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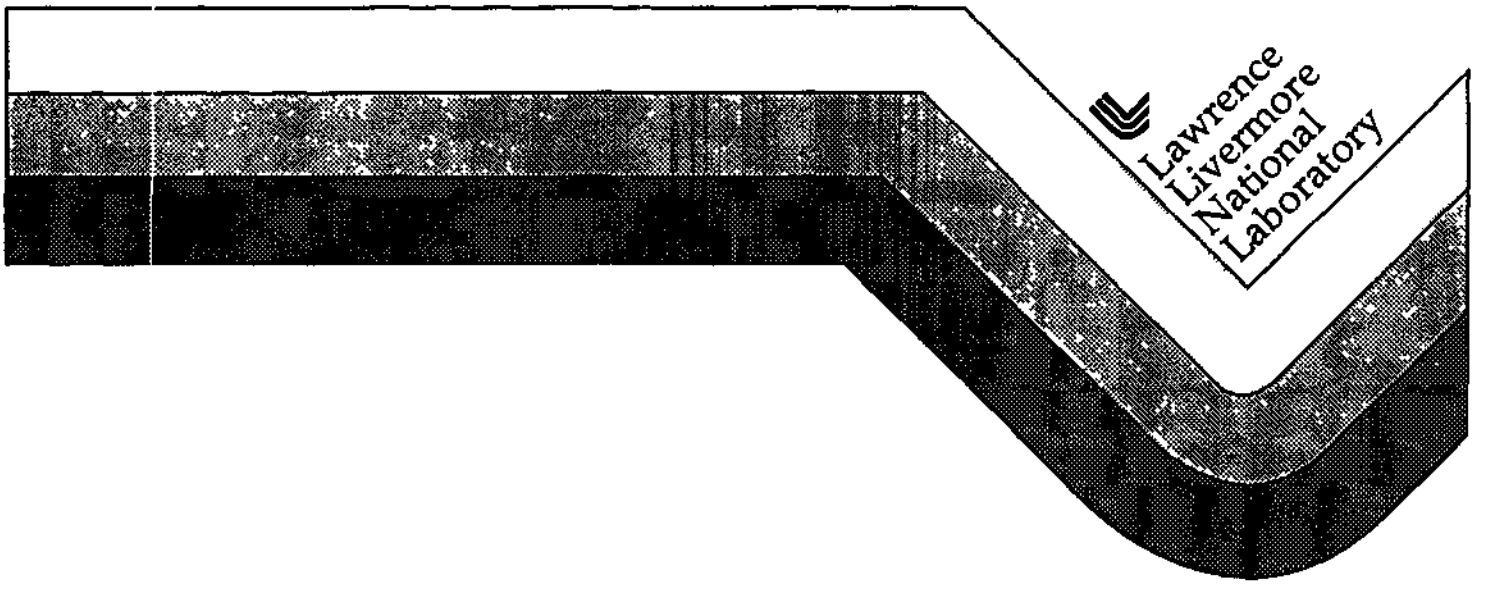

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Barometric Gas Transport Along Faults and Its Application to Nuclear Test-Ban Monitoring

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SUMMARY

Underground nuclear explosions produce a unique but evanescent set of radionuclide gases that potentially can be used in the context of an on-site, test-ban monitoring program to differentiate them from other detected events such as earthquakes or mining activity. In Part I of this report we describe an experiment to evaluate the upward transport of gases from an underground explosion using two gas tracers with very different diffusivities that were released in a 400-m-deep, chemical explosive detonation. The less diffusive (more massive) tracer was detected on a nearby geologic fault 50 days following the detonation while the more diffusive tracer was detected 375 days after release. Computer simulations indicate that the arrival time and the chromatographic behavior of transport are characteristic of barometrically induced flow in a fractured, porous matrix regime. For a hypothetical 1-kiloton fission explosion subject to the same weather and gas transport conditions of the chemical explosion, simulations predict the detectability of argon-37 after 80 days in spite of depletion by radioactive decay. Largely because of the earlier arrival of xenon-133, owing to its lower binary gas diffusivity, the exceedingly short lived isotope should also be detectable—arriving about 30 days earlier than argon.

In Part II we consider that our prediction of the detectability of argon and xenon is based upon the small volume (0.00001 m^3) sampling technique of the NPE tracer-gas sampling study while actual sampling for radionuclides would involve drawing much larger volume (possibly 0.1-1 m^3) gas samples from the near-surface.

Extraction of such a large volume of gas from 1-5 meter depths in the soil raises the possibility of significant atmospheric infiltration, leading to substantial dilution of the extracted gas sample. However, an infiltration experiment suggests that significant dilution would not, in fact, occur at the most prolific sampling stations of the earlier gas-tracer study. Of the soil gas being extracted at the shallowest sample site, less than 10% of the gas in the sample volume can be attributed to the infiltration of atmospheric gas.

