DOE/RL-96-62

Copy No. 29

Radioactive Air Emissions Notice of Construction for Vertical Calciner Operation at the Plutonium Finishing Plant

United States Department of Energy
Richland, Washington

Approved for Public Release

TRADEMARK DISCLAIMER

Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors.

This report has been reproduced from the best available copy. Available in paper copy and microfiche.

Available to the U.S. Department of Energy
and its contractors from
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831
(615) 576-8401

Available to the public from the U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
(703) 487-4650

Printed in the United States of America

DISCLM-5.CHP (8-91)

CONTENTS

FIGURES

TABLES

TERMS

ALARA	as low as reasonably achievable
BARCT	best available radionuclide control technology
CFR	*Code of Federal Regulations*
DCG	derived concentration guidelines
DF	decontamination factor
EPA	U.S. Environmental Protection Agency
HEPA	high-efficiency particulate air
MEI	maximally exposed individual
NOC	notice of construction
PFP	Plutonium Finishing Plant Complex
SEPA	*State Environmental Policy Act of 1971*
WAC	Washington Administrative Code

METRIC CONVERSION CHART

Into metric units			Out of metric units		
If you know	Multiply by	To get	If you know	Multiply by	To get
Length			**Length**		
inches	25.40	millimeters	millimeters	0.0393	inches
inches	2.54	centimeters	centimeters	0.393	inches
feet	0.3048	meters	meters	3.2808	feet
yards	0.914	meters	meters	1.09	yards
miles	1.609	kilometers	kilometers	0.62	miles
Area			**Area**		
square inches	6.4516	square centimeters	square centimeters	0.155	square inches
square feet	0.092	square meters	square meters	10.7639	square feet
square yards	0.836	square meters	square meters	1.20	square yards
square miles	2.59	square kilometers	square kilometers	0.39	square miles
acres	0.404	hectares	hectares	2.471	acres
Mass (weight)			**Mass (weight)**		
ounces	28.35	grams	grams	0.0352	ounces
pounds	0.453	kilograms	kilograms	2.2046	pounds
short ton	0.907	metric ton	metric ton	1.10	short ton
Volume			**Volume**		
fluid ounces	29.57	milliliters	milliliters	0.03	fluid ounces
quarts	0.95	liters	liters	1.057	quarts
gallons	3.79	liters	liters	0.26	gallons
cubic feet	0.03	cubic meters	cubic meters	35.3147	cubic feet
cubic yards	0.76456	cubic meters	cubic meters	1.308	cubic yards
Temperature			**Temperature**		
Fahrenheit	subtract 32 then multiply by 5/9ths	Celsius	Celsius	multiply by 9/5ths, then add 32	Fahrenheit
Force			**Force**		
pounds per square inch	6.895	kilopascals	kilopascals	1.4504×10^{-4}	pounds per square inch

Source: *Engineering Unit Conversions*, M. R. Lindeburg, PE., Second Ed., 1990, Professional Publications, Inc., Belmont, California.

RADIOACTIVE AIR EMISSIONS NOTICE OF CONSTRUCTION FOR VERTICAL CALCINER OPERATION AT THE PLUTONIUM FINISHING PLANT

1.0 INTRODUCTION

This document serves as a notice of construction (NOC) for construction, installation, and operation of a vertical calciner to stabilize plutonium at the Plutonium Finishing Plant (PFP) Complex, pursuant to the requirements of Washington Administrative Code (WAC) 246-247-060.

The PFP Complex is located in the 200 West Area of the Hanford Site (Figure 1). The PFP Complex consists of several large and small buildings that are grouped to form the processing complex (Figure 2). The PFP Complex activities are focused on the cleanout and stabilization of plutonium residue left from plutonium weapons material processing activities.

Approximately 4,800 liters of plutonium and transuranic solutions are stored at the PFP Complex, predominantly in 10-liter stainless steel vessels and some 10-liter polyethylene bottles, normally filled to 8.5 liters of solution. The total mass of plutonium and americium contained within this volume is approximately 335 kilograms. The polyethylene containers used to store the plutonium solutions were not designed for extended storage, and currently the polyethylene bottles are degrading. Plutonium solutions are acidic and corrosive and can damage the stainless steel vessels now used for storage, leading to leakage. Other potential vulnerabilities associated with the plutonium-bearing containers include the potential for radiolytic damage, hydrogen generation, and plutonium precipitation. The solutions are stored in containers with a favorable geometrical shape to prevent criticality.

