

Determination of the Radionuclide Release Factor for an Evaporator Process Using Nondestructive Assay

Prepared for the U.S. Department of Energy

FLUOR DANIEL HANFORD, INC.



Richland, Washington

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DETERMINATION OF THE RADIONUCLIDE RELEASE FACTOR FOR
AN EVAPORATOR PROCESS USING NONDESTRUCTIVE ASSAY

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Abstract

The 242-A Evaporator is the primary waste evaporator for the Hanford Site radioactive liquid waste stored in underground double-shell tanks. Low pressure evaporation is used to remove water from the waste, thus reducing the amount of tank space required for storage. The process produces a concentrated slurry, a process condensate, and an offgas. The offgas exhausts through two stages of high-efficiency particulate air (HEPA) filters before being discharged to the atmosphere.

40 CFR 61 Subpart H requires assessment of the unfiltered exhaust to determine if continuous compliant sampling is required. Because potential (unfiltered) emissions are not measured, methods have been developed to estimate these emissions. One of the methods accepted by the Environmental Protection Agency is the measurement of the accumulation of radionuclides on the HEPA filters.

Nondestructive assay (NDA) was selected for determining the accumulation on the HEPA filters. NDA was performed on the HEPA filters before and after a campaign in 1997. NDA results indicate that $2.1 \text{ E}+4$ becquerels of cesium-137 were accumulated on the primary HEPA filter during the campaign. The feed material processed in the campaign contained a total of $1.4 \text{ E}+16$ Bq of cesium-137. The release factor for the evaporator process is $1.5 \text{ E}-12$. Based on this release factor, continuous compliant sampling is not required.

I. Introduction

The U.S. Department of Energy, Richland Operations Office (DOE-RL) must comply with the requirements of the Clean Air Act and its implementing regulations. On December 15, 1989, the U.S. Environmental Protection Agency (EPA) promulgated the "National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities" (40 CFR Part 61, Subpart H). Subpart H requires continuous compliant stack monitoring at facilities with potential radionuclide emissions that could result in an effective dose equivalent (EDE) to the offsite maximum exposed individual (MEI) that exceeds $1.0 \text{ E}-6$ sieverts per year.

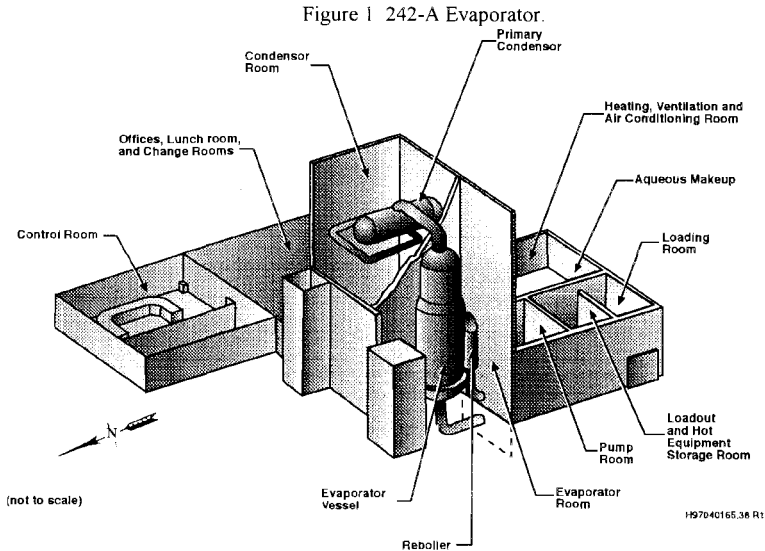
A Federal Facility Compliance Agreement (FFCA) between DOE-RL and the EPA, Region 10, was concluded on February 7, 1994. As part of the FFCA a strategy was developed to assess the potential radionuclide emissions for all stacks and bring these sampling systems into compliance. Because changes can occur in emissions, or as better assessment methods are

developed, the FFCA allows for re-assessment of stacks and for reduction of monitoring requirements. The 242-A Evaporator stack originally was assessed as a major stack (exceeding $1.0 \text{ E-}6 \text{ Sv/yr}$), based on the use of a conservative release factor of $1.0 \text{ E-}03$ in 40 CFR 61, Appendix D. Because the assessment was based on conservative numbers, and emission data from the facility indicated lower emissions than expected, methods for assessing the stack were examined to determine if a better methodology would yield more realistic potential emissions.

