

## LA-UR-13-20725

Approved for public release; distribution is unlimited.

Title: LANSCE: Overview of Radioactive Air Emissions Monitoring (RAEM) program

Author(s): Knight, William J.

Intended for: Presentation to European Spallation Source team



**Disclaimer:**

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

# *LANSCE*

Los Alamos Neutron Science Center

**BEAM ON**

**AREA A  
RUN PERMIT READY**

**CURRENT 996uA 09:07:15**

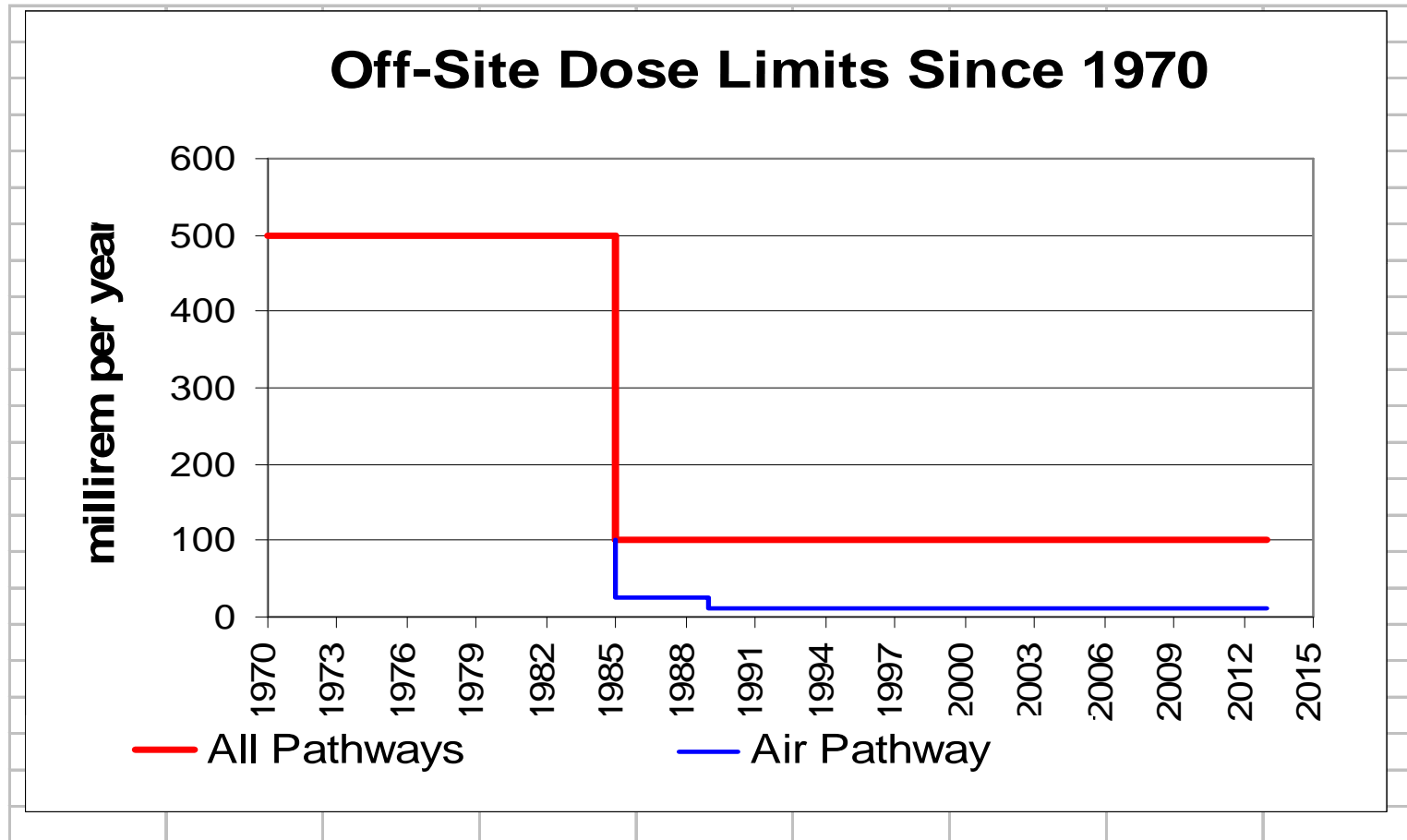
## Overview of Radioactive Air Emissions Monitoring (RAEM) program