Historically, the PFP Complex processed plutonium-based chemical solutions and converted these solutions into purified oxide and metal by way of a precipitation and filtration process, which yielded an acidic filtrate containing more plutonium than could be discarded. The prime purpose of the vertical calciner is to convert plutonium acid solutions to a more stable plutonium oxide. A test calciner has been developed and put in place in Room 188 of the 234-5Z Building. Development testing of this vertical calciner is ongoing. A new vertical calciner will be assembled for actual stabilization operation in Room 230C of the 234-5Z Building. The test calciner in Room 188 may be upgraded or replaced as an alternative to building a new calciner in Room 230C.

2.0 FACILITY LOCATION (Requirement 1)

U.S. Department of Energy, Richland Operations Office
Hanford Site
200 West Area, Plutonium Finishing Plant Complex
Richland, Washington 99352.

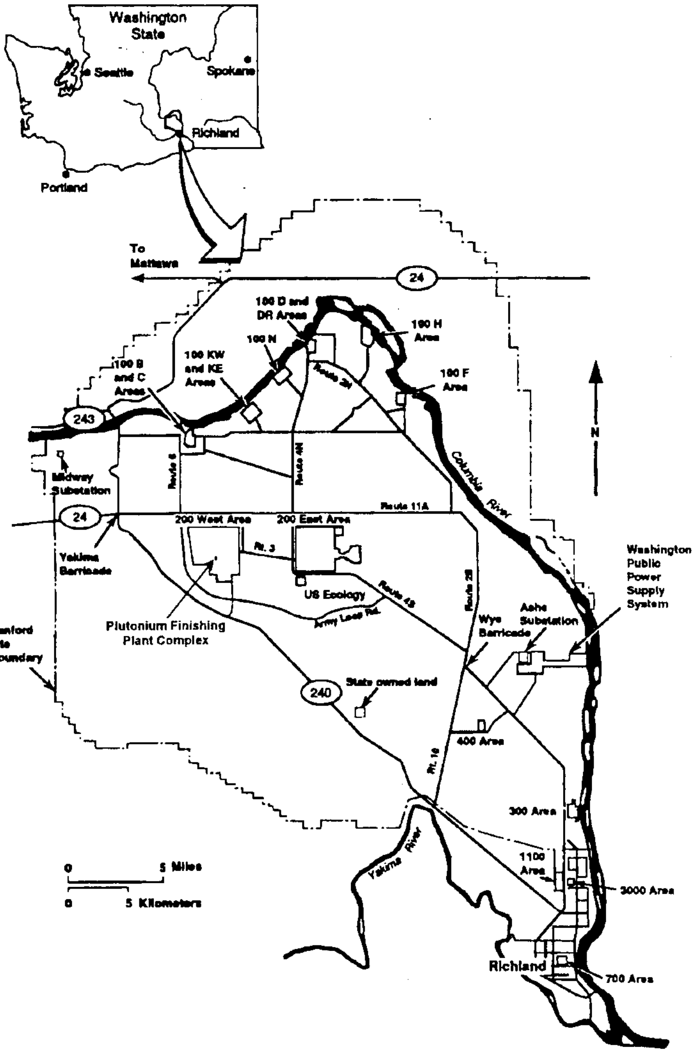

Figure 1. Hanford Site.

