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

Steven H Brown CHP
SHB Inc., Centennial, Colorado USA

Ian Shafer, US Department of Energy, Grand Junction, Colorado USA

Uranium Recovery Workshop
July 18-19, 2022
Denver, Co. USA

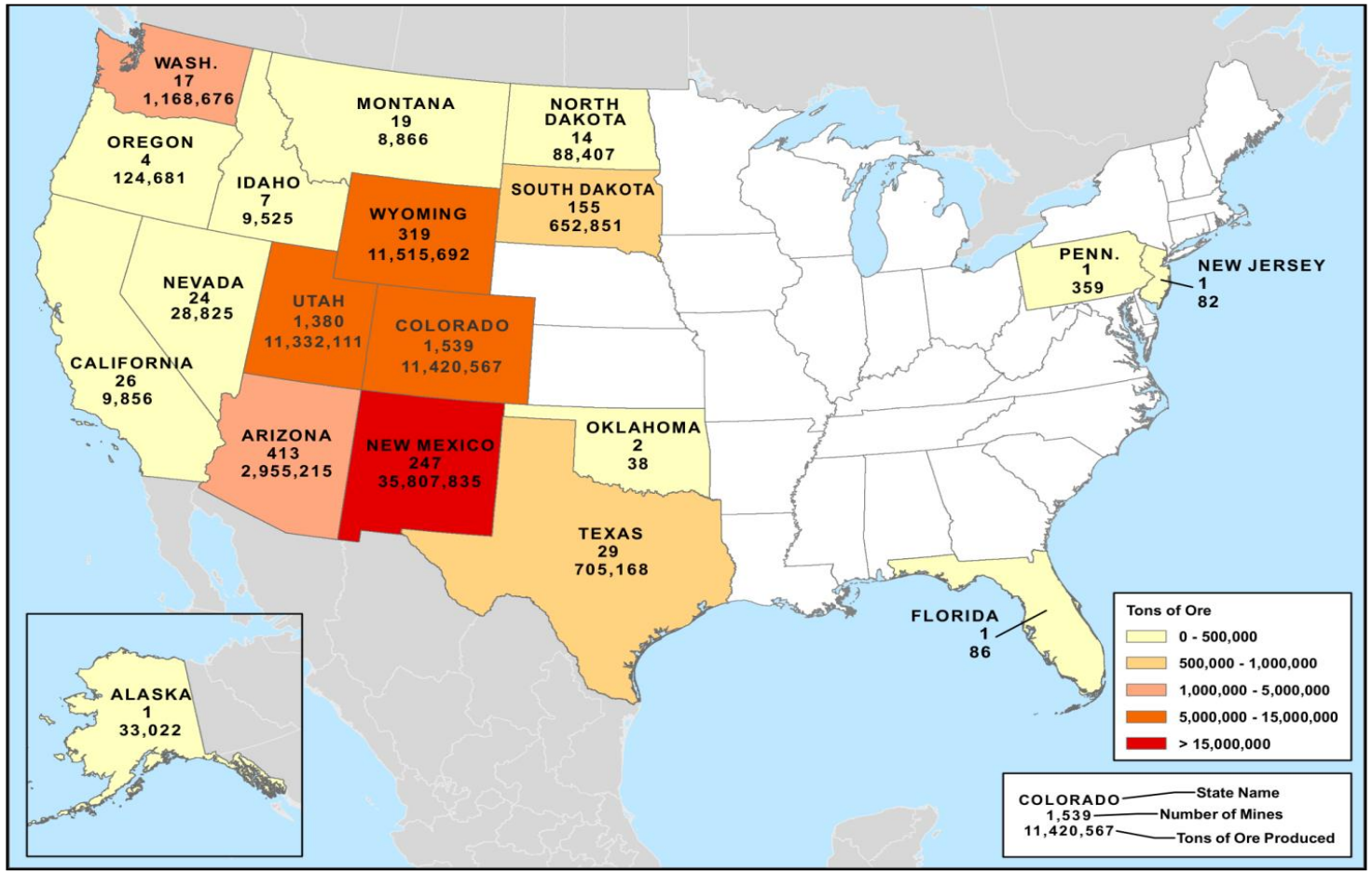
Outline

- Overview - US DOE Office of Legacy Management Defense Related Uranium Mine (DRUM) Sites
- Radiological Environment of DRUM Sites
- Determination of Radiological Screening levels
  - Recreationist and Resident
- Dose Assessment and Screening Level Methodology
- Gamma Survey Instrument Normalization Study
- Typical Results
- Conclusions

