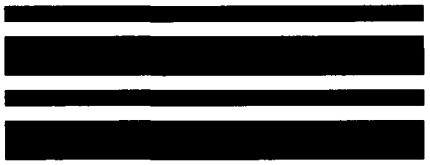

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Paper 1

THE MONTE BELLO RAT NOVEMBER 1953

by

D. W. H. BARNES
G. E. HARRISON
J. F. LOUTIT
W. H. A. RAYMOND

with appendices by

G. J. NEARY

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HARWELL, BERKS.
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Paper 1

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D. W. H. Barnes, G. E. Harrison, J. F. Loutit, W. H. A. Raymond

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Medical Research Council Radiobiological Research Unit

A.E.R.E. Harwell,
Berks.

July 1955.

"THE MONTE BELLO RAT, NOVEMBER 1953"

by

D.W.H. Barnes
G.E. Harrison
J.F. Loutit
W.H.A. Raymond

ABSTRACT

The black rat which inhabits Trimouille was trapped in November 1953 in the contaminated zone of the island. The lairs of the trapped rats were not found, but at least in their nightly foraging they were exposed to external β and γ radiation from the environment the estimated dose rate being 1000 m.rep/hr. and 30 mr/hr. respectively. (Appendix I). The pelt was contaminated which must have been a continual and additional source of external ~1 rep/day. (Appendix I). From the evidence of the intestinal contents, the rat lived on contaminated food. Nevertheless the post mortem appearances to the naked eye and the histological detail microscopically were essentially normal. The only evidence suggestive of damage from radiation was multiple foetal death in one pregnant animal.

Assay of the internal organs and tissues indicated that the soft tissues contained little radioactive material. The specific activity of these tissues was an order of magnitude less than that of the bones. In turn the bones were one or two orders of magnitude less in specific activity than the pelt and gut-contents which can be taken as representative of the external conditions. In two groups of animals, each group representing a different degree of environmental contamination, the specific activity of the bones was proportional to that of the environment. Over the course of a year the soft tissues and the gut and pelt decayed to just under one half of the original count-rate. Two samples of kidney and a larger number of bone-samples decayed more slowly to about 60% of the original count.

The measured rates of decay are in accord with a small number of radiochemical analyses. Those of soil and pelt indicated that almost all the radioactivity was attributable to ^{106}Ru and ^{144}Ce and daughter products with a notable deficiency of Sr and Zr. Bone on the other hand contained appreciable quantities of Sr. From the available figures of specific activity per gram of calcium it can be calculated that the availability of the radio elements in the environment was in descending orders of magnitude Sr > Ru > Ce. Assay of portions of the guts with contents suggest that the diet was of higher specific activity than the soil as sampled. It is tentatively concluded that the animal discriminated against strontium in favour of calcium.

A.E.R.E. Harwell.

15th June, 1955.

/BVW.

H/22088.

THE MONTE BELLO RAT

The rat inhabiting the Island of Trimouille was the so called black Asiatic rat. It was only seen at night, never in daylight.

Its nest was not found. No rat-holes were visible in the soft sand. In unblasted areas the ground-cover with vegetation, most of it dead, was considerable. It would have been eminently possible for rats to have nested in the dead grass. However in the area which had been blasted and which was the most contaminated zone, all vegetation had been cleared; the new growth was scanty and not sufficient to conceal nests. There was plenty of coral rock round the periphery of the island and the rock would have provided many holes and crevices. It would not have provided food and if rock were the habitat, journeys up to several hundreds of yards would have been necessitated for the rats to get food and to reach the sites where traps were successfully laid.

The diet can only be guessed. Green vegetation was, from the evidence of the stomach contents, one source. As with most animals in this torrid climate, vegetation must have been an important source of water and many of the xerophilic plants are succulents. Cannibalism cannot be neglected and at least one of the rats trapped was part eaten presumably by its like. Reptiles could also have been an appropriate diet for the carnivorous rat. One trap had obviously caught a snake or goanna, but only a small fragment of skin remained for identification. All the rats caught had full stomachs distended with greyish, green porridy material. The full stomach and the leanness of the animals suggested a bulky diet with low nutrient content.

The density of the population can only be guessed with the following observations in mind.

(i) Trapping was carried out on three sites. Site 1, which was virtually uncontaminated, was a base where the surface of the sand recorded only 8 c/s. (including a background of 2-3 c/s.) with a 1021 monitor. Site 2 included the remnants of various structures built of corrugated iron and was mildly contaminated. A 1313 monitor indicated a γ dose rate of 2-3 mr/h. at one yard above the ground. Site 3 was more heavily contaminated, the 1313 monitor gave readings of 30-100 mr/h. The site included various installations and remnants of galvanised iron where traps could be set under cover.

The results of the trapping were as follows:-

	Site 1	Site 2	Site 3
Rats caught / Traps set	4/4	7/20	7/47

These figures may, however, be misleading. A high proportion, which was not recorded, of the traps set in sites 2 and 3 were sprung with the bait gone. Rats were probably but not necessarily the agents; the almost wholly eaten reptile mentioned above suggests other possibilities.

(ii) The number of rat spoors was much greater in the sand in site 3 than elsewhere. This may be because there was much less vegetation and therefore more sand on which the spoors showed up more readily.

POSTMORTEM APPEARANCES

Eighteen rats were submitted to autopsy. The four from the base area (site 1) (negligible activity on the ground) were adults. Of the seven rats from the area of low activity (site 2, 2-3 mr/h.) all males, four were adolescents estimated to be between 100 and 200 gm. in weight and three were adult. Amongst the seven from the area of moderate activity (site 3, 30-100 mr/h.) - only one, a female, was a youngster of about 100 gm. weight.

In all cases the external appearances even in death suggested the rats had been healthy. The coat was thick and shining. Internally it is true the depot fat was very scanty but the musculature was firm, pink and glistening. None of the organs showed any deviation from the normal except in so far as could be attributed to the violent death. Two animals had been caught across the abdomen and showed lacerations of the liver: almost all showed congested lungs often with frank haemorrhages. One animal had been partially cannibalised with loss of some of the abdominal viscera.

