An Approach for Estimating the Total Effective Dose at Atmospheric Nuclear Weapons Test Sites - 9235

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ABSTRACT

Decisions about the response actions to be taken at several sites containing surface soils that were contaminated from historical atmospheric nuclear tests at the Nevada Test Site must be made. The regulatory framework for these decisions is described in a Federal Facility Agreement and Consent Order for the test site. Through agreement with the stakeholders, a decision criterion was established as a Total Effective Dose (TED) of 0.25 milli-sieverts per year (mSv/yr) to a receptor under three established exposure scenarios. Several historical sources of information regarding the distribution of radioactive contamination (which included actinides) and the areal concentration of that contamination are available, but none were found to be of sufficient quality for decision-making. In addition, traditional dose estimates using soil sampling and the RESRAD computer code would likely overestimate the internal dose due to the presence of significant amounts of trinity glass with large particle sizes. One site was selected for investigation and decision-making. The approach evaluated internal dose separately from the external dose and generated data that could be used to allow for improved confidence in the use of historical sources of information.

INTRODUCTION

Between January, 1951 and July, 1962, atmospheric nuclear tests were conducted at the Nevada Test Site [1]. More than 100 sites were affected by aboveground testing [2]. In 1996 the U.S. Department of Energy (DOE) and other parties entered into an agreement with the Nevada Division of Environmental Protection (NDEP) to, in part, "Identify[ing] sites of potential historic contamination and implement[ing] proposed corrective actions based upon public health and environmental considerations…". The agreement took the form of a Federal Facilities Agreement and Consent Order (FFACO)[3]. The agreement established a framework for decision-making for sites that warrant environmental investigations at the Nevada Test Site (NTS).

Efforts to evaluate sites that were primarily impacted by radioactive surface soil contamination accelerated in 2006 when stakeholders met and approved a decision criterion that was based upon dose, in

mSv/yr, to a receptor under three approved exposure scenarios. This focus on an exposure scenario, rather than upon comparisons of individual soil sample results to a set of radionuclide-specific Preliminary Action Levels (PALs), opened the door for more innovative methods to assess the sites.

A project team was assembled to develop the new approach to site evaluation, and a site was selected to demonstrate a "proof of concept" for the approach. Existing historical information, in the form of an aerial radiological survey and in-situ gamma spectroscopy measurements, was used to guide the initial site investigation. Data Quality Objectives (DQOs) were specified which would generate data of sufficient quality to make decisions about the site and which would justify an increased reliance on the historical information at future sites. This would facilitate the future investigations by reducing the number of soil samples needed to make a decision at a site. The team elected to determine the internal dose contribution (i.e., Committed Effective Dose) separately from the external dose contribution (i.e., Effective Dose) and to sum the results to determine the Total Effective Dose (TED) at the site. This approach would minimize the errors in the dose estimates due to the presence of large amounts of trinitite, and obviate the technical issues surrounding the need to size-reduce the trinitite during sample processing.

MATERIALS AND METHODS

Exposure Scenarios

A decision criterion that is expressed in terms of dose per year carries little meaning without stating the assumptions to be used in the dose estimates. The set of assumptions are referred to as an exposure scenario. The scenario describes the activities of the dose receptor (occupancy times, exposure duration, land-use, etc…) and the input parameters to be used in dose modeling calculations (in this case, RESRAD version 6.4).

At the NTS, three exposure scenarios had been agreed upon with stakeholders [4]; "Industrial Area", "Remote Work Area", and "Occasional Use Area". Since the NTS is situated within a desert, and based upon the expected future land-use for the NTS, all three scenarios excluded the plant ingestion, meat ingestion, milk ingestion, aquatic foods, drinking water, and radon exposure pathways. The external gamma, inhalation (without Radon), and soil ingestion pathways remained.

The most restrictive of these was the "Industrial Area" (IA) scenario. In this scenario, it is assumed that an industrial worker will be at the site for an entire career (225 days per year, 10 hours per day, for 25 years). For this project, the area of the contaminated zone was set to 100 square-meters (m^2) and the thickness of the contaminated zone was set at 0.05 meters (m) [5] [6].

