Conceptual Approach For Estimating Potential Air Toxics And Radionuclide Airborne Emissions From A Temporary Exhaust System For The 216-Z-9 Crib Removal Action

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ABSTRACT

The 216-Z-9 Crib, located at the Hanford Nuclear Reservation in southeastern Washington State, was the site of a successful mining effort to recover plutonium from the contaminated soils at the disposal site. A CERCLA Action Memorandum (AM) issued by the U.S. Department of Energy (DOE) requires the removal of the buildings associated with this mining effort to facilitate a remedial action planned for the near future. The decontamination and demolition of the 216 Z-9 Crib facilities is required under a consent order between the DOE, the U.S Environmental Protection Agency (EPA) and the Washington State Department of Ecology (Ecology).

Removal of the buildings located on and near the concrete cover slab over the 216-Z-9 Crib will require removal of the large soil-packaging glovebox located inside the 216-Z-9A Building. Prior to cleaning out the glovebox, it will be necessary to provide active filtered ventilation capability to ensure a negative pressure exists between the glovebox and the adjacent airspace while hands-on work proceeds within. The glovebox floor is open to the Z-9 crib cavern environment below. For this reason the crib and glovebox currently share a common airspace. The functional requirements for safely conducting work within the glovebox include provision of a negative pressure in the box of about 0.5 inches of water gage (nominal) less than the interior of the building. In addition, the building surrounding the glovebox will be maintained at a slight negative pressure with respect to outdoor ambient pressure.

In order to assess the relevant and appropriate clean air requirements for the new temporary ventilation system and associated emissions monitoring, it was necessary to reliably predict the nature of the exhaust air stream. Factors used to predict the presence and concentrations of certain radionuclide particulates and certain gases considered to be

air toxics, included reliability parameters, flow rates, radionuclide content, and off-gas compositions. Radionuclide content includes transuranic isotopes, primarily of plutonium and americium. Air toxics include carbon tetrachloride, butane, methanol, acetone and toluene. Flow rate prediction was based on available design and test data and considered equipment sizes, glovebox negative pressure requirements, and filter flow characteristics.

The approach used to predict the off-gas composition from the crib required experience-based predictive analysis combined with crib head space analytical results. Input information for emission estimates included: (1) gas composition sample data obtained from recent samples taken within the crib head space during static conditions, and (2) air in-leakage/dilution estimates based on physical characteristics of both the crib and the new temporary ventilation system.

The conceptual approach combined measurement-based data with conservative assumptions, and provides the estimates necessary to determine relevance and appropriateness of substantive requirements under federal and state laws and regulations.

INTRODUCTION

The 216-Z-9 Recuplex CAW (CA column waste) Waste Disposal Cavern, also known as the Z-9 Trench or the Z-9 Crib, is located near the Plutonium Finishing Plant (PFP) facility at the Hanford Site in southeastern Washington State. The 216-Z-9 Crib was used as a disposal site for effluent chemical and radiological wastes from the recovery of uranium and plutonium through extraction or RECUPLEX process, a method that recovered uranium and plutonium from liquid and solid wastes and scraps from other PFP processes.

The Z-9 Crib was constructed as an engineered trench with an open area beneath a concrete slab. It is 6.4m (21-ft) deep with a 23cm (9-in) thick concrete slab measuring 27.4m by 36.6m (90-ft x 120 ft). The volume of the crib air space is approximately 3,398 cubic meters or 120,000 cubic feet. The Z-9 Crib received approximately one million gallons (4×10^6 L) of liquid wastes from 1955 through 1962 during its operating life. Analyses of the crib soil in seven locations to a depth of up to six feet (two meters) beneath the crib floor indicated that the plutonium content of the crib soil ranged from a low value of 50 to a high value of 150 kg (the highest concentration measured was 34.5 g/L of soil), with the majority of the contamination contained within the top two feet of surface soil. Information regarding deposition of plutonium and other radionuclides as well as sampling and analysis and hazards evaluations regarding the crib soil has been documented by Teal.[1]

In 1973, a decision was made to remove the top 30 cm of contaminated soil as a means of reducing the risk of a criticality incident and to recover plutonium from the soil. The soil was removed using a small excavator arm, with the mined soil carried into the glovebox via a conveyor. Mining equipment operated continuously from 1976 through 1978. Approximately 50.9 cubic meters (1800 cubic feet) of contaminated soil were mined and transported from the crib floor to the surface through the packaging glovebox.

