

From: Taylor, Casmere H COL NNMC [Casmere.Taylor@med.navy.mil]
Sent: Tuesday, October 18, 2011 2:53 PM
To: Lawyer, Dennis; Ullrich, Elizabeth
Subject: Walter Reed Army Medical Center Decommissioning (Main Post) Follow-Up Information.
Attachments: WRAMC Main Post Surveys- Revised Field Schedule 30Sep11.pdf; Evaluation of Low Energy Beta Surface Contamination 30Sep11.pdf; Evaluation of Low Energy Beta Surface Contamination 30Sep11.docx

08-01738-02
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Mr. Lawyer & Staff;

Per our previous TELECON for your review are the following information;

1. Attachment 1 (WRAMC Main Post Surveys) is a time-line of our projected dates for the Main Post Survey for which you are free to come during any time of our survey. Please let us know at least the day before or the day of so we can ensure the individual names are noted with the security office.

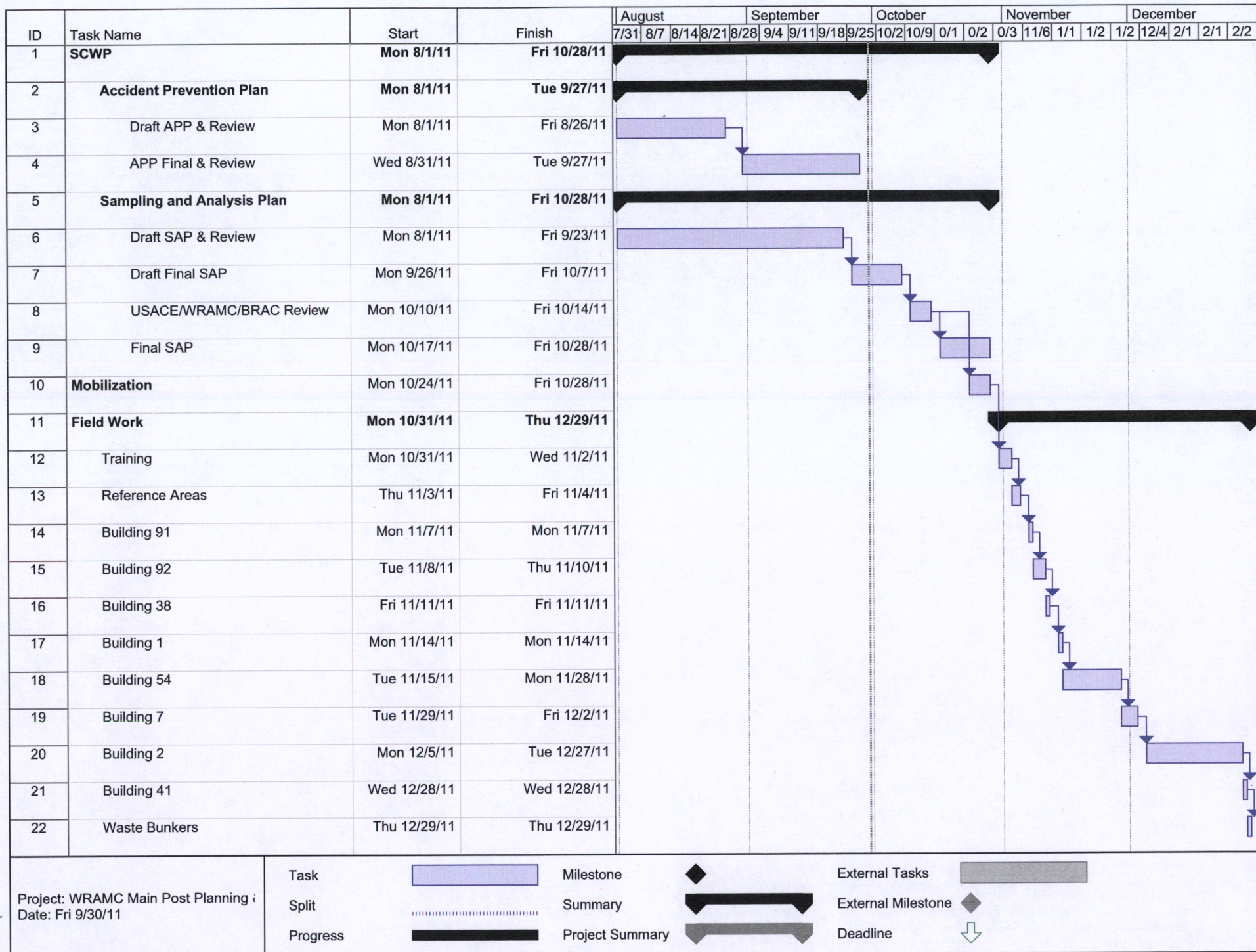
2. Attached is the Memo for your staff containing all of the information with respect to low-energy beta (LEb) detection for the Main Post surveys.

