWICH DETECTOR POSITION FOR SKULL MEASUREMENT

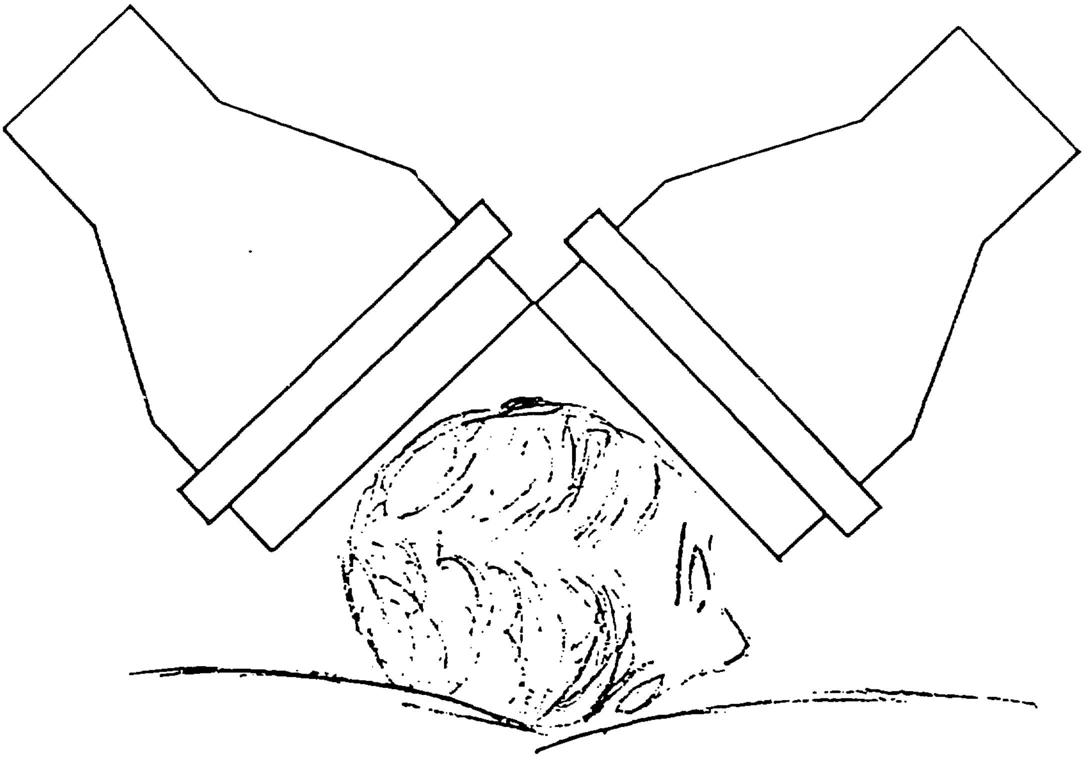
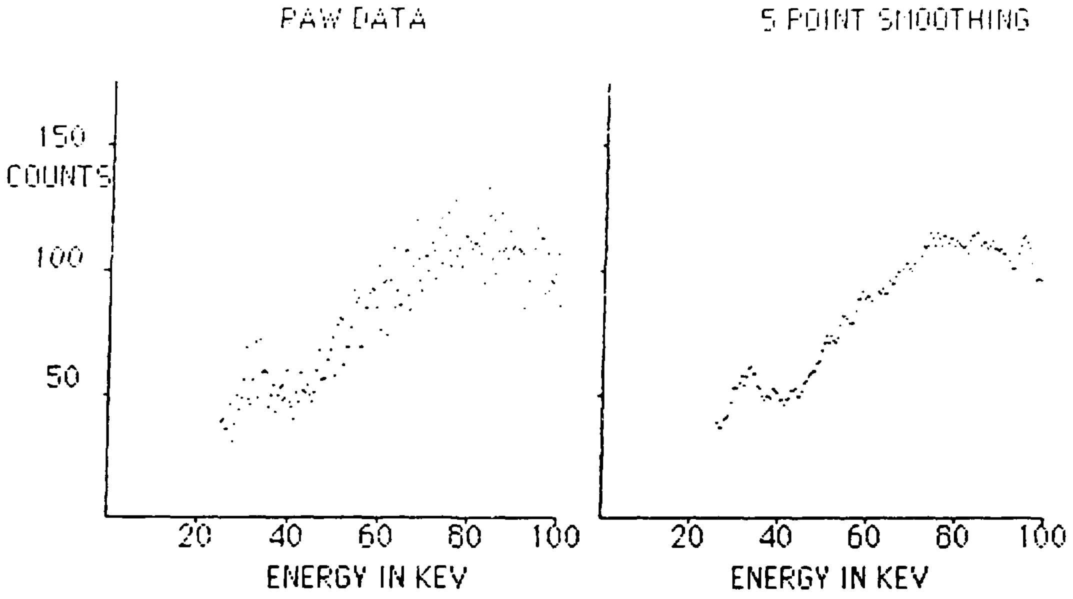