# Presentation Outline

---

- Facility Background
- Radioactive Air Emissions
  - Stacks
  - What is monitored & how
  - Emissions controls systems
  - Diffuse emissions
  - Historical perspective

# Emissions Limits



# LANSCCE Facility

---

- 800 meter long accelerator (1/2 mile)
  - generates high-energy ion beams
  - proton (H+) and negative-ion (H-) beams
  - high energy & velocity; 84% of light speed
  - high current; 1 milliampere of current (thru 1999)
    - = 6,240,000,000,000,000 protons per second
  - only facility of this power in nation
  - neutrons generated by impact of beam w/ target

# LANSCCE Research

---

- Accelerator Production of Tritium ('97-'99)
- Science-Based Stockpile Stewardship
  - Proton Radiography
- Materials Studies
  - crystal structure
  - neutron resonance spectroscopy
- Medical Radioisotope Production
- Pure nuclear physics research

# TA-53 Emissions Stacks

---

- Exhaust Stack 2 (ES-2)
  - Lujan Center (target 1), Weapons Neutron Research facility (targets 2 & 4), Line D beam tunnel. “South Side” of TA-53
- Exhaust Stack 3 (ES-3)
  - Area A, Area A-East (A6 beam stop), Line B, Line C, Beam Switchyard, Area C (HRS)
- Main accelerator tunnel held stagnant

# ES-2 Stack

---





# ES-3 Stack

---



# Radioactive Air Emissions

---

- Formed by spallation or activation
- “Target” atoms include air (nitrogen, oxygen), water vapor, beam line components
- Three types:
  - Gaseous Mixed Activation Products
  - Particulate & Vapor Activation Products
  - Tritiated Water Vapor

## Radioactive Material - Gases

---

When proton beam runs through ambient air...

- Generated by interaction of the ion beam, or secondary particles, with air & other materials. Mostly C-11, O-15, N-13
- Spallation: impact of high-energy particle with a target atom's nucleus, releasing protons or neutrons from target atom. Can also “shatter” nucleus.
- Activation: absorption of particle into nucleus

When beam line & targets are maintained under vacuum...

- Radiolysis: separation of cooling water into component parts under high incident radiation; subsequent activation/spallation. Results in mostly C-11 & O-15.

# Radioactive Material – Particulates & Vapors

---

## Particulate material

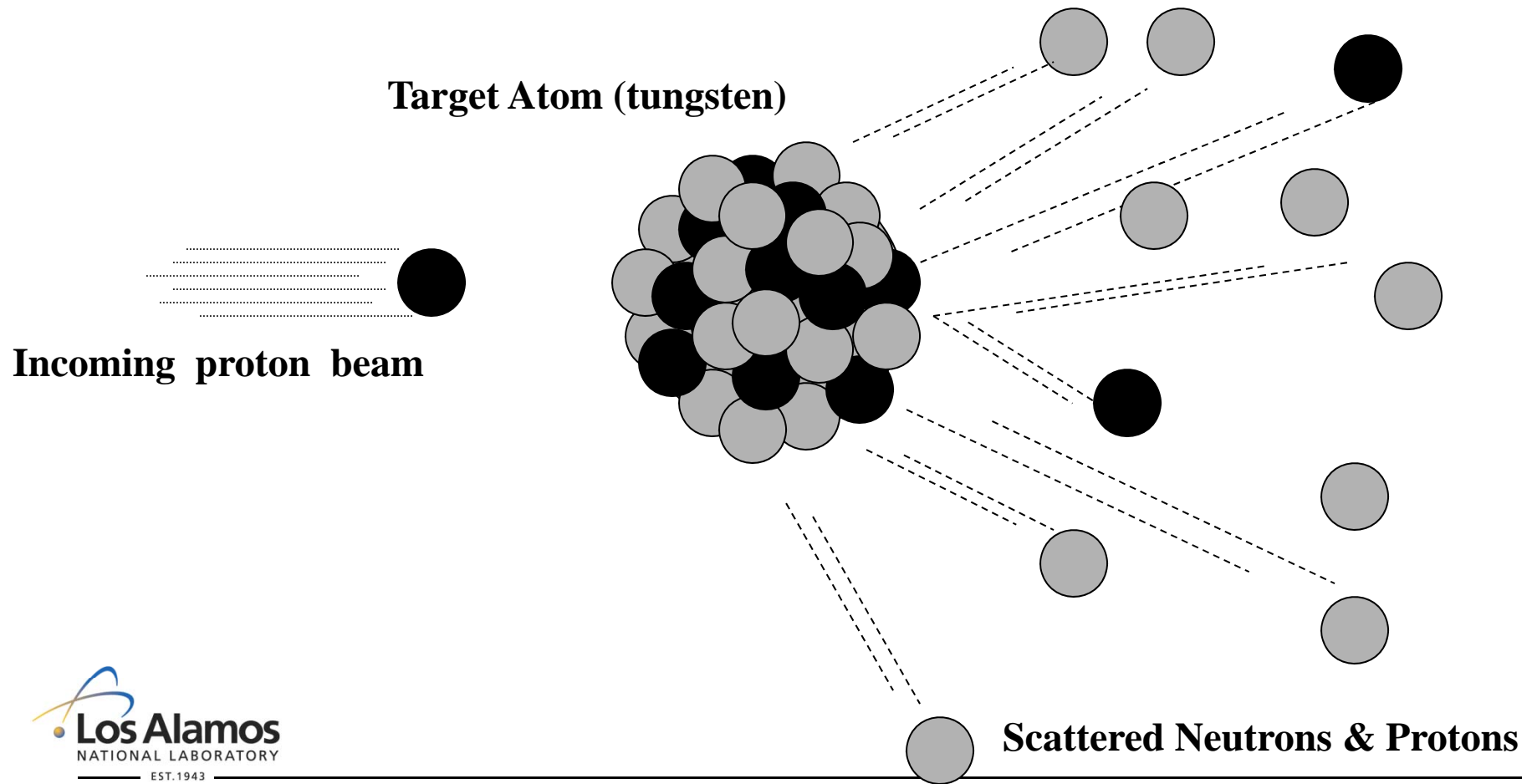
- Activation/spallation of beam line components or shielding  
Mn-54; Co-60; Na-22, Na-24
- Beryllium – cosmogenic nuclide, detections are usually rejected.  
But beryllium moderators; cannot assume naturally occurring.
- HEPA filtration on stack, particulates not seen in abundance