Nature of Contaminants

In an atmospheric nuclear weapons test, soils and surrounding structures may become radioactively contaminated through several routes. These routes can be simplified as neutron activation, fall-out of fission fragments, and fallout of un-fissioned nuclear fuel. Following fifty years of radioactive decay, radioactive contaminants from neutron activation consist primarily of europium (Eu)-152 and Eu-154 from the activation of trace amount of Eu in the soil. Cobalt (Co)-60 may also be present due to the activation of Co-59 in equipment and metal structures. Cesium (Cs)-137 and strontium (Sr)-90, which are fission fragments, may still be present at modest concentrations over large areas surrounding ground zero. Uranium (U)-235, plutonium (Pu)-238, Pu-239/240, and the associated Americium (Am)-241, which are constituents of the atomic device, may be present in notable concentrations over small areas near ground zero (GZ).

The distribution of contaminants around GZ is governed by the atmospheric conditions at the time of detonation, and by the relative boiling points of the individual chemical elements generated or contained within the fireball. For example, iodines (which have a low boiling point), may be found many miles from GZ. Plutoniums (which have a high boiling point), will quickly condense and fall-out very near the GZ. Depending on the wind direction and speed, the concentrations of fission fragments could vary significantly around GZ.

Trinitite (i.e., trinity glass) is the name given to desert soils that were mixed with radioactive debris from an atomic device and melted to form a green glass-like substance [7]. The radionuclides contained in trinitite may include fission products; activation products from neutron reactions with the nuclear fuel, the materials used in construction of the device, and the local GZ soil; the remnants of un-fissioned nuclear fuel; and radionuclides of natural origin in the soil. Radionuclides are expected to remain within the trinitite. The deposition of trinities around GZ is highly variable, and obvious "pools" of the material can be seen. Individual pieces of trinitite are generally quite large. Refer to figure 1.

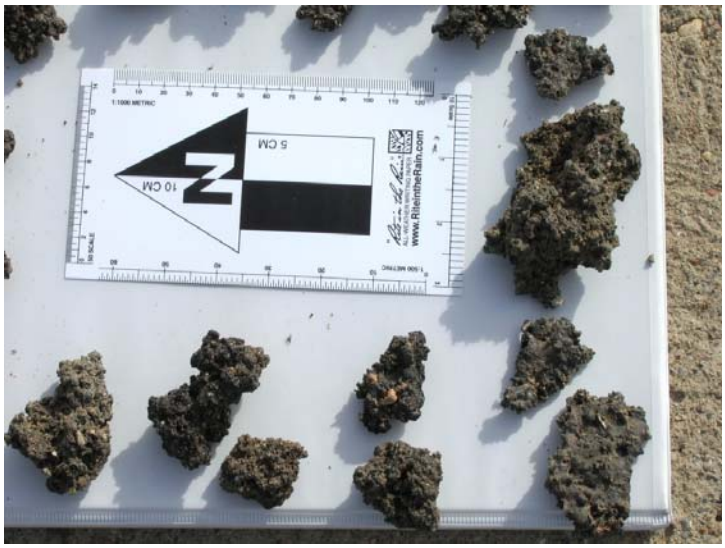

Figure 1 - Trinitite from the T-4 Site, NTS

Trinitite serves to "contain" a portion of the radioactive contamination at a testing site. The glass-like nature and large particle size effectively removes the contaminants from the inhalation and ingestion pathways, but not from the external gamma exposure pathway. A fraction of the europium radionuclides and a modest fraction of the fission fragments and un-fissioned fuel may also be found in surface soils (independent of the trinitite).

Site Selection

The project team selected the "T-4" testing site to demonstrate the new site evaluation approach. T-4 is the location of four near-ground surface atmospheric nuclear tests, conducted on towers, and utilizing the same GZ. The tests are summarized in table 1.