Two (≒ 14 and ≒ 15) of the four adult females from the area of moderate activity were obviously pregnant. In each case the entire uterus was fixed in alcohol and brought back to Harwell for examination. In ≒ 14, the left horn of the uterus contained 3 foetuses, apparently normal. These three embryos were digested for their radioactivity. The placentae and membranes were similarly assayed. The right horn of the uterus was examined by Dr. T.C. Carter at Harwell. It contained four embryos of 19-20 days gestation. In his opinion any embryo dying earlier than the tenth day would not have been identifiable under the conditions of fixation (i.e. in utero). In ≒ 15, the left horn contained five embryos. Four were viable and were digested: the placentae and membranes were similarly ashed. The fifth embryo according to Dr. Carter had died at about the 12-13th day. The right horn contained one live embryo of 14 days gestation, one embryo dead at the 13th day and one dead at the 9-10th day. In Dr. Carter's opinion three dead embryos out of a total of eight was by itself not remarkable, but two deaths of three occurring at the 12th or 13th day would probably be unusual in the absence of radiation. This isolated phenomenon was the only instance of a lesion suggestive of damage by radiation.

HISTOLOGY

Of the seven animals from each of the areas, low and moderate activity, the following tissues were taken for histological section:-

	Low Activity	Moderate Activity
Upper third of femur	(7)	(7)
Sternum	(4)	(6)
Liver	(6)	(7)
Colon	(1)	(1)
Thyroid	(1)	(1)
Kidney	(1)	(3)
Lung	(1)	(2)
Testis	(1)	(1)
Preputial gland	(1)	(1)
Adrenal	(0)	(1)
Spleen	(0)	(1)
Lymph gland	(0)	(1)
Skin	(0)	(1)

In no case was a lesion attributable to radiation detected microscopically.

Head of Femur

From the osteoblastic activity and the appearance of the epiphyseal plate it was possible to differentiate the young from the mature animals. This assessment tallied exactly with the post mortem report of the animal's size. No bone lesion was detected in any of the animals. The marrow was in all cases cellular and active in erythropoiesis and granulopoiesis. Megakaryocytes were normal in abundance and appearance. The degree of cellularity varied directly with the immaturity of the epiphyseal plate. The more immature the latter, the less was the amount of fat and the tigher was the packing of the haemopoietic elements.

Sternum

As with the femur, the age of the animal was determinable from the osteoblastic activity. There were no bony abnormalities and the activity of the marrow was closely parallel with that of the upper femur.

Colon

In one animal there was desquamation of the superficial portion of the mucosa. As post mortem changes were noted in other organs of this animal (#14) this change was also regarded as such.

Thyroid

In rat (#2) from the area of low activity the thyroid was unquestionably normal with most acini well filled with colloid. In rat (#4) from the area of moderate activity there were a few acini dilated with colloid in the periphery, but the bulk of the section showed acini filled with polygonal cells. The gland was not enlarged and the general post mortem appearances suggested no functional thyroid upset.

Kidney

The four kidneys examined showed no changes other than congestion and some petechial haemorrhages which were compatible with the mode of death.

Lung

In two of three instances there was massive congestion and haemorrhage again directly related to the violent death. Apart from formalin pigment in one case there was no evidence of pigment or particulates in the lung tissue.

Testis

In the two animals studied there was active spermatogenesis which appeared normal. In one, some tubules showed degenerative changes but this appeared to be an artefact of fixation.

Adrenal

In the single gland sectioned (#4), there was great congestion of medulla and of the reticular zone of the cortex. Nuclear changes - karyolysis and karyorrhexis - were present in both reticular and fascicular zones. Post mortem changes were noted elsewhere.

Spleen

In the single example there was congestion and some brown pigment deposits (? haemosiderin) in the pulp. The lymphoid tissue appeared normal.

Lymph gland
Skin
Preputial glands } There was nothing of note to report on in these sections.

METHODS OF RADIOACTIVE ASSAY

Dissection of Rats in the Field

The rats were pinned out with skewers through the four feet. With scissors the skin was incised centrally from pubis to the neck and from this incision laterals were taken along the limbs to wrist and ankle. With a clean knife skin flaps were reflected and pinned back. With clean scissors and forceps selected organs and tissues such as bone were dissected out and stored in 10% formolsaline or in 70% spirit. It is claimed that avoidance of contamination of the selected material from the skin was achieved.

In the case of one rat (# 14) after removal of the organs a complete skinning was attempted with the exception of the four feet which were bottled separately. It cannot be claimed that the carcase was uncontaminated by the pelt in the operation of skinning but a reasonable degree of 'no-touch' was attained. The pelt and carcass were bottled separately in spirit. In the laboratory at home, the separation of the preserved carcase into soft-tissue-carcase and skeleton was completed.

Preparation of Tissues

In the laboratory at home, the preserved organs and tissues were gently blotted and weighed. Portions were selected for histology and the weighed residuum boiled in concentrated nitric acid until completely digested. It was demonstrated that at this temperature there was no loss of activity from mixed fission products. When digestion was complete the liquid extracts were made up to known volumes with water and counted as liquids in a M6H Geiger Counter.

This regime was adopted with all but the first two specimens assayed. These were the stomach + small gut and large gut of rat 14 which were taken to dryness. It was shown that loss of activity occurred. The subsequent radiochemical assay of these two materials is therefore sound only for the non-volatile components.

RESULTS OF RADIOACTIVE ASSAY

Control Rat (≒1)

From this animal only two tissues were sampled - bone and liver. No radioactivity above background was detected in either.

Rats from Lightly Contaminated Area (≒2, 3, 5, 6, 7, 8, 9)

In the event the specific activity of the tissues from these animals was so low that the specimens, purposely selected not to be too large for adequate fixation, were not large enough for reliable counting. The largest samples of soft tissue from the liver were $\frac{3}{4}$ to 2 gm. in weight; yet the count-rate was barely above background. Only in the case of the long bones were significant counts obtained from the samples ($\frac{1}{2}$ to 3 gm.). Here, in the first week of January 1954, the specific activity ranged from 20 to 52 c/m/g (mean 35).

