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Plutonium Body Burden (Q) from Urine Assays

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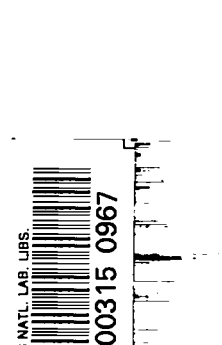

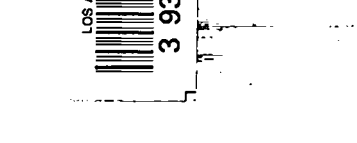

A HISTORY OF PUQFUA
Plutonium Body Burden (Q) from Urine Assays

by

James N. P. Lawrence

ABSTRACT

PUQFUA is a FORTRAN computer program that calculates *plu*tonium body burdens (*Q*) *f*rom *u*rine *a*ssay data. This report describes the historical development of the program at the Los Alamos Scientific Laboratory (LASL) since 1959. After a review of the basic techniques used in the original PUQFUA, its deficiencies are listed. The procedures used to improve the program and correct the deficiencies are described. Appendixes provide a detailed discussion of the evaluation made of the analytical errors in the plutonium urine assay program at LASL from 1944 to 1978.

I. INTRODUCTION

PUQFUA is a FORTRAN computer program that calculates plutonium body burdens from urine assay data. The original program, described in Ref. 1, became operational in 1959 at Los Alamos Scientific Laboratory (LASL). The mathematical basis of the technique is as follows.

PUQFUA is based on an article entitled, "The Application of Excretion Analyses to the Determination of Body Burden of Radioactive Isotopes," by Wright H. Langham.[4] Based on experimental evidence with humans, Langham developed a set of power function elimination equations for the excretion of plutonium over a five-year period. These equations are:

$$Y_u = 0.002t^{-0.74} \quad (1)$$

$$Y_{u+f} = 0.0079t^{-0.84} \quad (2)$$

where Y_u and Y_{u+f} are the *fractions* of the injected dose of plutonium excreted per day in the urine and in the urine plus faeces, respectively, and t is the time in days after injection. Langham emphasizes that the errors in the constants of the above expressions may be of the order of 10 per cent.

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[4]Wright H. Langham, British Journal of Radiology, Supplement 7, Part V, p. 95, 1957.

Since body elimination is by both urinary and faecal excretion, an integration of the expression Y_{u+f} from 1/2* to x + 1/2 days will give total fraction of the acute body burden which has been eliminated in x days (x=t). Subtracting this value from unity will give the fraction of the body burden retained (R_t) at x days after exposure.

The fraction of the original body burden (D_E) eliminated on a given day t, i.e., Y_u, will be equal to the amount of plutonium in the 24 hr urine on that day t divided by the original body burden, or

$$Y_u = U/D_E \tag{3}$$

where U is the amount of plutonium excreted on day t in the same unit as D_E is expressed. Combining the two expressions for Y_u, we obtain

$$D_E = 500 \cdot Ut^{0.74} . \tag{4}$$

Thus, by measuring the 24 hr urinary excretion on any day t, we are able to compute the original body burden from a single acute exposure.

In order to compute the additional body burden after another exposure, we now extend Langham's development. We calculate the urine sample to be expected from the original exposure on day t', where t' is greater than t. We then subtract this calculated daily urinary excretion from the measured value on day t' and compute D_E, using this difference as the value of U.

For successive exposures, the *sum* of the expected 24 hr urinary excretions is subtracted from the measured value and this difference is used in equation (4) as U to give the additional incremental body burden at the time of exposure.

By manipulating the equations previously given, the 24 hr urine specimen at some later time, t', is given by

$$U' = 0.002D_E\, t'^{-0.74} . \tag{5}$$

Thus, by a series of successive calculations of D_E, the expected partial 24 hr urinary excretion corresponding to each urine sample may be calculated.

Once all of the partial D_E's are calculated for all the urine specimens listed, the amount of plutonium retained by the body in each case is given by

$$D_R = D_E R_t = D_E \left(1 - 0.0079 \int_{1/2}^{x+1/2} t^{-0.94}\, dt\right) \tag{6}$$

or

$$D_R = D_E \left[1 - 0.1317(x + 1/2)^{0.06} + 0.1317\ (1/2)^{0.06}\right] \tag{6a}$$

where x is the number of days between the date of calculation and the date each partial body burden was received. Equation 6a accounts for the elimination of plutonium from the date of the exposure to the date on which the computation of body burden is made.

Hence, the total body burden on the date of calculation is given by the sum of all D_R's.[1]

*Arbitrarily chosen as a lower limit of integration since the power function is divergent for small values of t.

Input data for the original PUQFUA was arranged rather inefficiently. A series of data concerned with the calculation dates was followed by all data needed to estimate each individual's

body burden. Separate date entries were required for each urine sample date, each urine sample result, and the number of days before each urine sample date that an accident occurred. Updating or adding new urine data required extensive card filing and repunching of control cards for each individual. Identification information on individuals was quite meager, consisting of name, local identification number, LASL group code, and the last two digits of the person's birth year.

The original PUQFUA had a rather simple sample validation scheme, whereby each successive sample tested the validity of the sample preceding it. This testing was performed from the earliest dated sample to the latest. When a sample was deemed invalid, its date and any indication of a potential accident were deleted from the calculation of body burden.

Peer criticism of PUQFUA included the following.

(1) Integration of the power function from day 0 caused no mathematical problems as long as the exponent of Eq. (6) was not -1.0.
(2) Because PUQFUA started with the oldest samples and proceeded to the newer samples, a sample subsequently invalidated could validate the preceding sample.
(3) Accident dates could be lost during validation because they were entered as the number of elapsed days from the accident to the next urine sample.
(4) No margin of error was allowed in the validation process (that is, no account was taken of the accuracy of the urine assay results).

We recognized other deficiencies, also.

(1) The original urine data were transcribed at least three times. When urine assay data were taken directly from punched cards (from the analyst), for an interim revision of PUQFUA, we found that the original input data sometimes included analyses of feces, sputum, etc., for plutonium, and occasionally ^{241}Am urine assay results.
(2) The analyst's reporting units (counts/min) were not always converted properly to disintegrations per minute (dis/min).
(3) Zero assay results always caused invalidation of the previous samples, even if the zero result was nonrealistic.
(4) Only the fully documented accidents were included in the original data.

An interim program, PUQFUA1, addressed and eliminated some of the above deficiencies. Urine assay data were compiled from scratch using the punch card file of LASL Group H-5, who performed the urine assay, and were screened to prevent inclusion of any nonplutonium or nonurine assay data. Potential accidents were included by date

(1) when a person was in the room where plutonium was spilled or became airborne, whether or not that person had a high nose count;
(2) when the person had a wound while in a plutonium work area, whether or not contamination was detected at the wound site;
(3) when a person had a nose count of 50 dis/min in either nostril; or
(4) when a person was involved in the cleanup of a plutonium spill.

In general, the potential accidents were noted in an accident report or occurrence file, in the special urine sampling file, or in the person's medical record. This method resulted in a large increase in the number of included potential accidents. The validation process was modified to provide for analytical error and to proceed from the latest to the earliest samples (to form a self-consistent set of data). Appropriate corrections were made to consistently convert reported results to disintegrations per minute. Integration limits of Eq. (6) were changed to 0 to X.

Calculations using the interim program, PUQFUA1, resulted in significantly larger body burdens being estimated (than calculations using PUQFUA). This increase was due to the validation of more samples and the inclusion of more potential accidents. In general, an attempt was made to maximize the estimated body burden. When comparisons with autopsy body burdens

were possible, PUQFUA1 tended to be high by a factor of 2 to 8. The apparent overestimate was considered a fault that should be eliminated.

The latest version, PUQFUA2, addresses all the recognized deficiencies and orders the input data more efficiently. The following discussion treats most of the problems encountered and resolved in producing PUQFUA2.

II. URINE ASSAY PROGRAM AT LASL

At LASL, plutonium urine assays began in 1944. Since that date, several different chemical and counting techniques have been used. The urine assay results available on punch cards reflected the procedures used at the time of analysis, with no chemical blank, recovery factor, or counting geometry corrections made until 1957. From 1957 to 1977, chemical blanks, recovery factors, and geometry counting corrections were made and applied before the results were reported in disintegrations per minute per 24-h sample. (Since 1977, results have been reported in picocuries (pCi) per 24-h sample.) Before 1957, these correction factors were determined only sporadically. An analysis was made of the somewhat meager data to obtain an average value of the blank, recovery, and geometry corrections for each assay method. These factors were built into PUQFUA1 and PUQFUA2 to permit expressing the results in picocuries per 24-h sample. Taking into account the error in the reported result (determined from the spike analysis data), the error in the blank value (determined from the chemical blank analysis data) and the error in the recovery factor (determined from the recovery factor data), an expression (dependent upon the actual analysis technique and date) was derived to obtain the standard deviation of each urine assay result. This expression has been programmed into PUQFUA2 for use in the sample validation process. Appendixes A, B, and C give detailed discussions of the evaluation made of the analytical errors in the LASL urine assay program.

III. IDENTIFICATION OF POTENTIAL ACCIDENTS BY TYPE

In the input data to PUQFUA1, potential accident dates were identified only for the primary isotope involved. If the isotope was ^{239}Pu, the date was used for establishing the estimated date of intake only for ^{239}Pu. It was not used for ^{238}Pu intakes. If the isotope was ^{238}Pu, the potential accident date was used for establishing the estimated date of intake for both ^{238}Pu and ^{239}Pu.

To evaluate the proposed program changes, the detailed calculations were examined for 61 persons. Very detailed examinations were made of the data for four persons, with plotting of the urine, potential accident, and other data. The medical records and accident reports for all 61 persons were examined to determine the specific reason for inclusion of the potential accident date. We rarely found that urine results increased significantly during the next few years when nose swipes <1000 dis/min were the cause of including the potential accident date. Usually, the results increased when nose swipes exceeded 1000 dis/min, when wounds were excised, or when a wound was counted and found to be ≥0.2 nCi. In several cases, the urine results increased in the next couple of years following other types of potential accident dates, such as for high room air counts, spills, and acid burns.

PUQFUA was modified to permit identification of the type of potential accident as well as the isotope involved. The following classes of potential accidents and their designators are part of PUQFUA2.

