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AUTHOR(S): Joseph R. Wachter

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

WASTE MEASUREMENTS AT A PLUTONIUM SCRAP RECOVERY FACILITY

Joseph R. Wachter
Los Alamos National Laboratory
Los Alamos, NM U.S.A.

Abstract

Solid plutonium contaminated wastes are often highly heterogeneous, span a wide range of chemical compositions and matrix types, and are packaged in a variety of container sizes. NDA analysis of this waste depends on operator knowledge of these parameters so that proper segregation, instrument selection, quality assurance, and uncertainty estimation can take place.. This report describes current waste measurement practices and uncertainty estimates at a U.S. plutonium scrap recovery facility and presents a program for determining reproducibility and bias in NDA measurements. Following this, an operator's perspective on desirable NDA upgrades is offered.

1. Introduction

Los Alamos National Laboratory (LANL) is a U.S. national defense facility involved in the recovery and processing of plutonium. Wastes and residues are routinely generated here from many stages of plutonium metal fabrication including pyrochemical and aqueous processing of plutonium scrap. These processing steps produce a wide variety of leaner scrap and waste forms such as plutonium oxide from burned residues, Pu-bearing salts from production/reduction and metal purification processes, impure plutonium metal, metal reduction slags, ash, undissolved oxide heels, ceramics, cleaning rags, plastics, HEPA filters, and other remnants and apparatus generated from processing and cleanup tasks[1,2]. If the processed residue has a concentration such that plutonium recovery is economically justified, it is classified as scrap and retained for later treatment. If it is below economical recovery limits, it is disposed of as radioactive waste. Both liquid and solid wastes result from these processing steps. Liquid wastes include effluents from ion-exchange columns, oxalate filtrates, and caustic solutions generated by various head-end and purification operations. The solutions are filtered, treated in an evaporator, and chemically sampled to determine discard criterion. No Nondestructive Analyses (NDA) are performed on the liquid

wastes. Upon meeting the criterion, the filtrates and sludges are fixed in cement and sent to a waste storage complex. Solid wastes contaminated with plutonium are divided into two categories: Transuranic (TRU) waste containing greater than 3700 Bq/g (100 nCi/g) of radioactivity and Low Level Waste (LLW) containing less than this amount. TRU waste is obtained from various processing and cleanup steps within the processing areas while the LLW is derived from locations where contamination is expected to be light. Both solid waste categories receive NDA analyses to determine radioactive content. Presently, neither waste form is immobilized prior to disposal. The purpose of this report is to review the current status of solid waste measurement operations here, examine the reliability of those measurements, and consider desirable upgrades in support of future waste analysis. The first section of the report will briefly review current measurement practices at this facility. In this section, segregation of the waste prior to analysis, packaging, instrument selection, and quality assurance procedures will be discussed. Proper management of these functions often contributes as much to measurement reliability as NDA technique. Next, the report will discuss reproducibility and bias estimates for waste measurements performed here and how these estimates were justified. Finally, an operator's perspective on the most important needs in current NDA methodology will be presented. Listed here are software, hardware, instrument evaluation, and standards requirements needed to meet anticipated safeguards, quality assurance, and regulatory demands.

2. Waste Measurement Program

After the various processing steps, the resulting residue is segregated according to matrix type, inspected for hazardous materials (flammables, explosives, carcinogens, etc), and packaged into containers ranging from 10 cm diam x 10 cm tall to 208 l drums. It is then covered in plastic sheeting, removed from the processing glovebox, and delivered to the NDA laboratory for analysis. Materials such as paper, plastics, cellulosics, ash, powders, and other low density materials contaminated with plutonium are measured with Segmented Gamma Scanners (SGS) using high purity Ge detectors. High and intermediate density TRU waste, including metals, leaded gloves, tools, motors, and some lean salt residues are analyzed by passive neutron coincidence counting (NCC) methods. If the measurement establishes that an item is below economic discard limits and can be disposed of as TRU waste, it is

loaded into a drum with other items of similar matrix composition. Finally, the filled drums are sealed and given an NDA confirmation measurement in order to validate the initial measurements and verify that diversion has not taken place. If the confirmation measurement is within established limits, the drum is delivered to a waste holding area to await permanent disposal.

