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SURVEY OF DOE NDA PRACTICES FOR CH-TRU WASTE CERTIFICATION - ILLUSTRATED WITH A GREATER THAN 10,000 DRUM NDA DATA BASE

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1. Introduction

With the Waste Isolation Pilot Plant (WIPP) scheduled for opening in fiscal year 1989 the Department of Energy (DOE) recently sponsored a systematic assessment of the various DOE site nondestructive assay (NDA) practices used to certify contact-handled transuranic (CH-TRU) waste isotopic inventories. The bulk packaged CH-TRU wastes are certified according to the WIPP Waste Acceptance Criteria (WAC). These criteria include fissile criticality content, package heat generation, radioisotopic content, and gas generation safety assurances. NDA measurements provide data for the waste drum fissile criticality content, package heat generation (thermal power), specific activity, ^{239}Pu equivalent activity, and radioisotopic content.

NDA techniques now in widespread use throughout the DOE sites which provide the required WIPP-WAC certification include: Segmented Gamma Scanning (SGS), Passive Neutron Coincidence Counting (PNCC), and Differential-Dieaway-based Passive-Active Neutron (PAN). The general practice is to provide multiple NDA measurements at the various stages of waste packaging and certification; thereby, affording numerous examples of NDA technique assay result comparisons.

In particular, we have compiled a greater than 10,000 CH-TRU waste drum data base from seven DOE sites which have utilized such multiple NDA measurements within the past few years. Most of these NDA technique assay result comparisons have been performed on well-characterized, segregated waste categories such as cemented sludges, combustibles, metals, graphite residues, glasses, etc., with well-known plutonium isotopic compositions. Waste segregation and categorization practices vary from one DOE site to another. Perhaps the most systematic approach has been in use for several years at the Rocky Flats Plant (RFP), operated by Rockwell International, and located near Golden, Colorado. Most of the drum assays in our data base result from assays of RFP wastes, with comparisons available between the original RFP assays and PAN assays performed independently at the Idaho National Engineering Laboratory (INEL) Solid Waste Examination Pilot Plant (SWEPP) facility, operated by EG&G, Idaho. Most of the RFP assays were performed with hyperpure germanium (HPGe)-based SGS assay units. However, at least one very important waste category, processed first-stage sludges, is assayed at RFP using a sludge batch-sampling procedure, prior to filling of the waste drums.

Systematic assay comparisons for wastes generated at other DOE facilities are also available, including Westinghouse Hanford (WH) and Lawrence Livermore National Laboratory (LLNL). For these sites the types of NDA comparisons involve SGS (performed by the original waste generator) and both passive coincidence and differential-dieaway, performed with the site's PAN unit.

2. NDA System Certifications and Calibrations

SGS Measurements

The DOE facility standard guide used for preparation of SGS standards is ANSI N-15.35 (reference 1). The DOE facility standards for proper implementation and use of these standards are ANSI N-15.20 (reference 2) and ASTM C853-82 (reference 3). These standards call for preparation of the calibration material using intimate and stable mixtures of the TRU isotope with matrix material and for preparation of a suitable number of calibration standards to cover the isotopic concentration range of interest. In the case of 239Pu this range is 5 to 200g for 208 l drums. (Calibration standards spanning this approximate range are employed at all the DOE sites using SGS for weapons grade (WG) Pu assay).

Standards utilization practices vary somewhat from site to site; however, the general practice is to use at least two standards drums. One standards drum contains a TRU isotope mass at the low end of the range of interest, while the other contains a TRU isotope mass at the high end of the range. Both drums contain typical waste stream matrix mixtures and densities which are typically generated at the site. These standards drums are then measured by the assay instrument multiple times, both before and after each measurement session, to assure determination of a proper calibration constant. These calibrations are archived on magnetic media, as are all waste drum assays, for later reexamination, if required and for quality assurance documentation purposes.

Radiochemical Methods

Standard test methods (ANSI and ASTM) describe the radiochemical standard aliquot sampling procedure used in DOE facilities (e.g., RFP stage one sludges). Assay standards are prepared and used as indicated in the standard test methods. Assay accuracies at the final, filled-drum stage are difficult to estimate, since they depend primarily on the maintenance of homogeneous mixtures during the sampling, drying, cementing, and final drum-filling stages.

