# Application of MARSSIM and MARSAME Guidance to Footprint Reduction Projects at a DOE Site (LA-UR-15-28730) – 16216

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## ABSTRACT

Department of Energy regulations require that potentially contaminated items and materials being released to the public are monitored and that any residual radioactive contamination does not contribute more than 10 µSv/y (1 mrem/y). In footprint reduction and demolition projects, buildings with potential residual contamination must be carefully and systematically surveyed prior to recycling materials or releasing debris for burial in commercial landfills. Multi-agency guidance is available to assure statistically representative sampling of the items and materials, appropriate measurement quality objectives, and valid statistical approaches for release decisions. Additionally, implementing a cost-effective evaluation program for releases requires working closely with regulators and waste generators and developing a technically sound process to account for naturally occurring radioactive material (NORM) in building materials. Los Alamos National Laboratory has successfully implemented the available guidance for footprint reduction projects to help divert clean, recyclable materials from landfills and limit the possibility of mistakenly disposing of NORM as low level waste.

## INTRODUCTION

The footprint reduction program at Los Alamos National Laboratory (LANL) represents a critical step in facility modernization and significant cost savings which are broadly applicable to other Department of Energy (DOE) complex facilities. In the past five years, LANL has seen an increasing number of decontamination and decommissioning (D&D) projects for facilities with known or suspected contamination. This increase underscores the importance of defining and segregating radiological hazards from non-hazards in a timely, cost-efficient manner. Additionally, a defensible, reliable approach to radiological characterization can increase options for waste minimization and material recycling, which supports a culture of environmental stewardship.

DOE Order 458.1 *Radiation Protection of the Public and the Environment* [1] provides guidance for evaluating material releases and establishes a dose limit of 10 µSv/y (1 mrem/y) for the public release of personal property, including building materials. Additionally, DOE requires that the potential public dose from the release of the property meets as low as reasonably achievable (ALARA) guidance. When these requirements are met, LANL can release materials which would otherwise be disposed as low level radioactive waste (LLW). The cost difference between disposal pathways is significant, motivating footprint reduction project teams to meet the criteria for release.

Regulations for property release depend on the specific waste disposition pathway. At LANL, release of materials for reuse or recycle can use the surface contamination criteria provided in the former DOE Order 5400.5 [2] and reaffirmed in DOE Order 458.1 as pre-approved authorized limits (ALs) (see TABLE I). Currently, the DOE has not approved any ALs for volumetric contamination, though the American National Standards Institute (ANSI) and the Health Physics Society jointly produced guidance for material release that relies upon a dose limit to the public of 10 µSv/y (1 mrem/y) from a material's radionuclide content [3] (see TABLE II). Alternatively, release of materials to commercial or sanitary landfills from LANL is regulated by the State of New Mexico using a criterion of indistinguishable from background (IFB).

TABLE I. DOE authorized limits for release of material with surface contamination

| **Radionuclide groups and radionuclides common to LANL** | **Surface contamination limit $Bq/cm^2$ ($dpm/100cm^2$)** |
|---|---|
| U-natural, U-235, U-238 and associated decay products (Removable) | 0.2 (1,000) |
| U-natural, U-235, U-238 and associated decay products (Total) | 0.8 (5,000) |
| Transuranics, Ra-226, Ra-228, Th-230, Th-228, Pa-231, Ac-227, I-125, I-129 (Removable) | 0.003 (20) |
| Transuranics, Ra-226, Ra-228, Th-230, Th-228, Pa-231, Ac-227, I-125, I-129 (Total) | 0.02 (100) |
| Th-natural, Th-232, Sr-90, Ra-223, Ra-224, U-232, I-126, I-131, I-133 (Removable) | 0.04 (200) |
| Th-natural, Th-232, Sr-90, Ra-223, Ra-224, U-232, I-126, I-131, I-133 (Total) | 0.2 (1,000) |
| β/γ emitters (Removable) | 0.2 (1,000) |
| β/γ emitters (Total) | 0.8 (5,000) |
| Tritium and Special Tritium Compounds | 2 (10,000) |