Note: Should you have any questions or concerns please do not hesitate to contact the undersigned and I will insure your request are noted and follow-up received.

V/r

COL Caz Taylor
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EVALUATION OF LOW ENERGY BETA SURFACE CONTAMINATION

1.0 Purpose

The purpose of this paper is to establish the generally accepted industry practices for evaluation of radioactive surface contamination low energy beta emitters such as tritium and Ni-63, and then make application to surveys to be performed for decommissioning at Walter Reed Army Medical Center (WRAMC), Main Post, Washington, DC. References considered include international standards, peer reviewed documents, documents developed by federal agencies and manufacturers. Generally, the approach at WRAMC will include:

- Direct surface measurement of low energy betas with a gas flow windowless detector with indirect measurements (smears) at locations with directly measurable contamination.
 - MARSSIM and ISO-7503 calculations are required.
 - Calibration to H-3 energy required as the lowest energy to be detected.
 - Use of a H-3 performance check source as the lowest energy to be detected.
 - Performance and contamination checks conducted every four hours of use; upon failure, data rejected through previous survey period.
- Indirect measurements (smears) of low energy beta will be performed.
 - Smears to be collected across the useful range of direct measurements which were greater than the minimum detectable activity.
 - Smears to be collected at bias locations where detector cannot be used (e.g., drains and air exhausts).
 - Analyzed off site at a DoD Environmental Laboratory Accreditation Program (ELAP) accredited laboratory.
 - Analysis consists of liquid scintillation analysis for gross alpha and gross beta; no isotopic planned but will be considered as necessary.

2.0 Background

Radionuclides including tritium and nickel-63 used at WRAMC consisted of both sealed and unsealed radioactive material. The Nuclear Regulatory Commission (NRC) provides a look-up table as screening level release criteria in Appendix H of NUREG-1757, Vol. 2, *Consolidated NMSS Decommissioning Guidance*. The building surface screening level for tritium is $1.20\text{E}+08$ dpm/100cm² and for Ni-63 is $1.8\text{E}6$ dpm/100cm². NUREG-1757 requires that residual radioactivity on surfaces must be mostly fixed (not loose), with the fraction of loose (removable) residual radioactivity no greater than 10 % of the total surface activity. For cases when the fraction of removable contamination is undetermined or higher than 0.1, licensees may assume for screening purposes that 100% of surface contamination is removable, and therefore the screening values should be decreased by a factor of 10.

Instrumentation and techniques proposed at WRAMC for measurement of low energy beta surface contamination utilize a windowless gas flow proportional detector (Ludlum 44-110) and wetted smears for analysis by liquid scintillation techniques.

