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ABSTRACT

An inventory of radionuclides produced by 828 underground nuclear tests conducted at the Nevada Test Site by the Lawrence Livermore National Laboratory and the Los Alamos National Laboratory from 1951 to 1992 includes tritium, fission products, actinides, and activation products. This inventory provides an estimate of radioactivity remaining underground at the Nevada Test Site after nuclear testing. For the purposes of summary and publication, Los Alamos National Laboratory and Lawrence Livermore National Laboratory subdivided the inventory into five areas corresponding to the principal geographic test centers at the Nevada Test Site. The five areas roughly correspond to Underground Test Area "Corrective Action Units" for remediation of groundwaters. In addition, the inventory is further subdivided for the Yucca Flat region by tests where the working point depth is more than 328 feet (100 meters) above the water table and tests that were detonated below that level. Curie activities and atoms are reported as of September 23, 1992, the date of the last underground nuclear test at the Nevada Test Site. This inventory does not represent the total radioactivity dissolved in the groundwater beneath the Nevada Test Site, but is strictly a compilation of the residual radionuclide inventory remaining from those underground nuclear tests conducted by Lawrence Livermore National Laboratory and Los Alamos National Laboratory from 1951 to 1992. The written report is a companion to a personal computer-based database resident in the Analytical and Nuclear Chemistry Division of the Lawrence Livermore National Laboratory and the Chemistry Division of the Los Alamos National Laboratory. This computer database tabulates radionuclide totals, cavity radius, working point lithology, static water level, hole name, and firing data for each test. A companion classified report contains test-specific data from this computer database. This work has been sponsored by the Environmental Restoration Division at the U. S. Department of Energy, National Nuclear Security Administration Nevada Operations Office.

INTRODUCTION

This report provides an inventory of radionuclides residual from 828 underground nuclear tests conducted at the Nevada Test Site (NTS), Nye County, Nevada. The radionuclide inventory is part of a larger effort sponsored by the U. S. Department of Energy to comprehensively investigate the distribution and/or potential migration of these radionuclides in groundwaters at the NTS. Ultimately, the assessment of risk for receptors down-gradient from testing centers requires an accurate quantitative measure of potential contaminant species. Since 1951, the United States government has conducted 828 underground nuclear tests at the NTS (U.S. Department of Energy, Nevada Operations Office, 2000). These tests supported a program of strategic nuclear deterrence that served national defense policy during the four decades from 1950 to 1990. The U. S. Department of Energy and its predecessors sponsored these nuclear tests. The Los Alamos National Laboratory (LANL), the Lawrence Livermore National Laboratory (LLNL), and the U. S. Department of Defense with support from the national laboratories, conducted specific test series for the U. S. Government. In addition, a number of tests were conducted cooperatively on behalf of the United Kingdom. Objectives of the nuclear test program included ensuring operational readiness of stockpiled nuclear weapons, executing proof-of-principle experiments driven by nuclear weapon design, and studying the effects of enhanced radiation fields produced during nuclear explosions. Individual test schedules varied year to year and are summarized in Figure 1. From 1958 to 1961, nuclear testing was suspended by a voluntary international moratorium; in October 1992 a second voluntary testing moratorium was adopted by the U. S. Government and has remained in effect to the present. In this report we utilized data on the depths of burial and depths of the static water level (U.S. Department of Energy, Nevada Operations Office, 1997) compiled in support of environmental activities at the NTS.

THE NEVADA TEST SITE

The NTS is situated approximately 65 miles (105 kilometers) northwest of Las Vegas and comprises approximately 1,380 square miles (3,560 square kilometers) of north–south tending mountain ranges and mesas separated by broad alluvial basins typical of the Basin and Range physiographic province throughout much of Nevada, Arizona, and Utah. On the basis of its remote setting, favorable year-round weather, restricted access and prevailing wind patterns, the Nevada Proving Grounds—forerunner to the NTS—was established in the early 1950's as a continental proving ground for atmospheric nuclear tests. In 1951, the first underground nuclear test was fielded at the NTS and since late 1962, all U. S. nuclear tests have been conducted underground at the NTS. As a location for underground nuclear testing, the NTS is further characterized by extreme depths (>2,000 feet or > 600 meters), slow moving groundwater (1 to 300 feet/year or 1 to 100 meters/year), and an extremely arid climate.

THE RADIONUCLIDE SOURCE TERM

The potential for the contamination of groundwater beneath the NTS by nuclear testing has long been recognized (Batzel, 1959). Over the past two decades, studies of radiological contamination of soil and groundwater, notably those conducted by the Radionuclide Migration Program (presently the Hydrologic Resources Management Program), have complemented the nuclear test program. As a part of a Federal Facilities Agreement and Consent Order (1996) to address potential contamination at the NTS, the U.S. Department of Energy National Nuclear Security Administration Nevada Operations Office (NNSA/NV) designated the Underground Test Area (UGTA) to manage technical studies of groundwater contamination associated with underground testing.

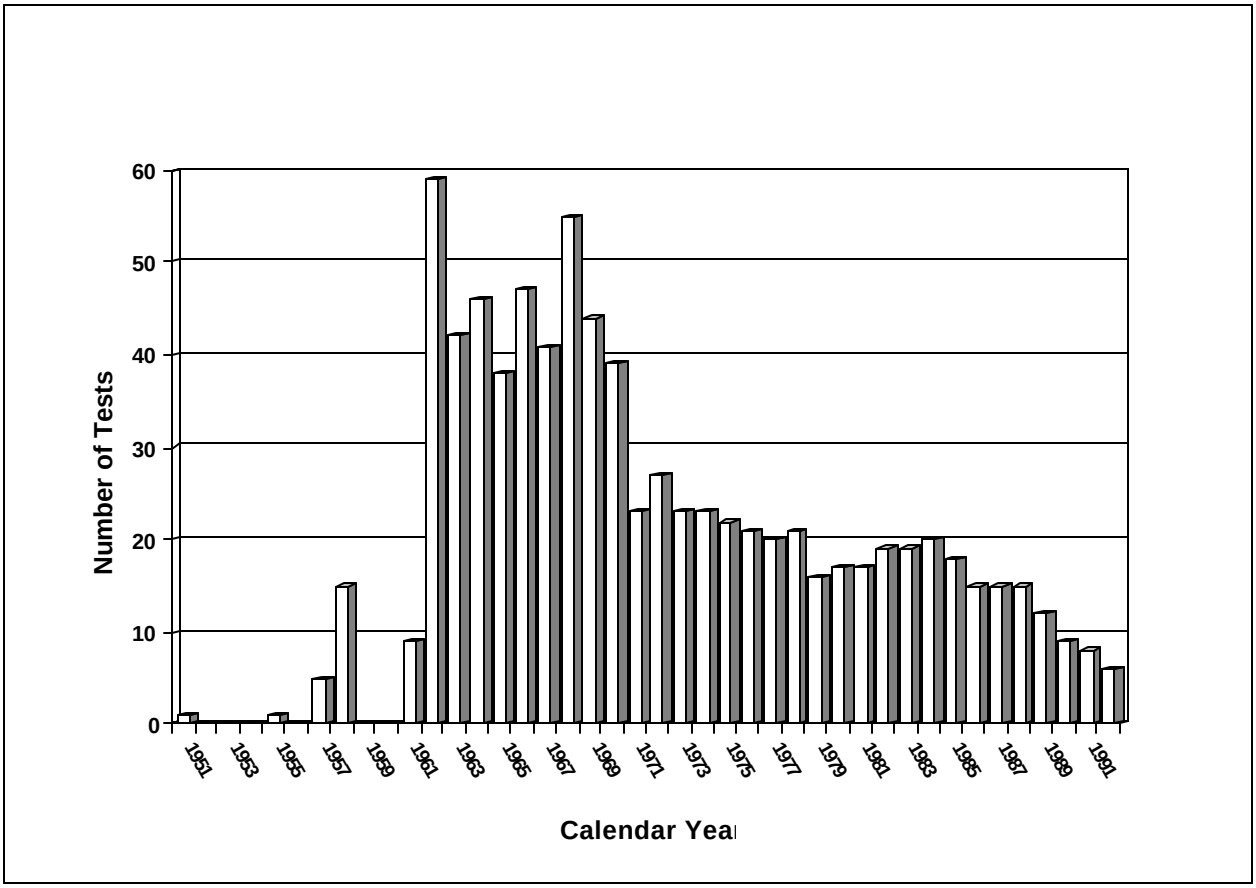

Figure 1: A histogram showing the number of underground nuclear tests conducted by year at the NTS.