TABLE II. Surface and volume contamination limits for the various groups of radionuclides. These limits are based on a 10 µSv/y (1 mrem/y) public dose.

| **Radionuclide groups and radionuclides common to LANL** | **Surface contamination limit $Bq/cm^2$ ($dpm/100cm^2$)** | **Volume contamination limit[a] Bq/g** |
|---|---|---|
| Group 1: High energy gamma emitters, radium, thorium, transuranics, and mobile beta- gamma emitters (e.g. Pu, Ra, Th) | 0.1 (600) | 0.1 |
| Group 2: Uranium[b] and selected beta emitters (e.g. Sr-90, U-234, U-235, U-238) | 1 (6000) | 1 |

| | | |
|---|---|---|
| Group 3: General beta-gamma emitters (e.g., Be-7, Pu-241) | 10 (60,000) | 10 |
| Group 4: Low-energy beta-gamma emitters (e.g. H-3) | 100 (600,000) | 100 |
| Group 5: Low-energy beta emitters (e.g. Sr-89) | 100 (600,000) | 1000 |

a Assuming an average surface to mass ratio of 1:1
b Natural uranium screening levels for clearance shall be lowered from Group 2 to Group 1 if decay-chain progeny are present

DOE Order 458.1 references the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) tool [4] and its supplement, the Multi-Agency Radiation Survey and Assessment of Materials and Equipment (MARSAME) tool [5] for release of real and personal property to the public. These release requirements are implemented at LANL through high-level policies and application-level procedures. As an academically-founded approach, MARSSIM/MARSAME data life cycle presents challenges at each stage. This paper presents a summary of LANL's experience in applied health physics for D&D projects. Rather than focusing on one specific project, this paper describes process and technical challenges that have been recurring themes in all projects.

## DESCRIPTION

Through policy and procedures, LANL applies the MARSSIM and MARSAME protocols to achieve representative sampling, define measurement quality objectives, and justify statistically-based decision making for release of property to the public. However, beyond technical procedure steps, successful implementation has required combining the academic MARSSIM/MARSAME approach with the realities of planned and ongoing D&D activities and dealing with the complexities in variable field measurements and rigorous statistical analysis.

In two years of working actively with D&D projects at LANL, numerous challenges have emerged. Specific concerns have included: (1) identifying process knowledge, (2) defining statistical parameters and decision criteria, (3) defining measurement quality objectives for field surveys, (4) providing statistical analysis of measurements for release decisions, (5) developing an IFB approach compensating for NORM interferences, (6) integrating a rigorous quality assurance protocol into the strict timelines of D&D projects, and (7) communicating with DOE oversight and coordinating independent verification activities to get the necessary approvals prior to material release.

## DISCUSSION

MARSSIM and MARSAME present a data life cycle for projects which consists of planning, implementation, assessment, and decision-making stages, as illustrated in Figure 1. Process and technical challenges have emerged within each of these stages. Below are discussions on the challenges, categorized by stage, and the LANL experience in each topic.