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

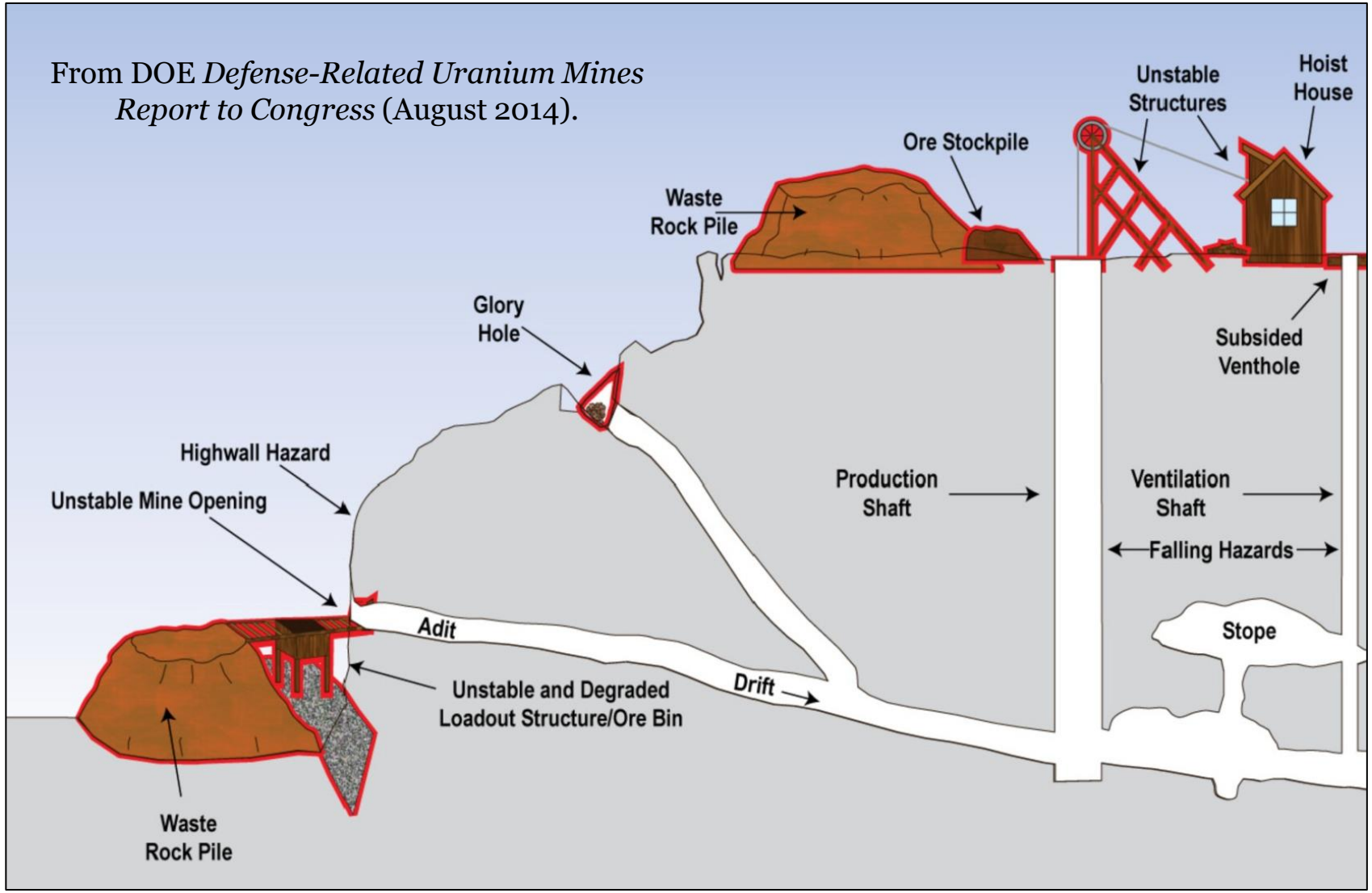
- US DOE-LM is responsible for validating and verifying (screening level characterization) approximately 3500 Defense Related Uranium Mining (DRUM) sites (many are small – but some are not)
- US Atomic Energy Commission purchased uranium ore prior to 1970 for defense programs.
- Most are in remote areas of SW US (CO, WY, UT, AZ, NM).
- DOE-LM has developed radiological screening criteria and wide area gamma survey protocols to assist in prioritizing this very large # of sites (along with physical and chemical hazards, human accessibility, other factors).

Number of DRUM Sites and Tons of Uranium Ore Produced by State



From DOE *Defense-Related Uranium Mines Report to Congress* (August 2014).

DRUM Site Features



Typical Environments – DRUM Sites(Legacy Uranium Mines)

Overview – DRUMs on Public Land

- For sites on Federal land - primarily Bureau of Land Management (BLM) and US Forest Service (USFS) - 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.
- Most sites are in arid environments with sparse vegetation; game animals are migratory and recreationists must bring own water and food.