Rats from Moderately Contaminated Area (≒4, 10, 11, 12, 13, 14, 15)

Even in the case of these animals the soft tissues contained little radioactivity and the calculated specific activities are therefore mere approximations. The following table gives the specific activities of tissues analysed.

SPECIFIC ACTIVITY OF TISSUES - c/m/g EARLY JANUARY, 1954

Animal	Skin	Colon & Contents	Long Bones	Sternum	Kidney	Liver	Lung	Foetus	Placenta	Testis
4	5,200	-	138*	-	58	22	36	-	-	-
10	-	-	212	84	25	28	-	-	-	21
11	-	-	500	276	-	60	-	-	-	-
12	-	-	950	135	-	59	-	-	-	-
13	-	-	383	217	-	45	-	-	-	-
14	15,400	8,200	256*	-	305	36	57	17	33	-
15	-	-	433	155	-	26	-	16	23	—

* mixed long and flat bones.

The specific activities of the bones are an order of magnitude greater than those of the soft tissues except for one high value for the kidneys. However the internal contamination is markedly less than that of the environment as exemplified by the pelt and the gut-contents.

In the case of #14 two other analyses were made (a) of the feet (b) of the soft-tissue-carcase. The specific activity of the feet came to 8600 c/m/g., about one half that of the rest of the integument. This is what one might expect from the mixture of skin, bone, cartilage and tendon of which the feet are composed. The specific activity of the soft tissue carcase was measured as 136 c/m/g. a figure higher than that of the other soft tissues except the kidney. The value for kidney is thought to be reliable as it was dissected cleanly. The carcase, however, as has been noted must have been partially contaminated in the operation of skinning and this soiling is most likely to be of soft tissue rather than the bones it covered.

In this group the specific activities of the bones are about an order of magnitude greater than those of the previous group from the area of low activity and this is in line with the ratio of the physical contamination for the two zones.

The decay of the various organs and tissues where the count rate was initially appreciable was followed for one year. The soft tissues (except kidneys) skin and guts decayed in parallel. The bones and less certainly the kidneys on the other hand decayed at a slower rate. This experimental finding is in exact accord with the prediction made from the few detailed chemical analyses made early in 1954 (vide infra).

Specimen	% of initial Count-Rate After 1 Year	
#= 4 Skin	47	Mean
#=14 Stomach and small intestine	47	
#=14 Large intestine	46	
#=14 Pelt	48	47 ± 0.3
#=14 Feet	48	
#= 2 Large intestine	48	
#=15 Liver	49	
#=14 Foetus	50	
#=14 Liver	47	
#=14 Carcase	49	
#=14 Lung	47	
#=13 Liver	44	47 ± 0.7
#=11 Liver	45	
#=10 Liver	44	
#= 4 Liver	47	
#=14 Kidney	55	56
#= 4 Kidney	57	
#=15 Long bones	59	
#=14 Mixed Bones	61	
#=13 Long bones	57	
#=13 Sternum	49	
#=12 Long bones	57	
#=12 Sternum	65	60 ± 1.3
#=11 Long bones	59	
#=11 Sternum	58	
#=10 Long bones	60	
#=10 Sternum	69	
#= 4 Mixed bones	65	
#= 9 Long bones	60	
#= 5 Long bones	62	
#= 3 Long bones	65	

RADIOCHEMICAL ANALYSIS OF THE GROUND FROM THE MODERATELY CONTAMINATED AREA AND THE SKELETON, PELT AND GUT CONTENTS FROM RAT 14

A chemical separation of the constituent activities in the soil of Trimouille was made on samples, collected in November 1952 and 1953, of a superficial layer of the ground from the moderately contaminated area. In addition, a similar analysis was made on nitric acid digests of the pelt, skeleton, stomach and small intestine and the large gut of rat 14. The possible fission-product activities at the time of the initial examination, February 1954 were:-

^{89}Sr, ^{90}Sr, ^{90}Y, ^{91}Y, ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{106}Rh, ^{137}Cs, ^{137}Ba,

^{144}Ce, ^{144}Pr and ^{147}Pm.

of which it was expected ^{106}Ru might be volatile in nitric acid. Although the general principles of the separation of carrier-free fission products by the use of ion-exchange columns had been well established by the work of

others[1], little had been published on the separation of ruthenium activities at the time of these experiments. Moreover, the nature of the biological samples, particularly acid digests of the skeleton with a high percentage of calcium and phosphate ion, made normal chemical methods of separation unworkable. The following method was evolved as a result of trials on several biological samples.

(a) Extraction of Ruthenium

A distillation of the nitroso compounds of ruthenium was made from a nitric acid digest of the soil or rat tissues. Figure 1 shows the still employed in which the outlet from a double surface condenser A was closed by a liquid trap B containing N soda, the distillate draining back being tapped off at C. The sample was put in flask D and about 10 ml. of concentrated nitric acid, Sp.G.1.42, containing about 2 mg. each of ruthenium, cerium, strontium and yttrium, as carriers, were slowly run in by turning tap at E. Heat was then applied to boil off the acid, the contents of the flask being kept near dryness throughout the operation. Further additions of nitric acid were made which occasionally contained 1 drop of a 1% solution of ruthenium chloride. Successive 10 ml. amounts of the distillate withdrawn from C were assayed on an M6H Geiger counter and the operation was continued until no activity was obtained in at least three consecutive distillates. The total activity due to the volatile fraction included a final water wash of the condenser and any activity in the trap which was always quite small.