## Vapors / Volatiles

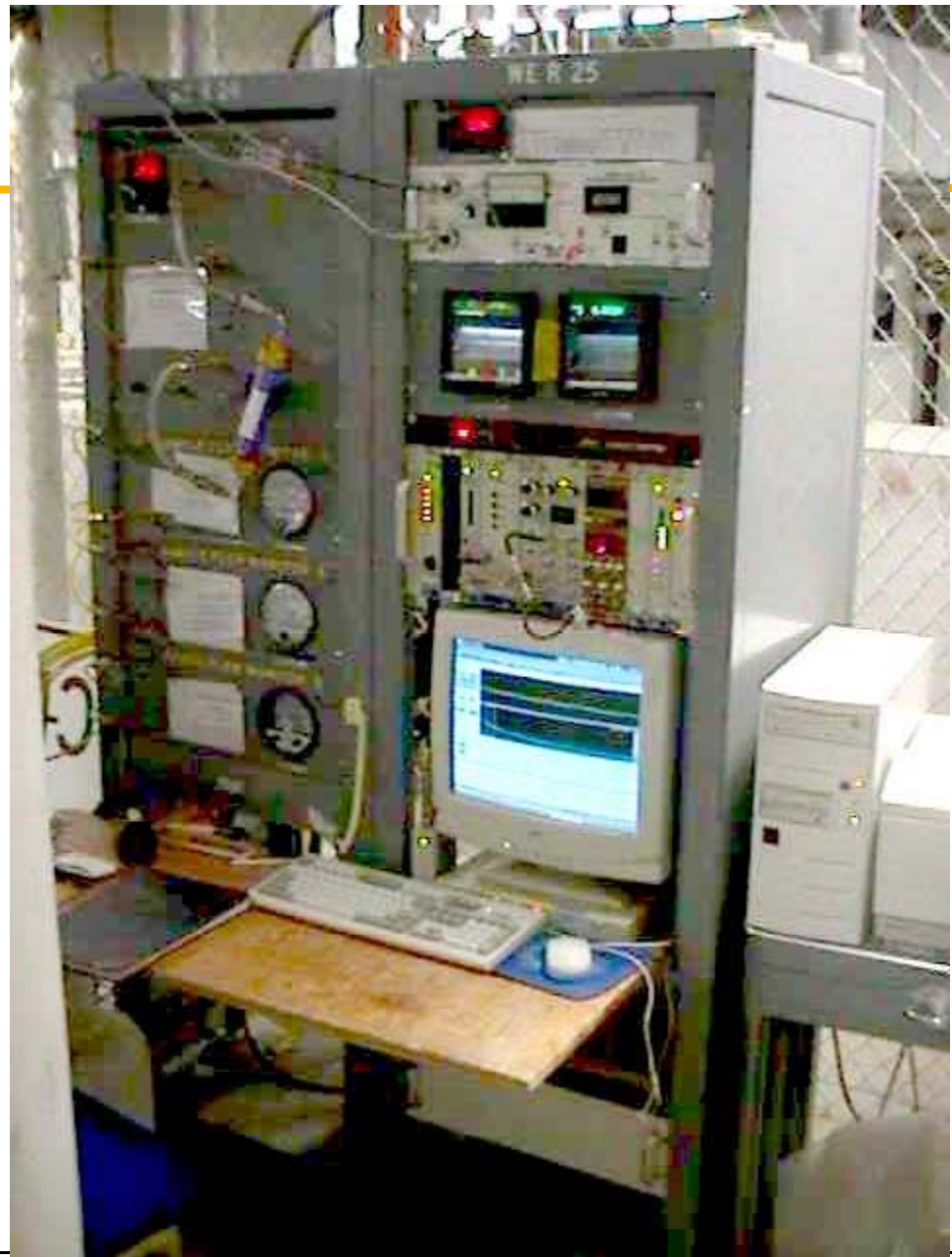
- Mercury isotopes – activation/spallation of shutter system.
- Bromine isotopes – naturally liquid/vapor
- Selenium & germanium – fluorine compounds, volatile