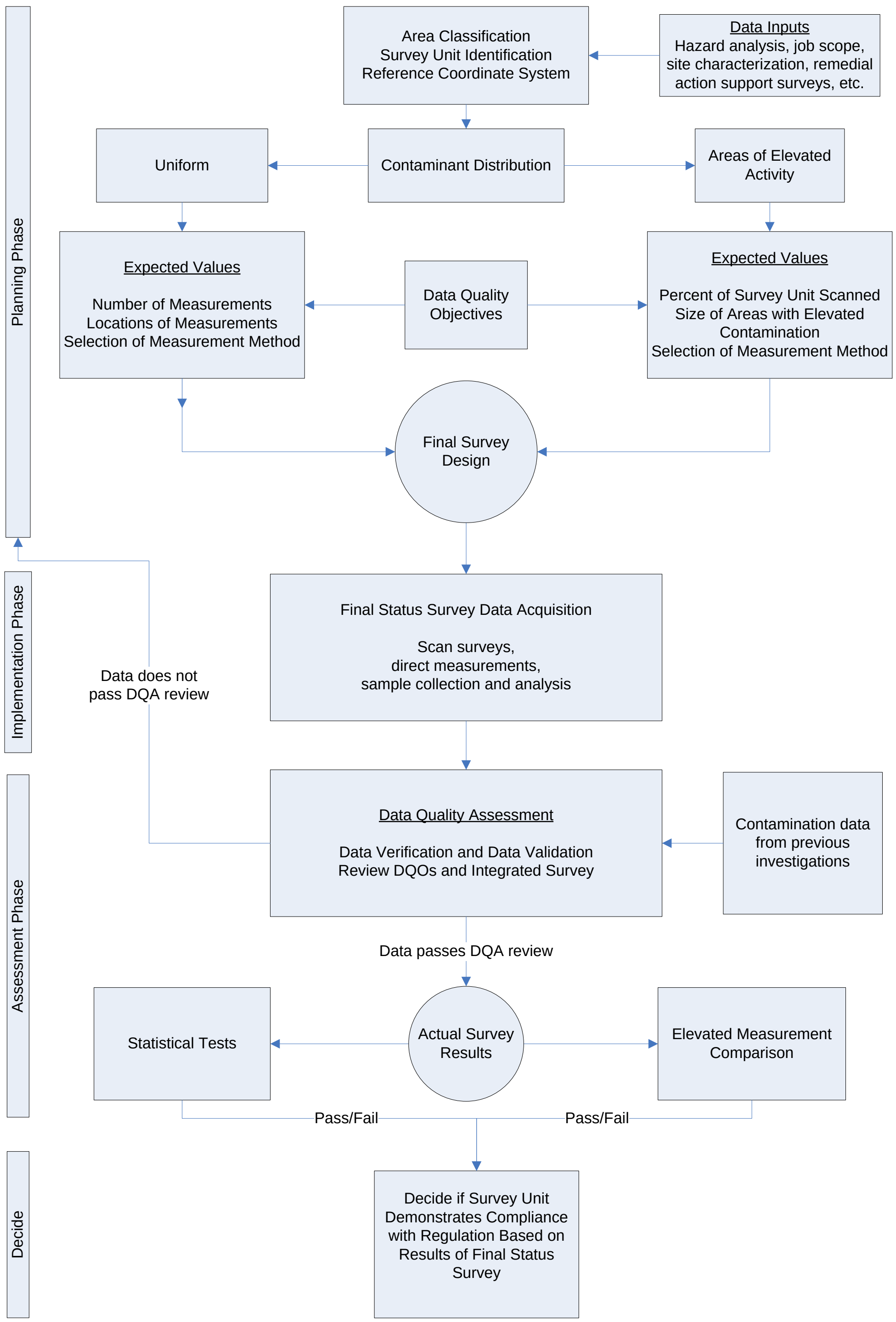


Fig. 1. Generalized process flow for the MARSSIM/MARSAME protocol

## Planning Phase and Development of Sampling Plans

*Challenge 1: Identifying process knowledge*

The initial action in scheduled D&D projects is acquiring process knowledge for the specific building, which informs fundamental assumptions about risk and defines the initial level of interest both for the survey plan and for DOE oversight. Many of the buildings at LANL are older, requiring information dating back decades. Finding workers with knowledge of past activities in these older buildings is increasingly difficult. However, searching for and interviewing these long-time employees has provided essential information. Additionally, finding archived radiological survey information dating back to historical facility operations has been problematic because early survey information is not in an electronic, searchable format. When process knowledge or survey data are unavailable, caution is exercised, and scoping or characterization surveys for residual radioactivity are used. These surveys rely heavily on judgement or biased sampling by radiological control technicians experienced in finding contamination. Information from these surveys is used to develop final release surveys and can also be used for cost estimates by contractors bidding for the D&D project.

