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RESEARCH GROUP

Physical measurements of activity in samples from Windscale.

D. V. BOOKER

Atomic Energy Research Establishment,

Harwell, Berkshire.

1958

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PHYSICAL MEASUREMENTS OF ACTIVITY IN SAMPLES FROM WINDSCALE

by

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ABSTRACT

Gamma scintillation spectrometer measurements in samples of milk, herbage and soil from the Windscale area are described. The geographical distribution of activity is studied, and the relative activities of I^{131}, Cs^{137} and Sr^{89} are discussed. A correlation between I^{131} in herbage and milk is recorded, and an estimate of the total deposition of I^{131} on herbage is made.

U.K.A.E.A. Research Group,
Atomic Energy Research Establishment,
HARWELL

October, 1958.

/RH

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1. Introduction

An accident in Windscale Pile No. 1, on October 10th, 1957, caused a release of activity from the pile stack. This activity was predominantly volatile fission products, especially I131, and radioactivity was found to be deposited on the surrounding countryside. Soil, herbage, water and food for human consumption from this area were sampled and tested for radioactivity at Windscale, Harwell and Woolwich.

This paper describes in detail the methods used for analysis by gamma spectroscopy of samples from Windscale, in one laboratory at Harwell, and the results obtained. In addition results from other laboratories both at Harwell and Windscale have been used to build up as complete a picture as possible of the distribution of fall-out activity. These results were made available to the author by Templeton (1958) at Windscale and by Clare and Humphries (1958) and Bryant et al (1958) at A.E.R.E.

In the first few days after the incident I131 activity in the samples was predominant to such an extent as to mask other activities. By retaining samples until the I131 had decayed the presence of other nuclides could be demonstrated. Cs137, Ru103 and 106, and Zr95 with their daughter products were found in herbage and soil, and Cs137 also appeared in milk.

2. Methods

Measurement of gamma activity in samples observed at Harwell were made using gamma scintillation spectrometers. Several different instruments were used in the measurements: a five-channel pulse amplitude analyser with a 3" x 3" diameter crystal of thallium activated sodium iodide; a one-hundred channel analyser with a 3" x $1\frac{3}{4}$" diameter crystal; another one-hundred channel analyser with a 2" x 4" diameter crystal.

2.1 Milk

At the time of the accident the five-channel analyser and 3" x 3" crystal were already set up for the study of Cs137 activity in dried milk (Booker 1957). It was very quickly adapted for measurement of I131 in liquid milk, and the first results were obtained within a few hours of the receipt of samples on the morning of October 12th.

In milk the only gamma-ray emitting contaminants which were observed were Iodine isotopes and Cs137. I131 is characterised by its 0.364 Mev gamma-ray (80% of disintegrations) in conjunction with gamma-rays of 0.080 Mev (2.2%), 0.284 Mev (53%), 0.637 Mev (9%) and 0.722 Mev (3%). (Strominger et al 1958). Cs137 emits a 0.662 Mev gamma ray in 92% of disintegrations and about 10% of these are internally converted. (Strominger et al 1958). One early sample of milk was observed to give a 0.53 Mev gamma-ray which was attributed to I133 (21 hr half-life), but its intensity was very small compared with the 0.637 Mev gamma-ray, from I131. Because of the 9% branching of I131 to give the 0.637 Mev gamma-ray, the 0.662 Mev gamma-ray from Cs137 in milk was swamped until about a month after the accident when the I131 level in milk had fallen by a factor of about fifty from its initial activity.

The I^{131} in milk was estimated by placing pint bottles of milk on the crystal. The five-channel analyser was adjusted so that the four lowest channels straddled the 0.364 Mev I^{131} peak which was of such intensity that it swamped counts from either Cs^{137} or naturally occurring K^{40} in milk. The counting rate from the sum of the four channels was compared with the rate obtained from a standard solution of I^{131} (Figure 1). The standard was made by dilution from a solution calibrated by N.P.L. It was found necessary to add several hundred milligrams of iodine carrier to the pint of the diluted solution together with some caustic soda in order to prevent iodine depositing on the glass surface of the bottle. All results quoted are in true source strength and not gamma source strength. Where nuclides have daughter products, the results are in terms of the activity of the parent nuclide.

2.2 Herbage

Herbage samples were measured for I^{131} content on the same crystal and analyser. Herbage, which had been cut from a known area, was weighed, thoroughly mixed, and a weighed portion (of about 100 g) put in sixteen ounce polyethylene jars. The geometry for counting was fixed, but the weight varied from sample to sample and fell as the herbage dried out. The density was low enough that any self absorption was very small. A comparison of counting rates was made using standards of the same volume containing vermiculite and a known volume of calibrated I^{131} solution. Although the polythene jars were not made air tight, repeated analysis of samples showed no evidence of loss of iodine by volatilization. Previous experience with surfaces contaminated with carrier-free iodine had shown that adsorption is very strong. In this case there was a possibility that the iodine was absorbed into the herbage tissue.

Other gamma-ray emitting nuclides occurred in the herbage, although in almost all samples the iodine gamma-ray predominated. The ratios of the counting rates in the four channels of the five-channel analyser looking at the 0.364 Mev peak of I^{131} in grass, were compared with the ratios from a standard I^{131} source. The constancy of these ratios in all but a few samples indicated that no other gamma-ray was giving an appreciable counting rate at this energy. Results for I^{131} in herbage on an arc from Pembroke to Norfolk were obtained by Clare and Humphries (1958) using a 100 channel analyser, which allowed an estimate of the contribution to the counting rate which was due to nuclides other than I^{131}.

As the I^{131} activity in the samples from the Windscale area decayed other gamma-ray emitting nuclides could be estimated using a one-hundred-channel analyser. The efficiency of the gamma spectrometer was found using I^{131} (364 Kev and 637 Kev), and Ba^{140} - La^{140} (335 Kev, 820 Kev and 1580 Kev) mixed with vermiculite.

2.3 Soil

Soil samples were obtained from cores of known depth and area. The samples were dried by exposure to air at room temperature and weighed, crushed and well mixed. A weighed quantity of soil was put in polythene bottles of sixteen ounce capacity. The samples were estimated for I^{131} on the five channel analyser when the activities were high, and by D. Humphries on a hundred-channel analyser for the low activities. A sample curve obtained from soil containing high I131 content is given in Figure 2, where the interference from other fission product gamma-rays and natural activities is small. Also illustrated is part of the spectrum from the same sample of soil measured six months later on a hundred-channel analyser, when the Cs137 content was being estimated. The interference due to the Zr95 - Nb95 gamma-rays at about 750 Kev and I131 gamma-ray at 637 Kev has decreased by decay. Using the known resolution of the instrument, an allowance has been made for the residual Zr95 - Nb95 activity and for the 606 Kev gamma-ray from RaC occurring naturally in soil. The contribution to the counting rate in the Cs137 peak due to Compton recoils from high energy gamma rays is also estimated. On soil samples where the interfering activities are small compared with the Cs137 activity, estimates of the Cs137 in soil are quite accurate; but estimates of Cs137 in soil far from Windscale are only approximate.

3. Results

A map of the district near Windscale is reproduced in Figure 3 giving the topography of the district.

3.1 Activity in milk

3.1.1 I131 in milk

The I131 activity in samples of milk measured in Harwell varied from less than 0.3 mμc/l (from North Cumberland, South-West England, and East Anglia) to well over 1000 mμc/l for a few samples from points close to Windscale.

A daily series of samples was obtained and analysed at Windscale to study the change in I131 activity in milk with time. Although the dissemination of I131 from the pile reached a peak in the early hours of October 11th, and ceased by about noon on that day, the activity did not appear in the milk of the local cows until the afternoon of the 11th. The maximum activity in milk was reached on the afternoon of the 12th or the morning of October 13th as shown in Table I.

TABLE I.

Change of I131 activity in milk with time

Time of milking		Sellafield μc/l	Seascale μc/l
11/10/57	am	0.003	-
	pm	0.47	0.4
12/10/57	am	0.48	0.85
	pm	1.32	0.6
13/10/57	am	0.39	0.94
	pm	1.12	0.63
14/10/57	am	0.54	0.51
	pm	0.72	0.67

After 14/10/57 the activity of milk fell off with time as shown in Table II.

TABLE II.

Apparent half-life of I131 in milk

Location	Apparent half-life	Period followed
	days	days
Seascale	4.7	Oct. 14 - Nov. 12
Sellafield	4.7	Oct. 14 - Nov. 11
Corney	5.5	Oct. 14 - Nov. 14

A graph of the fall of I131 in Seascale milk as a function of time is given in Figure 4.

The average value (5.0 days) for the apparent half-life is much shorter than the radioactive half-life of eight days. The difference may be due to the following reasons, but other explanations are possible.

(a) The I131 **per unit** weight of herbage was probably being reduced by biological processes and weathering.

(b) The cows from the time of the accident onwards were getting an increasing amount of concentrated food and would be grazing progressively less grass.

Using the recorded results produced by Windscale of all I131 in milk measurements made at Harwell and Windscale, the I131 concentrations in milk samples collected between October 13th and 19th have been tabulated and worked back to October 11th using a five-day half-life. In doing so the initial delay before the appearance of the I131 in milk has been ignored. The tabulated results have been plotted on maps of the area (Figures 5 and 6) and contours are drawn of the level of I131 activity in milk around Windscale and in North Lancashire. Many of the spot values of I131 in milk are the average values of determinations made on milk collected on different days during the period October 13th to October 19th. A similar tabulation of results for October 20th - 25th worked back to October 11th using a five-day half-life shows very similar features and supports the five-day half-life. Inspection of the contours in Figures 5 and 6 shows that some activity was blown ten or twenty miles in a north-easterly direction from Windscale, and then swept down in a southerly or south-easterly direction. As shown in Figure 6 some of the activity was carried across north-east Lancashire and into Yorkshire, but in general most of the cloud passed down the Midlands west of the Pennines. This picture of the fall-out distribution agrees with what might be expected from the meteorological conditions and with observations on air filters from different parts of England (Chamberlain and Dunster 1958, and Stewart and Crooks 1958).

3.1.2 Cs137 in milk

In the first two weeks after the accident large numbers of milk samples were counted, and were discarded after measurement of I131. A Seascale sample of 28th October was preserved, and when the I131 had decayed to a low enough level to enable the 0.662 Mev peak from Cs137 to be measured, the Cs137 content was found to be 16 mμc/l. In the period November 8th - 11th a more extensive series of milk samples was taken for Cs137 analysis. For this series, the pattern of distribution of Cs137 in milk, shown in Figure 7, was very similar to the I131 in milk pattern. A maximum of Cs137 activity of 15 mμc/l was found at Corney which is on rising ground in the direction of the plume and 15 Km from the factory. High levels of Cs137 in milk were found in a north-easterly and south-easterly directions, but levels in milk were much lower in other directions. Milk from the farm at Seascale previously sampled was included in the series and showed 11.5 mμc/l a decline of only 30% in the eleven day period since October 28th.

Continuation of the measurement of Cs137 in dried milk, which has been studied for the past two years (Booker 1957), has shown that the level of Cs137 in dried milk increased temporarily after the accident at all sampling stations south of Windscale but not at Carlisle or Mauchline which are north of the factory.

TABLE III

Cs137 in dried milk

Cs137 activity in $\mu\mu c$/gm Potassium

Location	Before 11/10/57 $\mu\mu c/g$.	After 11/10/57		Difference Due to Accident
		Date	$\mu\mu c/g$.	
Garstang	40	1/11/57	140	100
Carmarthen	80	5/11/57	105	25
Buckingham	30	9/11/57	55	25
Driffield	30	27/10/57	55	25
Frome	40	15/11/57	55	15
Carlisle	40	1/11/57	40	-
Mauchline (Scotland)	50	6/11/57	45	-

Results are quoted in terms of $\mu\mu c$ Cs137 per gram of potassium. The concentration of potassium in liquid milk is about 0.2% by weight so that the concentration of Cs137 due to the incident in liquid milk from Garstang and Buckingham for example would be approximately 0.2 $m\mu c$/l and 0.05 $m\mu c$/l respectively. These results are in reasonable relation to the Cs137 levels in Cumberland milk at about the same date (See para. 4.3). The high level in the Carmarthen milk is not explained. It is possible that some of the Cs137 may be due to high fall-out from bomb tests in Nevada during October. However no extra Cs137 appears north of Windscale. The excess Cs137 disappeared rapidly from the dried milk after November, and this was no doubt associated with the change in feeding of the cows from pasture to stored food.

