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STATISTICAL ANALYSIS OF RADIOCHEMICAL MEASUREMENTS
OF TRU RADIONUCLIDES IN REDC WASTE

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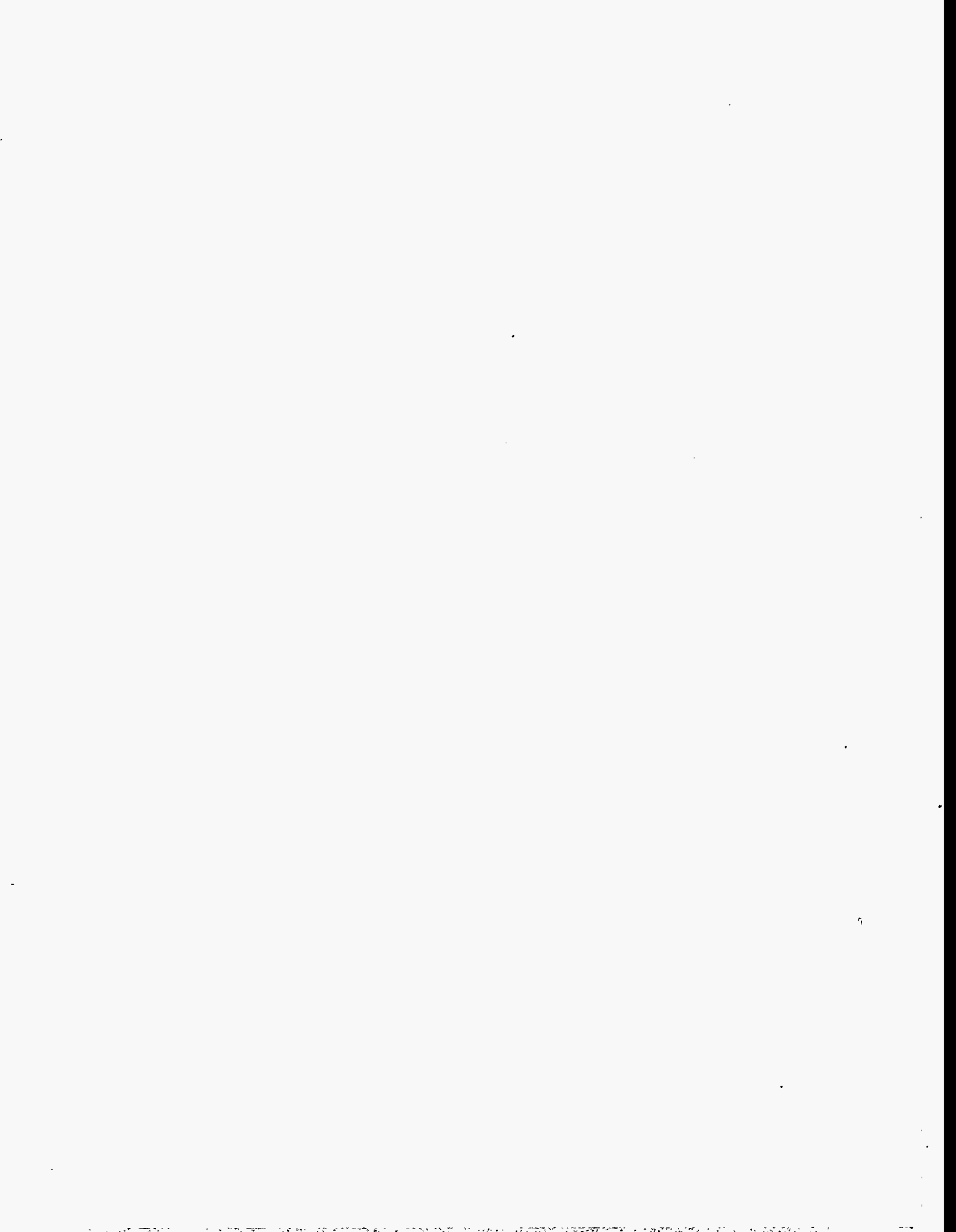
Contents

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List of Tables

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Abstract

This report summarizes the results of the study on the isotopic ratios of transuranium elements in waste from the Radiochemical Engineering Development Center actinide-processing streams. The knowledge of the isotopic ratios when combined with the results of various nondestructive assays, and in particular with the results of Active-Passive Neutron Examination Assay and Gamma Active Segmented Passive Assay, may lead to the significant increase in precision of the determination of TRU elements contained in ORNL generated waste streams.

Executive Summary

Radiochemistry measurements of contaminated-surface smears collected from within the REDC process and waste storage hot cells were statistically analyzed to estimate the proportion of gross alpha activity contributed by the transuranic radioisotopes Cm246, Pu238, Pu239, Pu240, Am241, and Am243[1]. Unlike weapons-grade plutonium, waste generated from the REDC is composed largely of non-TRU waste isotopes Cm244 and Cf252; hence the classification as TRU-waste by alpha-activity concentration alone, is difficult and requires a thorough evaluation of radiochemical results using statistical methods that address the unique aspects of the problem. This work was performed to analyze isotopic ratios of TRU radionuclides to gross alpha activity; test whether the ratios are constant over different types of debris waste; assist in the calibration of

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[1]TRU waste is, without regard to source or form, waste that is contaminated with alpha-emitting transuranium radionuclides with half-lives greater than 20 years and concentrations greater than 100nCi/g at the time of assay [DOE Order 5820.2a].

nondestructive assay instruments; and establish a basis for ascertaining whether the concentration of TRU radionuclides is less than the 100 nCi/g limit for classification as low-level waste.

Two sets of smears were collected from the hot cells. The majority of the first set of smears was collected in January 1996 near the end of target Campaign 69. Thirty-four smears were collected: 26 from cell 9; 1 each from cells 1, 2, 3, 4, 7, and 8; and 2 from cell 6. Waste material type (or component) was grouped into five classes: glass, metal, polyethylene, cell surface, and wipes. Smears were taken from each of these classes to test whether the proportion TRU was dependent on material type. The smears were analyzed by gross alpha counting in a gas flow proportional counter, alpha spectrometry, gamma-ray spectrometry, and gross neutron counting. Once the radiochemical results were available from the first set, it was realized that the plutonium sensitivity was insufficient to meet the needs of our analysis. It was concluded that a plutonium separation was required to improve the sensitivity by at least an order of magnitude from the set 1 results. Hence, a second set of smears was collected in May 1996, after Campaign 70 had begun. Twenty-six smears were collected this time, all from within cell 9, the primary cell from which waste is generated. Waste types from this set included glass, metal, and polyethylene. Having learned from our experience of the first sample set, we decided to collect a few duplicate samples from 2 separate items, a glass bottle and a piece of metal. This allowed an evaluation of the sample-to-sample variability. Additionally, a metal tool was smeared and then leached to compare a smear result with a leach result. A smear, followed by a leach, was also performed for three glass bottles. Plutonium was removed from an aliquot of leachate by solvent extraction and then analyzed by gross alpha counting and alpha spectrometry; thereby, improving the sensitivity to plutonium. In set 2 plutonium was detected in all 26 smears, versus only 9 of 34 in set 1. In summary, the samples collected in set 1 were from all of the cells—but mostly cell 9—with no duplicate smears taken. In contrast, set 2 was collected exclusively from cell 9; two subsets of duplicates were taken; and in two cases, smears were followed by an acid leach procedure, thus providing in two cases total radioactivity results per item.

Parametric and nonparametric statistical methods were performed on the data, which included a total of 60 smears and 2 leach results. Parametric methods require assumptions about the analytical form of probability distributions to be made and when possible verified on the statistical distribution of the data. Nonparametric methods use very mild assumptions like stochastic independence. The variable of interest was defined as the proportion TRU, the ratio of the radioactivity sum of measured TRU radionuclides (Bq) to gross alpha activity (Bq). For the parametric methods, normal and lognormal distributions were assumed for the distribution of the observed variable. However, no normality assumption was required for the nonparametric methods. Using parametric and nonparametric techniques, estimates of the mean and corresponding standard errors were computed for the variable, proportion TRU. Because all of the TRU radionuclides of interest were not detected in every sample, several data censoring techniques were used. Lower and upper bounds on the proportion TRU were obtained by making assumptions (low and high) about the contributing isotopes that were not detected in a smear. We defined a nondetect observation, i.e. a censored data point, as one in which at least one of the six TRU isotopes was not detected by radiochemical analysis. For a lower bound, the radioactivities of isotopes that were not detected were set to zero; for an upper bound, the radioactivity was set to 1% of the gross alpha activity, except for plutonium. Approximately 50% cases of plutonium isotopes were not detected in set 1 (recall no chemical separation), while in set 2 all plutonium isotopes were measured in all 26 smears. To address this experimental situation the complete sets of statistical analyses were repeated for three detection limits. First, the detection limit was set equal to 0.05% of the gross alpha. This limit corresponds to the lowest measured plutonium to gross alpha ratio in set 2. The second detection limit was chosen at 0.5% of gross alpha, which corresponds to typical measurement results obtained in set 2. In the third case we used the 1% detection limit, which coincides with precision typically accepted in alpha spectrometry. The latter choice is very pessimistic, and there exist cases in set 1 (see Table A1) where the reported plutonium to gross alpha ratio is less than 1%. Over 70% of the proportion TRU data points were censored, mostly due to inability to detect Cm246, a difficult isotope to measure.

Hence the importance of developing upper and lower bounds for the proportion TRU was increased under these measurement conditions. Of particular interest in this report is a description of the methodologies recommended for computing isotopic proportions, for dealing with censored data, and for rejecting outlier data points. In addition, regression analyses were performed to test the hypothesis that the proportion TRU is independent of gross alpha activity. Finally, the application of the proportion TRU to its utility in the calibration of nondestructive assay instruments is described. We did not reveal any practical discrepancy between the analyses performed with 3 different detection limits for plutonium. Therefore, only the results for the first choice (0.05%) were reported in the text. The results of the analyses for the other cases are available upon request. Three concluding plots (Figs. 10b, c, and d) contain information about all three cases to give a graphical impression of how similar the results are.

Of no surprise was the fact that the ratio of Cm244 alpha activity to gross alpha activity was greater than 0.90 in 93% of the observations. We found that the best estimate of the proportion TRU is determined by treating the data as lognormally distributed assuming interval censoring for the proportion TRU that was derived using the lower and upper bounds for the proportion TRU when nondetects were present. The mean estimate and its lower and upper bounds were computed according to our developed censoring methods. Special considerations were used for treating each of the two sample sets together, along with outlier observations, and between different waste materials. For a lognormal distribution, the statistically best estimate for proportion TRU is 0.0227 with a bounded range of [0.0180, 0.0267], (see Table 5(b), $\mu_4, \overline{\mu}_7, \underline{\mu}_7$). The lower and upper bounds are quite tight about the mean because there simply is not a large difference in treating an isotope that contributes to proportion TRU as zero versus detection limits. These results are compatible with documented process knowledge; the contribution of alpha activity from TRU radionuclides is very small, with the overwhelming majority of alpha radioactivity from Cm244. As one would expect, for the smears collected, the proportion of alpha activity that is TRU was not statistically significantly different for various materials, although this inference was based on minimal data for some material types.

- x -

Outlier observations were few in number, but quite pronounced since they contained anywhere from 3 to 5 orders of magnitude more alpha activity than the general cluster of smear data. For set 1, outliers were from smears of cell sumps, where a large portion of radioactivity can be expected to migrate. For set 2, outliers were from the leachate results. The high activity content on these observations was expected as well. When the outlier observations are included in the combined proportion TRU, the estimate increases a small amount, from 0.0227 to 0.0230 (Table 5(a)), in spite of a 5-magnitude increase in alpha activity for the outliers.

A linear regression showed that the proportion TRU decreases with increasing alpha activity. The following "linear" relationship (Table 6):

$$\ln\left(\frac{\text{TRU Activity}}{\text{gross } \alpha \text{ activity}}\right) = 1.33 - 0.19 \ln (\text{gross } \alpha \text{ activity}) \text{ [upper bound]}$$

$$\ln\left(\frac{\text{TRU Activity}}{\text{gross } \alpha \text{ activity}}\right) = 1.01 - 0.24 \ln (\text{gross } \alpha \text{ activity}) \text{ [lower bound]}$$

We must emphasize that while the relationship reported is statistically significant, in practice, it may not be important. The reason for this is two-fold: a) the results are very sensitive to how the large alpha activity results are treated mathematically (i.e. the gross alpha activity between smears and leach results are not 1:1); and b) the error of each data point, i.e. proportion TRU, is very large relative to the change in slope. That is to say, the relative change in slope is negligible relative to the Estimated Observational Error (EOE) for each of the data points. Additionally, we can state that while the amount of TRU activity increases with gross alpha activity, it increases at a smaller rate than the pronounced increase in gross alpha activity; thus leading to a negative relationship when expressed as proportion TRU.

When duplicate smears were made of the same item, the difference between measured activities was negligible compared to variations induced by sampling. In the case where an item (metal or glass) was smeared and then leached, the proportion TRU was within a few percent, even though the absolute alpha activity of the leach test was much larger.

The scope of this project offered a start at how these data might be used for

application to nondestructive assay. The variable of most interest for TRU waste is the TRU activity concentration, in nanocuries per gram of waste. This is the variable which is to be derived, in bulk, by nondestructive assay instrumentation. As mentioned, two leach tests were run: one on a metal tool, the other on three glass bottles. Leach results provide a more accurate measurement of total radioactivity per item, than do smear results. Dividing by the weight of the leached items gives one an estimate of TRU concentration "for that item". For the 3 glass bottles that were leached, the alpha activity concentration was 4E+04 nCi/g and the TRU concentration 496 nCi/g (see Table A3)—the proportion TRU was 0.0124. Smears were also collected on the glass bottles prior to leaching, yielding a gross alpha of 2.7E+04 nCi, each. Hence we now have one data point that allows us to express proportion TRU as a function of total alpha concentration and a conversion factor for relating gross alpha to nondestructive assay. By nondestructive assay measurements, gross alpha activity can be estimated, thereby providing an estimate of proportion TRU and hence, the TRU activity concentration (nCi/g). A link between sampling results and nondestructive assay has been established.

This report takes an exhaustive look at the treatment of currently available data where the desired signal (TRU radioactivity) is a small fraction of the total alpha radio activity. Handling of censored or nondetect observations, pooling data, statistical methods of estimation of isotopic proportions, and performing linear regression are covered in detail. The results of this work were well within nominal expectation. We achieved what we set out to do and now have a better understanding how the use of sampling data are to be integrated into a nondestructive assay program.

1. Introduction

1.1. Statement of the problem

A number of regulatory-based drivers have imposed stringent requirements on the characterization of radioactive waste; see [1, 2, 3]. For transuranic (TRU) waste, the Department of Energy's (DOE) Waste Isolation Pilot Plant (WIPP) requires, at a minimum, that the waste generator (a) provide methods for sampling and analyzing waste streams to establish an acceptable-knowledge baseline [4], (b) develop a recurring sampling plan that will assure that the data quality objectives can be met, and (c) calibrate and use nondestructive assay (NDA) instruments for certifying the waste, by bulk measurement, to the WIPP waste acceptance criteria (WAC) [5]. In order to meet these requirements for shipment of TRU waste to WIPP, existing waste characterization methods must be tested, evaluated, improved, and then ultimately approved by DOE and the Environmental Protection Agency (EPA).

Along with the technical and compliance related issues, cost is an important consideration in the identification of radioactive waste as low-level or TRU. This is due to the highly disparate disposal costs for low-level waste versus TRU waste. Current estimates suggest that the disposal costs for TRU waste may be a factor of 10 greater than that of low-level waste per unit volume. Thus, it is very critical - during the waste characterization process - that the radioactive waste be accurately identified as low-level or TRU to avoid any unnecessary cost penalties, while assuring compliance with regulations. Solid waste generated at the Radiochemical Engineering Development Center (REDC) located at Oak Ridge National Laboratory (ORNL) is of particular interest because it has produced approximately 90% by volume of the historic (legacy) remote-handled (RH) TRU waste, and all of the newly generated RH TRU waste generated at ORNL. Solid radioactive waste from REDC is currently managed as TRU waste, in spite of the fact that the largest fraction of radioactivity is due to the presence of Cm244 and Cf252, isotopes not considered TRU radionuclides [2]. TRU radionuclides of Pu and Am are known to be present in the waste, but there has not been satisfactory information in the past to conclusively demonstrate that the TRU concentration is less (or

greater) than the 100 nCi/g threshold [DOE Order 5820.2A]. It is imperative that a determination be made regarding the correct classification of REDC wastes in order to ensure the REDC correctly packages its TRU waste to meet the Waste Isolation Pilot Plant (WIPP) WAC [3].

The main objective of this work was to obtain better process knowledge of elemental and isotope structure of solid waste currently handled at REDC. The approach is essentially based on the application of various statistical techniques in the analysis of the collected radiochemical measurements.

The corresponding statistical information, combined with physical knowledge of the processes generating these wastes, allow us to enhance the accuracy and precision of the nondestructive waste characterization based on the technologies developed by the Applied Radiation Measurements Department (ARMD), including Active - Passive Neutron Examination Assay (APNea) and Gamma Active Segmented Passive (GASP); see [6 - 10].

If we know exactly the isotopic profile for the considered waste stream, then a reliable measurement of the total activity, or some individual components (e.g. gamma emitting isotopes), makes it possible to calculate the contents of all components of interest. The current situation is far from the above one; all components of interest are not known. Of course, waste inputs are stochastic in most respects and even routine inputs are poorly understood (compare with [10]). For instance, the solid waste streams may be "cross-contaminated" by items from another campaign's [2] activities; operational upsets can occur; the variation in composition of material after processing in REDC may depend upon operator's action; sampling measurements include a relatively small fraction of the items from a waste stream. Therefore, the main emphasis in this work is done on the statistical description and analysis of the isotope profiles. Sampling experiments are confined to the waste stream generated during Campaign 69.

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[2] A campaign at the REDC is the duration of a process to separate products from a particular target specification. Campaigns normally run from 1 to 2 years.

1.2. Material exposition

We start in Section 2 with a short description of waste generating processes and the main components (elemental or material) occurring in the considered waste stream. A thorough description may be found in [12]. Section 3 outlines the analytical methods used in this work to determine the individual isotopic activities. Section 4 contains an exploratory data analysis, which is based on visualization of available data. Various scatter plots and some simple statistical calculations help to detect a number of unusual entries and outlier observations, and to formulate the statistical hypotheses and models to be tested or verified.

After graphical summaries and evaluations, we proceed in Section 5 with several statistical methods and models to estimate the proportion TRU. The reason for considering many underlying models and methods was to assure the robustness of the final inferences with respect to the assumptions.

Appendix 1 contains a summary of the empirical information used in this study. The technical derivations and mathematical details are reported in two subsequent appendices. In particular, Appendix 2 provides the derivation of maximum likelihood estimators which should be used in the spectrometry to obtain individual isotopic activities. Finally, Appendix 3 focuses on the development of statistically efficient estimators of the proportion TRU under various assumptions concerning the random structure of the waste generation process and the associated observations.

2. Generating Processes and Main TRU Components.

During Campaign 69, the REDC processing facilities recovered and purified a number of elements from targets irradiated in the High Flux Isotope Reactor (HFIR) and remainders from past campaigns, for instance, the Savannah River Plant Reactor (SRPR); see [11, 12] for details. The list of potential radioactive elements is reported in Table 1.

Our work focused on plutonium, americium, curium, and californium isotopes. While Pu238/239/240, Cm246, and Am241/243 are by definition TRU radionuclides see [21], Cm244 and Cf252 are the primary isotopes (by activity) of REDC wastes.

Table 1: Potential Radionuclides in REDC Solid Wastes

Alpha emitters	Beta/gamma emitters
Cm242	Cs134
Cm243	Cs136
Cm244	Cs137
Cm245	
Cm246	Bk249
Cm248	
	Eu152
Cf249	Eu154
Cf250	Eu155
Cf252	
Cf253	Pm147
	Ni63
Ac227	Fe55
	Co60
Am241	I129
Am242m	Tc99
Am243	Sm151
	Sr90
Ra226*	Y90
	Sb125
Th228*	
Th229*	Ru106
Th232*	Rh106
Pu238	Ce144
Pu239	Pr144
Pu240	
Pu241	
Pu242	

* These radioisotopes are not subject to destructive analysis since radium targets are not currently processed at REDC.

- 4 -

Actually, the highly prominent presence of Cm244 and Cf252 makes the determination of TRU or not TRU by nondestructive assay (NDA) difficult, since one has to estimate a signal (emission produced by TRU waste elements) which constitutes a small fraction (usually less than 5% in samples available in this work) of the total signal.

For making the decision as to whether the waste is TRU or not TRU, NDA becomes technically admissible only when there is some prior information about the isotopic profiles of the basic elements. The immediate objective of this work was to provide this important information.

Physically, REDC wastes are very diverse and include, for instance, high efficiency particulate air filters from off-gas cleanup systems, gauges, discarded equipment, polyethylene bottles and smaller items from cell wastes. At present, the main concern is the characterization of a legacy population of 208 ℓ (55 gallons) drums, which are primarily loaded with cell wastes. Therefore, our efforts were directed to measurements with this latter type of wastes. The waste has the composition as described in Table 2.

Table 2: Waste Composition

Waste type	Waste items	Volume
Plastic	Melted poly bottles, poly blocks, MSM boots	$\approx 40\%$
Glass	Sample bottles	$\approx 30\%$
Metals	Tools	$\approx 20\%$
Cloth	Wipes	$\approx 10\%$

3. Sampling and Analytical Methods

3.1. Sampling

The selection of the sampling and analytical methods was based on the obvious compromise required between precision, time, and cost effectiveness. Most of the sampling was based on smears (relatively cheap, but not a very precise method). In the case of smears, we have a very significant contribution to the variability of the results because the smearing process itself adds to the total uncertainty through the selection and cov-

erage of smeared areas. Leaching (precise and expensive) was used only for "control" type sampling. Leaching results are valuable because they actually quantify the total activity on a waste item, allowing us to make the determination of whether or not the waste item(s) is TRU (i.e., greater than the defined limit of 100nCi/g).

Due to the very radioactive nature of the operations, all samples were collected remotely. Smears were taken using "Q-tip" swabs with wooden handles. After smearing any surface, the Q-tip head was placed in a labeled sample bottle, the extra wood broken off, and the bottle capped and removed from the REDC cell bank. This procedure made it possible to avoid contamination from the manipulator fingers and other sources. In the case of poly bottles, the inside of the bottle was smeared as well as the threads. In general, whenever it was possible, an operator tried to smear the most typical elements of every item.

Two sets of samples were collected for this study: 34 smear samples were taken for the first set, 26 smear and two leach samples were taken for the second set. The first set of samples was collected from waste items located in cell 9, where the waste is bagged out, and various locations in cells 1, 2, 3, 4, 6, 7, and 8 of the REDC facility. Cell 5 was not used in Campaign 69; therefore, no samples were taken from this cubicle. The second set of samples was taken from waste items in cell 9. Detailed sample locations and the sample tracking records are provided in Tables A4 and A5 of Appendix 1.

Waste items from each waste category were collected from Campaign 69 waste. Each waste item was selected at random from different poly buckets to ensure that the analytical results obtained from these samples represent the TRU wastes.

3.2. Analytical methods

The following methods were utilized in analyzing the "Q-tip" smears of waste for radionuclide content. In the first set, samples were analyzed using gross alpha, alpha spectrometry, gamma spectrometry, and gross neutron determination. Gross neutron determinations were performed in order to calculate Cf252 activities. The smears were individually leached in a 4N Nitric Acid solution for 2 hours at room temperature. The resulting leachate was made to a known volume prior to analyzing so that a total

smear activity determination could be made. Gamma spectrometry determinations were made by counting an aliquot of the leachate with a High Purity Germanium detector. The discrete gamma-ray energy peaks were identified and integrated to quantify the gamma-emitting radionuclides; compare with [13]. All of the fission and activation products identified in the smear samples were analyzed by gamma spectrometry, as well as Am241 and Am243. Although the two isotopes of americium are primarily alpha emitters, they were in general very small percentages of the total alpha activity and therefore could not be accurately measured using alpha spectrometry. Their concentrations were determined using gamma-ray spectrometry. The gross alpha determinations were made by evaporating an aliquot of the leachate on a stainless steel plate (in duplicate) followed by alpha counting on a gas proportional counter. One of these plates was then counted in an alpha spectrometer to identify the alpha-emitting radionuclides by pulse-height analysis. In order to quantify the major alpha emitters, each peak (energy) identified by alpha spectrometry was integrated to determine the individual peak areas. Once all peaks were integrated, the areas were summed to provide a total area. The individual peak areas were then divided by the total area to determine the percentage of alpha counts due to a particular alpha energy; compare with [14, 15]. The gross alpha and alpha spectrometry analyses thus provided quantitative activities for the different alpha energies. From the statistical point of view, this procedure may be considered as the first order approximation of the maximum likelihood estimator (see Appendix 2). The use of the exact maximum likelihood estimator may lead to a significant increase in peak area accuracy. The corresponding statistical software can be developed and applied at later stages of the TRU project.

Once compiled, additional information from gross neutron determinations (Cf252) and gamma ray analyses (Am isotopes) was used to determine the amount of contributions these nuclides made to their respective alpha energy peaks. As an example, Pu238 and Am241 both have primary alpha energies of nominally 5.50 MeV. By quantifying Am241 through gamma spectrometry, the activity contributed by Am241 to the 5.50 MeV peak was subtracted from the total activity at that energy to leave the Pu238 activity. Alpha-emitting radionuclides that were not registered by alpha spectrometry

were stated in data sets to be less than 1% of the total alpha activity. The selection of this detection level was based on the sensitivity of the alpha spectrometer used for the analysis.

Since this project was intended to profile alpha-emitting nuclides in the REDC waste stream (particularly TRU nuclides), no additional radiochemical separations were performed in order to quantify certain pure or nearly pure beta-emitters such as Ni63 and Sr90 that are also known to be present in the waste.

With the first sample set complete, it was apparent that the plutonium isotopes were often not detected by this method of analysis. It was agreed that an additional set of smears (26 samples) and two leached samples would be taken and, in addition to the analyses requested for the first sample set, a plutonium separation would be performed. The plutonium was removed from an aliquot of leachate by solvent extraction and then analyzed through a separate gross alpha and alpha spectrometry determination as described previously. Thus, plutonium isotopes of interest (Pu238,239,240,242) were separated from the other alpha emitters so that the limits of detection were greatly reduced, at least by an order of magnitude. This series of samples was complemented by two leaching experiments, in which the items previously smeared were used. All the data are compiled in Appendix 1.

4. Exploratory Analysis

4.1. Data description and notations

The analysis of the data has focused on the activity (Bq/total) observations for gross alpha and a collection of alpha-emitting radionuclides. Each observation vector from a given sample, which consisted of the observed activities with associated errors from a sample, was identified according to the cell source and waste type (i.e., plastic, metal, glass, etc.). The following alpha-emitting radionuclides were found: Am241, Am243, Cf252, Cm242, Cm244, Cm246, Pu238, and Pu239/240. The TRU activity for each sample was calculated from the sum of the activities of the following radionuclides: Am241, Am243, Cm246, Pu238, and Pu239/240. As mentioned in the previous section,

- 8 -

the data were obtained from two separate sample sets. The first sample set consisted exclusively of smear samples and a plutonium separation was not performed. The second sample set, in addition to smear samples, also had two leaching samples, on which plutonium separations were performed. The second sample set was required because of the high proportion of nondetects in the first sample set, i.e., observed activities known only to be "below a detection limit." A number of statistical and graphical methods were applied to the two data sets (34 observations in the first and 28 observations in the second). These methods were directed toward:

1. the detection of outliers or unusual observations;

2. a search for inconsistent observation vectors (e.g., individual radionuclide activities sum to a value greater than the gross alpha activity);

3. the analysis of observation vectors with nondetects; and

4. the graphical analysis of the proportion of TRU activity by waste type.

The isotopic ratios or proportions for the radionuclides of interest were calculated from each smear using the ratio of the observed radionuclide activity to the sample average gross alpha activity. Because the primary interest was in the TRU activity and its proportion, the analysis focused on the following components or proportions:

1. TRU (PTRU) (consists of Cm246, Pu238, Pu239/240, Am241,Am243);

2. Cm244 (PCm244);

3. Cf252 (PCf252);

4. the remaining radionuclides REST (PREST);

5. gross alpha (GA).

(The "P" in the notation for the above components means proportion, e.g., PTRU is the proportion of TRU activity in the sample.) We used the compound variables (i.e., aggregated radionuclide activities) to reduce the impact of the large number of nondetects. Actually, introduction of the variable REST is redundant in this particular

setting, because it comprises only one measurable radionuclide Cm242 (activities of other radionuclides listed in Table 1 were negligible). However, we use this notation to emphasize the idea of aggregation, which can be employed in similar situations but with a larger number of radionuclides. In what follows, we refer to set 1 as 34 samples from the first data set, and to set 2 as samples from the second data set.

Appendix 1 contains available information about these two data sets. It is evident that the number of nondetects, especially in set 1, is too large to use statistical approaches that do not account for this. Therefore, we had to apply alternative statistical procedures, which were developed to handle the nondetects properly. They are computationally intensive and demand some mathematical rigor. Before we proceed with those procedures, let us explore the data by relying upon visualization techniques.

4.2. Nondetects handling

As mentioned above, for many of the samples there were activities for some radionuclides (e.g., Cm246) known only to be below a detection limit, which was taken as 1% of the mean gross alpha activity. This bound is based on limitations of the analytical technique (see Section 3.2). However, for the plutonium radionuclides, the second set, which incorporated plutonium separations prior to analysis, showed that none of the samples contained individual plutonium radionuclides at a level lower than 0.05% of the gross alpha. Therefore, for statistical analyses, the lower limit of detection for these nuclides was set to 0.05% of the gross alpha (see comments about analyses for other choices of detection limits in the Executive Summary). A censoring flag (CNS) was defined for each radionuclide that equaled 0 for a detect and equaled 1 if the observed activity was a nondetect. For a nondetect, the activity and activity error were set equal to each other and this is what was used to designate a nondetect observation. Because of the presence of nondetects, the calculated TRU activity from the sum of the stated activities for the five radionuclides mentioned above actually represents an upper limit on TRU activity whenever nondetect components are present. A lower limit on the TRU activity was also calculated by replacing a nondetect TRU radionuclide component by 0. See the next section for more details.

- 10 -

4.3. Visual analysis

The observed activities for the radionuclides of interest ranged over orders of magnitude. Therefore, subsequent graphs and analyses were generally performed using the natural logarithm of the observed activities. The following plots were used in the preliminary evaluation of both data sets. Figures 1 (a-c) and 2(a-c) show the transformed TRU, Cf252, and Cm244 versus the transformed gross alpha activity for each experiment. The "x"'s in these figures indicate nondetect observations. In the case of TRU, it means that at least one of the TRU radionuclides was not detected. In this work, we have not explicitly addressed the problem of different numbers of nondetects in the compound variables. Some conclusions from a review of these figures are:

1. The significant number of nondetects (see last two columns in Tables 3-5 for exact numbers) will cause difficulties in the estimation of the TRU isotopic ratio in both sample sets (Figures 1a and 2a).

2. Cf252 and Cm244 activities do not have a significant problem with nondetects (Figures 1b and 2b).

3. Cm244 activity is the dominant activity in each sample. This can be seen in Figures 1c and 2c where most of the observations fall close to the 45° line $y = x$, meaning that the Cm244 activity is nearly equal to the gross alpha activity.

4. There are a couple of observations in each experiment that fall away from the bulk of the data, usually at the high gross alpha levels. We call these points leverage points and they are discussed in more detail later.

Figures 3 (a-c) and 4 (a-c) are similar to Figures 1 and 2 except that "G" stands for glass, "M" for metal, "P" for poly (melted or bottle), "C" for cell, and "W" for wipes.

Additional information becomes more illuminating if we introduce upper and lower bounds on an observed compound variable (see [16]), such as TRU activity, where for the i-th observation the upper bound is given by

$$u_i = \sum_{j=1}^{k} y_{ij}, \tag{1}$$

- 11 -

where j stands for radionuclide type, k is the number of radionuclides in the corresponding compound variable, y_{ij} is either an observed activity value, or a detection limit, discussed previously in Section 4.2. The lower bound is given by

$$\ell_i = \sum_{j=1}^{k'} y_{ij}, \tag{2}$$

where k' includes only the observed radionuclide activities in the compound variable, i.e., nondetects are omitted. "Two-sided" information is presented in Figures 5 and 6. We use "0" when $u_i = \ell_i$ (i.e., when there are no nondetects), and U or L to distinguish the upper and lower limits, respectively, when $u_i \neq \ell_i$.

Additional facts may be derived from a review of Figures 1 - 6.

1. There are several points, which stand apart from the majority, primarily at the highest activity levels. These are, so called, leverage points (see Figures 1a and 2a) and must be treated with some caution since these observations may dominate and distort any estimates or conclusions. Because of the extremely high gross alpha activity in two samples from each experiment (ID = RH4D and RH8A in sample set 1 and ID = LN-GL and LN-ML, the leached samples in sample set 2), the analyses were performed with and without these points to evaluate their effect on the results. RH4D was a smear from the sump in cell 4; RH8A from the sump in cell 8. The leached samples were glass bottles (GL) and metal (ML).

2. Two points (ID = RH4D and RH8A in sample set 1) correspond to smears made on the cell's surface. Actually, all observations of this type must be treated with some extra caution, because they can be affected more than others by previous campaigns. An attentive reader may notice that the majority of results marked with "C" in Figure 3 (a-c) stand apart from the rest of the data.

3. Observations from sample set 2 generally have higher gross alpha activities than those in sample set 1. The explanation could be very simple: two different operators collected samples for these two sets. Perhaps the second one spent more time in "smearing" items. However, other causes may be involved as well, for

example process changes during periods between the sampling times.

4. Observations are generally scattered along the straight line with unit slope. What could be the cause of that? For example, let us assume that (i stands for the observation number):

$$\frac{TRU_i}{GA_i} \equiv \text{const} = f. \tag{3}$$

Then

$$\ln TRU_i = \ln f + \ln GA_i, \tag{4}$$

which can be written as $y_{i1} = c + y_{i2}$ using the notation introduced in the comments to (1). Thus, the observed phenomenon is rather strong evidence that the ratio of activities of TRU and Cm244 is stable but affected by various "random" factors. More thorough analysis of this statement will be postponed until the following section.

5. Estimation of TRU Isotopic Ratio

5.1. Analysis of sample means

As an additional analysis of the evaluation of the radionuclide activities, different underlying mathematical models were used to estimate the mean PTRU in each experiment by waste type as well as combined. Only the individual TRU isotopic ratio from each sample, i.e., ratio of individual isotopic peak area to total peak area, as available for the analysis (see Section 3.2 for details). If the individual isotopic peak areas become available a more efficient statistical procedure, developed in the framework of this study and described in Appendix 3, could be used to evaluate PTRU and other isotopic ratios.

