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A STUDY OF WIPP RADIOLOGICAL TRANSURANIC WASTE CHARACTERIZATION REQUIREMENTS AND ACTIVITIES

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PECOS MANAGEMENT SERVICES, INC.

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ACRONYMS

AEC	Atomic Energy Commission
AK	acceptable knowledge
Am	americium
ASTM	American Society for Testing and Materials
ATWIR	annual transuranic waste inventory report
Ba	barium
CCA	compliance certification application
Cf	californium
CFR	Code of Federal Regulations
CH	contact handled
Ci	curies
Cm	curium
Co	cobalt
CRA	compliance recertification application
Cs	cesium
DA	destructive assay
DOE	Department of Energy
DOT	Department of Transportation
DSA	documented safety analysis
DTC	dose-to-curie
EPA	Environmental Protection Agency
Eu	europium
FEIS	final environmental impact statement
FGE	fissile gram equivalent
H	hydrogen
HLW	high-level waste
Kr	krypton
LLW	low level waste
LWA	Land Withdrawal Act
mrem/hr	millirem per hour
mrem/yr	millirem per year
MS	mass spectrometry

nCi/g	nanocuries per gram
NDA	nondestructive assay
NIST	National Institute of Standards and Technology
NMED	New Mexico Environment Department
Np	neptunium
NRC	Nuclear Regulatory Commission
ORIGEN	Oak Ridge Isotope GENeration
ORIGEN-ARP	ORIGEN Automatic Rapid Processing
ORNL	Oak Ridge National Laboratory
PA	performance assessment
PAN	passive-active neutron
PE-Ci	plutonium-239 equivalent activity
PECOS	PECOS Management Services, Inc.
Pm	promethium
Pu	plutonium
Ra	radium
rem	Röentgen equivalent man
rem/hr	rem per hour
rem/yr	rem per year
RH	remote handled
RTR	real-time radiography
SGS	segmented gamma scanning
Sr	strontium
$t_{1/2}$	half-life
TRAMPAC	TRU waste authorized methods of payload control
TRU	transuranic
TSR	technical safety requirements
TWBIR	transuranic waste baseline inventory report
U	uranium
VE	visual examination
WAC	waste acceptance criteria
WIPP	Waste Isolation Pilot Plant
Y	Yttrium

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I. SCOPE AND PURPOSE

Both contact handled (CH) and remote handled (RH) transuranic (TRU) waste must be characterized before they can be transported to the Waste Isolation Pilot Plant (WIPP), the geologic repository located in southeastern New Mexico, for permanent disposal. The following methodologies determine physical, chemical, and radiological parameters of TRU waste: visual examination (VE), real-time radiography (RTR), acceptable knowledge (AK), nondestructive assay (NDA), dose-to-curie (DTC) conversions, destructive assay (DA), waste sampling, and surface dose rate measurements. Of these, AK, DTC, NDA, DA, and surface dose rate measurements define the radiological characteristics of TRU waste bound for the WIPP.

As part of the scope of this task, PECOS Management Services, Inc. (PECOS) has reviewed documentation associated with TRU waste radiological characterization activities, including associated regulatory requirements, the basis for these requirements, and characterization program changes that have occurred over time. PECOS has also examined the full spectrum of radiological data acquired and tracked as part of the Department of Energy's (DOE) mission at WIPP.

The purpose of this task is to assess the extent of DOE's radioactive waste characterization activities with respect to regulatory requirements, as well as the significance and value of those activities as they relate to health and safety issues. Based on this assessment, this task intends to determine if DOE could simplify its TRU waste radiological sampling and analysis programs, data tracking, and reporting.

This report contains PECOS' conclusions and recommendations for proposed modifications that could potentially reduce waste characterization program costs and improve the efficiency of waste disposal operations at the WIPP without increasing associated health and safety risks.

II. BACKGROUND

At the WIPP, an integral part of the disposal process for TRU waste is waste characterization. Repository-based regulations and the manner in which TRU waste container-based radioactivity limits are defined drive TRU waste characterization requirements and procedures. These directives have evolved over the years due to regulations imposed on radioactive waste treatment, transportation, and disposal by a number of organizations, including the Department of Transportation (DOT), the Nuclear Regulatory Commission (NRC), the Environmental Protection Agency (EPA), and DOE.

Each of the above agencies upholds specific standards related to radioactive materials. DOE, DOT, and NRC regulations mitigate immediate concerns, while EPA handles long-term issues related to the public and the environment. Specifically, EPA dictates how TRU waste should be disposed of in order to ensure proper containment well into the future.[1, 2] DOE, however, establishes exposure limits for workers,[3] while DOT[4] and NRC[5] uphold a large set of regulations designed to ensure safe packaging and transporting of radioactive material. In essence, these transportation regulations are the primary drivers for TRU waste radiological characterization requirements, followed in importance by environmental performance assessment (PA) requirements.

Various regulatory agencies, each with a different agenda, approved characterization and disposal of CH TRU waste in the 1990s. Consequently, an extensive, stringent program was established and implemented to determine CH TRU waste container contents.[6, 7] This program equipped DOE, regulatory agencies, and concerned citizen groups with a firm safety basis during a time when DOE was learning how to prepare CH TRU waste for disposal at the WIPP. The RH TRU waste characterization program, developed in the 2000s, relies more on process knowledge and less on waste examination to maintain worker health and safety while reducing project costs.[7]

III. SUMMARY OF FINDINGS

The following material provides a synopsis of regulations and the manner in which they were developed. It also identifies portions of the radiological characterization program that DOE could possibly streamline while maintaining the crucial health and safety practices that minimize risks to workers, the public, and the environment.