As a confirmation that the distillate contained only ^{106}Ru, ^{106}Rh an extraction of solid ruthenium sulphide was made from the nitric acid solution. An enriched extract of ruthenium freed from the main bulk of nitric acid was made for this extraction by returning the distillate to a clean still and reheating to collect the initial fractions. Ruthenium sulphide together with a little sulphur, was then precipitated by passing hydrogen sulphide through the distillate diluted with one or two volumes of water. The sulphide was centrifuged off and plated on an aluminium planchette so that the decay and absorption of the activity in known thickness of aluminium could be compared with those of a source of ^{106}Ru, ^{106}Rh using an E.H.M. counter.

(b) Extraction of Strontium

The percentage of the total activity due to strontium was of special interest in the present experiments. In order the better to separate it from other radioactive species and because of the large excess of calcium, it was decided to preface any ion-exchange column separation by an extraction of strontium as nitrate in 19N nitric acid.

The residue in flask A was therefore transferred to a 50 ml. centrifuge tube by successive small washes with concentrated nitric acid. The insoluble material was removed by centrifuging and after washing, this was dried off on a planchette for assay on an E.H.M. counter. The supernatant was made 19N in nitric acid by the addition of fuming nitric acid until the original volume of the acid was increased by about 60%. The separation of strontium then proceeded as described by Harrison and Raymond 1955[2], the supernatants and washings being combined and retained for the separation of the rare-earth activities.

Strontium was finally plated as sulphate and the decay and absorption of the activity compared with that of a pure source of ^{90}Sr, ^{90}Y after allowing at least 11 days for the equilibrium of the daughter product, ^{90}Y, to be established.

(c) Separation of Zr, Nb, Ce and Other Rare Earths

The nitric acid supernatants were now taken to dryness the distillate being condensed and assayed for any residual ^{106}Ru, ^{106}Rh activity. The activity of this distillation was, as anticipated, practically zero. The residue was taken up in a minimum of 4N HCl by warming and the final acid solution made about 0.1N.

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others[1], little had been published on the separation of ruthenium activities at the time of these experiments. Moreover, the nature of the biological samples, particularly acid digests of the skeleton with a high percentage of calcium and phosphate ion, made normal chemical methods of separation unworkable. The following method was evolved as a result of trials on several biological samples.

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(c) Separation of Zr, Nb, Ce and Other Rare Earths

The nitric acid supernatants were now taken to dryness the distillate being condensed and assayed for any residual ^{106}Ru, ^{106}Rh activity. The activity of this distillation was, as anticipated, practically zero. The residue was taken up in a minimum of 4N HCl by warming and the final acid solution made about 0.1N.

A Zeo-carb 225, ion-exchange column was prepared some 20 cm. long and of about 1 sq. cm. section and recycled to the acid form by successive washings with water, soda and acid. The acid supernatant was passed down this column at a flow-rate of about ½ ml. per hour, the effluent being continuously monitored by an end window E.H.M. counter connected to a rate-meter and recording millivoltmeter. When the effluent activity was more than twice the background the effluent was collected in a separate flask until the recorded activity had once more returned to near the background. This collection was retained for oxidation and the subsequent preparation of a concentrate in the form of a dried extract so that activity and adsorption measurements could be made as for the ruthenium and strontium fractions. The activities appearing in the acid elution were of small activity showing that the samples contained only trace amounts of active anionic radicals. This stage was also effective in removing phosphate ion which otherwise was readily precipitated together with the calcium in the samples whenever attempts were made to separate the constituent activities by ordinary chemical methods by making the mother liquor alkaline.

The elution of zirconium and niobium with ½% oxalic acid and subsequently the rare earths in 5% citric acid over an ascending pH range from 3.0 to 4.5 followed the pattern described by Spedding et al.(1)

The peak activates in the eludate were again collected separately and after assay on the M6 counter they were oxidised with perchloric acid and plated for assay of the absorption and decay of the activity on the end window-counter.

RESULTS

All the chemical fractions were assayed for activity in the liquid form before they were plated as solid extracts for absorption and decay. Thus, the activity of these extracts is expressed as counts per min. on the M6 counter. The efficiency of this counter was determined for ^{144}Ce - ^{144}Pr; ^{106}Ru - ^{106}Rh; ^{89}Sr, and for ^{90}Sr - ^{90}Y so that these counts could readily be converted into disintegrations per min.

In correcting for the decay of strontium fractions, it was assumed that the relative distribution of $^{89}Sr/^{90}$ was normal.

The results for analysis on two samples of soil, one collected November 1953 and another collected November 1952 are given in Table I - III.

TABLE I

MODERATELY CONTAMINATED GROUND

SOIL - 1ST ANALYSIS 20.2.54.

Date	20.ii.54	1.xi.53				
Fission Products	M6ct/min/g x 10^{-4}	Calculated M6ct/min/g x 10^{-4}	Efficiency of Counter %	≠ dis/min/g x 10^{-5}	µC/g.	% Total Activity
$^{106}Ru^{106}Ru$	3.44	4.16	7.5	5.6	0.25	44.5
$^{144}Ce^{144}Pr$	3.63	4.62	7.5	6.2	0.28	50
$^{89}Sr^{90}Sr^{90}Y$	0.014	0.024	6.5	0.04	0.002	0.3
$^{95}Zr^{95}Nb$	0.021	0.062	~1.0	0.6	0.03	5.3
^{91}Y	0.007	0.027	7.0	0.04	0.002	0.3
TOTAL	7.11	8.9	-	-	0.56	-

≠ Total disintegrations/min/g for parent and daughter product.

TABLE II

SOIL - 2ND ANALYSIS 8.1.55.

Date	8.i.55	1.xi.53				
Activity	M6ct/min/g x 10^{-4}	Calculated M6ct/min/g x 10^{-4}	Efficiency of Counter %	dis/min/g x 10^{-5}	µC/g.	% Total Activity
$^{106}Ru\,^{106}Rh$	1.68	3.8	7.5	5.1	0.23	48
$^{144}Ce\,^{144}Pr$	1.45	4.2	7.5	5.6	0.25	52
$^{89}Sr\,^{90}Sr\,^{90}Y$	0.017	0.024	6.5	0.037	0.0017	0.3
TOTAL	3.15	8.0	-	-	0.48	-
	Calcium content of ground 36% $^{W}/_{W}$.					