# Spallation Example

proton beam incident on heavy target atom



# Stack Sampling Station ES-2



# Particulate & Vapor Activation Products (PVAP)

---

- Typically, spallation or activation of metal, concrete, etc.
- Airborne suspension of particles
- Particles subject to HEPA filtration; volatiles/vapors pass through
- Samples taken with paper filters (particulates) and charcoal filters (vapors)
- Exchanged weekly

## Issues

- Short-lived nuclides; ship off-site for analysis, 36 hours after removal.
- Decay correct results; assume generation scales with current.
- Changing conditions can yield different emissions species

# PVAP sample assemblies

---





# PVAP sampler in-line

---



# Tritiated Water Vapor (HTO)

---

- Activation of water vapor in the air
- Hydrogen + 2 neutrons = tritium
- Radioactive H<sub>2</sub>O referred to as HTO
- Water collected on indicating silica gel
- Samples exchanged monthly or upon saturation
- LANSCE - NOT required to monitor HTO
- Sampling operations ceased in 2001; use historical ratios to determine emissions

# HTO sample assembly



# Gaseous Mixed Activation Products (GMAP)

---

- Spallation & activation of air
- Makes up >90% of rad air emissions
- Primary Source: “air gap” at A6 Beam Stop (thru 1999)
- Primary source since 2000 – cooling water system at 1L target.
- Gaseous; NOT affected by filters
- Primary Constituents:
  - C-10, C-11, N-13, N-16, O-14, O-15, Ar-41
- All short-lived radionuclides
- Can cause off-site dose consequence if public receptors are nearby (1 km!)
- Water leaks, loss of vacuum can increase emissions (more “target” atoms)

