STRONTIUM-90 IN ION-EXCHANGE RESIN USED IN THE AUSTRALIAN FIEFS NETWORK

A Statistical Analysis of Strontium-90 Data from Measurements reported by HASL on Unexposed Ion-Exchange Resin from Australian Fallout Stations and on HASL Quality Control Blanks.

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1. Estimation of the Strontium-90 Activity in Ion-Exchange Resin Before exposure to Fallout

In order to determine monthly strontium-90 fallout deposited at the eight Australian FIEFS* stations, account must be taken of the level of strontium-90 contamination of the ion-exchange resin as prepared for use in the FIEFS. This procedure has always been important in monitoring strontium-90 fallout deposit in Australia because the level of strontium-90 contamination of ion-exchange resin, supplied by manufacturers in the Northern Hemisphere, has remained of the same order of magnitude as the monthly fallout deposit in the Southern Hemisphere.

Each shipment of ion-exchange resin received from the manufacturer** is treated to reduce ionic contamination, mixed thoroughly and then divided into separate lots for use month-by-month over the ensuing years. The separate monthly lots are protected against introduction of strontium-90 contamination before use. Estimates of the strontium-90 activity of the ion-exchange resin, as prepared for use in the FIEFS, are obtained by

- analysis of samples from the resin bulk, extracted before separation into monthly lots, to gain a measure of the level of

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* FIEFS is an acronym for the Funnel Ion-Exchange Fallout Sampler employed in the Australian fallout monitoring network (Devlin et al, 1971).

** Permutit cation exchange resin, Zerolit 225 (H) commercial grade; 100 kg of resin, as a narrow column, was washed with 500 kg of 3N HCl and 400 kg of demineralised water.

strontium-90 activity before the shipment is introduced into service and

- analysis by HASL of one or more samples of unused resin taken from each monthly lot in turn, as this is brought into use with the FIEFS, and submitted as blinds to HASL with the monthly set of eight fallout samples.

Surveillance of the analytical procedures is exercised by HASL by the addition of quality control materials, including blanks, with the monthly sets of fallout samples.

Throughout use of ion-exchange resin from a shipment, a single value is adopted as the currently available best estimate of the strontium-90 activity of the unexposed resin. At the outset, this value derives from the measurements made before the resin was brought into service; after that, the value is regularly reviewed in the light of the accumulating data from the analyses by HASL. Revision of the value leads to recalculation of all previous results on monthly fallout deposits obtained using resin from that particular shipment. The procedure is continued until the entire shipment of resin is exhausted. A final review is then made of all data on the unexposed resin and final results are calculated, and reported, for all of the monthly fallout deposits determined using the resin.

The present paper records the statistical analysis of accumulated data on strontium-90 activities reported by HASL

- for unexposed ion-exchange resins measured with the monthly sets of FIEFS samples for the period March 1973 to October 1976, all derived from the one shipment of resin, and
- for HASL quality control blanks, measured with the same monthly sets of FIEFS samples.

The measurement results are summarised in Table 1 and their frequency distributions are depicted in Figure 1. These results include the activity of chemical reagents used in the analyses (Harley, 1977).

2. Statistical Tests

Standard statistical tests - the one-way analysis of variance to test the equality of group means and Bartlett's test for the equality of group variances - were applied to the data for unexposed ion-exchange resin, discussed in section 3, and to the results for HASL quality control blanks, discussed in section 4.

Statistical arguments were applied to the frequency distributions of the measurement results for the unexposed ion-exchange resin, and for the HASL quality control blanks, to show that the lower tails of both distributions are anomalous; this is discussed in section 5. Correlation between the two sets of measurement results is discussed in section 6.

All statistical tests were made at the 5% significance level.

3. Unexposed Ion-Exchange Resin from the Australian FIFS Network.

Inspection of the measurement results on the unexposed ion-exchange resin indicates that

- all results are reported as positive and
- results for the six-monthly period January to June, 1974, are systematically lower than for any other six-monthly period.

For convenience in the analysis, the data were divided, as far as possible, into six-monthly groups as follows

Group	Number of Results
March to June, 1973	4
July to December, 1973	6
January to June, 1974	6
July to December, 1974	6
January to June, 1975	6
July to December, 1975	6
January to June, 1976	6
July to October, 1976	4

One-way analysis of variance and Bartlett's test establish that both the group mean and variance for the period January to June, 1974, are different from those for the other periods.