The Environmental Restoration Division of NNSA/NV, which sponsored the radionuclide inventory effort, administers UGTA. An early priority of the UGTA included determining an accurate measure of the total radiological inventory present at the NTS. The quantity of existing contaminants provides an upper bound on the

contamination underground at the NTS. This information is vital in the design of remediation strategies and effective resource management. Similarly, risk assessment developed for human health and the environment requires a reliable measure of radionuclides available for potential transport by groundwater to down-gradient receptors. The radionuclide inventory includes long-lived radioactive species produced by or remaining after underground nuclear explosions at the NTS during the period 1951 to 1992. The inventory represents a starting point for the estimation of radionuclides available for dispersal away from test centers. Not all radionuclides are equally available for transport. A necessary distinction must be drawn between the radionuclide source term that includes all radioactive material remaining after a nuclear test and the hydrologic source term that includes only those radionuclides dissolved in and/or transported by groundwater. The radionuclide inventory reported here does not represent the amount of radioactivity that is or ever will be dissolved in groundwater at the NTS. The hydrologic source term is considerably less than the total radionuclide source term. This report presents the radionuclide source term, which combines inventories compiled for underground nuclear tests conducted by LANL and LLNL, as well as tests supporting the U. S. Department of Defense (DOD).

UNDERGROUND NUCLEAR TESTING

As an aid to understanding the radionuclide source term, a review of the execution of underground nuclear tests and the phenomenology associated with nuclear explosions is presented. For the purposes of this report, underground nuclear tests may be considered as tests involving fissionable nuclear material placed underground and designed to produce a nuclear yield. The yield—or the amount of explosive energy released from a nuclear weapon—is typically measured in TNT equivalents. Yields for NTS underground nuclear experiments at the NTS range from <1 kiloton to >1 megaton.

Underground nuclear testing practice has evolved considerably since the first underground test in 1951 was detonated in a shallow hole. The Rainier test of September 1957 was the first nuclear test contained completely underground and was designed to prevent the release of radioactivity as well as to determine whether diagnostic information could be obtained from an underground nuclear test. In 1963, the United States signed the Limited Test Ban Treaty prohibiting nuclear testing other than underground. Containment scenarios largely eliminated venting of radioactive debris to the atmosphere. Experience gained through 1992 allowed containment to minimize the accidental release of radioactive gas to the surface without compromising device performance or test diagnostics.

A brief introduction to underground testing is valuable to place the radionuclide source term in context. Interested readers are referred to a comprehensive discussion of the containment of underground nuclear explosions provided by the U. S. Congress, Office of Technology Assessment (1989) and Pawloski (1999). Containment relies on the physical properties of surrounding geologic media including rock elastic strength and porosity, the device depth of burial, and impermeable seals and backfill, known as stemming, which prevent gas release out of the emplacement hole. The majority of

underground tests were either vertical shaft tests or horizontal tunnel tests. More than 90% of the underground tests were fired in vertical shafts a thousand feet or more below ground surface. Shaft tests were fielded primarily to test stockpiled weapons or design features in new weapons systems. NTS vertical shaft tests were conducted predominantly beneath Yucca Flat (Areas 1, 2, 3, 4, 6, 7, 8, 9, and 10) and Frenchman Flat (Area 5 and 11) for lower yield experiments, and beneath Pahute Mesa (Areas 19 and 20) for higher yield experiments. Approximately 30% of the shaft tests were fired beneath the static water level (water table). Generally, tests conducted on Yucca Flat were buried at depths of approximately 2,000 feet (600 meters) or less. Higher yield experiments might be buried at depths exceeding 4,000 feet (1200 meters) on Pahute Mesa. Horizontal tunnel tests occurred within tunnel complexes excavated in Rainier and Aqueduct Mesas (Area 12), Oak Spring/Butte (Area 15), and Shoshone Mountain (Area 16). Most tunnel tests were fired within zones of discontinuously perched groundwater beneath the Rainier and Aqueduct Mesas. Figure 2 is a map of the NTS showing the locations of underground nuclear tests.

NUCLEAR EXPLOSION PHENOMENOLOGY

A brief discussion of phenomenology is warranted in the context of the evolution of the radionuclide source term. The following builds on the summaries provided by U. S. Congress, Office of Technology Assessment (1989) and those compiled by Glasstone and Dolan (1977). The sequence of events following a nuclear explosion is illustrated in Figure 3. At firing (zero) time pressures within the weapons case can exceed several million pounds per square inch, and temperatures may be as high as 10^8 degrees Kelvin. The shock waves propagate outward away from the cavity forming a radius of fractured rock that extends approximately two to three times that of the spherical cavity. The shock wave loses energy until the surrounding rock is no longer crushed but is merely elastically compressed. Hydrodynamic calculations indicate that the shock wave vaporizes approximately 70 metric tons and melts 700 metric tons of rock for each kiloton of explosive yield (Olsen, 1967). Milliseconds after detonation, the weapon's case, rack and geologic media immediately surrounding the device are vaporized and a cavity forms in response to gas pressure and the explosive energy imparted to the surrounding rock. For tests fired at or below the water table, all standing water near the working point is also vaporized. As the gas continues to expand adiabatically, the cavity grows (approximately) spherically for a few tenths of a second until the gas pressure drops below the ambient lithostatic pressure. At this point, the cavity has reached its maximum radius and volume. Scaling laws based on empirical studies relate cavity radius to explosive yield (see Glasstone and Dolan, 1977 and U. S. Congress, Office of Technology Assessment, 1989).

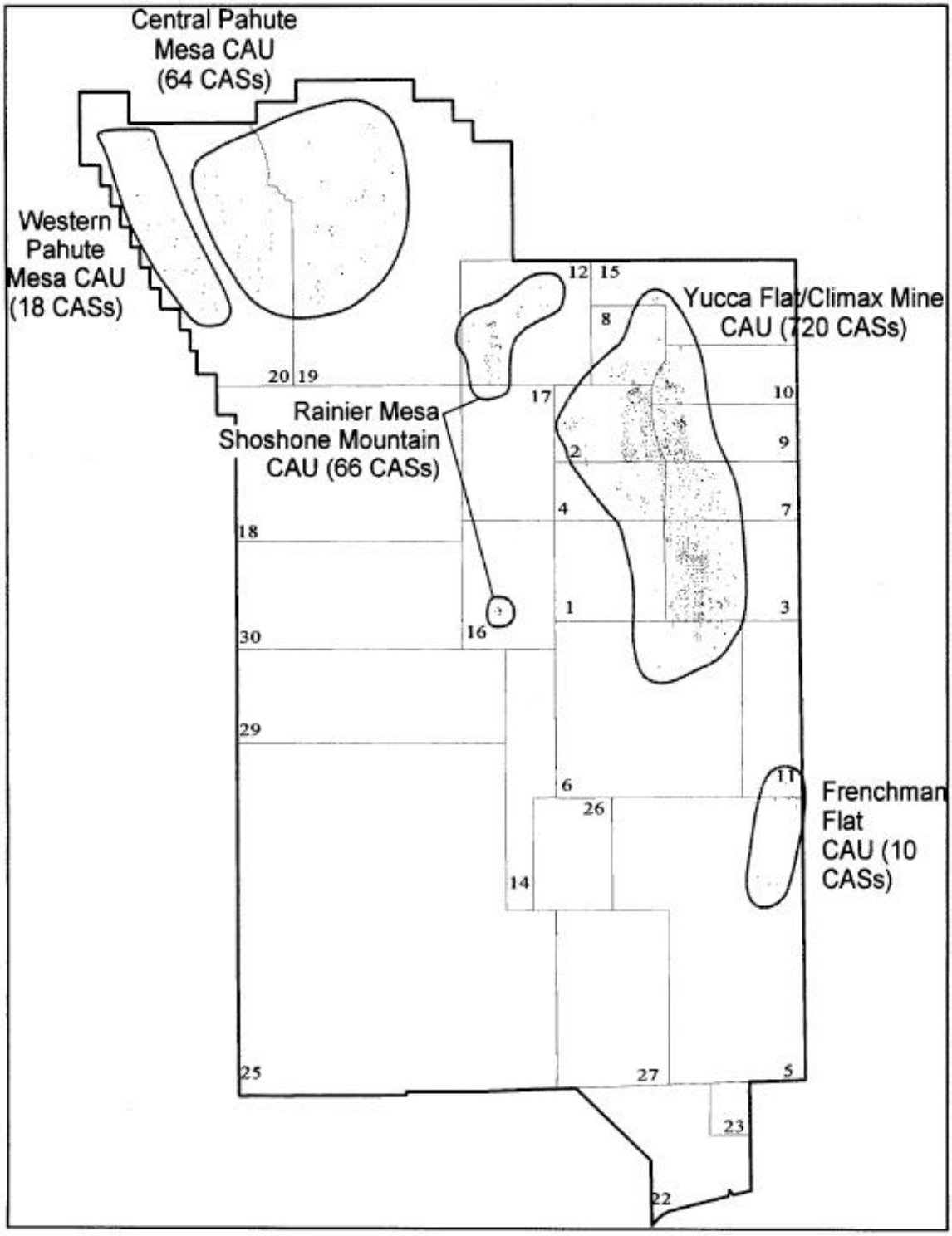

Figure 2: Map of the NTS showing the locations of underground nuclear tests.