3.2 Activity in herbage

3.2.1 I131 in herbage

Herbage samples were collected from many different points in the areanear Windscale and from various arcs up to two hundred miles from the stack. One particular field in Seascale, which was not being grazed, was selected for regular sampling in order to follow the "decay" of activity in herbage. The apparent half-life of I131 activity in herbage per unit area of ground was found to be approximately five days (Figure 8) in very close, although perhaps fortuitous, agreement with the apparent half-life in milk from the same farm (Figure 4). The details of the results are given in Table IX. It will be noticed that the weight of herbage cut from unit area decreased with a somewhat similar period (except for the first sample). This may be due to the fact that at this time of year the herbage is gradually dying back, but many other explanations can be brought forward. The observed figure of five days has been used for working back to the day of release of activity, the I131 activity in herbage from other areas. This procedure can be justified by:-

(a) Figures observed of total activity in herbage and soil at Seascale on October 28th (see para. 3.3) when worked back with the radioactive half-life of I131 give a total deposition of 19 $\mu c/m^2$ on October 11th as compared with an extrapolated figure of 17 $\mu c/m^2$ using a five-day half-life on herbage only (Figure 8).

(b) The figures for activity in herbage from the same area taken at different dates are consistent.

(c) Results can be compared with a gamma dose-rate survey carried out in the district. Chamberlain (1958) gives a correlation factor between the dose-rate as observed with a 1413A monitor on various dates, and total initial deposition of I131. Table IV gives a comparison of some results from the present herbage analyses and results using the correlation factor. The average ratio of these two estimates is 0.82.. The wide spread of ratios in the last column of Table IV may be due to:-

   (i) Survey measurements were not always done on the same day as the herbage was sampled and are not certain to be in exactly the same position as that from which the herbage was sampled.

   (ii) The 1413A monitor is sensitive to activity within a radius of at least thirty meters, while the herbage samples represent only a few square meters which would not necessarily give a figure equivalent to that from the surrounding area.

TABLE IV

Comparison of herbage activity with deposition calculated from the gamma dose-rate

Location	Map Reference	Herbage activity.*		Total deposited activity from gamma survey.		Ratio
		Date of sample	I131 μc/m^2 on 11/10/57	Date of survey	I131 μc/m^2 on 11/10/57	Herbage μc/m^2 / Total μc/m^2
ROF Drigg	061990	18/10	12	26/10	22.4	0.5
Drigg	052986	13/10	13.3	13/10	9.9	1.3
Ravenglass Jn.	090967	13/10	3.7	13/10	4.8	0.8
Seascale	047018	13/10	17	13/10	17	1.0
Swallowhurst	102910	13/10	7.5	14/10	4.6	1.6
Bridge End	122946	17/10	2.8	17/10	3.1	0.9
Corney	115914	17/10	5.7	24/10	10.3	0.6
Wasdale Head	187085	16/10	0.8	14/10	1.3	0.6
Bootle	110878	18/10	3.5	26/10	4.7	0.7
Duddon	198882	18/10	0.2	14/10	2.1	0.1
Whicham	133824	18/10	2.1	14/10	2.3	0.9
Torver	285943	18/10	1.35	29/10	1.6	0.8
Saltcoats	079972	18/10	4.6	18/10	17.4	0.3
Black Coombe	132832	24/10	5.1	24/10	4.0	1.2
Black Coombe	136855	24/10	7.3	24/10	12.5	0.6
Black Coombe	143872	24/10	15.6	24/10	19.8	0.8
Stoneside Hill	147894	24/10	11.7	24/10	9.2	1.3
						Mean 0.82

* Worked back to 11th October using five-day half-life.

Figures 9 and 10 give spot values of I131 in herbage in millimicrocuries per square metre of ground for samples collected between October 13th and 28th worked back to October 11th using a five-day half-life. It is possible to draw approximate contours of the levels of activity on herbage, and the shape of these contours is found to agree well with those obtained from the map of the activity of I131 in milk. In the region of the Lancashire - Yorkshire border, where there are no herbage measurements, some account of the I131 in milk contour is taken in drawing the herbage contour. The eastern parts of the 10 and 100 millimicrocuries per square metre contours are conjectural. On the assumption that the fallout of Cs137 was proportional to that of I131, the level of Cs137 in milk from the collecting centre at Driffield quoted above can be used to deduce that the I131 fall-out in the East Riding of Yorkshire was in the range 10 - 100 $m\mu c/m^2$. It is known from results of air samples (Stewart and Crooks 1958) and from gamma monitoring, that little activity reached County Durham and the North Riding, but the levels in the West Riding were comparable with, but lower than those in Lancashire.

3.2.2 Other activities in herbage

After sufficient time had elapsed for the I131 in herbage to decay to a negligible level, the following gamma-emitting isotopes were identified. The energies and half-lives of the different nuclides are taken from Strominger et al (1958).

(1)	Cerium	Observed gamma ray	140 Kev
	Ce141	Half-life 33 days.	Gamma ray energy 142 Kev
	Ce144	Half-life 285 days.	Gamma ray energy 134 Kev

The observed gamma ray was predominantly Ce141 as confirmed by following the decay curve of one sample.

(2)	Ruthenium	Observed gamma ray	500 Kev
	Ru103	Half-life 40 days.	Gamma ray energy 498 Kev
	Ru106→Rh106	Half life of Ru106 - 1 year	Gamma ray energy of Rh106 513 Kev

The observed gamma-ray was predominantly Ru103 as confirmed from the decay curve of one sample. The absence of a gamma-ray at 624 Kev which occurs in the spectrum of Rh106 at half the intensity of the 513 Kev gamma-ray is further evide..ce that the observed line is due predominantly to Rh103, although Cs137 peak at 662 Kev would partially mask the 624 Kev peak.

(3)	Caesium	Observed gamma ray	660 Kev
	Cs137	Half-life 33 years	Gamma ray energy 662 Kev
(4)	Zirconium-Niobium.	Observed gamma-ray 750 Kev.	
	Zr95	Half-life 65 days.	Gamma-ray energy 722 Kev
	Nb95	Half-life 35 days.	Gamma-ray energy 765 Kev

In the results Zr95 activities are quoted, assuming that, at the time of measurement, the Zr95 and Nb95 were in equilibrium. The ratio of activities of Zr95 and Nb95 when in equilibrium is 6:13.

Tables IX and X give a list of results for activities in herbage, as activity per gram wet weight of herbage, and activity per square metre of ground. For the Seascale herbage samples the series of results given in Table IX for Ru103 and 106, Cs137, and Zr95 microcuries per square metre are plotted in Figures 11, 12 and 13 respectively. The activity of each of these elements falls sharply with time for the first few weeks, followed by a gradual decrease in the decay rate. The Zr95 activity reaches an approximately constant level of 0.0025 $\mu c/m^2$ which is due probably to bomb-test fall-out*

The I131/Cs137 activity ratio in grass from Seascale is found to decrease exponentially with a half-life of 9.5 days (Figure 14). In other words the I131 was disappearing from the herbage (by physical and biological processes) at a slightly slower rate than the Cs137. Using this empirical half-life on I131/Cs137 ratios in grass from other places, it is found that the ratios worked back to October 11th were from 20-80 in the direction of the plume, and along Eskdale and Langdale, but some ratios of over 500 were found in other directions (see Table X). A ratio of I131/Cs137 activity of 64:1 was found in a filter paper air sample, which was in operation at Calder during the peak of the release at 10 am on October 11th (Chamberlain and Dunster 1958).

3.3 Activity in soil

Samples of soil were taken on October 28th and 31st from six different points from the fall-out area, with herbage samples from the same area. Later a control sample from Carlisle was taken. At each station ten sub-plots of one square yard each were marked out within an area of an acre. The herbage from these plots was cut and then a four inch diameter, four inch deep core of soil plus matt was taken from the centre of each square yard. Comparison of I131 in the herbage and the soil plus matt (Table V), shows that at the end of October an average of 16% (with range 7% to 31%) of the I131 was in the herbage. On the assumption (a) that originally 82% was in the herbage (Table IV), (b) that the activity per square metre in herbage decayed with a five-day half-life and (c) that the activity per square metre of herbage + soil + matt decayed with an eight-day half-life, it would be expected that, on October 28th, 34% of the activity would have been on the herbage.

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* During October and November 1957 a total of 0.007 $\mu c/m^2$ of Zr95 in fall-out was observed in rain water collected at Chilton Berks (Stewart 1958), a proportion of which would be retained by herbage. Morgan (1958) has found the Sr89 content in permanent grass at Chilton from bomb-test fall-out to be about 0.0025 $\mu c/m^2$ in each of the months September, October and November 1957 and most of this is ascribed to foliar uptake. Zr95 has a 40% greater yield and 20% longer half-life than Sr89. These results suggest that where the I131 fall-out from Windscale was low the Zr95 observed is due to bomb-test fall-out.

TABLE V

Activity in herbage and soil*

	Location	Seascale	Sellafield	Pennington	Corney	Bootle	Millom	Carlisle	Average
	Date	28/10	31/10	28/10	28/10	28/10	28/10	7/11	
I131	Soil $\mu c/m^2$ Herbage $\mu c/m^2$ Total $\mu c/m^2$	3.64 0.70 4.34	3.76 0.68 4.44	1.01 0.077 1.09	2.16 0.20 2.36	0.35 0.16 0.51	1.49 0.33 1.82	- - -	
	% in herbage	16	15	7	8	31	15	-	16
Cs137	Soil $m\mu c/m^2$ Herbage $m\mu c/m^2$ Total $m\mu c/m^2$	170 39 209	260 - -	89 - -	180 34 214	50 29 79	56 13.4 69	<15 - -	
	% in herbage	19	-	-	16	37	23	-	24
Sr90	Soil $m\mu c/m^2$ Herbage $m\mu c/m^2$ Total $m\mu c/m$	60.6 0.35 61.0	180 0.98 181	11.5 0.11 11.6	20.7 0.19 20.9	14.6 0.20 14.8	17.9 0.33 18.2	8.5 0.05 8.55	
	% in herbage	0.6	0.5	1.0	0.9	1.4	1.8	0.6	1.0

*Including the matt of live and dead roots with the soil.

The Cs137 activity in the soil is also shown in Table V. It seems that the fraction of Cs137 in herbage is higher at this date than the fraction of I131 in herbage, even though some of the Cs137 in the soil is due to bomb-test fall-out (probably less than 15 m c/m^2).

Also included in Table V are the results of Sr90 measurements made at Woolwich on the same samples (Bryant et al. 1958). The low proportion of Sr90 in the herbage relative to that in the soil indicates that most of the Sr90 in the soil is not associated with this incident.

4. Discussion of results

4.1 Foliar contamination

The results of the comparison of activity in herbage and gamma dose-rate (Table IV) and of activity in herbage and in soil plus matt (Table V) show that initially a high proportion of the I131 was deposited on the herbage. The degree to which metabolic processes enhanced the radioactive decay of activity from the herbage is not clear, but there is no evidence at all of appreciable translocation from soil to herbage, at this time of the year.

4.2 Relative activities of I131, Cs137 and Sr89

In Table VI are given the absolute and relative values of I131, Cs137 and Sr89 in herbage and milk at Seascale, and the relative activities in a filter paper exposed at Calder during part of the release. The Sr89 figures for the herbage and milk are quoted from Bryant et al. (1958) using a figure of 1.2% for the calcium content of milk. The Sr89 level of 0.0008 μc/l in milk on 13/10 was the highest observed at Seascale in the period following the accident. The Calder filter figures are quoted from Chamberlain and Dunster (1958).

TABLE VI

Relative activities of I131, Cs137 and Sr89 in Seascale herbage and milk

Nuclide	Calder Filter		Herbage activity			Milk activity		
	Date	Relative	Date	μc/m^2	Relative	Date	μc/l	Relative
I131	11/10	100	13/10	12.7	100	13/10	0.8	100
Cs137	11/10	2.6	13/10	0.18	1.4	28/10	0.0016	2.6
Sr89	11/10	1.0	13/10	0.024	0.19	13/10	0.0008	0.1

The relative amounts of these nuclides in the three types of sample are in reasonable agreement, but there is evidence that the activity of Sr89 in herbage and milk was less than would have been expected if the deposition to herbage and uptake by cattle had been as effective as that of I131 and Cs137.

4.3 Variation of deposition with distance.

The variation of I131 deposition can be followed in the approximate direction of the main plume from the figures in Table VII, which have been plotted in Figure 15. If the same decrease with distance is assumed for the Cs137 in milk as is observed with I131 for both herbage and milk, extrapolation would lead to an expected level of about 0.1mμc Cs137/l of milk at Wellingborough. This can be compared with a figure of 0.05mμc/l obtained with dried milk from Buckingham (see 3.1.2) at about the same time. Buckingham is some miles off the centre of the plume.