Excluding the measurement results for January to June, 1974, the mean strontium-90 activity is 0.55 $\pm$ 0.06 dpm; the uncertainty term is the standard deviation of the mean of the 38 results. A t-test shows that the mean activity is non-zero and positive.

4. HASL Quality Control Blanks

Inspection of the measurement results for the HASL quality control blanks indicates that

- all results are reported as positive,
- results for the six-monthly period January to June, 1974, are systematically lower than for any other six-monthly period and
- for nine monthly sets, a separate result for the HASL quality control blank was not available.

In order to compose data sets of sufficient size for statistical analysis, and because of the gaps in the monthly results, the data were divided into six groups as follows

Group	Number of Results
March to December, 1973	7
January to June, 1974	5
July, 1974 to June, 1975	6
July to December, 1975	6
January to June, 1976	6
July to October, 1976	4

One-way analysis of variance and Bartlett's test establish that both the group mean and variance for the period January to June, 1974, are different from those for the other periods.

Excluding the measurement results for January to June, 1974, the mean strontium-90 activity for the 29 measurements, and its uncertainty, are 0.37 $\pm$ 0.05 dpm; by t-test, the mean activity is non-zero and positive.

5. Examination of the Frequency Distributions

It is expected that the reported strontium-90 activities for the unexposed ion-exchange resin, and for the HASL quality control blanks, would each be distributed normally, with mean zero, or near-zero and positive; whereas, the frequency distributions of the two sets of measurement results, shown in Figure 1, indicate that

- the results for the unexposed resins are in two distinct groups : namely, a widely dispersed set of activities centred about 0.5-0.6 dpm and a much narrower group below 0.10 dpm,
- the results for the HASL quality control blanks are in two groups : namely, a broad distribution centred about 0.4-0.5 dpm and a narrow group below 0.15 dpm.

Neither set of measurement results, as reported, would appear to be normally distributed in the region of lowest activity. This can be demonstrated by comparing the

confidence limits for the frequency of reported results with the frequency that would be expected. Using the normal approximation of the binomial distribution for a series of n repetitions of an event, with probability p of the event occurring with relative frequency h, the confidence limits for p are given by

$$p' = \frac{hn + K^2/2 \pm K\,(h\,(1-h)\,n + K^2/4)^{1/2}}{n + K^2}$$

where 100 $(1 - \alpha)$% are the confidence limits and K is defined by

$$\frac{1}{\sqrt{2\pi}} \int_{-K}^{K} \exp\left(\frac{-t^2}{2}\right) dt = 1 - \alpha$$

For 95% confidence limits, K is 1.96.

The confidence limits for the frequencies of reported results are given in Table 2 and compared with the expected frequencies assuming that the reported activities are normally distributed. It is evident from Table 2 that far more results are reported in the regions of lowest activity than would be expected; that is, unless it is accepted that the low activity results represent the entire lower tail of the distribution, including negative values.

6. Correlation between Pairs of Results reported for Unexposed Ion-Exchange Resins and HASL Quality Control Blanks.

The pairs of measurement results are plotted as a scatter diagram in Figure 2.

The plot reveals that for the lower activities of HASL quality control blanks, the results on unexposed ion-exchange resin range from near zero up to 1.0 dpm; whereas, at the lower values of activity of unexposed resin, the reported dpm of the HASL blanks range from zero only up to 0.1 dpm. This is inconsistent with the expectation that both sets of results would have a similar spread.

Therefore, before testing the pairs of measurement results for correlation, a pair was deleted if the unexposed resin activity was below 0.10 dpm or the HASL blank was below 0.15 dpm. The correlation coefficient for the remaining 22 pairs of observations is 0.44; this is non-zero and positive at the 5% significance level and would appear to be due to the retention, in all reported results, of the activity of the chemical reagents used in the analyses.

7. Re-estimation of Mean and Standard Deviation

From the foregoing analysis, it is evident that the reported measurement results are anomalous in the lower activity region. Therefore, in order to re-estimate population mean and standard deviation for the strontium-90 activities of the unexposed resin, the lower tail of the distribution of reported results, including this region, is to be truncated.