Overview – DRUMS on Tribal or Private Land

- For sites on Tribal or private land, a full-time resident is assumed
- Residency for 350 days per year – 75% time at residence including periods of indoor and out of doors occupancy
- Public exposure limit of 1 mSv (100 mrem) / yr. applied to the annual exposure period to define a “high risk” site if exceeded.
- Most sites are in arid environments with sparse vegetation; game animals are migratory and groundwater is deep
- Residual radioactive material (NORM) contained in surface deposits, not at depth
- Accordingly, typical ground water exposure pathways (e.g. consumption from local water wells, vegetable gardens) are not impacted

Overview – Radiological Screening Criteria

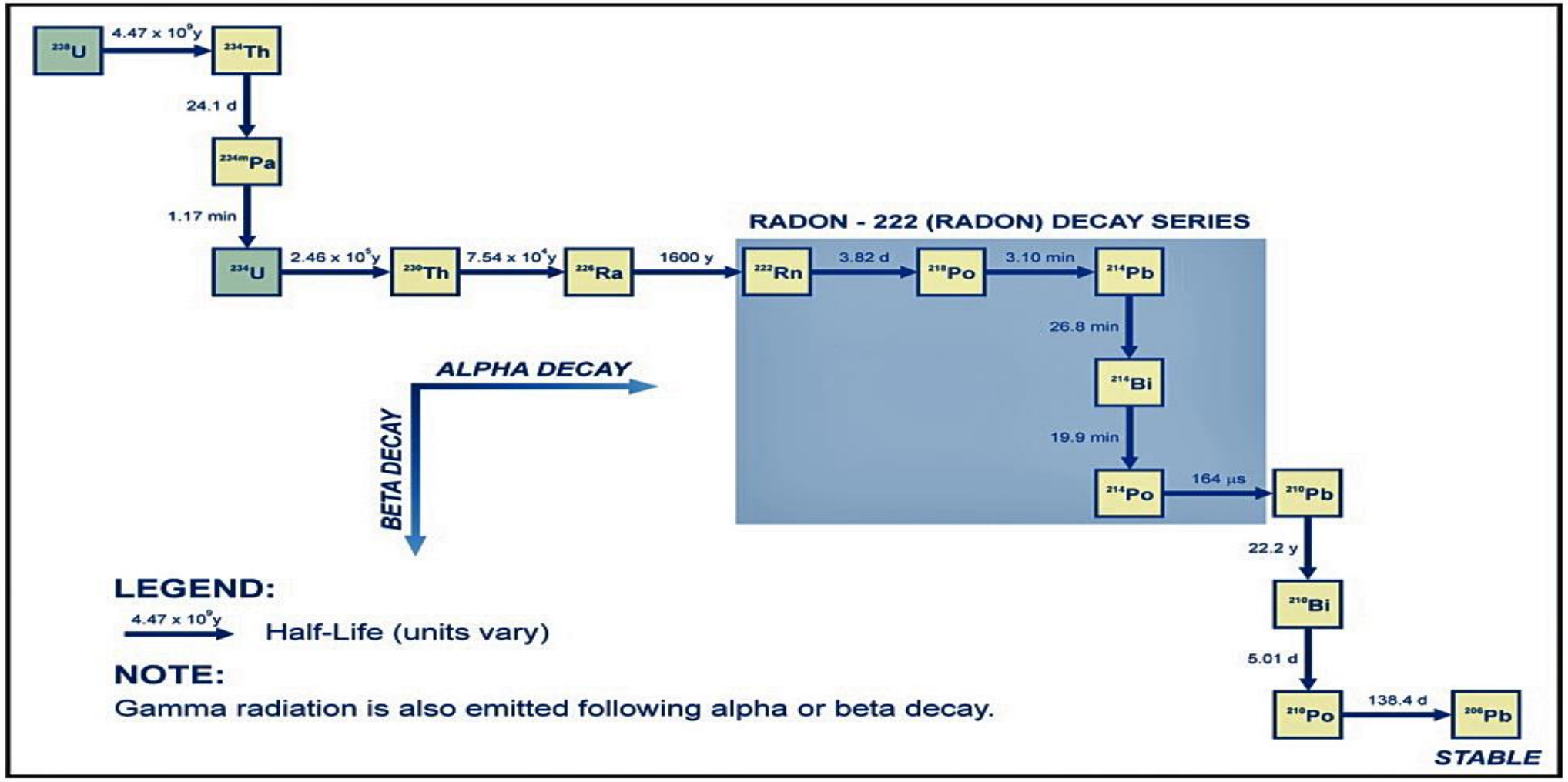
- Gamma exposure rates are primary screening criteria (uSv [uRem] per hr.)*
- Other federal agencies (i.e., U.S. Nuclear Regulatory Commission, U.S. Environmental Protection Agency, U.S. Army Corps of Engineers) use ^{226}Ra concentrations in soil as basic remediation criteria for legacy natural uranium (U) sites.
- Relationship of uSv (uRem) per hour values to Bq (pCi) per gram ^{226}Ra is secondary screening criteria.
- Purpose of screening criteria to assist in prioritization and hazard ranking of over three thousand sites

* 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

Radiological Environment Source Term Defined

- Contaminants of concern: natural U ore in soil, waste rocks, and spoils piles
- Full radioactive equilibrium in ^{238}U decay series
- Ratio of ^{235}U activity to ^{238}U activity is the natural abundance ratio (0.046)
  - Dose contribution from ^{235}U (actinium) series is small, but was considered
- Contribution from natural thorium (^{232}Th) series is negligible
  - Consistent with general ^{232}Th regional background

Uranium Series Radiological Source Term



Relative Abundances of Naturally Occurring Uranium* Isotopes

Isotope	Percent of Mass in U Natural	Percent of Radioactivity in U Natural	Half Life (years)
^{238}U	99.3	48.9	4.5 billion
^{235}U	0.72	2.2	704 million
^{234}U	0.005	48.9	245,000

*U.S. Public Health Service, Agency for Toxic Substances and Disease Registry. *Toxicological Profile for Uranium*. 2011.

