Michael Reimer, PhD
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January 15, 2018
RE: SUC 1593

Ms Amy Snyder, Senior Project Manager
Materials Decommissioning Branch (MDB)
Division of Decommissioning, Uranium Recovery, and Waste Programs (DUWP)
Office of Nuclear Material Safety and Safeguards (NMSS)
U.S. Nuclear Regulatory Commission (NRC)
Washington, D.C. 20555

Dear Ms Snyder,

I am sure you recognize the Army response of December 15, 2017 to the NRC request for additional information provides very little clarified material on sampling procedures at Pohakuloa Training Area in Hawaii (ML18009A456) and nothing on the primary question regarding possible isotope dilution using the composite sampling procedure.

The Army states “The composite sediment sampling procedure described in the site specific Environmental Radiation Monitoring Plan (ERMP) for Pohakuloa Training Area (PTA) was developed to be consistent with historical sampling procedures at PTA and the other 17 installations included in SUC-1593.”

This is an inadequate response because there is no assurance that previous sampling was proper and there is no compelling correlation offered that sampling elsewhere is valid at PTA.

PTA is a unique site among all the other installations where DU has been used and remains at the installation. It is an area composed of recently deposited basaltic material derived from oceanic materials. It has a climate predominantly influenced by its subtropical location and island setting. The latter greatly influences the formation of any soil from bedrock, soil typically being a major component of sediment, with very specific chemical and physical compositions. None of the other locations the Army oversees for DU has the attributes that are found at PTA.

The previous statement by the Army that no sediment exists at PTA (ML1206A506) and that of the NRC comment, including the observation that no soil exists at PTA, cannot be summarily overlooked or dismissed. The Army further states that there are no permanent streams at PTA so any sediment (claimed to be nonexistent) collected miles distant from the Radiation Controlled Areas (RCA) has never been shown to be derived from the RCAs. Again, the latter possibility of materials at the sampling site derived from material at the RCA is highly questionable when there are intermittent historical lava flows that could act as barriers to any direct water-flow transport from the RCAs.

A question posed by the NRC to the Army concerned the possibility of sample dilution by combining multiple aliquots of soil. This did not appear to be directly addressed in the response. There is a link to a report by Oak Ridge that NRC proffered that could be used as a basis for that response. Oak Ridge Institute for Science and Education, "Technical Bases and Guidance for the Use of Composite Soil Sampling for Demonstrating Compliance with Radiological Release Criteria," Oak Ridge, Tennessee, 2012 (Adams accession no. ML13101A090). This is a reasonable presentation of composite sampling, the method used at PTA, albeit for only one site.

I will use that report as a foundation to compare to what is being done at PTA for sediment sampling particularly involving dilution.

I am also concerned that the Army suggests modifying section 3.2 of the site specific ERMP. I believe such a suggestion should go through the amendment process and not be unilaterally changed. As it stands, the Army, as license holder, should not be allowed to modify the SUC-1593 license and its requirements without proper NRC procedural review.

Because this modification has been presented by the Army to the NRC for formal consideration, I will provide an alternative. This very germane alternative is included near the end of my commentary of the Army response to the NRC question on dilution (December 15, 2017 letter to the NRC). The alternative I provide has significant advantages as it also addresses the failure of the single site composite sampling to reach the objective of the sampling program and corrects for the dilution problem. My commentary should be sufficient to convey the concepts but I will be glad to provide greater details upon request.

The Army response indicates that three samples over time have already been collected. Would you be able to provide me with the reports or inform me of the location and availability of those analyses including the sampling collection data sheets and analytical QA/QC sheets including chain of custody documents?

Sincerely,

/s/

Michael Reimer, PhD
Retired geologist

Commentary on U.S. Army December 15, 2017 response to the NRC inquiry of November 29, 2017 for additional information regarding the 10CFR2.206 for the Pohakuloa Training Area, Source Materials License no. SUC-1593.
Prepared January 15, 2018

It appears that the Army did not directly address the question posed by NRC (ML18009A456) about sample dilution at Pohakuloa Training Area (PTA), Hawaii County, Hawaii. Rather, the Army replies that it is going to do what it has always done (historic sampling procedures) in regard to sampling. The basis for the request presented to the Army includes an Oak Ridge report (ML13101A090) that does address the issue of possible sample dilution.

The attempt to use a generic sampling method for DU detection at PTA is simply inappropriate. PTA is a unique site among all the other installations where DU has been used and remains at the installation. It is an area composed of recently deposited basaltic material derived from oceanic materials. It has a climate predominantly influenced by its subtropical location and island setting. The latter greatly influences the formation of any soil from bedrock with very specific chemical and physical compositions. No other location has those attributes that are found at PTA.

