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The Entry of Fission Products into Food Chains

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SUMMARY

(a) Investigations carried out both at Maralinga during Operation Buffalo and in the United Kingdom are described.

(b) The extent to which fallout was retained on pastures of types familiar in European agriculture was variable but, when the particle size of the deposition was relatively small, 15–20% of the deposit was found on the foliage. Less retention is, however, likely when a deposit is of large particle size.

(c) The retention of fallout by ripe ears of wheat appeared to be a minor source of hazard to man relative to the contamination of pastures grazed by dairy cattle.

(d) The solubility of fallout deposited on the ground surface, and also collected from radioactive cloud by aircraft, was examined. Relatively high solubility, indicative of ready biological availability, was observed in all cases except the near-in ground deposit after a tower shot. Even this material showed appreciable solubility.

(e) Experiments with rabbits and cows showed that the biological availability of fission products collected from the radioactive clouds of three weapons was not significantly reduced by its physical form. Some evidence was obtained that the same was true for the near-in ground deposit after a ground shot (Round 2).

(f) Studies of the metabolism of fallout by sheep, rabbits and cows confirmed expectations that isotopes of iodine and strontium are the major sources of hazard through ingestion. Shorter lived fission products such as tellurium and molybdenum were metabolized to an appreciable extent; they were, however, shown to be minor sources of hazard relative to the isotopes of iodine and strontium.

(g) The extent to which different isotopes of iodine contribute to the dose to the thyroid soon after the detonation of a weapon was examined.

(h) The results indicate that valid conclusions regarding the metabolism by animals of fission products from fallout can be reached by laboratory experiments using soluble preparations of chemically separated isotopes.

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SECTION 1: INTRODUCTION

THIS report contains the results of investigations of the movement of fission products into food chains which were undertaken in connexion with Operation Buffalo. The work was sponsored jointly by the Medical and Agricultural Research Councils, with the exception of the study of the contamination of cereals, which was proposed by the Ministry of Agriculture, Fisheries and Food.

The retention of fallout on vegetation, its solubility and its metabolism by animals were investigated at Maralinga by members of the Biological Target Response Group (Sections 2, 6 and 7 of this report). In addition, fallout was collected in filters carried by aircraft and sent by air to the United Kingdom for more detailed studies of metabolism by animals; these studies were conducted at the Medical Research Council Radiobiological Research Unit (Section 4) and the Agricultural Research Council Radiobiological Laboratory (Sections 3 and 5). In collaboration with the Ministry of Agriculture, Fisheries and Food, the latter laboratory also examined the extent of contamination of cereals which had been exposed at Maralinga (Section 3). The results of the investigation as a whole are reviewed in Section 8.

Apart from the members of the staff of the two Research Councils and their collaborators in the United Kingdom and Australia whose names appear as authors of Sections of this report, many others contributed to the work. The investigations with cultivated plants and with animals at Maralinga necessitated considerable preparations before the United Kingdom staff reached Australia. The Commonwealth Scientific and Industrial Research Organization coordinated these arrangements. Dr. F. R. G. White, then Chief Executive Officer, CSIRO, the late Prof. J. G. Wood, Chairman of the South Australian Committee of CSIRO and Mr. H. R. Marston, FRS, Chief of CSIRO Division of Biochemistry and General Nutrition, acted as general advisers; Mr Angus Packham, Technical Secretary of CSIRO Division of Biochemistry and General Nutrition, undertook administrative duties. Special metabolism cages for studies with sheep were designed by Mr. Marston who also provided the sheep from his laboratory and seconded two members of his staff, Messrs. I. G. Jarrett and B. J. Potter, to supervise their maintenance. Over 250 boxes containing growing pasture plants of European types were prepared in Adelaide under the direction of Prof. C. D. Donald. Messrs. N. Ford (CSIRO) and G. Sharman (University of Adelaide) collected biological specimens from a base at Emu during the operation.

Considerable voluntary assistance was given at Maralinga by officers of the Indoctrinee Force.

The writers of this report record their thanks for all the assistance they received.