*Challenge 2: Defining statistical parameters and decision criteria*

LANL uses two pathways for releasing items into the public domain. The first pathway is for situations requiring measurements indistinguishable from background (IFB). This pathway, described in MARSAME as Scenario B, includes soil or construction debris sent to commercial landfills. The second release pathway, MARSAME's Scenario A, is for items that have residual radioactivity above background, but at levels less than the AL (resulting in a dose rate less than 10 µSv/y (1 mrem/y) to a member of the public). Thus, the objective of the measurements is to provide quantitative information to determine if 1) if the item is free of detectable radioactivity (i.e., IFB), or 2) the amount of any residual radioactivity is below the appropriate ALs and consistent with ALARA considerations.

Statistical determination of the number of samples using the MARSSIM/MARSAME process requires selection of release criteria and knowledge of the levels and homogeneity of contamination expected in the property. Additionally, the capabilities and limitations of the instrumentation have to be accounted for, and finally, the Type 1 and 2 error probabilities should be selected with knowledge of the acceptable level of risk and input from institutional-level managers.

In MARSSIM/MARSAME, the sampling rigor (number of required measurements) is based heavily on a calculation of the relative shift, which is an estimator of the degree of difficulty of accurately differentiating a clean survey unit from a contaminated one. This calculation compares the expected survey unit standard deviation with a statistical "gray region," which is a measure of the difference between expected and acceptable radionuclide concentrations (see Eq.1).

$$Relative\ Shift = \frac{\Delta}{\sigma} = \frac{UBGR - LBGR}{\sigma} \quad \text{( Eq.1 )}$$

Where ($\Delta$) is the width of the gray region, ($\sigma$) is expected standard deviation of the radionuclide concentration in the survey unit, and UBGR and LBGR are the upper and lower bounds of the gray region, defined differently for the IFB and AL release pathways and described below.

Combined, these estimates and bounding assumptions are used to choose representative sampling in the survey unit. The following inputs are generally used for LANL D&D projects:

IFB release pathway:

- The null hypothesis, $H_0$, is that the survey unit is indistinguishable from background. "Passing" the survey unit, and releasing the material, would result from failing to reject the null hypothesis.
    - o Type 1 error (incorrectly rejecting the null hypothesis) would mean concluding the material was contaminated when it was IFB.
    - o Type 2 error (incorrectly failing to reject the null hypothesis) would mean concluding the material was IFB when it was contaminated.
- LBGR set to zero or at the expected value of uncontaminated reference materials. The chosen value of Type 1 error, generally 5% in LANL D&D projects, is defined for a true survey unit mean equal to the LBGR.
- UBGR set to a value that is statistically above the LBGR. The chosen value of Type 2 error, generally set at 10% in LANL D&D projects, is defined for a true survey unit mean equal to the UBGR.
- If data from uncontaminated reference materials are used, the choice of the material must be representative and the measurements made using the same type of instrument and survey technique.

AL release pathway:

- The null hypothesis, $H_0$, is that the survey unit is contaminated above the AL. "Passing" the survey unit, and releasing the material, would result from rejecting the null hypothesis.
    - o Type 1 error (incorrectly rejecting the null hypothesis) would mean concluding the item was below the AL, when in fact it was contaminated above the AL.
    - o Type 2 error (incorrectly failing to reject the null hypothesis) would mean concluding the material was contaminated above the AL when it was clean.
- LBGR set at the expected value (mean or median) measured in the scoping/characterization survey. The chosen value of Type 1 error, generally 5% in LANL D&D projects, is defined for a true survey unit mean equal to the LBGR.
- UBGR set to the AL. The chosen value of Type 2 error, generally set at 10% in LANL D&D projects, is defined for a true survey unit mean equal to the UBGR.

- Note that there are no preapproved ALs for volume contamination, and establishing volume ALs would require regulatory approval.