Activity and Decay of Ground Samples

Samples of the soil were assayed for total radioactivity on an end-window E.H.M. counter over a period of several months. The half-life of this activity was about 305 days during 1954 and from measurements on 11 such samples, the mean total specific activity was calculated to be 0.53 µC per g. on 1st November 1953, the mean square variation being 23%.

TABLE III

SOIL FROM THE SAME AREA COLLECTED NOVEMBER 1952

Date	23.iii.54	1.xi.53				
Activity	M6ct/min/g x 10^{-4}	Calculated M6ct/min/g x 10^{-4}	Efficiency of Counter %	dis/min/g x 10^{-5}	µC/g.	% Total Activity
$^{106}Ru\,^{106}Rh$	13.2	15.8	7.5	21.0	0.95	53.5
$^{144}Ce\,^{144}Pr$	10.4	13.3	7.5	17.8	0.80	44.9
$^{89}Sr\,^{90}Sr\,^{90}Y$	0.03	0.04	6.5	0.1	0.0045	0.25
$^{95}Zr\,^{95}Nb$	0.03	0.05	~1.0	0.5	0.022	0.1
^{91}Y	0.03	0.08	7.0	0.1	0.0045	0.25
TOTAL	23.7	29.3	-	-	1.78	-

The differential analyses agree very satisfactorily considering the variation in time between the analyses recorded in Tables I and II and considering the variation in samples.

The most notable feature of all the analyses is the paucity of strontium isotopes - determined no doubt by the nature of the circumstances at the time of the detonation. Also zirconium was not identified in the expected amount.

The material from rat 14 gave the results indicated in Table IV - VII.

TABLE IV

RAT 14 SKELETON (26.4 g. WET WEIGHT)

Date	10.i.54	1.xi.53				
Activity	M6ct/min x 10^{-4}	Calculated M6ct/min x 10^{-4}	Efficiency of Counter %	dis/min x 10^{-5}	$\mu C/g_4$ x 10^{4}	%Total Activity
^{106}Ru ^{106}Rh	0.60	0.685	7.5	0.91	15	75
^{89}Sr ^{90}Sr ^{90}Y	0.11	0.155	6.5	0.23	3.9	19.5
^{144}Ce ^{144}Pr	0.044	0.05	7.5	0.07	1.2	6.0
TOTAL	0.75	0.89	-	1.21	20	-

TABLE V

RAT 14 - PELT

Date	18.i.54	1.xi.53				
Activity	M6ct/min x 10^{-4}	Calculated M6ct/min x 10^{-4}	Efficiency of Counter %	dis/min x 10^{-5}	μC	%Total Activity
^{106}Ru ^{106}Rh	24.0	27.8	7.5	37.0	1.67	48.2
^{144}Ce ^{144}Pr	24.1	29.1	7.5	38.8	1.75	50.5
^{89}Sr ^{90}Sr ^{90}Y	0.05	0.08	6.5	0.12	0.005	0.15
^{91}Y	0.1	0.27	7.0	0.4	0.018	0.52
^{95}Zr ^{95}Nb	0.02	0.04	~1.0	0.4	0.018	0.52
TOTAL	48.3	57.3	-	76.7	3.45	-

TABLE VI

RAT 14 - STOMACH AND SMALL INTESTINE

Date	15.i.55	1.xi.53				
Activity	M6ct/min x 10^{-4}	Calculated M6ct/min x 10^{-4}	Efficiency of Counter %	dis/min x 10^{-5}	µC	%Total Activity
^{106}Ru ^{106}Rh	0.218	0.50	7.5	0.68	0.031	35.2*
^{144}Ce ^{144}Pr	0.320	0.94	7.5	1.26	0.057	64.5
^{89}Sr ^{90}Sr ^{90}Y	0.003	0.006	6.5	0.009	0.0004	0.5
TOTAL	0.54	1.45	-	1.95	0.088	-
		Calcium content 16.4 mg.				

* low because of losses of volatile components in digestion. (vide supra).

TABLE VII

RAT 14 - LARGE GUT AND BOWEL CONTENTS

Date	18.i.55	1.xi.53				
Activity	M6ct/min x 10^{-4}	Calculated M6ct/min x 10^{-4}	Efficiency of Counter %	dis/min x 10^{-5}	µC	%Total Activity
^{106}Ru ^{106}Rh	0.058	1.36	7.5	1.81	0.082	38.0*
^{144}Ce ^{144}Pr	0.075	2.18	7.5	2.90	0.13	61.0
^{89}Sr ^{90}Sr ^{90}Y	0.0017	0.032	6.5	0.05	0.0022	1.0
TOTAL	0.135	3.58	-	4.76	0.215	-
		Calcium content 35.8 mg.				

* low because of losses of volatile components in digestion. (vide supra).

The relative specific activities of the samples of soil (taking the analysis in Table I) to those of the rat's skeleton (Table IV) are therefore as in Table VIII.

TABLE VIII

Sample	Elements	Specific Activity μC/g., Specimen	SA Soil / SA Skeleton	Specific Activity μC/g.Ca.	SA Soil / SA Skeleton
Soil	$^{106}Ru^{106}Rh$	0.25	167	0.695	46
Skeleton	"	$1.5.10^{-3}$		$1.5.10^{-2}$	
Soil	$^{144}Ce^{144}Pr$	0.28	2540	0.77	700
Skeleton	"	$1.1.10^{-4}$		$1.1.10^{-3}$	
Soil	$^{89}Sr^{90}Sr^{90}Y$	2.10^{-3}	5	$5.5.10^{-3}$	1.5
Skeleton	"	$3.8.10^{-4}$		$3.8.10^{-3}$	

The ratio is also given in terms of activity per gram of Ca assuming that one-tenth of the wet weight of the skeleton is Ca. It can be presumed that the strontium is absorbed and retained to an extent greater by an order of magnitude than ruthenium which in turn differs by an order of magnitude from cerium. The figure of 1.5 for strontium may, from the point of view of metabolic behaviour and hazard, give an unduly pessimistic impression. It suggests that the animal utilised strontium equally well as calcium in its diet. When the similar ratios for stomach and small intestine with contents on the one hand and for large intestine and contents on the other are calculated, however, the figures given in Table IX emerge.