FIGURE 3A

PHOSWICH DETECTOR MEASUREMENT OF CONTROL 1 - HEAD



PHOSWICH DETECTOR MEASUREMENT OF CONTROL 1 - KNEE

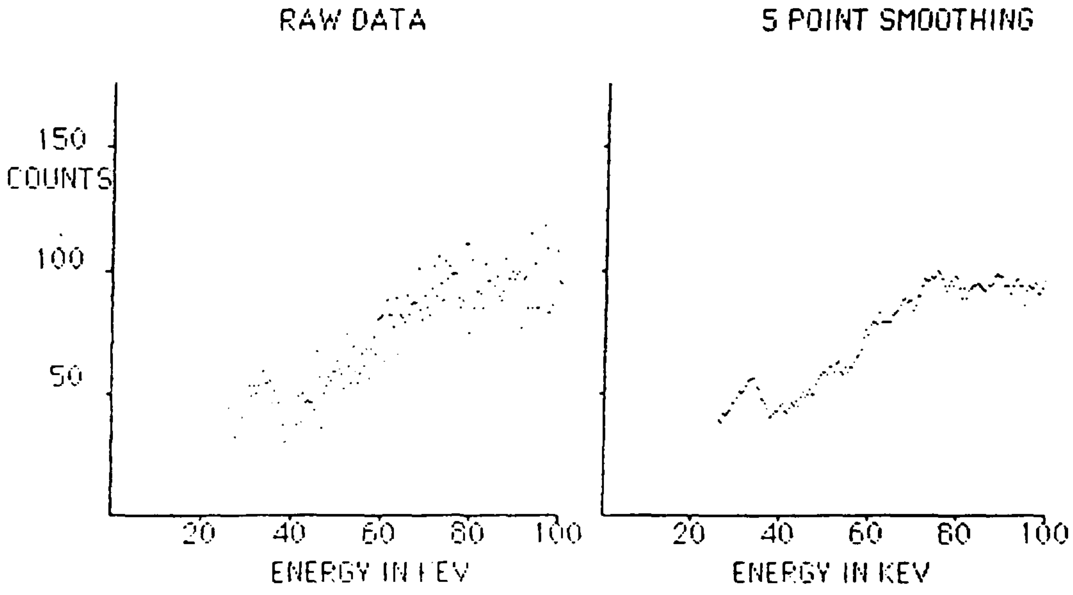
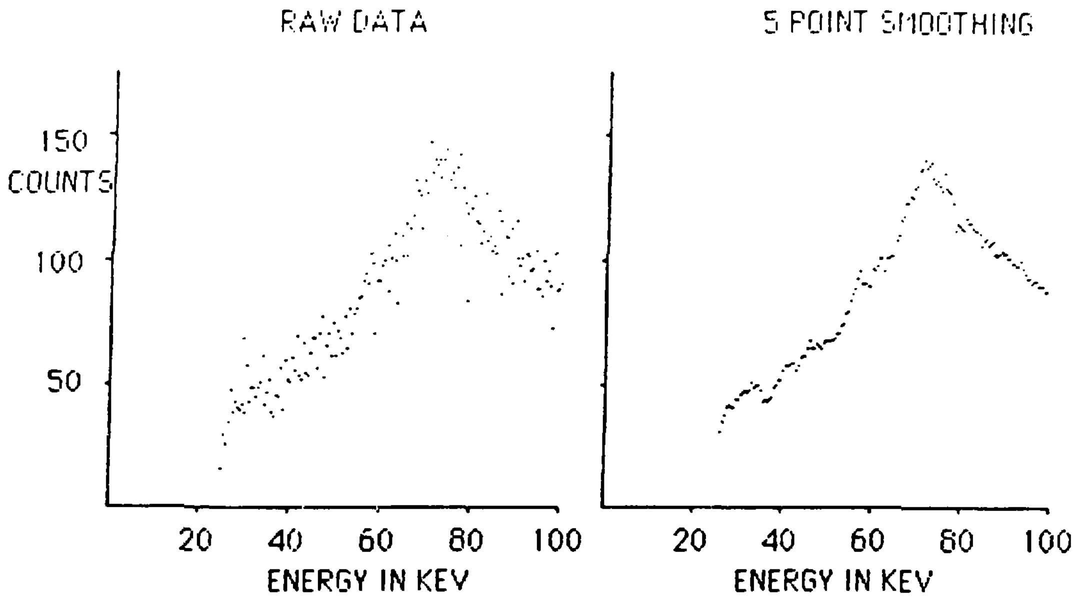