All of the models in this analysis made use of the nondetect or censored observations using both parametric and nonparametric methods. The basic parametric models assumed an underlying censored normal or lognormal distribution for the observed proportions. Different censoring mechanisms were also considered, e.g., one-sided or interval censoring. In the one-sided interval censoring case, the censored or nondetect observations are assumed to be less than the specified detection limit, i.e., the censored value could be any value from minus infinity to the detection limit (see $\hat{\mu}_1, \hat{\mu}_2$ and $\hat{\mu}_5$ in

- 13 -

Tables 3-5). In the closed interval censoring case, the censored nondetect observations are assumed to be between zero and the detection limit (see $\hat{\mu}_3$ and $\hat{\mu}_4$ in Tables 3-5). The nonparametric methods made use of estimates originally developed in the analysis of censored survival times. The nonparametric method uses the product limit estimate of the cumulative distribution function to obtain the estimated mean. The estimate was originally given by Kaplan and Meier and is described in [16]. All computations to obtain the censored estimates are based on procedures from the Statistical Analysis System (SAS [17]).

Tables 3, 4, and 5 provide a summary of the estimates of the mean of PTRU from the parametric and nonparametric models for the two sample sets individually and combined. The normal method noted in these tables assumes the original untransformed observations are normally distributed, whereas the lognormal method assumes the log-transformed observations are normally distributed. It should be emphasized that the Kaplan-Meier estimate does not need any assumptions about normality. Because of the 100% nondetects for the glass and metal components in sample set 1 and the poly component in sample set 2, no reliable estimates are available for these materials. When there is only one detect in the sample, the nonparametric method (Kaplan-Meier) cannot provide a standard error estimate in this case. The nonparametric method requires fewer assumptions than the parametric methods, and, therefore, gives us more defensible estimates; see [16]. However some of the parametric methods provide estimates of similar magnitudes. If additional sampling is planned, it would be advisable to obtain more detect observations from each waste component. A rough rule-of-thumb would be that if we desire a 50% reduction in the standard error, we should quadruple the sample size.

The first two parametric estimates reported in all tables are based on one-sided censoring, i.e. in the analyses we used only the fact that (see (1) in Section 4)

$$TRU_i \leq u_i. \tag{5}$$

As it can be seen, the use of only this fact generally leads to estimates (i.e., $\hat{\mu}_1, \hat{\mu}_2$)

which are unrealistically low, and must be discarded in future studies. As soon as we complemented (5) by an obvious constraint that

$$\ell_i \leq TRU_i, \tag{6}$$

all of the estimates reached the vicinity of 0.03 for the first sample set and 0.02 for the second one (see estimates $\hat{\mu}_3$ and $\hat{\mu}_4$).

The last two estimates (i.e., "Normal sample mean" and "Lognormal sample mean") in Tables 3 - 5 are determined as follows:

$$\overline{\mu}_6 = n^{-1} \sum_{i=1}^{n} u_i \ \ \text{and} \ \ \underline{\mu}_6 = n^{-1} \sum_{i=1}^{n} \ell_i, \tag{7}$$

$\overline{\mu}_7$ and $\underline{\mu}_7$ are the same as $\overline{\mu}_6$ and $\underline{\mu}_6$ except that the logarithm of the observed activities are used rather than the untransformed activities. If $\hat{\mu}$ is a hypothetical arithmetic mean constructed under the assumption that all nondetects are known, then obviously

$$\underline{\mu}_6 \leq \hat{\mu} \leq \overline{\mu}_6. \tag{8}$$

One can see that the discrepancy between $\underline{\mu}_6$ and $\overline{\mu}_6$ in all the tables is of order 0.01 or less in spite of a large number of nondetects the "true" but unknown $\hat{\mu}$ falls within a 30% range. The estimates which take into consideration the interval censoring (they are marked in the tables by the "Type Cens" entry "Interval") are regularly included in the interval $(\underline{\mu}, \overline{\mu})$. For instance, in all tables

$$\underline{\mu}_6 < \hat{\mu}_3 < \overline{\mu}_6. \tag{9}$$

Tables 3, 4, and 5 are the results of the statistical analysis of set 1, set 2, and sets 1 plus 2, respectively. The tables are partitioned into (a) and (b), corresponding to all observations and outlier observations omitted. The columns denote the method of estimation; the type of data censoring employed; the waste component type (wipes, poly, metal, glass, cell, or combined); the mean estimate for PTRU, the proportion TRU relative to gross alpha radioactivity; the standard error of the mean for PTRU;

Method	Type Cens	Waste Comp	Estimate	Std Err	n	# Cens
Normal	One-sided	Combined	0.0005	0.0128	34	30
μ_1		Cell	-0.0005	0.0215	9	8
		Glass	--	--	5	5
		Metal	--	--	4	4
		Poly	0.0044	0.0147	11	10
		Wipes	0.0220	0.0141	5	3
Lognormal	One-sided	Combined	0.0088	0.0042	34	30
μ_2		Cell	0.0072	0.0072	9	8
		Glass	--	--	5	5
		Metal	--	--	4	4
		Poly	0.0064	0.0060	11	10
		Wipes	0.0240	0.0085	5	3
Normal	Interval	Combined	0.0359	0.0046	34	30
μ_3		Cell	0.0331	0.0132	9	8
		Glass	0.0617	0.0132	5	5
		Metal	0.0341	0.0021	4	4
		Poly	0.0291	0.0036	11	10
		Wipes	0.0313	0.0061	5	3
Lognormal	Interval	Combined	0.0291	0.0032	34	30
μ_4		Cell	0.0217	0.0062	9	8
		Glass	0.0539	0.0132	5	5
		Metal	0.0340	0.0020	4	4
		Poly	0.0259	0.0042	11	10
		Wipes	0.0289	0.0051	5	3
Kaplan-Meier	One-sided	Combined	0.0274	0.0012	34	30
μ_5		Cell	0.0294	.	9	8
		Glass	--	--	5	5
		Metal	--	--	4	4
		Poly	0.0257	.	11	10
		Wipes	0.0348	0.0069	5	3
Normal Sample Mean	Ignored	Combined	0.0402	0.0048	34	30
(Upper bound)		Cell	0.0373	0.0141	9	8
$\bar{\mu}_6$		Glass	0.0669	0.0148	5	5
		Metal	0.0395	0.0029	4	4
		Poly	0.0332	0.0042	11	10
		Wipes	0.0343	0.0058	5	3
Normal Sample Mean	Set to 0	Combined	0.0317	0.0047	34	30
(Lower bound)		Cell	0.0289	0.0140	9	8
$\underline{\mu}_6$		Glass	0.0566	0.0147	5	5
		Metal	0.0290	0.0029	4	4
		Poly	0.0255	0.0032	11	10
		Wipes	0.0278	0.0076	5	3
Lognormal Sample Mean	Ignored	Combined	0.0336	0.0034	34	30
(Upper bound)		Cell	0.0272	0.0067	9	8
$\bar{\mu}_7$		Glass	0.0597	0.0148	5	5
		Metal	0.0392	0.0029	4	4
		Poly	0.0294	0.0052	11	10
		Wipes	0.0327	0.0048	5	3
Lognormal Sample Mean (Log)	Set to 0	Combined	0.0234	0.0035	34	30
(Lower bound)		Cell	0.0144	0.0064	9	8
$\underline{\mu}_7$		Glass	0.0479	0.0146	5	5
		Metal	0.0286	0.0029	4	4
		Poly	0.0230	0.0036	11	10
		Wipes	0.0245	0.0059	5	3

Method	Type Cens	Waste Comp	Estimate	Std Err	n	# Cens
Normal μ_1	One-sided	Combined	0.0005	0.0128	34	30
		Cell	-0.0005	0.0215	9	8
		Glass	--	--	5	5
		Metal	--	--	4	4
		Poly	0.0044	0.0147	11	10
		Wipes	0.0220	0.0141	5	3
Lognormal μ_2	One-sided	Combined	0.0088	0.0042	34	30
		Cell	0.0072	0.0072	9	8
		Glass	--	--	5	5
		Metal	--	--	4	4
		Poly	0.0064	0.0060	11	10
		Wipes	0.0240	0.0085	5	3
Normal μ_3	Interval	Combined	0.0359	0.0046	34	30
		Cell	0.0331	0.0132	9	8
		Glass	0.0617	0.0132	5	5
		Metal	0.0341	0.0021	4	4
		Poly	0.0291	0.0036	11	10
		Wipes	0.0313	0.0061	5	3
Lognormal μ_4	Interval	Combined	0.0291	0.0032	34	30
		Cell	0.0217	0.0062	9	8
		Glass	0.0539	0.0132	5	5
		Metal	0.0340	0.0020	4	4
		Poly	0.0259	0.0042	11	10
		Wipes	0.0289	0.0051	5	3
Kaplan-Meier μ_5	One-sided	Combined	0.0274	0.0012	34	30
		Cell	0.0294	.	9	8
		Glass	--	--	5	5
		Metal	--	--	4	4
		Poly	0.0257	.	11	10
		Wipes	0.0348	0.0069	5	3
Normal Sample Mean (Upper bound) $\bar{\mu}_6$	Ignored	Combined	0.0402	0.0048	34	30
		Cell	0.0373	0.0141	9	8
		Glass	0.0669	0.0148	5	5
		Metal	0.0395	0.0029	4	4
		Poly	0.0332	0.0042	11	10
		Wipes	0.0343	0.0058	5	3
Normal Sample Mean (Lower bound) $\underline{\mu}_6$	Set to 0	Combined	0.0317	0.0047	34	30
		Cell	0.0289	0.0140	9	8
		Glass	0.0566	0.0147	5	5
		Metal	0.0290	0.0029	4	4
		Poly	0.0255	0.0032	11	10
		Wipes	0.0278	0.0076	5	3
Lognormal Sample Mean (Upper bound) $\bar{\mu}_7$	Ignored	Combined	0.0336	0.0034	34	30
		Cell	0.0272	0.0067	9	8
		Glass	0.0597	0.0148	5	5
		Metal	0.0392	0.0029	4	4
		Poly	0.0294	0.0052	11	10
		Wipes	0.0327	0.0048	5	3
Lognormal Sample Mean (Lower bound) $\underline{\mu}_7$	Set to 0	Combined	0.0234	0.0035	34	30
		Cell	0.0144	0.0064	9	8
		Glass	0.0479	0.0146	5	5
		Metal	0.0286	0.0029	4	4
		Poly	0.0230	0.0036	11	10
		Wipes	0.0245	0.0059	5	3

TABLE 4(a). ESTIMATION OF TRU PROPORTION FROM SECOND SAMPLE SET
NO OBSERVATIONS OMITTED

Method	Type Cens	Waste Comp	Estimate	Std Err	n	# Cens
Normal $\hat{\mu}_1$	One-sided	Combined	0.0111	0.0036	28	20
		Glass	0.0134	0.0005	13	12
		Metal	0.0202	0.0034	12	5
		Poly	--	--	3	3
Lognormal $\hat{\mu}_2$	One-sided	Combined	0.0131	0.0019	28	20
		Glass	0.0134	0.0005	13	12
		Metal	.0.0188	0.0026	12	5
		Poly	--	--	3	3
Normal $\hat{\mu}_3$	Interval	Combined	0.0197	0.0021	28	20
		Glass	0.0134	0.0005	13	12
		Metal	0.0244	0.0028	12	5
		Poly	0.0322	0.0098	3	3
Lognormal $\hat{\mu}_4$	Interval	Combined	0.0173	0.0017	28	20
		Glass	0.0134	0.0005	13	12
		Metal	0.0225	0.0026	12	5
		Poly	0.0281	0.0085	3	3
Kaplan-Meier $\hat{\mu}_5$	One-sided	Combined	0.0157	0.0015	28	20
		Glass	0.0140	--	13	12
		Metal	0.0204	0.0032	12	5
		Poly	--	--	3	3
Normal Sample Mean (Upper bound) $\bar{\mu}_6$	Ignored	Combined	0.0315	0.0021	28	20
		Glass	0.0167	0.0006	13	12
		Metal	0.0266	0.0032	12	5
		Poly	0.0366	0.0115	3	3
Normal Sample Mean (Lower bound) $\underline{\mu}_6$	Set to 0	Combined	0.0161	0.0023	28	20
		Glass	0.0075	0.0008	13	12
		Metal	0.0225	0.0028	12	5
		Poly	0.0276	0.0123	3	3
Lognormal Sample Mean (Upper bound) $\bar{\mu}_7$	Ignored	Combined	0.0211	0.0016	28	20
		Glass	0.0166	0.0006	13	12
		Metal	0.0244	0.0031	12	5
		Poly	0.0333	0.0100	3	3
Lognormal Sample Mean (Lower bound) $\underline{\mu}_7$	Set to 0	Combined	0.0127	0.0017	28	20
		Glass	0.0070	0.0007	13	12
		Metal	0.0208	0.0024	12	5
		Poly	0.0227	0.0098	3	3

TABLE 4(b). ESTIMATION OF TRU PROPORTION FROM SECOND SAMPLE SET TWO OUTLIER OBSERVATIONS FROM LEACHING SAMPLES OMITTED

Method	Type Cens	Waste Comp	Estimate	Std Err	n	# Cens
Normal $\hat{\mu}_1$	One-sided	Combined	0.0114	0.0036	26	18
		Glass	0.0134	0.0005	12	11
		Metal	0.0202	0.0034	11	4
		Poly	–	–	3	3
Lognormal $\hat{\mu}_2$	One-sided	Combined	0.0131	0.0019	26	18
		Glass	0.0134	0.0005	12	11
		Metal	0.0188	0.0026	11	4
		Poly	–	–	3	3
Normal $\hat{\mu}_3$	Interval	Combined	0.0191	0.0022	26	18
		Glass	0.0134	0.0005	12	11
		Metal	0.0233	0.0028	11	4
		Poly	0.0322	0.0098	3	3
Lognormal $\hat{\mu}_4$	Interval	Combined	0.0169	0.0017	26	18
		Glass	0.0134	0.0005	12	11
		Metal	0.0215	0.0025	11	4
		Poly	0.0281	0.0085	3	3
Kaplan-Meier $\hat{\mu}_5$	One-sided	Combined	0.0158	0.0016	26	18
		Glass	0.0140	–	12	11
		Metal	0.0204	0.0032	11	4
		Poly	–	–	3	3
Normal Sample Mean (Upper bound) $\bar{\mu}_6$	Ignored	Combined	0.0225	0.0022	26	18
		Glass	0.0164	0.0006	12	11
		Metal	0.0252	0.0032	11	4
		Poly	0.0366	0.0115	3	3
Normal Sample Mean (Lower bound) $\underline{\mu}_6$	Set to 0	Combined	0.0156	0.0023	26	18
		Glass	0.0072	0.0008	12	11
		Metal	0.0216	0.0029	11	4
		Poly	0.0276	0.0123	3	3
Lognormal Sample Mean (Upper bound) $\bar{\mu}_7$	Ignored	Combined	0.0206	0.0016	26	18
		Glass	0.0163	0.0006	12	11
		Metal	0.0232	0.0030	11	4
		Poly	0.0333	0.100	3	3
Lognormal Sample Mean (Lower bound) $\underline{\mu}_7$	Set to 0	Combined	0.0123	0.0017	26	18
		Glass	0.0068	0.0007	12	11
		Metal	0.0200	0.0024	11	4
		Poly	0.0227	0.0098	3	3

TABLE 5(a). ESTIMATION OF TRU PROPORTION FROM BOTH SAMPLE SETS
NO OBSERVATIONS OMITTED

Method	Type Cens	Waste Comp	Estimate	Std Err	n	# Cens
Normal $\hat{\mu}_1$	One-sided	Combined	0.0079	0.0045	62	50
		Cell	-0.0005	0.0215	9	8
		Glass	0.0133	0.0005	18	17
		Metal	0.0199	0.0033	16	9
		Poly	0.0032	0.0151	14	13
		Wipes	0.0220	0.0141	5	3
Lognormal $\hat{\mu}_2$	One-sided	Combined	0.0116	0.0020	62	50
		Cell	0.0072	0.0072	9	8
		Glass	0.0134	0.0005	18	17
		Metal	0.0186	0.0025	16	9
		Poly	0.0061	0.0057	14	13
		Wipes	0.0240	0.0085	5	3
Normal $\hat{\mu}_3$	Interval	Combined	0.0285	0.0029	62	50
		Cell	0.0331	0.0132	9	8
		Glass .	0.0259	0.0064	18	17
		Metal	0.0268	0.0025	16	9
		Poly	0.0298	0.0035	14	13
		Wipes	0.0313	0.0061	5	3
Lognormal $\hat{\mu}_4$	Interval	Combined	0.0230	0.0019	62	50
		Cell	0.0218	0.0062	9	8
		Glass	0.0176	0.0034	18	17
		Metal	0.0248	0.0025	16	9
		Poly	0.0264	0.0037	14	13
		Wipes	0.0289	0.0051	5	3
Kaplan-Meier $\hat{\mu}_5$	One-sided	Combined	0.0165	0.0013	62	50
		Cell	0.0294	.	9	8
		Glass	0.0140	.	18	17
		Metal	0.0200	0.0030	16	9
		Poly	0.0257	.	14	13
		Wipes	0.0348	0.0069	5	3
Normal Sample Mean (Upper limit) $\bar{\mu}_6$	Ignored	Combined	0.0325	0.0030	62	50
		Cell	0.0374	0.0141	9	8
		Glass	0.0307	0.0066	18	17
		Metal	0.0299	0.0029	16	9
		Poly	0.0339	0.0039	14	13
		Wipes	0.0343	0.0058	5	3
Normal Sample Mean (Lower bound) $\underline{\mu}_6$	Set to 0	Combined	0.0247	0.0029	62	50
		Cell	0.0289	0.0140	9	8
		Glass	0.0211	0.0065	18	17
		Metal	0.0241	0.0023	16	9
		Poly	0.0260	0.0036	14	13
		Wipes	0.0278	0.0076	5	3
Lognormal Sample Mean (Upper bound) $\bar{\mu}_7$	Ignored	Combined	0.0272	0.0020	62	50
		Cell	0.0272	0.0067	9	8
		Glass	0.0237	0.0037	18	17
		Metal	0.0275	0.0030	16	9
		Poly	0.0302	0.0045	14	13
		Wipes	0.0327	0.0048	5	3
Lognormal Sample Mean (Lower bound) $\underline{\mu}_7$	Set to 0	Combined	0.0177	0.0019	62	50
		Cell	0.0144	0.0064	9	8
		Glass	0.0119	0.0028	18	17
		Metal	0.0225	0.0022	16	9
		Poly	0.0230	0.0033	14	13
		Wipes	0.0245	0.0059	5	3

TABLE 5(b). ESTIMATION OF TRU PROPORTION FROM BOTH SAMPLE SETS FOUR OUTLIER OBSERVATIONS OMITTED

Method	Type Cens	Waste Comp	Estimate	Std Err	n	# Cens
Normal $\hat{\mu}_1$	One-sided	Combined	0.0089	0.0044	58	46
		Cell	0.0066	0.0164	7	6
		Glass	0.0134	0.0005	17	16
		Metal	0.0199	0.0033	15	8
		Poly	0.0032	0.0151	14	13
		Wipes	0.0220	0.0141	5	3
Lognormal $\hat{\mu}_2$	One-sided	Combined	0.0121	0.0020	58	46
		Cell	0.0110	0.0078	7	6
		Glass	0.0134	0.0005	17	16
		Metal	0.0186	0.0025	15	8
		Poly	0.0061	0.0057	14	13
		Wipes	0.0240	0.0085	5	3
Normal $\hat{\mu}_3$	Interval	Combined	0.0270	0.0024	58	46
		Cell	0.0214	0.0029	7	6
		Glass	0.0265	0.0067	17	16
		Metal	0.0261	0.0025	15	8
		Poly	0.0298	0.0035	14	13
		Wipes	0.0313	0.0061	5	3
Lognormal $\hat{\mu}_4$	Interval	Combined	0.0227	0.0018	58	46
		Cell	0.0202	0.0029	7	6
		Glass	0.0177	0.0036	17	16
		Metal	0.0242	0.0025	15	8
		Poly	0.0264	0.0037	14	13
		Wipes	0.0289	0.0051	5	3
Kaplan-Meier $\hat{\mu}_5$	One-sided	Combined	0.0168	0.0014	58	46
		Cell	0.0294	.	7	6
		Glass	0.0140	.	17	16
		Metal	0.0200	0.0030	15	8
		Poly	0.0257	.	14	13
		Wipes	0.0348	0.0069	5	3
Normal Sample Mean (Upper bound) $\bar{\mu}_6$	Ignored	Combined	0.0309	0.0024	58	46
		Cell	0.0252	0.0033	7	6
		Glass	0.0313	0.0070	17	16
		Metal	0.0290	0.0029	15	8
		Poly	0.0339	0.0039	14	13
		Wipes	0.0343	0.0058	5	3
Normal Sample Mean (Lower bound) $\underline{\mu}_6$	Set to 0	Combined	0.0232	0.0024	58	46
		Cell	0.0174	0.0035	7	6
		Glass	0.0217	0.0069	17	16
		Metal	0.0236	0.0024	15	8
		Poly	0.0260	0.0034	14	13
		Wipes	0.0278	0.0076	5	3
Lognormal Sample Mean (Upper bound) $\bar{\mu}_7$	Ignored	Combined	0.0267	0.0019	58	46
		Cell	0.0240	0.0030	7	6
		Glass	0.0239	0.0039	17	16
		Metal	0.0267	0.0030	15	8
		Poly	0.0302	0.0045	14	13
		Wipes	0.0327	0.0048	5	3
Lognormal Sample Mean (Lower bound) $\underline{\mu}_7$	Set to 0	Combined	0.0180	0.0017	58	46
		Cell	0.0152	0.0032	7	6
		Glass	0.0120	0.0030	17	16
		Metal	0.0220	0.0022	15	8
		Poly	0.0230	0.0033	14	13
		Wipes	0.0245	0.0059	5	3

the number of samples (or observations); and the number of censored (or nondetect) observations. Taking into account that the logarithmic transform makes the distribution of the observations closer to the normal distribution, we *recommend* the use of $\hat{\mu}_4$ for further studies to estimate μ and to control (validate) the results with $\underline{\mu}_7$ and $\bar{\mu}_7$. The most obvious conclusion from a review of Tables 3 - 5 is that the estimates of the mean proportion TRU are systematically less in sample set 2 than in sample set 1. The estimates (Table 5) based on the combined information from both data sets neglect this fact and are recommended to be used only at preliminary stages of similar studies, for instance, when the sample sizes are relatively small (15 - 20 cases). Note, that the gross alpha activity was generally lower for set 1 than for set 2, leading to distinctly different estimates of PTRU. Thus, whenever there exists a possibility to stratify observations in accordance with the observed gross alpha activity, which is easy to measure, we *recommend* doing this stratification. The analysis of residuals from the estimates reported above leads to the recommendation of the estimate $\hat{\mu}_4$, which is the maximum likelihood estimate for the interval censored observations constructed under the assumption that the logarithms of observed values are normally distributed. The simultaneous use of estimates $\underline{\mu}_7$ and $\bar{\mu}_7$ gives an opportunity to evaluate the uncertainty imposed by the presence of nondetects. Their computation is strongly recommended when the fraction of nondetects is high, say greater than 10%.

5.2. Regression analysis

To analyze the tendency of the proportion TRU to decrease when the gross alpha activity increases, we regressed the proportion TRU (PTRU) activity on the gross alpha activity (GA) (both log transformed) for both sample sets individually and combined. The linear regression model for this is

$$y_{\alpha i} = \beta_{\alpha 0} + \beta_{\alpha 1} x_{\alpha i} + \varepsilon_{\alpha i} \tag{10}$$

and the quadratic regression model is given by

$$y_{\alpha i} = \beta_{\alpha 0} + \beta_{\alpha 1} x_{\alpha i} + \beta_{\alpha 2} x_{\alpha i}^2 + \varepsilon_{\alpha i}, \tag{11}$$

- 22 -

where $y_{\alpha i} = \ln \frac{TRU_{\alpha i}}{GA_{\alpha i}}$ may be considered as hypothetical observations with all nondetects replaced by actual (unfortunately frequently unknown) values, $x_{\alpha i} = \ln GA_{\alpha i}$, $GA_{\alpha i}$ is the mean gross alpha activity, for sample i (each sample was measured twice for gross alpha activity), α indicates the sample set number, β's are model parameters to be estimated; and $\varepsilon_{\alpha i}$ is the random noise component for observation $y_{\alpha i}$ with mean 0 ($i = 1, 2, \ldots, n_\alpha$ and $\alpha = 1, 2$). We did not construct different regression models for glass, metal, etc., for two reasons. First, the previous analysis did not reveal any statistically significant difference between TRU or PTRU for various materials. Second, we do not have the luxury to partition the data sets into smaller pieces because of the limited sample size, large number of nondetects within some waste types, and the additional parameters to be estimated.

Of course, both (10) and (11) are alternatives to the previously mentioned model (4) in Section 4, which in the current notation may be presented as

$$y_{\alpha i} = \beta_{\alpha 0} + \varepsilon_{\alpha i}, \ \ \beta_{\alpha 0} = \ln f. \tag{12}$$

Table 6 contains the regression results for the linear and quadratic models. Because not all $y_{\alpha i}$ are known, we performed analysis using the lower and upper bounds for TRU (compare with (5) and (6) of Section 5.1). Correspondingly, Table 6 contains two sets of parameter estimates for every considered case, one for the linear and one for the quadratic model. Similar to (8) of Section 5.1, we may assert that

$$\underline{\beta}_{\alpha 0} + \underline{\beta}_{\alpha 1} x_{\alpha i} \leq \hat{\beta}_{\alpha 0} + \hat{\beta}_{\alpha 1} x_{\alpha i} \leq \overline{\beta}_{\alpha 0} + \overline{\beta}_{\alpha 1} x_{\alpha i} \tag{13}$$

for all $x_{\alpha i}$. In the above expression, the underlined letters correspond to parameter estimates based on the lower bound observations and the letters with upper bars to the upper bound; compare with (7). The extension of (13) for the quadratic case is obvious.

What can be concluded from Table 6? Let us start with the quadratic case. The "rule of thumb", assuming normality, for determining significance of the parameters is that the ratio of the parameter's estimate (absolute value) to its standard error is

Table 6: Parameter Estimates from Regression of n (Proportion TRU) on n (Gross Alpha)

Exp	Data Set		Linear Model Parameters					Quadratic Model Parameters						
			EOE+	Intercept		Linear		EOE+	Intercept		Linear		Quadratic	
				Est.	Std Err	Est.	Std Err		Est.	Std Err	Est.	Std Err	Est.	Std Err
1	Upper	Reduced	0.4489	-1.2328	0.6263	-0.1956	0.0559	0.4262	5.7681	3.4356	-1.4099	0.5894	0.0517	0.0250
		Complete	0.5899	-2.6950	0.5261	-0.0604	0.0446	0.5488	2.7782	2.2909	-0.9030	0.3471	0.0308	0.0126
	Lower	Reduced	0.5549	-1.5337	0.7742	-0.1958	0.0691	0.5182	8.0258	4.1769	-1.8540	0.7165	0.0707	0.0304
		Complete	0.8109	-2.0396	0.7232	-0.1482	0.0613	0.8201	-0.2482	3.4235	-0.4240	0.5186	0.0101	0.0188
2	Upper	Reduced	0.3498	-1.2501	0.8683	-0.1910	0.0628	0.3553	-5.2986	7.9860	0.3938	1.1483	-0.0210	0.0411
		Complete	0.4135	-3.2484	0.7029	-0.0432	0.0495	0.3860	6.7486	4.5903	-1.3912	0.6143	0.0448	0.0203
	Lower	Reduced	0.6294	-0.2306	1.5626	-0.3022	0.1129	0.6179	18.8375	13.8883	-3.0566	1.9969	0.0988	0.0716
		Complete	0.7001	-3.1268	1.1902	-0.0880	0.0838	0.6239	17.3006	7.4204	-2.8424	0.9931	0.0915	0.0329
1 + 2	Upper*	Reduced	0.4007	-1.3296	0.3548	-0.1860	0.0285	0.3989	1.3083	2.1738	-0.6222	0.3558	0.0176	0.0143
		Complete	0.5282	-2.4301	0.3683	-0.0922	0.0285	0.4762	3.0792	1.4686	-0.9326	0.2197	0.0310	0.0080
	Lower*	Reduced	0.5828	-1.0077	0.5159	-0.2445	0.0414	0.5812	2.5572	3.1671	-0.8339	0.5183	0.0238	0.0209
		Complete	0.7619	-1.9585	0.5312	-0.1630	0.0411	0.7437	2.4939	2.2938	-0.8421	0.3432	0.0250	0.0126

* See page 26.
+ EOE = estimated observational error = square root of mean square error.

greater than 2 (in this case, the probability we will reject a parameter whose true value is zero is approximately 0.05). More than half of the cases in the last column of Table 6 satisfy this rule, implying that the quadratic coefficient is significant. However, we are very reluctant to recommend the use of quadratic models in the future for the reasons given in the following two paragraphs.

First, the presence of nondetects and the subsequent use of "upper bounds" and "lower bounds" makes the assumption about normality theoretically unacceptable, even for a relatively large number of observations, a case when normality is considered frequently as a corollary of the Central Limit Theorem (see, for instance, [19]). For a smaller number of observations, the assumption about normality of the underlying distribution is even less tenable because other types of distributions correspond better with our situation (see Appendix 3).

Second, the estimates of the quadratic coefficients are sensitive to the deleting of "leverage points", which were mentioned in Section 4 (see the last column of Table 6). The nonstability of the estimates indicates that the significance of the corresponding parameters may be spurious and induced by unknown factors, which have caused those two points at each of the sample sets to be far away from the main data clouds.

The linear model (10) is obviously a more reliable choice, and we recommend its use, but with some reservations: the practitioner has to remember that the standard regression analysis theory is not applicable to the full extent and the above remarks (for instance, about normality) are still valid here.

From all the linear models included in Table 6, it follows that the proportion of TRU is decreasing as the level of gross alpha activity increases. This tendency, being statistically significant, does not however change the proportion TRU too much. For instance, for the data sets with deleted outliers the slopes are in the range [-0.30, -0.19] in log-log space. Figures 7 - 10 give a visual presentation of the regression models, which were discussed above. For instance, one can see that the change in the logarithm of the proportion of TRU per unit change in the log scale of the gross alpha (i.e., the slope of the regression lines) is several times less than the estimated observational error (EOE). Actually, the corresponding ratios of the EOE to the estimated slope vary from 4.7 to

9.8; see Table 6, linear model results. We did not test the homogeneity of variance of the transformed observations because the number of observations was not sufficient for any statistically reliable inference. All the results were derived under the assumption that this variance is constant. It is well known that the introduction of weights to address non-homogeneity do not change the parameter estimates significantly and mainly lead to more realistic inferences about their standard errors.

We recommend use of the results of the regression analysis with the removed outliers. It is especially important for the second data set, in which both outliers correspond to leached items rather than smears (they are marked in Table A2 as LN-GL and LN-ML). The observed proportion TRU is in excellent agreement with the matching smears (LN-1, LN-11 and LN-7, LN-15 correspondingly) and can be used in constructing mean proportion TRU (i.e., estimates reported in Tables 3-4). However, the gross alpha readings for those two cases are not compatible with the corresponding readings for smears. Therefore, it can lead to a significant distortion of the estimate of the slope. This fact can be easily verified with the help of Fig. 8a and 8b. Following the above discussion, we recommend the linear models which are marked by asterisks in Table 6 and also presented in Fig. 9 (c and d) and Fig. 10 (a and b). In this table "reduced" refers to data sets with outlier observations deleted, i.e., two from set 1 and two from set 2.

5.3. Application to NDA

In order to apply the regression models to NDA, it is necessary that we convert $x =$ "gross alpha activities" to $Z =$ "total concentration for alpha emitters". This conversion can be made with the help of the proper calibration experiments. However, we have only one sample (LN-GL) where both variables, X and Z, are available which are matched with LN-1 and LN-11. Table A3 summarizes the results from the leaching sample LN-GL. In particular $Z \cong 4.0 \times 10^4 nCi/g$ which corresponds to the mean activity from two smears, which equals $1.1 \times 10^6 Bq$. From these results, the conversion factor can be roughly estimated as $K = 25.2$. Bounds on this factor may be obtained by using bounds on the estimated low and high end weights. Replacing x by $K \times Z$,

i.e., using the model

$$\ln{(PT\hat{R}U)} = \hat{\beta}_0 + \hat{\beta}_1 \ln{(K \times Z)}, \tag{14}$$

where the parameters $\hat{\beta}_0$ and $\hat{\beta}_1$ come from Table 6, we can estimate the proportion TRU knowing the total concentration for alpha emitters. The uncertainty of this equation can be partially evaluated by using the above bounds on K.

In order to make the conversion results statistically valid, we must have results from additional leaching experiments matched with smears. Having the corresponding results, one can construct a better estimate for the conversion factor. Of course, when the number of pairs "leaching - smearing" is large enough (say 10-15) then direct construction of model PTRU = F(Z) is possible. However, the cost of leaching is substantially higher than the cost of the smear analysis. In particular, this fact forced us to proceed in this work with the samples based on smearing. Separate conversions must be determined for each waste type (glass, metal, etc.) because of the different densities across waste types.

5.4. Comments on proportion TRU trend

Discussions with REDC operations personnel revealed that the trend found above may be valid from a physical point of view. One of the possible explanations is the following one.

The REDC processing campaign aims to not only separate the actinides from fission products, but to also partition the actinides from each other. The targets are made of predominantly Cm and are still predominantly Cm after irradiation in the HFIR. The calculations on the HFIR targets showed that, as of 3/5/96, the targets contained a total of approximately 0.13 grams of Pu (0.32 Ci) and 0.08 grams of Am (0.016 Ci) as compared to the 64.6 grams of Cm ($\sim$ 2300 Ci). This relatively large mass of Cm is the reason that all of the waste coming from the REDC cell bank is heavily contaminated with Cm244. One of the first steps in the process is to remove the rare earth fission products from the actinide products. Once this has been sufficiently accomplished, the actinides themselves are separated from each other. At this point in the process, there are different streams going into their respective tanks for further

- 27 -

processing. For example, the Cm product (most abundant and highest activity) will be relatively free of Pu, Am, Cf, Es, etc. after this initial processing. Any materials coming into contact with this Cm product (sample bottles, transfer lines, etc.) will be predominately contaminated with Cm, as there are only trace levels of other actinides as impurities in the product. Noting the total activities of Am, Pu and Cm at target discharge, if the product volumes are equal, then waste from the Cm stream will be much higher in total activity than waste from the Pu stream. Since this work has shown Am and Pu to be the greatest contributors to the TRU activity, it is not surprising that their relative percentages decrease when the activity (and hence Cm) increases. On the other hand, materials that have been in contact with the other product streams (Am, Pu) would be expected not to be as high in total activity, yet have a higher TRU percentage than a Cm product stream.

6. Summary

The primary objective of this work was to obtain an understanding and a proper statistical description of the elemental and isotopic structure of solid waste generated at REDC, for the ultimate purpose of classifying waste as TRU or not TRU. This was achieved through an investigation of the isotopic profile of alpha-emitting radionuclides. Prior to this study, we anticipated some stability in the isotopic profiles for various waste types or items. Our goal was to verify this and to estimate the corresponding profile, by measuring smears and leached samples. All of the statistical conclusions and inferences along with associated recommendations presented in this report are based on the assumed representativeness of the available data used.

Models and methods. Before analyzing the data, the main sources of uncertainty contributing to the variability in the observed data were identified and they included:

i. variability from one campaign to another (in this study, we were restricted to only Campaign 69);

ii. variability across different items or materials (e.g., plastic, glass, etc);

iii. variability in the selection of the item to be sampled or measured;

iv. randomness associated with the smearing process; and

- 28 -

v. randomness in the observation error for the various analytical techniques.