TABLE IX

Organ	Elements	Specific Activity μC/g.Ca	S_A gut and contents / S_A skeleton
Small Gut	$^{106}Ru^{106}Rh$	1.9	127
	$^{144}Ce^{144}Pr$	3.4	3100
	$^{89}Sr^{90}Sr^{90}Y$	.02	6
Large Gut	$^{106}Ru^{106}Rh$	2.2	150
	$^{144}Ce^{144}Pr$	3.6	3300
	$^{89}Sr^{90}Sr^{90}Y$	.061	16

The specific activities for Ce-Pr and Sr-Y are about four times or more those of the earth. Even for Ru-Rh where in the case of the gut samples, there were certainly some losses in the preparation, the specific activity is three times that of soil. If at least the stomach and small gut figures are representative of the diet, then the animal eats relatively more contamination than inactive soil. This would be expected if the surface of the ground had been relatively undisturbed by weather or extraneous circumstances. This finding may indicate that the animal does discriminate against strontium in favour of calcium. The higher ratio in the large gut (16) than in the small gut (6) may merely indicate error due to small activities - or perhaps the intestinal route of excretion by which this discrimination is effected.

DISCUSSION

These rats caught in the contaminated areas of Trimouille were essentially healthy. In spite of the contamination of the ground on which they foraged and of the herbage which they ate signs of damage from radiation were minimal or absent.

The dose rates to which they were exposed are calculated in Appendix I, which indicates that the γ dose rate from the contaminated ground was about 30 mr/hr. together with a β dose rate of about 1,000 m.rep/hr. It is a fair assumption that this dose applied for about eight hours a day if not for the whole twenty-four hours.

In addition they were subject to a smaller dose from contamination of the pelt - 1 rep per day - unless this is falsely high due to extra contamination of the pelt agonally. The dose to the skeleton is calculated to be less (Appendix II).

The notable conclusion is that in these circumstances the hazard from external irradiation is more considerable than from internal contamination.

REFERENCES

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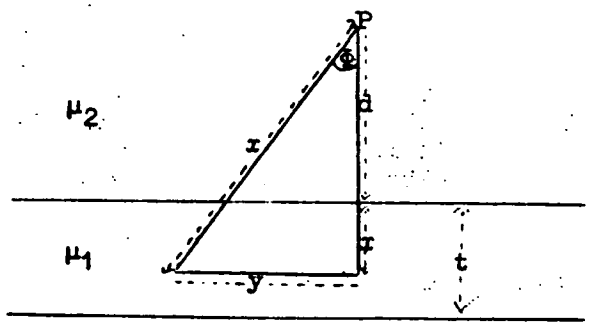
APPENDIX I

It is required to calculate the beta and gamma dose rates to a rat on contaminated soil, and the beta dose rate to a rat having contminated fur. These problems are basically similar.

(a) General Analysis

A slab of one medium, thickness t, and infinite area, contains a uniform specific activity m millicuries per cc. of an isotope which emits radiation (beta or gamma) having a specific dose rate k in units of r per millicurie-hour at 1 cm. The dose rate at a height d in an overlying medium is required. Expotential absorption of the radiation is assumed in both media, with linear coefficients μ_1 and μ_2; the fact that the beta range is not infinite is ignored, with negligible error.



The dose-rate of the primary radiation at P is

$$\int_{r=0}^{t}\int_{y=0}^{\infty} \frac{\kappa\, m\, 2\pi\, y\, dy\, dr}{x^2}\, e^{-(\mu_1 r \sec\theta + \mu_2 d \sec\theta)}$$

$$= 2\pi\kappa m \int_{r=0}^{t} dr \int_{v=(\mu_1 r\, \mu_2 d)}^{\infty} e^{-v}\frac{dv}{v}$$

$$= 2\pi\kappa m \int_{r=0}^{t} E\,(\mu_1 r\, \mu_2 d)\, dr$$

where E (Z) is the exponential integral tabulated by Jahnke and Emde as -Ei(-Z).

Integrating by parts,

$$\text{Dose-rate} = \frac{2\pi\kappa m}{\mu_1}\left[(\mu_1 t+\mu_2 d)\, E\,(\mu_1 t+\mu_2 d) - \mu_1 d.\, E\,(\mu_2 d) + e^{-\mu_2 d} - e^{-(\mu_1 t+\mu_2 d)}\right]$$

For gamma radiation, an approximate allowance can be made for scattered radiation by assuming that when the absorbing path is greater than the median free path, the rate of absorption of scattered radiation is equal to its rate of production (Chamberlain, A.E.R.E. HP/R.551). In other words the effective coefficient of conversion of primary energy flux to absorbed dose is, per electron, $(\sigma_a + \sigma_s)$ instead of σ_a when the absorbing path exceeds the median

free path. Thus it is necessary to add to the expression for primary dose-rate above, the quantity,

$$2\pi\kappa m \frac{\sigma_s}{\sigma_a} \int_{x=0}^{t} dx \int_{v=0.6931}^{\infty} e^{-v} \frac{dv}{v}$$

$$\frac{2\pi\kappa m}{\mu_1} \quad 0.3787 \frac{\sigma_s}{\sigma_a} \mu_1 t$$

A further approximate allowance may be made for the backscattering of gamma radiation from the soil lying beneath the active material. The first correction above for scattered radiation assumes that all the scattering is in the forward direction, and so no backscatter should be added to this. But for the area within this region, it is legitimate to add the empirically known amount of backscatter, which can be said to emanate from an area of radius equal to the median free path in the lower medium. Thus a second correction term should be added, namely

$$b.2\pi\kappa m \int_{x=0}^{t} dx \int_{v=(\mu_1 x+\mu_2 d)}^{v=\frac{\mu_1 x+\mu_2 d}{x+d}\sqrt{(d+x)^2+\left(\frac{0.6931}{\mu_1}\right)^2}} e^{-v} \frac{dv}{v}$$

where b is the fraction of radiation backscattered.