Table 1 - Summary of Tests at T-4

Test Name	Date	Yield (approximate)
Fox	5/25/52	11 kilotons
Nancy	3/24/53	24 kilotons
Apple 1	3/29/55	14 kilotons
Kepler	7/24/57	10 kilotons

Several factors were used in the selection of the T-4 site. These included:

- An atmospheric test with no crater (safety & access concerns)
- A site that remains relatively undisturbed (movement or removal of surface soils)
- A site that is near major roads and populated areas of the NTS, to facilitate mobilization and on-going work during the "testing" phase of the new approach
- A site that will not be unduly impacted by other NTS activities, such as military exercises
- A site that has not been impacted by radiological contaminants from other nuclear tests (e.g., plume overlap)

Historical Data - Radiological Flyover Surveys

A team from the Remote Sensing Laboratory conducted an aerial radiological survey of the NTS during August and September 1994. The survey measured the terrestrial gamma radiation to determine the levels of natural and man-made radiation and to examine the distribution of the radiation over the test site areas. The survey built upon previous similar surveys conducted from 1962 through 1993 [8]. The survey converted gross count-rate data into units of exposure rate (in micro-Roentgen per hour) and extrapolated the result to 1 m above ground level. The survey report provided contour maps of the exposure rate. Refer to figure 4 for an example from the T-4 site. The gamma ray spectrum from Am-241 was also mapped. Am-241 is present as a decay product of Pu-241, a contaminant seen in Pu-239, and is used as an indicator of the presence of Pu-239. Refer to figure 4 as well for an example from the T-4 site.

Historical Data - The RIDP Program

The Radionuclide Inventory and Distribution Program (RIDP) was a project to determine both the distribution and the total inventory of man-made radionuclides in the surface soils of the NTS [9]. The program used *in-situ* gamma spectroscopy for detailed information about specific areas, such as the GZs of atmospheric test sites. Some limited soil sampling was performed in order to estimate activities of non-gamma emitting radionuclides (e.g., Strontium-90). Data were expressed in units of nano-curies per square-meter (nCi/m^2) and the used to estimate the total radionuclide inventory at each site that was examined. The program concluded that "*in-situ* spectrometry gives better estimates of radionuclide activity than soil sampling, which tends to miss highly radioactive particles…"[10].

Project Soil Sampling Approach

The radiological flyover survey and the RIDP were each initiated for their own unique purposes, neither of which was the evaluation of total effective dose at a testing site. Because of this, it is not surprising that the data was insufficient, as a whole, to meet the current Data Quality Objectives for the T-4 site. Both the flyover survey and the RIDP still served as valuable resources that could be used to guide the investigation. The project team determined that a well-designed program of soil sampling and dose-rate measurements could be used to reach a decision at the T-4 site, and that such data could further be used to develop correlations between the "as-found" soil conditions and the RIDP estimates. This is particularly important because the RIDP collected very few confirmatory soil samples and because assumptions used to convert RIDP data from units of nCi/m^2 to picocuries per gram (pCi/g) needed to be validated. Some amount of correlation might also be established for the radiological flyover survey.

Soil sampling investigation areas (i.e., "sample plots") were established as 100 m^2 areas. Sample plots were positioned at locations that represented each of the major contour lines from the radiological flyover survey and the plots were centered, where available, over a RIDP *in-situ* measurement point.

Soil sample aliquots within each sample plot were collected over a depth of 5 centimeters (cm) using a trowel and a “stove pipe” type of circular template. The diameter of the stove pipe was selected such that nine aliquots, when composited into a single soil sample, would nearly fill a one-gallon paint can. The nine aliquots were collected using a random-start, systematic pattern that began near the southwest corner of each plot, covering the entire plot.

Each RIDP *in-situ* measurement determined the average activity of radionuclides over the RIDP detector’s field of view. Composite soil sampling was selected to mirror this averaging and to temper the potential sampling error that can occur due to the presence of actinide particles with high specific activities.

Four composite samples were collected from each sample plot so that the variability in each plot could be evaluated.