For a normal distribution truncated at x_1, the proportion of observations above x_1 is

$$1 - F\left(\frac{x_1 - \bar{x}}{\sigma}\right)$$

and the mean of observations above, and including, x_1 is given by

$$\bar{x} + \sigma \exp\left\{-\frac{(x_1 - \bar{x})^2}{2\sigma}\right\} \sqrt{2\pi} \left\{1 - F\left(\frac{x_1 - \bar{x}}{\sigma}\right)\right\}$$

where $F\left(\frac{x_1 - \bar{x}}{\sigma}\right) = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{x_1} \exp\left\{-\frac{(x - \bar{x})^2}{2\sigma^2}\right\} dx$

$\bar{x}$ is the mean of the normally distributed sample that should have been observed

σ is the standard deviation

$\bar{x}$ and σ are obtained from these expressions by use of tables of the area under the standard normal distribution.

For the unexposed ion-exchange resin, the re-estimated mean of the measurement results is 0.5 dpm and the standard deviation is 0.4 dpm. The values for the results on HASL quality control blanks are 0.3 dpm and 0.4 dpm, respectively. The t-test indicates that both mean values are non-zero and positive.

8. Lower Limit of Detection of Strontium-90 Fallout Deposit

Contamination of unexposed ion-exchange resin with strontium-90 imposes a lower limit on detectability of strontium-90 fallout deposit when the resin is employed in fallout monitoring with the eight Australian FIEFS stations. A lower limit of detection (LLD) for strontium-90 fallout deposit in the monthly fallout samples, measured by HASL, can be established by applying the reasoning of section D-08 of the HASL Procedures Manual (Harley, 1972) and assuming that the measurement standard deviation for unexposed resin, s, also applies to exposed resin at near-zero activities of strontium-90 fallout deposit. Then, LLD = 4.65s at a confidence level of 95% for detecting strontium-90 fallout deposit and at a 5% risk of falsely concluding that strontium-90 fallout activity is present, when it is not.

As s is estimated to be 0.4 dpm for the unexposed ion-exchange resin being employed with the Australian FIEFS stations, the LLD for strontium-90 activity in the fallout samples for these stations is 1.9 dpm; this corresponds to a strontium-90 fallout deposit of 0.012 mCi/km^2.

9. Conclusions

From the statistical analysis of the monthly measurement results reported by HASL for the unexposed ion-exchange resin used in the Australian FIEFS network from March 1973 to October 1976, and for HASL quality control blanks analysed with the unexposed resin, it is concluded that

(i) The means of both sets of measurement results, for unexposed resin and for quality control blank, are non-zero and positive.

(ii) The measurement results for the period January to June, 1974, are different to those reported for other periods. The reason for this difference is not known (Harley, 1977).

(iii) The lower tails of the frequency distributions of both sets of measurement results are anomalous; however, the anomalies are consistent with negative results being reported as positive, near-zero activities.

(iv) If (iii) is accepted, then re-estimation of the mean and standard deviation of the results on the unexposed resin gives 0.5 dpm and 0.4 dpm, respectively; the corresponding values for the results on the HASL blank are 0.3 dpm and 0.4 dpm, respectively.

(v) The two sets of reported measurement results are correlated with each other, evidently due to the retention, in all reported results, of the activity of the chemical reagents used in the analyses.

(vi) The lower limit of detection for strontium-90 fallout deposit, at the eight Australian FIFE stations employing the ion-exchange resin, is 0.012 mCi/km^2.

References

Devlin, D.A., Moroney, J.R., Munn, R.O. and Stewart, F.A. (1971). Defence Standards Laboratories Report AWTSC No. 1, 6-10.

Harley, J.H., ed., (1972) HASL Procedures Manual, USERDA report, HASL-300.

Harley, J.H. (1977) Private communication.

TABLE 1: SUMMARY OF [illegible] ANALYSED BY HASL FOR ANALYTICAL EXERCISE IN-CONJUNCTION WITH THE INTERNAL QUALITY CONTROL SCHEME.

