Natural Alpha-radioactivity in Bone, Liver and Blood Samples from the Various Species

by

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Abstract

In this thesis samples from the various species are studied for alpha-radioactive content. The experimental technique used is one of thick source "total" alpha counting with "fast pair"scaling enabling separation of "Thorium" series contribution. A study of the change in count rate over a time period enables Po-210 activity to be determined. Additional alpha spectroscopic techniques are used to identify specific alpha emitting isotopes in the samples.

Bone tissue alpha-activity levels are used as a basis for comparing levels between the species. Liver is studied as it shows interesting features, and blood is also studied. The method of sample preparation involves the removal of water from the samples at low temperatures and no additional chemical techniques are used.

Thirty six different species from four groups of animals were studied (herbivores, carnivores, omnivores and marine environment). Man shows very low levels of alpha-activity compared with other species of which the group "herbivores" show highest levels of alpha-activity, including large amounts of Po-210. The marine environment species show a large accumulation of Po-210 in liver.

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Chapter 1

NATURAL ALPHA-ACTIVITY IN BIOLOGICAL TISSUES

Much attention has been paid in recent years to the levels of natural radioactivity in human beings, and many measurements have been made on these levels in a variety of human tissues. The natural radioactive levels in other species have not received the same attention, as is reflected in a statement by W.V. Mayneord in the Rock Carling Fellowship monograph of 1964(1):

"... An almost untouched field of research lies in the study of tissues in various animal species. We have measured a few and find that ox bone may be 20 or 30 times more active than human bones, and so may certain sheep. Though we have few data, man seems to be among the less radioactive species. ..."

Mayneord's own group (Mayneord et al.(2) provided some data on this topic, but little additional data on the natural radioactive levels in animals has been forthcoming since then. The purpose of the present work is to compare the levels of activity in the tissues of various species and to establish man's position in an ordering of species according to radioactive levels. Specifically the alpha radioactive levels are studied as these along with K-40

are the important activity contributors in biological material {e.g. see Spiers(3) (Table 8.7)}.

As an introduction to this work the techniques and results of other workers are discussed briefly.

1.1 Techniques

The earliest measurements were made in order to estimate radium burden. Cremation ashes were used, the ashes being dissolved in concentrated acids and the Ra-226 gaseous daughter, the alpha emitter, radon, was driven into a counting chamber. This method was reported, for example, by Rajewsky(4) , in 1941. The technique was later used for measurements on individual tissues, the tissue being dry ashed at 600°C, and, more recently, wet ashed in concentrated acids at 250°C. This "radon emanation" technique has proved highly sensitive and still remains the most popular way of estimating Ra-226 in biological samples. A much-quoted refinement of the technique is that of Lucas(5). In this the radon is counted by an alpha-scintillation method, and the system is sensitive to 0.01 pc of Ra-226. The final accuracy is limited by counting statistics.

Ra-226 body burdens may also be estimated by measuring

the excretion of the gaseous radon from the body. This is done by measuring the radon content in exhaled breath per unit time and assuming a suitable value for the radon excretion fraction (typically 0.7). The method is reported by Stehney et al.(6) who further note in their paper that the radon exhaled is frequently less than atmospheric radon and hence the subject must initially breathe radon-free air. This method has an accuracy which is limited by the lack of accurate knowledge of the excretion fraction. The method cannot be used for estimating Th-228 from excreted thoron; thoron decays principally in the body and at most only 1% escapes in the breath {Ward and Jensen(7)}.

Since the late 1950's it has become apparent that other alpha emitters make an important contribution to the total activity in the body. In particular it was recognised that there was a contribution from the "natural fallout" material Pb-210 and/or its alpha emitting daughter Po-210. Chemical separation techniques with electro-deposition of the Po-210 onto metal discs are reported by Black(8) and Holtzman(9) using silver discs, Inouye et al.(10) using nickel discs and Krebs and Whipple(11) using copper discs.

The measurement of Th-228 is rather more complicated.

Radford et al.(12) chemically separated the radium content of their samples (containing Ra-226, Ra-224 and Ra-223). In this procedure Ra-224 (and hence Th-228) is determined from the short half-life alpha decay of its daughter Po-210. An alternative method of determining Ra-224 in the radium complex involves the differential decay or growth of the alpha activity and is reported by Stehney(13) and Goldin(14).

A technique by which the "total alpha activity" of the sample may be measured directly is that developed by Turner et al.(15). In this the sample is ashed and the product alpha counted, using a scintillation method, with a "fast-pair" technique enabling separation of the "Thorium" and "Uranium" series contributions. Counting is usually effected immediately after ashing and at the ashing temperature ($\sim 600^{\circ}C$) Po-210 is driven off and hence the contribution from "natural fallout" is not measured. Hill(16) showed that the Po-210 contribution could be calculated by noting the subsequent growth of alpha-activity with time {Po-210 growing in from parent Pb-210}. The development of large area low background ion chambers {Hill(17) , Osborne(18)} has further extended the ability to detect specific alpha-emitting nuclides.

Autoradiographic methods have also been used to detect "natural activity" in tissues but this technique has not found wide application due to the long exposure times required for the photographic emulsions.

1.2 Results

(a) Humans

There exists a large mass of data on the levels of alpha-radioactivity in the human body, though most of the data deal with the Ra-226 content of bone ash. Several compilations of the data have been made, notably by Walton et al.(19) , and the UNSCEAR report of 1962(20) and two recent texts which provide relevant tabulations and discussions are those of Spiers(3) and Bowen(21). In view of these compilations a comprehensive table will not be presented here and discussion will be confined to a summary of the data.

Total body burdens are usually estimated by measuring the activity in ashed samples and then assuming a suitable value for the ash content of the particular tissue in a "standard man". In this manner the UNSCEAR report propose 75 pc as the mean "total body" Ra-226 burden and Walton

et al. (who calculate a "world wide" bone ash average of 0.013 pc of Ra-226/gram bone ash) suggest that the average "total body" Ra-226 burden for a large fraction of the worlds population may be less than 100 pc. Similarly the Ra-228 average has been estimated, and though the data here is meagre the UNSCEAR report proposes a "total body" burden of 50 pc. Alpha-activity contribution in the "in vivo" state for the whole "Uranium" and "Thorium" series (below the two radium isotopes) may be calculated by assuming secular equilibrium. A correction must however be applied in the case of the "Uranium" series where much of the gaseous Ra-226 daughter, radon, is excreted from the body. No evidence has yet been found to suggest that the alpha-emitting series members above Ra-226 and Th-228 are present in any appreciable quantities {See, e.g. Spiers(3)}.

There is an additional contribution to the total body burden due to "natural fallout" and the UNSCEAR report proposes 200 pc as the total body Pb-210 content. It is as yet uncertain whether or not the alpha-emitting Po-210 is in equilibrium with its precursor Pb-210 (a beta emitter). Recent work by Groos et al.(22) and Stahlhofen(23) indicates that there may be Pb-210 , Po-210 equilibrium though work by Osborne(24) suggests that Pb-210 may be in excess of Po-210 {the above data have been published

subsequent to the UNSCEAR report} .

It is recognised that an accurate representative world wide average for body burdens is impossible since there will be real variations due to geographical distribution, feeding habits, individual metabolic patterns, etc. However, apart from areas of known high radioactivity and certain individuals whose activity is high due to some external factor i.e. radium dial painters, the values of Walton et al.(19) and the UNSCEAR report seem to be generally representative and the work of more recent authors (post 1962) supports this i.e. Hursh and Lovaas(25) ; Rajewsky and Stahlhofen(26) ; Chabra(27) Hallden et al.(28) ; Stahlhofen(29) ; Hunt et al.(30). Notable exceptions occur in data by Mayneord et al. {who calculate an Ra-226 value of 0.06 pc/gram bone ash from their "total alpha" counting method, cf. "world wide" average for Ra-226 of 0.013 pc/gram bone ash by Walton et al. } and Holtzman(31) who finds an average of 0.039 pc/gram bone ash for Ra-226 in studies an 128 individuals from a "normal" area.

Workers often use a single portion of a tissue for measurement and assume that the activity per gram ash in this is representative of the tissue as a whole. The validity of this is important especially in the case of

bone which is widely distributed throughout the body. Work by Walton et al.(19) suggests that this assumption may be correct (range for Ra-226 in bone ±15%) and Holtzman(31) confirms this for Ra-226 and Pb-210 (the latter, however, with somewhat less assurance as rib bone in general showed a high Pb-210 content). In contrast work by Hunt et al.(30) suggests that there may be significant variations in both the Ra-226 and Pb-210 content if "small portions" of bone are used. They report significantly different activities per gram ash for the two types of bone (spongy and compact) that make up the iliac crest.

The results of the work done in this thesis are presented in the form of pc/kgram wet tissue (note that this is not equivalent to the "in vivo" state as no correction has been made for radon excretion) as it is felt that this is the ultimately important quantity in measurements of "natural" activity in biological materials. For bone, we may make a comparison by accepting the UNSCEAR report proposed averages and by making the following assumptions:

(i) That ~80% of the activity lies in the bone. This is suggested by Mayneord et al.(2) and is accepted by Spiers(3) as being correct.

(ii) That the bone ash content of a "standard man" is 2500 grams.

(iii) That the average concentration factor from wet bone to bone ash is 2.8. This value is somewhat arbitrary: Stahlhofen(29) gives values ranging from 1.8 - 3.4 with a mean of 2.6 and Mayneord et al.(2) give a mean of 3.0. (A limited amount of data by Turner et al.(15) gives a range in concentration factors from 4 to 25).

On this basis, and, allowing for that fraction of Po-210 which is in equilibrium with Ra-226, a calculation of the "total alpha'activity in bone, using the UNSCEAR data, gives 68 pc/kgram wet bone. Measurements by Mayneord et al.(2) however, using a "total alpha" counting method technique similar to that used in this thesis give a mean value of 107 pc/kgram wet bone, and this value does not include the Po-210 contribution since their ashing technique (at 600°C) drives off polonium. Calculating a Po-210 contribution on the basis of the Ra-226/Pb-210 ratio from the UNSCEAR report, and allowing for that Po-210 that is in equilibrium with Ra-226 the "total activity" of Mayneord et al. increases to ~140 pc/kgram wet tissue. (Pb-210 , Po-210 equilibrium assumed) It must be noted that the "total alpha" counting technique has not yet been widely used and so the UNSCEAR data may be biased towards the emanation and chemical separation techniques, though

data by Stahlhofen(29) , using the "total alpha" counting technique shows general agreement for Ra-226 values with the "world wide" average of Walton et al.

Turning from bone to soft tissue data it is noted here that most of the data in this thesis concerns liver and blood (as well as bone). The major chemical breakdown of materials in the body occurs in the liver, which is also a "storehouse" for toxic materials that the body cannot excrete. Preliminary data obtained at the beginning of the experimental work here indicated liver as being an interesting tissue and it was chosen as a tissue for study on the basis of the above and not as being representative of soft tissues in general. There is little available data in the work of other authors, or liver as a specific tissue. Ra-226 levels in liver are given by Muth et al.(32) as 0.0034 pc/gram wet tissue and by Hursh and Lovaas(25) as 0.0002 pc/gram wet tissue. The Po-210 content has been measured by Hill(33) giving 0.015 pc/gram wet tissue; Osborne(24) , 0.010 pc/gram wet tissue; Stahlhofen(29) , 0.013 pc/gram wet tissue and Holtzman(31) 0.011 pc/gram wet tissue. Their results show a good degree of uniformity in Po-210 levels in the liver. Mayneord et al(2) find a "total alpha" activity of 0.33 pc/gram ash for liver, and, in the marked absence of other

data this appears to be the most suitable source of comparison with results in this work. A concentration factor of 66.7 for soft tissue ash is given by Mayneord et al. and this gives 0.005 pc/gram wet tissue for their "total alpha" activity in liver. Again, due to their technique, this does not include any contribution from Po-210. It is notable that this "total alpha-activity" value is a factor of 2 less than the average Po-210 levels as given by the above-mentioned authors (Hill etc.). Including this average Po-210 level in Mayneord et al.'s "total alpha-activity" gives a value of 0.017 pc/gram wet tissue for the "total alpha-activity" in liver.

The work of authors e.g. Hill(33) , Mayneord et al.(2) Holtzman(31) and Stahlhofen(29) indicates that activities in other soft tissues may be of a comparable level to those in liver. The "total alpha-activity" of blood has been measured by Hasson(34) who finds a value of 0.089 pc/gram whole blood. This is rather higher than that generally found in soft tissues and this value has not yet been confirmed (the experimental work for this was performed in the laboratories here at the University of Cape Town).

(b) Animals

The available data on levels of alpha-activity in animals other than man is sparse indeed, and such data as thereis concerns mainly the activity levels in bone tissue. Table 1 gives levels of activity for specific isotopes in various species, and a few conclusions may be drawn from the data. Levels are given by the various authors as pc/gram bone ash and, because of lack of information on concentration factors from bone ash to wet bone, no attempt has been made to convert the data to activity in wet bone. In comparing the data it must be assumed that the various ashing techniques used are uniform (i.e. ashing techniques as used on a specific bone by the different workers would give the same concentration factor). The data from work by Mayneord et al. has been calculated from their measurements of "total alpha-activity" and Th-228/Ra-226 ratios.

Table 1

Reference	Specie	No. of Samples	Th-228 pc/gram ash	Ra-226 pc/gram ash	Pb-210 pc/gram ash
Lucas & Ferrante (35)	Bovine	25	0.10-0.78	0.19-2.58	0.06-1.67
Holtzman (36)	Bovine	1			0.32
Hill (37)	Bovine	2			0.24-0.26
Petrow (38)	Bovine	4	0.15-0.16	0.31-0.40	2.25-2.48
Mayneord et al. (2)	Bovine	3?	∿0.09-0.32	∿0.27-0.79	
Mayneord et al. (2)	Sheep	2?	∿0.07-1.11	∿0.15-1.00	
Nelson & Rust (39)	Pig	6		0.015-0.075	
Lucas & Ferrante (35)	Deer	1		0.87	
Mayneord et al. (2)	Deer	2?	∿0.10-0.11	∿0.30-0.32	
Holtzman (36)	Reindeer	6		0.40-1.84	4.73-9.67
Holtzman (36)	Caribou	33		0.38-0.70	5.23-20.3
Holtzman (36)	Moose	1		0.20	0.70
Mayneord et al. (2)	Rabbit	1?	∿0.25	∿0.51	
Lucas & Ferrante (35)	Rabbit	1		0.41	

Table 1 (continued)

Reference	Specie	No. of Samples	Th-228 pc/gram ash	Ra-226 pc/gram ash	Pb-210 pc/gram ash
Lucas & Ferrante (35)	Horse	1		0.14	
Mayneord et al. (2)	Camel	1?	∿0.30	∿0.92	
Mayneord et al. (2)	Dog	1?	∿.003	∿.007	
Holtzman (36)	Wolf	2		0.10-0.11	0.06-1.39
Holtzman (36)	Seal	3		0.06-0.07	0.05-0.07
Holtzman (36)	Fish	2		0.02-0.03	0.05-0.29

Conclusions drawn from table 1 are:

(a) That the activity levels in animal bone are generally at a much higher level than that found in human bone cf. for example the Ra-226 values in table 1 with the "world wide average" of 0.013 pc/gram bone ash for Ra-226 given by Walton et al. Notable exceptions to this are Ra-226 levels in Pig, Dog, Seal and Fish bone.

(b) That the activity levels in bone in a single animal species may have a considerable range. This is well illustrated by the work on 25 bovine samples by Lucas and Ferrante and on 33 caribou samples by Holtzman.

(c) Pb-210 does not always appear to be in excess of its parent Ra-226 as is the case in human bones. The data available is however too limited to infer any reason for this.

Further useful information in terms of the present work on animal bone activity levels is restricted to isolated articles and may be tabulated:

(i) Petrow(38) in studies on bovine bone ash finds that Th-228 is not in equilibrium with its precursor Ra-228 (a beta emitter).

(ii) In work by Nelson and Rust(39)on six pigs, eleven different bone sample for each pig were analysed for Ra-226. The results showed a significant

variation of Ra-226 activity per gram bone ash in different bones of a single pig. This contrasts with the situation for humans (see earlier) wherein the Ra-226 activity per gram bone ash is considered uniform throughout the body.

(iii) Holtzman(36) in studies on Caribou and Wolf shows that for both these Po-210 is in approximate equilibrium with its precursor Pb-210.

The reported levels of activity in animal liver appear to be confined to a single value for the Pb-210 content of bovine liver by Holtzman(36) , who gives a value of 0.006 ± 0.009 pc/gram wet tissue.

Holtzman gives the Pb-210 content of a variety of other soft tissues from the same animal, the range of values being 0.0005 - 0.0067 pc/gram tissue. Little can thus be said about the alpha-activity levels in liver and blood for animal species.

Chapter 2

TOTAL ALPHA-COUNTING

2.1 Theory

The alpha counting technique used in this thesis is one of "thick source" alpha counting. In this the thickness of the sample is greater than the maximum range of the alpha particles emitted by the sample (typically 20-30 microns and seldom greater than 90 microns in solid materials).

