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THE DEPOSITION OF AIRBORNE RADIOACTIVE PARTICLES AND THE CLEANING EFFECT OF PRECIPITATION AT VALENTIA

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Summary

Nearly three years of measurements of the specific radioactivity of precipitation made at Valentia Observatory (Ireland) have been used to calculate the mean values for the first, second and third day after at least one day without precipitation. The observations were divided into two groups covering the periods before and after the practice was introduced of washing after dry days the surface of the rain collector. Mean values for different daily amounts of precipitation have also been computed. Mean values of airborne radioactivity have been calculated for days without measurable precipitation and for days with less than or equal to, and for greater than the median value of the daily precipitation. From these data and making reasonable assumptions, which should apply on the average, it has been found that the deposition of radioactive particles on dry days amounts to 20%, and the pollution collected by the first day's rain to 62% of the specific radioactivity of the precipitation observed on the second or third day; the specific radioactivity contained originally in the cloud drops is as high as 80%. The results are not only consistent within themselves but also provide some derived values, such as deposition velocity of particles, which are generally in agreement with those obtained elsewhere or appear to be reasonable.

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1. It is thought that the observed inconsistencies of radioactivity measurements of precipitation and of airborne particles in neighbouring places may be explained, at least in part, by differences, due mainly to climatological causes, of the cleaning of the atmosphere by precipitation.

There are few quantitative studies based on actual routine observations on the efficiency with which the precipitation removes the particles, liquid or solid, which pollute the atmosphere.

The present note gives the results obtained from an investigation based on available data [1] undertaken with a view to contributing to the clarification of this very complicated question.*

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*While this note was being written a number of important papers, published by the Norwegian Defence Research Establishment, became available thanks to the courtesy of the Superintendent, Mr. T. Hvinden. It has been possible to compare some of the results with those given in a most interesting paper by Mr. S.H. Small [3]

2. The specific radioactivity of precipitation on days following one or more days without precipitation at Valentia during the period 1st May 1956 to 31st December 1959 has been examined and mean values, in arbitrary units, of the specific activities 1, 2 and 3 days after one or more days without precipitation have been computed. The results are given in Table I.

3. The great variability of the specific radioactivity of precipitation during the period of observation is reflected in the table by the large value of the average deviation from the mean, but there is no doubt about the reality of the reduction of the specific radioactivity between the first and the second day with precipitation after a period without. If we apply Student's t-test it is found that the reduction of specific radioactivity from 235 to 125, a reduction of 53%, is significant ($t = 4.02$, $P < 0.001$); the difference between the values 125 and 136 obtained for the second and third day is not significant.

4. The radioactivity of the rain in a series of consecutive days with precipitation may be considered in the first place as the sum of the radioactivity present in the cloud drops plus the radioactivity which the rain drops collect in their fall to the ground. The mean specific radioactivity, c, of the cloud drops can be assumed to be in the average rather constant independently of whether there is precipitation or not at the ground, but the amount of radioactivity collected by the rain drops or snow flakes in their fall would depend on the radioactivity of the aerosols which pollute the lower layers of the atmosphere. It will be assumed that this pollution is being removed by the precipitation at a rate proportional to the value of the pollution at any one moment (neglecting any possible selective effect) and added to by deposition from higher levels of the atmosphere and by mixing with surrounding air from areas without precipitation. So that a moment will be reached when equality will occur between the addition (supposed constant) of radioactivity as pollution to the air and the removal of it by precipitation; after that moment the average specific radioactivity of precipitation should not change. From the values given in Table I it appears that at Valentia this equilibrium is reached after one day of precipitation as the corresponding specific radioactivities remain substantially constant.

5. The pollution of the air which has been deposited on the surface of the collector during the immediately preceding rainless days is removed by the precipitation which falls in the rain collector. Therefore the measured radioactivity of the precipitation after one or more days without rain includes not only, as stated in paragraph 4, the radioactivity originally in the cloud drops and that collected by the rain drops in their fall to the ground, but also the radioactivity accumulated on the surface of the collector during the day or days before the day on which precipitation has been measured.