Approximately ninety gross tons of soil containing approximately fifty-eight kilograms of plutonium was removed during the three-year operating period.

The 216-Z-9A Building has a small processing area containing the soil packaging glovebox and includes several support rooms. A portion of the floor under the glovebox is open to the crib interior below.

The total plutonium inventory (contained in the soil) of the glovebox has been measured by non-destructive assay as approximately five grams. Scheduled cleanup work at the Z-9 Crib includes removal of an estimated five to twenty-five liters of soil residues from the old packaging glovebox so that the glovebox can be removed from the 216-Z-9A building and disposed as low level waste.

Demolition of the 216-Z-9A building will require removal of the soil packaging glovebox located within the building. In order to support glovebox cleanout activities, the glovebox (Figure 1) must be reactivated (e.g., activities include glove replacement, disposition of glovebox internal waste, and removal of contaminated soil.). To assure the confinement and control of airborne contamination during conduct of clean-out work, an forced-draft HEPA ventilation system must be provided.

Figure 1. Soil Packaging Glovebox, 1976

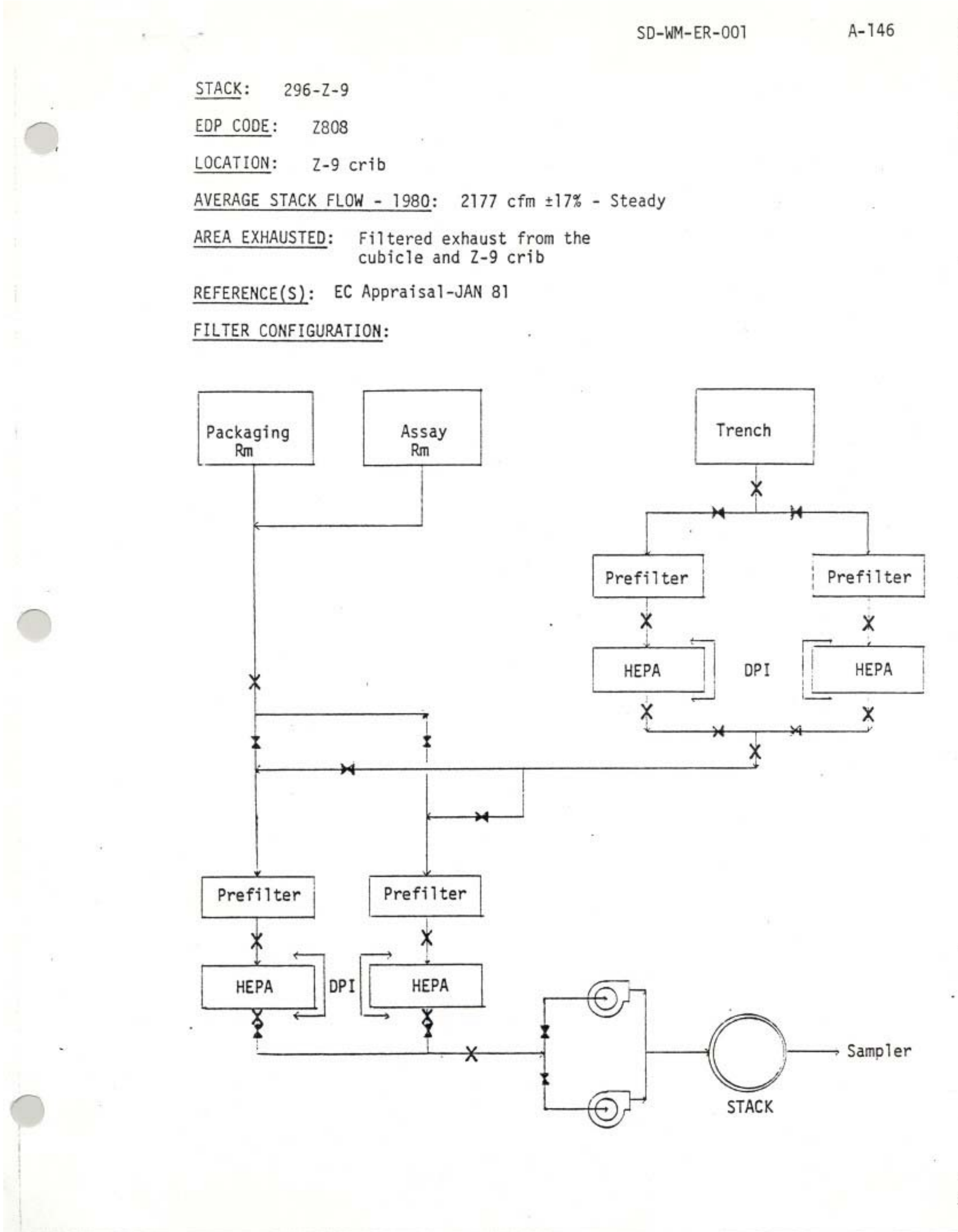
The major issue with applying negative pressure to the glovebox is that in order to pull a vacuum on the glovebox airspace which is integral to the crib airspace, a vacuum must also be pulled on the crib itself. Since the crib headspace contains toxic gases and radionuclide contaminated particulates, a small temporary exhaust system must be provided with appropriate abatement controls.