For Low Level Waste, compactible materials are first separated from noncompactibles. The two waste forms are then packaged into cardboard boxes, measured to determine that they meet LLW criterion, and removed from the processing facility for shallow land burial. LLW is analyzed with a multienergy detection scheme using a NaI detector to monitor the x-ray region for items which meet LLW criterion, and subsequently crossing over to progressively higher energy gamma rays to measure elevated activity levels. Oversize waste (i.e., waste that is too large to fit into standard LLW containers) is presently analyzed with four slab counters which surround the sample and measure the uncorrelated neutron signal.

At present, there are 15 gamma-ray and neutron assay instruments used to measure radioactive waste at this facility. They are operated and maintained by seven calibration and instrument repair personnel, ten measurement employees, and five technical support and oversight staff.

A wide variety of tests and measurements are performed to assure instrument performance and proper accounting of special nuclear materials. When a new NDA instrument arrives at this facility, it is subjected to a rigorous series of qualification checks and measurements under expected operating conditions. These include hardware, software, and algorithm check-out; short and long term instrument stability evaluation; assay range determination; assessment of chemical forms and matrix types which can be reliably assayed with the new instrument; resolution of measurement control parameters; establishment of calibration standards and frequency; documentation development; and operator training. New instrument qualification typically requires about six months of effort and is reviewed by safeguards, waste management, and quality assurance personnel. Upon completion of the qualification requirements, an instrument is put into regular service. Thereafter, instrument performance is monitored through daily and weekly stability tests, monthly review of stability data by operators and safeguards personnel, periodic calibration checks, and annual training and operating procedure updates. Instruments which are found to be operating outside the stability test limits are immediately pulled from service until the reasons for the failure have been diagnosed

and a new series of stability measurements have been successfully completed. Other elements in the medley of measurements and checks that assure instrument performance and plutonium accountability are the confirmation waste measurement protocol mentioned above, ad hoc studies of instrument performance, near real time process accountability, audits by outside agencies, and the Measurement Verification Program (MVP).

Despite the instrument qualification assessment, measurement control procedures, and waste segregation, nondestructive assays of the waste forms indicated above are often troublesome and may pose formidable challenges to the measurement specialist. Reasons for difficulties include:

- TRU waste comes in a variety of chemical compounds, physical sizes, isotopic proportions, and matrix compositions. Each of these may present complications for different measurement methodologies. For example, the presence of plutonium metal shot in the waste may impede gamma-ray measurements whereas multiplication and (α,n) effects can limit the credibility of neutron counter assays. Knowledge and quantification of these effects are often difficult for some waste forms.
- At this facility, most of the TRU and LLW waste is of a heterogeneous nature. Geometric variations in the spatial location of radiating materials affect instrument response and can limit assay reliability.
- Comparison measurements of SGS and NCC assays with highly accurate techniques such as calorimetry or spectrochemical analyses are a useful tool for establishing measurement efficacy. However, these are often not possible because of the diverse nature of the waste and the expense of the alternative analyses. Thus, an important supplementary tool for bias evaluation is not available for measurements of some waste forms.
- Representative standards for many kinds of waste materials do not exist and may prove impossible to fabricate. Indeed, the nature of many heterogeneous waste samples defy attempts to match them with representative standards.
- Oversize waste measurements are often hampered by background and geometry effects. These may limit assay sensitivity and introduce unacceptably large uncertainties into the measured values.