The choice of the appropriate underlying stochastic models for the observed activities were evaluated for a number of intuitively appealing sets of physical assumptions. All of them were proven to lead to the same type estimator of the isotopic proportion. The form of the statistically optimal estimator was specified as a ratio of means rather than a mean of ratios, which was used in this study because of the form the original data were provided. The derived maximum likelihood estimators provide efficient estimation of the proportion TRU and are recommended for future work. The results of the estimation problem using different physical assumptions are summarized in Appendix 3.

To address properly the randomness of the observational errors in analytical measurements, we analyzed the Poisson regression model in Appendix 2 and showed that the currently-used method of estimating individual isotopic activities is a simplified version of the exact, and statistically more efficient, maximum likelihood estimator. The latter leads to iterative regression methods and the corresponding numerical algorithms which are reported in Appendix 2 are recommended for future work.

Experimental results and analysis. The available data sets and, in particular, the data from set 1, contain a large number of cases where at least one of the TRU components was below the detection level, primarily CM246. The second sample achieved a higher resolution for the plutonium isotopes through chemical separation. The presence of the "less than" observations (or nondetects) necessitated the development of an approach, which includes several different statistical estimators, to find statistically reliable estimates of the proportion TRU (see Sections 4 and 5).

If the data from the two sample sets used in this study can be assumed to be representative samples for the campaign under analysis, then the results of the statistical analysis have shown the following:

i. No statistically significant difference in the proportion TRU across the different waste types (glass, metal, etc.) was detected, i.e., the variability from one waste type to another was comparable to the sampling variability within a given waste type. However, minimal data were available to make possible the statistical comparison for some of the

waste types.

ii. The hypothesis about the stability of the proportion TRU as a function of the sample gross alpha activity was rejected. Using regression analysis, a linear trend was revealed that showed the log proportion TRU to decrease as the log gross alpha activity increased. The reader should remember that statistical significance does not coincide generally with practical importance (see Section 5.2).

iii. The estimated mean proportion TRU ranged approximately from 0.01 to 0.05 within the range of the gross alpha activity which characterized the waste in the two sample sets.

iv. Cm244 was the dominant alpha-emitting radionuclide with more than 93% of the samples from the combined sample sets having a proportion Cm244 greater than 0.90.

v. Three choices of the detection limits for the plutonium isotopes (0.05%, 0.5% and 1% of gross alpha) did not reveal any practical discrepancy in the results, i.e., all conclusions remained unaffected. A graphical confirmation of that fact can be found from an examination and comparison of Figs. 10b, c, and d.

Although the trend in the proportion TRU as a function of the gross alpha activity mentioned above was based on the sample activity measurements from smear samples, when this result is used in the NDA, additional calibration of this dependency is needed using direct concentration rather than activity measurements. These concentration measurements are available only through leaching techniques, which were used with only one sample unit in this work. Hence, our recommendation is that at least several (> 4) leaching tests be run for every type of material preferably around the official 100 nCi/g threshold used for deciding whether the waste under consideration is TRU or not.

Non TRU radionuclide. The software and methodology developed in this work may be used for the analysis of any radionuclide listed in Tables A1 and A2, including beta-gamma emitters. Of course, we have to be very cautious with any formal statistical inferences when the number of nondetects is extremely high. Analysis and results for Cf252 have been performed for this project and are available upon request. They

are not reported in this publication because the results would double the size of this report. We note that for Cf252 all tendencies are very similar to those found for TRU. For instance, the proportion Cf252 decreases with increase of the total alpha activity, and this dependence is even more pronounced than in the TRU case. For beta-gamma emitters, the analysis of their correlation with TRU radionuclides can help in NDA. The corresponding results will be reported in [21].

Perspectives to complement NDA procedures. In spite of the presence of the above-mentioned linear trend, we can make statements about the average proportion TRU which may be applied to the entire range of sampled activity assuming a realistic censored underlying distribution for the observed proportions. From both the regression analysis and the estimates obtained from the censored distribution analysis, we conclude that the proportion TRU ranges from approximately 0.005 - 0.06. Under rather restrictive assumptions (e.g., log normality of the observations), this interval may be considered as a conservative bound on the 95% confidence interval for the mean proportion TRU, i.e., the actual confidence interval may fall within this interval. If additional leaching tests are performed, then the length of the confidence interval would be reduced by taking the linear trend into account. The previously mentioned interval will be replaced by 2 to 3 times the smaller interval: $\hat{PTRU} \pm 0.01$. The latter interval is a very simplistic approximation of what can be derived from the regression models constructed in this work after proper calibration in the nCi/g scale. This interval is crucial for the current NDA technology which does not allow for the reliable separation of activities for individual isotopes dominated by Cm244 and Cf252.

In this paper, we developed and applied a statistical approach to analyzing the relationship between proportion TRU and gross alpha activity. Obviously the analysis of other pairwise relationships (for instance, between Cf252 and gross alpha activity, proportion TRU and some beta/gamma emitters) may be accomplished within this framework. The main objective of those analyses would be to reveal statistically significant dependencies between TRU components and isotopes (or group of isotopes) whose concentrations can be relatively easily measured with NDA techniques.

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Appendix 1. Data

TABLE A1

THE 1st SET OF OBSERVATIONS (RH SERIES, 34 SAMPLES)

Analysis		RH9AA	RH9EE	RH9KK	RH9LL	RH9QQ	RH9SS
Gross Alpha	(Bq/total)	9.1E+03 +/- 1.9E+02	6.2E+05 +/- 7.4E+03	5.8E+04 +/- 1.0E+03	1.2E+04 +/- 2.5E+02	1.9E+05 +/- 2.5E+03	6.1E+04 +/- 1.1E+03
Alpha emitters							
^{244}Cm	(Bq/total)	8.3E+03 +/- 8.3E+02	6.0E+05 +/- 6.0E+04	5.2E+04 +/- 5.2E+03	1.1E+04 +/- 1.1E+03	1.8E+05 +/- 1.8E+04	5.8E+04 +/- 5.8E+03
^{238}Pu	(Bq/total)	1.7E+02 +/- 3.4E+01	< 6.2E+03	8.5E+02 +/- 1.7E+02	< 1.2E+02	< 1.9E+03	1.1E+02 +/- 2.2E+01
$^{239/240}$Pu	(Bq/total)	< 9.1E+01	6.2E+02 +/- 1.2E+02	5.8E+01 +/- 1.2E+01	< 1.2E+02	5.7E+02 +/- 1.1E+02	1.2E+02 +/- 2.4E+01
^{241}Am	(Bq/total)	1.7E+02 +/- 5.4E+00	2.8E+03 +/- 2.2E+01	2.5E+02 +/- 6.7E+00	8.5E+02 +/- 1.5E+01	5.6E+03 +/- 2.8E+01	6.2E+02 +/- 1.1E+01
^{243}Am	(Bq/total)	1.1E+01 +/- 2.0E+00	2.1E+02 +/- 6.9E+00	1.1E+01 +/- 2.3E+00	4.2E+01 +/- 4.7E+00	1.2E+02 +/- 8.3E+00	3.6E+01 +/- 3.8E+00
^{252}Cf	(Bq/total)	2.3E+02 +/- 4.6E+01	3.1E+03 +/- 6.2E+02	2.7E+03 +/- 5.5E+02	1.8E+02 +/- 3.6E+01	3.4E+03 +/- 6.8E+02	1.2E+03 +/- 2.3E+02
^{242}Cm	(Bq/total)	1.9E+02 +/- 3.8E+01	1.6E+04 +/- 3.2E+03	2.2E+03 +/- 4.4E+02	1.2E+02 +/- 2.4E+01	4.0E+03 +/- 8.0E+02	7.3E+02 +/- 1.5E+02
^{244}Cm	(Bq/total)	< 9.1E+01	6.2E+02 +/- 1.2E+02	< 5.8E+02	< 1.2E+02	< 1.9E+03	< 6.1E+02
Beta/gamma emitters							
^{60}Co	(Bq/total)	< 7.4E+00	< 2.1E+01	< 5.8E+00	< 2.6E+01	< 2.4E+01	< 1.5E+01
^{106}Ru	(Bq/total)	1.1E+02 +/- 2.5E+01	1.1E+03 +/- 6.8E+01	1.8E+02 +/- 2.5E+01	4.7E+03 +/- 1.0E+02	9.8E+02 +/- 9.0E+01	5.1E+02 +/- 4.7E+01
^{110m}Ag	(Bq/total)	< 5.7E+00	1.2E+02 +/- 4.9E+00	< 6.2E+00	1.2E+02 +/- 5.3E+00	1.1E+02 +/- 5.2E+00	< 1.0E+01
^{125}Sb	(Bq/total)	< 2.0E+01	4.7E+02 +/- 1.7E+01	1.2E+02 +/- 6.3E+00	8.6E+02 +/- 1.8E+01	3.5E+02 +/- 1.6E+01	3.2E+02 +/- 1.2E+01
^{134}Cs	(Bq/total)	1.6E+01 +/- 1.6E+00	1.9E+02 +/- 5.2E+00	1.9E+01 +/- 1.8E+00	1.8E+02 +/- 5.8E+00	1.4E+02 +/- 5.7E+00	1.1E+02 +/- 3.9E+00
^{137}Cs	(Bq/total)	9.8E+01 +/- 4.9E+00	1.9E+03 +/- 1.7E+01	1.8E+02 +/- 5.5E+00	1.6E+03 +/- 1.6E+01	1.3E+03 +/- 1.5E+01	7.0E+02 +/- 1.2E+01
^{144}Ce	(Bq/total)	2.2E+02 +/- 1.4E+01	2.6E+03 +/- 4.3E+01	1.3E+02 +/- 1.2E+01	1.8E+03 +/- 3.7E+01	2.0E+03 +/- 4.0E+01	1.7E+03 +/- 3.2E+01
^{152}Eu	(Bq/total)	< 3.4E+01	3.1E+02 +/- 1.1E+01	< 2.7E+01	1.6E+02 +/- 7.6E+00	1.5E+02 +/- 4.5E+01	2.8E+02 +/- 7.2E+00
^{154}Eu	(Bq/total)	1.5E+02 +/- 4.0E+00	1.3E+03 +/- 1.1E+01	1.1E+02 +/- 3.1E+00	8.3E+02 +/- 9.0E+00	2.2E+03 +/- 1.4E+01	7.9E+02 +/- 8.2E+00
^{155}Eu	(Bq/total)	9.1E+01 +/- 3.8E+00	6.1E+02 +/- 6.1E+01	5.1E+01 +/- 3.6E+00	4.3E+02 +/- 1.0E+01	3.3E+03 +/- 1.8E+01	3.3E+02 +/- 8.2E+00
^{237}Np	(Bq/total)	1.1E+01 +/- 3.5E+00	5.3E+02 +/- 2.0E+01	< 1.5E+01	< 4.4E+01	3.2E+02 +/- 5.0E+01	1.0E+02 +/- 2.4E+01

Analysis		RH9D	RH9E	RH9F	RH9J	RH9M	RH9A
Gross Alpha	(Bq/total)	4.8E+04 +/- 1.2E+03	2.6E+04 +/- 4.9E+02	1.0E+05 +/- 1.9E+03	5.2E+04 +/- 1.1E+03	1.0E+05 +/- 1.6E+03	1.1E+04 +/- 2.9E+02
Alpha emitters							
^{244}Cm	(Bq/total)	4.6E+04 +/- 4.6E+03	2.4E+04 +/- 2.4E+03	9.7E+04 +/- 9.7E+03	4.2E+04 +/- 4.2E+03	9.3E+04 +/- 9.3E+03	1.0E+04 +/- 1.0E+03
^{238}Pu	(Bq/total)	< 4.8E+02	3.0E+02 +/- 6.0E+01	< 1.0E+03	2.7E+02 +/- 5.4E+01	1.0E+02 +/- 2.0E+01	1.3E+02 +/- 2.6E+01
$^{239/240}$Pu	(Bq/total)	< 4.8E+02	< 2.6E+02	5.0E+02 +/- 1.0E+02	< 5.2E+02	4.0E+02 +/- 8.0E+01	< 1.1E+02
^{241}Am	(Bq/total)	1.0E+03 +/- 1.8E+01	3.5E+02 +/- 9.0E+00	4.6E+02 +/- 2.3E+01	7.2E+02 +/- 1.9E+01	1.4E+03 +/- 2.2E+01	2.2E+02 +/- 6.3E+00
^{243}Am	(Bq/total)	2.8E+01 +/- 6.0E+00	3.2E+01 +/- 5.0E+00	2.4E+01 +/- 6.4E+00	2.9E+01 +/- 5.7E+00	8.9E+01 +/- 9.0E+00	5.1E+00 +/- 1.8E+00
^{252}Cf	(Bq/total)	8.8E+02 +/- 1.9E+02	7.2E+02 +/- 1.4E+02	8.1E+02 +/- 1.6E+02	7.1E+03 +/- 1.4E+03	3.2E+03 +/- 6.4E+02	2.8E+02 +/- 5.7E+01
^{242}Cm	(Bq/total)	8.0E+02 +/- 1.6E+02	1.7E+02 +/- 3.3E+01	5.9E+02 +/- 1.2E+02	2.2E+03 +/- 4.4E+02	2.0E+03 +/- 4.0E+02	1.7E+02 +/- 3.4E+01
^{246}Cm	(Bq/total)	< 4.8E+02	< 2.6E+02	4.0E+02 +/- 8.0E+01	< 5.2E+02	< 1.0E+03	< 1.1E+02
Beta/gamma emitters							
^{60}Co	(Bq/total)	3.0E+01 +/- 5.0E+00	< 1.4E+01	< 1.9E+01	< 2.1E+01	2.0E+01 +/- 5.0E+00	5.0E+00 +/- 2.3E+00
^{106}Ru	(Bq/total)	1.8E+03 +/- 8.5E+01	2.3E+02 +/- 4.1E+01	4.6E+02 +/- 8.6E+01	1.9E+03 +/- 1.1E+02	7.6E+02 +/- 9.0E+01	1.5E+02 +/- 2.8E+01
^{110m}Ag	(Bq/total)	< 1.4E+01	3.9E+01 +/- 3.7E+00	4.6E+02 +/- 9.0E+00	< 2.2E+01	< 2.3E+01	< 7.0E+00
^{125}Sb	(Bq/total)	9.7E+01 +/- 1.8E+01	9.9E+01 +/- 1.3E+01	< 5.6E+01	2.9E+02 +/- 2.6E+01	5.3E+02 +/- 2.6E+01	< 2.1E+01
^{134}Cs	(Bq/total)	8.4E+01 +/- 5.7E+00	4.1E+01 +/- 4.3E+00	9.9E+01 +/- 6.6E+00	2.0E+02 +/- 8.1E+00	2.1E+02 +/- 8.5E+00	1.7E+01 +/- 2.0E+00
^{137}Cs	(Bq/total)	5.7E+02 +/- 1.5E+01	6.2E+02 +/- 1.4E+01	1.2E+03 +/- 2.0E+01	1.8E+03 +/- 2.4E+01	2.5E+03 +/- 2.9E+01	1.9E+02 +/- 6.0E+00
^{144}Ce	(Bq/total)	5.2E+02 +/- 3.2E+01	3.5E+02 +/- 2.7E+01	8.9E+02 +/- 5.0E+01	1.2E+03 +/- 5.5E+01	1.9E+03 +/- 5.4E+01	2.1E+02 +/- 1.5E+01
^{152}Eu	(Bq/total)	< 7.4E+01	< 6.5E+01	< 5.3E+01	< 8.8E+01	4.6E+02 +/- 1.6E+01	< 3.6E+01
^{154}Eu	(Bq/total)	4.3E+02 +/- 1.0E+01	2.8E+02 +/- 8.1E+00	2.4E+02 +/- 1.1E+01	4.7E+02 +/- 1.4E+01	1.6E+03 +/- 1.6E+01	1.4E+02 +/- 3.6E+00
^{155}Eu	(Bq/total)	4.2E+02 +/- 1.1E+01	1.3E+02 +/- 7.4E+00	2.5E+02 +/- 1.4E+01	3.0E+02 +/- 1.6E+01	7.8E+02 +/- 1.7E+01	1.2E+02 +/- 4.1E+00
^{239}Np	(Bq/total)	< 5.2E+01	< 3.3E+01	9.9E+01 +/- 2.0E+01	< 5.2E+01	1.3E+02 +/- 3.4E+01	< 1.9E+01

Analysis		RH9C	RH9G	RH9H	RH9I	RH9K	RH9L
Gross Alpha	(Bq/total)	3.5E+04 +/- 1.0E+03	2.9E+04 +/- 5.2E+02	5.1E+04 +/- 1.2E+03	6.3E+04 +/- 1.4E+03	1.1E+05 +/- 2.0E+03	6.0E+04 +/- 1.4E+03
Alpha emitters							
^{244} Cm	(Bq/total)	3.3E+04 +/- 3.3E+03	2.8E+04 +/- 2.8E+03	4.9E+04 +/- 4.9E+03	6.0E+04 +/- 6.0E+03	1.0E+05 +/- 1.0E+04	5.5E+04 +/- 5.5E+03
^{238} Pu	(Bq/total)	4.2E+02 +/- 8.3E+01	8.0E+02 +/- 1.6E+02	2.9E+01 +/- 5.8E+00	3.5E+01 +/- 7.0E+00	6.8E+02 +/- 1.4E+02	5.0E+02 +/- 1.0E+02
$^{239/240}$ Pu	(Bq/total)	< 3.5E+02	2.6E+02 +/- 5.2E+01	< 5.1E+02	2.5E+02 +/- 5.0E+01	4.4E+02 +/- 8.8E+01	3.0E+02 +/- 6.0E+01
^{241} Am	(Bq/total)	7.4E+02 +/- 1.1E+01	1.6E+02 +/- 5.4E+00	9.4E+02 +/- 1.2E+01	9.1E+02 +/- 1.3E+01	1.3E+03 +/- 1.6E+01	8.2E+02 +/- 1.2E+01
^{243} Am	(Bq/total)	2.1E+01 +/- 2.6E+00	8.9E+00 +/- 2.4E+00	4.6E+01 +/- 3.7E+00	4.2E+01 +/- 3.7E+00	8.1E+01 +/- 6.2E+00	3.4E+01 +/- 4.4E+00
^{252} Cf	(Bq/total)	6.6E+02 +/- 1.3E+02	1.5E+02 +/- 3.1E+01	7.0E+02 +/- 1.4E+02	9.4E+02 +/- 1.9E+02	3.1E+03 +/- 6.1E+02	1.9E+03 +/- 3.8E+02
^{242} Cm	(Bq/total)	5.3E+02 +/- 1.1E+02	1.1E+02 +/- 2.2E+01	4.3E+02 +/- 8.5E+01	7.6E+02 +/- 1.5E+02	2.4E+03 +/- 4.9E+02	1.3E+03 +/- 2.6E+02
^{246} Cm	(Bq/total)	< 3.5E+02	< 2.9E+02	< 5.1E+02	< 6.3E+02	3.3E+02 +/- 6.6E+01	< 6.0E+02
Beta/gamma emitters							
^{60} Co	(Bq/total)	< 8.0E+00	8.7E+00 +/- 1.9E+00	1.2E+01 +/- 2.6E+00	2.2E+01 +/- 3.7E+00	3.5E+01 +/- 4.1E+00	< 1.1E+01
^{106} Ru	(Bq/total)	3.1E+02 +/- 3.9E+01	1.2E+02 +/- 2.7E+01	4.5E+02 +/- 4.7E+01	4.9E+02 +/- 5.1E+01	4.2E+02 +/- 6.0E+01	3.2E+02 +/- 5.3E+01
^{110m} Ag	(Bq/total)	< 1.1E+01	2.3E+01 +/- 2.1E+00	6.2E+01 +/- 3.2E+00	4.7E+01 +/- 3.3E+00	< 1.4E+01	< 1.2E+01
^{125} Sb	(Bq/total)	1.4E+02 +/- 9.0E+00	5.0E+01 +/- 7.6E+00	1.5E+02 +/- 1.0E+01	3.2E+02 +/- 1.2E+01	3.0E+02 +/- 1.5E+01	< 4.0E+01
^{134} Cs	(Bq/total)	3.3E+01 +/- 3.1E+00	1.9E+01 +/- 2.0E+00	4.9E+01 +/- 3.0E+00	8.9E+01 +/- 3.8E+00	1.3E+02 +/- 5.0E+00	5.8E+01 +/- 4.3E+00
^{137} Cs	(Bq/total)	3.7E+02 +/- 8.0E+00	1.7E+02 +/- 6.0E+00	5.0E+02 +/- 9.3E+00	9.9E+02 +/- 1.2E+01	1.4E+03 +/- 1.7E+01	6.5E+02 +/- 1.1E+01
^{144} Ce	(Bq/total)	5.2E+02 +/- 2.1E+01	1.0E+02 +/- 1.2E+01	4.0E+02 +/- 2.1E+01	5.5E+02 +/- 2.3E+01	2.6E+03 +/- 4.0E+01	1.3E+03 +/- 2.7E+01
^{152} Eu	(Bq/total)	8.5E+01 +/- 5.7E+00	< 3.4E+01	< 4.4E+01	8.5E+01 +/- 1.4E+01	5.9E+02 +/- 9.0E+00	2.8E+02 +/- 1.5E+01
^{154} Eu	(Bq/total)	4.0E+02 +/- 5.7E+00	1.8E+02 +/- 4.5E+00	6.5E+02 +/- 6.8E+00	5.9E+02 +/- 8.0E+00	1.8E+03 +/- 1.2E+01	8.8E+02 +/- 8.6E+00
^{155} Eu	(Bq/total)	3.7E+02 +/- 5.9E+00	1.1E+02 +/- 4.2E+00	4.5E+02 +/- 8.0E+00	3.7E+02 +/- 7.4E+00	8.4E+02 +/- 1.2E+01	4.1E+02 +/- 1.2E+01
^{239} Np	(Bq/total)	5.9E+01 +/- 1.2E+01	< 1.9E+01	< 9.0E+01	7.4E+01 +/- 1.6E+01	1.3E+02 +/- 1.9E+01	7.2E+01 +/- 1.8E+01

Analysis		RH9N	RH9P	RH9Q	RH9S	RH9T	RH9U
Gross Alpha	(Bq/total)	1.9E+05 +/- 2.3E+03	9.9E+04 +/- 1.9E+03	8.7E+04 +/- 1.7E+03	1.5E+05 +/- 2.4E+03	1.6E+04 +/- 3.5E+02	1.6E+04 +/- 3.5E+02

Alpha emitters

Analysis		RH9N	RH9P	RH9Q	RH9S	RH9T	RH9U
^{244}Cm	(Bq/total)	1.7E+05 +/- 1.7E+04	9.4E+04 +/- 9.4E+03	8.2E+04 +/- 8.2E+03	1.5E+05 +/- 1.5E+04	1.5E+04 +/- 1.5E+03	1.5E+04 +/- 1.5E+03
^{238}Pu	(Bq/total)	< 1.9E+03	< 9.9E+02	< 8.7E+02	< 1.5E+03	3.9E+02 +/- 7.9E+01	< 1.6E+02
$^{239/240}$Pu	(Bq/total)	1.1E+03 +/- 2.3E+02	4.0E+02 +/- 7.9E+01	< 8.7E+02	1.5E+02 +/- 3.0E+01	2.9E+02 +/- 5.8E+01	< 1.6E+02
^{241}Am	(Bq/total)	1.1E+04 +/- 5.4E+01	2.2E+03 +/- 2.6E+01	3.0E+03 +/- 2.8E+01	2.1E+03 +/- 1.9E+01	8.7E+01 +/- 1.9E+01	3.2E+02 +/- 6.7E+00
^{243}Am	(Bq/total)	1.0E+02 +/- 1.5E+01	9.6E+01 +/- 7.1E+00	4.3E+01 +/- 7.1E+00	3.2E+02 +/- 8.6E+00	7.5E+00 +/- 8.6E+00	8.0E+00 +/- 2.3E+00
^{252}Cf	(Bq/total)	2.9E+03 +/- 5.9E+02	1.2E+03 +/- 2.4E+02	8.4E+02 +/- 1.7E+02	3.9E+02 +/- 7.8E+01	2.0E+02 +/- 4.0E+01	3.2E+02 +/- 6.4E+01
^{242}Cm	(Bq/total)	2.4E+03 +/- 4.7E+02	8.0E+02 +/- 1.6E+02	1.6E+03 +/- 3.2E+02	2.1E+02 +/- 4.2E+01	1.5E+02 +/- 3.0E+01	2.9E+02 +/- 5.8E+01
^{246}Cm	(Bq/total)	< 1.9E+03	< 9.9E+02	< 8.7E+02	< 1.5E+03	1.6E+02 +/- 3.2E+01	< 1.6E+02

Beta/gamma emitters

Analysis		RH9N	RH9P	RH9Q	RH9S	RH9T	RH9U
^{60}Co	(Bq/total)	< 2.2E+01	2.7E+01 +/- 5.5E+00	< 1.4E+01	3.7E+01 +/- 3.8E+00	< 1.4E+01	< 1.0E+01
^{106}Ru	(Bq/total)	9.1E+02 +/- 1.1E+02	3.7E+02 +/- 9.7E+01	< 1.5E+02	3.1E+02 +/- 5.8E+01	< 1.0E+02	1.9E+02 +/- 2.8E+01
^{110m}Ag	(Bq/total)	< 2.7E+01	< 1.9E+01	< 1.7E+01	< 1.5E+01	5.0E+01 +/- 3.5E+00	< 7.0E+00
^{125}Sb	(Bq/total)	8.8E+02 +/- 3.1E+01	6.0E+02 +/- 2.3E+01	< 5.0E+01	1.9E+03 +/- 3.2E+01	< 3.0E+01	< 2.4E+01
^{134}Cs	(Bq/total)	1.7E+02 +/- 1.0E+01	1.0E+02 +/- 6.5E+00	2.8E+01 +/- 4.6E+00	4.4E+02 +/- 6.9E+00	< 1.0E+01	1.3E+01 +/- 2.0E+00
^{137}Cs	(Bq/total)	1.5E+03 +/- 2.4E+01	1.2E+03 +/- 2.0E+01	3.1E+02 +/- 1.1E+01	6.9E+03 +/- 3.3E+01	6.3E+01 +/- 5.5E+00	1.4E+02 +/- 5.1E+00
^{144}Ce	(Bq/total)	1.9E+03 +/- 6.0E+01	7.6E+02 +/- 4.1E+01	5.9E+02 +/- 3.2E+01	9.9E+01 +/- 2.4E+01	1.2E+02 +/- 1.9E+01	1.4E+02 +/- 1.4E+01
^{152}Eu	(Bq/total)	4.4E+02 +/- 1.6E+01	< 9.9E+01	< 1.6E+02	1.1E+02 +/- 2.2E+01	< 4.8E+01	< 3.6E+01
^{154}Eu	(Bq/total)	5.4E+03 +/- 2.7E+01	9.7E+02 +/- 1.4E+01	1.4E+03 +/- 1.5E+01	3.4E+03 +/- 1.7E+01	6.8E+01 +/- 4.1E+00	1.2E+02 +/- 4.0E+00
^{155}Eu	(Bq/total)	9.6E+03 +/- 4.0E+01	3.9E+02 +/- 2.0E+01	2.2E+03 +/- 1.8E+01	1.3E+03 +/- 2.0E+02	3.7E+01 +/- 4.9E+00	1.0E+02 +/- 3.9E+00
^{239}Np	(Bq/total)	5.2E+02 +/- 5.2E+01	1.3E+02 +/- 1.5E+01	1.6E+02 +/- 4.0E+01	3.5E+02 +/- 5.3E+01	< 2.0E+01	< 1.8E+01

Analysis		RH9V	RH9O	RH9R	RH11B	RH12B	RH3B
Gross Alpha	(Bq/total)	5.9E+04 +/- 5.9E+02	5.3E+03 +/- 1.3E+02	2.2E+04 +/- 4.8E+02	5.4E+04 +/- 1.2E+03	6.3E+04 +/- 1.4E+03	4.3E+05 +/- 5.6E+03
Alpha emitters							
^{244}Cm	(Bq/total)	5.4E+04 +/- 5.4E+03	3.4E+03 +/- 3.4E+02	2.1E+04 +/- 2.1E+03	5.4E+04 +/- 5.4E+03	6.2E+04 +/- 6.2E+03	4.3E+05 +/- 4.3E+04
^{238}Pu	(Bq/total)	3.6E+02 +/- 7.2E+01	4.6E+02 +/- 9.1E+01	2.4E+02 +/- 4.8E+01	< 5.4E+02	< 6.3E+02	< 4.3E+03
$^{239/240}$Pu	(Bq/total)	3.5E+02 +/- 7.1E+01	< 5.3E+01	< 2.2E+02	< 5.4E+02	< 6.3E+02	< 4.3E+03
^{241}Am	(Bq/total)	7.6E+02 +/- 1.7E+01	5.4E+01 +/- 5.7E+00	2.2E+02 +/- 6.3E+00	3.8E+02 +/- 7.3E+00	6.4E+02 +/- 9.3E+00	3.9E+03 +/- 2.1E+01
^{243}Am	(Bq/total)	6.9E+01 +/- 7.2E+00	4.0E+00 +/- 2.8E+00	1.0E+01 +/- 2.4E+00	7.3E+00 +/- 2.3E+00	2.6E+01 +/- 2.7E+00	1.0E+02 +/- 3.4E+00
^{252}Cf	(Bq/total)	2.1E+03 +/- 4.2E+02	1.4E+03 +/- 2.8E+02	3.9E+02 +/- 7.8E+01	5.2E+01 +/- 1.0E+01	7.5E+01 +/- 1.5E+01	< 4.3E+03
^{242}Cm	(Bq/total)	9.5E+02 +/- 1.9E+02	< 5.3E+01	3.4E+02 +/- 6.7E+01	5.6E+01 +/- 1.1E+01	1.1E+02 +/- 2.3E+01	< 4.3E+03
^{246}Cm	(Bq/total)	1.8E+02 +/- 3.5E+01	< 5.3E+01	< 2.2E+02	< 5.4E+02	< 6.3E+02	< 4.3E+03
Beta/gamma emitters							
^{60}Co	(Bq/total)	1.1E+01 +/- 5.1E+00	< 1.0E+01	< 1.3E+01	< 8.0E+00	< 1.0E+01	< 7.0E+00
^{106}Ru	(Bq/total)	5.7E+02 +/- 8.2E+01	1.3E+03 +/- 7.1E+01	1.6E+02 +/- 3.7E+01	< 4.5E+01	< 5.6E+01	< 5.0E+01
^{110m}Ag	(Bq/total)	2.6E+01 +/- 4.7E+00	1.1E+03 +/- 9.3E+00	< 8.4E+00	< 6.0E+00	< 6.0E+00	< 5.0E+00
^{125}Sb	(Bq/total)	2.2E+02 +/- 1.8E+01	1.9E+02 +/- 1.3E+01	9.4E+01 +/- 8.2E+00	< 1.5E+01	< 1.6E+01	< 1.3E+01
^{134}Cs	(Bq/total)	1.3E+02 +/- 6.6E+00	< 1.2E+01	4.0E+01 +/- 2.7E+00	< 5.0E+00	< 6.0E+00	< 5.0E+00
^{137}Cs	(Bq/total)	8.5E+02 +/- 1.7E+01	9.9E+01 +/- 6.9E+00	4.0E+02 +/- 8.1E+00	3.1E+01 +/- 3.5E+00	4.9E+01 +/- 3.6E+00	2.1E+01 +/- 3.8E+00
^{144}Ce	(Bq/total)	1.3E+03 +/- 4.6E+01	1.5E+02 +/- 1.9E+01	2.5E+02 +/- 1.5E+01	3.8E+01 +/- 7.5E+00	6.4E+01 +/- 1.2E+01	4.7E+01 +/- 1.1E+01
^{152}Eu	(Bq/total)	1.4E+02 +/- 9.0E+00	< 3.4E+01	5.3E+01 +/- 3.7E+00	< 2.7E+01	< 2.7E+01	< 2.8E+01
^{154}Eu	(Bq/total)	7.3E+02 +/- 1.1E+01	< 3.7E+01	1.9E+02 +/- 4.3E+00	1.0E+02 +/- 3.4E+00	9.7E+01 +/- 3.7E+00	4.8E+01 +/- 2.9E+00
^{155}Eu	(Bq/total)	3.6E+02 +/- 1.2E+01	3.3E+01 +/- 5.1E+00	1.1E+02 +/- 4.7E+00	1.3E+02 +/- 4.5E+00	1.4E+02 +/- 4.6E+00	6.6E+01 +/- 7.0E+00
^{239}Np	(Bq/total)	1.2E+02 +/- 2.6E+01	< 1.8E+01	< 2.0E+01	4.3E+01 +/- 6.6E+00	5.4E+01 +/- 5.9E+00	2.3E+02 +/- 8.7E+00

Analysis		RH4D	RH6C	RH7D	RII8A
Gross Alpha	(Bq/total)	8.1E+07 +/- 1.1E+06	3.0E+06 +/- 3.0E+04	2.7E+06 +/- 3.5E+04	2.4E+08 +/- 3.1E+06
Alpha emitters					
^{244} Cm	(Bq/total)	6.9E+07 +/- 6.9E+06	2.9E+06 +/- 2.9E+05	2.5E+06 +/- 2.5E+05	2.4E+08 +/- 2.4E+07
^{238} Pu	(Bq/total)	< 8.1E+05	< 3.0E+04	< 2.7E+04	< 2.4E+06
$^{239/240}$ Pu	(Bq/total)	8.1E+04 +/- 1.6E+04	< 3.0E+04	5.4E+03 +/- 1.1E+03	< 2.4E+06
^{241} Am	(Bq/total)	1.1E+07 +/- 1.0E+04	8.8E+04 +/- 1.1E+02	2.3E+04 +/- 1.1E+02	2.1E+05 +/- 5.2E+02
^{243} Am	(Bq/total)	6.8E+04 +/- 2.6E+03	1.7E+03 +/- 1.7E+01	1.4E+03 +/- 2.8E+01	2.9E+04 +/- 2.0E+02
^{252} Cf	(Bq/total)	1.6E+05 +/- 3.3E+04	1.6E+04 +/- 3.3E+03	6.2E+04 +/- 1.2E+04	6.4E+05 +/- 1.3E+05
^{242} Cm	(Bq/total)	1.3E+06 +/- 2.6E+05	2.0E+04 +/- 3.9E+03	8.9E+04 +/- 1.8E+04	3.2E+05 +/- 6.5E+04
^{246} Cm	(Bq/total)	< 8.1E+05	< 3.0E+04	1.1E+04 +/- 2.2E+03	< 2.4E+06
Beta/gamma emitters					
^{60} Co	(Bq/total)	< 1.5E+03	2.7E+02 +/- 1.3E+01	1.4E+03 +/- 3.0E+01	1.4E+03 +/- 9.5E+01
^{106} Ru	(Bq/total)	< 2.9E+04	1.8E+04 +/- 2.8E+02	3.7E+04 +/- 5.4E+02	3.4E+05 +/- 2.3E+03
^{110m} Ag	(Bq/total)	1.2E+04 +/- 9.7E+02	2.9E+03 +/- 2.1E+01	9.7E+02 +/- 3.1E+01	4.1E+03 +/- 1.3E+02
^{125} Sb	(Bq/total)	< 8.1E+03 +/-	4.5E+04 +/- 2.3E+03	5.0E+03 +/- 5.0E+02	1.5E+04 +/- 6.9E+02
^{134} Cs	(Bq/total)	< 2.9E+03 +/-	2.1E+03 +/- 9.7E+01	1.1E+04 +/- 5.3E+01	8.4E+03 +/- 1.6E+02
^{137} Cs	(Bq/total)	3.1E+04 +/- 1.5E+03	2.0E+04 +/- 5.8E+01	8.3E+04 +/- 1.8E+02	5.3E+04 +/- 2.9E+02
^{144} Ce	(Bq/total)	4.1E+05 +/- 7.2E+03	1.1E+04 +/- 1.3E+02	4.2E+04 +/- 3.0E+02	8.3E+04 +/- 1.3E+03
^{152} Eu	(Bq/total)	4.1E+04 +/- 1.4E+03	4.3E+03 +/- 1.2E+02	1.2E+03 +/- 3.0E+02	8.5E+03 +/- 2.4E+02
^{154} Eu	(Bq/total)	3.9E+06 +/- 1.3E+04	1.8E+04 +/- 4.1E+01	1.5E+04 +/- 7.5E+01	1.2E+05 +/- 3.6E+02
^{155} Eu	(Bq/total)	7.6E+06 +/- 3.8E+05	1.8E+04 +/- 2.0E+02	8.9E+03 +/- 4.5E+02	1.5E+05 +/- 3.0E+03
^{239} Np	(Bq/total)	4.0E+05 +/- 2.0E+04	2.1E+03 +/- 2.0E+02	3.0E+03 +/- 3.0E+02	9.0E+04 +/- 9.6E+02