The Changing Definition of TRU Waste

In 1970, the Atomic Energy Commission (AEC), while attempting to accelerate creation of a salt-based repository in Kansas, established a definition for TRU waste. Prior to 1970, this type of waste was

referred to as plutonium (Pu)-contaminated or alpha (α)-contaminated. According to the AEC, TRU waste had to contain material contaminated with isotopes possessing an atomic number greater than 92 (the number of protons in a uranium [U] atom); it also had to exhibit a radioactivity level of at least 0.00000001 curies (Ci) per gram of waste (or 10 nanocuries per gram [nCi/g]), roughly the same level measured for radium-226 (Ra-226), which occurs naturally in the earth's crust.[8]

Then, in the late 1970s and early 1980s, EPA and DOE increased the activity concentration of TRU waste by an order of magnitude to 100 nCi/g. As noted by Rechard,[8] EPA investigations demonstrated that this new level would keep doses below 0.5 Röentgen equivalent man (rem) per year, or 500 millirem/yr (mrem/yr) for members of the general public.[9] Current EPA standards in Title 40 of the Code of Federal Regulations Part 191 (40 CFR 191) state that for the general public, continuous exposure cannot exceed 100 mrem/yr, while infrequent exposure cannot exceed 500 mrem/yr. (The occupational level for workers is higher: five rem/yr, according to 10 CFR 835.202.) Some sources[8] also speculate that the activity concentration limit was increased not only as a disposal activity cost-saving measure, but also because assay technology was not robust enough to segregate waste at the previous 10 nCi/g level.[10] By raising the limit, about 20 percent of TRU waste inventory was re-classified as low-level waste (LLW) without compromising long-term health and safety of workers, the public, or the environment.[8]

Subsequently during the development of the Nuclear Waste Policy Act of 1982, a condition was added asserting that the half-life ($t_{1/2}$) of contaminating isotopes in TRU waste must be greater than five years. While this did not exclude short-lived radionuclides within the waste, it did require that longer-lived radionuclides be disposed so they would be benign at the end of their isolation period.[8]

When 40 CFR 191 was issued in 1985, EPA once again modified the definition of TRU waste. In addition to previous descriptions, TRU waste was redefined as containing α-emitting TRU isotopes, which have the $t_{1/2} > 20$ years, not five years, further reducing the amount of waste that could be classified as TRU. EPA also stipulated that this type of waste could not be regarded as any of the following: high-level waste (HLW); waste that did not require geologic isolation as determined by DOE and EPA; or waste approved by NRC on a case-by-case basis per 10 CFR 61.

In September of 1988, DOE issued Order 5820.2A for Radioactive Waste Management, a version that superseded the 1986 DOE Order 5820.2 and included a revised definition of TRU waste. As a result of this updated order, waste could be classified as TRU without regard to its source or form as long as it contained more than 100 nCi/g of α-emitting TRU isotopes with $t_{1/2} > 20$ years at the time of assay. DOE could decide if other α-contaminated waste should be managed as TRU waste. The order further defined CH and RH TRU waste by designating a limiting surface dose rate of 200 mrem per hour (mrem/hr). Below this value, TRU waste would be considered CH; above it, TRU waste would be classified as RH.

Both DOT and the NRC formally adopted this limit and included it in their 1995 regulations for packaging and transporting radioactive materials.

Effect of Repository Limits on Waste Characterization Requirements

Repository limits for disposing TRU waste were first defined in the 1980s when DOE announced its decision to proceed with the phased development of the WIPP and subsequently produced the Consultation and Cooperation Agreement with the state of New Mexico. At that time, the WIPP was meant to accommodate 6,200,000 ft^3 (175,564 m^3) of CH TRU waste and 250,000 ft^3 (7,079 m^3) of RH TRU waste. Additionally, waste containers with a surface dose rate in excess of 100 rem/hr were prohibited at the WIPP, as stated in the final environmental impact statement (FEIS).

The Land Withdrawal Act (LWA)[11] ultimately defined repository limits for the WIPP in 1992, however, when it declared the following:

- Only defense-related TRU waste may be emplaced at WIPP.
- The surface dose rate of CH TRU waste containers must be less than 200 mrem/hr. No lower limit is specified in the LWA.
- The surface dose rate of RH TRU waste containers must be at least 200 mrem/hr, but cannot exceed 1,000 rem/hr (as opposed to 100 rem/hr stipulated in the FEIS).
- 95 percent of RH TRU waste volume is allowed in containers with a surface dose rate less than 100 rem/hr. The remaining five percent is allowed a surface dose rate as high as 1,000 rem/hr.
- The average total activity of waste inside an RH TRU waste canister must be less than 23 Ci/liter. This activity includes α, beta (β), gamma (γ), and neutron radiation at the time of disposal.
- The total activity of RH TRU waste allowed at the WIPP cannot be greater than 5,100,000 Ci. This includes α, β, γ and neutron radiation at the time of disposal. There is no corresponding limit for CH TRU waste stipulated in the LWA.
- The combined volume of CH and RH TRU waste allowed at WIPP must be less than or equal to 6,200,000 ft^3 (175,564 m^3), not 6,450,000 ft^3 (182,643 m^3) as had been established more than 10 years earlier.

Previous Characterization and Storage of TRU Waste

Along with defining TRU waste in 1970, the AEC also mandated that newly generated TRU waste be stored above ground (hence the term “retrievably stored”) to facilitate permanent disposal, estimated to occur within 10 to 20 years.[8] Prior to this decision, TRU waste was managed in much the same way as

LLW: pre-1970 waste was often simply buried in trenches or stored in tanks at generator sites. Consequently, while there is substantial information available regarding TRU waste generated since 1970, there are fewer data regarding the characteristics of radioactive waste generated prior to that era. (Earlier waste streams are collectively known as legacy waste.) What is known is limited mainly to historical AK.

Typically, AK is generated for a waste stream (not a specific drum), although it can be augmented using a variety of container-based waste characterization results. Information presented as AK includes process data (such as the date and purpose of waste creation), results of previously conducted sampling and analysis activities, and details collected when older waste was treated or repackaged in an effort to make it compliant with the WIPP Waste Acceptance Criteria (WAC). While AK consisted almost exclusively of historical information for legacy waste, it now includes data collected during production and packaging of newly generated waste.[7]

TRU Waste Health and Safety Considerations

Although there are numerous health hazards associated with characterizing, packaging/re-packaging, transporting, and disposing TRU waste, these risks are minimized when workers handle waste according to the WIPP WAC. The WAC incorporates requirements from the Hazardous Waste Facility Permit; Titles 10, 40, and 49 of the CFR; NRC certificates of compliance for transportation containers; the LWA; WIPP environmental impact statements; and WIPP CH and RH documented safety analyses (DSAs).

Implementation of 40 CFR 194, which established waste characterization guidelines as they apply to EPA compliance certification/recertification, reduces some radiation-related risks. Because these guidelines relate to the PA conducted for WIPP, this set of regulations ensures a low probability of long-term radiological releases that might impact the environment and members of the public. Short-term radiological risks associated with packaging and transporting TRU waste, on the other hand, are mitigated by certificates of compliance for TRU waste transportation containers, 10 CFR 71 (NRC regulations for packaging and transporting radioactive materials), and 49 CFR 171-180 (DOT regulations for packaging and transporting hazardous [radioactive] materials).