The method chosen for the reassessment was nondestructive assay (NDA) of high-efficiency particulate air (HEPA) filters. This method was performed before and after the second evaporation campaign in 1997 (Campaign 97-2) to identify a more accurate release factor based on quantities of gamma ray emitting radionuclides present. The results of the reassessment are presented in this paper

II. Facility Description

The 242-A Evaporator (Figure 1) is located on the Hanford Site in southeastern Washington State. Radioactive waste solutions from fuel reprocessing are stored in underground waste tanks awaiting final treatment and disposal. The mission of the 242-A Evaporator is to reduce the volume of waste in these tanks to provide additional space for receipt of waste solutions generated during current and future waste treatment activities. The 242-A Evaporator currently operates in discrete campaigns, each evaporating between 2 and 8 million liters of waste, depending on initial waste volume and concentration. Since 1994, six campaigns have achieved a total waste volume reduction of 36 million liters.

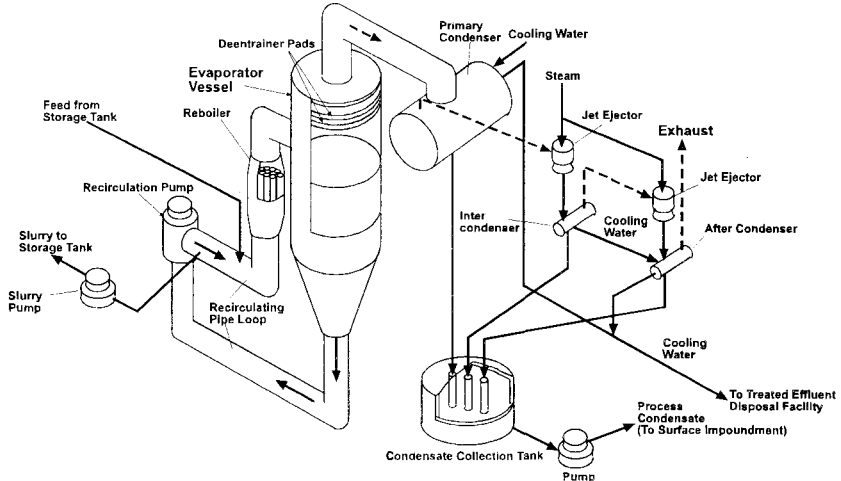


III. Evaporator Process

The 242-A Evaporator process uses a forced-circulation, vacuum evaporation system to reduce the volume of radioactive waste solutions. The incoming waste stream from the tanks is separated by evaporation into two liquid streams: a concentrated slurry stream and a process condensate stream. The slurry contains the majority of the radionuclides and inorganic constituents. When the evaporation rate is approximately 50 percent of the waste feed rate, the slurry pump is started to begin transferring the slurry stream back to the underground waste tanks. Vapor from the evaporation process is condensed, producing process condensate, which is primarily water with trace amounts of organic material and greatly reduced concentrations of radionuclides. The process condensate is collected in the condensate collection tank and transferred to a surface impoundment for storage before final treatment and disposal to the soil column. Figure 2 shows a simplified schematic of the 242-A Evaporator process.

The evaporation rate is limited to about 50 percent of the feed flowrate by the need to keep the slurry pumping rate high enough to avoid solid precipitation in the transfer line. If more volume reduction is desired, the slurry can be transferred from the underground storage tanks back to the 242-A Evaporator for additional evaporation. The overall volume reduction is limited by a variety of factors, including maximum density, solids content, and radionuclide content.

Figure 2 242-A Evaporator process flow schematic.