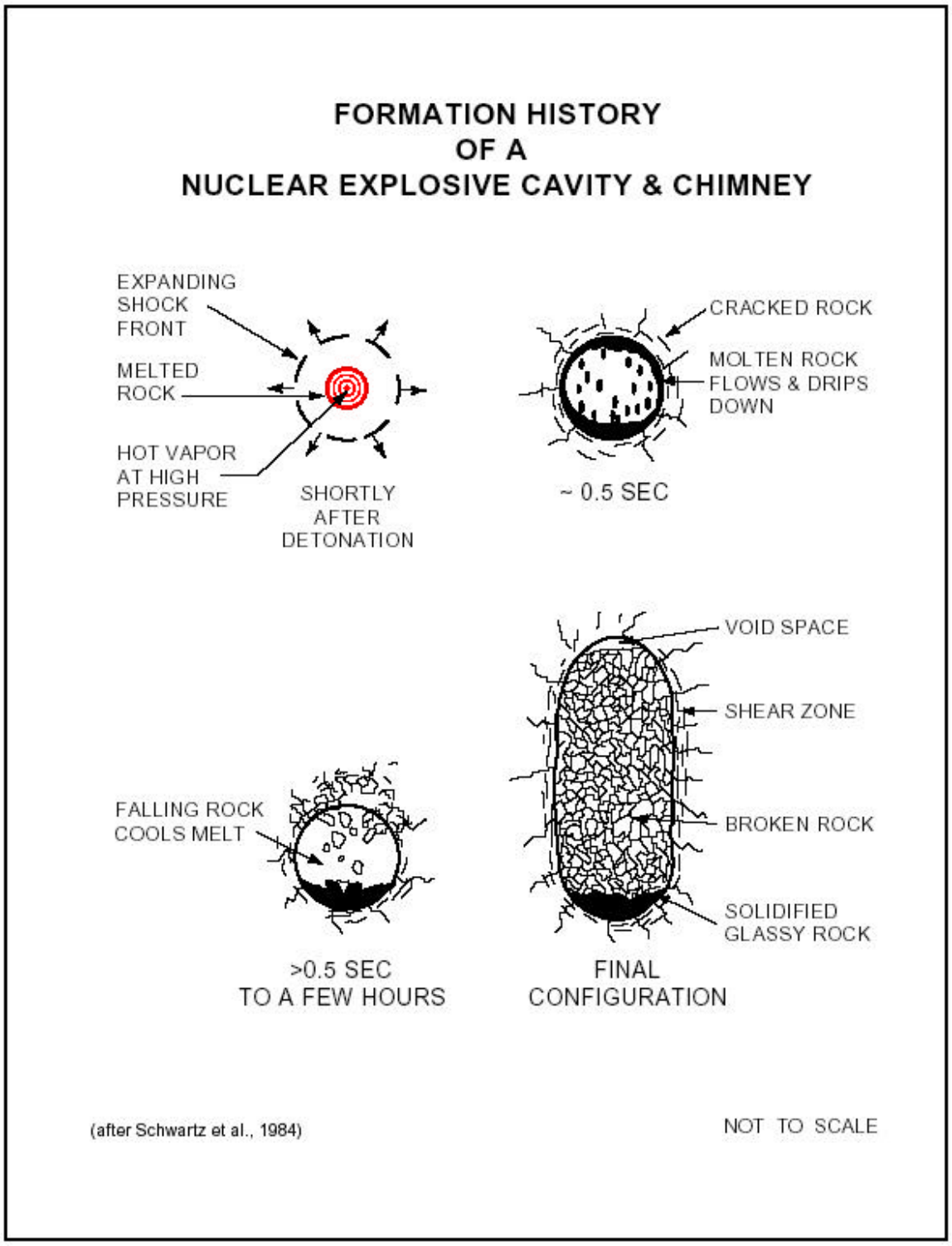

Figure 3: Idealized formation history of an underground nuclear explosion in siliceous rock. Stages are in elapsed time from detonation.

During the period of cavity growth, the vaporized material, consisting largely of volatilized silicate phases, condenses as it cools and mixes with molten rock that lines the circumference of the cavity. The melt flows down the walls and begins to coalesce in a puddle and solidify on the floor of the cavity. At this time, condensable gases change phase and cavity pressure drops. The gas pressure in the cavity diminishes to the point where the cavity can no longer support the weight of the overlying rock. The cavity collapses in on itself, often with blocks of rock chaotically incorporated in the still partially molten melt glass. As blocks of rock fill the cavity void, the process is perpetuated upward as the rubble continues to fall downward. This process creates a rubble chimney that propagates upward from the working point until the void volume in the chimney is completely filled with rubble debris and the strength of the overlying rocks can support the overburden, or until the chimney reaches the surface and creates a subsidence crater. Typically the collapse and chimney formation occurs within a few hours of zero time, but it may occur as late as a few days or months after the explosion. For tests fired at or below the water table, hydrologic heads immediately perturbed by the displacement of water during the explosion will begin to return to original preshot static water levels within days to years, depending on adjacent hydrologic conductivities.

DISTRIBUTION OF RADIONUCLIDES

Temperature and pressure histories associated with the explosion strongly influence the distribution of radionuclides within the cavity-chimney complex (Borg *et al.*, 1976). Immediately after a nuclear explosion, all of the radioactive species exist as a plasma. As the cavity ceases to expand, heat is transferred to the wall rock and the cavity temperature and pressure begin to drop. The melt that flows to the floor of the cavity entrains the refractory radionuclides with higher boiling points (rare earth elements, alkaline earths, Zr, and Pu). Most of these refractory species are trapped in the cooling melt; a small proportion is incorporated with the collapsed chimney rubble as splash or fine droplets entrained with escaping cavity gases. Volatile species with lower boiling points (tritium, alkalis, Ru, U, Sb, Cl, I) circulate up cracks in the rubble chimney. Activation products are concentrated around the working point and will be largely incorporated in the melt or debris that borders the cavity. Volatile species, particularly ^{90}Kr and ^{137}Xe, are transported as gases through the rubble and will be concentrated higher in the cavity and in the chimney relative to the refractory radionuclides. Drillback samples systematically exhibit higher volatile to refractory radionuclide ratios (Cs/Eu) for returns collected higher in the cavity and chimney (Thompson, 1996). In addition, recent studies of nuclear tests fired above the water table indicate that material may be transported by prompt injection through explosion induced fractures arranged radially away from ground zero (Smith et al., 1996). By this mechanism, gaseous species, particularly tritium and ^{137}Cs (^{137}Cs has a gaseous ^{137}Xe precursor with a 3.8 minute half-life), may be deposited several cavity radii away from the working point. There is some evidence that refractory species may be similarly transported by prompt injection (Nimz and Thompson, 1992). Ultimately, the amounts and types of radionuclides resulting from a specific nuclear explosion will depend on the amount of fissile material, the fission/fusion ratio, and the device yield. On a test-by-test basis, these data are classified. This report attempts to

quantify the abundance of these residual radionuclide species for five principal geographic test centers at the NTS.

RADIONUCLIDE INCLUSION CRITERIA

Radionuclides to be considered for inclusion in the source term inventory are: 1) residual and unburned fissile fuel and tracer materials, such as U isotopes, Pu isotopes, Am isotopes, and ^{244}Cm; 2) fission products such as ^{137}Cs and ^{90}Sr; 3) tritium (^{3}H); and 4) activities induced by neutrons in device parts, in external hardware, and in the surrounding geologic medium (such as ^{14}C, ^{36}C1, and ^{41}Ca). Not all of the radionuclides produced during a nuclear test are included in the source term inventory. Many of the nuclides have half-lives so short (microseconds to hours) that they decay to undetectable levels soon after the test. Other nuclides are produced in such low initial abundance that they never exceed levels deemed unsafe or non-permissible by regulatory agencies. Criteria were developed to exclude such nuclides from the inventory; this permits the user to focus attention on the nuclides of interest from the perspective of risk assessment. Excluded were nuclides produced in such low amounts that if all of the amount produced during a nuclear test were dissolved into a volume of water equal to the volume of the detonation cavity for the test, 100 years from 1992 the resulting aqueous concentration (activity) in μCi/mL (one μCi = 2.22 x 10^{6} disintegrations per minute) would be less than one-tenth of the values for the maximum permissible concentrations (MPC's) proposed for drinking water by the U. S. Environmental Protection Agency in the Federal Register (1991). This effectively excludes almost all radionuclides with half-lives less than ten years. However, if a radionuclide exceeded these criteria for at least one test, it was included for all other tests for which estimates are available even if concentrations were below the 0.1 MPC criterion. The MPC of a nuclide listed in this compilation is that concentration in drinking water that will impart a dose of 4 mrem/year to a person drinking an average of 2 liters of water per day. The requirement of 100 years into the future eliminates many nuclides that are produced in great abundance in nuclear detonations but have half-lives sufficiently short that they will have decayed below the 0.1 MPC value by that time. For nuclides with no listed values, we conservatively assumed a value of 10^{-8} Ci/mL for the MPC. We have compiled the list of nuclides shown in Table I that have half-lives of >10 years (with the exception of ^{154}Eu at 8.6 y) and could be produced in nuclear detonations. Listed values of MPC's were obtained from the Federal Register reference. Table I includes the principal radiochemical sources or production mechanisms of test-derived radionuclides. Where multiple sources are identified, each is listed.