To estimate the relative value of the radioactivity of the sample due to these different origins it would be necessary to have a series of observations of samples when one or other of the mentioned sources of radioactivity had been eliminated. To achieve this end it was found impracticable to have the rain collector covered when there was no precipitation; so the next best method was adopted on and from April 1958 of washing the exposed surface of the collector whenever there had been no precipitation in the previous day. This practice would result in the reduction of the specific radioactivity of the precipitation on the first day by that part that is due to the deposition of pollution during the previous rainless day or days. Thus, it would be expected that the reduction of the specific radioactivity of the precipitation from the first to the second day after a period without precipitation would be greater before than after washing was introduced. This is shown to occur in Table II.

The difference between the average values of the specific radioactivity of the first and second days with precipitation is a real one: $t = 3.43$, $P < 0.001$ before washing and $t = 2.23$, $P < 0.02$ after washing was introduced. The difference in specific radioactivity between the second and third rainy days is not significant in either case.

The average level of radioactivity up to April 1958 inclusive was substantially smaller than from May 1958 to the end of 1959. For purposes of comparison the values are also given in Table II as a percentage of the mean of the radioactivities measured on the second and third day.

6. From the percentage data in the table for the period after washing was introduced it is deduced, as stated in paragraph 4, that the radioactive pollution captured by the precipitation during the first day, w_1, together with the deposited pollution of the atmosphere on the first wet day, d_1, and the radioactivity of the cloud droplets, c, amount to 159% of the radioactivity measured on the second day with precipitation. Therefore

$$w_1 + d_1 + c = 159 \qquad (1)$$

As this relative amount of radioactivity was also present in the rain collected before the practice of washing the collector was introduced*, the difference of 47% between the value of 206 before washing was introduced and 159% can be attributed to be due to the dry deposition. During the period concerned the average number of dry days before a day with precipitation was 2.3 so that the mean deposition in a dry day was d_o, $= \frac{47}{2.3}$ or 20% of the specific radioactivity on the second rainy day.

*The mean daily rainfall on the first day with precipitation after one or more dry days was substantially constant during the whole period considered here: 5.6 mms before and 5.3 mms after the practice of washing the collector was introduced.

7. As stated before, the specific radioactivity of the rain on the third day is not significantly different from that on the second day, and, as scavenging and deposition occurs on the third day of precipitation in the same manner as it occurs on the second day it follows that

$$w_2 + d_2 + c = 100,$$

$$\text{and} \quad w_3 + d_3 + c = 100$$

where the letters have the same signification as in paragraph 6 and the sub-indices indicate that they refer to the second and third days with precipitation.

The radioactivity added by deposition from higher levels and by mixing with other air is equal to the amount removed by the precipitation and by deposition in the collector once the equilibrium state has been reached. Therefore

$$w_2 + d_2 = w_3 + d_3 = d_o = 20$$

$$\text{and} \quad c = 100 - 20 = 80$$

i.e. the average specific radioactivity of the cloud droplets amounts to 80% of the specific radioactivity of the precipitation of the second day.

8. It is natural to assume that the depositions on a wet and a dry day are in the same ratio as the average radioactivity of the pollution of the atmosphere if the nature and size spectrum of the particles is supposed similar. It has been found (see paragraph 12) that during the period February 1957 to December 1959 the airborne radioactivity observed at Valentia in days without measurable precipitation* was 140 pC per 100 cu.m. and in days with measurable precipitation (mean value of the order of 7 mm) 100 pC per 100 cu.m. The ratio 1.40 between these two values must be considered as characteristic of the nature and size of the particles which pollute the air and of the type, amount and duration of the precipitation. This ratio which has been remarkably constant at Valentia during the period covered by this investigation (see Table IV) will be used, in the absence of a better possibility to compute an approximate value of d_2. It follows therefore that $d_2 = 20/1.40 = 14$, that is to say 14% of the specific radio-activity of rain on the second day with precipitation is due to deposition during the previous rainy day. This gives for the pollution captured by the precipitation after the first rainy day $w_2 = w_3 = 20 - 14 = 6$ or 6% of the specific radioactivity of the precipitation of the second rainy day.

9. Substituting for c in equation (1) paragraph 6, gives

$$w_1 + d_1 = 159 - 80 = 79$$

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*Days with less than 0.1 mm.

