Development of Radiological Screening Levels and Associated Gamma Survey Methodologies for Radiological Characterization at US DOE Office of Legacy Management Defense-Related Uranium Mine Sites

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Overview: US DOE Defense Related Uranium Mining (DRUM) Sites on Federal Land

- DOE - LM is responsible for validating and verifying (screening level characterization) approximately 2400 Defense Related Uranium Mining (DRUM) sites located on Federal land (many are small – but some are not).
- Uranium ore purchased by the Atomic Energy Commission prior to 1970 for defense programs.
- Most are in remote areas of SW US (CO, WY, UT, AZ, NM).
- LM has developed radiological screening criteria and wide area gamma survey protocols to assist in prioritizing this very large # of sites (along with physical hazards, human accessibility, other factors).
Overview - continued

- For sites on Federal land (primarily Bureau of Land Management and US Forest Service) - public access for recreational activities (camping, hiking, fishing, etc.) is typically limited to 2 weeks per year

- Public exposure limit of 1 mSv (100 mrem) / yr. applied to the 2 week / yr. exposure period to define a “high risk” site if exceeded

- BLM has concurred with exposure scenario and general approach

- Most sites are in semi-arid environments with sparse vegetation; game animals are migratory and recreationists must bring own water and food
Gamma exposure rates are primary screening criteria (uSv [uR] per hr.)*

Other federal agencies (i.e., U.S. Nuclear Regulatory Commission, U.S. Environmental Protection Agency, U.S. Army Corps of Engineers) use $^{226}$Ra concentrations in soil as basic remediation criteria for legacy natural uranium (U) sites.

Relationship of uR/hr. values to Bq (pCi) / gram $^{226}$Ra is secondary screening criteria.

* Although International units and nomenclature are used throughout this presentation, it must be recognized that US regulations and associated radiological quantities are expressed in common US units and the “screening criteria” had to be expressed in comparative units.
Typical Environments – DRUM Sites (Legacy Uranium Mines)
Radiological Environment of DRUM Sites

- Contaminants of Concern = natural uranium ore in soil, waste rocks and spoils piles.
- Full radioactive equilibrium in both $^{238}\text{U}$ and $^{235}\text{U}$ decay series.
- Ratio of $^{235}\text{U}$ to $^{238}\text{U}$ activity is the natural abundance ratio (0.046).
- Contribution from natural thorium ($^{232}\text{Th}$) series is negligible - consistent with general $^{232}\text{Th}$ regional background.
Pathway Analysis for a Recreational User of DRUM Sites

<table>
<thead>
<tr>
<th>Pathway</th>
<th>Circumstances for Recreational Use of DRUM Sites</th>
</tr>
</thead>
<tbody>
<tr>
<td>External (gamma) from soil</td>
<td>U ore residues and waste rock, spoils piles, soil, adits and portals, etc.</td>
</tr>
<tr>
<td>Inhalation of soil dusts</td>
<td>Incidental and OHV rider</td>
</tr>
<tr>
<td>Inhalation of radon and progeny</td>
<td>Little time for progeny ingrowth; outdoors – assume user not in adits or portals (safety)*</td>
</tr>
<tr>
<td>Ingestion of soil</td>
<td>Incidental</td>
</tr>
<tr>
<td>Ingestion of water</td>
<td>Not applicable (NA) – must bring own water</td>
</tr>
<tr>
<td>Ingestion of food</td>
<td>NA – must bring own food; vegetation generally sparse, particularly on/around ore/spoil piles; migratory game animals (did not spend life in “contaminated zone”)</td>
</tr>
</tbody>
</table>

* Priority of reclamation activities is removal of physical hazards; many open portals and adits will have been sealed or otherwise remediated
Methodology For Developing Radiological Risk Screening Levels*

“Put” unit concentration of \( ^{238} \text{U} \) in soil - e.g., R Bq/gram; all other nuclides in decay series = 1 Bq/gram (equilibrium)*

\[
T = Ex + S_{inj} + I_d + I_{ohv} + I_{Rn}
\]

\( T \) = Total Effective Dose Equivalent (TEDE) for a 14 day recreational exposure scenario per Bq/gram of each of the \( ^{238} \text{U} \) plus \( ^{235} \text{U} \) series radionuclides in soil.

