Evaluation and Measurements of Radioactive Air Emission and Off-Site Doses at SLAC

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Abstract:

SLAC, a high-energy (GeV) electron accelerator facility, performs experimental and theoretical research using high-energy electron and/or positron beams that can produce secondary neutron and gamma radiation when beam losses occur. Radioactive air production (mainly $^{11}$C, $^{13}$N, $^{15}$O, $^{41}$Ar) and release is one of the environmental protection program issues. DOE Order 458.1 requires that 40 CFR 61 Subpart H’s NESHAPs requirements be followed. These regulations prescribe a total dose limit of 10 mrem y$^{-1}$ to the Maximally Exposed Individual (MEI) of general public, a requirement for a continuous air monitoring system if a release point within a facility can cause > 0.1 mrem y$^{-1}$ to the MEI, and a requirement for periodic confirmatory measurements for minor sources which give releases that contribute ≤ 0.1 mrem y$^{-1}$ to the MEI. At SLAC, all air release points for current operations are evaluated to be minor sources. This paper describes SLAC’s evaluation following NESHAPs requirements and measurements using the Air Monitoring Station (AMS) as periodic confirmatory measurements.

Keywords: NESHAPS; Maximally Exposed Individual; Electron Accelerator; Air Monitoring; Radioactive Air; $^{11}$C; $^{13}$N; $^{15}$O; $^{41}$Ar

Introduction:

Radioactive material is inevitably produced by the operations of high-energy (GeV) particle accelerators near high normal beam loss points. Along the beamlines, some beam particles strike certain accelerator components and produce induced radioactivity in them. Secondary radiation in the form of high-energy photons or neutrons can also interact with materials. When high-energy photons or neutrons interact with the nuclei present in air molecules, radionuclides such as $^{11}$C (half life of 20 minutes), $^{13}$N (10 minutes), $^{15}$O (2 minutes), and $^{41}$Ar (110 minutes), may be produced [1]. This paper describes SLAC’s evaluation following NESHAPs requirements and measurements using the Air Monitoring Station (AMS) as periodic confirmatory measurements.
Evaluation:

SLAC uses CAP88-PC Version 2 [2] to calculate potential effective dose equivalent (EDE) to MEIs and to the population from the estimated airborne radioactivity released by SLAC.

The potential doses to the MEI and public population are calculated as follows:

1) Saturation activity is calculated using a conservative beam loss on an optimum target, e.g., from the yield values (Ci/W per meter of air path) in the IAEA Report 188 [1] and a reasonable air path for each release point of a facility. SLAC has more than 10 facilities and each facility may have more than one release point. In some cases, Monte Carlo calculations (e.g., using FLUKA, MARS or MCNPX codes) by simulating actual beam-target-housing geometry can be performed to obtain more accurate production.

2) Accelerator operational scenarios (length of operation per year and annual beam losses) and reasonable air exchange rates for accelerator housing are used to estimate the total radioactivity produced and release per year.

3) For each of the release points, the number of times during the year that potentially activated air associated with the release point is vented to the atmosphere is determined. If the air volume is essentially sealed from the atmosphere during normal operations, then the number of releases is conservatively assumed to be 1 air change per day to take into account any building ventilation leakages. The total volume of contained air is assumed to be released per each air exchange. In those cases where the potentially activated air isn’t sealed from the atmosphere, the number of releases is calculated based on the rate of air changes (from actual measurements or design specifications) per unit time for the volume of interest.

4) The location of MEI (where off-site public currently resides) that is relevant to each air release point within SLAC is then determined. There are a total of more than 10 MEI locations.

5) Based on the annual airborne radioactivity release, the doses to the MEI and the collective doses to the population up to 80-km from SLAC are calculated using the EPA-approved atmospheric dispersion and radiation dose calculation computer code, CAP88. The CAP88 modeling program calculates the average off-site radiation dose to individuals at specified distances and directions from the SLAC facilities, and collectively to individuals within each population segment around the SLAC facilities.

6) The Bay Area meteorological data provided for San Francisco Airport (SFO) and the CY2000 census population distribution are used in the calculations, while default agricultural values are used. CAP88 also requires information on stack height and diameter. SLAC conservatively specifies that all releases of airborne radioactivity at SLAC take place from ground-level points, i.e., from stacks with heights and diameters of 0 meters.

Finally, the maximum MEI dose and the collective dose to public population are then reported in the annual NESHAPs. In CY2011, the resulting dose to the MEI was calculated to be
1.04 x 10^{-3} mrem (1.04 x 10^{-5} mSv). This is well below the regulatory limit of 10 mrem (0.1 mSv) in any one year. In addition, there is no individual release point exceeding the 0.1 mrem y^{-1} (0.001 mSv y^{-1}) limit for the continuous monitoring requirement (the maximum value was 1.03 x 10^{-3} mrem y^{-1} from the Positron Vault release point during FACET operations). The collective effective dose equivalent to the population within 80 km of SLAC’s site boundary (estimated 5 x 10^6 persons) due to releases of airborne radioactivity at SLAC in CY11 was calculated to be 5.4 person-mrem.

