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PNNL-30202

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U.S. DEPARTMENT OF ENERGY

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Abstract

In June 2018 radioactive (^{37}Ar and ^{127}Xe) and stable (SF_6) tracers were injected into the chimney of the Disko Elm underground nuclear explosion, located in the U-12p tunnel complex. After the injection sampling was performed from locations within the tunnel, a borehole drilled from the surface, and from the surface of the mesa. Additionally, the ^{127}Xe activity within the chimney was continuously monitored for two months after the injection. In contrast to previous gas migration experiments, none of the injected radiotracers were detected in the monitoring borehole, indicating that the specific geology of Aqueduct Mesa limits the impact of natural transport of these gases in the subsurface. However, elevated levels of ^{39}Ar (an activation product produced by underground nuclear explosions) were detected and quantified in each sample. This observation has significant implications for nuclear explosion monitoring since ^{39}Ar is a long-lived radioisotope which can be expected to be present in significantly elevated levels for many years after an underground nuclear explosion.

Summary

In an effort to better understand the transport of radioactive gases after an underground nuclear explosion (UNE), several gas migration experiments have been conducted at the Nevada National Security Site (NNSS). The most recent experiment was conducted at the site of the Disko Elm historic UNE, located in the U12p tunnel complex. This experiment, part of the Underground Nuclear Explosion Signatures Experiment (UNESE), involved drilling back into the Disko Elm chimney and emplacing a set of recirculating gas tubes and a tube for the injection of tracer gases. As part of the experiment, a borehole was also drilled from the surface of Aqueduct Mesa (the mesa containing the U-12p tunnel complex) and completed with four recirculating gas sampling zones. This document reports the details of the UNESE Phase II noble gas migration experiment to support future analysis of the results. The goal is to provide relevant information for the development of simulations which use this experiment as a test case for models which aim to improve understanding of containment of UNEs and the fate of nuclear debris after a UNE.

In June 2018, three tracer gases were injected into the Disko Elm chimney. Two of the tracers were radioactive gases, ^{37}Ar ($t_{1/2} = 35.04$ days) and ^{127}Xe ($t_{1/2} = 36.4$ days), while the third tracer, sulfur hexafluoride (SF_6) is stable. The total tracer injections equaled 0.65±0.15 Ci of ^{37}Ar,0.56±0.14 Ci of ^{127}Xe, and 51 kg of SF_6. An effort was made to minimize the amount of carrier gas injected with the tracers, with a total of 43.3 m^3 of make-up air injected along with the tracers.

After the injection the transport of the tracers was measured by collection of discrete samples and by real-time radiation detection. The concentration of ^{127}Xe in the chimney was monitored for two months using a radiation detector looking at a continuously recirculating flow of gas from the chimney. A second real-time monitor was located farther down the tunnel looking at gas from the historic re-entry tunnel. Discrete 1 L samples were also collected from those two locations along with a third location near the tunnel ventilation intake. The use of real-time detectors was a significant improvement on previous gas migration experiments which relied solely on point measurements to observe the migration of tracers.

At the surface of Aqueduct Mesa, both large (2 m^3) and small (1 L) volume samples were collected from the four sampling zones in the surface borehole. Large volume samples of air were also collected just above the ground surface. The large volume samples were analyzed for the injected radiotracers as well as for ^{39}Ar, an activation product produced by the historic UNE. The small samples were analyzed for the injected stable tracer SF_6 as well as for Freon 13B1, which was injected previously as part of a suitability study of the site.

The injected radiotracers were never observed in the samples collected from the surface borehole, which stands in contrast to previous gas migration experiments conducted at a different location on the NNSS. This indicates that, despite similarities in the overall geologies between the two sites, natural transport drivers do not act as quickly in the specific geology of Aqueduct Mesa and that any gas transport would be completely dominated by the initial pressure and temperature increases produced in UNE. Measurements of the stable tracers at part-per-trillion (ppt) levels did detect the presence of both injected stable tracers in the borehole, however, the analysis is complicated by the drilling and completion of the sampling borehole immediately prior to the injection.

While the injected radiotracers were never observed in the samples collected from the surface borehole, elevated ^{39}Ar levels were measured in every sample collected around P-tunnel and Aqueduct Mesa. This has significant implications for nuclear explosion monitoring. Elevated levels of ^{39}Ar will negatively impact ^{37}Ar detection limits, and with a half life of 269 years ^{39}Ar can be expected to be present in significantly elevated levels for many years post UNE.

Acknowledgments

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Acronyms and Abbreviations

GC-MS	Gas chromatograph – mass spectrometer
ID	Inner diameter
MDA	Minimum detectable activity
MDC	Minimum detectable concentration
NGME	Noble Gas Migration Experiment
NNSS	Nevada National Security Site
NPE	Non-Proliferation Experiment
OD	Outer diameter
PNNL	Pacific Northwest National Laboratory
PPB	Part-per-billion
PPT	Part-per-trillion
SCUBA	Self-contained underwater breathing apparatus
SGZ	Surface ground zero
UNE	Underground nuclear explosion
UNESE	Underground Nuclear Explosion Signatures Experiment

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1.0 Introduction

The Underground Nuclear Explosion Signatures Experiment (UNESE) was a multi-year research and development project created to apply a broad range of research and development (R&D) techniques and technologies to nuclear explosion monitoring and nuclear nonproliferation. As part of UNESE, two noble gas migration experiments were conducted at the Nevada National Security Site (NNSS) to simulate the transport of radioactive noble gases that would be created during an Underground Nuclear Explosion (UNE). The focus of these experiments was to study the transport of gases through a UNE produced fracture network using only natural transport mechanisms. The purpose of this document is to document the second of these experiments.

UNESE was broken into two phases. Phase I focused on activities surrounding the site of the Barnwell underground nuclear explosion, which occurred at the U20az borehole on the NNSS. This was a vertically emplaced historic nuclear test. Phase II focused on activities at the U-12p tunnel complex at the NNSS, also known as P-tunnel, and specifically on the site of the Disko Elm UNE. This historic nuclear test was horizontally emplaced.

This report focuses on the noble gas migration experiment conducted in P-tunnel as part of UNESE Phase II. In this experiment, radioactive (^{37}Ar and ^{127}Xe) and stable (SF_6) gas tracers were injected into the Disko Elm chimney and allowed to transport naturally. Samples were collected from various locations in the tunnel and on the mesa surface and analyzed for the injected tracers as well as for ^{39}Ar, a long-lived activation product of interest.

1.1 Previous migration experiments

In 1993, the Non-Proliferation Experiment (NPE) was conducted in Rainier Mesa at the NNSS, releasing ^{3}He and SF_6 in conjunction with the detonation of 1.3 million kg of chemical explosives (Carrigan et al. 1996). This experiment compared the transport of the two tracers, measuring the arrival time and relative dilution of each tracer as it reached the surface. It highlighted the need to further examine the different transport rates of various tracer gases since it was initially assumed that the tracers would transport equally. While the NPE looked at the long-distance transport of tracer gases, the tracers in the NPE were advectively driven by the pressure and temperature increases caused by the chemical explosion, rather than being allowed to transport naturally.

In an effort to better understand the case when tracers transport through the subsurface with no explosive driving force, two previous noble gas migration experiments have been conducted at the NNSS. The first was the 2013 Noble Gas Migration Experiment (NGME), in which the radioactive tracers ^{37}Ar and ^{127}Xe and the stable tracer SF_6 were injected into the site of the Barnwell historic underground nuclear explosion (Olsen et al. 2016). 100 days after the tracer injection, the injection point was pressurized with air, forcing the rapid transport of the tracer gases to the surface where samples were being collected. This experiment highlighted that radioactive noble gases transport at a different rate in the subsurface than SF_6, despite both tracers being considered non-reactive.

The second experiment was the UNESE Phase 1 experiment, also known as NGME16, which repeated the injection of ^{37}Ar, ^{127}Xe, and SF_6 into the Barnwell chimney (Johnson et al. 2019; McIntyre et al. 2017). In this experiment, however, the pressurization of the injection point was not repeated, and boreholes were used to collect samples from various depths surrounding the

injection point. In contrast to the first NGME where radioargon and radioxenon were seen to transport together, the results of this experiment were that radioxenon was depleted relative to both radioargon and SF_6 after transporting through the subsurface. It is believed that the different transport rates were observed in the 2016 experiment since that experiment was indicative of a long-term seepage event while the 2013 experiment was more representative of bulk advective transport of gas from the injection point.

1.2 Tracers of interest

Samples collected as part of the UNESE Phase II migration experiment were analyzed for three radioactive gases (^{37}Ar, ^{39}Ar, ^{127}Xe) and two stable tracers (SF_6 and CF_3Br (Freon 13B1)). Three of the tracers, ^{37}Ar, ^{127}Xe, and SF_6 were used in previous transport experiments including UNESE Phase I. The tracers SF_6 and Freon™13B1 were used to act as stable surrogates for the radioactive tracers since they do not suffer loss due to radioactive decay. However, the results of the previous tracer experiments and more recent laboratory experiments show that the behavior of these tracers are not identical (Johnson et al. 2019; Olsen et al. 2016; Paul et al. 2018).

1.2.1 Stable tracers

The tracer Freon 13B1 (CF_3Br) was injected into the Disko Elm chimney 6-months prior to the noble gas migration experiment discussed in this report. During that injection, the chimney was pressurized, forcing the tracer to transport much more rapidly than the subsequent injection. A subset of samples collected during the experiment were analyzed for Freon 13B1 to determine if the combined advective and diffusive transport had caused it to reach our sampling locations.

The stable tracer SF_6 was injected in conjunction with the two radioactive tracers. A subset of the samples collected as part of this experiment were analyzed for the SF_6 concentration. A challenge with using SF_6 in this experiment is its legacy presence in P-tunnel from use in electrical equipment. Because of this, there is potential for a background of SF_6 to be present in the surrounding geologic media.

1.2.2 Radioactive tracers

Radioactive isotopes of xenon (^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe) are produced in large quantities during a UNE as fission products. The longest-lived isotope, ^{131m}Xe, has a half-life of 11.9 days, which makes it challenging to inject sufficient quantity to be detectable over a year-long migration experiment. Instead, a different radioisotope of xenon was used, ^{127}Xe, which has a half-life of 36.4 days which is more favorable for use in a months long experiment.

The radiotracers used in this experiment were produced by irradiating a mixture of stable ^{36}Ar and ^{126}Xe at the University of Texas at Austin's TRIGA reactor over the course of 18 days.

During UNESE Phase I, a persistent high-activity background was detected in samples collected for measurement of the ^{37}Ar background. The culprit was identified as ^{39}Ar, an activation product produced when neutrons interact with potassium in the surrounding geology (McIntyre et al. 2017). Since that initial detection, significant work has been performed at PNNL to improve our ^{39}Ar measurement capability, particularly of the high-activity samples (up to ~21 kBq/m^3 in Phase II) which were collected during Phase I and Phase II. Additionally, work is ongoing to understand the potential signatures provided by ^{39}Ar. In support of this work, the

samples collected as part of the UNESE Phase II migration experiment were each analyzed for their ^{39}Ar concentration.