SECTION 2: PHYSICAL CHARACTERISTICS OF FALLOUT AND ITS RETENTION ON HERBAGE

By R. Scott Russell* and J. V. Possingham†

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2.1. INTRODUCTION

Two questions of fundamental importance arise in the assessment of the hazards which would result from the deposition of fallout on land and its entry into food chains: its biological availability and the extent to which it lodges on

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edible vegetation. Variations in its composition due to "fractionation" in the cloud could in certain circumstances be a further important factor. Work undertaken to obtain information on these subjects is here described.

The "relative biological availability" of a fission product in fallout may be defined as the ratio of the extent to which it enters biological systems to that of a soluble preparation of the same isotope. A value of 1 thus indicates that the physical form of the fission product in question has not reduced the extent to which it can be absorbed; lower values indicate that the form of the fallout has significantly reduced its availability. No absolute meaning can, however, be ascribed to such measurements; they are qualitative rather than quantitative. The possibility cannot be ignored that whereas one organism may absorb fallout and soluble carrier-free preparations of the same isotope to the same extent, another may discriminate between them. However, both for the practical purpose of Civil Defence and for the planning of research such measurements are of considerable value; if the "relative biological availability" of fallout material approximates to 1, information on the metabolism of the fission product can be obtained validly and simply from investigations in which chemically separated isotopes are used as tracers. The biological availability of fission products can be studied in two ways, namely:

(a) By comparing the quantities of a given isotope which test organisms absorb when provided with weapon debris and with chemically-separated material prepared from an appropriately irradiated target.

(b) By examining the solubility of weapon debris in water, carriers or buffer solutions.

The latter provides considerably less information but it has the advantage of speed and simplicity. Results obtained by this method are given first below; biological experiments are described in later sections of this report.

The extent to which fission products are retained by the leaves of plants is another major factor which determines the magnitude of ingestion hazards in the period immediately following the detonation of atomic weapons. No quantitative information on this question was available when Operation Buffalo was planned. The retention of contamination by herbage of the types which grazing animals consume is a question of particular importance as the most widespread ingestion hazard to the human population is likely to arise through the contamination of milk; furthermore, the exposure of the animals themselves merits consideration. As the nature of herbage affects the extent to which it retains fallout, the arid native Australian vegetation was unsuitable for assessing the situation which would occur in European pastures. Accordingly, pasture species were grown in boxes to produce swards comparable to good pastures in temperate regions. These boxes, which served also to collect fallout for experiments on the metabolism of animals, were exposed in the fallout area. The herbage was subsequently assayed and autoradiographs were prepared.

There is considerable evidence that the rapidity with which fission products condense can influence the extent to which they are trapped in large particles and become deposited close to Ground Zero. Species which have gaseous or volatile precursors may therefore be deposited to a relatively small extent near the target. The extent to which they are deposited may be expected to be correspondingly enhanced at great distances. ^{89}Sr (a product of the decay of

TABLE 1

Total Deposition of Fission Products per Unit Area for Rounds 1 and 2 at Collecting Sites

Values are shown only for rounds on which the sites were used.

Collection site ref.	Fission products deposited per unit area, (mc/m^2) at 1 hr	
	Round 1	Round 2
A	—	847
B	—	799
C	—	364
D	—	186
E	—	176
F	69	67
G	320	111
H	417	153
I	1515	242
J	1390	211
K	—	223
L	—	89
M	—	34
N	—	22
O	—	21
P	—	12
Q	21	—
R	118	—
S	—	2·97
T	—	2·18
U	<1·39	—
V	<1·39	—
W	1·81	—
X	3·61	—

^{89}Kr through ^{89}Rb) and ^{90}Sr (a product of the decay of ^{90}Kr) are therefore likely to be depleted in the localities close to the target. While little effort could be deployed to investigate this question, some work was undertaken.