Internal dose at the T-4 site was determined via soil sampling and use of the RESRAD code, as will be discussed below. Soil aliquots were sieved through a #4 mesh screen prior to compositing, to remove the vast bulk of the trinitite from the sample prior to analysis. Since trinitite exists as very large non-respire-able and non-ingestible particles, internal dose would be over estimated if trinitite were included. This approach was reasonable since experience at the NTS indicates that in excess of 90% of the anthropic radioactivity in soil exists in the fine fraction of soil (exclusive of that in trinity glass)[9].

Soil samples were sent to a commercial analytical laboratory. After processing in the laboratory, each sample was first subjected to high resolution gamma spectroscopy. Aliquots were then removed from that gamma spectroscopy sample and analyzed for Sr-90, isotopic Pu, isotopic uranium (U), and isotopic thorium (Th). Actinide contamination from atmospheric weapons testing consists of small particles of high specific activity. Experience at the NTS has shown that analytical results for plutonium can vary by an order of magnitude or more for individual 1 gram aliquots taken from the same soil sample. One reference [11] suggested that 100 gram aliquots for isotopic Pu were most appropriate, but this was found to not be feasible for a commercial analytical laboratory. For this project, the team was able to specify 5 gram sample aliquots.

Internal Dose

Site evaluation and comparison to decision criteria can be performed via soil sampling and dose estimation via the RESRAD computer code. The code is capable of evaluating both the potential internal and potential external dose. Trinitite contains Cs-137, Eu-152 and Eu-154 (among other radionuclides) and contributes to external dose in a significant way. The inclusion of trinitite in the soil samples would be appropriate for estimates of the external dose. As was previously mentioned, it is not reasonable to expect that trinitite would be inhaled or ingested. The inclusion of trinitite in the soil samples would significantly over estimate the internal dose and would not be appropriate. For this reason, the project team elected to determine the internal dose and the external dose for sample plots via separate means and then sum the result to yield a total effective dose.

The RESRAD code was run for the Industrial Area exposure scenario, with the assumptions and input parameters as described in reference 4 [4], but with the external gamma pathway suppressed. Each radionuclide potential contaminant of concern for an atmospheric weapons testing site was input at a base activity-concentration of 0.037 becquerels/g (Bq/g). The output was a set of derived concentration guidelines (DCGs), in units of (mSv/yr) per (Bq/g). A soil sample with an individual radionuclide result that is equal to its DCG would contribute 0.25 mSv/IA-yr internal dose to a receptor under the industrial

area exposure scenario. The industrial area scenario was selected since it was the most conservative of the three.

External Dose

External dose was measured directly through the use of thermo-luminescent dosimeters (TLDs). Thirty Panasonic UD-802AT2 TLDs were obtained from the Environmental Technical Services group at the NTS and placed at twenty-six locations within the T-4 site. The locations were selected to correspond with RIDP *in-situ* measurement points and the center of soil sample plots. TLDs were hung at a height of 1 m above the ground surface in a manner that duplicated the NTS environmental monitoring program. TLDs were emplaced early in the project schedule so that they could be exposed over a period that encompassed the entire 2,250 hours of the Industrial Area exposure scenario.

External dose was also measured through the use of a Bicron model micro-rem per hour (micro-rem/hr) meter. Dose rate measurements were collected at a height of 1 meter above the ground at the center of each soil sample plot and at each TLD location. In addition, the entire T-4 site was gridded-off, with a spacing of 50 meters, and a dose rate measurement was taken at a height of 1 meter at each grid intersection.

RESULTS AND DISCUSSION

Internal Dose

Seventy-eight soil samples were collected an analyzed for the T-4 site. This encompassed twenty planned sample plots, investigation of three Am-241 plumes (refer to figure 4), investigation by GPS-assisted gamma walkover survey of two areas of elevated activity found in a rill-eroded area in the southeast portion of the site (refer to figure 2), and field quality control samples.

