

Dose Assessment from Potential Radionuclide Emissions from Stacks on the Hanford Site

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management



Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

Copyright License By acceptance of this article, the publisher and/or recipient acknowledges the
U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper.

Approved for Public Release

Dose Assessment from Potential Radionuclide Emissions from Stacks on the Hanford Site

W. E. Davis
J. M. Barnett

Date Published
April 1995

Presented at
Challenges and Innovations in the Management of Hazardous Waste
Air & Waste Management Association

May 10-12, 1995

To Be Published in
Robotics and Remote Systems Division Proceedings

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management



**Westinghouse
Hanford Company**

P.O. Box 1970
Richland, Washington

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

Copyright License By acceptance of this article, the publisher and/or recipient acknowledges the
U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper.

Approved for Public Release

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED *WJ*

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. 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. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Dose Assessment From Potential Radionuclide Emissions From Stacks on the Hanford Site

W. E. Davis and J. M. Barnett
Westinghouse Hanford Company
Richland, Washington 99352

ABSTRACT

On February 3, 1993, the U.S. Department of Energy, Richland Operations Office (RL), received a Compliance Order and Information Request from the Director of the Air and Toxics Division of the U.S. Environmental Protection Agency (EPA), Region 10. The Compliance Order required RL to (1) evaluate all radionuclide emission points at the Hanford Site to determine which points are subject to the continuous emission sampling requirements of Title 40, Code of Federal Regulations, Part 61 (40 CFR 61), Subpart H, and (2) continuously sample radionuclide emissions in accordance with requirements in 40 CFR 61.93. The Information Request required RL to provide a written Compliance Plan to meet the requirements of the Compliance Order. A Compliance Plan was submitted to EPA, Region 10, on April 30, 1993.

The Compliance Plan specified that a dose assessment would be performed for 84 Westinghouse Hanford Company (WHC) stacks registered with the Washington State Department of Health (Washington [State] Administrative Code [WAC] 246-247) on the Hanford Site. Any stack identified in the assessment as having potential emissions to cause an effective dose equivalent (EDE) to a maximum exposed individual (MEI) greater than 0.1 mrem y^{-1} must have a compliant sampling system. In addition, a Federal Facility Compliance Agreement (FFCA) was signed on February 7, 1994. The FFCA required that all unregistered stacks on the Hanford Site be assessed. This requirement increased the number of stacks to be assessed to 123 stacks. Six methods were developed and were approved by EPA, Region 10, for performing the assessments:

- Release fractions from Appendix D of 40 CFR 61
- Back calculations using a HEPA filtration factor
- Nondestructive assay of HEPA filters
- A spill release fraction
- Upstream of HEPA filter air concentrations
- Limiting inventory.

The first two methods were found to be extremely conservative for estimating releases. The third method, which used a state-of-the-art portable gamma spectrometer for in situ measurements, yielded surprising results on the distribution of radionuclides on HEPA filters. All six methods are described in the paper.

An initial assessment using only the HEPA filtration factor for back calculations identified 32 stacks that would have emissions which would cause an EDE to the MEI greater than 0.1 mrem y^{-1} . When the other methods were applied the number was reduced to 20 stacks. The paper discusses reasons for these overestimates.

INTRODUCTION

On December 15, 1989, the U.S. Environmental Protection Agency (EPA) promulgated in the Code of Federal Regulations (CFR) the *National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities* (40 CFR 61, Subpart H¹). Subpart H requires that the emissions of radionuclides from U.S. Department of Energy (DOE) facilities shall not exceed those amounts which would cause any member of the public to receive an

effective dose equivalent (EDE) of 10 mrem in a year. In addition, potential emissions of radionuclides from a facility which could cause an EDE to a maximum exposed individual (MEI) to exceed 0.1 mrem y^{-1} require continuous monitoring.