Potential Accident Classes	Class Designators For ^{238}Pu	Class Designators For ^{239}Pu
Unspecified type	8	9
Wound with excision	K	U
Wound count ≥0.2 nCi	L	V
High room air count, if next years' urines show obvious increase	M	W
High nose counts, if next years' urines show obvious increase	N	X
Nose count >1000 dis/min (either side)	ϕ	Y
Other types, if next years' urines show obvious increase	P	Z

In PUQFUA2, the ^{239}Pu designators restrict the application of the potential accident dates to estimating intake dates to ^{239}Pu data, whereas the ^{238}Pu designators apply to data for both isotopes. When the input data are prepared for PUQFUA2, potential accidents are coded by the appropriate designator when possible. That is, the classes [wound with excisions, wound count ≥0.2 nCi, and nose count over 1000 dis/min (either side)] are coded appropriately. All other potential accidents mentioned in occurrence reports would be coded only by isotope, until sufficient urine assays over the next few years permit determination that other designators are more appropriate or that the potential accident date should be eliminated entirely.

IV. IDENTIFICATION OF UNSUITABLE URINE ASSAY RESULTS

From the inception of the plutonium urine assay program until mid 1975, knowing when requested samples actually had been submitted and analyzed was a problem. In part, this problem was the result of having the analyses performed by Group H-5, whereas the requests for analysis originated in the Health Physics Group, H-1. In most cases, the analysis results were reported by Group H-5. But in a few cases, no reports were issued. Reasons for these cases included

- failure to collect a sample,
- no analysis performed because of insufficient urine volume,
- no results because sample was lost through analysis errors,
- no results because of bad pulse height analysis (PHA) spectra,
- no results because of low chemical recovery,
- no results because of suspected contamination, and
- no results because no spike was added to sample and chemical recovery could not be calculated.

After discussions between Groups H-5 and H-1, it was recognized that in many of these cases the possibility of reporting unreliable results existed, and that such a report would call to Group H-1's attention the need for another sample.

A mutually acceptable arrangement was established between the groups. This arrangement called for a report by Group H-5 to be made on *every* plutonium urine sample requested by Group H-1 or anyone else. If the results were completely unavailable (that is, no sample collected), a report would be made indicating the circumstances. If the results were unreliable, the report would indicate the results obtained but be coded as unreliable. The reports by Group H-5 would also indicate whether another sample had been rescheduled by that group.

PUQFUA2 accepts urine-assay-result input with coded designators to indicate valid results for ^{238}Pu or ^{239}Pu, no results, or unreliable results. The program also permits coding of the assay results to indicate the effect of chelating agents. These features became operational in January 1976.

V. USE OF CALCULATED STANDARD DEVIATIONS FOR REPLICATE ANALYSES

As indicated in Sec. II, PUQFUA2 assigns a standard deviation to each urine assay result in the input data. The formulae used to make these assignments are given in Appendixes A, B, and C.

In a few cases, Group H-5 has run replicate analyses on certain samples and reported each result. The general observation is that the standard deviation of the replicate results is different from that calculated by the formulae. All versions of PUQFUA have been designed to accept only a single result on any given date. (This design is appropriate because the results are expressed in activity per 24 h.) To handle replicate results reported on the same date before PUQFUA2, the input data were adjusted by hand or by an auxiliary computer program to assign the results to different consecutive dates, starting with the reported date. PUQFUA's estimates of plutonium intake from this form of data depend quite strongly on the ordering of the samples; that is, whether the highest results are first, last, or in the middle. To reflect the actual situation more accurately and to eliminate the unpredictable variability of body burden estimates, PUQFUA2 now accepts replicate results on the same date, calculates their average and standard deviation, which then are used as a single sample on the date they were submitted for calculating body burden.

To make this concept operational, it was necessary to modify the input data file by restoring the replicate results to the same date, that is, by removing the consecutive dates that had been generated.

VI. SAMPLE VALIDATION TECHNIQUES

A major feature of the original PUQFUA was the sample validation concept. This feature was incorporated because contaminated urine specimens were quite possible, especially before 1957. Some modifications of the original process have taken place over the years.

In PUQFUA1, the interim version of PUQFUA, two successive techniques, primary and secondary, were used.

A. Interim Primary Technique

Starting with the latest sample and working towards earlier samples, successive pairs are examined. The later sample of each pair is used to test the validity of the earlier sample. If the earlier sample is validated, it is used as the later of the next pair of samples to be tested. If the earlier sample is invalidated, that is, set equal to zero for the calculations, the later sample of that pair remains the later of the next pair to be tested. The sample next earlier than the invalidated sample becomes the one to be tested.

Let

U_e = earlier dated urine result,

U_l = later dated urine result,

σ_e = standard deviation of U_e,

σ_l = standard deviation of U_l,

D_e = integer date of U_e,

D_l = integer date of U_l,

E_i = estimated integer date of intake,

U_c = calculated urine result expected from U_e on D_l,

σ_c = calculated standard deviation of U_c, and

$Z_{1-\alpha}$ = standard normal variable for $(1 - \alpha)$ one-sided confidence interval.

Then,

$$U_c = U_e[(D_e - E_i)/(D_l - E_i)]^{0.74} \tag{1}$$

and

$$\sigma = \sigma_e[(D_e - E_i)/(D_l - E_i)]^{0.74} . \tag{2}$$

The statistical test used by PUQFUA1 and PUQFUA2 is found in Ref. 2. In terms of the variables defined above, the hypothesis to be tested is: Does the calculated urine result U_c exceed the measured result U_l? The U_l is permitted to exceed U_c because the larger value of U_l may have resulted from an additional intake between D_e and D_l. Basic to this test is the assumption that in the event of *no* additional intake, the urine level at any later time can be calculated from Langham's urinary elimination equation, provided the date of intake has been established. Equations (1) and (2) are applications of Langham's urinary excretion equation.

In applying the test in PUQFUA1 and PUQFUA2, the significance level of the test (α) is chosen as 0.10. The standard normal variable for the 90% one-sided (because U_ℓ is allowed to exceed U_c) confidence interval is 1.282 ($Z_{1-\alpha}$). If

$$(U_c - U_\ell) > Z_{1-\alpha} \sqrt{(\sigma_c^2/n_c) + (\sigma_\ell^2/n_\ell)},$$

where $n_c = n_\ell = 1$, we conclude that U_c does exceed U_ℓ at the chosen level of significance, *and* sample U_e on date D_e is rejected as invalid (because U_c was calculated from U_e).

Estimated date of intake is

- halfway between dates of consecutive pairs of samples if no potential accidents are recorded between samples,
- 1/2 day before later sample of pair if potential accident is recorded on date of later sample,
- the earliest potential accident date if several occur between the paired samples,
- 1/2 day before initial sample if potential accident is recorded on date of initial sample,
- the earliest potential accident date before the initial sample if any potential accidents occurred before initial sample, or
- 15 days before the initial sample if no potential accidents occurred before initial sample.

Estimated date of intake of the later sample of a pair is reassigned to be the estimated date of intake of the earlier sample, *if* the earlier sample is invalidated and a potential accident date occurred on the same date as the earlier sample.

B. Interim Secondary Techniques

These techniques are applied, *only* when the primary test would invalidate the earlier sample *and if* the later (validating) sample is ≤0.1 pCi.

The first secondary test uses the four earlier and untested samples preceding the sample being tested. (At least two samples are required.) The *average* predicted urine value and standard deviation on date of the sample being tested are calculated. If the actual value of the sample being tested minus the average predicted urine value exceeds $Z_{1-\alpha}$ times the square root of the sum of the square of the standard deviation of the average predicted standard deviation divided by number of the samples averaged, the final test is performed. Otherwise, the sample is validated.

The final secondary test requires for validation that the three samples preceding the sample being tested all be retained by the primary technique *and* that the sample being tested be ≤1.5 times the raw average (no calculated reduction) of the two samples preceding it.

These secondary tests were an attempt to prevent intuitively unreasonable invalidations. In practice they cause retention of only a few samples. Because they used untested data (earlier samples), the secondary techniques were abandoned completely in PUQFUA2.

C. Inadequacies of Interim Techniques

Detailed examination of the body burden calculations for some 60 persons with significant body burdens exposed apparent faults in the validation techniques of PUQFUA1. One such observation was that a sample (result N) could have been validated by the sample (result N + 1) immediately following it in time, but this following sample would have been invalidated by several consecutive samples (results N + 2, N + 3, N + 4) further on in time. We also frequently observed that a sample, which seemed consistent with the trend of samples before and following it, was invalidated by a negative, zero, or near-zero sample result that immediately followed it.

We also found occasions when a result was two or more times larger that the approximately constant level (analysis result) of the 4 to 10 samples of both earlier and later dates.

When a negative, zero, or near-zero result occurred in the midst of a series of significantly positive results, it seems most probable that the near-zero result was caused by faulty analysis techniques (that is, the activity was lost in the chemical procedures) or the sample analyzed belonged to some person other than it was identified as belonging to. In either case, the negative, zero, or near-zero sample should be eliminated from the PUQFUA calculation to prevent its causing invalidation of the sample immediately preceding it.

A sample's being two or more times larger than the several samples on either side (in time) of it is also suspect. Only if documented records suggest an inhalation or wound (injection) accident involving *soluble* plutonium should we expect to see Langham's predicted elimination pattern. Usually, in inhalation or wound accidents, the samples indicate a slowly rising activity level followed by an even more slowly decreasing activity level. Thus, for those cases where records do not support an inhalation or wound involving soluble plutonium, the samples should be eliminated from the PUQFUA calculations to prevent the addition of a false incremental increase to the body buuden.

During the cupferron era, that is, until October 1, 1949, it was particularly difficult to make objective decisions as to which samples were truly representative of plutonium in an individual. As discussed in Appendix A, analysis blanks and chemical recovery tests were performed infrequently during the cupferron era. Out of 191 blanks run by the cupferron method, ~12% indicated >1.4 counts/min. If this 12% of the blank analysis data were treated as personnel urine data (that is, if an average blank were subtracted from the gross count, and the result were divided by the average chemical recovery factor), these 12% would appear as samples of 0.8 pCi or larger. Assuming personnel urine assays to be statistically in the same class as blank analyses, we might expect that ~12% of all personnel urine assays run by the cupferron method would indicate 0.8 pCi or more, even if there were no plutonium actually present.

