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UNITED STATES ATOMIC ENERGY COMMISSION

CONTROL CHART METHOD APPLIED TO ERRORS IN RADIOACTIVE COUNTING

by

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CONTROL CHART METHOD APPLIED TO RADIOACTIVE COUNTING

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ABSTRACT

The Control Chart statistical methods, developed by Shewhart for the control of quality of menufactured products, are applied to the control of Geiger-Willer counting instruments. Experiments are reported to show the use of the Control Chart method for detecting disturbances in instrumental behavior and for detecting radioactive effects so weak that they are near the limit of detection of the instruments. As a corollary, the control chart can be used to reduce to its practical limit the time required for tests. CONTROL CHART METHOD APPLIED TO ERRORS IN RADIOACTIVE COUNTING

William G. Schlocht

INTRODUCTION

The study of weakly radioactive rocks and minerals by the Section of Chemistry and Physics of the Geological Survey has included determinations of radioactivity by Geiger-Müller counters. Many of the samples examined are so weakly radioactive that their effect on a counting instrument is of about the same size as that of the surroundings. The radioactivity to be determined thus may be near the natural limit of precision of the instrument, which is imposed by the local background effect. There is also another serious source of error in the determinations; the counting apparatus itself is delicate and is subject to many disturbances, some of them so subtle as to easily escape notice unless constantly watched for. It is therefore important that ne precaution be overlooked that will help to avoid mistakes.

A series of experiments made in this laboratory shows that the modern statistical methods developed by Dr. Walter A. Showhart at the Bell Telephone Laboratories are very useful for handling problems arising from both of the sources of error mentioned, background fluctuations and instrumental defects. The ContSol Chart method is a practical approach both for indicating defective behavior of the counting instrument and for determining very weak radioactivities.

QUALITY CONTROL

The Shewhart methods were developed to provide an economic basis for control of manufacturing and inspection of products used by the American Telephone and Telegraph Company. They have been widely adopted in industry under the sponsorship of the War Department during the second World War. The application of statistical methods to quality centrel has made possible an adequate and comprehensive practical test of statistical theory. In manufacturing processes, operations and tests are repeated many thousands of times, providing at no extra expense masses of data that could not be collected elsewhere; no one could finance the repetition of experiments so many times unless they were incidental to some economic process. The statistical methods found practicable and dependable in control of manufactured product are also useful in the interpretation of scientific experiments, because the same basic problems are often involved. They are problems of predicting, from past data, where future points would fall if there were no change in the process.

The outstanding features distinguishing Shewhart's quality control methods from the older statistical methods are the grouping of observations and the retention of their sequence. These are accomplished by the centrel chart, in which successive groups of measurements, that is, samples, are plotted in the order in which they were made. This gives a test of the randomness of the fluctuations in results, which previous methods did not provide. Any patterns of regularities in the arrangement of the points on the chart indicates that the process is "out of

control," that is, the variations are not rendom. If the variations are random, control limits can be calculated, outside of which observations will seldom fall unless the process is changed. This provides a sensitive test for disturbances in the process. The most useful limits are the "three-sigma" limits; if a point falls outside these control limits, the assumption that some disturbance has taken place in the process will almost always be correct. The randomness test gives the other check of control; the appearance of any pattern or regularity on the chart shows that the process has changed.

Experience with statistical methods in industry has made us realise that the ideas of absolute precision and perfect certainty are a particularly expensive--and also pointless--form of uncompromising idealism. By spending enough time and money we may greatly increase the precision of a measuring process -/; but there

/ Before the last revolution in physics, it was considered that we might in this way approach as close as we please to the "true" value of a quantity; but since actual constructive knowledge has been deduced by recognizing that there is a natural limit to precision in the description of a physical system, the idea of perfectly exact values of quantities must be considered meaningless.

is a point beyond which it is not worthwhile to go. This degree of certainty is almost always that defined by the 3σ limits; if we not as if a point outside the 3σ limit is assignable to a change in the process, we shall seldom be wrong. It ign't worthwhile to be any righter; we can't afford it.