In order to determine compliance, Section 61.93 of 40 CFR 61 provides requirements for monitoring radionuclide emissions from point sources. These monitoring requirements became effective upon promulgation of the regulation on December 15, 1989. Also, DOE, Richland Operations Office (RL) is an "owner or operator" of a "facility" (i.e., the Hanford Site [see Figure 1]) as defined in 40 CFR 61. On February 3, 1993, RL received a Compliance Order and Information Request from the Director of the Air and Toxics Division of the EPA, Region 10. The Compliance Order required RL to evaluate all radionuclide emission points at the Hanford Site to determine which are subject to continuous emission measurement requirements in 40 CFR 61, Subpart H, and to continuously measure radionuclide emissions in accordance with 40 CFR 61.93. The Information Request required RL to provide a written Compliance Plan to meet the requirements of the Compliance Order. A Compliance Plan was submitted to EPA, Region 10, on April 30, 1993. The Compliance Plan set as one of the milestones: Complete assessment of the 84 Hanford Site registered stacks under Westinghouse Hanford Company (WHC) management by December 17, 1993.

This document presents the dose assessment results for the registered stacks on the Hanford Site for potential emissions, i.e. emissions with no control devices in place. Further, the document identifies those stacks requiring continuous monitoring, i.e. the EDE from potential emissions $> 0.1 \text{ mrem y}^{-1}$.

SCOPE

The stack assessment of potential emissions was performed on 84 registered and 39 unregistered stacks on the Hanford Site (registration refers to registration under Washington Administrative Code (WAC) 246-247 with the Washington State Department of Health [DOH]). Unfortunately, potential emissions are not sampled for most stacks. For these stacks, other methods were developed and submitted to EPA, Region 10, for approval. The methods used in assessing the potential emissions from the stacks are described below.

METHODS FOR CALCULATING POTENTIAL STACK EMISSIONS

The calculations of potential emissions for the registered stacks were made using six methods approved for use by EPA, Region 10:

- 1) Release fractions from Appendix D of 40 CFR 61
- 2) Back calculations using a high-efficiency particulate air (HEPA) filtration factor
- 3) Nondestructive assay (NDA) of HEPA filters
- 4) A spill release fraction
- 5) Upstream of HEPA filter air concentrations
- 6) Limiting inventory.

Calculations based on the first two methods are considered extremely conservative. The last four methods provide a more realistic estimate of the actual potential releases. For example, if the assessment of stack emissions from either Method 1 or 2 indicate that the potential emissions would result in an EDE exceeding 0.1 mrem y^{-1} , but any of the other four methods result in an EDE less than 0.1 mrem y^{-1} , the more realistic method results would be accepted.

Method 1. Release Fractions from Appendix D of 40 CFR 61. The potential emissions for a system without control devices can be estimated based on factors presented in 40 CFR 61 Appendix D: for gases the release factor is 1, for liquids or particulate solids the release factor is 1×10^{-3} , and for solids the release factor is 1×10^{-6} . This method is extremely conservative, since

accidents involving liquids and loose particulates have a release fraction orders of magnitude less than the 1×10^{-3} release fraction. In an accident condition, a spill of powder from a 1-m height would have a maximum release fraction of 4×10^{-5} . A liquid spill from the same 1-m height would have a maximum release fraction of approximately 1×10^{-6} . A 1×10^{-3} release fraction is used for powders and liquids. In cases where the assessment was performed on waste tanks, if the inventory was covered by supernate the supernate was evaluated using the release factor for liquids while the covered salt cake was estimated using a release factor for solids.

Method 2. Back Calculations Using a High Efficiency Particulate Air Filtration Factor.

This method for estimating potential emissions assumes the Nuclear Air Cleaning Handbook² decontamination factor (DF) of 3000 ($DF \sim 1/[1-0.9997]^1$) for $0.3\text{-}\mu\text{m}$ particles by a HEPA filter. The method assumes that each bank in series acts independently of a preceding bank and will remove contamination with the same 3000 DF. For a system with "n" banks of HEPA filters in series, the DF is 3000^n . This method can be conservative for a contaminated system. When processing no longer occurs, the resuspension of contamination downstream of the HEPA filters can dominate the airborne releases from a facility. Multiplying these releases by 3000^n will overestimate the potential emissions by orders of magnitude.

Method 3. Nondestructive Assay of High-Efficiency Particulate Air Filters.