## Implementation Phase

*Challenge 3: Defining measurement quality objectives for field surveys*

Working with the radiological technicians to develop and meet measurement quality objectives is important. The requirements for instrumentation, survey technique (scan, scalar counts, smears, etc.), measurement sensitivity, units of measurements (e.g., dpm/100 $cm^2$ rather than NDA), quality assurance measures, and formats for documenting the results are all critical components of a successful survey. The selection of instruments must match the measurement quality objectives. Field measurements are inherently variable due to changing environmental conditions (e.g., temperature). Also, differential shielding of emissions from nearby NORM sources and/or external radiation fields not related to the building materials can interfere with measurements and must be accounted for.

Once the survey is complete, the statistical evaluation of the data can proceed. Scoping or characterization surveys are used to estimate homogeneity of any contamination and to classify the building as to potential for contamination and levels.

## Assessment and Decision Making Phases

*Challenge 4: Providing statistical analysis of measurements for release decisions*

Comparisons of measurements against MDAs, ALs, or reference measurements of NORM in building materials are made using statistical techniques. Generally, non-parametric tests are used since measurements of radioactivity in/on materials are often result in skewed distributions. The assessment phase requires establishing decision criteria for hypothesis testing. LANL's decision criteria include:

IFB release pathway:

- If all measurements are: 1) ≤ detectable levels, or 2) < reference background values such as the 95% Upper Confidence Level of the background mean (95% UCL), then no further action is required and the items are candidates for unrestricted release.
- If all measurements are > 95% UCL of background, then the item is not a candidate for release through the IFB pathway and the item can be considered for decontamination or decay in storage followed by resampling before it can be released.
- If the mean for a set of measurements is below the 95% UCL of background, but some individual measurements are above the 95% UCL, then statistical analysis is needed. Generally, non-parametric statistical approaches are used to evaluate the null hypothesis. If contamination is present in background,

the Wilcoxon Rank Sum test is suggested, and if contamination is not present in background, the Sign Test is suggested.

AL release pathway:

- If all measurements are ≤ AL, then no further action is required and the items are candidates for unrestricted release.
- If all measurements or the 95% UCL are > the AL, then the item is not a candidate for release through the AL release pathway and the items can be considered for decontamination or decay in storage followed by resampling before it can be released.
- If the 95% UCL for a set of measurements is below the AL, but some individual measurements are above the AL, then statistical analysis is needed. Generally, non-parametric statistical approaches are used to evaluate the null hypothesis. If contamination is present in background, the Wilcoxon Rank Sum test is suggested, and if contamination is not present in background, the Sign Test is suggested.

*Challenge 5: Developing an IFB approach compensating for NORM interferences*

Using the IFB release approach requires searching for and quantifying very low levels of potential contamination in materials with variable NORM content. Differentiating clean and contaminated materials is complicated by two primary factors. First, typical field instruments are count rate meters and do not provide isotopic information. These instruments have low-level detection capability, but the measured count rates are influenced by NORM. Building materials can contain NORM at levels comparable to, or in some cases exceeding, regional soils, rock, and sediment, and there is substantial variation in the concentrations of these radionuclides. Second, measurements for residual surface contamination are complicated by buildup of additional radioactivity through deposition of radon decay products. These longer lived natural radionuclides include Pb-210, which contributes to beta emission with Bi-210 beta decay in equilibrium, and Po-210, which contributes to alpha emission [6][7]. The NORM within building materials and the retention of deposited Pb-210, Bi-210, and Po-210 result in both alpha and beta counts in measurements for surface activity. This background interference complicates the detection of anthropogenic radionuclides deposited due to facility operations. Therefore, it is critical to characterize the measurement background when making release determinations.

Notably, the traditional measurement approach for surface contamination includes smears for removable contamination as well as direct counts of total contamination. Measurements for removable contamination on uncontaminated surfaces are not expected to be positive because NORM in the building material is not removable. Therefore, smear counts are compared to the instrument’s minimum detectable concentration for IFB release decisions.