The 3σ limits are empirical. They were chosen not because the chance of exceeding them is some particular value, but because they work best in practice. The adoption of this eptimum condition, arrived at by experience, may be thought of as a definition of "practical certainty." As long as the process is in control, we are "practically certain" that a point will never fall outside the control limits; if it does, we are "practically certain" that the evidence of a change in the

process is significant. Although the calculations are formally based on the normal distribution, this rule of behavior holds for any distribution likely to be met with in practice.

Specific directions for making and using cantrol charts are published in two pamphlets by the American Standards Association (1, 2); these and the A. S. T. M. Manual on Presentation of Data (3), Supplement B, contain a table of fractors for computing control limits. The meaning of the procedure is clearly and concisely described in an article by Colonel Simon (4). Dr. W. Edwards Deming has given a practical discussion (5), and Dr. Shewhart's published lectures (6) give a more detailed and philosophical discussion of Quality Control methods. Shewhart (7) and Simon (8) have written comprehensive textbooks on the subject.

The Control Chart method is clearly described in the literature listed, so no attempt is made in this report to give detailed directions for using the method. It involves much computational labor, but this cannot be avoided by any method if observations are to be fully utilized, to the limit of their precision.

Useful as statistical methods are, it cannot be too strongly emphasized that they are no substitute for technical knowledge and experience. When an instrument does not operate properly, the hunch of a competent technician may be far more efficient than the soundest statistical methods in locating the trouble. Nevertheless, they are a valuable aid to the technician who understands the instrument; and insofar as they are based on common sense, they can be of help to the untrained operator. The

control chart does not usually suggest what is wrong, but it does give a sensitive indication that <u>something</u> is wrong. Furthermore, it provides a practical test of hypotheses as to what may be wrong.

CONTROL CHART FOR INSTRUMENTAL BEHAVIOR

Statistical methods can be applied to errors of instruments only if the errors are random. It is often considered that if the variations in a measurement have the same distribution function as those following a random law, the actual variations are random. The condition that fluctuations have a frequency distribution like that of some random law is necessary but not sufficient to establish their randomness, because the frequency distribution is not a complete description of the random law.

The comparison of frequency distributions is thus an extremely delicate method for detecting certain kinds of defects in instruments; an outstanding use of it has been the application of interval counters to detection of spurious counts in Geiger-Küller counters (9, 10, 11). If a counting tube gives spurious pulses, its frequency distribution for a given source will show too many intervals containing high counts, or too many short intervals between pulses. Thus if a counting tube does not pass the test with an interval selector, it is defective. But a counter that does pass this test is not necessarily free from all defects. Figure 1 shows run charts illustrating this point as applied to an experiment with an interval selector. Davis and Curtiss (Ref. 11, fig. 4) give a curve for distribution of intervals between alpha-particle counts in an ionisation chamber, with a mean of 59 counts per second. Figure 1, A shows a sequence of 50 one-second counts from a hypothetical random

source following the Poisson law, and having a mean of 59 counts per second. Such a process would have the distribution of intervals shown in Pavis and Curtiss' curve, and the run chart gives no reason to question its randomness. Figure 1, B and C shows sequences of counts having the same frequency curve as the random sample, as shown by the histograms on the right. They would give the same distribution of intervals, but a counter showing such strange behavior in sequence of counts would certainly call for further investigation.

A frequency-distribution comparison is not a complete test for randomness, because the sequence of the fluctuations is discarded in collecting together all the observations in a given size range. The frequency test lacks the essential condition to detect "runs" or patterns of regularity in the sequence.

"Run charts" are used in Quality Control to watch for patterns in the sequence. An empirical rule sometimes found useful is (12) that if as many as seven successive observations are all above the mean, all below the mean, or successively increase or decrease without alternation, the process is out of control. This test is possibly too delicate, that is, it may lead to suspicion of disturbances somewhat oftener than is justified.

The important feature of the Shewhart method is the use of a control chart. ^Ubservations in sequence are grouped into samples, to get an estimate of the dispersion about its mean of the unlimited population of possible samples from which they were taken. The control

limits are placed at a distance of three times this estimated standard deviation on either side of the central line. The estimate of the 30[±] control limits for the sample means is made from the average value of either the sample ranges or the sample standard deviations. Figure 2 is a control chart for 100 samples of 4, of which the first 50 individual counts are shown in the run chart of figure 1A, taken from the hypothetical random population with mean of 59. It shows that the estimated mean and control limits for the parent population of samples are not determined exactly, but are estimated more precisely as the number of samples increases.