In September 1992, the EPA, Region 10, concurred with a WHC proposal for a test method to measure radioactive particulate emissions (particularly gamma emitters from HEPA filters) in stack exhaust systems across the Hanford Site (Barnett 1994³). An NDA (i.e., in situ) method was developed, tested, and implemented using portable low-resolution (NaI) gamma spectroscopy instrumentation⁴ to evaluate gamma emissions from operating HEPA filters. Guidance for the developed method comes from the 40 CFR 61, Subpart H, and Appendix B, Method 114⁵.

Method 4. Upstream of High-Efficiency Particulate Air Filter Air Concentrations.

The upstream air concentrations provide direct information on the potential emissions from a facility. This method can be applied by using Continuous Air Monitoring (CAM) data, inserting air sample probes for upstream measurements, or radiological analysis of removed HEPA filters. CAM data can be used if the data are taken from the process area that contains the radionuclide inventory. Air measurements may also be collected in the ventilation system upstream of the HEPA filters. In this case, a sampling port is selected, a probe inserted, and air concentrations are measured. The final method is the sampling of the furthest upstream HEPA filters for the facility. In this case one or more HEPA filters are removed and the radiological content is analyzed either by destructive assay or by gamma spectroscopy. The airflow during the time the HEPA filter is online is used to determine the annual release rate. All of these methods are technically based in that the measurements represent the potential concentrations emitted from a facility without control devices.

Method 5. A Spill Release Fraction. In the facility venting to the 296-Z-5 stack, the only potential emissions occur when a canister of PuO_2 powder is repackaged. No emissions are expected from contamination in the ventilation system for the facility since it is essentially uncontaminated. When a canister is to be repackaged, the double canisters containing 1500 g of PuO_2 are opened in a hood and the powder is poured into another canister. The canister is sealed and put into another canister and it is sealed. The pouring could cause a release of PuO_2 to the air. To estimate the potential maximum release, observed spill release fractions for depleted uranium oxide spills (Sutter et al. 1981⁶) was used. A maximum release fraction value of 4.0×10^{-5} was used. This number is extremely conservative since it represents a release fraction for a 1-m release height for the spill. The actual release height is a maximum of 0.15 m. The spill release fraction was only used for the 296-Z-5 stack.

Method 6. Limiting Inventory. The actual emissions were used from a facility with the CAP88 dose model (Beres 1991⁷) to predict the dose to the MEI. A multiplicative factor was then calculated between the predicted dose and the 0.1 mrem y⁻¹ dose. Once the factor was determined, an assumption was made concerning the nature of the contamination in the facility. Multiplying the release data by the multiplicative factor divided by the release factor provided the limiting inventory in the facility which, if released without control devices in place, would produce a 0.1 mrem y⁻¹ dose to the MEI. By comparing the actual inventory with the limiting inventory, a facility could be determined to be either above or below the 0.1 mrem y⁻¹ dose to the MEI. This method was only applied to one facility: 105-N Building 14-ft Decontamination Room.

CAP88 DOSE MODELING

A potential source term was calculated for the 123 stacks using one of the six approved methods. Once the potential source terms were determined, the source term, location, and stack height were used with Hanford meteorology in the CAP88 computer model (Beres 1991⁷) to predict the EDE for the MEI.

The CAP88 model is able to incorporate plume rise from the stack based on the flow rate and stack diameter. After leaving the stack, the plume is modeled to disperse based on meteorological conditions. Modeled concentrations are decreased based on dry deposition (dry deposition velocity = 1.8 cm s⁻¹). Wet deposition is ignored because of the low incident of precipitation on the Hanford Site. The modeled concentrations are used to calculate an EDE for an offsite MEI by summing the product of the EDE for each isotope with its activity.

$$\dot{H}_{total} = \sum (EDE_{(i)} \times A_{(i)}) = \text{mrem y}^{-1} \quad (1)$$

where:

- H = potential unabated dose to the MEI
- EDE_(i) = effective dose equivalent for each radionuclide
- A_(i) = total activity for radioisotope i
- i = radioisotope

Normalized source terms were used in CAP88 for each of the major areas on site, i.e., 100, 200, 300, 400, and 600 Areas. Five-year averaged meteorological data (1983-1987) taken on the Hanford Site were used to compute normalized air concentrations, i.e., X Q⁻¹, where X is the air concentration (Ci m⁻³) normalized by the source term Q (Ci s⁻¹). Two stack release heights were used: 89 m (200-ft stack with a 100-ft plume rise) and 10 m (33 ft). To model an individual stack the following input was needed:

- The potential radiological source terms
- The stack release height
- The location of the stack in one of the major areas.