2.1 MARSSIM and International Standards Organization (ISO) Concepts

ISO-7503-1 states that as a result of plausible and conservative assumptions, the values for source efficiency (ϵ_s) should be used in the absence of more precisely known values:

- $\epsilon_s = 0.5$ [beta emitters ($E_{\beta\text{max}} \geq 0.4 \text{ MeV}$)]
- $\epsilon_s = 0.25$ [beta emitters ($0.15 \text{ MeV} < E_{\beta\text{max}} < 0.4 \text{ MeV}$)]

The source efficiencies described are not applicable to H-3 with beta energies of 0.0186 MeV max and 0.0056 MeV average. Neither are they applicable to Ni-63 with beta energies of 0.066 MeV max and 0.017 MeV average. Because of this, ISO developed ISO-7503-2 specifically for tritium and ISO-7503-3 for other low beta-emitters ($E_{\beta\text{max}} < 0.15 \text{ MeV}$).

ISO-7503-2 applies to the evaluation of tritium contamination on surfaces of equipment and facilities, containers of radioactive materials and sealed sources.

- Tritium surface contamination is defined as the total activity of tritium adsorbed upon and absorbed into the surface.
- Directly measurable tritium is defined as the fraction of the tritium surface contamination available for direct measurement.
- The removable tritium surface contamination is defined as the fraction of tritium surface contamination which is removable or transferable under normal working conditions.

ISO-7503-2 provides both a caution and position regarding tritium measurements: "The total surface contamination cannot be accurately evaluated by direct or indirect methods. Direct measurements are carried out with contamination-measuring instruments which will not respond to all activity absorbed below the surface. Indirect measurements performed by wet smear tests generally provide a reasonable estimate of the removable surface contamination at the time of collection. .. smear test evaluation is an adequate method of assessing the actual radiological hazard arising from incorporation in the course of contact with surfaces by tritium."

ISO-7503-3 indicates that adequate instruments for direct measurement of Ni-63 include a windowless gas flow proportional counter and for indirect (smears) detection includes liquid scintillation counters. As neither ISO-7503-2 nor -3 provide a suggested source efficiency for direct measurements, one option is to use a total (4π) efficiency factor provided with the calibration of the instrument. A second option is to apply a removable factor of 0.1 for smears indicated in ISO-7503-1 and -2.

2.1.1 Direct Measurements

Measuring instruments that directly detect low energy beta surface contamination may respond to both removable activity and part of the activity absorbed into the surface; particularly, this is addressed for tritium. The ISO specifically addresses the use of windowless gas proportional and scintillation detectors.

- The ISO indicates that the background should be checked frequently because the detector is liable to become contaminated. This would yield a false positive.
- The correct functioning (performance check) should be performed using a suitable check source. The stated frequency is daily for instruments in frequent use; otherwise before each use. Deviations of more than 25% from an agreed value shall give rise to a recalibration. There is no specificity on what is a “suitable” check source for tritium measurements. ISO-7503-3 lists Ni-63 as an appropriate radionuclide for evaluation.

The basic formula from MARSSIM for calculating the dpm per 100 cm² is repeated here as follows:

$$\frac{dpm}{100cm^2} = \frac{\frac{C_S}{t_S} - \frac{C_B}{t_B}}{\epsilon_{total} \left(\frac{a}{100} \right)}$$

Where

- C_S = integrated counts recorded by the instrument
- C_B = background counts recorded by the instrument
- t_S = time period over which the counts were recorded
- t_B = time period over which the background counts were recorded
- ϵ_{total} = total efficiency
- a = physical probe area

2.1.2 Indirect Measurement (Smears)

MARSSIM implies that removable activity is at best semi-quantitative and is difficult to interpret quantitatively.

MARSSIM Section 8.5.4, Removable Activity, is quoted: “Some regulatory agencies may require that smear samples be taken at indoor grid locations as an indication of removable surface activity. The percentage of removable activity assumed in the exposure pathway models has a great impact on dose calculations. However, measurements of smears are very difficult to interpret quantitatively. Therefore, the results of smear samples should not be used for determining compliance. Rather, they should be used as a diagnostic tool to determine if further investigation is necessary.