\( Ex \) = Dose Equivalent (DE) from external exposure from soil/rocks

\( S_{inj} \) = Committed Effective Dose Equivalent (CEDE) from incidental ingestion of soil

\( I_d \) = CEDE from inhalation of dusts

\( I_{ohv} \) = CEDE from inhalation of dusts during use of OHV

\( I_{Rn} \) = CEDE from inhalation of \( ^{222} \text{Rn} \) and progeny

Results

\[ \text{TEDE} = \text{Ex} + S_{\text{inj}} + I_d + I_{\text{ohv}} + I_{\text{Rn}} \]

\[ T = 168 + 2.8 + E^{-3} + 0.3 + 19.6 = 191 \text{ uSv (0.19 mSv) per Bq /g} \]
\[ \text{(0.68 mrem per pCi/g)} \]

\[ 1 \text{ mSv / 0.19 mSv per Bq/g} = 5.3 \text{ Bq/gram at 1 mSv per yr. (147 pCi \{e.g. }^{226}\text{Ra\} per gram at 100mrem per yr.)} \]

Contribution to the TEDE from external gamma exposure = \( 168 / 191 = 88\% \)

This relationship was used directly to establish average gamma exposure rate above background that ensures < annual limit: \( 0.88 \times 1 \text{ mSv during 2 weeks per year of recreational use} = 2.56 \text{ uSv (256 }\mu\text{rem) / hr. above bkg.} \)

Accordingly, Upper Screening Level = 256 \(\mu\text{rem / hr.} \)
Portable gamma-ray spectrometer based on advanced microprocessor technologies.

Detector equipped with thallium-activated sodium iodide (NaI) crystal - 1-liter volume (approximately a 4 by 4-inch).

Designed for portable / mobile radiological and geophysical surveys, including in rugged environments

For DRUM Program, instrument is used in backpack configuration worn by the RCT during performance of the gamma surveys

System also includes GPS link and a data logger connected to a portable electronic device, such as smartphone, tablet, or notebook.
PGIS – 2: Standard and Backpack Configurations
Historical Issues with NaI based Measurements In Uranium Series Photon (Gamma) Fields

- NaI detector based instruments have been the standard field gamma survey approach in the Uranium industry for 40+ years
- Rugged, easy to use with wide range of survey instruments
- However – two primary operational characteristics that often must be addressed when interpreting their output, based on data needs and objectives of survey program:
  - Energy Dependence
  - Tissue Equivalence
Energy Dependence

- Amplitude of electronic signal from photomultiplier tube is function of energy of the incident gamma ray.
- Particular relevance for measurement of uranium-series radionuclides since NaI traditionally calibrated to Cs137 - higher energy photon than average energy of photons in a uranium-series field.
- Accordingly, in uranium-series photon fields, NaI will typically “over respond” (provide results greater than actual values).
Energy Dependence of NaI Detectors

Energy Response for Ludlum Model 44-10

Response Normalized to $^{137}$Cs

Gamma Energy (keV)

Am-241, Co-57, Ba-133, Cs-137, Co-60

COURTESY LUDLUM INSTRUMENTS @ HTTPS://LUDLUMS.COM/PRODUCTS/ALL-PRODUCTS/PRODUCT/MODEL-44-10
Tissue Equivalence

- Unless specifically addressed in the physical design or associated software, traditional NaI-based detector systems measure **exposure rates in air** in roentgen (R) units (uR / hr.)
- However, human exposure limits and criteria usually expressed in tissue-equivalent (effective-dose) units - uSv / hr. or µrem / hr. (US regulations)
- Exposure rate in air 10-15% > dose in tissue (density air vs. human tissue)
- The LM DRUM program screening levels are derived to limit exposure to future human occupants of these sites – therefore gamma survey results should be comparable in tissue-equivalent (effective-dose) units.
Need for Instrument "Normalization" Study

- Unless same equipment and scanning geometry are used for all gamma surveys over time (e.g. project lifespans of many years) - necessary to normalize the data to a common basis of comparison.

- Normalization helps ensure results of future gamma scans, which might use different detectors / different detector geometries / measurement protocols can be compared in a meaningfully way against the initial survey screening-level values established for this project.
Normalization Approach

- Field studies performed to verify instrument performance vs. manufacturers published specifications of software’s ability to achieve energy independence and tissue equivalence.

- Accomplished via “side by side” comparisons to a primary standard (Reuter Stokes high-pressure ionization chamber - HPIC) and to another instrument well documented as tissue-equivalent (Bicron Micro Rem/Sievert Tissue Equivalent Survey Meter).
Instruments “Side by Side” in Field

- Blackstone 6
  - DRUM Site

- Walker Field (GJ Airport)
  - Calibration Pads

- GJ Disposal Site
  - Calibration Pad H
Quantifying “Body Shielding” Factor in Backpack Configuration

• Observed that PGIS-2 response varied slightly based on position of body relative to calibration sources and at DRUM sites
• Conducted study to quantify “body shielding effect”
• Determined could approach 15 %* depending on specifics of the geometry (position of body relative to direction and extent of source

*Note that soil sampling including quantification of U and Ra concentrations further define the radiological environment of these sites
Quantifying “Body Shielding” Factor in Backpack Configuration - continued
## Results – PGIS-2 vs. HPIC

