TRU-ly Clean--What Does It Mean? -8286

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ABSTRACT

The evolution and genesis of the definition of transuranic waste (known as TRU) and its application to the cleanup criteria applied to soils contaminated with transuranics, specifically plutonium, has been a matter of discussion at contaminated sites in the United States and elsewhere. Cleanup decisions and the processes that led up to those decisions have varied at several plutonium contaminated sites within the United States and without the pacific region. The sites with radionuclide soil action levels include Bikini and Enewetak Atolls, Republic of the Marshall Islands; Johnston Atoll, Hawaii; the Hanford Site in Washington State; the Nevada Test Site; the Rocky Flats Environmental Technology Site in Colorado; the Chariot Site in north Alaska; and the Maralinga Site in Australia.

The soil-action level developed for Rocky Flats by the U.S. Department of Energy, U.S. Environmental Protection Agency, and the Colorado Department of Public Health and Environment for plutonium is one of the higher soil-action levels approved by regulatory agencies that is considered protective for future use of land at a cleanup site. The Republic of the Marshall Islands has adopted a relatively conservative cleanup standard to accommodate the subsistence lifestyle of the islanders, while the Rocky Flats Environmental Technology Site has been transferred to the U.S. Department of the Interior to be used as a fish and wildlife refuge, a land use that resulted in a less conservative plutonium soil cleanup level.

INTRODUCTION

A transuranic element is defined by the Nuclear Regulatory Commission as an artificially made, radioactive element that has an atomic number higher than uranium in the periodic table of elements such as neptunium, plutonium, americium, and others. Transuranic waste or TRU waste is defined as material contaminated with transuranic elements that is produced primarily from reprocessing spent fuel, for the production of plutonium.

The definition of TRU was further developed as a result of a meeting of U. S. scientists. In early 1970, a meeting was held in a Stouffer's Inn near Denver's old Stapleton Field to discuss and establish a lower limit value for defining transuranic wastes that were accumulating at United States Atomic Energy Commission (AEC) sites involved in production and use of plutonium. The meeting was chaired by Dr. Wayne Bills, an AEC employee from the Idaho National Engineering Laboratory, and included participants from the AEC production sites. An outcome of the meeting was consensus that special

consideration should be given to wastes contaminated with trans-uranium elements if the level of radioactivity exceeded 10 nanocuries per gram of waste. That value was derived from comparison with the highest known levels of radium deposits in the earth's crust.

Later the definition of "transuranic radioactive waste" was codified in 40 CFR 191 of the United States code of federal regulations. The definition describes waste containing more that 100 nanocuries of alpha-emitting transuranic isotopes with half-lives greater than twenty years per gram of waste. High level radioactive waste is excluded from this definition by statute.

Scientists recognized the necessity for long lived radionuclides to be interred in geologic formations that could be reliably predicted to endure and to remain undisturbed until the radioactive elements of interest have decayed to innocuous forms. The bedded salts area located in southern New Mexico was eventually selected as a viable option and the Waste Isolation Pilot Plant was established at that location. Soil cleanup levels have been developed primarily considering exposure and dose.

The testing of plutonium weapons, aircraft and testing accidents and disposal or spillage of production wastes from the generation and fabrication or machining of plutonium has resulted in large scale contamination of several sites world-wide. Because plutonium is a very long-lived and toxic material, the clean-up level for plutonium in soils has received much discussion and debate.

Government Accounting Office investigators reported in a 2000 letter and report to Congress [1] that there is no uniform cleanup standard applicable to radioactive materials in the United States. The report describes discrepancies between Nuclear Regulatory Commission (NRC) and Environmental Protection Agency (EPA) regulations regarding exposure limits and regulatory cleanup levels as applied to decommissioning and cleanup sites where both agencies have jurisdiction. EPA's risk based approach sets a relatively restrictive risk goal of one in a million, or 10-6, that an individual will develop cancer in a lifetime as a remediation goal. NRC cleanup standards are dose-based and specific to identified radionuclides. Cleanup goals and standards have not only varied in the U.S.; they vary world-wide.

SUMMARY OF BASIS FOR STANDARDS APPLIED

The difference in cleanup approaches is based on applying standards that are risk based versus dose based. Other factors include designation of current and future land use and the uniform application of risk based exposure scenarios. The use of different computer models by different parties and agencies as well as differing in judgement over input parameters to the computer models leads to variable risk and dose assessments and therefore different soil cleanup standards.

