

## WHAT WOULD HAPPEN IF HIGH-LEVEL NUCLEAR WASTES WERE STORED NEAR THE SURFACE OF THE EARTH

H. Lawroski, N. A. Chipman, W. A. Freeby, and G. G. Simpson  
Allied Chemical Corporation  
Idaho National Engineering Laboratory  
Idaho Falls, Idaho

W. A. Rodger and R. L. Frendberg  
Nuclear Safety Associates  
Bethesda, Maryland

### INTRODUCTION

In the evaluation of long-term management of defense high-level radioactive wastes, studies are being made to assess the potential environmental impacts of various options. These include several waste forms and storage or repository concepts.

The work described here is an abbreviated version of a much more comprehensive statement originally scheduled for publication as a draft by DOE in December 1978. The evaluation represents an extensive effort of DOE contractors and is not designed to establish a position for waste management. However, some potential solutions and their calculated impacts are presented.

The high-level wastes discussed in this paper are those generated by the recovery of highly enriched uranium fuels (e.g., aluminum, zirconium, and stainless-steel-clad elements). The wastes include first-, second-, and third-cycle raffinates from the Idaho Chemical Processing Plant (ICPP) as well as concentrated intermediate level liquid wastes generated at the Idaho National Engineering Laboratory (INEL).

At present, the high-level liquid wastes (HLLW) are held in interim storage in stainless steel tanks. The earliest tanks have been in use for some 20 to 25 years with minor indication of corrosion or deterioration.<sup>1</sup> Approximately 2.5 million gallons of high-level wastes are being held for solidification.

Major processes at the ICPP involve fuel receipt and storage, dissolution and extraction for uranium recovery, interim liquid waste storage, and, finally, calcination of the high-level liquid wastes into granular-powder solids for storage in stainless steel bins within reinforced concrete vaults. A simplified diagram of the fuel reprocessing sequence is shown in Fig. 1.

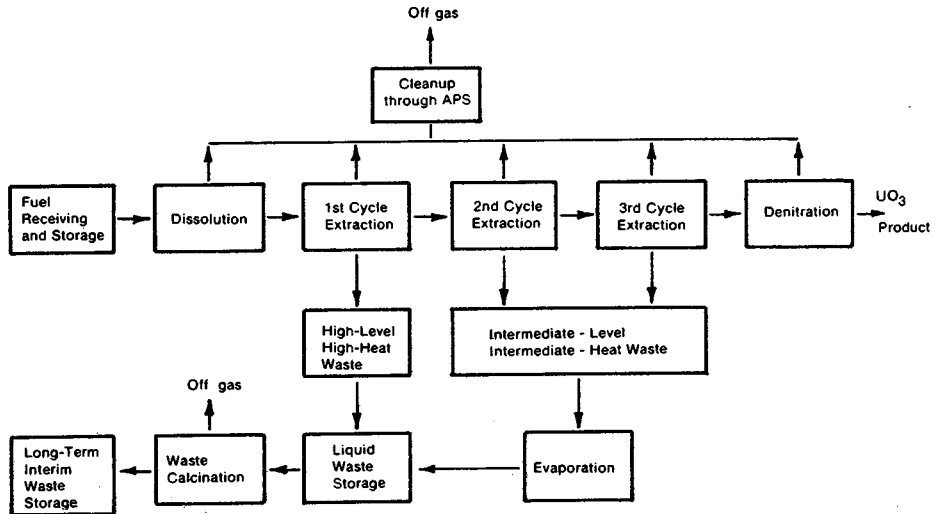


FIG. 1. DIAGRAM OF FUEL REPROCESSING SEQUENCE

ACC-A-3370

The fuel is brought to the plant in shielded casks as assemblies or portions of assemblies sized so that dissolution can be performed without dissection or further reduction in physical size. The spent fuel is stored under water (except for a complement of graphite fuels, stored dry). Charges of fuel are dissolved by acid, either nitric or hydrofluoric, depending on the fuel cladding. In specific cases, catalysts or an electrolytic dissolver are used to enhance the dissolution process. The spent graphite fuels will be burned, and the residues will be dissolved to recover and separate the enriched uranium. The acid raffinates from the extraction processes are transferred to the stainless steel tanks noted above. These high-level liquid wastes are then calcined in a fluidized bed at about 400 to 500°C to drive off the  $\text{NO}_x$  and water to form oxide calcines. The decay heat loads of the processed wastes are matched to the calcine bin design to prevent excess calcine temperatures, thereby preventing undesirable sintering. The calcines are now being held in interim storage until the final decision is made for disposal.

#### COMPOSITION OF WASTES

The high-level liquid wastes generated at the ICPP consist of the liquid waste from the uranium-fission product extraction cycles. The raffinate from the first-cycle extraction column contains about 99.9% of the radioactive fission products. The radionuclides of major concern in this waste are isotopes of cesium, strontium, cerium, uranium, plutonium, and other transuranics. Toxic chemicals containing mercury, cadmium, and fluorides are also present in the waste.

The actual composition of the wastes varies greatly, depending on the type of fuel being reprocessed, the irradiation level of the fuel, and the length of time the fuel is stored prior to reprocessing. Due to the high decay heat from the fission products, first- and second-cycle raffinates are stored in cooled tanks to prevent self-boiling. Raffinate from the third-cycle extraction is also classified as HLLW but does not require cooling; it is stored in the same tanks as the waste from the process equipment waste system.

The wastes (primarily nitrates and fluorides) are kept in an acid environment. Additional radioactive liquid wastes are also generated from decontamination and analytical activities at the ICPP. These wastes are concentrated by evaporation in the process equipment waste system.

The liquid wastes are calcined and reduced in volume by a factor of 7 to 8 resulting in a less mobile waste form. In the calcination process, HLLW is sprayed into a heated, fluidized bed of solidified waste granules. The granules are kept in movement (fluidized) by streams of heated air to provide efficient heat transfer. The granules are maintained at an operating temperature of 500°C (932°F) by the combustion of kerosene within the fluidized bed. This supplies the heat required to evaporate water and decompose nitrates to oxides. The HLLW, consisting mainly of nitrates and fluoride salts, is calcined into such solid compounds as  $Al_2O_3$ ,  $CaF_2$ ,  $ZrO_2$ , and  $Fe_2O_3$ .

In Table I are shown the properties of the typical calciner product. Water vapor, oxides of nitrogen, and other volatiles leave with the calciner off-gas. The calcine is withdrawn continuously from the bed and transferred pneumatically to the calcined solids storage facility (CSSF) in heavy-walled stainless steel lines buried in concrete. The calciner off-gas is cleaned in the process off-gas cleanup system and sent through the atmospheric protection system before release through the ICPP stack.