These regulations have been established and implemented because TRU waste radioactivity typically occurs in the form of α-particles and γ-rays, although β-particles and neutrons also contribute to the total activity. A sheet of paper, the dead outer layer of the skin, or several inches of air can stop α-particles; however, α-contaminated material is harmful if sufficient quantities are swallowed or inhaled, a risk that diminishes once waste composed of the primary α-emitting TRU radionuclides (americium-241 [Am-241], Pu-238, Pu-239, and Pu-240) is packaged in approved containers.

Gamma radiation often occurs when α- or β-particles are released during the fission of U-235, followed by the subsequent decay of fission product radionuclides such as cesium-137/metastable barium-137 (Cs-137/Ba-137m). Gamma radiation is highly penetrating, requiring steel, lead, or concrete to shield workers from its hazardous effects.

CH TRU waste contains mostly α- and very little γ-radiation. Protective measures combined with α-radiation's poor penetration ability keep workers safe while characterizing CH TRU waste. Because the waste containers then block this α-radiation, the primary radiological risk to workers transporting and disposing of this waste arises from γ-radiation penetrating the container.

RH TRU waste, on the other hand, produces a larger quantity of γ-rays and is therefore associated with greater health risks for workers. However, DOE procedures call for minimal handling of this waste for the purpose of waste characterization, relying instead on AK. Workers transporting and disposing of RH TRU waste are prohibited from coming into direct contact with the waste containers, reducing their chances of encountering the γ-rays that penetrate the surface. Moreover, γ-emitting RH TRU radionuclides are relatively short-lived, decaying after about 300 years to a level comparable to that of longer-lived, α-dominated CH TRU waste.

Current Radiological Waste Characterization Requirements

Radiological waste characterization is a requirement for ensuring the potential for worker exposure and criticality accidents is reduced to as low a level as possible. To this end, DOE performs NDA on some RH TRU waste containers and on 100 percent of CH TRU waste containers. These tests are part of the process of obtaining/calculating several radiological parameters which include the following: quantity and activity of radionuclides present; Pu-239 fissile gram equivalent (FGE); Pu-239 Equivalent Activity (PE-Ci); the surface dose rate; and the decay heat for each container. Each of these quantities has a basis in regulations for either transporting or disposing TRU waste.

For example, to keep the amount of fissile material in a container below a point that might induce criticality, the FGE must be determined. Even though Pu-239 is the main source of fissile material in WIPP-bound waste, other radionuclides are also fissile sources. Therefore, to calculate the total FGE for each container, these non-Pu-239 sources are reported in terms of their Pu-239 FGEs. In adhering to container-based regulatory limits, the probability of a criticality accident is reduced to less than one chance in one million per year.

Furthermore, in their respective regulations (CFRs), DOT and NRC have provided safe packaging and transportation limits for over 380 radionuclides shipped in Type A packages (such as 55-gallon drums,

standard waste boxes, and 10-drum overpacks that are routinely used for TRU waste).[4, 5] By determining the main radioactive components of the waste in each drum, the remaining radionuclides can be deduced and the activity kept within packaging and transportation limits.

PECOS has prepared a summary of the primary parameters of interest for WIPP-bound TRU waste, as presented in *Table 1, Primary Radiological Waste Characterization Requirements as Summarized from Regulatory Documents*. The information contained in this table has been selected from various sources, including 40 CFR 191 and 194; 49 CFR 173; 10 CFR 71; DOE Order 435.1; LWA, as amended; Atomic Energy Act of 1954, as amended; TRUPACT-II, HalfPACT and RH TRU 72-B Certificates of Compliance; CH/RH Technical Safety Requirements (TSR); CH-TRAMPAC; and RH-TRAMPAC.

Table 1. **Primary Radiological Waste Characterization Requirements as Summarized from Regulatory Documents.**

PARAMETER OF INTEREST	WHY IT IS REQUIRED	HOW IT IS OBTAINED
Dose rates for Type A packages (disposal containers) and Type B packages (TRUPACT-II, HalfPACT, RH TRU 72-B, etc.)	Determines if disposal containers have TRU waste that is RH or CH in nature via the 200 mrem/hr limit.[1, 2, 11] Used to determine if surface dose rate of disposal containers exceeds 1,000 rem/hr limit for RH TRU waste.[11] Ensures radiation levels are not exceeded at different distances from the disposal and/or transportation containers, ensuring safe waste transportation from generator sites to WIPP.[5, 4, 12]	An approved instrument with a nationally traceable calibration standard should not register more than 200 mrem/hr at the surface of the package. The surface dose rate of a package can be up to 1,000 mrem/hr, as long as an approved instrument measures less than 200 mrem/hr at the outer surface of the transport vehicle, 10 mrem/hr two meters away from the lateral surface of the vehicle, and 2 mrem/hr in the cab. For Type B packages in an accident condition, an approved instrument held one meter from the surface of the package should not register a dose rate of more than 1,000 mrem/hr. The CH-TRAMPAC states that surface dose rate limits are lower for S100 and S300 pipe overpacks.
TRU α activity	Determines TRU α activity concentration (see below).	AK supported by NDA measurements for one or two key CH TRU radionuclides plus supplemental correlations/computations to identify the remaining α-emitting radionuclides and their activity. AK for RH TRU waste plus NDA measurements if AK is deemed insufficient.
TRU α activity concentration	Determines if waste has more than the minimum 100 nCi/g of activity related to α-emitting TRU radionuclides.[1, 2, 3, 11, 13]	Divide the TRU α activity by the mass of the waste in a disposal container (mass of the filled container minus the mass of the empty container).