It was realized that Operation Buffalo would provide a particularly favourable opportunity for investigating these questions; the contrasting manners in which the first three rounds were to be detonated would cause wide differences in the physical properties of both airborne fission products and the ground deposition. Since round 1 was a tower shot the fallout would, to varying extents, be entrapped in fused material from the ground surface which had been drawn up into the fireball. Round 2 was to be detonated on the ground surface; American experience[1] had shown that fallout from such explosions was mixed with considerable quantities of unfused debris and that its biological availability was therefore greater than in material from tower shots. Round 3, which was to be dropped from an aircraft and detonated above the ground, would produce fallout associated with little, if any, surface debris, and local fallout was expected to be relatively slight. These circumstances guided the planning of the work: the major effort was devoted to work on round 2, since it appeared likely to produce the worst conditions.

Samples of airborne debris collected in filters carried by aircraft were obtained from rounds 1, 2 and 3. Collections of vegetation contaminated with fallout were made after rounds 1 and 2 only.

The nature of the terrain made it necessary to confine the greater part of the ground collection to the road system accessible from Maralinga. This limited the collection of herbage from distant sites but arrangements were made for some samples to be collected 100–150 miles from Ground Zero. Difficulties of transport and delays in the firing schedule, however, caused the number of such samples collected to be small. It was impracticable to set out herbage boxes between the time of detonation of the weapons and the deposition of fallout, and the relatively wide limits of acceptable local wind conditions for firing made it impossible to predict accurately the path of the short and medium range fallout. Thus it was necessary to set the boxes in an arc exceeding 90°. In consequence only a few were contaminated. The extent of fallout at the collection sites is shown in Table 1.

2.2. EXPERIMENTAL METHODS

2.2.1. *Types of Herbage Investigated*

(a) *European pasture species grown in boxes*—Boxes of surface area 0·29 m^2 (approximately 75 cm long by 40 cm wide) and 20 cm deep were planted with ryegrass (*Lolium perenne*) or ryegrass and clover (*Trifolium repens*) 9 months before the trial (Fig. 1). The boxes were prepared at WRE, Salisbury, S. Australia, under the guidance of Prof. C. M. Donald, University of Adelaide. A peat–soil mixture of high waterholding capacity was employed so that the boxes could retain sufficient water to prevent the wilting of plants in a period of 2–3 days. One month before the trial the boxes were transported by rail to Maralinga and were conveyed to the field in the first day of "Standby". Carrier slings were provided so that the boxes, which weighed approximately 200 lb, could be carried by two men (Fig. 2). Wooden frames 10 cm high and covered with wire netting could be fixed to the tops of the boxes so that they could be stacked in transit without injury to the foliage. It was considered that the covers might be necessary to protect the boxes from kangaroos and other native fauna in the fallout area. No such protection, however, proved necessary and a comparison of the contamination of boxes with and without covers during round 1 indicated no significant differences.

(b) *Native species*—Native vegetation of types consumed by grazing animals were sampled, viz. *Atriplex* sp., *Kockia* sp., *Erodium* sp., *Danthonia* sp. and *Stipa* sp. In practice no significant differences were observed between species; however, the extent of contamination of plants of the same species was so variable that it would have been possible to establish the significance only if very large differences occurred. The general appearance of the vegetation is shown in Figs. 3 and 4.

2.2.2. *Sampling of Herbage*

All samples were placed in polythene bags immediately they were collected. In round 1 a significant but very variable quantity of fallout became dislodged in the bags. This was ignored, it being assumed that similar dislodgement of particles would be caused by grazing animals or wind. After round 2 such losses were small.

Samples from herbage boxes were severed at ground level. Native species were represented as closely as possible by the tissues grazing animal would consume, i.e. the new season's growth of grasses or young twigs of *Atriplex* sp.

2.2.3. *Collection of Fallout*

Double layers of surgical gauze 1 m^2 in area were attached to sheets of polythene 1 m wide and 1½ m long and laid out on the ground surface. To prevent the gauze being moved by wind the ends of the polythene sheets were rolled around lengths of angle iron (Fig. 4). Surface soil samples were collected from some localities after round 2.

2.2.4. *Assay of Total Activity of Herbage Samples*

The prime requirement in this work was to relate the activity in the samples to that deposited on the ground surface. Accordingly, a procedure as similar as possible to that adapted by the Radiological Measurements Group for the assessment of sticky papers was adopted. Proportional counters and a β-scintillation counter (anthracene) were compared in preliminary work and the latter was selected on grounds of speed and reliability.