Potential dose data was produced by the CAP88 model for each registered stack.

RESULTS OF DOSE ASSESSMENT

Of the total of 123 stacks assessed, 45 were assessed using release fractions from Appendix D (Method 1), 35 using back calculations (Method 2), 15 using NDA (Method 3), 28 using upstream air samples (Method 4), 1 using the powder release fraction (Method 5), and 1 using the limiting inventory (Method 6). Twenty stacks (see Table 1) were identified to have potential emissions which could cause an EDE > 0.1 mrem y⁻¹. One of the original seven designated stacks, the 291-T-1 stack, was assessed by NDA and reduced to a nondesignated stack⁸.

To evaluate five of the methods (Method 6, Limiting Inventory was excluded), each method was compared to the results of back calculations (Method 2) for all stacks. The back calculations were performed for 84 of the 123 stacks and the results indicated emissions from 32 stacks would have caused an EDE which would have exceeded 0.1 mrem y^{-1} and required the stacks to provide continuous monitoring. When a stack reported a nondetectable release, the minimum detection limit was used as the release for the stack. A comparison made for a subset of 30 stacks using release factors from Appendix D (Method 1) with back calculations showed no correlation between the two methods (Figure 2). Further, 21 of the 30 back calculations potential emission exceeded the release fractions emissions. In only four comparisons did the release fractions from Appendix D identify a designated stack when the back calculation indicated a nondesignated.

The comparison between NDA and back calculations showed that back calculations overestimated the potential releases for all nine stacks by at least four orders of magnitude (Table 2 and Figure 3). Four of the stacks (see Table 3) reported nondetection of a release. For these stacks, the minimum detection limit was used for back calculations. If back calculations had been used instead of NDA, all nine of these stacks would have required continuous monitoring. Upstream air sample (Method 4) for two of the stacks showed that the back calculations overestimated the emissions by three orders of magnitude. For the single stack that used the powder release fraction, the back calculations overestimated the release by four orders of magnitude.

By using all six methods approved by EPA, Region 10, instead of only the back calculation method for assessing the 123 WHC stacks, the number of stacks requiring continuous monitoring was reduced from 32 to 20.

When upstream measurements or NDA were performed, an apparent DF was computed for eight nondesignated stacks having releases above the detection limit. The term apparent has been used since an actual DF could not be computed. An actual DF would have to be calculated for a variety of particle sizes with air concentration measurements before and after the filters. The apparent DF was formed using either the estimated annual air concentration from upstream measurements or the loading on the filters ratioed with the measured releases. The apparent DFs ranged in value from 0.5 to a maximum value of 250 (Table 3). For one stack, 308-ET-EX, a DF of 0.5 was computed, i.e., more released than was assessed on the HEPA filters.

CONCLUSIONS

By using the six approved methods approved by EPA, Region 10, instead of only the original back calculation method for assessing the 84 WHC stacks, the number of stacks requiring continuous monitoring was reduced from 32 to the 20.

The intercomparison between results showed that no correlation existed between back calculations and release fractions from Appendix D results. Also the NDA, upstream air samples, and powder release fraction method results were at least three orders of magnitude lower than the back calculations results.

The most surprising results of the assessment came from NDA. NDA when it could be applied was found to be an easy method for assessing potential emissions. For the nine stacks assessed by NDA, all nine of the stacks would have required continuous monitoring when assessed by back calculations. However, when NDA was applied all stacks had potential emissions that would cause an EDE below the $>0.1 \text{ mrem y}^{-1}$ standard.

Apparent DFs were calculated for eight nondesignated stacks emissions above the detection limit. These apparent DFs ranged from 0.5 to 250.

ACKNOWLEDGMENTS

This work would not have been possible without the support of Larry Diediker of Westinghouse Hanford Company, Judson Kenoyer of Science Applications International Corporation, John Glissmeyer of Pacific Northwest Laboratory, and the dedicated air emission cognizant engineers from Westinghouse Hanford Company.