2.0 Disko Elm Tracer Injection

In 2017 a reentry borehole (designated RE-7) was drilled from the tunnel wall into the chimney of the Disko Elm nuclear test as shown in Figure 1. The borehole was angled upwards at 31° in order to strike the chimney at a point well above any potential nuclear debris. The borehole was drilled using a coring bit to a diameter of 9.65 cm.

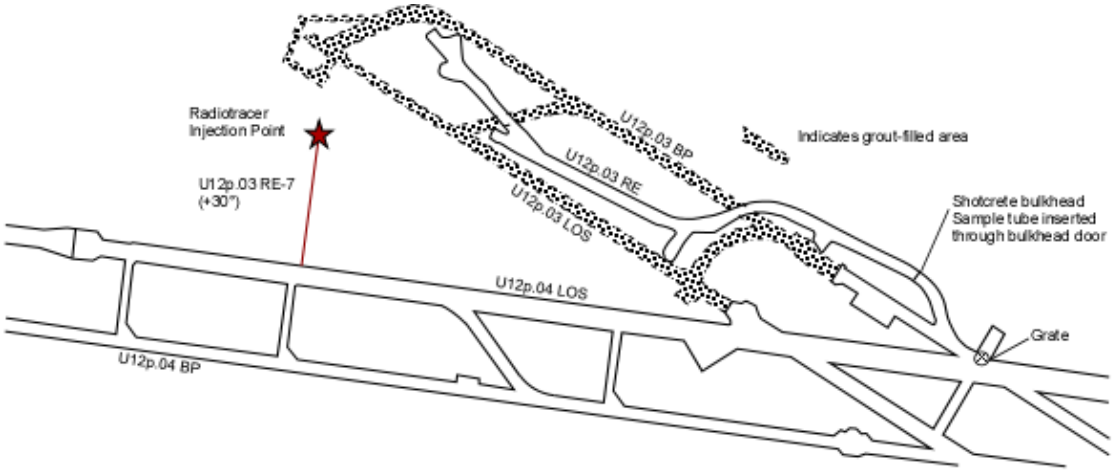

Figure 1. A diagram of the area of U-12p Tunnel surrounding the Disko Elm nuclear test showing the location of the radiotracer injection.

The RE-7 borehole was then completed as shown in Figure 2 and Figure 3. Two steel tubes were welded together every 9.1 meters then inserted into the borehole. The larger injection tube had an OD of 4.83 cm, an ID of 3.85 cm, and was connected using flush joints. Approximately 1 meter from the end of the injection tube a rubber packer cup was inserted to block grout from flowing into the chimney. The smaller of the two steel tubes terminated several meters below the injection tube terminus and was used as a grout return line. Grout, specifically MC300 Ultrafine Cement, was pumped into the borehole from the collar and allowed to flow until the grout return line was filled, indicating that grout had filled the hole to at least the end of the grout return line.

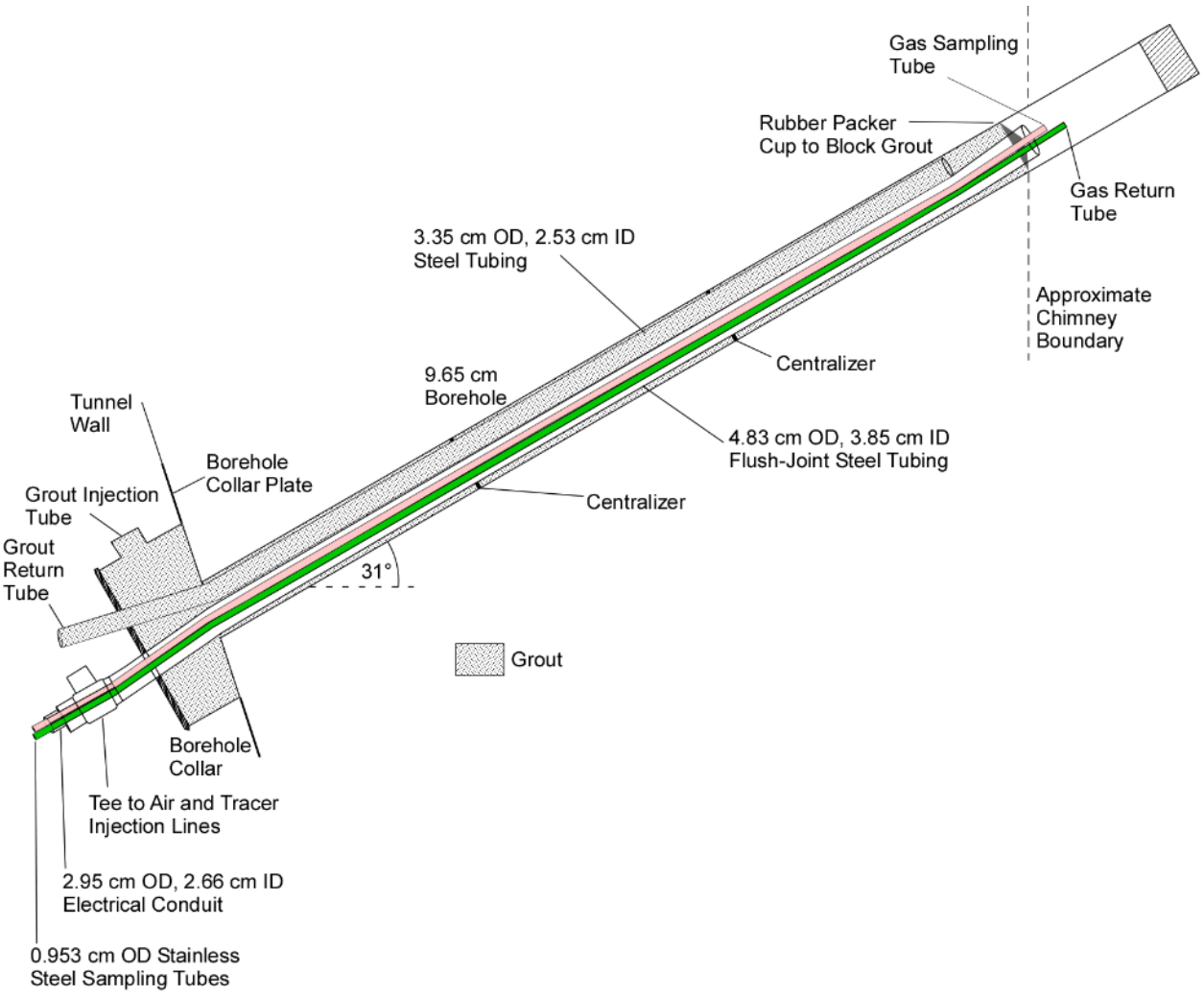

Figure 2. A side view of the RE-7 borehole completion.

The injection tube penetrates approximately 1 meter past the chimney boundary. Inside this tube was inserted two smaller stainless-steel tubes, wrapped in electrical conduit. These smaller tubes were used as gas sampling and return lines. A cross-section of the injection and sampling tube completion is shown in Figure 3. Tracer gasses were injected through the 4.83 cm OD steel tubing. Sampling was performed using the pair of 0.953 cm stainless steel tubes, with one tube used to draw sample from the chimney and the other used to recirculate the gas back into the chimney.

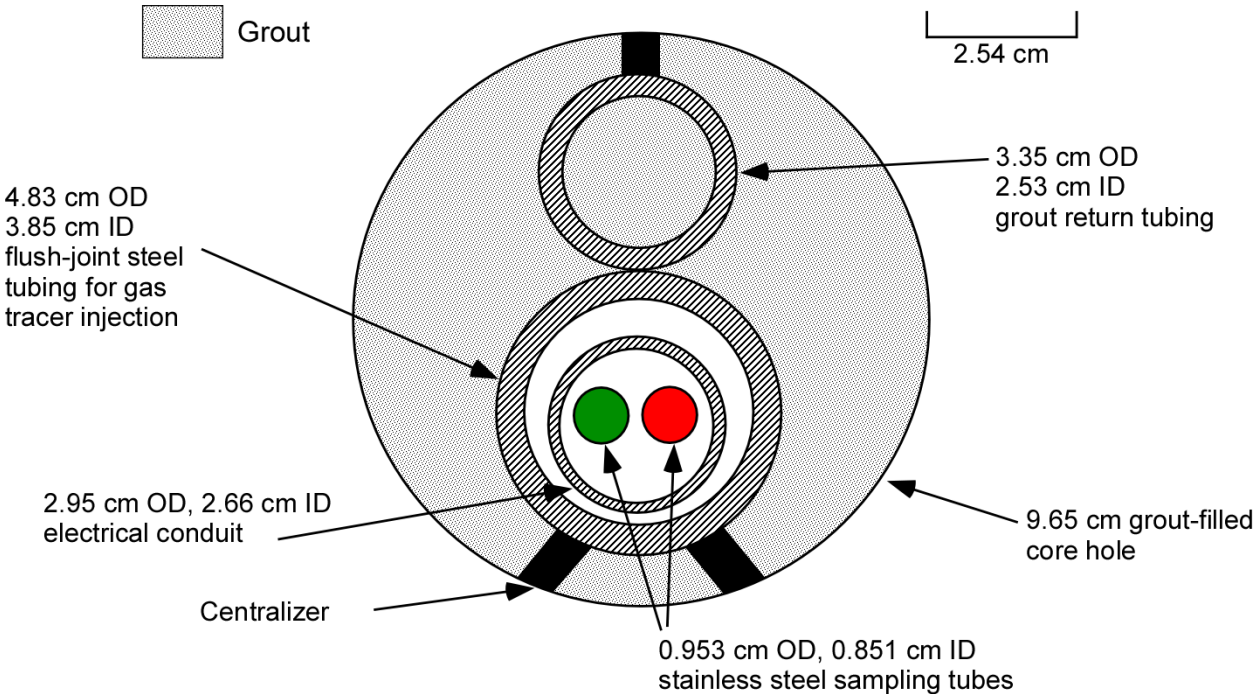

Figure 3. A cross section of the borehole completion for the RE-7 injection borehole.

2.1 June 2018 tracer injection

Two radioactive tracers (^{37}Ar and ^{127}Xe) and one stable tracer (SF_6) were injected as part of the June 2018 injection. The total tracer injections equaled 0.65±0.15 Ci of ^{37}Ar,0.56±0.14 Ci of ^{127}Xe, and 51 kg of SF_6. The three tracer gases were injected into the P-tunnel chimney on June 20, 2018 from 09:15 to 14:15 local time (PDT). A diagram of the injection system setup is shown in Figure 4. The radiotracers were delivered in three lead shielded flow-through volumes. The three sample volumes were connected to the injection system in parallel using quick connect fittings and were secured in place using vice clamps as shown in Figure 5. Nitrogen gas was used as the carrier gas for the radiotracers. The injection of the nitrogen and radiotracers was digitally throttled and adjusted throughout the injection to provide a near-constant activity injection rate. At the end of the injection, the nitrogen flow rate was increased to blow out the remaining radiotracers.