Dried herbage samples weighing 1–10 g were placed flattened in polythene bags approximately 5 in. square. The bags were sealed and held by a spring-loaded tray approximately ⅜ in. below the anthracene. The equipment for this purpose was modified by the Nuclear Instruments Section from that devised for counting sticky papers.

The equipment was calibrated with sections of "sticky paper" and herbage samples, which had been assayed by the Radiological Measurements Group. The mean of eight such calibrations showed the efficiency for gross β-activity to be 17%. The background counting rate was approximately 280 counts/min.

Geometric errors were investigated by changing the position of samples within the bags and recounting them for both sides. The degree of reproducibility varied somewhat depending on the nature of the samples; agreement within 2% was common and the mean variation between replicate counts was less than 5%. Self-absorption factors were determined by placing different weights of uncontaminated herbage on top of filter papers saturated with fallout. Since the herbage samples would be contaminated on both sides it was assumed that self-absorption would attenuate radiation from a contaminated sample of a given weight to the same extent as an uncontaminated sample of half that weight attenuated the radiation from a source placed below it. A correction curve for self-absorption could thus be drawn; it was approximately linear. Duplicate counts of all samples were recorded on a minimum of three occasions spaced over at least 12 days; in the majority of cases more frequent counting was possible. Decay curves were fitted graphically; the values for the point in time midway between the first and last counting occasions were extrapolated back to 1 hr using decay factors determined by the Radiological Measurements Group for fallout from the weapon in question.

This assay procedure was clearly open to greater errors than if samples had been ground or ashed and presented to a counter in a more fixed geometry. However, the errors of the method were shown to be small relative to variation in the activity of replicate samples. Furthermore, the procedure had the advantage of speed, high sensitivity and easy cross-calibration with the measurements of deposition per unit area by the Radiological Measurements Group.

Some specimens collected towards the close of the Operation could not be assayed prior to the return of the party to England. These samples were digested in nitric acid and counted as solutions with glass Geiger–Müller counters (type M.6) by Dr. Helen M. Squire, at ARC Radiobiological Laboratory, Compton. Fourteen samples which had been assayed in the β-scintillation counter at Maralinga were similarly treated to provide a basis for cross-calibration. Determinations of ^{89}Sr and ^{140}Ba were made on a limited number of samples.

2.2.5. *Preparation of Autoradiographs of Contaminated Herbage*

Specimens of pasture and native herbage were mounted between sheets of thin Cellophane on wadding pads backed by plywood (25 × 20 cm). The Cellophane was drawn tightly around the boards to retain the specimens in position. The approximate exposure times were determined with a Geiger–Müller counter attached to a type 1021 radiation monitor and the specimens were then exposed to X-ray film in a "deep freeze". Lead sheets covered in polythene were employed both to keep the film in close contact with the mounted specimens and to prevent radiation from one specimen penetrating to the films exposed to other specimens in the same container.

2.3. SOLUBILITY OF FALLOUT

2.3.1. *Ground Collections Examined Soon After Detonation*

In Table 2 results are shown for fallout collected on gauze sheets after round 1. Only three sheets were contaminated. They were extracted for successive 24-hr periods in distilled water (pH 6·5) and in 0·01 M strontium and barium nitrates. Considerable variability between replicate samples from the same sites was observed. Material from site R, the site of lowest contamination, showed over 9% solubility of the total of both treatments. The sites of higher contamination gave values of about half that figure.

The fallout from round 2, the ground shot (Table 3), was soluble to the extent of up to 25% in water and carrier solutions. The decay rates of the extracts of material collected at two sites differed markedly, the decay factors from day $D + 4$ to day $D + 18$ being: site B, 14; site I, 40. Solubility in buffer at pH 4 was also examined in material from one site; the mean value was 49%. The higher solubility in round 2 than in round 1 is attributed to the fact that it was less associated with fused material.