Accepting for d_1 a value which is the mean of d_o and d_2 it follows that

$$d_1 = 17\%$$

$$\text{and} \qquad w_1 = 62\%$$

of the mean specific radioactivity observed on the second day with precipitation.

10. In Table III the different percentages found are summarized.

For the determination of d_1 the average value $d_o = 20\%$ of the deposition in dry days has been used. It is obvious that the deposition in a dry period is an increasing function of time until an equilibrium value is obtained. Therefore the value of d_o used in paragraph 8 for computing d_1 is only a first approximation. A method of successive approximation could be used but it is considered that the character of this investigation does not justify trying to obtain greater accuracy.

11. It is not contended that the percentages found for the radioactivity of different origins should be, on average, the same in all circumstances and localities. For instance, the assumptions regarding pollution at the stratosphere, upper troposphere and near the ground and their relationships which are implied in previous paragraphs, may vary considerably, not only due to the different types of bombs and the levels at which their explosions have taken place, but also due to climatological factors and, perhaps, also to the nature and size of the atmospheric aerosols which capture some, at least, of the radioactive particles. It is expected that similar studies with data from other stations where the measurement of the radioactivity of precipitation and settled dust is carried out under different local climatological conditions may produce different values for these percentages. From these differences some indication may be obtained of the cleaning of the atmosphere by precipitation under different circumstances.

In this connection it is of interest to ascertain the degree of reliability to be attached to the values given in Table III.

12. From what has been stated in previous paragraphs it is to be expected that the airborne radioactivity would be greater on dry days than on days with precipitation and that its value on rainy days from the second onwards would be rather constant. As the differences between the mean values of the airborne radioactivity at Valentia for the first, second, etc. days of a period with precipitation during the months February 1957 to December 1959 are not statistically significant, average values have been computed for days without measurable precipitation and for days with precipitation equal or less than 3 mms. and greater than 3 mms. The value of 3 mms. has been selected because it is the median value of the daily precipitation for the period. The results are given in Table IV. The mean precipitation in the three different groups are 0.0, 1.2 and 10.1 mms.

Figure 1 drawn using these mean values confirms that the amount of airborne radioactivity tends to become constant with increased precipitation. The values of 140 and 100 pC per 100 cubic metres of air adopted in paragraph 8 for the average airborne radioactivity in dry days and the second or third rainy days is also hereby justified.

13. The daily average specific radioactivity of precipitation has been classified according to the amount of precipitation in the 24-hour period to which they refer and mean values computed for the periods May 1956 to April 1958 and May 1958 to September 1959 and for the whole period of observations. The results are given in Table V and Figures 2 and 3.

The curves in these figures show a marked decrease of the specific radioactivity with increasing amount of precipitation up to about 5 mm. independently of whether the practice of washing the collector was in operation or not. As was to be expected the decrease is less when the washing of the collector had removed the radioactivity deposited on it. The figure also shows that the decrease is much slower or even may cease in both cases when the amount of precipitation is greater than about 5 mm. As the average precipitation per wet day was 5.6 mm. for the first period and 5.3 mm. for the second period, the near constancy of the mean specific radioactivity after the first day of rain is easily explained.

14. It appears that the decrease of the specific radioactivity with increased amount of precipitation is due (a) to the washing effect of the rain drops falling on the surface of the collector and (b) to the continuous elimination of pollution from the atmosphere by the falling precipitation.

(a) Experiments have been made at Valentia on four occasions in order to determine the rate of progressive contamination of the water used for washing the raingauge. The amount of water used, usually one litre, was divided on those occasions in four equal parts which were successively used for washing the collector. The radioactivity in each portion was then determined in the usual way and Table VI gives the average results. These are also given in such a form that the effect of different amounts of washing water can be appreciated. It is, of course, impossible to say whether the manual washing of the collector was more or less effective on every occasion than the washing by falling drops but it may be noted that the points plotted in Fig. 3 using the values of Table VI fit quite closely the extreme left hand portion of the curves. On general grounds it is evident that if the amount of water used for washing and the number of parts in which it is divided were increased the new values obtained would also give a reasonable fit to the remainder of the curves for natural precipitation.