The work scope also includes the removal of the Mining Operator’s Cubicle (216-Z-9B). The Operators’ Cubicle was originally provided with an HVAC system that supported both worker comfort and the removal of heat from the high intensity crib lighting system. No HEPA filtration was provided in the original design since the operator’s airspace had no direct communication with the contaminated crib environment. Use of a temporary HEPA exhaust system to ventilate Building 216-Z-9B is not required.

The estimate for radionuclide release is based upon a specified set of assumptions from the Washington State regulations which require that estimation of the potential to emit airborne radionuclides must reflect a system operating for a year with no filtration of the exhaust air, but with operations otherwise normal.

DESIGN OF HVAC SYSTEM

The original ventilation exhaust system was installed on the 216-Z-9 Crib as part of construction project HCP-687 in 1975. A simplified flow diagram of the forced-draft ventilation system installed by the HCP project is described in Figure 2. (Note: The repackaging glovebox is not shown in Figure 2; it received supply air from the repackaging room and was exhausted through the crib.) A picture of the old system as taken from the outside of the building is presented in Figure 3. The operating basis of the old system involved withdrawing air from the crib cavity in order to impose a negative pressure in the cavity of approximately one-half inch measured on a water gauge. This pressure level is equivalent to a depression on a mercury-based barometer of 0.0385 inches of mercury.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STACK: 296-Z-9

EDP CODE: Z808

LOCATION: Z-9 crib

AVERAGE STACK FLOW - 1980: 2177 cfm ±17% - Steady

AREA EXHAUSTED: Filtered exhaust from the cubicle and Z-9 crib

REFERENCE(S): EC Appraisal-JAN 81

FILTER CONFIGURATION:

Figure 2. Schematic of Original HVAC Parameters

Figure 3. Original Deactivated HVAC System

Prior to terminating the active exhaust, the original crib vent filters were modified in late 1981. One vent filter was capped. The other was replaced with a new (passive) HEPA-type filter.

Review of the system design information reveals the former supply and exhaust systems were isolated in the spring of 1982. The building air supply system has been blanked near the supply fan discharge. The process (crib) exhaust is blanked on the duct exiting the crib (see FCN 55738.). The room exhaust was blanked at exhaust inlet grilles located within the building (see FCN 56493). The glovebox inlet HEPA filters were also blanked. The glovebox inlet filter assembly was modified to attach a second passive HEPA filter vented to atmosphere (see FCN 56485). The current design ensures the dominant flow path during barometric pressure changes is between the two breather filters. However, given the large surface area of the crib cover, additional unfiltered leakage paths are possible and expected. In support of the Z-9 Crib deactivation activities, existing passive vent filters will be replaced.