For an individual urine assay result, it is obviously not possible to determine whether the sample falls in the high 12%. But it is reasonable to examine carefully all samples in the cupferron period that exceeded 0.8 pCi. Since about January 1946, the number of urine assays performed allow a rejection decision procedure that takes into account the trend of the assay results. In 1944 and 1945, few urine assays were performed because the process was being developed. For the years 1944 and 1945, we plan that all urine assays results >0.8 pCi be tested by the following techniques unless records can be located that show an exposure to soluble plutonium took place. If records strongly suggest exposure to *soluble* plutonium* by inhalation or wounds, the samples should be considered for retention, even if normal testing would reject them. High nose counts with no mention of soluble plutonium should *not* be considered evidence for retention of a urine assay result >0.8 pCi.

Since 1946 (cupferron era), direct evidence of an exposure to soluble plutonium should be sufficient evidence to retain a sample result >0.8 pCi, even if the sample is rejected by tests A, B, and C.

Tests A and B, described later, are used to reject outlying high samples. In addition to the need for eliminating the improbably high results, there is also a need to eliminate certain negative, zero, or near-zero results to prevent them from causing invalidation of the immediately preceding results. We suspect that many negative sample results were the result of the loss of plutonium from the sample being analyzed during the analysis procedure and the counting of an essentially unused counting plate. In part, this conjecture is based on the observation that blank analyses during the cupferron era averaged ~0.6 pCi with only 16 of 191 results <0.1 pCi. Test C, described below, is used to test the improbably low samples.

*In the normal chemical sense.

During the $BiPO_4$ era (that is, from October 1, 1949, to January 28, 1957), similar, but less extreme, problems with high and low samples are encountered. As discussed in Appendix A, analysis blanks and chemical recovery tests were performed infrequently during the $BiPO_4$ era. Out of 188 blanks run by the $BiPO_4$ precipitate (ppt.) method, about 6% indicated >0.4 counts/min. Treating this 6% of the blank analyses as personnel urine data would result in the appearance of samples of 0.4 pCi or larger.

Reasoning similar to that presented for the cupferron method suggests that all $BiPO_4$ results >0.4 pCi should be considered suspect and subjected to tests A and B. If direct evidence in the records indicates an exposure to soluble plutonium, the sample probably should be retained. (Exceptions could occur if all later samples were near zero.) Similarly, test C should be used on certain negative, zero, or near-zero samples.

For the NTA-ZnS (nuclear track film type A and ZnS scintillator technique) and PHA analysis eras (see Appendixes B and C), data do not exist in a form to support the development of a control limit on improbably high results. The urine assay results during the NTA-ZnS and PHA eras seem more consistent (with smaller error bars). Because the chemistry was the same as in the $BiPO_4$ era, the check level should not be any higher than in the $BiPO_4$ era. To keep the additional checking to a reasonable amount, the level adopted for the $BiPO_4$ era has been established for the NTA-ZnS and PHA eras; that is, all samples >0.4 pCi will be tested successively by tests A and B. Because the error bars on the results are so small for near-zero results in the NTA-ZnS and PHA eras, all positive results <0.075 pCi, all zero, and all negative results should be tested by test C, keeping all results used in the test within the NTA-ZnS or PHA eras.

D. Finalized Validation Techniques

In the application of tests A, B, and C, *all compared results are to be in the same analysis era.* For example, results in the $BiPO_4$ era are not to be used to test results in the cupferron era. *Tests A and B* are used for the suspected high results. They *are applied sequentially* (test A first, then test B if required) *to the largest results first, then the second largest, etc., until all results exceeding the upper test level have been tested. Test C is then applied to the results less than LEAST (defined below), chronologically.*

All three tests compare sample results in excess of certain designated values. Samples are considered positive if they exceed the value of LEAST. In the cupferron era, LEAST is 0.10 pCi; in all other eras, LEAST is 0.075 pCi. Samples are considered high if they exceed 0.8 pCi in the cupferron era and if they exceed 0.4 pCi in the other eras. In test C, comparisons are also made against 0.0 pCi in some cases.

1. **Test A.** This test is used for high sample results. Its purpose is to treat those cases where the trend level of the results is somewhat elevated. Passage of test A is sufficient for retention of the sample; that is, if a sample result passes test A, test B should not be applied.

If there are four or more positive samples (that is, greater than LEAST) within ±1 yr of the sample being tested and in the same analysis era, continue with test A; otherwise, start test B.

Select the four sample results greater than LEAST (within ±1 yr either earlier or later) that were closest in date of collection to the sample being tested. Find (1) the average of these four sample results and (2) the average of the standard deviations of these four sample results. If the sample result being tested is less than the four-sample average plus one-fourth of the four-sample standard deviation average, retain the sample being tested; otherwise administer test B.

In the computer program of test A during the evaluation period, the standard deviation of the four-sample *average* was calculated by the standard method for the analysis era, and a statement was printed if the sample would pass the test using the calculated standard deviation of the four-sample average. The actual pass or fail criterion now used in the computer code is as stated in the preceding paragraph. The alternative calculation was not incorporated in PUQFUA2 because it did not result in a significant number of changed validation results of the test.

The factor of one-fourth of the four-sample standard deviation average was determined empirically as a reasonable criterion during the detailed hand-calculation evaluation of the technique. (The hand calculations were made on the data of four high body burden cases.) The stated procedure caused invalidation of samples that intuitively appear out of line.

2. **Test B.** This test is used for high sample results.

For the sample result being tested, determine whether one or more retained potential accident dates occurred between the sample being tested and the next earlier positive sample. If the sample being tested was the first sample result, all retained potential accident dates preceding that sample must be considered. If more than one such date is found between the paired samples, the calculations specified below must be performed for each retained potential accident date. If no retained potential accident date was found, for an initial sample or an initial positive sample, use a date 15 days before the initial positive sample as the assumed exposure date, and for cases where there was an earlier *positive* sample, use a date midway between the two samples as the assumed exposure date. Using the dates found by the above procedures, the date and sample being tested, and the dates of the next three *positive* samples, calculate the excretion levels predicted by Langham's equation on the dates of the next three positive samples. Calculate the fractional reduction in the urine level on each of those dates, and apply this reduction factor to the standard deviation of the sample being tested to obtain a pseudo standard deviation for all the calculated levels. If the actual-measured urine excretion level of two out of three of these later samples is greater than the calculated urine excretion level minus one-third of the pseudo standard deviation on the appropriate dates, retain the sample being tested. Otherwise, reject the sample as a judgment invalidation. If two or more retained potential accident dates result in conflicting decisions, the general practice should be to retain the sample. If test B was required and there were not three positive samples within the same analysis era after the sample being tested, the sample is validated by default.

Note: The equation to be used for the above calculation is

$$\text{Reduction Factor RF} = [I/(E + I)]^{0.74},$$

where I is the number of days between the assumed exposure (or the retained potential accident date) and the sample being tested, and E is the number of days between the sample being tested and the date of the later sample.

$$\text{Calculated urine level} = CL = RF \times UR, \text{ and}$$

$$\text{Pseudo standard deviation} = PS = RF \times U2S,$$

where UR is the result being tested and U2S is the standard deviation of the result being tested.

The computer program to accomplish test B validates those samples for which the retain test is passed in 50% or more of the individual cases tested (when there is more than one potential accident date before the sample being tested).

The factor of one-third of the pseudo standard deviation used in test B was determined empirically by hand calculations. It also caused invalidation of intuitively out-of-line samples.

3. Test C. This test is used for all samples <0.1 pCi in the cupferron era and for all samples <0.075 pCi in other eras.

Test C eliminates those low or negative urine results that, if left valid, would cause subsequent invalidation of high results that precede them (see comment below). It is not intended that all low or negative sample results be invalidated, particularly if no high results have been recorded. *All low and negative samples are retained until the first sample exceeding LEAST is encountered, then* the following tests are applied.

In parts of test C, both the value of the sample being tested (VS) and the value of the sample plus one standard deviation (VSPIS) are used for testing. Part of the testing is related to the analysis eras because the accuracy varies strongly with the type of analysis.

In the cupferron era, even if there is at least one sample >0.1 pCi in the set and VSPIS is <0, the sample is rejected as too small. Also in the cupferron era, even if there are three or more samples >0.1 pCi in the set and VSPIS is <0.1, the sample is rejected as too small. In the $BiPO_4$ era, samples are rejected as too small, even if (1) there is at least one sample >0.075 pCi and VSPIS is <0, or (2) even if there are three or more samples >0.075 pCi and VSPIS is <0.075 pCi. In the NTA-ZnS or PHA eras, samples are rejected as too small, even if (1) there are at least two samples >0.075 pCi and VS is ≤0, or (2) even if there are at least *five* samples >0.075 pCi and VSPIS is ≤0.04 pCi. The number of samples mentioned above was determined empirically in the detailed hand-calculation evaluation of the procedure on the four test cases.

Samples surviving the above tests are examined in relation to the earliest date of a validated sample >0.8 pCi in the cupferron era or >0.4 pCi in other analysis eras. The date of the earliest sample exceeding these high criteria is determined. That date, regardless of the era when it occurred, is used to test the surviving samples. All surviving samples dated before the first high samples are kept with the added notation of no highs.

Samples occurring after the first high sample are tested further. The average of the four retained samples greater than LEAST (0.1 pCi in cupferron era; 0.075 in other eras) and closest in time to the sample being tested is calculated. No time limit within the analysis era of the sample being tested is imposed on the dates of the retained samples greater than LEAST. If VSPIS exceeds this average, the sample being tested is kept; if not, the sample is rejected. The sample may be rejected as too small if VS is <0 and if VSPIS is less than LEAST. If there are not four retained samples greater than LEAST, the sample being tested is kept by default.

One of the print files generated by PUQFUA2 lists the details of these validation tests so that interested and knowledgeable persons can see the calculational details that resulted in sample validation or invalidation.

Comment: During the PUQFUA2 evaluation period, body burden calculations were performed on data from 60 persons having significant body burdens, using different combinations of the above validation techniques. After examining the calculations in detail, we decided that, in PUQFUA2, tests A, B, and C would be applied to all data for each individual, and the surviving samples then would be subjected to the PUQFUA1 primary validation technique.