Generally speaking, in the dose assessment methodology, dose is calculated by multiplying a dose conversion factor (unit dose/unit intake) for a specific radionuclide by the total intake/exposure to the radionuclide. The factor can then be multiplied by a

probability coefficient to arrive at a risk value. This is the effective dose equivalent approach and is the approach favored by the NRC and the DOE. EPA's preferred methodology for determining risk based cleanup levels is to use the cancer slope factor approach. In performing a risk assessment using the cancer slope factor approach, risk is calculated directly by assigning a unit of risk for every unit of exposure or probability of adverse effect in pico curies (pCi) and then multiplying by the total exposure.

NRC and DOE follow the effective dose equivalent approach in currently operating facilities. The NRC extended the dose approach to cover cleanup. NRC cleanup levels were derived using dose conversion factors to back-calculate radionuclide concentrations (activity/mass) corresponding to a target dose. EPA's approach derived from studying cancer-causing chemicals expressing future risks in terms of excess cancer probabilities. EPA extended this method to radionuclide contamination. As previously stated, the EPA CERCLA approach for risk assessment is the cancer slope factor approach although both the EPA and the NRC require exposures to be modeled. To do this, exposure scenarios are used as input parameters to computer models that are used to calculate risk or dose. Input parameters include pathway- external exposure, inhalation, direct ingestion of soil, ingestion of contaminated food and ingestion of drinking water and durations based on land use scenarios such as resident, farmer or visitor.

The dose assessment approach for establishing cleanup levels is based on an annual exposure and using the effective dose equivalent (EDE). For example, annual dose (inhalation pathway) = dose conversion factor (DCF) x radionuclide concentration in air x breathing rate x exposure duration. DCFs are set by the International Commission on Radiological Protection (ICRP) and are expressed in dose per unit measure.(see ICRP Publication 30 [2] or the new ICRP Publication 72 [3]) Each radionuclide has a unique DCF base on the type, strength, target and cancer induction rate.

Guidance for evaluating risks from exposure to radioactive substances to human health and the environment is found in *Risk Assessment Guidance for Superfund (RAGS): Part A (EPA, 1989)*[4]. This process is specified in the National Contingency Plan (NCP) and provides a framework for assessing baseline risks, developing preliminary remediation goals and evaluating risks associated with cleanup alternatives. In these instances, a cancer slope factor is used. A slope factor is similar to a dose conversion factor, but instead of assigning a unit dose for each unit of exposure, mrem/pCi, a unit of risk is assigned for every unit exposure or probability of adverse effect/pCi. The slope factor is an estimate of the probability of the individual developing cancer per exposure to a radionuclide carcinogen over a lifetime. For example, if the standard inhalation pathway is used for the risk scenario, the equation is: Risk for inhalation pathway=(inhalation slope factor)x(radionuclide concentration in air)x(breathing rate)x(exposure duration).

EPA has calculated slope factors for most radionuclides and provided them in *Health Effects Assessment Summary Tables (HEAST)* EPA, 2001b [5]. These are found in EPA's Preliminary Remediation Goals (PRG) electronic calculator. EPA guidance documents equate the 15-mrem annual dose to a 3x10E-4 risk (This is 15mrem/yrx30yrs x 7.3xE-2/sievert x E-5sievert/mrem=3xE-4) [6]

Computer Models, Input Parameters and Exposure Scenarios

Computer models are used to determine exposures at a site. Equations in the model sum the exposure from the ingestion, inhalation and external irradiation pathways. Each of these pathways has an intake, exposure period and a dose conversion factor or a cancer slope factor. Modifying factors can also impact the results. Computer models include the following: RESRAD for RESidual RADiation, which was developed by DOE and used at DOE sites, MMSOILS (EPA) and DandD (NRC). In addition, new *RESRAD-OFFSITE,* (DOE, August, 2007) calculates the potential radiological dose and corresponding excess lifetime cancer risk using the predicted radionuclide concentrations in the environment, and derives soil cleanup guidelines corresponding to a specified dose limit. Nine exposure pathways are available according to DOE.

The input parameters to the models have a significant influence on the cleanup level determined and are responsible for some of the variability seen in cleanup values at different sites. These include future land use, the exposure scenarios used and certain modifying factors based on special situations.