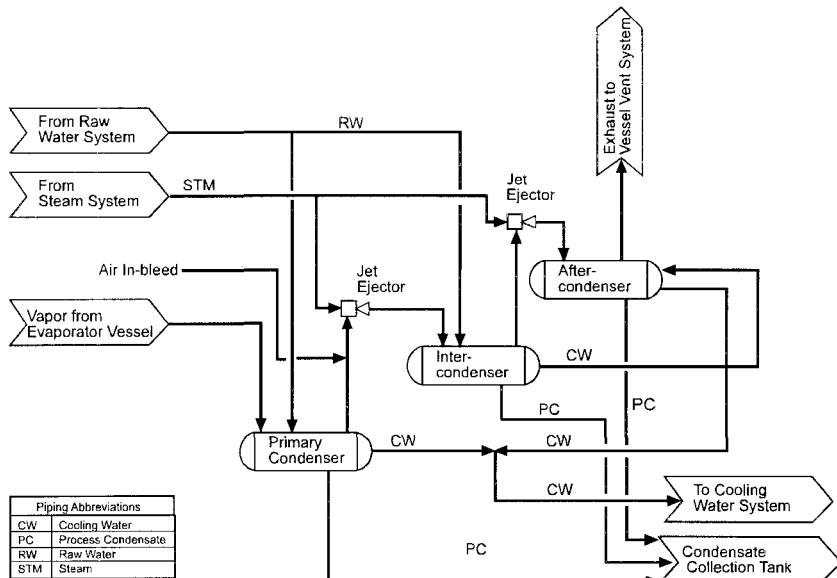
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Evaporator Vacuum Condenser System

Vapor in the evaporator vessel passes through two wire mesh deentrainment pads, which remove liquid droplets and particulates before these can enter the vacuum condenser system. The lower deentrainment pad is 10 centimeters thick and the upper deentrainment pad is 15 centimeters thick. Vapor drawn from the evaporator vessel flows into the primary condenser where the majority of the condensation takes place. The primary condenser is a large four-pass, tube-in-shell heat exchanger with a capacity of 6.4 megawatts. Uncondensed vapor exits the primary condenser to a jet ejector, while the condensed vapor (process condensate) drains to the condensate collection tank. Figure 3 shows a schematic of the vacuum condenser system.

Vacuum for the evaporator vessel and the vacuum condenser system is provided by two steam jet ejectors, arranged in series, which provide an absolute pressure of 5.5 kilopascals. The intercondenser and aftercondenser condense steam from the jet ejectors. Exhaust from the condensers exits to the vessel ventilation air cleaning system, while condensed vapor (process condensate) drains to the condensation collection tank. The intercondenser and aftercondenser are small heat exchangers, each with capacity of about 300 kilowatts. Water is used as the cooling fluid in all condensers.

Figure 3 242-A Evaporator vacuum condenser system.



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Air Cleaning

Exhaust from the vacuum condenser system enters the vessel ventilation system, which consists of two demister pads, a heater, two HEPA filters arranged in series, and an exhaust fan. After the fan, the exhaust exits the building through an 8-inch diameter pipe and exhausts to the atmosphere at an elevation of 19 meters above ground level. Volumetric flowrate through the vessel ventilation system is about 18 cubic meters per minute.

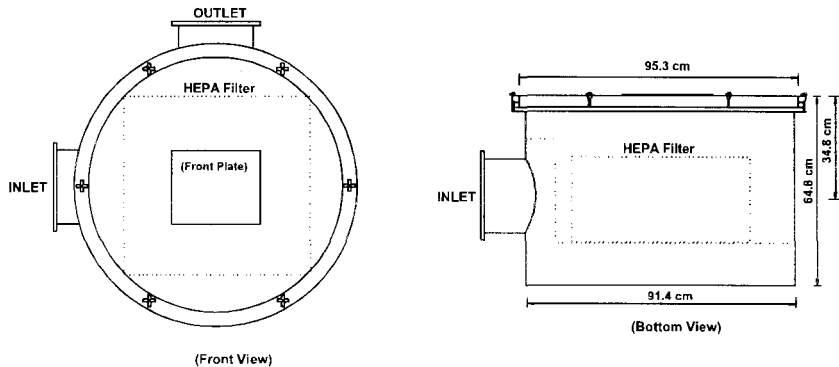
The demisters are identical 18-centimeter-thick wire mesh pads, which catch entrained droplets from the aftercondenser and drain the liquid to the condensate collection tank. The first pad could be flushed during outages. The heater consists of electric resistance coils, which heat the offgas stream to 66°C to prevent condensation in the HEPA filters.

