Application of Statistical Techniques to Environmental Radiation Surveillance

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IN AN environmental surveillance program, such as that operated by the Division of Radiological Health, Public Health Service, a primary function is the analysis and measurement of radionuclide concentrations in a variety of samples. Typical media analyzed under this program include air, water, milk, selected food items, and various types of biota. As with any other experimental work, there is an error associated with each determination. The purpose of this paper is to review some of the statistical techniques employed to estimate and minimize these errors.

Sources of Error

Sampling. Since, in many instances, the data obtained by radiochemical assay are used to estimate the radioactive intake of the general population, the samples must be representative. Although the selection of samples for surveillance programs is usually beyond the immediate control of the analytical staff, it is important to realize that invalid sampling can be a major source of error.

In the Pasteurized Milk Network of the Public Health Service, collection procedures were designed to obtain reasonably representative samples. In this program, collectors in 62 major U.S. cities submit weekly 1-gallon milk samples, which represent 80–100 percent of all fluid milk consumed in that city. Ideally, the sample is collected by drawing from each major milk processing plant a volume proportional to its share of the market. Table 1 illustrates the operation of this method for Cincinnati in March 1963. Similar considerations are applied in the selection of samples for other networks operated by the Public Health Service.

Sampling procedures are being investigated in a study of the data from the network sample in Boston and from each of the six major milk distributors serving the Boston area. The agreement between the weighted average of these individual measurements of cesium 137 and the analytical results of the composite network sample is shown in figure 1.

Counting. Although improper sample selection could be a major source of error, the laboratory is more directly concerned with the analytical errors, which are more amenable to control.

A major source of experimental error arises from the random manner in which atomic nuclei disintegrate. The probability that any single atom will disintegrate in a given time is small and constant. Such processes are described by the Poisson distribution which may be expressed by the relationship,

$$f(x)=\frac{e^{-\mu}\,\mu^{x}}{x!}.$$

The distribution function, $f(x)$, gives the probability of observing x counts in a given time when μ, which is usually not known, is the true

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average number of counts estimated from a large number of replicate counts. The probability of observing x_i or fewer counts is given by the cumulative distribution function,

$$F(x \leq x_i) = \sum_{x=0}^{x_i} \frac{e^{-\mu}\mu^x}{x!}.$$

The Poisson distribution is characterized by the fact that the standard deviation is equal to the square root of the mean (*1*).

The corresponding normal distribution, with mean μ and standard deviation $\sqrt{\mu}$, is a good approximation to the Poisson distribution with mean μ except when the number of counts is small (<30). This permits application of statistical theory of normal populations to counting statistics. The Poisson and normal distributions are described in standard statistics texts (*2*). Symbols and their definitions used throughout this paper are given in the nomenclature key.

The true values of μ and σ are never known; therefore, in experimental work they are replaced by their best estimates, $\bar{x}$ and s'_x. In most radiation counting, the count rate, which is equal to x/d or y, is used in preference to the number of total counts. The distribution of the counting rates is also approximately normal with a standard deviation of $\sqrt{x}/d = \sqrt{x/d^2} = \sqrt{y/d}$. Thus approximately 68 percent of replicate observations of a counting rate should fall within the interval, $\bar{y} \pm \sqrt{\bar{y}/d}$, and approximately 95 percent within $\bar{y} \pm 1.96\sqrt{\bar{y}/d}$.

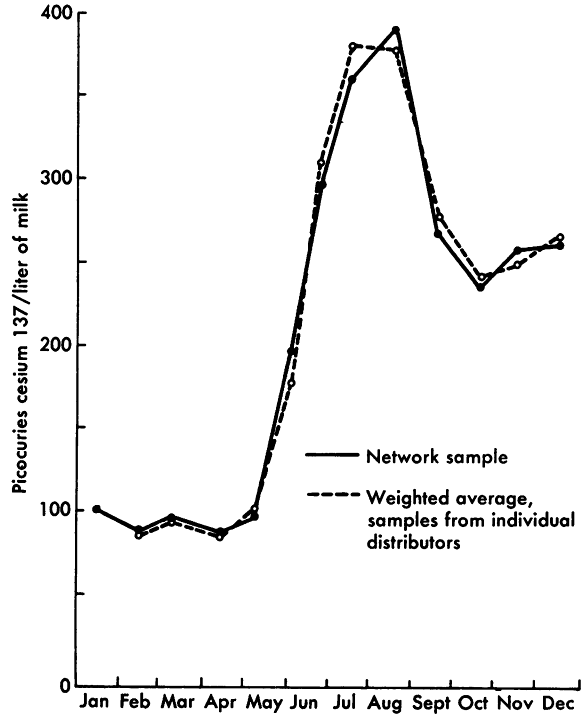
Table 1. Composition of pasteurized milk network sample for Cincinnati