PART I: NPE GAS TRACER EXPERIMENT

Introduction

The DOE Comprehensive Test Ban Treaty (CTBT) Research program was created to further knowledge about technologies that could be used for on-site inspections and confidence-building measures that may be called for under the provisions of the CTBT, which was signed by President Clinton on September 25, 1996. The philosophy of the LLNL On-Site Inspection (OSI) project was to investigate technology areas which showed promise to be of great value for an OSI or confidence building, but were insufficiently developed. Four technologies, three of which apply to OSI and one which applies to confidence building, were identified for funding during FY95 and FY96. The OSI technologies are aftershock monitoring, noble gas transport modeling and sampling, and remote sensing of disturbed ground. This report covers the results of the trace-radionuclide detection study carried out as part of the Non-Proliferation Experiment (NPE) involving a one-kiloton chemical explosion within Rainier Mesa at the Nevada Test Site.

Temporal changes in subsurface gas concentrations in zones of seismic activity have previously been studied as precursors of earthquakes (1) while spatial

variations of soil gases have been used for mineral exploration (2). For monitoring compliance with a nuclear test-ban treaty, the ability to detect the localized emanations of short lived, fission-produced radionuclides from the subsurface is an important diagnostic for international, on-site, challenge inspections of the suspected detonation sites of treaty signatories (3,4). Under the protocol of a treaty, the detection of such radionuclides may be a basis for declaring a violation. The primary purpose of the experiment described here was to determine if a deep underground explosion could be detected by gas sampling along nearby existing geologic faults. In the event of detection, another goal of the experiment was to provide a basis for predicting the detectability of rapidly decaying, fission-produced radionuclides that can be linked unambiguously to a nuclear detonation.

The Barometric Pumping Model

It is well known that binary gas diffusion is much too slow to move detectable concentrations of short half-life (^{37}Ar: 34.8 days, ^{133}Xe: 5.2 days) radionuclides from depths of hundreds of meters or kilometers to the surface. Even flows induced by a long term pressurized (3 kPa or 30 mb) subsurface source are surprisingly ineffective for pushing gases to the surface because of the typically anisotropic permeability of geologic layering that results in mostly horizontal gas flow from the source (5). On the other hand, an extended 3 kPa decrease in the pressure at the surface, which might be associated with a large storm, can be highly effective for drawing gas toward the surface along a fracture system. But the connected porosity of the fracture walls, allowing diffusive and convective exchanges between the porous matrix of the host regime and the fracture, makes the upward transport of a trace gas more complicated than a simple channel-flow model would predict.

The simplified numerical model of Rainier Mesa, which is supported by the results of gas-tracer sampling, is a uniform porous medium traversed by a narrow,

millimeter-width fracture. Gas flows from the detonation point via this fracture toward the surface during periods when the barometric pressure falls below the ambient gas pressure in the formation. As gas flows upward in the fracture, it also has the opportunity to diffuse into the surrounding rock matrix. Thus, the concentration of a tracer will typically decrease during upward transport as a result of diffusional losses into the the porous walls. The higher the diffusivity of the tracer, the more rapidly that it will be lost into the pore space of the fracture walls. When the barometric pressure increases to higher values than the surrounding soil-gas pressure, gas is pushed downward along fractures as well as into the porous walls. Tracer gas is retained in the walls of the fracture until the next period of low pressure. At this time tracer-bearing gas from the wall is freed into the fracture and continues toward the surface. The presence of diffusion prevents the transport from being totally reversible. As a result there is a net "ratcheting" of the tracer concentration front upward even if pressure variations at the surface are perfectly sinusoidal. The efficiency of induced upward transport of a given tracer or radionuclide depends not only on the amplitude of a pressure drop but also on its frequency. In our model for Rainier Mesa, short term barometric fluctuations (tens of minutes to hours) tap the shallow, gas-filled porosity of the fracture walls more effectively than the deeper regime adjacent to the explosion cavity. Longer term (12 hours to days) pressure drops of the same amplitude can draw gas *directly* from deeper wall porosity or the explosion source. Then the upward transport is limited entirely by the diffusivity of the tracer. A tracer that has a low diffusivity, such as sulfur hexaflouride, will travel much further vertically during the period of a given barometric low. If a tracer's diffusivity is low enough and the descending period of the barometric low is long enough, tracer or bomb products can reach the surface on the first barometic event. The sulfur hexaflouride (SF_6) tracer has a low diffusivity and may reach the surface during a big barometric low. For high diffusivity tracers

or isotopes and shorter barometric fluctuations, a finite concentration will still reach the surface by the *less* direct "ratcheting" process (6,4). The ^{3}He tracer used in this experiment ultimately reached the surface by this means. Radioisotopes of interest tend to fall between these two tracers in terms of the rapidity with which they reach the surface.