TABLE VII

Variation of contamination with distance from Windscale

Location	Distance Km.	I131		Cs137	
		On ground μc/m² on 11/10	In milk μc/l on 11/10	On ground mμc/m²	In milk mμc/l on 8/11
Seascale	3.2	18.9	1.04	209	11.4
Corney	15	10.5	0.52	214	14.6
Millom	28	7.9	0.5	69	3.7
Pennington	36	4.8	0.5	89	3.6
Oswaldtwistle	104	0.6*	0.052	-	-
Macclesfield	150	0.14*	-	-	-
Wellingborough	290	0.08*	0.007.	-	-

* Herbage only

4.4 Correlation of milk and herbage activities

When the results for I131 in herbage are compared with the level of activity in milk from the same area a correlation can be obtained between the herbage activity and the milk activity. The median of thirty ratios shows that 11 μc I131/sq. metre of herbage produced 1 μc/l of milk (Figure 16). The spread of results is from 2 to 35 with a standard error of $\pm$ 2 μc/m².

4.5 Total deposition

Integration of the areas between each contour of the maps of Windscale and of England given in Figures 9 and 10 gives an indication of the total amount of I131 which was deposited on herbage in Great Britain. The geometric mean of the contours has been taken for estimating the fall-out between contour-lines. The details in Table VIII give a total of about 11,000 curies. As the whole plume was not covered when taking herbage samples this figure can only be approximate. A further deposition of about 2000 curies can be estimated in the Irish Sea south of the Furness peninsular. Stewart and Crooks (1958) give figures for the exposure of different towns in Europe due to I131 from Windscale. Comparing these with the figures of deposition on herbage shows that very approximately a further 5000 curies of I131 may have been deposited over the sea and in northern Europe giving a total figure of about 20,000 curies of I131 for the activity deposited from the Windscale accident.

TABLE VIII

Estimate of total I131 deposited in Great Britain

Level of activity mμc/m²	Area Sq. Km.	Assumed average activity mμc/m²	Total activity curies
> 10,000	30	15,000	450
1,000 - 10,000	480*	3,160	1,500
100 - 1,000	16,200	316	5,100
10 - 100	>123,000	31.6	> 3,900
			Total > 10,950

*Excluding contribution from deposition in Irish Sea south of Barrow.

5. Conclusions

5.1 Techniques

The versatility of the gamma-scintillation spectrometer has been demonstrated by the measurement of the gamma-ray activity of numerous biological samples in many different forms and of widely varying specific activity. One advantage which this method holds over conventional chemical separations is the small amount of preparation required for samples.

Measurements of the I131 content of milk were made at Windscale using crystals of one inch diameter and thickness, in conjunction with a single channel analyser. This was found to be sensitive enough to measure activities as low as 2 mμc of I131 per litre of milk when using samples of about 500 cc. The larger crystal which was used in Harwell (3" x 3") was found to be more sensitive, and levels of 0.2 mμc/ml could be estimated using one pint of milk. The five channel analyser provided a useful indication of the stability of the apparatus.

The multichannel analysers were found to be essential when several gamma-ray emitting isotopes were present in the samples. The large crystals (3" x 3" and 4" x 2") were necessary for measurement of activities in samples of low specific activity but small crystals would have been quite adequate for samples of soil and herbage collected near Windscale.

Owing to the nature of the Windscale accident, that the predominating release of activity was I131, gamma spectrometer measurements on milk with a single channel analyser were satisfactory. However should an accident occur in which the release was a normal fission-product mixture, it is possible that other gamma-ray emitters (such as Ba - La140) would appear in milk and a multichannel-analyser would be required for activity measurements. Soil and herbage measurements would certainly have to be made using a multichannel analyser.

5.2 Results

Measurements of I131 on herbage quoted in Table IV, where they are compared with the gamma dose-rate from the ground, show that initially a large proportion of the I131 was deposited on the herbage.

Figures quoted in Table VI show that I131 and Cs137 behaved very similarly in deposition from the air to herbage and in passage through the cow to milk, at a point about two miles south of Windscale. Sr89 behaved differently in the fact that less of it appeared in milk relative to the amount disseminated but most of the difference appears to be due to a lower rate of deposition of Sr89 from air to herbage.

The rate of decrease of I131 and Cs137 in herbage and milk with distance from Windscale appears to be similar as shown in Table VII. Even at a distance of about 200 miles from Windscale, evidence from the Cs137 content of dried milk shows that the ratio of Cs137/I131 in milk was very similar to that nearer to Windscale.

Contour maps of the I131 in milk (Figs. 5 and 6) show very similar features to the contour maps of I131 per square metre of herbage. (Figs. 9 and 10). An estimate of the total deposition of I131 has been made from the herbage contour maps, giving a figure of about 20,000 curies. This is in good agreement with estimates of total release made by Crabtree (1958) using the results of air sampling by Stewart and Crooks (1958).

Acknowledgements

I wish to thank Mr. D. W. Ironside of the National Agricultural Advisory Service and Mr. W. L. Templeton of Windscale and many other members of the U.K.A.E.A. staff at Windscale for the collection of samples. Many of the measurements of I131 in milk were made at Windscale, and the work done by Windscale staff is gratefully acknowledged.

I also acknowledge the assistance given by Dr. J. Rundo, especially with the use of his 100 channel analyser; and thank Mr. N. G. Stewart and Dr. F. J. Bryant for permission to quote unpublished results obtained by their groups.

I thank especially Mr. A. C. Chamberlain, with whom I have had many useful discussions about the interpretation of results given here.

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TABLE IX

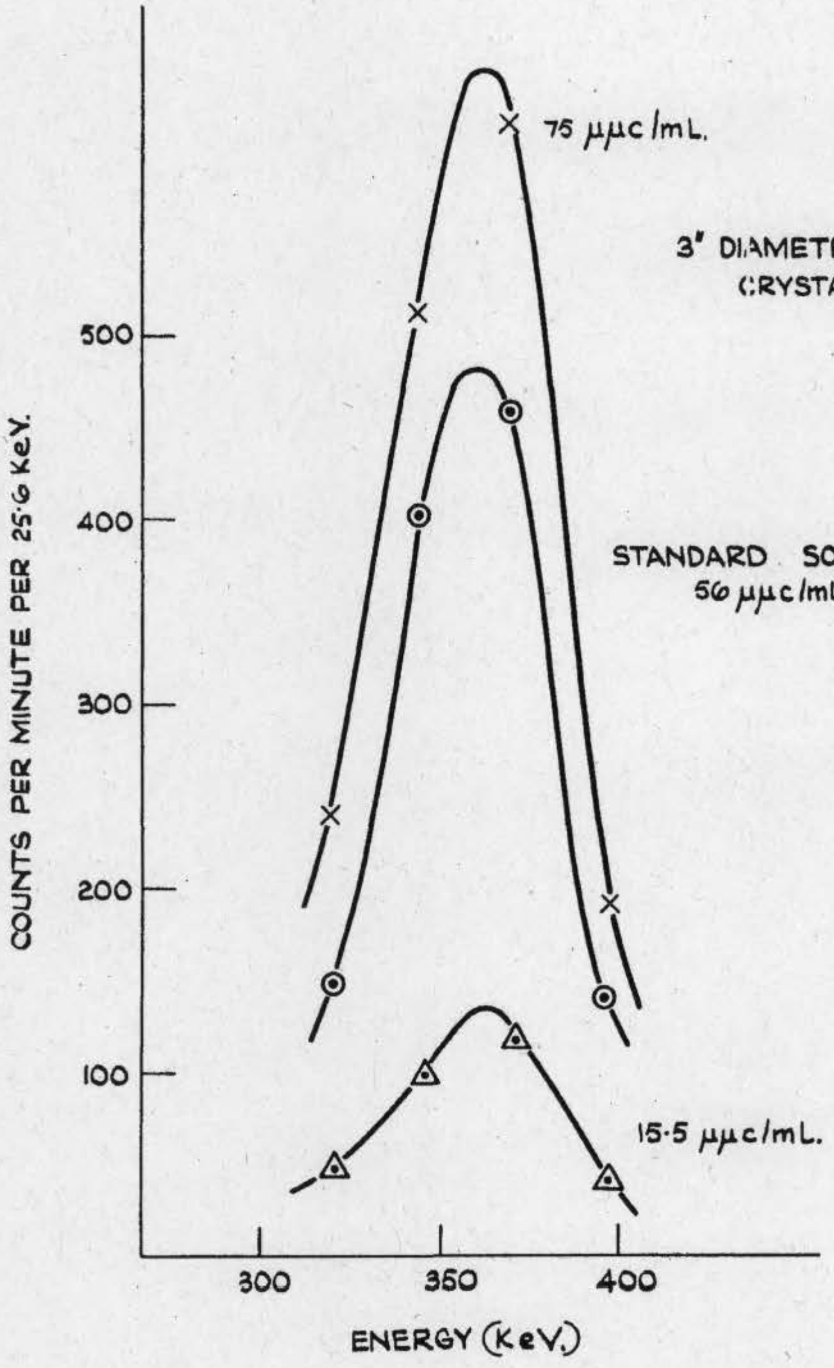
Weights and activities of herbage cut at Parkers Seascale

Location	Map Reference	Date of Collection	Wet wt.	Dry wt./Wet wt.	I^{131}		$Ru^{103} + Ru^{106}$		Cs^{137}		Zr^{95}		I^{131}/Cs^{137}	[illegible]
			gm/m^2	%	μμc/gm wet wt.	μc/m^2	μμc/gm wet wt.	μc/m^2	μμc/gm wet wt.	μc/m^2	μμc/gm wet wt.	μc/m^2		Worked back to Oct. [illegible]
Parkers Seascale	047018	13/10/57	348	29	36000	12.7	660	0.23	520	0.18	81	0.028	71	[illegible]
		18/10/57	1970	14	4000	7.8	96	0.19	100	0.20	47	0.093	38	64
					5300	10.5	140	0.27	105	0.20	20	0.039	51	84
		19/10/57	1580	36	2200	3.5	85	0.13	63	0.10	8.5	0.013	35	62
		21/10/57	1410	-	2100	2.95	81	0.11	66	0.094	9.1	0.013	32	65
		22/10/57	940	35	2400	2.3	56	0.052	61	0.057	9.5	0.0088	40	88
		23/10/57	1030	41	3500	3.6	95	0.098	100	0.11	16	0.016	34	80
		24/10/57	555	33	3200	1.8	88	0.049	100	0.055	25	0.014	32	83
		25/10/57	320	31	2600	0.83	-	-	-	-	-	-	-	-
		26/10/57	820	26	4000	3.3	98	0.081	135	0.11	21	0.018	30	87
		27/10/57	490	29	3300	1.6	90	0.044	120	0.066	37	0.018	27	85
		28/10/57	490	19	1400	0.70	45	0.022	53	0.026	14	0.0069	27	92
		28/10/57	775	25	1300	1.0	-	-	-	-	-	-	-	-
		29/10/57	386	36	2200	0.86	99	0.038	100	0.039	21	0.0082	22	81
		30/10/57	411	48	1200	0.50	51	0.021	61	0.025	12	0.0050	20	77
		31/10/57	780	34	1140	0.88	-	-	-	-	-	-	-	-
		1/11/57	690	27	640	0.44	39	0.027	46	0.032	6.3	0.0044	14	62
		4/11/57	436	-	1450	0.63	71	0.031	87	0.038	31	0.0013	17	94
		7/11/57	204	-	1100	0.23	73	0.015	95	0.019	18	0.0038	12	81
		11/11/57	118	-	2700	0.32	240	0.028	340	0.040	84	0.0098	8.0	75
		14/11/57	119	-	1200	0.15	220	0.026	190	0.023	54	0.0063	6.4	74
		17/1/58	424	-	-	-	9.7	0.0041	9.7	0.0041	9.1	0.0038	-	-
		18/2/58	118	-	-	-	24	0.0028	18	0.0021	23	0.0028	-	-
		10/3/58	66	-	-	-	32	0.0021	47	0.0031	41	0.0025	-	-