HEPA Filter Assemblies

The HEPA filter assemblies are self-contained canisters; each canister contains a single HEPA filter that can be replaced as necessary through a sealable access cover. A drawing of an individual filter housing assembly with relevant dimensions is shown in Figure 4. A configuration drawing with appropriate dimensions of the filter assemblies and associated piping is given in Figure 5.

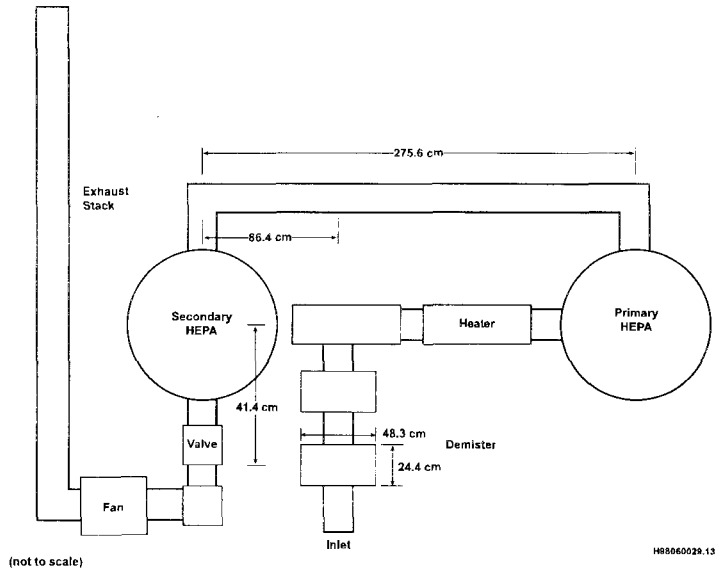
The primary and secondary HEPA filter housings are constructed from stainless steel sheet metal. The sheet metal housing is 14 gauge (1.91 millimeters) stainless steel. The size of the rectangular HEPA filter cartridge is 61 centimeters by 61 centimeters by 29 centimeters deep.

Figure 4 HEPA filter housing.



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Figure 5 242-A Evaporator exhaust train.



IV. Nondestructive Assay

The first NDA measurements performed insitu on HEPA filters on the Hanford Site 4 years ago used low resolution spectroscopy.^(1,2,3) This method was developed to determine the insitu radionuclide content on HEPA filters for use in assessing potential radionuclide emissions from stacks. The NDA measurements reported here have three major improvements: (1) high resolution from a high purity germanium (HPGe), detector allows the processing of uncollided gamma rays yielding no ambiguity concerning shielding;⁽⁴⁾ (2) a differential shielding technique allows for accurate measurements of a target source in the presence of very high levels of background radiation that cannot be totally shielded; and (3) the data were preprocessed onsite during the measurements to adjust for unforeseen sources of conflicting background radiation.

Equipment

A HPGe detector with collimation shielding and multiple measurement configurations was used on September 15, 1997 and November 6, 1997 to identify gamma ray emitting radionuclides and to establish their content on primary and secondary HEPA filters and the demister unit. The NDA measurements were performed in accordance with 40 CFR 61, Subpart H. The method analyzes the radionuclide content of the HEPA filters directly through the use of a portable gamma-ray

spectroscopy. The NDA measurement method for particulate radionuclides is described in 40 CFR 61, Appendix B, Method 114, "Test Methods for Measuring Radionuclide Emissions from Stationary Sources" for gamma emitting radionuclides using high resolution gamma ray spectroscopy.