T960702

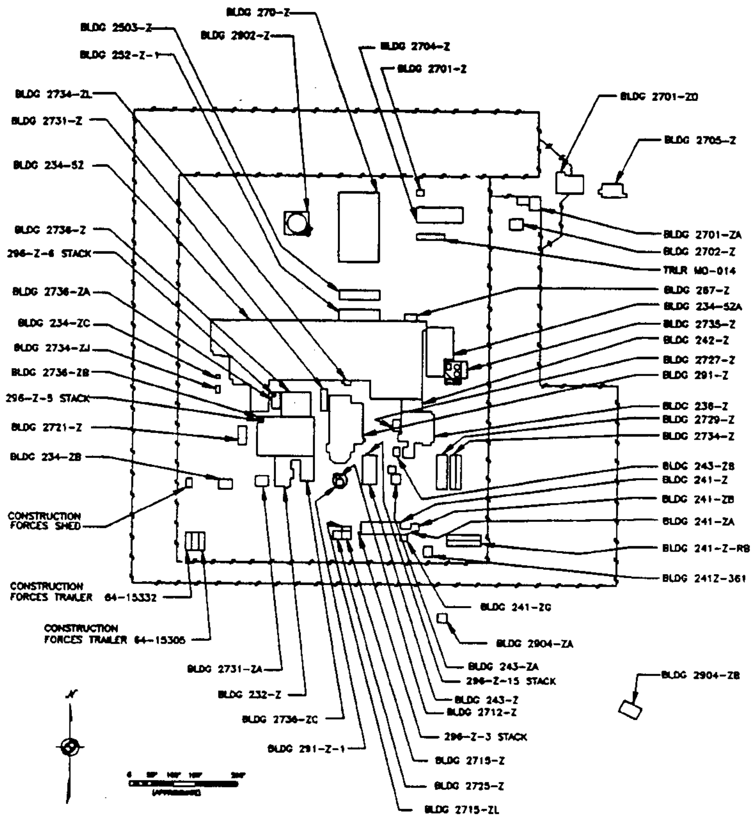

Figure 2. Plutonium Finishing Plant Complex.

The geodetic coordinates for the main stack, 291-Z-1 are as follows:

Latitude: 46° 32' 59" North Longitude: 119° 37' 59" West

3.0 RESPONSIBLE MANAGER (Requirement 2)

Mr. J. E. Mecca, Director,
Transition Program Division
U.S. Department of Energy, Richland Operations Office
P.O. Box 550
Richland, Washington 99352
(509) 376-7471.

4.0 TYPE OF PROPOSED ACTION (Requirement 3)

The proposed action is a modification of the existing process within the PFP Complex. It is proposed that a vertical calciner be constructed and installed in Room 230C of the 234-5Z Building and operated to stabilize the plutonium. A potential increase in emissions will occur at emission point 291-Z-1. Historically, the emissions at this point have remained relatively constant whether or not there were any processes operating. No actual increase in emissions is anticipated from the operation of the vertical calciner.

5.0 STATE ENVIRONMENTAL POLICY ACT (Requirement 4)

The Washington State Department of Health has determined that this activity is categorically exempt from the *State Environmental Policy Act* (SEPA) *of 1971* process. Separately, under the *National Environmental Policy Act* (NEPA) *of 1969*, the U.S. Department of Energy, Richland Operations Office (DOE-RL) has included the activity in the PFP Stabilization Environmental Impact Statement (EIS) (DOE-RL 1996). A Record of Decision was issued by DOE-RL on June 25, 1996, which included a pretreatment ion exchange process and vertical calcination as part of the preferred alternative for stabilization of plutonium-bearing solutions.

6.0 PROCESS DESCRIPTION (Requirements 5 and 7)

A vertical calciner was tested at the PFP Complex during the early 1960's as a method to continuously convert plutonium nitrate solution to plutonium dioxide. This method has advantages over more complex stabilization methods involving precipitation, filtration, and calcination steps because this involves only the calcination step.