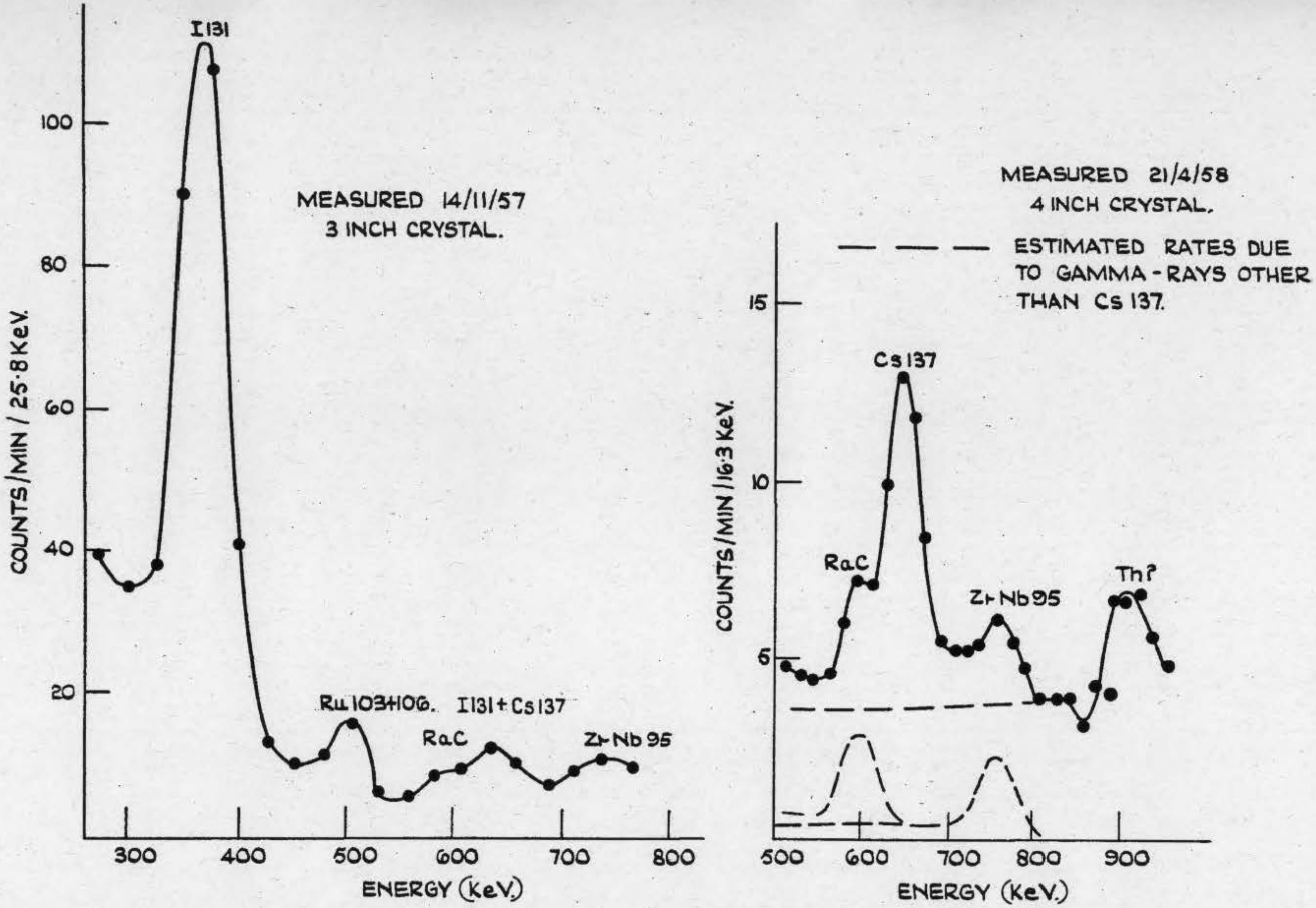
TABLE X

Spot activities of herbage cut near Windscale

Location	Map Reference	Date of Collection	Wet wt.	Dry wt./Wet wt.	I^{131}		$Ru^{103} + Ru^{106}$		Cs^{137}		Zr^{95}		I^{131}/Cs^{137}	I^{131}/Cs^{137}
			gm/m²	%	µµc/gm wet wt.	µc/m²	µµc/gm wet wt.	µc/m²	µµc/gm wet wt.	µc/m²	µµc/gm wet wt.	µc/m²		Worked back to Oct. 11
Drigg	052986	13/10/57	500	50	20000	10.1	420	0.21	490	0.245	78	0.035	41	48
Ravenglass Junction	090967	13/10/57	284	40	9900	2.8	240	0.069	160	0.046	44	0.013	61	71
Greenmoor Side	025056	13/10/57	241	29	14000	3.3	110	0.027	42	0.010	25	0.0059	330	380
Beckermet	014061	13/10/57	235	27	5600	1.3	53	0.012	23	0.0054	32	0.0074	240	280
Medlbank	080025	13/10/57	370	18.5	1350	0.49	38	0.014	3.0	0.001	26	0.0094	450	520
Swallowhurst	102910	13/10/57	553	23	10300	5.7	300	0.17	225	0.125	5.0	0.0032	46	54
Whitehaven	990183	17/10/57	1980	26	86	0.17	14	0.027	<0.5	<0.001	12	0.024	>170	>260
Rowrah	049187	17/10/57	2540	17.5	17	0.044	12	0.031	<0.5	<0.001	11	0.028	>34	>52
Cravensdale Farm	094174	17/10/57	1100	18	16	0.018	13	0.014	<0.4	<0.0005	13	0.015	>40	>62
Gillerthwaite Farm	136141	17/10/57	1300	14.5	53	0.068	12	0.015	0.8	0.001	7.5	0.010	66	100
Greendale Farm	152057	17/10/57	442	26	330	0.15	36	0.016	1.7	0.00075	22	0.010	190	290
Eskdale Green	148998	17/10/57	2230	14	1020	2.3	32	0.072	1.4	0.003	17	0.039	730	1100
Knott End	134976	17/10/57	1210	15.5	770	0.93	38	0.046	5.9	0.007	16	0.019	130	200
Bridge End	122946	17/10/57	1450	15.5	850	1.2	24	0.035	10.8	0.016	8.2	0.012	79	120
Corney	115914	17/10/57	1170	-	2400	2.8	96	0.11	89	0.10	13	0.015	27	41
Drigg	060988	18/10/57	1220	-	3700	4.5	190	0.23	230	0.28	15	0.019	24	40
Saltcoats	078972	18/10/57	1160	-	1750	2.0	110	0.13	140	0.16	5.0	0.006	16	26
Haverigg	156791	18/10/57	720	-	740	0.53	110	0.078	8.5	0.006	2.0	0.015	120	200
Croft End	247912	18/10/57	515	25	190	0.099	8	0.004	<0.6	<0.0003	7.9	0.0038	>320	>530
Bootle	110878	18/10/57	1030	18	1300	1.3	36	0.037	31	0.032	9.1	0.0094	41	68
Duddon Bridge	197882	18/10/57	760	17	100	0.077	4	0.003	1.4	0.001	4.4	0.0031	70	110
Parsonage	133824	18/10/57	1040	13.5	760	0.79	19	0.019	22	0.023	7.9	0.008	35	57
Torver	285943	18/10/57	1140	24	450	0.51	16	0.018	9.5	0.001	12	0.013	470	780
Coniston	366988	18/10/57	660	17	71	0.047	9	0.006	<0.6	<0.0004	6.3	0.0041	>120	>200
Raven Crag	400080	18/10/57	436	20	150	0.065	11	0.005	3.3	0.0014	5.4	0.0023	45	74
Beyond Coniston	302978	18/10/57	840	16.5	31	0.026	13	0.01	<0.6	<0.0005	9.8	0.0082	> 56	> 93
Corney	118919	28/10/57	1200	28	250	0.30	-	-	29	0.034	24	0.029	8.7	30
Millom	178850	28/10/57	950	15	350	0.33	-	-	14	0.013	17	0.016	24	83
Bootle	082906	28/10/57	630	15	260	0.16	-	-	46	0.029	21	0.013	5.7	18
Seascale Golf Course	030020	15/11/57	525	-	2000	1.0	250	0.13	150	0.080	160	0.082	13	160
Braystones	016058	17/11/57	396	-	74	0.029	12	0.0048	1.8	0.0007	7.3	0.0028	41	600
Beckermet	021064	17/11/57	189	-	150	0.028	115	0.022	1.9	0.0004	8.2	0.0016	78	1100
Ponsonby	053052	17/11/57	190	-	77	0.015	21	0.004	4.1	0.0008	13	0.0025	19	270
Lane Head	042022	18/11/57	1120	-	780	0.88	57	0.064	57	0.065	2.5	0.0028	14	210
Moss Side Wide	050026	18/11/57	253	-	260	0.066	32	0.008	4.5	0.0011	16	0.0041	56	660
Calder Bridge	044065	18/11/57	295	-	34	0.010	14	0.004	2.9	0.0009	8.5	0.0025	11	180



A.E.R.E. HP/R 2607. FIG. 1. SAMPLE CURVES OF MEASUREMENT OF I131 IN MILK USING THE FIVE CHANNEL ANALYSER.



A.E.R.E. HP/R.2607. FIG. 2. SAMPLE CURVES OF MEASUREMENTS OF SOIL ACTIVITY. SEASCALE SOIL.

