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Abatement Case Study

March 2024

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Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory Richland, Washington 99354

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1.0 Introduction

Radioxenon emissions from industrial sources such as fission based medical isotope production (MIP) facilities and nuclear reactors are generally known to be well below levels of public health and safety concern. However, the global background of radioxenon produced by MIP interferes with nuclear explosion monitoring by the International Monitoring System (IMS) developed for the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO PrepCom) (CTBTO PrepCom, 2024). It was calculated that xenon emissions levels of 5×10⁹ Bq/day ¹³³Xe were low enough to have minimal impact on International Monitoring System (IMS) stations (Bowyer et al, 2013). There are several technologies currently used to abate radioactive xenon emissions to meet regulatory release levels, and some alternative methods have been investigated to reduce xenon release levels well below required regulatory levels (Doll et al, 2014, Gueibe, et al, 2014). While MIP producers are sympathetic to the issue of radioxenon interference with IMS monitoring, the cost to implement and maintain additional abatement systems has resulted in limited implementation. Therefore, more cost-effective options for xenon abatement are needed to help reduce the impact of these emissions on nuclear explosion monitoring.

In this paper study, two options for lowering ¹³³Xe emissions at a hypothetical MIP facility were investigated. The releases at the facility were reduced from regulatory levels the facility was designed to meet, to significantly lower voluntary 5×10⁹ Bq/day ¹³³Xe release levels to minimize the impact on nuclear explosion monitoring (Bowyer et al, 2013) – this is considerably lower than the required regulatory levels that typical facilities are designed to achieve. The two options considered in this study are 1) doubling the number of charcoal delay beds and 2) cooling the vault that contains charcoal delay beds in order to increase the holdup time of radioxenon. This longer holdup time allows more of the short-lived xenon isotopes to decay prior to being released to the environment, and therefore reduces the total xenon inventory released from the facility. As part of this comparison, we estimate costs to install each option and discuss their potential impact on the operational facility.

2.0 Background

The IMS incorporates seismic, hydroacoustic, infrasound and radionuclide stations to detect potential nuclear explosions. When fully implemented, 40 of the 80 total radionuclide stations will have highly sensitive noble gas monitoring capability (currently there are 39 noble gas systems planned with the 40th location to be named later) (CTBTO PrepCom, 2024). Radioactive xenon backgrounds were first measured when testing noble gas detection systems for the IMS. Upon investigation, it was discovered that the major source of these backgrounds was MIP with other sources such as nuclear power plant (NPP) facilities contributing at a lower level (Bowyer, 2021). Because the radioxenon background from MIP can be very similar in concentration and isotopic signatures of the four treaty relevant xenon isotopes produced during fission (^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe), it is difficult to distinguish between xenon released from MIP and underground nuclear testing. While MIP facilities do incorporate methods to reduce the levels of radioxenon released to meet health and safety requirements, emissions are regularly detected by the IMS nearly every day across the globe. Releases from MIP are typically between 2.0x10⁹ Bq/day and 1.1x10¹³ Bq/day (Miley, 2023), depending on the production level and xenon abatement methods incorporated at that facility, which at the higher end is orders of magnitude above the voluntary xenon emissions level of 5×10⁹ Bg/day ¹³³Xe which has been suggested would reduce the impact radioxenon released from industrial sources.

There are several methods that have been incorporated for reduction of radioxenon from industrial facilities but charcoal delay beds and holding tanks are the most abundantly utilized (Doll et al, 2014). These methods take advantage of the relatively short half-live of xenon isotopes used for nuclear explosion monitoring (^{131m}Xe-11.84d, ^{133m}Xe-2.19d, ¹³³Xe-5.24d, and ¹³⁵Xe-9.1h) by slowing the release from the facility to allow for the xenon isotopes to decay to lower concentrations. As xenon and iodine isotopes (which decays to form xenon isotopes), flow through a charcoal bed, the molecules interact with the charcoal which slows the progress through the bed. Reducing the temperature increases these interactions, resulting in longer holdup times. Therefore, either increasing the number of charcoal beds or cooling the charcoal will result in longer holdup times, allowing the radioxenon more time to decay and reducing the xenon concentrations released.