We believe that the validation techniques in PUQFUA2 are superior to those in either of the earlier PUQFUA versions. However, we recognize that unique situations can and do occur

- when an individual urine result should be retained despite its being invalidated by the test procedures and
- when an individual urine result should be invalidated despite its being validated by the test procedures.

Provisions for both cases have been provided in PUQFUA2 by special coding of the sample data.

VII. REDUCTION OF BODY BURDEN OVERESTIMATES

One criticism of the earlier versions of PUQFUA has been that the PUQFUA body burden estimates usually exceed autopsy estimates by significant amounts, particularly for the lesser body burdens. This problem is due, in part, to a combination of two factors,

(1) the Langham elimination equation, which was basically derived to fit the elimination pattern for ~140 days and was slightly modified, on the basis of data on three industrial exposures, to ~1700 days; and
(2) the feature of PUQFUA that assigns intake dates halfway between consecutive validated urine samples.

When the time between samples is 6 months or less, the incremental body burdens calculated with Langham's equations are quite reasonable. However, when 10 to 20 yr elapsed between samples, significant incremental body burdens are calculated from results that are at about the lower limit of reliable detection (that is, twice the standard deviation of a blank sample).

It was recognized that this upward ratchetting of body burden estimates could be reduced if urine sample results were obtained more frequently. A computer test program was written to generate fictitious urine results at specified intervals between paired, validated real samples. A justification for using such fictitious results is that the absence of urine samples in the intervening time period is a strong indication that no significant plutonium exposures took place (that is, urine samples would have been collected after known significant plutonium exposures).

Two forms of the generation program were tested originally. When the second (later dated) sample was larger than the first (earlier dated) sample, both methods performed the same. Intermediate urine values were generated on a linear ramp from the value of the earlier sample to the later one. The programs differed when the second sample of each pair was smaller than the first. The program LINEAR GENUR generated the fictitious results on a linear ramp downward from the earlier to the later sample. The program LANGHAM GENUR used the two adjacent results to estimate a date of intake (before the earlier sample) that would place both actual samples on the Langham elimination curve ($D_E = 500\ U\ t^{0.74}$). Using this estimated date of intake, Langham's equation, and the analysis results of the earlier sample, the expected results were calculated on the dates for the specified interval. These fictitious results and dates were used with the actual urine results to estimate the body burden.

Note: In LANGHAM GENUR, two exceptions were made to the procedure given above for establishing the estimated date of intake. If the estimated date of intake (by the above procedures) was earlier than the dates of the following situations, the dates corresponding to the following situations were used as the estimated date of uptake for the generation of fictitious urine results.

(1) If the earlier sample of the pair was the first sample submitted for plutonium analyses, the estimated date of intake for purposes of calculating the fictitious urine results was set equal to the date 15 days before the first sample.
(2) If the calculated date of uptake was at an earlier date than the validated sample preceding the earlier sample of the pair, the date of uptake was set equal to the day after the date of the sample preceding the earlier sample of the pair.

Note: No fictitious results were generated between any potential accident date and the date of the first validated sample following the potential accident date.

The effect on body burden estimates of these programs was evaluated by calculation on ~20 real sets of urine data, using several different, specified intervals between the fictitious results. In general, lower body burden estimates resulted from the shortest interval selected. To be consistent with the long-established PUQFUA procedure of assuming uptake 15 days before the initial sample, we decided that a 30-day interval between fictitious samples was satisfactory. The 30-day interval could be handled adequately by the computers available at LASL. In the test

calculations, LANGHAM GENUR resulted in somewhat smaller body burden estimates than LINEAR GENUR or no data generation. Because the purpose of the test was to devise a method for reducing the estimates, the LANGHAM GENUR was selected for incorporation in PUQFUA2. One further change was made in LANGHAM GENUR to handle the data generation when the later sample of the pair was larger than the earlier sample. Instead of using the linear rise from the earlier to the later sample, an exposure date one day after the next earlier sample (sample preceding the pair being treated) was assumed, and generated samples were calculated at the specified interval, to result in the observed excretion rate on the date of the earlier of the pair of samples being treated. This modification also resulted in a slight lowering of calculated body burden when LANGHAM GENUR was applied to the 20 test cases. PUQFUA2 incorporates this feature.

VIII. CALCULATION OF nCi-YEARS AS WELL AS nCi BODY BURDEN

As an aid to the "Health Study of Workers in the Nuclear Industry," we decided to include nCi-years. The nCi-years were determined for each current incremental body burden, as the product of the current incremental body burden times the number of years from the estimated date of the incremental uptake to the date of the calculation. For a deceased person, the date of calculation is the date of death. For living persons, the date of calculation is usually the first day of the month after the latest sample result, but the date is entered as input and can be adjusted at will. The current incremental nCi-years are summed to provide the total nCi-years.

IX. NEW PRESENTATION OF DATA BY MICROFICHE

In the older PUQFUA versions, some details of the calculation were printed. The typical printout was about 22 in. of 11- by 16-in. computer print paper for the complete data on all past and present LASL employees. The information in this printout included the person's identification information, column headings, a line for each ^{239}Pu analysis, which gave normal and integer dates of the samples, normal and integer dates of estimated uptake, incremental uptake estimates and current incremental uptake estimates, and a statement of the total incremental body burden. Data for ^{236}Pu followed in the same format. When the validation scheme rejected a sample, the reason for rejection was not given.

While evaluating the proposed changes in the computational techniques, we realized that more detailed printouts were needed for occasional closer scrutiny. For example, it was desirable to see the details of the validation calculations on individual samples, in part as proof that the program was performing appropriately.

To save printing time, paper, and storage space for the printed output, we planned to record the details of the PUQFUA2 calculations on microfiche. The 22 in. of 11- by 16-in. paper would be reduced to about 25 microfiche cards. Because we planned to include more detailed information in PUQFUA2, more than 25 microfiche cards would be produced at the comparable savings of space.

The detailed output of PUQFUA2 first tabulates all data for ^{239}Pu, then tabulates data for ^{236}Pu if any analyses for ^{238}Pu have been made.

The output consists of

(1) The person's name and identification information: that is, the person's Z number, last name plus initials, social security number, employment group code, birth date (MM/DD/YY), sex, first name, place of birth (town, state, country), notation of alive or dead, and date of employment termination or death.

(2) Details of the validation calculations for each urine sample requiring validation by the new techniques, followed by details on invalidation by the old PUQFUA primary validation technique. Also noted are those samples designated valid by prior judgment.
(3) Detailed listing of urine sample dates, results (pCi), and validation indication; potential accident dates and designators; generated data with dates; estimated dates of exposure; incremental increases in body burden and nCi-years; and summation of body burden (nCi) and nCi-years, under the headings of the variable names used in the program.
(4) Repeat of the person's name and identification information, a statement of the calculation date (input date unless person is deceased, then date of death) and the isotope identification, followed by these column headings: line number, calender date, integer date, urine results (pCi), isotope or validation indication, potential accident designator, incremental initial body burden (nCi), incremental current body burden (nCi), accumulative current body burden (nCi), incremental nCi-years, accumulative nCi-years, and line number, followed by a line-by-line listing of the actual urine sample and potential accident dates, with the appropriate body burden calculation results.

REFERENCES

1. James N. P. Lawrence, "PUQFUA: An IBM-704 FORTRAN Code for Determining Plutonium Body Burden From Urine Assays," Los Alamos Scientific Laboratory report LA-2329 (April 1960).

2. Mary Gibbons Natrella, "Variability in Performance of Each of A and B Is Known From Previous Experience and the Standard Deviations Are σ_A and σ_B, Respectively," in *Experimental Statistics*, National Bureau of Standards Handbook 91 (US Government Printing Office, Washington) 1963, pp. 3-37, -38.

APPENDIX A

PLUTONIUM URINE ASSAYS FROM 1944 TO JANUARY 1957

Blanks, recoveries, conversion to picocuries, and standard deviation of picocurie results

From 1944 to October 1949, the plutonium urine assays were run by the cupferron method. From October 1949 to January 28, 1957, the plutonium urine assays were run by the $BiPO_4$ precipitate (ppt.) method.

All analysis blanks (urine or water, run through the complete analysis procedure) and recovery data (direct-plate counts of the spikes and the spiked urine analyses) were recorded from the LA Notebooks* used to record the urine assay data. These records were separated into cupferron and $BiPO_4$-ppt. sets for analysis.

*LASL internal documents.

Unfortunately, the blank and recovery procedures were performed on a nonuniform time basis, as shown by Table A-I. The dates given show the time span of all blank and recovery samples run that year. During the date interval, samples were run sporadically. Figure A-1 shows the frequency distribution of the cupferron blank results. The number of blanks >1.8 counts/min was two in 1944, nine in 1945, none in 1946, five in 1947, three in 1948, and none in 1949. By observation, we determined that the distribution was not normal, but skewed toward the higher values. The problem of which values to retain in calculating an average blank for the cupferron procedure was not straightforward. We felt that at least the 14 counts/min blank should be removed from the set to be averaged, but we did not know how many others to remove.

Chauvenet's criterion was not used because it should not be applied successively and because it should be applied to normal distributions.

It was decided to look at the criterion of eliminating all samples in excess of $\bar{x} \pm n\sigma$, where n was 3 or 4 (that is, eliminate all samples in excess of three or four standard deviations from the mean). New means and standard deviations were calculated after each large value was deleted. For n = 3 (that is, three standard deviations), all blanks >1.7 counts/min would be eliminated. For n = 4, all blanks >2.9 counts/min would be eliminated. Because of the skewed distribution, we decided to use the four-standard-deviation criterion for eliminating the outlier results.