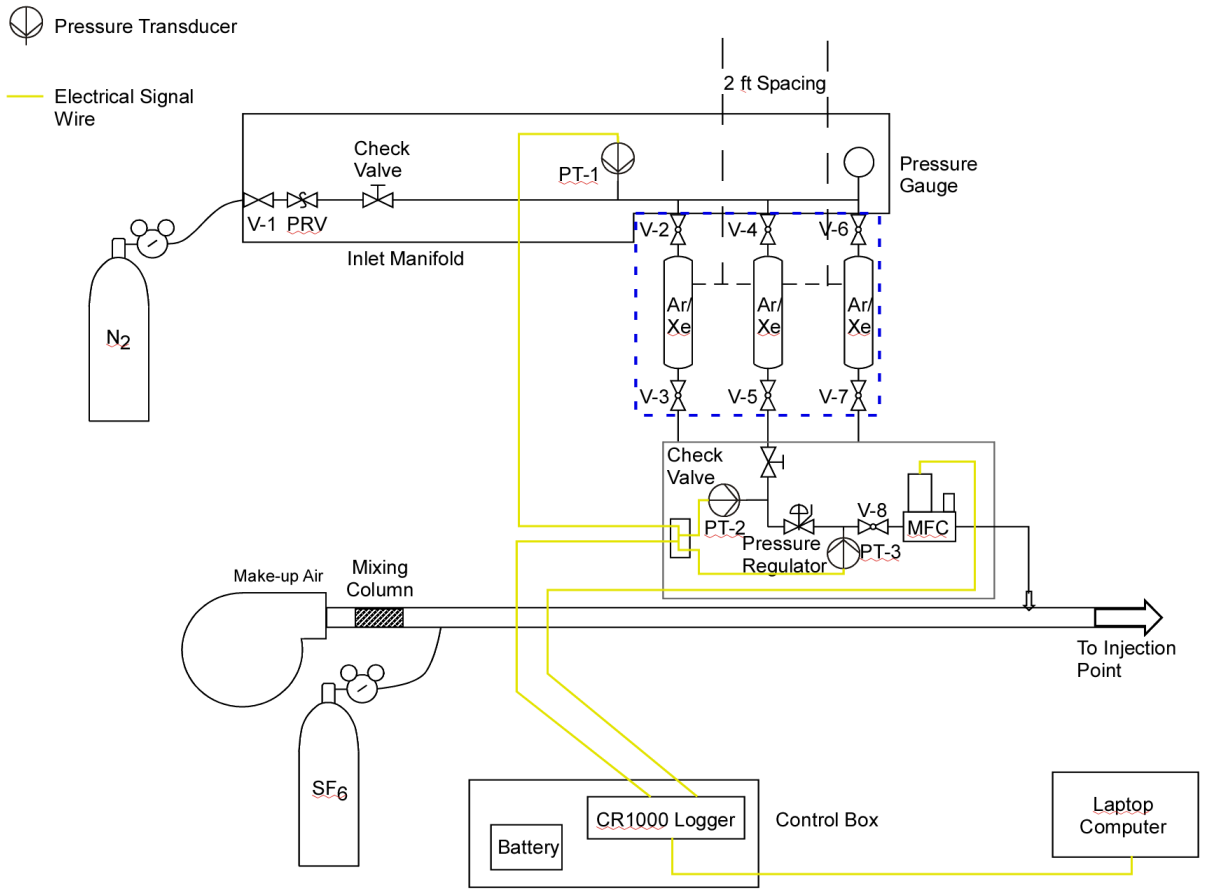

Figure 4. Diagram of the tracer gas injection system used for the June 2018 injection. Everything outside of the blue dashed line was pre-assembled and pressure tested prior to the tracer injection

Figure 5. Canisters (as shown in the blue box in Figure 4) containing the mixed ^{127}Xe and ^{37}Ar tracer as deployed for the June 2018 tracer injection.

The stable SF_6 gas was delivered in a standard gas cylinder and was connected to injection line downstream of the radiotracers and make-up air. The SF_6 cylinder was wrapped in heat tape to prevent the bottle from freezing as shown in Figure 6. The flow of SF_6 and make-up air were each controlled by rotometers and the flow-rates recorded in Table 1. A total of 51 kg, or 8.5 m^3 at STP of SF_6 were injected. In order to improve mixing of the air, radiotracers, and SF_6, the make-up air was flowed through a mixing column to increase turbulence and induce mixing. The total make-up air volume was 43.3 m^3.

Table 1. Flow rates of Make-up air, SF_6, and radiotracers as recorded during the tracer injection.

Time [PDT]	Make-up air flow [L/min]	SF_6 flow [L/min]	Radiotracer flow [mL/min]
1000	170	78	69
1010	170	75	70
1020	170	80	70
1030	170	80	69
1045	170	80	69
1100	170	80	69
1115	170	80	69
1130	170	70	69
1131	170	80	69
1139	170	80	73
1158	170	70->80	74
1200	170	80	77
1215	165->170	75->80	78
1233	172	75->80	77

1245	170	80	77
1257	170	80	209
1320	170	80	209
1334	170	75->80	209
1346	170	75->80	209
1358		60->dropping	208
		Letting SF_6 bleed dry	
1402		50	
1403		40	
1405		0->shutoff	
1406			1008
1409			2000
1410			3013

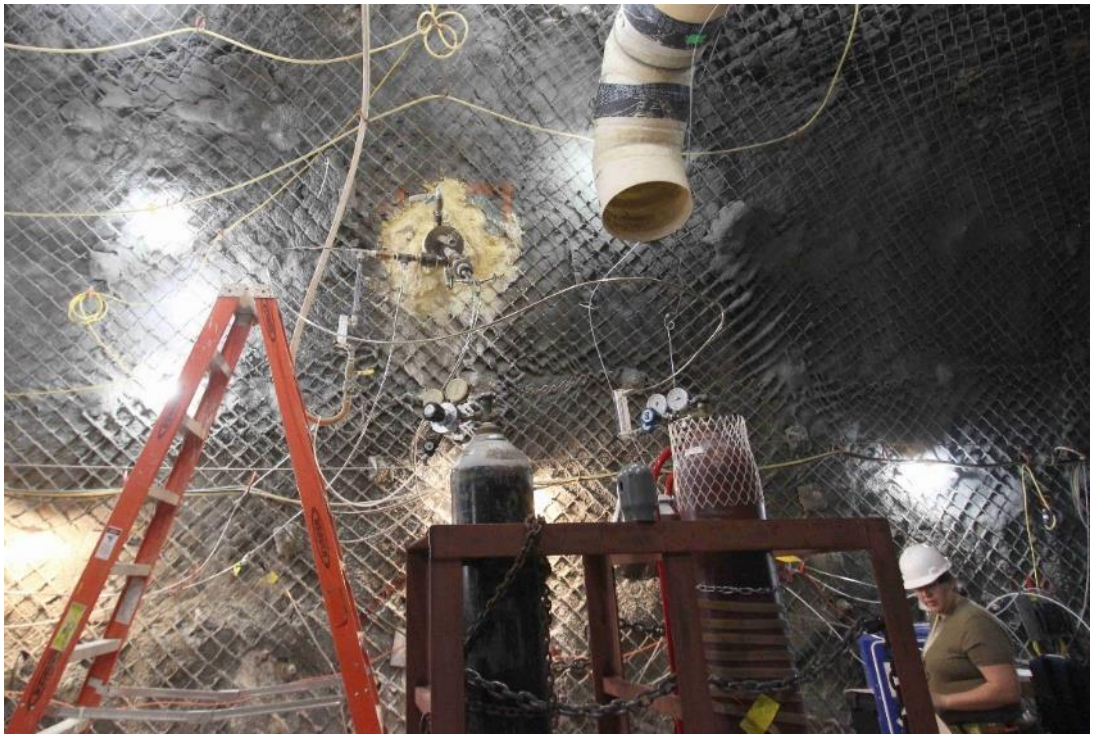

Figure 6. Cylinders containing nitrogen gas (green, left) and sulfur hexafluoride (red, right) as deployed for the June 2018 tracer injection. The heat tape used to prevent freezing of the SF_6 cylinder is seen wrapped around the red SF_6 cylinder at the lower right.

3.0 P-tunnel Sampling

Gas samples were collected from three locations inside P-tunnel. Two of the sampling locations allowed for both real-time measurement of the ^{127}Xe activity in air and collection of discrete samples into 1 L Tedlar bags. The third location only allowed for discrete sample collection. The locations of these three sampling points is shown in Figure 7 overlaid on a map of the surrounding tunnel.

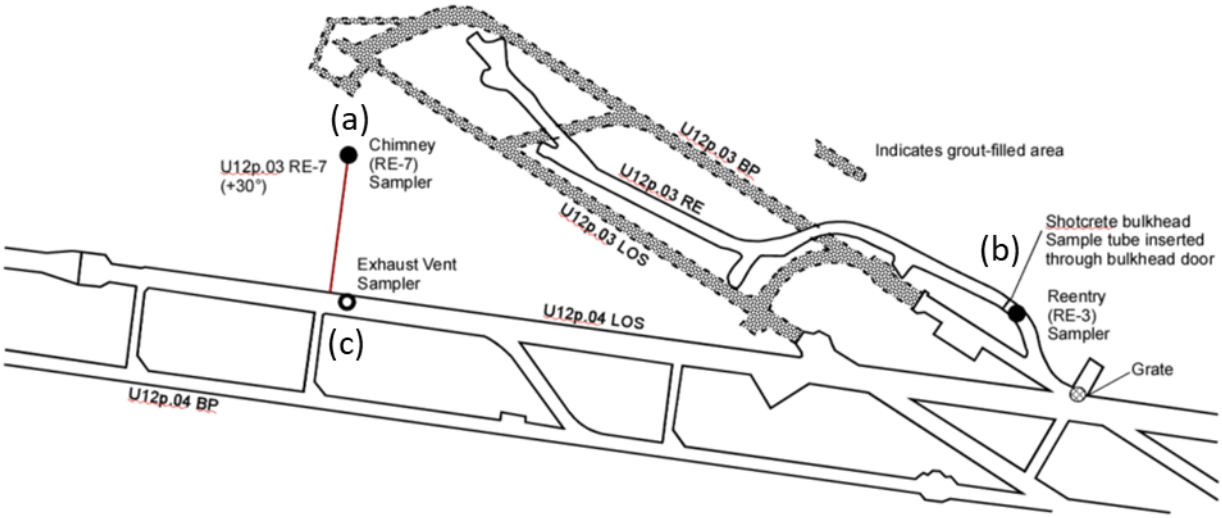

Figure 7. A diagram of the area of U-12p Tunnel surrounding the Disko Elm nuclear test. Recirculating gas samplers are shown as filled circles, while the open circle represents the location of a grab sampler.