PNCC Measurements

The PNCC method for determination of Pu assay in product materials has been used for Nuclear Safeguards verification purposes within the DOE complex for more than 20 years. PNCC has also been applied to the assay of TRU-bearing wastes and scraps for many years (reference 2, sub-sections 20-28). In addition, the USNRC Regulatory Guide 5.11 describes NDA techniques acceptable to the NRC for assay of wastes and scraps, which includes PNCC assay measurement technique.

These standards and regulatory guides are used to ensure proper application by the DOE of PNCC to scrap and waste assay. It should also be realized that the passive assay portion of the PAN assay technique described below is an adaptation of the PNCC method using sophisticated "self-measurement" matrix corrections (see reference 4).

PAN Measurements

The PAN assay systems are very recent additions to NDA instrumentation, and as a consequence, ASTM and ANSI standards have not as yet been developed for the PAN assay systems. (Of course, as mentioned previously, the passive coincidence portion of PAN is the same as PNCC and, therefore, PNCC standards are followed for that portion of the PAN system). LA-10774-MS (reference 4) discusses the standards used in all present PAN units, for which all passive and active calibration standards have NBS-traceable or NBS-referenceable origins.

Following an extensive absolute and matrix standards calibration at the Los Alamos National Laboratory (LANL) prior to installation at the various sites, the PAN units were each provided a set of secondary standards (placed in "Pink Drums" for conspicuous identification) consisting of standard, NBS-referenceable ^{252}Cf passive assay and ^{235}U active assay materials (secondary standards). A baseline reference for both passive and active assays was established for each PAN unit with these unique "Pink Drum" standards, and each unit has subsequently performed standard Pink Drum assays prior to each set of PAN waste drum assays. A typical set of these standards measurements performed with the SWEPP/INEL unit and extending for almost a three year period is shown in Figure 1. As can be seen, the individual passive and active standards measurements fall well within a +/-10% window, with no measurable systematic drift during the three-year operational history. In LA-10774-MS (reference 4) are presented the corresponding Pink Drum measurements for Hanford, SRP, and the Mobile PAN Assay units. All measurements demonstrate the same basic stability of response.

More extensive discussions of each assay method described in Section 2 are given in reference 5.

3. Systematic Assay Technique Comparisons

Figures 2a, 2b, and 2c show a set of data resulting from the assays of 200 CH-TRU waste drums. The assays were performed in 1986 at LLNL. These drum data sets are not segregated according to matrix content, but are of mostly combustible and general, non-sludge, laboratory waste. Figure 2a shows the comparisons between SGS (LLNL unit) and PNCC (PNCC portion of the LANL Mobile PAN Assay System). Figure 2b shows the identical SGS data compared with the active assay (differential-dieaway) portion of the LANL Mobile PAN Assay System. Figure 2c shows an internal PNCC comparison between the two independent (high- and low-sensitivity) PNCC detector package systems incorporated into the LANL Mobile PAN Assay System. In all three comparisons of the same data set the agreements are excellent. No systematic assay bias is apparent to at least the+/- 10% level.

Figure 3 shows an assay comparison of the PNCC and differential-dieaway (DDA) portions of Hanford's PAN system, which was performed with a set of 300 general-category waste drums. The nominal cosmic ray PNCC background at the Hanford installation is sufficiently low that credible WG Pu (i.e., 6% ^{240}Pu) assays of waste drums may be performed on a routine basis down to below 0.1 g total Pu mass. Of course, the lower limit of detection (LLD) for the active or differential-dieaway PAN assay is about 0.001 g total Pu mass at all installations, including the one at Hanford. Note that the data of Figure 3 is plotted on a log-log scale so as to emphasize the dynamic range covered; that is, 0.01 to 10 g total Pu mass. The agreement between PNCC and DDA is excellent, with no apparent systematic bias to at least the +/- 10% level.

Figure 4 shows a set of assays of 300 segregated waste drums from RFP. Each of these waste drums contains a matrix described as "graphite molds". As such, they are both a low material-density and low neutron-absorption matrix. Consequently, all three NDA methods with which these drums were assayed (SGS, PNCC, and DDA) would not be subject to any significant matrix effects or corrections. For PAN assays the larger the matrix correction calculated, the larger the relative error associated with the assay measurement. See reference 4 for a detailed discussion of the PAN matrix correction calculations. In effect, this set of assay comparisons is an excellent one with which to judge the calibration accuracy and reliability of the three independent NDA system techniques. The calibrations have been maintained over a several year (almost three years) period of routine operations at two different DOE sites. The SGS assays were performed at RFP using one or more site units. The PNCC and DDA measurements were performed at SWEPP (INEL) using that site's PAN unit. It should be noted that the DDA measurements were corrected for fissile self-absorption effects using a semi-empirical model described in reference 4. For this data set there again appears to be excellent comparison agreement at the +/- 10% level.