The cavity volume was determined using the cavity radius for each test where it was known and converting this to a volume in mL, assuming a spherical cavity. To arrive at a concentration value, we assumed that the radionuclides produced in the test were distributed uniformly throughout the cavity as if they were dissolved in a volume of water equal to the cavity volume. For tests with very low total yields (<0.1 kiloton), cavity volumes were negligible and the concentration criterion was not invoked.

TABLE I

Candidate Radionuclides for Inclusion into Source-Term Inventory
(MPG values from Federal Register, 1991)

Element	Nuclide	Half-life (y)*	MPC (μCi/mL)	Main Source(s) (FP=fission product)
Hydrogen	^{3}H	12.32	6.1×10^{-5}	device component; ^{6}Li (n,α) T
Carbon	^{14}C	5715	3.2×10^{-6}	^{14}N (n,p); ^{13}C (n,γ); ^{17}O (n,α)
Aluminum	$^{26}A1$	7.1×10^{5}	----	$^{27}A1$ (n,2n)
Chlorine	$^{36}C1$	3.01×10^{5}	1.8×10^{-6}	$^{35}C1$ (n,γ); ^{39}K (n,α)
Argon	^{39}Ar	269	----	^{39}K (n,p); ^{38}Ar(n,γ)
Potassium	^{40}K	1.27×10^{9}	----	natural
Calcium	^{41}Ca	1.03×10^{5}	----	^{40}Ca (n,γ)
Nickel	^{59}Ni	7.6×10^{4}	2.7×10^{-5}	^{58}Ni (n,γ)
	^{63}Ni	100	9.9×10^{-6}	^{62}Ni (n,γ), ^{64}Ni (n,2n), ^{63}Cu (n,p)
Krypton	^{85}Kr	10.76	----	FP; ^{84}Kr (n,γ)
Strontium	^{90}Sr	28.78	4.2×10^{-8}	FP
Zirconium	^{93}Zr	1.5×10^{6}	5.1×10^{-6}	FP; ^{92}Zr (n,γ); ^{94}Zr (n,2n)
Niobium	^{93m}Nb	16.1	1.0×10^{-5}	^{93}Nb (n,n′)
	^{94}Nb	2.0×10^{4}	7.1×10^{-7}	FP; ^{93}Nb(n,γ)
Technetium	^{99}Tc	2.13×10^{5}	3.8×10^{-6}	FP; ^{99}Ru (n,p)
Palladium	^{107}Pd	6.5×10^{6}	3.7×10^{-5}	FP; ^{106}Pd (n,γ)
Cadmium	^{113m}Cd	14.1	----	FP
Tin	^{121m}Sn	~55	2.3×10^{-6}	FP; ^{120}Sn (n,γ)
	^{126}Sn	2.5×10^{5}	2.9×10^{-7}	FP
Iodine	^{129}I	1.57×10^{7}	2.1×10^{-8}	FP; ^{129}Xe (n,p)
Cesium	^{135}Cs	2.3×10^{6}	7.9×10^{-7}	FP
	^{137}Cs	30.07	1.2×10^{-7}	FP; ^{137}Ba (n,p)
Samarium	^{151}Sm	90	1.4×10^{-5}	FP; ^{150}Sm (n,γ)
Europium	^{150}Eu	36	----	^{151}Eu (n,2n)
	^{152}Eu	13.54	8.4×10^{-7}	^{151}Eu (n,γ); ^{153}Eu (n,2n)
	^{154}Eu	8.593	6.7×10^{-7}	^{153}Eu (n,γ)
Holmium	^{166}Ho	1.2×10^{3}	----	FP; ^{165}Ho (n,γ)
Thorium	^{232}Th	1.40×10^{10}	9.2×10^{-8}	natural and device component
Uranium	^{232}U	69.8	1.0×10^{-8}	device component; ^{233}U (n,2n)
	^{233}U	1.592×10^{5}	2.6×10^{-8}	device component; radiochemical tracer
	^{234}U	2.46×10^{5}	2.6×10^{-8}	natural and device component
	^{235}U	7.04×10^{8}	2.6×10^{-8}	natural and device component
	^{236}U	2.342×10^{7}	2.7×10^{-8}	device component; ^{235}U (n,γ); ^{238}U (n,2n)[2]
	^{238}U	4.47×10^{9}	2.6×10^{-8}	natural and device component

Element	Nuclide	Half-life (y)*	MPC (μCi/mL)	Main Source(s) (FP=fission product)
Neptunium	^{237}Np	2.14 x 10^{6}	7.2 x 10^{-9}	radiochemical tracer; decay of ^{237}U
Plutonium	^{238}Pu	87.7	7.2 x 10^{-9}	device component; radiochemical tracer; ^{239}Pu (n,2n); ^{237}Np (n,γ)
	^{239}Pu	2.410 x 10^{4}	6.5 x 10^{-8}	device component; decay of ^{239}U
	^{240}Pu	6.56 x 10^{3}	6.5 x 10^{-8}	device component; ^{239}Pu (n,γ); decay of ^{240}U
	^{241}Pu	14.4	----	device component; ^{240}Pu (n,γ); decay of ^{241}U
	^{242}Pu	3.75 x 10^{5}	6.8 x 10^{-8}	device component; radiochemical tracer; ^{241}Pu (n,γ); decay of ^{242}U
Americium	^{241}Am	432.7	6.4 x 10^{-9}	device component; radiochemical tracer; decay of ^{241}Pu
	^{243}Am	7.37 x 10^{3}	6.5 x 10^{-9}	device component; radiochemical tracer
Curium	^{244}Cm	18.1	1.0 x 10^{-8}	radiochemical tracer

* Half-lives obtained from GE Chart of the Nuclides, Fifteenth Edition (1996).

DATA STRUCTURE

The complete underground source-term inventory for the NTS combines separate inventories compiled by LANL and LLNL. By agreement, the Chemistry Division of LANL will maintain the Microsoft® Access database and provide the Analytical and Nuclear Chemistry Division of LLNL with any updated version of the database. Although the present document reports a summary of the inventory, it is LANL's and LLNL's intention that their respective inventories reside in digital format at both laboratories. The databases will be improved through refinements in pre-shot and post-shot data input, error checking, and refinements of computational algorithms — particularly for fission products and tritium. The databases are designed to be "living" files that can be manipulated to serve the requirements of specific investigators. By this definition, LANL and LLNL computer databases, not this or any supporting document, carry the current and comprehensive radionuclide inventory.

The LANL database resides in Microsoft® Access that runs on a Windows NT computing platform. Computer hard drives incorporate the active databases that are regularly backed-up on tape-drive storage media. The database is organized by test name, hole, number, sponsoring laboratory, fire date, total yield, depth of burial, cavity radius, static water level, working point geology, and test-specific radionuclide amounts (reported in atoms and curies at test time) for 43 isotopes meeting the above criteria for inclusion. Several parent isotopes with shorter half-lives are also compiled in the database for decay calculations. Because the databases compile test-specific nuclear performance data including yield, residual nuclear fuels, and thermonuclear and fission products, by necessity they are classified as Secret Restricted Data. The databases are accessible to

investigators holding an active U. S. Department of Energy Q security clearance with a valid need-to-know.

For the purposes of summary and publication, LANL and LLNL subdivided the inventory into five areas corresponding to the principal geographic test centers at the NTS (see Table II). The five areas roughly correspond to UGTA "Corrective Action Units" for remediation of groundwaters. In addition, the inventory is further subdivided for the Yucca Flat region by tests where the working point depth is more than 328 feet (100 meters) above the water table and tests that were detonated below that level. Generally, testing sites within each of the five areas have similar geologic and hydrologic properties.

TABLE II

Combination of NTS Areas into Principal Geographic Test Centers

NTS Principal Geographic Test Centers	NTS Areas Included
Yucca Flat*	1,2,3,4,6,7,8,9,10,15
Pahute Mesa – 19	19
Pahute Mesa – 20	20
Frenchman Flat	5,11
Rainier Mesa/Shoshone Mt	12,16,18,30

*Further subdivided into above and below water table.

Summaries derived from this database list the aggregate activities in curies and atoms of radionuclides that meet the selection criteria. The values are decay-corrected to September 23, 1992, the date of the last underground nuclear test at the NTS, and designated as above or below the reference point static water level plus 328 feet (100 meters) for Yucca Flat. These tables represent the long-lived radionuclide source term for all tests conducted underground at the NTS by LANL, LLNL, and the DoD between 1951 and 1992.