**Confirmatory Measurements:**

Eight AMSs have been used to monitor the amount of airborne radioactivity generated at key SLAC facilities when needed. Confirmatory measurements can also be verified with grab air sample using plastic bag of about 30 liters capacity and count the balloon with High Purity Germanium Detector gamma spectrometry.

The locations of the AMS are where activation of the air is more likely, i.e., beam losses are higher. For example, AMS-20 is located above the Positron Vault (PV), a high normal beam loss location, at the 2-mile-long LINAC Sector 20. The AMS description, measurement method, and results of Positron Vault are presented below.

**AMS**

Each AMS consists of a large aluminum box mounted on a concrete slab (see Fig 1) on top of accelerator housing soil berm. Housed within the aluminum box is a 10.45”- long Geiger-Müller (GM) tube (Model TGM N107) gamma detector inside a MG-2 Gas Sampler shielded by 2”-thick lead to reduce ambient radiation background. The air pump pushes air through the Gas Sampler and the in-line particulate air filter. The air pump has an air flow gauge. The air pump operating conditions have to be tested annually when the system is in use and at least every two years otherwise. GM signals are sent to a CAMAC (Computer Automated Measurement and Control) crates in the Main Control Center (MCC).
Figure 1. Top view of the AMS. Shielded gas sampler with GM tube detector inside. Air flow meter, particulate filter air head, 12-V DC power supply, HV and amplifier unit, manometer with field readout, intake and exhaust Tygon tubing, electronic manometer with pressure data output to MCC and Eberline air pump are all shown.

AMS measurement process:

Confirmatory measurements using AMS is a four-step process, as described below (also see Table 1)

Step 1: $^{137}$Cs free-in-air calibration for the AMS GM tube

The first step is to perform $^{137}$Cs free-in-air calibration for the AMS GM tube. This allows GM signal in cpm versus the open-field exposure rate in mR h$^{-1}$ to be obtained.

GM detector was removed from the AMS gas sampler tank and positioned on a stand. NIST 3.79 mCi $^{137}$Cs source was positioned at 1 m distance above the GM detector to avoid scattering contribution from the surrounding materials.

Exposure rate at the GM tube location was calculated using the $^{137}$Cs gamma ray constant of 0.33 mR hr$^{-1}$ at 1 m per 1-mCi, and confirmed with reference ion chamber measurements.

The GM response factor is 5180 cpm per 1 mR hr$^{-1}$ for a free-in-air exposure with $^{137}$Cs.

Step 2: Radioactive air decay curve measurements

The second step is to perform radioactive air decay curve measurements using the AMS by fitting the theoretical decay curves of $^{11}$C, $^{13}$N, $^{15}$O, $^{41}$Ar to the measured, composited decay
curve. This allows the determination of the signal (in cpm) for each isotope inside the gas sampler.

Before the air measurements, the average beam power has to be recorded, i.e., electrons energy, number of pulses per seconds (Hz), and the average beam current. Pump air flow rate was set at 30 liters per hour (lph), which is low enough to insure minimal pressure difference between gas sampler and pressure in PV. In addition, because of the low air flow and the large air volume in the PV (9740 ft$^3$, 350 m$^3$), the condition of equilibrium concentration between gas sampler and PV is reached.

Background signal determined with no radioactive air inside gas sampler needs to be subtracted first. The background value with AMS-20 pump off and GM detector inside of the gas sampler was 0.45 ± 0.03 cps. The measurements started with pumping activated air in the PV through the AMS-20 gas sampler for at least 20-min to allow equilibrium signal to be reached and then both the gas inlet and outlet were closed (this is time zero for the decay curve measurements). Radioactive air decay curve was then measured with 1-minute increments up to 25 minutes and then 6-minute increments up to 3 hours.

The measured decay data were plotted and fitted with each isotope’s decay curve (determined by the isotope’s theoretical half-life). The 6-min data with decay time beyond 2-h were used to determine the amount of $^{41}$Ar (which has the longest half-life) with the assumption of other 3 isotopes, if any, already fully decayed. The signal of $^{41}$Ar at time zero was derived as the average of the extrapolated values from each data point beyond 2-h decay time.

The next step is to subtract the $^{41}$Ar signal from the total signal. The 6-min data is too coarse for the fitting of short half-lived isotopes. To determine contributions of $^{11}$C, $^{13}$N and $^{15}$O, the scaler data with 1-min intervals recorded for 25 minutes were used.

The beam powers during the 6-min and 1-min measurements were different. Therefore, a beam power ratio of 1.58 was used to normalize the data between the 6-min and 1-min measurements. The results were normalized for the beam power of 1.09 kW.

Each radioisotope contribution was determined by fitting a theoretical curve into experimental decay graph. Figure 2 shows the PV’s measured decay curves of $^{41}$Ar-subtracted signal. Both 6-min and 1-min data are presented, but 1-min data were used to derive the $^{13}$N signal.