(All results include the activity of chemical reagents used in the analyses)

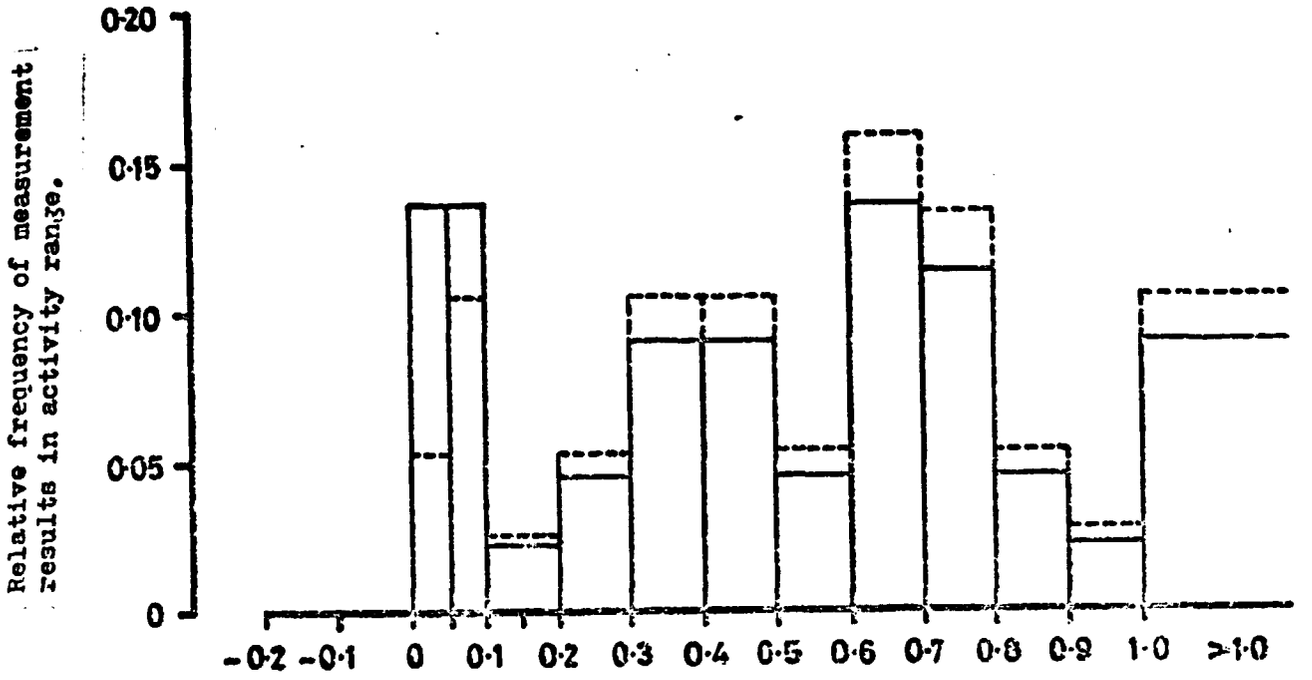
	1973		1974		1975		1976	
Monthly Set	Unexposed resin dpm	HASL blank dpm	Unexposed resin dpm	HASL blank dpm	Unexposed resin dpm	HASL blank dpm	Unexposed resin dpm	HASL blank dpm
January			0.07	0.07	0.06	0.09	0.50	0.47
February			0.02	0.02	0.37		0.64	0.23
March	1.13	0.11	0.02	0.01	0.46	0.27	1.04	0.09
April	0.06		0.02	0.02	0.50	0.06	0.85	0.76
May	0.64(1)	0.16(2)	0.06	0.02	0.05		0.63	0.37
June	0.80	0.61	0.02		0.32	0.44	0.95	0.56
July	0.75		0.82	0.02	0.52	0.32	0.73	0.66
August	0.39	0.52	0.02		0.11	0.08	0.56	0.88
September	0.67	0.20	0.65		0.74	0.13	1.18	0.75
October	0.72	1.01	0.27	0.37	1.43	0.70	0.70	0.24
November	0.16	0.11	0.37		0.50	0.48		
December	0.10	0.11	0.29		0.10	0.10		

(1) Used only in analysis of variance.

(2) Not used in any statistical analysis.