REFERENCES

1. 40 CFR 61, Subpart H, *National Emission Standards for Hazardous Air Pollutants*, "National Emission Standards for Emissions of Radionuclides other than Radon from Department of Energy Facilities," Code of Federal Regulations, as amended.
2. Buchsted, C. A., Fuller, A. B., and Kahn, J. E., *Nuclear Air Cleaning Handbook*, ERDA 76-21, UC-11, Oak Ridge National Laboratory, Oak Ridge, Tennessee, 1976.
3. Barnett, J. M., "Potential Radionuclide Emissions From Stacks on the Hanford Site, Part II: Dose Assessment Methodology Using Portable Low-resolution Gamma Spectroscopy," in *23rd DOE/NRC Nuclear Air Cleaning and Treatment Conference Proceedings*, July 25-28, 1994, Buffalo, New York.
4. BTI, *BTI Portable Gamma-Ray Spectrometer Microspec-1™ with Gamma-Ray Probe*, Bubble Technology Industries, Inc., Chalk River, Ontario, Canada, 1991.
5. 40 CFR 61, Appendix B, Method 114, *National Emission Standards for Hazardous Air Pollutants*, "Test Methods for Measuring Radionuclide Emissions from Stationary Sources, Code of Federal Regulations, as amended.
6. Sutter, S. L., Johnson, J. W., and Mishima, Jofu, *Aerosols Generated by Free Fall of Powders and Solutions in Static Air*, NUREG/CR-2139, PNL-3786, Pacific Northwest Laboratory, Richland, Washington, 1981.
7. Beres, D. A., *The Clean Air Act Assessment Package — 1988 (CAP-88). A Dose and Risk Assessment Methodology of Radionuclide Emissions to Air*, Vols. 1-3, U.S. Environmental Protection Agency, Washington, D.C., 1990.
8. Mizner, J. H. Jr., and Barnett, J. M., *The 291-T-1 Exhaust Stack Petition for Redesignation*, WHC-SD-WM-ER-292, Westinghouse Hanford Company, Richland, Washington, 1993.
9. WHC, *Environmental Releases for Calendar Year 1992*, WHC-EP-0527-2, Westinghouse Hanford Company, Richland, Washington, 1993.

Table 1. Designated stacks^a.

#	Stack Number	Facility	Number of HEPA Filters
1.	291-A-1 ^b	PUREX	3
2.	296-A-1	PUREX	3
3.	291-B-1 ^b	B PLANT	2
4.	291-Z-1 ^b	PFP	1-3 ^c
5.	296-A-17	TANK FARMS	2
6.	296-A-22 ^b	TANK FARMS	2
7.	296-A-25	TANK FARMS	2
8.	296-A-27	TANK FARMS	2
9.	296-A-29	TANK FARMS	2
10.	296-A-40 ^b	TANK FARMS	2
11.	296-B-28	TANK FARMS	2
12.	296-C-05	TANK FARMS	2
13.	296-P-16	TANK FARMS	2
14.	296-P-23	TANK FARMS	2
15.	296-P-26	TANK FARMS	2
16.	296-P-28	TANK FARMS	2
17.	296-S-15	TANK FARMS	2
18.	296-S-22	TANK FARMS	2
19.	296-T-18	TANK FARMS	2
20.	340-NT-EX ^b	340 BUILDING	2

HEPA high-efficiency particulate air.

PFP Plutonium Finishing Plant.

PUREX Plutonium Uranium Extraction.

^aStacks with potential emission to cause a dose >0.1 mrem y⁻¹.

^bOriginal seven designated stacks.

^cStages of filtration depends on exhaust pathway.

Table 2. Nondestructive assay comparison with back calculations.

Stack Number	NDA (mrem y ⁻¹)	Back Calculation (mrem y ⁻¹)	Log ₁₀ (difference)
291-T-1	4x10 ⁻³	4x10 ²	5
296-T-13	3x10 ⁻⁴	3x10 ¹	5
296-B-13	5x10 ⁻⁸	2x10 ^{0(a)}	8
296-G-1	3x10 ⁻⁸	3x10 ^{-1(a)}	7
296-S-2	1x10 ⁻⁵	1.2x10 ⁻¹	4
296-S-16	2x10 ⁻⁶	1.0x10 ⁻¹	5
296-W-3	2x10 ⁻⁹	3x10 ^{0(a)}	9
308-ET-EX	3x10 ⁻⁵	1x10 ²	7
437-MN&ST	2x10 ⁻³	2x10 ^{1(a)}	4

NDA nondestructive assay.