# A6 Beam Stop

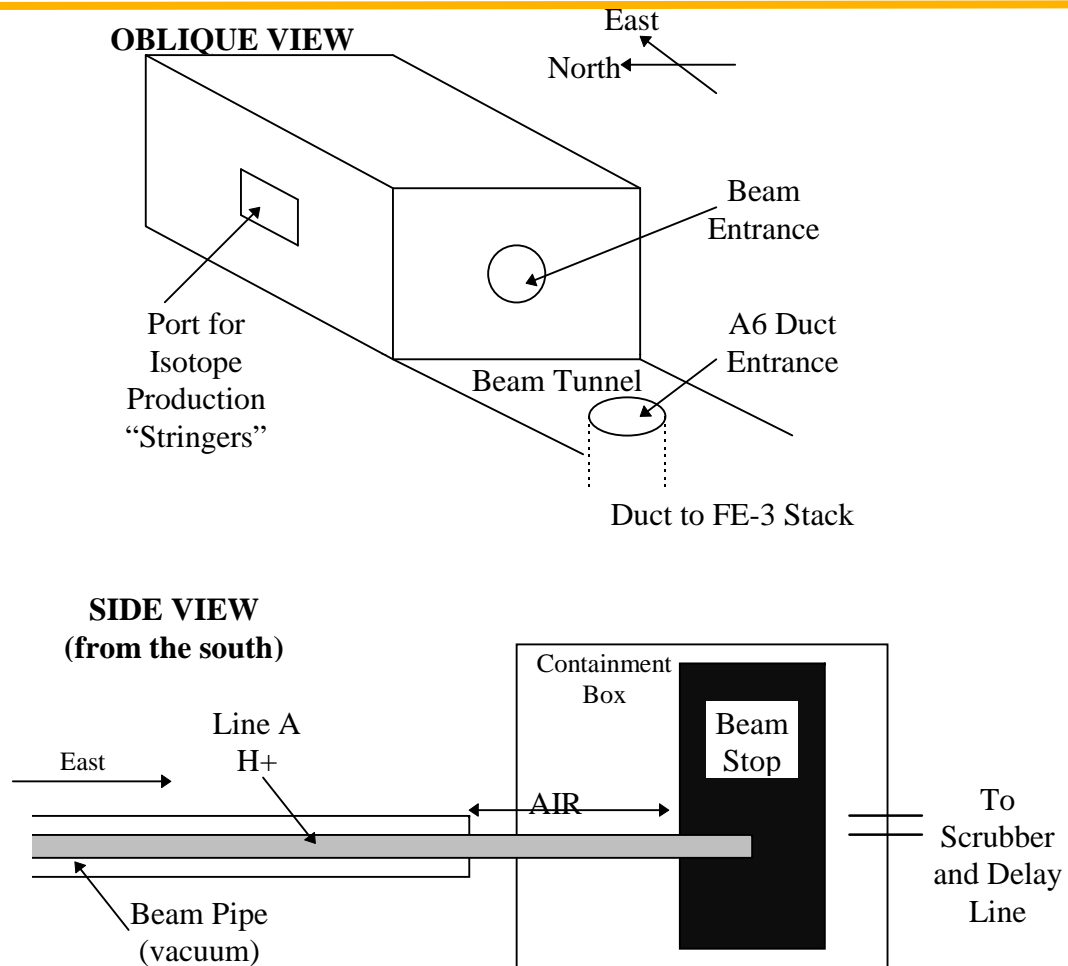


Figure 3: Block Diagrams of Containment Box

# GMAP Monitoring

---

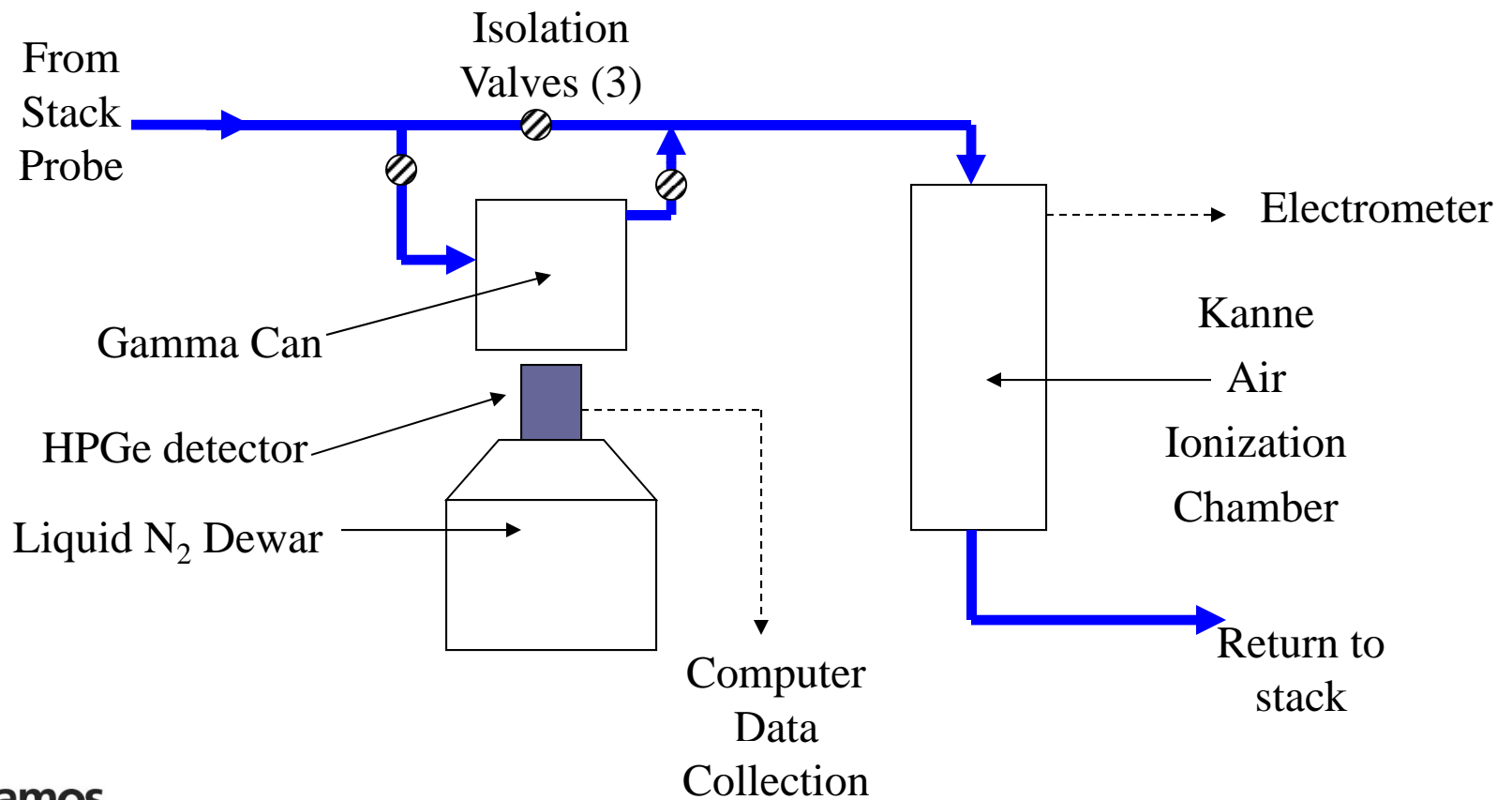
- Two detector systems in series
- Flow-through ionization chamber
  - Kanne air ionization chamber chamber
  - 300 volt bias
  - Measures “gross emissions,” total magnitude
  - Continuous operations – minimal down time due to Excel-based acquisition files; <5 mins/week
  - Total charge collected (picocoulombs) relates to magnitude of emissions (curies)
  - Tracks integrated current (charge) over time

# Gas Composition Measurements

---

- High-Purity Germanium detector (HPGe)
  - “quasi-continuous”; 3-7 day acquisitions
  - measures relative composition of stack air
  - Isolate system for “decay curves”  
Differentiate half-lives of pure 511 keV emitting radionuclides
  - All nuclides related to 511 keV line
  - Ratio, 511 keV peak area in HPGe to pC collected on ion chamber during acquisition.
  - Emission calculation uses total charge collected, 511 keV peak area per pC collected ratio, stack flow, ion chamber volume to determine curies of positron-emitting radionuclides emitted.
  - Other radionuclides calculated via ratio to 511 keV nuclides.