FIGURE 3B

PHOSWICH DETECTOR MEASUREMENT OF CONTROL 2 - HEAD



PHOSWICH DETECTOR MEASUREMENT OF CONTROL 2 - KNEE

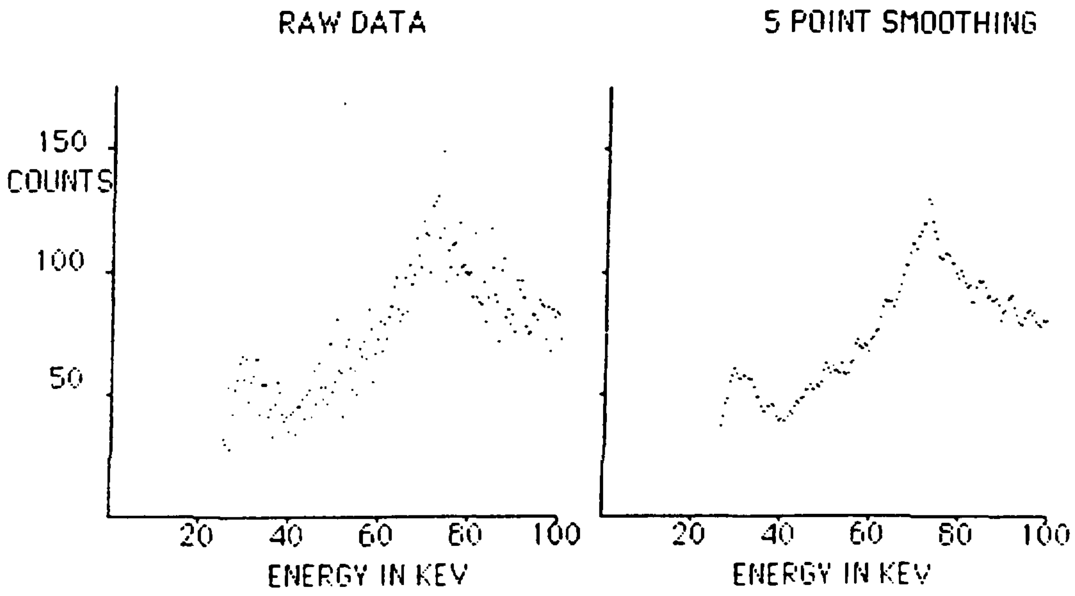