A variety of numerical models have been evaluated and show the clear dominance of the ratcheting process over upward transport by pure gaseous diffusion alone. Uniform pressure fluctuations of ± 5 mbar having a 20 hour period moved an SF_6 concentration front about 100 m in 20 days (4). At the surface, the concentration of the tracer or radionuclide oscillates in time having a growing amplitude with each repetition of the ratcheting cycle. This model implies that the long term lows associated with large storms are not necessary to get a tracer gas to the surface. On the other hand, incremental gas transport by ratcheting alone is almost certainly inadequate to get very short-lived radionuclides to the surface in measurable quantities.

The Experiment

Sited at a depth of 400 m in the bedded tuffs (7) of Rainier Mesa (Fig. 1a) at the Nevada Test Site (NTS), a seismic experiment called the Non-Proliferation Experiment (NPE), involving a one-kiloton chemical explosion provided a unique opportunity for simulating the release and transport of gases from a nuclear event that would be either exceptionally difficult or impossible to detect by observations limited to surface changes (8). Approximately 8 m^3 of sulfur hexaflouride (SF_6), a relatively inert, high molecular weight (146) compound having good thermal stability (9), was released by the detonation. Gas chromatography was used to detect SF_6 in air samples which have a low atmospheric background level, less than 3 parts-per-trillion (ppt by volume), at the test site. A 1.3 m^3 volume of ^{3}He was also

released into the cavity by the NPE explosion. The low atmospheric background level of ^3He (7.34 ppt) also permits a very low threshold for detection using mass spectrometry.

Approximately 200 gas samples were analyzed over a 508 day period during which a wide variety of barometric events were recorded by a NOAA weather station on Rainier Mesa (elev. 2286 m). Most of the sampling occurred in the fall and spring, when large barometric depressions occur, although a few negative measurements on mild summer days were also obtained. The presence of deep snow during the late fall, winter and early spring made the mesa inaccessible. On the surface near the detonation point (Fig. 1b), typically three or more stations were sampled during a barometric event (see Table 1). Stainless steel tubes with porous end caps were driven into the ground to depths between 1.5 and 5 m along fractures and faults at a number of sample points. The ground was often tarped around these points to limit atmospheric infiltration. At other locations, the ground was tarped for trapping soil gas that reached the surface. The tubes driven into fractures and fault zones were found to be far more effective for detection of the tracers than simply the random tarping of the surface over the detonation. A tracer was detected only once at a site where gas was withdrawn from under a tarp and not from a tube in the ground (Table 1, Suite 18). This is consistent with the model of fractures as providing a "fast track" to the surface for trace gases.

Results of Gas Tracer Experiment

Following several large barometric depressions during the fall of 1993 (Fig. 2a), sulfur hexaflouride was first detected in concentrations of 280 - 600 ppt in fractures along a fault several hundred meters from surface ground zero (Fig 1b) only 50 days after the detonation (Fig. 2b). The breakthrough of SF_6, measured at several ppt over background, evidently depended little on higher frequency, low amplitude ratcheting during the 50 days. According to our numerical model of the

Rainier Mesa fracture-matrix system, even if the gas had been released 83 days before the actual release time and there had been only small amplitude pressure variations, the gas would not have arrived much earlier. This implies that several large barometric events that occurred prior to arrival were primarily responsible for moving gas from the source region to the surface. Indeed, the numerical model for SF_6 arrival predicts concentration increases of several orders of magnitude, to detectable levels, during some large barometric lows that occurred just after the detonation in the late fall of 1993.

After its arrival, SF_6 was found to be well above background a number of times in later sample suites as indicated by the "detect" symbols on the timeline (Fig. 2b). However, it is also evident that SF_6 was not detected in other sample suites. The evanescent behavior of the tracer as indicated by the "no detects" may be the result of near-surface dispersion into a porous and highly fractured surface regime that introduces additional sampling variability through winds, atmospheric infiltration and rain (10). Our models do not include such a time-dependent dispersive interaction between the atmosphere and the weathered near-surface layer and, thus, probably overestimate the concentrations near the surface.

Helium-3 was initially detected at levels exceeding two standard deviations (taken conservatively to be 1 ppt in excess of the 7.34 ppt atmospheric level for ^3He) 325 days following the first detection of SF_6 and 375 days after the detonation. Several weeks following detection, a sample was obtained on the fault passing near the cavity (Fig. 1a) with the maximum observed concentration of about 22 ppt. The numerical models for arrival of both tracers (Fig. 2c and 2e) appear to be in excellent agreement with the apparent arrivals. Both models are the same except for differences in diffusivity and initial concentration as determined by the amount of tracer placed in the cavity. Surprisingly, the most likely explanation for the very large delay between the arrivals of the two tracers is the 8-fold larger binary gas

diffusivity of ^3He compared to SF_6 (11). In this fracture-matrix regime, the most diffusive gas arrives last at the surface according to our models. This behavior can also be inferred from analytical models of barometric pumping efficiency (9). Such a result may seem counterintuitive until one recognizes that the primary means of transport to the surface is advection by fracture flow and not diffusion. The rapid transverse diffusion of gas from a fracture into the matrix means that gas leaves the flow before it has moved very far vertically into a regime of lower concentration. Indeed, substantially reducing matrix porosity prevents the loss of helium from fracture flow. In this case, the models predict that the two tracers will arrive at about the same time a few weeks after the detonation.