# GMAP Detector Schematic





# Controlling GMAP Emissions

---

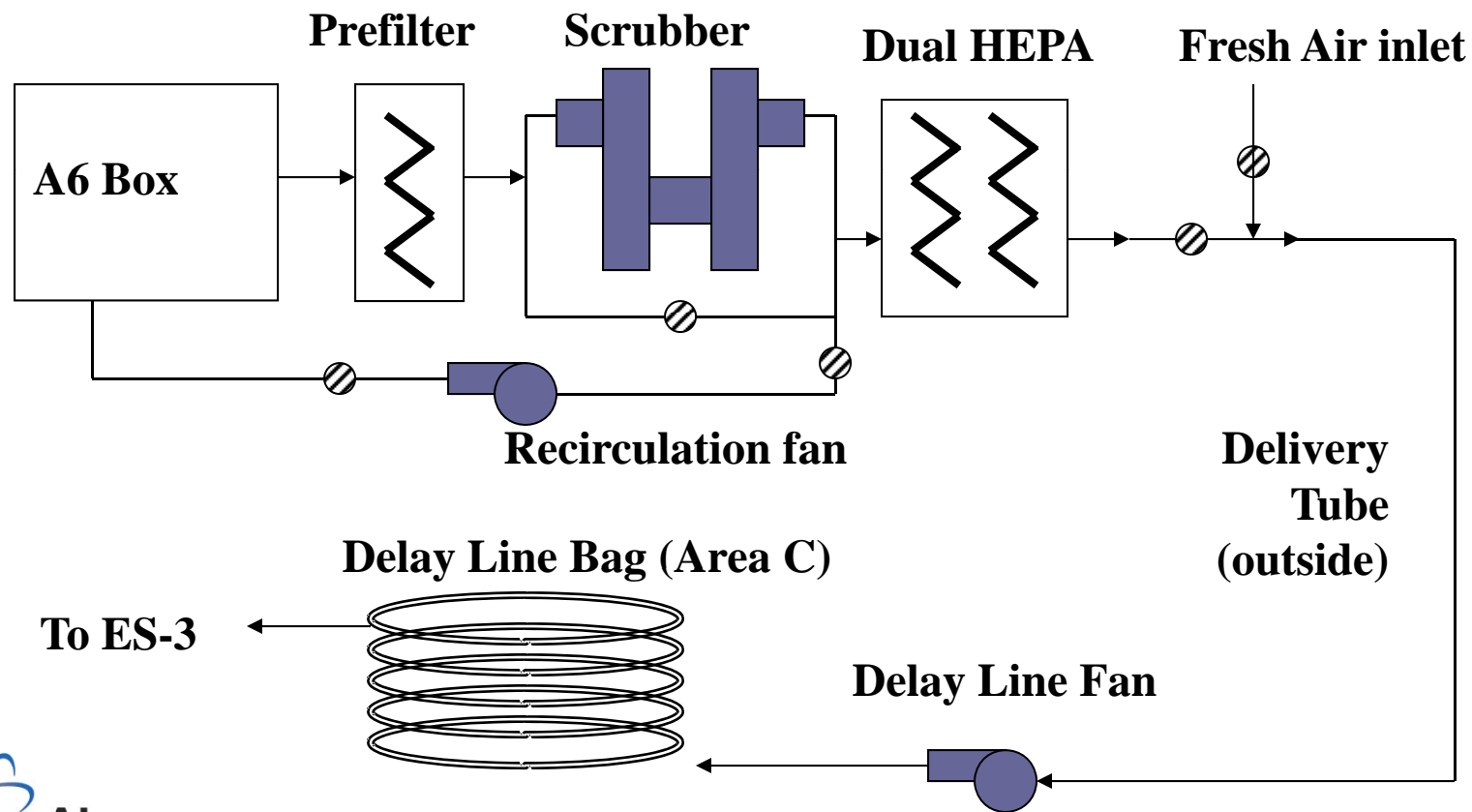
- Short half-lives: nuclides subject to rapid decay, if containment is possible
- A6 Delay Line system: 1994-1998
  - Pulls low flow rate (~100 cfm) from A6 containment box
  - 4000 feet duct, 2' diameter
  - > 100 minutes delay before ventilation out ES-3
  - Reduces A6 emissions by over 80%
- 1L Delay Line System: 2004-2005
  - Very low flow - < 1 cfm
  - Uses pressure buildup in cooling water loop as driving force
  - Pressure relief system vents into large-diameter copper tubing
  - Migrates through tubing, discharge to stack

## More GMAP controls – A6 system

---

- Filters in Delay Line system (Line A, 1000 microamp beam)
  - shielded, disposable prefilter – removes major contamination from system
  - dual certified HEPA filter system – removes all remaining particulates
- Wet air scrubber system
  - two packed-bed towers
  - removes excess water (tritium), acid, carbon dioxide, vapors, particulates