For the idealized case of a single isotope homogensously distributed in a homogeneous sample we have

$$C = \frac{NAR\rho}{4} \qquad (1)$$

where

N - no. of alpha particles emitted per unit time per unit mass of sample

A - source area

R - range of alpha particles in the sample

ρ - density of the sample

C is the count rate per unit time in the detector provided that alpha-particles down to zero energy are detected with 100% efficiency, and that there are no contributions from other sources (e.g. beta's, gamma's, noise etc.).

This formula was discussed in an early paper by

Finney and Evans(40) and has been successfully applied by a number of authors {e.g. Nogami and Hurley(41); Turner et al.(15)}. Where the sample contains a mixture of alpha emitting isotopes a summation must be taken over R and N.

$$C = \frac{A\rho}{4} \sum_{i} N_i R_i \qquad (2)$$

This summation demands that some prior assumptions be made on the nature of the activity present in the sample. Results of previous workers and the limited data of the alpha spectrographic method discussed later (see chap. 3) give an indication of the alpha emitting elements present and also a rough (not quantitative) idea of the relative quantities present. For the purposes of the basic calculations in the present work we shall assume:

(i) That the activity contribution in the material is from the "natural" Uranium, Thorium and Actinium Series.

(ii) That the head members of the Uranium and Thorium Series are Ra-226 and Ra-228 (a beta emitter) respectively. This assumption follows that of Mayneord et al.(2) , and further a review by Spiers(3) . It is also supported by the few samples examined spectroscopically (see chap. 5

sect. 5.3). Since the long lived series members above Ra-226 and Ra-228 are assumed to be present in insufficient quantities to support the observed activity it is also presumed that the two long lived Actinium series members U-235 and Pa-231 are present in negligible quantities and thus the contribution from the entire Actinium series is deemed negligible. This latter assumption is a minor one, since even if U-235 is present the Actinium series contribution to the total alpha activity is normally only ∿5% of the Uranium series contribution.

(iii) That radioactive equilibrium exists in both the Uranium and Thorium series below Ra-226 and Th-228 respectively, apart from a possible disequilibrium situation below Pb-210 (see (iv) below). All samples were left to settle for at least three weeks to ensure attainment of this degree of equilibrium.

(iv) That there can be a contribution to the alpha activity from the Uranium series daughter Po-210 over and above its contribution in (iii). This excess Po-210 results ultimately from the emanation of gaseous radon into the atmosphere. The "natural fallout" of Pb-210, and daughter has been discussed

by many authors {Burton and Stewart(42); Hill (16) etc.} and the degree of equilibrium between the beta-emitter Pb-210 and its alpha emitting daughter Po-210 is variable.

Equation (2) now becomes

$$C = \frac{A_\rho}{4}\left[N_{226} \sum_i R_i + N_{228} \sum_j R_j + N_E R_{Po\text{-}210} \right] \quad (3)$$

where N_{226}, N_{228} and N_E are the activities per gram of Ra-226, Th-228 and the "excess" Po-210 respectively.

In order to extract the specific alpha activity of the sample as a whole from the above equation it is necessary to obtain further information about the various contributions. The Th-228 activity may be determined independently by the "pairs technique" as suggested by Hurley and Shorey(43) and developed by Turner et al.(15) and others. This technique is based on the fact that there are in the thorium series two successive alpha decays from Em-220 to Po-216 and from Po-216 to Pb-212 separated by a short time interval (the half-life of Po-216 is 0.158 seconds). There will therefore be "pairs" of alpha particles emitted with a very short time interval between the successive disintegrations. By including an electro-

mechanical register of long dead time (about 0.4 secs) in addition to the normal "fast" scalar, the "pairs" rate may be determined. The pairs rate formula then gives Cherry(45)

$$C_p = \frac{nAR_1\rho}{8}\left[1 - \frac{R_1}{3R_2}\right] \qquad (4)$$

where

C_p - number of alpha "pairs" emerging from area A per unit time

R_1 , R_2 are ranges of two alpha particles involved where $R_1 < R_2$

n - no. of Em-220 disintegrations per unit mass of sample per unit time.

On the basis of the assumption of equilibrium given in (iii) above, we have $n = N_{228}$.

The next problem concerns the estimation of N_E . For samples containing "excess" amounts of lead-210 and/or Po-210 activity, three possibilities arise.

(a) the activity will show an exponential decrease on a 138-day time scale if the Po-210 activity is in excess of the Pb-210 activity.

(b) the activity will show an increase if it is supported

by Pb-210 in excess of Po-210 activity. This increase will be an exponential rise on the time scale of the Po-210 half life.

(c) if Pb-210 and Po-210 are in equilibrium there will be no appreciable time variation over a period of the order of a year or less (the approximate time interval involved in the present work).

The contribution in (a) above is readily calculated by noting the fall off in the count rate over a specific time (generally $\geq$ 4 months). The count rate due to unsupported Po-210 at time of sample collection is given by

$$C_{Po\text{-}210} = \frac{\Delta C}{1-e^{-\lambda \Delta t_1}} \cdot \frac{1}{e^{-\lambda \Delta t_2}}$$

where

ΔC is change in count rate

Δt_1 is time (in days) elapsed between two successive counting periods

Δt_2 is time elapsed from sample collection to first counting period

λ decay constant of Po-210.

The contribution due to (b) may be calculated on a similar basis. Only a few of the many samples examined showed this count rate increase feature and they are studied individually

under the results in chapter 5.

The technique as outlined this far (which provides the bulk of the data in this thesis) does not provide a means of determining the contribution due to (c). The work of previous authors e.g. Hill (33), Holtzman(31) ; using additional separation techniques indicates that there can often be an "excess" of Po-210 contribution which is supported by Pb-210 (but not by Ra-226), and it must be recognized that out total alpha counting data does not provide information on this point. The primary purpose of this work is, however, to compare total alpha-activities in various samples, and the presence of excess Pb-210 is of major concern only if it affects the accuracy of the results presented. The total alpha-activity data will be subdivided into thorium series activity, uranium series activity, and a time dependant contribution (representing the cases (a) and (b) above). The contribution to be labelled "uranium"series activity will therefore depend upon whether we ignore the possibility of "excess" Po-210 which is in equilibrium with Pb-210,or not. Fortunately the maximum possible error involved is small, since the average alpha-particle range (in air) for the whole Uranium series below Ra-226 is 4.52 cms whereas that for Po-210 alone is 3.85 cms. In the calculations an arbitrary

decision to exclude the possibility of excess Po-210 in equilibrium with Pb-210 has been made and it must be recognised that the "uranium series activity" values might therefore be too high by as 17.4%.

We next turn to the problem of estimating the ranges R which occur in formulas (3) and (4). The problem of the range of alpha particles in various media has been studied by many authors {Whaling(44), Cherry (45) etc.} though mostly for single elements. The range in composite media is usually calculated by the atom fraction method {e.g. Nogami and Hurley(41)}. A number of empirical formulae exist for the calculation of the range. A formula by Glasson(46) assumes that the stopping power (and not the range as used by Turner et al.) is proportional to $Z^{2/3}$ and is used in the present work. According to this formula we have

$$R_\rho = (.032)(10^{-3})\, R_{air} \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}}$$

where

f_i - atom fraction of element

w_i - atomic weight of element

Z_i - atomic no. of element

R_{air} - range (in cms) of alpha particles in air at N.T.P.

The alpha particle ranges in air for the various isotopes are well known and are listed in Table 2, using data from Siri(50).

As an empirical check on the validity of the Glasson range formula and the experimental procedure in general several blood samples were spiked with known amounts of old thorium sulphate (i.e. with the complete thorium series in equilibrium). The results showed that the calculations based on the above gave results about 10% lower than expected. This is probably due to

(i) The ZnS phosphor being granular and hence there not being 2π geometry in the sample phosphor system. This results in small correction factors which differ for the counts and pairs respectively.

(ii) The Glasson formula for range being in error.

The count and pairs rates are thus multiplied by factor k and k_p respectively where k and k_p are determined from the "spiked" experiment (appendix III) and are found to be $k = 1.13$ and $k_p = 1.11$.

The justification for using the Glasson stopping-power formula is somewhat arbitrary but it has been chosen because other spiking experiments (e.g. on sea water, by Gericke(47) have shown that the k and k_p factors show less

Table 2

Alpha ranges in Standard Air. {Siri(50)}

Alpha Emitting Isotope	Range in "Standard Air" cms
URANIUM SERIES	
Uranium-238	2.65
Uranium-234	3.21
Thorium-230	2.85
Radium-226	3.13
Emanation-222	4.05
Polonium-218	4.66
Polonium-214	6.91
Polonium-210	3.85
THORIUM SERIES	
Thorium-232	2.80
Thorium-228	3.67
Radium-224	4.08
Emanation-220	5.00
Polonium-216	5.64
Bismuth-212 (33.7%)	4.73
Polonium-212(66.3%)	8.57

"Mean"air ranges for alpha particles from isotopes in Uranium series below Ra-226 = 4.52 cms.

"Mean"air ranges for alpha particles from isotopes in Thorium series below Th-228 = 5.14 cms.

variation from one sample medium to another than if alternative formulae (e.g. the $Z^{2/3}$ range dependence used by Turner et al., or the well known Bragg-Kleeman formula) are used.

Finally we obtain

$$C = 8\times10^{-5} k A \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} \left[5.14 N_{Th} + 4.52 Nur + 3.85 N_{Po210}\right]$$

$$C_p = 2.82\times10^{-5} k_p A \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} N_{Th}$$

$$C_{Po210} = \frac{\Delta C}{(1-e^{-\lambda \Delta t_1})} \cdot \frac{1}{e^{-\lambda \Delta t_2}}$$

where

C, C_p are observables in counts/hour and pairs/hour respectively.

N_{Th} is the no. of alpha disintegrations per hour per gram resulting from the thorium series

Nur is the no. of alpha disintegrations per hour per gram resulting from the uranium series per hour

C_{Po210} is the count rate per hour due to "excess" in supported polonium 210, at time of sample collection.

In conclusion it is worth noting that the sensitivity of this method is limited by

(a) The degree to which the specific activity of the sample can be concentrated and

(b) the maximum value of A (determined by the size of the photocathode on the photomulitplier tube).

2.2 Sample Preparation

Blood and liver have a low specific activity, and in their natural state are liquid or moist. Concentration and drying are thus required. Bones, on the other hand, have sufficient specific activity to eliminate the necessity of concentration and only require drying from their natural state and removal of any adhering fat, tissue, or marrow. The procedures adopted for sample preparation are as follows:

Blood and Liver

It is desirable to concentrate the activity without recourse to chemistry and the method of freeze drying (lyophilisation) was chosen as the most suitable technique. Flosdorf et al.(48), Merryman(49), and Hasson(34) give detailed reviews of this process. This essentially involves the sublimation of moisture from the sample under a vacuum.

The livers are pulverized to liquid form in a Braun food mixer prior to freeze drying. About 60 ml of the weighed sample (liver or blood) are placed in a 2 litre round-bottomed Pyrex flask and shell frozen onto the sides of the flask by rotating in a mixture of dry ice and alcohol. When the sample is completely frozen a

characteristic cracking sound is heard as the sample separates from the walls of the flask. The flask is then connected to the outlet manifold (four outlets in the case of our apparatus) of a large condenser, and a vacuum is applied to the condenser using a large capacity rotary oil pump. The condenser is insulated with polystyrene and cooled to -70°C with a dry-ice, alcohol mixture (fig. 1).

Sublimation of the water from the sample keeps it in a frozen state and no further external cooling is necessary. The sublimated water freezes out in the condenser, thus protecting the pump. As the sample dries, the atmospheric ice layer, which forms on the outer walls of the flask recedes, and the temperature of the sample rises to room temperature. At this stage more than 95% of the water content has been removed (fig. 2).

In this manner four samples at a time can be treated, drying time being approximately 10 hours. Three runs can be undertaken before the condenser needs defrosting. The advantages of this method in the present work are:-

(i) Because of the low temperatures involved, loss of volatile constituents, other than water, is minimized.

(ii) The sublimation of the water does not involve any

Fig.1. Freeze Drying Apparatus

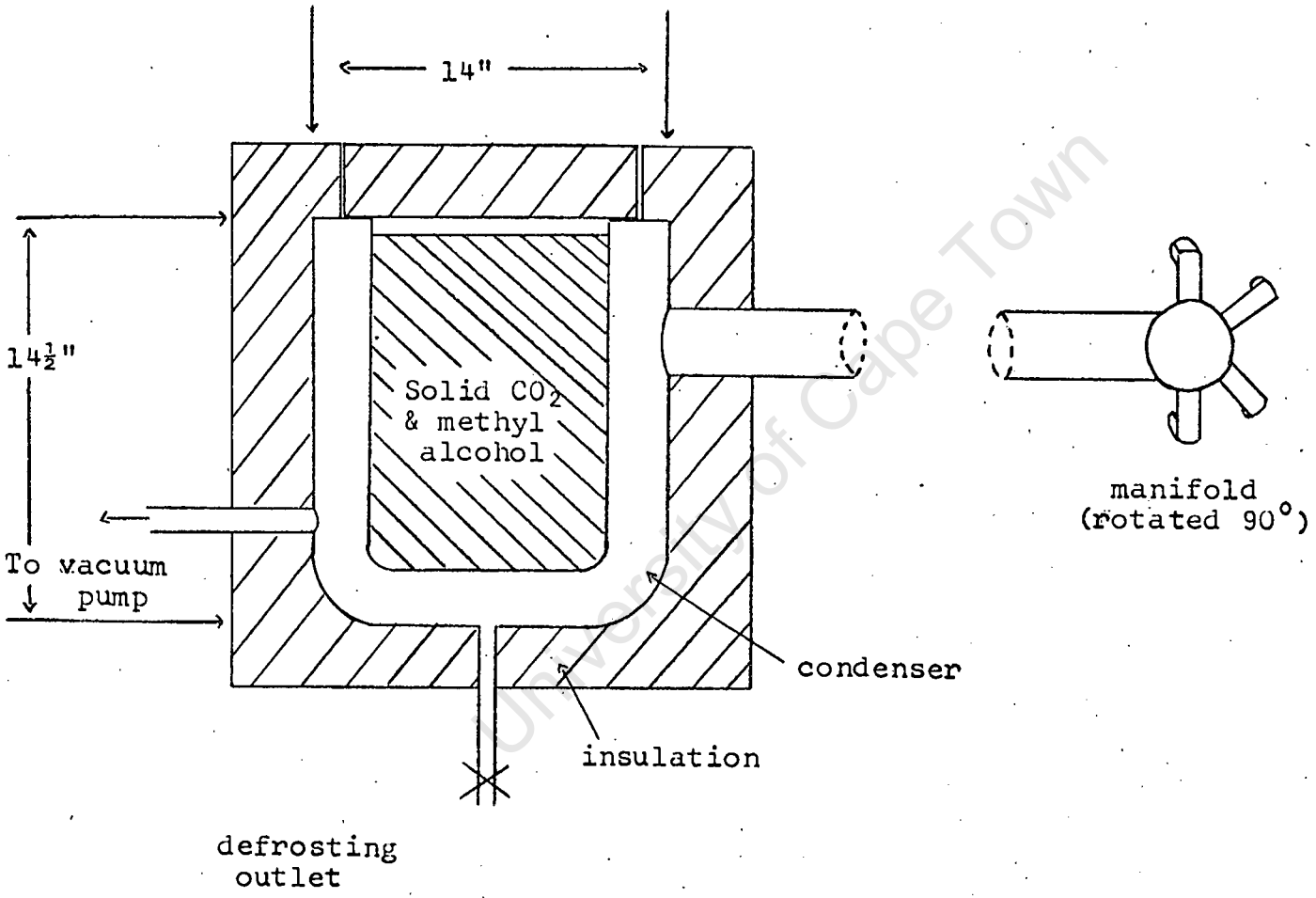
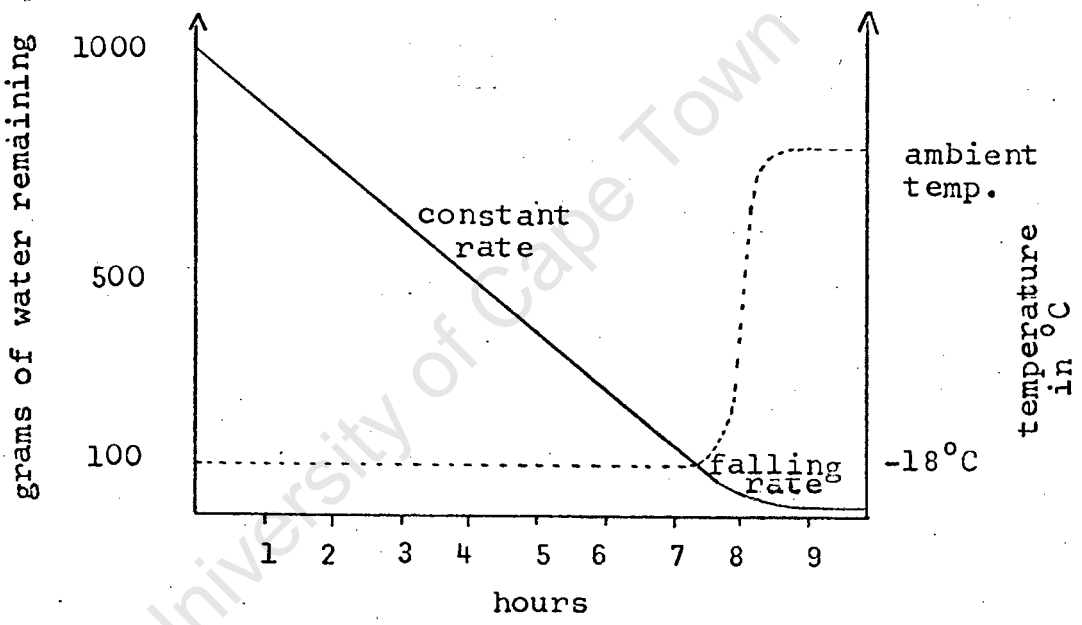


Fig. 2

Drying Curve for Freeze Drier



Rate of drying of 10% plasma protein solution frozen in a uniform flat layer 10mm thick

{Details from Hasson(34)}

physical movement of the solid material and the original structure of the sample is retained.