3.0 Medical Isotope Production Xenon Abatement System (Hypothetical)

A hypothetical MIP facility with a source term of approximately 2x10¹⁴ Bq/day each of ¹³³Xe and ¹³⁵Xe was modeled which employs a train of eight charcoal abatement beds to meet their regulatory requirements for the abatement of gaseous radioactive xenon and iodine species. PNNL simulated steady state operation of the abatement beds, using the equations described in (Ritzmann et al. 2024), assuming the incoming gas and the vault holding the beds are maintained at 70°F. The temperature distributions based on the simulations are shown in Figure 1 and illustrate how the first three beds experience minor elevations in temperature. The highest temperature expected is 73.5°F in the first bed. All the beds have peak temperatures within 15 inches of inlet at the top of the bed. Unsurprisingly, the peak temperatures occur at the center (radially) of the bed.



Figure 1. Temperature (°F) distribution in each of the eight abatement beds. From right-to-left, top: beds 1-4, bottom 5-8.

The model predicts an overall holdup time of 35.1 days for ¹³³Xe with other xenon species in the same range (35.0-35.2 days) for the steady state assumption. The long holdup time for xenon means that any isotope/species with a short half-life (e.g., ¹³⁵Xe, ^{135m}Xe) is reduced nearly completely due to decay. The steady state simulations provide important context regarding the performance of the as-designed noble gas abatement system. The model results suggest a holdup time slightly lower than the 5 days per bed expected by the producer; however, the adsorption isotherm used in the models leads to predicted holdup times shorter than what has been observed. The influence of this, and other, modeling choices on the predicted holdup time is discussed in (Ritzmann et al. 2024).

4.0 Methods to Reduce Xenon Emissions

4.1 Doubling the charcoal beds

4.1.1 Overview

The first method investigated to reduce xenon emissions is increasing the number of charcoal beds used for xenon holdup. The additional beds will slow the release of radioxenon and allow additional decay to stable isotopes forms prior to being released to the environment. For an existing facility there would unlikely be space within the facility. Therefore, employing this method would require construction of a standalone building to house the additional charcoal beds outside of the main facility and modifications to redirect the flow exiting the existing charcoal beds to the new beds.

4.1.2 Modeling to determine the additional number of charcoal beds

The number of additional charcoal beds required to reach 5×10^9 Bq/day ¹³³Xe released was based on modeling of the facilities abatement system outlined in Section 3. It was determined that doubling the bed size would increase the xenon holdup time from approximately 35 to 70 days and decrease the emissions from the facility below 5×10^9 Bq/day ¹³³Xe.

4.1.3 Cost estimate for additional charcoal beds

PNNL created a cost estimate for expansion of the original 8 adsorbent bed configuration. The cost estimate basis assumes construction of a building with 8 additional beds with an identical configuration to the base case 8 bed design to meet regulatory requirements along with modifications to the current facility. This will result in a total of 16 adsorbent beds from a greenfield project that includes the marginal cost of building the additional 8 beds.

The adsorbent vessels for the charcoal beds are based on drawings of the existing carbon adsorbent beds (6' diameter x 12'6" height) with 316L stainless steel metallurgy. The cost of each bed was estimated by Aspen Capital Cost Estimator (ACCE version 14) at \$350k/vessel. This Piping and instrumentation were then added to each drum cost. The piping and instrumentation configuration was obtained from Piping and Instrumentation Diagrams (P&IDs). All 8 drums with the associated equipment, piping and instrumentation had a total installed cost of \$2.8M. The drum inventory (activated carbon) was estimated to cost \$10/lb. The packing density was assumed to be around 39 lb/ft³, slightly in excess of the packing density in the technical bulletin by NUCON® (NUCON 2019) but consistent with the parameters used in the modeling. The total inventory required was calculated from the bed dimensions was approximately 11,000 lbs, resulting in a fill cost of \$110k/bed or \$900k for the 8 beds.