A study was done to characterize the influence of NORM on surface contamination measurements on common building materials [9]. For this, 60-second direct scalar measurements of alpha and beta radiation were made using an Eberline SHP380AB probe coupled to an E600 instrument. These instruments can be programmed to display net activity by subtracting out the instrument background from the gross counts obtained during the direct survey. Measurements using several SHP380AB instruments were made on uncontaminated surfaces of building materials at a variety of locations to characterize background levels of alpha and beta activity. The building materials selected included painted and bare concrete, metal (painted, rusted, and galvanized), wallboard, stucco, tile, wood (bare and painted), ceiling tile, and carpet. In many cases, two repeated measurements were made in each location with the same instrument to assess measurement variability for quality assurance. Similarly, measurements at the same locations were repeated with a second instrument to assess inter-instrument variability.

Measurement results of uncontaminated, reference building materials are shown in Figure 2. Beta values generally range between 500 and 5000 dpm/100 cm$^2$ and alpha values range between 10 and 100 dpm/100 cm$^2$. The biggest differences across the material types were found in the rusted metals, where the alpha activities appear higher and the beta activities lower than the other materials. These distributions of measurements on uncontaminated building materials can be used to statistically determine if field measurements on building materials are IFB.

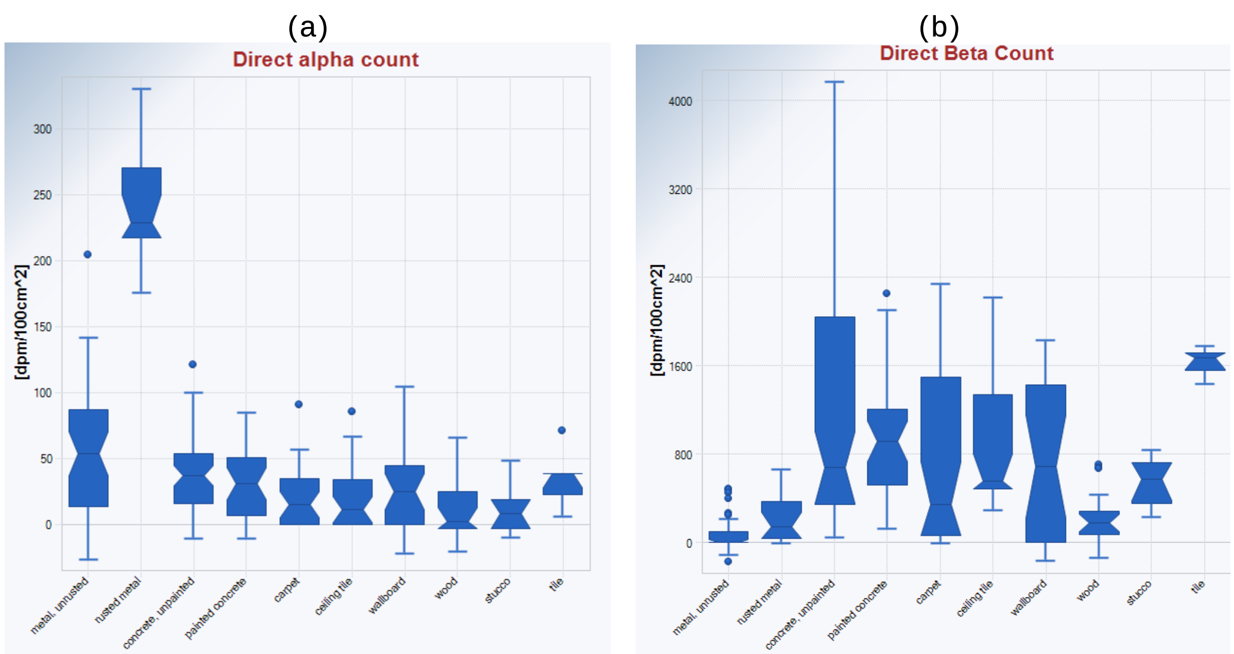


Fig. 2. Surface contamination measurements for alpha (a) and beta (b) radioactivity of NORM on uncontaminated building materials.