3.1 Recirculating gas measurement system

Two gas recirculation systems were installed in P-tunnel to continuously circulate gas from the sampling location to a radiation detector before being returned to the sampling region. This allowed for near-realtime measurements of the ^{127}Xe concentration. The first recirculation system was installed on the RE-7 borehole, see (a) in Figure 7, to sample gas from the injection point. This allowed for monitoring of the ^{127}Xe activity during and after the tracer injection. The second system was used to analyze gas taken from the RE-3 reentry tunnel which can be seen at point (b) on the right side of Figure 7. This sampling point was installed on the far side of a bulkhead installed to separate the legacy Disko Elm re-entry drift from the main tunnel complex.

Gas was collected from the sampling point and pulled through a 1.2 L Marinelli beaker and sent back to the sampling region with a flow rate of 2 L/min. Gamma-ray detectors were then used to count the decays of ^{127}Xe in the gas volume. The detector setup is described in greater detail in Section 5.3.

3.2 Small volume samples

Small volume samples (1 L) were collected from three locations in P-tunnel. Two of the samplers drew directly from the gas lines which fed the recirculating gas systems while a third system drew samples from the tunnel ventilation intake located close to the RE-7 chimney sampler as seen at point (c) in Figure 7. Each system was configured to automatically collect

samples on a pre-set interval. Samples were collected in 1 L Tedlar bags, with a sampling period of one-minute. Each sampler was capable of collecting six samples before needing to be serviced. On days when a single sample was collected the samples were typically collected at 1:00am local time. Large volume background samples

Prior to the tracer injection, a series of large-volume air samples were collected from P-tunnel in large bladders (~2 m^3) as shown in Figure 8. These samples were analyzed for the presence of the injected tracers and measured for ^{39}Ar. An additional atmospheric sample was collected on the tunnel apron via direct compression into a SCUBA bottle.

Figure 8. Large volume (~2 m^3) background samples being removed from P-tunnel by rail car.

4.0 Aqueduct Mesa Borehole and Sampling

Sampling was performed from the surface of Aqueduct Mesa (which contains P-tunnel) for a year following the tracer injection. Samples were collected in one of three ways:

- Large volume (~2 m^3) samples compressed directly into a SCUBA bottle
- Large volume (~2 m^3) samples collected in a large volume grey water bladder bag before being compressed into a SCUBA bottle
- Small volume (1 L) samples collected in Tedlar bags.

4.1 Aqueduct Mesa borehole

A borehole was drilled from the surface of Aqueduct Mesa to a depth of 262.8 m and completed with four gas sampling zones as shown in Figure 9. Each sampling zone was completed with a layer of sand both above and below a layer of gravel which surrounded a pair of stainless steel mesh screens connected to the sampling and return lines. The sampling lines were 9.525 mm OD stainless steel, while the return lines were 6.35 mm OD stainless steel. The mesa borehole was approximately 80 m from the Disko Elm surface ground zero (SGZ).

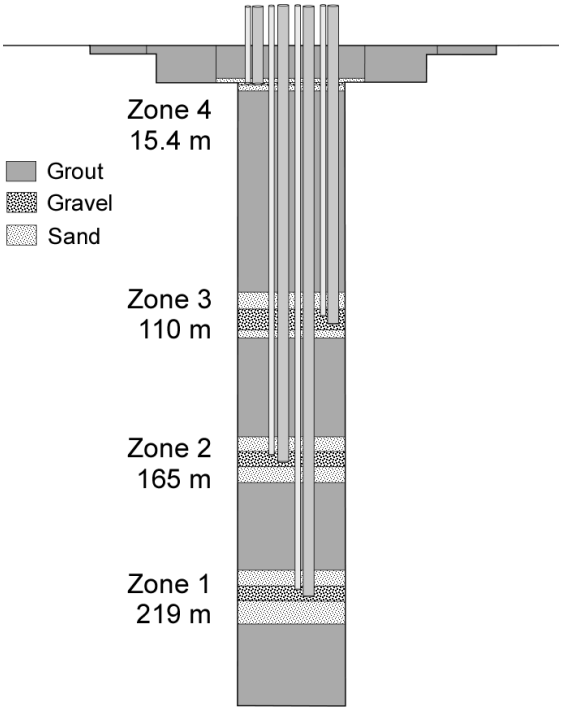

Figure 9. A schematic of the sampling borehole drilled from the surface of Aqueduct Mesa.

Large volume samples were collected into bladder bags approximately monthly from each zone, while small volume samples were collected at an interval such that six samples were collected between each large volume sample. After collection, the large volume samples were compressed into SCUBA bottles with a dive-air compressor. Assuming that the highest activity samples would come from the deepest borehole region (Zone 1), sample compression was performed in reverse order to minimize the probability of cross contamination from residual gas

inside the compressor. The compressor was also purged with atmospheric air between each sample.

4.2 Atmospheric sampling

Atmospheric samples were occasionally collected from a location downwind of the Aqueduct Mesa borehole. Theses samples were collected via direct compression into SCUBA bottles. The typical atmospheric sample collection method is shown in Figure 10, with the air intake positioned ~30 cm above the ground surface. When atmospheric samples were collected, they were always the first sample compressed to minimize the possibility of memory effect in the compressor contaminating the samples.

Figure 10. SCUBA compressor setup to collect an atmospheric sample near the mesa borehole.

4.3 Tarp sampling

Collection of shallow subsurface samples was accomplished using a surface tarp sampling location, shown in Figure 11.This tarp was positioned near the Disko Elm SGZ and the edges were both buried in the soil and covered with sandbags to minimize atmospheric infiltration. Samples were then collected from a tube placed underneath the center of the tarp into a large bladder bag. After sampling was complete the air in the bladder bag was compressed into a SCUBA bottle.

Figure 11. Surface tarp location near surface ground zero (SGZ) of Disko Elm (white flag).

5.0 Tracer Measurement Systems

The various radioactive tracers measured as part of this experiment are listed in Table 2 along with their half-lives and primary mode of decay. Because of the variety of decay modes, different detectors were needed to make the measurements of each isotope.

Table 2. The radioactive isotopes which were measured during the UNESE Phase II experiment, their half-lives, and primary decay modes.

Isotope	Half-Life	Primary Decay Mode
^{37}Ar	35.0 days	Electron capture
^{39}Ar	269 years	Beta-decay
^{127}Xe	36.3 days	Simultaneous emission of gamma-rays, X-rays, Auger and conversion electrons

The samples collected during this exercise where processed on a number of analysis system both onsite and at PNNL. Onsite measurements were made of both argon isotopes for two weeks after injection using a fieldable proportional counter system. All later measurements were made on the same system but were performed at PNNL. Xenon measurements were performed onsite in recirculating gamma detector systems for two months following injection. All large volume samples were measured on laboratory beta-gamma detectors for the duration of the experiment. All analysis of stable gas tracers were performed at PNNL.

5.1 Measuring ^{37}Ar

In order to quantify ^{37}Ar, approximately 400 L of sample air was introduced to the Argon Field System (Figure 12) for processing and quantification (Hayes et al. 2019). This system was used for onsite measurement of samples during the two weeks following the tracer injection, then was returned to PNNL and used to measure samples that were shipped back to the lab for the remainder of the experiment.

Measurement was accomplished using a 250 cc proportional counter housed inside of a cosmic ray veto shielding comprised of active shielding (PVT panels) and passive shielding made of lead and borated polyethylene. The analysis was accomplished by measuring the 2.82 keV auger electron emitted from the decay. A high gain is set on the proportional counter to observe the region between 0 and 15 keV. A gaussian fit is used over the search region to quantify activity. The system is well described in (Alexander et al. 2019).

Figure 12. The ^{37}Ar measurement system deployed to the NNSS being used to measure a sample of air collected from P-tunnel (bottle denoted with orange hang-tag).

5.2 Measuring ^{39}Ar

Samples were measured for ^{39}Ar by recalibrating the gain of the proportional counters used in the ^{37}Ar measurement to observe the whole ^{39}Ar beta spectrum, looking from 0 to 650 keV. The analysis was accomplished by subtracting a known background for the detector at the sample pressure and comparing the difference with a spectrum from a known activity “efficiency” sample. Often, samples from the P-Tunnel borehole were too high in ^{39}Ar activity to adequately quantify on the system, so a dilution of sample gas and P10 background gas was used to bring the samples within the known range of the counters (<500 Hz.)

5.3 Measuring ^{127}Xe

Measurements of ^{127}Xe were performed using two detector types. Onsite measurements were performed using a recirculating gas flow around a NaI(Tl) gamma ray detector. Laboratory measurements were performed using large volume gas samples which were counted in beta-gamma coincidence detectors.

5.3.1 Laboratory measurements of ^{127}Xe

Samples were measured for ^{127}Xe using beta-gamma coincidence spectroscopy. Large volume samples were first processed to extract and purify xenon from the bulk air sample. This form of sample analysis required larger sample volumes to extract the necessary amount of xenon from the whole air samples. The gas purification system uses a method similar to that described in (Bowyer, Abel, and Hubbard 1999) and beta gamma detectors typically use a scintillating plastic beta cell and a NaI(Tl) gamma detector (Cooper et al. 2007).

5.3.2 Measurements of ^{127}Xe in recirculation systems

Two recirculating gas systems were installed in the tunnel and used to monitor the ^{127}Xe concentrations in the chimney and in the RE-3 drift. For the system installed on the chimney

(the RE-7 sampler), a 3x3 NaI(Tl) detector was placed inside the Marinelli beaker and programmed to collect a gamma spectrum an average of every 10 minutes. The chimney detector system is shown in Figure 13.

On the re-entry system (the RE-3 sampler), the detector inside the Marinelli beaker was a 3x3 CsI(Tl) which was programmed to collect a gamma spectrum an average of every 15 minutes.

Figure 13. (Left) The recirculating xenon detector installed on the RE-7 borehole where the tracer injection occurred. (Right) The recirculating detector system installed in P-tunnel with Tedlar bags attached to collect discrete samples.

5.3.3 Analysis of recirculating gas spectra

The minimum detectable activity (MDA) was calculated for each recirculating gas detector and from this MDA the equivalent minimum detectable concentration (MDC) was derived.

For the chimney detector, a 3x3 NaI(Tl) detector, the MDA was calculated to be 11 ± 3 Bq ^{127}Xe in 1.15 L. This is equivalent to an MDC of $(9.1 \pm 2.5) \times 10^3$ Bq/m^3. Because of the high activity of ^{127}Xe observed by this detector, significant detector dead-time had to be accounted for. This, in part, led to a decrease in the MDA when compared to the re-entry detector.

The re-entry detector, a 3x3 CsI(Tl) detector, the MDA was calculated to be 5.3 ± 1.3 Bq ^{127}Xe in 1.15 L. This is equivalent to an MDC of $(4.6 \pm 1.2) \times 10^3$ Bq/m^3.