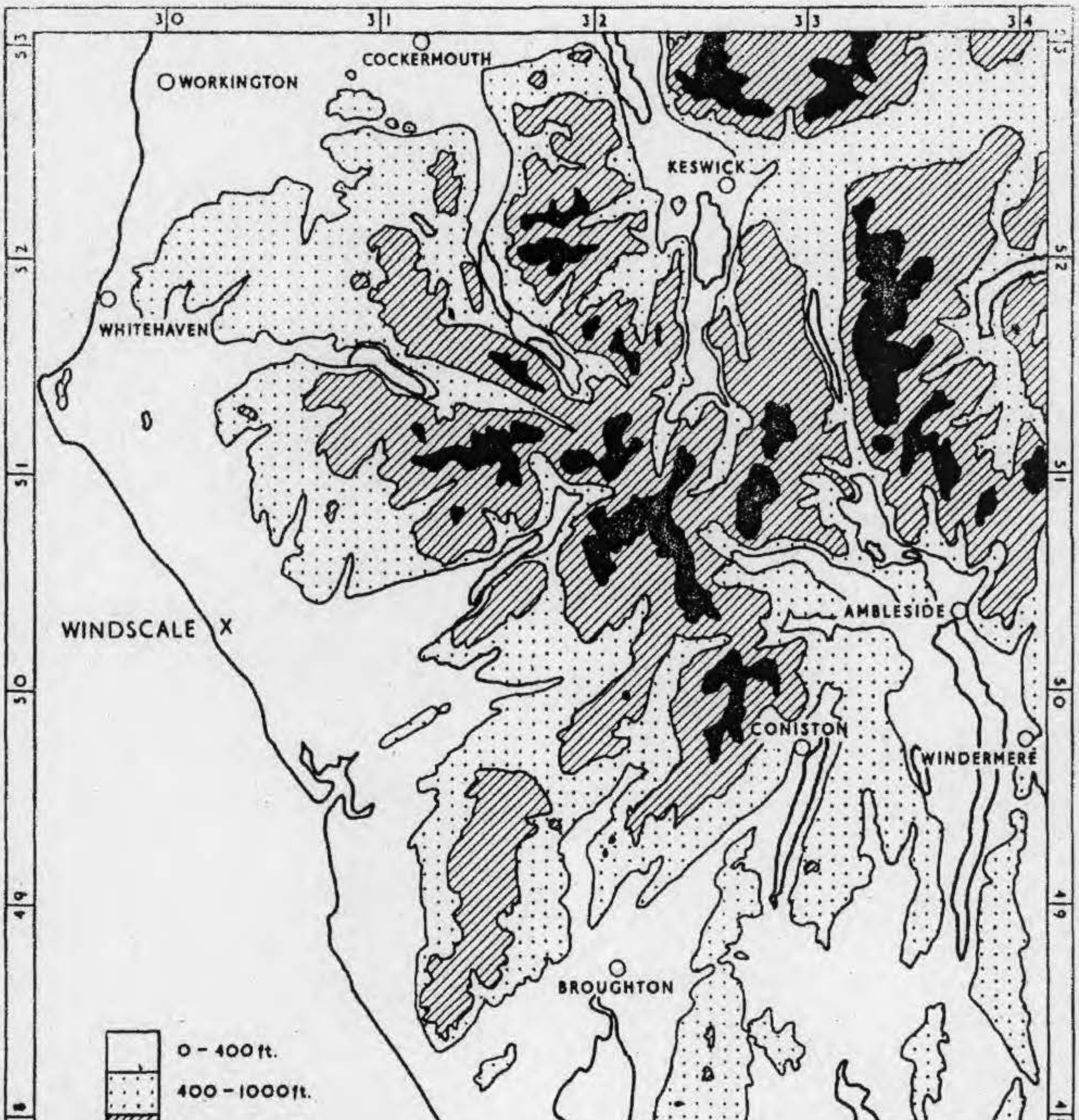
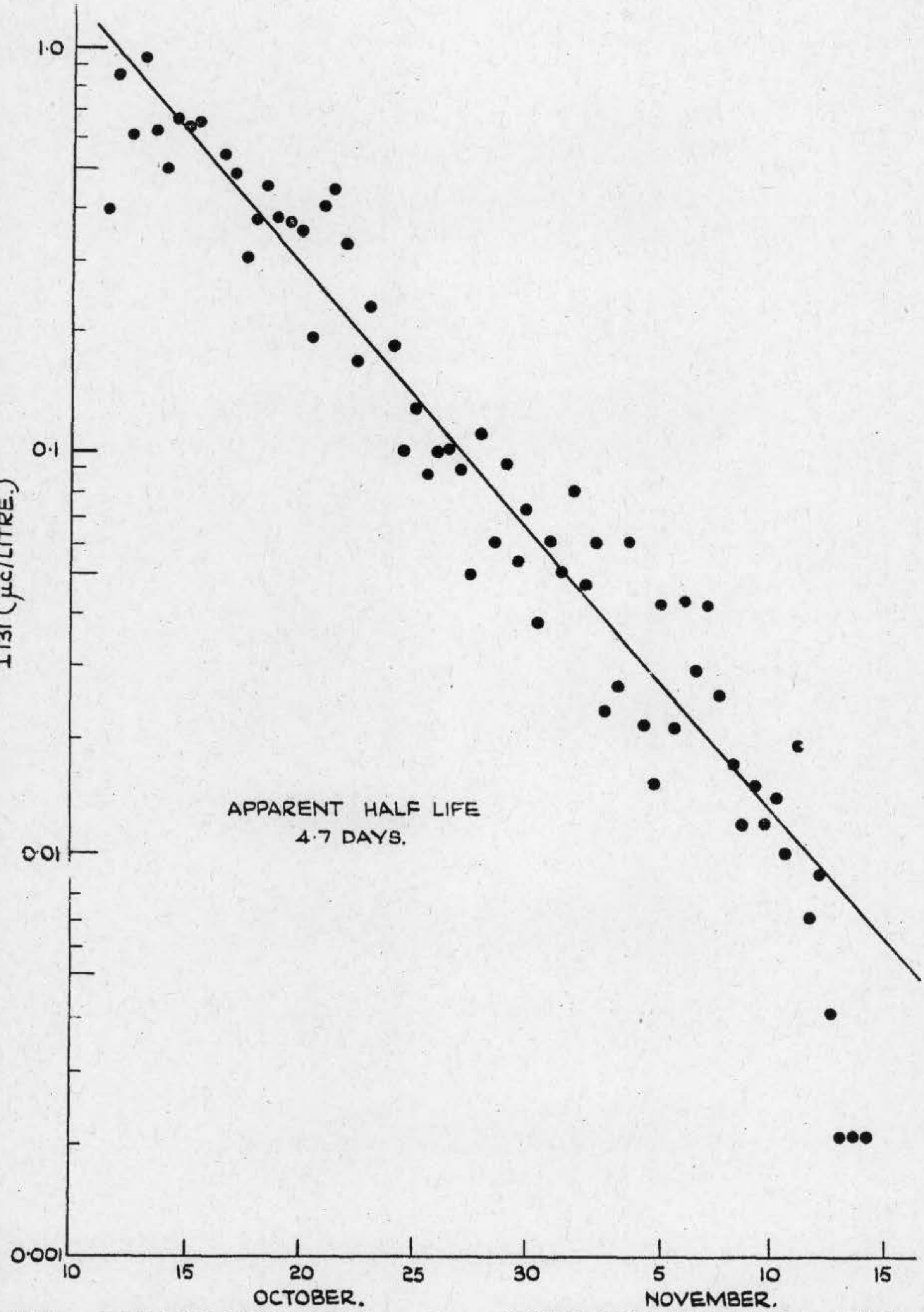
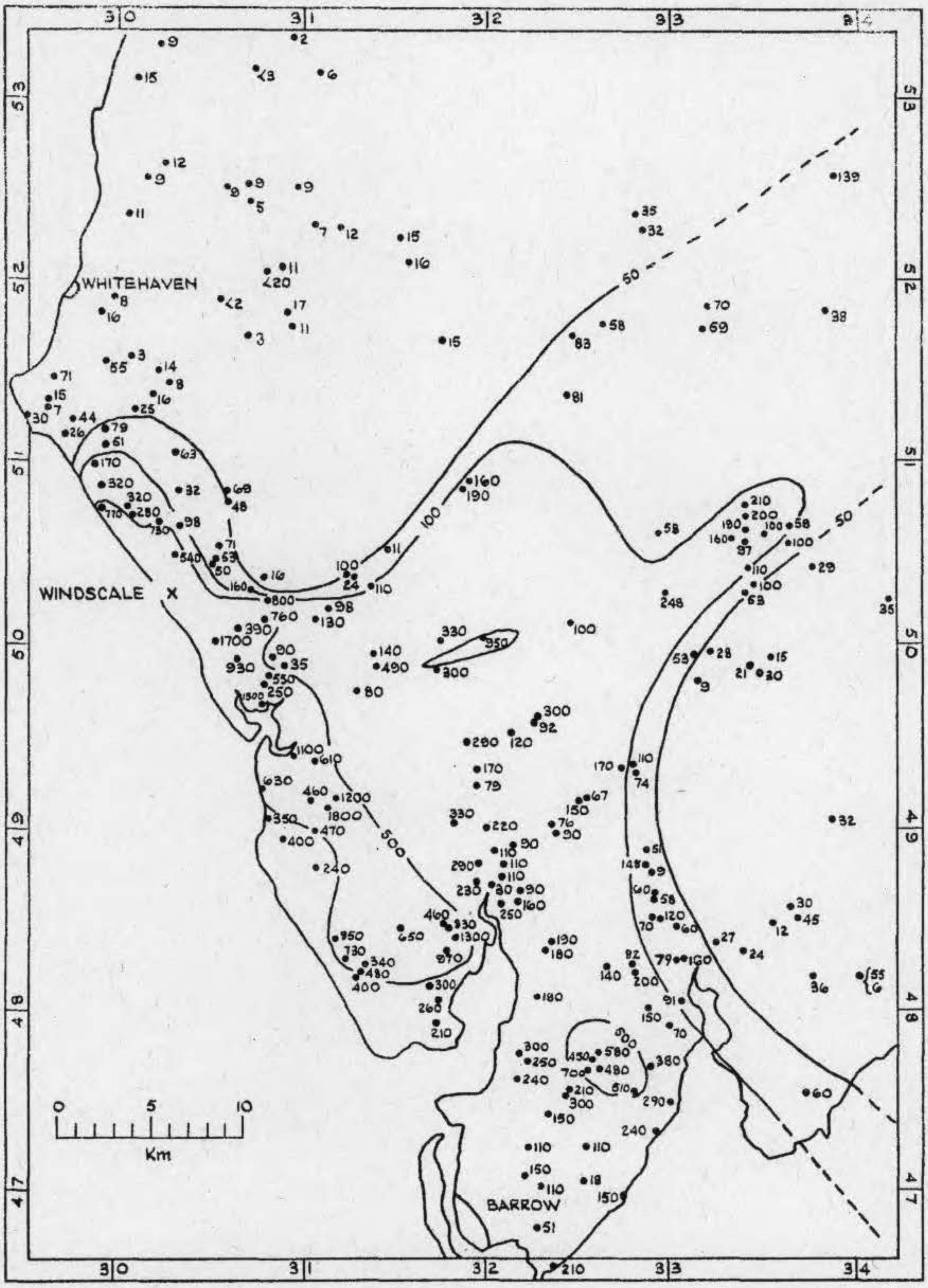


FIG. 3 CONTOUR MAP OF WINDSCALE AREA

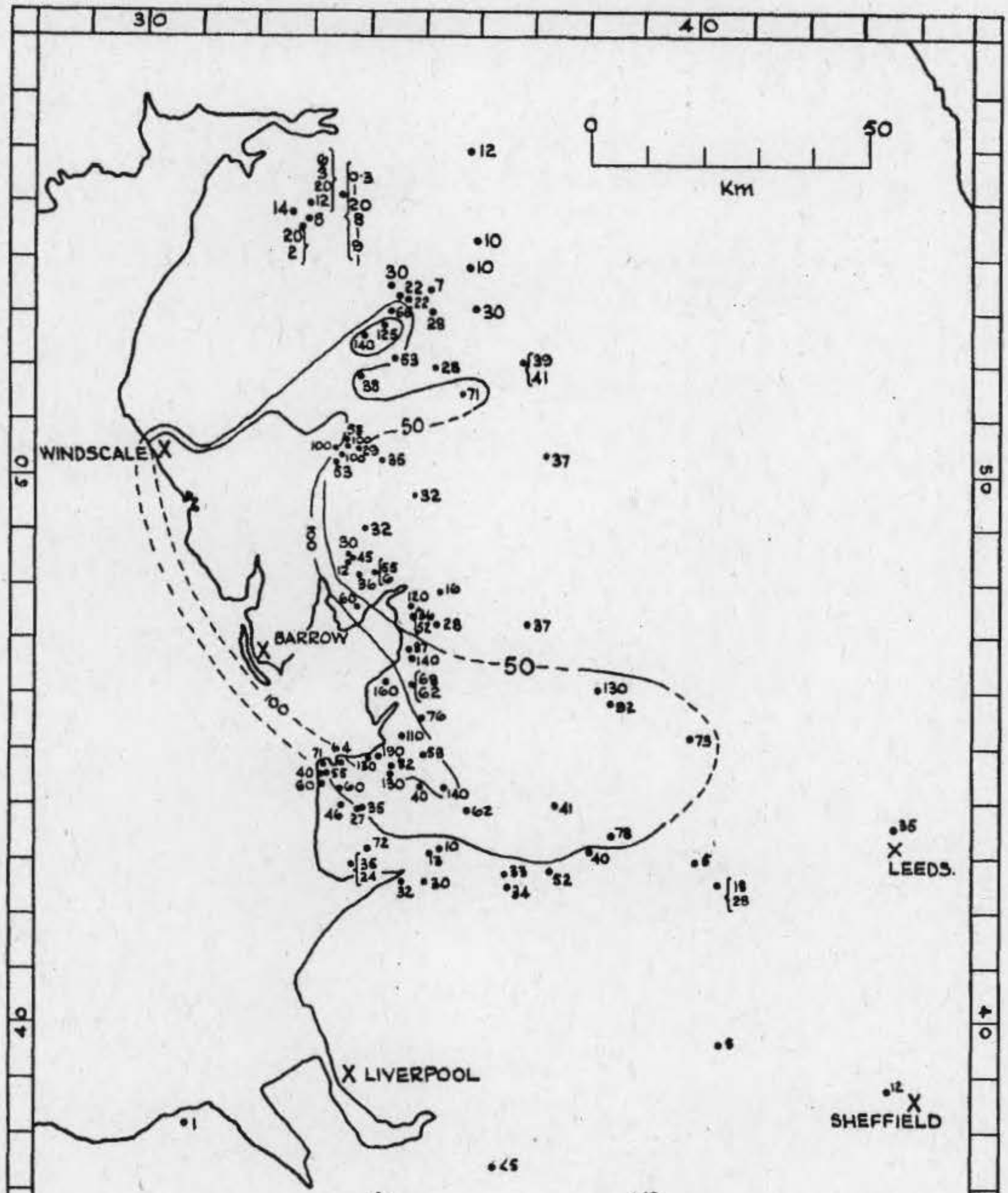


A.E.R.E. HP/R.2607. FIG. 4. DECREASE OF I 131 WITH TIME IN SEASCALE MILK.



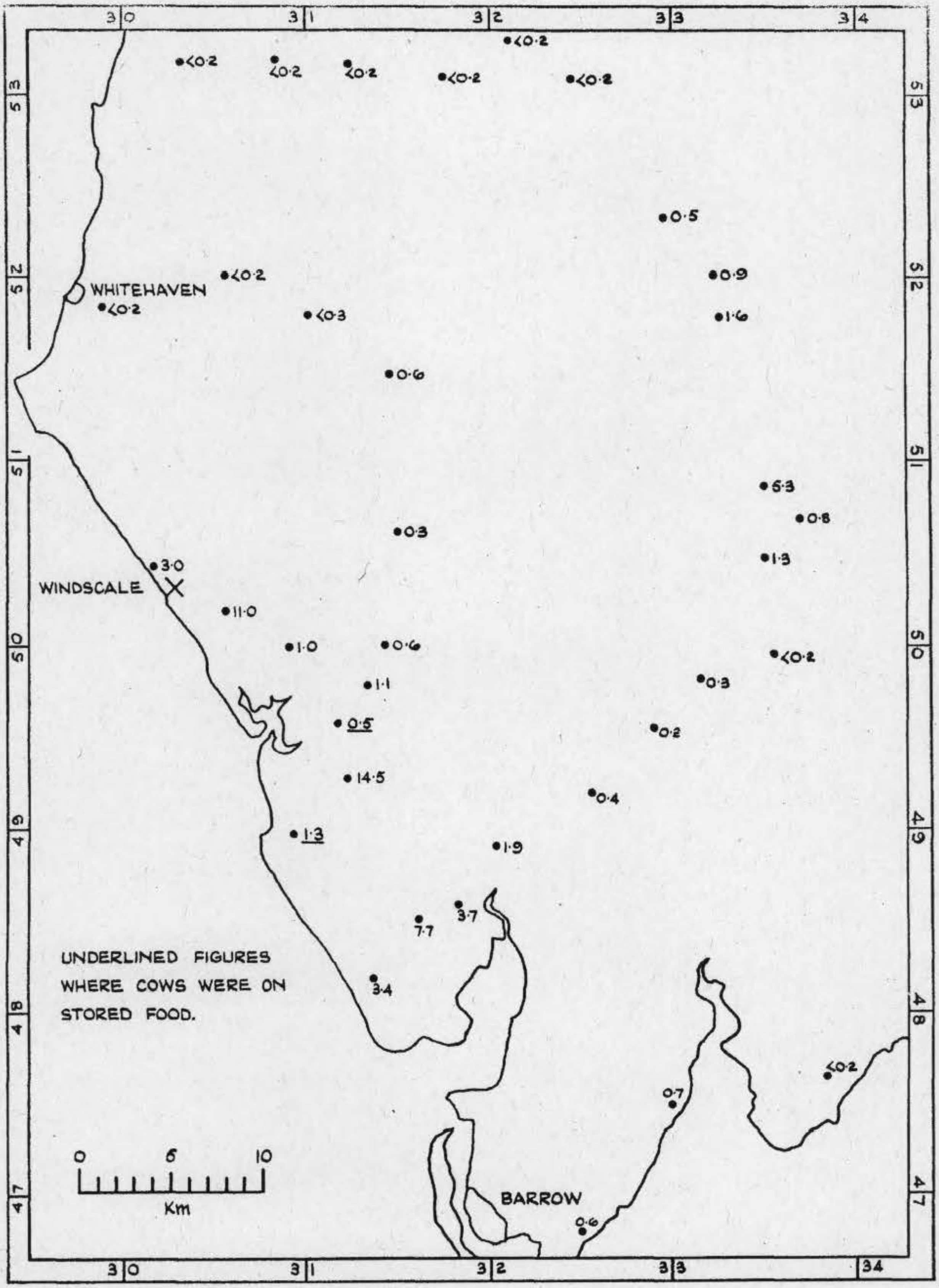
A.E.R.E. HP/R 2607 FIG. 5 MAP OF WINDSCALE AREA SHOWING I 131 IN MILK IN MILLIMICROCURIES PER LITRE.