If $\mu_1 x$ and $\mu_2 d$ are small compared with unity, most of the primary dose comes from within the area which contributes backscattered radiation. A sufficiently precise backscatter correction can therefore be made assuming that the total primary dose is increased in the proportion (1+b). Thus the total gamma radiation dose rate is in this case

$$\frac{2\pi\kappa m}{\mu_1} \left\{ (1+b)\left[(\mu_1 t+\mu_2 d)\, E(\mu_1 t+\mu_2 d) - \mu_2 d.\, E(\mu_2 d) + e^{-\mu_2 d} - e^{-(\mu_1 t+\mu_2 d)} \right] + 0.3787 \frac{\sigma_s}{\sigma_a} \mu_1 t \right\}$$

For beta activity in soil, it is sufficient to multiply the primary dose-rate by a simple backscatter factor, while for beta activity in an animal's fur, backscattering can be neglected.

(b) Gamma Radiation Dose-Rate

The experimentally observed gamma dose-rate in Site 3 was about 30 mr/hr. in November 1953. A sample of the soil (mainly calcium carbonate) showed a total specific activity of about 1 μc/gm. (Tables I, II, III), consisting principally of ^{106}Ru and ^{144}Ce in equal amounts with daughter products. The mixed radiations of the different isotopes can be represented by the equivalent values in Table A.

	Equivalent Maximum Beta Energy, Mev.	Gamma	
		Equivalent Energy Mev.	Excitation
^{106}Ru	.04	-	-
^{106}Rh	3.2	0.61	38%
^{144}Ce	0.26	0.08	30%
^{144}Pr	3.0	-	-

TABLE A Equivalent Radiations

The specific gamma ray dose-rates for the two gamma energies are 3.6 and 0.5r/mc.hr. at 1 cm. respectively. The mass absorption coefficients in the soil are $8.85 \times 10^{-2} cm^2/g.$ and $1.74 \times 10^{-1} cm^2/g.$ respectively and the density of soil may be taken as $2.0 g/c^3$. The lower energy gamma ray is comparatively unimportant; a sufficient approximation is to add half its specific dose rate per quantum to that of the 0.61 Mev. gamma ray, and assume a single gamma ray energy of 0.61 Mev. Thus in the expression for total dose-rate

$$m = \frac{0.38}{4} \times 10^{-3} \times 2 = 1.9 \times 10^{-4} mc/cc, \quad k = (3.6 + \tfrac{1}{2} \times \tfrac{30}{38} \times 0.5) = 3.8/mc./hr.$$

$$\mu_1 = 1.77 \times 10^{-1}, \quad \mu_2 = 1.07 \times 10^{-4} cm^{-1} \quad (1+b) = 1.12, \quad \sigma_s/\sigma_a \quad 2.68.$$

The thickness t of the layer of active soil is unknown, though it was not expected to be more than 1 cm. The thickness of the sample of soil removed was of the order of 1 cm; some inactive soil could have been included and clearly the specific activity of the sample sets a lower limit to the possible specific activity in the real active layer. Four possible values of t have been assumed, from 0 to ∞, and the corresponding specific activities in the active layer required to produce 30mr/hr. at 1 metre have been calculated. By comparing these figures with the specific activity of the sample, estimates may be made of the thickness of soil removed in taking the sample. The dose-rate at a point $2\frac{1}{2}$ cm. above the soil has been calculated in each case, to correspond to the mean dose-rate to the rat. Self-shielding is negligible and it may be shown that the mean dose-rate to the rat is very closely equal to the dose-rate at the mid-height, in spite of the possible high values of dose-rate at the soil surface for a superficial distribution of activity. The various calculations are summarized in Table B.

Assumed Thickness of Active layer cm.	Required Gamma Activity Per Unit Area	Required Specific Gamma Activity in Active Layer	Calculated Sample Thickness	Dose-Rate at 2.5 cm.
0	0.229 $\mu c/cm^2$	-	1.2 cm.	52.6 mr/hr.
0.1	0.260 $\mu c/cm^2$	2.605 $\mu c/cm^3$	1.3 cm.	36.2 mr/hr.
1.0	0.384 $\mu c/cm^2$	0.384 $\mu c/cm^3$	2.0 cm.	32.2 mr/hr.
∞	∞	0.200 $\mu c/cm^3$	Any Value	30.3 mr/hr.

TABLE B Observed dose-rate at 1 metre = 30 mr/hr. and measured soil sample specific gamma activity 0.38 $\mu c/cm^3$.

It is apparent that there is reasonable agreement between the observed dose-rate at 1 metre and the measured sample activity on any assumption about the true thickness of the active layer. More important, the dose-rate to the rat is in any case of the same order as that measured at 1 metre. A perfect two-dimensional surface distribution of activity is unlikely a priori, and even if present, small irregularities of the surface would reduce the dose-rate appreciably. It is thus likely that the dose-rate to the rat in November 1953 was close to 30 mr/hr.

(c) Beta Radiation Dose-Rate From Soil

Of the various beta groups in Table A, the ^{106}Ru beta rays can be disregarded since the maximum range even in air is only about 2 cm. The ^{106}Rh and ^{144}Pr rays can be lumped as a single groups with maximum energy 3.1 Mev; the ^{144}Ce rays form a second group. The relevant constants are given in Table C.

Group	Max. Energy, Mev.	k r/mc. hr. at 1 cm.	$\mu 1$, cm^{-1}	$\mu 2$, cm^{-1}
1	3.1	400	7.7	4.6×10^{-3}
2	0.26	800	230	0.14

TABLE C Beta Radiation Characteristics

The beta-ray activities associated with the possible levels of gamma-ray activities given in Table B may be derived by reference to Table A. The corresponding beta-ray dose-rates to a rat at height 2.5 cm. for the various cases are given in Table D, allowing a factor of $\frac{1}{2}$ for self shielding by the rat. The backscatter for a surface layer of activity has been taken as 20 per cent, and zero for a thick layer.