TABLE A2

THE 2nd SET OF OBSERVATIONS (LN SERIES, 28 SAMPLES)

Analysis		LN-1	LN-3	LN-4	LN-6	LN-7	LN-8
Gross Alpha	(Bq/total)	1.1E+06 +/- 1.9E+04	2.9E+06 +/- 4.4E+04	1.3E+06 +/- 2.1E+04	2.3E+06 +/- 3.9E+04	2.1E+05 +/- 1.2E+04	1.6E+06 +/- 3.2E+04
Alpha emitters							
^{244}Cm	(Bq/total)	1.1E+06 +/- 1.1E+05	2.8E+06 +/- 2.8E+05	1.3E+06 +/- 1.3E+05	2.3E+06 +/- 2.3E+05	2.0E+05 +/- 2.0E+04	1.6E+06 +/- 1.6E+05
^{238}Pu	(Bq/total)	4.7E+02 +/- 9.4E+01	1.4E+03 +/- 2.8E+02	9.8E+02 +/- 2.0E+02	1.7E+03 +/- 3.4E+02	3.4E+03 +/- 6.8E+02	7.9E+02 +/- 1.6E+02
$^{239/240}$Pu	(Bq/total)	5.1E+02 +/- 1.0E+02	6.3E+03 +/- 1.3E+03	4.0E+03 +/- 8.0E+02	6.3E+03 +/- 1.3E+03	9.5E+02 +/- 1.9E+02	4.7E+03 +/- 9.4E+02
^{242}Pu	(Bq/total)	2.0E+01 +/- 4.0E+00	1.5E+01 +/- 3.1E+00	2.0E+01 +/- 4.0E+00	1.6E+01 +/- 3.2E+00	4.8E+01 +/- 9.7E+00	4.4E+01 +/- 8.8E+00
^{241}Am	(Bq/total)	7.3E+03 +/- 9.5E+01	2.5E+03 +/- 5.4E+01	2.2E+03 +/- 5.2E+01	7.1E+03 +/- 8.1E+01	2.5E+03 +/- 4.7E+01	3.2E+03 +/- 5.3E+01
^{243}Am	(Bq/total)	4.5E+02 +/- 2.9E+01	5.7E+02 +/- 2.4E+01	4.1E+02 +/- 2.5E+01	6.8E+02 +/- 3.1E+01	8.0E+01 +/- 1.0E+01	3.7E+02 +/- 2.2E+01
^{252}Cf	(Bq/total)	1.6E+04 +/- 3.3E+03	2.6E+03 +/- 5.2E+02	2.5E+03 +/- 4.9E+02	3.2E+03 +/- 6.4E+02	4.8E+03 +/- 9.7E+02	2.8E+03 +/- 5.7E+02
^{243}Cm	(Bq/total)	8.8E+03 +/- 1.8E+03	3.1E+03 +/- 6.2E+02	2.7E+03 +/- 5.5E+02	3.7E+03 +/- 7.4E+02	3.8E+03 +/- 7.6E+02	3.6E+03 +/- 7.1E+02
^{246}Cm	(Bq/total)	< 1.1E+04	< 2.9E+04	< 1.3E+04	< 2.3E+04	< 2.1E+03	< 1.6E+04
Beta/gamma emitters							
^{60}Co	(Bq/total)	4.2E+02 +/- 3.0E+01	4.8E+01 +/- 1.2E+01	9.7E+01 +/- 1.2E+01	8.2E+01 +/- 1.6E+01	< 3.3E+01	1.5E+02 +/- 1.6E+01
^{106}Ru	(Bq/total)	5.6E+04 +/- 7.1E+02	2.2E+03 +/- 2.7E+02	6.6E+03 +/- 2.6E+02	1.9E+03 +/- 3.3E+02	8.8E+02 +/- 1.9E+02	1.8E+03 +/- 2.3E+02
^{110m}Ag	(Bq/total)	2.2E+03 +/- 3.7E+01	< 5.4E+01	1.2E+02 +/- 1.5E+01	< 7.8E+01	1.5E+02 +/- 1.2E+01	< 5.1E+01
^{125}Sb	(Bq/total)	1.7E+03 +/- 1.5E+02	2.5E+03 +/- 7.1E+01	< 1.7E+02	1.7E+03 +/- 1.8E+02	< 1.5E+02	1.1E+03 +/- 7.3E+01
^{134}Cs	(Bq/total)	1.7E+03 +/- 3.6E+01	6.0E+02 +/- 2.0E+01	3.3E+02 +/- 1.9E+01	2.9E+03 +/- 3.6E+01	< 3.9E+01	7.3E+02 +/- 2.1E+01
^{137}Cs	(Bq/total)	2.0E+04 +/- 1.3E+02	8.8E+03 +/- 8.2E+01	3.6E+03 +/- 5.3E+01	4.8E+04 +/- 1.9E+02	8.2E+02 +/- 2.9E+01	1.1E+04 +/- 9.4E+01
^{144}Ce	(Bq/total)	9.9E+03 +/- 2.5E+02	1.4E+03 +/- 1.2E+02	1.5E+03 +/- 1.1E+02	2.0E+03 +/- 1.6E+02	7.1E+02 +/- 8.6E+01	1.5E+03 +/- 1.0E+02
^{152}Eu	(Bq/total)	6.6E+02 +/- 3.3E+02	< 2.3E+02	4.0E+02 +/- 3.2E+01	5.0E+02 +/- 2.0E+02	4.2E+02 +/- 7.9E+01	< 2.6E+02
^{154}Eu	(Bq/total)	4.5E+03 +/- 6.8E+01	3.8E+03 +/- 3.8E+01	1.9E+03 +/- 3.6E+01	3.6E+03 +/- 4.8E+01	1.5E+03 +/- 2.8E+01	1.8E+03 +/- 2.9E+01
^{155}Eu	(Bq/total)	2.5E+03 +/- 8.7E+01	1.6E+03 +/- 5.6E+01	9.0E+02 +/- 5.5E+01	1.9E+03 +/- 6.6E+01	9.5E+02 +/- 4.2E+01	1.1E+03 +/- 4.8E+01
^{237}Np	(Bq/total)	7.2E+02 +/- 9.1E+01	1.5E+03 +/- 6.3E+01	8.4E+02 +/- 9.4E+01	1.2E+03 +/- 1.0E+02	< 1.7E+02	9.5E+02 +/- 5.1E+01

Analysis		LN-9	LN-11	LN-13	LN-15	LN-19	LN-25
Gross Alpha	(Bq/total)	2.0E+06 +/- 3.8E+04	1.1E+06 +/- 1.9E+04	2.3E+06 +/- 3.9E+04	4.1E+05 +/- 8.4E+03	9.1E+05 +/- 1.2E+04	6.7E+05 +/- 1.1E+04
Alpha emitters							
^{244}Cm	(Bq/total)	2.0E+06 +/- 2.0E+05	1.1E+06 +/- 1.1E+05	2.3E+06 +/- 2.3E+05	3.8E+05 +/- 3.8E+04	9.0E+05 +/- 9.0E+04	6.6E+05 +/- 6.6E+04
^{238}Pu	(Bq/total)	6.6E+02 +/- 1.3E+02	1.3E+03 +/- 2.5E+02	1.1E+03 +/- 2.3E+02	6.3E+03 +/- 1.3E+03	1.1E+03 +/- 2.1E+02	4.4E+02 +/- 8.8E+01
$^{239/240}$Pu	(Bq/total)	4.9E+03 +/- 9.9E+02	2.0E+03 +/- 4.0E+02	8.9E+03 +/- 1.8E+03	1.8E+03 +/- 3.6E+02	4.0E+03 +/- 8.0E+02	1.1E+03 +/- 2.2E+02
^{242}Pu	(Bq/total)	3.9E+01 +/- 7.8E+00	2.0E+01 +/- 4.0E+00	2.0E+01 +/- 4.0E+00	6.6E+01 +/- 1.3E+01	3.2E+01 +/- 6.4E+00	8.0E+00 +/- 1.6E+00
^{241}Am	(Bq/total)	1.5E+03 +/- 5.7E+01	6.2E+03 +/- 6.5E+01	2.8E+03 +/- 5.4E+01	5.9E+03 +/- 6.8E+01	1.9E+03 +/- 4.5E+01	4.9E+03 +/- 5.5E+01
^{243}Am	(Bq/total)	3.1E+02 +/- 2.5E+01	4.1E+02 +/- 2.3E+01	3.0E+02 +/- 1.7E+01	2.9E+02 +/- 1.7E+01	1.4E+02 +/- 1.3E+01	2.2E+02 +/- 1.3E+01
^{252}Cf	(Bq/total)	1.6E+03 +/- 3.3E+02	5.5E+03 +/- 1.1E+03	2.5E+03 +/- 4.9E+02	1.0E+04 +/- 2.1E+03	2.1E+03 +/- 4.2E+02	1.1E+03 +/- 2.2E+02
^{242}Cm	(Bq/total)	6.4E+03 +/- 1.3E+03	1.6E+04 +/- 3.3E+03	2.1E+03 +/- 4.3E+02	5.7E+03 +/- 1.1E+03	6.1E+02 +/- 1.2E+02	2.3E+03 +/- 4.5E+02
^{246}Cm	(Bq/total)	< 2.0E+04	< 1.1E+04	< 2.3E+04	3.3E+03 +/- 6.6E+02	< 9.1E+03	2.7E+03 +/- 5.4E+02
Beta/gamma emitters							
^{60}Co	(Bq/total)	< 3.9E+01	1.1E+02 +/- 2.1E+01	< 3.8E+01	2.9E+01 +/- 1.6E+01	< 3.8E+01	< 3.9E+01
^{106}Ru	(Bq/total)	2.3E+03 +/- 2.9E+02	2.2E+04 +/- 5.2E+02	1.0E+03 +/- 1.8E+02	1.0E+03 +/- 2.0E+02	1.7E+03 +/- 2.2E+02	1.2E+03 +/- 1.6E+02
^{110m}Ag	(Bq/total)	< 5.5E+01	8.8E+02 +/- 2.8E+01	< 4.6E+01	< 5.5E+01	1.5E+02 +/- 1.5E+01	< 3.4E+01
^{125}Sb	(Bq/total)	< 2.0E+02	1.4E+03 +/- 1.2E+02	< 2.0E+02	1.0E+03 +/- 5.4E+01	8.6E+02 +/- 5.7E+01	5.8E+02 +/- 4.3E+01
^{134}Cs	(Bq/total)	3.1E+02 +/- 2.1E+01	2.9E+03 +/- 3.9E+01	5.4E+02 +/- 2.2E+01	8.0E+01 +/- 1.7E+01	3.3E+02 +/- 1.6E+01	8.3E+01 +/- 9.0E+00
^{137}Cs	(Bq/total)	5.2E+03 +/- 7.3E+01	4.5E+04 +/- 1.8E+02	8.6E+03 +/- 8.4E+01	1.0E+03 +/- 3.4E+01	4.6E+03 +/- 5.8E+01	1.1E+03 +/- 3.0E+01
^{144}Ce	(Bq/total)	3.9E+02 +/- 1.0E+02	7.3E+03 +/- 2.1E+02	1.2E+03 +/- 1.2E+02	2.2E+03 +/- 1.0E+02	1.0E+03 +/- 8.9E+01	5.8E+02 +/- 6.3E+01
^{152}Eu	(Bq/total)	< 1.8E+02	< 3.0E+02	< 1.9E+02	8.3E+02 +/- 3.2E+01	< 1.4E+02	< 1.5E+02
^{154}Eu	(Bq/total)	7.9E+02 +/- 4.6E+01	3.4E+03 +/- 5.4E+01	1.3E+03 +/- 3.5E+01	3.4E+03 +/- 3.8E+01	1.1E+03 +/- 6.6E+01	6.8E+02 +/- 2.0E+01
^{155}Eu	(Bq/total)	6.7E+02 +/- 4.6E+01	2.4E+03 +/- 7.4E+01	6.2E+02 +/- 5.5E+01	2.6E+03 +/- 6.2E+01	5.2E+02 +/- 4.0E+01	3.2E+02 +/- 2.6E+01
^{219}Np	(Bq/total)	7.5E+02 +/- 7.6E+01	8.9E+02 +/- 9.2E+01	1.1E+03 +/- 5.9E+01	6.4E+02 +/- 5.5E+01	7.1E+02 +/- 4.6E+01	5.9E+02 +/- 3.5E+01

-43-

Analysis		LN-26	LN-28	LN-33	LN-35	LN-36	LN-38
Gross Alpha	(Bq/total)	1.5E+06 +/- 2.2E+04	1.1E+06 +/- 1.9E+04	1.4E+06 +/- 2.2E+04	1.5E+06 +/- 2.2E+04	9.5E+06 +/- 7.9E+04	2.1E+05 +/- 5.9E+03
Alpha emitters							
^{244}Cm	(Bq/total)	1.4E+06 +/- 1.4E+05	1.1E+06 +/- 1.1E+05	1.4E+06 +/- 1.4E+05	1.4E+06 +/- 1.4E+05	9.4E+06 +/- 9.4E+05	2.0E+05 +/- 2.0E+04
^{238}Pu	(Bq/total)	1.1E+03 +/- 2.2E+02	1.4E+03 +/- 2.9E+02	6.4E+03 +/- 1.3E+03	1.4E+04 +/- 2.9E+03	2.3E+03 +/- 4.6E+02	6.9E+02 +/- 1.4E+02
$^{239/240}$Pu	(Bq/total)	6.9E+03 +/- 1.4E+03	6.6E+03 +/- 1.3E+03	3.6E+03 +/- 7.2E+02	5.5E+03 +/- 1.1E+03	3.8E+03 +/- 7.6E+02	4.8E+02 +/- 9.7E+01
^{242}Pu	(Bq/total)	6.5E+01 +/- 1.3E+01	4.9E+01 +/- 9.7E+00	5.0E+01 +/- 1.0E+01	2.0E+02 +/- 4.0E+01	2.4E+01 +/- 4.9E+00	2.8E+01 +/- 5.5E+00
^{241}Am	(Bq/total)	1.7E+03 +/- 5.4E+01	2.2E+03 +/- 4.9E+01	1.5E+04 +/- 9.3E+01	5.8E+04 +/- 2.0E+02	8.3E+04 +/- 2.4E+02	1.7E+03 +/- 4.1E+01
^{243}Am	(Bq/total)	2.4E+02 +/- 2.0E+01	2.9E+02 +/- 1.4E+01	1.3E+03 +/- 3.1E+01	3.7E+02 +/- 2.7E+01	1.0E+03 +/- 2.9E+01	1.7E+02 +/- 1.7E+01
^{252}Cf	(Bq/total)	5.8E+03 +/- 1.2E+03	2.1E+03 +/- 4.2E+02	1.3E+04 +/- 2.6E+03	2.1E+04 +/- 4.2E+03	4.5E+03 +/- 9.0E+02	1.9E+03 +/- 3.8E+02
^{242}Cm	(Bq/total)	5.8E+03 +/- 1.2E+03	1.2E+03 +/- 2.4E+02	9.4E+03 +/- 1.9E+03	1.9E+04 +/- 3.9E+03	5.0E+03 +/- 1.0E+03	1.5E+03 +/- 2.9E+02
^{246}Cm	(Bq/total)	< 1.5E+04	< 1.1E+04	< 1.4E+04	< 1.5E+04	2.9E+04 +/- 5.7E+03	8.4E+02 +/- 1.7E+02
Beta/gamma emitters							
^{60}Co	(Bq/total)	1.9E+02 +/- 2.0E+01	2.1E+01 +/- 1.2E+01	2.1E+02 +/- 2.4E+01	1.7E+02 +/- 2.2E+01	< 4.6E+01	9.5E+01 +/- 1.3E+01
^{106}Ru	(Bq/total)	2.1E+04 +/- 4.8E+02	2.3E+03 +/- 2.9E+02	2.7E+04 +/- 6.2E+02	4.1E+03 +/- 4.1E+02	1.3E+03 +/- 2.0E+02	2.6E+03 +/- 2.6E+02
^{110m}Ag	(Bq/total)	3.3E+03 +/- 4.0E+01	< 5.2E+01	3.7E+02 +/- 3.0E+01	3.5E+02 +/- 2.8E+01	< 5.4E+01	1.4E+02 +/- 1.4E+01
^{125}Sb	(Bq/total)	1.5E+03 +/- 8.3E+01	9.9E+02 +/- 6.3E+01	3.2E+03 +/- 1.2E+02	2.9E+03 +/- 9.9E+01	1.2E+03 +/- 6.4E+01	6.6E+02 +/- 4.9E+01
^{134}Cs	(Bq/total)	5.5E+02 +/- 2.9E+01	4.6E+02 +/- 1.9E+01	7.1E+02 +/- 3.7E+01	3.3E+02 +/- 3.4E+01	4.1E+02 +/- 2.0E+01	2.3E+02 +/- 1.5E+01
^{137}Cs	(Bq/total)	3.5E+03 +/- 6.4E+01	6.9E+03 +/- 7.6E+01	7.8E+03 +/- 8.5E+01	3.8E+03 +/- 6.6E+01	6.3E+03 +/- 6.8E+01	1.9E+03 +/- 4.0E+01
^{144}Ce	(Bq/total)	8.6E+03 +/- 1.9E+02	1.1E+03 +/- 1.1E+02	1.0E+04 +/- 2.3E+02	6.0E+03 +/- 1.9E+02	1.1E+03 +/- 1.3E+02	1.6E+03 +/- 9.6E+01
^{152}Eu	(Bq/total)	< 1.8E+02	< 2.1E+02	1.5E+03 +/- 3.0E+02	1.3E+03 +/- 2.6E+02	< 2.1E+02	< 2.3E+02
^{154}Eu	(Bq/total)	1.0E+03 +/- 4.1E+01	1.3E+03 +/- 2.5E+01	1.7E+04 +/- 9.7E+01	8.9E+03 +/- 6.7E+01	1.6E+03 +/- 3.5E+01	2.0E+03 +/- 3.1E+01
^{155}Eu	(Bq/total)	6.4E+02 +/- 5.9E+01	7.0E+02 +/- 4.6E+01	9.6E+03 +/- 1.2E+02	8.3E+03 +/- 1.1E+02	7.6E+02 +/- 5.8E+01	1.0E+03 +/- 3.8E+01
^{239}Np	(Bq/total)	1.4E+03 +/- 8.8E+01	7.0E+02 +/- 8.5E+01	2.1E+03 +/- 1.2E+02	1.4E+03 +/- 1.2E+02	4.8E+03 +/- 1.7E+02	2.4E+02 +/- 3.3E+01

-44-

Analysis		LN-39	LN-40	LN-41	LN-44	LN-46	LN-48
Gross Alpha	(Bq/total)	1.3E+07 +/- 1.3E+05	1.4E+06 +/- 2.2E+04	5.5E+05 +/- 9.7E+03	3.5E+05 +/- 7.6E+03	1.9E+05 +/- 1.6E+03	3.9E+05 +/- 1.0E+04
Alpha emitters							
^{244}Cm	(Bq/total)	1.3E+07 +/- 1.3E+06	1.4E+06 +/- 1.4E+05	5.3E+05 +/- 5.3E+04	3.4E+05 +/- 3.4E+04	1.9E+05 +/- 1.9E+04	3.8E+05 +/- 3.8E+04
^{238}Pu	(Bq/total)	3.0E+03 +/- 6.1E+02	3.8E+02 +/- 7.5E+01	2.0E+03 +/- 3.9E+02	3.1E+03 +/- 6.3E+02	1.1E+03 +/- 2.2E+02	1.6E+03 +/- 3.1E+02
$^{239/240}$Pu	(Bq/total)	6.2E+03 +/- 1.2E+03	2.9E+02 +/- 5.7E+01	2.1E+03 +/- 4.2E+02	1.4E+03 +/- 2.9E+02	6.8E+02 +/- 1.4E+02	9.3E+02 +/- 1.9E+02
^{242}Pu	(Bq/total)	9.3E+00 +/- 1.9E+00	8.7E+00 +/- 1.7E+00	3.3E+01 +/- 6.6E+00	4.6E+01 +/- 9.2E+00	1.7E+01 +/- 3.4E+00	1.3E+01 +/- 2.5E+00
^{241}Am	(Bq/total)	1.1E+05 +/- 2.7E+02	1.1E+04 +/- 5.9E+02	7.2E+03 +/- 7.7E+01	2.2E+03 +/- 4.3E+01	2.0E+03 +/- 4.2E+01	3.7E+03 +/- 5.6E+01
^{243}Am	(Bq/total)	1.2E+03 +/- 3.1E+01	5.8E+03 +/- 3.3E+02	2.1E+02 +/- 2.0E+01	1.9E+02 +/- 1.4E+01	1.1E+02 +/- 1.6E+01	1.5E+02 +/- 1.3E+01
^{252}Cf	(Bq/total)	5.2E+03 +/- 1.0E+03	1.8E+04 +/- 3.5E+03	6.0E+03 +/- 1.2E+03	1.2E+03 +/- 2.4E+02	1.8E+03 +/- 3.5E+02	2.5E+03 +/- 4.9E+02
^{242}Cm	(Bq/total)	7.8E+03 +/- 1.6E+03	1.3E+04 +/- 2.6E+03	1.7E+03 +/- 3.4E+02	4.1E+03 +/- 8.1E+02	3.2E+02 +/- 6.5E+01	1.4E+03 +/- 2.9E+02
^{246}Cm	(Bq/total)	3.9E+04 +/- 7.8E+03	< 1.4E+04	4.8E+03 +/- 9.6E+02	< 3.5E+03	3.8E+02 +/- 7.6E+01	< 3.9E+03
Beta/gamma emitters							
^{60}Co	(Bq/total)	5.0E+01 +/- 1.2E+01	< 3.7E+02	1.4E+02 +/- 1.4E+01	3.9E+01 +/- 1.1E+01	1.1E+02 +/- 1.4E+01	6.7E+01 +/- 1.1E+01
^{106}Ru	(Bq/total)	2.2E+03 +/- 2.1E+02	6.2E+05 +/- 6.7E+03	7.3E+03 +/- 3.4E+02	7.8E+02 +/- 1.3E+02	3.3E+03 +/- 2.1E+02	3.0E+03 +/- 2.8E+01
^{110m}Ag	(Bq/total)	2.0E+02 +/- 1.8E+01	3.1E+03 +/- 2.6E+02	1.6E+02 +/- 1.6E+01	2.6E+02 +/- 1.3E+01	< 4.8E+01	< 4.6E+01
^{125}Sb	(Bq/total)	1.4E+03 +/- 7.1E+01	< 3.0E+03	3.9E+03 +/- 1.1E+02	5.1E+02 +/- 3.9E+01	5.3E+02 +/- 4.1E+01	8.9E+02 +/- 6.3E+01
^{134}Cs	(Bq/total)	3.0E+02 +/- 2.0E+01	< 1.2E+04	4.9E+02 +/- 3.1E+01	2.6E+02 +/- 1.2E+01	2.5E+02 +/- 1.4E+01	3.5E+02 +/- 2.2E+01
^{137}Cs	(Bq/total)	4.4E+03 +/- 5.9E+01	1.7E+04 +/- 5.2E+02	6.3E+03 +/- 6.9E+01	3.9E+03 +/- 5.5E+01	2.2E+03 +/- 4.2E+01	4.6E+03 +/- 6.4E+01
^{144}Ce	(Bq/total)	7.0E+02 +/- 8.6E+01	1.9E+04 +/- 1.8E+03	1.2E+03 +/- 9.9E+01	1.0E+03 +/- 8.7E+01	1.2E+03 +/- 8.1E+01	1.3E+03 +/- 9.8E+01
^{152}Eu	(Bq/total)	< 2.2E+02	< 1.5E+03	< 2.1E+02	< 1.7E+02	< 1.8E+02	< 2.0E+02
^{154}Eu	(Bq/total)	1.5E+03 +/- 3.6E+01	4.1E+04 +/- 1.4E+03	2.8E+03 +/- 4.2E+01	6.0E+02 +/- 1.7E+01	1.4E+03 +/- 2.9E+01	2.1E+03 +/- 3.5E+01
^{155}Eu	(Bq/total)	3.5E+02 +/- 3.9E+01	1.7E+04 +/- 7.3E+02	6.8E+02 +/- 4.1E+01	3.2E+02 +/- 3.2E+01	1.0E+03 +/- 4.2E+01	1.0E+03 +/- 5.3E+01
^{239}Np	(Bq/total)	3.5E+03 +/- 1.2E+03	< 4.3E+03	< 2.7E+02	3.6E+02 +/- 3.5E+01	2.6E+02 +/- 4.0E+01	4.5E+02 +/- 5.1E+01

Analysis		LN-49	LN-54	LN-GL	LN-ML
Gross Alpha	(Bq/total)	1.2E+05 +/- 4.4E+03	6.0E+06 +/- 1.0E+04	1.2E+08 +/- 1.7E+06	7.3E+07 +/- 1.4E+06
Alpha emitters					
^{244} Cm	(Bq/total)	1.2E+05 +/- 1.2E+04	5.9E+05 +/- 5.9E+04	1.2E+08 +/- 1.2E+07	6.9E+07 +/- 6.9E+06
^{238} Pu	(Bq/total)	4.0E+02 +/- 8.0E+01	1.4E+03 +/- 2.8E+02	2.7E+05 +/- 5.5E+04	1.1E+06 +/- 2.2E+05
$^{239/240}$ Pu	(Bq/total)	4.0E+02 +/- 8.0E+01	1.1E+03 +/- 2.3E+02	3.2E+05 +/- 6.4E+04	2.9E+05 +/- 5.8E+04
^{242} Pu	(Bq/total)	< 8.2E+01	3.4E+01 +/- 6.8E+00	4.2E+03 +/- 8.4E+02	1.3E+04 +/- 2.5E+03
^{241} Am	(Bq/total)	8.9E+02 +/- 2.8E+01	5.2E+03 +/- 5.9E+01	6.2E+05 +/- 6.6E+03	9.3E+05 +/- 5.5E+03
^{243} Am	(Bq/total)	8.8E+01 +/- 8.4E+00	3.2E+02 +/- 1.5E+01	7.1E+04 +/- 1.8E+03	4.9E+04 +/- 1.1E+03
^{252} Cf	(Bq/total)	7.8E+02 +/- 1.6E+02	2.5E+03 +/- 4.9E+02	1.3E+06 +/- 2.6E+05	2.0E+06 +/- 4.0E+05
^{242} Cm	(Bq/total)	3.0E+02 +/- 6.0E+01	2.3E+03 +/- 4.7E+02	5.0E+05 +/- 1.0E+05	4.1E+05 +/- 8.1E+04
^{246} Cm	(Bq/total)	< 1.2E+03	1.8E+03 +/- 3.6E+02	< 1.2E+06	< 7.3E+05
Beta/gamma emitters					
^{60} Co	(Bq/total)	3.8E+01 +/- 1.0E+01	7.0E+01 +/- 1.3E+01	4.7E+04 +/- 1.6E+03	6.5E+03 +/- 1.0E+03
^{106} Ru	(Bq/total)	1.1E+03 +/- 1.5E+02	2.6E+03 +/- 2.2E+02	5.2E+06 +/- 4.6E+04	3.7E+05 +/- 2.2E+04
^{110m} Ag	(Bq/total)	< 3.1E+01	1.5E+02 +/- 1.3E+01	7.7E+04 +/- 2.1E+03	1.7E+04 +/- 1.1E+03
^{125} Sb	(Bq/total)	2.7E+02 +/- 3.1E+01	5.6E+02 +/- 4.7E+01	2.3E+05 +/- 3.5E+04	1.1E+05 +/- 4.0E+03
^{134} Cs	(Bq/total)	9.1E+01 +/- 9.8E+00	2.2E+02 +/- 1.8E+01	5.0E+05 +/- 3.2E+03	1.4E+04 +/- 1.0E+03
^{137} Cs	(Bq/total)	8.7E+02 +/- 3.1E+01	3.0E+03 +/- 4.9E+01	6.7E+06 +/- 1.4E+04	1.5E+05 +/- 2.5E+03
^{144} Ce	(Bq/total)	5.1E+02 +/- 6.0E+01	2.7E+03 +/- 1.1E+02	5.5E+05 +/- 1.6E+04	3.3E+05 +/- 8.0E+03
^{152} Eu	(Bq/total)	< 1.7E+02	3.2E+02 +/- 1.3E+02	5.6E+04 +/- 3.1E+03	1.4E+05 +/- 2.6E+03
^{154} Eu	(Bq/total)	7.8E+02 +/- 1.8E+01	2.6E+03 +/- 3.6E+01	3.9E+05 +/- 3.4E+03	5.2E+05 +/- 2.9E+03
^{155} Eu	(Bq/total)	3.8E+02 +/- 2.9E+01	1.3E+03 +/- 5.0E+01	2.4E+05 +/- 4.6E+03	4.4E+05 +/- 4.4E+04
^{239} Np	(Bq/total)	1.6E+02 +/- 3.0E+01	5.2E+02 +/- 4.2E+01	1.5E+05 +/- 9.6E+03	9.0E+04 +/- 1.8E+04

TABLE A3

EXAMPLE (LN-GL) OF INFORMATION AVAILABLE FOR THE LEACHING TESTS

LN-GL
3 glass bottles weighing 80.2g

Analysis		Result	Error
Gross Alpha	(Bq/total)	1.2E+08 +/-	1.7E+06

Alpha emitters

^{244} Cm	(Bq/total)	1.2E+08 +/-	1.2E+07
^{238} Pu	(Bq/total)	2.7E+05 +/-	5.5E+04
$^{239/240}$ Pu	(Bq/total)	3.2E+05 +/-	6.4E+04
^{242} Pu	(Bq/total)	4.2E+03 +/-	8.4E+02
^{241} Am	(Bq/total)	6.2E+05 +/-	6.6E+03
^{243} Am	(Bq/total)	7.1E+04 +/-	1.8E+03
^{249} Cf	(Bq/total)	4.0E+03 +/-	8.0E+02
^{250} Cf	(Bq/total)	5.1E+04 +/-	1.0E+04
^{251} Cf	(Bq/total)	2.3E+02 +/-	4.7E+01
^{252} Cf	(Bq/total)	1.3E+06 +/-	2.6E+05
^{253} Cf	(Bq/total)	3.5E+04 +/-	7.0E+03
^{254} Cf	(Bq/total)	6.9E+03 +/-	1.4E+03
^{242} Cm	(Bq/total)	5.0E+05 +/-	1.0E+05
^{245} Cm	(Bq/total)	4.4E+03 +/-	4.4E+02
^{246} Cm	(Bq/total)	1.7E+05 +/-	1.7E+04
^{247} Cm	(Bq/total)	1.2E+00 +/-	1.2E-01
^{248} Cm	(Bq/total)	3.5E+02 +/-	3.5E+01

Beta/gamma emitters

^{60} Co	(Bq/total)	4.7E+04 +/-	1.6E+03
^{106} Ru	(Bq/total)	5.2E+06 +/-	4.6E+04
^{110m} Ag	(Bq/total)	7.7E+04 +/-	2.1E+03
^{125} Sb	(Bq/total)	2.3E+05 +/-	3.5E+04
^{134} Cs	(Bq/total)	5.0E+05 +/-	3.2E+03
^{137} Cs	(Bq/total)	6.7E+06 +/-	1.4E+04
^{144} Ce	(Bq/total)	5.5E+05 +/-	1.6E+04
^{152} Eu	(Bq/total)	5.6E+04 +/-	3.1E-03
^{154} Eu	(Bq/total)	3.9E+05 +/-	3.4E+03
^{155} Eu	(Bq/total)	2.4E+05 +/-	4.6E+03
^{239} Np	(Bq/total)	1.5E+05 +/-	9.6E+03

Total Alpha Concentration =	4.0E+04 nCi/g
TRU Concentration =	495.7 nCi/g

COMMENTS TO TABLE A3

Three glass sample bottles (LN-GL) were removed from a waste can in cubicle 8 in order to leach. The bottles were placed in a beaker filled with 400 mL of 8N Nitric Acid and allowed to leach for approximately four hours. Following the leach an aliquot of the total leachate was taken from the beaker for radiochemical analyses. The mass of the three bottles had to be estimated because there are no current means of weighing inside the cell bank. Three sets of three sample bottles identical to those leached were weighed resulting in an average of 80.2 grams. This mass was combined with the radiochemical analyses to provide a TRU Concentration for the set of bottles.