Dairy	Gallons of milk produced weekly	Percent of total produced	Number of pints in sample	Percent of total sample
A	25, 000	19. 41	1. 50	18. 75
B	29, 000	22. 52	1. 75	21. 88
C	21, 000	16. 30	1. 25	15. 62
D	15, 000	11. 65	1. 00	12. 50
E	11, 000	8. 54	. 50	6. 25
F	8, 500	6. 60	. 75	9. 38
G	8, 000	6. 21	. 50	6. 25
H	8, 000	6. 21	. 50	6. 25
I	3, 300	2. 56	. 25	3. 12
Total represented in sample	128, 800	100	8	100

Key to Nomenclature

Symbol	*Formula*	*Meaning*
μ	----------------	true average number of total counts.
σ^2	μ	variance of total counts
σ	$\sqrt{\mu}$	standard deviation of total counts
x_i	----------------	number of total counts in i^{th} observation, $i = 1, 2, \ldots, n$.
n	----------------	number of observations
$\bar{x}$	$\sum_{i=1}^{n} x_i/n$	observed average number of total counts, best estimate of μ
s'_x	$\sqrt{\bar{x}}$	estimate of theoretical standard deviation of the x_i's, best estimate of σ.
s	$\sqrt{\frac{\Sigma(x_i-\bar{x})^2}{(n-1)}}$	observed standard deviation of the x_i's
$s_{\bar{x}}$	$s'_x/\sqrt{n}$	estimate of theoretical standard deviation of $\bar{x}$
y_i	x_i/d	number of counts per minute (cpm) in i^{th} observation.
d	----------------	counting duration
$\bar{y}$	$\bar{x}/d = \sum_{i=1}^{n} y_i/n$	observed average number of cpm
s'_y	$\sqrt{\bar{y}/d}$	estimate of theoretical standard deviation of the y_i's
s_y	$\sqrt{\frac{\Sigma(y_i-)\bar{y}^2}{(n-1)}}$	observed standard deviation of the y_i's
$s_{\bar{y}}$	$s'_y/\sqrt{n}$	estimate of theoretical standard deviation of y

Figure 1. Comparison of cesium 137 content of network milk sample with the weighted average cesium 137 content of milk samples from individual distributors, Boston, Mass.



SOURCE: Unpublished data compiled by the Division of Radiological Health, Public Health Service.

Since all counting instrumentation has a background counting rate, the net count rate due to the sample alone can be obtained only by subtracting this background counting rate from the gross sample-plus-background rate. Consequently, it is necessary to work with the difference of two independently obtained quantities. The error of such a difference, by propagation of error theory, is equal to the square root of the sum of the squares of the error in each quantity.