SOURCES OF RADIONUCLIDE DATA

Values for the total inventory of radionuclides were determined in two principal fashions for this compilation: measurements and calculations. Many nuclides were historically measured from small samples recovered from the underground environment as part of the radiochemical diagnostics effort. These nuclides were those most relevant for diagnostic purposes: fission products, residual fuel species, and radiochemical detector and tracer isotopes. These measurements have also served to establish an understanding of the processes occurring in the underground environment and were often reduced to computer codes used in the prediction of the production of nuclides. These codes were used to

stand in where measurements were not made at the time of the test or where interpretation of the data for total inventory was difficult.

Radiochemical diagnostics depend upon the recovery of a small portion of the debris from the underground nuclear test site. Both LANL and LLNL did this in a similar fashion, using slightly different methods to choose and collect samples. Only small portions of the residual materials were recovered. Recovered material was dissolved in strong acids to produce a solution of device material; analysis proceeded to determine atoms of particular nuclides per milliliter (or gram) of this solution. These quantities can be converted into total inventory by dividing by the relative proportion of the device in the sample.

As part of the radiochemical diagnostics effort, knowledge of the pre-test inventory of materials going into the test device is required. Complete inventories of fissile isotopes (limited in early times by measurement techniques) were recorded for essentially all tests. Similarly, most tritium data are also available. Based upon complete suites of measurements, it is possible to establish mathematical relationships (implemented by computer codes) to mimic isotopic changes when measurements were not made. A large amount of the information required for this study was available in existing databases maintained by the Analytical and Nuclear Chemistry Division at LLNL and the Chemistry Division at LANL. Other information was derived from publications or databases from other organizations. Information collected in this study has been entered into an Access database, with each laboratory responsible for maintaining its contribution. These data have been collated for the summary described here, and are described in detail in the comprehensive report (Miller, et al., 2001).

METHODS OF ESTIMATING POSTSHOT RADIONUCLIDE CONTENT

Postshot radionuclides present underground at the NTS can be conveniently grouped into four different categories: fission products, actinide elements, tritium, and activation products/natural materials. Each of these groups requires different methodologies for estimating the postshot quantities of the nuclides. In addition, historical differences in the radiochemical diagnostic approaches employed by LANL and LLNL lead to slightly different techniques for each laboratory. The following describe the methods used in this study.

FISSION PRODUCTS

Most of the fission products of interest to the UGTA are formed in such low yield or have such long half-lives that they were not measured radiochemically following the tests. In addition, many of these isotopes experience chemical fractionation and are not representative of the bomb fractions. These volatile radionuclides are of little or no diagnostic value, but are of importance to remedial investigations because of their potential mobility.

For a large number of tests, high-yield fission products were measured in order to determine the fission yield of the device. Other fission products were also measured to characterize the fission split or the distribution of fissions among the fissile materials. For the remaining tests, sufficient precedent or other diagnostic measurements were available to state the fission yield and split. Thus determined, the fission yields and splits have served as the basis for the calculation of the amounts of the various fission products given in this study.

We have chosen to calculate the postshot amounts from fission yields based on the type of fissile fuel and the neutron energy spectrum. In most cases, these characteristics were available in the documentation associated with each individual test. In other cases, we reasoned by analogy. The overall, effective yield of a fission product for each test is the weighted average of the yields from that test's mix of fissions from each fissile fuel and neutron energy spectrum. For the majority of fission products, the fission yields of England and Rider (1994) were used by both LANL and LLNL.

ACTINIDES

Los Alamos' Chemistry Division and Livermore's Analytical and Nuclear Chemistry Division have chosen consistent paths in the manner in which they have treated their actinide data. This is in part driven by the available data and in part by the technical biases that developed in their individual histories of weapons radiochemical diagnostics.

For many isotopes, we based our determination of the residual amounts of radionuclides from the measurements on actual core samples. These measurements were made historically in the course of the underground test program. In most cases, the postshot amount of a particular isotope or suite of isotopes was known, based on device performance. This permitted the determination of the fraction of the device present in each particular sample (i.e., the bomb fractions). Through ratio measurements to other species, the absolute abundances of the isotopes could be calculated.

A different approach was used when measurements were not available or were compromised by underground fractionation phenomena. Based on knowledge of the device, its performance, and production/destruction characteristics, we calculated the amounts of postshot isotopes, using algorithms implemented in the database structure. Some approximations were very simple: tracer isotopes were treated as unchanged from the ingoing amount loaded with the device. Other approximations were derived from many years of observations combined with explosion code information.

TRITIUM

On most U. S. nuclear tests, for times as long as 100 years following the detonation, tritium has the highest activity of any radionuclide with a half-life longer than ten years (see Figure 4 on page 19). However, tritium was not measured on most U. S. nuclear tests. Because of this we have expended considerable effort in estimating the residual tritium on the individual nuclear tests. We have also sought to achieve agreement

between LANL and LLNL for this isotope. Because of their earlier start on the underground inventory effort, LLNL has provided most of the algorithms for tritium production and destruction. LANL has served to confirm these methods through independent explosion code calculations.

The amount of residual tritium from a nuclear test is the net result of the amount loaded on the device and the amounts produced and destroyed in the nuclear explosion. A wide variety of sources and reactions are involved in this production/destruction. Each unique material and device type was examined to provide methods consistent with the limited suite of tritium measurements and the results of explosion code calculations.

ACTIVATION PRODUCTS AND NATURAL ISOTOPES

Reactions of neutrons with parts of the device other than the fuel, such as the structural materials, the stemming, and the geologic medium, produce several nuclides with half-lives longer than 10 years. There are about 50 such nuclides, but many of them can be excluded using the source term inventory criteria, because of extremely long half-life of the nuclide or very small production rate. Several nuclides do meet the selection criteria, however. The contributions of these nuclides to the source term were estimated using available data on other slow- and fast-neutron activation products, relevant cross section information, and neutron transport calculations.

For most of the nuclides of interest, activation of the geologic medium by reactions of slow neutrons is an important production mechanism. To estimate this, we made use of extensive data at both laboratories on the production of ^{160}Tb by ^{159}Tb (n,γ). Both the Analytical and Nuclear Chemistry Division at LLNL and the Chemistry Division at LANL have measured ^{160}Tb in debris samples from a large number of underground nuclear tests as an indicator of neutron activation of soil constituents. These measurements were used to correct other observations for such soil contributions. Empirical relationships were independently derived by the laboratories to estimate the ^{160}Tb production in cases where measurements were not made.

The amounts of other nuclides produced by the slow neutron reactions in the soil relative to ^{160}Tb were estimated using relationships derived at LLNL using neutron transport calculations. Monte Carlo neutron transport calculations were performed using the MCNP code (LANL, 1981); in addition, the Monte Carlo transport calculations by Lessler and Guy (1965) were also useful. Average trace element abundances (C.F. Smith, 1993) in the geologic media for several drill holes were used in these calculations (see Table III). Adjustments were made for the different composition of the LANL magnetite stemming material. These approximations were considered adequate in view of the rather large uncertainties in the calculation of the activation products and the fact that the activation products in general are not major components of the radionuclide inventory.

The production of nuclides in device parts was, in some cases, calculated relative to other nuclides that were actually measured as part of the radiochemical diagnostics of the test. In addition, naturally-occurring radioactive isotopes (^{40}K, ^{232}Th, ^{234}U, ^{235}U, and ^{238}U)

were included and represent the amount in the rock melted during a detonation (700 tons/kiloton of yield).

TABLE III

Soil and Magnetite
Elemental Abundances

Element	Soil Abundance (wt%)	Magnetite Abundance (wt%)
Al	4.9	0.78
Cl	0.011	<0.012
K	4.2	0.24
Ca	2.8	1.04
Ni	0.0094	0.0094
Nb	0.001	0.001
Eu	9.0×10^{-5}	2.3×10^{-4}
Tb	6.8×10^{-5}	2.2×10^{-4}

ACCURACY OF THE REPORTED INVENTORY

The accuracy quoted on the content of the inventory depends to a large part on the sources of the included nuclides. Clearly, nuclides whose abundances were measured directly are more accurately reported than those for which estimates had to be made based on device characteristics and performance. There are also tests in the inventory for which little or no postshot information exists; estimates of nuclidic content for these tests are considerably more uncertain and increase the overall uncertainty for a given nuclide. In Table IV, we show the different groups into which the inventory nuclides have been combined, along with our estimates of the accuracies of the test-by-test amounts we obtained for the nuclides in each group. The accuracies of fission product amounts from very low yield tests are not included in Table IV because the contribution of such tests to the total fission product inventory is negligible. Details of the accuracies of individual nuclide amounts are given after the table.