The 1988 ISO notes that a smear wetted with glycerol is preferable for the determination of tritium surface contamination. Later studies which will be discussed recommend distilled water.

Some clarity is provided through the Health Physics Society which has posted Answer to Question #5897 Submitted to "Ask the Experts and is quoted in part:

“The liquid scintillation counting (LSC) efficiency can be greatly affected by the transparency of the wipe, the amount of dirt/soiling on the wipe, and the presence of quenching

chemicals on the wipe. There have been numerous studies of various aspects of the problem by a number of individuals, but I don't believe an overriding consensus has been reached as to whether wet wipes are better than dry, what solvents are the best if wet wipes are done, or what is the preferred wipe material. Based on what I have read, I believe it is accurate to say that for removal of many forms of tritium contamination, wipes wetted with distilled water will generally do a better job of removal of contamination from most surfaces than will dry wipes."

A 1992 article by R. Klein et al., "Detecting Removable Surface Contamination" (*Health Physics*, v. 62[2], 186-189), compared glass fiber filters and cotton swabs for their effectiveness in removal of surface contamination and in LSC. They concluded that the glass fiber filters were noticeably better and that the filters wet with water were generally at least twice as efficient as dry wipes (glass fiber filters were Whatman Type GF/A) in removal of surface contamination."

The selected DoD ELAP accredited laboratory, GEL Laboratories, will establish a minimum detection limit of 30 dpm as indicated by MARSSIM for liquid scintillation analysis for low energy beta analysis of smears.

The generally applicable method for measuring tritium smear samples is by liquid scintillation counting. The use of proportional detectors (windowless or internal gas-flow types) is more appropriate for dry smears.

The activity per unit area, A_R , of the removable tritium contamination of the surface being wiped is given by the equation:

$$A_R = \frac{A}{F \times S}$$

where

A is the activity of the smear sample assessed by standard LSC technique;

F is the removal factor;

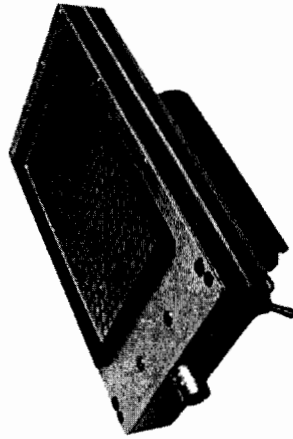
S is the area smeared in centimeters squared.

The removal factor should be determined experimentally, but if not a value of $F=0.1$ is to be used.

3.0 Description and Characteristics of Ludlum 44-110 Detector

A photograph of the detector is shown below and the manufacturer provides the following specifications:

Figure 1 - Photograph of Ludlum 44-110 Detector



Indicated use: tritium surface survey/windowless beta survey is designed to be placed on a flat surface and then purged with P-10 gas for 20 seconds before counting

Detector: windowless gas flow proportional

Recommended Counting Gas: P-10 (10% methane, 90% argon)

Gas Purge Time: approximately 20 seconds

Window: windowless

Window Area: 126 cm² (19.5 in²) active; 100 cm² (15.5 in²) open

Efficiency (4 π): 25%—³H

Background: typically 400 cpm

Suggested instruments: Models 12, 2000, 2200, 2221, 2350-1, 177-61

Operating Voltage: typically 1650-1700 volts

Counter Threshold Setting: typically 4.0 mV

Size: 12.4 x 11.7 x 22.9 cm (4.9 x 4.6 x 9 in.) (H x W x L)

Weight: 0.9 kg (2 lb)