The extraction procedures used were unlikely to displace all labile ions. The results are, however, indicative of considerable solubility. The increased values induced by buffer extraction are believed to be attributable to the fact that the underlying rocks which contained 80% $CaCO_3$ were the major source of particulate material in the fallout.

TABLE 2

Round 1: *Percentage Solubility of Fallout Deposited on the Ground*

Examined 4–16 days after detonation; duration of extraction in each solution was 24 hr.

Collection site ref.	Extractant	Number of replicates	Percentage solubility	
			Range	Mean
G	Water	4	2·2–5·4	3·3
	Carrier	2	0·5–0·6	0·6
	Total			3·9
H	Water	4	3·8–7·2	5·9
	Carrier	2	0·5–1·2	0·9
	Total			6·8
R	Water	2	4·8–8·9	6·8
	Carrier	2	2·2–2·8	2·5
	Total			9·3

TABLE 3

Round 2: Percentage Solubility of Fallout Deposited on the Ground

Examined 5–18 days after detonation; duration of extraction in each solution was 24 hr.

Collection site ref.	Extractant	Number of replicates	Percentage solubility	
			Range	Mean
B	Water	2	12·3–16·9	14·6
	Carrier	2	3·4–5·6	4·5
	Total			19·1
I	Water	3	18·7–27·2	22·3
	Carrier	2	3·7–10·6	7·2
	Total			29·5
I	Buffer (pH 4)	3	43·0–57·7	49·0

2.3.2. *Ground Collections Examined* 6 *Months After Detonation*

Gauzes exposed at four sites during round 2 were subjected to prolonged extraction with water and buffers of decreasing pH, 6 months after the trial. The results are shown in Table 4. Extraction at each pH was continued until further removal was slight; over the prolonged period of extraction (over 50 days) at least two-thirds of the activity was removed.

TABLE 4

Round 2: Percentage Solubility of Fallout Collected on the Ground and Subjected to Successive Extraction with Buffer Solutions 6 Months After the Trial

The figures in brackets indicate the duration of extraction in days. The extraction was completed at day $D + 238$ in each case.

Site ref.	pH of buffer			Total extraction (%)
	6·5	4	3	
B	1·0	20·0	54·2	75·2
	(2)	(27)	(36)	
I	1·3	25·0	47·5	73·8
	(2)	(27)	(36)	
L	1·3	1·9	63·6	66·8
	(7)	(8)	(36)	
M	0·9	0·5	73·5	74·9
	(7)	(8)	(36)	

2.3.3. *Contaminated Soil*

Samples of soil from the 0–1, 1–4 in. horizons at site A were collected after round 2 and returned to the United Kingdom for examination. Prolonged shaking with successive aliquots of ammonium acetate at pH 3 removed 33–34% of the total activity; approximately half this quantity is extracted at pH 5. Because of the difficulty of reaching definite conclusions regarding the availability of ions in soil from results of chemical extraction, this work was not carried out on a more extensive scale. The present findings are, however, suggestive of appreciable availability at this site. The particle size of fallout was smaller at all other sites covered by these investigations; it was therefore likely to be at least as available.

It had been intended to carry out plant culture experiments so that the absorption of ^{90}Sr by plants from fallout could be compared with that of chemically separated preparations; a precise measurement of the relative biological availability of the weapon fallout would thus be obtained. In the summer of 1957 when it had been intended to carry out this work the total activity of ^{89}Sr plus ^{90}Sr in the soil from the 0–1 in. horizon was considerably less than 0·5 mμc/g. Only a few per cent of the ^{90}Sr in soil is absorbed by plants in a single season and this level of contamination was therefore too low for accurate results to be obtained using the facilities then available. Both for this reason and because considerable evidence of high biological availability had already been obtained in experiments with animals this work was not undertaken.

2.3.4. *Air Collections*

Airborne debris collected in aircraft filters after round 2 was examined at Maralinga on $D + 18$. The solubility in water (24 hr) followed by carriers (24 hr) for material from high and low levels in the cloud was 42·5 and 49·5%, respectively. The corresponding values for extraction in buffer solutions were 66·0 and 59·4%.