To maintain additional confidence in the assays performed at

LANL, studies are routinely conducted to monitor the reliability of the NDA measurements of the plutonium product materials, scrap, residues, and waste generated here. During the past three years, for example, studies have been carried out on neutron and gamma-ray measurements of hydrofluorination residues, matrix effects in passive neutron counters, comparative NDA analyses of molten salt extraction residues, evaluations of three gamma-ray isotopic codes for high ^{241}Am materials, appraisal of a self attenuation correction for SGS analysis, and NCC measurements of direct oxide reduction salts, among others. In addition to these studies, there is an ongoing program for evaluating NDA measurements over the broad spectrum of process residue and TRU waste forms generated at this facility. This Measurement Verification Program was originally intended as a mechanism for resolving inventory differences and for uncovering unreliable instrument performance. But the data collected over the five year span of the program, to date, have also revealed information on random and systematic uncertainties in the measurements that would be difficult to determine by other means. Although the program generally concentrates on scrap and residue materials, that is, on materials which have a higher concentration of plutonium than waste; their chemical composition, matrix form, instrument selection, and packaging are generally equivalent to TRU waste here. Therefore, examinations of instrument performance for scrap and residues can be used to provide insight into the performance of the instruments for measuring waste. There are several segments to the program; however only the two that pertain to waste measurements will be included in this report. One pertinent segment compares SGS and NCC analyses with previous measurements made by the same instrument, while a second segment compares the SGS and NCC measurements with reference values obtained from calorimetry + isotopics (CI) analysis. A discussion of the results obtained from these segments of the program is given below.

3. Measurement Verification Program

The MVP is applied to the entire range of NDA instruments that measure TRU waste. Process materials containing plutonium as an oxide, a salt, or metal; and embedded in the wide variety of matrices typical of a scrap recovery facility, are measured with these instruments. That is, rags, tools, crucible parts, plastics, rubber and leaded gloves, furnace parts, non-plutonium metal, etc are all analyzed with these instruments. For the program, the

materials are segregated and instrument method is selected in the manner indicated above. Plutonium content varies according to chemical and matrix form but ranges from 1 gram to several hundred grams for oxides and salts and less than 200 g for metals. The SGS and NCC assays for both segments of the program are single measurements of 15 minutes, or less, duration. During the course of the program reported here, all instruments were under the measurement control procedures cited above. For that segment in which the same item is assayed on the same instrument, measurement intervals ranged from one week to as much as seven years. On average however, the two measurements were separated by about 18 months. Providing the instrument used for the assays has been operating in a stable fashion over the time period separating the measurements, as guaranteed by the measurement control procedures, this comparison gives an estimate of the long term reproducibility of the measurement for the NDA technique and type of process material under review. For that segment of the MVP which compares NCC or SGS assays to reference CI analyses to evaluate measurement bias, the two measurements are generally separated by about 3.5 years, although some differ by as much as ten years. Previous studies have indicated that the reference measurements are accurate to within ± 1 % of the nominal plutonium value in the sample.

Table I summarizes the raw results from analysis of these two segments of the MVP data. Ratio R_1 in the table is the average of the mass weighted ratios of a single SGS or NCC measurement to a second measurement made on the same instrument at a later date. The ratio R_2 is the average of the mass weighted ratios of a single SGS or NCC measurement to that of a reference CI measurement made at a later time. Results from 3 SGSs and 6 NCCs were folded together to obtain the reported ratios. These instruments have been used to measure over 95 % of the waste generated here. The uncertainties associated with the ratios are the standard deviation (1σ) of the combined data. Plutonium content for the SGS measurements ranged from 1 g to 350 g, whereas for NCC measurements, the range was 1 g to 370 g.

The table indicates that R_1 is 1.04 for SGSs; that is, SGS remeasurements averaged 4 % lower than the initial measurement. This ratio is consistent with settling of heavier plutonium particles inside the containers over time. Settling has been observed to occur for some of the salts measured by this technique. When the denser plutonium particles fall to the bottom of the container between the first and second measurements, the resulting self attenuation and end effect counting losses tend to bias the

later assay low. The ratio R_2 for SGSs is 0.94, indicating that these measurements are lower by 6 %, on average, than the reference measurements. Because most sources of SGS error, such as end effects and self attenuation, tend to bias these assays low, this result is not surprising. Note that the standard deviations attached to both SGS ratios are greater than 20 % of the ratio values, indicative of the large variability in measurements of process residues and waste.