#### STORAGE OF CALCINE

The calcine is presently stored in stainless steel bins. The CSSF consists of vertical stainless steel bins within reinforced concrete vaults anchored in bedrock. There are three facilities in use; the first and second are filled, and the third is being filled. Construction of a fourth is complete, the fifth is in advanced design, and the sixth and seventh are in early design. The present and future facilities are compared in Table II.

The design, whether annular or cylindrical, is determined by the decay heat characteristics of the calcine to be stored. These calcines with higher heat loads are stored in annular-configuration bins to maintain temperatures well below sintering. Cutaway views of the two types of bins are shown in Fig. 2.

The structural capabilities of the bins and vaults have been studied with respect to a hypothetical earthquake used for design of INEL facilities. This study indicated that neither the concrete vault structure nor the bins would be damaged by the severe ground acceleration accompanying such an earthquake. Other postulated natural disasters such as floods or tornadoes were examined for potential effects, and the integrity of the CSSF would be maintained. Corrosion data indicate that the bins have a serviceable lifetime of well over 500 years.

TABLE I  
TYPICAL PROPERTIES OF CALCINER PRODUCT

<u>Physical Properties</u>	<u>Aluminum Waste</u>	<u>Zirconium Waste</u>	<u>Future Waste</u>
Mass median particle diam, mm	0.56 to 0.70	0.6 to 0.8	0.6 to 0.8
Bulk density, g/cm <sup>3</sup>	1.0 to 1.2	1.7	1.3
Composition, wt%			
Zirconium as ZrO <sub>2</sub>	---	21.4	*
Calcium as CaF <sub>2</sub>	---	54.2	*
Aluminum as Al <sub>2</sub> O <sub>3</sub>	88.2 to 89.1	21.9	*
Sodium as Na <sub>2</sub> O	1.3 to 2.0	---	*
Nitrogen as N <sub>2</sub> O <sub>5</sub>	3.9 to 4.1	1.9	*
Mercury as HgO	2.9	---	*
Water	2.0	0.6	*
Gross fission-product oxides	0.6	---	*
Radioactive Properties:			
Heat generation, Btu/hr-kg	0.14 to 0.36	0.05 to 0.09	1.2
Principal radioisotopes, Ci/kg			
<sup>90</sup> Sr	2.23 to 3.3	0.36 to 1.14	13
<sup>134</sup> Cs	0.09 to 0.18	---	3.3
<sup>137</sup> Cs	2.4 to 3.6	0.45 to 1.4	13
<sup>144</sup> Ce	0.6 to 7.9	---	8.2
<sup>106</sup> Ru	0.02	0.004 to 0.01	0.97
<sup>95</sup> Zr- <sup>95</sup> Nb	0 to 0.23	---	---
<sup>147</sup> Pm	1.5 to 4.8	---	12
<sup>238</sup> Pu	~0.002	~0.01	0.07
<sup>239</sup> Pu	~0.0004	~0.0001	0.0007
<sup>240</sup> Pu	~0.0001	~0.00006	0.00065

\*The chemical composition of future calcine will be similar to the composition of zirconium waste.

TABLE II  
CHARACTERISTICS OF THE CALCINE SOLIDS STORAGE FACILITIES

<u>CSSF</u>	<u>No. of Bins</u>	<u>Total Volume</u>	<u>Bin Description</u>	<u>Retrieval</u>	<u>Vault Shape</u>	<u>Cooling</u>
1	4	220 m <sup>3</sup>	Annular	No retrieval access provided	Rectangular	Forced or natural convection
2	7	850 m <sup>3</sup>	Cylindrical	Retrieval access provided	Cylindrical	Natural convection
3	7	1130 m <sup>3</sup>	Cylindrical	Retrieval access provided	Cylindrical	Natural convection
4	3	480 m <sup>3</sup>	Cylindrical	Retrieval access provided	Cylindrical	Natural convection
5*	7	990 m <sup>3</sup>	Annular	Retrieval access provided	Cylindrical	Natural convection
6	7	990 m <sup>3</sup>	Annular	Retrieval access provided	Cylindrical	Natural convection
7	Design basis not complete					

\*Based on a conceptual design.

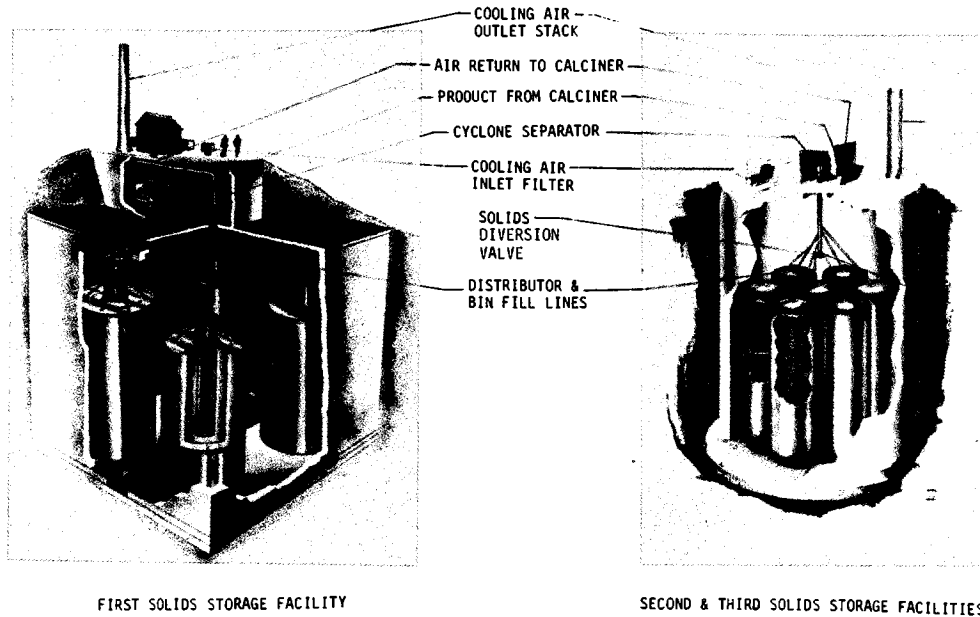


FIG. 2. CUTAWAY VIEWS OF THE HIGH-LEVEL SOLIDS STORAGE FACILITIES

## MAJOR ALTERNATIVES FOR LONG-TERM DISPOSAL

A programmatic assessment of environmental impacts associated with reasonably available options for the ultimate disposition of the waste was made. The six major candidates were selected by evaluating some 13 alternatives presented in ERDA 77-43.<sup>2</sup> It should be realized that many other alternatives were examined and were considered less desirable. The major candidate long-range waste management options are:

- Option 1: Continue current operations, which is disposal of calcine in the CSSF at the INEL.
- Option 2: Convert the calcine to pellets and dispose of the pellets at the INEL.
- Option 3: Convert the calcine to glass and dispose of glass at the INEL.
- Option 4: Remove actinides from the wastes, convert the actinides to a glass, dispose of glass at an off-site Federal repository and dispose of actinide-depleted waste at the INEL.
- Option 5: Stabilize the calcine by removing remaining nitrates and water, and dispose of calcine at an off-site Federal repository.
- Option 6: Convert the calcine to a glass and dispose of the glass at an off-site Federal repository.