A control chart is also made for the scatter or dispersion within the samples, expressed either as range (difference of the extreme values within a sample) or as sample standard deviation. The control charts for ranges and standard deviations have been found more sensitive to short-time changes (changes within the subgroups) in the process than the control chart for means.

CONTROL CHARTS FOR GEIGER-MULLER COUNTERS

Laboratory Background Counts

Experiments were made to see if the Control Chart method is useful for estimating the errors in Geiger-Müller counting apparatus, for detecting disturbances in the instruments, and for detecting weak radioactivities with the apparatus. If the control chart shows that the fluctuations in counts from a constant source are random, statistical formulas for error may legitimately be applied. A sample point falling outside the control limits is an indication of a change in the process,

and the instrument is not reliable until the cause of the disturbance is removed. Finally, if a counter is known to be "in control," any change in the conditions that will reproducibly throw it out of control may be assumed to be the cause of the loss of control; so if a rock specimen brought near the counter results in points outside the control limits for standard conditions, it may be considered significantly different in radioactivity from the standard.

For low counting rates, a constant background is a suitable standard condition; for high rates, control charts might be made for operation with a radioactive standard in the specimen holder.

The laboratory experiments were made with an A. C. operated scaleof-eight counter, registering on Cenco High Voltage Impulse Counters. The field counts were made by counting clicks heard in the earphones of a battery-operated portable gamma-ray counter. Counts were made for five-minute periods.

Because of the limited data available, the groupings of observations were restricted to the smallest sized samples practicable, so as to have as many points as possible on the control charts. For the laboratory experiments each point on the control charts represents a group (sample) of four observations, each observation being a 5-minute count; for the field observations each sample point represents a group of three 5-minute counts. These are the smallest sample sizes that give significant results.

Laboratory background counts

Figure 3 shows background counts with a gamma-ray counting tube containing organic vapor, made by the Geophysical Instrument Company for use in the "Series G" portable counter. In operation with the laboratory scale-of-eight circuit, the counter was not shielded by the brass tube used in the field work. Each point represents a sample of four 5-minute counts, reduced to counts per minute. The successive points represent background counts taken on different days over a 2-1/2 months period. The charts show satisfactory control; the control limits for samples 37 to 52 agree with those predicted by the previous samples (see Appendix 1).

Figure 4 shows the control charts for background with a beta-particle counting tube that changed its characteristics after some satisfactory performance. It was a Herbach and Rademan window tube, Type GLB 20. Each point represents a sample of four 5-minute counts, expressed in counts per minute. The control chart shows how a point outside the control limits gave warning of a change in the mean background count and in the control limits.

Figure 5 shows control charts for samples of four 5-minute back-Fround counts, in counts per minute, with another Herbach and Rademan GLB 20 beta-counting window tube, taken over a 7-weeks period. The first 43 points represent samples taken on successive days; the later samples were taken at the rate of 2 to 6 points each day. The apparatus was in control, and the actual performance after the 45th sample was not significantly different from that predicted. This tube was used in

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the experiments on the detection of weak radioactivity, because of its satisfactory record of control. Control charts for ranges and standard deviations are both given, to show the close similarity in their behavior. The range is much simpler to calculate, and is just as good as the standard deviation for indicating the state of control. The run chart for these data is shown in figure 6; it shows no patterns or runs. Hach of its points represents a single 5-minute count, reduced to counts per minute.

Field background counts

In the experience of the Geological Survey, battery-operated field counters have seldom given evidence of being in control. The batteries run down and recover in irregular ways, and the instruments are not equipped to maintain constant circuit conditions. The instruments are often moved from one location to another before enough background counts can be made to establish control with a chart. If a counter is kept in ene place for many days, and is found to be in control, the control chart method may be used to reduce the time needed for testing specimens, or to enable weaker specimens to be tested.

Figure 7 shows control charts for background from 219 5-minute counts made by Kenneth Brill with a Series G portable field counter, kept at one field station, over a period of 2-1/2 weeks. Each point represents the mean of a sample of three 5-minute counts, expressed as counts in five minutes.