For NCCs, R_1 equals 0.99, indicating that remeasurements using these instruments averaged 1 % higher than the original values. Here, the magnitude of the remeasurement difference is about one fourth the magnitude of its SGS counterpart. For neutron counters, in contrast to SGSs, a high remeasurement value may result from settling because it could lead to closer packing of the plutonium within the containers. This would tend to increase neutron multiplication and bias the second assay high with respect to the first. Although it is known that settling does take place within some of these containers, this interpretation is speculative since there is no complementary evidence, at present, to suggest that the plutonium particles are more closely coupled in these containers. The ratio R_2 for NCCs is 1.03, indicating an average high bias of 3 % in these measurements. Again, this result is not surprising since multiplication and (α,n) effects tend to overestimate neutron counter measurements. While the standard deviations for both the reproducibility and bias data are again large, they are significantly smaller than their SGS analogues.

Two additional points should be mentioned in regards to the averages presented in Table 1. First, each instrument of the same type shows similar bias and reproducibility tendencies. That is, for all the SGSs used in the study, the later measurements are consistently smaller than the earlier measurements. Thus, in addition to the overall R_1 average being greater than unity, the same is true for each of the individual SGSs. Also, all the SGSs used in the study are biased low compared to the reference CI measurements. The same is true for NCC measurements. Bias and reproducibility tendencies for the individual instruments all trend in the same direction as the overall averages. The ratios, therefore, reflect consistent instrument susceptibilities. The second point is that, although three different gamma-ray isotopics codes were used to establish reference values in this study, additional data was also taken to assure that each code gave essentially the same isotopic percentages for the materials reported here. Thus, the biases are due entirely to inadequacies in

the SGS or NCC measurements, not to error in the CI analyses.

The data summarized in Table I incorporate all the measurements performed over the duration of the MVP program, to date. However, during the course of the program, certain bias trends were noted and corrections were applied to future assays of those material types. In some cases, the MVP data indicated that certain process residues could not be reliably assayed by SGSs, so the efficacy of NCC measurements was investigated. In other cases, bias correction factors had to be applied to NCC assays to obtain unbiased results. Yet again, the use of an instrument to measure certain matrix forms had to be limited to no more than 50 g of plutonium in order to assure credible assays. Therefore, some of the raw data summarized above represent measurements which were subsequently determined to be unreliable and the averages in Table I include results which were later discarded for improved measurement methodologies. For this reason, the summarized data were retabulated, but with those measurements known to be biased omitted. The revised results are shown in Table II. These results include the measurements using the improved methodologies.

The revised table indicates that the ratio R_1 for SGSs has now been reduced to 1.01; that is, later SGS measurements are lower than the original assays by 1 %, on average. This is approximately one fourth the difference seen in Table I. The revised value may intimate that some settling is still occurring in a fraction of the materials receiving SGS measurements, but that those with the greatest proclivity for settling are no longer receiving this measurement. Moreover, because the average difference is only 1 %, whereas the dispersion in R_1 remains high at 21 %, then any interpretations on the causes for the disparity are highly speculative. Substantial improvement is also seen in R_2, the average ratio of the SGS measurements to reference CI values. An average measurement is now biased low by about 2 % which is less than half of the bias seen in Table I. The variability in this ratio has also improved markedly.

For NCCs, the revised reproducibility ratio R_1 indicates a difference of 1 % between the two measurements, about the same as in Table I. Apparently, those materials which were omitted from the original data because of unsuitable chemistries or matrices were not the cause of the differences in the two NCC analyses. Again, it should be noted in this regard, however, that the 1 % difference is small compared to the variability (14 %) in this data. The bias ratio R_2 for NCCs, on the other hand, does show substantial improvement. An average measurement is now biased 1 %, down

substantially from the 3 % average seen in Table I. There has been little change in the standard deviation for this ratio over the previous data.