There are variations of the above options that could enhance long-term characteristics of disposed waste. For example, in Option 4, instead of converting the actinides into a glass, the actinides could be fissioned in a high-neutron energy reactor. The fission process would produce power and at the same time destroy the long-half-life actinides. Recent calculations<sup>3</sup> on actinide burnup made by scientists at the University of Arizona indicate that the neutronics are quite favorable.

The six above options were analyzed to estimate possible individual and population exposures if any one of these six options were implemented. The exposures were calculated for incremental periods up to 100 million years after processing.

### METHODOLOGY

Each of the options were examined for potential scenarios that might result in pathways to man. The basic scenarios were:



- (1) Operational releases, either routine or accidental
- (2) Migration, such as leaching of isotopes from the disposed waste to a water source and subsequent use of the water
- (3) Intrusion into the waste, such as by an archaeologist or other person(s).

The scenarios of the pathways to man are illustrated in Fig. 3. Both radioactive and nonradioactive isotopes were considered.

For the computed doses to individuals, the methodology and parameters are contained in Regulatory Guide 1.109. The estimates were made for adults only because refinement among age groups was not warranted primarily due to the comparative nature of the study. The doses calculated were 50-year dose commitments from exposures received during one year. To account for pathways where radioactivity may build up during interim storage, the doses were estimated for the last year of ICPP operation. Doses were calculated for each option, for each pathway, and for time periods extending out to 100 million years. Decay schemes and chronologically ambient isotopic inventories were used in all computations.

As part of this evaluation, estimates were made on population doses to those within 50 miles of the INEL site. Realize that for some scenarios, the calculated values apply to doses for only a limited number of people and will not apply to widespread population exposures. Further, some of the scenarios could result in smaller doses to a large number of people. In the calculations of population doses by these modes, population distribution was taken into account. Where the maximum individual receptor is close to the point of discharge of an airborne release, the average dose to individuals within 50 miles can be reasonably approximated by using a value of 1% of the maximum individual dose. A more rigorous treatment (not justified in these computations) requires the matching of site meteorology and population distribution. Table III shows the modes of population exposure that were considered for each pathway. For those exposure modes that result only in doses to individuals, assumptions were made as to the number of individuals involved to estimate the size of the population for that pathway and time period. Additional assumptions needed to carry out these calculations are shown in Table IV.

The extrapolation of population and utilization of area-wide meteorology presented some difficulty for application to the

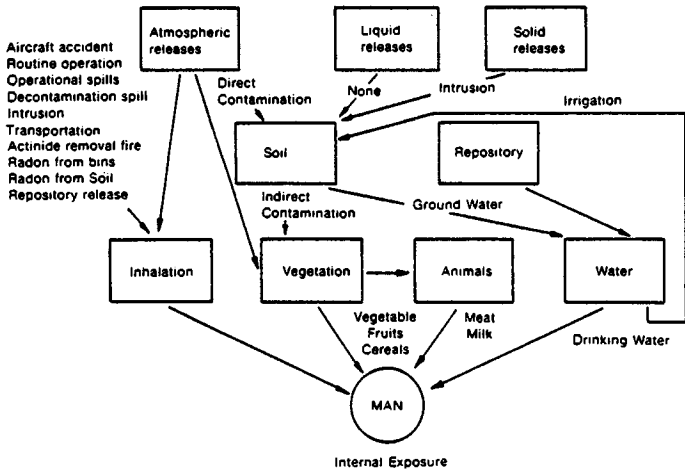
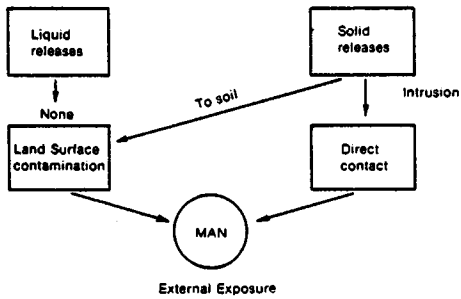


FIG. 3. PATHWAYS TO MAN

TABLE III  
MODES OF POPULATION EXPOSURE

<u>Pathway</u>	Results in Exposure to Population		
	<u>Individuals</u>	<u>Groups</u>	<u>Workers</u>
Operational Releases:			
Routine operations		X	
Spillage of calcine		X	
Decontamination spills		X	
Fire		X	
Transport to repository	X	X	X
Occupational exposure			X
Migrational Losses:			
Leaching into groundwater		X	
Radon emanation from bin	X		
Intrusions:			
Airplane accident		X	
Individual intruder			
Inhalation	X		
Direct Exposure	X		
Contaminated ground			
Food	X		
External exposure	X		
Radon	X		

TABLE IV  
 ASSUMPTIONS USED TO CALCULATE POPULATION DOSES  
 DUE TO EXPOSURE OF INDIVIDUALS

<u>Pathway</u>	<u>No. of Individuals Involved</u>	<u>Events Per Year</u>
Groundwater	5	0.01
Intrusion	10	0.01
Food	5	0.0001
External radiation	5	0.0001
Radon - Bin	5	0.01
Radon - Soil	5	0.00001

scenarios. The population was scaled up linearly by a factor of 5 over the next 150 years and was assumed to be constant thereafter. Since the source terms were particulates, depleted dispersion factors were used. A deposition velocity of 0.01 m/s was utilized to determine deposition of particulates.

#### SOURCE TERM

The source term for the calculations was determined by considering the inventories of all important isotopes in the wastes as a function of time. These included solid and liquid wastes to be calcined through the year 2020. At the base year of 2020, the total quantity of wastes is  $2 \times 10^7$  kg of equivalent calcine. The concentrations of radionuclides used in all these calculations are shown in Table V.

A computer code was utilized to calculate concentrations as a function of time. The WALTERS<sup>4</sup> code accumulates and decays all of the parent radionuclides, allows for the in-growth of daughter products and for each time period of interest gives for each nuclide the total quantity (curies) and concentration ( $\mu\text{Ci}/\text{kg}$  of equivalent calcine). The code also gives the average quantity and concentration integrated over that time period. It is the latter set of values that is used in making calculations for any particular time period.

As a matter of practical interest, when an existing radioactive nuclide (parent) decays, it frequently produces one or more subsequent nuclides (daughters) that are themselves radioactive. Some of these radioactive daughters are more toxic than their parents. Thus, in determining the inventories of radionuclides as a function of time, it is necessary to consider the buildup (and decay) of the daughter products.

#### PATHWAYS

Evaluations were made for the more conceivable types of releases from the facility. Operational releases, migrational losses, and intrusional modes were considered. The parameters used were based on actual operational experience, Regulatory Guide 1.109, reported soil data, and other applicable references. These references and the details of the calculations can be found in Appendices B and C of the *Draft Environmental Impact Statement, Long-Term Management of Defense High-Level Radioactive Waste, Idaho Chemical Processing Plant* (May 1979) (base document).