Assumed Thickness of Active Layer cm.	Activity		Dose-Rate, rep/hour		
	Group 1 ^{106}Rh - ^{144}Pr	Group 2 ^{144}Ce	Group 1 ^{106}Rh - ^{144}Pr	Group 2 ^{144}Ce	Total
0	1.205 $\mu c/cm^2$	0.602 $\mu c/cm^2$	7.1	1.4	8.5
0.1	13.71 $\mu c/cm^3$	6.86 $\mu c/cm^3$	0.55	0.17	1.8
1.0	2.02 $\mu c/cm^3$	1.01 $\mu c/cm^3$	0.59	0.025	0.62
∞	1.05 $\mu c/cm^3$	0.52 $\mu c/cm^3$	0.31	0.013	0.32

TABLE D Beta Radiation Dose-Rates to Rat

It appears likely that the beta radiation dose-rate to the rat from the soil in November 1953 was about 1 rep/hr.

(d) Beta Radiation Dose-Rate from Fur

The composition of the active material on the fur of the rat was virtually the same as that in the soil (Table V). The total activity on the pelt consisted in effect of 3.45 μc of high energy beta radiation (Group I, Table E) and 1.72 μc of low energy beta radiation. The skin dose is to be estimated for a conventional depth of 5 mg/cm^2, and two possibilities are considered:-

(i) All the activity is strictly on the skin surface.

(ii) The activity is uniformly distributed in a $\frac{1}{2}$ cm. layer of fur whose overall density is 0.1.

The surface area of the rat is about 400 cm^2. The fur of the rat now corresponds to the infinite plane of active soil while the tissue corresponds to the air in the first problem. The results are given in Table E.

Assumed Thickness of Active Fur cm.	Activity		Dose-Rate, rep/hour		
	Group 1	Group 2	Group 1	Group 2	Total
0	0.00863 $\mu c/cm^2$	0.00432 $\mu c/cm^2$	0.074	0.010	0.084
0.5	0.0173 $\mu c/cc.$	0.00863 $\mu c/cc.$	0.041	0.0086	0.050

TABLE E Beta Radiation Dose-Rates to Rat from Contaminated Fur

The dose-rate at the basal layer of the skin is thus not greatly dependent on the quantity of fur; a representative figure for November 1953 would appear to be 0.05 rep/hr. or 1 rep/day.

APPENDIX II

DOSE-RATE TO SKELETON, RAT 14

(a) Beta Radiation Dose-Rate

The isotopes present in the rat skeleton in November 1953 are given in Table IV. It may be estimated that at this time, ^{89}Sr and ^{90}Sr are present in equal amounts, each corresponding to $\frac{1}{3}$ of the combined amount (3.9×10^{-4} μC/g); the Y^{90} accounts for the remaining $\frac{1}{3}$. The individual activities, with the maximum energy of the beta ray spectrum, are given in Table F.

Isotope	^{106}Ru	^{106}Rh	^{89}Sr	^{90}Sr	^{90}Y	^{144}Ce	^{144}Pr
Max. Energy Mev.	0.04	3.2	1.5	0.6	2.4	0.26	3.0
Activity 10^{-4}μc/g.	7.5	7.5	1.3	1.3	1.3	0.6	0.6

TABLE F ACTIVITIES IN SKELETON, RAT 14, 1.xi.53

The absorption coefficient of the hardest beta rays (^{106}Rh) is about 7 cm^{-1} in bone. The dimensions of the bone in the rat skeleton are a few mms., and so it is a reasonable approximation to assume complete absorption of all the beta ray energy in the bone. The corresponding dose-rate is 5.9×10^{-2} rep/day. At the bone surface the dose-rate is one-half this value.

(b) Gamma Radiation Dose-Rate

The gamma radiation activity in the rat skeleton consists of 2.85×10^{-4} μc/g. for the gamma ray of 0.61 Mev and 0.18×10^{-4} μc/g. for the gamma ray of 0.08 Mev (See Table IV and Table A of Appendix I). It may be estimated that for a bone thickness of 2 or 3 mms, the energy absorbed in the bone is only about 1 per cent for the 0.61 Mev gamma ray, and 2 per cent for the 0.08 Mev gamma ray. Therefore the dose-rates in the bone due to these gamma rays are about 10^{-4} rep/day and 2×10^{-6} rep/day respectively. More precise estimates are unnecessary because these dose-rates are negligible compared with those due to the beta radiations.

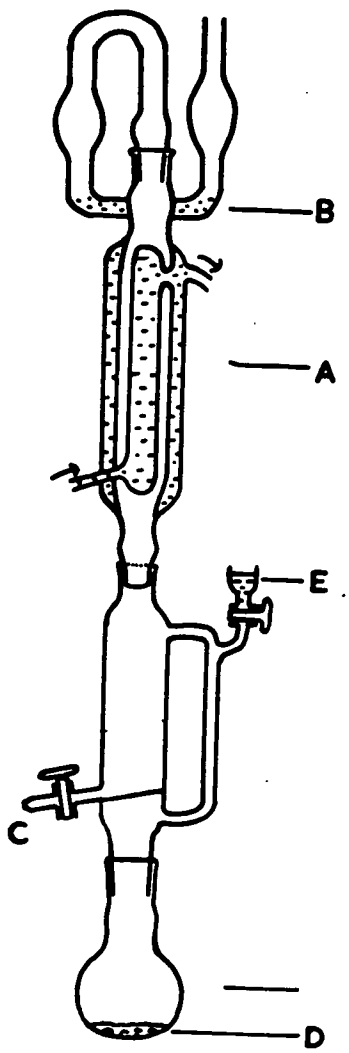


FIG. 1 Distillation apparatus.

A - double surface condenser
B - liquid trap
C - draining tap
D - Flask
E - nitric acid condenser.