(iii) The final product is easily ground to a homogeneous powder in a pestle and mortar.

(iv) Samples of a different nature may be dried on the apparatus at the same time.

(v) The concentration factor attained in the drying process is easily determined.

Bones

The bones are cleaned of all surplus adhering tissue and then steamed in a pressure cooker for approximately 20 minutes to remove any fat and marrow and to loosen ligament endings. The resulting bones are clean and sharply defined (no soft outer layer) and are then left to "dry out" completely in a dust free room for 2 to 3 weeks. The dried bone is then crushed in a hydraulic ram to pieces about ½ inch square, the sample being well wrapped in plastic sheeting to avoid contamination by contact with the metal ram surfaces. The crushed sample is then finely ground in a Braun coffee grinder.

In order to check whether or not the degree to which the samples were ground affected the count rate several samples were prepared in two lots, one being fine ground

(separated out to less than 100 microns by sieving) and the other being very coarse. The results are shown in Table 3, and indicate that the particle size does not affect the count rate to a significant degree.

The problem of ascertaining the concentration factor of bone is rather more difficult and to a certain extent depends on the definition of bone (in vivo). As a basis for this work bone (in vivo) is defined as being that hard material, free from any soft adhering tissue which makes up the skeleton of animals. Several bone samples were scrupulously cleaned, weighed, prepared for crushing as above and then reweighed. The results are shown in Table 4 Since by their very nature the bone sheath, tendons and marrow cannot easily be removed from bone in vivo,(although they all come free on boiling) the figures shown for the concentration factor are likely to be slightly higher than the true value in each case. The results show good uniformity except for the one pig bone which was a trotter (foot) and impossible to free from much of the adhering tissue, and one ox bone which was porous and filled with interstitial marrow. A mean value of 1.3 is taken as the concentration factor of bone from the in vivo state to that as defined above.

The packing of the ground liver, blood and bone surplus

Table 3

Count rate dependance on particle size for bone

Sample	Corrected count rate. Sample fine ground (particle size < 100 microns) c/hr	Corrected count rate. Sample coarse ground (particle size >> 100 microns) c/hr
Cow bone	98 ± 2	99 ± 2
Pig bone (1)	3.4 ± 0.6	3.1 ± 0.6
Pig bone (2)	4.9 ± 0.6	4.6 ± 0.6
Donkey bone	69 ± 2	69 ± 2

Table 4

Bone Concentration Factors

Specie	Type of bone	Concentration Factor
Ox	Tibia	1.33
Ox	Tibia	1.25
Ox	Tibia	1.21
Ox	Tibia	1.24
Ox	Femur	1.32
Ox	Femur	3*
Ox	Femur	1.41
Ox	Femur	1.26
Pig	Tibia	1.41
Pig	Tibia	1.20
Pig	Metatarsals	2.96*
Human	Femur	1.37
Human	Femur	1.23
Human	Femur	1.39
	Mean	1.30

Note: Values marked * excluded (see text 2.2 Bones)

for alpha-counting follows the method described by Turner et al.(15). This method is simple and economical, and involves minimal contamination and emanation loss problems. A perspex ring is firmly placed on the adhesive surface of (Scotch Brand) cellulose adhesive tape. Zinc sulphide phosphor power is then sprinkled uniformly over the adhesive surface. The phosphor used in this work is Levy-West type G231 silver activated zinc sulphide, which has a specified grain size of between 20 and 40 microns.

The powdered sample is packed directly onto the zinc sulphide, covered with a labelled cardboard disc, and sealed with a further layer of adhesive tape. Samples are then stored in a desiccator containing phosphorous pentoxide and allowed to settle for two to three weeks before counting to allow secular equilibrium to be attained. Use of a drying agent in the desiccator was found by experience to be necessary to stop any absorption of moisture by the sample. It is probable that absorbed water dissolves some of the alpha emitting elements which are in turn preferentially adsorbed onto the surface of the phosphor. The homogeneous thick source assumption then no longer holds and anomalous and non reproductive results are obtained. Storage of samples in a desiccator was found to permit reproducible results for samples stored for a period of at

least eighteen months.

All glassware used in preparation of the samples is routinely cleaned with hydrochloric acid, then washed with teepol and finally rinsed with distilled water. Checks on possible contamination by the Braun grinder were made by grinding pharmaceutical grade Dextrose which has a very low activity. No observable contamination was noted during the course of the experiments.

2.3 Counting Technique

The sample discs are placed on an EMI 9530B photomultiplier with a 5" photocathode housed in a light-tight metal can. Low noise level is attained by "dark adaption" {Gordon et al(51)} and by tuning the cathode to first dynode resistor {Pagano et al.(52)}. Actual changing and positioning of samples on the photomultiplier takes place in the dark with the E.H.T. switched off.

The pulses from the photomultiplier are fed through a conventional pulse amplifier followed by a discriminator, and thence fed simultaneously into an electronic scaler with a resolving time of 200 μsecs and an electromechanical register with a dead time of about 0.4 secs. The total alpha count is given by the scaler whilst the alpha pairs rate is given by the difference between the scaler and electromechanical register readings.

The E.H.T. supply voltage to the photomultiplier and the discriminator bias voltage must be set so that, ideally, all the alpha counts from the sample are recorded and all the other (e.g. beta and gamma rays) are rejected. This is done by plotting the count rate from a suitable alpha-emitting source (the blood samples spiked with old Thorium sulphate was used) as a function of E.H.T. for a fixed discriminator bias with a suitable choice of E.H.T. voltage

it is possible to obtain a satisfactorily long "plateau" and to determine the true alpha count rate to within a few percent. The operating points selected corresponded to E.H.T. settings at the lower end of the thorium sulphate plateau. This reduces the operating E.H.T. (and hence the photomultiplier noice) to a minimum while still providing high efficiency for alpha particles.

The corrections that must be applied to the observed alpha-pairs and total alpha count rate, due to the nature of the counting equipment are:-

(a) Count rate corrections

(i) Background correction due to electronic noise. This is determined periodically by operating the counting set up without any sample disc present. A typical value is 0.5 ± 0.2 c/hr.

(ii) Background phosphor count rate. In order to determine this one should ideally count a sample whose material contains zero activity. It was not possible to find any such material. However, certain materials, notably pharmaceutical grade Dextrose gave consistent, very low count rates of the order of 2.0 ± 0.3 c/hr. This value was taken as the background phosphor count rate.

The above corrections must be subtracted from the observed count rate.

(b) Pairs rate correction.

(i) Background due to electronic noise. This correction, determined as in (a)(i) above was found to be negligible.

(ii) The phosphor background pairs rate was determined using the same material as in (a)(ii) above and was found to be 0.02 ± 0.02 p/hr.

(iii) Spurious alpha pairs formed by coincidences of unrelated counts. Following Cherry (45) the spurious pairs rate is $N^2\tau$ where N is the total observed count rate and τ the dead time of the electromechanical register. τ was determined periodically for all the counting set-ups in use by means of a uranium source of suitable activity.

The above corrections must all be subtracted from the observed pairs rate. Further, since the register has a finite dead time not all the alpha pairs are recorded. This is allowed for in a multiplicative factor of form:-

$$f = \frac{1}{1 - \exp\left[\frac{-0.693\tau}{T_{\frac{1}{2}}}\right]}$$

where $T_{\frac{1}{2}}$ is the half-life of the Po216 nucleus responsible for the pairs i.e. 0.158 seconds.

These pairs rate corrections are all small and the statistical error in the pairs rate is normally larger than any of these.

Chapter 3

ALPHA SPECTROSCOPY

For alpha spectroscopic studies source thickness needs to be limited to about 100 $\mu g/cm^2$ in order to avoid excessive contributions to the line width. If alpha-spectroscopic measurements on biological samples are to be made without prior chemical concentration, this limitation requires that a detector with a large source area be used. Cherry(53) has reviewed the subject of alpha-particle detectors as applied to samples from the natural environment, and has compared the performance of alpha-spectroscopic detectors designed by several workers for this purpose, and it is clear that the ion chamber of Hill(17) is the most sensitive instrument. The ion chamber used in the present work follows Hill's design, and the construction followed from plans kindly made available by him. Basic features of the Hill ion chamber are:

(i) Two concentric cylindrical electrodes of length 90 cms and radii 0.45 cms and 30 cms respectively.

(ii) The material to be analysed is mounted on the metallized surface of a sheet of approximately 0.1 mlm thick aluminized cellulose acetate lining

the inner surface of the outer electrode and providing a useful area of 15,000 sq cms for the source.

(iii) A negative high tension of 5000 volts is applied to the source, the central collecting electrode being at ground potential.

(iv) A set of four concentric guard rings is placed at each end in order to maintain approximately radial field conditions throughout the length of the chamber.

(v) A standard calibrating source is provided on one side of a rotatable strip placed in a notch cut in the source holder, with provision for turning the calibrating source into or out of the active counting volume of the chamber.

(vi) Gas tight stainless steel chamber in order to achieve low radon emmanation.

(vii) Provision for evacuating the chamber to a pressure of less than 0.5 mlm Hg before filling with 98% Argon 2% Methane counting gas.

(viii) A continuous gas purification system in order to ensure constant conditions of electron collection efficiency and electron collection time, and also to remove any radon which might accumulate. The

gas is first passed over a column of metallic calcium turnings maintained at a temperature between 300 and 350°C (to remove oxygen). It then passes over a column of charcoal cooled to -70°C in a bath containing a mixture of solid CO_2 and acetone, to remove radon.

(ix) The chamber is not gridded. Hill calculates theoretically for his chamber that without a grid and in the range 4-9 MeV the line width due to "positive ion effect" only ranges from $2\frac{1}{2}$-5% of total line width.

Certain modifications were made in our apparatus in order to facilitate operation and in an attempt to improve chamber function. (Figs. (3) and (4) show our apparatus and counting assembly) These were:-

(i) Use of 90% Argon 10% methane. This is obtainable commercially in a high purity form (used extensively in proportional counters) and eliminated difficulties experienced here in obtaining separate cylinders of sufficiently high purity Argon and methane.

(ii) A 6 Kv high tension supply is used. For 90% Argon 10% methane, this gives a collection efficiency approaching 100% without any gas multiplication. (This feature was checked experimentally). It also

Fig. 3. Gas chamber and gas circulation system

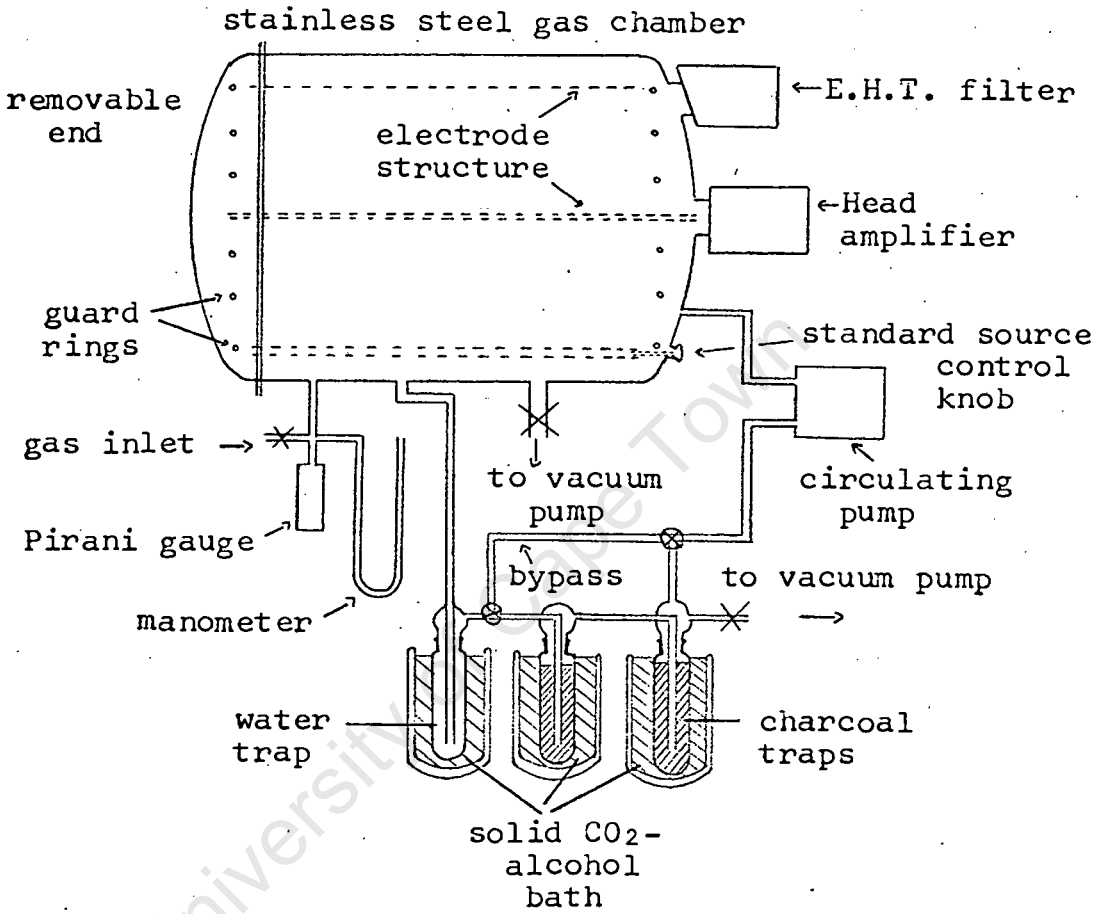
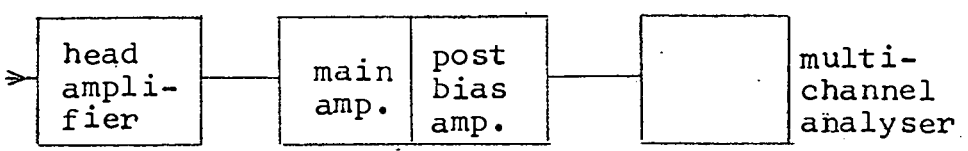


Fig. 4. Block diagram of electronics



gives a fast drift velocity (i.e. a short electron collection time) (Fig. 5).

(iii) The gas purification system was altered to consist of a water trap and two charcoal traps. A circulating pump with a greater range of flow rates than that of Hill's was used. Careful attention to sealing rendered gas leaks negligible, and with the high purity gas used, the heated calcium column was deemed unnecessary. In a series of tests it was found that gas characteristics were highly sensitive to the presence of water vapour and that adsorption of water onto charcoal impaired its radon adsorption efficiency. For this reason a water trap was included. With this system it was possible to maintain stable gas conditions and good resoltuion in the chamber for periods of up to 100 hours. Table 5 shows peak positions at intervals over a 100 hour run.

A problem with our chamber is that the stainless steel walls have a relatively high radon emanation. Other methods of removing the radon (i.e. freezing out, since radon solidifies at -71°C whereas Argon and methane liquify at temperatures < -150°C) were investigated but were not

Fig.5 Drift velocities of electrons in argon +10% methane mixture {English & Hanna(54)}

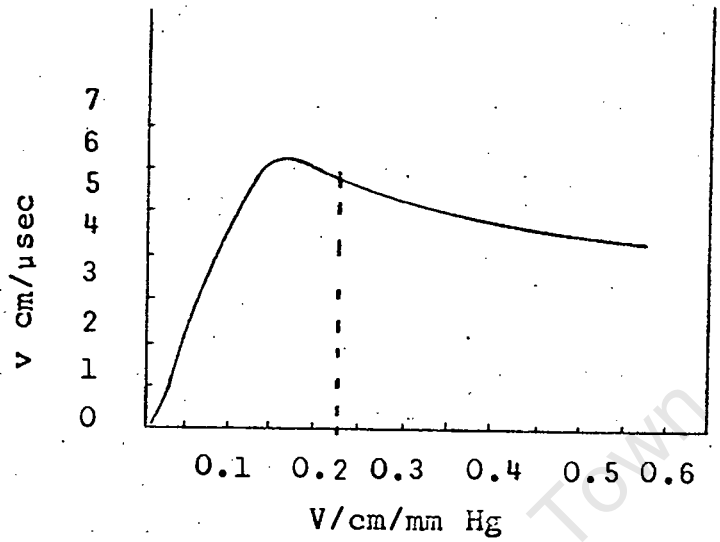


Table 5

Peak positions of Test Sources in 100 hour run of Ion chamber (U-238 and U-234 test sources)

Date	Time	Peak position U-238	Peak position U-234
		channel no.	channel no.
4/2/69	11.15 am	64	83
"	4.20 pm	64	83
"	10.30 pm	64	83
5/2/69	8.20 am	65	84
"	1.30 pm	64-65	83
"	11.30 pm	64	83
6/2/69	8.30 pm	64	83
"	2.15 pm	64	83
"	11.30 pm	64	83
7/2/69	8.20 am	64	83
"	5.05 pm	64	83
8/2/69	8.30 am	64	83

Date from 256 channel pulse height multi-channel analyser at 32 keV per channel

considered feasible. Experiments with charcoal showed that the radon adsorption rate varies inversely with the flow rate. This disadvantage was partially overcome by increasing the amount of charcoal instead of increasing the flow rate. Charcoal also adsorbs methane, but it was found that the slight variation in gas mixture as a result of this, did not affect the gas characteristics to any degree. This observation is also confirmed by other workers i.e. Kocharov(55).