(b) The removal of pollution from the atmosphere by falling drops and flakes can be assumed to be proportional to the amount of precipitation and to the amount of radioactivity present in the polluted atmosphere at any one moment. If it is further assumed that the addition of radioactivity by deposition from higher levels and by mixing with other air occurs at a constant rate, it is clear that after a certain amount of precipitation, the specific radioactivity of the water should tend to remain constant.

15. It is noteworthy that the curves of Fig. 2 are very similar to curves drawn for Harwell in Great Britain [2] and for Kjeller in Norway [3]. This may be a general feature of the removal of pollution by precipitation at least in Northwest Europe.

For these two places the slope of the curve also changes considerably at about 5 mm. of precipitation. However it cannot be deduced from this that the specific radioactivity of precipitation after the first day of rain would tend to become constant everywhere a similar curve obtains. This can only occur if and when the mean precipitation in a wet day is at least equal to 5 mms., i.e. the value of the precipitation at which the change in the slope occurs.

16. As stated before, from May 1958, the surface of the collector used for radioactivity measurements was washed whenever possible at normal sampling time, when no precipitation had been observed in the previous day and the water used for the washing was then evaporated and counted in the same manner as ordinary precipitation. These data can be used to estimate the dry deposition per unit of exposed area. For the period May 1958 to December 1959 the average radioactivity of the washing water expressed in the same arbitrary units as the radioactivity of the precipitation, was 122 per day. The average rainfall for wet days during the period was 5.3 mm., so we obtain that the contribution of this radioactivity to the specific radioactivity of the first day of rainfall would have been 122/5.3 = 23 units. This represents 74% of the figure obtained, 31 units (20% of 153 units) by applying the percentage determined in paragraph 6 for deposition on dry days. Given the difficulty of this type of measurements, the agreement is considered as satisfactory.

17. The velocity of deposition of pollution has been defined as

$$V = \frac{\text{Rate of deposition of radioactivity per unit area per unit time}}{\text{Radioactivity of airborne particles per unit volume of air}}$$

An estimate for the conditions obtaining at Valentia can be obtained as follows: The 122 units for dry deposition per day obtained from the washed water should be multiplied by 1.63 to take into account losses due to evaporation etc. and by 2.958 to convert the arbitrary units into pC per square metre. The result is 588.2 pC per square metre per day or 6.81×10^{-7} pC per cm^2 per second. The concentration of airborne radioactivity per cubic metre during the relevant period was 169 pC per 100 cu.m.

or 1.69×10^{-6} pC per cm^3. Taking the efficiency of the filter as 90%, we obtain -

$$V = 0.36 \text{ cm/sec.}$$

In Great Britain [1] it has been found that V = 0.07 cm/sec using gummed paper for determining the rate of deposition. For Norway values ranging between 0.2 and 3.4 cm/sec (average 0.72) are given in [2]. They have been obtained by a method more germane to the one described here.

18. During the period May 1958 to December 1959 the pollution due to airborne particles was $1.69 \times 10^{-6}/0.9 = 1.88 \times 10^{-6}$ pC/cm^3. If this pollution falls with an average speed of 0.36 cm/sec, then the radioactivity deposited on a dry day is 588.2×10^{-4} pC per cm^2. Assuming constant concentration with height this radioactivity was originally contained in a layer of air about 312 m. thick.

The mean amount of radioactivity captured from the polluted atmosphere and brought down to the ground from the base of the clouds by one millimetre of precipitation over one square metre during the months May 1957 to December 1959, amounted for the first wet day to 62% of 153 units or

$$153 \times 0.62 \times 1.63 \times 2.958 = 457 \text{ pC.}$$

In a day with average precipitation (5.6 mm.) the mean amount of radioactivity brought down was 0.256 pC per cm^2. This corresponds to the radioactivity contained in $\frac{0.256}{1.88 \times 10^{-6}} = 1.36 \times 10^5$ cm = 1360 m.

This would represent the mean height of the base of cloud before precipitation starts at Valentia, which seems too large by a factor of about 2.

Another value for the mean height of the base of cloud can be obtained as follows: the percentage amount of radioactivity captured by precipitation in the first day (62%) is 3.1 times the percentage deposition in a dry day (20%). Assuming full capture of the pollution by the precipitation the layer of the lower atmosphere cleaned by the precipitation would be 312 x 3.1 = 967 m. which appears to be also too large by a factor of 1.5.