TABLE A4

SAMPLE TRACKING RECORD

First data set

SAMPLE NUMNBER	CELL LOC.	SAMPLING DATE	WASTE CATEGORY	WASTE ITEM DESCRIPTION	TYPE OF SAMPLE
RH9AA	9	10/26/95	Plastic	Melted Poly	Smear
RH9EE	9	10/26/95	Plastic	Poly Valve	Smear
RH9KK	9	10/26/95	Plastic	Bottle lid	Smear
RH9LL	9	10/26/95	Glass	Sample bottle	Smear
RH9QQ	9	10/26/95	Metal	Wire	Smear
RH9SS	9	10/26/95	Cloth	Wipes	Smear
RH9A	9	1/11/96	Plastic	Melted Poly	Smear
RH9C	9	1/11/96	Plastic	Melted Poly	Smear
RH9D	9	1/11/96	Plastic	Bain Marie bucket	Smear
RH9E	9	1/11/96	Plastic	Social FDV poly valve	Smear
RH9F	9	1/11/96	Plastic	PVC Valve	Smear
RH9G	9	1/11/96	Plastic	PVC Valve	Smear
RH9H	9	1/11/96	Plastic	Light cord	Smear
RH9I	9	1/11/96	Plastic	MSM boot	Smear
RH9J	9	1/11/96	Plastic	Poly bottle lid	Smear
RH9K	9	1/11/96	Plastic	Bottle lid	Smear
RH9L	9	1/10/96	Glass	Sample bottle	Smear
RH9M	9	1/11/96	Glass	Sample bottle	Smear
RH9N	9	1/11/96	Glass	Glassware	Smear
RH9O	9	1/11/96	Glass	Filter housing	Smear
RH9P	6	1/11/96	Metal	Metal tool	Smear
RH9Q	9	1/11/96	Metal	PreAmp lead can	Smear
RH9R	9	1/11/96	Metal	Stainless steel lid	Smear
RH9S	9	10/26/95	Cloth	Wipes	Smear
RH9T	9	1/11/96	Cloth	Wipes	Smear
RH9U	9	1/11/96	Cloth	Wipes	Smear
RH9V	9	1/11/96	Cloth	String	Smear
RH1B	1	1/11/96		MSM Boot	Smear
RH2B	2	1/11/96		MSM Boot	Smear
RH3B	3	1/11/96		MSM Boot	Smear
RH4D	4	1/11/96		Cubicle Sump	Smear
RH6C	6	1/11/96		Cubicle Sump	Smear
RH7D	7	1/11/96		Cubicle Sump	Smear
RH8A	8	1/11/96		Cubicle Sump	Smear

Total number of samples = 34

TABLE A5

SAMPLE TRACKING RECORD

Second data set

SAMPLE NUMNBER	CELL LOC.	SAMPLING DATE	WASTE CATEGORY	WASTE ITEM DESCRIPTION	TYPE OF SAMPLE
LN-1	9	5/1/96	glass	bottle	Smear
LN-3	9	5/1/96	glass	bottle	Smear
LN-4	9	5/1/96	glass	bottle	Smear
LN-6	9	5/1/96	glass	bottle	Smear
LN-7	9	5/1/96	metal	tool	Smear
LN-8	9	5/1/96	glass	bottle	Smear
LN-9	9	5/1/96	glass	bulb	Smear
LN-11	9	5/1/96	glass	bottle	Smear
LN-13	9	5/1/96	glass	bottle	Smear
LN-15	9	5/1/96	metal	tool	Smear
LN-19	9	5/1/96	glass	bottle	Smear
LN-25	9	5/1/96	glass	bottle	Smear
LN-26	9	5/1/96	glass	bottle	Smear
LN-28	9	5/1/96	glass	bottle	Smear
LN-33	9	5/1/96	poly	valve	Smear
LN-35	9	5/1/96	poly	bottle	Smear
LN-36	9	5/1/96	metal	cell	Smear
LN-38	9	5/1/96	metal	tool	Smear
LN-39	9	5/1/96	metal	cell	Smear
LN-40	9	5/1/96	poly	hose	Smear
LN-41	9	5/1/96	metal	column	Smear
LN-44	9	5/1/96	metal	cap	Smear
LN-46	9	5/1/96	metal	can	Smear
LN-48	9	5/1/96	metal	tool	Smear
LN-49	9	5/1/96	metal	socket	Smear
LN-54	9	5/1/96	metal	case	Smear
LN-GL	9	5/1/96	glass	bottles	Leach
LN-ML	9	5/1/96	metal	tool	Leach

Total number of samples = 28

Notes:

1. LN-ML (Metal leach of a tool) has duplicate smears performed on the tool prior to leaching: LN-7 and LN-15
2. LN-GL (Glass leach of 3 bottles) has duplicate smears performed on the bottles prior to leaching: LN-1 and LN-11
3. LN-19 and LN-28 are duplicate smears of the same glass bottle
4. LN-36 and LN-39 are duplicate smears of a metal weighing cell

-50-

Appendix 2. Linear Regression for Poisson-Distributed Data with Special Reference to Spectrometry

This appendix outlines the method used to obtain the individual radionuclide activities from the sample spectrograph using methods based on linear Poisson regression. Let $\mathbf{y}$ be the $n \times 1$ vector of observed counts from the n channels of interest in the spectrograph. If we assume the elements in $\mathbf{y}$ to be the realization of n independent Poisson random variables, then the likelihood function of the sample vector $\mathbf{y}$ is given by

$$L(\mathbf{y}, \boldsymbol{\theta}) = \prod_{i=1}^{n} \frac{exp(-\mu(x_i, \boldsymbol{\theta}))\mu(x_i, \boldsymbol{\theta})^{y_i}}{y_i!} \tag{1}$$

or

$$l(\mathbf{y}, \boldsymbol{\theta}) = \log(L(\mathbf{y}, \boldsymbol{\theta})) = -\sum_{i=1}^{n} \mu(x_i, \boldsymbol{\theta}) + \sum_{i=1}^{n} y_i \ln \mu(x_i, \boldsymbol{\theta}) + \text{constant} \tag{2}$$

where

$$\mu(x_i, \boldsymbol{\theta}) = \theta_1 \psi_1(x_i) + \ldots + \theta_m \psi_m(x_i) = \boldsymbol{\theta}^T \Psi(x_i) \tag{3}$$

Our immediate objective is to maximize $l(\mathbf{y}, \boldsymbol{\theta})$ with respect to $\boldsymbol{\theta}$. It should be noted that in spectrometry it is natural to assume that all components of the vector $\boldsymbol{\theta}$ are non-negative. Since $\mu(x, \boldsymbol{\theta})$ is a linear function of the estimated parameters, the maximum likelihood estimator is found as the solution to the system of equations

$$\frac{\partial \ell(\mathbf{y}, \boldsymbol{\theta})}{\partial \theta_\alpha} = -\sum_{i=1}^{n} \psi_\alpha(x_i) + \sum_{i=1}^{n} \frac{y_i \psi_\alpha(x_i)}{\boldsymbol{\theta}^T \psi(x_i)}, \ \alpha = 1, \ldots, m \tag{4}$$

or

$$\sum_{i=1}^{n} w_i y_i \psi_\alpha(x_i) = \sum_{i=1}^{n} \psi_\alpha(x_i), \tag{5}$$

where

$$w_i^{-1} = \boldsymbol{\theta}^T \Psi(x_i). \tag{6}$$

After some simplification, this same system can be written in the following matrix notation:

$$\Psi^T W \Psi \boldsymbol{\theta} = \Psi^T W \mathbf{y} \tag{7}$$

where W is an $n \times n$ diagonal matrix with the i-th diagonal entry w_i; the $n \times (m+1)$ matrix Ψ has the $(i, \alpha)^{th}$ entry given by $\psi_\alpha(x_i)$; θ is the $(m+1) \times 1$ vector with α^{th} entry θ_α; and y is the $n \times 1$ vector with i-th entry y_i for $i = 1, 2, \ldots, n$ and $\alpha = 0, 1, \ldots, m$. Let $M(\theta)$ equal $\Psi' W \Psi$ and $Y(\theta)$ equal $\Psi' W y$, then the solution for θ is given by

$$\hat{\theta} = M^{-1}(\theta) Y(\theta). \tag{8}$$

The above equation for the solution of θ is a typical object for the "fixed point" method in numerical mathematics or for the "iterative reweighted least squares" estimation method in statistics; compare with [14, 15]. At the s-th iteration, the new estimate of θ is obtained from

$$\hat{\theta}_{s+1} = M^{-1}(\hat{\theta}_s) Y(\hat{\theta}_s). \tag{9}$$

Under some reasonably achievable regularity conditions, it can be shown that

$$\lim_{s \to \infty} \hat{\theta}_s = \hat{\theta} \tag{10}$$

where $\hat{\theta}$ is the maximum likelihood estimator.

When the spectra of all elements in the mixture are well disjoint, i.e. their peaks do not overlap, then the following assumption looks very reasonable. There are disjoint sets I_α, such that

$$\psi_\alpha(x_i) = \begin{cases} \phi_\alpha(x_i) \geq 0, & i \in I_\alpha \\ 0, & \text{otherwise} \end{cases} \tag{11}$$

where $I_\alpha \cap I_\beta = 0$ when $\alpha \neq \beta$. Under this set of assumptions, $M(\theta)$ is a diagonal matrix with diagonal elements given by

$$M_{\alpha\alpha}(\theta) = \theta_\alpha^{-1} \sum_{i \in I_\alpha} \phi_\alpha(x_i), \tag{12}$$

for $\alpha = 1, 2, \ldots, m$. Similarly, for this case, the elements in the vector $Y(\theta)$ are given by

$$Y_\alpha(\theta) = \theta_\alpha^{-1} \sum_{i \in I_\alpha} y_i \tag{13}$$

- 52 -

again for $\alpha = 1, 2, \ldots, m$. From equations (12) and (13), we have

$$\hat{\theta} = \frac{\sum\limits_{i \in I_\alpha} y_i}{\sum\limits_{i \in I_\alpha} \phi_\alpha(x_i)} \tag{14}$$

and

$$Var(\hat{\theta}_\alpha) = \frac{\sum\limits_{i \in I_\alpha} Var(y_i)}{(\sum\limits_{i \in I_\alpha} \phi_\alpha(x_i))^2} = \frac{\theta_\alpha}{\sum\limits_{i \in I_\alpha} \phi_\alpha(x_i)} \tag{15}$$

for $\alpha = 1, 2, \ldots, m$. An estimate of $Var(\hat{\theta}_\alpha)$ can be found by substituting $\hat{\theta}_\alpha$ for θ_α. Being a sum of random variables, which have Poisson distributions, $\hat{\theta}_\alpha$ also has the Poisson distribution. Thus, we conclude that the method which is standardly used to estimate activities (see Section 3) may be considered as an approximate one and there exist statistically more efficient methods. The iterative procedure described above is easily realized with modern software. The superiority becomes especially evident when the background noise is comparable with the height peaks and peaks from various elements are overlapping.

Appendix 3. Models and Analysis of Sampling Experiments

Models. In the sampling experiment there are at least three types of uncertainties, which have to be considered and modeled:

1. Sampling of items from the population of items of different types, exposure, cleaning treatment, etc.

2. All emitting processes are random. Correspondingly, all the observed "counts" must be treated as random variables.

3. Observational errors, for instance, the presence of a background noise, can be a significant addition to the above mentioned sources of uncertainty.

The uncertainties of type (a) are probably the largest one. A proper inventory and stratification procedure may reduce them, but not remove them completely.

One of the simplest models used in many applied studies may be described in the "waste characterization" setting as follows:

Let x_0 be a contamination level (e.g. TRU activity) of an item at some stage of its active life. Then the probability that this level will be increased by x does not depend upon the existing contamination level x_0. In other words, there are equal chances to increase contamination by a given amount either for a "dirty" or "clean" item. In the language of probability, our assumption can be described as

$$\text{Probability } (x_0 \leq X < x_0 + x) = \text{Probability } (0 \leq X < x). \qquad (1)$$

The corresponding probability density function (pdf) that satisfies equation (1) is well known:

$$p(x) = \sigma^{-1} e^{-x/\sigma}, \qquad (2)$$

and is called the exponential distribution; see [15] for more details. For the pdf (2), the expected value of contamination equals σ. We understand that model (1) and (2) are an approximate description of reality. Nevertheless, we prefer to use this model versus the more traditional Gaussian or normal distribution, because unlike the latter it has

a better physical explanation in our setting. In addition, it results in relatively simple estimation procedures.

The probability models for the uncertainties of type (b) are well known in nuclear physics and we confine our choice to the Poisson distribution where the probability that we observe x counts in a given unit of time is

$$p(x) = \frac{\lambda^x e^{-\lambda}}{x!}. \tag{3}$$

In this particular setting, the observational errors are comparatively small and the choice of the models can vary for different measurement procedures. In this appendix we address the first two types of uncertainties.

Analysis of proportion. Let us assume that in a sampling experiment we can measure two components x_1 and x_2. In our instance, α-activity of TRU may be selected as x_1, and α-activity of all other isotopes as x_2. Let us further assume that the ratio of x_1 and x_2 is rather stable. In terms of model (2) it means that

$$\sigma_2 = k\sigma_1, \tag{4}$$

where $\sigma_\alpha, \alpha = 1, 2$, is the expected value of the corresponding distribution.

Assuming that the realizations x_{1i} and $x_{2i}, i = 1, \ldots, n$, are independent and known exactly, we can introduce the following maximum likelihood function

$$L(x_1, x_2, \sigma, k) = \prod_{i=1}^{n} \sigma^{-2} k^{-1} \exp\left(\sigma^{-1}(x_{1i} + k^{-1} x_{2i})\right) \tag{5}$$

where $\sigma = \sigma_1$.

Straightforward maximization of (5) leads to the maximum likelihood estimator:

$$\hat{k} = \frac{\overline{x}_2}{\overline{x}_1}, \tag{6}$$

where $\overline{x}_\alpha = \frac{1}{n} \sum_{i=1}^{n} x_{\alpha i}, \alpha = 1, 2$.

When x_α are activities of some elements, then they may be measured through some

- 55 -

counting procedures, i.e. instead of $x_{\alpha i}$ we know $y_{\alpha i}$, which are distributed (see (3)) as

$$p(y_\alpha | x_{\alpha i}) = \frac{x_{\alpha i}^{y_\alpha} \, e^{-x_{\alpha i}}}{y_\alpha !} \tag{7}$$

and

$$
\begin{aligned}
p(y_\alpha | \sigma_\alpha) &= \int_0^\infty p\,(y_\alpha | x_{\alpha i}) p(x_{\alpha i} | \sigma_\alpha) \; dx_{\alpha i} \\
&= \int_0^\infty \frac{x_{\alpha i}^{y_\alpha} \, e^{-(1+1/\sigma\alpha)x_{\alpha i}}}{y_\alpha ! \sigma_\alpha} dx_{\alpha i} \\
&= \frac{\sigma_\alpha^{y_\alpha}}{(1+\sigma_\alpha)^{y_\alpha + 1}}.
\end{aligned}
\tag{8}
$$

In (8) we used the formula

$$\int_0^\infty x^{y-1} e^{-ax} dx = a^{-y}\Gamma(y).$$

Since $\sigma_2 = k\sigma_1$ and from (8) we have that

$$L(\mathbf{y}_1, \mathbf{y}_2, \sigma, k) = \prod_{i=1}^n \frac{\sigma^{y_{1i}}(k\sigma)^{y_{2i}}}{(1+\sigma)^{y_{1i}+1}(1+k\sigma)^{y_{2i}+1}} \tag{9}$$

where $\mathbf{y}_\alpha$ is the $n \times 1$ vector of $y_{\alpha i}, \alpha = 1, 2$, and $\sigma = \sigma_1$.

Again, straightforward maximization of (9) shows that the maximum likelihood estimator for k is

$$\hat{k} = \frac{\overline{y}_2}{\overline{y}_1} \tag{10}$$

where $\overline{y} = \frac{1}{n}\sum_{i=1}^n y_{\alpha i}$ and $\alpha = 1, 2$.

Alternative model. In the previous section, we assumed that x_{1i} and x_{2i} are "generated" independently of each other. The link between σ_1 and σ_2 is actually some loose link between the means of these two random variables. Perhaps in the case of contaminated objects, it is expedient to assume a stronger relationship between x_{1i} and x_{2i}. Namely, we assume that

$$x_{2i} = kx_{1i} \tag{11}$$

- 56 -

In other words, the ratio, x_{2i}/x_{1i}, between various components for all selected items is the same. Do not forget that we observe "counts" y_{1i} and y_{2i}, in general. Unlike the previous section, where

$$p(y_1, y_2, |\sigma) = p(y_1|\sigma)\, p(y_2|\sigma) \tag{12}$$

i.e. y_1 and y_2 are independent, now observations are dependent and we cannot use (8) to derive the common probability density for y_1, and y_2.

However, it still has a simple form, which may be found through direct integration:

$$
\begin{aligned}
p(y_1, y_2|\sigma) &= \int_0^\infty p(y_1, y_2|x_1)\ p(x_1|\sigma)dx_1, \\
&= \int_0^\infty \frac{x_1^{y_1} e^{-x_1}}{y_1!}\ \frac{(kx_1)^{y_2} e^{-kx_1}}{y_2!}\ \frac{1}{\sigma}e^{-x_1/\sigma}dx_1, \\
&= \frac{(y_1+y_2)!}{y_1! y_2!}\ \frac{(k\sigma)^{y_2}\sigma^{y_1}}{(k\sigma+\sigma+1)^{(y_1+y_2+1)}}
\end{aligned}
\tag{13}
$$

Now the maximum likelihood function equals

$$L(\mathbf{y_1}, \mathbf{y_2}, \sigma, k) = \prod_{i=1}^n \frac{(y_{1i}+y_{2i})!}{y_{1i}! y_{2i}!} \cdot \frac{\sigma^{y_{1i}}(k\sigma)^{y_{2i}}}{(k\sigma+\sigma+1)^{y_{1i}+y_{2i}+1}}. \tag{14}$$

Amazingly, after direct maximization of (14), we have the following maximum likelihood estimator for k:

$$\hat{k} = \frac{\overline{y}_1}{\overline{y}_2}. \tag{15}$$

Thus, in all three cases considered, we have the same formula for $\hat{k}$. In all three cases we need to know the arithmetic means ($\overline{y}_1$ and $\overline{y}_2$) to get the maximum likelihood estimators. For the reader with some statistical background, we note that $\overline{y}_1$ and $\overline{y}_2$ are sufficient statistics for k and σ. At a less technical level, one can say that $\overline{y}_1$ and $\overline{y}_2$ contain all the information in the frame of the considered models about k and σ.

Nuisance parameters approach. In the previous sections, it was assumed that the contamination of items has the exponential distribution. In spite of the existence of very reasonable physical background to use this distribution, the validity of the assumptions, which were made, can be questioned. In this section we present another approach, which is free from those assumptions (see (1), (2), (4)). However, the assumptions (3)

- 57 -

and (11) are essential for the reported result. These two assumptions state that the distribution of activity (counts) measurements have the Poisson distribution and that the ratio of activities is constant for all items. Our main objective is to find this ratio.

From (3) and (11) we can derive that

$$L(\mathbf{y_1}, \mathbf{y_2}, x_1, k) = \prod_{i=1}^{n} \frac{x_{1i}^{y_{1i}} e^{-x_{1i}}}{y_{1i}!} \frac{(kx_{1i})^{y_{2i}} e^{-kx_{1i}}}{y_{2i}!} . \tag{16}$$

Variables $x_{1i}, i = 1, \ldots, n$, may be considered as nuisance parameters. Direct maximization of (16) shows immediately that we can construct the maximum likelihood estimator for k, which does not depend upon x_{1i}, and is given by

$$\hat{k} = \frac{\overline{y}_1}{\overline{y}_2}. \tag{17}$$

Again, we have come to the same formula.

PLOTS

FIGURES 1 - 10

Legend Symbols Used in Figures

×	Observations where at least one component is below detection level (Figures 1 and 2);
*	Observations where all components are known (Figures 1 and 2);.
C, G, M, P, and W	Observations whose source is cell (C), glass (G), metal (M), poly (P) or wipe (W) (Figures 3 and 4);
U, L, or 0	Observation derived using upper component activity value (V), lower component value (L), or derived when $U = L$ (Figures 5 and 6);
+	Observation (upper or lower bound) used in regression analysis (Figures 7 - 10).
	Observations marked by their "sample ID" are outlier observations or associated with the observations which were discussed in text; (see Table A5).

FIG. 1a   LN TRU ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=1
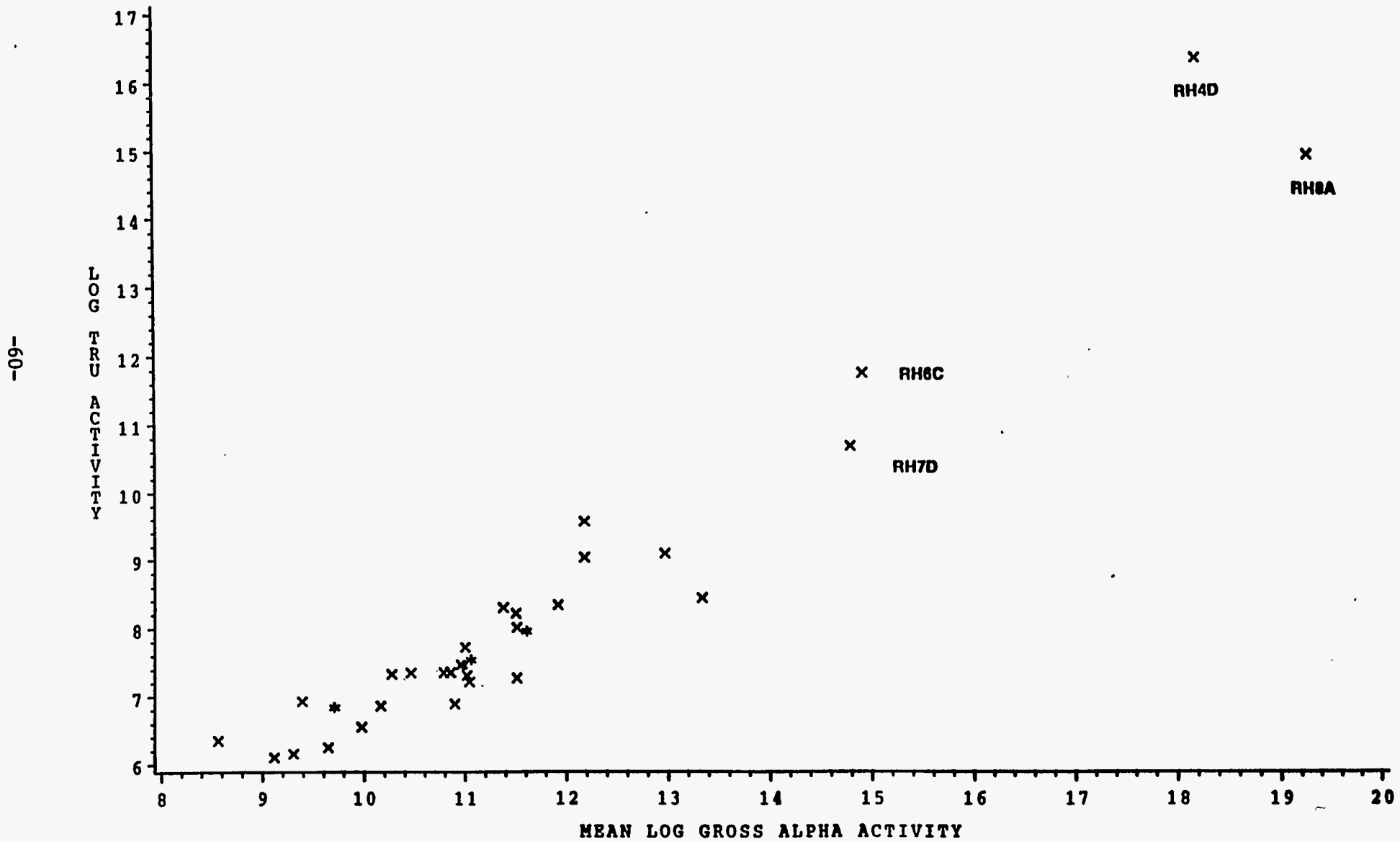
FIG. 1b   LN CF252 ACTIVITY VS LN GROSS ALPHA
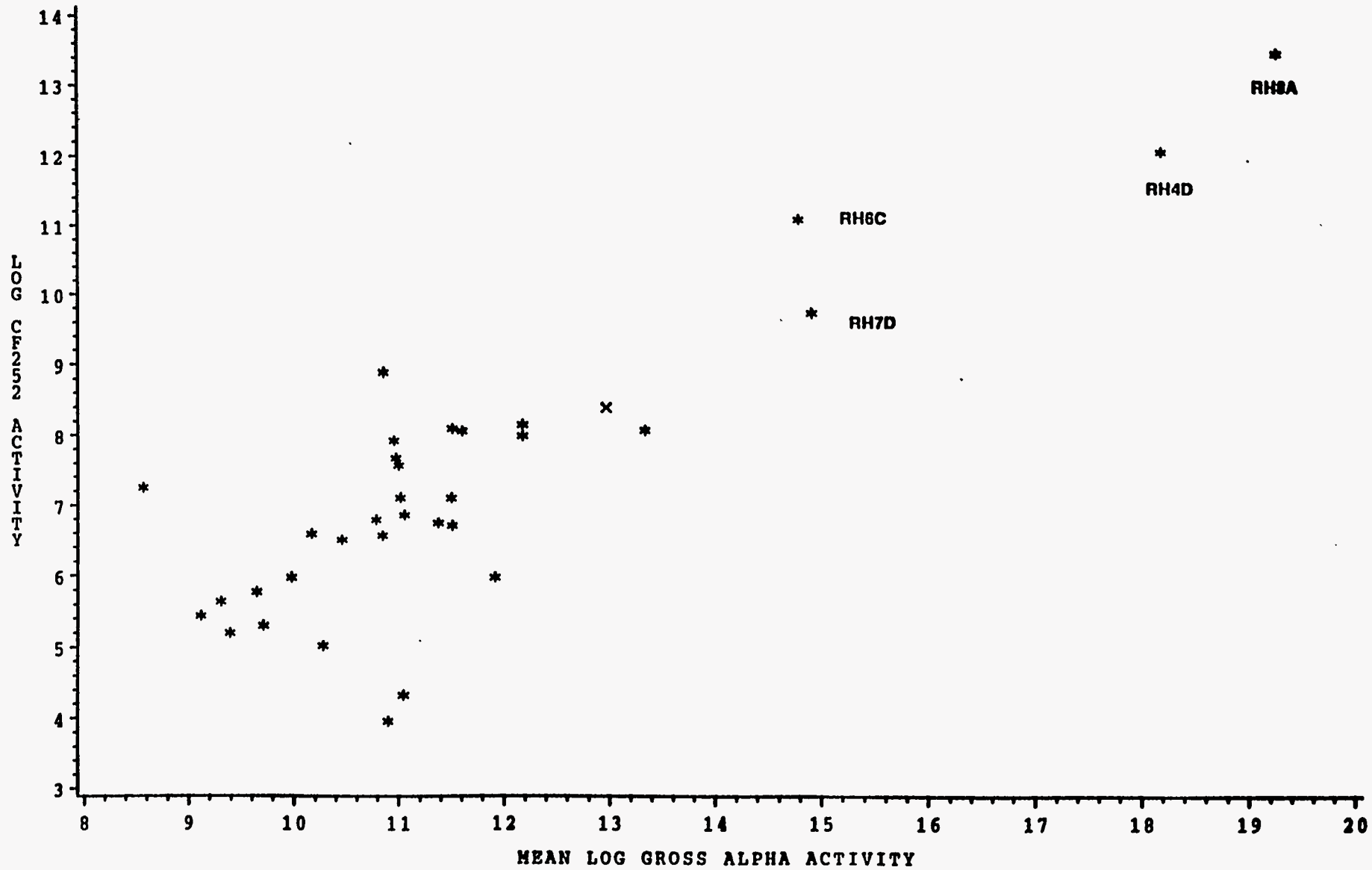
COMPLETE DATA SET
EXP=1

-61-

FIG. 1c   LN CM244 ACTIVITY VS LN GROSS ALPHA
COMPLETE DATA SET
EXP=1
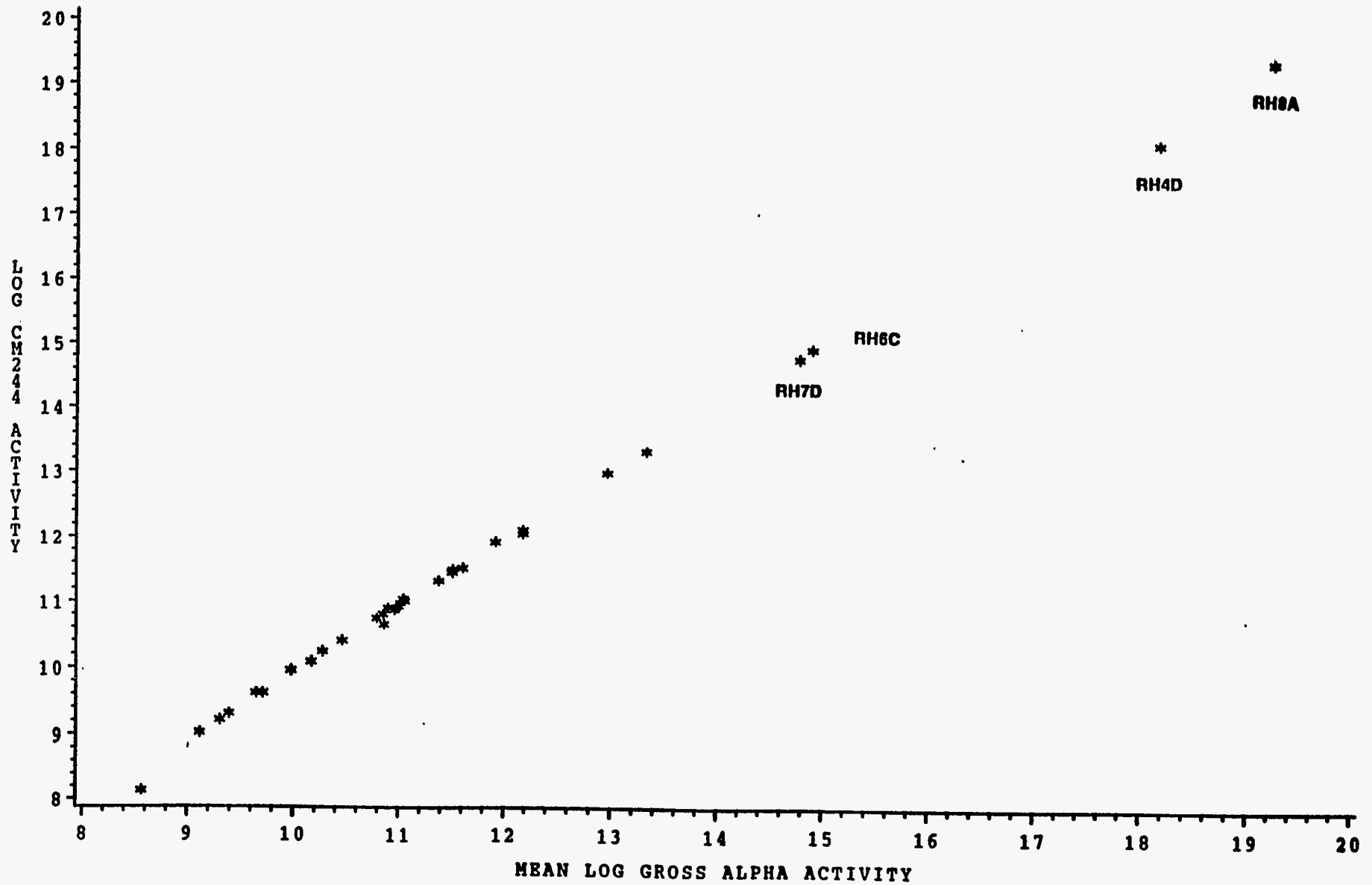
LOG CM244 ACTIVITY

MEAN LOG GROSS ALPHA ACTIVITY

RH8A
RH4D
RH6C
RH7D

-62-

FIG. 2a   LN TRU ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=2

MEAN LOG GROSS ALPHA ACTIVITY
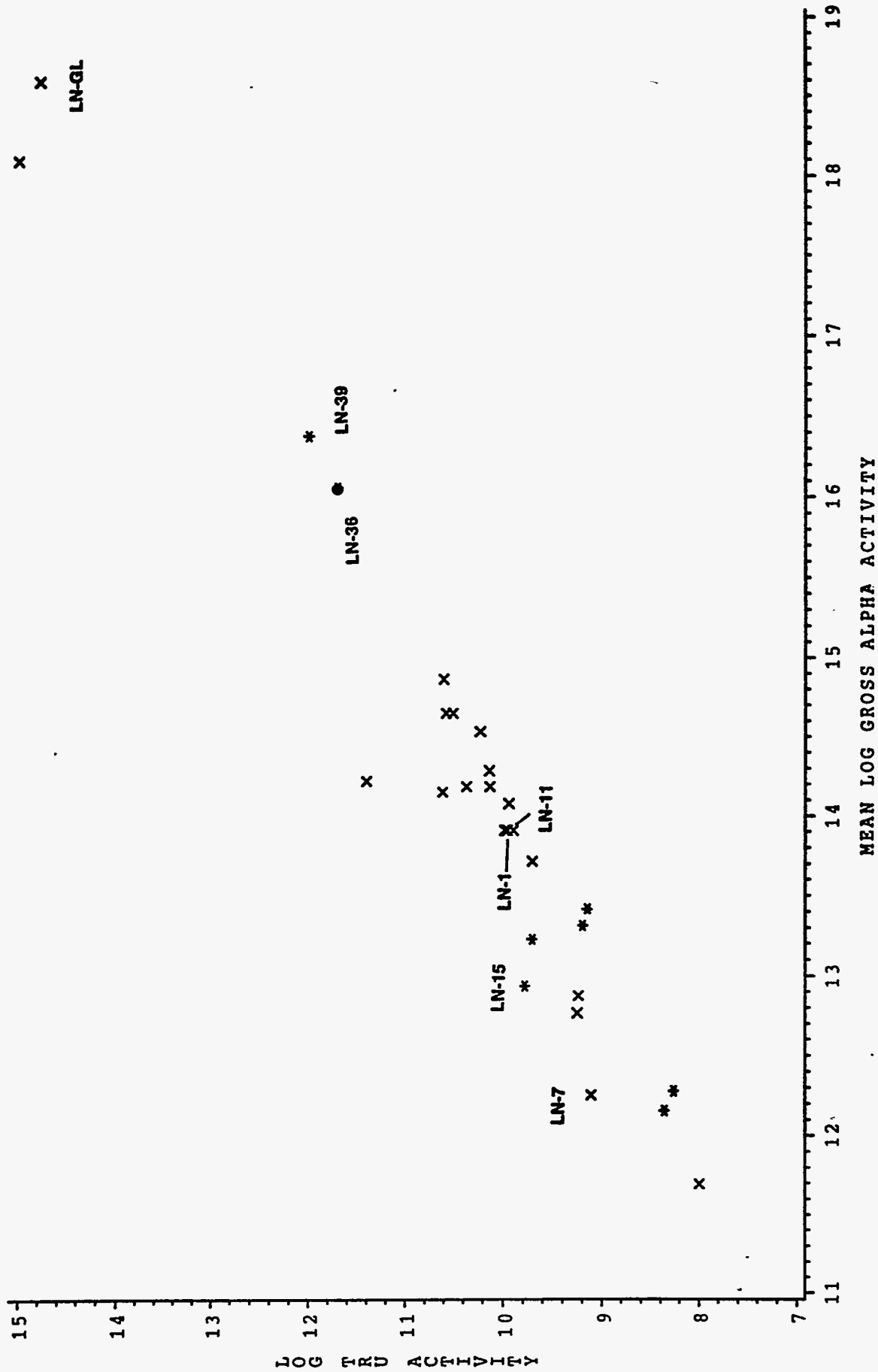
LOG TRU ACTIVITY