<table>
<thead>
<tr>
<th>(A) Location</th>
<th>(B) HPIC Mean Exposure Rate (µR/hr)</th>
<th>(C) PGIS-2 Mean Dose Rate (µrem/hr) Corrected with Shielding Factor of +15%(^{(1)})</th>
<th>(D) PGIS-2 Equivalent Mean Dose Rate Converted to µR/hr - Column (C) × 1.14 µR per µrem</th>
<th>(E) Difference - Column (D)/ Column (B)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>13.7</td>
<td>8.6</td>
<td>9.8</td>
<td>0.71</td>
</tr>
<tr>
<td>2</td>
<td>17.8</td>
<td>11.7</td>
<td>13.4</td>
<td>0.75</td>
</tr>
<tr>
<td>3</td>
<td>22.2</td>
<td>15.0</td>
<td>17.1</td>
<td>0.77</td>
</tr>
<tr>
<td>4</td>
<td>26</td>
<td>18</td>
<td>20.5</td>
<td>0.78</td>
</tr>
<tr>
<td>5</td>
<td>25.1</td>
<td>17.1</td>
<td>19.5</td>
<td>0.78</td>
</tr>
<tr>
<td>6</td>
<td>34.3</td>
<td>21.3</td>
<td>24.3</td>
<td>0.71</td>
</tr>
<tr>
<td>7</td>
<td>57.5</td>
<td>54.5</td>
<td>62.1</td>
<td>1.08</td>
</tr>
<tr>
<td>8</td>
<td>100.6</td>
<td>71.2</td>
<td>81.2</td>
<td>0.80</td>
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<tr>
<td>9</td>
<td>113.7</td>
<td>97.7</td>
<td>110.7</td>
<td>0.97</td>
</tr>
<tr>
<td>10</td>
<td>141</td>
<td>111.5</td>
<td>127.1</td>
<td>0.90</td>
</tr>
<tr>
<td>11</td>
<td>448.6</td>
<td>409(^{(1)})</td>
<td>466</td>
<td>1.04</td>
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<tr>
<td>Average Difference</td>
<td></td>
<td></td>
<td></td>
<td>0.85/0.95(^{(1)})</td>
</tr>
</tbody>
</table>

\(^{(1)}\) 0.85 = average of all 11 locations; 0.95 = average of locations at 5 highest exposure rates
Results – PGIS-2 vs. BICRON

<table>
<thead>
<tr>
<th>Location No.</th>
<th>PGIS-2 Mean Dose Rate (µrem/hr.) Corrected With Shielding Factor of +15%</th>
<th>Bicron Micro Rem Survey Meter Dose Rate (µrem/hr.)</th>
<th>Difference (PGIS-2 Corrected vs. Bicron Micro Rem Survey Meter)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8.6</td>
<td>8</td>
<td>1.07</td>
</tr>
<tr>
<td>2</td>
<td>11.7</td>
<td>15</td>
<td>0.78</td>
</tr>
<tr>
<td>3</td>
<td>15.0</td>
<td>18</td>
<td>0.83</td>
</tr>
<tr>
<td>4</td>
<td>18.0</td>
<td>20</td>
<td>0.90</td>
</tr>
<tr>
<td>5</td>
<td>17.1</td>
<td>20</td>
<td>0.87</td>
</tr>
<tr>
<td>6</td>
<td>21.3</td>
<td>25</td>
<td>0.85</td>
</tr>
<tr>
<td>7</td>
<td>54.5</td>
<td>50</td>
<td>1.09</td>
</tr>
<tr>
<td>8</td>
<td>71.2</td>
<td>98</td>
<td>0.73</td>
</tr>
<tr>
<td>9</td>
<td>97.7</td>
<td>90</td>
<td>1.08</td>
</tr>
<tr>
<td>10</td>
<td>111.5</td>
<td>120</td>
<td>0.93</td>
</tr>
<tr>
<td>11</td>
<td>N/A – Backpack at contact with Pad Surface</td>
<td>500</td>
<td>0.82</td>
</tr>
</tbody>
</table>

Difference 0.91
Example 1: Gamma Survey Output
Example 2: Gamma Survey Output
Summary and Conclusions

- Future exposure scenario used 2-weeks per year for a recreational camper with associated exposure pathways and 1 mSv dose constraint.
- Consistent with limitations of use established by BLM for the climate and ecosystems associated with these generally remote and semi-arid sites.
- Gamma exposure pathway dominates the dose under the recreational exposure scenario (>85 percent) and can be readily measured in the field.
- Wide area gamma survey protocols developed using standard industry practices.
- DOE chose NaI based portable gamma-ray spectrometer with advanced microprocessor technologies.
Conclusions (continued)

- Normalization field studies used energy independent primary standard (Reuter Stokes HPIC) and BICRON tissue equivalent survey meter in “side by side” comparisons with the PGIS-2
- Within limitations of field circumstances, PGIS-2 demonstrated generally good energy independence and tissue equivalence
- Field survey protocols applying the previously developed radiological screening levels with these instruments have completed > 700 DRUM site wide area gamma surveys through May 2019
Questions?

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