MILK COLLECTED OCTOBER 13-19th. ACTIVITY WORKED BACK TO OCTOBER 11th. USING A FIVE-DAY HALF-LIFE.



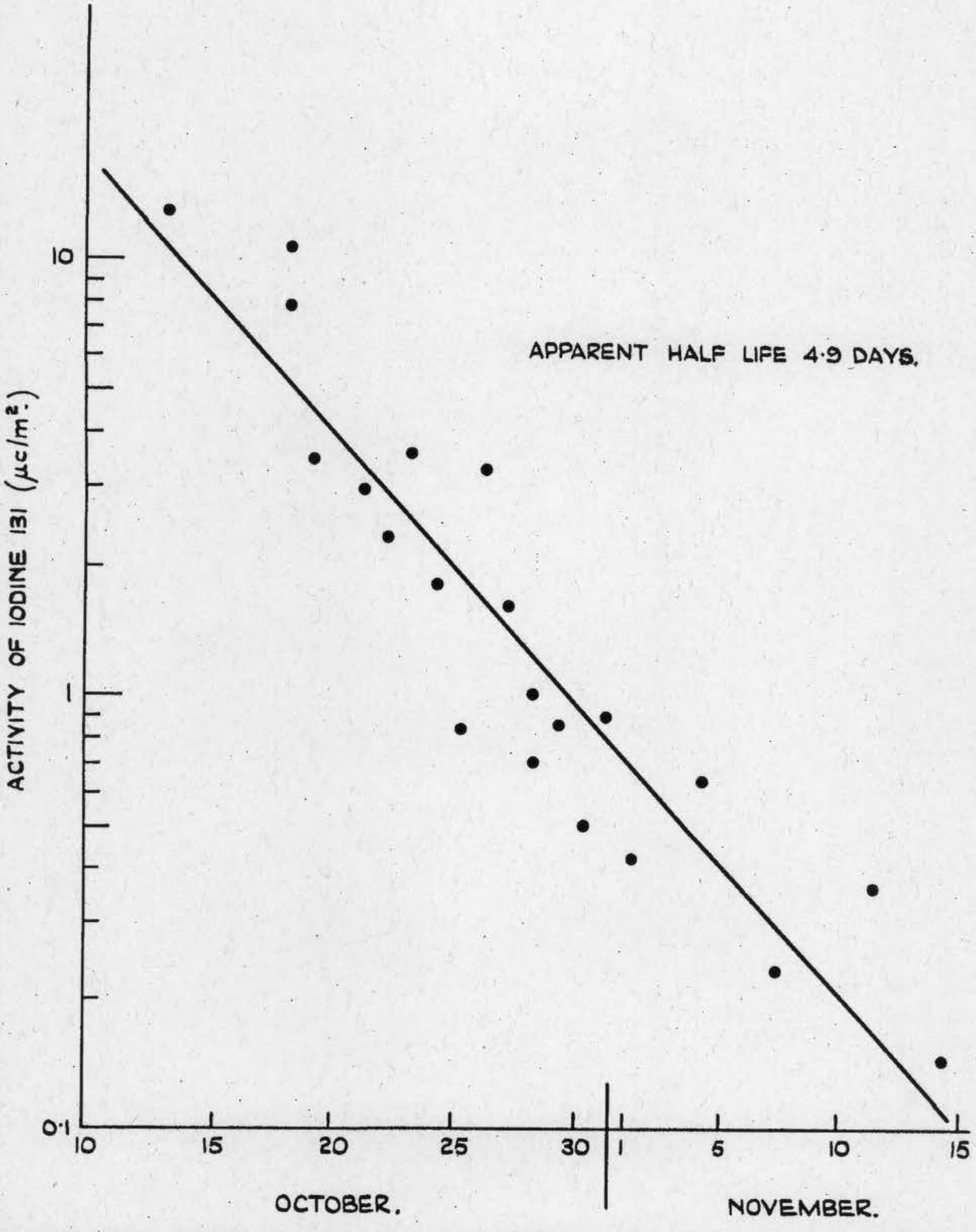
A.E.R.E. HP/R 2607. FIG. 6. MAP OF NORTH ENGLAND SHOWING I 131 IN MILK IN MILLIMICROCURIES PER LITRE.

MILK COLLECTED OCTOBER 13-19th. ACTIVITY WORKED BACK TO OCTOBER 11th. USING A FIVE DAY HALF-LIFE.

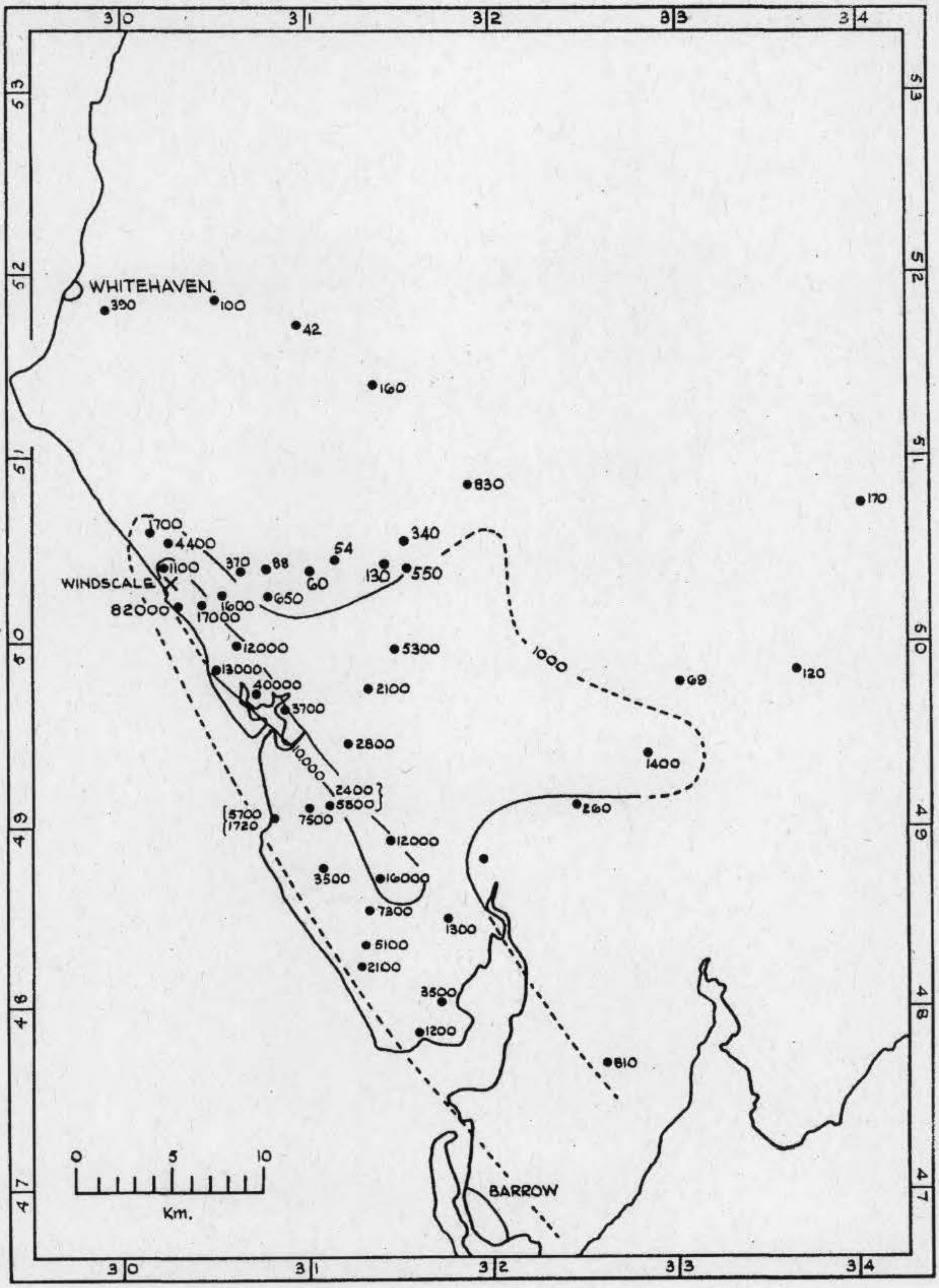


A.E.R.E. HP/R 2607. FIG. 7. MAP OF WINDSCALE AREA SHOWING Cs 137 IN MILK IN MILLIMICROCURIES PER LITRE.

MILK COLLECTED NOVEMBER 8-11th.

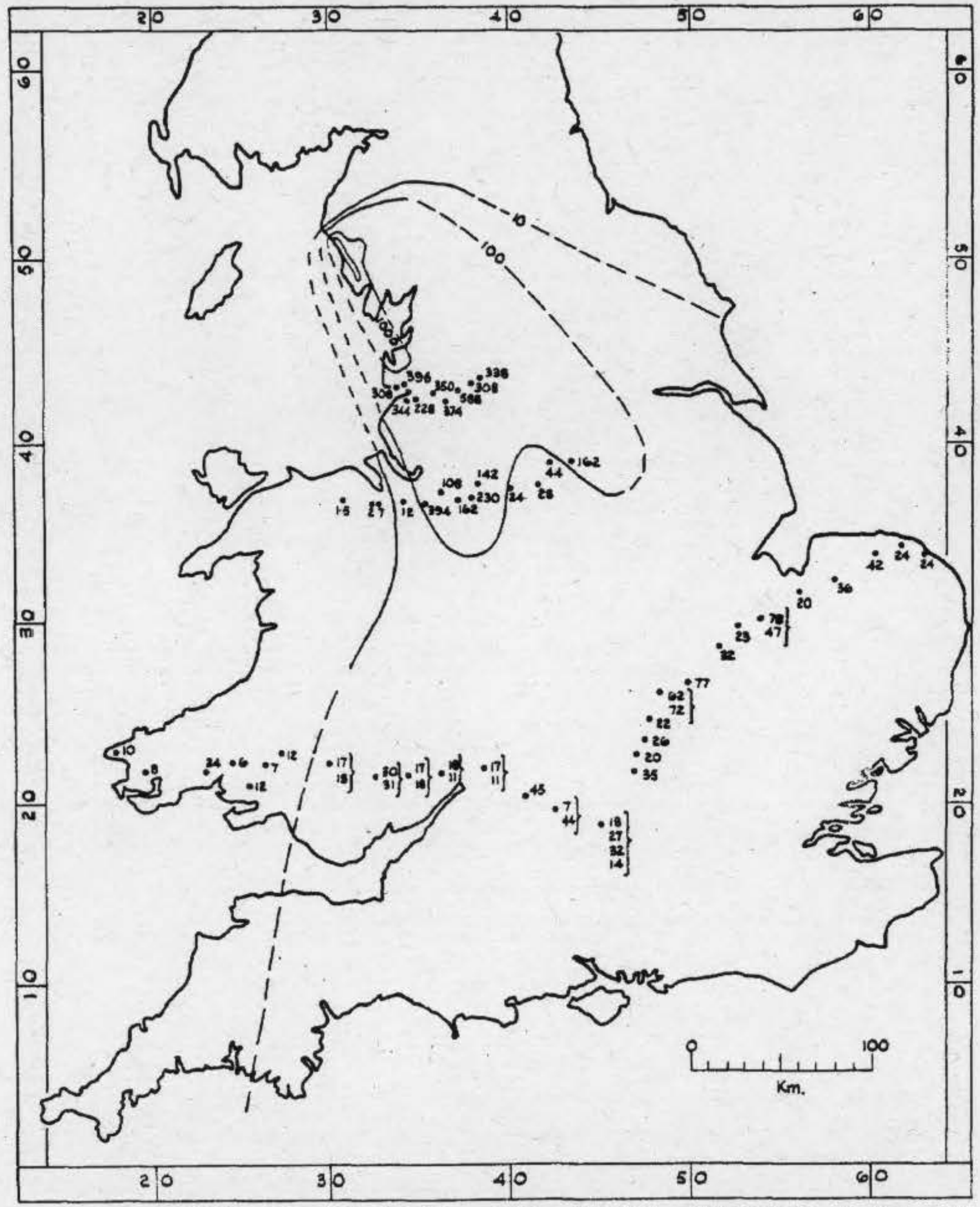


A.E.R.E. HP/R 2607. FIG. 8. APPARENT DECAY OF I 131 IN SEA-SCALE HERBAGE.