FIGURE 3c

PHOSWICH DETECTOR MEASUREMENT OF CONTROL 3 - HEAD

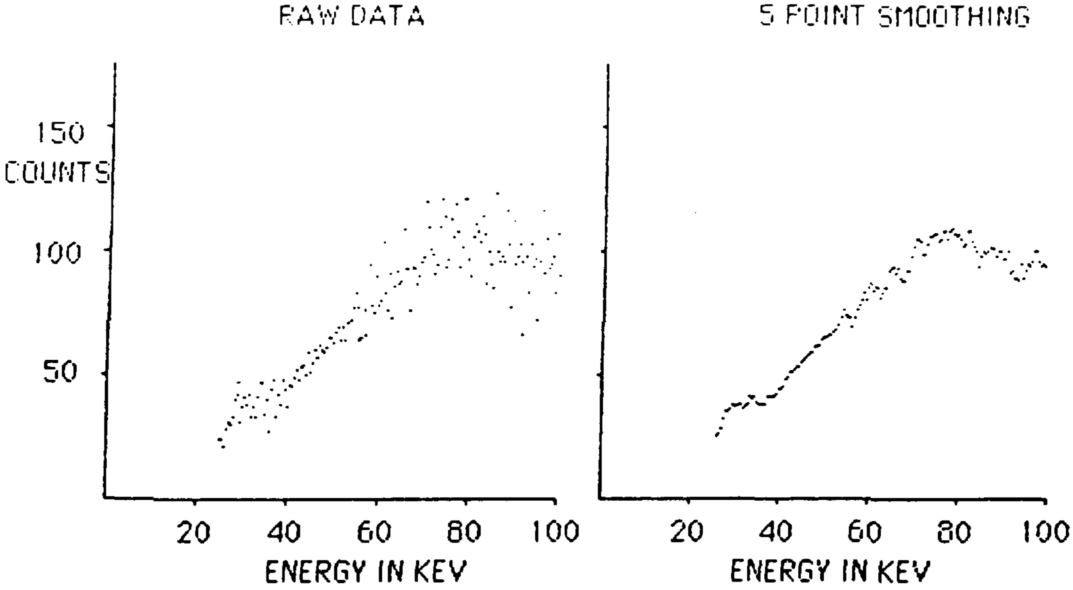
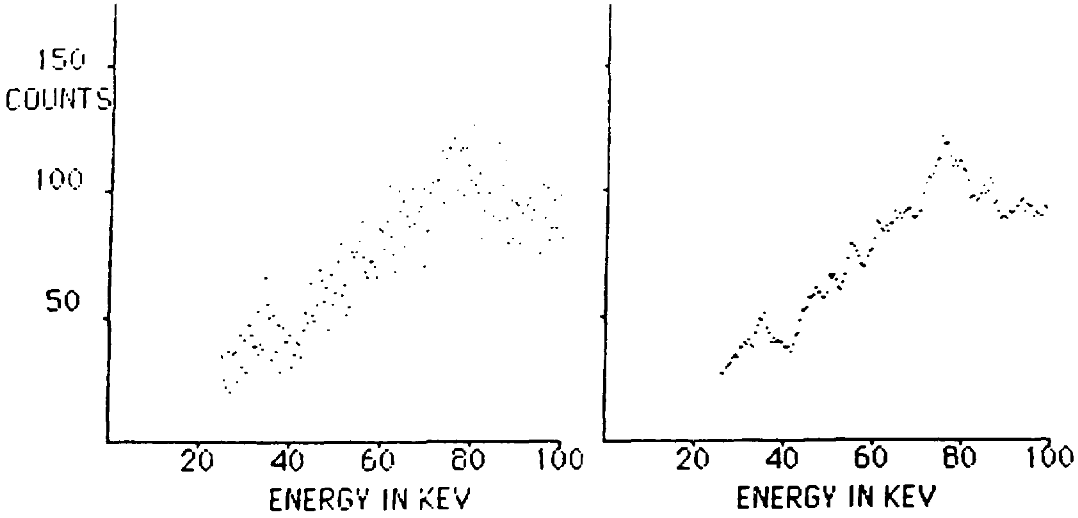