Prediction of ^{37}Ar and ^{133}Xe Arrival from Analogous One-Kiloton Nuclear Test

Given the very good agreement between our gas-transport model and the observations, we use it to predict the arrival of detectable concentrations of ^{37}Ar and ^{133}Xe assuming a 1-kt fission detonation. Argon-37 and ^{133}Xe are preferred for the detection of nuclear explosions for two reasons: (1) they are not produced naturally in significant quantities so that natural background levels are exceedingly low, and (2) their short half-lives of 34.8 days and 5.3 days, respectively, can be used to infer the recentness of an event. The model specific to argon included the concentration of ^{37}Ar produced by fission-neutron bombardment of ^{40}Ca in the soil, the diffusivity of argon (between that of ^3He and SF_6), solubility of argon in groundwater, and its half life (12). The model predicts that ^{37}Ar would have been detectable approximately 80 days following detonation (13) at a concentration exceeding 10^{-18} moles of radionuclide per mole of air, or 6 Bq/m^3 (one becquerel (Bq) equals one decay-per-second) in this particular case. However, the decay of ^{37}Ar to ^{37}Cl will limit the sampling "window" during which surface detection is possible. Using an analogous model for ^{133}Xe, we found that its rather short half life does not prevent

it from reaching the surface in detectable quantities and predict concentrations at the surface exceeding 10^{-18} moles of xenon/mole of air (41 Bq/m^3 for ^{133}Xe) about 50 days after detonation (14). The much larger initial quantity of ^{133}Xe produced relative to that of ^{37}Ar combined with its lower diffusivity, which is comparable to that of SF_6 causing it to be transported more rapidly than ^{37}Ar to the surface, offsets its loss by rapid decay.

Estimates of the temporal sampling "windows" for both ^{37}Ar and ^{133}Xe are shown in Fig. 3 for the hypothesized one-kiloton nuclear detonation. They are based upon the predicted concentrations at the earliest arrivals of the radionuclides. These concentrations are then subject to decay as determined by the half lives of the gases. For this case, Fig. 3 suggests how wide the window is for the occurrance of barometric lows that are responsible for bringing radionuclides to the surface in detectable quantities. For argon the window is about 200 days and for xenon the width is about 150 days. These estimates assume the same size barometric lows as these responsible for the arrivals at 50 and 80 days after the detonation.

The predictions of the concentrations of ^{37}Ar and ^{133}Xe at their respective arrival times are likely to be conservative for two reasons. In our radionuclide transport simulations, we have assumed production rates that are low for both radionuclides: 7×10^{-5} moles (9.7×10^{12} Bq) of ^{37}Ar per kiloton, which is a quantity less than half of that produced by an explosion that is characterized by an intentionally high attenuation of fission neutrons (15) needed for ^{37}Ar production, and 7.7×10^{-3} moles (6.7×10^{15} Bq) of ^{133}Xe per kiloton, corresponding to 50% of the fission chain yield of this isotope. Further, our predictive models for radionuclide transport are calibrated using the observed arrivals of the SF_6 and ^{3}He tracers rather than their actual arrivals. The observed arrivals of the two tracers can occur later than their actual arrivals owing to imperfect or sparse sampling. As a result, our

models will tend to underestimate transport rates of trace gases through the overburden.

We conclude that even deep and very well-contained underground nuclear explosions, which do not propagate fractures to the surface, are liable to detection by the barometrically driven emission of short-lived radionuclides from nearby surface-breaking faults, natural and mining-induced fracture networks, and cooling joints, which tend to be found (16) in subsurface environments common to mining districts. Deep barometric lows associated with large storm fronts are needed for rapidly transporting the higher atomic mass gases, which include the radionuclides of interest, to the surface. Thus, selecting the timing of a challenge inspection to include the arrival of weather fronts may be necessary to optimize the possibility of detection. Owing to the heterogeneity of pathways for gas transport, we found that it was a better strategy to locate sampling stations on existing faults and fractures, even at distances of hundreds of meters from surface ground zero, than to select a sampling station using proximity considerations alone.