Prior to running any samples a background blank of plain aluminized cellulose acetate was run in order to ascertain the contribution from chamber emanation and from the natural activity of the acetate. Results of this and of a typical test source run are shown in Figs.6 & 7.The background count over range 4-9 MeV was 218 c/hour. This is somewhat higher than that obtained by Hill (~90 c/hour) and as explained above is attributable to the nature of the stainless steel walls of the chamber. The three test sources used are U-238 (4.195 MeV), U-234 (4.768 MeV) and Pu-239 (5.160 MeV). The uranium sources were prepared in our laboratories and have a considerable line thickness. The Pu-239 source was supplied by the Radiochemical Centre, Amsterdam has a relatively small line thickness. The resolution of the Pu-239 source in chamber operation was

Fig. 6 4 Minute Test Source Run

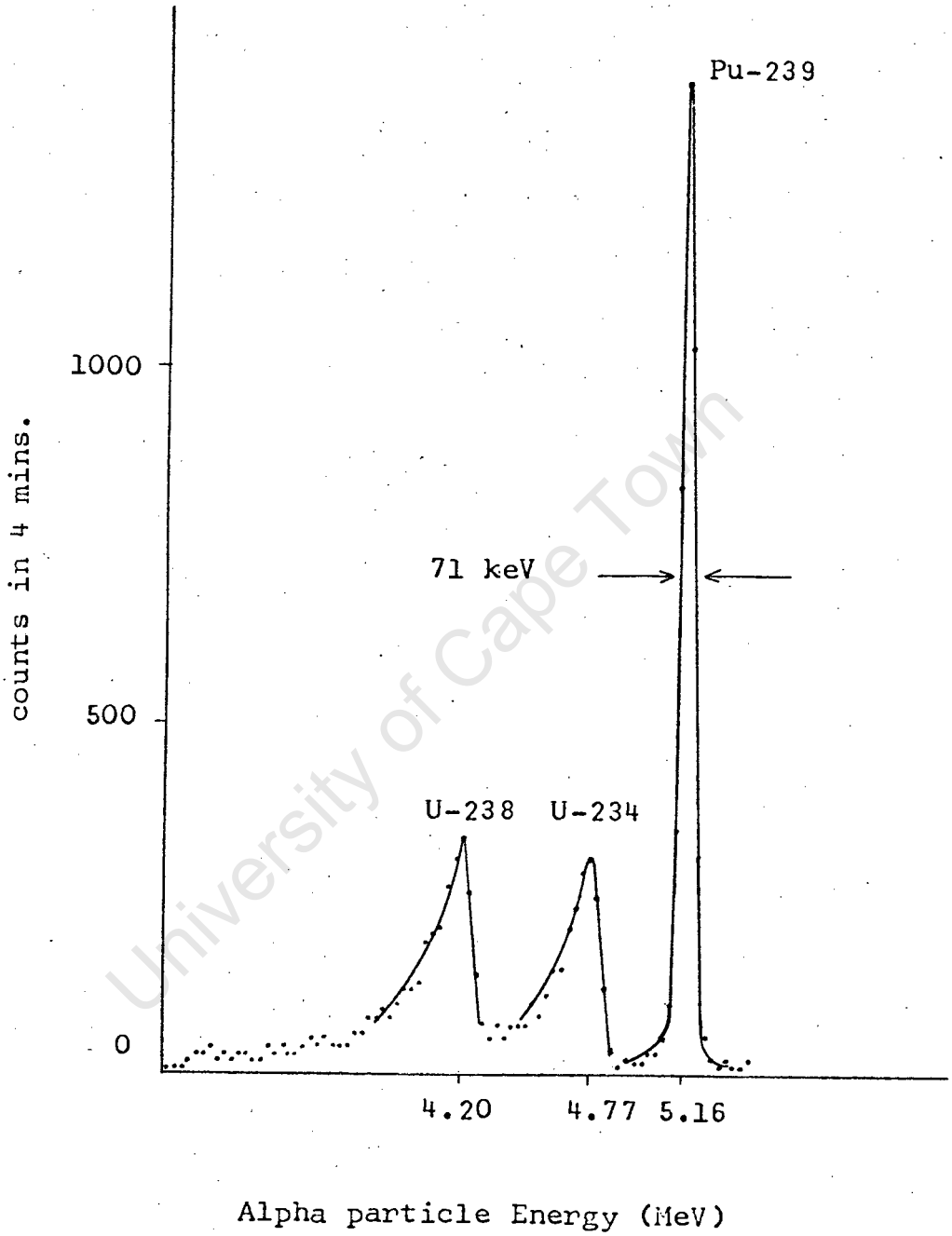
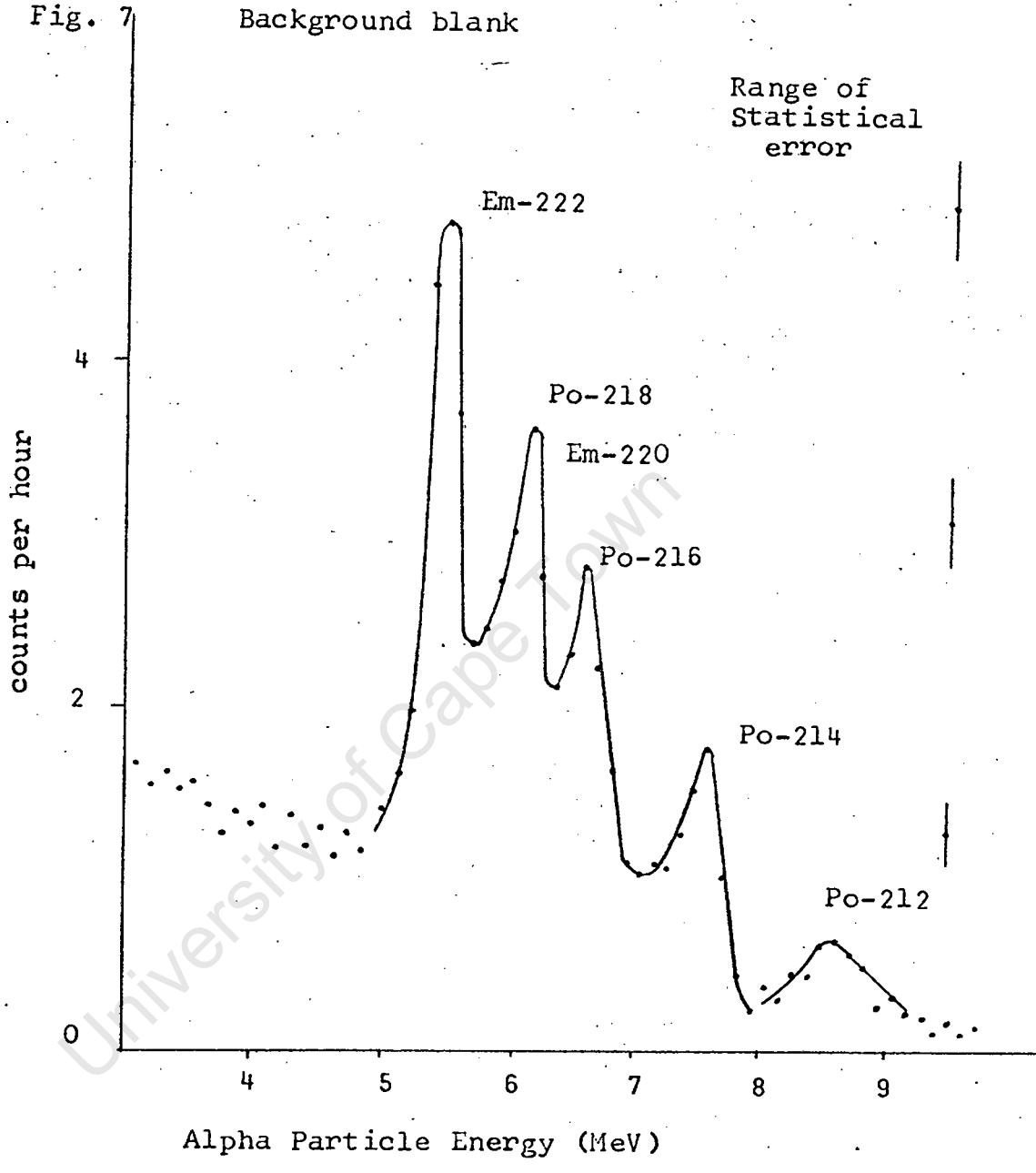


Fig. 7 Background blank



consistently in region of 70 keV (full width at half maximum height) as compared with resolutions of 110 keV obtained by Hill.

Routine operation of the chamber proceeds as follows:

(i) The chamber is pumped down by a large capacity rotary oil pump to approximately 180 μHg. At the same time the charcoal traps, isolated by a system of taps, are heated to drive off adsorbed gasses, and pumped down by a second pump.

(ii) The counting gas is "bled" in from the cylinder until the pressure in the chamber is atmospheric plus 1 cm Hg (for ease of leak detection). The gas is then circulated, through the water trap only, for approximately 10 hours to condense any water vapour present (the charcoal traps are out of circuit during this process).

(iii) The charcoal traps are introduced into the circuit, the high tension is applied and periodic checks are performed with test sources until their position in the analyser spectrum remains stable.

(iv) A four minute "test source" run is performed. The test sources are then turned out of the counting volume and the source is allowed to "run" for ∿50 hours. At the end a test source run is again

performed to check for any possible shift in the analyser spectrum.

The results so obtained are converted to counts/hour/channel and the background, treated in the same fashion is subtracted from source. The resulting counts/hour/channel are then plotted on a graph and by fixing the known energy points of the calibration source, the peaks in the source may be identified.

In order to obtain a thin source of our chamber, of source area 15,000 sq cms, approximately one gram of material, finely ground is used. The grinding technique, when used in minerals takes the particle size down to < 5 microns. Bone material is slightly fibrous as compared with the crystaline structure of mineral matter and hence not so readily ground, but it is estimated that the resulting bone particle size is of the order of 10 microns or less.

For bone the fire ground material as obtained from the Braun grinder in total alpha sample preparation is used as starting material. This is ground in a mechanical agate mortar and pestle in an alcohol suspension. Follow-

ing grinding, which takes about 20 hours the suspension is allowed to stand for 45 minutes to separate out the remaining large particles. The fine suspension is then decanted, diluted further with alcohol to about 100 ml and then sprayed onto the aluminised cellulose acetate sheet using a perfume spray.

In the case of livers, which are slightly fibrous, and of low density and hence difficult to grind under suspension, approximately one gram of the freeze dried material is first dissolved in concentrate nitric acid. The solution is carefully and slowly evaporated to dryness in a fume cupboard at a controlled temperature of 60°C. The resulting residue goes into suspension in alcohol with ease. This again is diluted to 100 ml and then sprayed onto the acetate sheet.

Once the alcohol has evaporated from the surface of the aluminised cellulose acetate, leaving a thin uniform covering of material, the sheet is fitted to a perspex cylindrical frame and placed in the chamber.

Chapter 4

RESULTS

The bulk of the raw data obtained in the experimental work done in this thesis is tabulated in Appendix A typical sample calculation is given in Appendix In calculating these values it has been assumed that the error due to counting statistics is of the Poisson form $\sqrt{N}$, where N is the observed count, and that this is valid for the "pairs" as well as for the total counts. This assumption is not strictly valid for the very low "pairs" rates which are frequently encountered but it is unlikely to be a major issue in the discussion that follows.

The data presented in this chapter are the summarized results from which the main conclusions will be drawn. In preparing these results attention was paid to the most suitable form of "mean value" for a set of samples and also to the best way of formulating the error. The two basic classes of "means" considered were weighted means, and arithmetic means.

(i) Weighted means and errors

In this the individual values are weighted according to their respective errors. The standard formula for

calculating the weighted mean of a set of values $x_i \pm \sigma_i$ is

$$\bar{x} = \frac{1}{\sum_i \sigma_i^{-2}} \sum_i \frac{x_i}{\sigma_i^2}$$

The standard error of the weighted mean may be calculated either by assuming "internal consistency" in which the standard error entirely depends upon the standard errors of the individual values, or by assuming "external consistency" in which the standard error depends upon the differences between the individual values. {Topping(56)} The formulae for these are:

Internal consistency

$$\bar{\sigma}^2 = \frac{1}{\sum_i \sigma_i^{-2}}$$

External consistency

$$\bar{\sigma}^2 = \frac{\sum_i \frac{(x_i - \bar{x})^2}{\sigma_i^2}}{(n-1)\sum_i \sigma_i^{-2}} \qquad n = \text{no. of values}$$

In calculating the individual values in the appendix tables the main error was that due to counting statistics, and errors quoted are due to counting statistics only. As previously stated, a Poisson distribution for the counting was assumed, giving an error of $\sqrt{N}$, where N is the

observed count. This means effectively that high count rates, i.e. high activities, will have low errors and that where there is a large range of values the weighted mean is biased in favour of the higher values. For this reason weighted means were considered unsuitable for the presentation of this work. The effectiveness of weighted mean errors was judged by comparing these errors to the actual range of the individual values. For the error to have any significance it must reflect in some manner the range of individual values (generally range $\bar{x} \pm \sqrt{n}\ \bar{\sigma}$, where n is number of values, should include approximately 66% of the individual values). Weighted means and internally consistent weighted mean errors were calculated for several sets of the data in Appendix I and it was found that these weighted mean errors did not adequately reflect the range of individual values except in cases where the range of values was small e.g. Pig livers. Weighted mean errors calculated by the method of internal consistency were thus considered unsuitable for this work. On the other hand the "external consistency" weighted mean errors were in all cases greatly in excess of the "internal consistency" weighted mean errors, indicating that for all the data there was a <u>real</u> range of values and that the range was not merely a consequence of the

individual errors.

(ii) Arithmetic means

For a set of values $x_i \pm \sigma_i$ this is

$$\bar{x} = \frac{\sum_i x_i}{n} \qquad n = \text{no. of values.}$$

The formula does not take into account the error of the individual readings. However, since the values are taken from a population in which there is a real variation of values, no one individual value can be anymore correct (within the range of its statistical error) than any other value. Hence an arithmetic mean is probably the best method of representing the population. The summarized data are therefore presented in the form of arithmetic means. The extent of the "sampling" of the population is given by quoting the range from the lowest to the highest value obtained for each set of data studied.

The samples studied were drawn from a number of areas in the Republic of South Africa and Rhodesia. For obvious practical reasons the number of soft tissue samples for the "wild animals" is somewhat limited. A full index of the origin of the individual samples is

given as a preface to the tables in Appendix I. A brief sketch of the nature and feeding habits of the "wild animals" studied will be useful and is given here.

Buck - herbivore; similar to English deer
Zebra - herbivore; member of "horse" family
Elephant - herbivore; long lived
Wild dog - carnivore; predator; member of "dog" family
Hyena - carnivore; scavenger; member of "dog" family
Jackal - carnivore; scavenger; member of "dog" family
Leopard - carnivore; predator; member of "cat" family
Lion - carnivore; predator; member of "cat" family
Cheetah - carnivore; predator; member of "cat" family
Baboon - omnivore ; diet mainly insects and roots

In presentation of the following tables, the species are ordered in descending order of activity level.

N.B.

(i) Where Po-210 excess levels are not given this implies that no such Po-210 excess was dectable within the limits of counting statistics.

(ii) In order to reduce confusion in tabulation the % values given have been calculated from the mean values, and ranges of %'s have not been given. Details of these can be obtained by

referring to Appendix I. The frequently high errors associated with the pairs data (which are used for calculating the thorium series percentages) should constantly be kept in mind.