*Minimum detection limit used for back calculations.

Table 3. Apparent decontamination factors for nondesignated stacks.

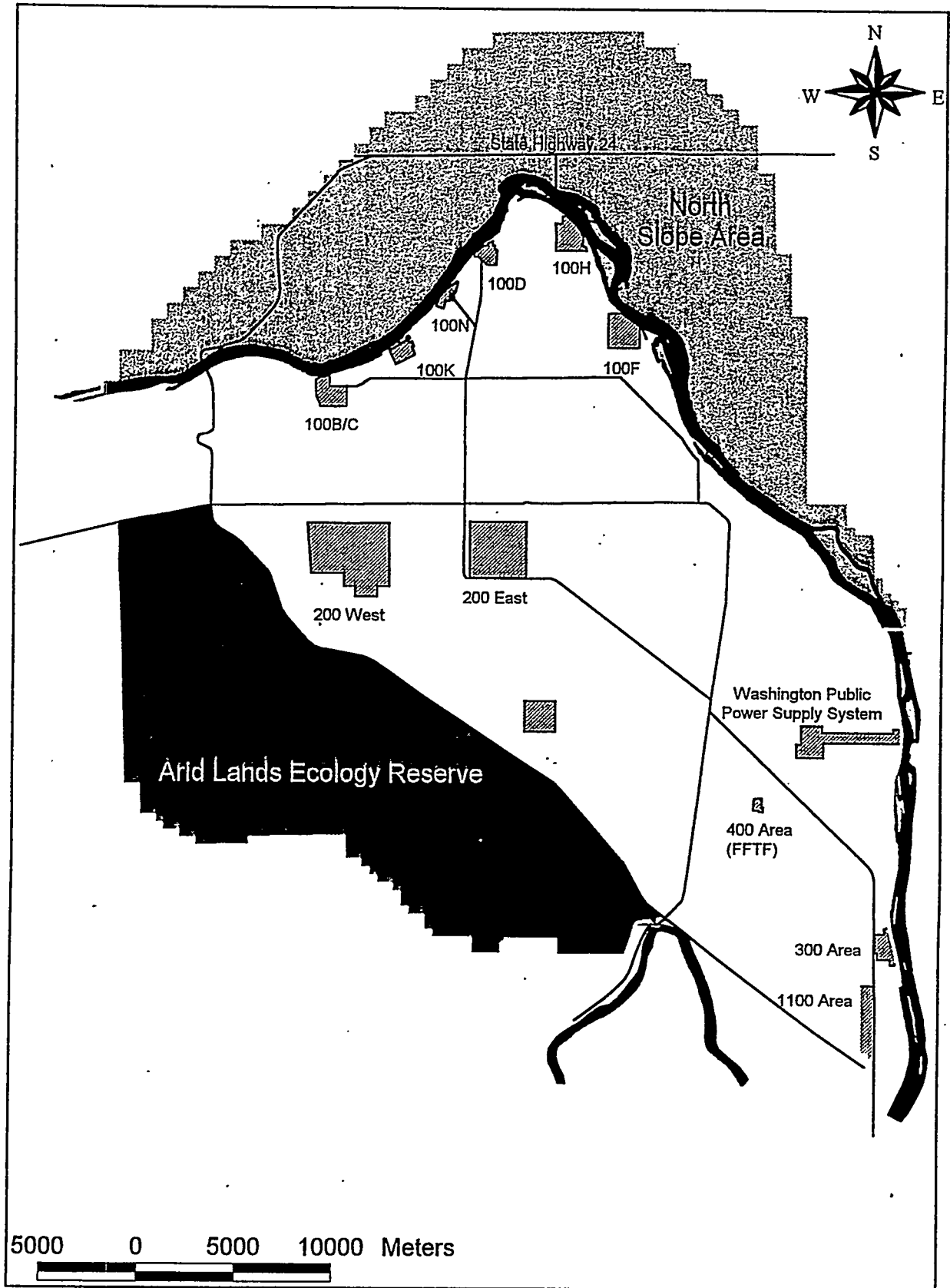
Stack Number	Method	Number of HEPA	Nuclide	Unabated (Ci)	Release (Ci) ⁹	DF
291-T-1	NDA	Prefilter, 2 HEPAs ^a	⁹⁰ Sr	1.6x10 ⁻³	1.2x10 ⁻⁵	190
			¹³⁷ Cs	1.7x10 ⁻³	1.3x10 ⁻⁵	130
			^{239,240} Pu	2.0x10 ⁻³	2.2x10 ⁻⁵	110
			²⁴¹ Am	2.0x10 ⁻⁴	2.0x10 ⁻⁶	100
296-T-13	NDA	2	^{239,240} Pu	1.7x10 ⁻⁴	2.0x10 ⁻⁶	90
296-T-11	Upstream	2	total beta	1.5x10 ⁻⁵	4.3x10 ⁻⁷	30
296-S-2	NDA	2	total alpha	2.1x10 ⁻⁶	8.4x10 ⁻⁹	240
			total beta	3.5x10 ⁻⁶	1.4x10 ⁻⁸	250
296-S-7W	Upstream	2	^{239,240} Pu	3.2x10 ⁻⁵	7.5x10 ⁻⁷	40
			²⁴¹ Am	3.9x10 ⁻⁴	4.2x10 ⁻⁶	90
296-S-16	NDA	1	total alpha	3.1x10 ⁻⁷	1.7x10 ⁻⁹	185
			total beta	9.8x10 ⁻⁶	5.3x10 ⁻⁸	180
308-ET-EX	NDA	2	total alpha	9.3x10 ⁻⁷	9.2x10 ⁻⁸	10
			total beta	1.8x10 ⁻⁷	3.4x10 ⁻⁷	0.5
437-MN&ST	NDA	2	total alpha	2.0x10 ⁻⁵	1.9x10 ⁻⁷	105
			total beta	7.8x10 ⁻⁵	7.4x10 ⁻⁷	106