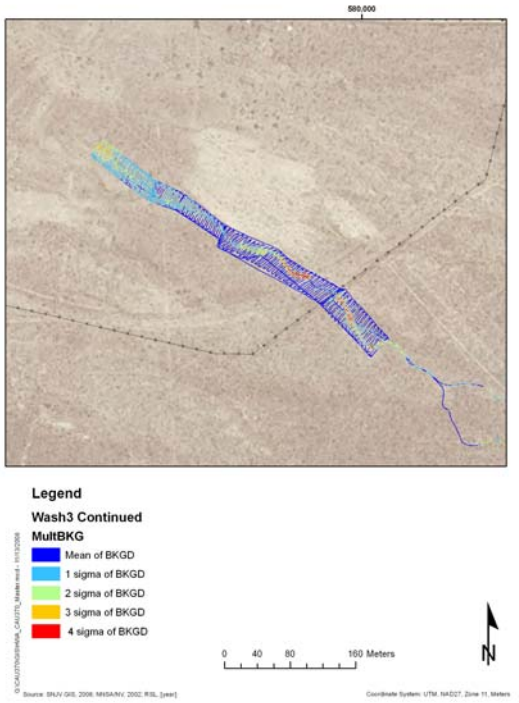

Figure 2 - Investigation of Southern Wash at T-4

Internal doses, in units of mSv/IA-yr, were calculated for each of the 78 soil samples. The maximum internal dose was 0.096 mSv/yr (9.6 mrem/yr). The mean was 0.014 mSv/yr (1.4 mrem/yr) with a standard deviation of 0.018 mSv/yr (1.8 mrem/yr). The mean plus one standard deviation is only 13% of the decision criterion.

External Dose

TLDs were emplaced for a total of 2,544 hours or 113% of the 2,250 hours described in the industrial area exposure scenario. The TLD results were normalized to 2,250 hours and then a background of 0.313 mSv/IA-yr (31.3 mrem/IA-yr) was subtracted. The background was taken from the 2006 Nevada Test Site Environmental Report for an area that is similar to the T-4 site [12].

The maximum net external dose was 2.18 mSv/IA-yr (218 mrem/IA-yr). The mean was 0.747 mSv/IA-yr (74.7 mrem/IA-yr) with a standard deviation of 0.674 mSv/IA-yr (67.4 mrem/IA-yr). The mean plus one standard deviation is 568% of the decision criterion. Seventeen of the twenty-six locations exceed the decision criterion.

TLD results could be paired with Bicron micro-rem/hr instrument measurements at twenty-six locations at the T-4 site. The Bicron reading was converted to units of mSv/hr, multiplied by the 2,250 hours of the industrial area exposure scenario, and then a background of 0.311 mSv was subtracted. This yielded dose estimates in terms of mSv/IA-yr which could be compared directly to the TLD results. The correlation was excellent, with a R^2 value of 92.0%. It is feasible that dose rate measurements could be used to evaluate the external dose directly, without the need for the TLDs.

Bicron micro-rem/hr measurements were taken at 319 grid survey points. By inspection, there appear to be significant differences between the Bicron dose rate measurements and the corresponding dose rate as calculated and projected by the aerial radiological survey. The aerial radiological survey appears to be an effective tool for survey planning, but external dose should be determined via TLD or Bicron measurement. Refer to figure 3 (values in figure 3 are listed in units of milli-rem/hr to allow direct comparison to the units provided in the aerial radiological survey results).

Total Effective Dose

The total effective dose (TED) in mSv/IA-yr was calculated for each of the fifteen planned sample plots and the five areas of investigation by summing the results for the internal dose with that of the external dose. Thirteen of the fifteen planned sample plots exceeded the decision criterion. Two investigation areas exceeded the decision criterion. Results are presented in figure 4.

