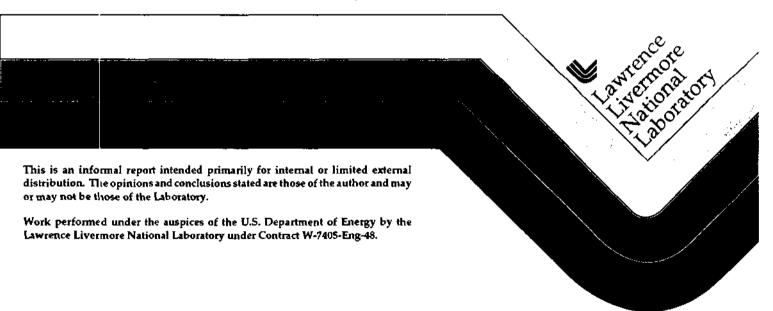


INIS-XA-N--327

# The NPE Gas Tracer Test and the Development of On-Site Inspection Techniques

Charles Carrigan, Ray Heinle, and J. J. Zucca

#### **April 13, 1995**



# The NPE Gas Tracer Test and the Development of On-Site Inspection Techniques

Charles Carrigan, Ray Heinle, and J. J. Zucca

April 13, 1995

# Summary

Tracer gases emplaced in or near the detonation cavity of the 1-kiloton Non-Proliferation Event required 1.5 and 13.5 months for sulfur hexaflouride and helium-3, respectively, to reach the surface of Rainier Mesa from an emplacement depth of 400 meters. The sites that first produced tracer gases are those located in known faults and fractures. Numerical modeling suggests that transport to the surface is accomplished within this time frame through atmospheric pumping along high permeability pathways such as fractures. The difference in travel time between the two tracers is due to differences in gas diffusivity and can also be explained by our numerical modeling.

### Introduction

Negotiations are being conducted in Geneva under the auspices of the Conference on Disarmament that could lead to a comprehensive test ban treaty. A future treaty will most likely contain a provision for on-site inspection (OSI) of ambiguous events. A key aspect of OSI will be to search for radioactive gases that are indicative of nuclear explosions. Of these gases, five are potential targets of collection during an OSI: Xenon-135 and -133, Argon-37, Krypton-85, and Tritium which have half lives of 9 hours, 5 days, 35 days, 11 years, and 13 years respectively. Argon-37<sup>1</sup> is the most attractive target since its half-life is long enough that it will still be detectable after several months, and has a small world-wide background.

The Nonproliferation Experiment (NPE) involved the underground detonation in Rainier Mesa at the Nevada Test Site of an ammonium nitrate and fuel oil blasting agent that released one kiloton of explosive energy. The primary purpose of this experiment was to ascertain the current capability of treaty verification technology to seismically discriminate between nuclear explosions and single-point chemical explosions. However, this experiment also provided the opportunity to carry out OSI related studies on an overburied and tamped event having many characteristics of the type of detonation that might be the subject of an OSI under a treaty. The chemical explosive was emplaced in a cavity that was connected to the outside by a system of tunnels. Therefore, the surface ground zero was not disturbed prior to detonation. Furthermore, after the explosion there was no evidence of surface deformation such as cracking and/or settling.

Argon-37 is a result of irradiating the calcium in a device and surrounding soil at the site of an underground nuclear explosion with neutrons produced by fission and/or fusion according to the reaction  $n + {}^{40}\text{Ca} - {}^{>41}\text{Ca} - {}^{>37}\text{Ar} + \alpha$ 

# **OSI Gas Sampling Experiment**

Since the NPE involved the detonation of chemical explosives, no radioactive gases were produced. To simulate the production of detectable gases, bottles of two different tracer gases, Helium-3 (3He) and sulfur hexaflouride (SF<sub>2</sub>), were placed in or near the explosive cavity. The bottles were crushed at the time of the explosion releasing the gases. <sup>3</sup>He is a stable isotope that is a good tracer because of its low background abundance, its inert nature and its chemical stability. The 'He bottle contained about 1300 liters (at standard temperature and pressure). Laboratory mass spectrographic techniques permit detection of 'He down to 0.1 ppt (parts per trillion). The volume of <sup>3</sup>He released in the NPE was about 5000 times greater than the amount of Argon-37 (<sup>17</sup>Ar) produced in a nuclear explosion of comparable yield. However, the greater volume of <sup>3</sup>He released in the NPE is offset by the much greater sensitivity of methods used in detecting <sup>37</sup>Ar. The SF<sub>x</sub> bottle contained about 115 lb. of tracer. This tracer is relatively inert and although it breaks down at high temperature, NPE containment studies suggested that most of the tracer would survive the explosion. It is detectable using gas chromatograph analyses also in the ppt range and analyses are less costly than for <sup>3</sup>He. To minimize the chance of thermal decomposition, the bottle of sulfur hexaflouride was placed immediately outside the detonation cavity against the bulkhead cover. Release of two different tracers having very different molecular weights allows the arrival times of tracers of intermediate molecular weight, such as 37Ar, to be bounded