This original HVAC system was inspected and evaluated for use during the demolition of the mining complex of buildings. System walk-downs revealed significant corrosion of the carbon steel forced-air ventilation equipment including filter housings and ducts. A determination was made that reactivating the original system is not a viable option for supporting the deactivation work. Therefore, installation of a smaller-capacity temporary forced-air HEPA-filtered exhaust system is planned. A system is planned that would be operated for short intervals, rather than a continuously operating system. However, the exhaust system must be capable of operating continuously throughout the work evolution. The design must allow easy transition between active and passive ventilation modes with little or no operator intervention. A proposed system design concept is shown in Figure 4. The design is based on preliminary model estimates for predicting the concentration and behavior of several toxic gases and particulates of soil contaminated with radionuclides. The proposed design provides sufficient airflow to ventilate the glovebox and crib, addresses potential moisture in the exhaust air stream due to condensation, and provides an appropriate number of testable HEPA filter stages to ensure low emissions.

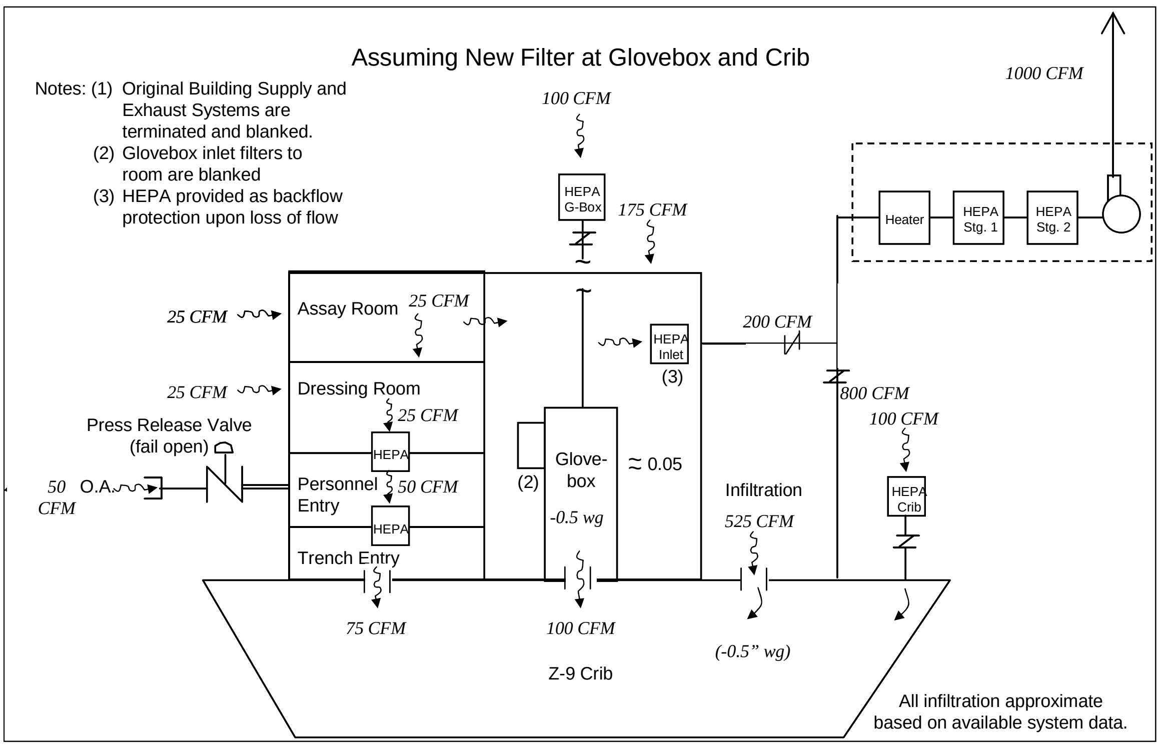

Figure 4. Schematic of Proposed New HVAC System

Toxic Air Emissions Estimate

To determine the concentration of toxic air pollutants in the headspace of the crib, Summa canister samples of the headspace gases were taken on four occasions from December 2006 through June of 2007. Results were documented in *Facility Condition Analysis for the 216-Z-9 Trench and Cover Slab to Support Removal of the Above-Grade Soil Mining Structures*, Hopkins, 2007[2]. Analytical values for carbon tetrachloride, butane, methanol, acetone and toluene were evaluated for regulation as air toxics to be considered for monitoring. Additional information on characterization and structural analysis of the 216-Z-9 Crib can be found in *Structural Characterization Work Plan for the Photographic Characterization of the 216-Z-9 Trench by Cameras and Crawler*, and *Decommissioning The 216-Z-9 Crib Plutonium Mining Facility At The Plutonium Finishing Plant: Issues Characterization*, Hopkins, 2007.[3, 4]

In order to design the HVAC system with appropriate HEPA filtration, a model for predicting the inventory and behavior of gases within the crib air space and possibly from floor soil when a negative pressure is imposed on the crib cavity and the effects of increased flow through the crib was needed. Two approaches were considered for estimating potential air toxic emissions: 1.) Generation rate is constant and independent of flow; and, 2.) Generation rate varies directly with flow.