Air filter material from rounds 1 and 3 was examined by the Medical Research Council Radiobiological Research Unit, Harwell, on $D + 5$. Values for solubility in water during extraction periods of 24 hr were 34 and 32% for rounds 1 and 3, respectively. Chemical analysis indicated that the solubilities of ^{132}Te and ^{99}Mo were approximately 30% while the values for ^{140}Ba and ^{89}Sr were 80% and 45% respectively. Results for the extraction of activity from these filters by carrier solutions are shown in Section 5 of this report.

2.4. THE RETENTION OF FALLOUT ON HERBAGE

2.4.1. *Round* 1

Most of the herbage boxes were reserved for round 2, as the experiments with animals were then to take place. Only twenty were therefore exposed to round 1

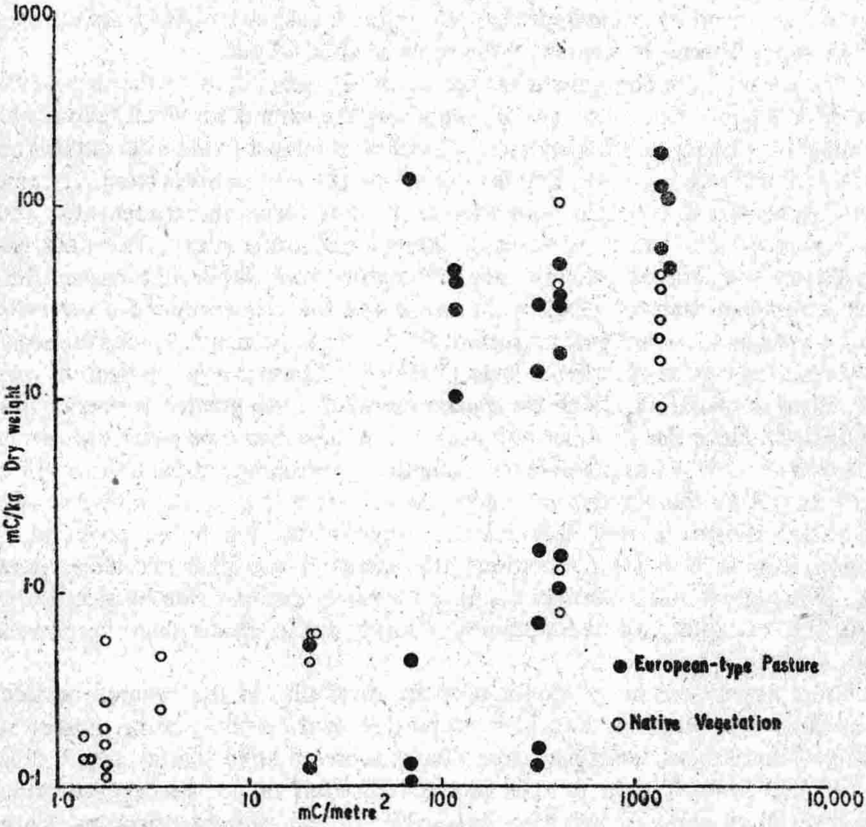

FIG. 5. Round 1. The relationship between the activity per unit weight of dry herbage and the deposition per m² of ground surface.

and of these but ten, which had been exposed at seven sites, were contaminated to an adequate extent. Replicate samples were taken from these boxes but the greater part of the work on round 1 was devoted to the examination of native plants of the following species: *Stipa*, *Atriplex* and *Erodium* spp. In Fig. 5 the activity per unit dry weight of herbage samples is related to the deposition per m^2 of ground surface. A very wide scatter is apparent in the contamination of herbage exposed to the same level of fallout.