TABLE V

INITIAL RADIONUCLIDE CONCENTRATIONS OF INPUT SOLIDS WASTES,<sup>5</sup> ALL OPTIONS

Nuclide	Activity (Ci/kg)	Nuclide	Activity (Ci/kg)	Nuclide	Activity (Ci/kg)
<sup>79</sup> Se	6.4E-05*	<sup>87</sup> Rb	3.6E-09	<sup>90</sup> Sr	1.3E+01
<sup>90</sup> Y	1.3E+01	<sup>93</sup> Zr	3.1E-04	<sup>93m</sup> Nb	7.5E-05
<sup>99</sup> Tc	2.1E-03	<sup>106</sup> Ru	9.7E-01	<sup>106</sup> Rh	9.7E-01
<sup>107</sup> Pd	2.0E-06	<sup>126</sup> Sn	3.2E-05	<sup>126m</sup> Sb	3.2E-05
<sup>126</sup> Sb	3.2E-05	<sup>134</sup> Cs	3.3	<sup>135</sup> Cs	7.5E-05
<sup>137</sup> Cs	1.3E+01	<sup>137m</sup> Ba	1.2E+01	<sup>144</sup> Ce	8.2
<sup>144</sup> Pr	8.2	<sup>144</sup> Nd	0.0	<sup>147</sup> Pm	1.2E+01
<sup>147</sup> Sm	0.0	<sup>151</sup> Sm	1.7E-01	<sup>154</sup> Eu	1.8E-01
<sup>226</sup> Ra	0.0	<sup>230</sup> Th	0.0	<sup>233</sup> Pa	0.0
<sup>233</sup> U	1.2E-12	<sup>234</sup> U	4.3E-10	<sup>235</sup> U	1.8E-09
<sup>236</sup> U	1.0E-08	<sup>237</sup> U	4.8E-12	<sup>238</sup> U	1.0E-14
<sup>237</sup> Np	4.8E-08	<sup>239</sup> Np	0.0	<sup>238</sup> Pu	7.0E-02
<sup>239</sup> Pu	7.0E-04	<sup>240</sup> Pu	6.5E-04	<sup>241</sup> Pu	1.6E-01
<sup>242</sup> Pu	1.8E-06	<sup>241</sup> Am	9.8E-04	<sup>243</sup> Am	8.3E-06
<sup>242</sup> Cm	6.5E-04	<sup>244</sup> Cm	5.2E-04		
Total	8.5E+01				

$$*6.4E-05 = 6.4 \times 10^{-5} = 0.000064$$

For operational releases, the following pathways were examined:

- |                            |                                  |
|----------------------------|----------------------------------|
| (1) Routine releases       | (4) Fire in cell                 |
| (2) Spillage of calcine    | (5) Occupational exposure        |
| (3) Decontamination spills | (6) Transportation to repository |

It should be remembered that in all cases evaluated, the starting point was calcine in stainless steel bins housed in reinforced concrete vaults just below ground.

Migrational losses considered were as follows:

- (1) Groundwater migration
- (2) Radon emanation

In examining potential effects of migrational losses due to groundwater effects, it was assumed that the stainless steel bins, the surrounding concrete, and the reinforced concrete vault will have deteriorated to the extent that incident rainwater could enter the bins and come in contact with the stored waste. The vertical distance to the aquifer is about 140 m. From logs of well drilling near the ICPP, there are several layers of lava and soil of which there is a total of approximately 105 to 120 m of lava with the remainder being soil. For calculational purposes, a depth of 15 m of soil was considered for ion exchange capability. A cutaway of the site location is shown in Fig. 4. More details of the assumptions and calculations are in the base document noted previously.

In the evaluation of radon emanation, it was assumed that sometime in the future, homes are built unknowingly directly over the buried CSSF. An additional assumption was that if a home were built on soil that has elevated concentrations of radon coming from it, the gas can be kept inside the dwelling long enough to allow daughter products of radon to build up, resulting in increased lung doses.

Intrusional modes included the following:

- (1) Airplane accident
- (2) Physical intrusion

In the airplane accident calculation, the event was postulated to occur prior to the year 2100. The rationale behind this assumption was that prior to 2100 A.D., decay heat from the stored waste precluded entombment of the bins in concrete. Further, no additional

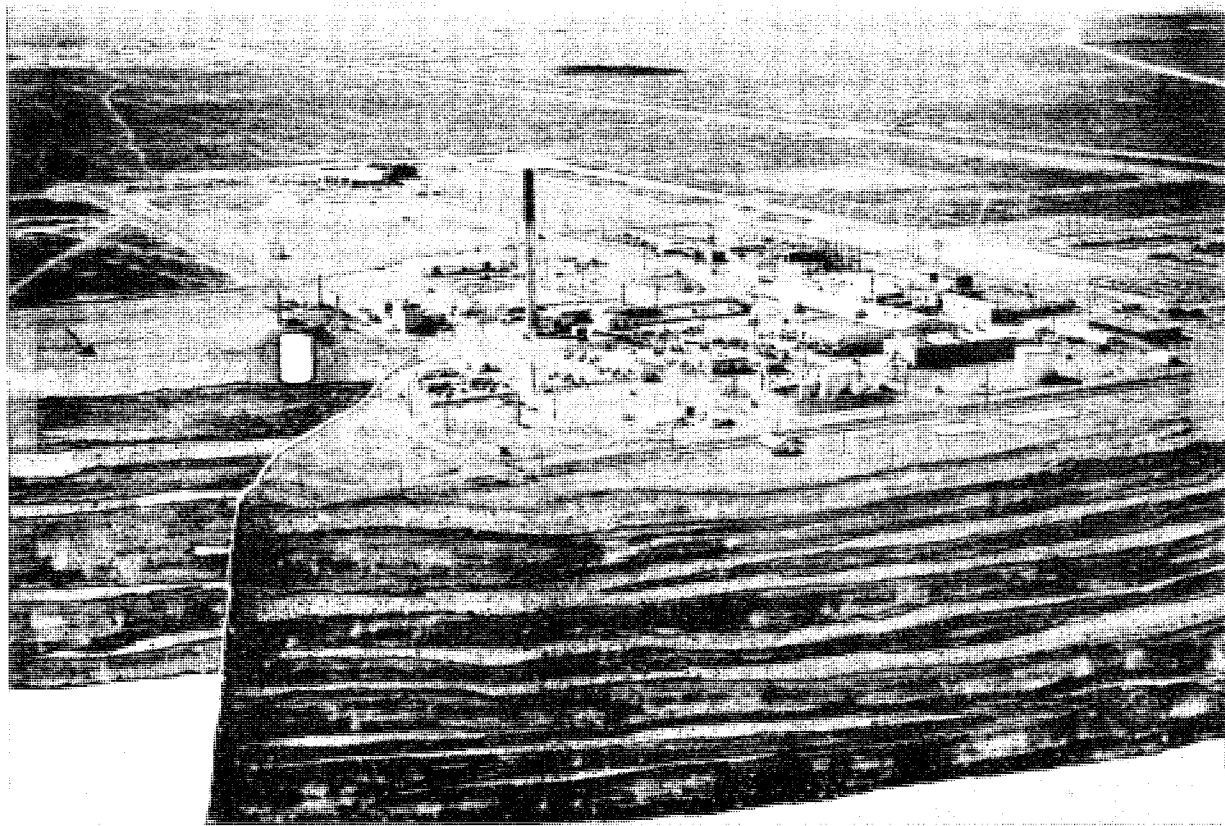


FIG. 4. CUTAWAY OF SOIL STRATA BENEATH THE ICPP BINS.