19. The results given above appear to be fairly consistent within themselves and with those obtained in other countries. Although the accuracy of the mean values is not high, not only due to the inherent uncertainty of the calibration of the counters but also to the difficulties of obtaining and preparing representative samples for counting, the results may be considered as satisfactory. The use of percentages eliminates or reduces the influence of most of the inaccuracies mentioned. On the other hand the inaccuracies acquire greater importance when comparing the data obtained using different techniques.

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References:

[1] Measurements of radioactivity of precipitation and airborne particles in Ireland. Published by the Irish Meteorological Service, Department of Transport and Power (In preparation)

[2] A.E.R.E., HP/R 2354 (1957)

[3] S.H. Small, Wet and dry deposition of fallout materials at Kjeller, Norwegian Defence Research Establishment, Intern. Rapport, F-0391, October 1959.

TABLE I

Mean specific radioactivity of precipitation at Valentia on the first, second and third day after one or more days without precipitation

Arbitrary Units | All cases

No. of days after at least one day without precipit-ation	One or more days with precipitation		
	Mean specific Radio-activity	Mean deviation	No. of cases
One	235	237	156
Two	125	118	118
Three	136	121	92

TABLE II

Mean specific radioactivity of precipitation at Valentia on the first, second and third day after one or more days without precipitation

No. of days after at least one day without precipitation	No washing				Washing			
	Mean Specific Radio-activity		Mean deviation	No. of cases	Mean Specific Radio-activity		Mean deviation	No. of cases
	Arbitrary Units	%			Arbitrary Units	%		
One	227	206	243	81	243	159	221	75
Two	102)	100	85	63	152)	100	138	55
Three	120)		107	49	154)		138	43

TABLE III

Mean specific radioactivity as percentage

Cloud drops, c,			80
Deposition per	dry day, d_0		20
	first wet day, d_1		17
	second, third, etc., wet day, d_2, d_3, ...,		14
Air Pollution captured by precipitation	first day, w_1		62
	second, third, etc., day, w_2, w_3, ...,		6
Specific radioactivity of precipitation	first day	Collector washed $c+w_1+d_1$	159
		Collector not washed $c+w_1+d_1+2,3d_0$	206
	second, third etc. day	$c+w_2+d_2$, $c+w_3+d_3$, ...	100

TABLE IV

Mean radioactivity of airborne particles at Valentia in pC per 100 cubic metres

Period	Days with no measurable precipitation			Days with measurable precipitation					
				≤ 3.0 mm.			> 3.0 mm.		
	Mean	Mean deviation	No. of cases	Mean	Mean deviation	No. of cases	Mean	Mean deviation	No. of cases
February 1957 to April 1958	100	-	128	80	-	172	72	-	151
May 1958 to December 1959	169	-	177	136	-	208	116	-	225
February 1957 to December 1959	140	104	305	111	91	380	98	89	376

TABLE V

Mean specific radioactivity of precipitation at Valentia as a function of the amount of daily precipitation

May 1956 to April 1958			May 1958 to September 1959			All Observations		
Mean daily Precipitation	Specific Radioactivity		Mean daily Precipitation	Specific Radioactivity		Mean daily Precipitation	Specific Radioactivity	
mms.	pC per litre	%	mms.	pC per litre	%	mms.	pC per litre	%
0.5	877	100	0.5	1487	100	0.5	1117	100
2.0	306	35	2.0	764	51	2.0	487	44
5.0	227	26	5.0	328	22	5.0	268	24
10.0)	168)	19)	10.0)	296)	20)	10.0)	222)	20)
18.0) 15.0	195) 178	22) 20	18.0) 14.0	241) 284	16) 19	18.0) 15.0	215) 222	20) 20
35.0)	178)	20)	31.0)	366	25)	33.5)	247)	22)

TABLE VI

Radioactivity of the water used for washing the collector

Arbitrary units

Actual amount of water used in successive washings	Actual Radioactivity	Cumulative data			
		Amount of water	Radioactivity		
			Actual	Per unit volume	%
c.c.		c.c.		c.c.	
250	166	250	166	0.66	100
250	77	500	243	0.49	73
250	49	750	292	0.39	59
250	21	1000	313	0.31	47