EPA RISK BASED CLEANUP STANDARDS AT CERCLA SITES

Several guidance documents state EPA's position regarding cleanup of radionuclides at CERCLA sites. EPA's 1997 memorandum on establishing cleanup levels at CERCLA sites states that the risk range for all carcinogens established in the NCP (10-4 to 10-6) should be used to establish cleanup levels when ARARs are not available or are deemed insufficiently protective. It further clarifies that the NRC limit of 25/100 mrem/yr dose level should not be used for cleanup level determinations at CERCLA sites. (*Establishment of Cleanup Levels for CERCLA Sites with Radioactive Contamination,* August 22, 1997, OSWER Directive 9200.4-18 [7] and *Clarification of the Role of Applicable, or Relevant and Appropriate Requirements in Establishing Preliminary Remediation Goals Under CERCLA,* August 22, 1997, OSWER Directive 9200.4-23.)[8]

Apparently there is enough confusion in the field on establishing certain cleanup practices that EPA published a memorandum requesting EPA Regional Offices to consult with Headquarters on CERCLA response decisions that may set precedents related to radionuclide cleanup. (see *Headquarters Consultation for Radioactively Contaminated Sites,* July 26, 2000, OSWER Directive 9200.1-33P)[9]

PLUTONIUM CLEAN-UP LEVELS AT SELECTED CLEAN-UP SITES

Bikini and Enewetak Atolls, Republic of the Marshall Islands

Atmospheric testing in the South Pacific by the United States resulted in large scale contamination of several atolls in the Marshall Islands including Enewetak. The Enewetak Atoll is a ring of 40 islands with a lagoon of twenty miles in diameter.

The cleanup of Enewetak was the first cleanup that targeted plutonium and the first time the U.S. attempted to set plutonium cleanup standards. A 1979 draft EIS to discuss further cleanup options was published in which soil with plutonium concentrations between 1.48 Bq and 14.8 Bq (40 and 400 pCi/g) would be considered on a case-by-case basis and all soil contaminated at levels above 14.8 Bq (400 pCi/g) would be removed. The cleanup during 1977-78 resulted in the 40pCi/g level for residential islands and 160 pCi for the rest. [6] On Bikini, the radionuclides of concern include cesium-137 and strontium-90, plutonium isotopes and americium-241. Although surface soils are contaminated with plutonium, the most important pathway for human exposure on the islands is ingestion of cesium-137 contained in locally grown food crop products such as coconuts.

The Republic of the Marshall Islands has adopted a cleanup standard of 0.15 mSv or 15 mrem per year above background.

The *combined option* adopted by the Rongelap Atoll Local Government (RALGOV) specifies the treatment of agricultural areas of the island with potassium fertilizer to reduce the uptake of cesium-137 into plants, and removal and replacement of contaminated surface soil around the village and housing areas. The same approach is recommended for Bikini Island. (LLNL web site: Marshall Islands Dose Assessment & Radioecology Project,)

Native islanders expressed concern about radiation exposure living on Enewetak and the safety of using the northern reaches of the lagoon for fishing and food gathering. In 2000 the DOE began construction of a lab with capability for whole body counting and a program for plutonium urinalysis. Islanders were trained to perform the whole body counting (See Figure 1.).

Figure 1. Enewetak, Marshall Islands: Radiological Laboratory and Lagoon

Johnston Atoll (Kalama Island), Hawaii

Johnston Atoll or Kalama Island is located between the islands forming the State of Hawaii and the Marshall Islands. The island has no inhabitants and has been used as a military reservation since 1941.

The plutonium contamination on Johnston Atoll, (aerial view, Figure 2, was caused by three separate accidents: two aborted rocket launches during atmospheric tests, and a rocket explosion on the launch pad. Pieces of plutonium metal "rained" onto the island from the former two accidents and were scattered across the island by the third. The plutonium contamination was further documented in an assessment of chemical and radionuclide contamination of Johnston performed in 1988 and 1989..(see Hopkins, *Independent Assessment, Johnston Atoll*.)