INEL-S-15 840



precautions were made to protect the bins by placing a berm around the shielding housing of the distribution system of the bins. Penetration of the bins due to impact was assumed to perpetrate the release of calcine. The discussed event is only of significant consequence while the waste is calcine prior to encasement or conversion to other less dispersible solid forms.

For evaluation of wastes residing for a long period of time near the surface, it must be assumed that intrusion would occur. This could be a well driller, an archaeologist, a prospector, or simply someone who is curious. Once the intruder violates the storage complex, he would be exposed to radiation in two ways: receipt of direct penetrating radiation and inhalation of dust that would contain radioactivity. Both potential exposure pathways were evaluated.

After a physical intrusion has occurred, it is likely that some of the waste will have been brought to the surface as a result of that intrusion. Subsequent residents could then be exposed to this surface contamination in at least three ways. The pathways considered are: growing of food in the contaminated soil and consumption of that food by the resident, direct exposure to penetrating radiation from the ground-plane contamination, and exposure to radon daughters in a home built on the contaminated land.

#### TIME PERIODS

For those cases where occupational exposures were calculated, the time period was from inception of the activity until it was completed, i.e., for most cases, up to 2020 A.D. The airplane accident scenario was assumed to be operating until 2100 A.D. at which time the encasement of the bins was completed.

The time periods examined for all other scenarios ended at 1,000, 2,000, 5,000, 10,000, 20,000, 50,000, 100,000, 200,000, 500,000, 1 million, 2 million, 5 million, 10 million, 20 million, 50 million, and 100 million years after encasement.

There has been considerable discussion about institutional control and what length of time could be assumed. Since the mode of storage would basically be below the surface, an arbitrary time of 1,000 years was used as a combination of institutional control and recorded history. The stainless steel bins and the encasement concrete were assumed to break down and potentially allow migrational effects to commence in about 500 years.

Note that in these evaluations there were no particular attempts to ameliorate consequences. With relatively little effort, it is conceivable to be able to reduce the calculated doses.

## RESULTS AND DISCUSSION

The calculational results of primary interest are those in which (1) the calcine might be left as is, with encasement of the bins in the vaults after the major decay heat load has been dissipated; (2) the calcine might be treated further and solidified as pellets or glass; and (3) the actinide content of the wastes might be reduced to an innocuous level. In any case, further processing beyond the present calcining results in additional exposures due to routine releases, processing spills, and transportation events.

During the period in which the stainless steel bins have untreated calcine and before they are encased in concrete, the major conceivable impact appears to be an airplane accident. This was considered because at present at least one of the vaults that houses the stainless bins extends above the ground even though the concrete vault rests on bedrock. In the postulated accident, the impacting airplane penetrates the vault and releases the calcine. The calculated maximum individual dose is estimated to be 100 rem. If protective measures were effected, such as a substantive berm around the vaults, the consequences could be substantially reduced. It should also be recognized that the probability of an airplane accident is also quite small, i.e., estimated at approximately  $2 \times 10^{-7}/\text{yr}$ .

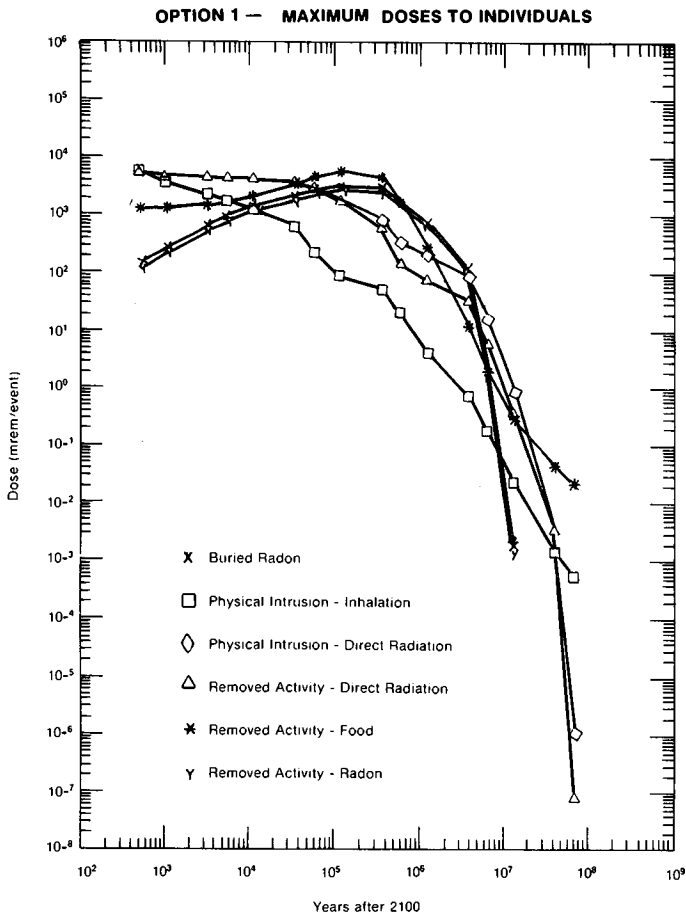
The calculated airplane accident scenario indicated the second highest dose (100 rem) to an individual. The consequences from a postulated transportation accident involving separated actinides was the highest calculated dose (180 rem) to an individual. All other scenarios portrayed significantly lower doses to individuals (less than 5 rem).

The short-term (less than 1,000 years) major isotopes were Sr-90 and Cs-137. As decay chains operate, major isotopes for each pathway change depending on mobility and method of human involvement.