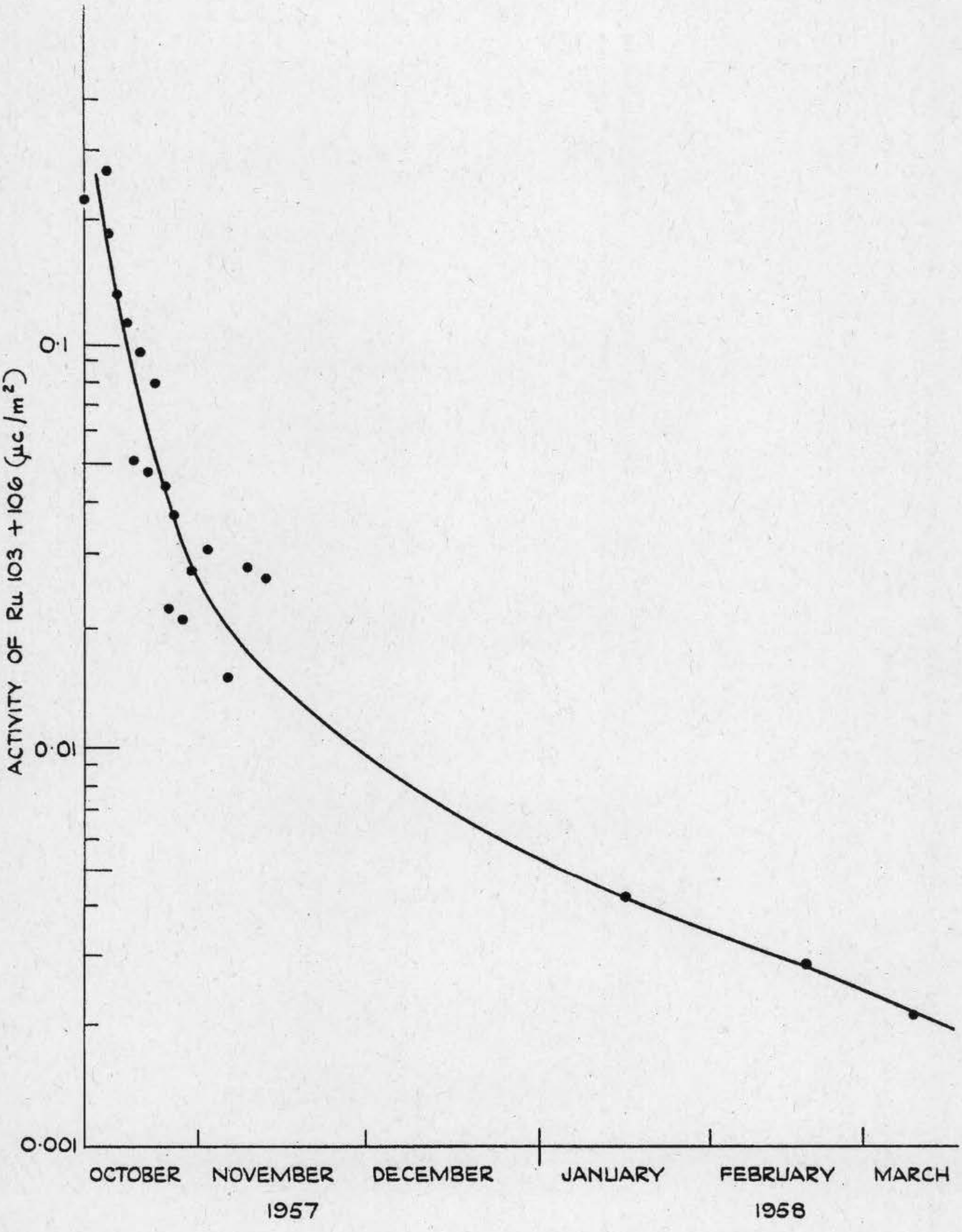
A.E.R.E. HP/R 2607. FIG. 9. MAP OF WINDSCALE AREA SHOWING I131 IN HERBAGE IN MILLIMICROCURIES PER SQUARE METER.

HERBAGE COLLECTED OCTOBER 13-28th. ACTIVITY WORKED BACK TO OCTOBER 11th. USING A FIVE-DAY HALF-LIFE.

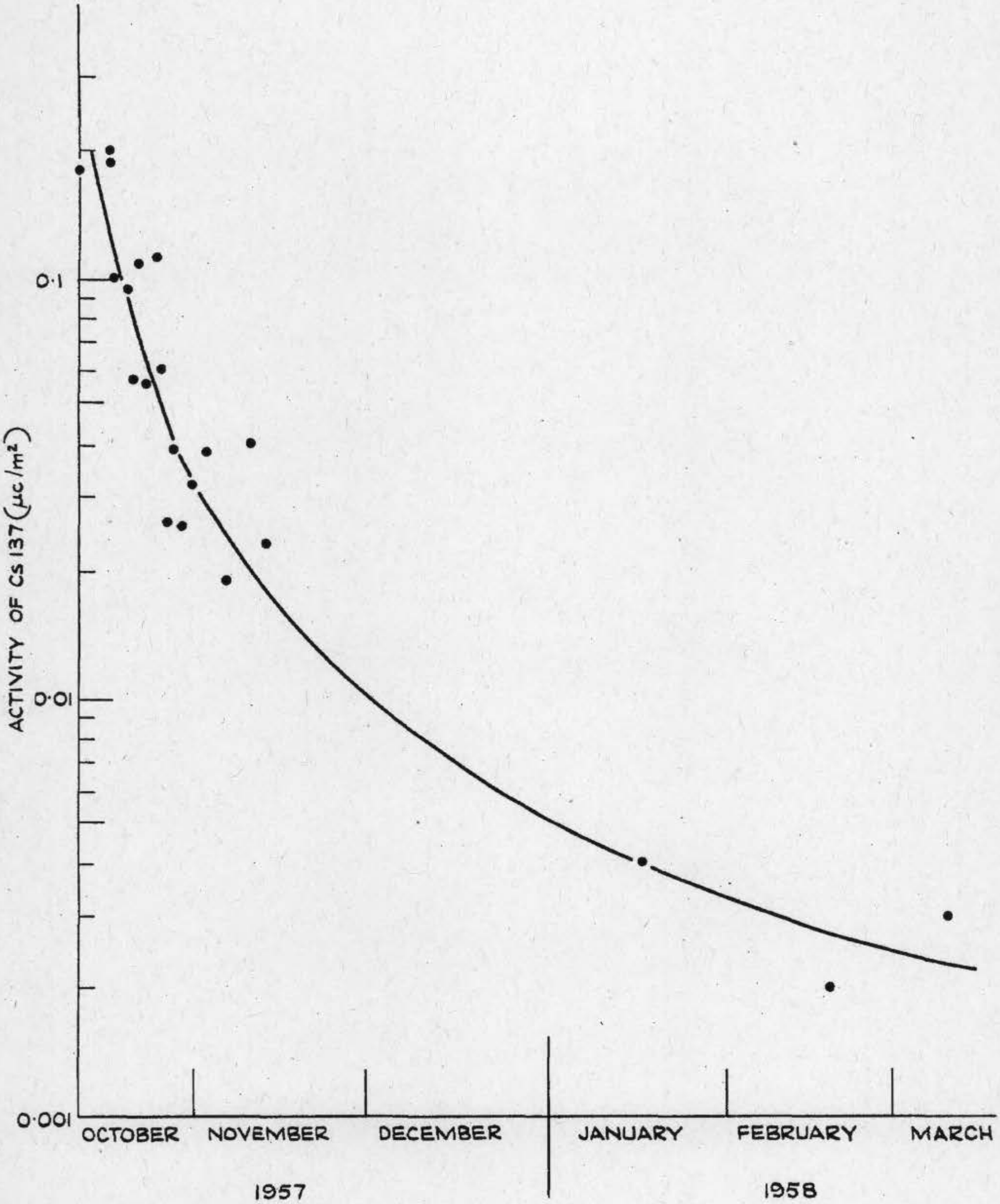


HP/R 2607. FIG. 10. MAP OF ENGLAND AND WALES SHOWING I 131 IN HERBAGE IN MILLIMICROCURIES PER SQUARE METER.

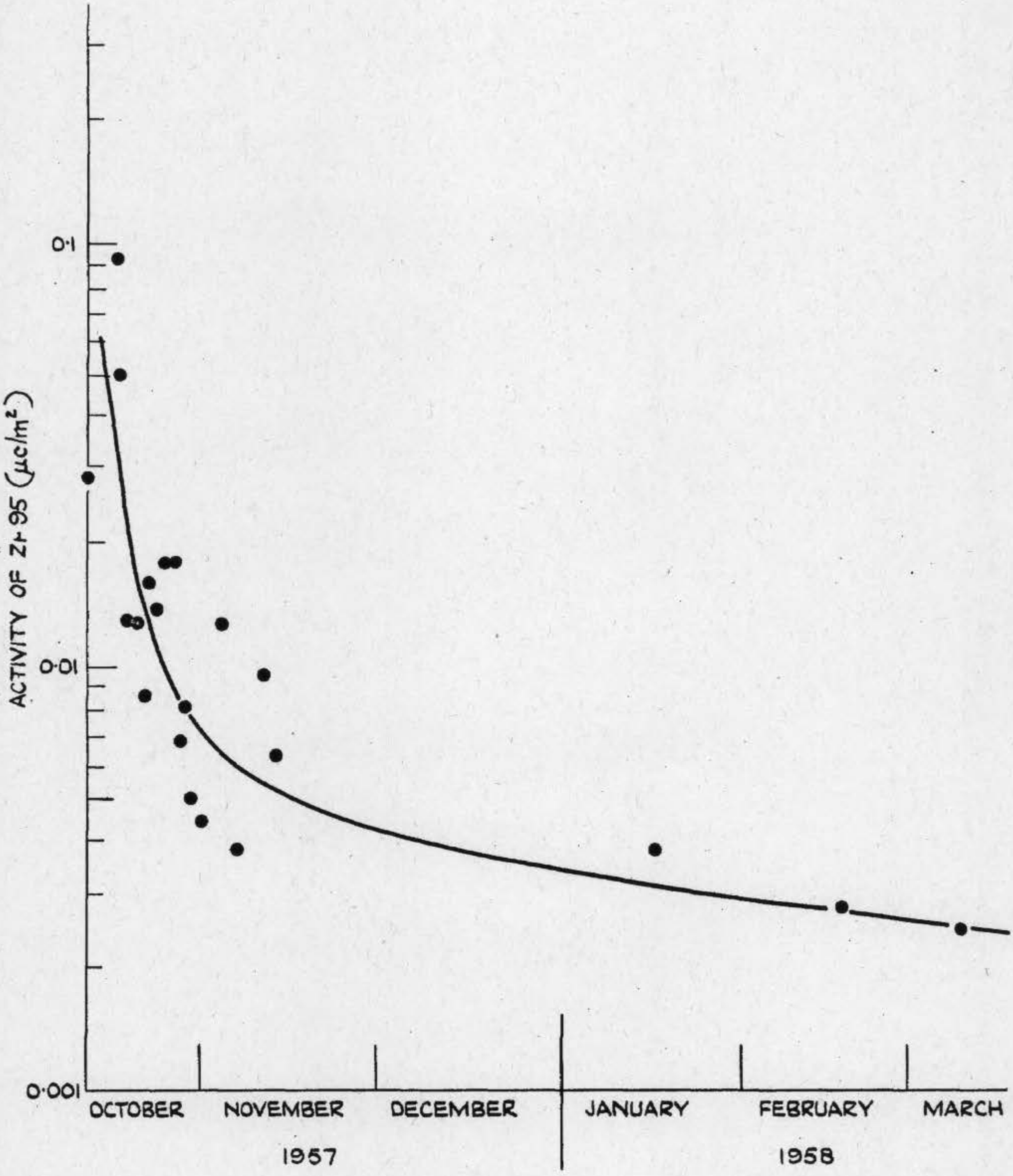
HERBAGE COLLECTED OCTOBER 13th - 20th. ACTIVITY WORKED BACK TO OCTOBER 11th USING A FIVE DAY HALF-LIFE.