What is presented by the Army in its response creates an even more irrational and confusing scenario than what existed previously.

Regarding composite sampling, that which is composed of collected increments, I refer to Table 2.1 in the Oak Ridge Institute for Science and Education (ORISE) Report (ML13101A090) provided by the NRC in what appears to be general guidance for the Army to consider. Table 2.1 summarizes when composite sampling is advantageous and lists the disadvantages that must also be considered and addressed in the planning and data life cycle. For convenience, I replicate it here.

Table 2.1. Composite Sampling Overview

Advantages	Disadvantages
1. Reduces analytical costs.	1. Should not be used for establishing surrogate ratios
2. Provides a better estimate of mean concentration in the study area.	2. Information is lost on the individual sample increments that make up a composite. This loss of information is a concern when testing to determine if a ROC exceeds a threshold,e.g., a $DCGL_{EMC}$ over a specific area because of possible dilution to one or more increments with elevated activity concentrations by the other composite increments.

Table 2.1 (continued)

3. Identifying units that have the highest contaminant levels.	3. Cannot be used when action levels ($DCGL_{WS}$) are near analytical detection limits or the natural natural background concentration levels.
	4. For non-homogenous contaminant distributions temporal or spatial variability information is lost.
4. With an appropriately adjusted contaminant benchmark/investigation level, composite sampling can increase the ability to detect hot spots by increasing the number of locations sampled.	5. Cannot be used when integrity of individual sample values change, such as loss of volatile contaminants, due to the physical compositing mechanism.

Acronyms: ROC radionuclides of concern
$DCGL_W$ average derived concentration guideline level
DCG_{EMC} elevated measurement comparison derived concentration guideline level

Table 2.1 continued

Uses and Considerations for Applying Composite Sampling

1. Useful when the size of the pattern or feature of interest, such as hot spots, is smaller than the spacing between the statistically required random sampling locations.
2. User must account for potential introduction of large additional errors due to heterogeneous nature of the contaminant in the matrix, or the matrix itself.
3. Aliquots used to form the composite must be of equivalent weight/volume and the individual aliquots and the composite itself must be well homogenized.
4. Must account for the dilution factor when evaluating the result against a threshold, most commonly a hotspot or legal action threshold. Necessitates a modified investigation level (MIL).
5. In most cases, the user must maintain the ability for re-testing of individual samples (increments) making up the composite to retrieve potentially lost information.

From Table 2.1, the composite procedure used by the Army is present, without exception, in all 5 statements of disadvantages in the Overview. That makes it a completely improper procedure. Keep in mind that this observation is based on totally independent guidelines prepared in 2012 by ORISE.

Here is the breakdown:

Disadvantage 1. The issue concerns uranium that has been depleted in 235U. The analytical procedure uses the ratio of 238U/234U. Because 235U is not measured, 234U becomes a surrogate for 235U. Therefore composite sampling is not a proper analytical technique at PTA.

Disadvantage 2. Information is lost on the individual sample increments that make up a composite. This is a clear statement that can stand alone.

Disadvantage 3. As any action level at PTA will be near natural background concentration levels, compositing is improper and should not be used at PTA.

Disadvantage 4. Any DU contamination is likely to be of particulate nature and therefore heterogeneous in the sample matrix. Consequently, composite sampling at PTA is not warranted.

Disadvantage 5. The sampling procedure described by the Army causes the loss of sample integrity. This is a major flaw in the current composite sampling process. Therefore, composite sampling is improper at PTA. I will amplify this point later after continuing discussion of Uses and Considerations for Composite Sampling in Table 2.1.

<u>Comment on Uses and Considerations for Applying Composite Sampling Table 2.1</u>

Issue 1. As only one sample site is being used, issue 1 does not apply.

Issue 2. As with disadvantage number 4 above, the radionuclides of interest are likely to be heterogeneous in the sample matrix. This issue must be addressed and modification of the sampling procedure will be required.

Issue 3. The sample increments must be equivalent in weight/volume and must be well homogenized. The procedure used by the Army does not provide for such sample homogeneity. There appears to be nothing in the current procedure to address equivalent sample weight/volume.

Issue 4. The heterogeneity of the sample ROC will likely provide dilution effects for analysis and minimize threshold concentrations. This issue has not been addressed by the Army or the analytical laboratory. What must be considered an arbitrary selection of 238U/234U of 3 must be discarded and replaced with any ratio value greater than 1 (with reasonable deviation).