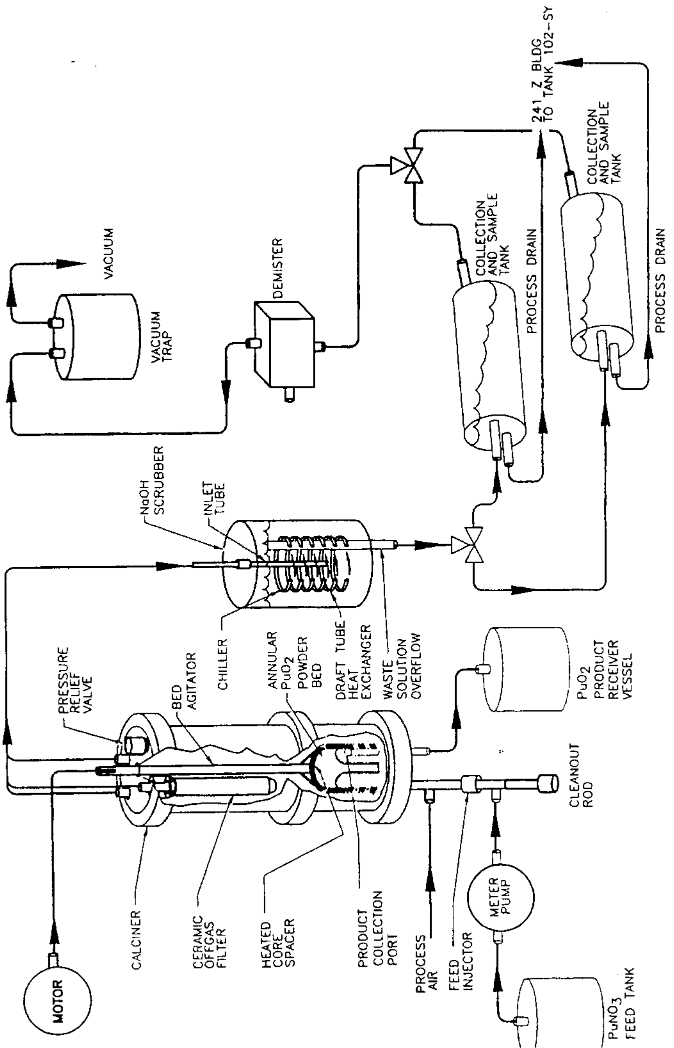

Figure 3. Process Flow Diagram of the Vertical Calciner.

The vertical calciner design (Figure 3) proposed for stabilizing the plutonium solutions under the EIS (Section 5.0) is similar to the calciner tested in the 1960's. The temperatures in the calciner will be 900 to 1050°C, appreciably higher than those employed historically. The 1960's model vertical calciner was operated at temperatures that completely decomposed the plutonium nitrate while preserving plutonium reactivity for purposes of subsequent weapons processing. The higher temperatures of the current design will minimize plutonium reactivity. Other operational improvements have been made that primarily improve the handling methods of the effluents from the process. Approximately 75 percent of the plutonium-bearing solutions, containing roughly a third of the source term, will require a pretreatment ion exchange process in Room 228A of the 234-5Z Building.

The vertical calciner will be assembled in a glovebox in the 234-5Z Building. The equipment will be sized to process 1 to 2 liters per hour of solution for concentrated feeds and 3 to 4 liters per hour for dilute feeds. The feed rate is variable to ensure the plutonium introduced into the vertical calciner has sufficient residence time to produce a suitable oxide.

The plutonium solutions will be slowly introduced into a stirred bed of previously generated product solids. This feed material will be slowly metered into the bottom of the vertical calciner. The feed rate will depend on the concentration of plutonium in the solution being processed. The feed will be roughly 1 to 2 liters per hour for concentrated feeds (>25 grams plutonium per liter) and 3 to 4 liters per hour for dilute feeds (<25 grams plutonium per liter).

The vertical calciner is a combination calciner and a 2-micron offgas filter. Plutonium solutions are calcined in the lower unit and the vapors are filtered in the upper unit.

The calcination unit can be described as two concentric pipes. Calcination takes place in the annular space between these two pipes. As the liquid feed is introduced into the calciner, the feed rapidly evaporates in the hot powder bed, subsequently undergoing drying, denitration, and final heat treatment to a stable impure plutonium oxide powder.

Heat is supplied to the calcination unit by heaters wrapped around the outer pipe wall and inside the inner pipe wall. The heaters maintain the calciner temperature between 900 and 1050°C. These temperatures are considerably higher than required for decomposition of nitrate (130 to 225°C). The higher temperature serves to greatly reduce the plutonium reactivity, to promote complete formation of plutonium dioxide, and to facilitate a packageable plutonium product.

During calcination, the hot plutonium powder bed is stirred continuously by a U-shaped agitator that fits over the inner, capped pipe. The agitator speed affects both the particle size of the plutonium dioxide powder and the rate of heat transfer between the powder and the calciner wall.