NDA Measurements

The NDA measurements of the primary and secondary HEPA filters and the lower demister pad were performed with the detector inside both one and two collimation shields (Figure 6) yielding two measurement configurations at each location. When the outer shield was removed, the shield was positioned away from the measurement location so as not to interfere with the gamma ray flux field or other background gamma ray emitting sources. The collimation shield was positioned at each filter housing to record gamma ray activity from only the filter being measured, excluding all other filter assemblies. The detector location and cone of projection for each filter stage are shown graphically in Figure 7. The two configurations allow solving two unknowns at each measurement location. The two unknowns are the magnitude of radioactivity within the cone of projection and the background activity (or all radioactivity not within the cone of projection).

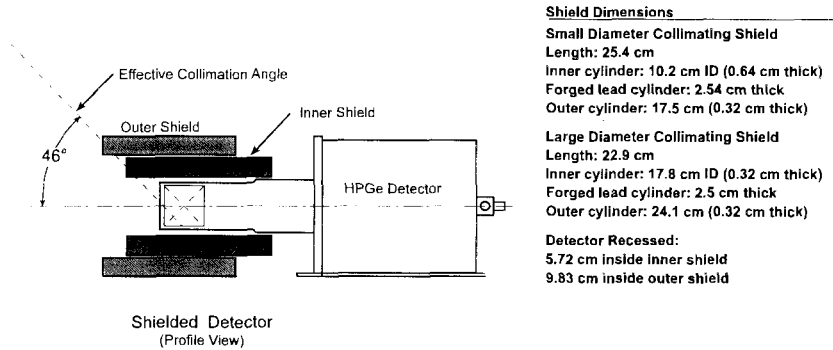
The counting system deadtime for all NDA measurements with the HPGe detector was less than 5 percent. The system linearity to increasing count rate activity was measured analytically and determined to require no secondary correction for deadtimes of less than 40 percent.⁽⁵⁾

The results of the NDA measurements of the primary and secondary HEPA filters are shown in Tables 1 and 2. The activity levels in the demister unit and the primary HEPA filter prevented accurate measurement of the radioactivity contained in the secondary HEPA filter. Consequently, the radioactivity has been estimated conservatively for the secondary HEPA filter.

There were two adjustments to the original measurement program made as a result of the site pre-analysis. Even though the lower demister pad is not considered a component of the exhaust control equipment, an NDA measurement was obtained when a preliminary survey identified the module as having higher gamma ray emissions than the primary HEPA unit. Because it was suspected that this unit had the potential of interfering with the post-campaign NDA measurements, results for the demister unit are included in Tables 1 and 2.

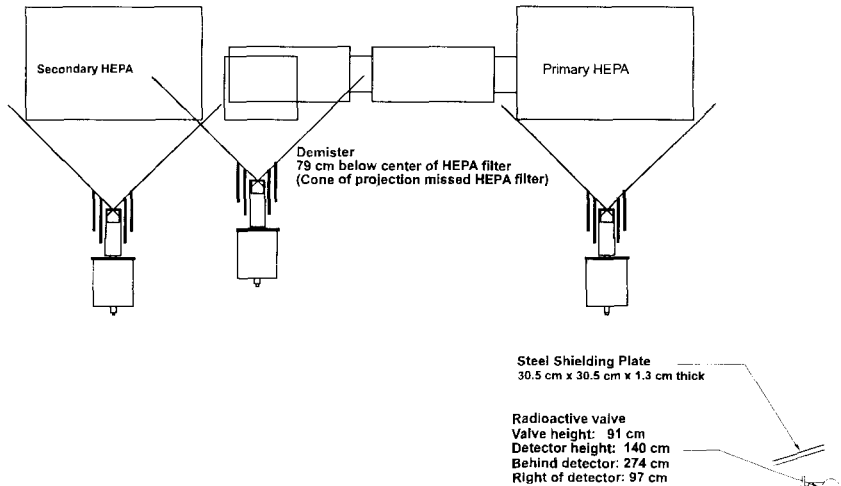
Another example of the value of site estimates of the NDA measurements involved a remote non-evaporator process valve on the other side of the room. During measurement of the primary HEPA filter, a valve containing higher activity than the primary HEPA filter was identified and suspected to be contributing to the unattenuated portion of the spectra measurement. As a result, additional measurements with two shielding thickness were placed between the valve and the detector. A minor contribution from the valve contamination to the unattenuated signal was identified and the computations eliminated this source of background interference.