Thus if

$$y=y_1-y_2$$

then

$$s_y'^2=s_{y_1}'^2+s_{y_2}'^2$$

$$=\frac{y_1}{d_1}+\frac{y_2}{d_2}$$

and

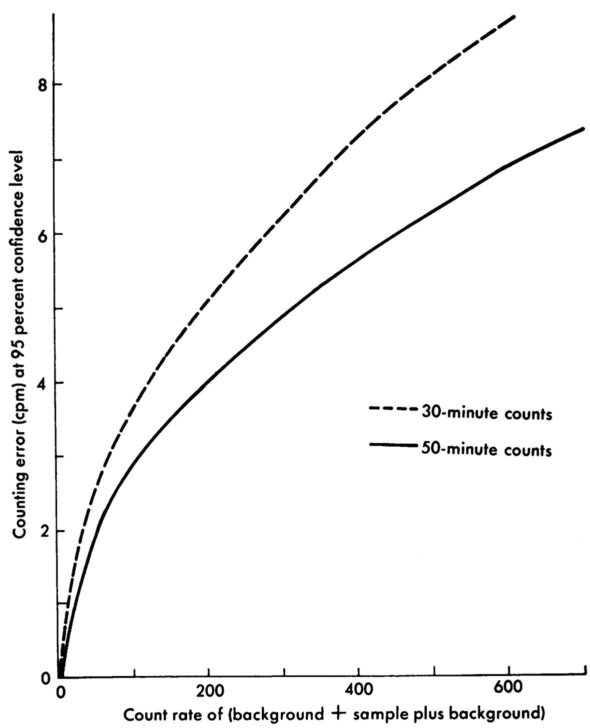
$$s_y'=\left[\frac{y_1}{d_1}+\frac{y_2}{d_2}\right]^{1/2}$$

The standard deviation of the net count rate is used to find the counting error and the lower limits of detectability. Graphs and nomographs for finding counting errors may be found in "Statistical Methods Used in the Measurement of Radioactivity with Some Useful Graphs and Nomographs" by Alan A. Jarret (*3*) and in the "Radiological Health Handbook" (*4*). If samples and backgrounds are routinely counted for a set time, it is more convenient to construct graphs for the counting error for these specific conditions. At the Northeastern Radiological Health Laboratory, these standard counting times are 50 minutes for gamma spectrometry and 30 minutes for low-level beta counting. Figure 2 shows the error for 30- and 50-minute counts of sample and of background.

As an example of the use of these curves, assume the following counting data:

Quantity	*Background*	*Sample plus background*
Counting time	50 minutes	50 minutes.
Count rate	50 cpm	100 cpm.

Figure 2. Counting error (cpm) at the 95 percent confidence level for 30- and 50-minute counts of sample and background



The sum of the count rates of background and sample plus background is 150 cpm; from figure 2 the corresponding counting error at the 95 percent confidence level is 3.45 cpm.

Evaluation of Error

Often one may be interested in knowing if the variation in repeated measurements of a sample may be attributed wholly to the randomness of the disintegration process, or whether some other source of variability, such as instrument malfunction, exists. To determine this, the χ^2 (chi-square) statistic is used.

If x is normally distributed with variance σ^2, and s^2 is the sample variance based on n replicate measurements, then $(n-1)\,s^2/\sigma^2$ has a χ^2 distribution with $(n-1)$ degrees of freedom (*5*). The justification for use of the chi-square test is the fact that the Poisson counting distribution is approximated by the normal distribution for large $\bar{x}$. Thus if,

$$\sigma^2 \cong \frac{\sum x_i}{n},$$

and

$$s^2 = \frac{\sum (x_i - \bar{x})^2}{n-1},$$

then

$$\frac{\chi^2}{n-1} = \frac{(n-1)s^2}{\sigma^2} \cong \frac{(n-1)\sum (x_i-\bar{x})^2/(n-1)}{\sum x_i/n}$$

or

$$\chi^2 \cong \frac{\sum_{i=1}^{n} (x_i-\bar{x})^2}{\bar{x}}.$$

An alternate expression for chi-square may be obtained by substituting

$x_i = y_i\, d,$

$$\chi^2 \cong \frac{\sum (y_i d - \bar{y} d)^2}{\bar{y} d} = \frac{d^2 \sum (y_i - \bar{y})^2}{\bar{y} d}$$

$$\chi^2 \cong \frac{\sum (y_i - \bar{y})^2}{\bar{y}/d}.$$

Application of Control Concepts

As an illustration of the application of these concepts to routine laboratory operations, the procedures for determining the stability of the background of low-level beta counters and gamma spectrometers at the Northeastern Radiological Health Laboratory are presented.

Beta counters. The laboratory's two low-level beta counters are each equipped with 50-position automatic sample changers. Formerly, a separate background determination was made daily for each position being used. To determine whether it would be possible to use a single background for all positions of each counter over a long period of time, a study of background variation was undertaken.