During the thermal denitration, solutions containing alkaline metal nitrates (i.e., potassium nitrate) tend to become increasingly viscous and pasty before decomposition. These mastic-phase formations are undesirable, as these formations can form hard deposits on equipment surfaces and on particulate material through the reaction zone. Mastic formation is avoided by converting the feed solution to dry oxide as quickly as possible, or by removing the impurities through the ion exchange pretreatment process. The slow feed rate into the calciner will facilitate a rapid transition to oxide and avoid problems with the mastic phase for those solutions which do not undergo the pretreatment ion exchange process. The plutonium oxide product is discharged into a product receiver as described in the following paragraphs.

The plutonium oxide is collected as it overflows through a slot in the inner pipe. The product powder consists of relatively large, uniform particles. The grinding action of the agitator prevents the oxide particles from becoming too large. The oxide proceeds down an overflow tube and into a product bed receiver vessel. The height of the overflow tube can be adjusted to control the bed volume within the calciner. When the product receiver vessel is full, the oxide will be bagged out of the glovebox. If the plutonium product does not meet requirements of the U.S. Department of Energy long-term storage standard 3013-94 (DOE 1994), it will be transferred to a muffle furnace capable of meeting the nominal 1,000°C firing temperature required by the standard. After thermal stabilization in the muffle furnace is completed, a sample of the oxide will be sent to the engineering laboratory to verify that the loss on ignition is <0.5 weight percent. Following this final verification, the plutonium product will be transferred to the PFP Complex vaults until a final plutonium disposition decision is made.

The calciner offgas stream consists of a mixture of air, water, nitric acid, oxides of nitrogen (NO_x), and a small component of organic impurities. Aqueous hydrochloric solutions run through the calciner will yield chloride in the offgas, which will be scrubbed out as sodium chloride. In addition, fine plutonium oxide powder could be entrained in the offgas stream. The offgas stream is run through two micron filters to remove the entrained oxide powder. The filters periodically are blown back with compressed air to knock particles from the filter back into the base of the calciner. The filters are blown back alternately so that one filter is always in operation. A vacuum applied to the filters draws off vapors that are run through a combined condenser and scrubber. This removes the majority of water and nitric acid in the offgas and any plutonium that might pass the filters. A basic scrubbing solution removes acid gases such as hydrogen chloride as well as nitrogen oxides. The nonreacted, noncombustible offgases remaining after the scrubber/chiller processing proceed through two existing stages of high-efficiency particulate air (HEPA) filtration before being released out the 291-Z-1 main stack. The offgases consist of water and air, with only trace amounts of NO_x. The resultant aqueous scrub solution consists of sodium hydroxide, water, sodium chloride, sodium nitrate, and sodium nitrite.

7.0 ANNUAL POSSESSION QUANTITY AND PHYSICAL FORM

(Requirements 8, 10, 11, and 12)

The total amount of radioactive material in the plutonium nitrate solutions is estimated at 335 kilograms (DOE-RL 1996). To determine the annual possession quantity, the isotopic distribution in Table 1 (WHC 1995) was applied to 335 kilograms.

Both fuels grade and weapons grade material were processed at the PFP Complex. Because the feed solutions to the calciner are associated with weapons grade plutonium, distribution of isotopes for weapons grade material was used to estimate a conservative potential-to-emit and offsite dose for the activity. All isotopes in the inventory of each radionuclide are presumed to be in liquid form.

Table 1. Isotopic Distribution for Weapons Grade Production Plutonium.

Radioisotope	Abundance* (weight percent)
Pu-238	0.01
Pu-239	93.75
Pu-240	5.95
Pu-241	0.27
Am-241	0.20

*Weapons Grade Distribution, "Plutonium Finishing Plant Safety Analysis Report" (WHC 1995).

Note: This is typical data from numerous N stock samples and will not total to 100 percent.

8.0 CONTROL SYSTEM (Requirement 6)

No additional emission control equipment is planned for the main stack as the existing plant equipment is adequate (Figure 4). There are no volatile radioisotopes in the inventory and all of the material is in solution. The HEPA filter system in the building is accepted as best available radionuclide control technology (BARCT) in the absence of volatile substances. This was confirmed by Washington State Department of Health in 1992 (Moon 1992).