#### **Experimental Results/Field Observations**

The timeline for gas sampling and NPE detonation is shown in Figure 1. We collected gas samples before the NPE detonation to establish background levels, which were on the order of a few ppt by volume for both tracer gases. Following the NPE detonation on September 22, 1993, gas samples were taken during periods of low barometric pressure—i.e. when soil gases are flowing out of the ground. We sampled gases at various times from a total of about 30 sites which consisted of metal tubes, called dry points, driven into faults and cracks as well as from tarped surface sites located away from cracks at radial locations from the NPE surface ground zero. The distribution of sites is plotted on the topographic contour map of Rainier Mesa shown in Figure 2. The first evidence for a tracer gas arrival is the 300 ppt SF<sub>6</sub> sample on November 10, 1993 at station OS-2 in the vicinity of the nearby Hunter's Trophy event. The sample was taken near a fault that dips down toward the NPE working point (Maggie-Baldwin, 1993, personal

Figure 1: Timeline for OSI Gas Tracer Experiment

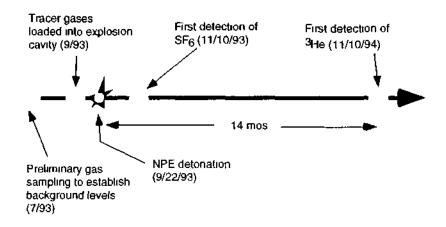
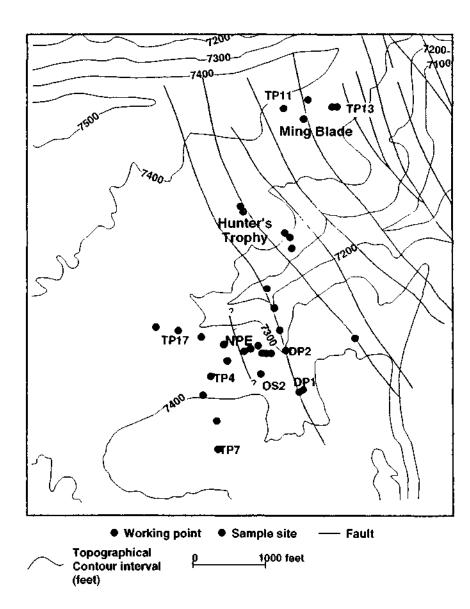


Figure 2: Map illustrating distribution of sampling sites on top of Rainier Mesa. Some sites are marked where tracers have been detected.



#### UNDERGROUND NUCLEAR EXPLOSIONS

#### GAS TRACERS AND ON-SITE INSPECTIONS

communication). Additional samples with high concentrations of SF<sub>6</sub> were obtained several months afterward in March 1994 at the same site and at other sites around the NPE.

Helium-3 was not detected until November 10, 1994 approximately one year following the initial detection of SF<sub>6</sub>. The site, DP-1, is located about 300 m southeast of the NPE surface ground zero. It contained approximately 21 ppt 'He which is about three times the natural background level. The site has also produced amounts of SF<sub>6</sub> equal to 15 times the background level. Two sites (TP-11 and TP-13) approximately one kilometer away have also produced 'He significantly above the background. However these sites are near the surface ground zero of an explosion that released a significant amount of 'He in 1983. Thus it is likely that we are detecting gases from this event. Unfortunately we do not have predetonation background levels of gas at this station.

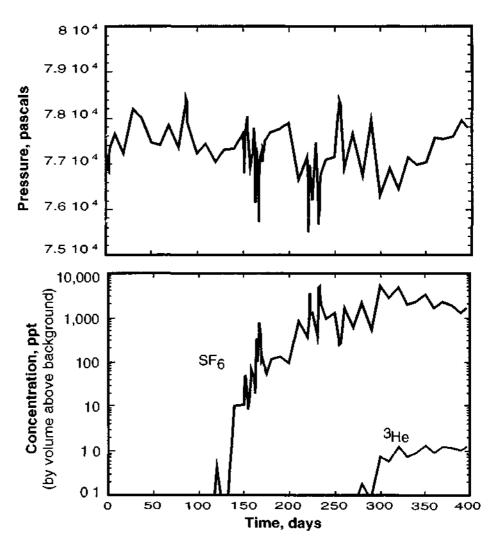
# Numerical Models of Gas Tracer Flow and the Effect of Barometric Pumping