Several conclusions can be drawn from a comparison of the two tables. First, it is apparent that the MVP program has been successful in determining unreliable instrument performance. From the initial data in Table I, five different material types were dissociated from their original measurement method and reassayed with another technique. The resulting improvements in reproducibility and bias seen in Table II validate their dissociation and markedly improve overall measurement credibility. Because those materials which most tended to shift the bias and reproducibility ratios away from unity have now been identified and alternative techniques have been selected, future improvements will be more difficult. This is compounded by the large standard deviations found in the ratios which will make identification of small biases within this dispersion increasingly troublesome. The large variability seen in the standard deviations is not isolated to just a few material types, but instead is spread across the entire spectrum of materials that established the revised values in Table II. It is also true that for some materials, no technique is entirely appropriate. That is, there is a high probability for a small bias no matter which NDA technique is used. Certain salts generated at this facility fit this category. The salts contain product slag mixed with magnesium sand and crucible remnants resulting from PuF_4 reduction. Residue and waste from this process contain CaF_2 salt, unknown amounts of PuF_4, and plutonium metal shot. This matrix presents difficulties for NCC analysis because of large (α,n) effects resulting from the presence of fluorine atoms. Likewise, SGS assays are hindered by attenuation of the plutonium gamma rays in the shot. Identification and treatment of these measurement kinds of measurement problems must be done on an individual basis. A second conclusion to be drawn from this program is that the averages and standard deviations determined here provide a better assessment of measurement uncertainty for process residues and waste than most established estimates. Generally, precision and bias estimates are made on the basis of repeated measurements of standards (reference materials) or are based on an educated guess. Reference materials are usually designed to suit the NDA technique to which they are applied. That is, the chemical and matrix form of the standard, the stability of the plutonium within the container, and its homogeneity are well adapted to the measurement technique which uses them. Precision and

bias values for these reference materials provide useful estimates of optimal instrument performance under the facility's operating conditions, but do not reflect the measurement uncertainties that should be applied to process material assays. The latter depend on the matrix and chemistry of the material, packaging, location of the plutonium within the sample, the presence of radioactive impurities such as ^{241}Am, calibration variability, etc. That is, they depend on a wide assortment of properties whose effects on the measurements are not clarified by precision and bias values associated with analysis of reference materials. Often these properties are beyond the observation and control of the NDA instrument operator, such as the presence of interferants or the homogeneity of the plutonium particles within a container. In these cases, estimates of measurement uncertainty based on reference materials measurements are particularly unsuitable and NDA instrument operators must resort to opinions based on their experience or process knowledge, that is, an educated guess. These latter estimates are sometimes useful, but all too frequently, are later proven to be false. Moreover, they require verification to be of value for safeguards purposes. Generally, this is difficult to provide.

With these difficulties in estimating uncertainties in mind, Table III was composed. This table lists several different assessments of bias and reproducibility that are either calculated at the Los Alamos plutonium scrap recovery facility or are in general use among U.S. TRU waste measurement agencies[3]. The first row of entries under Reproducibility in the table are the percent standard deviations (% SD) in 15 sequential measurements of a plutonium standard containing 56 g of ^{239}Pu. These are typical precision values taken randomly from a series of weekly measurements on instruments used to assay TRU waste here and are often used as indicators of measurement precision among safeguards personnel at this facility. The second entries under the same heading are the average % SDs of the daily standards measurements taken over the five year span of the MVP program. These numbers represent one estimate of the long term precision in an instrument's measurements. Conventional Reference Values (CRV), the third listings under this heading and the second row of entries under the Bias heading are taken from Reference 3. The reproducibility value is defined as the standard deviation in a series of repeated measurements; i.e., the same estimate that was used to establish the second row of entries under this heading. The spread in values for this CRV is determined from statistical