Figure 5 assay result comparisons were obtained from measurements of a set of 1300 stage one sludge drums which had been generated at RFP. The graph shows a comparison of batch-average Pu assay values performed at RFP (using a grab-sampling radiochemical method (see Section 2) applied prior to filling of the drums) with the corresponding DDA-batch averages obtained from the average over all individual drums in each batch. The latter assays were performed at SWEPP/INEL using their PAN unit. The linear regression analysis performed on this data set indicates an excellent systematic assay agreement, to within +/- 5%, even though the scatter displayed for individual comparisons is several times this amount. Basically, the systematics of both assay techniques appear to agree excellently.

Not all assay comparisons of specific waste forms provide such excellent agreements as those discussed above. One such waste category type is provided by RFP category 320, tantalum (Ta) crucibles. What one observes for these CH-TRU waste drums is that the PNCC assay values agree well with SGS measurements, but that DDA measurements are systematically lower than either the PNCC or SGS results. A detailed paper discussing not only code 320 comparisons but also assay comparisons for a large number of other RFP segregated waste categories as well is being prepared by J. R. Smith.

One very interesting feature of this particular waste category type is the systematic behavior of the DDA assays relative to the PNCC and SGS: the ratio between DDA and either of the other two assays is a constant, independent of Pu mass. A careful study (using real-time radiography, RTR) of this waste matrix revealed that these drums normally consisted of stacked Ta crucibles positioned along the drum central axis with low-density cellulosic material placed around them to keep them centered. In this type of geometry (see reference 4) the PAN absorption index neutron flux monitor fails to sense the actual thermal neutron absorbing material (i.e., the Ta), although it produces a considerable effect on the assays. However, since these drums are prepared uniformally, the SWEPP assay algorithm has been modified to process them with the appropriate correction factor applied to all DDA measurements of this category waste drum.

4. Conclusions

Generally speaking, excellent systematic agreement among all currently in-use DOE assay techniques is obtained. Where systematic disagreements do occur, the reason is generally apparent, and corrections to assay algorithms may be made if proper matrix segregation practices are implemented.

5. References

1. ANSI N15.35, "Guide to Preparing Calibration Material for Nondestructive Assay Systems that Count Passive Gamma Rays."

2. ANSI N15.20-1975, "American National Standard Guide to Calibrating Nondestructive Assay Systems."

3. ASTM C 853-82, "Standard Test Methods for Nondestructive Assay of Special Nuclear Materials Contained in Scrap and Waste."

4. J.T. Caldwell, R.D. Hastings, G.C. Herrera, W.E. Kunz, E.R. Shunk, "The Los Alamos Second-Generation System for Passive and Active Neutron Assays of Drum-Size Containers", Los Alamos Formal Report LA-10774-MS, September 1986.

5. F. J. Schultz and J. T. Caldwell, DOE Assay Methods Used for Certification of CH-TRU Waste, ORNL 6485, in publication, October 1988.