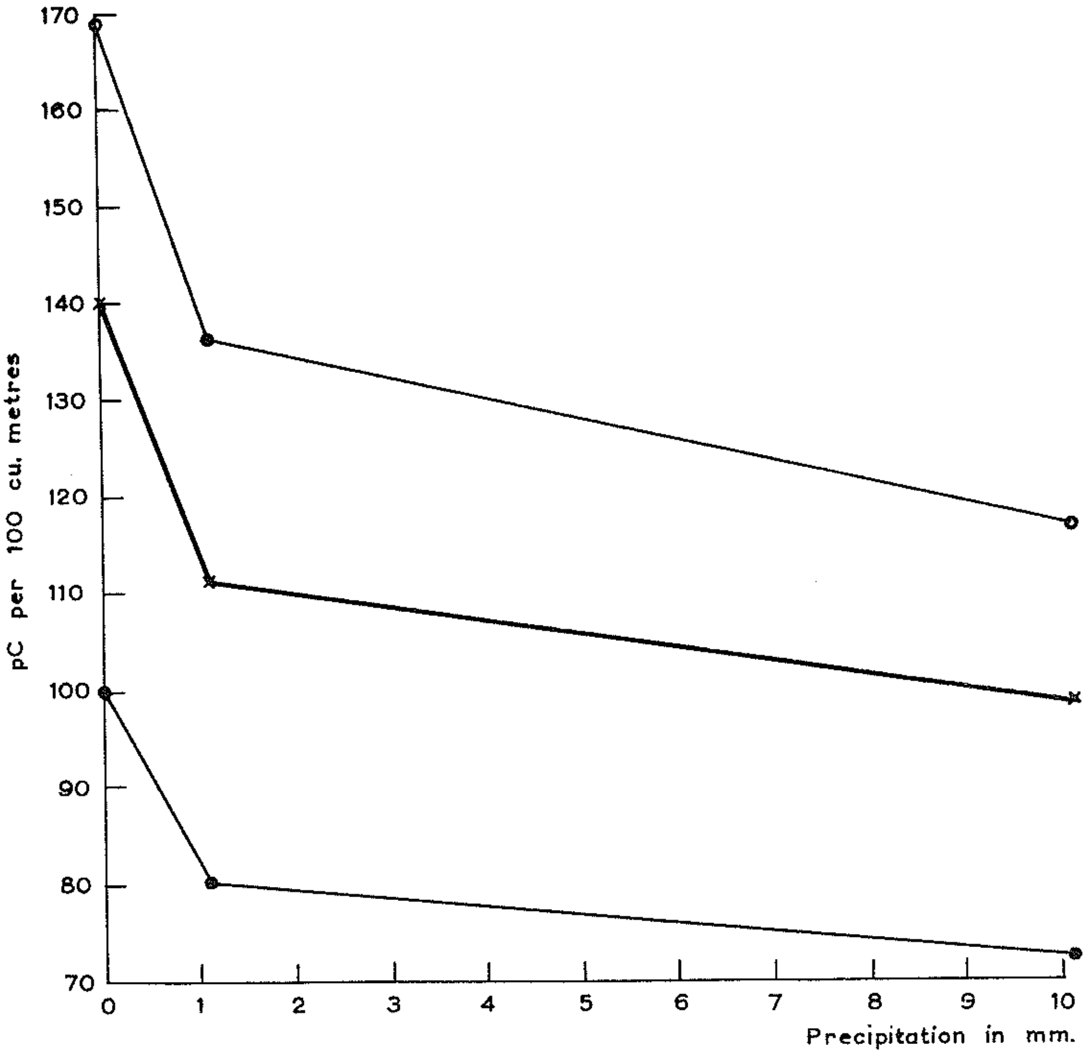

Fig. 1. Mean radioactivity of airborne particles at Valentia as a function of the precipitation. ●—● February 1957 to April 1958, ⊖—⊖ May 1958 to December 1959, ×—× February 1957 to December 1959.