FIG. 2b   LN CF252 ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=2

-64-
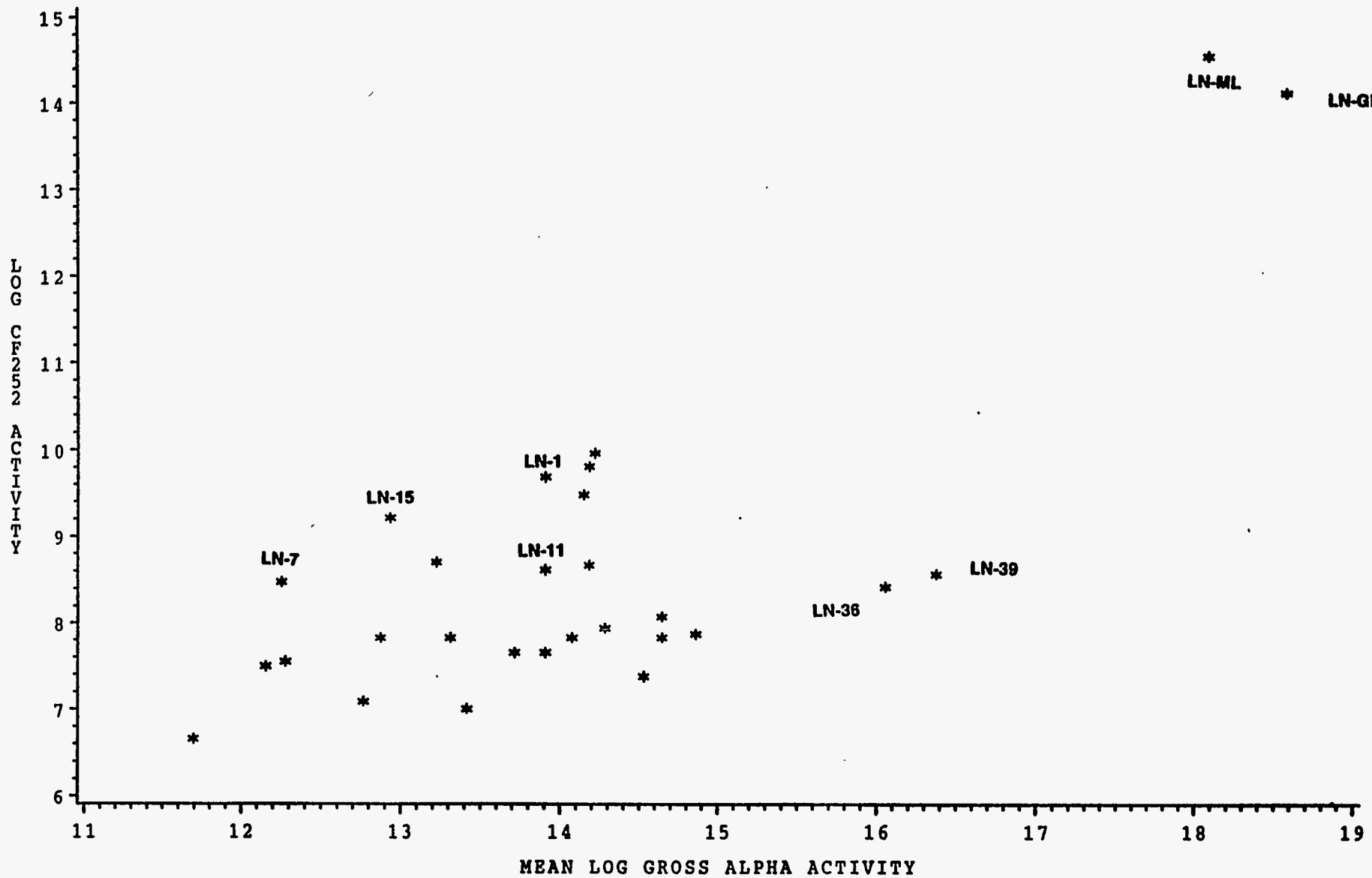
FIG. 2c   LN CM244 ACTIVITY VS LN GROSS ALPHA
COMPLETE DATA SET
EXP=2
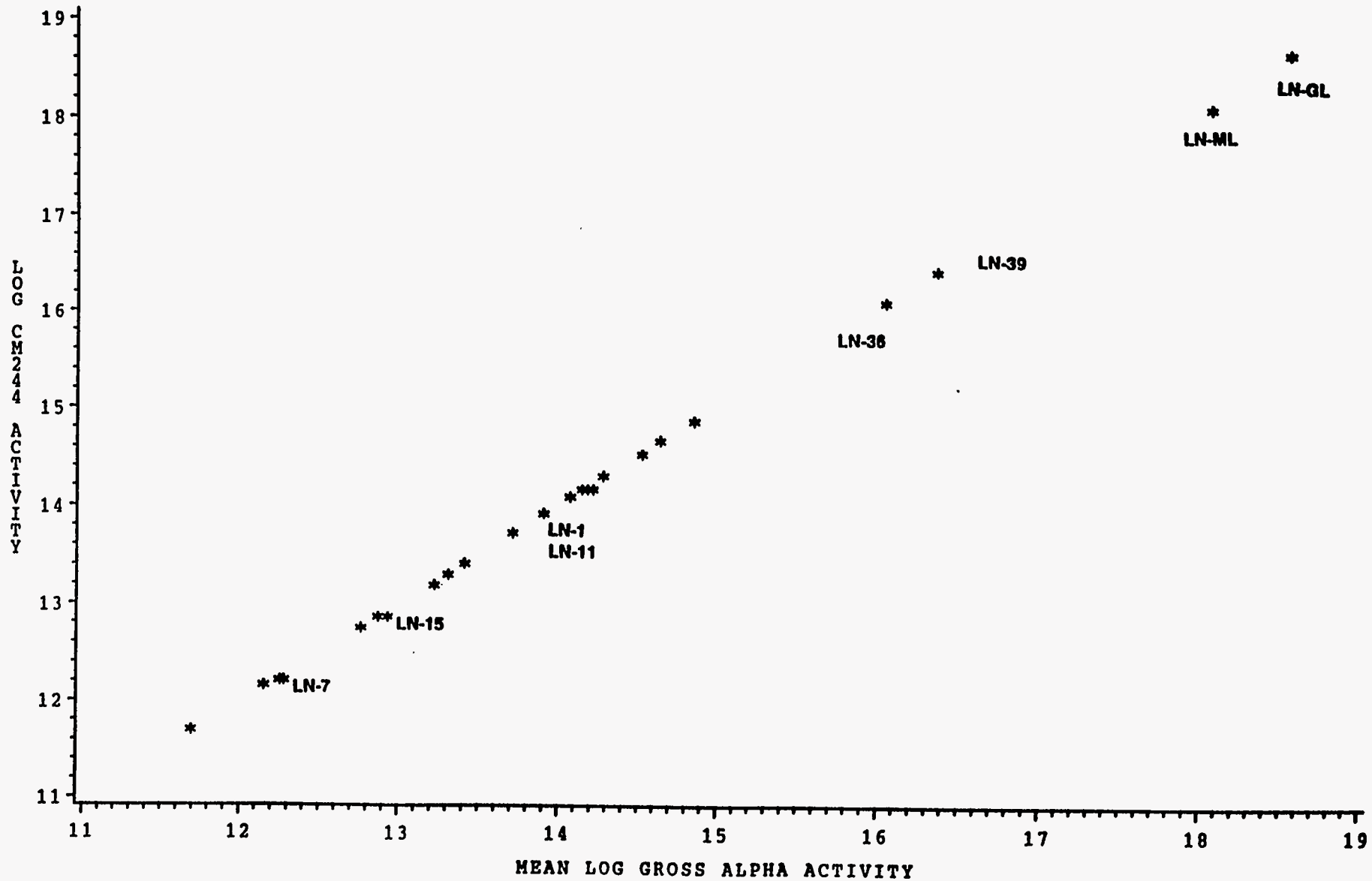
-65-

FIG. 3a   LN TRU ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=1

-99-

LOG TRU ACTIVITY (y-axis)

MEAN LOG GROSS ALPHA ACTIVITY (x-axis)

FIG. 3b   LN CF252 ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=1

Scatter plot with Y-axis labeled "LOG CF252 ACTIVITY" ranging from 3 to 14, and X-axis labeled "MEAN LOG GROSS ALPHA ACTIVITY" ranging from 8 to 20.

Notable labeled points: C (≈19, 13.2), RH8A (≈18.5, 12.7), C (≈18, 11.8), RH4D, C (≈14.8, 11), RH6C, C (≈15, 9.6), RH7D.
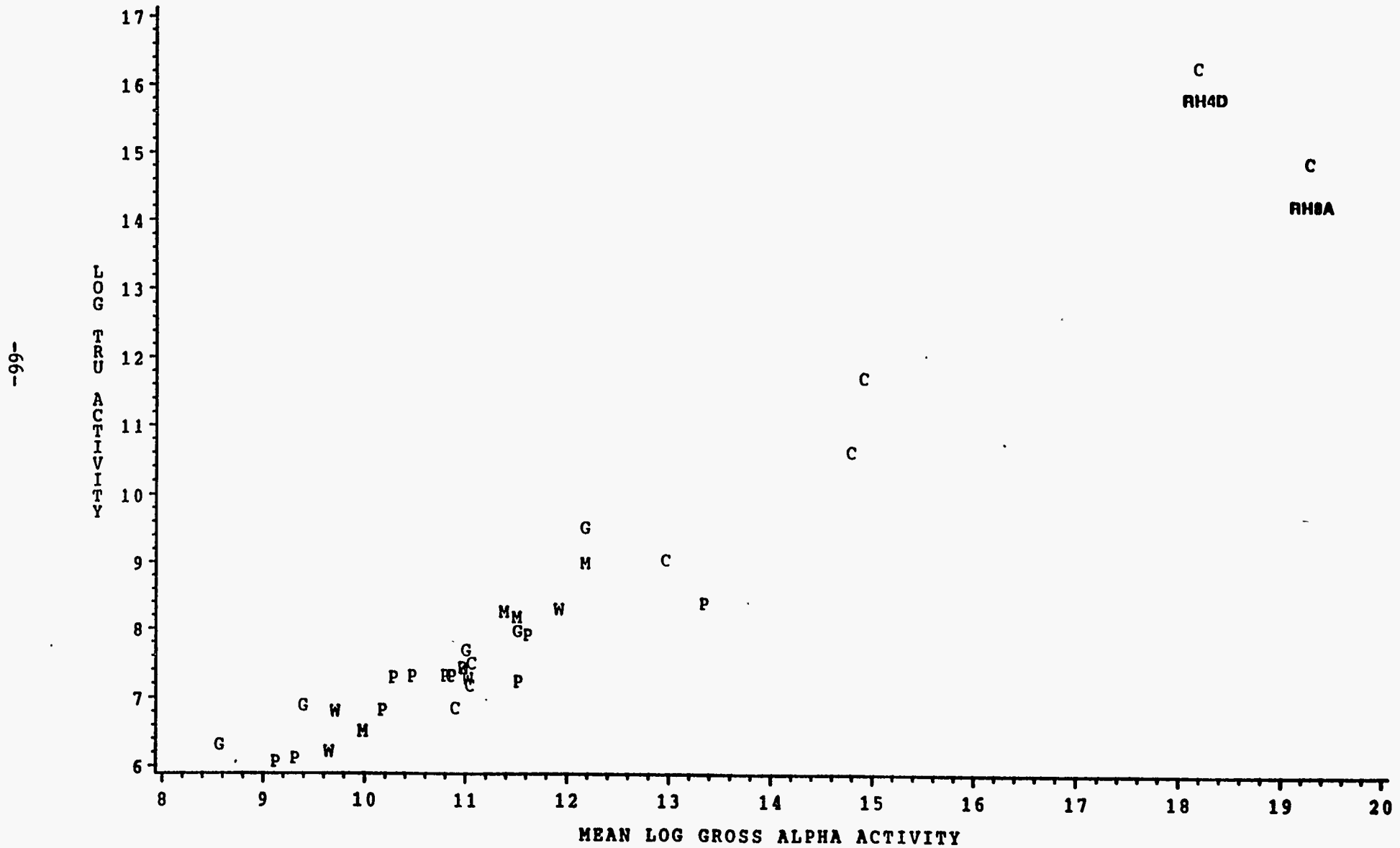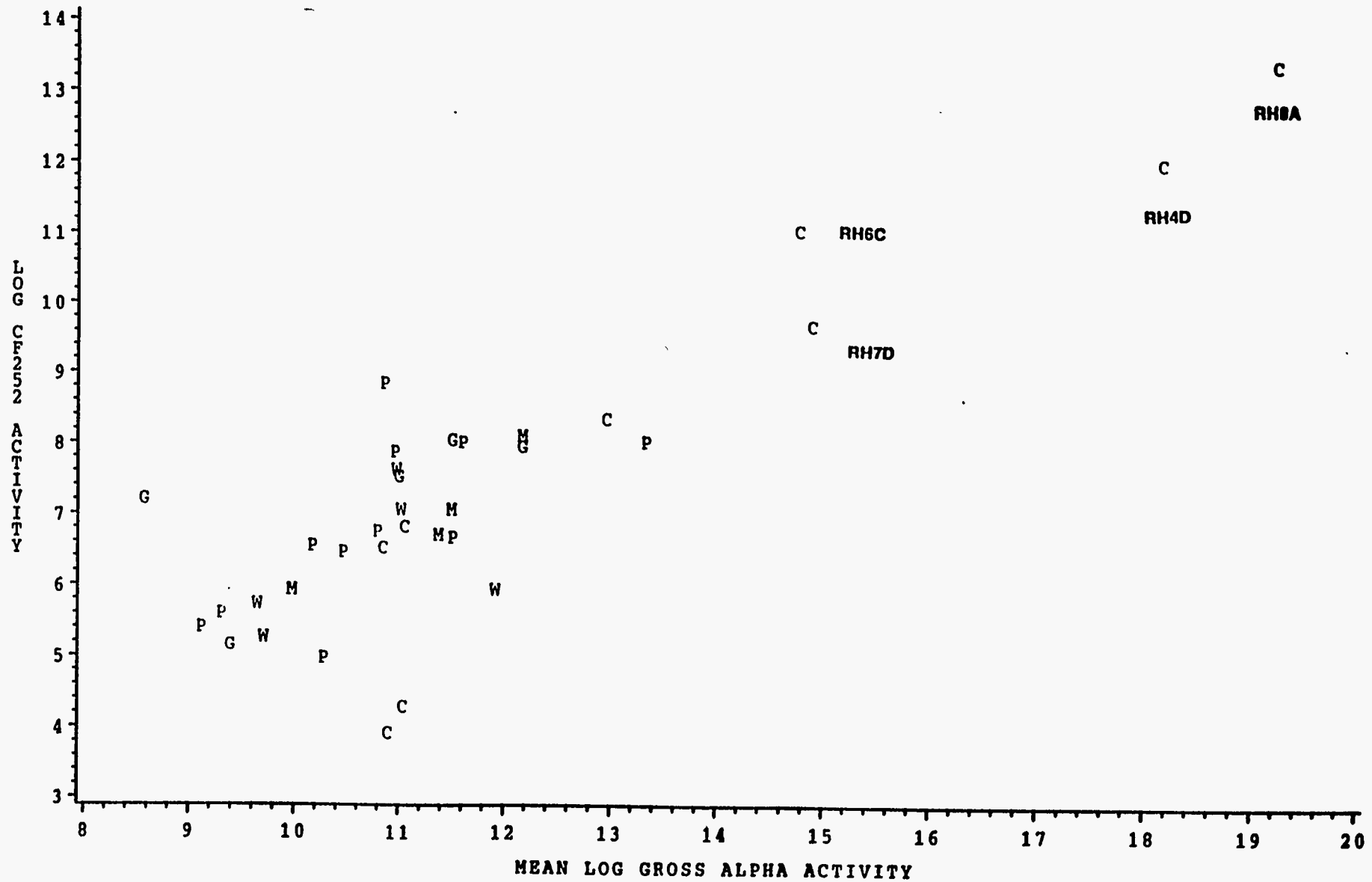
FIG. 3c   LN CM244 ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=1

-89-

MEAN LOG GROSS ALPHA ACTIVITY
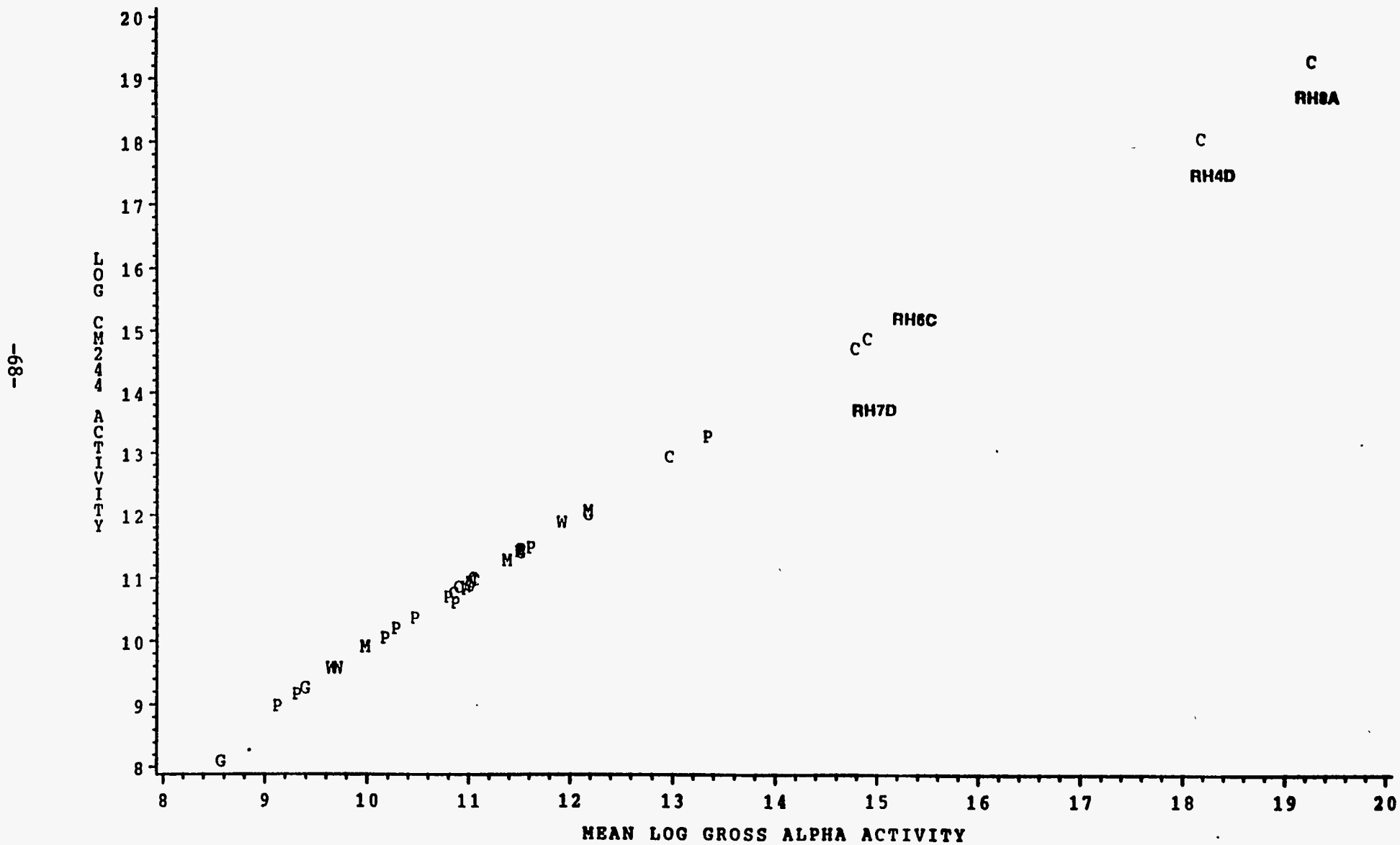
FIG. 4a   LN TRU ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=2

FIG. 4b   LN CF252 ACTIVITY VS LN GROSS ALPHA

COMPLETE DATA SET
EXP=2

-70-
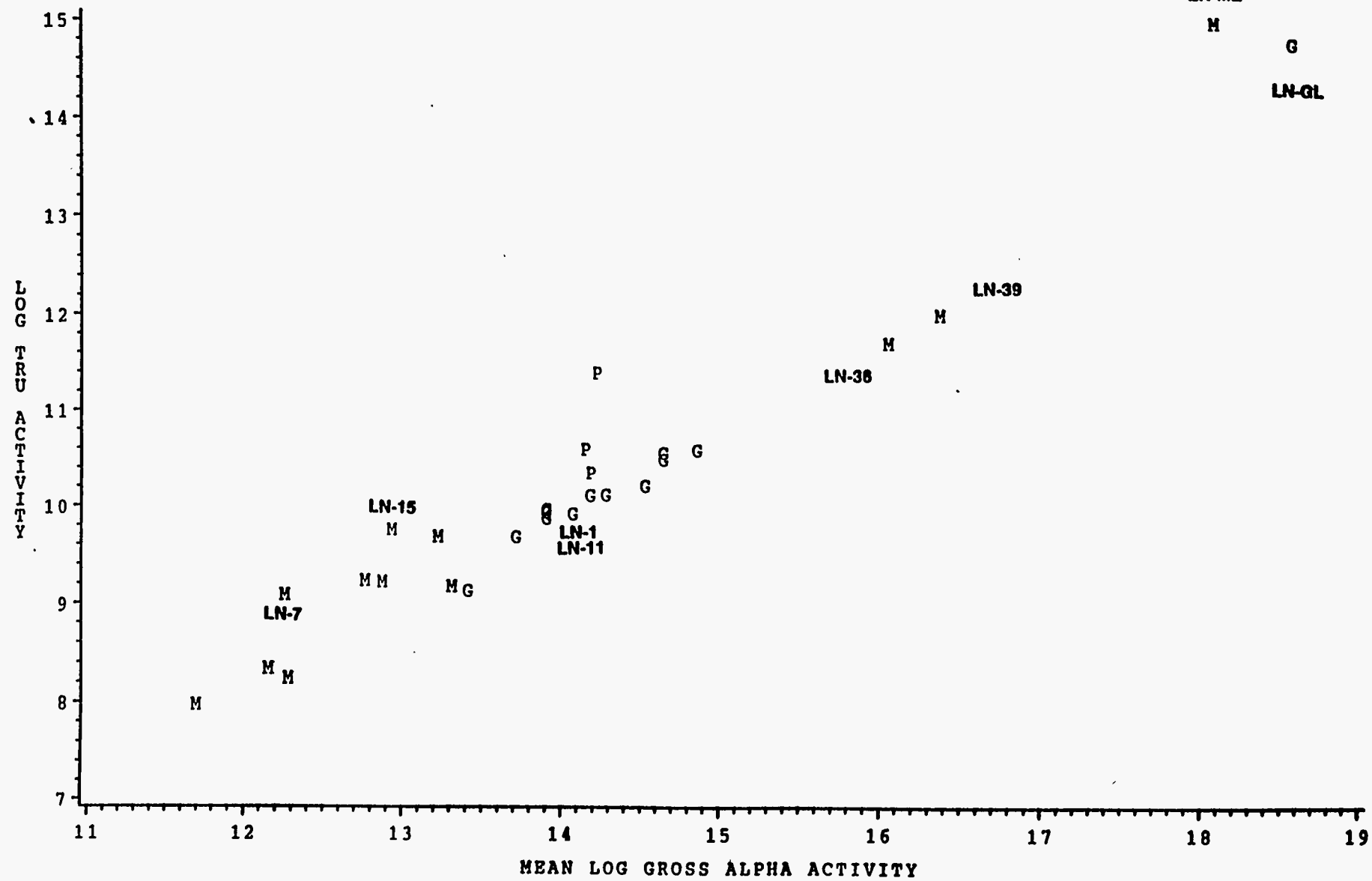
MEAN LOG GROSS ALPHA ACTIVITY
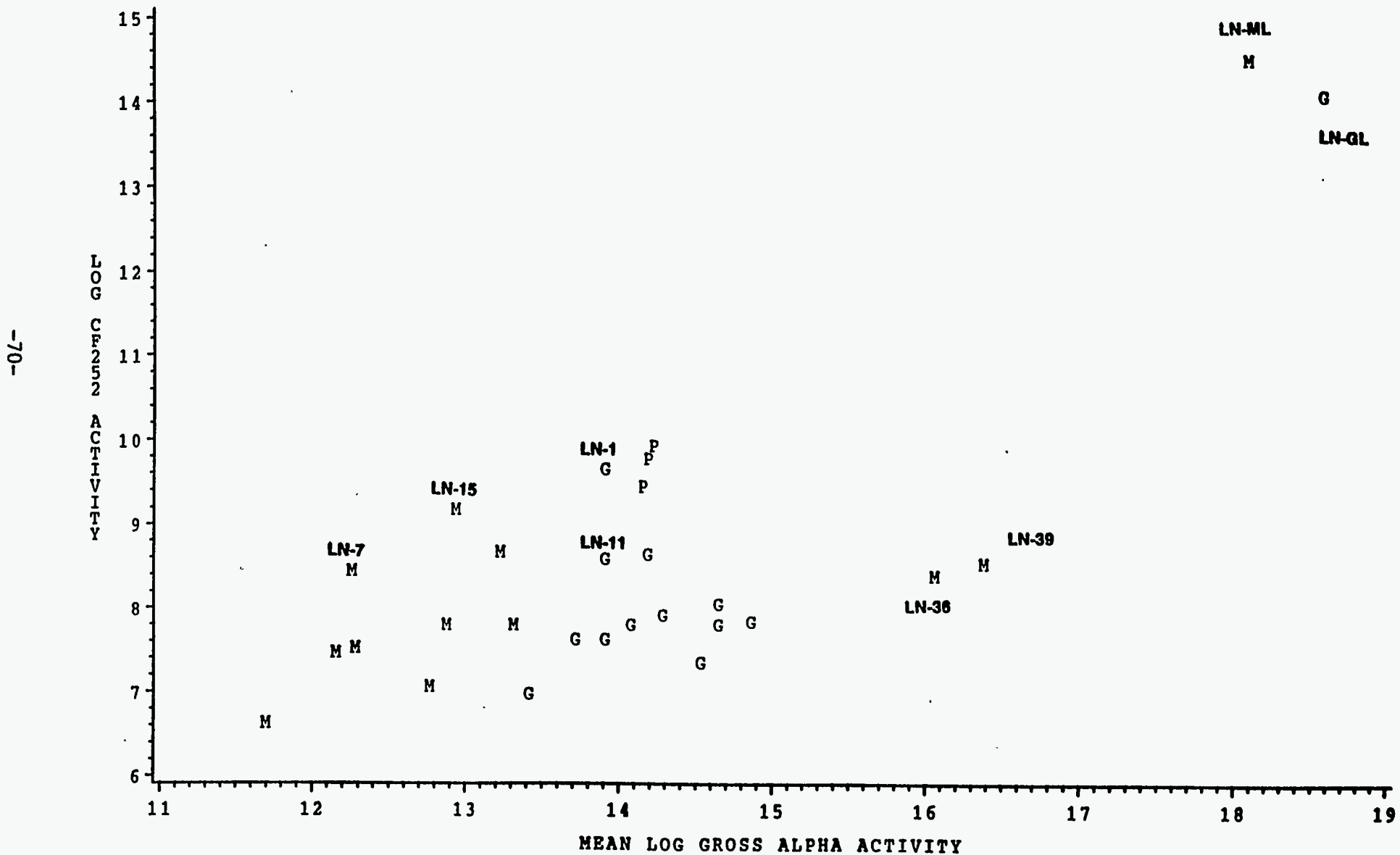
FIG. 4c . LN CM244 ACTIVITY VS LN GROSS ALPHA
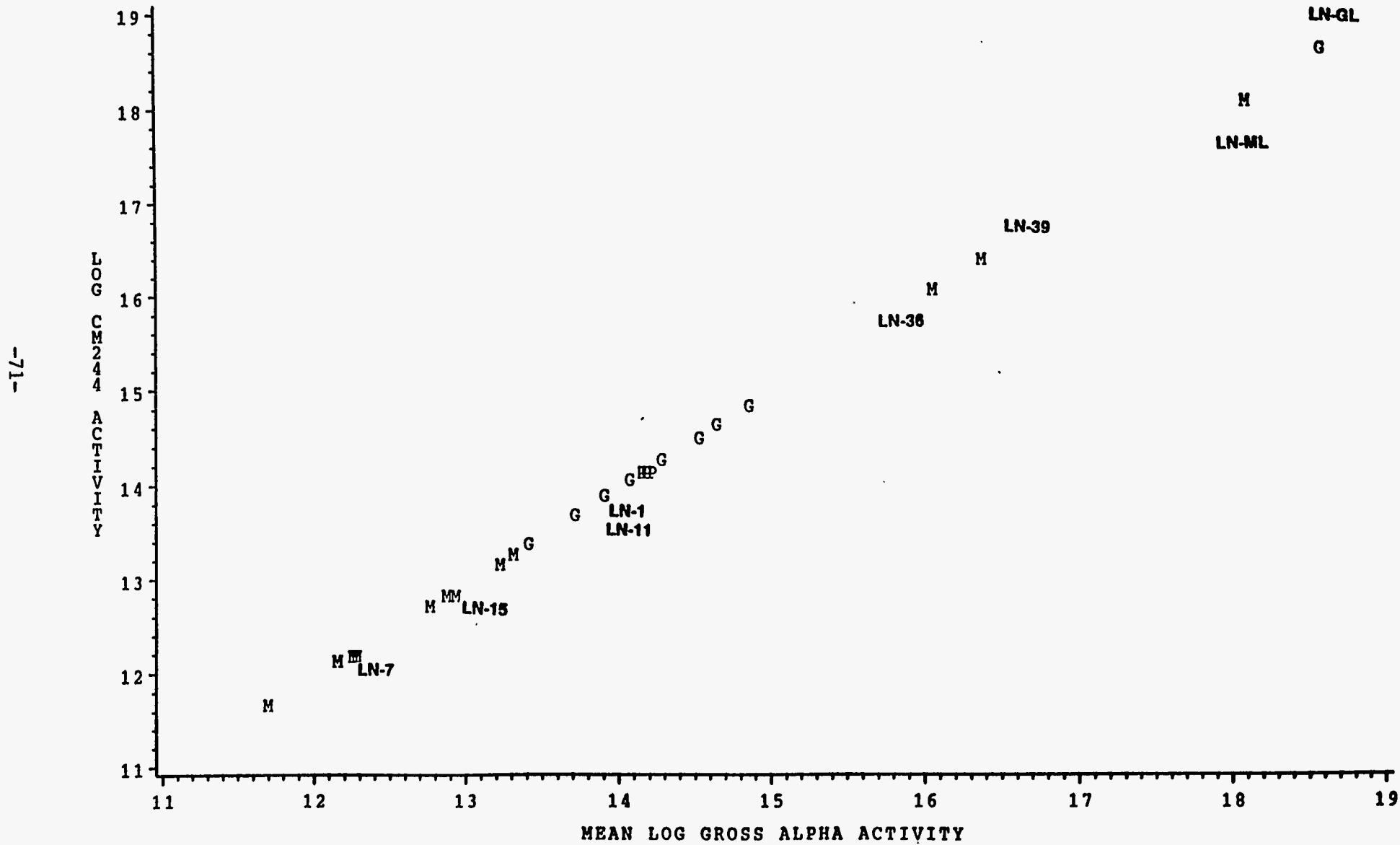
COMPLETE DATA SET
EXP=2

FIG. 5 . UPPER AND LOWER LN TRU ACTIVITIES VS MEAN LN GROSS ALPHA
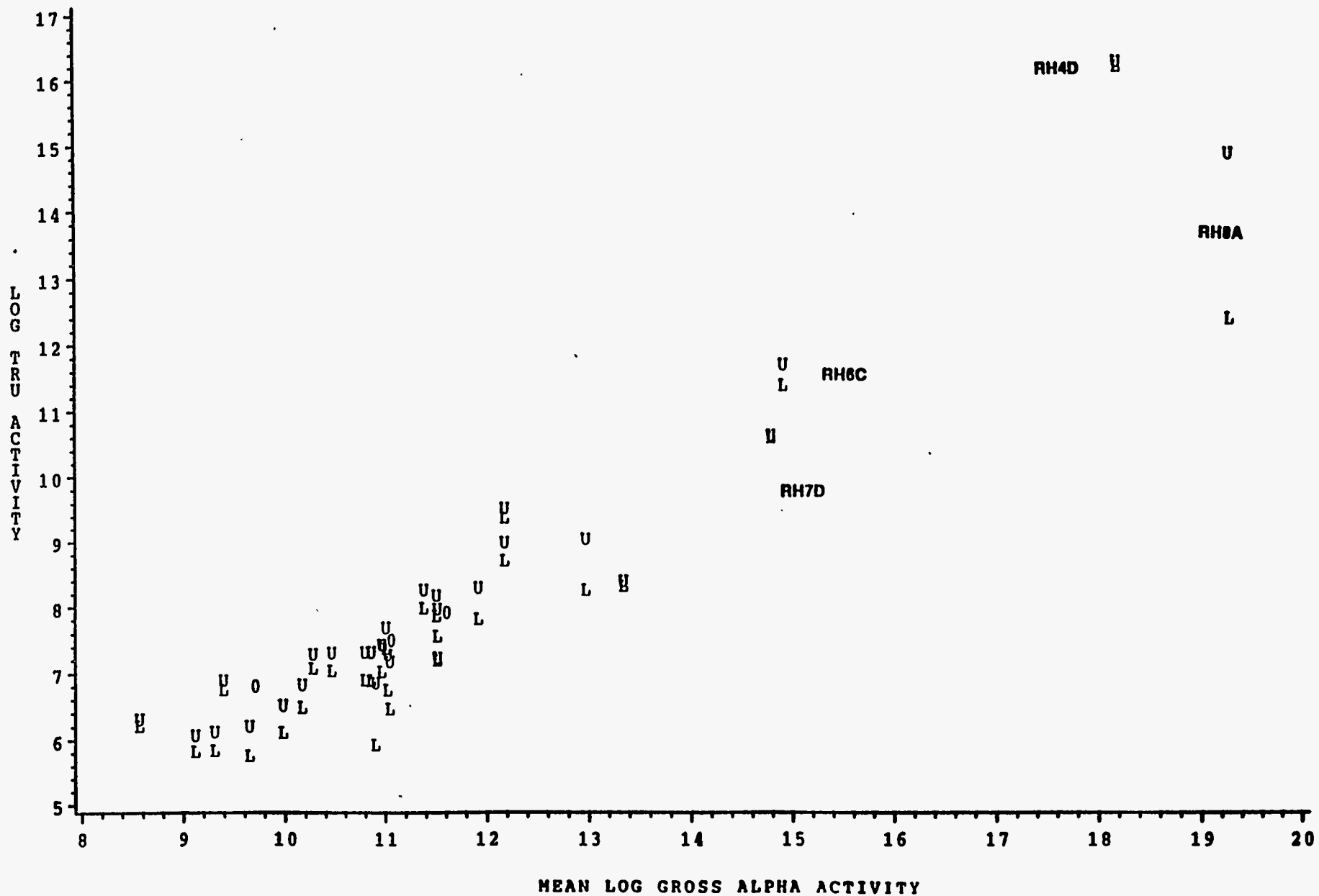
COMPLETE DATA SET
EXP=1