Table 6

Bone

Species	No. of samples	Range of "total alpha activity pc/kgram wet bone	Mean "total alpha activity" pc/kgram wet bone	% Po-210 excess	% Uranium series activity	% Thorium series activity
Cattle	3	6400-7000	6600	34%	32%	34%
Sheep	2	2200-5800	4000	43%	25%	32%
Horse	1	-	1900	79%	5%	16%
Donkey	1	-	1900	44%	26%	40%
Buck	1	-	1600	32%	30%	38%
Fish	10	6-7800	1200	*	85%	11%
Dog	4	780-1100	910	N.D.	60%	40%
Cormorant (sea bird)	1	-	800	79%	5%	16%
Jackal	1	-	720	N.D.	22%	78%
Hyena	1	-	680	N.D.	36%	64%

Table 6 (continued)

Bone

Species	No. of Samples	Range of "total alpha activity" pc/kgram wet bone	Mean "total alpha activity" pc/kgram wet bone	% Po-210 excess	% Uranium series activity	% Thorium series activity
Dove (land bird)	1	-	580	N.D.	44%	56%
Chicken	1	-	400	N.D.	76%	24%
Baboon	1	-	390	N.D.	38%	62%
Lion	1	-	290	N.D.	64%	36%
Cheetah	1	-	270	N.D.	4%	96%
Seal	4	170- 340	240	N.D.	36%	64%
Man	7	180- 310	240	N.D.	50%	50%
Pig	7	110- 190	140	N.D.	70%	30%

* Not calculated because only one bone of ten showed Po-210 excess.

Table 7

Bone (Aged)*

Species	No. of samples	Range of "total alpha activity" pc/kgram wet bone	Mean "total alpha activity" pc/kgram wet bone	% Po-210 excess	% Uranium series activity	% Thorium series activity
Lion	1		1100	N.D.	42%	58%
Wild dog	2	640- 690	670	N.D.	35%	65%
Buck	4	160-1300	470	N.D.	38%	62%
Leopard	1	-	420	N.D.	62%	38%
Elephant	3	190- 690	380	N.D.	34%	66%
Zebra	3	300- 360	320	N.D.	68%	32%
Jackal	1	-	210	N.D.	47%	53%

* These bones were collected without accurate knowledge of the date of death of the individual animals. The samples however were studied at least nine months after the "supposed" date of death.

Table 8

Liver

Species	No. of samples	Range of "total alpha activity" pc/kgram wet tissue	Mean "total alpha activity" pc/kgram wet tissue	% Po-210 excess	% Uranium series activity	% Thorium series activity
Fish	10	160-4000	1300	71%	23%	6%
Seal	4	89-2400	930	96%	3%	1%
Cattle	2	140- 180	160	39%	..	61%
Buck	4	23- 460	150	43%	4%	53%
Dog	4	53- 120	83	N.D.	64%	36%
Pig	9	20- 210	67	N.D.	47%	53%
Sheep	2	53- 80	66	N.D.	68%	32%
Man	10	15- 43	26	N.D.	85%	15%

Table 9

Blood

Species	No. of samples	Range of "total alpha activity pc/kgram wet tissue	Mean "total alpha activity" pc/kgram wet tissue	% Po-210 excess	% Uranium series activity	% Thorium series activity
Seal	2	64- 110	86	98%		2%
Cattle	5	12- 57	30	N.D.	83%	17%
Buck	5	11- 31	18	N.D.	64%	36%
Pig	11	7- 23	15	N.D.	72%	28%
Sheep	5	3- 20	10	N.D.	52%	48%
Dog	4	4- 14	9	N.D.	~100%	
Baboon	1	-	8	N.D.	75%	25%
Horse	2	6- 8	7	N.D.	69%	31%
Donkey	2	4- 8	6	N.D.		~100%
Man	5	0- 10	5	N.D.	38%	62%

Table 10

Dog tissue analysis

Species	No. of samples	Range of "total alpha activity" pc/kgram wet tissue	Mean "total alpha activity" pc/kgram wet tissue	% Po-210 excess	% Uranium series activity	% Thorium series activity
Bone	4	780-1100	910	N.D.	60%	40%
Liver	4	53- 120	83	N.D.	64%	36%
Kidney	4	59- 93	73	N.D.	59%	41%
Heart	4	31- 80	55	N.D.	88%	12%
Lung	2	50- 50	50	N.D.	63%	37%
Flesh	3	39- 46	43	N.D.	70½	30%
Testes	2	34- 51	42	N.D.	80%	20%
Blood	4	4- 14	9	N.D.	~100%	N.D.

The following immediate conclusions can be drawn from these results:

(i) Blood and liver show much lower levels of alpha activity than bone. From the tissue analysis of dogs it appears that soft tissues in general have lower levels of alpha activity than bone. This is of course in agreement with the well established conclusions of earlier workers for humans e.g. Mayneord et al.(2). Bone tissue comprises a considerable propotion of body weight and thus most of the alpha activity in animal bodies lies in the bone. Bone activity levels may therefore be used as a basis for comparison of activity levels over different species.

(ii) An important exception to the above are the species from the marine environment e.g. fish and seals. These species tend to accumulate excess Po-210 in the liver.

(iii) Using bone alpha activity levels as a basis for comparison the highest level of activity is found in herbivores. Carnivores have a significantly lower level of activity and the omnivores man and pig show very low levels of activity.

(iv) There is a considerable range in the activity levels of the members of a single species.

(v) Po-210 is significantly in excess of its precursor Pb-210 in herbivores. The excess Po-210 further constitutes an appreciable fraction of the total alpha activity in herbivores.

These conclusions will be discussed in detail in the next chapter. Further data will be presented and in particular the alpha spectra for various samples will be displayed. A critical examination of the technique will also be undertaken in this next chapter.

Chapter 5

DISCUSSION

5.1 Critical Examination of Technique

It is pertinent at this point to examine the validity of the technique used and to see if the alpha activity as measured reflects the position in the "in vivo" state of the animals.

The routine freeze drying of samples followed the procedure developed in these laboratories by Hasson(34) In an extensive study he showed that contamination of the samples, in the course of this procedure, was highly unlikely. Furthermore, extensive data from a wide variety of samples measured in this laboratory have shown that the method of counting gives reproducible results and that heavy radionuclides in the sample are not adsorbed onto the ZnS phosphor (which would give anomalously high results) provided that the samples are stored in a desiccator as previously described. The question of inhomogeneity in the samples has been considered previously in this work and it has been shown that this problem does not occur for bone. Hasson has likewise shown that blood samples prepared by the freeze drying

technique are homogeneous.

The concentration factors for the soft tissues were determined individually for each sample and separate portions of the same sample gave comparable concentration factors when freeze dried separately. It is thus unlikely that a significant error arises here. For the bone samples however, a standard concentration factor of 1.3 for all bones was assumed. This was done because of the difficulty in freeing "wet bone" from all adhering tissue, and, in order to prevent this task from overwhelming the work of this thesis, the bones were boiled for twenty minutes to remove adhering tissues. The method of determining the concentration factor for bone, described in Chapter 2 Section 2.7 was considered to give the most accurate representation of the method of preparing bone samples but it is possible that the concentration factor is too large and hence the bone activity levels may be slightly higher than the calculations show {Woodard(57)} finds the average water content of bone to be 12.5% i.e. a concentration factor of 1.1}.

In calculating the various contributions to the "total alpha" activity it is assumed that the fall off in count rate accurately indicates the excess Po-210 content of the sample. This may however be incorrect

for the following reason. In the "in vivo" state a certain fraction of the gaseous Ra-226 daughter Ra-226 daughter radon is excreted from the body. When a specific sample is collected and packed for alpha counting the excreted radon is no longer lost. The radon decays through a series of daughters to Pb-210, and alpha emitter Po-210 builds in from the Po-210 (on a time scale of the Po-210 half life of 138 days). Thus there will be an increase in the count rate over time as this now "contained" radon excretion fraction decays to Pb-210 (this part of the chain attains equilibrium within the three week interval between sample packing and counting) and then builds in to Po-210. This increase in count rate is masked by the greater fall off in count rate due to excess Po-210. It does however mean that the observed fall off in count rate is less than that actually due to the excess Po-210 and hence the calculated excess Po-210 activity is lower than the correct value. The excretion fraction for the various species of animals is not known and no correction for this can be made. However, taking the radon excretion fraction of 0.7 for man as being generally true for animals it may be calculated that the error in excess Po-210 for "average" bovine bone is ∿13%. This is a small error as compared to the statistical

error in the excess Po-210 values.

The activity as calculated for the various samples is for "wet tissue" and the most significant difference between this calculated activity and the activity in the "in vivo" state is that due to radon excretion from the live animal. Essentially this means that the "uranium" series alpha-activity in the "in vivo" state is less than that calculated for the "wet tissue" state (since the method "contains" this previously excreted radon). Again, radon excretion fractions are not known for the various species and so no accurate correction from "wet tissue" to "in vivo" state can be made. However, if the radon excretion fraction of 0.7 for man is again assumed to be true for animals then a calculation for "average" bovine bone shows that the "total" alpha-activity for the bone as calculated in this work is 18% greater than the total-activity in vivo. This error is then also small.

It may be considered therefore that the technique as used is valid, but that if the "in vivo" state is to be reflected certain small corrections to the alpha activity calculated for the "wet tissue" state must be made. However these corrections must be applied to all samples and hence the values calculated for the "wet

tissue" state may be used as a basis for ordering the species according to alpha activity levels.

5.2 Comparison of results obtained with those of other workers

It is useful to compare the results of this work with those of other workers as reflected in Chapter 1.

(a) Animals

Table 1 chapter 1 gives Ra-226 and Th-228 activities in bones as measured by different authors and these specific isotope activities may be calculated for this work from the data in Appendix I. The Ra-226 specific activity may however only be accurately calculated for those samples which show a high activity as the "uranium" series for low activity samples contains any excess Po-210 present. Ra-226 and Th-228 are taken to be $\frac{1}{5}$ of total "uranium" and "thorium" series respectively (each has five alpha emitters below head member). Taking the concentration factor from "wet bone" to bone ash" to be 2.8 (see Chapter 1 section 1.2 the following comparative points may be noted (within the range of data)

(i) Good agreement with Ra-226 and Th-228 values calculated from data by Mayneord et al. for

cattle and sheep bone.

(ii) Good agreement with Ra-226 values for bovine bones as measured by "Ra emanation" technique {Lucas and Ferrante; Petrow } but poor agreement with Th-228 for bovine bones measured by Petrow.

(iii) Good agreement with Ra-226 in horse bone as measured using "Ra emanation" technique by Lucas and Ferrante.

(iv) While noting that for "uranium" series activity for low activity samples in the work may contain excess Po-210 contribution the Ra-226 values for seal and pig bone agree well with those given by Holtzman and Nelson and Rust respectively (both using "Ra emanation" techniques).

It is difficult to draw conclusions on the basis of these limited points of comparison but it does appear that the Ra-226 levels as measured by the "Ra emanation" technique and the "total alpha" counting technique show substantial agreement.

(b) Man

There is a large difference between the calculated value of 68 pc/kgram wet bone from the UNSCEAR report data (Chapter 1) and the mean value of 240 pc/kgram wet bone as measured in this work. The value calculated from data by Mayneord et al. viz. ~140 pc/kgram wet bone, using a "total alpha" counting technique is somewhat closer to the value obtained here.

The liver tissues studied also showed higher levels of activity than is generally assumed for soft tissues, although as is noted in Chapter 1, there is little available specific data on liver activities. The mean human blood activity from the present work is 5pc/kgram wet blood and it is interesting that Hasson(34) using the same technique in these laboratories finds a mean value of 89 pc/kgram wet blood. The possible reasons for this discrepancy are still under consideration

A possible reason for the discrepancy between the results this work and those of other workers may lie in the method of concentration of sample material used by the workers. Hasson(34) has shown that, for blood, heating to even low temperatures of the order of 100°C results in an appreciable loss of activity which cannot

be ascribed purely to loss of the volatile Po-210. The ashing techniques variously used by other authors concentrates the material by heating to temperatures between 250 and 600°C, and it is possible that a certain amount of activity is lost by this process of heating.

An alternative possibility is worth noting: if a higher level had been assumed for the background blank in the present work the value for human bone activity (and, indeed, all the low activity samples) would have been lowered to a more acceptable value, while not appreciably changing the level of the higher activity samples (e.g. herbivores). The background blank was chosen conservatively on the basis of the lowest count rate observed over a large range of materials. Many of the count rates observed in the present work are only fractionally above this assumed background and any raising of this level would mean that an unacceptable number of the samples studied would, on correction, show negative activity levels. It is felt then that the background blank count rate used is an upper limit of the true value, and as such, could not be the cause for the levels of activity in e.g. human bone being higher than that found by other workers.

A further possibility for the generally higher levels

of activity observed in species may be a high natural activity in the areas from which the samples originated. Overall, the samples studied from the various areas do not indicate any difference in natural activity levels between the areas from which samples studied in this work were drawn, but it may be that the southern part of Africa as a whole has a higher level of natural activity than that normally found elsewhere.

In conclusion here it may be noted that the ordering of species according to bone activity levels agrees substantially with a similar table (on fewer data and fewer species) by Mayneord et al.(2).

During the course of this experiment during which ~ 200 samples were counted a few of the samples studied gave anomalous results. These were possibly as a result of poor statistics, and have not been included in the summary. They are however discussed here for the sake of completeness.

(i) Three bone samples studied showed a fall off in count rate over time accompanied by a significant fall off in the pairs rate over time. The samples included a horse bone, donkey bone, and a sheep

bone. This fall off in the pairs rate over time is indicative of disequilibrium between Ra-228 and Th-228, and suggests that Th-228 is in excess of Ra-228, in contrast to work by Petrow(38) which suggests that Ra-228 is in excess of Th-228. Th-228 has a 1.9 year half life and it has been considered advisable to keep these samples under consideration for a further period before coming to any conclusion on the matter.

(ii) Three bone samples viz. two buck bones and one cow bone, showed an increase in count rate over time. This is indicative of unsupported Pb-210 in excess of Po-210. The two buck bones however were very oily when packed and the readings for them are possibly due to heavy radionuclides being adsorbed from an oily suspension onto the ZnS (considered in Chapter 2). The cow bone showed no such condition and is being examined further over a time period in order to check statistics.

5.3 Detailed discussion

(a) Marine Environment

The interesting feature of the fish and seals studied is the high concentration of excess Po-210 activity in the liver. This high concentration of excess Po-210 in the liver is not accompanied by a similar high concentration in bone and thus total activity in the livers of these creatures is generally higher than levels in bone. The mean liver/bone total activity ratios for fish and seal from the data presented in table are 1.1 and 3.8 respectively. Work done on Pb-210/Po-210 ratios in sea water and plankton by Shannon et al.(58) show that plankton tends to selectively "absorb" Po-210. Plankton forms the basis of the marine environment "food chain" and it seems that the high concentrations of Po-210 in the livers of these marine specie may be due to this selective "absorption" of Po-210 by the plankton. Fish gills and flesh have further been studied (Appendix and it appears that little Po-210 is present in these tissues, and, from the absence of Po-210 in the bones of both fish and seal it appears that there is some mechanism in the livers of these species by which ingested Po-210 is retained in the liver. This high concentration of Po-210 in liver (or digestive gland, as it often is in

the lower forms of life) unaccompanied by similar high concentrations in bone, or shell has been observed by Shannon(59) in other marine species, i.e. rock lobster, mussels. This contrasts radically with the case for herbivorous land animals in which excess Po-210 is found both in liver and bone.

The seal livers are further interesting in that the level of excess Po-210 concentration in the livers seems to bear some relation to their age. Since seals (at least up to the age studied) are continuously growing, the activity level is possibly related to quantity of food intake.

Excess Po-210 in seal livers

Sample and ∿ age	excess Po-210 activity pc/kgram wet tissue
Foetus 8 months (- 2 months)	85 ± 18
Pup ∿ 10 months	180 ± 30
Female ∿ 3 years	960 ± 50
Female ∿ 4 years pregnant	2320 ± 70

The foetus was removed from the ∿4 years old pregnant female seal, and on the time scale by which the others have been dated i.e. from birth, it is -2 months old. Alpha spectroscopy was performed on the livers of the ∿3 years old and ∿4 years old seals. Both spectra showed a Po-210 peak only, and that for the ∿4 years old seal is displayed here.