TABLE IV

Estimated Accuracies for Individual Nuclides in
the Various Groups of Radionuclides in this Inventory

Fission products:	~ 10 to 30 % for most of the fission products
Unspent fuel materials:	~ 20 % or better
Fuel activation products:	~ 50 % or better
Residual tritium:	~ 300% or better
Activation products:	~ a factor of 10

FISSION PRODUCTS

For reported amounts of fission products, accuracy depends on the accuracy of the following for that test: 1) the fission yield (kilotons), 2) the fission split, and 3) the yields of the fission product (atoms per fission) from the various fissions for the test. Not counting some of the tests with very low yields (<0.1 ton), the fission yields of almost all the tests are known to within 5 to 30%. For those tests with radiochemical measurements, the fission splits among the fissionable nuclides usually are known accurately. The accuracies for the fission product yields we quote here are from England and Rider (1994). The least accurately known yields are those for the fission products ^{94}Nb, ^{113m}Cd, ^{121m}Sn, ^{152}Eu, and ^{166m}Ho. Their yields could be in error by a factor of 2 to 100 or more. Fortunately, these are also the fission products with the lowest yields, so they are very minor contributors to the radionuclide inventory. Furthermore, the total amounts of ^{94}Nb, ^{152}Eu, and ^{166m}Ho produced as activation products are at least 1000 times larger than the amounts from fission. The accuracies of the ^{126}Sn yields are 4 to 16%. The accuracies of the yields of all the other fission products in our inventory are 0.4 to 11%. For tests on which no radiochemical measurements were made, uncertainty for the fission products can exceed 200% unless the test was known to be similar in design and performance to one measured.

ACTINIDES

Most of the tabulated amounts of unspent fission fuel and tracer materials are accurate to 1 to 10%. A few, including some of the values for tests for which the device performance is known only approximately, have uncertainties of up to 20%.

The tabulated amounts of minor isotopes and activation products of fission fuel, when derived from actual measurements, are accurate to 20% or better. However, when the amounts are estimates from approximate device performance information and estimated preshot compositions, the uncertainties for some of the isotopes may rise to 50%. The total ^{232}U inventory is good only to a factor of 2 or 3 because only about 15% of it is based on actual measurements. Uranium-237 data for some of the older tests are missing

and still need to be found or replaced by estimates. Because of this, our ^{237}Np inventory is probably low by a few tens of percent.

TRITIUM

The estimates of residual tritium have a wide range of uncertainties: from 1 - 300%. The 1 or 2% uncertainty applies to those tests on which there was no thermonuclear yield and negligible tritium production, in which case the postshot amount of tritium equals the preshot amount. The 300% uncertainty applies to devices with unusual designs and tests for which performance is only known approximately.

ACTIVATION PRODUCTS

Our activation product estimates are likely accurate only to a factor of 10 in most instances. This relatively large error is the result of the approximate nature of the methods we used to obtain the estimates described above, such as using data for nuclides from similar reactions and approximate Monte Carlo neutron transport models employing average elemental abundances for soil activation. The part of the europium isotope inventory that is based on direct measurements is more accurate, with uncertainties of 10 to 30%.

RADIONUCLIDE INVENTORY WITH TIME

Figures 4 and 5 illustrate the relative percentage and total activity (plotted on log scale) of residual tritium, fission product, and actinide activities remaining with time (from 1994, in years) from underground nuclear tests conducted below the water table at the NTS. Tritium initially dominates the radionuclide inventory but is overtaken by fission products after approximately 100 years. After 500 years, actinides populate the majority of the remaining inventory, while the total activity has decreased approximately three orders of magnitude.

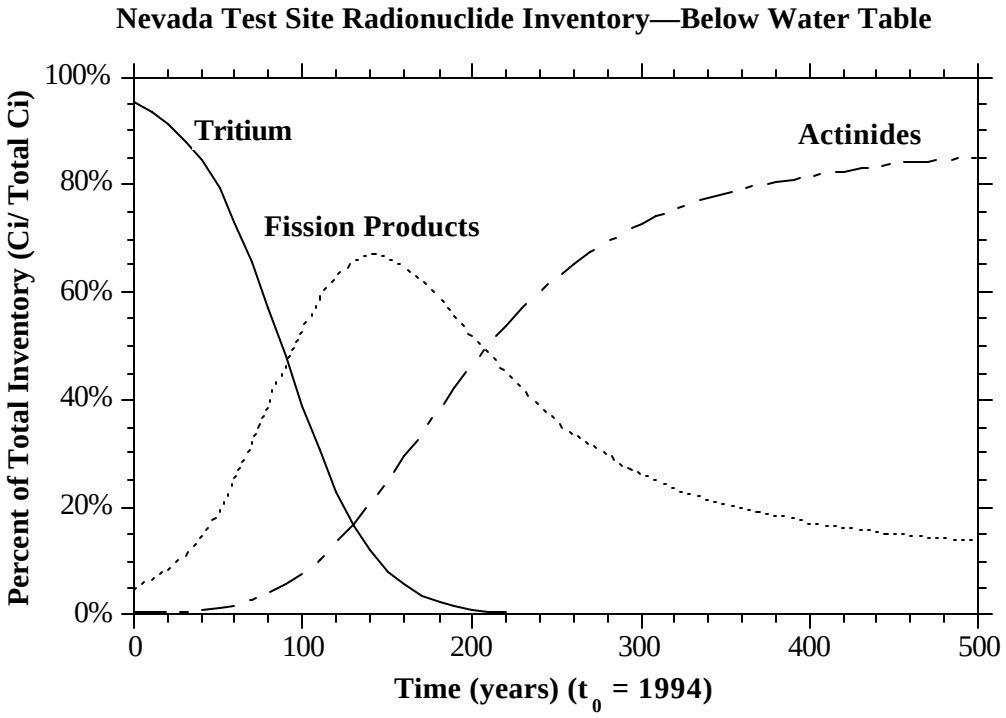

Figure 4: Relative percentage of residual tritium, fission product, and actinide activities below water table remaining with time.

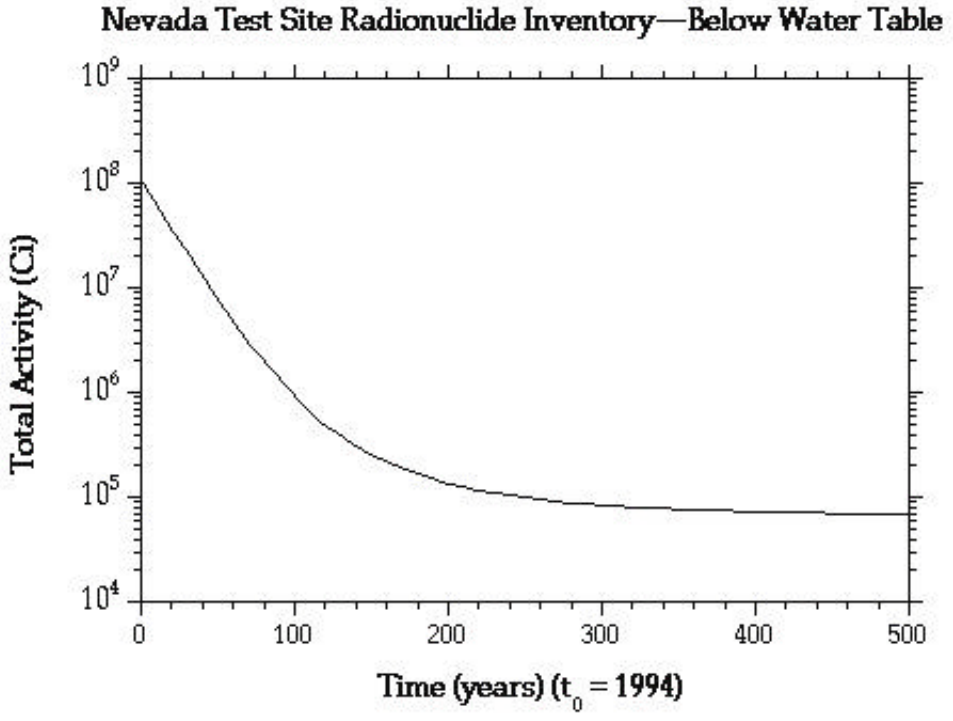

Figure 5: Total activity in curies below water table decayed over 500 years.

EXPLANATION OF RADIONUCLIDE INVENTORY TABLES

Table V contains the summary of radionuclide totals in Curies as they existed on September 23, 1992. Table VI contains the summary of the radionuclide totals in atoms as they existed on September 23, 1992. These data are grouped according to six principal geographic test centers of the NTS (five areas with two sub-divisions in Yucca Flat). Radionuclides are arranged within each table according to their atomic number and atomic mass.

NOTES ON CALCULATIONS

We give below several basic equations useful in relating activities and numbers of radioactive atoms at specified times. It should be noted that activities (A) and decay constants (λ) must be in the same time units in these equations. In Table V activities are given in curies (disintegrations/second) and half-lives in years, so time conversions are required. We assumed 1 year = 365.24 days = 3.156E+7 seconds.