LOG TRU ACTIVITY

MEAN LOG GROSS ALPHA ACTIVITY

-72-

FIG. 6 . UPPER AND LOWER LN TRU ACTIVITIES VS MEAN LN GROSS ALPHA

COMPLETE DATA SET
EXP=2

MEAN LOG GROSS ALPHA ACTIVITY
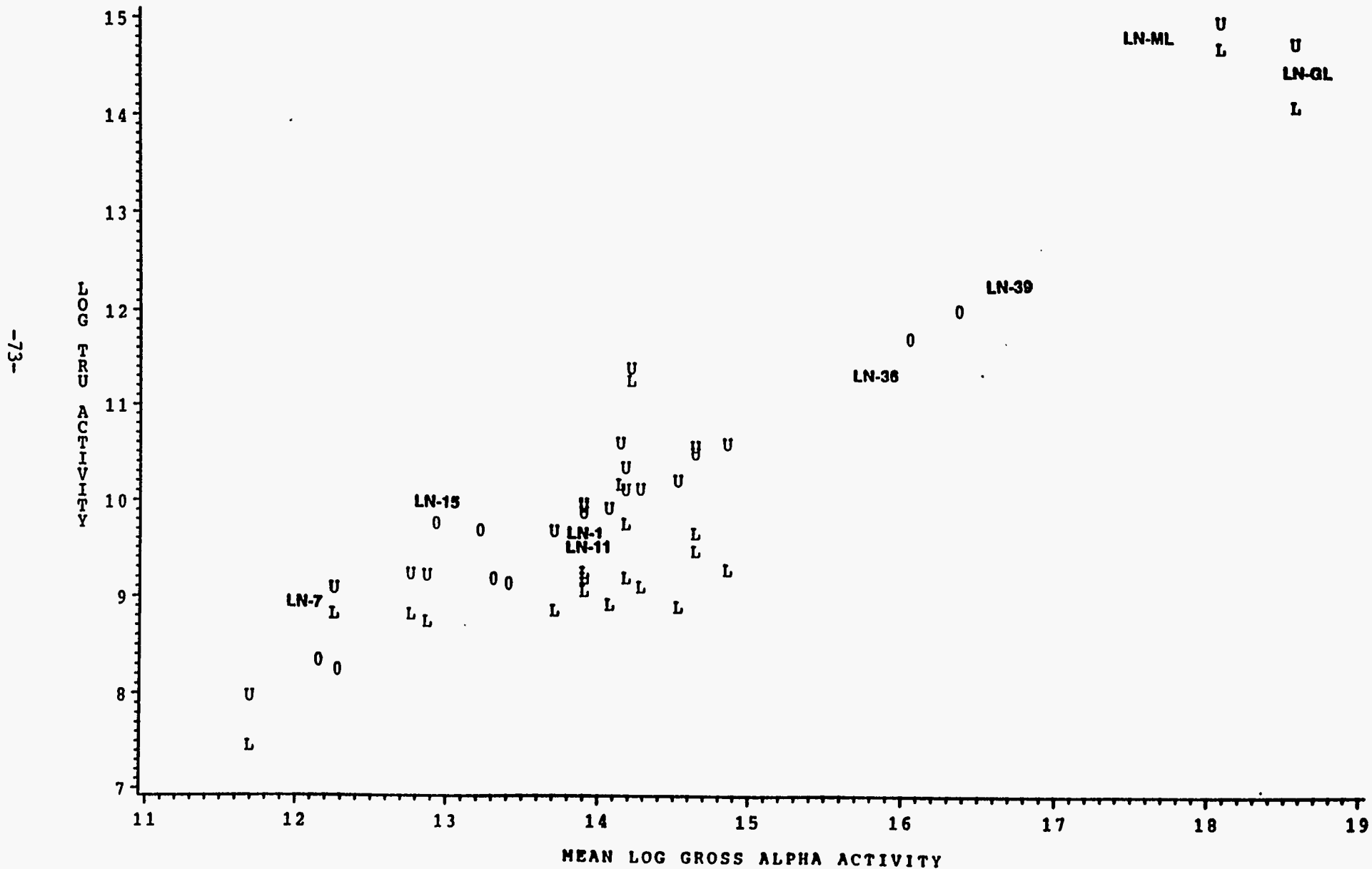
FIG. 7a.   1ST & 2ND ORDER REGRESSION OF LN TRU (LOWER) ON LN GROSS ALPHA
EXPERIMENT 1--NO OBSERVATIONS OMITTED
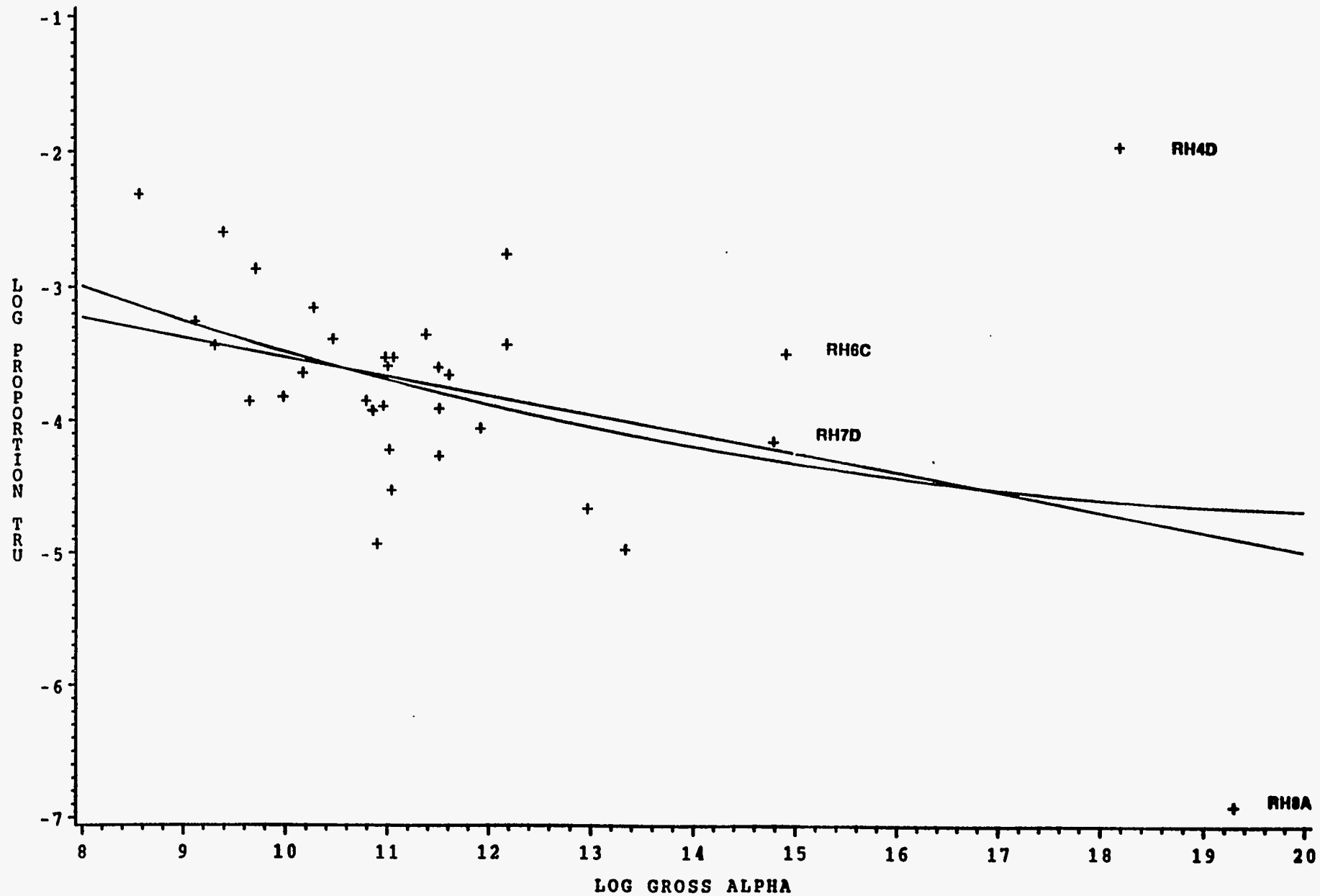
FIG. 7b. 1ST & 2ND ORDER REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA
EXPERIMENT 1--NO OBSERVATIONS OMITTED

-75-
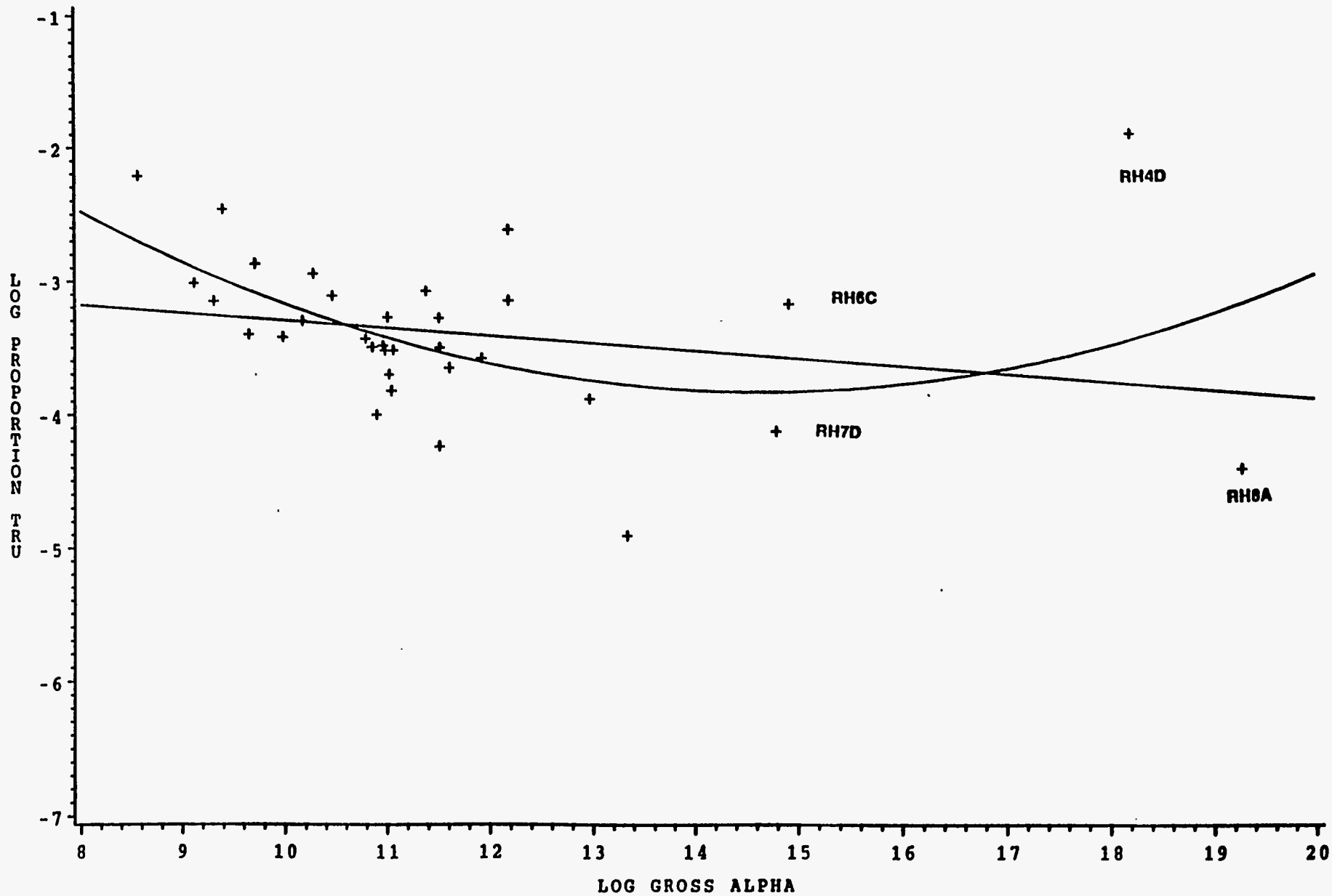
FIG. 7c. 1ST & 2ND ORDER REGRESSION OF LN TRU (LOWER) ON LN GROSS ALPHA

EXPERIMENT 1--2 OUTLIER OBSERVATIONS OMITTED
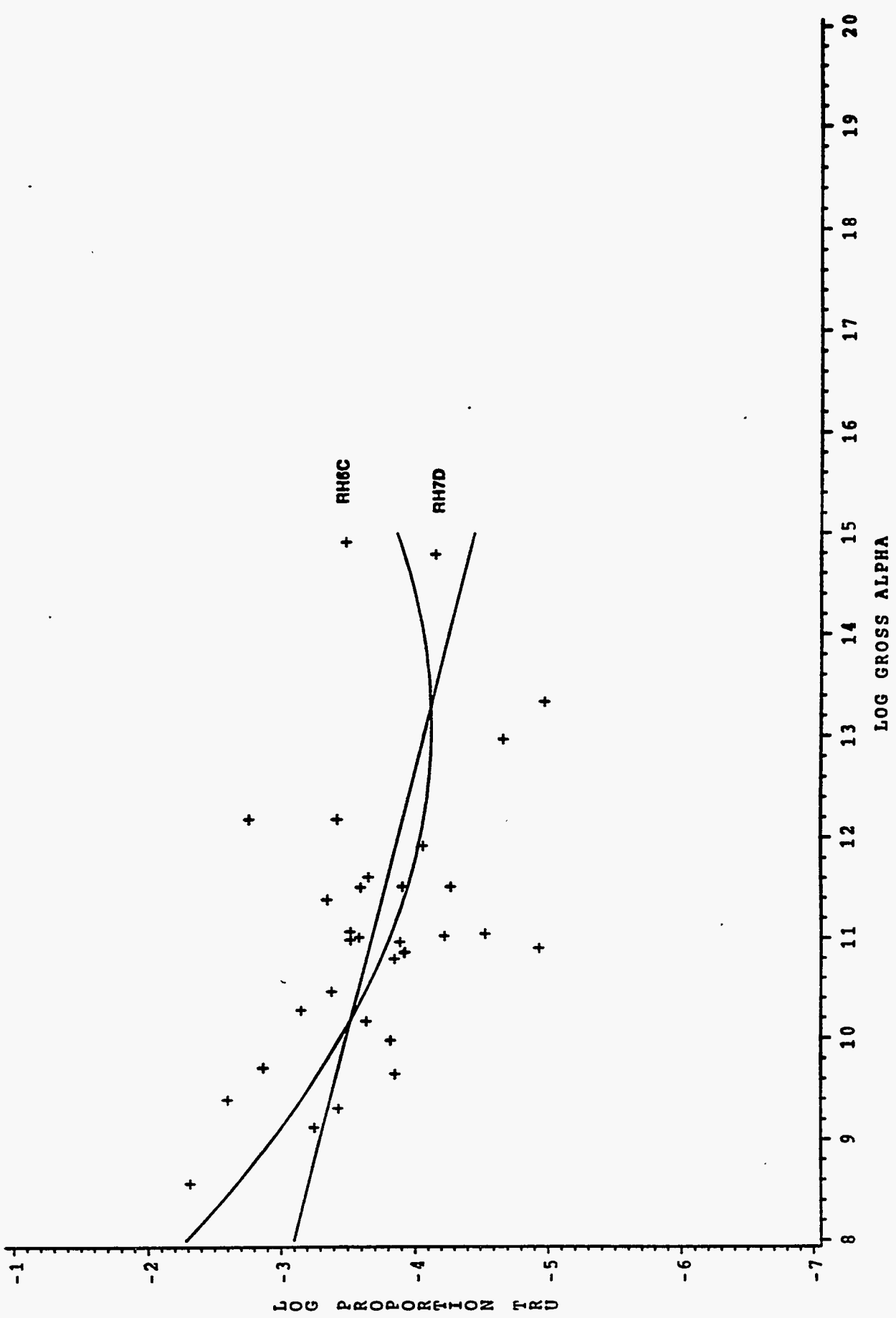
FIG. 7d.   1ST & 2ND ORDER REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA
EXPERIMENT 1--2. OUTLIER OBSERVATIONS OMITTED
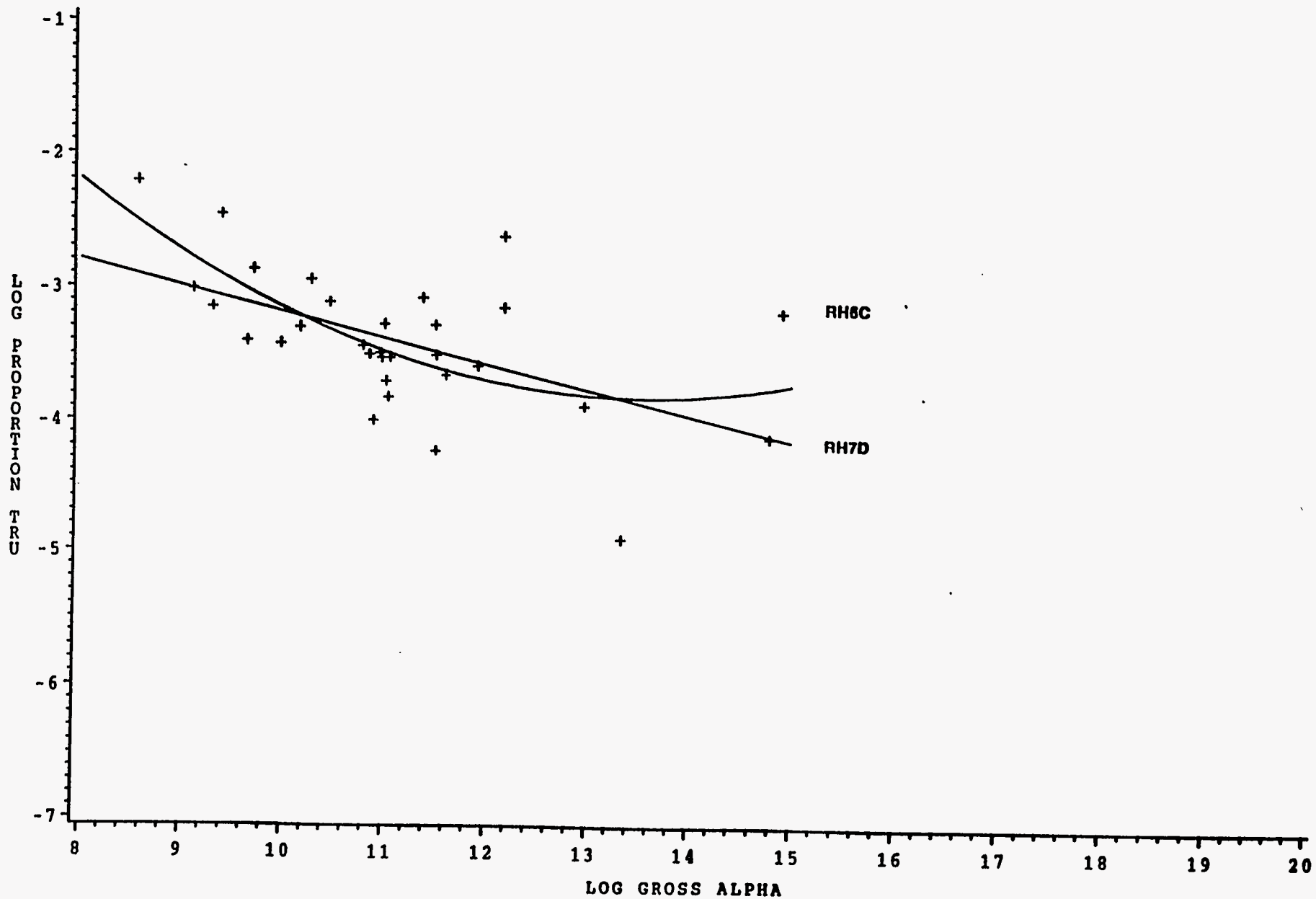
-77-

FIG. 8a.   1ST & 2ND ORDER REGRESSION OF LN TRU (LOWER) ON LN GROSS ALPHA

EXPERIMENT 2--NO OBSERVATIONS OMITTED

-78-
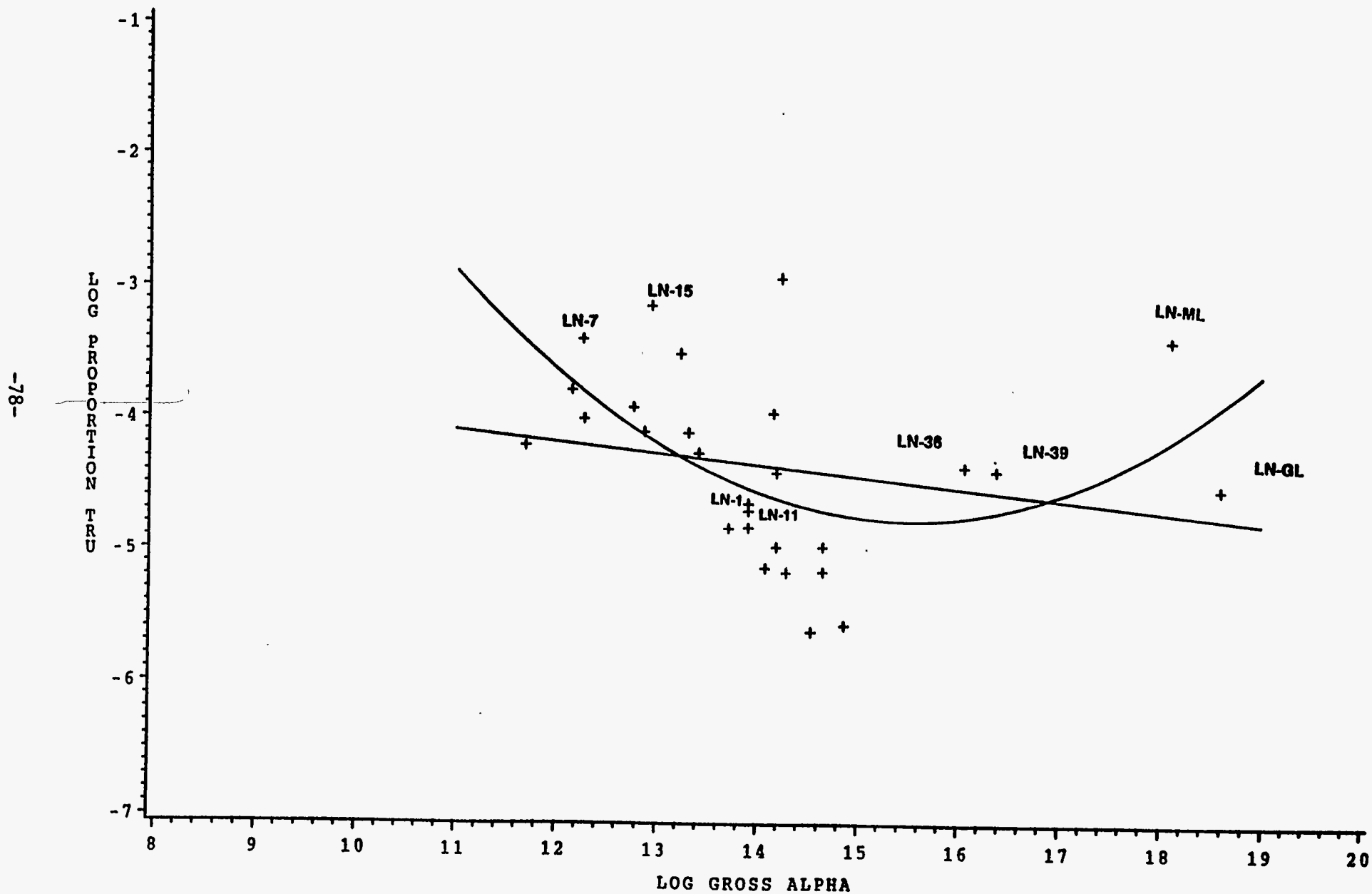
FIG. 8b.   1ST & 2ND ORDER REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA
EXPERIMENT 2--NO OBSERVATIONS OMITTED
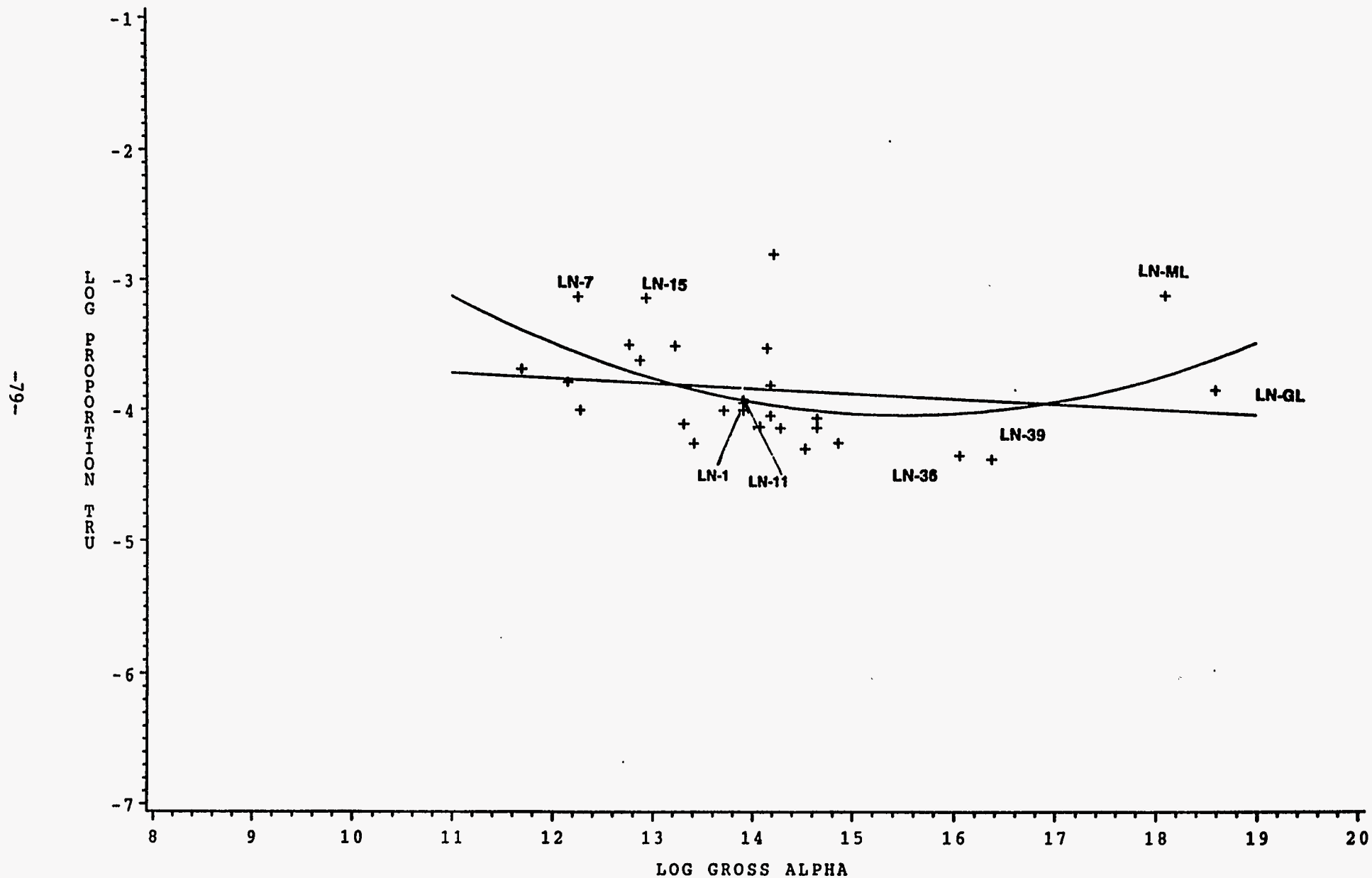
FIG. 8c. 1ST & 2ND ORDER REGRESSION OF LN TRU (LOWER) ON LN GROSS ALPHA

EXPERIMENT 2--2 OUTLIER OBSERVATIONS OMITTED
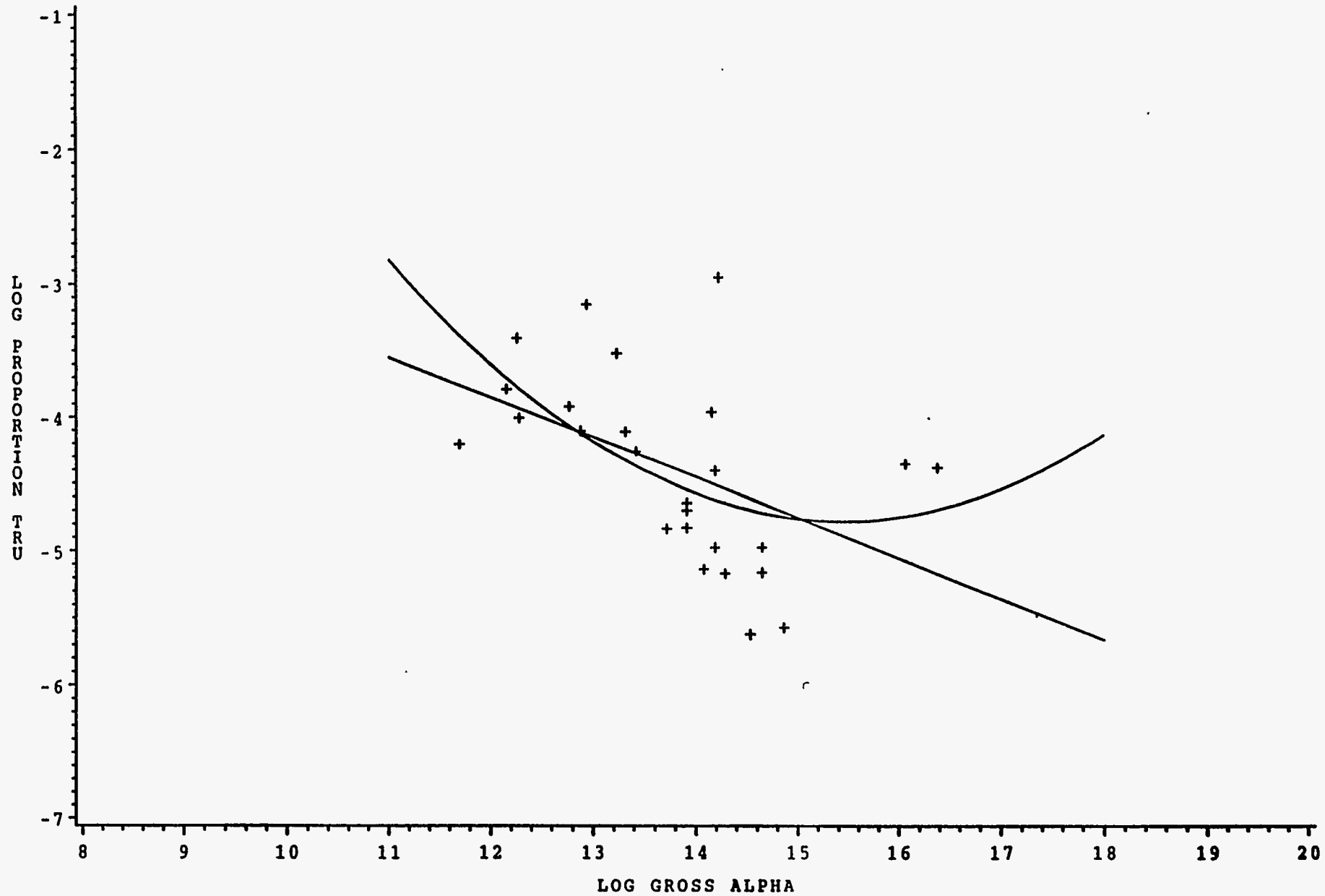
FIG. 8d.   1ST & 2ND ORDER REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA
EXPERIMENT 2--2 OUTLIER OBSERVATIONS OMITTED
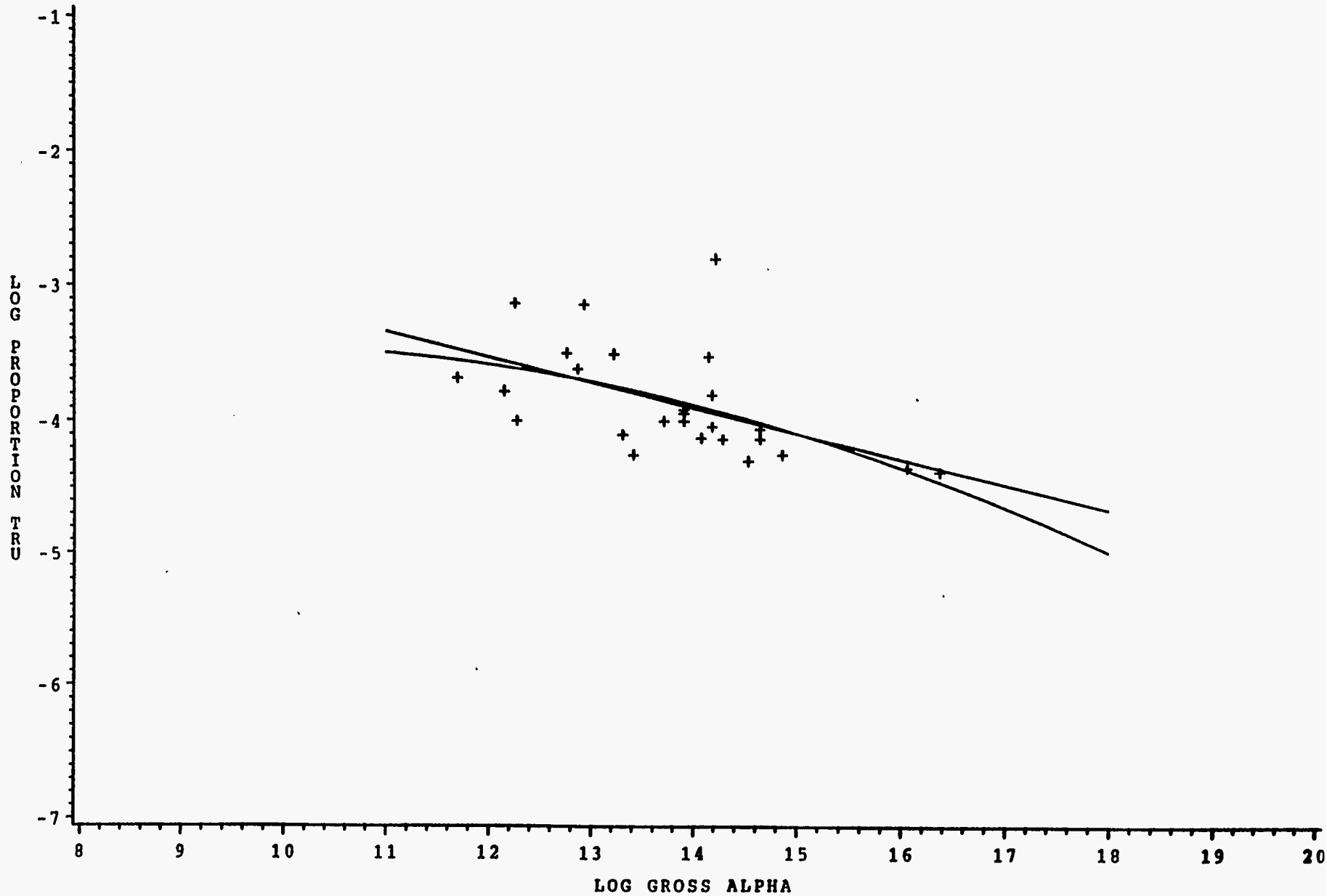
FIG. 9a.   1ST & 2ND ORDER REGRESSION OF LN TRU (LOWER) ON LN GROSS ALPHA
BOTH EXPERIMENTS COMBINED--NO OBSERVATIONS OMITTED