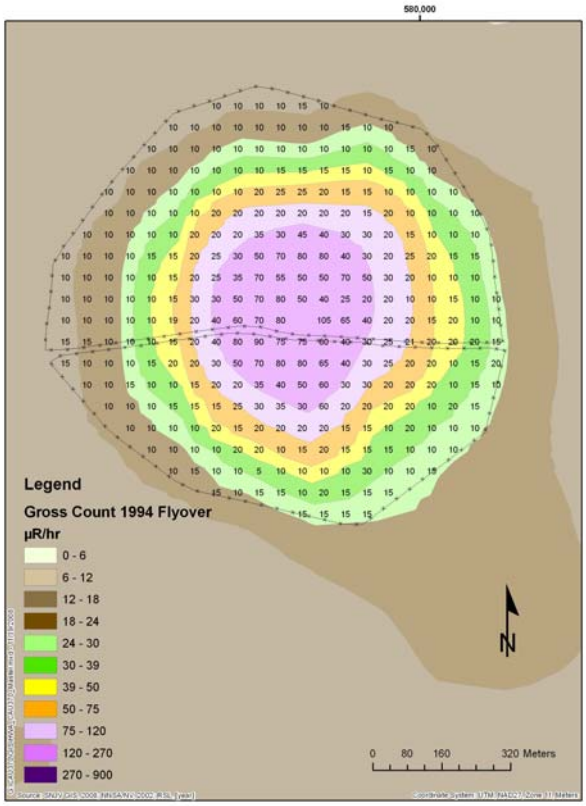

Figure 3 – Aerial Radiological Survey, Overlain By Grid Survey Results (in micro-rem/hr)

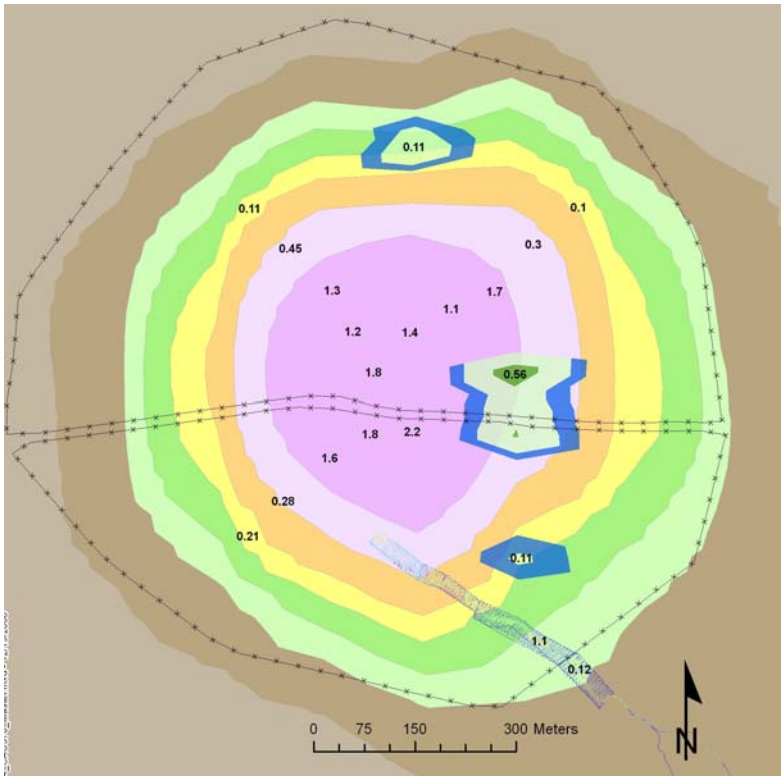

Figure 4 - Total Effective Dose (mSv/IA-yr) at T-4

Correlations to RIDP

Eight of the fifteen planned soil sample plots were selected at locations that corresponded with a RIDP *in-situ* gamma spectroscopy measurement location. The remaining seven soil sample plots were selected to gain additional information about one or more isopleths from the 1994 radiological flyover survey.

At the eight soil sample plots, isotopic-specific soil sample results were compared to the corresponding RIDP *in-situ* measurement to evaluate the relationship. Isotopic-specific results were available for four (in some cases five) composite soil samples at each of the sampling plots. The results were averaged for each sample plot. The 1980's isotopic-specific RIDP data was decayed to October 1, 2008 and converted from units of nCi/m^2 to pCi/g. The "hard to detect" radionuclides (e.g., Pu-238, Pu-239, Sr-90) were inferred from indicator radionuclides (e.g., Am-241, Cs-137) based upon the isotopic ratios as cited in the original RIDP report. Soil sample results were then compared to the RIDP results through the use of the MiniTab version 15 statistical software package. The coefficient of determination (R^2) values are presented in table 2.