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PARAMETER OF INTEREST	WHY IT IS REQUIRED	HOW IT IS OBTAINED
Isotopic composition of the waste	Determines Pu-239 FGE and decay heat (see below).	AK supported by NDA measurements for one or two key CH TRU radionuclides plus supplemental computations to identify remaining α-emitting radionuclides and their activity. AK for RH TRU waste plus NDA measurements if AK is deemed insufficient.
Radionuclide activities (and masses) of the waste	Determines FGE and decay heat (see below). DOE Order 435.1 states this is one of the minimum waste characterization components needed for waste certification.	AK and/or direct NDA/DA measurements of one or two key radionuclides plus additional correlations/calculations to determine the quantity of remaining radionuclides.
Decay heat per disposal container	Determines that decay heat is less than 40 watts (TRUPACT-II), 30 watts (HalfPACT), 50/300 watts (organic and inorganic waste) and additional limits stated in the RH-TRAMPAC (RH TRU 72-B).[17, 18, 19]	After determining isotopic composition and radionuclide quantities for a disposal container, a look-up table is used to acquire the decay heat for each radionuclide. Individual decay heats are summed to get the value for the container. Calorimetry can be used for containers less than 50 liters in volume.
Pu-239 FGE per disposal container	Determines that FGE limits are not exceeded, ensuring safe waste transportation from generator sites to WIPP.[12, 14, 15, 16, 17, 18, 19] The RH/CH TSR states this regulation exists to "protect assumptions for Nuclear Criticality Safety Evaluations that show criticality in transport containers is not credible."[18, 19]	After determining the isotopic composition and radionuclide quantities for a disposal container, a look-up table is used to acquire the FGE for each radionuclide (not all radionuclides contribute to the FGE). Individual FGEs are summed and total measurement uncertainties are factored to arrive at the container's FGE.
Pu-239 Equivalent Activity (PE-Ci)	Determines it is safe to transport waste from generator sites to WIPP.[12, 14, 15, 16, 17] Also mandated by the CH/RH TSR to "protect basic inventory assumptions."[18, 19]	Calculated for CH waste in S100, S200 and S300 pipe overpacks and RH waste (transported in an RH TRU 72-B) using the activity of TRU radionuclides (including U-233) and a nuclide-specific worker safety weighting factor (some of which are included in *Appendix B* of Revision 6.3 of the WIPP WAC; the remainder can be calculated).

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PARAMETER OF INTEREST	WHY IT IS REQUIRED	HOW IT IS OBTAINED
Activity for Type A packaging (disposal containers) and Type B containers.	Determines activity for individual radionuclides in each disposal container is below the regulatory limit, ensuring safe waste transportation from generator sites to WIPP.[4, 5] The CH TRAMPAC states that a TRUPACT-II or a HalfPACT cannot exceed 406 Ci if they are transporting S100 and S300 overpacks. When transporting S200 overpacks, there are radionuclide-dependent limits (and activity plus error $\leq 10^5$ A_2 per 10 CFR 71). A_2 is the maximum activity of radioactive material (i.e., TRU waste) permitted in Type A packages).[5]	Once the radionuclide activities are known for each disposal container, they are compared to the limits in 49 CFR 173.435 and 436 (or 10 CFR 71).
Activities (and masses) of the 10 EPA-mandated radionuclides	Determines compliance with EPA regulations for radioactive waste disposal.[1, 2]	AK and/or direct measurements of one or two key radionuclides plus additional calculations to determine the quantity of remaining radionuclides.
Activities and masses of additional radionuclides contributing to 95 percent of the radioactive payload hazard	Ensures safe waste transportation from generator sites to WIPP.[4]	AK and/or direct measurements of one or two key radionuclides plus additional calculations to determine the quantity of remaining radionuclides.
RH canister activity concentration	Determines if total activity is less than the 23 Ci/liter limit.[11]	AK for the waste stream is examined to determine if the limit could be reached. If so, DTC or NDA/DA can be used to determine total activity for representative containers for that waste stream.
Activity of all radionuclides impacting repository limits	Determines compliance with EPA certification.[1, 2]	AK, DTC, NDA/DA can be combined with modeling packages to determine activities of radionuclides and their daughters.
Activity of all radionuclides impacting the PA	Determines compliance with EPA certification.[1, 2]	AK and NDA/DA can be combined with modeling packages to determine the activities of radionuclides and their daughters.

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While it is imperative to adequately characterize waste bound for WIPP, DOE must also factor in project costs and operational considerations when determining the best course of action for its CH and RH characterization plans. For CH TRU waste, radiological characterization methods encompass both AK and 100-percent confirmatory NDA.[6, 7, 20] However, for RH TRU waste, DOE demonstrated the appropriateness of using AK as the main characterization method, with confirmatory NDA conducted only when necessary.[7, 21] Regulatory agencies accepted this approach because it precluded workers' exposure to the higher levels of radioactivity inherent in RH TRU waste. Moreover, the majority of RH TRU waste bound for WIPP has either not yet been generated or will require re-packaging, allowing DOE the opportunity to secure exceedingly accurate AK records for this type of waste.[7]

Several DOE orders, CFRs, and generator site/WIPP documents contain regulations and guidance designed to protect workers, the general public, and the environment from radiation exposure now and 10,000 years into the future. For example, the WIPP WAC[22] provides the criteria used for waste acceptance at WIPP by incorporating several sources of regulations and procedures. With respect to radiological waste characterization, the WAC references the CH TRU Waste Authorized Methods for Payload Control (CH-TRAMPAC) and RH-TRAMPAC for requirements regarding nuclear properties of the waste. The CH-TRAMPAC in turn, refers to the CH TRU Payload Appendices for specifics regarding NDA. CH TRU Payload Appendices Section 5.2 does contain this information, though it is in essence, a scaled-down version of an August 1991 paper by Schultz and Caldwell of Oak Ridge National Laboratory (ORNL).

In reviewing these documents, PECOS has determined that only a few radiological parameters directly comprise the waste characterization process at generator and storage sites across the DOE complex. The remaining parameters are deduced from AK and/or NDA measurements combined with knowledge about isotopic ratios and software capable of extrapolating information pertaining to container contents.