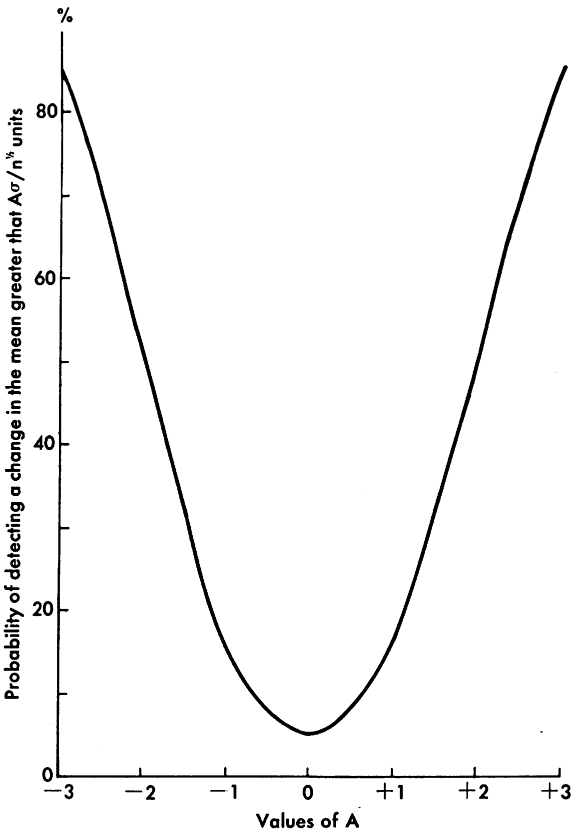
The chi-square test was utilized to test the hypothesis that the variation in background determinations was wholly caused by the randomness of the disintegration process. Data used in this evaluation were the average and variance of approximately 100 30-minute backgrounds taken over a 3- to 4-day period. Chi-square tables are usually available only up to 30 degrees of freedom; therefore, the standard normal deviate t was obtained by use of the expression $t=\sqrt{2\chi^2}-\sqrt{2n'-1}$ where n' is the number of degrees of freedom (*2a*). Since t is normally distributed, the above hypothesis would be accepted at the 95 percent confidence level when $|t| \leq 1.96$. Table 2 summarizes the data which were obtained for the two beta detectors. Because the values of t in each case were <1.96, the background may be attributed entirely to the randomness of the disintegration process, with no significant contribution from instrument malfunction or electrical disturbances.

Once it had been established that the background was consistent within expected random counting deviation, background variations were

Table 2. Background analysis data on low-level beta counters

Quantity	Counter 1	Counter 2
n, number of backgrounds	116	71
$\bar{y}$, average background	.98	.93
y/d, expected variance	.0325	.0310
$\sqrt{\bar{y}/d}$, expected standard deviation	.18	.18
d, time counted (minutes)	30	30
$\sum \frac{(y_i-\bar{y})^2}{(n-1)}$, observed variance	.0394	.0344
$\chi^2 \cong (y_i-\bar{y})^2/(\bar{y}/d)$	140.63	78.80
$t=\sqrt{2\chi^2}-\sqrt{2n'-1}$	1.63	.78

Figure 3. Power function for testing the hypothesis that the observed mean is equal to the expected mean (two-tailed test, σ=.05)



monitored with control charts for daily operation. Both the mean and range of the background determinations were monitored.

The background counting rate of each of 16 sample positions on each instrument was determined nightly. This number of determinations was selected, on the basis of power curves, so as to provide approximately a 60 percent probability of recognizing a change of 0.10 cpm in the mean (*5a*). The power curve (fig. 3) shows the power of the test—that is, the probability of recognizing a change in the mean when the change is greater than $A\ \sigma/\sqrt{n}$—as a function of A. Thus, when this change is 0.10 cpm,

$$0.10=\frac{A(0.18)}{\sqrt{16}}$$

or

$$A=\frac{(0.10)4}{0.18}=2.22$$

The power corresponding to A=2.22 was found from the graph to be approximately 60 percent. The probability of detecting any other change, Δ, can be obtained by calculating the corresponding A and using figure 3 to determine the power. The probabilities are given in the following table.