The equipment is housed in a concrete and steel bunker with dimensions of 18'Wx38'Lx20'H. Assuming 24" thick concrete walls and an average of 1" thick steel for every wall (including the floor and ceiling), the bunker construction cost is \$1.2 million (\$0.2 million for concrete and \$1 million for steel). There are also additional costs to the main building, which sits over the bunker space. Assuming the building floor area increases by 700 sf (18'x38') with a floor space cost of \$1,000/sf, the additional building cost is \$700k. The estimated total cost to build a new vault containing 8 beds is provided in Table 1.

| Item | Cost (Millions) |
|-----------------------------|-----------------|
| Carbon vessels | \$2.8 |
| Activated carbon material | \$0.9 |
| Building | \$0.7 |
| Bunker (concrete and steel) | \$1.2 |
| Initial total cost estimate | \$5.6 |
| | |
| 30% contingency | \$1.7 |
| Estimated total cost | \$7.3 |

| Table 1. Summary of total m | arginal cost | for additional | charcoal beds. |
|-----------------------------|--------------|----------------|----------------|
|-----------------------------|--------------|----------------|----------------|

4.2 Cooling the delay beds

4.2.1 Overview

The second method investigated to reduce xenon emissions was cooling of the vault that contains the 8 charcoal beds to meet regulatory emissions limits. Cooling the charcoal beds will slow down the interactions between xenon and charcoal, resulting in longer holdup times on the same amount of adsorbent. To facilitate cooling of the vault where the charcoal beds are located, modifications to the vault will need to be made to add insulation and cooling capabilities. Additional drying of the process gases prior to entering the cooled charcoal beds should also be considered to prevent condensation and potential freezing of water. Those costs are not included in this estimate.

4.2.2 Modeling for cooling of delay beds

Modeling of the current charcoal beds used for xenon abatement was performed at various temperatures to predict the temperature that the system would reduce the emissions from the facility to 5×10^9 Bq/day ¹³³Xe and the amount of heat generated and released into the vault due to decay of radioisotope held on the charcoal beds. The model discussed above (Section 3) was used to model the performance of the abatement beds when the walls of the charcoal beds were cooled below 70°F. The inlet gas was maintained at 70°F. This represents the case where the vault holding the beds is cooled by an HVAC system. Steady state operation was subsequently simulated in a sweep starting at 70°F (the base case presented in Section 3) and stepping the wall temperature down in 0.5°F increments to a final temperature of -4°F. Radioactive decay energy heats the carbon bed and the process gas resulting in elevated temperatures as the gas exits each bed. The heated process gas transfers some energy to the colder vault as it passes through the piping between beds.

The results for several temperatures determined in this model are tabulated in Table 2 and shown in Figure 2. The results indicate that at 21° F (-6°C) the system would reach the 5×10⁹ Bq/day ¹³³Xe emissions level given inputs of ~2.0×10¹⁴ Bq/day ¹³³Xe and ¹³⁵Xe. It is worth noting that at the coldest temperatures, the ^{131m}Xe emissions may be exceed the ¹³³Xe emissions due to the longer half-life of ^{131m}Xe.

| Ambient Temperature | ¹³³ Xe Emissions (Bq/day) |
|---------------------|--------------------------------------|
| 70°F (21°C) | 1.3×1012 |
| 32°F (0°C) | 2.8×1010 |
| 21°F (-6°C) | 5.0×10 ⁹ |
| -4°F (-20°C) | 1.8×10 ⁷ |