PART II: ATMOSPHERIC INFILTRATION EXPERIMENT

Introduction

In many respects the site location and sampling techniques employed at sites on Rainier Mesa during the 18 months following the NPE detonation are the same as those that would probably be used during an on-site inspection in some other part of the world. The most important difference is the sample volume of gases extracted from tubes driven short distances (1-5 m) into the surface soil by hand or with the aid of a truck-mounted hydraulic ram. For the NPE, the samples obtained to determine the presence of SF_6 and ^3He varied in size from 10-50 cubic centimeters (1 to 5 x 10^{-5} cubic meters). For an on-site inspection using ^{133}Xe and ^{37}Ar radionuclide counters, gas samples of from 0.1-1 cubic meters may be required to obtain adequate sensitivity. Drawing such large volumes from tubes that penetrate at most several meters below the surface raises the possibility that significant atmospheric infiltration will occur. This is because the air removed from the soil must be replaced by air from elsewhere, and the atmosphere is the nearest source. The main effect of including significant amounts of air with the sample soil gases is a potentially significant decrease in the concentration of radionuclides in the sample.

Computer Simulations of Atmospheric Infiltration During Near-Surface Sampling

Computer simulations of the effect of infiltration on soil gas concentrations were performed using NUFT, the porous flow program used for the gas transport simulations described above. Infiltration in both a fractured and uniform porous medium was studied (Fig. 4). The effect of

infiltration in a fractured environment is illustrated by the plot in Figure 4. The fracture readily channels gas from the atmosphere downward (left cross-section, Fig. 4) to the inlet of the tube which is placed in the fracture. Compared to a sample taken at the same depth, but in a homogeneous porous environment (right cross-section, Fig. 4), the fractured environment permits a four-fold dilution of the concentration of the tracer. Emplacing the tube more deeply, 7.1 m instead of 2.1 m, in the fracture partially mitigates the effect of atmospheric dilution. Such models are suggestive of how dilution of a tracer or radionuclide signal might be affected by the state of the near-surface environment. In the uniform porosity limit, atmospheric dilution never plays a significant role. In the limit of withdrawing gas from a fracture, atmospheric dilution can easily reduce a radionuclide signal below the detection threshold.

Atmospheric Infiltration Field Experiment

To estimate the effect of infiltration at sites that are similar to those sampled during an on-site inspection, a series of field experiments were conducted at two sampling stations on Rainier Mesa used during the NPE gas tracer experiment described in Part I. The stations selected produced significant tracer signals during the course of the gas-transport experiment. These sites were used because they are assumed to be associated with open-fracture systems that are thought to provide pathways from both the source and the atmosphere to the sample-tube inlet. Site DP-1 (Fig. 1b) was emplaced in a fault using a truck-mounted hydraulic ram. The tube was inserted into

the root system of a pinion tree to a depth of about 5 meters. While this sample point is emplaced within a fault zone that cuts fractured and jointed tuff formations, no nearby surface breaking fractures were found. Site OS-3 is located approximately 200 m from DP-1. It is probably not in a fault zone but is associated with an old surface fissure that has been filled in. Evidence of a deeper, open network is suggested by rodent holes that appear in the vicinity of OS-3. The sample tube, which is shallower than that of DP-1, penetrates the dirt-filled fissure to a depth of about 1.5 meters.

The first experiment used dilution of radon gas as an indicator of the amount of infiltration. Radon-222 is a noble gas that is produced more-or-less uniformly by the decay of uranium, which occurs naturally in the soil. Radon counters supplied by the NTS contractor, Bechtel, were connected to the tubes at the sample sites. The counters draw in gas using a small pump with a capacity of about 1 liter/min which is comparable to the flow rate assumed in the computer models. The concentration of radon in the detector is actually inferred from counting the decay of the daughter product of radon, polonium-218. Daughter isotopes polonium-214 and -216 were also counted. Both sample points were characterized by different levels of the radon and its daughters entering the detector, but over the counting period the concentration of radon or its daughters did not appear to change significantly. This result suggests that even in an identified fracture regime, if surface fractures are not apparent, infiltration may not be a significant effect. The constancy of the near-surface gas concentrations were quite similar to that of the homogeneous model described in Fig. 4.

Finally, the surface around the sample points was tarped with plastic sheeting and the edges of the tarps were covered with soil. A bottle containing

an isotope of xenon gas was released under the tarps. Again, soil gases were pumped from the ground into the detectors. In this experiment, which is far more sensitive for detecting atmospheric infiltration than the radon experiments, any infiltration of the atmosphere would be indicated as an increase in the xenon concentration. At DP-1, with its deeper sample tube (5 m depth), insignificant increases in xenon were detected during the 0.1 cubic meter extraction test. Slightly higher levels of xenon were detected at the OS-3 site with its shallower sample tube (approximately 1 m depth). Given the differences in the depth of emplacement of the sample tubes, it is not surprising that infiltration was clearly more at the OS-3 site. But the very modest increase in xenon levels suggests that, of the gases captured in a 0.1 cubic meter sample, less than 10% of the sample volume is attributable to atmospheric gas drawn into the soil during the extraction period.