(b) Herbivores

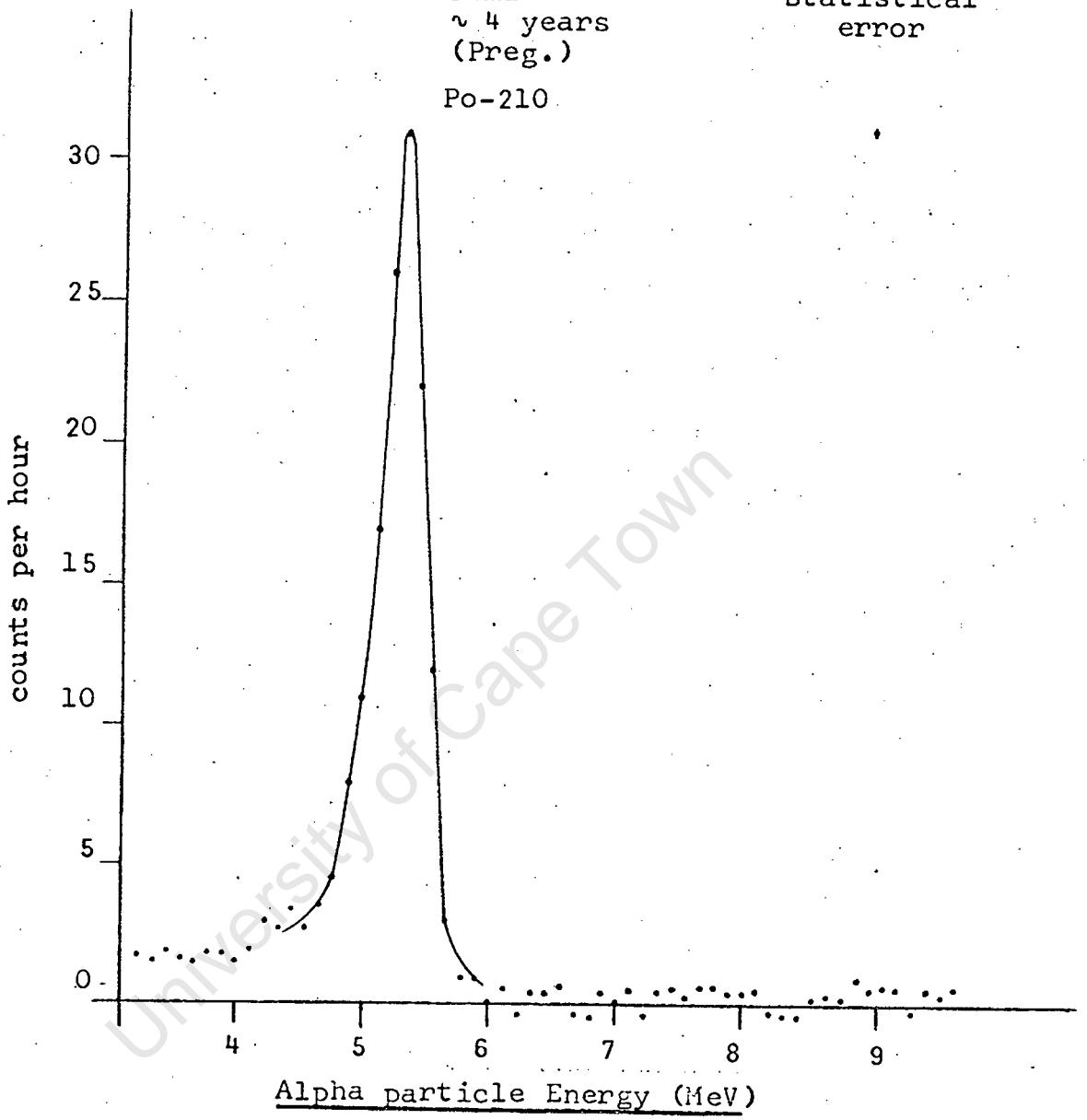
These species show an excess of Po-210 in both liver and bone and this "excess" Po-210 in certain herbivore tissues has also been found by Hill(It is presumed that the cause of this excess Po-210 derives ultimately from "natural fall out" of Pb-210 and Po-210. Work by Hill(60) shows that for certain grasses Pb-210 is in excess of Po-210, and this is confirmed by the work of Holtzman(61). The mechanism by which Po-210, which is not in excess in the main source of supply of food for these herbivores, becomes in excess in certain tissues of the animals presents a problem. This has been discussed by both Hill(60) and Holtzman(61) though both concentrate on the situation in man, which is a somewhat different problem due to the fact that man is an omnivore. It seems possible that there is a redistribution of Pb-210,

Figure 9

Seal liver - Female ~ 4 years (Preg.)

Po-210

Range of Statistical error



"Total" alpha counting results

Po-210 activity = 2300±70 pc/kgram wet tissue

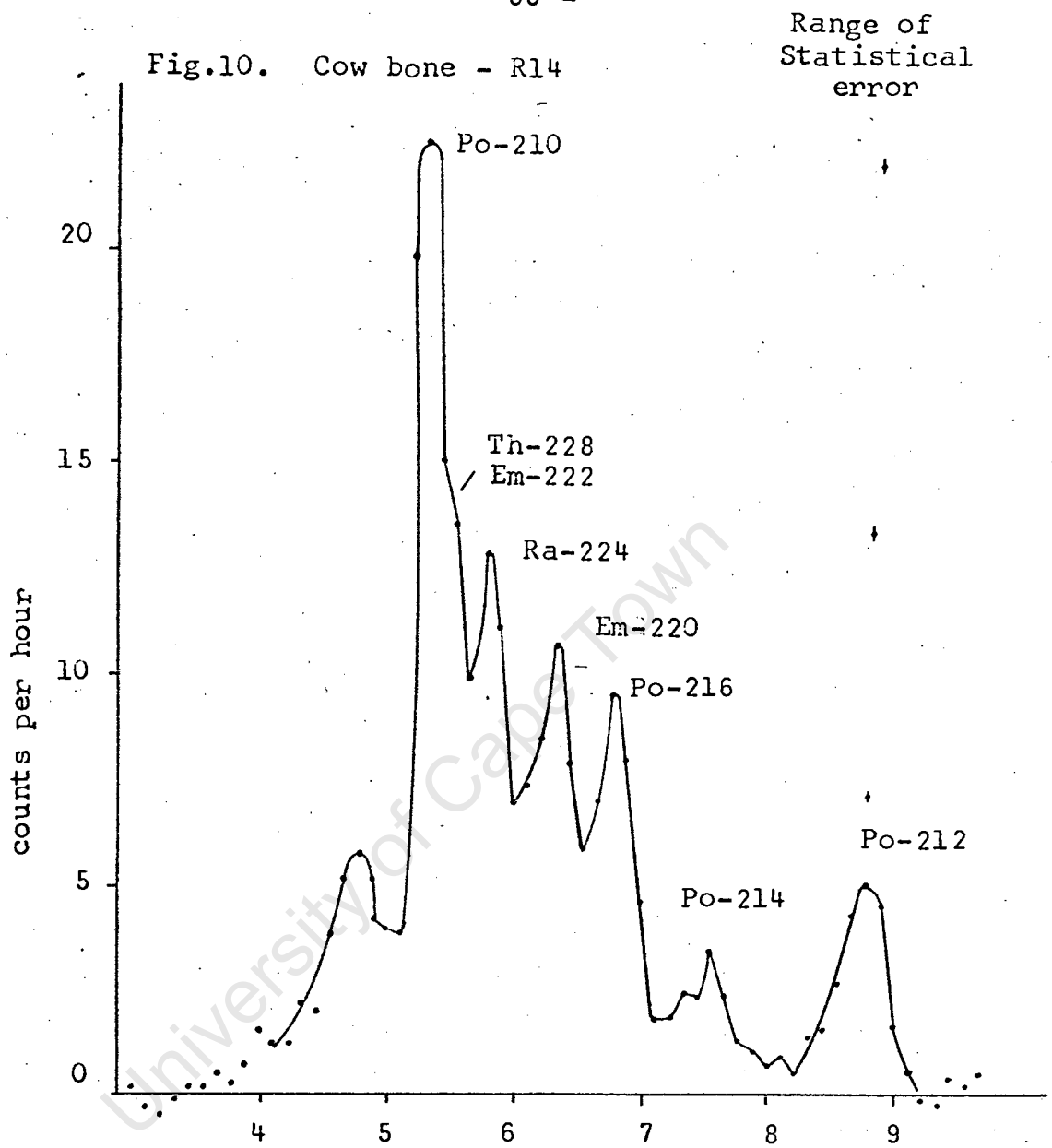
"Uranium" series activity = 110±80 pc/kgram wet tissue

"Thorium" series activity = 8±60 pc/kgram wet tissue

Po-210 in the body as is suggested by Holtzman(61) for man. Lead, however, is a toxic poison and is not readily excreted from the body once it is ingested into the blood system. The primary breakdown of foodstuffs, once it passes into the blood system, occurs in the liver, and the work done here indicates that in herbivores, Po-210 is in excess even at this point. It seems possible that redistribution occurs then at some point in the alimentary canal possibly by a selective absorption through the gut wall. It is difficult to make any assertions on this point, but it is clear that there is a need for further examination of the problem.

The alpha spectra for a sheep bone, cow bone, and reed buck liver are presented here. Peak identification shows the presence of the "uranium" and "thorium" series members as well as the excess Po-210 peak. In particular the Ra-226 and Th-228 daughters are clearly present. There is little data with which to compare these spectra but it is interesting that a spectrum of normal human liver ash by Hill(33) shows only a Po-210 peak and no "thorium" or "uranium" series daughters, whereas his spectra for human bone ash does show some indication of the presence of the "uranium" and "thorium" series daughters.

Fig.10. Cow bone - R14



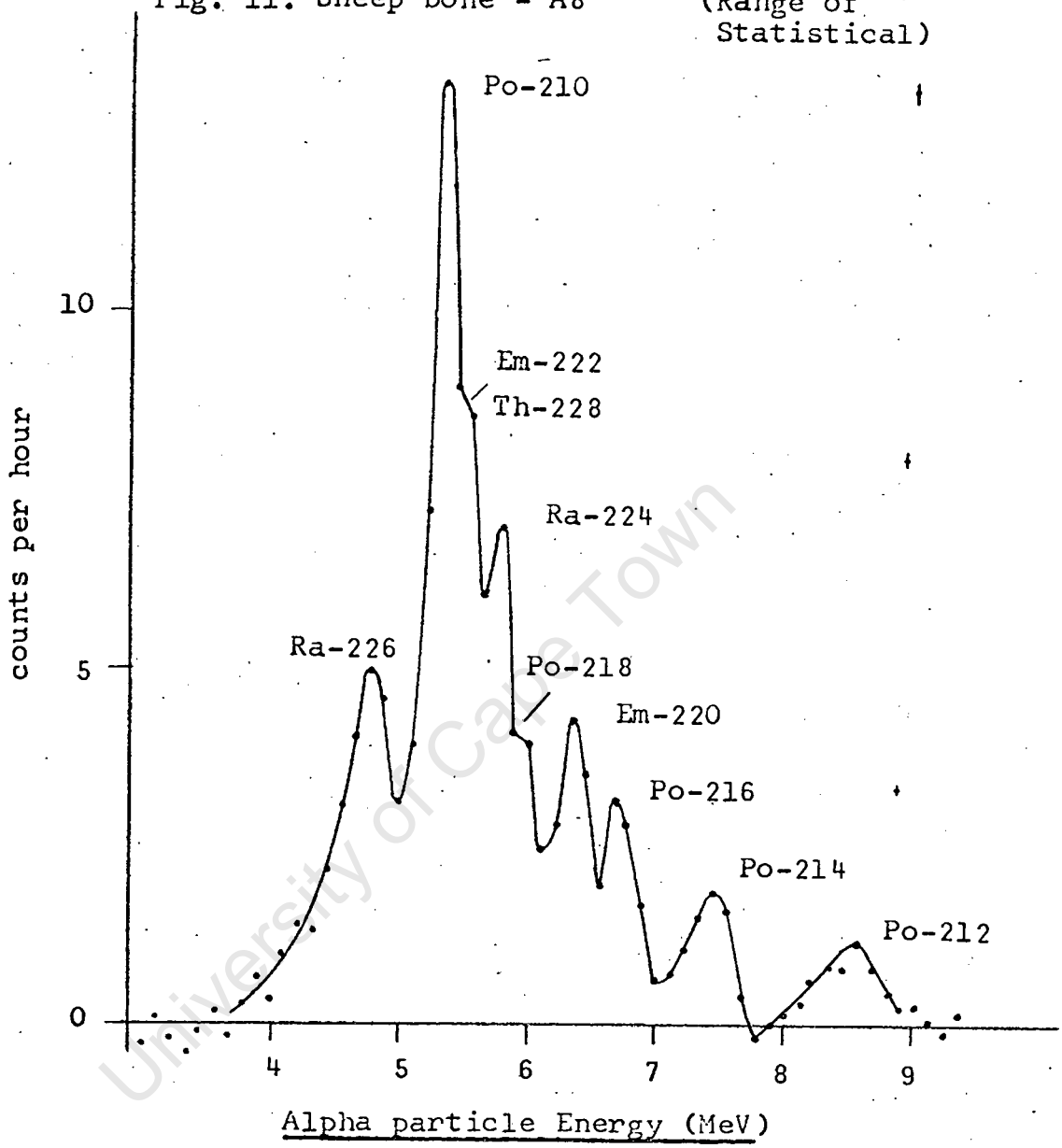
"Total" alpha counting results

Po-210 activity = 3200±700 pc/kgram wet bone

"Uranium" series activity = 600±750 pc/kgram wet bone

"Thorium" series activity = 2600±600 pc/kgram wet bone

Fig. 11. Sheep bone - A8



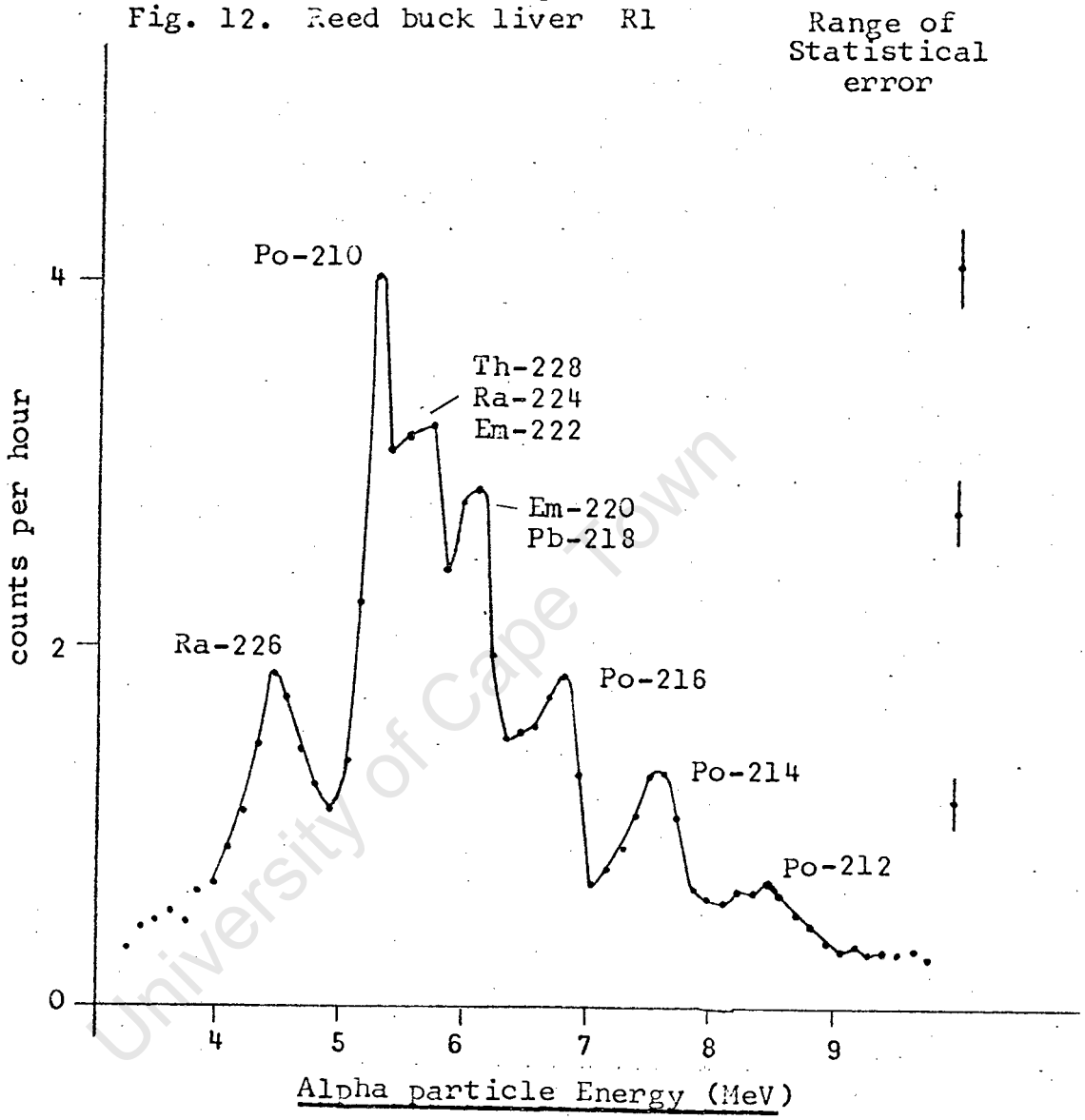
Total alpha counting results

Po-210 excess = 811.9±350.0 pc/kgram wet bone

"Uranium series" =1261.3±231.1 pc/kgram " "

"Thorium series" = 165.1±102.5 pc/kgram " "

Fig. 12. Reed buck liver R1



"Total" alpha counting results

Po-210 excess = 200±70 pc/kgram wet tissue

"Uranium" series activity = -50±80 pc/kgram wet tissue

"Thorium" series activity = 300±50 pc/kgram wet tissue

(c) Carnivores

The relatively low activity found in these samples is likely to be as a result of their diet, which is mainly the soft tissues of herbivores. Since most of the alpha activity in herbivores concentrates in the bones, the levels of activity found in carnivores, as ingested from the soft tissues of herbivores should be generally low, and this is found to be the case. There seems to be no marked distinction "activity level"-wise between the two major branches of carnivores studied, i.e. "cat" family and "dog" family.