- Activity $\Leftrightarrow$ atoms

$A = \lambda N$

A = activity in disintegrations/unit time

λ = ln(2)/half-life

N = number of atoms

- Activity or atoms at various times

$A(t) = A(0)e^{-\lambda t}$

$N(t) = N(0)e^{-\lambda t}$

A(0), N(0) are at reference time

A(t), N(t) are at time-of-interest

λ = ln(2)/half-life

t = difference between reference time and time-of-interest

TABLE V

Radionuclide Summary in **Curies** Decay Corrected to September 23, 1992
for the Six Principal Geographic Test Centers

Radio-nuclide	Frenchman Flat	Pahute Mesa-19	Pahute Mesa-20	Rainer Mesa Shoshone Mt	Yucca Flat - Above	Yucca Flat - Below	**Total**
H-3	1.744E+05	1.778E+07	5.903E+07	7.645E+05	1.472E+07	3.316E+07	**1.256E+08**
C-14	6.653E+01	2.193E+02	4.693E+02	1.102E+02	1.137E+03	8.389E+02	**2.841E+03**
Al-26	7.035E-03	8.975E-04	8.370E-03	4.548E-04	5.573E-02	3.595E-02	**1.084E-01**
Cl-36	8.907E+00	9.108E+01	1.573E+02	1.130E+01	1.163E+02	2.309E+02	**6.158E+02**
Ar-39	6.166E+00	6.398E+02	1.247E+03	3.663E+01	3.204E+02	9.551E+02	**3.205E+03**
K-40	1.649E+00	1.588E+02	3.171E+02	9.233E+00	8.219E+01	2.422E+02	**8.112E+02**
Ca-41	6.542E+01	5.050E+02	1.273E+03	7.063E+01	8.545E+02	1.661E+03	**4.429E+03**
Ni-59	1.634E+00	1.596E+01	2.976E+01	2.021E+00	2.139E+01	4.265E+01	**1.134E+02**
Ni-63	1.679E+02	1.724E+03	3.126E+03	2.118E+02	2.334E+03	5.229E+03	**1.279E+04**
Kr-85	1.285E+02	4.981E+04	5.706E+04	1.344E+03	1.137E+04	5.805E+04	**1.778E+05**
Sr-90	1.879E+03	5.804E+05	6.835E+05	1.592E+04	1.499E+05	7.479E+05	**2.179E+06**
Zr-93	1.118E-01	1.887E+01	2.372E+01	7.990E-01	6.852E+00	2.607E+01	**7.641E+01**
Nb-93m	0.000E+00	2.969E+03	5. I00E+03	2.667E+00	6.246E+02	6.730E+03	**1.543E+04**
Nb-94	6.968E-01	7.938E+01	9.852E+01	9.248E-01	2.296E+01	1.975E+02	**3.999E+02**
Tc-99	1.167E+00	1.344E+02	1.782E+02	7.817E+00	6.153E+01	1.875E+02	**5.706E+02**
Pd-107	1.949E-02	5.957E-01	1.002E+00	1.164E-01	7.634E-01	9.226E-01	**3.420E+00**
Cd-113m	2.991E+00	5.017E+02	7.469E+02	2.545E+01	1.566E+02	4.994E+02	**1.933E+03**
Sn-121m	1.646E+01	1.782E+03	2.667E+03	1.081E+02	6.738E+02	1.918E+03	**7.165E+03**
Sn-126	8.193E-02	8.085E+00	1.188E+01	5.200E-01	3.402E+00	9.161E+00	**3.313E+01**
I-129	4.542E-03	4.153E-01	5.596E-01	2.920E-02	2.079E-01	5.422E-01	**1.759E+00**
Cs-135	1.362E-01	1.393E+01	1.838E+01	8.966E-01	6.926E+00	1.970E+01	**5.997E+01**
Cs-137	5.045E+03	6.971E+05	8.957E+05	3.773E+04	2.919E+05	9.299E+05	**2.857E+06**
Sm-151	2.949E+02	2.307E+04	3.568E+04	1.939E-i-03	1.388E+04	3.189E+04	**1.068E+05**
Eu-150	9.859E-03	7.805E+01	1.069E+03	2.057E-03	1.354E+04	1.099E+02	**1.479E+04**
Eu-152	7.569E+02	l.151E+04	2.970E+04	1.703E+03	3.634E+04	7.083E+04	**1.508E+05**
Eu-154	2.622E+02	7.099E+03	1.327E+04	9.090E+02	2.968E+04	5.480E+04	**1.060E+05**
Ho-166m	2.024E+00	3.083E+01	2.892E+01	3.354E+00	2.665E+01	5.514E+01	**1.469E+02**
Th-232	1.196E-01	1.147E+0l	2.319E+01	6.757E-01	5.969E+00	1.752E+01	**5.895E+01**
U-232	l.027E-02	8.730E+01	1.738E+02	9.188E-01	9.004E+01	3.690E+02	**7.211E+02**
U-233	1.334E-03	6.508E+01	1.176E+02	1.107E+01	1.202E+02	1.525E+02	**4.664E+02**
U-234	4.3 16E-01	1.42 1E+02	1. 179E+02	1 .037E+01	1 .648E+02	2.8 14E+02	**7. 169E+02**
U-235	8.570E-03	1.293E+00	l.343E+00	1.717E-01	2.557E+00	3.220E+00	**8.593E+00**
U-236	2.995E-03	2.213E+00	2.647E+00	1.483E-01	9.123E-01	3.458E+00	**9.381E+00**
U-238	9.507E-02	6.826E+00	1.250E+01	6.919E-0l	8.674E+00	1.570E+01	**4.449E+01**
Np-237	1.379E-02	1.196E+0l	2.476E+01	6.027E-02	1.140E+00	1.072E+01	**4.865E+01**
Pu-238	3.232E+02	2.857E+03	4.768E+03	2.659E+03	1.774E+04	1.115E+04	**3.950E+04**
Pu-239	1.415E+03	7.684E+03	1.262E+04	1.085E+04	9.997E+04	2.746E+04	**1.600E+05**
Pu-240	3.489E+02	2.041E+03	4.405E+03	2.763E+03	2.532E+04	7.045E+03	**4.193E+04**
Pu-241	4.408E+03	2.946E+04	6.952E+04	4.315E+04	3.415E+05	1.034E+05	**5.914E+05**
Pu-242	2.882E-02	1.367E+00	2.279E+00	3.962E-01	7.485E+00	4.621E+00	**1.618E+01**
Am-241	5.022E+02	1.299E+03	3.567E+03	2.555E+03	2.309E+04	6.088E+03	**3.710E+04**
Am-243	0.000E+00	1.203E-02	1.772E-01	7.900E-01	2.682E+00	3.416E+00	**7.078E+00**
Cm-244	0.000E+00	1.190E+03	2.197E+03	4.961E+01	1.586E+03	2.506E+03	**7.529E+03**
Total	**1.901E+05**	**1.920E+07**	**6.086E+07**	**8.867E+05**	**1.578E+07**	**3.523E+07**	**1.321E+08**

TABLE VI

Radionuclide Summary in **Atoms** Decay Corrected to September 23, 1992
for the Six Principal Geographic Test Centers