Table 2 - Coefficient Of Determination (R^2) Values

Radionuclide	R^2
Am-241	91.4%
Co-60	71.6%
Cs-137	95.5%
Eu-152	6.2%
Eu-154	18.5%
Pu-238	96.3%
Pu-239/240	92.7%
Sr-90	23.0%
U-235	Not Detected

In general, there was a good correlation between the decayed and inferred RIDP *in-situ* isotopic sample results and the average of the project soil samples at the eight sample plots.

The R^2 value for Co-60 is poor. The concentration of Co-60 in the soil samples was quite low and near the analytical detection limit.

The R^2 value for both Eu-152 and Eu-154 is very poor, indicating difficulty with either the RIDP measurement or the soil sampling processes. The most likely source of this discrepancy is the soil sampling approach, which collected soil over a depth of 5 cm. The RIDP reports indicated that isotopes of europium could be present at depths of up to 60 cm or more due to neutron activation of the soil, particularly near GZ. Since europium was likely to contribute little to internal dose, especially at depths > 5 cm, the project team made a decision to limit soil sampling depth to 5 cm to more accurately reflect the true activity-concentrations of the actinides (which reside in the top 5 cm of soil). A second factor in discrepancy that can be attributed to the soil sampling approach was the removal of trinitite from the samples in the field. Trinitite may contain modest amounts of europium. This europium in trinitite would be included in the RIDP *in-situ* measurements, but physically excluded from the soil sample results.

The R^2 value for Sr-90 is also very poor. Sr-90 was not directly measured by the RIDP, with the exception of a few soil samples that were collected to verify contaminant ratios and to determine the depth of contamination. In many cases, the soil samples were distant from the GZ. The Sr-90 values provided from the RIDP data are inferred from the Cs-137 values using a Sr-90 to Cs-137 ratio as determined from those limited RIDP soil samples. The R^2 value can be improved to 77.9% by using an

average Sr-90 to Cs-137 ratio of 0.087 taken from the T-4 project soil samples, and by removing the ratio from sample plot "A" from the average. Sample plot "A" was at the GZ and its ratio was significantly different from the other samples.

For the T-4 site, it appears to be reasonable to utilize the decayed and inferred RIDP in-situ gamma spectroscopy data to make estimates of the potential internal dose. The poor correlation between RIDP results and soil samples for Co-60, Eu-152, Eu-154, and Sr-90 is mitigated by the fact that these radionuclides contribute doses that are much less than $1/100^{th}$ of the decision criterion of 0.25 mSv/IA-yr at the T-4 site.

CONCLUSIONS

When trinitite is present at an atmospheric atomic device testing site, evaluation of the site using the traditional soil sampling with evaluation under the RESRAD code will overestimate the internal dose. Exclusion of the trinitite in the traditional approach will underestimate the external dose. A modified approach where internal dose is determined via soil sampling and external dose is determined via TLD appears to be reasonable, efficient, and effective.

The soil sampling protocols selected for the T-4 site evaluation yielded high quality data with relatively low variability among samples in a sample plot. These protocols require further evaluation at sites where actinides may be present at significantly higher activity concentrations.

The evaluation of external dose via TLDs was found to be cost-effective, but significant lead-time may be necessary to obtain the desired exposure time of 2,250 hours. The use of a micro-rem/hr meter might serve as a suitable substitute, yielding rapid results and decisions.

The RIDP and aerial radiological survey serve as excellent planning tools. With additional data collection, the decayed and inferred RIDP data might be combined with dose rate measurements at future sites to make preliminary dose estimates in terms of TED. This could then be verified with a limited number of soil samples, which would significantly reduce the time and expense of site evaluation.

The TED at the T-4 site was dominated by the external dose. This may not be the case at other atmospheric testing sites, therefore the approach used at T-4 should be further evaluated.

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