Issue 5. This issue requires that the increment sample collections be maintained so further testing can be accomplished. This is not being done in the Army procedure and the composite samples may be discarded after 6 months by the laboratory. The Army states “The samplers do not send individual sub-samples to the laboratory for analysis.”

In fact, they do not even maintain the integrity of the sub-samples as they are mixed in the field.

There is no question here that the composite sampling procedure utilized by the Army is woefully inadequate and improper. Every disadvantage listed in the ORISE Report is contained in and negatively impacts the current Army sampling plan. NRC must insist on proper procedures to address the issue of DU migration away from the RCAs.

The Army sampling procedure causes loss of sample integrity. The Army procedure for PTA is mostly described in its RESPONSE in the section labeled 3.2 SEDIMENT SAMPLING. Some details are in the presentation preceding this section. Often, reference is made to Annex 19 (ML16265A233) in the ERMP. A review of Annex 19 will show that very little actual information is supplied for sampling. This issue was provided previously to NRC and Annex 19 was shown to be not helpful in understanding the full details of the sampling or analytical procedure.

That annex addresses Quality Assurances for the Project Plan through a series of worksheets. The information provided on the data collection sheet for QA/QC is not fully explained and appears to be highly inadequate. For example, more information on weather conditions especially precipitation events and the time before sampling, the amount of moisture in the sample, the general size distribution of the sample and increments (it should really be measured and can be done so by the laboratory) as this is important to provide characteristics of the chemical and physical composition of the sample. The information collected should be greatly expanded. Basically, the field aliquots should all individually be sent to the laboratory for individual analysis.

Overall, Annex 19 is quite lacking in specifics. For example Worksheet 18, ‘Sampling Locations and Methods’ refers the reader to worksheet 21, ‘Field SOPs’ (standard operating procedure), but every entry on worksheet 21 is listed as TBD (to be determined). It would be of value to have the TBDs filled in.

The sample sent to the laboratory has already combined subsamples in the field and the samplers do not send subsamples to the laboratory. This is in absolute contrast to the sampling criteria in the quoted ORISE reference. A worksheet (number 28) in Annex 19 lists the analytical procedure for sediment samples. One entry addresses running every 1 in 10 samples as a duplicate, a truly difficult feat when only one composite sample is provided. The Army plan represents a waste of time and money. There are so many improper procedures and unknown variables in the methodology that nothing of value can be derived from any approach that employs these methods. Data comparison from one collection time to another will be impossible.

In the Army response, a brief overview of procedures is given. Ten increments of sample are collected from ten sites within a one-meter radius scraping off the surface material but not exceeding 3 centimeters in depth. It states that a sufficient amount of sample will be collected for QA/QC. The sediment is then placed into a disposable tray or plastic bag, large pebbles and organic matter are removed. Excess water is then

removed from the sediment and the sample is mixed in the bag or on the tray. Then the appropriate sample container (provided by the analytical lab) is filled without any explanation of the transfer procedure to the 8 ounce container.

The problem is how the sample is handled when collected and processed. The sample is mixed with no explanation of the mixing method, in the bag or on the tray, and then transferred to an 8 ounce jar provided by the analytical lab. No specific information on breakdown of sample size is given, no guarantee that equal quantities of sample from the ten increments are obtained, and no explanation of why water might be present in the sample when the ERMP suggests that samples will only be collected during dry periods.

The Army contends that "…compositing will ensure the collection of samples comprised of varying particle sizes…" To the contrary, its procedure is guaranteed to corrupt that very quality of sample integrity. Placing the sediment material in a bag or on a tray will cause small particles to adhere to the tray or bag surface. It cannot be completely transferred from the bag or tray to the sample jar provided by the laboratory. Small particles will be left behind as they will cling to the sample mixing material. Any action of pouring off any water present will also remove small suspended particles. The Army says some time may be required for settling of particles to occur, but in actuality, that time could be hours if not days for nanometer size particles. (https://www.hindawi.com/journals/jnm/2016/7061838/).

In other words, sample integrity is lost. This introduces a bias in the sample by removing the small particles that may well be the size group that contains the DU. Do the readers here want proof? Conduct your own experiment. When you get home from work today, take a plastic storage bag and put some confectioners' (powdered) sugar into it. Then dump it out. Is there any material left in the bag? Particle size of powdered sugar ranges from 10 to 40 micrometers. Smaller particles, likely those representative of DU and its oxides, could even be more tenacious in adhering to the surface of some intermediate container.