The control chart shows no patterns, and only three points actually out of control, but too many points are near and on the control limits. Since the points look nearly in control, Mr. Rupert Gause suggested the

use of an unpublished method derived by him, for estimating the average non-random variation in certain Army Ordnance test measurements, made with instruments subject to a drift in their zero reading or "blank." From estimates of the variation within the samples and of the zet of samples as a whole, a coefficient is calculated that represents the average non-random drift, so that a long-run estimate can be made of what part of the fluctuation is random (See Appendix 2).

For this field counter, Gause's test gives an estimate that about 0.4 of the total fluctuation is non-random. Thus for this particular instrument and location, the mean background calculated from the long-run behavior is at least as good an estimate as that of any particular subgroup of observations.

DETECTION OF WEAK RADIOACTIVITY

Choice of Action Limits

If a counting instrument is in control, the control chart method can be used to shorten the time required to test a specimen, or to test weaker specimens than usual. Any mineral specimen that throws the instrument out of control is practically certain to be radioactive. The definition of practical certainty may be modified to suit the purpose of the test. The control limits are action limits, and the choice of action limits depends on the purpose of the action. If we wish to be right the optimum number of times in testing specimens, we adopt the 3s² control limits for the criterion of practical certainty. If we want to avoid any further examination of specimens that might not be radioactive, as in searching for high grade ores, wider control

limits are used. More usually, if we are looking for very weakly radioactive materials, we may want to be sure that no samples are discarded without further testing, if there is the least chance that they may be radioactive. When it is more important that no weak speciments be overlooked than it is to waste time in repeating tests on some non-radioactive specimens, narrower limits can be used. For example with 20° limits, about 5 percent of the specimens chosen for further testing will be non-radioactive.

Statistical Tests of Significance

In specimens too weak to give a point outside the background upper control limit, the radioactivity may still be strong enough to give a run of points above the mean. In principle, such weak radioactivity may be detected with practical certainty if enough observations are made. The various refined statistical tests of significance may be applied to runs, but must be interpreted cautiously in view of their limitations (ref. 13, pp. 137-8).

Examples

Use of the control chart method to detect weak radioactivity is illustrated by some experiments made with the laboratory scale-of-eight counter, using the beta-counting tube that gave the record of controlled hackground fluctuations shown in figure 5. Between the background counts shown on the control chart, counts were made by putting the specimens, in a 30 ml beaker, about 4 mm below the window of the vertically mounted counting tube. Samples of four 5-minute counts were recorded, taken on different days when possible.

A specimen of river gravel was sorted into three fractions, according to their mineral composition, by Robert L. Smith of the Geological Survey. Counts were made to determine which of the fractions is most radioactive. The largest part, Fraction B, weighing 108 grams, was compared with an equal bulk of a standard mixture containing 0.17 e/e uranium; figure 8 shows this comparison along with background counts made during the same period. The standard gave a mean of 11.8 counts per minute above mean background count, so the factor $\frac{0.17}{11.8} \pm 0.014 \leq U$ for each count per minute above background was used to calculate the radioactivity of the specimen. Four of the six points for Fraction B are above the upper background control limit, so it is certainly radioactive; the mean is 3.2 counts per minute above mean background count, giving a radioactivity equivalent to 3.2 x 0.014 = 0.04 $\leq U$.

The next largest fraction, C, weighed only 52 grams, so it was compared with an equal weight of fraction B, about equal to it in bulk. As shown in figure 9, all the points for fraction C are inside the background control limits, so it is not certainly radioactive. The mean count for fraction C is 0.4 counts per minute above mean background; a significance test suggested by Dr. W. Edwards Deming shows that the difference is not certainly real. The control limits for the specimen are 1.7 counts per minute above and below the mean. The width of the control limits for the five points is divided by the square root of 5, giving control limits 0.75 counts per minute above and below its mean for a sample five times as large as that represented by each original point. The mean background is inside these control limits, so that the

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background is not certainly different from the counts made on the specimen. These control limits are shown enclosing the larger circle, representing the lumped points, at the right of the control chart.

The effect from 52 grams of Fraction ^B is certainly real, because 3 of the 5 points are above the upper background control limits. If the activity of Fraction C, 0.4 counts per minute is real, it is only about 1/7 that of Fraction B, which is 2.8 counts per minute above background.