-82-
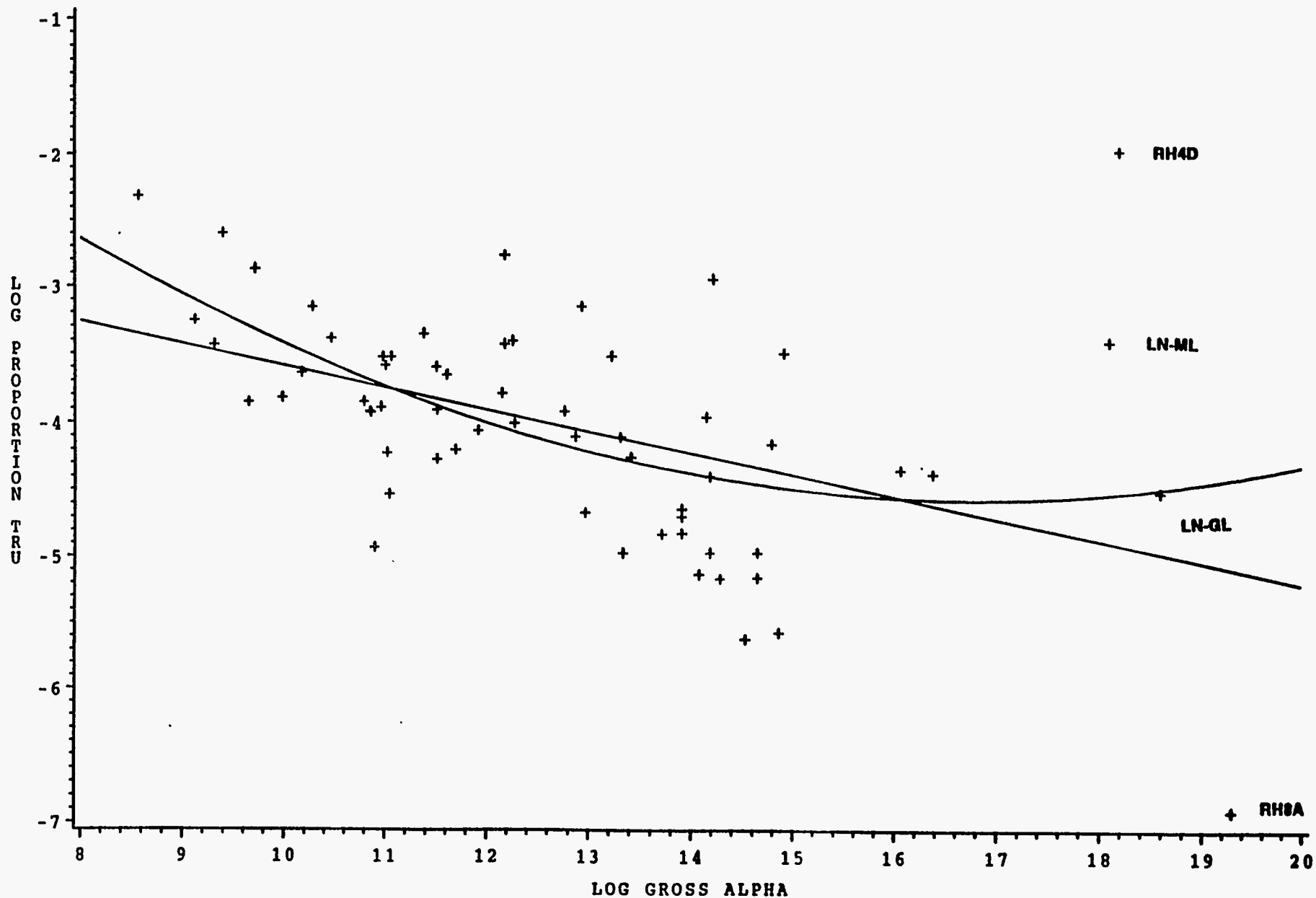
FIG. 9b.   1ST & 2ND ORDER REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA

BOTH EXPERIMENTS COMBINED--NO OBSERVATIONS OMITTED
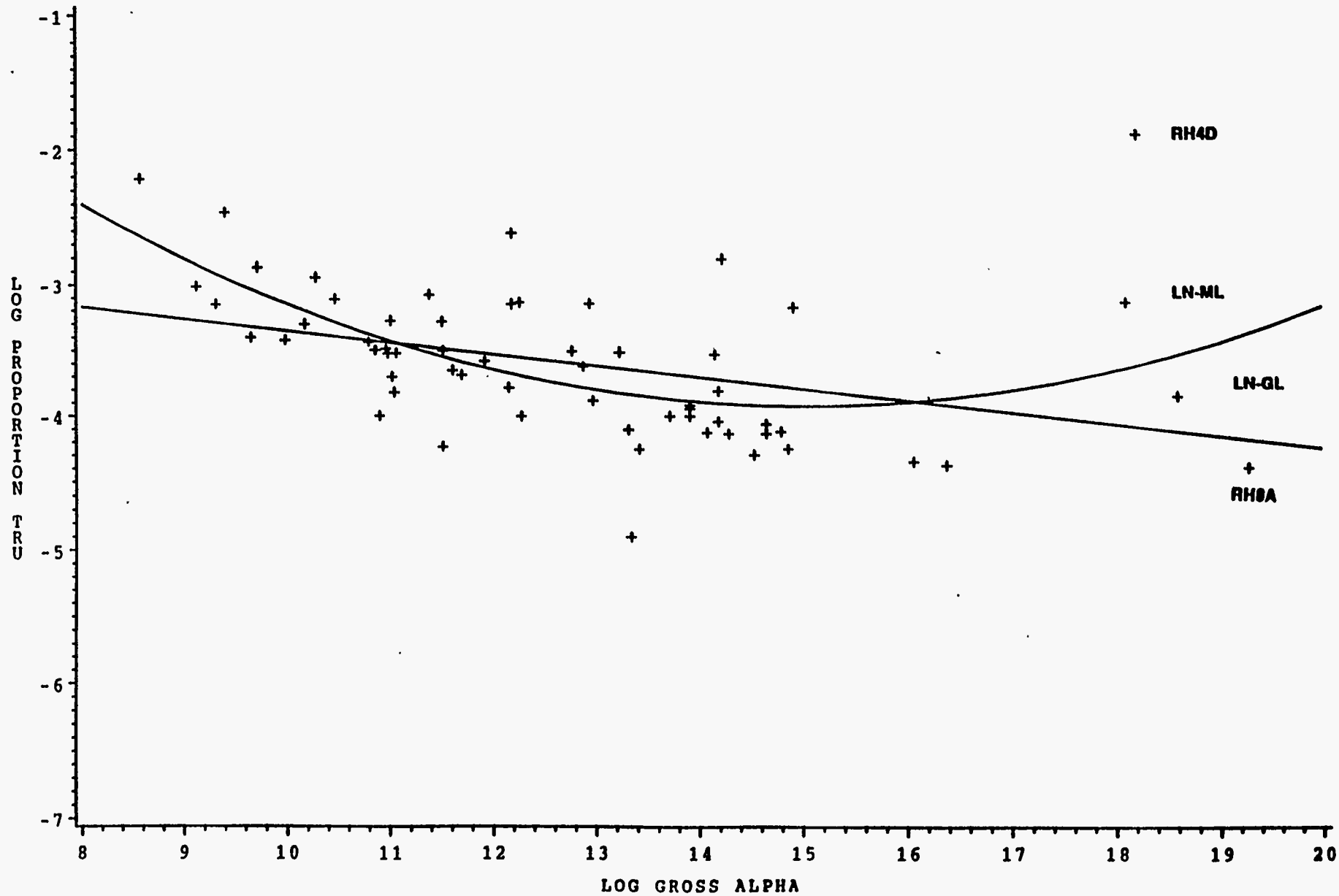
FIG. 9c. 1ST & 2ND ORDER REGRESSION OF LN TRU (LOWER) ON LN GROSS ALPHA
BOTH EXPERIMENTS COMBINED--4 OUTLIERS OMITTED
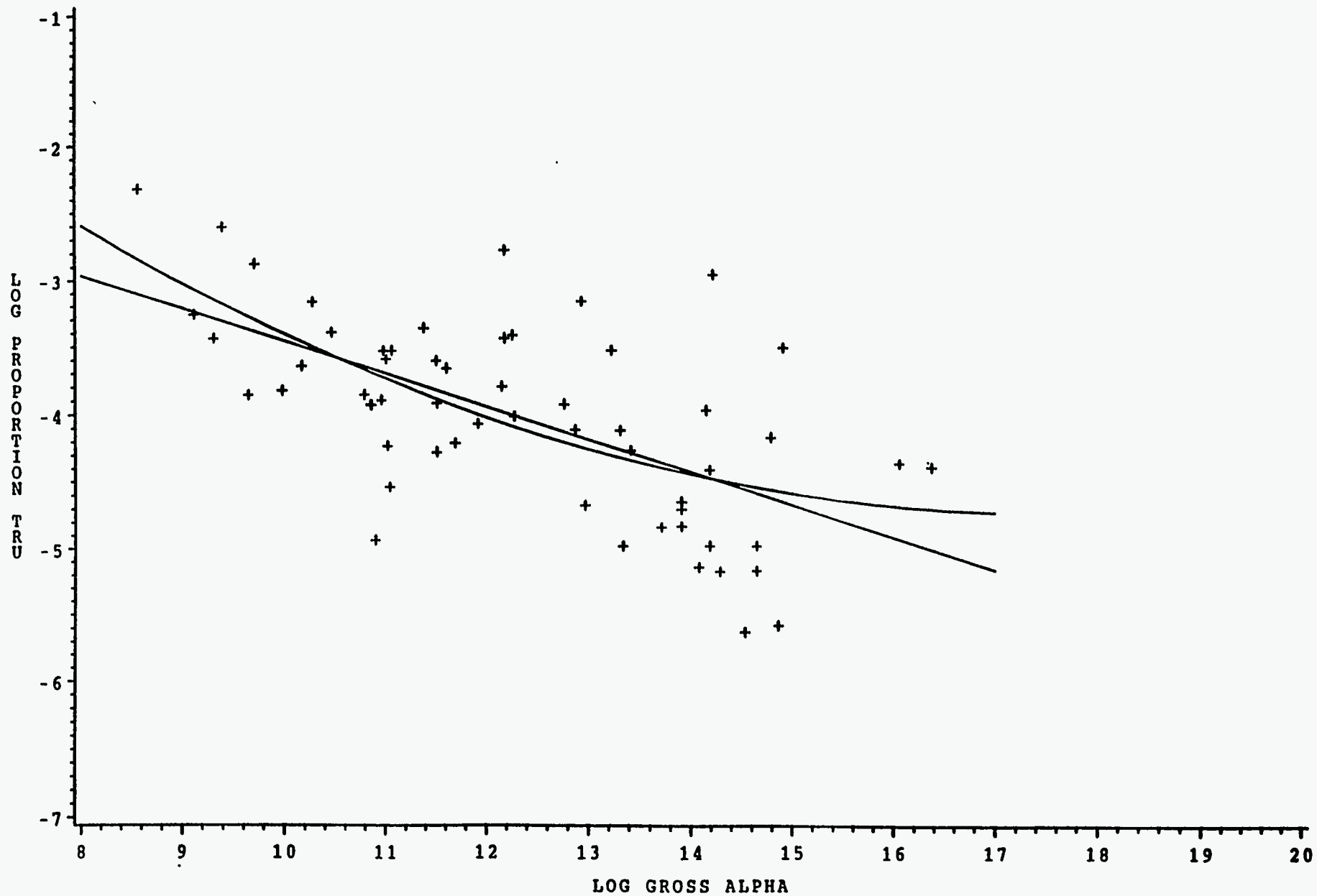
FIG. 9d.   1ST & 2ND ORDER REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA
BOTH EXPERIMENTS COMBINED--4 OUTLIERS OMITTED
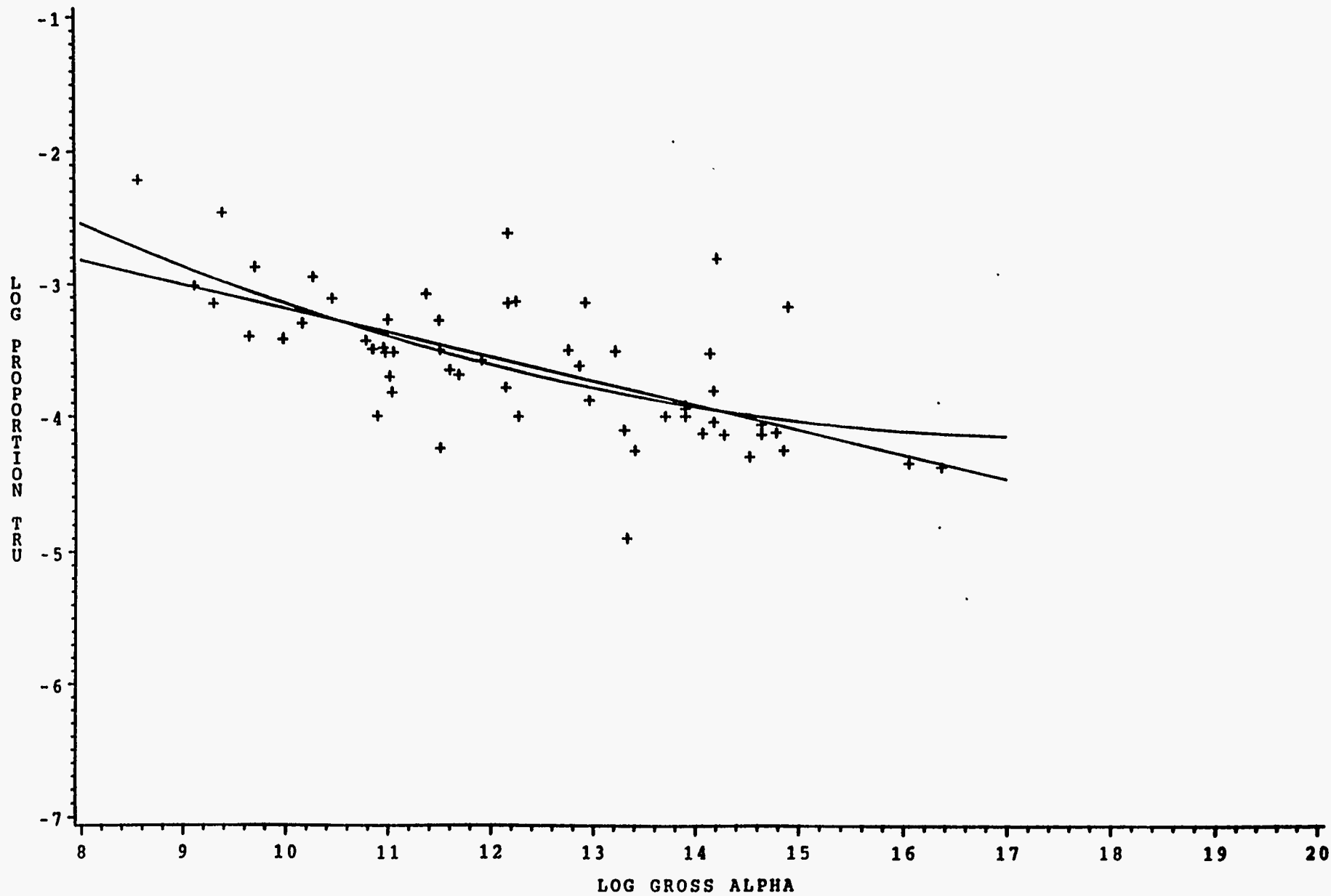
-85-

FIG. 10a. LINEAR REGRESSION OF LN TRU (LOWER) ON LN GROSS ALPHA

BOTH EXPERIMENTS COMBINED--4 OUTLIERS OMITTED
DASHED LINES BOUND 95% CONFIDENCE LIMITS ON MEAN
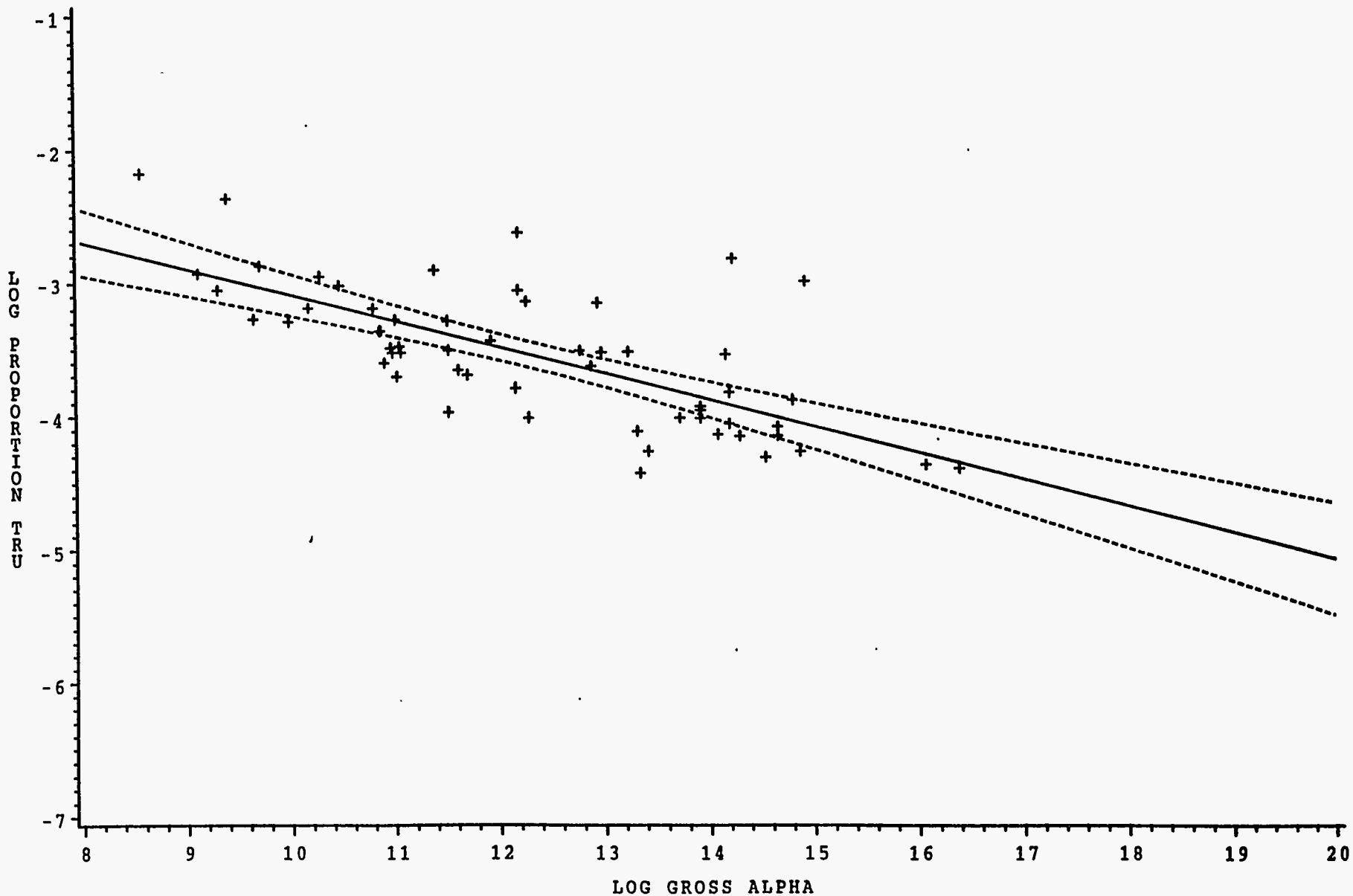PU DETECTION LIMIT = 0.05% OF GROSS ALPHA

-98-
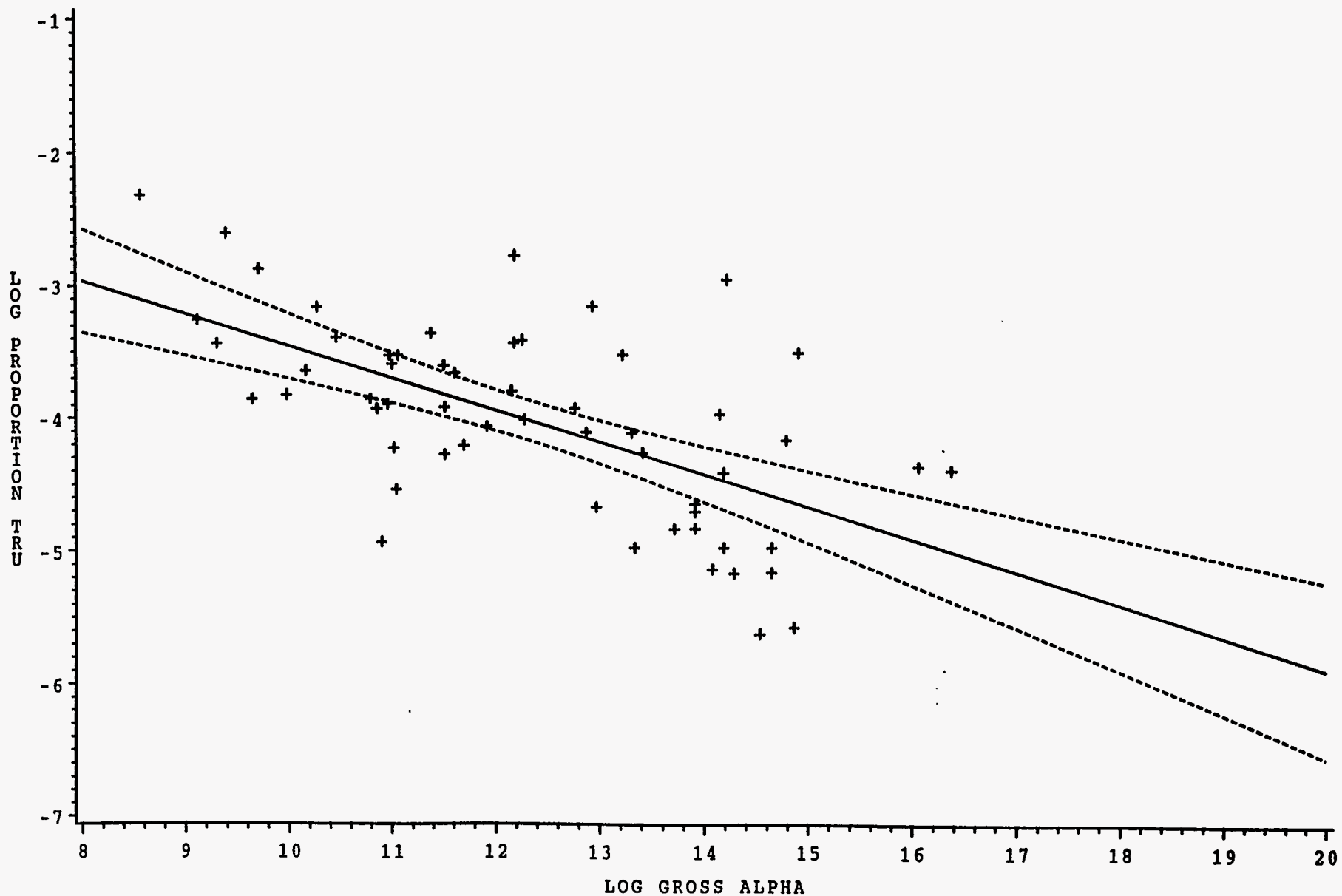
FIG. 10b. LINEAR REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA

BOTH EXPERIMENTS COMBINED--4 OUTLIERS OMITTED
DASHED LINES BOUND 95% CONFIDENCE LIMITS ON MEAN
PU DETECTION LIMIT = 0.05% OF GROSS ALPHA
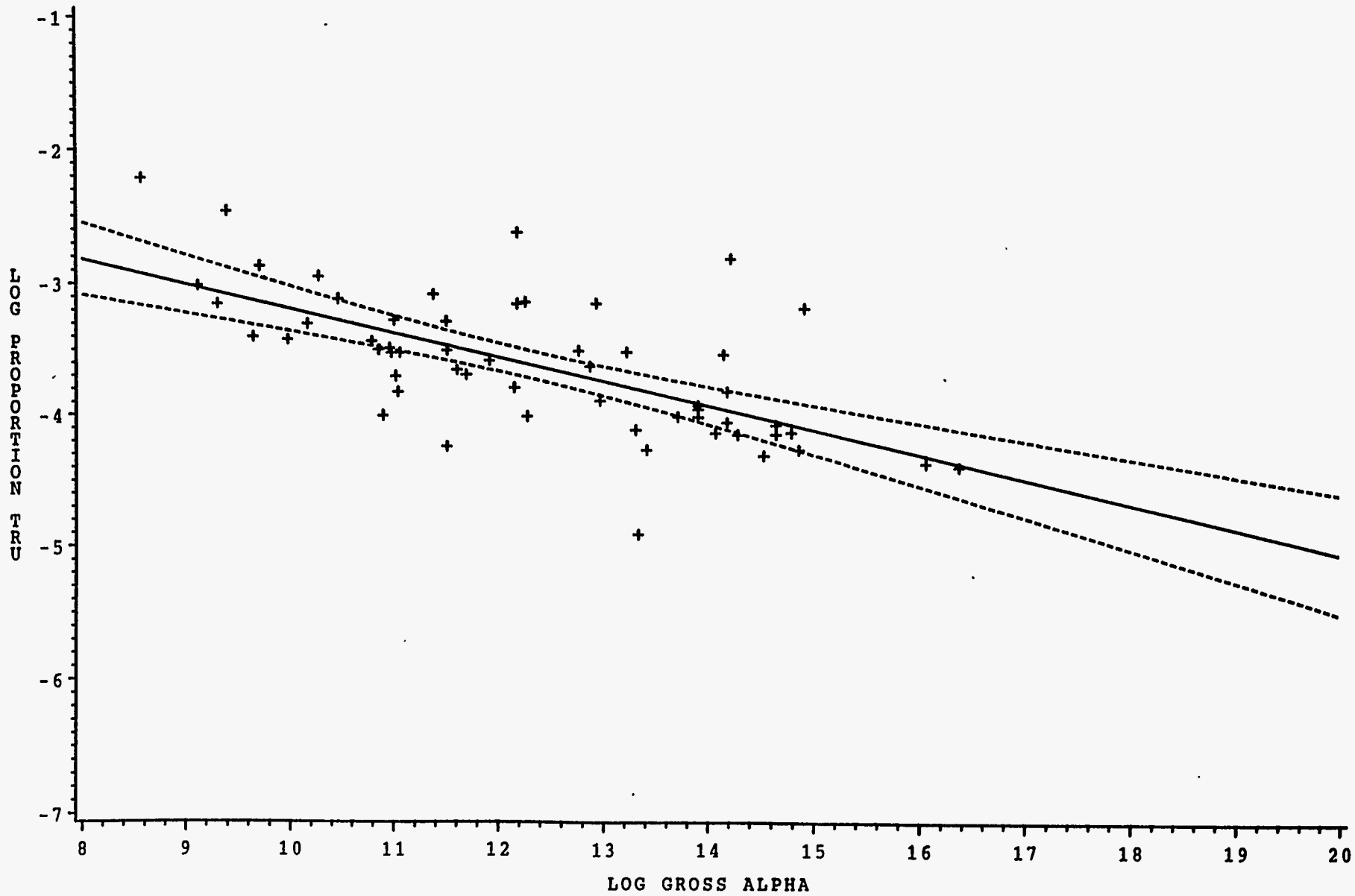
FIG. 10c. LINEAR REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA

BOTH EXPERIMENTS COMBINED--4 OUTLIERS OMITTED
DASHED LINES BOUND 95% CONFIDENCE LIMITS ON MEAN
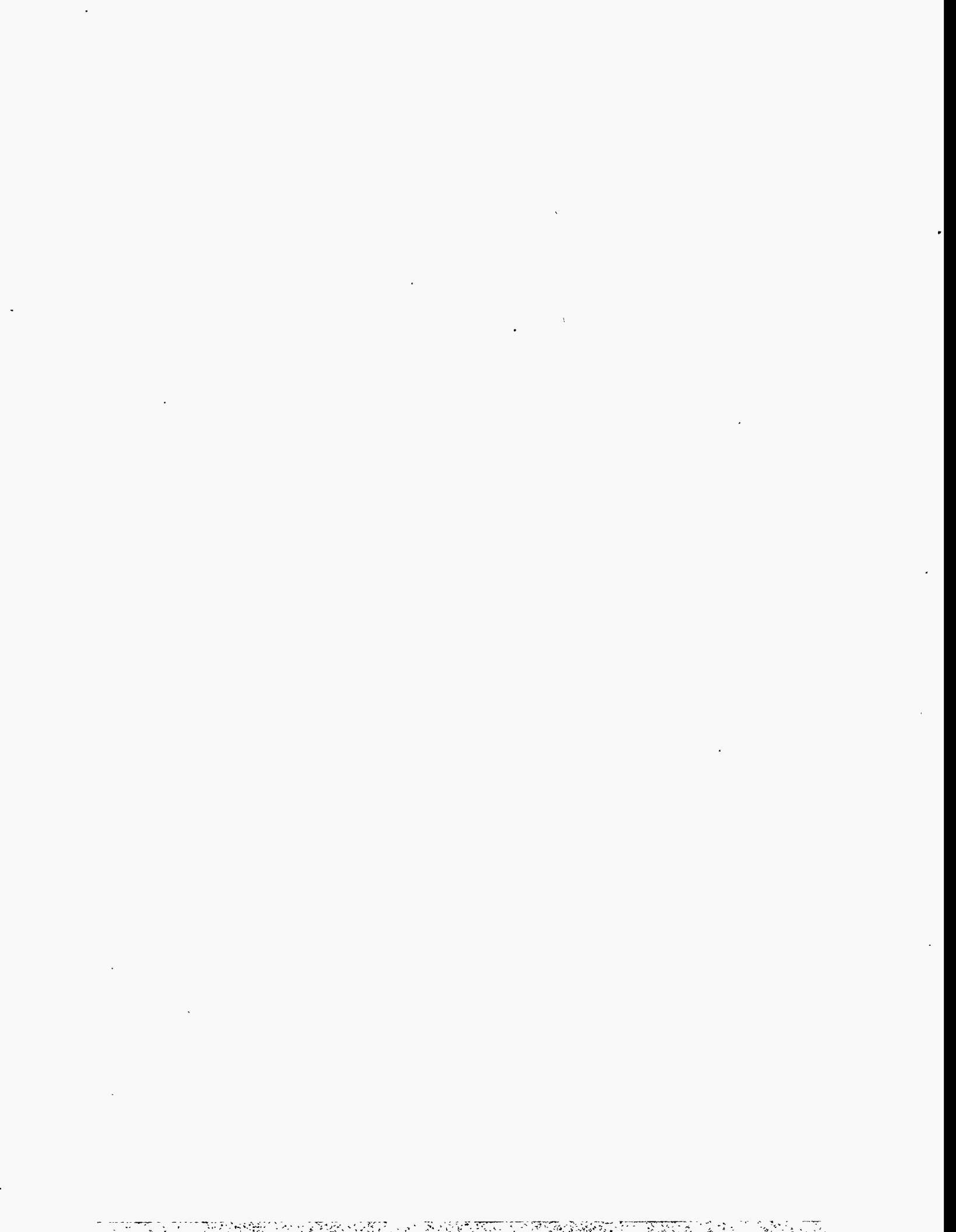### PU DETECTION LIMIT = 0.5% OF GROSS ALPHA

FIG. 10d. LINEAR REGRESSION OF LN TRU (UPPER) ON LN GROSS ALPHA

BOTH EXPERIMENTS COMBINED--4 OUTLIERS OMITTED
DASHED LINES BOUND 95% CONFIDENCE LIMITS ON MEAN
PU DETECTION LIMIT = 1.0% OF GROSS ALPHA

-69-
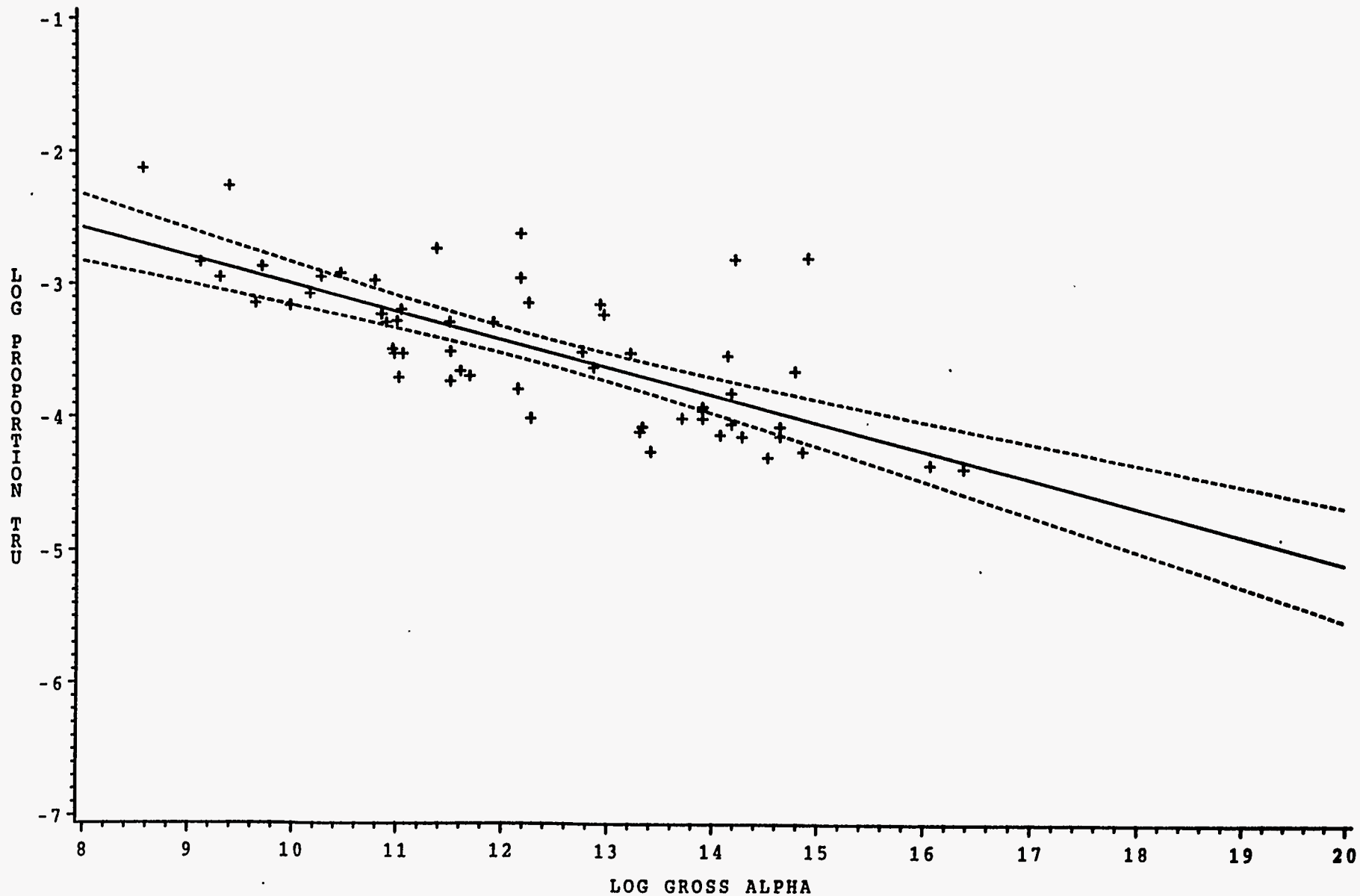
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