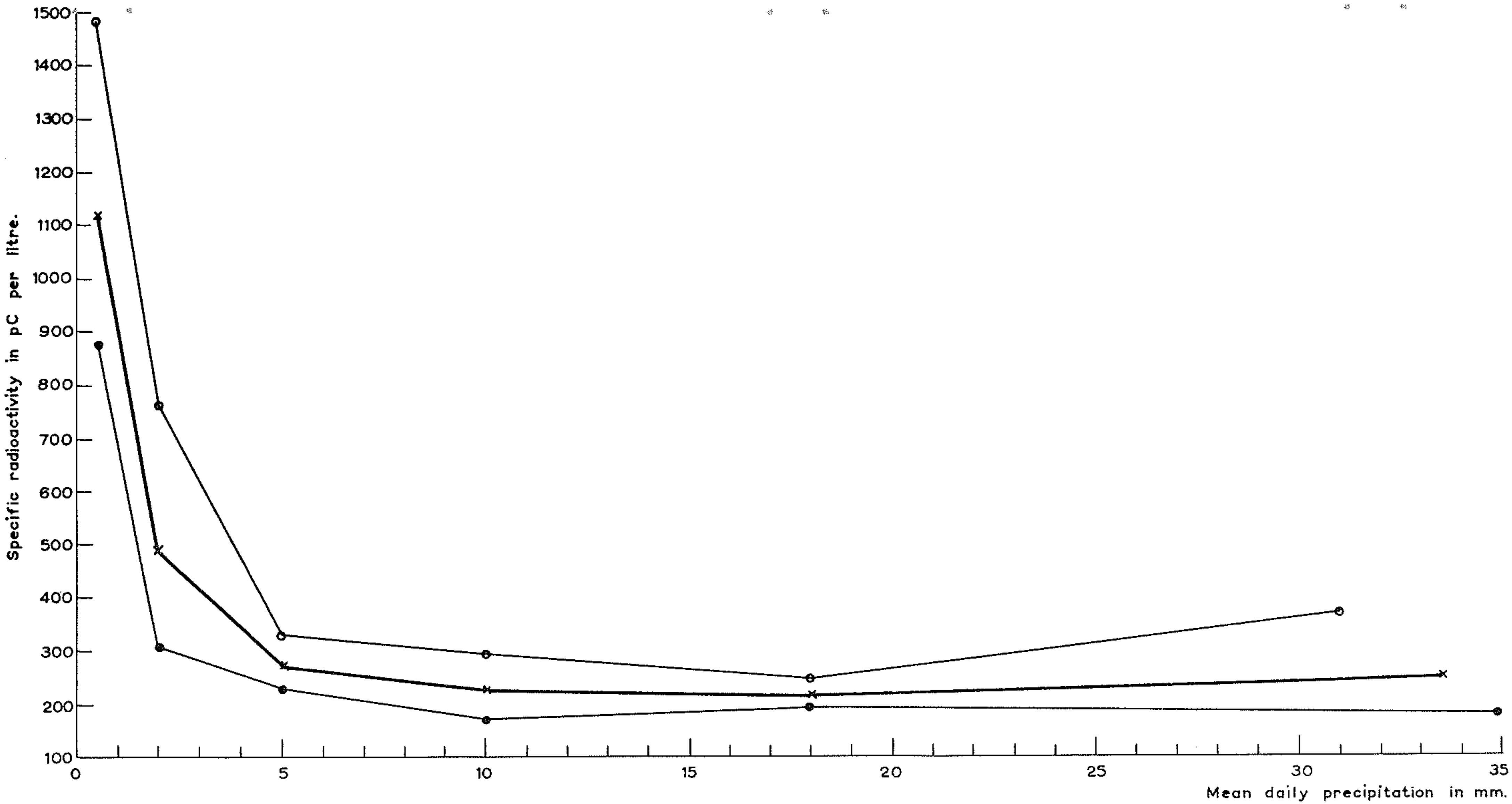

Fig. 2. Mean specific radioactivity of precipitation at Valentia as a function of the amount of daily precipitation.
●—● May 1956 to April 1958, ○—○ May 1958 to September 1959, ×—× May 1956 to September 1959.