	Unexposed ion-exchange resin activity range: 0 to 0.10 dpm		NBSL quality control blank activity range: 0 to 0.15 dpm	
	Jan-Jun 1974 included	Jan-Jun 1974 excluded	Jan-Jun 1974 included	Jan-Jun 1974 excluded
Reported measurement results				
total number of measurement results	44	38	34	29
number of measurement results in the range	12	6	15	10
frequency of measurement results in the range	0.27	0.16	0.44	0.35
95% confidence limits on frequency	0.16, 0.42	0.07, 0.30	0.29, 0.61	0.20, 0.53
Expectation for normally distributed activities				
mean activity*	0.48 dpm	0.55 dpm	0.32 dpm	0.38 dpm
unbiased standard deviation*	0.36 dpm	0.35 dpm	0.28 dpm	0.28 dpm
expected frequency in the activity range	0.05	0.04	0.15	0.12
expected frequency for the entire lower tail of the distribution below the upper limit of the activity range	0.15	0.10	0.27	0.21

* estimated from Table 1; see sections 2 & 3 of the text.



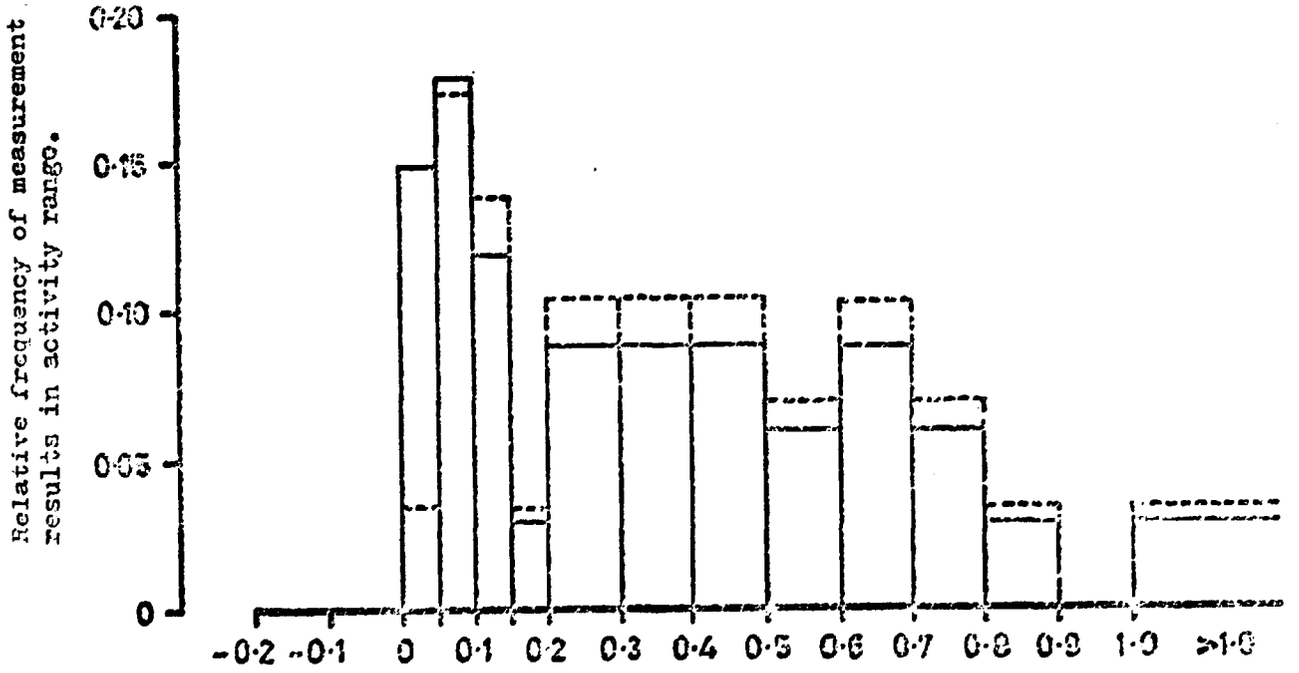


FIGURE 1: Frequency distributions of the monthly measurement results, from March 1973 to October 1976, reported by HASL for Australian unexposed ion-exchange resins and HASL quality control blanks

——— including results for January to June, 1974

------ excluding results for January to June, 1974.