In the latter part of the 1980's, a dose based cleanup level of 13.5 pCi/g was determined based on EPA's draft guidance specifying a 10-mrem dose. The *Johnston Atoll Radiological Survey* (2000) describes how RESRAD 5.82 was used to determine cleanup goals. To determine cleanup goals for plutonium, four exposure scenarios were studied: a Fish and Wildlife worker, a cleanup worker, an ecotourist and an uninvited homesteader. The soil ingestion exposure pathway was most prominent in contributing dose.[6]

In September 2000, EPA Region 9 recommended the cleanup level of 13.5pCi/g, which represents a 7.1 x 10-6 cancer risk to an island resident.

Figure 2. Aerial of Johnston Island (Kalama Island) Hawaii, USA

Rocky Flats Environmental Technology Site (Rocky Flats), Colorado USA

Rocky Flats is located approximately sixteen miles outside of Denver, Colorado in the U.S. The Rocky Flats site was part of the nuclear weapons complex and was the site of the fabrication of nuclear weapons components from plutonium and uranium. Releases of radionuclide contaminants resulting from fires and accidents contaminated the site. The cleanup of Rocky Flats covered several years and the agreements associated with radionuclide soil action levels were the most contentious. The 1996 recommendation for plutonium cleanup levels for the wildlife refuge worker scenario was 780 pCi/g [6] adjusted to 651pCi/g. An independent panel, the Rocky Flats Soil Action Level Oversight Panel, recommended that the action level for plutonium be reduced to 35pCi/g stating that the most protective scenario is the subsistence farmer scenario. (*Independent Radionuclide Soil Action Level Evaluation at the Rocky Flats Environmental Technology Site*,http://www.racteam.com) The radionuclide soil action level was determined independently after analysis of available data. Uncertainties included, for example, the effect of prairie fires on the resuspension of material. The independent study resulted in a technically based soil action level for plutonium using a 15mrem per year dose limit of 35pCi/gram.[John E. Till, PhD, et al. *Final Report, Technical project Summary, Radionuclide Soil Action Level Oversight Panel*, February, 2000][10]

Nevada Test Site, Nevada USA

The Nevada Test Site located outside of Las Vegas, Nevada was used for a multitude of atmospheric and subterranean nuclear weapons tests. In addition, so-called “Safety Shots” were performed at the Tonopah Test Range which resulted in the dispersion of plutonium particles over a wide area. Safety shots generally consist of detonating high explosives near nuclear devices to determine whether the explosives will cause the device to activate.

In 1996, the Nevada Division of Environmental Protection (NDEP) and DOE signed a consent order. The “Clean Slates” sites was the focus of plutonium contamination cleanup where plutonium concentrations ranged from background to more than 12, 800 pCi/g (474 Bq/g). A dose assessment was performed by DOE which resulted in the recommendation of a soil cleanup level of 200 pCi/g (7.4 Bq) based on the rancher, farmer, rural resident and industrial worker exposure scenarios. [6]

Figure 3. Aerial of Nevada Test Site, USA With Sedan Crater

Hanford Nuclear Reservation, Washington USA

DOE's Hanford Site is part of the national nuclear weapons complex that was contaminated primarily as a result of plutonium processing operations. The site covers approximately 600 square miles in the southeastern portion of Washington state. The site

borders the Columbia River and has one of the pristine remaining examples of shrub-steppe habitat.

DOE, EPA and the Washington Department of Ecology signed an order on consent under the Federal Facilities Compliance Act , in 1989. This agreement is also referred to as the Tri-Party Agreement.

In determining risk and exposure scenarios at the Hanford Site, the Native American lifestyle is an important risk scenario as future land use may include hunting, fishing and cultural practices.

Cleanup levels are still being developed at Hanford and remediation activities in the 100 area of the Site provide an example of how cleanup values have developed. In 1995, EPA issued the 100 Area Record of Decision (ROD). The intent of the ROD was to establish cleanup levels that allowed unrestricted use by a rural resident based on a 15-mrem annual dose calculated by using RESRAD. The Remedial Action Goal (RAG) for the 100 Area, for direct exposure for plutonium-238 is 37.2 pCi/g (1.37 Bq/g) and for plutonium 239/240 it is 33.9 pCi/g.(1.25 Bq/g).