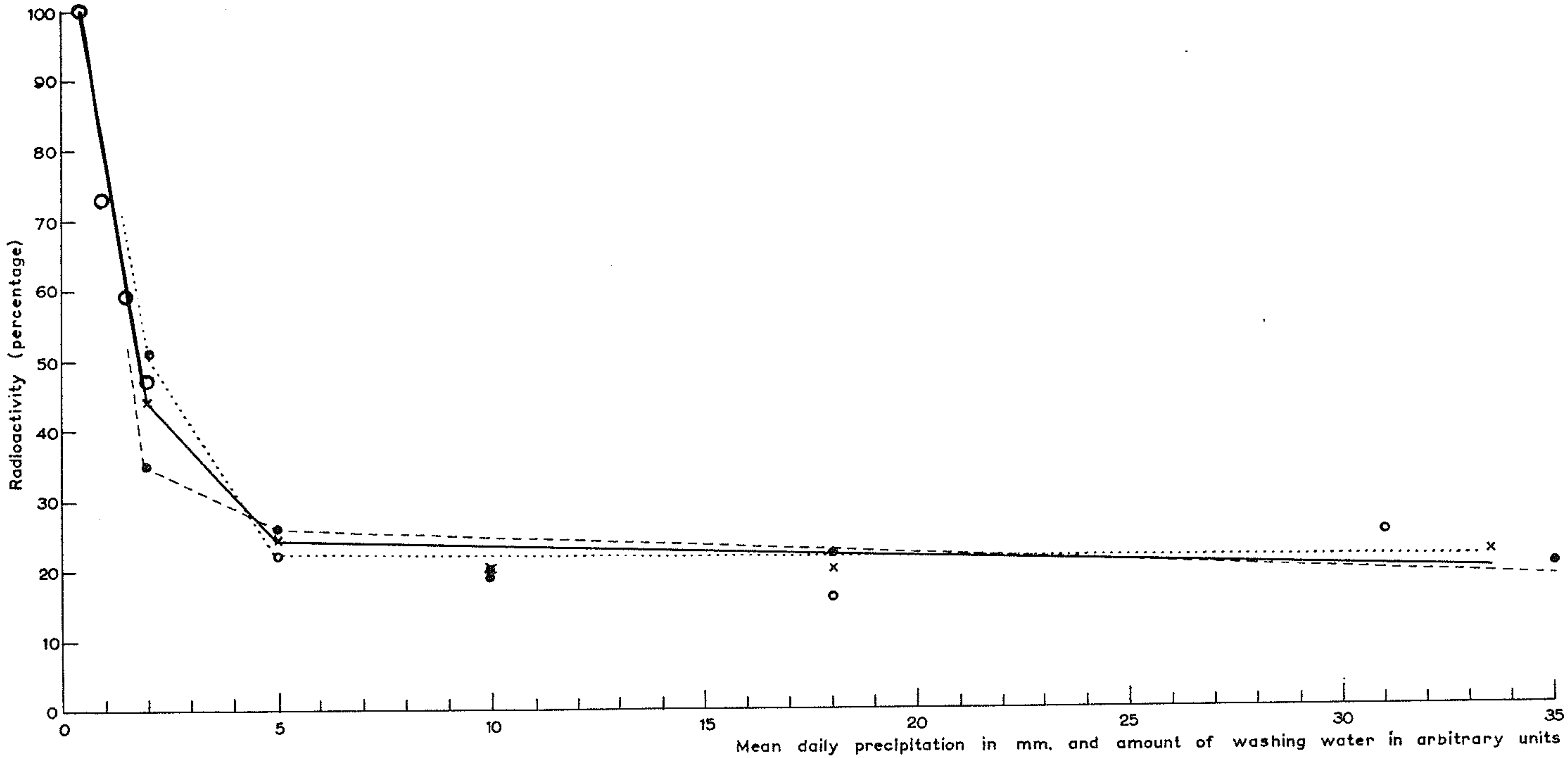

Fig. 3. Mean specific radioactivity of precipitation at Valentia (percentage) as a function of the amount of daily precipitation and radioactivity of washing water as a function of its amount. ●- -● May 1956 to April 1958, o----o May 1958 to September 1959, ×——× May 1956 to September 1959, O Washing observations.