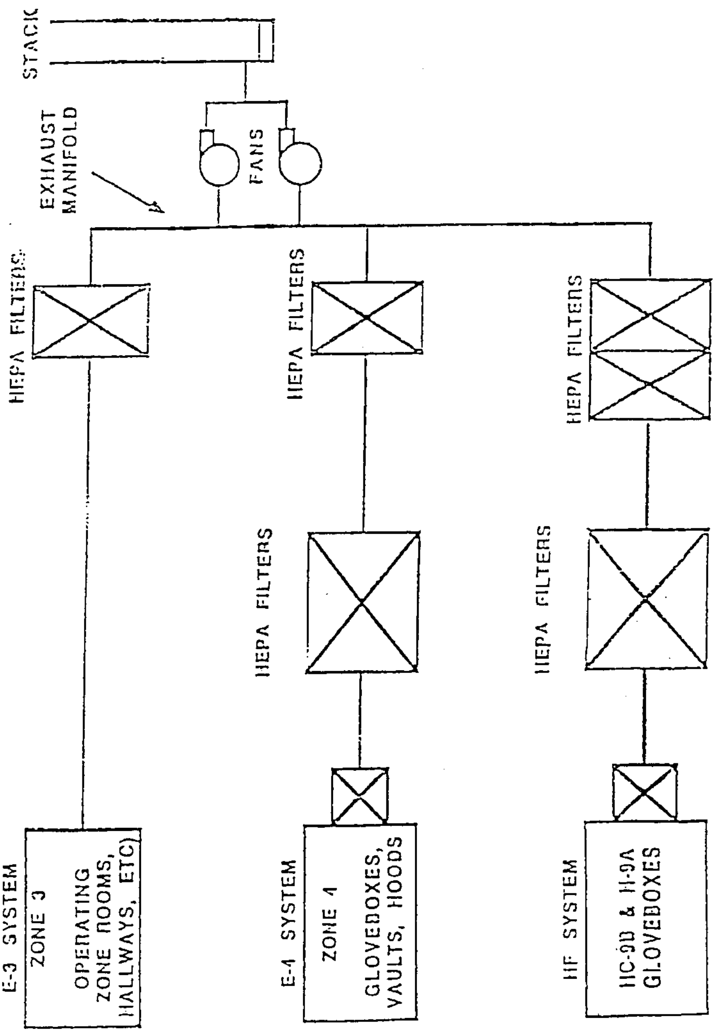

Figure 4. Applicable Control Technology Components.

9.0 MONITORING SYSTEM (Requirement 9)

The 291-Z-1 main stack is equipped with a stack sampler consisting of an alpha continuous air monitor and a record sampler. The Washington State Department of Health, Radiation Protection Division, inspected this unit during a comprehensive inspection in July 1991. There were no findings specific to the system and the system meets the requirement for continuous sampling as required by 40 CFR 61, Subpart H.

10.0 RELEASE RATES (Requirement 13)

The emissions noted in Table 2 were calculated based on the total inventory of approximately 335 kilograms. The emission of a given isotope is the product of the total inventory amount multiplied by the abundance percentage and the activity rate and adjusted by the release factor of 10^{-3} for liquids (based on the discussion in Section 7.0 of the release form of each radionuclide). This is illustrated typically as follows using Pu-238:

$$(335 \text{ kg}) \times (1000 \text{ g/kg}) \times (.0001) \times (17.1 \text{ Ci/g}) \times 10^{-3} = 5.73\text{E-01 Ci/yr.}$$

Table 2. Potential Annual Unabated Emissions.

Radioisotope	Weight (grams)	Activity rate (curies per gram)	Release fraction	Unabated emissions (curies per year)
Pu-238	3.35 E+01	1.71 E+01	1 E-03	5.73 E-01
Pu-239	3.14 E+05	6.20 E-02	1 E-03	1.95 E+01
Pu-240	1.99 E+04	2.30 E-01	1 E-03	4.58 E+00
Pu-241	9.05 E+02	1.10 E+02	1 E-03	9.95 E+01
Am-241	6.70 E+02	3.20 E+00	1 E-03	2.14 E+00

The abated emissions were calculated from the unabated emissions listed in Table 2 and the decontamination factor (WHC 1991, page 5). A decontamination factor of 2,000 was used in the calculations based on the 99.95 percent efficient HEPA filter. The abated emissions equals the unabated emissions divided by the decontamination factor. The potential annual abated emissions from the activity are shown in Table 3.