In order to simplify the analysis of the spectra, the total number of counts in the spectrum was used to calculate the ^{127}Xe activity. This assumption can be made since the number of background counts is negligible when compared to the number of ^{127}Xe counts in the detector. A spectrum collected by the chimney detector before and after the radiotracer injection is shown in Figure 14.

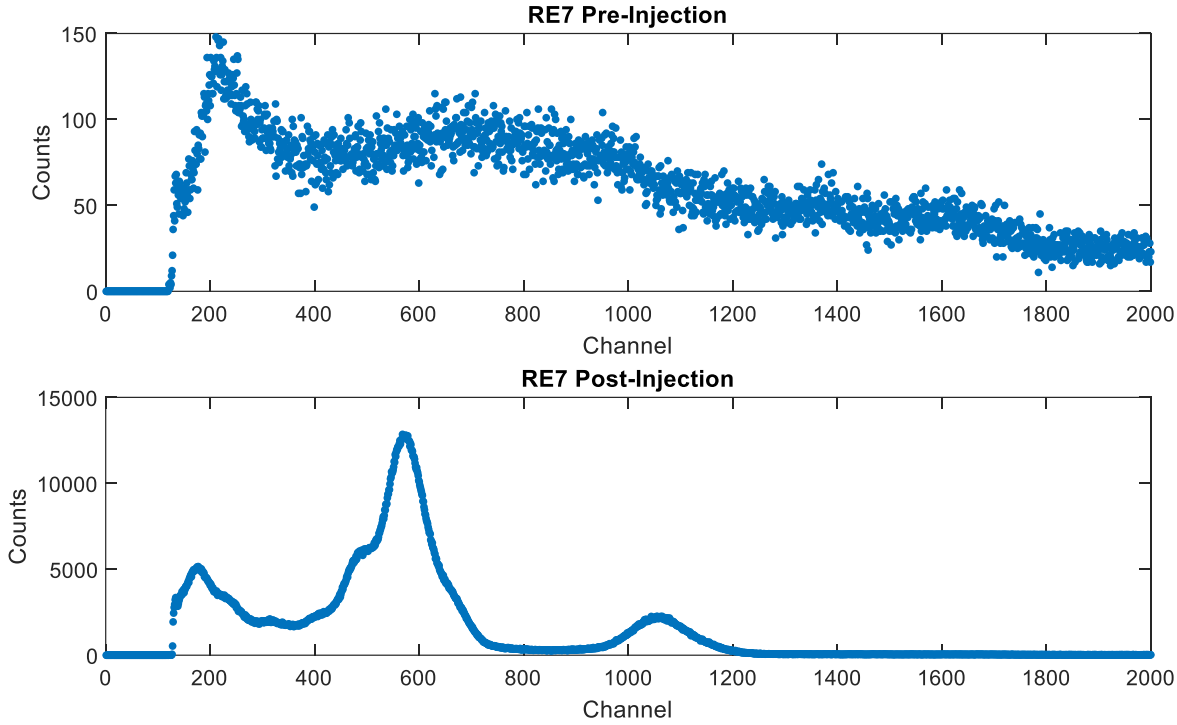

Figure 14. 10-minute spectra collected by the chimney (RE-7) gas detector before and after the radiotracer injection.

5.4 Measuring the stable tracers

Initial measurements of the stable tracers were performed on an Agilent Technologies gas chromatograph – mass spectrometer (GC-MS) (Agilent 7890A – Agilent 5975C) system with a helium carrier gas. Samples were initially injected into the GC-MS using a 0.25 mL sampling loop, then the compounds were separated using a 60 m long GasPro PORAPLOT column. The GC column temperature was initially held at 40°C for 4 minutes, then ramped up to 230°C at a rate of 15°C/min. The temperature was held at 230°C for five minutes. Five-level calibration was performed using standards prepared with mixture of both tracers at concentrations of 2, 10, 40, 200, and 1000 ppb_v in helium.

After several months of no detections of the injected tracer gases, the decision was made to have commercial analysis performed on a subset of the small volume samples to reduce the detection limit to part-per-trillion (ppt) levels. Samples were shipped to Lagus Applied Technologies and analyzed for SF_6 and Freon 13B1.

The reported limit of detection of the Largus analysis for Freon 13B1 was 40 ppt, and the limit of quantification was 100 ppt. The approximate calibration ranges for Freon 13B1 spanned 0.25 – 300 ppb. The reported limit of detection for SF_6 was 10 ppt, and the limit of quantification was 12 ppt. The approximate calibration ranges for SF_6 spanned 0.012 – 500 ppb.

6.0 Results

The following section lists the results of each measurement made as part of the UNESE Phase II gas migration experiment at P-tunnel.

6.1 Measurements of ^{39}Ar

After the discovery of the a large background of ^{39}Ar during UNESE Phase 1 (McIntyre et al. 2017), the decision was made to measure each sample for ^{39}Ar as well as for the injected tracers. The results of those measurements are shown here for both subsurface and atmospheric samples.

6.1.1 Subsurface ^{39}Ar Measurements

Each large volume sample collected on Aqueduct Mesa was analyzed to determine the concentration of ^{39}Ar present in the sample. The results of these measurements are shown in Tables 3-6 below and are plotted as both a function of depth from the ground surface and distance from the historic Disko Elm UNE in Figure 15.

Table 3. Measured ^{39}Ar concentrations in Borehole Zone 1

Sampling Date	^{39}Ar Concentration [Bq/m3]	+/-
7/23/2018	19988	93.4/0.0
8/16/2018	21295	560/467
10/15/2018	11768	93.4/0.0
11/12/2018	18960	560/560
12/10/2018	19054	560/467
4/17/2019	19054	467/467

Table 4. Measured ^{39}Ar concentrations in Borehole Zone 2

Sampling Date	^{39}Ar Concentration [Bq/m^3]	+/-
6/20/2018	16532	93.4/93.4
7/22/2018	17839	0.0/93.4
8/16/2018	13636	374/374
9/13/2018	18213	374/467
10/15/2018	14570	93.4/93.4
11/12/2018	15691	374/374
12/10/2018	14757	374/374
4/17/2019	16812	467/374

Table 5. Measured ^{39}Ar concentrations in Borehole Zone 3

Sampling Date	^{39}Ar Concentration [Bq/m^3]	+/-
6/19/2018	1135	20.4/15.3
7/22/2018	338.1	0.9/0.9
8/16/2018	304.5	2.80/1.87
9/13/2018	328.8	8.41/8.41
10/15/2018	574.4	3.74/3.74
11/15/2018	449.3	12.1/11.2
12/13/2018	418.4	10.3/10.3
4/17/2019	577.2	14.0/14.0

Table 6. Measured ^{39}Ar concentrations in Borehole Zone 4

Sampling Date	^{39}Ar Concentration [Bq/m^3]	+/-
6/20/2018	122.4	0.9/0.9
7/22/2018	55.20	0.3/0.2
8/16/2018	300.8	7.47/7.47
9/13/2018	26.06	0.65/0.65
10/15/2018	25.78	0.47/0.37
11/12/2018	23.07	0.56/0.56
12/10/2018	32.50	0.93/0.84
4/17/2019	13.73	0.28/0.37

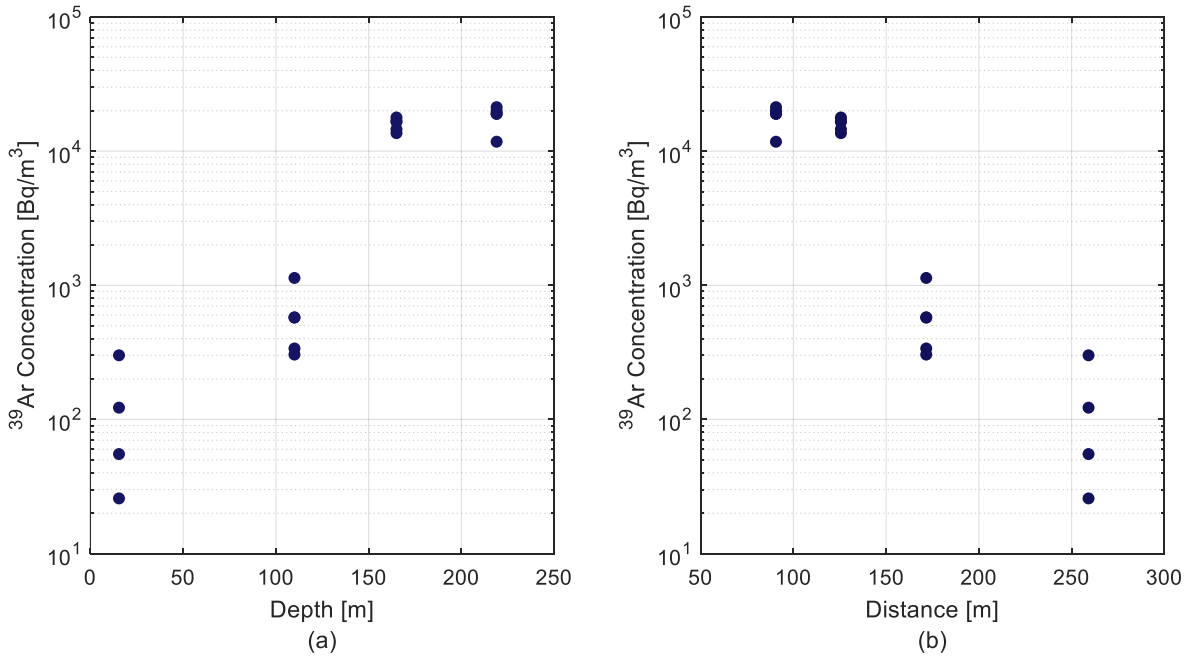

Figure 15. The measured ^{39}Ar concentrations as a function of (a) depth from the ground surface and (b) distance from the Disko Elm UNE. Collected from the Aqueduct Mesa borehole.

6.1.2 Atmospheric ^{39}Ar Measurements

In addition to the borehole gas samples shown above, atmospheric samples were also collected and analyzed from various locations and are reported in Table 7.

Table 7. Atmospheric concentrations of ^{39}Ar measured on Aqueduct Mesa and at the entrance to P-tunnel

Location	Sampling Date	^{39}Ar Concentration [Bq/m^3]	+/-
P-tunnel Apron	6/12/2018	5.931	0.08
Aqueduct Mesa	7/24/2018	0.524	0.01
Aqueduct Mesa	4/17/2019	0.779	0.02

6.2 Measurements of ^{37}Ar

Because of the large background of ^{39}Ar present in the geology surrounding Disko Elm, the probability of detecting ^{37}Ar decreased significantly. None of the injected ^{37}Ar was measured in samples collected from the mesa borehole. The minimum detectable activity of ^{37}Ar from each borehole sampling region is shown in Table 8 for a representative sample set (samples collected on 22 July 2018). For reference, the background levels of ^{39}Ar are also shown.

Table 8. The ^{37}Ar MDA in each borehole sampling zone on 22 July 2018. The background ^{39}Ar activity is also shown for each sample.