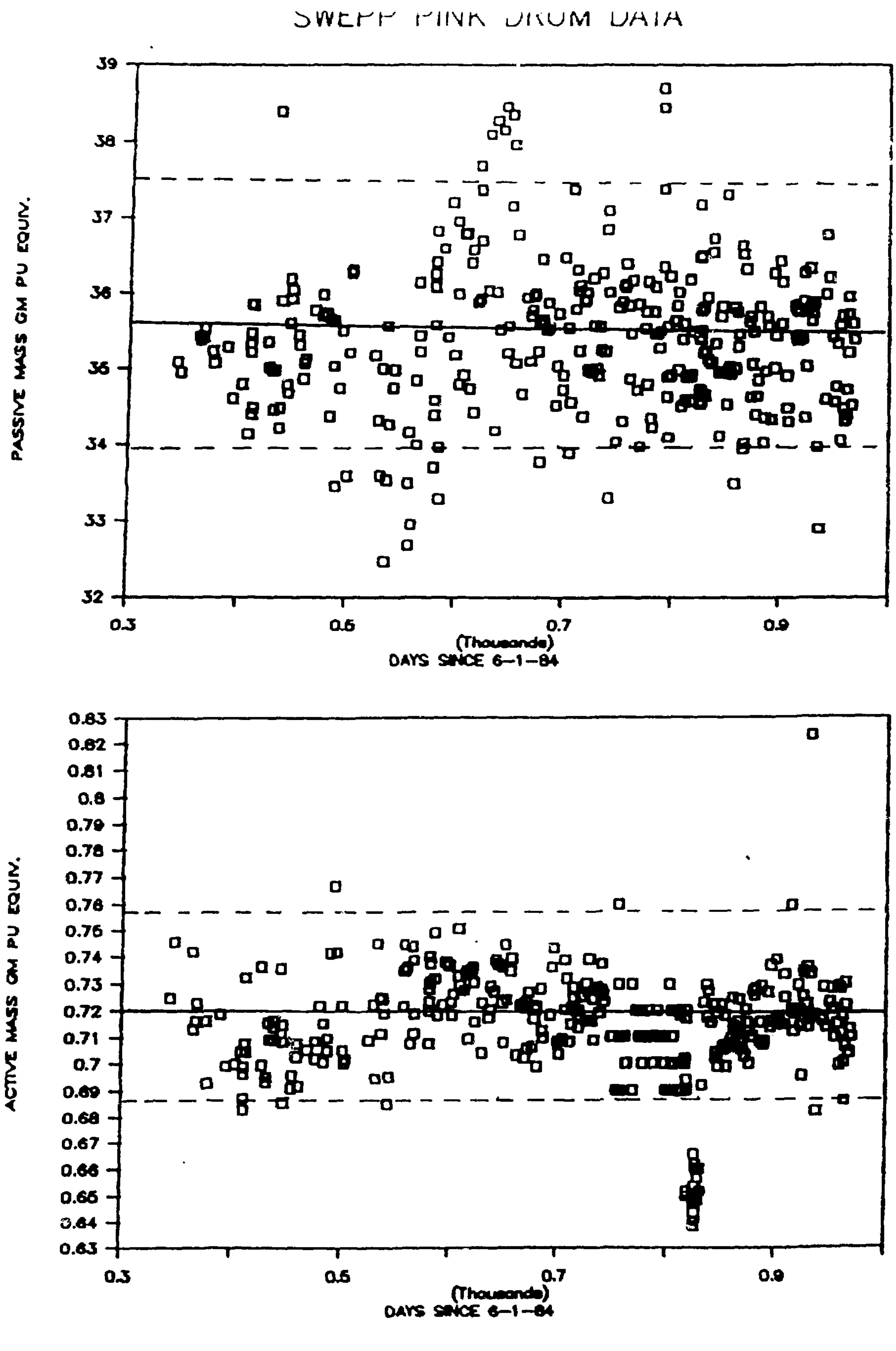
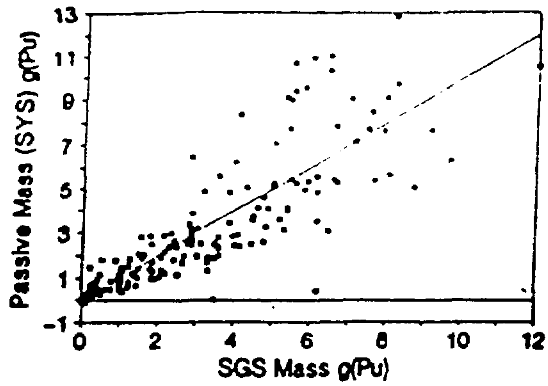
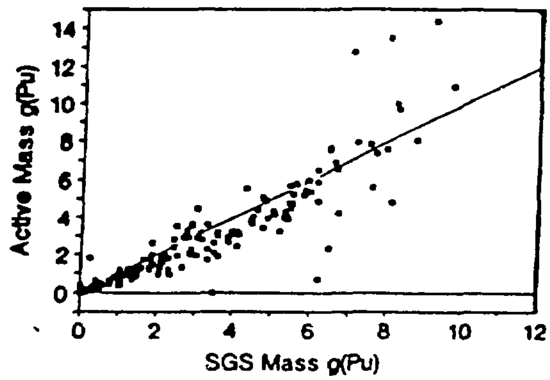


Figure 1 . SWEPP PAN Standards Measurements (Pink Drum) Performed Over a Three Year Period. Top Shows Passive Standard and Bottom Shows Active Standard. Dashed Lines Indicate a +/-5% Measurement Error Band About Expected Standards Assay Values.





