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Potential Radionuclide Emissions From Stacks on the Hanford Site, Part 1: Dose Assessment

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POTENTIAL RADIONUCLIDE EMISSIONS FROM STACKS ON THE HANFORD SITE, PART 1: DOSE ASSESSMENT

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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 requires RL to evaluate all radionuclide emission points at the Hanford Site to determine which are subject to continuous emission monitoring requirements in Title 40, Code of Federal Regulations, Part 61, (40 CFR 61) Subpart H, and to continuously monitor 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 (WAC 246-247) on the Hanford Site. Stacks that have the potential emissions to cause an effective dose equivalent (EDE) to a maximum exposed individual (MEI) greater than 0.1 mrem y^{-1} must be monitored continuously for radionuclide emissions. Five methods 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, and Upstream of HEPA Filter Air Concentrations. The first two methods were extremely conservative for estimating releases. The third method, which used a state-of-the-art portable gamma spectrometer, yielded surprising results from the distribution of radionuclides on the HEPA filters. All five methods are described.

Assessments using a HEPA Filtration Factor for back calculations identified 32 stacks that would have emissions that would cause an EDE to the MEI greater than 0.1 mrem y^{-1} . The number was reduced to 15 stacks when the other methods were applied. The paper discusses reasons for the overestimates.

I. Introduction

On December 15, 1989, 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 H⁽¹⁾. Subpart H requires the emissions of radionuclides from the U.S. Department of Energy (DOE) facilities shall not exceed those amounts that 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 that could cause an EDE to a maximum exposed individual to exceed 0.1 mrem y^{-1} require continuous monitoring.

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To determine compliance, section 61.93 provides requirements for monitoring of 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) as defined in 40 CFR 61 is an "owner or operator" of a "facility," i.e., the Hanford Site (see Figure 1). On February 3, 1993, 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 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 management by WHC by December 17, 1993.

Figure 1 Hanford site.

The purpose of this document is to present 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 will identify those stacks requiring continuous monitoring (i.e., the EDE from potential emissions >0.1 mrem y^{-1}).

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II. Scope

The stack assessment of potential emissions was performed on 84 registered stacks on the Hanford Site. These emission sources represent individual point sources presently registered under Washington Administrative Code 246-247 with the Washington Department of Health. Unfortunately, for most stacks potential emissions are not sampled. 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.

III. Methods for Calculating Potential Stack Emissions

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

1. Source Term Estimation Based on release factors in Appendix D
2. Back Calculations Based on a Decontamination Factor of 3000^n , where n equals the number of HEPA filter banks in series
3. Nondestructive Assessment (NDA)
4. Upstream of HEPA Filter Air Concentration Measurements
5. Spill Release Fraction (296-Z-5)

Calculations based on the first two methods are considered extremely conservative. The last three methods should provide a realistic estimate of the actual potential releases. When the assessment of stack emissions based on Appendix D or from back calculations could indicate that the potential emissions would result in an EDE exceeding 0.1 mrem y^{-1} , while one of the last methods would result in an EDE less than 0.1 mrem y^{-1} , the more realistic method results would be accepted over the first two conservative methods.

1. Appendix D Release Factors

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, because 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 of the entire inventory would release under extremely conservative conditions 4×10^{-5} release fraction and a liquid spill from the same would release approximately 1×10^{-6} release fraction. 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.

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2. Back Calculations Based on a Decontamination Factor of 3000ⁿ

This method for estimating potential emissions assumes the Nuclear Air Cleaning Handbook⁽²⁾ decontamination factor (DF) of 3000 ($DF = 1/(1-0.9997)$) for 0.3μ 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 decontamination factor. For a system with n banks of HEPA filters in series, the decontamination factor 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.

3. Nondestructive Assessment (NDA)

In September 1992, EPA, Region 10, concurred with a WHC proposal for a test method to measure radioactive particulate emissions (particularly gamma emitters from high-efficiency particulate air [HEPA] filters) in stack exhaust systems across the Hanford Site (see Part II, Barnett, 1994⁽³⁾). A nondestructive assessment (i.e., in situ) method was developed, tested, and implemented using portable low-resolution (NaI) gamma spectroscopy instrumentation⁽⁴⁾ 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 114⁽⁵⁾.

4. Upstream of HEPA Filter Air Concentration Measurements

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 is 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 a gamma spectrometer. The airflow during the time the HEPA filter is online is used to determine the annual release rate. These methods are technically based in that the measurements represent the potential concentrations emitted from a facility without control devices.

5. Spill Release Fraction (296-Z-5)

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 because it is essentially uncontaminated. When a canister is repackaged, the double canisters containing 1,500 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

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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. 1981⁽⁶⁾) was used. A maximum release fraction value of 4.0×10^{-5} was used. This number is extremely conservative because 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.

IV. CAP88 Dose Modeling

A potential source term was calculated for the 84 registered stacks using one of the five approved methods. Once the potential source terms were determined, the source term, location, and stack height were used with Hanford Site meteorology in the CAP88 computer model (Beres 1991⁽⁷⁾) to predict the EDE for the maximum exposed individual.

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.) However, 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.