Table 2. Predicted ¹³³Xe emissions at various temperatures



Figure 2. Predicted ¹³³Xe emissions with varying charcoal bed temperature

To better determine the amount of cooling required for the vault, estimated heat flows from the bed were considered. At steady state, heat generated in the beds must be balanced by heat removed from the beds either through increasing the temperature of the outlet gas or transferring heat from the bed to the surroundings. As greater xenon abatement is achieved, there is a greater amount of decay energy that must be removed from the system. The calculated heat flows show that this is the case, with the warmer beds emitting only 42 W at temperature (70°F) while increasing to 2500 W at the coldest temperature investigated (-4°F). As the temperature goes down, the fraction of the heat produced in the first bed increases, see Table 3. This is because cooling increases the holdup time in each of the beds causing a greater amount of xenon to decay in the first bed.

| T _{ambient} | $\dot{Q_1}$ | $\dot{Q_2}$ | $\dot{Q_3}$ | $\dot{Q_4}$ | $\dot{Q_5}$ | $\dot{Q_6}$ | $\dot{Q_7}$ | $\dot{Q_8}$ | Q_{Total} |
|----------------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| 70°F (21°C) | 19 | 11 | 6 | 3 | 2 | 1 | 0 | 0 | 42 |
| 32°F (0°C) | 800 | 93 | 17 | 4 | 1 | 0 | 0 | 0 | 920 |
| 21°F (-6°C) | 1200 | 120 | 18 | 2 | 0 | 0 | 0 | 0 | 1400 |
| -4°F (- 20°C) | 2260 | 200 | 20 | 2 | 0 | 0 | 0 | 0 | 2500 |

| Table 3. Hea | at flow (W | V) transferre | ed out of | each bed | at various | temperatur | es along | with the | total |
|--------------|-------------|---------------|-----------|------------|------------|-------------|-----------|-----------|--------|
| he | eat flow fr | om all the l | oeds into | the vault. | Heat flows | s are given | to two si | gnificant | digits |

One of the challenges that these results highlight is that uniformly cooling a vault will not be easy with one bed acting as the main heat source. This will lead to localized heating that may result in the first bed experiencing a higher ambient temperature than the remaining beds. The design of the cooling system and layout of the beds may prove an important design criterion when considering this abatement approach.

4.2.3 Cost estimate for cooling charcoal beds

Estimates for the vault size to be cooled were made based on the schematics in Figure 3. The vault for the charcoal beds is 15.5'x36.5'x20' and cooled to $21 \degree F (-6\degree C) \pm 5\degree F$ controlling the relative humidity below condensation levels. For reliability, redundant cooling units shall be employed and installed outside of the building. A system shall be included to monitor the vault temperature and alarm staff if outside of the specified temperature range. Radiation decay from the 8 charcoal beds is expected to generate 1400W of heat within the vault which must be accounted for when designing the cooling system – most of the heat will originate from the first 2 charcoal beds, see Table 3. The walls and ceiling will be insulated but not the floor due to the charcoal beds in the vault and one standard sized maintenance access door will be included, which will only be opened for maintenance. It should be noted that the floor should be insulated below grade during construction to avoid surface cracking of the floor due to water permeating from the ground to the floor surface. The estimated dimensions and other parameters used for acquiring quotes for cooling the vault are summarized in Table 4.





| Parameter | Value | Comments |
|----------------------------|---------------------|---|
| Number of delay beds | 8 | |
| Tank height | 12.6' | |
| Tank width | 6' | |
| Interior vault width | 15.6' | |
| Interior vault length | 36'4" | |
| Interior vault height | 20' | |
| Standard door | | (no or little access) |
| Cool to | (-6°C) 21°±5° F | Maximum temperature to meet 5×10 ⁹ |
| | | Bq/day ¹³³ Xe releases |
| Heat flow in space | 1400W | (most heat from first 2 beds) |
| Relative Humidity | Maintain RH below | |
| | condensation levels | |
| Chart recorder/alarm | | |
| Redundant coolers | | For reliability |
| No insulation on the floor | | Due to charcoal beds being in place |
| Vault wall thickness | 24" Concrete | |

Table 4. Estimated parameters for charcoal bed vault.