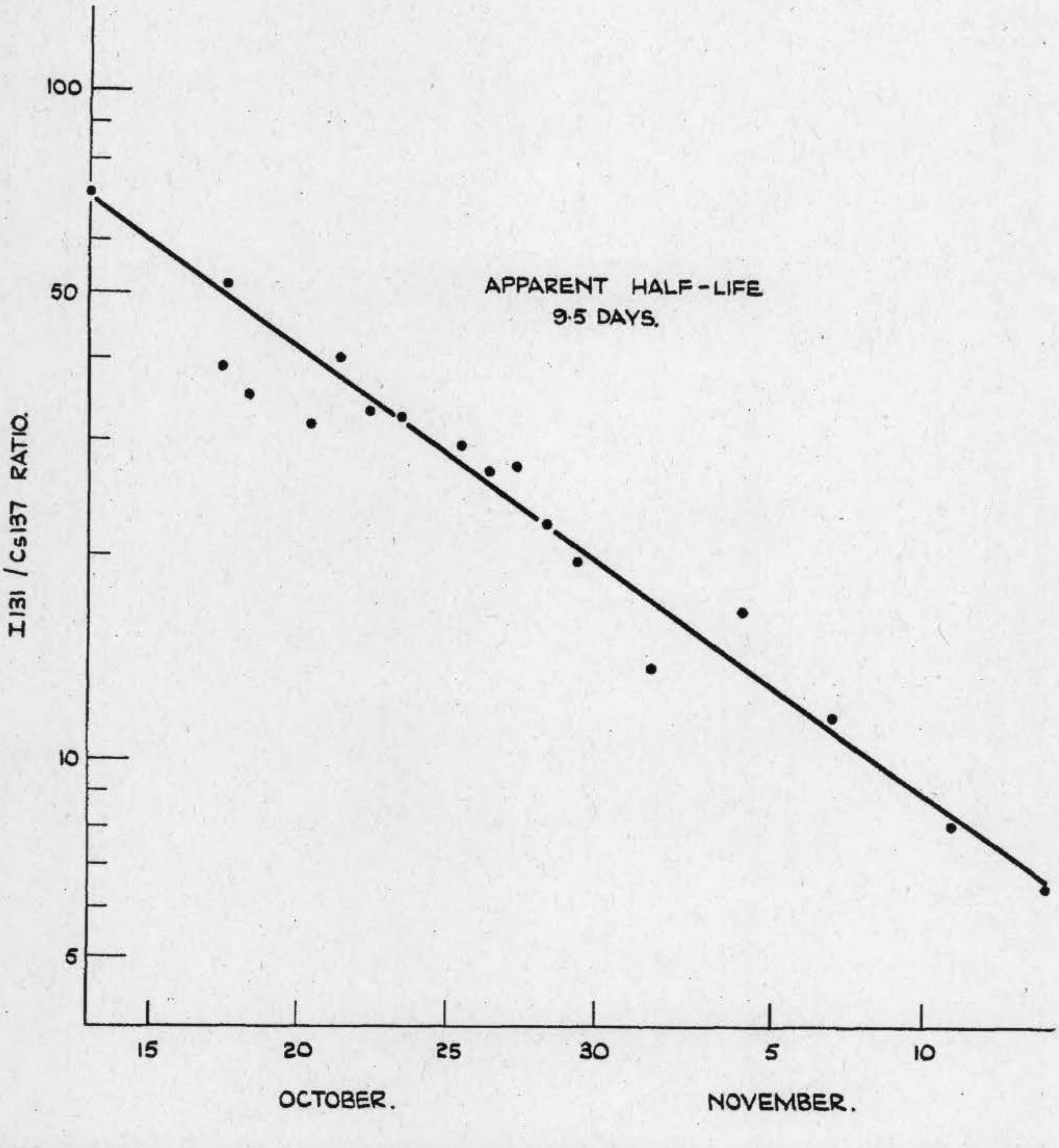
A.E.R.E. HP/R.2607. FIG. 11. APPARENT DECAY OF Ru 103 + Ru106 IN SEASCALE HERBAGE.



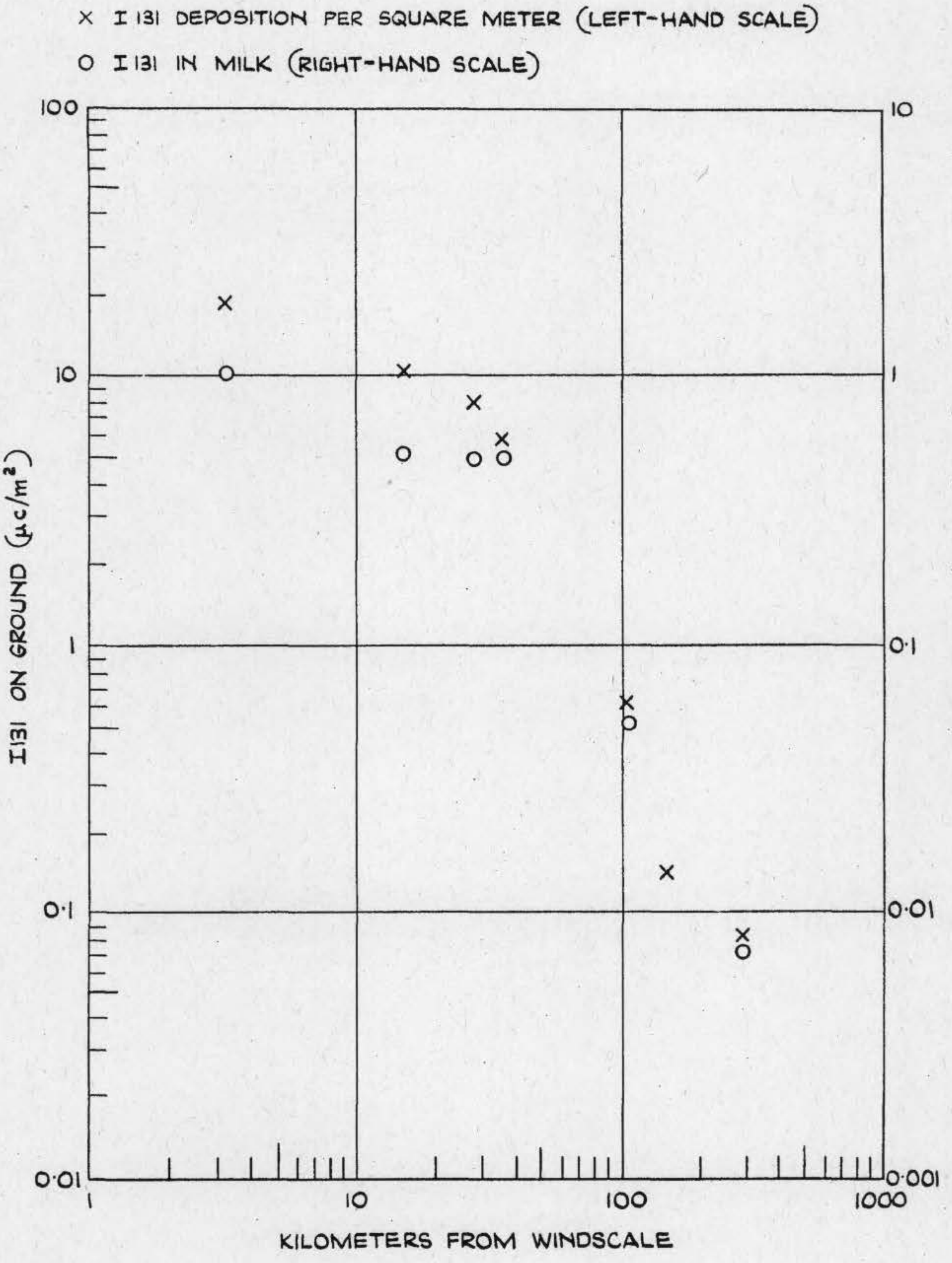
A.E.R.E. HP/R2607. FIG.12. APPARENT DECAY OF Cs 137 IN SEASCALE HERBAGE.



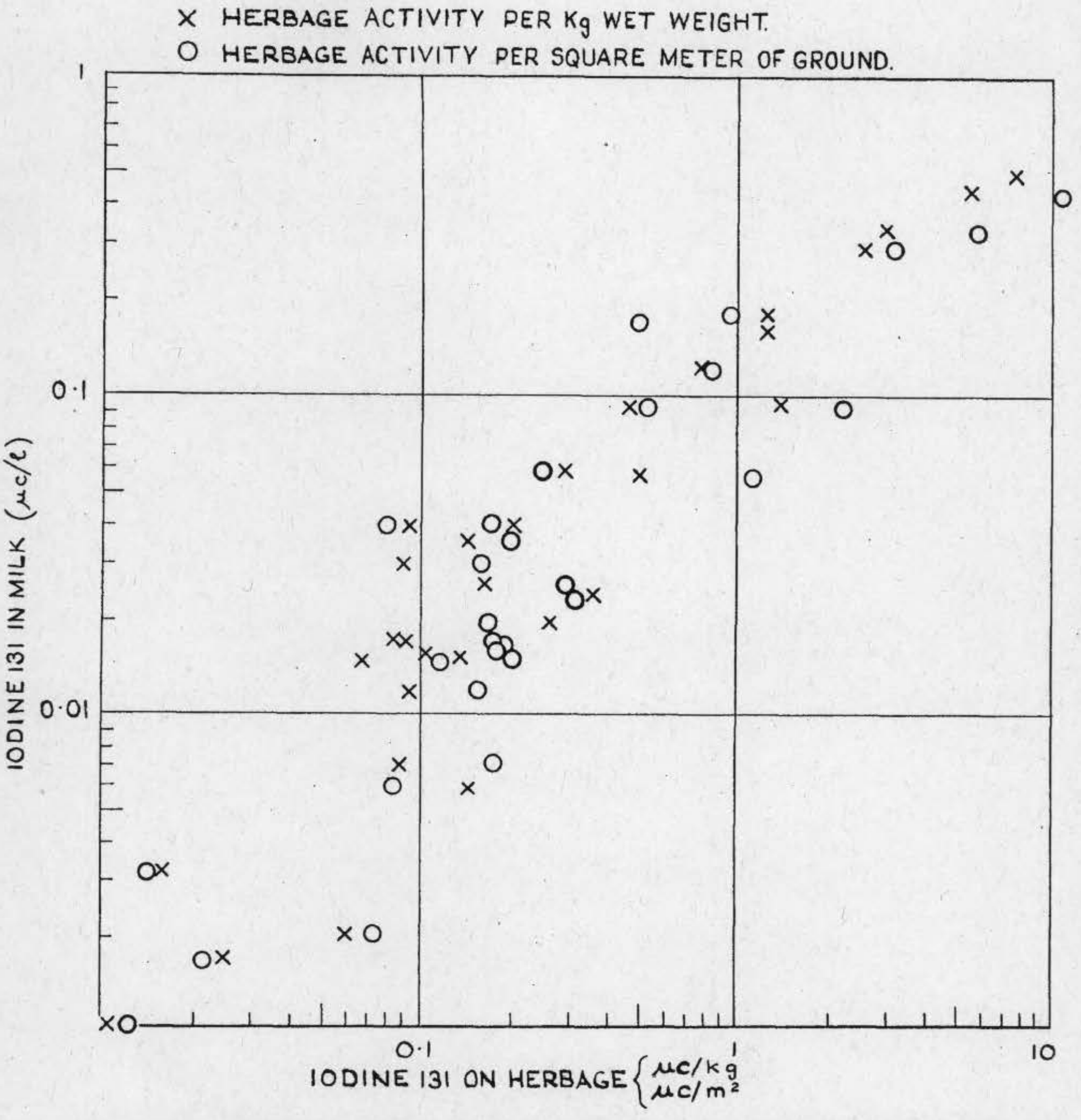
A.E.R.E. HP/R 2607. FIG. 13. APPARENT DECAY OF Zr 95 IN SEASCALE HERBAGE.



A.E.R.E. HP/R2607. FIG.14. CHANGE OF RATIO OF I131 AND Cs 137 ACTIVITIES WITH TIME IN SEASCALE HERBAGE.



A.E.R.E. HP/R 2607. FIG.15. DECREASE OF I131 DEPOSITION AND I131 IN MILK WITH DISTANCE FROM WINDSCALE.



A.E.R.E. HP/R 2607. FIG. 16. CORRELATION BETWEEN I 131 IN HERBAGE AND MILK.