Waste Characterization Methods

The characterization process used to determine physical, chemical, and radiological parameters of CH TRU waste is far more rigorous than that used for RH TRU waste. According to reports[6, 7] by the National Research Council, this discrepancy is not related to any health or safety matters. DOE simply wanted to "maintain an effective working relationship" with the New Mexico Environment Department (NMED) at the outset of the disposal mission at the WIPP. Thus, when the first WIPP WAC was generated in the 1980s, DOE opted for a more conservative approach and specified characterization activities that exceeded state and federal requirements.[6, 7]

Characterization methods for RH TRU waste were not approved at the same time as were the original plans for CH TRU waste. While this delayed RH TRU waste emplacement, it ultimately allowed DOE to propose an RH TRU waste characterization program that unlike the CH TRU waste program, was not assay-dominated. As a result, AK may be found at the core of RH TRU waste characterization, and according to 40 CFR 194.22, the RH TRU characterization program allows DOE to complete/validate data using one of several methods: confirmatory testing, corroborating data, peer review, or a determination of quality assurance program equivalency. Despite this latitude offered by EPA, DOE seems to rely heavily on confirmatory testing.[6]

Confirmatory testing for radiological parameters includes surface dose rate measurements, DTC conversion, DA and NDA methods, and modeling derived from sampling. Often, a combination of NDA methods such as γ-ray spectrometry and passive neutron coincidence counting is used to calculate the quantity of radionuclides in waste destined for the WIPP. The primary confirmatory testing methods [12, 17, 21, 22] are discussed below.

1) Surface Dose Rate Measurements

The dose rate at the surface of each container is determined using a calibrated instrument traceable to the National Institute of Standards and Technology (NIST). The amount of β, γ, and neutron radiation at any point at the container surface cannot exceed regulatory limits in the LWA and CFRs (α radiation is stopped by the container and therefore not measured).

2) Dose-to-Curie Conversion

After the surface dose rate is known, the amount of radioactivity in a waste container is determined using a combination of AK and computer modeling based on known isotopic decay schemes. Assuming that the radioactive source material is the same across the waste stream, empirically-derived conversion factors are used to relate the quantity of γ-emitting radionuclides in the waste (such as Cs-137) to the quantity of other radionuclides on a container-by-container basis.

3) Destructive Assay Methods

DA methods such as radiochemical assay are used when it is necessary to use a chemical analysis to determine radiological parameters of irradiated fuel or non-solid waste forms like sludge. With sludge, for example, waste is first homogenized, and samples are drawn, prepared, and analyzed using α-or γ-spectrometry, as described in the next section. Results can help deduce the Pu and Am content for drums of de-watered or solidified sludge after packaging.

4) Nondestructive Assay Methods

- Gamma spectrometry is used to determine the isotopic composition of waste inside a container, including γ-emitting isotopes of U, Pu, neptunium (Np), and curium (Cm). However, Pu-242 cannot be measured directly because it has no useful γ emissions. Analyses are performed using American Society for Testing and Materials (ASTM) C1030 or an equivalent method.
- Mass spectrometry (MS) is used to determine isotopic composition for the Pu-239 FGE for CH containers and reveals what isotopes are in the waste, though not their quantities. Testers perform analyses using ASTM C696, ASTM C697, and ASTM C759, or an equivalent method.
- Alpha spectrometry has limited use in determining radiological characteristics of TRU waste. When MS does not provide reliable results because the amount of Pu-238 in a sample is too small, α-spectrometry can determine the isotopic abundance of this particular radionuclide. These analyses are performed using ASTM C1415 or an equivalent method.
- The calorimetric method is a form of NDA that can directly measure the total decay heat of a package. Using the measured value, the mass of radionuclides in the package can be determined if the isotopic composition is also known. This is especially useful when assaying weapons-grade Pu (Pu-239) whose heat release comes from α-and β-decay. This technique provides one of the most effective means for determining the quantity of Pu and Am-241 in a container, although it can take 20 minutes to 24 hours to collect the requisite data. In addition to long data acquisition times, this method is not suitable for 55-gallon (208-liter) drums. Instead, it accommodates packages with a volume typically less than 50 liters. Analyses are performed using ASTM C1458 or an equivalent method.
- Passive γ measurements are used for larger containers (such as 55-gallon drums) to determine the quantity of radionuclides present in the waste. Segmented gamma scanning (SGS) and segmented passive gamma scanning devices can detect U-233, Pu-238, Pu-239, Np-237, Am-241, and Am-243 because at least one high-energy γ-ray is emitted from the waste container with enough intensity that quantities of nuclides emitting low-energy γ-rays can be estimated when the isotopic composition is known. Analyses are performed using ASTM C1133 or an equivalent method.
- Passive neutron coincidence counting provides results quickly when the relative abundance of Pu-238, Pu-240, and Pu-242 is known and the total Pu mass must be determined. Testers perform analyses using ASTM C1207 or an equivalent method.
- Passive-Active Neutron (PAN) assay can quantify nuclei that are fissile (U-233, U-235, Pu-239 and Pu-241) or capable of spontaneous fission (Pu-238, Pu-240, Pu-242, Cm-244, Cm-248, and californium-252 [Cf-252]), although only one fissile radionuclide is actually measured. Because

the relative abundance of Pu or U must first be known, the total Pu or U mass can be determined following a 10-minute assay. The use of PAN can also segregate LLW and TRU waste at the 100 nCi/g concentration cutoff limit. Analyses are performed using ASTM C1493 or an equivalent method.

Radionuclides of Interest

DOE uses the data derived from the radiological characterization activities discussed above to build the TRU waste radionuclide inventory, to verify that TRU waste is inside the containers permanently disposed of at WIPP, and to confirm that these containers adhere to transportation and repository limits. However, only a few key radionuclides can be measured directly through NDA. Extrapolation with "Oak Ridge Isotope GENeration" (ORIGEN) — a computer code ORNL created in the 1970s — must determine the remainder.[23] ORIGEN benefited from updates in 1991 (ORIGEN2 version 2.1) and again in 2002 (ORIGEN2 version 2.2). The 2002 release included substantial improvements to the number of radionuclides (increased from 418 to 2,101) and photon lines (increased from 12,000 to 115,000) referenced by the software and serves as the basis for ORIGEN-ARP (Automatic Rapid Processing), the version developed for modern computing systems.[24]

Using radiological parameters provided by AK and confirmatory testing, ORIGEN simulates how a given amount of a parent radionuclide will decay/transform over time, enabling DOE to determine the content and characteristics of its radioactive waste. DOE then uses the data for specific radionuclides from the data generated by ORIGEN to prove that WIPP-bound waste meets all regulatory requirements.