counting errors resulting primarily from counting time limitations and plutonium loading. Under the Bias heading, the Conventional Reference Value is defined as the closeness of a measured value to its true value and is estimated by the difference between a measured average and its accepted reference value. Factors such as matrix type, chemical form, homogeneity, packaging, etc determine the spread in these values. The CRV entries presently serve as precision and bias guidelines for measurements of TRU waste destined for the WIPP radioactive waste disposal site in the U.S. The fourth Reproducibility entries in Table III are the % SD associated with the R_1 ratios in Table II. Under Bias in Table III, the average percent relative differences (% RD) resulting from the daily measurements of a 56 g plutonium reference standard over the five year period are shown in the first row. Finally, the third entries under the Bias heading list the % RD values excerpted from the R_2 ratios in Table II.

The first observation that can be drawn from Table III is that the criterion presently used here to determine SGS and NCC measurement reproducibility has limited practical value. The reproducability % SDs from 15 successive measurements of a standard show only how stably the instrument performed over a short time span. They provide little insight into measurement reproducibility taken over an extended period of time or for process material measurements. The second row of entries under Reproducibility are, through comparison with the first row, useful for pointing out differences between the short term and long term dispersions in a reference standard's measurements. The two SGS values are seen to have a particularly large difference, indicative primarily of their sensitivity to random electronic and microphonic noise in the environment. It is also useful to contrast the long term reproducabilities with the Conventional Reference Values. Because of the measurement times and plutonium content used to determine the second row of entries in Table III, the minimum CRV values should correspond closely to those values. At this facility, the long term reproducibility of the SGS and NCC measurements are both slightly below the CRVs, which demonstrates that these instruments compare favorably with other instruments using the same technique. The final entries under Reproducibility display the values determined from residue and waste measurements at this facility. The very large differences between these values and the other NCC and SGS reproducibility values in the table reveal that the other estimates are clearly inadequate for determining uncertainties in residues and waste. These last reproducibility values are about 5

to 10 times larger than the other estimates. The large differences are important for NDA Laboratory operators and safeguards personnel who are required to assess the degree to which the measurement of waste and residues can be repeated. The large uncertainties shown in the fourth row of the table more realistically reflect this repeatability whereas the other estimates are more closely related to optimal instrument performance.

Under the Bias heading in Table III, the average SGS and NCC biases in measuring reference standards (the first row of entries) are seen to have small positive values. These values serve as useful indicators that the instruments at this facility are being operated under a reasonably effective measurement control program, but should not be interpreted as estimates of the bias to be expected in waste and residue measurements. The CRV values in the second row under this heading are larger than the values in the first row, by comparison. Again, this indicates that the instruments used here compare favorably with other NCCs and SGSs, although the comparison is somewhat strained since the values in the first row were determined from repeated measurements while the CRV definition is an estimate of expected bias in a single measurement. In any case, a better estimate of bias in waste and residue assays can be obtained from the third row of entries in this category. These indicate that, at this facility, SGS assays are biased low by about 2 % whereas the NCC assays have been biased 1 % high. These values provide the most useful estimate of bias in waste measurements here because they take into account matrix and chemical variations, inhomogeneities, packaging differences, the presence of impurities, etc. In addition, they can be used to calculate the expected plutonium inventory difference due to inaccuracies in TRU waste measurements. Since the corrections in assay methodology indicated above have been implemented, the plutonium content in the waste has been underestimated by less than 15 g/year, on average.

Comparable bias and reproducibility estimates for low level waste measurements have not yet been obtained. These estimates will be difficult to acquire because of the labor involved in determining reference values for this waste form. Only chemical analyses can provide the sensitivity to achieve useful reference values for the small quantities of plutonium found in this waste, and the expense and resource allocation required to prepare the number of chemical samples that mimic the range of LLW matrix categories is prohibitive.