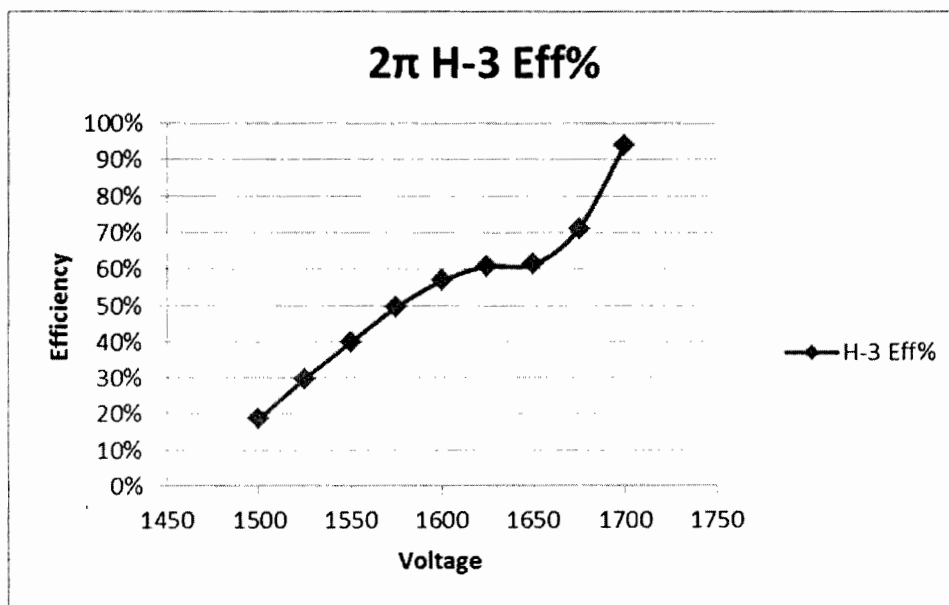
The operating manual indicates that in order to prevent residue forming on the chamber and anode wires, the detector should not be used on very dirty or dusty surfaces. Extreme humidity in the air (in excess of 85%) and moisture on surfaces can cause the detector to behave erratically. Therefore, it is not recommended to use the detector in high humidity or moist conditions.

Static electric on a surface is sufficient to prevent some tritium beta particles from escaping from the surface, thus reducing calculated surface emission.

3.1 Calibration Efficiency Curve

A typical calibration efficiency curve for H-3 is provided below; an operating range for each detector will be made and the operating voltage and efficiency will differ slightly from detector to detector.

Figure 2 – H-3 Efficiency Voltage Plateau



3.2 Static Minimum Detection Concentration

NUREG-1507 provides a rigorous derivation of the expression for instrument sensitivity, typically stated as the minimum detectable concentration (MDC). Determining the MDC in cpm requires knowledge of the survey instrument efficiency, the material source efficiency and the background rates. The following is an *a priori* analysis and actual MDCs will be determined with background and calibration data for instruments to be used. Per the MARSSIM "Roadmap" for direct measurements and sample analyses, minimum detectable concentrations (MDCs) less than 10% of the DCGL are preferable while MDCs up to 50% of the DCGL are acceptable.

For static measurements, the background will be taken on similar material that has not been impacted by radioactive material for 10 counts. The following equation for the MDC from NUREG-1507, (Equation 3-10), as modified for efficiency and detector area, applies:

$$\text{Static MDC} = \frac{3 + 4.65 * \sqrt{C_B}}{(\epsilon_{total}) * T * (\frac{a}{100})}$$

where:

$$\begin{aligned}C_b &= 200 \text{ counts in analysis time of 30 seconds} \\ \epsilon_{\text{total}} &= \text{total efficiency from manufacturer for tritium: 0.25} \\ T &= 0.5 \text{ minute} \\ a &= 126 \text{ cm}^2, \text{ probe area}\end{aligned}$$

With these parameters, the static MDC is approximately 435 dpm/100 cm². The MDC is significantly less than 10% of the NRC's screening level or any proposed reduction. As tritium emits the lowest energy beta, a factor 3.5 less than Ni-63, response and efficiency to the tritium beta will be conservative.