Sampling Zone	^{37}Ar MDA [Bq/m^3]	^{39}Ar Concentration [Bq/m^3]
Zone 1	3.635	19988
Zone 2	3.065	17839
Zone 3	0.313	338.1
Zone 4	0.165	55.20

6.3 Measurements of ^{127}Xe

6.3.1 Evolution of ^{127}Xe at the injection point

A primary goal of the recirculating gas detection system was to observe the evolution of the ^{127}Xe tracer in the Disko Elm chimney after injection. In Figure 16, the ^{127}Xe concentration in the chimney is shown for ~40 days post-injection.

Prior to the injection, sporadic ^{127}Xe concentrations appear above detection limits. These detections correspond to when the shielded source canisters were brought into the tunnel and placed near the detector. While the detector was shielded with a thin layer of lead, the nearby 0.6 Ci source of ^{127}Xe was still detectable.

The intent of the injection was to maintain a fairly constant rate of injection of the radiotracer activity. In Figure 17, the measured ^{127}Xe concentration in the chimney during the injection shows that the activity in the sampling region remained fairly constant during the injection, save for a fluctuation around 13:00 that corresponds to a change in the injection flow rate.

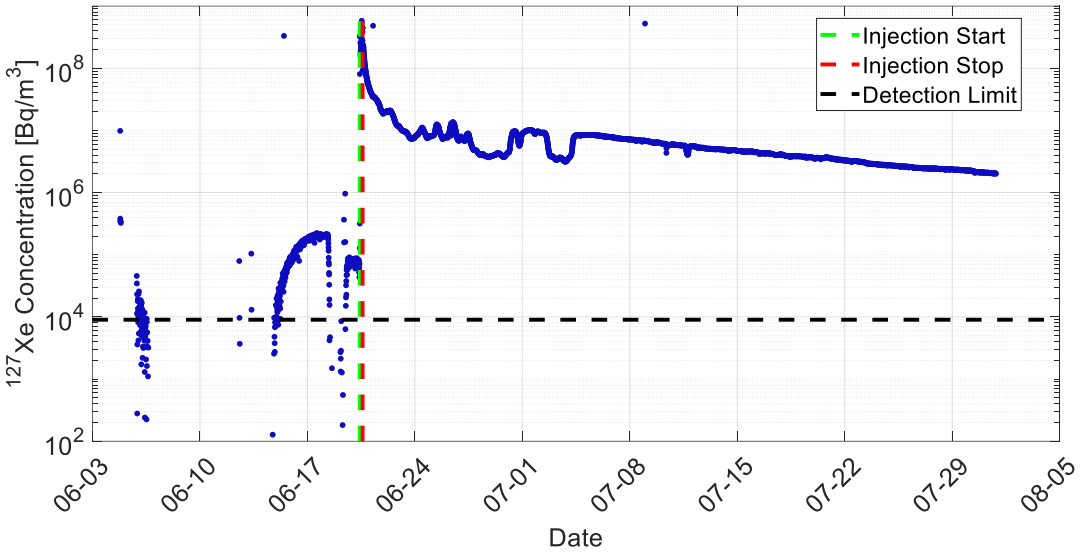

Figure 16. Measurements of the ^{127}Xe activity in the Disko Elm chimney.

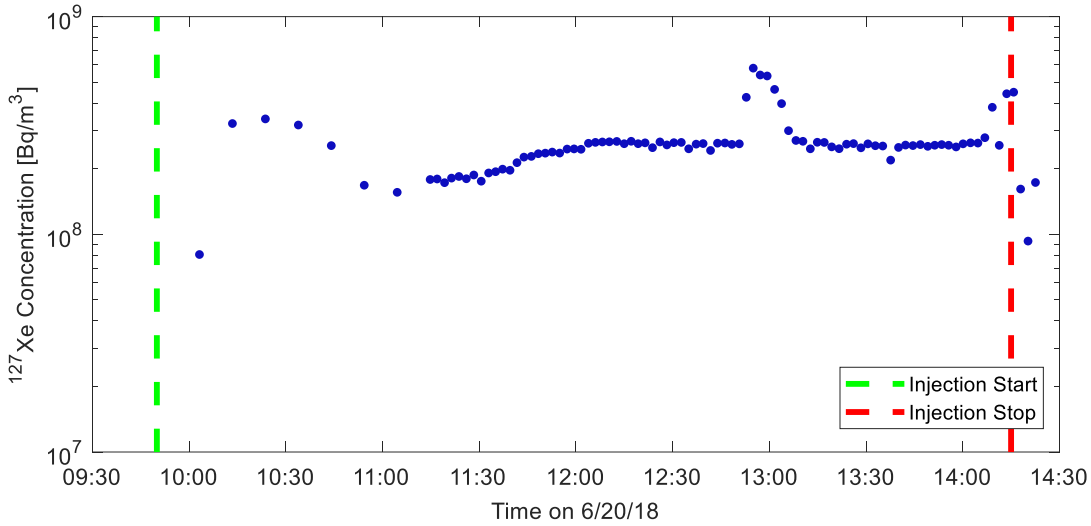

Figure 17. Measurements of the ^{127}Xe concentration in the Disko Elm chimney during the radiotracer injection.

The maximum concentration of ^{127}Xe measured by the detector was 5.8×10^6 Bq/m^3. After the injection the ^{127}Xe concentration in the chimney decreased rapidly for 36-hours as the tracer expanded to fill the available space and mixed with the air that was already present. During this time the concentration decreased from 5.8×10^8 Bq/m^3 to 2.7×10^7 Bq/m^3. This equates to an expansion of the tracer into an air volume of approximately 600 m^3.

After the initial drop, the concentration initially stabilized at approximately 5×10^6 Bq/m^3. At this point a series of fluctuations in the concentration are observed which generally correspond to tunnel openings and the shutdown and startup of the tunnel ventilation system.

In Figure 18 the late time evolution is shown next to the evolution that would be expected if the decrease was a function of radioactive decay alone. This result indicates that radioactive decay is not the only loss mechanism for ^{127}Xe in the chimney.

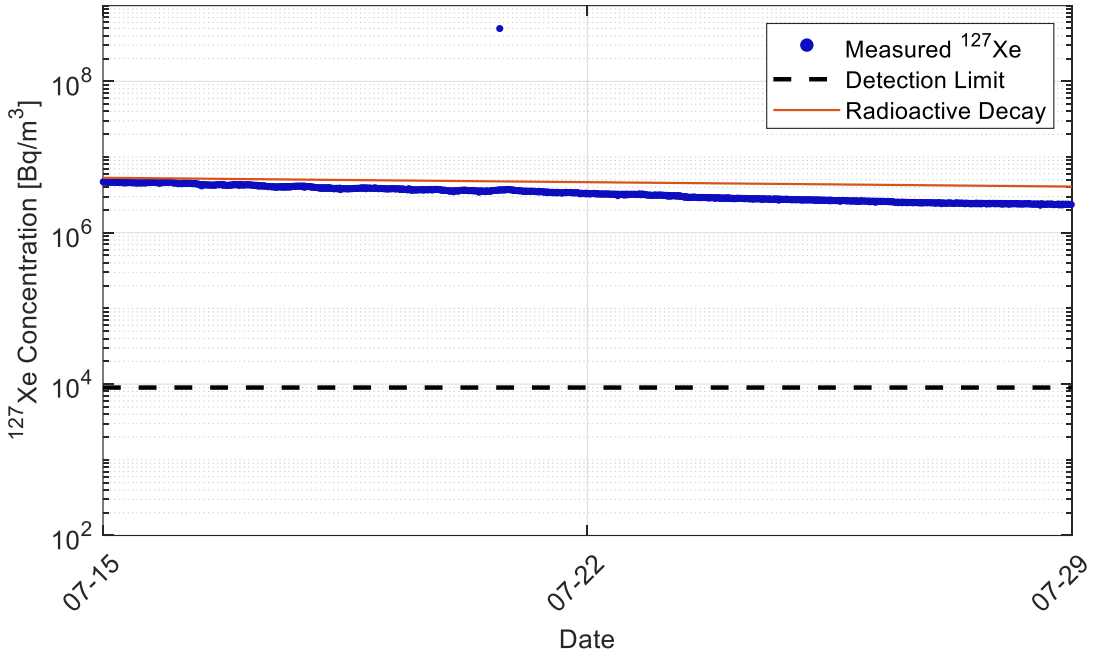

Figure 18. Late time decrease in ^{127}Xe activity concentration compared with the radioactive decay curve for ^{127}Xe.

6.3.2 Large volume ^{127}Xe samples from Aqueduct Mesa

None of the analyzed samples contained detectable quantities of ^{127}Xe. Two example spectra from the analysis of samples collected from borehole zones 1 and 2 on 4/17/19 are shown in Figure 19 and Figure 20. Neither spectrum shows any indication of the presence of radioxenon.

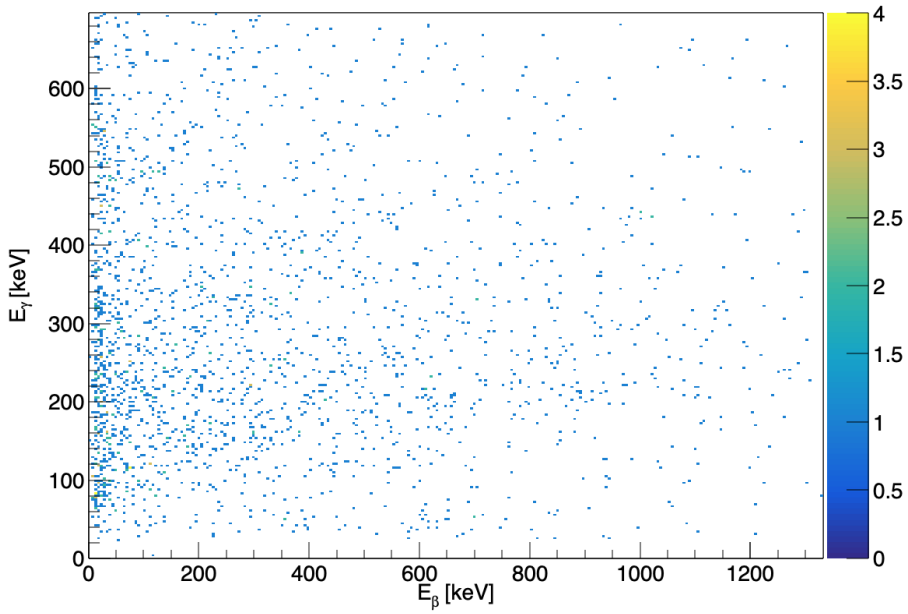

Figure 19. A beta-gamma spectrum of the sample collected from borehole Zone 1 on 17 April, 2019.