This preliminary infiltration experiment suggests minimal to nonexistent atmospheric infiltration for 0.1 cubic meter samples drawn at a rate of one-liter-per-minute from sample tubes at two of the most productive sampling sites that were emplaced at depths between 1-5 meters. The effect of much higher pumping rates was not studied in the field experiments because the radon-counter pumping rate could not readily be changed, but numerical modeling was used to estimate the behavior of increased withdrawal rate in the model of sampling at 2.1 meter depth in a fracture. For a four-fold increase in pumping rate, the high extraction rate model predicts instantaneous tracer mass flux values that are very slightly larger than for the lower extraction rate case (see Fig. 4). However, the total extraction rate, that is, total air flow, is much larger which means that the actual tracer or radionuclide gas concentration decreases for the case involving high-extraction rate from a fracture. Increasing the extraction rate by the same

amount for the uniform permeability case yields a concentration that is more or less unchanged during acquisition of the 0.1 cubic meter sample. While higher extraction rates may be preferable from the perspective of the amount of time required to obtain a sample, we conclude that significant sample dilution may occur in cases where a sample tube is drawing gases from a fracture network that is connected to the atmosphere. The amount of dilution will depend on the characteristics of each sample site. The two sites used in the radon and xenon field studies (DP-1, OS-3) appear to be more like the uniform porosity case of Fig. 4. Thus, samples produced at these sites would be less affected by increasing extraction rates beyond one-liter/minute.

RECOMMENDATIONS FOR SAMPLING AND CTBT PROTOCOL BASED ON EXPERIMENTS OF PARTS I & II

Using the experiments described here and in a paper published in the open literature (18), several recommendations about possible on-site inspections may be made.

1. Following a suspected detonation, the prompt (hours to days) arrival of an OSI team may not be necessary to insure detection since some time may be required for gases to reach the surface. However, the very early arrival of a sampling team may permit pressurized gas seeps to be detected in some cases.

2. Given the detection sensitivities for argon and xenon isotopes assumed in this report, the occupation of a suspected test site may be potentially productive for 100-200 days. Of course these times depend on the initial amount of argon and xenon produced. A number of factors, including

detonation size, play a role in determining the initial quantities of these isotopes. The times given here assume a conservative production of xenon and argon from a modest 1-kt detonation.

3. Gas sampling at the site of a suspected underground nuclear test should coincide with the arrival of barometric lows. Sampling during periods of high pressure is likely to be a wasted exercise. In fact, gas sampling during the period immediately following a pressure minimum, that is, when the surface pressure is low but is increasing, usually did not result in any detects.

4. The siting of surface sampling stations should not be based on the proximity to the anticipated surface ground zero alone. Sites with fractures hundreds of meters from surface ground zero may be preferable for detecting bomb products. Fracture networks serve as a "fast track" for the flow of gas to the surface. The sampling of fractures, even those hundreds of meters from surface ground zero, yielded a number of detects. On the other hand, randomly tarping near surface ground zero was mostly unsuccessful for producing detects.

5. Sampling tubes should be emplaced as deeply as practicable into fault zones or fissures to eliminate infiltration from the atmosphere. Fissures that are open to the air should be covered with plastic tarps whose edges are covered with sand or dirt to eliminate infiltration. To reduce infiltration as much as possible, tarps should be used at all sites even if the fractures are already soil filled.

6. In cases involving very high permeability pathways to the surface, very high extraction rates (tens of liters/min) may cause significant amounts of air to infiltrate and dilute the soil-gas sample.

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12. Regarding the pertinent differences in chemical and nuclear source functions, the amount of gas produced per kiloton in a detonation is estimated to be 1.5×10^4 m^3 for a nuclear event and 3.6×10^5 m^3 for a chemical event. A conservative estimate of ^{37}Ar production by neutron bombardment of ^{40}Ca per kiloton is 7×10^{-5} moles. Argon's intermediate value of diffusivity is $D(^{37}Ar)=2.0 \times 10^{-5}$ m^2/s.

13. Arrival concentrations of 1×10^{-18} after 80 days, including decay effects, require quite achievable sensitivity thresholds that are somewhat higher than those attained in above-ground laboratories according to

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14. For ^{133}Xe, 7.7×10^{-3} moles/kt are estimated. Diffusivity is 1.24×10^{-5} m^2/s making it comparable to SF_6 in transport behavior. Detection sensitivities are assumed to approximate those for ^{37}Ar. At about 50 days, the time of predicted detection in this case, the total amounts of each radionuclide left in the fracture-matrix system are comparable even though much more xenon was produced initially. If the presence of groundwater is taken into account, the delaying effect on transport owing to the large solubility of xenon in water appears to be more than offset by an increase in the tortuosity of the connected pore space, which reduces the effective binary diffusivity, so that xenon actually arrives at the surface sooner in nearly saturated environments than it does in dry ones.