Typical Pathways Evaluated for Future Exposure

Most common

- External exposure from deposition onto/in soil (unique for radiological sites)
- Incidental inhalation of dust (re-suspended soil)
- Direct inhalation of radon/progeny (U sites)
- Incidental ingestion of soil and dusts (e.g., vegetable gardens, children playing in garden, construction activities)
- Direct ingestion of water from contaminated zone
- Direct ingestion of foods grown or raised in contaminated zone (e.g., vegetables, meat, milk, and fish) specific to the area

Pathway Analysis for a Recreational User of DRUM Sites

Pathway	Circumstances for Recreational Use of DRUM Sites
External (gamma) from Soil	Ore residues and waste rock, spoils piles, soil, adits and portals, etc.
Inhalation of Soil Dusts	Incidental and OHV rider
Inhalation of Radon and Progeny	Little time for progeny ingrowth; outdoors - assume user not in adits or portals (safety)*
Ingestion of Water	Not applicable (NA) - must bring own water
Ingestion of Food	NA - must bring own food; vegetation generally sparse, particularly on/around ore/spoil piles; migratory game animals (did not spend life in "contaminated zone")

* Priority of reclamation activities is removal of physical hazards; many open portals and adits will have been sealed or otherwise remediated

Pathway Analysis for a Residential User of DRUM Sites

Pathway	Circumstances for Residential Use of DRUM Sites
External (Gamma) from Soil	Ore residues and waste rock, spoils piles, soil, adits and portals, etc.
Inhalation of Soil Dusts	Incidental
Ingestion of Soil	Incidental
Inhalation of Radon and Progeny	Not Applicable (NA) - circumstances of home construction, foundation and NORM content in construction materials dominate potential for exposure within the home
Ingestion of Water and Food	NA - "contaminated zone" only at surface - no impact on deep ground water and therefore no impact on vegetable gardens, etc.

Perform Dose Analysis for Each Applicable Exposure Pathway

- General approach
  - Place a unit concentration of the reference radionuclide(s), (e.g., 1 Bq {pCi} / g) in the soil along with the associated ratios of the other dosimetrically important radionuclides - For natural U ore, ratio is 1 for all progeny
  - Perform intake and dose modeling to establish the annual dose associated with the reference nuclide and mixture in each relevant pathway (per Bq per gram in soil) and sum results
  - This defines the reference dose per unit concentration in soil
    - A simple ratio of the annual dose limit (e.g., 1 mSv) to the reference dose defines the concentration in soil that ensures the dose limit will not be exceeded
    - Establish % contribution from external pathway
    - Ratio of annual dose to applicable exposure hours = exposure rate that establishes operational screening level (e.g., uSv per hr.)

Methodology For Developing Radiological Risk Screening Levels for Recreationist*

"Put" unit concentration of ^{238}U in soil - e.g., 1 Bq per gram; all other nuclides in decay series = 1 Bq per gram (equilibrium)*

$$\mathbf{T = Ex + S_{inj} + I_d + I_{ohv} + I_{Rn}}$$

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

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

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

$\mathbf{I_d}$ = CEDE from inhalation of dusts

$\mathbf{I_{ohv}}$ = CEDE from inhalation of dusts during use of Off Highway Vehicle (OHV)

$\mathbf{I_{Rn}}$ = CEDE from inhalation of ^{222}Rn and progeny

* Details presented in Brown et al. Establishing Radiological Screening Levels for Defense-Related Uranium Mine (Drum) Sites on BLM Land Using a Recreational Future-use Scenario. Health Physics. June 2018, Volume 114, Number 6

Methodology For Developing Radiological Risk Screening Levels for Resident

The Total Effective Dose Equivalent (TEDE) is the sum of the dose equivalents from each pathway:

$$T = E_x + S_{ing} + I_d$$

Where:

T = TEDE for a 350 days per year residential exposure per Bq/gram including each of the ^{238}U plus ^{235}U series radionuclides in soil.

E_x = Dose Equivalent (DE) from external exposure from the soil – sum of exposure while outdoors ($Ex_{outside}$) + indoors (Ex_{inside})

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

I_d = CEDE from inhalation of fugitive dusts

Why No Groundwater and Radon / Progeny for Resident ?

- Groundwater - These sites only involve residual surface deposits, climate is arid, groundwater is deep – drinking water and local vegetable garden pathways not applicable
- Radon / Progeny – Structural, construction and ventilation related aspects of homes have much greater impact on exposure than ^{226}Ra soil levels underneath
- Homes built atop soil with identical levels of radium can have orders of magnitude differences in indoor radon levels; reducing the radium content in the soil may not result in any reduction in indoor radon levels and therefore evaluation of this exposure pathway is not necessary. (US EPA 2001)

Results - Recreationist

TEDE = Ex + S_{inj} + I_d + I_{ohv} + I_{Rn}

T = 162 + 2.8 + E-3 + 0.3 + 19.6 = 184 uSv per Bq per gram (0.68 mrem per pCi per gram)

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

Contribution to the TEDE from external gamma exposure = 162 / 184 = 88%

This relationship was used directly to establish average gamma exposure rate above background that ensures < annual limit: 0.88 X 1mSv during 2 weeks per year of recreational use = 2.56 uSv (256 µrem) per hr. above bkg.