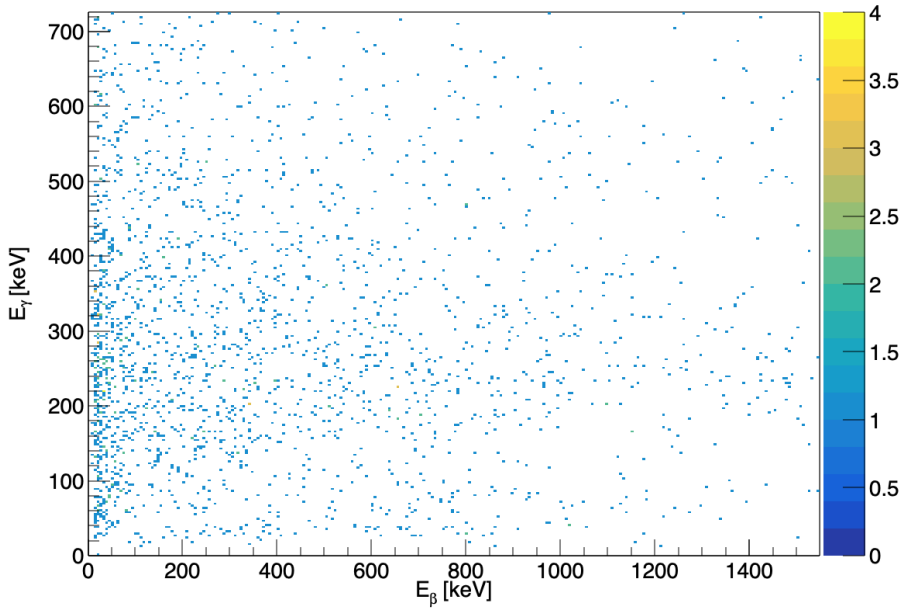

Figure 20. A beta-gamma spectrum of the sample collected from borehole Zone 2 on 17 April, 2019.

6.4 Measurements of stable tracers

6.4.1 Stable tracers in P-tunnel

In Table 9 through Table 11, the concentrations of SF_6 and Freon 13B1 measured in 1 L Tedlar samples collected in P-tunnel are shown. All of the SF_6 measurements made in RE7 after the date of the injection, 6/20/18, are well outside of the detector calibration range and should be viewed qualitatively rather than quantitatively.

Table 9. Measured stable tracer concentrations in RE7. The time of sampling is provided (where known) when multiple samples were collected on the same date. The (*) represents a duplicate analysis.

Sampling Date	SF_6 [ppb_v]	CF_3Br (Freon 13B1) [ppb_v]
2/13/18	NM	7.46
2/15/18	NM	10.9
3/22/18	NM	3.5×10^6
3/22/18	NM	2.0×10^6
3/22/18	NM	3.0×10^6
6/20/18 13:35	2.3×10^8	1.4853×10^4
6/20/18 13:35*	1.66×10^8	NM
6/20/18 21:42	7.760×10^6	2.147×10^5
6/22/18 18:22	2.46×10^7	2.267×10^5
6/21/18 9:30	2.05×10^7	1.051×10^6
6/21/18 22:09	2.107×10^6	2.181×10^5
6/24/18	6.923×10^6	1.164×10^6
6/22/18 10:38	2.325×10^6	2.189×10^5
6/25/18	4.239×10^6	1.122×10^6
6/26/18	1.196×10^5	2.2172×10^4

Table 10. Measured stable tracer concentrations in RE3. The time of sampling is provided (where known) when multiple samples were collected on the same date.

Sampling Date	SF_6 [ppb_v]	CF_3Br (Freon 13B1) [ppb_v]
6/18/18 12:00	<1	67467
6/20/18 13:10	<1	55107
6/20/18 17:10	2.82	55724
6/21/18 1:10	9.19	51827
6/21/18 5:10	14.17	50117
6/21/18 13:10	18.89	55967
6/21/18 17:10	13.32	46751
6/22/18 1:10	3.05	47168
6/22/18 9:10	2.34	52909
6/23/18 12:10	1.96	132
6/24/18 13:10	1.06	290

6/25/18 12:10	0.573	45000
6/25/18 16:10	0.641	33000
7/11/18	13.39	38048
7/11/18	<2.0	34938
7/13/18	13.06	35177
7/13/18	<2.0	32297
7/15/18	51.61	673.24
7/17/18	13.02	37219
7/19/18	17.17	17417
7/21/18	13.99	31821
7/29/18	7.84	15000
8/14/18	7.43	8000
8/22/18	2.72	30000
9/3/18	0.246	2.82
9/11/18	153	25000

Table 11. Measured stable tracer concentrations in the P-tunnel ventilation system. The time of sampling is provided (where known) when multiple samples were collected on the same date.

Sampling Date	SF_6 [ppb_v]	CF_3Br (Freon 13B1) [ppb_v]
6/20/18 9:00	1.46	10.01
6/20/18 9:45	642.74	2.95
6/20/18 13:10	574.27	3.21
6/20/18 17:10	341.03	7.84
6/20/18 21:10	130.61	17.14
6/21/18 1:10	157.9	6.92
6/21/18 5:10	106.37	9.9
6/21/18 9:05	162.12	8.47
6/21/18 13:10	77.2	28.08
6/22/18 1:10	36.22	14
6/22/18 9:10	36.52	9.7
6/23/18 0:00	22.97	7.44
6/23/18 12:10	17.76	30.02
6/24/18 0:00	15.05	17.2
6/25/18 0:10	12.36	9.58
6/25/18 12:10	25.06	14.51
6/25/18 16:10	14.84	6.25

6/25/18 20:10	14.49	8.7
6/26/18 0:00	14.91	5.45
7/11/18 0:00	18.74	24.76
7/13/18 0:00	28.32	15.76
7/15/18 0:00	142.17	54.72
7/17/18 0:00	7.29	26.26
7/19/18 0:00	12.57	13.2
7/21/18 0:00	19.36	13.99
8/6/18 0:00	0.549	7.09
9/7/18 0:00	0.349	4.95

6.4.2 Stable tracers in the mesa borehole

In Table 12 through Table 15 below, the concentration of stable tracers SF_6 and Freon 13B1 measured in samples collected from the Aqueduct Mesa borehole are shown. Most of these measurements were made on the 1 L Tedlar bag samples collected in the periods between the monthly large volume samples. Because of the elevated water level in the borehole during the early part of the experiment, no small volume samples were collected from Zone 1 until October 2018. At that point, the automated sampler previously used for Zone 4 was transitioned to collect samples from Zone 1.

Table 12. Measured stable tracer concentrations in borehole Zone 1.

Sampling Date	SF_6 [ppb_v]	CF_3Br (Freon 13B1) [ppb_v]
10/17/2018	0.158	0.263
10/21/2018	0.019	0
10/25/2018	0.086	0.000
10/29/2018	0.052	0.000
11/2/2018	0.144	0.057
11/6/2018	0.963	0.076
11/18/2018	1.50	0.120
11/22/2018	NM	0.000
11/26/2018	NM	0.111
11/30/2018	NM	0.000
12/4/2018	1.460	0.00
12/8/2018	1.060	0.00
12/23/2018	1.39	0.436
4/17/2019	0.858	1.9

Table 13. Measured stable tracer concentrations in borehole Zone 2. The time of sampling is provided (where known) when multiple samples were collected on the same date.

Sampling Date	SF_6 [ppb_v]	CF_3Br (Freon 13B1) [ppb_v]
6/21/18 11:42	0.066	0.041
6/21/18 12:22	<1	<1
6/22/18	0.776	0.177
6/23/18 8:00	0.504	0
6/23/18 22:00	<1	<1
6/24/18 12:00	0.821	0.122
6/24/18 2:00	<1	<1
6/27/18	0.987	0.089
7/1/18	0.431	0
7/5/18	0.097	0
7/13/18	0.478	0.093
7/17/18	0.786	0.089
7/29/18	0.518	0
8/6/18	0.436	0
8/14/18	0.459	0
9/15/18	0.467	0
9/15/18	0.987	0.114
9/23/18	0.069	0
10/1/18	0.503	0.089
10/25/18	0.395	0.134
11/2/18	0.383	0.121
11/6/18	0.527	0.22
11/26/18	0.428	0.08
11/26/18	0.345	0.077
12/4/18	0.456	0.063
4/17/19	0.139	0

Table 14. Measured stable tracer concentrations in borehole Zone 3. The time of sampling is provided (where known) when multiple samples were collected on the same date.

Sampling Date	SF_6 [ppb_v]	CF_3Br (Freon 13B1) [ppb_v]
6/19/18	0.2	0.288
6/21/18 12:00	0.042	0
6/21/18 12:22	<1	<1

6/22/18 4:00	<1	<1
6/22/18 18:00	0.908	0.226
6/23/18 8:00	0.613	1.83
6/23/18 22:00	0.277	0.07
6/24/18 2:00	<1	<1
6/24/18 12:00	0.915	0.157
6/27/18	1.03	0.156
7/5/18	0.199	0.073
7/9/18	0.585	0.081
7/13/18	0.031	0
7/17/18	0.042	0
8/6/18	0.077	0.05
9/15/18	0.389	0.078
9/19/18	0.234	0.064
9/23/18	0.307	0
9/27/18	0.199	0.073
10/1/18	0.075	0
10/5/18	0.075	0.063
10/17/18	0.025	0.148
10/21/18	0.018	0.44
10/25/18	0.054	0
10/29/18	0.059	0.057
11/2/18	0.048	0.055
11/6/18	0.049	0.053
11/26/18	0.137	0.109
4/17/19	0.06	0

Table 15. Measured stable tracer concentrations in borehole Zone 4. The time of sampling is provided (where known) when multiple samples were collected on the same date.

Sampling Date	SF_6 [ppb_v]	CF_3Br (Freon 13B1) [ppb_v]
6/21/18 12:00	<1	<1
6/21/18 12:20	0.411	0.108
6/21/18 12:40	<1	<1
6/21/18 13:00	<1	<1
6/21/18 13:20	<1	<1
6/22/18 4:00	<1	<1

6/22/18 18:00	<1	<1
6/23/18 8:00	<1	<1
6/23/18 22:00	<1	<1
6/24/18 2:00	2.2	1.58
6/24/18 12:00	<1	<1
6/27/18	<2	<2
7/1/18	<2	<2
7/5/18	<2	<2
7/9/18	0.223	0.047
8/6/18	0.279	0.091
8/14/18	<2	<2
9/27/18	0.167	0
10/1/18	0.18	0
10/5/18	0.049	0.042
4/17/19	0.015	0

The evolution of the stable tracer concentrations in the surface borehole for 40-days post-injection is shown in Figure 21. Elevated tracer concentrations are observed in samples collected on 6/23 and 6/24, however the elevated concentrations are observed more strongly for the Freon 13B1 than for SF_6. Since the Freon 13B1 was injected in January 2018, rather than with the other tracers on 6/20/18, this should not be indicative of the arrival of the injected tracers. The quick stabilization of these tracer concentrations at later times are also more indicative of relaxation back to background levels then the arrival of new tracer gas.