The smallest part, Fraction A, weighed 21 grams, and so was compared with the same weight of Fraction B (with about the same bulk). 21 grams of fraction C was also tested. In figure 10, three of the five points for fraction B are shown above the upper background control limit; so that its effect is real even in a 21 gram portion. The mean effect is about $\frac{0.04}{2.4} = 0.017 \%$ U for each count per minute above background. Fraction A gives points all within the background control limits, but it almost passes the lumped control limit test; if a few more points were obtained that did not change the mean and control limits appreciably, the difference of 0.9 counts per minute above background, equivalent to 0.9 x 0.017 = 0.02 % U, could be considered significant.

As shown by the control charts, if Fraction A is radioactive, it is more so than Fraction C whose mean difference from background, 0.6 counts per minute, is equivalent to 0.6 x 0.017 = 0.01 % U. The control limits for Fraction C differ by 2.1 counts per minute from its mean; the number of points necessary to be sure that the mean of 0.6 counts per minute above background is significant can be estimated by the qondition that the width of the lumped control limits be less than 0.65

$$\frac{2.1}{\sqrt{n}} < 0.6, so \quad n > \left(\frac{2.1}{0.6}\right)^2 = 12.$$

At least 7 more points would have to be obtained, <u>all</u> consistent with the control limits and mean already found, to be practically certain that Fraction C is radioactive.

The control chart method thus shows in which of the three fractions the radioactivity of this small sample of gravel is concentrated, and also permits a rough estimate of its activity to be made. Still more information can be had if it is worth the time.

A simple rough test of significance of runs is illustrated in figure 11. Control charts are shown for two specimens whose points are inside the background control limits, but are all above a cortain level. We take the lowest point of the run and calculate the chance that a sample mean, from a normal distribution with the same mean and control limits as the background, will exceed this lowest value. It is the area P under the normal curve and above the given limit; and the chance that a successive points will all be above the given limit is the n^{th} power of P. If Pⁿ is very small, it is unlikely that the run is a background fluctuation. To correspond to the 3_{0^-} limit for a normal distribution, we may say that a run is certainly different from background if Pⁿ is smaller than 0.001.

Five samples of four 5-minute counts were made with a slab of manganese ore under the counter tube window. The control chart for sample means, in counts per minute, shows that none of the five points are below 10 counts per minute; the area of the background distribution curve above this level (14) is P = 0.09. The chance that the background will give such a run is therefore not greater than $P^5 = 10^{-5}$; so the difference is probably significant.

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Another set of five count samples was made with a placer specimen, No. 492, under the tube window. The lowest point on the control chart is only 0.4 counts per minute above the background mean. The chance that a background sample will exceed this value is P = 0.3, and so $P^5 = 0.003$; the run is not certainly different from background by this test. A few more points would be nesessary to settle the question.

SUMMARY

The examples given show the advantages of the control chart method in planning for the optimum or most economical use of counting instruments, and in extracting the greatest possible amount of information from the observations. When materials of fairly high activity are to be tested, the desirable procedure is to use methods of greater sensitivity than the expected variations. With weaker activities, this may not be possible because available instruments are not sensitive enough, or because it would take too long. Then the control chart method makes it possible to test as many specimens as possible in a given time at predetermined levels of certainty, or to test them with the optimum precision that is economically desirable.

ACKNOWLEDGEMENTS

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-19- **Appendix** 1 Sample calculation of control limits Samples of 4 5-minute counts, in counts per minute. For an example we test the limits, predicted by the first 30 sample points on the control chart for the r-ray counter (fig. 3), by calculating the means and control limits for the counts actually observed in the next 13 samples of 4. For the first series of 30 sample points, the mean is 3.50 counts per minute with control limits of 4.72 and 2.28; the mean range is 1.69 with control limits 3.85 and 0.