The concept that gas generation rate is constant and independent of crib air flow (Approach 1) was considered as a possible approach for quantifying the potential emissions of toxic air pollutants exiting the new HVAC system. In order to fully evaluate and validate this approach, more data regarding crib air exchange as a function of varying barometric pressure, soil mass transfer diffusion coefficients and other data were required. Although this approach is discussed, the requisite data were not available to completely validate the approach. The second approach, "generation rate varies directly with air flow", is the approach chosen for determining the gas emissions estimates and provides a more conservative approach to estimating potential toxic air pollutant emissions. Both approaches are discussed below.

Approach 1:
Generation Rate Is Constant And Independent Of Crib Air Flow Rate

For Approach 1, if the assumption is that the generation rate of toxic air pollutants is constant and independent of air flow, the air toxics can be calculated based on the existing passive turnover of the crib air volume.

Supporting the assumption is the fact that the crib environment has been minimally affected by external forces other than barometric pressure changes during the last 30 years. The bulk of any liquid volatile organics near the surface should have long ago evaporated creating a quasi-steady state condition within the crib environment with only minor changes in airborne concentrations possible due to dilution effects associated with crib breathing. Presence of free liquids in the floor of the crib is unlikely and was not observed in recent high resolution photography of the crib.. It is assumed that molecular diffusion through the soil column due to concentration gradients is the primary driver for current chemical airborne levels. These levels are not expected to significantly increase with increased ventilation through the crib; therefore, the air toxics generation rate may be viewed as essentially constant. An estimate of air toxic emissions can be developed using the highest measured concentrations and existing metrological data.

Effects on the crib system that would be imposed by barometric pressure variations were reviewed for impact. The total effect was estimated and identified to be negligible with regard to affecting the output of air and gases from the new ventilation system proposed. Investigations of barometric pressure relationships to Hanford waste tanks has been documented in *Barometric Pressure Variations,* M.D. Crippen, 1993 [5] and identifies an average historical barometric pressure of 29.92 inches of mercury and a total yearly breathing of approximately 1.69 atmospheres (49.4 inches of water). The proposed HVAC system will impose an additional -0.5 inches of water vacuum on the crib environment. Because the crib environment will track to normal barometric pressure, the soil column has been exposed to the full range of normal atmospheric pressures, the additional -0.5 inch vacuum from an active ventilation system will have negligible effect. Neglecting temperature, by applying the ideal gas law, an estimate of total air exchange rate between the Z-9 crib and the environment can be calculated based on the volume of the crib and the total pressure change per year (49.4 inches of water.). Then determining the relative concentrations of the air toxic constituents in the total volume of crib air is possible.

An active ventilation system on the crib will greatly increase the crib air exchange rate from the current from passive atmospheric breathing rate. Based on infiltration estimates of the original design and available test data, approximately 800 cubic feet per minute (cfm) must be exhausted to achieve a crib pressure of approximately -0.5 in. water gage. Most of this air will be introduced as infiltration and will occur near the top of the crib through small cracks and crevices. Air velocity through these infiltration sites can be quite high, but the resulting higher-velocity air currents should degrade rapidly after entering the crib environment leaving little potential for suspension of contaminants at the crib floor level (location of concentrated chemical and plutonium-laden soil.)

Introduction of cold or hot air into the crib has the potential to create thermal currents and significantly increase air movement within the crib. Increased air movement could disturb a small percentage of surface contamination present on the crib support columns, abandoned mining equipment, crib ceiling, etc. However, because much of the contamination has bonded to the soil particles, the bulk of the disturbed particles should settle out and fall to the floor. The increased flow rate could also result in a more cyclic and wider spread of crib air temperature ranges. As such, moisture content of both the soil and crib airspace will rise and fall based on ambient conditions.