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CAPTIONS

FIG. 1a Cross section of layered tuffs of Rainier Mesa at the Nevada Test Site showing the location of the cavity relative to the surface and surrounding faults including the main fault on which the most prolific sample stations, DP-1 and OS-1 (Fig. 1b), are situated. These stations are several hundred meters from the point on the fault which is nearest to surface ground zero. Figure adapted from ref. 5.

FIG. 1b Planview of the distribution of sample points relative to existing faults and surface ground zero of the detonation which has become known as the Non-Proliferation Experiment (NPE). The most prolific site DP-1 is located on the fault that is thought to pass close to the detonation cavity. This station has produced maximum values of ^3He and the largest values of SF_6 during the final sampling period (fall 1994). The station was emplaced only at the beginning of this period. Large SF_6 values, in the 500-600 pptv range, were obtained at OS-1 which is only a few meters from DP-1 and is in the fault zone. OS-1 was the first station to be emplaced. High values were also obtained along a fault about 550 m (1.3 cavity depths) north of the NPE surface ground zero. This site (OS-6), near an underground nuclear test called Hunter's Trophy, has produced the earliest detections of both tracers with the other sites, closer to the NPE, yielding detections shortly thereafter. [On the date ^3He was first detected, a site on the fault near OS-1 produced a concentration for ^3He that fell just below the 1 pptv excess required to declare a detection.] A site marked "TP-4" is one of about 15 stations surrounding the NPE surface ground zero where a plastic tarpaulin was laid out on the soil to trap gas as it reached the surface (all other unlabelled points are such tarped

stations). Of all measurements from these tarped sample points, only TP-4 once yielded an SF_6 concentration above background. All sites labeled "OS" (Original Site) and "DP" (Drive Point) are located on pre-existing faults or fissures. The former have sample tubes that were driven to 1-2 m depths manually while the latter have sample tubes or drive points installed to depths of 5 m using a truck mounted hydraulic ram.

FIG. 2 Sampling histories (b) and (d) for both tracers along with models of near-surface-concentration histories (c) and (e) are plotted in conjunction with the history of the driving barometric pressure variations (a). The horizontal line indicates the average pressure, 773 millibars, over the sampling period. In (b) and (d) the suite of samples taken during a given barometric low is summarized either as a "detect" (at least one sample of a suite above background - ) or a "no detect" event () with the numbers beneath the symbols corresponding to the sample suite numbers of TABLE 1. Numerical models were used to produce plots (c) and (e) of the tracer concentrations in excess of background values in parts-per-trillion-by-volume at standard temperature and pressure (pptv) as functions of time. For reference, the thresholds of argon and xenon detection are given by the horizontal lines in plots. The concentration of SF_6 is plotted on a log-linear plot because of the wide variation in values. The model exhibits a large change in concentration at the surface at a time corresponding to observations of the apparent arrival of SF_6 (b). For ^3He, the apparent arrival as interpreted from the observations (d) is also in excellent agreement with the linear plot of concentration as a function of time (e) as indicated by the model. [Note: two null events () immediately precede the arrival () although on the scale of the diagram they appear as one null event.] Hydrologic parameters used in

the models, such as gas-phase porosity (0.10), permeability (10^{-15} m^2), fracture separation (6.4 m) and tortuosity (0.1), are thought to be appropriate in a bulk sense for the fractured tuffs of Rainer Mesa and nearby Yucca Mountain with its well-studied hydrologic system. An integrated finite difference, multi-phase, flow and transport computer program(17) performs the gas transport simulations on a 2100 element, 6.4 m wide by 400 m deep biased grid using the hydrologic parameters as input. The same fracture-matrix model is used for both the SF_6 and ^3He concentration histories; only the diffusivities and initial concentrations are different. The initial concentration distribution of SF_6 and ^3He occupies a region in the fractured porous medium extending upward to 200 m beneath the surface of the mesa. This fracture model is consistent with models used in detonation containment studies [8,9]. Parameter sensitivity studies show that large changes in model parameters such as fracture width (0.001 - 0.004 m, 0.00125 m used), fracture separation (3.3 - 12.8 m, 6.4 m used), and initial halo concentration of SF_6 ($3.0x10^{-7}$ to $2.2x10^{-5}$, $2.2x10^{-5}$ used) all resulted in changes to the arrival time of 13 days or less for SF_6.

FIG. 3 Illustrated is an estimate of the signal amplitude of both xenon and argon as a function of time. This rough estimate is based on the predicted strength of the signals at their originally predicted arrival times, 50 days for xenon and 80 days for argon, assuming the barometric pressure variations and soil transport conditions appropriate to the model of Rainier Mesa. Had the actual barometric lows responsible for the arrivals been delayed, causing the gases to arrive after their originally predicted arrivals, we can estimate the signal strength of the gas arrivals as a function of time from the graph. Considering the indicated detection limits for both gases, we would expect that it should be possible to detect the xenon signature out to about 175 days

following detonation and the argon signature out to 225 days. Because of the shallower slope of the argon decay line, improvements in the detection limit will have a greater effect on extending the "window" of time during which radionuclide signatures can be detected.