4.0 Commercially Available Performance Source

For this work, an H-3 beta reference source will be used. The source has an activity of 10kBq and will be calibrated for emission only (100-300 β/s). The source has an active diameter of 50 mm with overall dimensions of 60 mm x 3 mm. The reference source is manufactured by Isotrak in Germany, is fully traceable to NIST, and distributed by:

Eckert & Ziegler Analytics
1380 Seaboard Industrial Blvd.
Atlanta, GA 30318
Tel. 404-425-5014

5.0 Conclusions and Survey Approach

As there is no scan technique the density of direct measurements will be increased by limiting the size of the survey units compared to MARSSIM permitted sizes.

It should be noted that any stated "Residual Activity" is conservatively assumed to be entirely due to Ni-63 which has a lower DCGL. In actual fact, a portion (or even all) of the gross residual count rate may be attributable to other radionuclides of concern. However, for the sake of evaluating potential contamination, this assumption was deemed appropriate.

If the fraction of removable contamination cannot be determined as significant measurable contamination may not exist or if determined higher than 0.1, surface contamination will be considered as 100% removable and the screening values will be decreased by a factor of 10. As the screening level is conservatively at 1.8E6 dpm/100 cm² and the direct MDC is under 500 dpm/100cm², the difference remains quite large for statistical evaluations.

The approach at WRAMC will include:

- Direct surface measurement of low energy betas with a gas flow windowless detector with indirect measurements (smears) at locations with directly measurable contamination.
 - MARSSIM and ISO-7503 calculations are required.
 - Calibration to H-3 energy required as the lowest energy to be detected.
 - Use of a H-3 performance check source as the lowest energy to be detected.

- Performance and contamination checks conducted every four hours of use; upon failure, data rejected through previous survey period.
- Indirect measurements (smears) of low energy beta will be performed.
 - Smears to be collected across the useful range of direct measurements which were greater than the minimum detectable activity. A minimum of two smears will be collected in each survey unit regardless of direct measurement results to provide additional quality control of the technique.
 - Smears will be wetted with distilled water.
 - Smears to be collected at bias locations where detector cannot be used (e.g., drains and air exhausts).
 - Analyzed off site at a DoD Environmental Laboratory Accreditation Program (ELAP) accredited laboratory.
 - Analysis consists of liquid scintillation analysis for gross alpha and gross beta; no isotopic planned but will be considered as necessary.

References

- 1 *Evaluation of Surface Contamination-Part1, Beta Emitters (maximum beta energy greater than 0.15 MeV) and Alpha Emitters* (ISO 7503-1), International Organization for Standardization, 1988.
- 2 *Evaluation of Surface Contamination-Part2, Tritium surface contamination* (ISO 7503-2), International Organization for Standardization, 1988.
- 3 *Evaluation of Surface Contamination-Part3, Isomeric transition and electron capture emitters, low energy beta-emitters ($E_{\beta_{max}} < 0.15$ MeV)* (ISO 7503-3), International Organization for Standardization, 1996.
- 4 *Minimum Detectable Concentrations with Typical Radiation Survey Instruments for Various Contaminants and Field Conditions*, NUREG 1507, US NRC, June, 1998.
- 5 *Multi-Agency Radiation Survey and Site Investigation Manual*, EPA/402/R-97-016, Revision 1. NUREG-1575, US NRC, August, 2000.
- 6 *Consolidated NMSS Decommissioning Guidance*, NUREG-1757 Volumes 1 and 2, US NRC, 2003.
- 7 *Ludlum User Manual – Ludlum Model 44-110 and Model 44-110-1*, September 2006, Ludlum Measurements, Inc., Sweetwater, Texas, 79556.
- 8 Department of Defense Quality Systems Manual (DoD QSM), Version 4.2 (Dec 2010); provides baseline requirements for the establishment and management of quality systems for environmental testing laboratories performing services for the Department of Defense.