Δ	A	*Probability (percent)*
0.13	2.89	83
.12	2.67	77
.11	2.45	68
.10	2.22	60

The mean chart (fig. 4), for a sample of size 16, is very simply prepared. The central value $\bar{y}$ is the average background; lines are drawn above and below $\bar{y}$ at distances corresponding to 1, 2, and $3\sigma_{\bar{y}}$ where $\sigma_{\bar{y}}$ is the expected standard deviation (0.18) divided by $\sqrt{16}$. The range chart (fig. 5) is drawn as follows. Coefficients d_2, D_U and D_L are taken from

Figure 4. Control chart for average background of low level beta counter 2

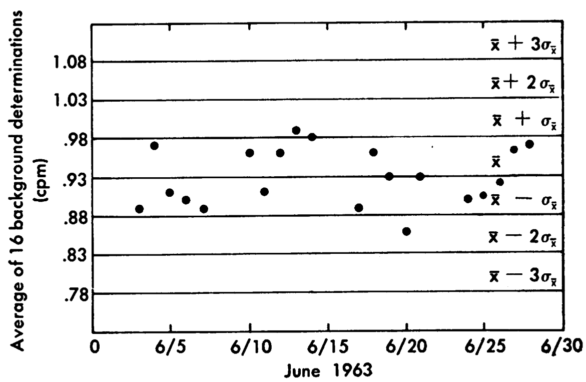
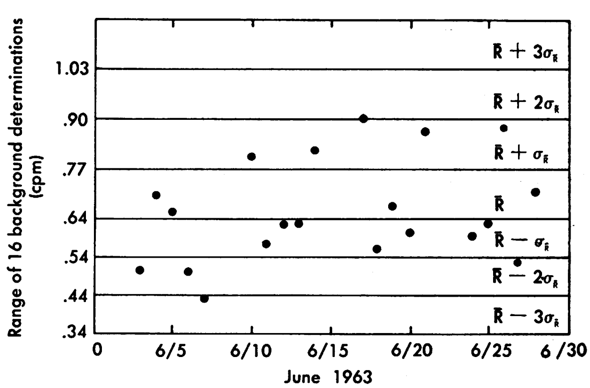


Figure 5. Control chart for range of backgrounds of low level beta counter 2



standard statistical tables prepared for the construction of range control charts. For $n=16$, $d_2=3.54$, $D_U=1.624$, and $D_L=0.529$ (*5b*, *5c*). The average or expected range $\overline{R}$ is calculated by $\overline{R}=d_2\sigma=0.64$. The upper 99 percent confidence level for the range ($\overline{R}+3\sigma_{\overline{R}}$) is calculated by $\overline{R}+3\sigma_{\overline{R}}=D_U\overline{R}=1.04$. The lower 99 percent confidence level for the range ($\overline{R}-3\sigma_{\overline{R}}$) is calculated by $\overline{R}-3\sigma_{\overline{R}}=D_L\overline{R}=0.34$. The σ and 2σ limits for the range are obtained by dividing the difference between the 3σ limits and the average range into three equal parts.

Figures 4 and 5, the control charts for the second low-level beta counter for the month

Figure 6. Control chart for background of Northeast Radiological Health Laboratory 4, taken in iodine 131 region (0.31–0.40 Mev)