4. Operator's Perspective on Desirable NDA Upgrades

From an operator's perspective, no report is complete without a list of what he believes to be the most pressing NDA needs. Although these types of lists often reflect a local perspective, that is, the needs at the operator's own facility, I have attempted to incorporate impressions gathered from visiting other U.S. waste measurement facilities. In general, the needs do not express a desire for radical improvements in NDA hardware design. Instead, they concentrate on methods for improving measurement quality through upgrades to existing designs. The needs are driven by NDA workers' desires for dependable instrument operation, requirements for improved and verifiable non destructive measurement and uncertainty analysis, and in response to anticipated demands for increased automation. A brief description of these needs is given below:

- NDA instrument electronics should be more robust to withstand the difficult work environments encountered at many processing facilities. Frequently, these environments are dirty, noisy (both RF and microphonic noise may be present), and connected to fluctuating AC power supplies. In addition, some instrument designs incorporate noisy cabling and stepping motor arrangements. These degrade measurement quality and lead to increased instrument failure rates. The effects are particularly acute for gamma-ray instruments. At this facility, SGS failure rates have been as high as 3 -/month for some instruments. NDA instrument developers should consider more robust design or improved methods for isolating detectors, amplifiers, cabling, and computers from noise and power fluctuations.
- Assays of oversize TRU and LLW waste are often highly problematic at waste measurement facilities. Typically this waste is dense, consisting of metals or compacted materials, so neutron counting methods offer the best prospect. These methods are susceptible to measurement error resulting from fluctuating backgrounds, unknown spatial location of neutron sources within the waste, and matrix effects. In most waste measurement facilities within the U.S. weapons complex, instruments to assay oversize waste are either entirely lacking or perform only marginally. The resulting unreliability in the estimates of plutonium content is beginning to come under increasing regulatory scrutiny.

Instrument developers should consider increased developmental efforts in this area.

- Several improvements in SGS analysis are needed. Peak fitting and peak stripping routines should become a regular part of the SGS assay. In the U.S., several attempts have been made to incorporate these capabilities into SGS software, however few units have been fully tested and integrated into a production environment. This capability is of major importance for older waste forms that are inadequately documented or whose origins are poorly understood. For this waste, operators often have only a limited understanding of the nature of the spectral interferences they may encounter. Peak fitting and stripping algorithms can mitigate error associated with these measurements and enhance the operator's ability to perform reliable assays of these waste forms. In addition, continued research into corrections for self-attenuation and end effects must be sustained. Because waste is often heterogeneous and subject to settling inside containers, improved measurement algorithms which incorporate these corrections will prove especially useful.
- Standards, standards, standards. All U.S. waste measurement facility operators complain of the inadequacy of their own standard reference materials and of their inability to procure new ones. Obstacles to procurement include lack of fabrication facilities for new standards, expense, location of pure standards source materials, shipping problems, and lack of on-site storage capability. Because auditors and regulators increasingly require verification of NDA measurements through comparisons to standards measurements, this need will continue to escalate in the future.
- Safeguards and waste disposal regulators are becoming increasingly concerned with NDA measurement uncertainty. Presently, most instruments include statistical counting variations as the only component in the measured uncertainty. However, this is usually the minimum uncertainty in the measurement and does not reflect variability in calibration, background, container effects, the influence of spatial variations on detector efficiency, matrix effects, etc. Algorithms should be developed to include all significant random and bias sources as part of the total uncertainty reported with an NDA measurement.
- Heightened attention to expert evaluation of new software and hardware designs is needed. Our facility has experienced

numerous errors in measurement algorithms, hidden software glitches, and poor or noisy hardware design in recent instrument purchases. In addition, some commercial vendors tend to oversell the measurement capabilities of their instruments. Discussions with operators at other U.S. facilities have reenforced these impressions. To correct this condition, a center where new instrument designs can be independently evaluated is required. The center would test software and hardware components under common operating conditions and appraise measurement algorithms for a wide variety of materials, including waste. Their assessment of an instrument's performance will lead to improved confidence among operators that their measurement needs are met, alert designers and manufacturers of flaws and limitations in their products, and assure auditors and regulators of the merits of measurements performed with these instruments.