The mean for all blanks <2.9 counts/min was 0.690 counts/min with a standard deviation of 0.533 counts/min. When no blanks were run with a cupferron recovery measurement, in the

TABLE A-I

BLANK AND RECOVERY DETERMINATIONS

Cupferron Blanks		Cupferron Recoveries	
Date	Number Samples	Date	Number Samples
11/09 - 11/27/44	3	10/24 - 12/07/44	17
02/16 - 11/17/45	67	02/27 - 11/28/45	35
02/11 - 09/04/46	37	04/05 - 09/16/46	13
01/28 - 09/10/47	67	01/28 - 12/10/47	88
03/08 - 07/14/48	10	03/09 - 12/16/48	15
03/10 - 08/22/49	7	03/10 - 08/22/49	8

$BiPO_4$ Ppt. Blanks		$BiPO_4$ Ppt. Recoveries	
Date	Number Samples	Date	Number Samples
10/17/49 - 06/26/50	26	10/17/49 - 06/26/50	46
1951	0	1951	0
1952	0	06/24 - 06/26/52	12
09/25 - 11/02/53	2	08/14 - 12/03/53	27
01/28 - 12/03/54	74	01/28 - 06/17/54	94
02/55 - 05/20/55	86	02/55 - 01/02/56	47

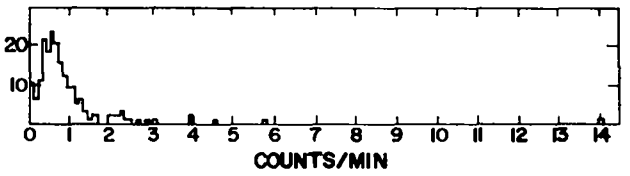

Fig. A-1.
Frequency distribution of cupferron blanks.

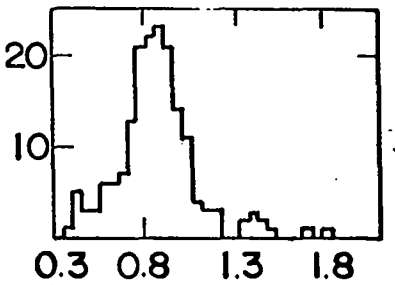

Fig. A-3.
Fractional cupferron recovery.

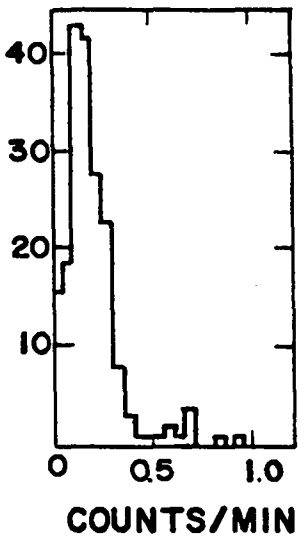

Fig. A-2.
Frequency distribution of $BiPO_4$ ppt. blanks.

calculation of fractional recovery, a blank of 0.690 counts/min was subtracted. (When blanks were run with a cupferron recovery measurement, the average of those blanks was used, regardless of whether the set included blanks >2.9 counts/min.)

Figure A-2 shows the frequency distribution of the $BiPO_4$ ppt. blanks. The number of blanks >0.55 counts/min was two in 1950, one in 1953, and four in 1954. Again by observation, we determined that the distribution was not normal but skewed toward the higher values. Again because of the skewed distribution, we decided to use the four-standard-deviation criterion for eliminating the outlier results. This criterion eliminated all $BiPO_4$ ppt. blanks >0.55 counts/min.

The mean for the remaining blanks was 0.155 counts/min with a standard deviation of 0.099 counts/min. When no blanks were run on a $BiPO_4$-ppt.-recovery measurement in the calculation of fractional recovery, a blank of 0.155 counts/min was subtracted. (When blanks were run with the $BiPO_4$ recovery measurement, the average of those blanks was used, regardless of whether they included blanks >0.55 counts/min.)

For both types of analyses to calculate the fractional recovery, if several direct-plate spikes accompanied the recovery measurement, the average value of the direct plates was used to calculate the fractional recovery.

Figure A-3 shows a frequency distribution of the fractional cupferron recoveries. This distribution appears normal, except for the two high values. Because of this more normal appearance, the criterion for eliminating samples was chosen to be those in excess of $\bar{x} \pm 3\sigma$. Successively, the 1.731, 1.666, and 1.456 samples were eliminated. The resultant mean was 0.823 with a standard deviation of 0.194 for the fractional recovery for the cupferron method.

Figure A-4 shows a frequency distribution of the fractional $BiPO_4$-ppt. recoveries. Again, the distribution appears normal, except for the three *negative* recovery values. Again, the $\bar{x} \pm 3\sigma$ criterion for eliminating samples was chosen. Successively, −1.125, −0.148, and −0.011 were eliminated. The resultant mean was 0.674 with a standard deviation of 0.209 for the fractional recovery by the $BiPO_4$-ppt. method.

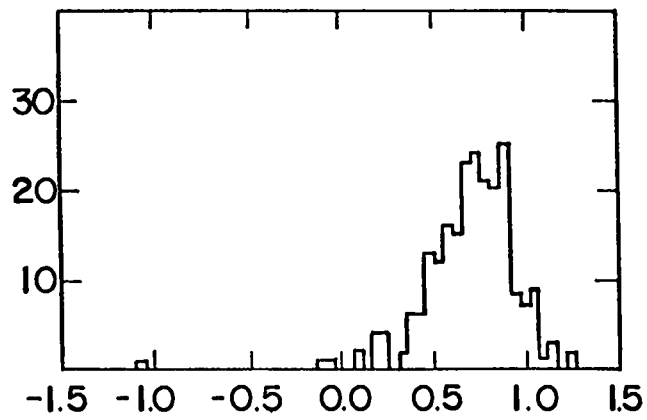

Fig. A-4.
Fractional $BiPO_4$-ppt. recovery.

It was possible to determine the standard deviation of individual recoveries for only one set of $BiPO_4$ data. In that case, several samples were run, and several counts were made on each recovery analysis. Thus, the standard deviation of each component of the recovery factor was available, and the standard technique of propagation of error for mathematical relations could be applied. For each set of data, the two extreme values had a minimum recovery factor of 0.444 with a standard deviation of 0.174 and a maximum recovery factor of 0.874 with a standard deviation of 0.252. These values bracket the $BiPO_4$-ppt. average of 0.674 with standard deviation of 0.209.

To obtain an indication of the relation between the mean of a set of assumed-identical samples and the relative percentage standard deviation (coefficient of variance), the blank data and sets of recovery data (which were identically spiked) for the cupferron method were tabulated and plotted on log-log paper (Fig. A-5). Although the spread of points was large, there appeared to be a trend. The data were least squares fitted to a power function equation with the result

$$100\ \sigma/\bar{x} = 30.10\ \bar{x}^{-0.4081}\ \text{(cupferron)}\ .$$

The same procedure was followed for the $BiPO_4$-ppt. method (Fig. A-6). Again, the spread of points was large, and there appeared to be a trend. These data were also least squares fitted to a power function with the result

$$100\ \sigma/\bar{x} = 23.84\ \bar{x}^{-0.3803}\ (BiPO_4\ \text{ppt.})\ .$$

The major effect observed in this exercise is that the standard deviation varies approximately as the square root of the mean. If all counting times had been the same for all data, this effect would have been expected. The multiplicative coefficients (30.10 and 23.84) apparently account for the other factors influencing the standard deviation.

As a result of this study, the following equation was used for the cupferron-method data to convert the personnel urine assay data from the reported unit (counts/min) to picocuries per 24-h sample.

$$P(\text{pCi/24-h sample}) = (2.0/2.2)\{[(C \pm \sigma_C) - (B \pm \sigma_B)]/(R \pm \sigma_R)\}\ \text{(cupferron)}\ ,$$

where

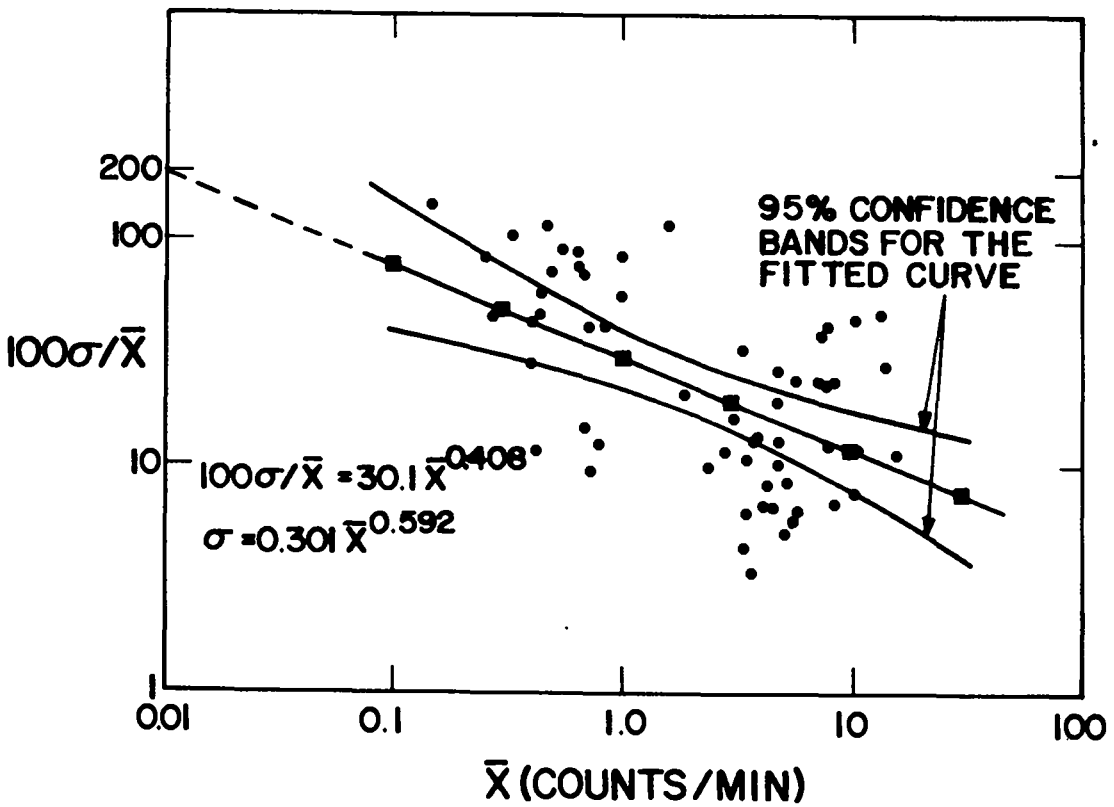

Fig. A-5.
Mean vs relative percentage standard deviation of blank and recovery data for cupferron method.