MARSAME describes appropriate comparisons and statistical approaches to test for differences in field measurements from background. These IFB analysis techniques can include direct comparison of field measurements to the 95% Upper UCL of the mean of the background measurements, or comparing the distribution of field measurements to the distribution of background measurements using non-parametric statistical tests. Due to expected counts from NORM, LANL's IFB analyses use the Wilcoxon Rank Sum (WRS) test, which compares the counts of survey unit material to counts of reference area material. In the WRS test, all measurements are ranked in increasing order, and the sum of the survey unit ranks is compared to a critical value calculated using Eq. 2.

$$WRS\ Critical\ Value = \frac{m(n+m+1)}{2} + z\sqrt{\frac{nm}{12}\left[(n+m+1) - \sum_{j=1}^{g}\frac{t_j(t_j^2-1)}{(n+m)(n+m+1)}\right.} \qquad (\text{Eq.2})$$

Where (m) is the number of measurements in the survey unit, (n) is the number of measurements in the reference area, (z) is determined by the acceptable statistical error, (g) is the number of groups of tied measurements, ($t_j$) is the number of tied measurements in the $j^{th}$ group.

This IFB approach has the advantage of providing decision makers guidance on release decisions using only net count rates without isotopic information. Hypothetically, it is possible that some fraction of the measured counts is due to anthropogenic radionuclides, so it is instructive to compare the reference count rates in Figure 2 with other standards for context. For example, the maximum count rates in Figure 2 are lower than the 1 mrem/yr (10 μSv/y) reference values in ANSI 13.12 (see TABLE II) and generally below the authorized release values presented in DOE Order 5400.5 (see TABLE I). The only exception is for transuranics, which have a total alpha surface contamination release limit of 100 dpm/100cm$^2$, which is lower than the upper range of NORM alpha values we measured for metals. Though metals are known to collect and retain radon progeny, some care should be exercised when interpreting count rate data for metals in areas with potential transuranic contamination.

*Challenge 6: Integrating a rigorous quality assurance protocol into the strict timelines of D&D projects*

As LANL's footprint reduction program continues to grow, an ongoing challenge in compliance with DOE Order 458.1 is keeping track of D&D projects and maintaining relationships with the project teams to ensure that each project meets regulatory requirements. Cost and schedule are often the primary drivers in this process, so one of the key techniques for ensuring compliance is being engaged early and communicating the time requirements of subject matter expert review. LANL's experience has indicated that clearly communicating compliance requirements is beneficial. For example, a checklist of necessary surveys, reports, reviews, and signatures can help restrict demolition and movement of material until internal and DOE oversight authorizations are in place. Although MARSSIM and MARSAME specify the steps necessary to making a valid statistical decision, practical application requires scheduling time for historical fact finding, iterative survey development (characterization surveys and any remedial action support surveys), statistical analysis, and DOE independent verification.

*Challenge 7: Communicating and coordinating with DOE for independent verification*

Finally, DOE Order 458.1 requires that property releases to the public have some level of independent verification (IV) by DOE. The IV is implemented based on a graded approach that can vary from simple document reviews up to side-by-side measurements, and the scope and timeline of IV activities must be communicated between the project leaders, internal health physics reviewers, and DOE oversight.

## CONCLUSION

As LANL moves forward with its footprint reduction program, the diversity of building size, complexity, contamination potential, and available process knowledge continues to grow. Fortunately, MARSSIM and MARSAME are very flexible tools. Applying the statistical basis of MARSSIM and MARSAME to footprint reduction projects of various scales has allowed the successful release of clean building materials from LANL for recycle and as IFB waste. On the other hand, surveys of some potentially contaminated buildings have found distinguishable contamination and supported decisions to send waste for LLW disposal. Areas of ongoing work include evaluating materials with potential volumetric contamination, planning and interpreting surveys for unique and mixed building materials, and increasing the data set for reference materials.

LANL's experience continues to merge the academic advantages of a MARSSIM-based statistical design with the schedule constraints of D&D projects in a way that preserves public safety and demonstrates regulatory compliance in a cost effective manner. Challenges and successes experienced at LANL may be useful to DOE facilities or other entities seeking to avoid the cost associated with LLW disposal or meet an increasing demand for environmental sustainability.

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