- Several developments are underway in U.S. waste management facilities which may affect NDA instrumentation design and analysis. These include automated waste handling, waste stabilization, and requirements for measurements of non-nuclear properties of waste. Driving the development for automated waste handling is a desire to reduce the risks from radiation exposure and accidents that come about from human contact. In the future, automated equipment will be used with greater frequency to move, load, measure, and dispose of waste. NDA instrument developers should be aware of these developments and alert to their implications. New instruments may have to be designed that are compatible with automated conveyor systems, can be loaded with robots, and interface easily with computers that control other measurement and loading equipment. Instrument developers may be asked to address such issues as standardization of software protocols, reliability of non NDA equipment in automated systems, and integrated system design. Stabilization of a greater variety and number of TRU waste forms is being motivated by regulatory concerns and delays in the opening of a permanent disposal repository in the U.S. Cementation, bitumenization, polymerization, and vitrification are all processes which have either been successfully implemented or show promise for waste immobilization. NDA measurement equipment may be useful for analysis of stabilized waste, however existing research on assay reliability is sparse. Increased attention should be given to the effect of different stabilization schemes on

measurement accuracy and to development of improved instrumentation for its analysis. In addition to NDA measurements, most TRU waste in the U.S. will also be subjected to radiography analysis, alpha particle monitoring, weight measurements, ultrasound testing (for drum integrity), and headspace analysis by gas chromatography (for VOCs and hydrogen). Integration of these five tasks into a single measurement station would reduce moving and loading requirements and could mesh well with improved automated waste handling systems discussed above.

5. Summation

Reported above are a review of waste measurement operations at a U.S. plutonium scrap recovery facility, uncertainty estimates for those measurements, and some recommendations for future NDA development efforts. In the first section, waste packaging and handling activities are briefly discussed, then instrument selection and quality assurance procedures are reviewed. The next section considers the results of a program to estimate reproducibility and bias in measurements of residues and waste here. SGS measurements of these materials were found to be reproducible to within 21 % and to be subject a bias that averaged -2 % over the course of the program. Neutron counter measurements were reproducible to within 14 % and had a +1 % bias when compared to reference values. Reasons for the differences in assay results and various estimates of measurement uncertainty were also discussed. Finally, this report closed with an identification of NDA needs that, from an operator's perspective, require future development efforts. The needs present software, hardware, instrument evaluation, and standards requirements for improved safeguards, quality assurance, and disposal of radioactive waste.

6. References

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Table I. Results from analysis of all SGS and NCC MVP data are summarized below. R_1 is the ratio of a single SGS or NCC measurement to a second measurement made on the same instrument at a later time. R_2 is the ratio of the SGS or NCC measurement to the CI result. The numbers in parentheses are the number of measurements used to determine each of the ratios.

	R_1	R_2
SGS	1.04 +/- 0.21 (333)	0.94 +/- 0.20 (75)
NCC	0.99 +/- 0.14 (277)	1.03 +/- 0.12 (93)

Table II. The revised MVP results are summarized below. These results exclude all assays known to be biased from the original data.

	R_1	R_2
SGS	1.01 +/- 0.21 (248)	0.98 +/- 0.14 (56)
NCC	0.99 +/- 0.14 (249)	1.01 +/- 0.12 (45)

Table III. Various estimates of reproducibility and bias in NDA analyses. % SD is the percent standard deviation of the measured data whereas % RD is the percent relative difference between the measured and reference values.

REPRODUCIBILITY	SGS	NCC
% SD of 15 Sequential Measurements	1.0 %	2.6 %
% SD of Daily Measurements	2.1 %	2.8 %
Conventional Reference Values	3 % - 100 %	3 % - 50 %
% SD from Table II	21 %	14 %

BIAS		
Average % RD of Daily Measurements	+ 0.4 %	+ 0.4 %
Conventional Reference Values	+/- 0.5 - 10 %	+/- 0.5 - 10 %
Average % RD from Table II	- 2 %	+ 1 %