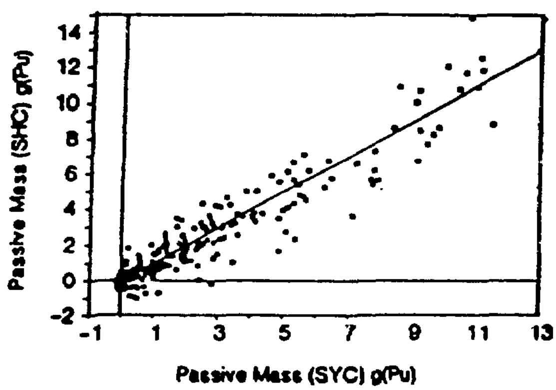


Figure 2

Intercomparison Assays of a Set of 200 TRU Waste Drums (LLNL). Top Shows Passive Neutron (PAN) Compared to SGS. Middle Shows Active Neutron (PAN) Compared to SGS. Bottom Shows Comparison of the Two Independent PAN Passive Neutron Assay Systems.

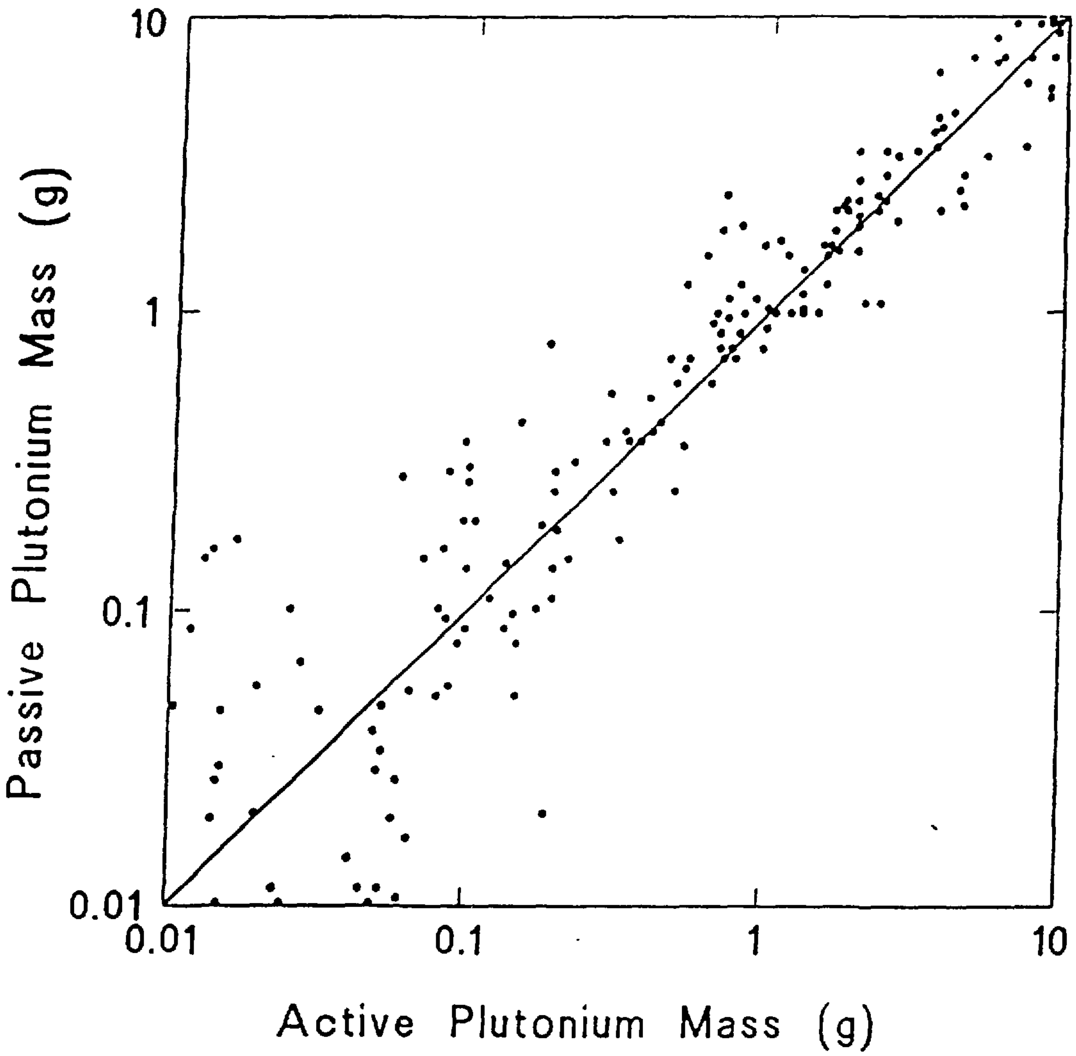


Figure 3. Comparison of 300 Hanford TRU Waste Drum Assays Done with the PAN System, Passive Neutron Compared to Active Neutron.