Figure 6 HPGe collimation shielding.



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Figure 7 HPGe detector measurement locations for 242-A Evaporator.



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NDA Results

Gamma ray emitting radionuclides identified from the NDA measurements were cesium-137 and antimony-125. The radioactive content of the primary and secondary HEPA filters (excluding demister unit) for the pre-campaign NDA measurements was low at 2.2 E+4 becquerels (Bq) and 1.2 E+4 Bq total for ¹³⁷Cs and ¹²⁵Sb, respectively. The incremental increase in the gamma ray emitting radionuclides (post-campaign minus pre-campaign NDA measurements) in the HEPA filters was 2.1 E+04 Bq for ¹³⁷Cs. The uncertainty of the measurement results was 30%. The main factor contributing to the high uncertainty level was the low count rate recorded from the radionuclides in the filter housings. There was no statistically significant change between pre- and post-campaign measurements for the secondary HEPA filter or for ¹²⁵Sb. During the NDA measurements, the radiation from the demister was found to be the dominant background contributor.

Table 1 Pre-campaign results.

| Filter | Detector Position (centimeters) | Shield (single and double) ¹ | ¹³⁷ Cs (counts per second) | ¹³⁷ Cs Activity ² Bq | ²⁵ Sb (counts per second) | ²⁵ Sb Activity ² Bq |
|-----------|---------------------------------|---|---------------------------------------|--|--------------------------------------|---|
| Primary | 48.5 | Single | 1.64 | | .30 | |
| | " | Single | 1.88 | | .36 | |
| | " | 1 + Fe ³ | 1.67 | | .35 | |
| | " | 1 + Fe ³ | 1.72 | | .37 | |
| | " | Double | 1.97 | | .24 | |
| | " | Double | 1.95 | 2.2 E+04 | .33 | 1.1 E+04 |
| Secondary | 48.9 | Single | 0.16 | | 0.37 | |
| | " | Double | 0.03 | 3.3E+02 | 0.04 | 8.9 E+02 |
| Total | | | | 2.2 E+04 | | 1.2 E+04 |
| Demister | 33.2 | Single | 9.38 | | 4.02 | |
| | " | Double | 9.30 | 5.2 E+04 | 4.22 | 5.6 E+04 |

¹ Ordered for report uniformity, not necessarily order of measurement.² Uncertainties (1 sigma): Accuracy=30%; Precision=20%.³ Single shield plus iron shielding placed between process valve and detector.

Table 2 Post-campaign results.

| Filter | Detector Position (centimeters) | Shield (single and double) ¹ | ¹³⁷ Cs (counts per second) | ¹³⁷ Cs Activity ² Bq | ²⁵ Sb (counts per second) | ²⁵ Sb Activity ² Bq |
|-----------|---------------------------------|---|---------------------------------------|--|--------------------------------------|---|
| Primary | 48.8 | Single | 3.39 | | .29 | |
| | " | 1 + Fe ³ | 3.42 | | .37 | |
| | " | 1 + Fe ³ | 3.46 | | .34 | |
| | " | 1 + Fe ³ | 3.42 | | .23 | |
| | " | Double | 3.42 | 4.3 E+04 | .34 | 8.5 E+03 |
| Secondary | 48.8 | Single | 0.15 | | - | |
| | " | Single | 0.12 | | - | |
| | " | Double | 0.02 | <2.2 E+02 | - | - |
| Total | | | | 4.3 E+04 | | 8.5 E+03 |
| Demister | 48.8 | Single | 6.66 | | 2.64 | |
| | " | Double | 6.18 | 8.4 E+04 | 2.66 | 7.9 E+04 |

¹ Ordered for report uniformity, not necessarily order of measurement.

² Uncertainties (1 sigma): Accuracy=30%, Precision=20%.

³ Single shield plus iron shielding placed between process valve and detector.