Packaging, transportation, and disposal regulations have driven the selection of these radionuclides of interest. One set of radionuclides was chosen to insure that public and worker health and safety are not jeopardized during TRU waste packaging and transportation. Therefore, radionuclides that contribute to the Pu-239 FGE, decay heat, and waste activity (including the PE-Ci determination) are included in this grouping.[22] According to both the CH and RH TRAMPAC, 17 radionuclides can contribute to the FGE. Decay heat, on the other hand, might come from any of more than 200 radionuclides listed in these two documents. (The extent of the contribution depends on the amount of each radionuclide present in the waste container.) Numerous radionuclides can also impact the calculation for activity compliance stated in 49 CFR 173 and 10 CFR 71, although only 13 radionuclides are listed as main contributors to the PE-Ci calculation according to Appendix B ("Pu-239 Equivalent Activity") of the WIPP WAC.

A second set of radionuclides was selected to ensure compliance with TRU waste disposal regulations.[22] Foremost in this group are the 10 EPA-mandated radionuclides agreed upon by DOE and EPA during the Compliance Certification Application (CCA) process in 1996. These radionuclides represent the most

significant sources of potential environmental contamination if released from the repository, and are not limited to TRU radionuclides (strontium-90 [Sr-90] and Cs-137 are prime examples). In addition, 40 CFR 191 stipulates that containment release limits exist for many additional radionuclides.

In *Table 2, Activity for EPA-mandated Radionuclides as Reported by DOE in TRU Waste Inventory Reports for 1995 to 2007*, compiled by PECOS, data for the EPA-mandated radionuclides are presented in the most recent inventory reports beginning with the TRU Waste Baseline Inventory Report (TWBIR) from 1996 through the 2008 Annual TRU Waste Inventory Report (ATWIR).

Table 2. **Activity (decayed to the inventory cut-off date) for EPA-mandated radionuclides as reported by DOE in TRU waste inventory reports for 1995 to 2007.**
*(** indicates a TRU radionuclide)*

RADIONUCLIDE (and Half-Life)	CH AND RH ACTIVITY (Ci) IN TWBIR REV 3 (Data for 19 Sites as of 12/31/1995)		CH AND RH ACTIVITY (Ci) IN TWBIR-2004 (Data for 21 Sites as of 12/31/2001)		CH AND RH ACTIVITY (Ci) IN ATWIR-2007 (Data for 17 Sites as of 12/31/2006)		CH AND RH ACTIVITY (Ci) IN ATWIR-2008 (Data for 18 Sites as of 12/31/2007)	
Sr-90 (28.9 yr)	2,220	43,600	55,000	340,000	1,720	642,000	482	83,000
Cs-137 (30.08 yr)	3,120	18,900	5,500	440,000	7,640	739,000	470	101,000
Pu-238 (87.7 yr)**	756,000	169	1,300,000	3,900	1,480,000	7,730	809,000	3,580
Am-241 (432.6 yr)**	240,000	482	430,000	14,000	393,000	20,700	431,000	3,910
Pu-240 (6,561 yr)**	68,800	179	86,000	1,600	112,000	707	138,000	863
Pu-239 (24,110 yr)**	351,000	559	540,000	5,300	475,000	2,870	489,000	2,300
U-233 (159,200 yr)	1,200	436	1,000	150	625	440	111	32
U-234 (245,500 yr)	107	12	170	32	157	33	98	2.69
Pu-242 (375,000 yr)**	493	0.01	10	0.49	39	0.69	70	1.18
U-238 (4,468,000,000 yr)	6.08	0.12	61	140	57	56	26	0.13
Calculated Top 10 Activity at Inventory Cut-Off (CH, RH and Total)	1,422,946	64,337	2,417,741	805,122	2,470,238	1,413,536	1,868,257	194,689
	1,487,283		3,222,863		3,883,775		2,062,946	
Reported Total Inventory Activity at Cut-Off	2,649,000		6,000,000		7,330,000		3,949,000	
Percent of Total Inventory Activity Represented	56.15%		53.71%		52.98%		52.24%	

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In order to prove that WIPP has a reasonable expectation of achieving long-term containment of disposed waste, Sandia National Laboratories conducts a PA for DOE, the results of which are submitted to EPA as a part of the compliance certification/recertification application. For the CCA, more than 65 radionuclides were included.[25] For the Compliance Recertification Application (CRA) submitted for 2004, the number was reduced to 29.[26] This remains unchanged in the CRA-2009.[27]

Finally, the LWA requires that the total activity of RH TRU waste at the time of disposal does not exceed 5,100,000 Ci. This, therefore, necessitates tracking radionuclides in RH waste canisters that contribute to α-, β-, γ- and neutron radiation levels in the repository. Conversely, there is no similar disposal requirement for CH TRU waste.[11]

As summarized by PECOS in *Table 3,* ATWIR-*2008 Radionuclides with More than 100 Ci of Combined CH and RH Activity as of December 31, 2007*, the TRU waste inventory consequently accounts for not only the 10 EPA-mandated radionuclides, but also decay products and any other radionuclides contributing to the decay heat or activity of waste transported to and placed inside the repository. In the years since the TWBIR was submitted for the CCA[28], the net number of radionuclides has grown from 135 to 155 (27 were added and seven were removed) now reported in the ATWIR-2008.[29]

Table 3. **ATWIR-2008 Radionuclides with more than 100 Ci of Combined CH and RH Activity as of December 31, 2007. (** indicates a TRU radionuclide)**

NUCLIDE INFORMATION			EPA CERTIFICATION INFORMATION				TRANSPORTATION INFORMATION			ATWIR-2008 INFORMATION	
Nuclides reported in the ATWIR-2008	Primary Decay Mode	Half-life	EPA Top-10 Nuclide?	Used in the CCA PA?	Used in the CRA-2004 PA/PABC?	Used in the CRA-2009 PA/PABC?	Used to Determine Decay Heat?	Used to Determine Pu-FGE?	Used to Determine PE-Ci?	Contributes to 99% of Activity?	Nuclide Total Activity (Ci)
Pu-241	β-	14.290 y		✓	✓	✓	✓	✓	✓	✓	1,702,900
Pu-238**	α	87.7 y	✓	✓	✓	✓	✓	✓	✓	✓	812,580
Pu-239**	α	24,110 y	✓	✓	✓	✓	✓	✓	✓	✓	491,300
Am-241**	α	432.6 y	✓	✓	✓	✓	✓	✓	✓	✓	434,910
Pu-240**	α	6,561 y	✓	✓	✓	✓	✓	✓	✓	✓	138,863
Cs-137	β-	30.08 y	✓	✓	✓	✓	✓			✓	101,470
Ba-137m	Isomeric transition	2.55 m		✓			✓			✓	94,935
Sr-90	β-	28.90 y	✓	✓	✓	✓	✓			✓	83,482
Y-90	β-	2.67 d		✓			✓				82,455
H-3	β-	12.32 y					✓				4,550
Cm-244	α	18.1 y		✓	✓	✓	✓	✓	✓		2,969
Pm-147	β-	2.6234 y		✓	✓	✓	✓				289
Co-60	β-	5.27 y					✓				286
Eu-155	β-	4.753 y					✓				273
Eu-154	β-	8.593 y					✓				249
Kr-85	β-	10.73 y					✓				219
Cs-134	β-	2.0652 y					✓				168
U-233	α	159,200 y	✓	✓	✓	✓	✓	✓	✓		143
U-234	α	245,500 y	✓	✓	✓	✓	✓				101