Although diffusion coefficients through soils have some temperature dependency, relatively minor changes in average soil temperatures should have little impact on the overall diffusion rate. It is believed, that once the crib is actively ventilated, the average VOC concentration in the crib airspace will asymptotically approach zero over time. This approach assumes that gas generation rate from the soil is constant. Therefore, annual total emissions can be estimated by simply adding the calculated annual emissions in the passive mode plus one volumetric air exchange of the crib.

Obtaining accurate estimates of soil diffusion coefficients for the various chemical constituents would require additional sampling and testing. Current data are not available to validate the assumption that gas generation rate remains constant over the duration.

Approach 2: Generation Rate Varies Directly with Crib Airflow Rate

Approach 2 assumes the generation rate will increase in direct relationship to flow in order to maintain a constant crib concentration. Using the maximum measured concentrations and a crib exhaust airflow rate of 1000 cfm with 800 cfm passing through the crib air space, total emissions can be determined by multiplying the airborne concentration by the amount of air displaced over a years' time.

Because no mechanism was identified that supports the assumed direct relationship between air toxics generation rate and crib airflow rate, Approach 2 clearly provides a more conservative estimate of air toxic emissions and compensates for uncertainties associated with Approach 1.

Air Monitoring Plan Requirements

The 216-Z-9 radioactive air monitoring plan establishes the method of controlling and monitoring radioactive air emissions for the D&D of the 216-Z-9 Crib soil mining structures. The plan includes a description of the ventilation exhaust system required for reactivation of the Z-9A glovebox. The plan is required by the CERCLA and addresses the substantive requirements of *Washington Administrative Code* 246-247 and 40 *Code of Federal Regulations* Part 61 Subpart H. Substantive requirements for air pollutant emissions, controls, and monitoring are derived from regulations promulgated under the federal Clean Air Act of 1990 and Amendments (42 USC 7401 et seq.), and the Washington Clean Air Act (RCW 70.94).

The federal and delegated state implementing regulations addressing the *National Emission Standards for Hazardous Air Pollutants* Subpart H (40 CFR Part 61) require that the combined radionuclide airborne emissions from the U.S. DOE Hanford Site shall be controlled so as not to exceed amounts that would cause an exposure to any member of the public of greater than 10 millirem per year (mrem/yr) effective dose equivalent. The same regulations address point sources (i.e., stacks or vents) emitting radioactive airborne emissions, requiring monitoring of any such sources with a major potential for radioactive airborne emissions (i.e., those with a potential to provide greater than 0.1 mrem/yr effective dose equivalent to the maximum public receptor), and requiring periodic confirmatory measurement of any lesser source emissions, sufficient to verify low emissions. Handling radiologically-contaminated materials during the Z-9 demolition activities has the potential to generate radioactive air emissions.

Radionuclide inventory is based on the crib soil inventory located in the glovebox itself resulting from soil spillages. Plutonium inventory was determined by gamma spectroscopy analysis.[2] Based on anticipated radiological conditions in the trench, conservative estimates of potential emissions were evaluated based on the unit dose factors in *Calculating Potential to Emit Radiological Releases and Doses* [6] to determine the annual potential to emit (PTE) to the hypothetical maximum exposed public individual (MEI). The approved dose models show the MEI for PFP would be located at the Laser Interferometer Gravitational Wave Observatory (LIGO), situated 18,310 meters East/Southeast of the 200 West Area. The primary radionuclides of concern are plutonium 238, plutonium 239, plutonium 240, plutonium 241, plutonium 242, americium 241, uranium 234, uranium 235, uranium 236, uranium 237, uranium 238, and neptunium 237. Americium 241 values are used to represent all the transuranic isotopes above for conservative calculation of the PTE.

Results of PTE and Air Toxics Calculations Using HVAC New System Conceptual Design

The crib concentration and volume of potentially regulated gases were directly analyzed and the radionuclide inventory of the glovebox was measured non-destructively. The amount of potentially contaminated soil which could be lofted was estimated. With regard to the presence of radionuclides that would be discharged from a new HVAC system, two approaches were considered for estimating the quantities of radionuclides that would be discharged. To satisfy regulatory requirements, the analysis assumed that the system would operate continuously for one year and that the system would have no HEPA filters. This approach, based on WAC 246-247-030 (22), requires an assumed release fraction for particulate solid radionuclides of 1E-03 of available material. The available material is based on the plutonium content of the contaminated soil: 4.75 grams from the glovebox and a possible 0.6 grams from dropped materials reaching the crib floor. The total available material is 5.35 grams.[2] Therefore, the PTE release fraction is .00535 g plutonium or 5.35 x 1 E-3.