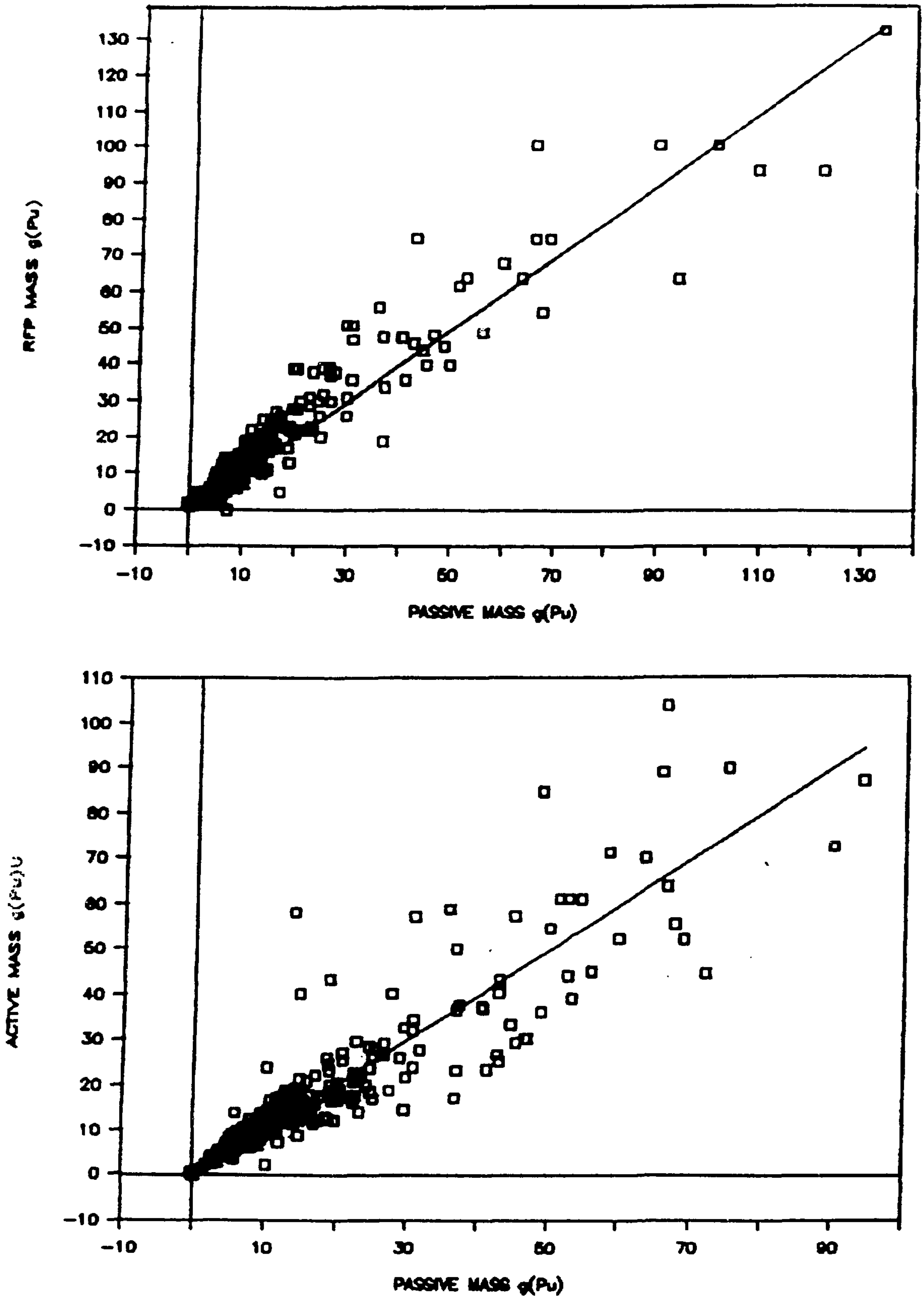


Figure 4. Assay Intercomparisons of a Set of 300 RFP TRU Waste Drums (Graphite Molds Matrix) Showing RFP SGS System Assays Compared to SWEPP PAN Unit Passive Neutron Assays (Top) and SWEPP PAN Active Neutron and Passive Neutron Comparisons (Bottom).