Project Chariot Site, North Slope Borough, Alaska, USA

The Project Chariot site is a remote and isolated area located in the Ogoturuk Valley along the Ogoturuk Creek in the Cape Thompson region of northwest Alaska. This region is approximately 200 miles north of the Arctic Circle and is bordered by the Chukchi Sea. Extreme weather conditions make it inaccessible most of the year. Native Inuit hunters harvest seal, walrus, and caribou in the area. The area is also used for gathering berries and other flora to support a subsistence type lifestyle. The U.S. DOE conducted a remedial action at the Project Chariot Site in the summer and fall of 1993.

The site was contaminated with soil brought in from the Nevada Test Site's Sedan Crater. The Sedan Crater was formed in 1962 from the test of a 100 kiloton nuclear device that was placed approximately 600 feet in a shallow area of the test site such that a crater would form after detonation of the device. The crater is on the National Registry of Historic Places Contaminated Sedan Crater soils along with radioactive tracers were brought to the Chariot site in order to conduct studies assessing the uptake of radionuclides in tundra biota including flora and fauna.

A risk assessment was performed to determine soil cleanup levels for plutonium and other long lived radionuclides using cesium 137 as an indicator radionuclide for the presence of the contaminated soils and performing analysis with gamma spectroscopy. The risk from exposure to the radionuclides was calculated using EPA's Human Health Evaluation Manual. The exposure scenario modeled the contaminated soil as containing 1.1 Bq/gram (30pCi/g) Cs-137. A preliminary remediation goal of 0.7 Bq/g (20pCi/g) was calculated based on a 1/10-4 risk of developing cancer over a person's lifetime and an exposure time for ten percent of the year due to the remote nature of the site. The clean-up level for Cs-137 was identified as 0.4 Bq/g (10pCi/g) resulting in a plutonium

cleanup level of .03pCi/g. (see Hopkins, *Project Chariot Remedial Action Work Pla*n [11] and *Project Chariot Site Assessment and Remedial Action Final Report* [12] and Hopkins, *Project Chariot Remediation.*)[13]

Maralinga, Australia

The Maralinga site in Australia, located in the remote outback, was used for the United Kingdom's nuclear testing program. Safety tests on the nuclear devices were performed which resulted in the dispersal of plutonium throughout the local environment. The plutonium was present in the form of particles, fragments and dust.

The cleanup criteria for the Maralinga site were determined by using a scenario involving the Aborigines in the area living an outstation lifestyle. The Maralinga Technical Advisory Committee (MARTAC) established three cleanup criteria: soil-removal, clearance, and unrestricted land use. These were based on three dose pathways: inhalation of re-suspended dust, ingestion of soil or contaminated food, and wound contamination.

The soil removal cleanup level was set where the levels of dispersed Am-241 exceeded 40 kBq/m2 (1.08xE6 pCi/m2) averaged over 10, 000 square meters. The clearance criteria, where soil was removed, was to leave soil that did not exceed 3 kBq/m2 of Am-241 (8.1xE4pCi/m2) averaged over 10, 000m2. The unrestricted land-use criteria considered permanent occupancy and unrestricted land use and required the Am-241 soil contamination level to be less than 3kBq/m2 Am-241 (8.1xE4pCi/m2) averaged over 3000m2. (*Maralinga Site Cleanup*, Australian Radiation Protection and Nuclear Safety Agency, web site detailed fact sheet).

The calculated dose to an individual living an Aboriginal semi-traditional lifestyle at the 3 kBq of Am-241/m2 (8.1x E4 pCi) contour is 5 mSv/a (500mrem), which is five times that allowed for a member of the public. This level of effective dose was accepted by the Tjarutja. It is the level at which the risk of contracting a cancer by age 50 is one in ten thousand. (Parkinson, Alan, *Maralinga Rehabilitation Project*, 2000.)

Figure 4. An Out-station in Maralinga, Australia

CONCLUSIONS

In summary, it appears that regulations regarding radioactive contamination cleanup levels in soils are still evolving, are complex, and vary both within the U.S and world-wide. The use of two different risk assessment approaches-dose and cancer risk (cancer slope factors) result in different cleanup values at sites. Dose conversion factors, cancer slope factors and computer models continue to be revised and updated. This results in soil cleanup levels becoming a moving target. The selection of input parameters to computer models can make a very large difference in the cleanup level that is derived from the risk assessment. This is especially true of determinations of future land use. And, if the cleanup at a site is not considered protective, the use of long-term stewardship methods must be considered.

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