4.2.3.1 Cooling system and installation

An estimate to supply a custom sized cooling and insulation system for the charcoal bed vault was prepared by American Walk In Coolers (AWIC) in Tucson, AZ based on the charcoal bed vault parameters and cooling requirements listed in Table 4. This estimate includes 4" thick high-density R-32 rated insulation panels to cover the walls and ceiling, a standard 36" x 78" door, two outdoor 8 hp UL approved condensing units for redundancy (with a 5-year warranty), two low profile evaporator coils, a temperature display with alarm capability, and associated components to complete the installation of the system. The total estimate of the equipment for this vault cooling system is \$99,000 delivered not including installation. Detailed cost estimate information received from American Walk In Coolers is in Appendix A.

A cost estimate for installation of the AWIC cooling system was created with the assistance of PNNL construction project management and is based on installing in a facility with protocols similar to the Radiological Processing Laboratory (RPL) facility at PNNL which is a Category II non-reactor research facility. This estimate was for installation during construction of the hypothetical facility with the cold vault implemented into the original construction plans. The estimate includes craft labor (electrician, carpenter, etc.), supplies not provided by AWIC, and overhead costs. The total installation cost was estimated to be \$110,000. A more detailed cost breakout for the cost of installation can be found in Figure 4 and specific details related to the estimate are attached in Appendix A.

| Description | Amount | Totals | Hours | Rate | Cost Basis | Cost per Unit | Percent of Total | |
|---|--------|---------|-------------|-----------|------------|---------------|------------------|---------|
| Labor | 28,456 | | 481.745 hrs | | | | 25.82% | |
| Material | 10,491 | | | | | | 9.52% | |
| Subcontract | | | | | | | | |
| Equipment | 2,809 | | 137.486 hrs | | | | 2.55% | |
| Other | 2,079 | | | | | | 1.89% | |
| Construction Direct | 43,835 | 43,835 | | | | | 39.78% | 39.78% |
| Conceptual Stage Scope Allowance | 13,150 | | | 30.000 % | т | | 11.93% | |
| Final Clean-Up Allowance | 370 | | | 0.650 % | T | | 0.34% | |
| Small Tools Allowance @ 3% of Labor | 854 | | | 3.000 % | C | | 0.77% | |
| General Conditions Allowance | 14,552 | | | 25.000 % | T | | 13.21% | |
| Construction Factors & Allowances | 28,926 | 72,761 | | | | | 26.25% | 66.03% |
| | | | | | | | | |
| Construction Base | | 72,761 | | | | | | 66.03% |
| Home Office Overhead | 10 914 | | | 15,000 % | т | | 9.90% | |
| Profit/Fee | 8 367 | | | 10.000 % | Ť | | 7 59% | |
| Construction Markups | 19,281 | 92.042 | | 10.000 /0 | | | 17.50% | 83.53% |
| | | | | | | | | |
| Performance Bond-Buildings/Insurance | 1,565 | | | 1.700 % | т | | 1.42% | |
| Construction Bonding & Insurance | 1,565 | 93,607 | | | | | 1.42% | 84.95% |
| Salaa/I laa Tay @ 9.7% of Matariala 8 Equipment | 1 157 | | | 9 700 % | 0 | | 1.05% | |
| Sales/Ose Tax @ 0.7% of Materials & Equipment | 1,157 | | | 0.700 % | C T | | 1.03% | |
| WA B&O Tax @.471% | 440 | 05 040 | | 0.471 70 | 1 | | 0.41% | 00 409/ |
| Construction Taxes | 1,603 | 95,210 | | | | | 1.40% | 00.40% |
| Construction Total | | 95,210 | | | | | | 86.40% |
| Aguisition Burdon | 14,986 | | | 15.740 % | т | | 13.60% | |
| Total | , | 110 106 | | | | | | |

Estimate Totals

Figure 4. Summary of estimated cost to construct a building with 8 additional charcoal beds. Additional details can be found in the attached file in Appendix A.