The ratio of the contamination per kg of dry vegetation to the deposition per m^2 is a convenient measure for comparing the extents to which fallout was retained by herbage at different sites. This ratio is related to the total deposition of fission products per m^2 in Fig. 6. Variations between samples from the same site with respect to the ratio were often as great as variation between sites, and there was no consistent evidence of interspecific differences. The data are inadequate for detailed analysis; accordingly no more elaborate treatment has been attempted than to calculate the range and the mean values for the ratio mc/kg herbage to mc/m^2 ground surface for herbage and native species in localities contaminated to different extents (Table 5). There is some indication that the retention of fallout, by native species especially, was greater at sites of low deposition. Since the yield per unit area of the European type pasture grown in boxes was known it was possible to calculate the percentage of the fallout which was retained by the vegetation. As would be expected from the above results the values showed a very wide scatter. Eight of the ten boxes gave figures ranging from 0·06 to 1·8% retention; the values for the other two were 5 and 6%. The highest and lowest results were for boxes exposed side by side at the same site; variability was therefore not primarily due to the distance of the boxes from Ground Zero.

There appears to be no doubt that the variability in the results obtained from this round were due to a high proportion of the activity being present in relatively infrequent large particles. Truly representative figures could thus be obtained only if each sample assayed consisted of the herbage covering relatively large areas. This was impossible in the present operation since 10–20 g dry weight was the maximum sample size convenient for assaying.

Autoradiographs were prepared of representative samples (Figs. 7 to 9). The general characteristics were:

(a) Contamination consisted entirely of discrete particles; in no case was the outline of the leaves apparent as would be the case if material had been absorbed into the tissues.

(b) The distribution of particles on the leaf surface wa highly irregular but contamination appeared to be particularly great in angles where particles could lodge. This was especially conspicuous in the sheathing bases of grass leaves (Fig. 7). The dissection of specimens showed that the contamination per unit weight of tissue was considerably greater in the basal portions of the plants.

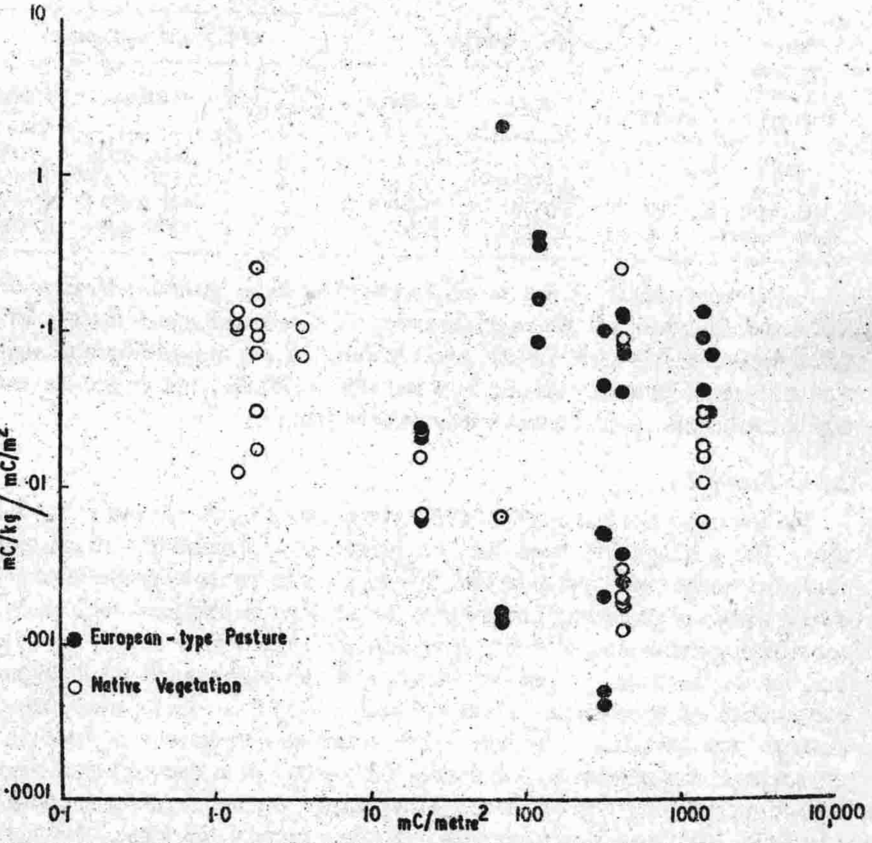

FIG. 6. Round 1. The relationship between the total fission product deposition per m² of ground surface and the ratio of contamination per kg of dry herbage to the deposition of fission products per m².