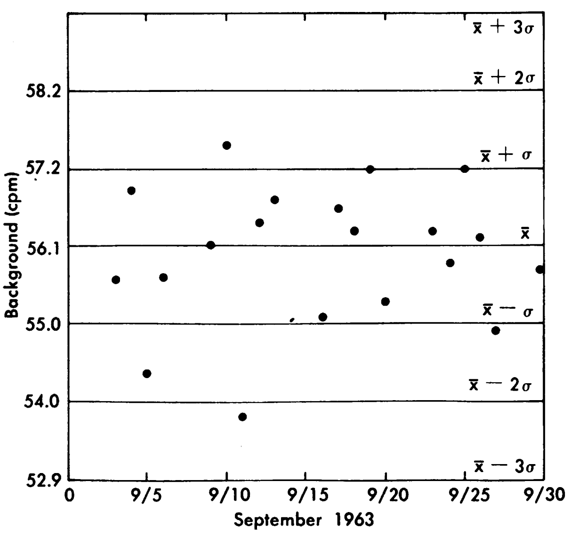
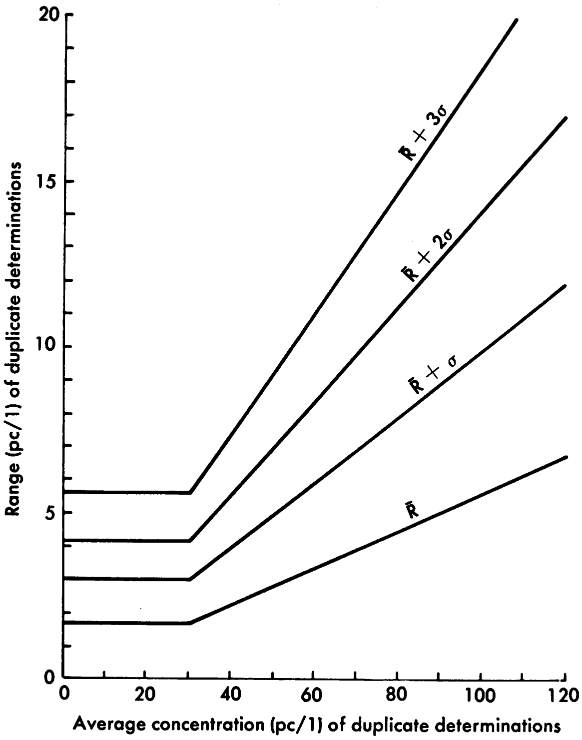


Table 3. Expected standard deviation of nuclides found in milk

Nuclide and concentration	Standard deviation
Iodine 131, barium 140, cesium 137:	
≤100 picocuries per liter	5 picocuries per liter.
>100 picocuries per liter	5 percent.
Potassium: Any value	0. 06 grams per liter.
Calcium: Any value	.02 grams per liter.
Strontium 90:	
≤30 picocuries per liter	1.5 picocuries per liter.
>30 picocuries per liter	5 percent.
Strontium 89:	
≤60 picocuries per liter	3.0 picocuries per liter.
>60 picocuries per liter	5 percent.

Figure 7. Control limits for range of strontium 90 (pc/1) in milk samples analyzed in duplicate



of June 1963, show that the backgrounds remained within statistical control during this period.

Gamma spectrometers. Chi-square tests were performed on the background determinations of four 200-channel gamma spectrometers operated by the laboratory staff. Formerly backgrounds for the sample containers had been run and calculated each day for nuclides in the following energy ranges: iodine 131, 0.31 to 0.40 Mev; barium 140, 0.43 to 0.56 Mev; cesium 137, 0.59 to 0.72 Mev; and potassium 40, 1.37 to 1.50 Mev.

After these tests had established that the backgrounds were within statistical control, it was decided to employ the background in the iodine 131 region (0.31 to 0.40 Mev) as an indicator of continued control since it is the lower energy range that is particularly subject to variation. Because the detectors used in gamma spectrometry do not have multiple sample positions, it was not necessary to establish a control chart for ranges. Figure 6, the control chart for one spectrometer system in the iodine 131 region for the month of September 1963, indicates that this background remained within statistical control.

Intralaboratory quality control. Application of statistical techniques is not limited to instrumentation. These techniques may be used whenever it is possible to estimate the expected precision of a determination. At the laboratory this condition was assumed to be satisfied when the analytical procedures involved in preparing the samples for nuclear counting were fairly well established and when the instruments used for counting were reasonably stable and properly standardized. This is to say that the errors arising from extraneous causes, such as sample preparation and counter instability, had been reduced to a point where under everyday operating conditions they were insignificant in comparison to the expected analytical error. The purpose of the control chart program was to ascertain that the two conditions previously mentioned are maintained.

The number of samples selected for reanalysis should be sufficient to inspire confidence in the reliability of the results but not so great as to impose an excessive sample load. At this laboratory every fifth milk sample is resubmitted for gamma spectrometry and every fourth milk sample for radiochemical analysis.

The expected standard deviations were determined on the basis of experience with the method, including the average values for background, efficiency, chemical yield, and decay. The range is then computed and used as a criterion for accepting or rejecting the reliability of the data.