Options 1, 2, 3, and 4 basically involve disposal of high-level solid wastes near the surface. The results of the calculation of doses to individuals for Option 1 are shown in Table VI. Figure 5 illustrates the calculated event consequences at various points in time. Note that physical intrusion and removed activity scenarios

TABLE VI  
PATHWAYS SUMMARY FOR LEAVING PRESENT CALCINE IN BINS ON SITE (OPTION 1)

Years Elapsed	PATHWAYS							
	Ground Water	Buried Radon	Airplane	Physical Intrusion		Direct Radiation	Removed Activity	
				Inhalation	Direct Radiation		Food	Radon
	Consequence, mR (Major Isotope)							
1 000	0.9 ( <sup>79</sup> Se)	1.2 x 10 <sup>2</sup>		4.2 x 10 <sup>3</sup> ( <sup>241</sup> Am)	3.7 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	4.2 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.0 x 10 <sup>3</sup> ( <sup>99</sup> Tc)	1.0 x 10 <sup>2</sup>
2 000	9.3 x 10 <sup>-5</sup> ( <sup>90</sup> Sr)	2.1 x 10 <sup>2</sup>		2.6 x 10 <sup>3</sup> ( <sup>239</sup> Pu)	3.3 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.7 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.0 x 10 <sup>3</sup> ( <sup>99</sup> Tc)	1.8 x 10 <sup>2</sup>
5 000	2.7 ( <sup>99</sup> Tc)	5.2 x 10 <sup>2</sup>		1.7 x 10 <sup>3</sup> ( <sup>239</sup> Pu)	3.1 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.5 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.1 x 10 <sup>3</sup> ( <sup>99</sup> Tc)	4.4 x 10 <sup>2</sup>
10 000	0	7.3 x 10 <sup>2</sup>		1.3 x 10 <sup>3</sup> ( <sup>239</sup> Pu)	3.0 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.4 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.2 x 10 <sup>3</sup> ( <sup>99</sup> Tc)	6.3 x 10 <sup>2</sup>
20 000	2.0 ( <sup>135</sup> Cs)	1.1 x 10 <sup>3</sup>		9.2 x 10 <sup>2</sup> ( <sup>239</sup> Pu)	2.9 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.2 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.6 x 10 <sup>3</sup> ( <sup>99</sup> Tc)	9.2 x 10 <sup>2</sup>
50 000	1.9 x 10 <sup>3</sup> ( <sup>226</sup> Ra)	1.6 x 10 <sup>3</sup>		4.7 x 10 <sup>2</sup> ( <sup>239</sup> Pu)	2.6 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	2.8 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	2.3 x 10 <sup>3</sup> ( <sup>99</sup> Tc)	1.3 x 10 <sup>3</sup>
100 000	0	2.0 x 10 <sup>3</sup>		1.8 x 10 <sup>2</sup> ( <sup>239</sup> Pu)	2.1 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	2.2 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.4 x 10 <sup>3</sup> ( <sup>226</sup> Ra)	1.8 x 10 <sup>3</sup>
200 000	0	2.3 x 10 <sup>3</sup>		70 ( <sup>239</sup> Pu)	1.4 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.4 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	4.1 x 10 <sup>3</sup> ( <sup>226</sup> Ra)	2.0 x 10 <sup>3</sup>
500 000	0	2.2 x 10 <sup>3</sup>		42 ( <sup>226</sup> Ra)	6.1 x 10 <sup>2</sup> ( <sup>126</sup> Sb)	4.8 x 10 <sup>2</sup> ( <sup>126</sup> Sb)	3.4 x 10 <sup>3</sup> ( <sup>226</sup> Ra)	1.9 x 10 <sup>3</sup>
1 000 000	0	1.4 x 10 <sup>3</sup>		17 ( <sup>226</sup> Ra)	2.6 x 10 <sup>2</sup> ( <sup>93</sup> Nb)	1.2 x 10 <sup>2</sup> ( <sup>93</sup> Nb)	1.3 x 10 <sup>3</sup> ( <sup>226</sup> Ra)	1.2 x 10 <sup>3</sup>
2 000 000	0	6.1 x 10 <sup>2</sup>		3.6 ( <sup>226</sup> Ra)	1.6 x 10 <sup>2</sup> ( <sup>93</sup> Nb)	64 ( <sup>93</sup> Nb)	2.2 x 10 <sup>2</sup> ( <sup>226</sup> Ra)	5.3 x 10 <sup>2</sup>
5 000 000	0	1.0 x 10 <sup>2</sup>		0.64 ( <sup>237</sup> Np)	68 ( <sup>93</sup> Nb)	27 ( <sup>93</sup> Nb)	10 ( <sup>135</sup> Cs)	90
10 000 000	0	1.7		0.17 ( <sup>237</sup> Np)	13 ( <sup>93</sup> Nb)	5.2 ( <sup>93</sup> Nb)	1.7 ( <sup>135</sup> Cs)	1.5
20 000 000	0	.002		0.02 ( <sup>237</sup> Np)	0.74 ( <sup>93</sup> Nb)	0.34 ( <sup>93</sup> Nb)	0.26 ( <sup>135</sup> Cs)	0.0017
50 000 000	0	0		0.0015 ( <sup>236</sup> U)	0.0035 ( <sup>93</sup> Nb)	0.0022 ( <sup>233</sup> Pa)	0.04 ( <sup>87</sup> Rb)	0
100 000 000	0	0		0.00059 ( <sup>235/238</sup> U)	0.00001 ( <sup>107</sup> Pd)	10 <sup>-7</sup> ( <sup>107</sup> Pd)	0.02 ( <sup>87</sup> Rb)	0



**FIG. 5. OPTION 1 EVENT CONSEQUENCES  
TO INDIVIDUALS AFTER 2100**

ACC-A-3448

present the highest calculated doses (4200 millirem). The groundwater pathway doses appear to be unusual in fluctuation. This is primarily because there is a wave of activity due to the physical phenomenon of ion exchange and time scales. In fact, the postulated doses would be somewhat smooth oscillations.

With respect to the food chain, Tc-99 appears to be the key isotope for a period of time of approximately 50,000 years for this scenario, then radium from the decayed uranium becomes the predominant isotope. It is of interest to note that only in inhalation during physical intrusion do the actinides play a major role, and even then the calculated dose is not large.

The potential long-range effects of further processing the calcine are examined in Options 2 and 3. Option 2 involves converting the calcine to pellets. Conservatively, it is estimated that leach resistance is improved by a factor of 100 resulting in reduction of most groundwater doses. The pellets would also be less susceptible to release of radioactivity to plants and subsequent food chains. A review of the food chain pathway indicates a substantial reduction in doses compared to untreated calcine. There would also be a reduction in the amount of actinides that might become airborne during an intrusion. The calculated effects for Option 2 are listed in Table VII.

Option 3 considers converting the calcine to glass and placing the glass in comparable storage to that of calcine in Option 1. In the evaluation of Option 3, the major benefits are the reductions in calculated doses from physical intrusion (inhalation) and from removed activity (food). These results are listed in Table VIII. For the scenario of an airplane accident involving glass in storage, the doses calculated were insignificant because no dispersion occurred for all practical purposes.