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IV. CONCLUSIONS

After reviewing the numerous regulatory bases of TRU waste characterization, PECOS draws the following conclusions regarding current DOE programs for CH and RH radiological waste characterization, data tracking, and reporting:

- The requirement of 100 percent NDA for CH TRU waste does not appear to be grounded in either a scientific or a health and safety foundation. Moreover, no similar requirement exists for RH TRU waste.
- A key piece of documentation about NDA methods used in characterizing CH TRU waste, namely Appendix 5.2 of the CH TRU Payload Appendices, is out of date. Corresponding NDA documentation for RH TRU waste is nonexistent.
- Of the 10 radionuclides mandated by EPA for long-term regulatory compliance, only six have substantial impact on overall activity in the repository.
- While a fairly extensive accounting of radionuclides exists in the TRU waste inventory database, it is unnecessary to list most radionuclides in the inventory reports published by DOE because they, in total, contribute insignificantly to repository radionuclide activity and to regulatory limits.

CH Versus RH TRU Waste Radiological Characterization Activities

With respect to health and safety aspects of radiological waste characterization, while DOE has implemented programs for both CH and RH TRU waste that are protective of workers, there is a major difference noted between the two programs: the CH program contains a 100-percent confirmatory NDA requirement. However, as DOE has established with the RH program, NDA of all waste containers is unnecessary.

An approach similar to the one implemented for the RH characterization program could be used for CH TRU waste, especially when one considers the LWA only requires reporting the total activity of RH TRU waste, not CH TRU waste. One would still generate information appropriate to run the PA by performing NDA on a statistically valid sampling of CH TRU waste containers. At the same time, DOE could reduce project costs while maintaining quality control/quality assurance objectives and minimizing risks to worker health and safety.

NDA Documentation

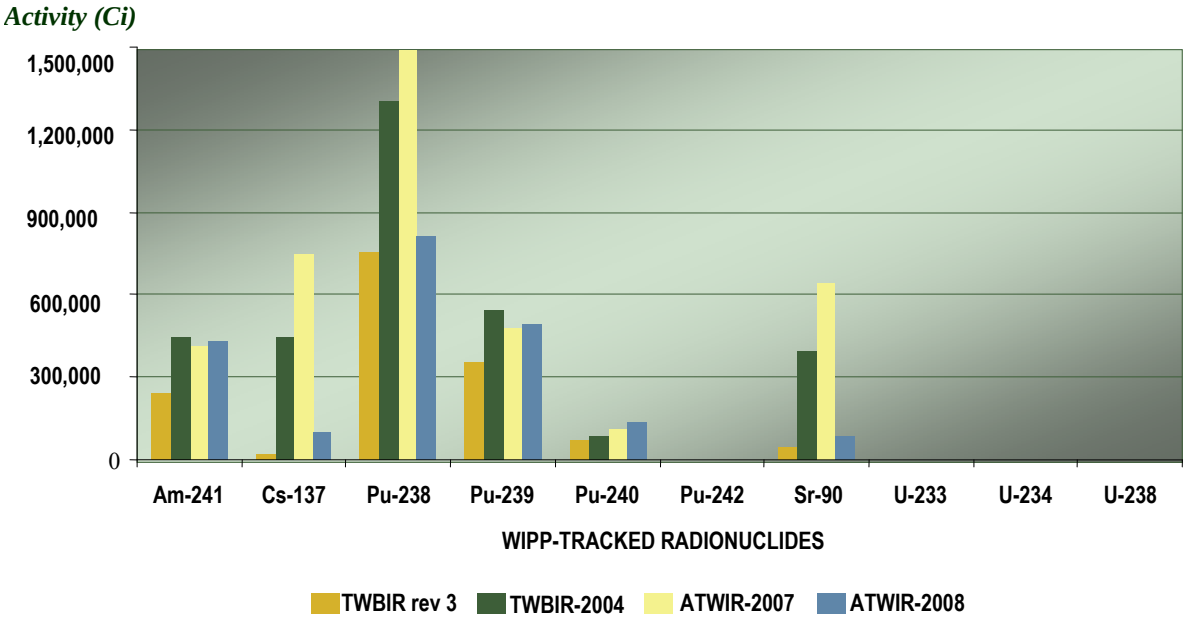
Even though Section 5.2 of the CH TRU Payload Appendices contains information concerning radiological waste characterization, this section is no more than a scaled-down version of the August 1991

paper authored by Schultz and Caldwell. As such, it cannot fully address DOE's existing characterization methods because NDA hardware, software, and analysis methods have evolved during the intervening 18 years. This technological progression has been demonstrated not only in numerous papers published by personnel at generator sites and the WIPP, but also in audit reports for waste characterization activities at TRU waste generator sites. Beyond this, there is no similar section in the RH TRU Payload Appendices, despite the fact NDA is also performed on RH TRU waste.

Radionuclides Used to Determine Compliance with EPA Certification

EPA mandated that DOE track the activity of 10 specific radionuclides as part of the original WIPP certification process. Data collected thus far shows that six isotopes have made significant contributions, and that the longest-lived radionuclides, U-233, U-234, U-238 and Pu-242, exhibit little activity (See *Figure 1,* below, and *Table 2*). In fact, according to the ATWIR-2008, these four radionuclides account for only 342 Ci of combined CH and RH TRU activity as of the inventory cutoff date. This number represents less than 0.017 percent of activity for the 10 EPA radionuclides, which themselves account for less than 53 percent of total inventory activity.