Sample Number	Observations				Mean	Range
	Xi	×2	X ₃	Xų	X	R R
37	4.4	3.0	3.8	4.4	3.90	1.4
39	3.4	4.4	3.4	2.8	3.50	1.6
40	3.4	2.2	3.2	3.8	3.15	1.6
41	2.8	4.0	2.6	4.0	3.33	1.4
43	3.6	3.8	4.2	2.2	3.45	2.0
44	5.0	3.4	2.4	3.2.	3.50	2.6
45	3.2	2.8	4.2	4.4	3.65	1.6
47	4.0	3.4	3.6	5.2	3.80	2.6
48	3.4	3.2	2.8	2.8	3.05	0.6
49	3.8	2.8	3.4	5.0	3.75	2.2
50	4.0	3.6	3.8	2.6	3.50	1.4
51	3.4	4.4	1.8	4.2	3.45	2.6
52	3.6	2.6	3.0	3.4	3,15	1.0
Totals Means		-	anaa ay ahaa ahaa ahaa ahaa ahaa ahaa ah		13 45.20 $\overline{X} = 3.48$	$\frac{13[22.6]}{R=1.74}$

Observed means, last 13 samples of 4

The observed means agree satisfactorily with the predicted ones.

Mean	Predicted	Observed	
Mean, 🕱	э.50	3.48	
Range, R	1.69	1.74	



the factor for control limits for the means is
$$A_2 = 0.729$$
, and
the factors for control limits for the ranges are $D_3 = 0$ and
 $D_4 = 2.28$.
Control limits for the means:
 $\overline{X} \pm A_2 \overline{R} = 3.48 \pm 0.729 \times 1.74 = 3.48 \pm 1.27 = \begin{cases} 4.75\\ 2.21\end{cases}$
Control limits for the ranges:
Upper control limit: $D_4 \overline{R} = 2.28 \times 1.74 = 3.97$
Lower control limit: $D_4 \overline{R} = 0 \times 1.74 = 0$

The observed control limits agree satisfactorily with the predicted ones.

Control limits for	Predicted	Observed	
Means, X	4.72	4.75	
Ranges, R	3.85	3.97 0	

APPENDIX 2. Gause's estimate of non-random drift.

An estimate of non-random drift in the sero-reading of an instrument is made from the control chart for mean sero-reading, by estimating a, the fraction of the fluctuation that is caused by non-random effects.

 σ_y^2 , a measure of the average fluctuation within samples of n observations, can be estimated from the mean range \overline{R} . It is an estimate of the average <u>random</u> fluctuation, and its ratio to the average <u>total</u> (random plus non-random) fluctuation, $\sigma_{\overline{X}}^2$, gives the fraction of the total variation that is purely random.

For points each representing a sample of n observations, $\sigma y^2 = \frac{\overline{R}^2}{n d_2^2}$ is calculated from the mean range \overline{R} and the factor d_2 (found in Table 1 of reference 1, p. 50), as a measure of the random part of the fluctuation of the points about the mean line $\overline{\overline{x}}$.

 $\mathcal{O}_{\overline{X}}^2$, the variance (standard deviation squared) of the sample points \overline{X} about the mean line $\overline{\overline{X}}$, is taken as a measure of the total fluctuation, random and non-random.

Gause's estimate of the fraction of non-random fluctuation is

$$a = 1 - \frac{\sigma_{1^{2}}}{\sigma_{1^{2}}} = 1 - \frac{\frac{\bar{R}^{2}}{n \, d_{2}^{2}}}{\sigma_{1^{2}}}$$





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ages remained at the standard values.

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se they are included in the control limit calculations.



proces etton of a . confirming the predi trol limits





There are no patterns or "runs" to indicate lack of control. ackground in count obtervition of five minutes. shows in figure 5.



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FRACTION C, 52 g.

Figure 9. Weakly radioactive river gravel, employ of four 5-minute counts. 52 grans of Fraction 6 compared with an equal amount of Fraction B, plotted in counts per minute on the background control chart for means. B is practically certainly radioactive, while 6 is not; the mean background line falls inside the lumped control limits for the five sample means of 6, taken as a single sample of Em4 = 20.



Figure 10. Weekly radioactive river gravel, samples of four 5-minute counts.







PLACER SAMPLE NO. 492

Figure 11. A rough test of significance of runs. Samples of four 5-minute counts, plotted in counts per minute on the background control shart for means. The shaded area 2 under the background distribution surve is the chance that a background sample will be greater than the lowest sample mean found for the specimen; so the chance that a successive background samples are greater than this minimum value is \mathbb{R}^n .

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