For duplicate analyses the average range ($\overline{R}$) is estimated by 1.12σ and the upper and lower 99 percent confidence levels by 3.267 $\overline{R}$ and 0.000 $\overline{R}$ respectively. Table 3 gives the

Table 4. Summary, quality control, July 1964

Nuclide	$\pm\sigma$	$\pm 2\sigma$	$\pm 3\sigma$	$>3\sigma$
Iodine 131	18	1	(1)	(1)
Barium 140	15	4	(1)	(1)
Cesium 137	19	(1)	(1)	(1)
Potassium	16	2	1	(1)
Calcium	7	(1)	(1)	(1)
Strontium 90	5	1	1	(1)
Strontium 89	6	1	(1)	(1)

[1] No replicate with this deviation.

Table 5. Quality control, gamma spectroscopy, July 1964

Sample number	Date collected	Date analyzed	Analyzer number	Iodine 131	Barium 140	Cesium 137	Potassium stable
M0664B	June 30	July 2	1	11. 4	−3. 2	120. 0	1. 61
P345		do	4	3. 9	8. 5	115. 3	1. 50
M0673B	July 13	July 15	3	. 3	−4. 9	86. 0	1. 56
P376		July 16	3	−2. 6	5. 1	83. 2	1. 58
M0674B	July 14	July 15	4	−4. 7	6. 3	110. 0	1. 43
P381		do	2	−2. 3	1. 1	118. 2	1. 42

NOTE: All deviations were within acceptable limits.

Table 6. Quality control, chemistry, July 1964

Sample number	Date collected	Date counted	Counter	Calcium	Strontium 90	Strontium 89
M0655A	June 12–19	July 2	2	1. 22	31. 1	−2. 1
P(283–305)		July 8	1	1. 22	33. 9	−3. 8
M0668A	June 26–July 3	July 22	2	1. 09	23. 7	1. 4
P(326–352)		do	2	1. 10	26. 5	−1. 0

NOTE: All deviations were within acceptable limits.

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expected standard deviations of the various nuclides in pasteurized milk samples. Figure 7 illustrates the limits for the range for strontium 90 obtained by using these values. The results of duplicate analyses of samples for the control program are reported monthly (tables 4, 5, and 6).

One final point is the limitations of these statistical tests. They will indicate the precision of the results of n determinations about their mean. However, these tests will not indicate whether the observed mean approximates the true mean. This must be accomplished by other methods.

Conclusion

The degrees of variation in repeated measurements of background and environmental samples on nuclear counting equipment can be minimized by resolving and removing sources of variance peculiar to the particula apparatus. Once under control, optimum operating conditions can be monitored through the use of the appropriate statistical techniques. Application of these techniques to low-level beta counters and gamma spectrometers at the Northeastern Radiological Health Laboratory has confirmed the success of this approach.

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(*3*) Jarret, A. A.: Statistical methods used in the measurement of radioactivity with some useful graphs and nomographs. U.S. Atomic Energy Commission, Division of Technical Information AECU-262, June 17, 1946, pp. 15–25.

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(*5*) Dixon, W. J., and Massey, F. J., Jr.: Introduction to statistical analysis. McGraw-Hill Book Co., Inc., New York, 1957, p. 80; (*a*) p. 249; (*b*) p. 404; (*c*) p. 130f.

Courses in Chemical and Biological Defense

To aid the Public Health Service in meeting some of its responsibilities to provide selected health and medical personnel with general knowledge in the technical aspects of chemical and biological defense, 1-week classes in public health and the medical aspects of chemical and biological defense will be conducted at the U.S. Army Chemical School at Fort McClellan, Ala., during the period April 26–30, 1965.

The participants, whose positions require knowledge in chemical and biological defense, will include representatives of State and local health departments, Veterans Administration, Public Health Service, faculty members of affiliated schools in the Medical Education for National Defense Program, and other interested persons. Security clearance is not required. Those who attend will be housed in governmental quarters at a cost of $1.50 per night. Government eating facilities are available at about $3 per day.

Applications should be made on enrollment forms available from the Deputy Chief, Training Branch, Division of Health Mobilization, Office of the Surgeon General, Public Health Service, U.S. Department of Health, Education, and Welfare, Washington, D.C., 20201.