# Delay Line Schematic – 1990's A6 Beam Stop



# Delay Line Bag (1998)

---



# Delay Line 1998: Top View

---



# Delay Line: Inlet & Outlet

---



# Scrubber Schematic

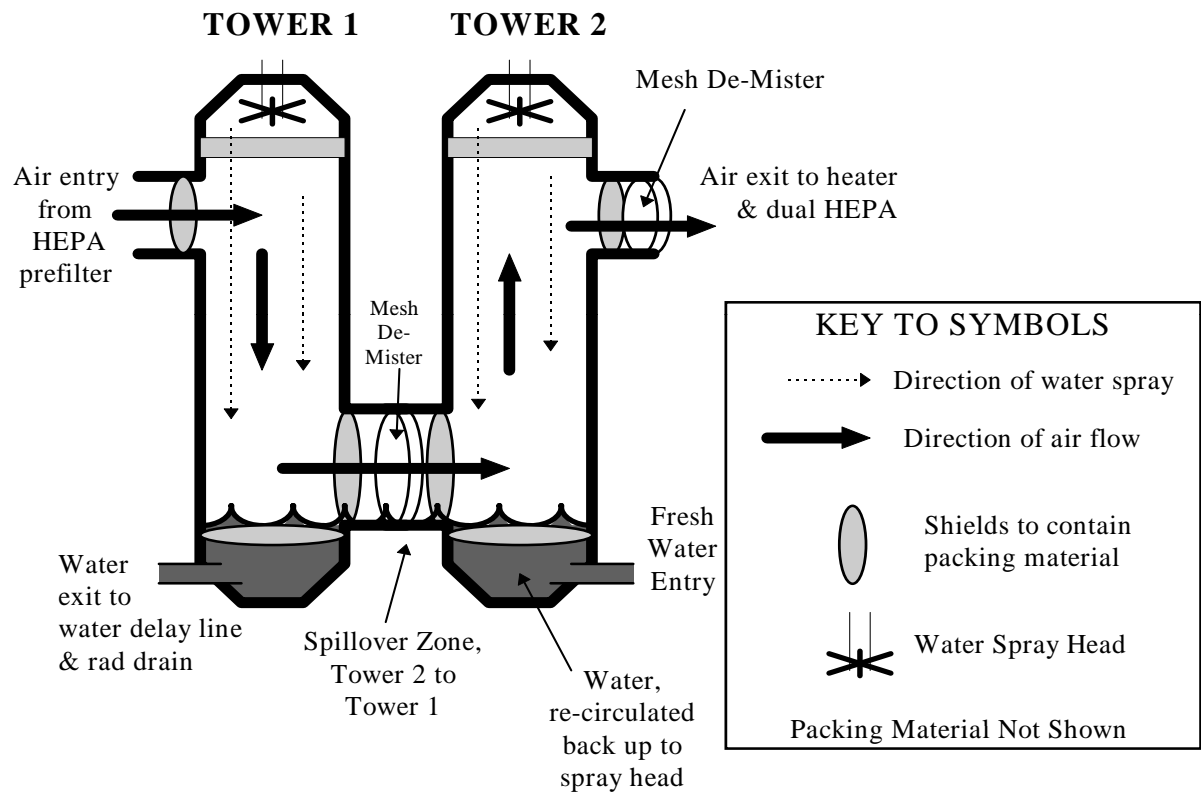


Figure 6: Diagram of Scrubber

## Issues with Emissions Controls

---

A6 Beam Stop – high-intensity beam, air gap

- Air (nitrogen) activation leads to high-acid environment; damages diagnostic equipment.
- Scrubber water becomes acidic; control pH prior to discharge
- Direct radiation source from activated air in system
- “Transient” state nuclides bypass filters; Ne-24 (gas) to Na-24 contamination downstream

1L Target – 120 microamp beam, under vacuum

- No air activation, but radiolysis of water is major source
- Pressure buildup in cooling system
- Cracked valve at delay system entrance in 2005 – 300 Ci/day, 6 mrem off-site dose



# Diffuse Emissions

---

- Radioactive air migrates from beam line area into buildings & rooms along Line A
- Measure concentration of radioactive air in room
- Measure, estimate outflow of air
- GMAP only, conservative composition
- Minimized by delay line & sealing efforts

# Diffuse Emissions Monitor

50 Liter Kanne chamber

---



# Diffuse Emissions Sites

---

- Area A-East
- Isotope Production building
- Beam Switchyard
- Area A (during A1 or A2 operations)
- 1L Service Area
- Other areas monitored for potential emissions, personnel protection, and diagnostic purposes

## Issues

- Changing equipment & operations leads to different monitoring needs
- Generation rate can be subject to beam performance (spill)
- Emission rate can be subject to meteorological conditions

# Summary

---

- Emissions are dominated by gaseous species – difficult to control
- Types & quantity of emissions can vary with facility conditions
- Changing regulations can increase importance of emissions management
- Different experiments can drive different emissions monitoring needs

- Thanks for your time

David Fuehne  
davef@lanl.gov  
(505) 665-3850

Bill Knight  
knight@lanl.gov  
(505) 665-4503