FIGURE 3D

PHOSWICH DETECTOR MEASUREMENT OF MINER 1 - HEAD

RAW DATA

5 POINT SMOOTHING



PHOSWICH DETECTOR MEASUREMENT OF MINER 1 - KNEE

RAW DATA

5 POINT SMOOTHING

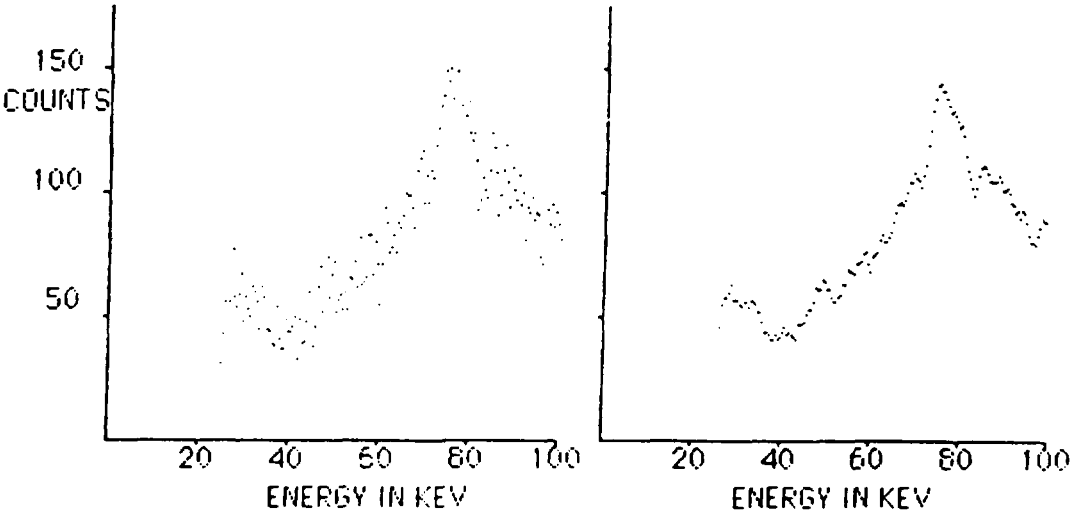
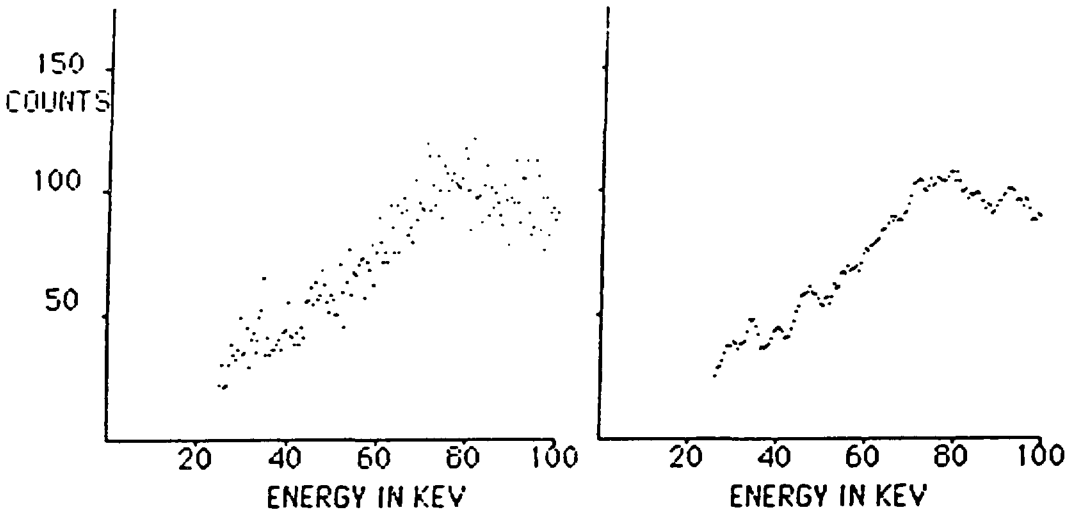