Table 3. Potential Annual Abated Emissions.

Radioisotope	Unabated Emissions (curies per year)	Decontamination factor	Abated emissions (curies per year)
Pu-238	5.73 E-01	2.0 E+03	2.86 E-04
Pu-239	1.95 E+01	2.0 E+03	9.74 E-03
Pu-240	4.58 E+01	2.0 E+03	2.29 E-02
Pu-241	9.95 E+01	2.0 E+03	4.97 E-02
Am-241	2.14 E+00	2.0 E+03	1.07 E-03

The air emissions data for the 291-Z-1 stack listed in Table 4 were taken from the Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 1994 (DOE-RL 1995a, page 2-4).

Table 4. 1994 Radionuclide Air Emissions Data for the 291-Z-1 Stack (PFP).

Radioisotope	Annual emissions (curies)
Pu-238	1.5 E-05
Pu-239	3.0 E-04
Pu-240	2.0 E-03
Pu-241	5.5 E-05
Am-241	1.6 E-05

Note: Discharge height (m) - 61.0.
Emission control (stages) - HEPA (1-3).
Total flow (m^3) - 3.5 E+09 m^3/yr.
Annual average flow rate(m^3/s) - 113.8.

11.0 OFFSITE IMPACT (Requirements 14 and 15)

This section contains information regarding the total effective dose equivalents (TEDE) to the maximally exposed individual (MEI) offsite resulting from unabated and abated emissions from the activity. The MEI is located 24 kilometers west of the 200 West Area. The potential unabated and abated TEDE to the MEI of 69.6 and 0.0348 millirem per year, respectively, are summarized in Table 5 and Table 6. The unit dose factors included in the tables were submitted previously to the Washington State Department of Health.

The information required to develop the unit dose factors was from the "Clean Air Act Assessment Package 1988" computer code (WHC 1991).

Table 5. Potential Annual Unabated Offsite Dose.

Radioisotope	Unabated emissions (curies per year)	Unit Dose Factor (millirem per curie)	Unabated dose (millirem per year)
Pu-238	5.73 E-01	2.16	1.24 E+00
Pu-239	1.95 E+01	2.42	4.71 E+01
Pu-240	4.58 E+00	2.41	1.10 E+01
Pu-241	9.95 E+01	0.0486	4.84 E+00
Am-241	2.14 E+00	2.49	5.34 E+00
Total dose	N/A	N/A	6.96 E+01

Table 6. Potential Annual Abated Offsite Dose.

Radioisotope	Abated emissions (curie per year)	Unit dose factor (millirem per curie)	Abated dose (millirem per year)
Pu-238	2.86 E-04	2.16	6.19 E-04
Pu-239	9.74 E-03	2.42	2.36 E-02
Pu-240	2.29 E-03	2.41	5.52 E-03
Pu-241	4.97 E-02	0.0486	2.42 E-03
Am-241	1.07 E-03	2.49	2.67 E-03
Total dose	N/A	N/A	3.48 E-02

12.0 FACILITY LIFETIME (Requirement 17)

Stabilization of plutonium solutions using the calciner is estimated to require 1 to 2 years to complete. The lifetime of the PFP Complex and this vertical calciner will depend on the future mission as determined by the DOE-RL.

13.0 TECHNOLOGY STANDARDS (Requirement 18)

The 291-Z-1 stack is a registered stack with the Washington State Department of Health. The stack design and operation will not be modified to complete the stabilization of the plutonium solution using the vertical calciner process.

The U.S. Environmental Protection Agency (EPA), Headquarters, was requested to approve the 291-Z-1 stack sampling/monitoring system as compliant with 40 CFR 61, Subpart H, on September 8, 1995 (DOE-RL 1995b). On September 19, 1995, the EPA approved the 291-Z-1 stack to be in compliance with the requirements of 40 CFR 61, Subpart H (DOE-RL 1995c).