One potential explanation for these elevated measurements is the relaxation of gas after completion of the borehole. Significant quantities of water were pumped into the mesa borehole during drilling, particularly at the deeper levels, which would serve to force gases away from the borehole while the pore space was water filled. Additionally, the borehole was left open while drilling and completion was conducted, which would naturally serve to reduce the concentration of any background gases. Background levels of SF_6 were elevated in and around P-tunnel due to its extensive prior use in equipment deployed throughout the tunnel.

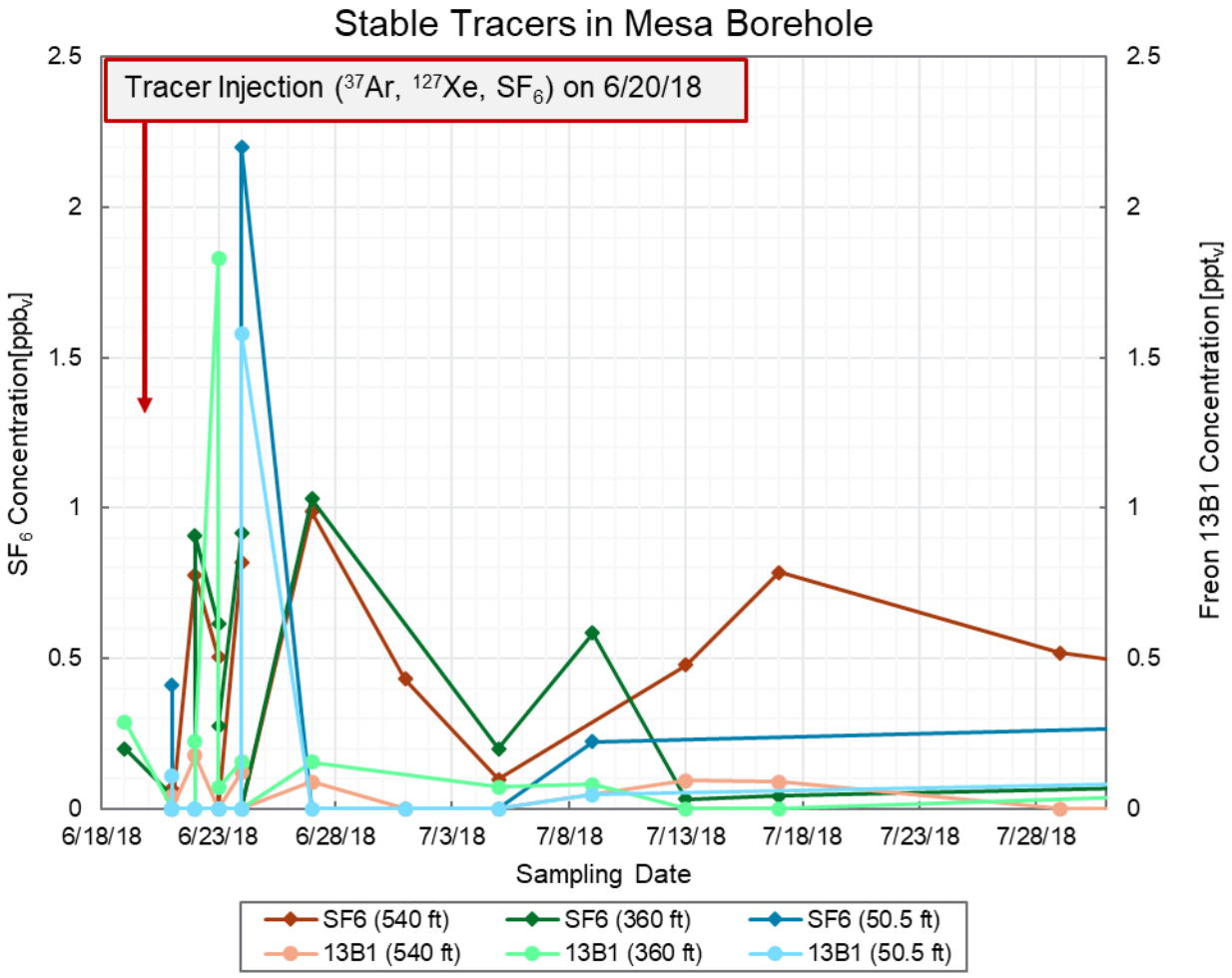

Figure 21. Evolution of SF_6 and Freon 13B1 in mesa borehole zones 2-4 for 40 days post-injection.

The longer-term evolution of the stable tracer concentrations in all levels of the mesa borehole is shown in Figure 22. Here the concentrations in Zones 2-4 appear to be nearly steady state, with small fluctuation likely due to perturbations caused by large volume sampling or significant weather events.

The significant increase in SF_6 concentrations in Zone 1 beginning November 2018 is also difficult to interpret. While the sharp increase in SF_6 concentration might indicate the arrival of the injected SF_6, the subsequent increase of Freon 13B1 (which was injected six months earlier) complicates the analysis. Since Zone 1 was submerged in water for several months post-injection, the question remains whether the arrival indicates the relaxation of the borehole back to background concentrations of the tracers or if the observed SF_6 arrival is truly the injected tracer. The lack of corresponding ^{127}Xe detections also complicates the analysis.

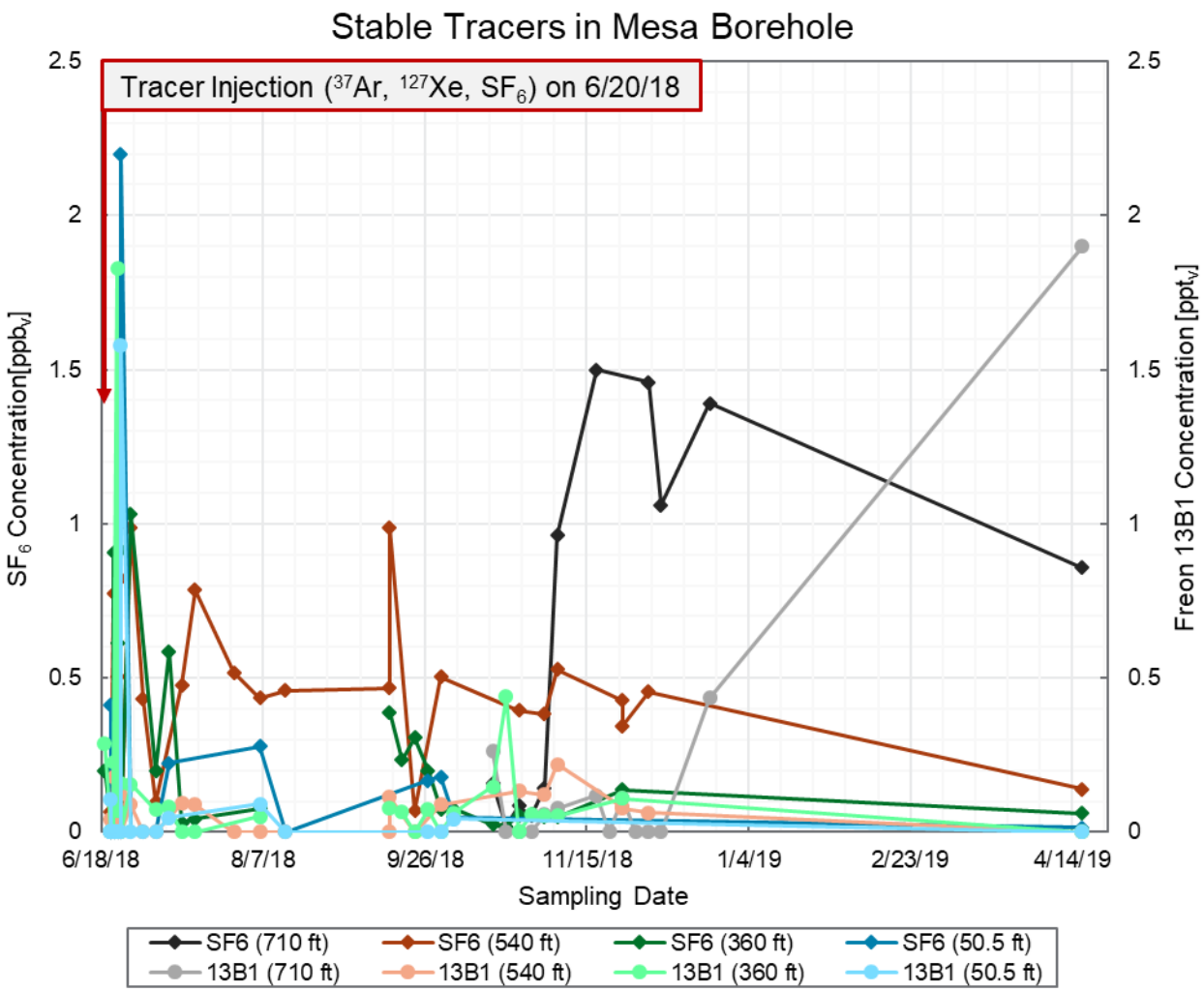

Figure 22. Evolution of SF_6 and Freon 13B1 in all mesa borehole zones from June 2018 through April 2019.

7.0 Conclusions

The injection of ^{37}Ar, ^{127}Xe, and SF_6 into the Disko Elm historic UNE chimney was successfully accomplished in June 2018. The introduction of a recirculating gas/detector system allowed for the observation of the evolution of ^{127}Xe in the chimney. This was a major improvement on previous gas migration experiments which relied upon point measurements of the tracer materials from points surrounding the injection.

While large volume samples were collected monthly from the mesa borehole for the six-month period following the tracer injection, and once more in April 2019, no positive detections were made of ^{37}Ar or ^{127}Xe in any of these samples. This indicates that natural transport drivers do not act quickly in the tuff of Aqueduct Mesa and that any gas transport post-UNE would be dominated by the initial pressure and temperature increases provided by the explosion. This is in contrast to the tuff of Rainier Mesa, where the UNESE Phase I noble gas migration experiment observed tracer transport 300 meters away from the injection point 5 months after injection and at the ground surface nine months after injection.

Future gas migration experiments should carefully consider the impact of borehole installation on the subsurface environment. The short period between completion of the surface borehole and the tracer injection introduced significant uncertainties that make interpreting the stable tracer data difficult. This highlights the need to allow for sufficient time in future experiments for the system to return to equilibrium between borehole completion and the commencement of sampling.

The presence of elevated ^{39}Ar levels in every sample collected from P-tunnel and Aqueduct Mesa has significant implications for nuclear explosion monitoring. First, elevated ^{39}Ar levels will negatively impact the MDC for ^{37}Ar measurements. While this impact is likely to be limited immediately post-UNE, at later times (on the order of 1-2 years) the ^{39}Ar signature will likely limit the current ability to detect ^{37}Ar. Additionally, these measurements highlight the potential to use ^{39}Ar as a long-term signature of UNE's. While the traditional radioactive noble gases of interest have relatively short half-lives, meaning that they will decay to non-detectable levels within a few years, ^{39}Ar has now been shown to be present in significantly elevated levels decades after a UNE.

8.0 References

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