Based on decomposition of measured decay curve, the signal of $^{41}$Ar, $^{13}$N, $^{15}$O at time zero were derived as 20 cpm, 160 cpm, and 213 cpm, respectively. (see Fig. 2).
Figure 2: The Positron Vault’s measured decay curves of $^{41}{\text{Ar}}$-subtracted signal (both 6-min and 1-min data are presented, but 1-min data was used to derive the $^{13}{\text{N}}$ signal). Theoretical $^{13}{\text{N}}$ and $^{15}{\text{O}}$ decay lines with a half-life of 10-min and 2-min, respectively, fit well with the signal curves, while 20-min decay of $^{11}{\text{C}}$ cannot be seen. The beam power is at 1.09 kW.

**Step 3: Microshield modeling of the AMS gas sampler**

The third step is to perform modeling of the AMS gas sampler with Microshield code to determine the relationship between each isotope concentrations ($\mu$Ci ml$^{-1}$ in the gas sampler) and its dose rate contribution to the GM tube response (mR h$^{-1}$).

Microshield-5® model of the gas sampler with the GM detector was created and then used to determine each nuclide’s contribution to the signal of GM tube. Microshield modeling calculations were performed for 1-Ci activity for each nuclide in the 11-liter sampler. The exposure rates at 5 dose points along the GM detector longitudinal axis were calculated and the average exposure rate result was used.

The average of the five dose points along the GM tube was $7.5 \times 10^4$ mR h$^{-1}$ and the dose-concentration conversion factor was 1.27 $\mu$Ci L$^{-1}$ per mR hr$^{-1}$ for $^{41}\text{Ar}$. The positron-emitting
isotopes of $^{11}$C, $^{13}$N, and $^{15}$O have the same conversion factor (1.42 $\mu$Ci L$^{-1}$ per mR hr$^{-1}$) as they emit the same energy of 511 keV photons.

The activity concentration inside AMS gas sampler is calculated by dividing the count rates for each isotope at time zero by the $^{137}$Cs response and then multiplying the MicroShield-calculated dose factor. The saturation activity concentration inside AMS gas sampler (almost the same as that of PV) at zero decay time was for 0.040 $\mu$Ci L$^{-1}$ for $^{13}$N, 0.054 $\mu$Ci L$^{-1}$ for $^{15}$O, and 0.005 $\mu$Ci L$^{-1}$ for $^{41}$Ar and zero for $^{11}$C at a beam power of 1 kW. The results are shown in column 5 of Table 1.

**Step 4: PV Saturation Activity Calculations**

The last step is to relate the air radioactivity concentration in gas sampler to that in PV.

The AMS pressure was measured to be 710 mmHg. The concentration in AMS is converted to that in PV by simply correcting for their air pressure difference (concentration is linearly proportional to air pressure). Multiplying the estimated PV air volume (350,000 liters) with PV activity concentration, the saturation activity at 1 kW was derived and shown in Table 1.

Table 1: Summary of results and measured saturation activities and comparison with International Atomic Energy Agency (IAEA) references values.

<table>
<thead>
<tr>
<th></th>
<th>Step 1: GM Calibration with $^{137}$Cs (cpm per mR h$^{-1}$)</th>
<th>Step 2: GM Signal (cpm) at 1.09 kW</th>
<th>Step 3: Microshield Modeling ($\mu$Ci L$^{-1}$ per mR hr$^{-1}$)</th>
<th>Step 4a: AMS Activity Concentration at 1-kW ($\mu$Ci L$^{-1}$)</th>
<th>Step 4b: PV Activity Concentration at 1-kW ($\mu$Ci L$^{-1}$)</th>
<th>Experiment Saturation Activity in PV at 1-kW (mCi)</th>
<th>IAEA Saturation Activity (mCi)</th>
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<tr>
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<tr>
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<td>0.043</td>
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<td>213</td>
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<td>0.054</td>
<td>0.057</td>
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<td>74</td>
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<tr>
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<tr>
<td>PV</td>
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**Discussions and Conclusions:**

Theoretical $^{13}$N and $^{15}$O decay lines with a half-life of 10-min and 2-min, respectively, fit well (see the parallel line between fit lines and the measured data in Fig. 2), while 20-min decay of $^{11}$C cannot be seen in the plot. Absence of $^{11}$C is expected as it has a very small parent’s $^{12}$C abundance in the air and a very small activation reaction cross section.
Since the AMS is located right above the vault and connected directly to the vent, there is no decay in the air path between PV and AMS gas sampler. Because of the air volume in PV is much greater than air pumped into AMS, there was no activity “depletion” in the vault. Also, it was assumed that after an hour of beam hitting the positron target, saturated equilibrium is reached.

Confirmatory measurement values were about a factor of 3 lower than the conservative, calculated values in SLAC NESHAPs reports [3], except $^{41}$Ar (which is thermal neutron-induced isotope and whose production strongly depends on the room size and shape and had a factor of 2 higher), which were calculated based on optimum yield values from the IAEA report [1]. Both measurements and calculations show that the $^{15}$O has the highest yield in PV (20 mCi at 1-kW, GeV hitting a copper target inside PV from measurements).

**References:**


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