$B \pm \sigma_B = (0.690 \pm 0.533)$ counts/min ,

$R \pm \sigma_R = 0.823 \pm 0.194$,

and

$C \pm \sigma_c = (C \pm 0.3010C^{0.5919})$ counts/min-24-h sample

because

$100\ \sigma_c/C = 30.10\ C^{-0.4081}$.

Individually, the standard deviation (σ_p) of calculated picocuries per 24-h sample could be computed for each reported counts per minute per 24-h sample (C), by the standard technique of propagation of error for the equation. That is,

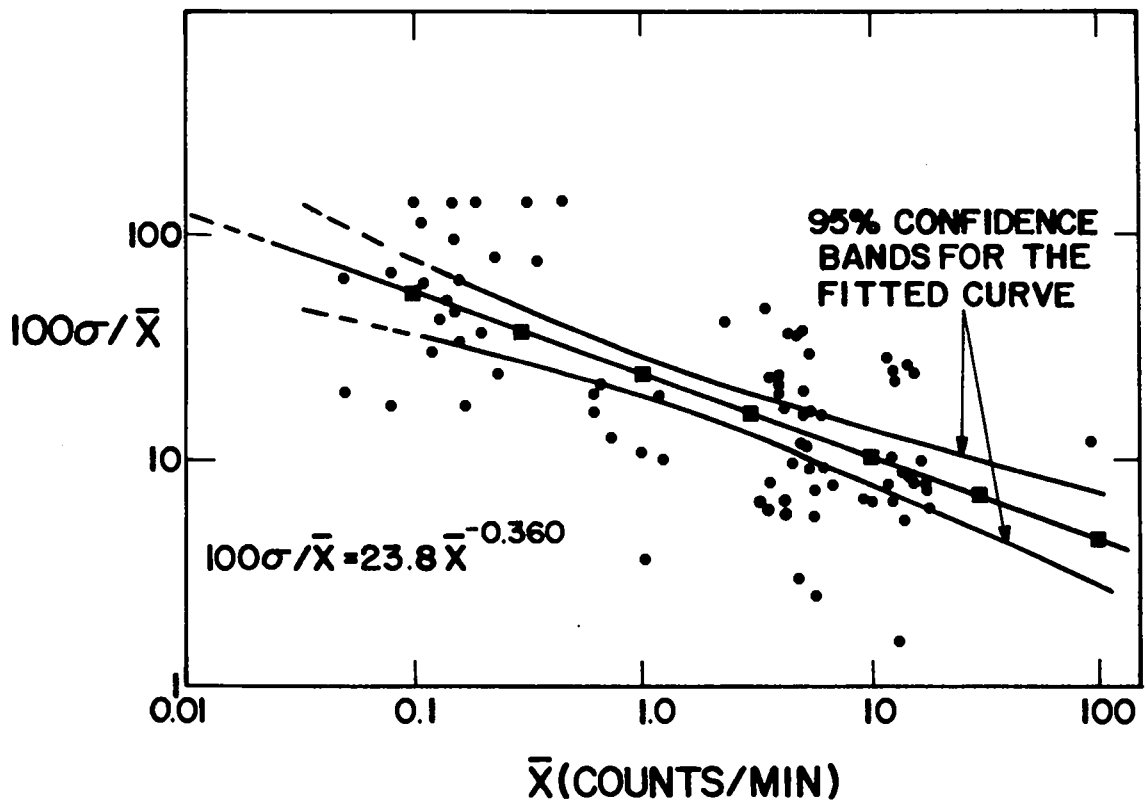

Fig. A-6.
Mean vs relative percentage standard deviation for blank and recovery data for $BiPO_4$-ppt. method.

$$\sigma_p/P = 1/P\sqrt{[(\partial P/\partial C)\sigma_C]^2 + [(\partial P/\partial B)\sigma_B]^2 + [(\partial P/\partial R)\sigma_R]^2}$$

$$= \sqrt{[\sigma_C/(C - B)]^2 + [-\sigma_B/(C - B)]^2 + (-\sigma_R/R)^2}$$

$$= \sqrt{[0.3010C^{0.8919}/(C - 0.69)]^2 + [0.533/(C - 0.69)]^2 + (0.194/0.823)^2} .$$

We hoped that a simpler equation could be developed. Over the range of 0.1 to 30 counts/min per 24-h sample, the standard deviation (σ_p) of the calculated picocuries per 24-h sample (P) was computed and plotted vs C on log-log paper (Fig. A-7). The data were fitted to the curve

$$\sigma_p = 0.3797C^{0.8893} + 0.6539(0.4278)^C .$$

For the $BiPO_4$-ppt. personnel urine assays, the reporting unit was disintegrations per minute per 24-h sample (D). This actually was only twice the counts per minute per 24-h sample, because no blanks had been subtracted and no recovery factor had been applied. Thus, the picocurie per 24-h sample conversion equation for the $BiPO_4$-ppt. assays is

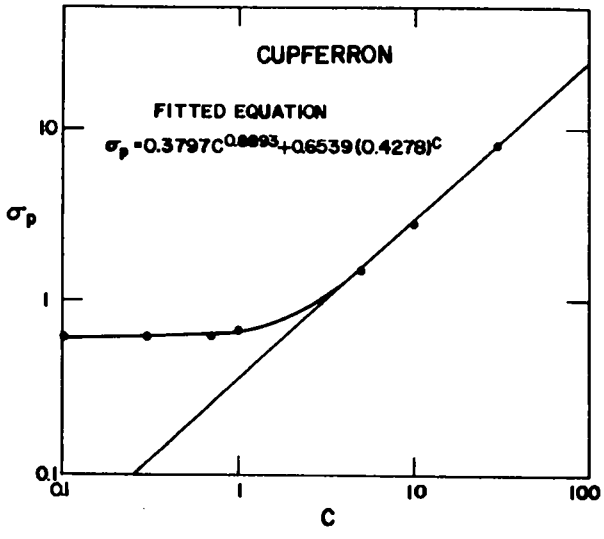

Fig. A-7.
Standard deviation of calculated pCi/24-h sample vs observed counts per minute.

$$P(\text{pCi/24-h sample}) = 1/1.1\ \{[(C \pm \sigma_C) - (B \pm \sigma_B)]/(R \pm \sigma_R)\} \quad (BiPO_4 \text{ ppt.}) ,$$

where

$$B \pm \sigma_B = 0.155 \pm 0.099 ,$$

$$R \pm \sigma_R = 0.674 \pm 0.209 ,$$

and

$$C \pm \sigma_C = C \pm 0.2384C^{0.8397} ,$$

because

$$C = D/2$$

and

$$100\ \sigma_C/C = 23.84C^{-0.3603} .$$

Figure A-8 shows the computed values of σ_p vs C, over the range 0.06 to 30 counts/min per 24-h sample. The fitted equation for σ_p is

$$\sigma_p = 0.4834C^{0.9588} + 0.1366(0.05192)^C .$$

Note: After these equations are used to calculate σ_p, the result is rounded to only two or three places after the decimal point to be consistent with the reported counts per minute per 24-h sample.

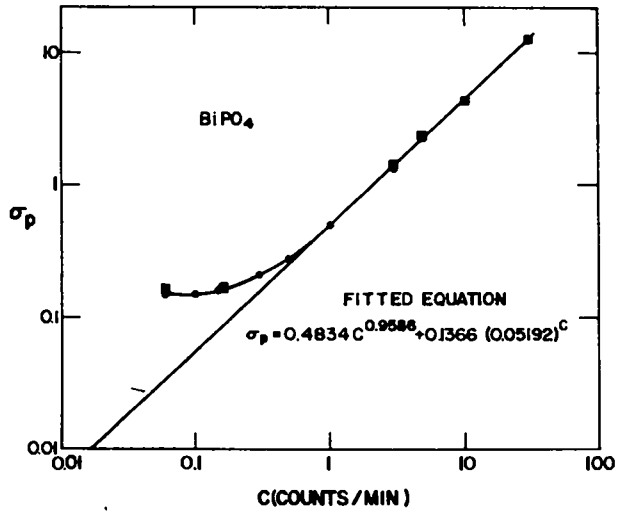

Fig. A-8.
Computed values of σ_p vs C.

APPENDIX B

PLUTONIUM URINE ASSAYS FROM JANUARY 1957 TO JANUARY 1967

Blanks, recoveries, conversion to picocuries, and standard deviation of picocurie results

From January 1957 to April 1966, the plutonium urine assays were run by the NTA method. The NTA method is not a complete description as the chemical procedures were changed at least once during that period. One change in chemical procedures is, perhaps, reflected in the significantly higher average backgrounds, as recorded in the LA Notebooks for October 1957 through October 1959. From April 1966 to January 1967, the ZnS method was used for plutonium urine assays. The chemical procedures remained the same as had been used for the NTA method, with only the counting technique changed.

Starting in September 1957, 2 blanks and 2 recoveries were run with each set of 12 personnel urine samples (making 16 samples in a complete set). This procedure was followed until November 1964 when apparently blanks were discontinued. (At least no blanks were recorded in the LA Notebooks after that date.) During most of the time from September 1957 to November 1964, the value of the spike used for determining the recovery was 0.5 dis/min of ^{239}Pu, and this value was sometimes recorded in the LA Notebooks. When different quantities of spike were used, the new values were usually recorded. From April 1963 to January 1964 and from November 1964 to January 1966, the practice of recording the actual disintegrations per minute (uncorrected for recovery) for the spiked samples was discontinued. No explanation was given and perusal of the LA Notebooks suggests that the actual recovery calculations were done elsewhere. Only the percentage recovery was recorded in the LA Notebooks.

From January 1966 for the remainder of the NTA analyses and for the ZnS method (until January 1967), other laboratory notebooks were used to record the actual counting data and to make the recovery calculations. In most cases, only the final answers were recorded in the LA Notebooks.

Starting with the NTA method, it became standard practice for the urine analysis results to be expressed in disintegrations per minute per 24-h sample with corrections for blank and recovery already applied. The problem for the body burden calculation is then to determine the standard deviation of the reported results. A technique similar to that used for cupferron and $BiPO_4$ ppt. was followed except that the final standard deviation relation was correlated with the picocuries per 24-h sample (Group H-5's reported disintegrations per minute per 24-h sample divided by 2.22).