To obtain a comparison for what the effects of the presence of actinides might be, an evaluation of actinide-depleted calcine was made. This was part of Option 4. The results of the calculations are shown in Table IX. The apparent primary effect was to reduce the inhalation dose during physical intrusion.

As part of the overall study, probabilities were estimated for each of the pathways. Also, population doses were calculated in the conventional manner. The resulting doses were very small fractions of the natural radiation background.

Although the calculated doses vary considerably from option to option, there do not appear to be any significantly high doses indicated in any of the scenarios or pathways studied here. The studies also indicate that a limited number of fission product isotopes are key. In the food chain, it is Tc-99, and in the direct radiation pathways, Sb-126. As stated previously, Ra-226 becomes prominent after 20,000 years.

TABLE VII

PATHWAYS SUMMARY FOR CONVERTING CALCINE TO PELLETS AND LEAVING IN BINS AT SITE (OPTION 2)

Years Elapsed	P A T H W A Y S						
	Ground Water	Buried Radon	Physical Intrusion		Removed Activity		
			Inhalation	Direct Radiation	Direct Radiation	Food	Radon
	Consequence, mr (Major Isotope)						
1 000	$6 \times 10^{-3}$ ( $^{79}\text{Se}$ )	79	$3.3 \times 10^2$ ( $^{241}\text{Am}$ )	$2.9 \times 10^3$ ( $^{126}\text{Sb}$ )	$2.8 \times 10^3$ ( $^{126}\text{Sb}$ )	6.9 ( $^{99}\text{Tc}$ )	83
2 000	$5.9 \times 10^{-3}$ ( $^{79}\text{Se}$ )	140	$2.1 \times 10^2$ ( $^{239}\text{Pu}$ )	$2.6 \times 10^3$ ( $^{126}\text{Sb}$ )	$2.5 \times 10^3$ ( $^{126}\text{Sb}$ )	6.8 ( $^{99}\text{Tc}$ )	150
5 000	$3.5 \times 10^{-2}$ ( $^{99}\text{Tc}$ )	350	$1.4 \times 10^2$ ( $^{239}\text{Pu}$ )	$2.5 \times 10^3$ ( $^{126}\text{Sb}$ )	$2.3 \times 10^3$ ( $^{126}\text{Sb}$ )	7.4 ( $^{99}\text{Tc}$ )	370
10 000	$3.1 \times 10^{-2}$ ( $^{99}\text{Tc}$ )	500	$1.1 \times 10^2$ ( $^{239}\text{Pu}$ )	$2.4 \times 10^3$ ( $^{126}\text{Sb}$ )	$2.3 \times 10^3$ ( $^{126}\text{Sb}$ )	8.3 ( $^{99}\text{Tc}$ )	520
20 000	$1.3 \times 10^{-1}$ ( $^{135}\text{Cs}$ )	730	74 ( $^{239}\text{Pu}$ )	$2.3 \times 10^3$ ( $^{126}\text{Sb}$ )	$2.1 \times 10^3$ ( $^{126}\text{Sb}$ )	11 ( $^{226}\text{Ra}$ )	770
50 000	$1.8 \times 10^2$ ( $^{226}\text{Ra}$ )	$1.1 \times 10^3$	37 ( $^{239}\text{Pu}$ )	$2.0 \times 10^3$ ( $^{126}\text{Sb}$ )	$1.9 \times 10^3$ ( $^{126}\text{Sb}$ )	16 ( $^{226}\text{Ra}$ )	$1.1 \times 10^3$
100 000	0	$1.4 \times 10^3$	14 ( $^{239}\text{Pu}$ )	$1.6 \times 10^3$ ( $^{126}\text{Sb}$ )	$1.5 \times 10^3$ ( $^{126}\text{Sb}$ )	23 ( $^{226}\text{Ra}$ )	$1.5 \times 10^3$
200 000	0	$1.6 \times 10^3$	5.6 ( $^{226}\text{Ra}$ )	$1.1 \times 10^3$ ( $^{126}\text{Sb}$ )	$9.1 \times 10^2$ ( $^{126}\text{Sb}$ )	28 ( $^{226}\text{Ra}$ )	$1.7 \times 10^3$
500 000	0	$1.5 \times 10^3$	3.3 ( $^{226}\text{Ra}$ )	$4.5 \times 10^2$ ( $^{126}\text{Sb}$ )	$3.2 \times 10^2$ ( $^{126}\text{Sb}$ )	23 ( $^{226}\text{Ra}$ )	$1.6 \times 10^3$
1 000 000	0	980	1.4 ( $^{226}\text{Ra}$ )	$1.8 \times 10^2$ ( $^{93m}\text{Nb}$ )	82 ( $^{93m}\text{Nb}$ )	8.9 ( $^{226}\text{Ra}$ )	$1.0 \times 10^3$
2 000 000	0	420	0.3 ( $^{226}\text{Ra}$ )	$1.1 \times 10^2$ ( $^{93m}\text{Nb}$ )	43 ( $^{93m}\text{Nb}$ )	1.4 ( $^{226}\text{Ra}$ )	$4.4 \times 10^2$
5 000 000	0	71	0.05 ( $^{237}\text{Np}$ )	46 ( $^{93m}\text{Nb}$ )	18 ( $^{93m}\text{Nb}$ )	0.07 ( $^{135}\text{Cs}$ )	75
10 000 000	0	1.2	0.01 ( $^{237}\text{Np}$ )	8.5 ( $^{93m}\text{Nb}$ )	3.5 ( $^{93m}\text{Nb}$ )	0.01 ( $^{135}\text{Cs}$ )	1.2

TABLE VIII  
PATHWAYS SUMMARY FOR STORAGE OF CALCINE AS GLASS AT THE SITE (OPTION 3)