Figure 1. **Activity of EPA-Mandated Radionuclides as Reported in the Inventory for 1995, 2001, 2006, and 2007.**



In addition to these 10 radionuclides, EPA is concerned with WIPP's ability to maintain radioactive waste containment over the long term. To support this effort, only CH TRU waste data is included in the PA. RH TRU waste is insignificant to the PA because it contains radionuclides that substantially decay within 300 years of WIPP closure (well short of EPA's 10,000 year compliance period) and that will account for

no more than four percent of WIPP waste volume and less than 15 percent of the radioactivity (via restrictions in the LWA). While CH TRU waste is a factor for potential long-term radiological impacts on the environment, performing NDA on every container is excessive. Rather, completing NDA on a representative sample from each waste stream would provide adequate data for modeling something as inherently uncertain as repository performance 10,000 years into the future.

Radionuclides in Inventory Reports

A review of the ATWIR-2008 shows DOE tracks 155 radionuclides. Having these radionuclides in the TRU waste inventory database is understandable from both a scientific and a regulatory perspective. Specifically, CH TRU waste radionuclides are included in the inventory because they account for most of the activity (10 times more radioactivity than RH TRU waste); and they also impact the PA. The LWA, on the other hand, states that the radioactivity of RH TRU waste must be tracked to ensure it does not exceed 5,100,000 Ci. Additionally, 149 of the roughly 380 radionuclides evident in packaging and transportation-related tables in 10 CFR 71 and 49 CFR 173 are applicable to TRU waste and are contained in the inventory database.

Acquisition of this extensive amount of data is only possible through the use of ORIGEN in conjunction with AK and supplemental radiological characterization data. Equipped with a few key pieces of radiological information, ORIGEN automatically determines the radionuclides that should be present in the waste. The ATWIR-2008 consequently reports these numerous radionuclides, even if they make insignificant contributions to container/repository decay heat and activity.

It also appears that ORIGEN2 version 2.2 is the source of the 27 radionuclides that have been introduced to inventory reports since the CRA-2004. Altogether, these 27 add 1.89 Ci of activity. The decay heat is increased by 5.15 milliwatts because 21 of 27 nuclides are used in that particular calculation. One of the 27 is a TRU nuclide. None are used to determine Pu-239 FGE or PE-Ci, and none factor into the PA for the CRA-2004 or the CRA-2009.

Upon closer inspection of the most recent inventory report (ATWIR-2008), it contains 56 entries for radionuclides with a combined CH and RH activity of less than 1 Ci across the entire DOE complex, as summarized by PECOS in *Table 4,* below. Of these, 23 (over 41 percent) are the newly reported radionuclides mentioned above. Moreover, when looking at contributions that are each less than 1 Ci of activity, the list expands to 72 radionuclides, adding a mere 8.24 Ci of activity to the WIPP as a whole.

In fact, the first eight radionuclides account for over 99 percent of the repository activity; this subset also includes the six most active EPA-mandated radionuclides mentioned earlier. When comparing the radionuclides needed for EPA-driven regulations with those necessary to meet transportation

requirements, *Table 4* clearly shows the extent of DOE's profusion of irrelevant radionuclide information that is published in its inventory reports.

PECOS noted that DOE has eliminated radionuclides from inventory reports in the past. Seven nuclides listed in the inventory report for the CCA were not included in any subsequent report. One nuclide added to the ATWIR-2007 was deleted the following year in the ATWIR-2008. Thus, DOE's need to present data for numerous radionuclides contributing small levels of radioactivity and decay heat remains unclear.

Table 4. **Regulatory Applicability of Radionuclides in the ATWIR-2008.**

	BASIC INVENTORY INFORMATION			EPA CERTIFICATION			PACKAGING & TRANSPORTATION		
	Number of Nuclides	Number of TRU Nuclides	Total Activity (Ci) at Inventory Cut-Off	Number of Top 10 EPA Nuclides	Number of Nuclides Added Since CCA Inventory	Number of Nuclides Used in CRA-2009 PA	Number of Nuclides Used to Determine Decay Heat	Number of Nuclides Used to Determine Pu-239 FGE	Number of Nuclides Used to Determine PE-Ci
Entire ATWIR-2008	**155**	**17**	**3,953,181**	**10**	**27**	**29**	**149**	**17**	**13**
Top 99% of ATWIR-2008 Activity	**8**	**4**	**3,860,440**	**6**	**0**	**7**	**8**	**5**	**5**
Nuclides with > 100 Ci of Activity	**19**	**4**	**3,952,142**	**8**	**0**	**11**	**19**	**7**	**7**
Nuclides with < 1 Ci of Activity	**72**	**6**	**8.24**	**0**	**27**	**8**	**56**	**3**	**2**
Nuclides Totaling < 1 Ci of Activity	**56**	**6**	**0.92**	**0**	**23**	**5**	**50**	**3**	**1**

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V. RECOMMENDATIONS

PECOS recommends the following programmatic changes:

- Because the requirement of 100 percent NDA for CH TRU waste does not appear to have a scientific or a health and safety basis, PECOS urges DOE to work with the appropriate regulators to reduce this requirement and perform only a statistically valid sampling. This potential cost-saving measure would maintain good quality control and quality assurance procedures while not adversely impacting worker health and safety.

- DOE should update the CH assay methods in Appendix 5.2 of the CH TRU Payload Appendices and include a similar section for RH NDA methods, which currently are not detailed in any DOE documents.
- DOE should use the opportunity afforded by the current EPA recertification process to eliminate the requirement to track and report activity for U-233, U-234, U-238 and Pu-242. When examining *Table 3* of this report, it is also apparent there are no significantly active, long-lived radionuclides that would suffice as replacements for these four nuclides.
- While a fairly extensive accounting of radionuclides exists in the TRU waste inventory database, PECOS believes it is unnecessary to include most of these radionuclides in the inventory reports published by DOE. Instead, DOE should list only the most significant radionuclides, perhaps those contributing to the top 99 percent of the repository activity (the eight most active radionuclides).

  This adjustment will result in continued streamlining of the inventory report, a process initiated with the ATWIR-2008. Moreover, stakeholder confidence could increase if these eight nuclides were shown in an ATWIR Main Body table depicting the level of radioactivity at the inventory cut-off date *and* at the WIPP closure in 2033.

  If DOE opts to preserve the existing tables of data for the 155 radionuclides, the information might be better presented as an Appendix to the ATWIR, or perhaps included in the supplemental inventory report produced to support the PA (a new document mentioned in the ATWIR-2008).

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