V. Dose Estimate Based On NDA Results

The incremental increase for the campaign was used to estimate potential emissions and their resultant offsite dose. The offsite dose assessment was performed using the ratio of ¹³⁷Cs with other radionuclides in the feed material to calculate the total loading of radionuclides on the HEPA filters for the campaign. The resultant loading on the HEPA filters was assumed to be released and used as input to CAP88 for dose modeling.^(6,7) The sum of the individual doses is used to produce the total EDE to the MEI. The potential (non-filtered) offsite dose for the campaign is shown in Table 3.

The NDA study showed that 2.1 E+4 Bq of ¹³⁷Cs were accumulated on the primary HEPA filter during this campaign (compared to 2.2 E+04 Bq accumulation from the previous five campaigns, which had lower radionuclide content). The feed material was processed in a single pass, and contained a total of 1.4 E+16 Bq of ¹³⁷Cs. The release factor for the evaporator process is defined as the amount released divided by the amount processed. In this case, 2.1 E+4 divided by 1.4 E+16 produces a release factor of 1.5 E-12. The same release factor was found when the pre-campaign NDA measurement of ¹³⁷Cs was compared to the total quantity of ¹³⁷Cs processed during the first five campaigns.

Table 3 Dose assessment results.

| Radionuclide | Bq processed | Isotopic ratio ¹³⁷ Cs=1 | Loading ² on HEPA (Bq) | CAP88 dose factor (Sv/Bq) | Potential offsite dose (Sv) |
|------------------------------------|--------------|---------------------------------------|---|---------------------------------|--------------------------------|
| ⁹⁰ Sr | 2.0 E+14 | 1.4 E-02 | 2.9 E+02 | 1.2 E-17 | 3.5 E-15 |
| ⁹⁹ Tc | 5.4 E+12 | 3.8 E-04 | 8.0 E+00 | 2.9 E-19 | 2.3 E-18 |
| ¹³⁷ Cs | 1.4 E+16 | 1.0 E+00 | 2.1 E+04 | 6.5 E-18 | 1.4 E-13 |
| ²³⁸ Pu ¹ | 9.4 E+11 | 6.7 E-05 | 1.4 E+00 | 2.2 E-15 | 3.1 E-15 |
| ^{239/240} Pu ¹ | 3.7 E+12 | 2.7 E-04 | 5.7 E+00 | 2.3 E-15 | 1.3 E-14 |
| ²⁴¹ Pu ¹ | 1.5 E+14 | 1.1 E-02 | 2.3 E+02 | 3.7 E-17 | 8.5 E-15 |
| ²⁴¹ Am ¹ | 4.5 E+11 | 3.2 E-05 | 6.7 E-01 | 3.5 E-15 | 2.4 E-15 |
| Total | | | | | 1.7 E-13 |

¹ Feed material less than detection limit, calculated using detection limit.

² ¹³⁷Cs measured, other radionuclides calculated.

VI. Conclusions

A study was conducted on the HEPA filters of the 242-A Evaporator to determine if potential emissions from the stack were less than those that would require continuous compliant stack sampling. The study was performed using insitu NDA of the HEPA filters. Results of the NDA indicated that 2.1 E+4 Bq of ¹³⁷Cs were accumulated on the HEPA filters during an evaporator campaign. When divided by the feed material processed in the campaign, 1.4 E+16 Bq of ¹³⁷Cs, an airborne release factor for the evaporator process was calculated to be 1.5 E-12.

When the CAP88 model was run using the accumulated radionuclides on the HEPA filters during a campaign, an offsite EDE to the MEI of 1.7 E-13 Sv was calculated. This EDE is approximately 7 orders of magnitude below the 1.0 E-6 Sv/yr regulatory limit (0.1 mrem/yr). Using the maximum annual throughput and the release factor calculated for the 242-A Evaporator would increase the offsite EDE to 3.2 E-11 Sv/yr, which is approximately 4 orders of magnitude below the regulatory limit. Based on these results, the 242-A Evaporator stack was downgraded to a minor stack.

Given these results, two points can be made. First, the evaporator process is extremely efficient and yields a high volume reduction in the waste with minimal air emissions. Second, the successful application of insitu NDA measurements allowed a reclassification of the stack to minor status; continuous compliant sampling is not required.

Acknowledgments

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