(d) Omnivores

These specie show a very low level of alpha activity. Again this may be due to the feeding habits of these specie. Fruit and vegetables are shown by Mayneord et al.(2) to have a very low specific alpha activity, and the main meat contributant to the diet is flesh, which also has a low activity. A point of interest in examining the omnivores, is the known physiological similarities between pig and man {Bustad(62)}.

It is of interest to estimate the resulting radiation dose to human bone from the levels calculated here. For a 5 MeV alpha particle 1 pc/kgram uniformly distributed,

corresponds to a dose rate of ∿0.1 millirad per year {Hill(33)} and, assuming a relative biological efficiency (R.B.E.) of 10 for naturally occurring alpha's this gives a dose of 1 millirem per year (i.e. expressions in pc/kgram are equivalent to mrem/year). On this basis the average does to human bone, using the mean value calculated here, is 230 millrem per year. This result is greatly in excess of values given by Eisenbud(63) for the total dose rate to bone from all external sources viz. 100 millrem/year. [It should here be noted that the value calculated is from that for "wet bone". The "in vivo" state as discussed previously will be slightly lower and the value of 230 millrem per year is thus an upper limit of the dose rate.]

One may further compare the dose rates to human tissues on the basis of the present work with the dose to human tissues from the naturally occurring K-40 which is often assumed to be the major source of internal irradiation in humans. Spiers(3) gives the dose rate to human bone and soft tissues, from K-40, as 6 mrad/year and 19 mrad/year respectively. For an R.B.E. of one for β and γ particles this corresponds to 6 mrem/year for bone and 18 mrem/year for soft tissues. From the date in this work (using the approximation described above) the dose

to human tissues from alpha particles is 230 mrem/year for bone and 26 mrem/year for liver, which both are in excess of the K-40 levels. [Similar levels of K-40 in bovines are assumed by Lucas and Ferrante and for these animals (and hence in herbivores in general) the alpha particle dose calculated from the work of this thesis is greatly in excess of the dose due to K-40.]

A general conclusion may be drawn: that the alpha activity levels in the various classes of species are related to their feeding habits. In an attempt to correlate further feeding habits and alpha activity levels several pigs were obtained from one of the local agricultural colleges, these pigs being separated into two classes each of which had a different diet. Unfortunately the experiment was designed for purposes other than radioactivity measurements and samples of the food showed no significant variation in alpha activity between the two diets. No comparison between similar animals or different diets could therefore be made. However the alpha activity in the pig food was measured to be about 0.6 pc/gram food, which for an intake of ~3200

grams per day results in a nett-intake of 1920 pc per day. From data on general foodstuffs by Mayneord et al. (Table 3)(64) an approximate estimate of the average alpha activity of the general range of foodstuffs consumed by humans is 0.1 pc/gram, which for a human consuming ~2000 grams of food per day means an alpha activity intake of 200 pc per day. Work done by Jones(65) in these laboratories, on foodstuffs, gives results which also indicate a daily human intake of 200 pc of alpha activity. This is higher than the minimum amount of 5 pc/day, quoted by Mayneord et al.(64)in the same paper for humans, but it can be seen that this is still very much lower than the daily alpha activity intake of the pigs studied. Pigs nonetheless show an equal or lower level of activity than man (see Table 6) and it is possible that there is some discriminatory mechanism present which rejects or excretes much of the alpha activity intake of pigs. Although the data given here supports in a general way the supposition that the inherent alpha-activity of a species is determined mainly by the activity of its food intake, it is clear that the possibility of this supposition being modified by physiological mechanisms cannot be excluded.

Chapter 6

SUMMARY AND PROPOSALS FOR FUTURE WORK IN THE FIELD

The work of this thesis has shown that there is a large variation in the levels of alpha radioactivity, not only between different species, but also between members of a single specie. Bone tissue activity levels have been found to be a suitable basis for comparing the activity levels of different animals and it has been shown that the class of animals, herbivores, show the highest levels of alpha activity, and that the livers and bones of these herbivores contain large activity contributions from unsupported Po-210. The carnivores show a significantly lower level of activity than herbivores and man and pig (omnivores) show lowest levels of activity. By considering the feeding habits of these three main classes of animals it can reasonably be assumed that the levels of activity in the animals are in a large measure dependant on diet. However, physiological mechanisms may play a large part in determining the levels of activity in the animal, and this is well illustrated in marine environment where Po-210 tends to accumulate in

the liver.

An examination of the experimental technique used shows that while results may be slightly high, since activities are measured for the "wet tissue" rather than the "in vivo" state, the technique is in general valid for the range of tissues studied. A certain discrepancy is noted between the results obtained here and those of other workers (especially on human tissues). It is suggested that this discrepancy may be due to "ashing" techniques used by other workers, though it is considered possible that the southern part of Africa as a whole has a higher level of natural radioactivity than "normal".

Finally a list of desirable projects which follow from the work of this thesis are:-

(i) A study of Po-210 in the "food chain" of the marine environment and a closer examination of the livers of marine species.

(ii) Examination of the Pb-210, Po-210 disequilibrium situation in herbivores, especially with respect to explaining the presence of Po-210 in excess of Po-210 in these animals.

(iii) The performing of controlled feeding experiments on various animal species to study the possible correlation between the intake of alpha activity

and body alpha activity levels.

(iv) Determination of the radon excretion fraction for major species of animals in order that "in vivo" levels of activity may be calculated.

(v) Further examination into the possibility of alpha activity being lost during the "ashing" processes in common use.

(vi) Development of a method of sample concentration which does not involve the use of heat yet gives concentration factors of the same order of magnitude as the "ashing" techniques (concentration factors of 30-50) [The feasibility of using radio frequency ashing techniques is at present being examined in these laboratories.]

Appendix I

TABLES OF RAW DATA

The specific origin of samples studied is as follows:-

Humans, dog, fish, seals and herbivores prefixed A

Cape Town and vicinity

Herbivore samples prefixed H

Hermanus, Cape Province

Carnivore samples prefixed K

Kruger National Park, Transvaal

Samples prefixed R

Beitbridge and Fort Victoria areas, Rhodesia

Pig samples without a prefix

Elsenburg, Cape Province

N.B. Where Po-210 excess activity levels are not indicated this means that no such Po-210 excess was detectable within the limits of counting statistics.

Table 12

Cattle (bovine)

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
R14	6.0		19.0±7.4	0± 2.9	19.0± 7.9
A4	5.5		56.5±9.6	0.8± 3.4	57.3±10.2
A5	6.0		31.7±8.3	12.4± 7.6	44.1±11.3
H3	6.2		17.1±7.1	1.4± 3.5	18.5± 7.9
H4	5.9		0.7±7.4	11.2±12.7	11.9±14.7
Arith. mean			25.0	5.2	30.2
Liver					
H3	4.0	110.3 ±33.3	-50.0±44.8	123.7±30.4	184.0±63.6
H4	3.6	40.6 ±38.3	-19.7±49.0	118.1±31.9	139.0±69.9
Arith.mean		75.5	-34.9	120.9	161.5
Bone					
R14		3151.0±674.5	602.4±745.5	2641.0±569.2	6394.4±1155.3
A4		1271.0±371.2	2971.3±399.1	2038.1±285.6	6280.4±615.3
A5		2313.6±357.6	2658.3±394.6	2026.0±285.1	6997.9±604.0
Arith. mean		2245.2	2077.3	2235.0	6557.6

Table 13

Sheep

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
A1	6.1		1.1± 2.3	2.2±3.0	3.3± 3.8
A8	6.2		9.6± 6.7	10.5±4.7	20.1± 8.2
A7	6.3		1.3±10.6	5.5±6.9	6.8±12.6
H1	8.6		6.7± 4.2	2.6±2.7	9.3± 5.0
H2	7.3		6.9± 4.6	3.0±3.0	9.9± 5.5
Arith. mean			5.1	4.8	9.9
Liver					
H1	3.9		51.6± 9.2	1.1± 4.8	52.7±10.4
H2	3.3		38.6±19.8	40.9±12.7	79.5±23.5
Arith mean			45.1	21.0	66.1
Bone					
A8		811.9±350.0	1261.3±231.1	165.1±102.5	2238.3±431.8
H1		2621.1±361.5	707.2±301.3	2442.0±199.2	5770.3±511.0
Arith. mean		1716.5	984.2	1303.6	4004.3

Table 14

Horse

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
A10	5.2		4.7±7.4	3.4±4.0	8.1± 8.4
A12	3.9		5.2±5.77	1.1±4.0	6.3±10.0
Arith. mean			5.0	2.3	7.2
Bone					
A12		1539.0±485.2	91.2±147.9	313.1±81.4	1943.3±513.7

Donkey

Blood					
A9	5.5		-1.9±9.9	9.5±5.1	7.6±11.1
A11	6.2		0±8.9	4.2±5.6	4.2±10.5
Arith. mean			-1.0	6.9	5.9
Bone					
A11		779.7±171.2	368.2±198.5	711.9±177.7	1859.8±409.6

Table 15

Buck

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
Grysbok	17.5		-.9± 6.3	16.3±5.5	15.4± 8.4
Impala					
R4	7.9		27.8± 7.0	3.3±2.2	31.1± 7.3
Reedbuck					
R1	6.3		23.4± 8.0	-2.8±2.8	20.6± 8.5
R3	8.0		-1.7±10.2	13.0±7.5	11.3±12.7
Duiker					
R5	4.9		9.8±12.3	3.5±7.0	13.3±14.2
Arith. mean			11.7	6.7	18.3
Liver					
Impala					
R4	4.6	*	38.1±11.0	3.8± 3.8	41.9± 11.6
Reedbuck					
R1	4.5	204.5±67.2	-47.6±81.1	302.8±48.4	459.7±115.9
R3	4.8	56.1± 9.4	9.9±12.3	12.7±10.9	78.7± 18.9
Duiker					
R5	4.6	*	21.3±10.7	1.9± 5.7	23.2± 12.1
Arith. mean		65.2	5.4	80.3	150.9
Bone					
R3 Reedbuck		517.7± 119.0	483.5±602.3	609.1±102.5	1610.3±622.4

Table 16

Fishbone

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Red Roman		*	176.4±45.5	113.8± 36.2	290.2± 58.1
Mussel-cracker		*	109.1±29.0	22.8± 20.2	131.9± 35.3
Black Parrot		*	323.1±72.6	264.7± 60.5	587.8± 94.5
Bream		549.9±183.3	0±414.7	28.5± 34.2	578.4±454.7
Dassie		*	17.6± 16.6	-11.4± 8.1	6.2± 18.5
Butterfish		*	419.2± 88.6	449.7± 74.5	868.9±115.8
Red Stump-nose		*	285.2± 28.9	2.9± 16.1	288.1± 33.1
John Brown		*	7676.5±216.6	187.9±181.2	7844.4±282.3
Galjoen		*	778.2± 65.9	156.5± 50.3	934.7± 82.9
Snoek		*	125.1± 29.0	14.3± 20.2	139.4± 35.3
Arith. mean		not calc.	990.9	123.0	1168.9

Fish Livers

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Red Roman	2.9	308.4±124.9	141.8± 62.5	12.0± 38.9	462.2±145.1
Mussel-cracker	3.6	0±161.3	131.1± 38.8	26.5± 28.9	157.6± 48.4
Black Parrot	2.6	318.9±159.5	180.9±121.9	46.7± 36.7	546.5±243.9
Bream	2.2	0±221.6	418.6± 49.5	90.7± 35.5	499.3± 60.9
Dassie	3.9	709.4±111.4	29.6± 99.0	62.3± 37.8	801.3±153.8
Butter-fish	2.1	1242.7±231.5	519.2±183.6	210.8± 74.4	1972.7±304.7
Red Stump-nose	2.0	1317.0±188.1	-98.9± 36.0	108.5± 56.4	1326.8±199.6
John Brown	2.0	1006.8±223.7	679.1±198.2	56.4± 60.8	1742.3±305.0
Galjoen	2.0	3524.0±298.3	365.4±280.3	134.5±112.8	4023.9±424.6
Snoek	2.0	616.5±198.2	602.7±174.3	13.0± 47.7	1232.2±268.2
Arith. mean		904.4	297.0	76.1	1277.5

Fish Flesh

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Red Roman	4.3		4.1± 9.6	9.1± 7.1	13.2±11.9
Mussel-cracker	4.3		35.6±11.5	8.1± 8.6	43.7±14.4
Black Parrot	3.3		23.7±11.4	5.3± 7.5	29.0±136.
Bream	4.2		38.6±10.1	7.2± 7.1	45.8±12.3
Dassie	4.5		44.8± 7.2	0± 4.1	44.8± 8.3
Butter-fish	4.3		11.7± 6.4	-4.0± 2.8	7.7± 7.0
Red Stump-nose	4.0		5.3± 7.9	2.2± 4.6	7.5± 9.1
John Brown	4.0		15.6±12.4	16.3± 9.9	31.9±15.9
Galjoen	4.0		27.2±15.6	18.5± 9.9	45.7±18.5
Snoek	4.0		69.6±24.1	87.9±19.9	157.5±31.3
Arith. mean			27.6	15.1	42.7

Fish Gills

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Red Roman	3.2	*	210.4±35.2	97.7±25.9	308.1± 43.7
Musselcracker	6.6	384.0±50.7	-53.7±39.9	2.6± 9.2	332.9± 65.2
Black Parrot	3.2	*	27.1±31.5	126.1±26.9	153.3± 41.4
Bream	5.2	228.3±76.1	161.2±59.9	0±10.0	389.5± 97.4
Dassie	3.1	*	168.7±23.9	33.6±17.8	202.3± 29.8
Butterfish	2.1	*	252.1±47.5	177.7±39.0	429.8± 61.5
Red Stumpnose	3.0	*	180.0±15.1	-4.4± 7.2	175.6± 16.7
John Brown	3.0	*	805.8±32.1	26.0±22.5	831.8± 39.2
Galjoen	2.5	296.1±98.7	10.2±85.8	34.7±24.3	341.0±133.0
Snoek	3.0	*	119.8±13.2	-1.5± 6.2	118.3± 14.6
Arith. mean		90.8	188.2	49.3	328.3

Table 17

Dog

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
F	5.6		6.0±5.7	-1.6±2.2	4.4±6.1
D	6.0		13.9±8.5	-2.9±2.9	11.0±9.0
E	4.8		10.5±4.8	-3.6±3.6	6.9±6.0
A	8.4		14.4±3.3	0±2.1	14.4±3.9
Arith. mean			11.2	-2.0	9.2
Liver					
D	3.6		35.6±32.8	79.6±26.3	115.2±42.3
B	3.3		66.7±18.0	10.5±10.5	77.3±20.8
A	3.9		76.0±15.2	8.9± 8.9	84.9±17.6
C	3.8		32.1±15.4	20.6±11.5	52.7±19.2
Arith mean			52.6	29.9	82.5
Heart					
D	3.5		75.1±14.3	5.0± 5.0	80.1±15.1
B	4.4		32.8± 9.2	-2.0± 4.0	30.8±10.0
A	4.8		42.6± 9.8	-3.6± 3.6	39.0±10.4
C	3.6		43.6±16.1	26.5±11.9	70.1±20.0
Arith. mean			48.5	6.5	55

Dog (continued)

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Kidney					
D	3.8		28.7±28.7	63.9±23.0	92.6±36.8
B	3.6		22.5±20.5	48.2±14.5	70.7±25.1
A	3.6		68.0±17.0	0± 4.8	68.0±17.7
C	4.6		51.4±10.8	7.5± 7.5	58.9±13.1
Arith. mean			42.7	29.9	72.6
Lung					
B	4.5		32.4±13.9	17.4±9.7	49.8±16.9
C	5.4		31.0±10.9	19.3±8.1	50.3±13.6
Arith. mean			31.7	18.4	50.1
Testes					
B	5.8		27.5± 9.6	6.0±6.0	33.5±11.3
A	5.1		40.7±11.8	10.2±6.8	50.9±13.6
Arith. mean			34.1	8.1	42.2
Flesh					
D	4.8		13.2±13.1	25.3±9.1	38.5±16.0
B	3.8		41.7±12.9	4.6±6.9	46.3±14.6
A	3.7		35.9±15.1	9.4±9.4	45.3±17.8
Arith. mean			30.3	13.1	43.4

Dog (continued)

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Bone					
D			150.3±120.3	626.2±112.8	776.5±164.9
B			627.9± 97.9	233.4± 79.6	861.3±126.2
A			790.0±102.7	341.5± 78.5	1131.5±129.3
C			617.2± 98.8	238.6± 78.7	855.8±126.3
Arith. mean			546.4	360.0	906.3