The LA Notebooks from September 1957 through January 1966 were examined. The following data, where available, were recorded.

(1) Blank results (expressed in disintegrations per minute, that is, 50% counting geometry applied).
(2) Gross disintegrations per minute for spiked samples and spiked value when different from 0.5 dis/min.
(3) Recorded percentage recovery.

The data were kept separated according to the LA Notebook number, to see if any significant differences could be determined with the passage of time. For each LA Notebook, the average and the standard deviation were calculated for the blanks, the gross disintegrations per minute for each value of the spike, and the percentage recovery. The 4-σ (four-standard-deviation) criterion was used to eliminate outliers for the blank data, and the 3-σ criterion was used to eliminate outliers for the gross disintegrations per minute and percentage recovery data.

There were no statistically significant differences in the average percent recoveries over the entire time. Therefore, the average of all the recovery measurements was determined to be (70.71 ± 17.23)%. This value and the standard deviation were used in the following determination of the overall standard deviation of the picocuries per 24-h sample.

From October 1959 to January 1966, there were no statistically significant differences in the average blanks. The average of all blanks during this time was determined to be (0.0067 ± 0.0049) dis/min. However, the average blanks calculated for the three LA Notebooks from September 1957 to October 1959 were significantly different. For LA Notebook N-25 (September 1957 to July 1958), the blank (average ± one standard deviation) was (0.0258 ± 0.0282) dis/min. For LA Notebook N-28 (July 1958 to February 1959) the blank (average ± 1 σ) was (0.0395 ± 0.0473) dis/min. For LA Notebook N-34 (February 1959 to October 1959) the blank (average ± 1 σ) was (0.015 ± 0.0181) dis/min. These values of the average blanks and standard deviations were used in the following determination of the overall standard deviation of the picocuries per 24-h sample.

There were no statistically significant differences in the average gross disintegrations per minute for the spiked samples. These data and the individual blank averages for each LA Notebook were plotted vs their respective relative percentage standard deviations. A curve was least squares fitted to these data (Fig. B-1) with the result

$$\sigma_D/D = 0.2023D^{-0.2802} .$$

Although not following the square root of the disintegrations per minute relation as closely as the cupferron and $BiPO_4$ data, this equation nevertheless was used in the following determination of the overall standard deviation of the picocuries per 24-h sample.

The general equation relating the uncorrected disintegrations per minute per 24-h sample D, the blank B (in disintegrations per minute), and the fractional recovery R to the picocuries per 24-h sample P is

$$P = (1/2.2)\,[(D \pm \sigma_D) - (B \pm \sigma_B)]/(R \pm \sigma_R) \quad \text{(NTA)} ,$$

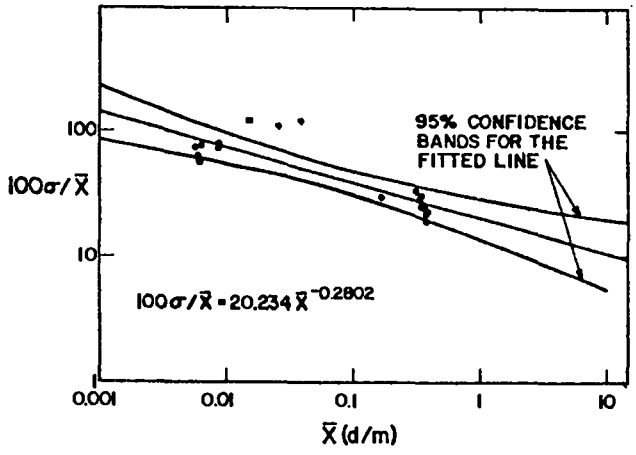

Fig. B-1.
Mean vs relative standard deviation for NTA method.

where

$(B \pm \sigma_B)_{N\text{-}25} = (0.0258 \pm 0.0282)$ dis/min ,

$(B \pm \sigma_B)_{N\text{-}28} = (0.0395 \pm 0.0473)$ dis/min ,

$(B \pm \sigma_B)_{N\text{-}34} = (0.0151 \pm 0.0181)$ dis/min ,

$(B \pm \sigma_B)_{NTA+ZnS} = (0.0067 \pm 0.0049)$ dis/min ,

$R \pm \sigma_R = (0.7071 \pm 0.1723)$,

and

$D \pm \sigma_D = D \pm 0.2023D^{0.7198}$.

The overall standard deviation (σ_p) in the calculated picocuries per 24-h sample was calculated by the method described in the cupferron data analysis. Figure B-2 shows the computed values of σ_p vs P over the range 0.0001 to 3 pCi/24-h samples. Equations fitted to the four blank curves were

$\sigma_{p_{N\text{-}25}} = 0.3168P^{0.8850} + 0.01915(0.000\ 107\ 1)^P$,

$\sigma_{p_{N\text{-}28}} = 0.3168P^{0.8850} + 003214(0.000\ 166\ 2)^P$,

$\sigma_{p_{N\text{-}34}} = 0.3168P^{0.8850} + 0.01197(0.000\ 000\ 918\ 1)^P$,

$\sigma_{p_{NTA+ZnS}} = 0.3168P^{0.8850} + 0.004414(0.000\ 085\ 14)^P$,

Note: After these equations are used to calculate σ_p, the result is rounded to only two or three places after the decimal point to be consistent with the reported disintegrations per minute per 24-h sample.

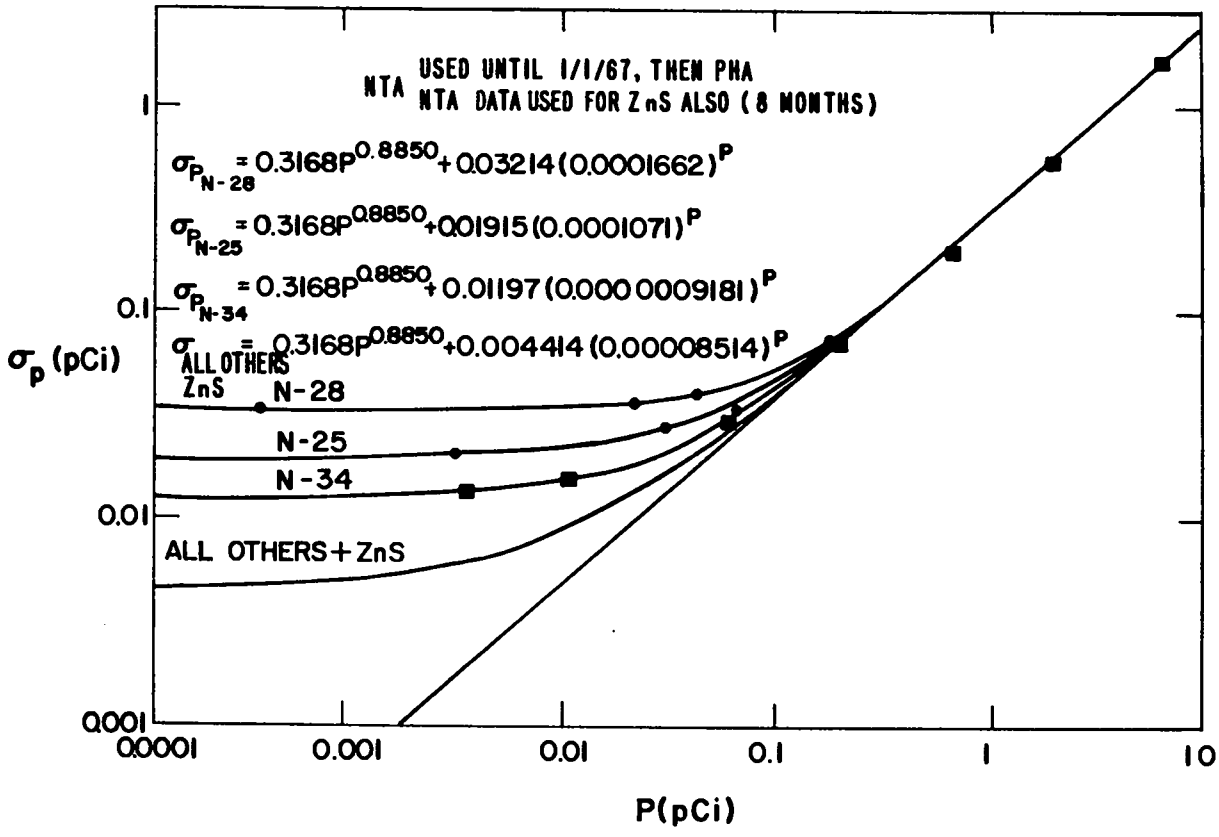

Fig. B-2.
Computed values of σ_p vs P.

APPENDIX C

PLUTONIUM URINE ASSAYS FROM JANUARY 1967 THROUGH DECEMBER 1977

Blanks and standard deviation of picocuries results

From January 1967 to June 1971, the plutonium urine assays were run either by the ZnS method or by the alpha PHA method. Only if samples were requested to be analyzed for both ^{238}Pu and ^{239}Pu was the alpha PHA method used. The chemical procedures are assumed to be essentially the same for both methods.

During this period, blank and spike data were minimal and have not been examined. The actual calculation of results was not fully computerized, but results were reported in dis/min per 24-h sample, with appropriate corrections made for blanks and recovery. For body burden calculations, the standard deviation vs reported pCi (actually dis/min per 24-h were reported

and converted to pCi), as determined in Appendix B for the ZnS method, continued to be used until June 1971.

Starting in 1971, routine quality control (QC) procedures were instituted by Group H-5, at about the same time that all plutonium urines were analyzed routinely by the alpha PHA method with added spikes of ^{242}Pu for individual determinations of chemical recovery. At first, the quality control consisted of running blanks. In 1971, only two overspikes with ^{239}Pu were recorded. From November 1972, blanks overspiked with ^{239}Pu were run routinely along with true blanks. Usually, 2 QC samples were analyzed with each set of 14 personnel samples. This practice continued through 1973. In 1974, true QC blanks were essentially discontinued. Instead, half the QC samples were overspiked with ^{239}Pu and the other half were overspiked with ^{238}Pu. This permitted a determination of blanks with respect to the isotope *not* used in the overspike and determination of the accuracy of the overspike for the isotope used to overspike. Infrequently, true blanks and overspikes with both plutonium isotopes were run in the QC program.