Accordingly, Upper Screening Level = 2.56 µSv / hr. above bkg..

Approach – Resident

- Residential scenario screening levels are still under development and will be reported in the future
- Approach essentially identical to that used for Recreationist with 1 mSv per yr. as dose constraint
- Establish a unit concentration of 1 pCi (Bq) per gram of ^{238}U in soil
- All other radionuclides in the uranium decay series will be present at 1 pCi (0.036 Bq) per gram since secular equilibrium is assumed.
- The exposure as a Total Effective Dose Equivalent (TEDE) over a 1-year period is the sum of the dose rates from each of the relevant nuclides within each relevant exposure pathway as previously identified
- With a few exceptions, all parameter inputs are being derived from default values provided in US EPA *Soil Screening Criteria for Radionuclides* (2001)

Historical Issues with NaI based Measurements In Uranium Series Photon (Gamma) Fields

- Sodium iodide 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 – instrument response function of average energy of photon field (calibrated to Cs 137, higher energy than U series average) - instrument will “over respond” in U gamma field
  - Tissue Equivalence – instrument measuring exposure rate in air, not absorbed dose in tissue (density air vs. tissue) – screening levels based on “dose” to humans

Instrument Performance Verification

- DOE –LM is using NaI (Tl) based spectrometer but with advanced microprocessor technologies in a backpack configuration
- Field studies conducted 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 (high-pressure ionization chamber - HPIC) and to another instrument well documented as tissue-equivalent (Micro Rem/Sievert Tissue Equivalent Survey Meter).