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

where:

$\dot{H}$ = 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). The 5-year average of Hanford Site meteorological data (1983-1987) taken on the site was used to compute normalized air concentrations, i.e., X/Q , where X is the air concentration (curies/ m^3) normalized by the source term Q (curies/s). Two stack release heights were used: 89 m (200 ft stack with a 100 ft plume rise) and 10 m (33 ft). The following input was needed to model an individual stack: the potential radiological source terms, the stack release height was chosen, and the location of the stack in one of the major areas. Potential dose data was produced by the CAP88 model for each registered stack.

V. Results of Dose Assessment

Of the 82 registered stacks assessed, 41 were assessed using release fractions from Appendix D, 31 using back calculations, nine stacks using NDA, two using upstream air samples, and one using the powder release fraction. Fifteen stacks (see Table 1) were identified to have potential emissions that could cause an effective

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Table 1 Designated stacks.

Stack Number	Facility	Number of HEPA Filters
291-A-1 ^b	PUREX	3
296-A-1	PUREX	3
291-B-1 ^b	B Plant	2
291-Z-12	PFP	1-3 ^c
296-Z-3	PFP	2
296-A-22 ^b	Tank Farms	2
296-A-27	Tank Farms	2
296-A-29	Tank Farms	2
296-A-40	Tank Farms	2
296-P-16	Tank Farms	2
296-A-22 ^b	Tank Farms	2
296-P-23	Tank Farms	2
296-S-7W	ERO	2
296-S-15	Tank Farms	2
308-GL-EX	308 Building	3
340-NT-EX ^b	340 Building	2

^aStacks with potential emission to cause a dose >0.1 mrem y^{-1} .

^bOriginal seven designated stacks.

^cStages of filtration depends on exhaust pathway.

dose equivalent >0.1 mrem y^{-1} . One of the original seven designated stacks, 291-T-1, was assessed by NDA and reduced to a nondesignated stack⁽⁸⁾.

To evaluate the five different methods, each method was compared to the results of back calculations for all stacks. The back calculations were performed for all stacks and the results indicated emissions from 32 stacks would have caused an EDE that would have exceeded 0.1 mrem y^{-1} and required the stacks to provide continuous monitoring. When a stack reported a non detectable 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 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.

Figure 2 Doses from back calculations compared to release fractions from Appendix D (* - Release Fraction Appendix D, • - Back calculations).

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 stacks would have required continuous monitoring. Upstream air sample method 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.

Table 2 NDA Comparison with back calculations.

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

^aMinimum detection limit used for back calculations.

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Figure 3 Doses from NDA compared to back calculations.

Table 3 Apparent decontamination factors for nondesignated stacks.

Stack Number	Method	Number of HEPA	Nuclide	Unabated (Ci)	Release (Ci)	DF
291-T-1	NDA	Prefilter, 2 HEPAs	Sr90	1.6E-3	1.2E-5	190
			Cs137	1.7E-3	1.3E-5	130
			Pu239,240	2.0E-3	2.2E-5	110
			Am241	2.0E-4	2.0E-6	100
296-T-13	NDA	2	Pu239,240	1.7E-4	2.0E-6	90
296-T-11	Upstream	2	total beta	1.5E-5	4.3E-7	30
296-S-2	NDA	2	total alpha	2.1E-6	8.4E-9	240
			total beta	3.5E-6	1.4E-8	250
296-S-7W	Upstream	2	Pu239,240	3.2E-5	7.5E-7	40
			Am241	3.9E-4	4.2E-6	90
296-S-16	NDA	1	total alpha	3.1E-7	1.7E-9	185
			total beta	9.8E-6	5.3E-8	180
308-ET-EX	NDA	2	total alpha	9.3E-7	9.2E-8	10
			total beta	1.8E-7	3.4E-7	0.5
437-MN&ST	NDA	2	total alpha	2.0E-5	1.9E-7	105
			total beta	7.8E-5	7.4E-7	106

When upstream measurements or NDA were performed, an apparent decontamination factor was computed for eight nondesignated stacks having releases above the detection limit. The term apparent has been

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used because an actual decontamination factor could not be computed. An actual decontamination factor would have to be calculated for a variety of particle sizes with air concentration measurements before and after the filters. The apparent decontamination factor was formed using either the estimated annual air concentration or the loading on the filters ratioed with the measured releases. The measured releases were $<2\text{E}-5$ curies per year for a radioisotope for all the stacks. The unabated emissions for these stacks were less than $3\text{E}-3$ curies y^{-1} . For one stack 308-ET-EX a DF of 0.5 was computed (i.e., more released than was assessed on the HEPA filters). For some of the designated stacks the DFs, if measured, would produce larger DFs than noted in Table 3.

VI. Conclusions

By using the five 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 15.

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 was found to be an easy method for assessing potential emissions. For the nine stacks assessed by NDA, all nine would have required continuous monitoring when assessed by back calculations. However, when NDA was applied, all stacks had potential emissions that would cause an effective dose equivalent below the >0.1 mrem y^{-1} standard.

Apparent decontamination factors were calculated for eight nondesignated stack emissions above the detection limit. These apparent decontamination factors ranged from 0.5 to 250.

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