Years Elapsed	P A T H W A Y S						
	Ground Water	Buried Radon	Physical Intrusion		Removed Activity		
			Inhalation	Direct Radiation	Direct Radiation	Food	Radon
	Consequence, jr (Major Isotope)						
1 000	$2.2 \times 10^{-5}$ ( <sup>79</sup> Se)	5.1	14 ( <sup>241</sup> Am)	210 ( <sup>126</sup> Sb)	$2.8 \times 10^3$ ( <sup>126</sup> Sb)	0.07 ( <sup>99</sup> Tc)	83
2 000	$2.2 \times 10^{-5}$ ( <sup>79</sup> Se)	9.2	8.5 ( <sup>239</sup> Pu)	190 ( <sup>126</sup> Sb)	$2.5 \times 10^3$ ( <sup>126</sup> Sb)	0.07 ( <sup>99</sup> Tc)	150
5 000	$1.2 \times 10^{-4}$ ( <sup>99</sup> Tc)	23	5.7 ( <sup>239</sup> Pu)	180 ( <sup>126</sup> Sb)	$2.3 \times 10^3$ ( <sup>126</sup> Sb)	0.07 ( <sup>99</sup> Tc)	370
10 000	$1.1 \times 10^{-4}$ ( <sup>99</sup> Tc)	32	4.3 ( <sup>239</sup> Pu)	170 ( <sup>126</sup> Sb)	$2.3 \times 10^3$ ( <sup>126</sup> Sb)	0.08 ( <sup>99</sup> Tc)	520
20 000	$4.7 \times 10^{-4}$ ( <sup>135</sup> Cs)	47	3.0 ( <sup>239</sup> Pu)	160 ( <sup>126</sup> Sb)	$2.1 \times 10^3$ ( <sup>126</sup> Sb)	0.1 ( <sup>226</sup> Ra)	770
50 000	0.7 ( <sup>226</sup> Ra)	69	1.5 ( <sup>239</sup> Pu)	140 ( <sup>126</sup> Sb)	$1.9 \times 10^3$ ( <sup>126</sup> Sb)	0.2 ( <sup>226</sup> Ra)	$1.1 \times 10^3$
100 000	1.2 ( <sup>226</sup> Ra)	90	0.6 ( <sup>239</sup> Pu)	110 ( <sup>126</sup> Sb)	$1.5 \times 10^3$ ( <sup>126</sup> Sb)	0.2 ( <sup>226</sup> Ra)	$1.5 \times 10^3$
200 000	1.6 ( <sup>226</sup> Ra)	100	0.2 ( <sup>239</sup> Pu)	74 ( <sup>126</sup> Sb)	910 ( <sup>126</sup> Sb)	0.3 ( <sup>226</sup> Ra)	$1.7 \times 10^3$
500 000	1.4 ( <sup>226</sup> Ra)	96	0.1 ( <sup>226</sup> Ra)	30 ( <sup>126</sup> Sb)	320 ( <sup>126</sup> Sb)	0.2 ( <sup>226</sup> Ra)	$1.6 \times 10^3$
1 000 000	0.6 ( <sup>226</sup> Ra)	63	0.06 ( <sup>226</sup> Ra)	11 ( <sup>93m</sup> Nb)	82 ( <sup>93m</sup> Nb)	0.09 ( <sup>226</sup> Ra)	$1.0 \times 10^3$
2 000 000	0.09 ( <sup>226</sup> Ra)	27	0.01 ( <sup>226</sup> Ra)	6.1 ( <sup>93m</sup> Nb)	43 ( <sup>93m</sup> Nb)	0.01 ( <sup>226</sup> Ra)	440
5 000 000	0	4.6	$2 \times 10^{-3}$ ( <sup>237</sup> Np)	2.6 ( <sup>93m</sup> Nb)	18 ( <sup>93m</sup> Nb)	$7 \times 10^{-4}$ ( <sup>135</sup> Cs)	75
10 000 000	0	0.08	$6 \times 10^{-4}$ ( <sup>237</sup> Np)	0.5 ( <sup>93m</sup> Nb)	4 ( <sup>93m</sup> Nb)	$1 \times 10^{-4}$ ( <sup>135</sup> Cs)	1.2



TABLE IX

PATHWAYS SUMMARY FOR CALCINE WITH ACTINIDES REMOVED IN BINS ON SITE (OPTION 4)

Years Elapsed	PATHWAYS						
	Ground Water	Buried Radon	Physical Intrusion		Direct Radiation	Removed Activity	
			Inhalation	Direct Radiation		Food	Radon
	Consequence, mr (Major Isotope)						
1 000	0.9 ( <sup>79</sup> Se)	2.3	6.6 ( <sup>99</sup> Tc)	3.2 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.5 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.0 x 10 <sup>3</sup> ( <sup>99</sup> Tc)	2.0
2 000	9.3 x 10 <sup>-5</sup> ( <sup>90</sup> Sr)	4.2	6.3 ( <sup>99</sup> Tc)	3.2 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.5 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	9.7 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	3.6
5 000	2.7 ( <sup>99</sup> Tc)	10	6.1 ( <sup>99</sup> Tc)	3.1 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.4 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	9.7 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	8.8
10 000	0	15	5.9 ( <sup>99</sup> Tc)	3.0 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.4 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	9.4 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	12
20 000	1.8 ( <sup>135</sup> Cs)	21	5.7 ( <sup>99</sup> Tc)	2.9 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	3.2 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	9.1 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	18
50 000	0.4 ( <sup>226</sup> Ra)	31	5.3 ( <sup>99</sup> Tc)	2.6 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	2.8 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	8.2 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	27
100 000	0	41	4.6 ( <sup>99</sup> Tc)	2.0 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	2.2 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	6.8 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	35
200 000	0	47	3.6 ( <sup>99</sup> Tc)	1.4 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	1.4 x 10 <sup>3</sup> ( <sup>126</sup> Sb)	4.9 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	40
500 000	0	43	1.9 ( <sup>99</sup> Tc)	600 ( <sup>126</sup> Sb, <sup>93m</sup> Nb)	4.7 x 10 <sup>2</sup> ( <sup>126</sup> Sb)	2.4 x 10 <sup>2</sup> ( <sup>99</sup> Tc)	37
1 000 000	0	29	0.6 ( <sup>99</sup> Tc)	260 ( <sup>93m</sup> Nb)	120 ( <sup>93m</sup> Nb)	76 ( <sup>99</sup> Tc)	25
2 000 000	0	12	0.08 ( <sup>99</sup> Tc)	160 ( <sup>93m</sup> Nb)	59 ( <sup>93m</sup> Nb)	18 ( <sup>135</sup> Cs)	10
5 000 000	0	2.1	0.009 ( <sup>93m</sup> Nb)	66 ( <sup>93m</sup> Nb)	25 ( <sup>93m</sup> Nb)	5.9 ( <sup>135</sup> Cs)	1.8
10 000 000	0	0.03	0.002 ( <sup>135</sup> Cs)	12 ( <sup>93m</sup> Nb)	4.5 ( <sup>93m</sup> Nb)	1.7 ( <sup>135</sup> Cs)	0.03

## REFERENCES

1. T. L. Hoffman, "Corrosion Evaluation of Stainless Steels in High-Level Radioactive Wastes," Material Performance, Vol 15 No. 3, pp 34-38, Mar 1976
2. ERDA-77-43, Alternatives for Long-Term Management of Defense High-Level Radioactive Waste, Idaho Chemical Processing Plant, Sept 1977
3. W. M. Farr and D. J. Murphy, Jr., "Actinide Burner Studies," (Study for Sandia Laboratory), Univ of Arizona, Jul 1978
4. Nuclear Safety Associates, "Comparison of Alternatives for Long-Term Management of Defense High-Level Radioactive Wastes from the Idaho Chemical Processing Plant," NSA-305, June 1978
5. G. G. Simpson and N. A. Chipman, Radioactive Source Terms for Environmental Impact Statement - Long-Term Management of Defense High-Level Radioactive Wastes - Idaho Chemical Processing Plant, Idaho Falls, Idaho, ACI-359 (Dec 1978).