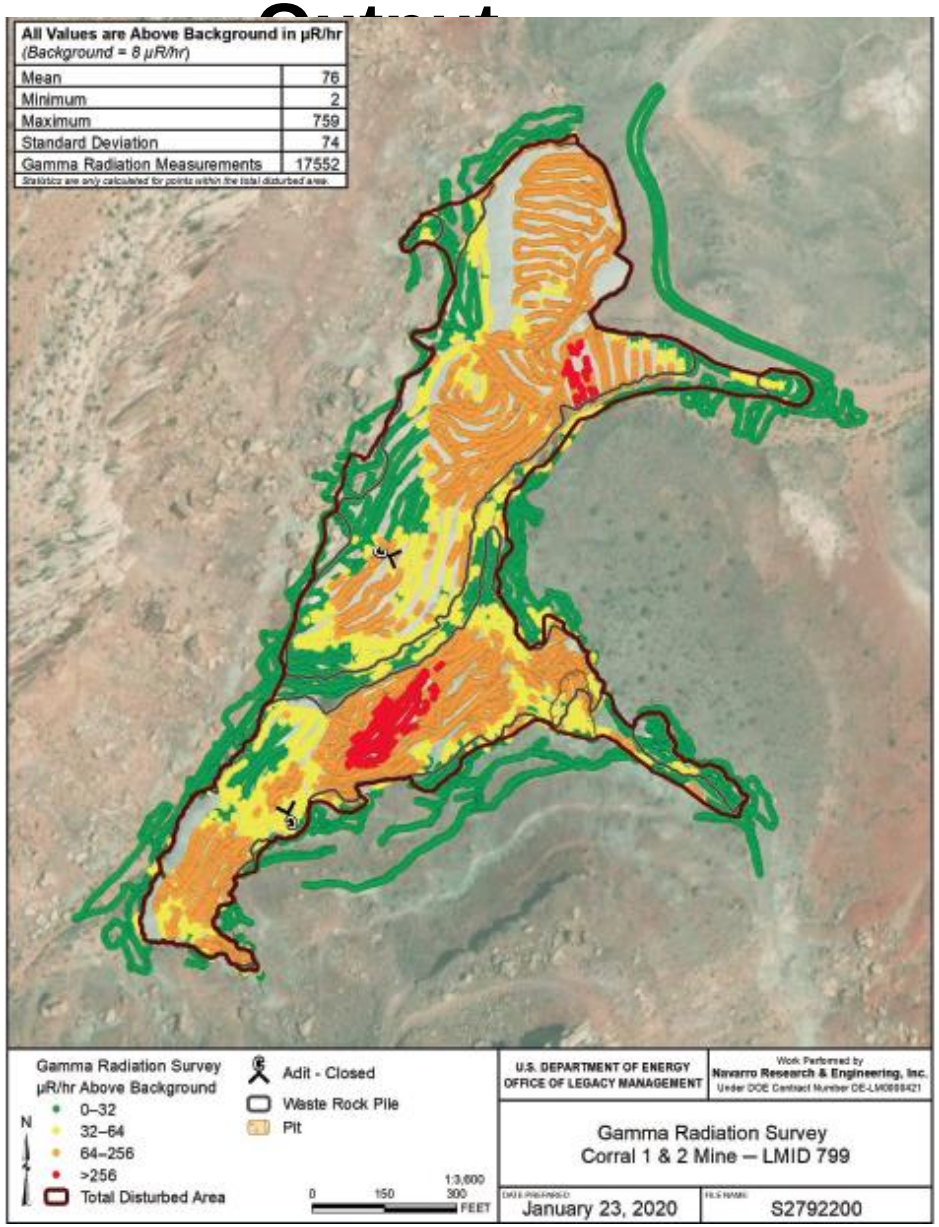
Instruments “Side by Side” in Field

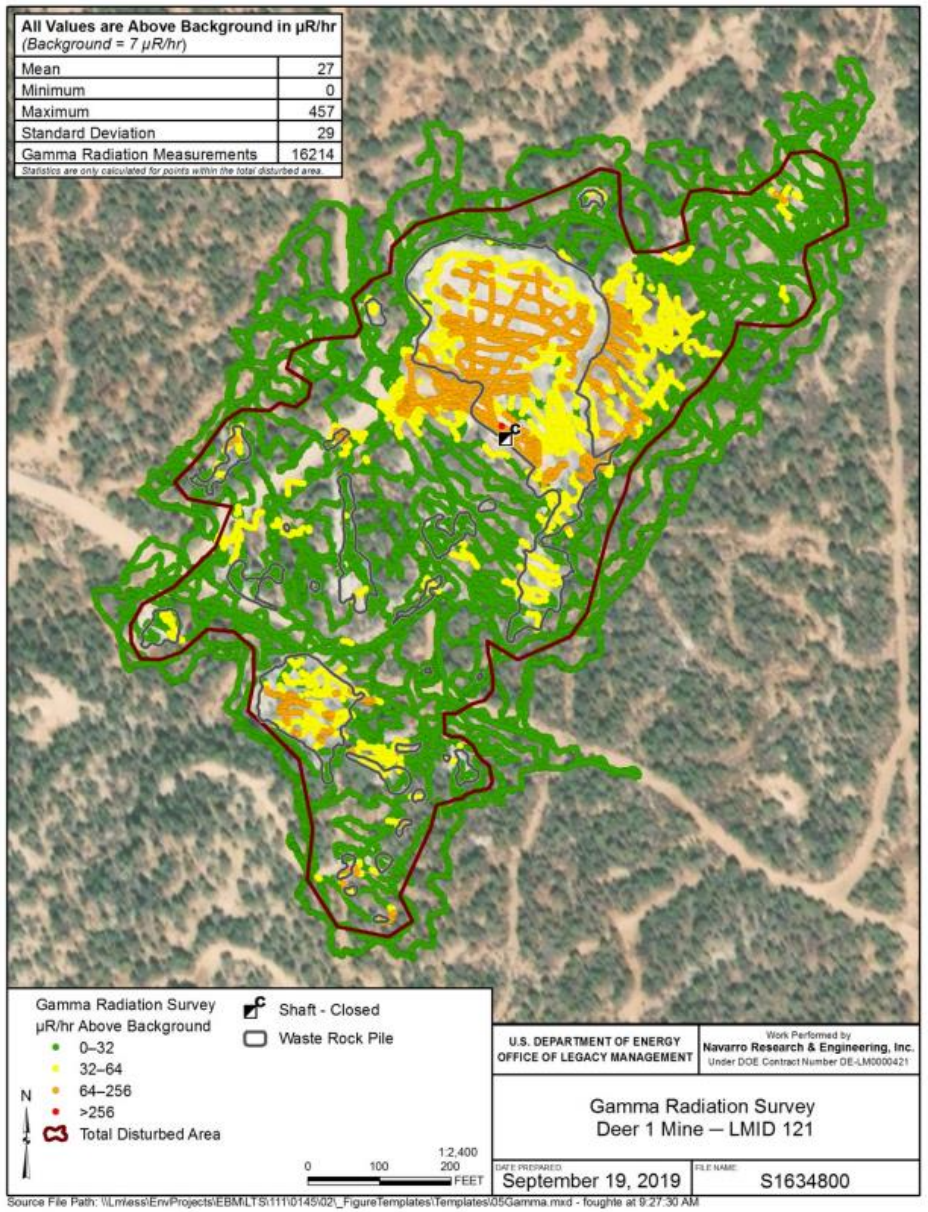
Blackstone 6
DRUM Site

Walker Field (GJ Airport)
Calibration Pads

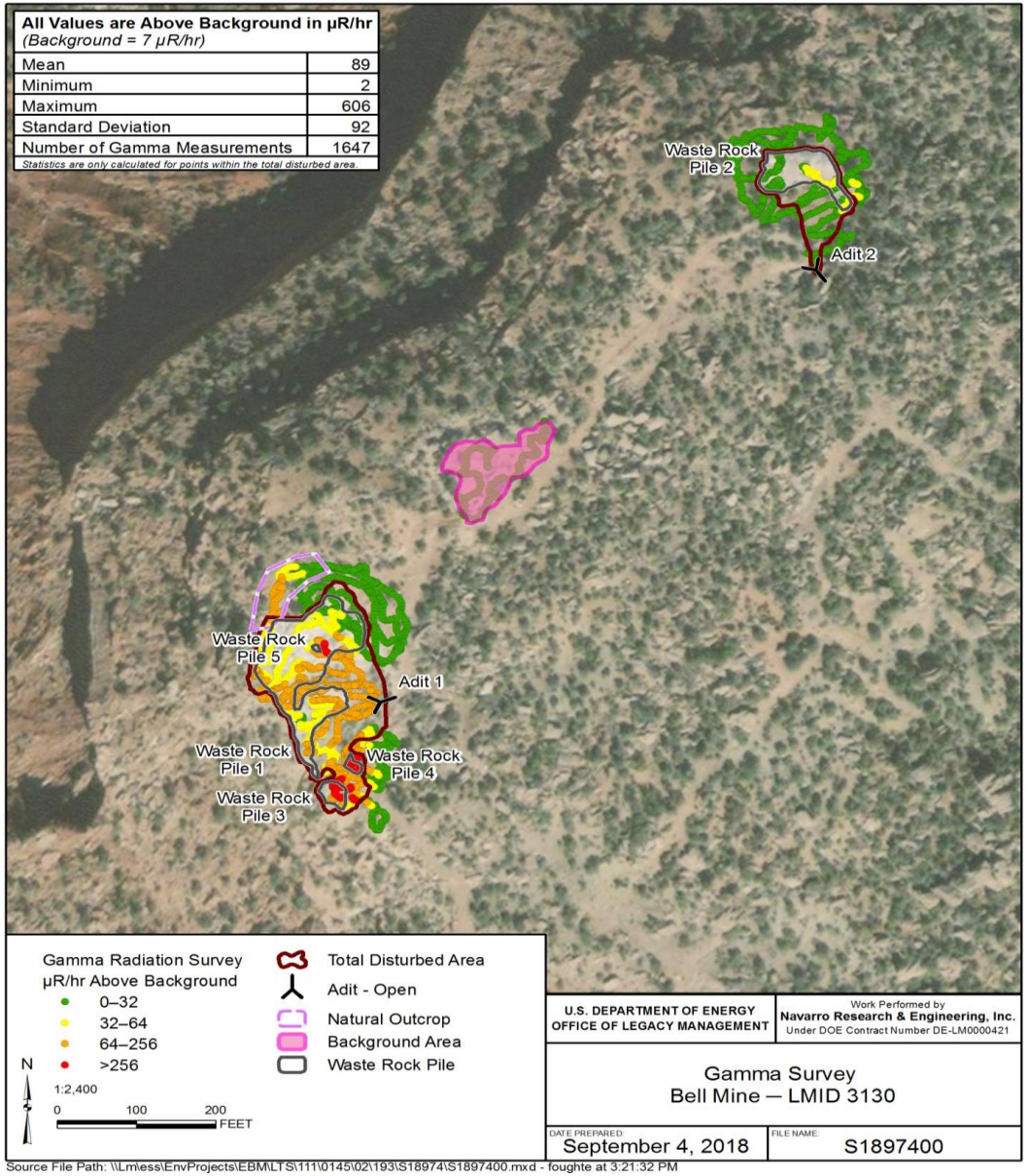
GJ Disposal Site
Calibration Pad H

Example 1 and 2: Gamma Survey Output

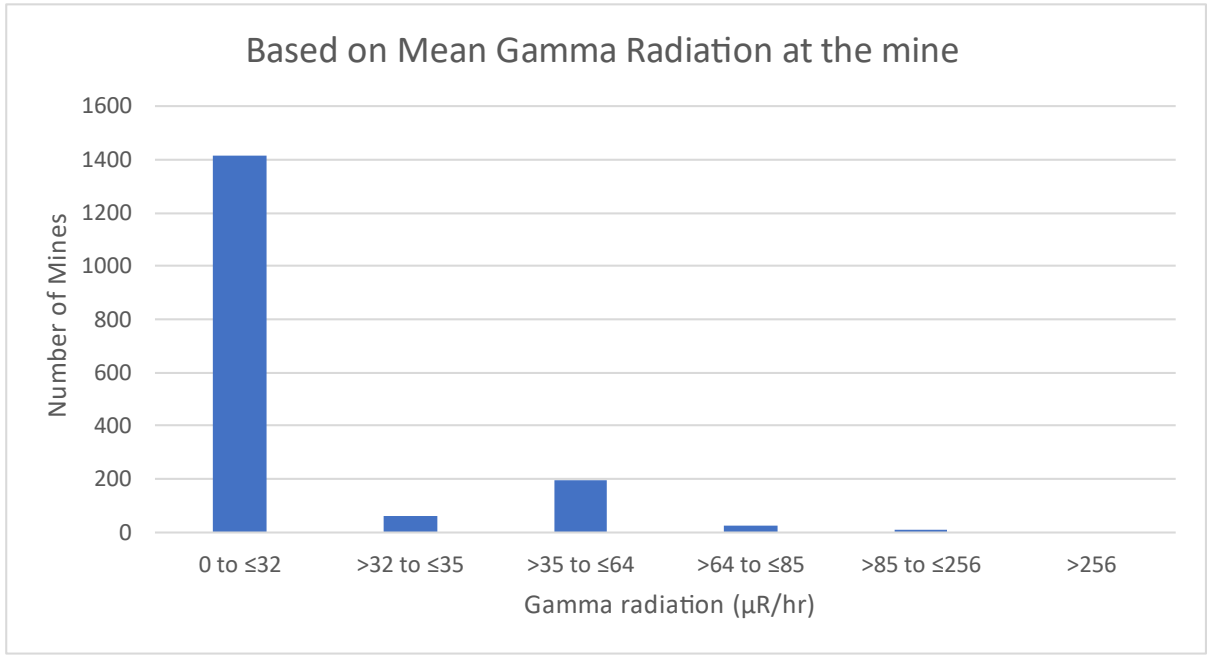


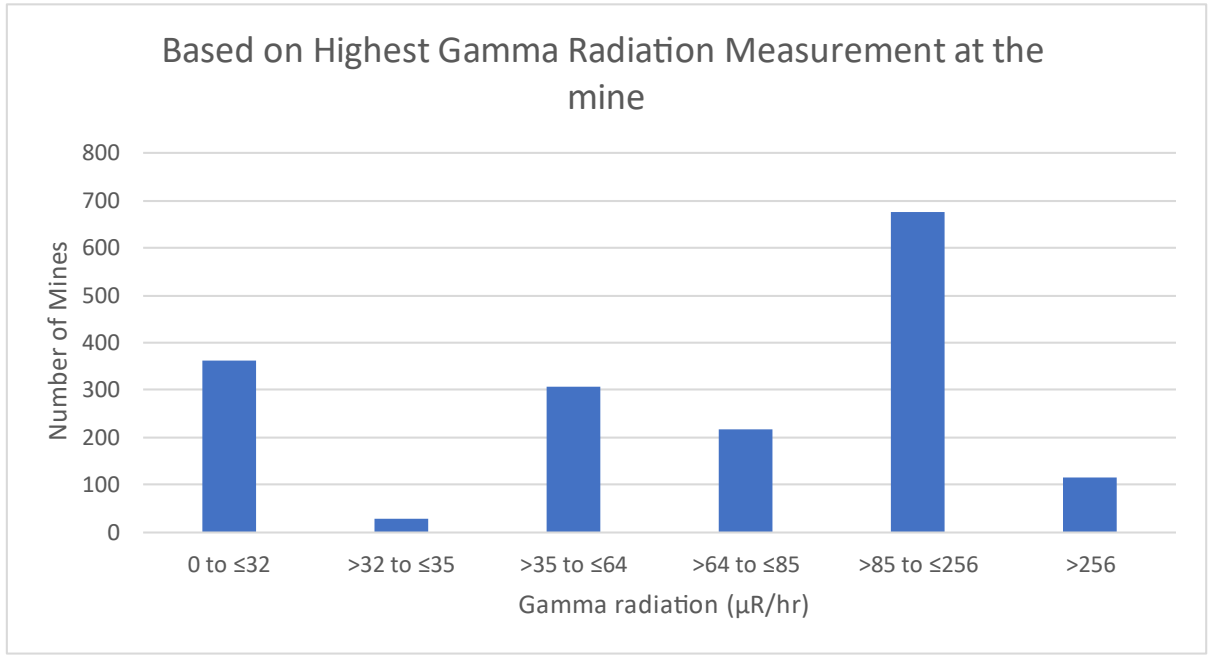


Example 3: Gamma Survey Output



Radiological Data Summary – about 1800 DRUM sites Thru CY 2021





100 uR = 1 uSv

Summary and Conclusions

- Exposure scenarios defined for a recreational camper and residential use with associated exposure pathways and annual 1 mSv dose constraint.
- Consistent with limitations of use established for the climate and ecosystems associated with these generally remote and arid sites.
- Gamma exposure pathway dominates the dose under both scenarios (>88 percent) and can be readily measured in the field.
- Wide area gamma survey protocols developed using standard industry practices.
- DOE chose NaI(Tl) based portable gamma-ray spectrometer with with advanced microprocessor technologies.

Conclusions (continued)

- Performance verification field studies used energy independent primary standard (HPIC) and tissue equivalent survey meter in “side by side” comparisons with the NaI(Tl) based gamma instruments.
- Within limitations of field circumstances, generally good energy independence and tissue equivalence was demonstrated .
- Field survey protocols applying the previously developed radiological screening levels with these instruments have completed ~1800 DRUM site wide area gamma surveys by the end of CY 2021.

Questions?

Point of Contact:
Steve Brown, CHP
SHB Inc.
Centennial, CO. USA
+1 303 524 1124 (office)
+1 303 941 1506 (mobile)
shb12@msn.com
www.shbinc.us