FIGURE 3E

PHOSWICH DETECTOR MEASUREMENT OF MINER 2 - HEAD

RAW DATA

5 POINT SMOOTHING



PHOSWICH DETECTOR MEASUREMENT OF MINER 2 - KNEE

RAW DATA

5 POINT SMOOTHING

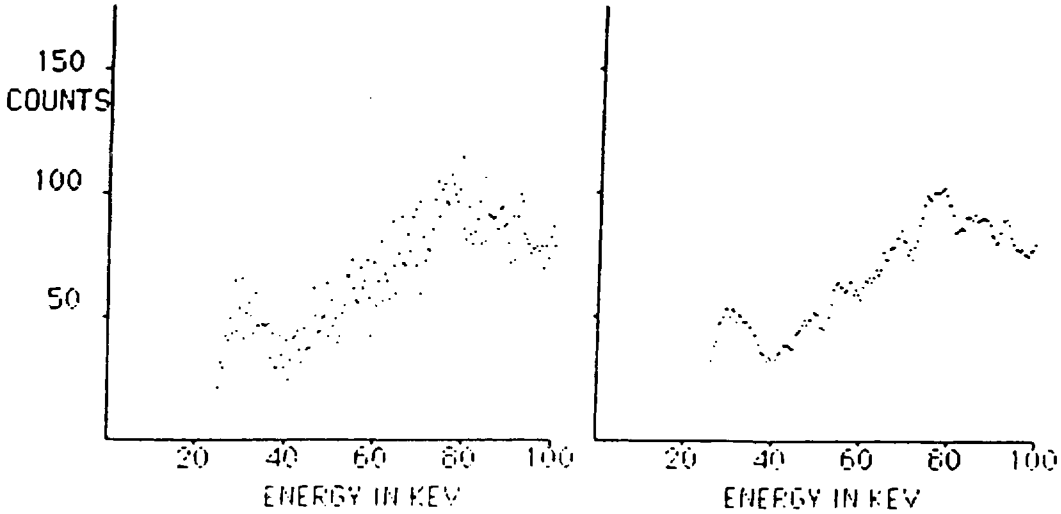
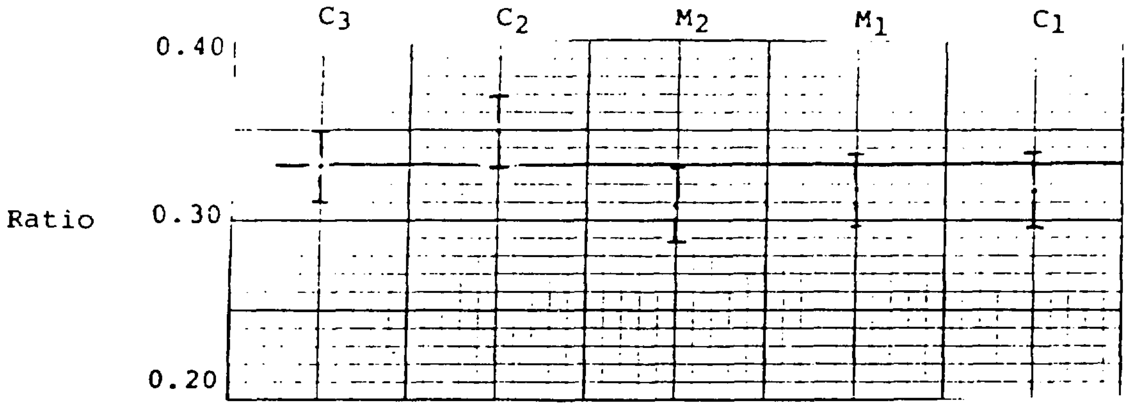