14.0 DISCUSSION OF BEST AVAILABLE RADIONUCLIDE CONTROL TECHNOLOGY

The HEPA filter system in the building is considered BARCT in the absence of volatile substances. The Washington State Department of Health, Radiation Protection Division, inspected this unit during a comprehensive inspection in July 1991. There were no findings specific to the system.

15.0 REFERENCES

DOE, 1994, *Criteria for Safe Storage of Plutonium Metal and Oxides*, DOE-STD-3013-94.0.

DOE-RL, 1995a, *Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 1994*, DOE/RL-95-49, U.S. Department of Energy, Richland Operations Office, Richland, Washington.

DOE-RL, 1995b, Letter, "Request for Approval for the 291-Z-1 Stack Sampling/Monitoring System", dated September 8, 1995, U.S. Department of Energy, Richland Operations Office, Richland, Washington.

DOE-RL, 1995c, Letter, Richard W. Poeton, EPA, to James E. Rasmussen, DOE-RL, dated September 18, 1995, Letter # AT-082, U.S. Environmental Protection Agency, Region 10, Seattle, Washington.

DOE-RL, 1996, *Plutonium Finishing Plant Stabilization Final Environmental Impact Statement*, DOE/EIS-0244-F, U.S. Department of Energy, Richland Operations Office, Richland, Washington.

Moon, T. W., 1992, October 5, 1992, Telephone Conference Memorandum, to K. Fox-Williams of the State of Washington Department of Health, Air & Water Permitting, memo 81173-92-101, Westinghouse Hanford Company, Richland, Washington.

WHC, 1991, *Unit Dose Calculation Methods and Summary of Facility Effluent Monitoring Plan Determinations*, WHC-EP-0498, Westinghouse Hanford Company, Richland, Washington.

WHC, 1995a, *Hanford Site Radionuclide National Emission Standards for Hazardous Air Pollutants Registered and Unregistered Stack (Powered Exhaust) Source Assessment*, WHC-EP-0894, Westinghouse Hanford Company, Richland, Washington.

WHC, 1995b, *Plutonium Finishing Plant Safety Analysis Report*, WHC-SD-CP-SAR-021, Rev. 0C, Westinghouse Hanford Company, Richland, Washington.

DISTRIBUTION

OFFSITE

A. W. Conklin, Head
Air Emissions and Defense Waste Section
Division of Radiation Protection
State of Washington Department of Health
Airdustrial Park Building 5, LE-13
Olympia, Washington 98504-0095

J. Leitch, Chief
Radiation and Indoor Air Section
U.S. Environmental Protection Agency
Region 10
1200 Sixth Avenue
Seattle, Washington 98101

J. Wilkinson
Confederated Tribes of the Umatilla
Indian Reservation
P. O. Box 638
Pendleton, Oregon 97801

D. Powaukee
Nez Perce Tribe
P. O. Box 305
Lapwai, Idaho 80540

R. Jim, Manager
Environmental Restoration/
Waste Management Program
Confederated Tribes and Bands
of the Yakama Nation
P. O. Box 151
Toppenish, Washington 98948

ONSITE

U.S. Department of Energy, Richland Operations Office	**MSIN**
C. M. Bell	A5-52
R. M. Carosino	A4-52
J. E. Mecca	R3-79
R. S. Ollero	R3-79
H. M. Rodriguez	A5-15
D. W. Templeton	R3-79
OSTI (2)	E6-44
Reading Room (2)	H2-53

960715.1741

DISTRIBUTION (cont)

ONSITE		MSIN
	<u>Pacific Northwest National Laboratory</u>	
	Hanford Technical Library	K1-11
	<u>Westinghouse Hanford Company</u>	
	P. K. Aardal	A3-01
	T. E. Frazier	H6-25
	R. E. Johnson (5)	H6-25
	D. J. McBride (2)	T5-54
	P. J. Sullivan	T5-54
	Air Operating Permit File	H6-25
	DPC	A3-94
	EDMC (2)	H6-08
	Central Files	A3-88