Data on QC was excerpted from the LA notebooks. Table C-I indicates the number and type of QC samples according to year.

To observe any significant trends over these years, the blank determinations for each isotope were averaged in groups of 25-50, and the standard deviations (σ) calculated. The only data rejected were those determinations exceeding +5 standard deviations. The data are plotted in Fig. C-1 for the average blank $\pm 2\ \sigma$ vs the year (or part of year). In general, no consistent bettering or worsening of the results appears over the years. There appears to be no difference in the ^{238}Pu and ^{239}Pu determinations. However, there is a 0.010-dis/min average positive bias in the determinations, which is encompassed by the average standard deviation of ± 0.022 dis/min.

The overall accuracy of plutonium in urine determinations was examined by calculating the ratio of the evaluated result divided by the overspike (Eval/Spike). Seven values of overspike have been used for ^{238}Pu and ten values for ^{239}Pu. Averages and standard deviations of Eval/Spike were determined for each spike value in each year. Correlations of these averages and standard deviations with the year did not appear significant for either ^{238}Pu or ^{239}Pu evaluations. The average Eval/Spike for all overspikes (regardless of magnitude) over all years was 1.02 for ^{239}Pu and 1.03 for ^{238}Pu. However, a trend was observed in the standard deviation of the average with respect to the magnitude of the overspike. These data are shown in Fig. C-2, where the single-σ values are plotted vs the magnitude of the overspike. Also shown are the weighted least squares fits of the ^{239}Pu, ^{238}Pu, and combined data. The numbers beside the points indicate the number of individual samples used to compute the standard deviation (and for weighting).

TABLE C-I

NUMBER AND TYPE OF QC SAMPLES PER YEAR

Year	^{239}Pu Blank	^{239}Pu Spike	^{238}Pu Blank	^{238}Pu Spike
1971	8	2	8	0
1972	95	17	85	0
1973	164	99	256	0
1974	150	134	148	136
1975	185	176	211	152
1976	200	206	211	194
1977	149	153	154	147

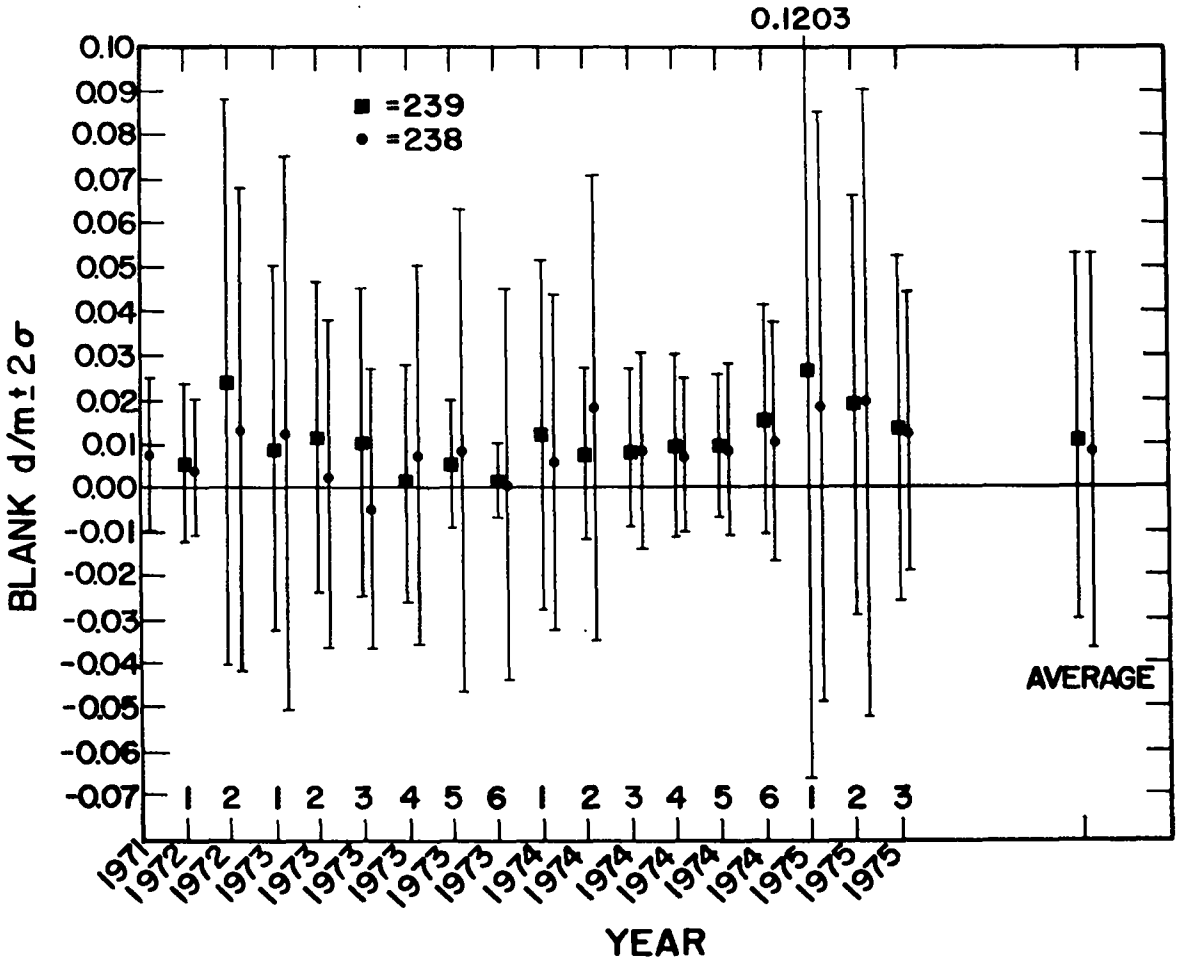

Fig. C-1.
Average blank ± 2 σ vs time.

Although there appears to be a difference in the ^{239}Pu and ^{238}Pu curves, there were fewer ^{238}Pu data and fewer data at the larger overspike values. We suspect that this scarcity has influenced the curve fitting. It was decided to use the composite least squares fit for determining the assignment of the one standard deviation for reported personnel urine results. To be consistent with the expression of standard deviation used for the NTA and ZnS methods, the fitted curve was renormalized to

$$\sigma_{P_{PHA}} = 0.0876P^{0.8790} ,$$

where $\sigma_{P_{PHA}}$ and P are in pCi. This expression equals the blank standard deviation at P = 0.041 pCi. For body burden calculations, the above equation is used to determine $\sigma_{P_{PHA}}$ for reported results >0.041 pCi. For reported results <0.041 pCi, the $\sigma_{P_{PHA}}$ is set at 0.01 pCi for all ^{238}Pu and ^{239}Pu results from June 1971 onward.

Note: The QC and blank data from April 1975 through December 1977 were examined in 1978. Only slight reductions in the average standard deviations and blanks were found. These differences were not sufficient to warrant changing the expressions given above.

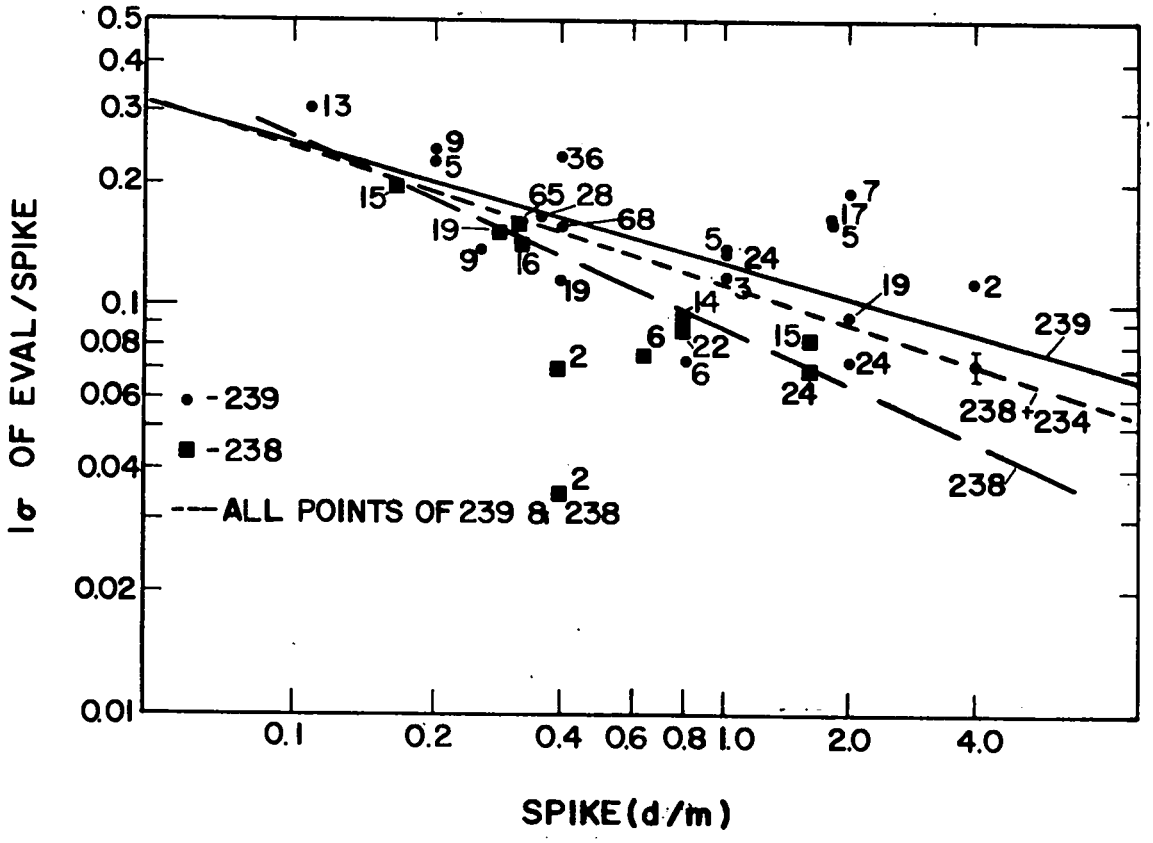

Fig. C-2.
Single-σ values vs overspike.

Note: After using these equations to calculate σ_P, the result is rounded to only two or three places after the decimal point to be consistent with the reported disintegrations per minute (or pCi) per 24-h sample.

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