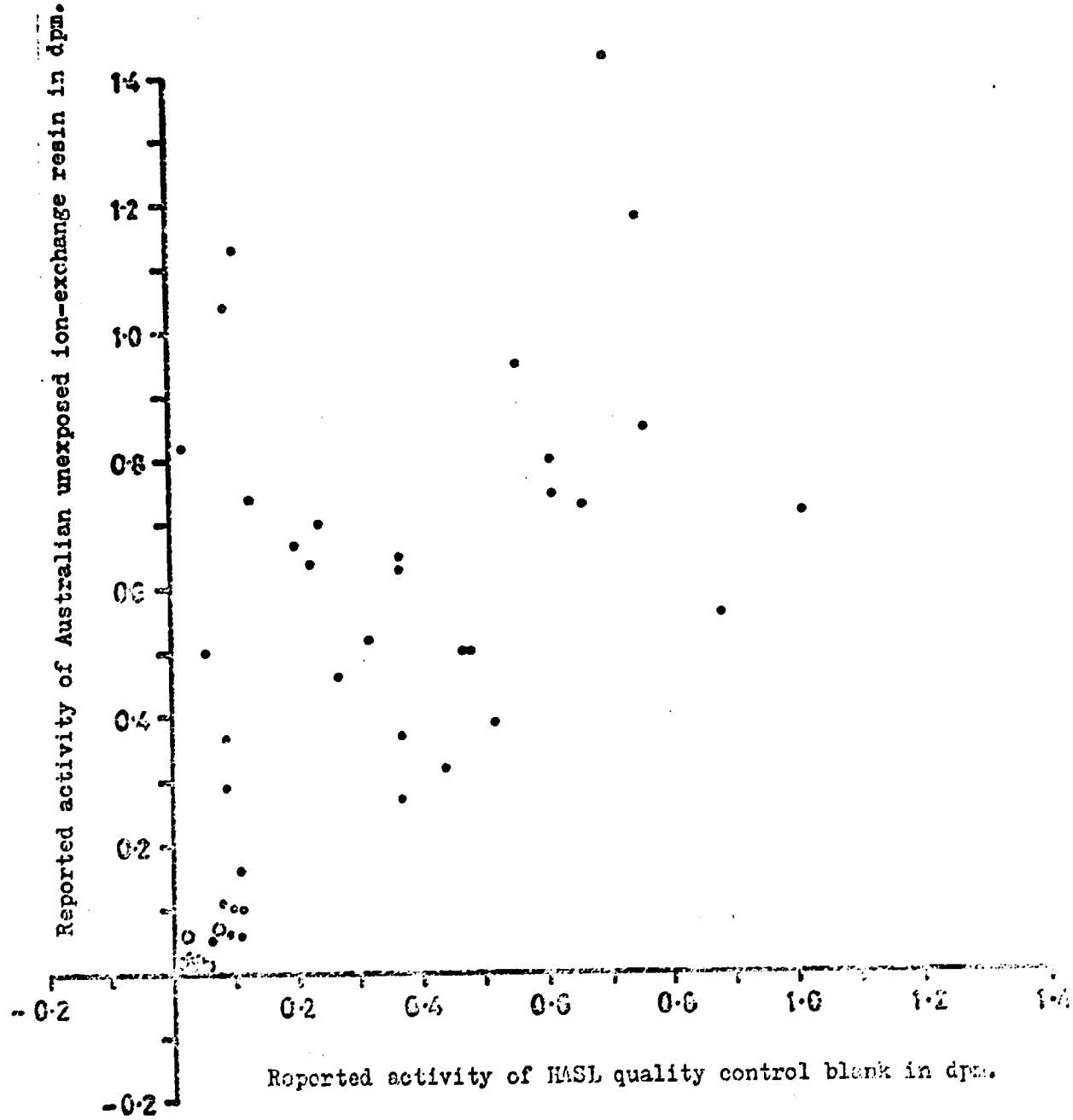


FIGURE 2: Scatter diagram of the pairs of measurement results, from March 1973 to October 1976, reported by HASL for Australian unexposed ion-exchange resin and HASL quality control blanks. The pairs of results for the period January to June, 1974, are distinguished as o.

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