We performed numerical simulations to determine the effect of barometric pumping on the surface arrival time of gases and also to determine the dependence of the tracer gas surface arrival time on the initial conditions such as concentration and pressure of cavity gases using the computer program NUFT (Nitao, 1993) developed at LLNL. We modeled the rock formation comprising Rainier Mesa as a geometrically idealized coupled, fracture/matrix system. We tested models with two different initial conditions: 1) the explosion cavity is undamaged by the explosion and pressure driven flow does not initially move the gases away from the cavity and 2) the explosion fractures the surrounding medium which allows gas to be driven under pressure approximately 200 m from the cavity walls. This second or "halo" model, assumes that trace nuclear gases are initially uniformly distributed within the pore space of the material surrounding the point of detonation and that the halo is penetrated by uniformly spaced fractures extending to the surface. We used values of the matrix porosity and permeability of 0.1 and 1 mD (milli Darcy, 1x10<sup>-15</sup> m<sup>2</sup>), respectively. For the penetrating fractures, the values of their thickness and spacing are assumed to be 0.001 m and 6.4 m. The values characterizing the matrix are appropriate for volcanic tuff formations at NTS and the fracture characteristics fall within the range considered in the study of Nilson et al. (1991) on similar materials at NTS.

Our modeling included the effect of barometric pumping caused by the weather systems passing over Ramier Mesa. This effect tends to cause increased flow along high permeability pathways such as fractures. We used the observed variations in barometric pressure for the top of Rainier Mesa, which were obtained through NOAA. Unlike earlier calculations that assume an idealized coupling between fracture flow and matrix diffusion, our modeling involved a full solution of the fracture and matrix flow and diffusion equations.

The "halo" model produced the closest fit to our observed data. It qualitatively predicts the time scale for arrival of the two tracers as well as the apparent earlier arrival of SF<sub>6</sub>. If barometric pumping is left out of the calculations, many years are required for diffusion alone to bring the gas to the surface. Figure 3 is a plot of both surface barometric pressure and the predicted concentrations (background removed) from our model of both <sup>3</sup>He and SF<sub>6</sub> measured near fractures at the surface as a function of time. The most significant feature is the difference in arrival times of the two tracer

Figure 3:
Result of numerical simulation illustrating surface pressure variation (observed data from Rainier Mesa) and predicted concentrations of both tracers as a function of time
Background values for SF and <sup>3</sup>He are 7 and 3 ppt respectively



gases predicted by the model. According to the model, it takes almost three times longer for the <sup>3</sup>He to reach the surface than the SF<sub>6</sub> which agrees qualitatively with our observations. This difference is apparently the result of the different molecular weights of the two tracers. The greater weight of the SF<sub>6</sub> tracer causes it to have a lower gas diffusivity than the <sup>3</sup>He tracer. When barometric pumping is included in the calculations, flow is mainly along fractures but diffusion of gas also occurs into the porous matrix of the fracture wall. Gases with higher diffusivity (i.e. lower molecular weight) diffuse at a greater rate into the walls of the fracture. Thus, the higher diffusivity gas is depleted from any upward fracture flow soon after the flow is initiated which in turn, delays its arrival at the surface. The molecular weight of the radionuclide gas of main interest to OSI, Argon-37, is bracketed by our results and should have intermediate arrivals time to <sup>3</sup>He and SF<sub>6</sub>

### Conclusions

To develop OSI gas sampling, analysis and modeling technology, we have taken advantage of the NPE detonation to simulate the transport of cavity gases from an overburied one-kt nuclear event to the surface of Rainier Mesa. The tracer gases that we emplaced in the NPE explosion cavity, <sup>3</sup>He and SF<sub>6</sub>, have been observed at the surface of Rainier Mesa with first arrival times of 1.5 and 13.5 months, respectively, from the NPE detonation time. Our computer simulations qualitatively agree with this observation and with the observation that gas is produced most rapidly by barometric pumping along high-permeability pathways, such as fractures and faults. Besides the development of new technology, the major impact of our work on OSI is the recognition that gas sampling within natural fractures and faults is likely to be far more effective for detecting gases from a very recent event than tarping large surface areas. The latter approach was used extensively in earlier work on underground explosions at sites where the cavity gases had many years, in some cases, to reach the surface. The new results indicate that cavity gases can be "fast tracked" to the surface along natural fractures so that detection under the appropriate barometric conditions is possible only a few months following a well contained event.

# Acknowledgments

This work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract number W-7405-ENG-48, in support of the Comprehensive Test-Ban Treaty Research and Development Program sponsored by the DOE Office of Non-Proliferation and National Security.

## References

Nitao, J.J. (1993), Reference Manual for the NUFT Flow and Transport Code, Version 1.0, *UCRL-ID-113520*, *Lawrence Livermore National Laboratory*, Livermore CA.

Nilson, R.H., Peterson, E.W., Lie, K.H., Burkhard, N.R. and others (1991), Atmospheric Pumping: A Mechanism Causing Vertical Transport of Contaminated Gases Through Fractured Permeable Media, J. Geophys. Res., 96, 21933-21948.