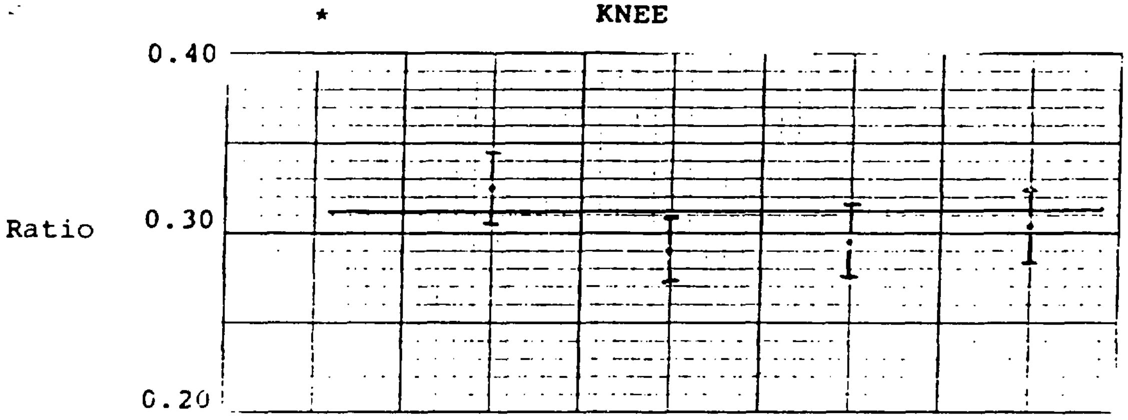
FIGURE 4

RATIOS OF INTEGRALS

OF 38 - 55 kev INTERVAL/79-111 kev INTERVAL



HEAD



KNEE

* no knee data for C3

TABLE 1

STATISTICS ON SUBJECTS

SUBJECT	HEIGHT cm	WEIGHT kg	AGE YR.	WLM	EXPOSURE YEARS	HEAD CIRCUM cm	HEAD COUNTS 38-55keV	KNEE CIRCUM cm	KNEE COUNTS 38-55 keV
M1	170	71	56	1766	'48-'60	58.2	1748(±84)	35	1696(±82)
M2	159	71	55	1235	'50-'68	57.5	1698(±82)	35	1449(±76)
C1	170	82	36	0	'72-'73	59.5	1939(±88)	40	1651(±81)
C2	182	69	50	258	'53-'59	57.5	2057(±91)	36	1662(±81)
C3	168	78	39	0	-	59.5	1864(±86)	38	-

TABLE 2

INTEGRALS OF GAMMA SPECTRA

		ENERGY INTERVALS			
SUBJECT		38-55 keV	79-111 keV	RATIO (± 1σ)	
C1	HEAD	1939	6184	0.314	±0.009
	KNEE	1651	5442	0.303	±0.009
C2	HEAD	2057	5918	0.348	±0.009
	KNEE	1662	5122	0.325	±0.009
C3	HEAD	1864	5652	0.330	±0.009
	KNEE	-	-		
M1	HEAD	1748	5524	0.316	±0.009
	KNEE	1696	5770	0.294	±0.009
M2	HEAD	1698	5521	0.308	±0.009
	KNEE	1449	5002	0.290	±0.009
CONTROL AVERAGE	HEAD			0.331	(±0.017)
	KNEE			0.314	(±0.016)

C CONTROL

M EX-MINER

TABLE 3

INTEGRALS OF GAMMA SPECTRA

SUBJECT	A_1	A_2	A_3	NET 47 keV AREA	STANDARD DEVIATION
C_1 HEAD	605	737	929	-30	33
KNEE	547	659	734	19	31
C_2 HEAD	695	805	945	-15	35
KNEE	516	687	738	60	32
C_3 HEAD	585	725	851	7	33
KNEE					
M_1 HEAD	510	706	842	30	32
KNEE	539	696	756	49	32
M_2 HEAD	541	692	769	37	32
KNEE	434	578	680	21	29