Table 18

Bones

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Cheetah					
K1			10.3±79.1	259.0±62.2	269.3±100.6
Lion					
K2			185.9±57.6	103.6±37.3	289.5±68.6
Hyena					
K3			246.1±100.9	429.2±81.5	675.3±129.7
Jackal					
K4			154.8±108.3	562.4±90.0	717.2±140.8
Dove					
B1			257.3±95.2	325.9±88.2	583.2±130.0
Cormorant					
B2		646.7±109.0	23.3±96.4	130.9±51.2	800.9±154.3
Chicken					
B3			303.5±133.5	96.1±40.4	399.6±139.5
Baboon					
R2			148.0±99.2	239.1±81.3	387.1±128.3

Table 19

Bones (Aged)

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Buck					
Impala					
R7			-133.2± 75.9	227.7± 63.8	158.3± 99.2
R9			124.5± 48.5	48.5± 34.4	173.0± 59.5
Eland					
R18			533.0±117.3	797.7±199.4	1330.7±231.3
Kudu					
R8			190.2± 57.0	96.8± 40.7	287.0± 70.0
Arith. mean			178.6	292.7	471.3
Zebra					
R17			244.7±51.4	45.5±34.1	300.2± 61.7
R19			219.0±89.8	142.3±52.7	361.3±104.1
R20			194.2±48.6	125.2±35.1	319.4± 59.9
Arith. mean			219.3	104.3	323.6
Elephant					
R21			283.1±101.9	411.5±98.8	694.6±141.9
R11			6.8± 59.8	182.2±47.4	189.0±76.3
R10			104.1± 55.2	153.7±41.5	257.8± 69.1
Arith. mean			131.8	249.1	380.2

Bones (Aged) (continued)

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Jackal					
R22			100.7±68.5	113.8±51.2	214.5±85.5
Lion					
R16			482.8±130.4	660.3±125.5	1143.1±181.0
Leopard					
R14			258.3±54.2	159.4±41.4	417.7±68.2
Wild dog					
R15			272.8±150.0	421.2±75.8	694.0±168.0
R12			190.5± 91.4	449.7±76.4	640.2±119.1
Arith. mean			231.7	435.5	667.2

Table 20

Seal

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
S1	5.6	65.3±22.7	-10.2±13.2	9.3±6.2	64.4±27.0
S2	7.0	103.2±12.6	0.7±12.8	3.7±5.0	107.6±18.6
Arith. mean		84.3	-4.8	6.5	86.0
Liver					
Female ~ 3 years	3.2	963.8±46.6	48.2±49.2	-16.3±16.3	995.7±69.7
Pup ~ 10 months	3.1	175.8±29.3	-36.2±35.8	44.8±22.4	184.4±51.4
Foetus 8 months	5.2	84.6±17.7	-2.2±17.2	6.7± 6.7	89.1±25.6
Female preg. ~ 4 years	3.1	2316.0±69.8	107.6±78.9	8.4±58.8	2432.0±120.6
Arith. mean		885.1	29.4	10.9	925.3

Seal (continued)

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Bone					
Pup ~ 10 months			19.3±64.7	318.8±54.3	338.1±84.4
Foetus 8 months			62.4±58.6	233.4±48.3	295.8±75.9
Female preg. ~ 4 years			147.4±26.5	19.9±16.1	167.3±31.0
Seal S1			121.5±33.3	45.5±22.8	167.0±40.4
Arith. mean			87.7	154.4	242.1

Table 21

Man

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
0723	5.7		7.2±6.9	3.0±4.6	10.2± 8.3
0721	6.1		0±5.4	0±2.8	0± 6.1
0717	5.7		-5.5±8.2	6.1±6.1	0.6±10.2
0715	6.4		11.4±5.1	-2.7±2.7	8.7± 5.8
0782	6.4		-3.1±7.3	9.5±5.4	6.4± 9.1
Arith. mean			2.0	3.2	5.2
Liver					
F38	4.2		24.8±11.0	16.5±8.0	43.3±13.6
M37	4.6		19.1±6.8	0±4.0	19.1± 7.9
M61	3.6		39.7±6.6	-4.8±3.4	34.9± 7.4
M46	4.2		14.0±8.8	6.2±5.8	20.2±10.5
M26	4.0		16.2±6.9	4.3±6.1	20.5± 9.2
F20	4.2		10.5±6.4	0±4.3	10.5± 7.7
F72	3.7		26.9±7.4	0±5.0	26.9± 8.9
M2	2.9		15.2±9.4	0±6.4	15.2±11.4
M14	4.4		19.0±8.4	13.8±5.6	32.8±10.0
M57	4.4		35.3±7.7	3.9±2.8	39.2± 8.2
Arith. mean			22.3	4.0	26.3

Man (continued)

Sample Number	Conc. factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Bone					
F38			186.4±44.7	28.5±22.8	214.9±50.2
M37			244.8±44.1	62.6±28.2	307.4±52.3
M61			133.7±55.3	116.7±34.3	250.4±65.0
M46			60.3±44.2	227.7±37.3	288.0±57.8
M26			-16.4±60.0	273.3±50.9	256.9±78.7
F20			113.9±35.8	86.0±29.1	199.9±46.1
M57			125.4±33.8	51.2±22.5	176.6±40.6
Arith. mean			121.2	120.9	242.1

Table 22

Pig

Sample Number	Conc. Factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Blood					
A2	6.7		0±7.6	6.5±3.0	6.5± 8.2
A3	5.9		7.2±3.2	10.3±9.9	17.5±10.4
H5	6.5		3.7±6.7	8.0±4.9	11.7± 8.3
H6	6.1		10.9±4.6	0±2.0	10.9± 5.0
12	7.1		17.3±8.6	2.5±5.0	19.8± 9.9
13	6.0		8.5±7.7	11.6±5.8	20.1± 9.6
9	6.4		26.0±7.8	-2.7±2.7	23.3± 8.3
2	5.6		10.7±9.0	-3.1±3.1	7.6± 9.5
8	6.3		10.4±7.4	4.1±5.5	14.5± 9.2
14	7.0		11.8±5.5	5.0±3.8	16.8± 6.7
6	6.9		8.5±7.9	3.8±6.3	12.3±10.1
Arith. mean			10.5	4.2	14.6

Pig (continued)

Sample Number	Conc. Factor	Po-210 activity pc/kgram	"Uranium" series α-activity pc/kgram	"Thorium" series α-activity pc/kgram	Total α-activity pc/kgram
Liver					
H5	4.1		23.1± 8.5	5.3± 5.7	28.4±10.2
H6	3.7		28.4±12.7	16.4±10.0	44.8±16.2
9	3.6		-4.3±45.7	156.7±39.2	152.4±60.2
2	3.2		56.7±18.9	5.4±10.8	62.1±21.8
12	3.7		129.3±37.5	77.6±30.3	206.9±48.2
13	3.2		21.0±10.3	0± 5.4	21.0±11.6
8	3.4		0±13.7	20.4±10.2	20.4±17.1
14	3.5		15.1±13.3	12.4± 3.3	27.5±13.7
6	3.4		15.1±19.5	20.4±15.3	35.5±24.8
Arith. mean			31.6	35.0	66.6
Bones					
A2			112.3±44.9	74.0±28.1	186.3±53.0
A3			93.1±32.6	11.4±17.1	104.5±36.8
12			150.1±28.5	-11.4±11.4	138.7±30.7
13			75.0±45.8	39.8±34.2	114.8±60.7
9			29.5±56.6	85.3±45.2	114.8±72.4
2			101.8±33.6	11.4±22.8	113.2±40.6
6			113.0±56.5	79.7±39.9	192.7±69.2
Arith. mean			96.4	41.5	137.9

Appendix II

Sample Calculation - Donkey bone A-11

Hasson apparatus,

Sample area 81 sq cms

Date of sample collection 30/3/68

Date of first counting period 7/5/68

Date of second counting period 5/1/69

	7/5/68	5/1/69
Total Alpha Count	988	788
No. of Alpha Pairs	31	25
Counting Time	23.7 hours	24.0 hours
Observed count rate (N)	$\frac{988 \pm \sqrt{988}}{23.7}$	$\frac{788 \pm \sqrt{788}}{24}$
	= 41.68 ± 1.33 c/hr	= 32.83 ± 1.17 c/hr
Observed pairs rate	$\frac{31 \pm \sqrt{31}}{23.7}$	$\frac{25 \pm \sqrt{25}}{24}$
	= 1.31 ± 0.24 p/hr	= 1.04 ± 0.21 p/hr
Electromechanical register dead time (τ)	0.45 secs	0.37 secs

Pairs Rate Corrections

	7/5/68	5/1/69
a) Background (blank + noise)	0.02 ± 0.02 p/hr	0.02 ± 0.02 p/hr
b) Spurious pairs = $N^2\tau$	0.22 p/hr	0.11 p/hr
c) Multiplicative factor (f)	1.17	1.26
∴ Pairs rate	1.25 ± 0.24 p/hr	1.15 ± 0.21 p/hr

Count Rate Corrections

Background (noise + blank)	2.40 ± 0.26 c/hr	2.40 ± 0.26 c/hr
∴ Count rate	39.28 ± 1.36 c/hr	30.43 ± 1.20 c/hr

Fall off in count rate 7/5/68 - 5/1/69 8.85 ± 1.81 c/hr= ΔC

Fall off in pairs rate 7/5/68 - 5/1/69 0.10 ± 0.32 p/m

The fall off in count rate is significant whereas the fall off in pairs rate is not. This in indicative of an excess of Po-210 and is treated accordingly.

Time interval 7/5/68 - 5/1/69 is 243 days = Δt_1

Time interval 30/3/68- 7/5/69 is 38 days = Δt_2

Po-210 count rate contribution at line of first counting

$$= \frac{\Delta C}{1-e^{-\lambda \Delta t_1}} = 12.59 \pm 2.57 \text{ c/hr}$$

Po-210 count rate contribution at time of collection

$$C_{Po\text{-}210} = \frac{\Delta C}{1-e^{-\lambda \Delta t_1}} \cdot \frac{1}{e^{-\lambda \Delta t_2}} = 15.22 \pm 3.11 \text{ c/hr}$$

For an activity which is entirely "Thorium series" the count/pairs ratio is 14.6 (see appendix III).

∴ "Uranium series" count contribution at time of first counting is

[count rate at first counting - 14.6 × pairs rate - Po-210 count contributions at time of first counting]

∴ Cur ("Uranium series" count contribution) = 8.44 ± 4.55 c/hr

Thus we have

$C_{Po\text{-}210}$ = 15.22 ± 3.11 c/hr

Cur = 8.44 ± 4.55 c/hr

C_p = 1.25 ± 0.24 p/hr

and for bone $\frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} = 4.0$

$k = 1.13$

$k_p = 1.11$

concentration factor for bone = 1.3

For Po-210

From theory equation

$$C_{Po-210} = 8 \times 10^{-5}\ kA\ \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} \times 3.85\ N_E$$

where N_E disintegrations/gram/hour

Substituting for known values, changing N_E to pc/kgram and dividing by concentration factor, we get

Po-210 activity = 779.7 ± 171.2 pc/kgram wet bone

For "Uranium series"

From theory equation

$$Cur = 8 \times 10^{-5}\ kA\ \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} \times 4.52\ Nur$$

and treating as above

"Uranium series" activity = 368.2 ± 198.5 pc/kgram wet bone.

For "Thorium series"

From theory equation

$$C_p = 2.82 \times 10^{-5} \times k_p \times A \times \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} \quad N_{Th}$$

and hence

"Thorium series" activity = 711.9 ± 177.7 pc/kgram wet bone.

The "total alpha activity" which is the sum of the contributions above is thus:

1859.8 ± 409.6 pc/kgram wet bone

For soft tissues $\frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} = 3.4$

The concentration factor was determined for each individual soft tissue sample.

An anticoagulant was added to the blood samples at time of collection and hence a correction for this was applied to the blood samples. The anticoagulant solution used was a citrate solution as used by the Blood Transfusion unit of Groote Schuur Hospital. The solution is prepared by dissolving 69.5 grams Tri-sodium citrate, 25.0 grams Citric acid and 69.0 Dextrose in 5 litres of distilled water. The anticoagulant is added to the fresh blood in ratio 1 : 4.

A sample of the anticoagulant was freeze dried and counted, giving 2.5 c/hr and 0.01 p/hr above background. Taking the density of both blood and anticoagulant solution to be 1 the following corrections were applied to the blood

samples.

Count rate correction

$$C' = \frac{(C - 2.5\ (\frac{1}{5} \times 0.033)}{(1 - \frac{1}{5} \times 0.033)}$$

where C is observed count rate after background corrections.

C' corrected count rate.

Pairs rate correction

$$C'_p = \frac{C_p - 0.1\ (\frac{1}{5} \times 0.033)}{(1 - \frac{1}{5} \times 0.033)}$$

where C_p is observed pairs rate after background and multiplicative factor corrections.

C'_p corrected pairs rate.

The correction in the case of the count rate was very small in all cases, and that due to the pairs rate was in all cases negligible.

Appendix III

"Spiked" sample calculation for k and k_p and calculation of counts/pairs ratio for Thorium series

(i) "Spiking"

Theory

For total counting, assuming series equilibrium

$$C = \frac{NA\rho}{4} \sum R_i$$

R_i - ranges
ρ - density
A - area
N - no. of dis/gram/hour
C - counts/hour.

Because of equilibrium N is same for each member of a series and, for a single series,

$$N = \eta\lambda_H$$

λ_H decay constant of head member of series in hours^{-1}

η no. of atoms of head member of series per gram of sample

$$\eta = \frac{(H)(10^{-6})}{(A_H)(1.66)(10^{-24})}$$

H - no. of ppm of head member of series
A_H - atomic mass of head member.

$$\therefore \quad N = \frac{(H)(.693)(10^{18})}{(T_{\frac{1}{2}})(A_H)(1.66)(365)(24)}$$

$T_{\frac{1}{2}}$ - half life of head member in years

∴ For a sample containing thorium series only, headed by Th-232

$N_{232} = 14.77\ Th$

Th - no. of ppm of Th-232

$T_{\frac{1}{2}}$ for Th-232 = 1.4×10^{10} years

A_H for Th-232 =

Thus

$$C = \frac{14.77\ Th \times A}{4} \sum_{Th-232} R_\rho$$

and $$\sum_{Th-232} R_\rho = 0.32 \times 10^{-3} \times \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} \sum_{Th-232} R_{air}$$

For thorium series below Th-232 $R_{air} = 28.48$ cms.

$$\text{For blood } \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} = 3.4$$

$A = 81$ sq cms.

$$\therefore \quad C = 9.27\ Th.$$

For "alpha pairs"

$$C_p = \frac{nAR_1\rho}{8}\left(1 - \frac{R_1}{3R_2}\right)$$ where R_1 range Em-220

R_2 range Po-216

For series in equilibrium $n = N_{232}$

and, treating as above

$$C_p = 0.574\ Th$$

The "spiked" blood sample contained "old" thorium sulphate and the thorium content was determined from gamma spectroscopy as 0.415 grams thorium per gram thorium sulphate. The sample was "spiked" to contain $3.8(10^{-4})$ grams thorium sulphate in 45 grams wet blood. The sample was freeze dried and the concentration factor found to be 5.77.

∴ Thorium content in ppm in dried sample

$$= \frac{(.415)(3.8)(10^{-4})(5.77)(10^6)}{45}$$

$$= 20.22 \text{ ppm.}$$

Thus:

Calculated C = 9.27 × 20.22 = 187.4 c/hour

Calculated C_p = 0.574 × 20.22 = 11.6 p/hour.

The sample was counted at intervals of ∿ 1 week throughout the course of this work. After correcting for background etc.

Mean observed C = 166.6 c/hour

Mean observed C_p = 10.5 c/hour

The observed C and C_p are lower than the calculated C and C_p by factors k and k_p respectively where

$$k = \frac{187.4}{166.6} = 1.13$$

$$k_p = \frac{11.6}{10.5} = 1.11$$

(ii) Counts/pairs ratio for Thorium series

For total alpha counting

$$C = \frac{NA\rho}{4} \sum_i R_i$$ - symbols as above

and for "alpha-pairs"

$$C_p = \frac{nAR_1\rho}{8} \left(1 - \frac{R_1}{3R_2}\right)$$ - symbols as above.

For the work done in this thesis it is presumed that the thorium series is headed by Th-228 (see Chapter 2)

Thus for an idealised sample containing only thorium

series, in equilibrium

$N = N_{228}$, $n = N_{228}$, where N_{228} is no. of dis/gram/hour of Th-228

and

$$\frac{C}{C_p} = \frac{\frac{N_{228} A \rho}{4} \sum_i R_i}{\frac{N_{228} A R_1 \rho}{8} 1 - \frac{R_1}{3R_2}}$$

substituting for $R\rho$, as in theory above

$$\frac{C}{C_p} = \frac{2 \left| (0.32) \times (10^{-3}) \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} \sum_{\text{Th-228}} R_{air} \right.}{(0.32)(10^{-3}) \frac{\Sigma f_i w_i}{\Sigma f_i Z^{2/3}} R_{1air} 1 - \frac{R_1}{3R_2}}$$

$\therefore \sum_{\text{Th-228}} R_{air} = 25.68 \quad R_{1air} = 5.00 \quad \frac{R_1}{R_2} = 0.887$

$\therefore \frac{C}{C_p} = \frac{2 \times 25.68}{5.00 \ (0.704)} = 14.6$

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