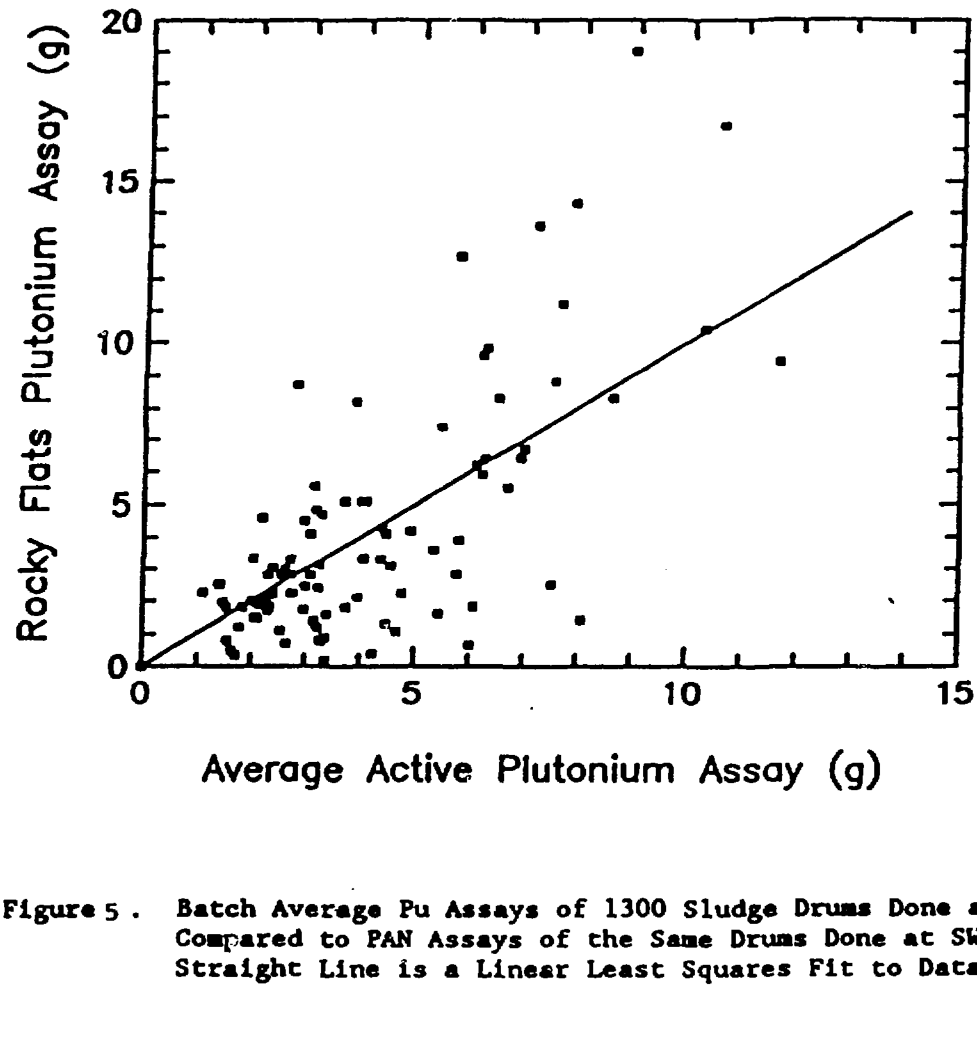


Figure 5. Batch Average Pu Assays of 1300 Sludge Drums Done at RFP Compared to PAN Assays of the Same Drums Done at SWEPP/INEL Straight Line is a Linear Least Squares Fit to Data.