Given the relatively low potential to emit and the anticipated limited operating duration of the system, installation of continuous air monitoring is not warranted. In lieu of stack air monitoring or sampling, it is proposed that the final forced-air filter be removed upon system removal for analysis to support estimates of emissions for annual reporting purposes.

Two approaches to estimating the quantities of air toxics that would be released by operation of the new system were evaluated. Approach 1 assumed a constant soil generation and release rate of chemical constituents while Approach 2 assumed a constant airborne concentration stack release of air toxics entrained in the 800 cubic feet per minute (cfm) coming from the crib air space based on the highest measured value obtained through sampling with no credit for dilution. The calculations and results are found in the removal action work plan for the crib mining complex. Given the low potential to emit toxic air pollutants, no engineered controls are warranted to capture chemical emissions.

Effect of Barometric Pressure on Crib Gas Dynamics

Effects on the crib system that would be imposed by barometric pressure variations were reviewed. The total impact was found to be negligible with regard to affecting the output of air and gases from the ventilation system proposed. Investigations of barometric pressure relationships to Hanford waste tanks has been documented in *Barometric Pressure Variations*, M.D. Crippen, 1993 [5] which identifies total yearly breathing as approximately 2.21 inches of mercury or approximately 50 inches of water. The HVAC system will impose -0.5 inches of vacuum on a water gage. This represents approximately 1.0% of the total pressure variation imposed by the natural barometric pressure changes at the crib over a one-year time interval.

Removal of Old Ventilation System

The removal of the old ventilation system and filter boxes will not require a forced-air emission control system. Approved procedures for use of fixing contamination with foam or other state of the art fixants, wrapping, blanking and glove bags and other containments will be used to remove the old ventilation system in sections. Once the potential for minor emissions is confirmed, HEPA vacuums will be used as necessary during the removal of the old ventilation system at the Z-9A Building during decommissioning activities. These will vary in size and primarily be small portable units currently in use on the Hanford site with flow capacities between 50 and 300 cfm, but a larger capacity unit with flow rates could also be used. These units would be used to supplement the approved work package contamination controls. Vacuuming using one of these devices would be controlled based on the specifics of the situation.

To verify low emissions, emissions will be analyzed to assure minor status, and a contamination survey of the outlet of the HEPA-type filtered device will be performed at the completion of each use to verify low emissions. Due to the nature of the activities using HEPA-type-filtered air movers, significant abated release associated with these devices is not anticipated, and the existing near field monitoring network will be used to measure air emissions for the activities associated with these temporary point sources in conjunction with the fugitive unit. The Hanford site protocol established for near-facility monitors will be followed for data collection, sampling frequencies, sample analysis, and data reporting (DOE/RL-91-50, or latest revision). Emissions will be reported as part of the Hanford Site annual reporting.

CONCLUSIONS

Removal of the buildings located on and near the concrete cover slab over the 216-Z-9 Crib will require removal of the large soil packaging glovebox located inside the 216-Z-9A Building. Prior to cleaning out the glovebox, it will be necessary to provide an active filtered ventilation capability to ensure a negative pressure with respect to the airspace while hands-on work proceeds within the glovebox. The functional requirements for safely conducting work within the glovebox include provision of a negative pressure in the box of about -0.5 inches of water gage with respect to the interior of the building. In addition, the building surrounding the glovebox will be maintained at a slightly negative pressure with respect to outdoor ambient pressure.

In order to assess whether certain clean air requirements are relevant and appropriate to the new temporary ventilation system and associated emissions monitoring, it was necessary to reliably predict several details of the nature of the exhaust air stream. These details included reliability parameters, flow rates, radionuclide content, and off-gas compositions to predict the presence and concentrations of certain expected gases considered to be air toxics. The radionuclide content will include transuranic isotopes, primarily plutonium and americium. The air toxics will include carbon tetrachloride, butane, methanol, acetone and toluene. Flow estimates were based on infiltration rates assumed in the original ventilation system and as supported by available system data.

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