DF decontamination factor.

HEPA high-efficiency particulate air.

NDA nondestructive assay.

Figure 1. Map of the Hanford Site.



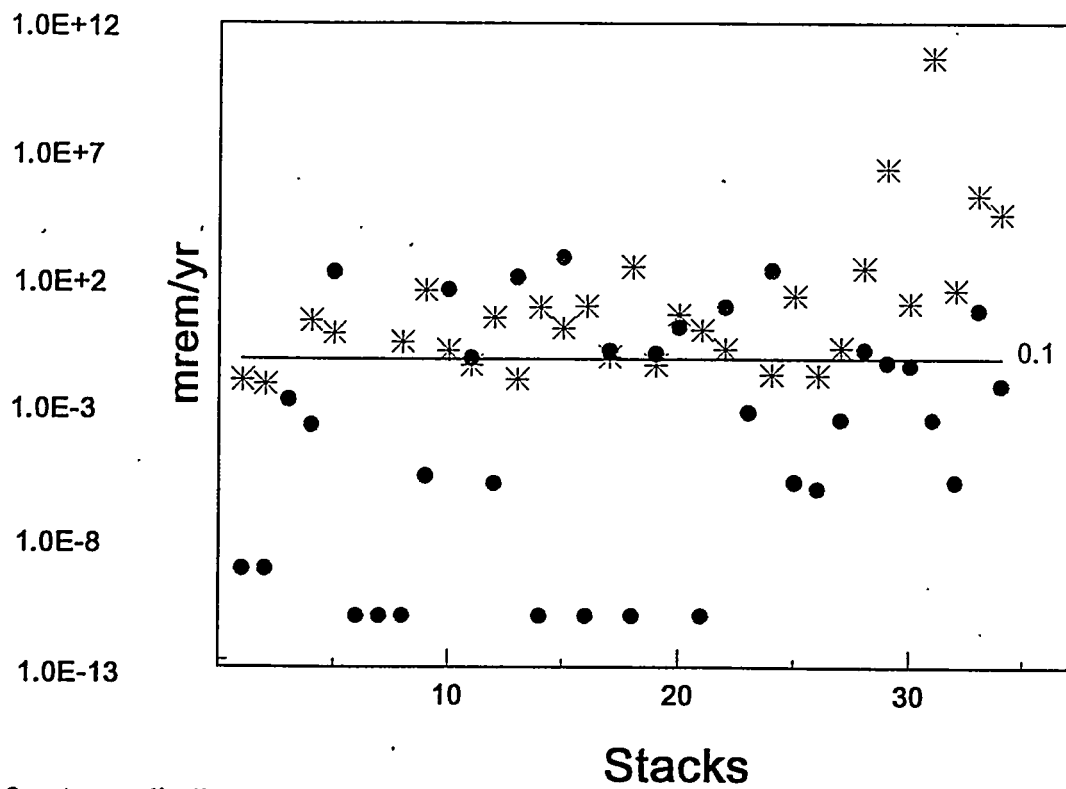


Figure 2. Appendix D versus back calculations comparison.

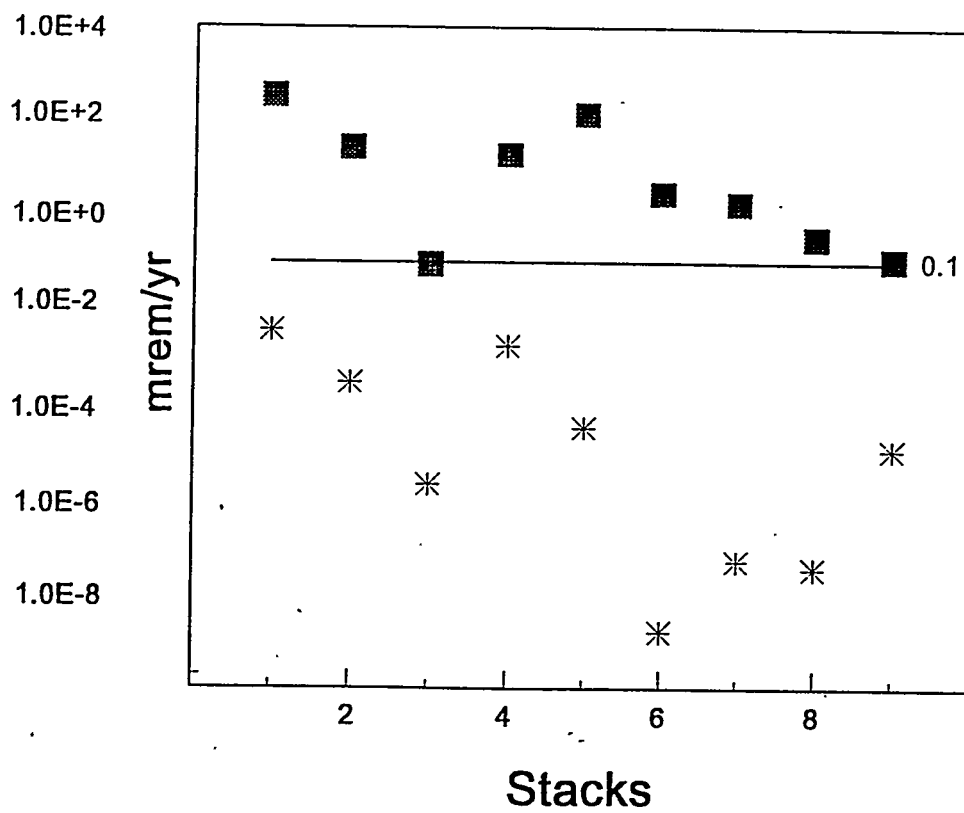


Figure 3. Comparison of nondestructive assay with back calculations.

KEY WORDS

NESHAP

Radionuclide air concentration

Nondestructive assay

High-efficiency particulate air filters

HEPA

DISTRIBUTION

ONSITE

6	<u>Westinghouse Hanford Company</u>	<u>MSIN</u>
	J. M. Barnett	T1-30
	L. P. Diediker	T1-30
	W. E. Davis	H6-20
	Central Files	L8-04
	OSTI (2)	L8-07

This page intentionally left blank.