Radio-nuclide	Frenchman Flat	Pahute Mesa-19	Pahute Mesa-20	Rainer Mesa Shoshone Mt	Yucca Flat - Above	Yucca Flat - Below	**Total**
H-3	3.620E+24	3.689E+26	1.225E+27	1.586E+25	3.055E+26	6.881E+26	**2.607E+27**
C-14	6.405E+23	2.111E+24	4.518E+24	1.061E+24	1.094E+25	8.076E+24	**2.735E+25**
Al-26	8.413E+21	1.073E+21	1.001E+22	5.439E+20	6.665E+22	4.300E+22	**1.297E+23**
Cl-36	4.516E+24	4.618E+25	7.973E+25	5.730E+24	5.899E+25	1.171E+26	**3.122E+26**
Ar-39	2.794E+21	2.899E+23	5.652E+23	1.660E+22	1.452E+23	4.328E+23	**1.452E+24**
K-40	3.527E+27	3.398E+29	6.783E+29	1.975E+28	1.758E+29	5.181E+29	**1.735E+30**
Ca-41	1. 135E+25	8.763E+25	2.208E+26	1.225E+25	1.483E+26	2.882E+26	**7.685E+26**
Ni-59	2.092E+23	2.043E+24	3.810E+24	2.587E+23	2.738E+24	5.460E+24	**1.452E+25**
Ni-63	2.828E+22	2.904E+23	5.266E+23	3.568E+22	3.932E+23	8.808E+23	**2.155E+24**
Kr-85	2.329E+21	9.028E+23	1.034E+24	2.437E+22	2.060E+23	1.052E+24	**3.222E+24**
Sr-90	9. 109E+22	2.8 14E+25	3.3 14E+25	7.7 17E+23	7.265E+24	3.626E+25	**1 .057E+26**
Zr-93	2.825E+23	4.767E+25	5.993E+25	2.019E+24	1.731E+25	6.587E+25	**1.931E+26**
Nb-93m	0.000E+00	8.053E+22	1.383E+23	7.234E+19	1.694E+22	1.825E+23	**4.184E+23**
Nb-94	2.347E+22	2.674E+24	3.3 19E+24	3.1 16E+22	7.734E+23	6.652E+24	**1.347E+25**
Tc-99	4.189E+23	4.821E+25	6.394E+25	2.805E+24	2.208E+25	6.727E+25	**2.047E+26**
Pd-107	2.135E+23	6.523E+24	1.097E+25	1.274E+24	8.359E+24	1.010E+25	**3.744E+25**
Cd-113m	7.104E+19	1.192E+22	1.774E+22	6.044E+20	3.719E+21	1.186E+22	**4.591E+22**
Sn-121m	1.525E+21	1.651E+23	2.470E+23	1.001E+22	6.243E+22	1.777E+23	**6.638E+23**
Sn-126	3.450E+22	3.405E+24	5.002E+24	2.190E+23	1.433E+24	3.858E+24	**1.395E+25**
I-129	1.201E+23	1.098E+25	1.480E+25	7.723E+23	5.498E+24	1.434E+25	**4.65 IE+25**
Cs-135	5.277E+23	5.397E+25	7.120E+25	3.474E+24	2.683E+25	7.633E+25	**2.323E+26**
Cs-137	2.555E+23	3.53 1E+25	4.537E+25	1.91 1E+24	1.478E+25	4.7 10E+25	**1.447E+26**
Sm-151	4.471E+22	3.498E+24	5.409E+24	2.940E+23	2.105E+24	4.835E+24	**1.619E+25**
Eu-150	5.979E+17	4.733E+21	6.483E+22	1.247E+17	8.209E+23	6.664E+21	**8.972E+23**
Eu-152	1.726E+22	2.626E+23	6.774E+23	3.885E+22	8.288E+23	1.615E+24	**3.440E+24**
Eu-154	3.796E+21	1.028E+23	1.921E+23	1.316E+22	4.297E+23	7.932E+23	**1.535E+24**
Ho-166m	4.092E+21	6.231E+22	5.846E+22	6.781E+21	5.387E+22	1.1 15E+23	**2.970E+23**
Th-232	2.821E+27	2.706E+29	5.468E+29	1.594E+28	1.408E+29	4.133E+29	**1.390E+30**
U-232	1.208E+18	1.026E+22	2.044E+22	1.080E+20	1.059E+22	4.338E+22	**8.478E+22**
U-233	3.578E+20	1.745E+25	3.154E+25	2.969E+24	3.223E+25	4.090E+25	**1.251E+26**
U-234	1.788E+23	5.888E+25	4.885E+25	4.298E+24	6.828E+25	1.166E+26	**2.971E+26**
U-235	1.016E+25	1.533E+27	1.592E+27	2.036E+26	3.032E+27	3.819E+27	**1.019E+28**
U-236	1.181E+23	8.730E+25	1.044E+26	5.851E+24	3.599E+25	1.364E+26	**3.701E+26**
U-238	7.159E+26	5.140E+28	9.41 1E+28	5.210E+27	6.531E+28	1.182E+29	**3.350E+29**
Np-237	4.969E+22	4.310E+25	8.923E+25	2.172E+23	4.108E+24	3.863E+25	**1.753E+26**
Pu-238	4.775E+22	4.220E+23	7.043E+23	3.929E+23	2.621E+24	1.647E+24	**5.835E+24**
Pu-239	5.744E+25	3.1 19E+26	5. 123E+26	4.406E+26	4.058E+27	1.1 15E+27	**6.495E+27**
Pu-240	3.855E+24	2.256E+25	4.867E+25	3.053E+25	2.798E+26	7.785E+25	**4.633E+26**
Pu-241	1.069E+23	7.145E+23	1.686E+24	1.047E+24	8.284E+24	2.508E+24	**1.435E+25**
Pu-242	1.821E+22	8.637E+23	1.440E+24	2.503E+23	4.728E+24	2.919E+24	**1.022E+25**
Am-241	3.661E+23	9.468E+23	2.600E+24	1.863E+24	1.683E+25	4.437E+24	**2.704E+25**
Am-243	0.000E+00	1.493E+20	2.200E+21	9.807E+21	3.330E+22	4.241E+22	**8.787E+22**
Cm-244	0.000E+00	3.629E+22	6.700E+22	1.513E+21	4.836E+22	7.641E+22	**2.296E+23**
Total	**7.158E+27**	**6.646E+29**	**1.324E+30**	**4.164E+28**	**3.901E+29**	**1.057E+30**	**3.483E+30**

DISCUSSION

The total underground radionuclide inventory for the NTS provides a reasonable estimate of residual radioactive products present after nearly four decades of nuclear testing. The source term inventory is unique for three reasons. First, this compilation provides a quantified radionuclide source term specifically derived from each test. Second, the NTS radionuclide inventory includes longer-lived radionuclides of lesser importance for test diagnostics. Third, the inventory incorporates a significant portion of radionuclides introduced from natural sources in addition to those expected from anthropogenic sources.

As mentioned earlier, radionuclide totals for the NTS represent an upper limit of radionuclides potentially available for transport. The radionuclide source term will never be transported in its entirety; the hydrologic source term comprises those species that are dissolved in or transportable by groundwater. The mobility of radionuclides is moderated both by chemical kinetics and hydrology. Numerous experimental and field studies accompanying the nuclear test program (see Borg *et al.*, 1976, Nimz and Thompson, 1992, and Smith, 1998) indicate, in general, long-lived radionuclides (actinides, fission products) are relatively insoluble in NTS groundwater. Furthermore, these are attenuated during transport by ion-exchange and surface reactions with sorptive minerals—particularly zeolites—characteristic of the NTS volcanic stratigraphy. With the exception of tritium, which is efficiently dispersed in groundwater as molecular HTO, the hydrologic source term will be less than the radionuclide source term. Approximately two thirds of the devices fired at the NTS were detonated above the static water level and probably will not substantively contribute to the hydrologic source term. Finally, the hydrology of the NTS consists of a complex and variably transmissive stratigraphy of regional Paleozoic carbonate (transmissivities up to 10^5 gallons/day/foot) and overlying Tertiary volcanic (transmissivities of up to 10^4 gallons/day/foot) aquifers divided by prevailing flow directions and by discharge areas into three groundwater subbasins. While a comprehensive description of radionuclide migration and NTS hydrology is outside the scope of this report, radionuclide transport likely will not occur uniformly away from the testing centers at the NTS. Different geologic and hydrologic properties will greatly affect potential radionuclide transport.

The source term summed over 828 underground nuclear tests provides radionuclide totals for five principal geographic test centers. By summing radionuclide totals for individual tests into regional areas, radionuclide production from an individual test is integrated into the sum for that region. The effect, particularly for tests with higher yield, is to minimize the contribution from any one test. Because the radionuclide source term is device dependent, local contributions will be disproportionately skewed toward the regional average. Geographic divisions were made arbitrarily and should not be treated as mutually exclusive; ideally, each test should be considered individually relative to both yield and specific radionuclide production.

The radionuclide inventory is dominated by residual radioactivity introduced by nuclear testing, but also includes a significant proportion of natural radioactivity. Volcanic tuffs

and rhyolites erupted from volcanic centers over the past 15 million years are highly evolved rocks that contain oxides of uranium, thorium, and potassium—all of which are naturally radioactive. Past studies (Olsen, 1967) on core samples from underground nuclear tests indicate that 700 tons of geologic media are melted per kiloton of nuclear explosive yield. This value was recently confirmed by examining the radiochemical diagnostic data for the most device-debris-rich core sample from each of 18 LLNL tests between 1970 and 1988. Therefore, natural ^{40}K, natural ^{232}Th, and natural uranium activities are mixed into the melt created by all the tests in each of the five principal geographic test centers. The average potassium, thorium, and uranium concentrations of NTS working point media are 4%, 22 ppm, and 3.7 ppm by weight, respectively. Using these values, we calculate that 0.0229 Ci of ^{40}K, 0.00169 Ci of ^{232}Th, and 0.00177 Ci of uranium are incorporated into the melt per kiloton of yield. We include this contribution in our totals for each for the five principal geographic test centers defined for the inventory. Background radioactivity from naturally occurring radionuclides constitutes an effective lower limit for monitoring or remedial levels proposed for the NTS.

FUTURE WORK

It should be recognized that the database represented by this work is an evolving entity, subject to improvements and corrections as more sophisticated interpretations evolve and additional information is gleaned from the historic record. We expect, with continued support, that the values of the underground nuclide inventory will improve. Large-scale changes in the sums are not expected.

LANL expects to actively maintain the database, commensurate with the resources available. Routine communication of updates to LLNL concerning changes in the database will occur, ensuring that the most comprehensive and current values are available at both locations. Improvements to the database structure and implementation are envisioned to facilitate greater utility and quality assurance.

Eleven underground nuclear tests sponsored by the laboratories occurred outside the boundaries of the NTS, but within the boundaries of the United States. These offsite tests are not included in this inventory. Residual radionuclide inventories for these tests are available as a separate classified report (Goishi, *et al.*, 1995).

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