The total estimated cost to implement a cold vault for charcoal beds including equipment, labor and a 30% contingency was determined to be \$272,000, see Table 5. This assumes that the system is installed as part of the original construction of the hypothetical facility. It is likely that equipment such as a pressure swing adsorption system to dry the process gas prior to entering the cooled charcoal beds will be required to prevent condensation and potential freezing of water. This estimate does not consider the addition pressure swing adsorption equipment, but this should be evaluated if implementing a cooled vault to determine if it is required. Retrofitting an existing facility will likely be difficult since access to the vault will be restricted, space within the vault will be limited, required modifications to install electrical and lines for the cooler may not be possible, and changes to an operational nuclear facility may require regulatory approvals. The costs associated with retrofitting a cooled vault in an existing facility will be much higher due to these factors and are outside the scope of this study.

| Item | Cost |
|-----------------------------------|-----------|
| Cooling and insulation equipment | \$99,000 |
| Installation of cooling equipment | \$110,000 |
| Initial total cost estimate | \$209,000 |
| | |
| 30% contingency | \$63,000 |
| Total cost estimate | \$272,000 |

Table 5. Summary of total marginal cost for cooling upgrades.

5.0 Conclusions

Two options for lowering ¹³³Xe emissions at a hypothetical MIP facility from release levels designed for regulatory limits to a significantly lower level of 5×10⁹ Bq/day ¹³³Xe to minimize the impact on nuclear explosion monitoring were considered in this study. The options investigated were increasing the number of charcoal delay beds and cooling the vault containing charcoal delay beds required to meet regulatory limits. These methods reduce the amount of radioxenon released by increasing the holdup time and allow the short-lived xenon isotopes to decay prior to being released to the environment.

The total cost estimated to double the number of charcoal beds along with building the structure to contain the beds was approximately \$7.3 million; the cost to upgrade a vault containing the charcoal beds to a cold vault capable of holding the temperature of the vault to -6°C will be approximately \$272,000 plus any additional cost if addition of a gas dryer system to cool and dry the process gas prior to entering the cooled charcoal beds is required. For these estimates, both options were assumed to be part of the original construction of the facility. While doubling the number of charcoal beds will cost more upfront, this is a passive system that will require minimal maintenance of the building, whereas, cooling of the charcoal beds will require routine monitoring of the cooling performance, regular maintenance of cooling system, ongoing costs due to power consumption, and replacement of cooler units (approximately every 15 years). Even with the extra upkeep, cooling the charcoal beds will likely be cheaper over a 20 to 30-year period.

Retrofitting an existing facility will be more difficult and significantly more expensive than incorporating either of the options into the original design of the facility. This is due to many factors which include: restricted access to the vault after the facility is active, limited space within the vault for additional cooling equipment, difficulty in adding required modifications to install electrical or lines for the cooler that were not designed into the facility, and the required regulatory approvals for modifications to the facility. If modifying an existing facility, it may be less expensive to build the extra space for additional delay beds adjacent to the facility since this will require fewer changes the existing structures and be less disruptive to ongoing medical isotope processing. Costs associated with retrofitting were not considered in this study since they will be facility specific.

There are other options not considered in this study to reduce radioxenon emissions from MIP or other industrial sources such as cold trapping or the use of alternative adsorbents to charcoal. In addition to alternate options, combinations of the options may be viable. For instance, cooling of charcoal beds could be combined with additional charcoal beds to reduce cost and size of system. In addition, if cooling the charcoal beds to -6°C is not feasible, cooling the vault to a lower temperature can still have a significant impact – for our hypothetical model there was approximately an order of magnitude reduction in ¹³³Xe for 10°C drop in temperature.

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Appendix A – Documents for cold vault purchase and installation

Cost estimate for cold vault from American Walk In Coolers and associated documents





DRAWINGS_Q19892 2402-9135R_REF. SPECS..pdf

PDF

Detailed cost estimate for installation of cold vault.



Cooler installation estimate.pdf

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