Further, mixing the sample of unknown quantity and placing some or all of it in the jar requires a very thorough and careful mixing process. None has been described. Mixing in the plastic bag by shaking or squeezing, or stirring material on a tray would not be acceptable practice.

The sample collected from 10 sites must be at least of sufficient quantity for the laboratory to analyze. The laboratory presumably is comfortable with the eight ounce jar it provides. Given the sediment material is likely composed of basaltic fragments, the bedrock material from which soil matrix can be derived, then a probable bulk density of 1.6 gm/cm^3 is reasonable. Eight ounces (U.S. liquid measure) would give a volume of approximately 237 ml or about 380 gm of sample if the bottle were filled. This points out that on average the incremental samples would be less than 25 ml or about 38 grams each, using the bulk density of 1.6 gm/cm^3. This quantity is far short of what could reasonably be called a representative sample, especially when there will be a particle

size differential within the sample. This is exacerbated when the Army states a procedure that “The samplers do not have to fill each jar because the laboratory is not concerned if a smaller-size aliquot is all that is available.”

Overall, this continues to demonstrate a very inadequate reply by the Army to the NRC question and imparts conflicting methodologies.

When the Army proposed to collect air samples, it claimed that sediment samples did not exist (ML1206A506). NRC also stated that soil samples did not exist. When the Army decided not to collect air samples, it proposed to collect sediment samples at a site that was miles away from the RCAs. Such a location has doubtful connectivity to the RCAs because of inter-situated lava flows. This conundrum has never been explained but the Army is now collecting sediment samples.

The issue the NRC raised concerning the dilution of sample by collection and combination consolidation of sample from different sites different sites has not been addressed in this response.

The sampling plan will never address the objective it is purported to achieve and the data will be so suspect to uncertainties that nothing of merit will ever be recoverable. The problem is created in that there does not appear to be consistency in the sample collection process or adherence to standard protocols. That makes it much more difficult if not impossible to interpret and compare the data.

RESPONSE TO SUGGESTION THAT SITE SPECIFIC ERMP 3.2 BE AMENDED

The Army asks in its response of December 15, 2017 to the NRC to consider amending the ERMP associated with license SUC-1593. Specifically, the request is for a modification to Section 3.2, Sediment Sampling. Such issues should go through the amendment process. If the NRC is considering making the suggestion part of the license, an enhanced alternative is presented here that is superior and must be implemented in place of the one presented by the Army.

Sampling shall consist of collecting 10 discrete aliquots of sediment material each to be placed separately in the sample containers provided by the analytical laboratory and each to be analyzed individually. The analytical laboratory shall obtain from the individual samples provided, and following standard protocols, an aliquot of each that is needed for analysis, that portion being representative of the sample in its entirety including particle size distribution and suspected physical and chemical composition variations of possible different sediment types. The sampling location shall be situated at a location in a stream bed that has been visually confirmed to have flow and deposition from the radiation controlled areas after typical rainfall events at Pohakuloa Training Area. The samples shall be collected from a one-meter radius of the identified sample location each providing at least the minimum required sample quantity for analysis as requested by the laboratory. Each sample collection submitted shall include

at least one aliquot sample in ten that is a possible background sample but not identified to the laboratory as such and one aliquot sample in ten that is a same aliquot site duplicate provided by the sampler but not identified as such to the laboratory.

The major advantage of this slightly enhanced alternative is that it addresses the dilution question and eliminates sample loss of critical particle sizes while still providing information that a composite sample would. In this case, the composite is of the individual analyses rather than a composite of the sample materials. Sampling after a rainfall event would be required and the isotopic ratio to be used for determining the presence of DU should still be modified but can be done so on the basis of the acquired analytical results. There should be a requirement that some samples should be analyzed by ICP; they could be the highest spectrometric isotope ratio of 238U to 235U (or surrogate 234U) and an occasional random sample.

Of course, some of the following 9 items in section 3.2 would have to be changed as the responsibility of sample changes, such as removal of organics and pebbles would fall under the laboratory's purview.

By providing the Oak Ridge reference as part of the basis for the question concerning dilution, the NRC clearly recognizes the importance of this issue and the impact it can have on the entire ERMP. Without those enhancements and attention to detail, the ERMP sampling program is useless. This is the time and opportunity for the NRC to make necessary changes and adjustments in the sampling plan and analytical procedure for Pohakuloa Training Area to make them both meaningful for the objectives of the ERMP.