FIG. 4 Computer simulations of soil-gas extraction are illustrated for several different near-surface, gas-flow environments. A sample tube (left graphic) extracts from a 2.1 meter depth in an narrow fracture (0.00125 m). Extracting only 0.1 cubic meters causes significant infiltration along the fracture as shown in the figure. The plot of concentration versus time shows that infiltration of atmosphere along the fracture (curve labelled 2.1 m depth-fracture) dilutes the concentration of tracer by a factor of four compared to extraction from a tube emplaced in a uniform porous medium at the same depth (right graphic). Increasing the depth of the sampling tube in the fracture (7.1 m) increases the tracer concentration (decreases dilution) in the sample only slightly.

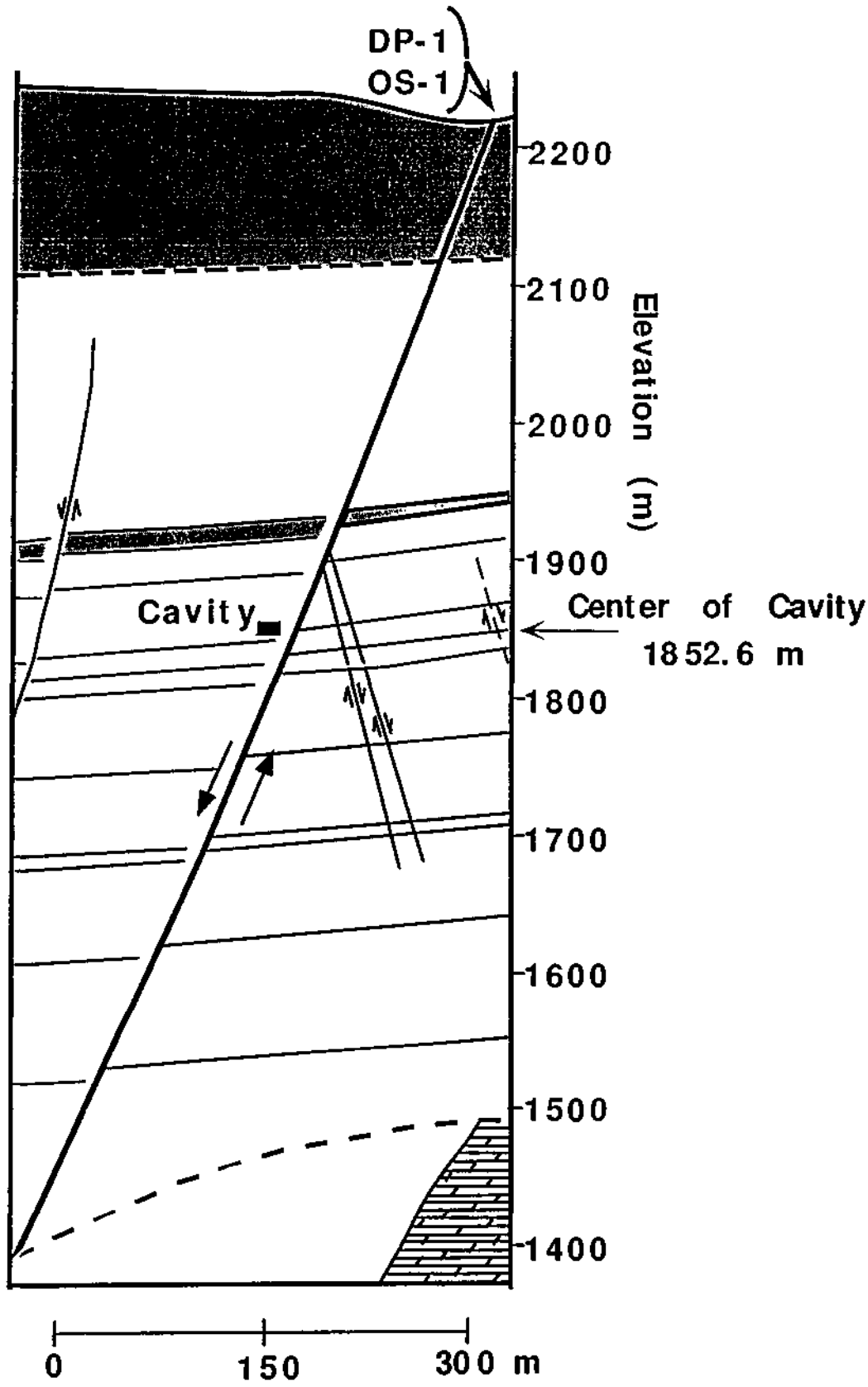
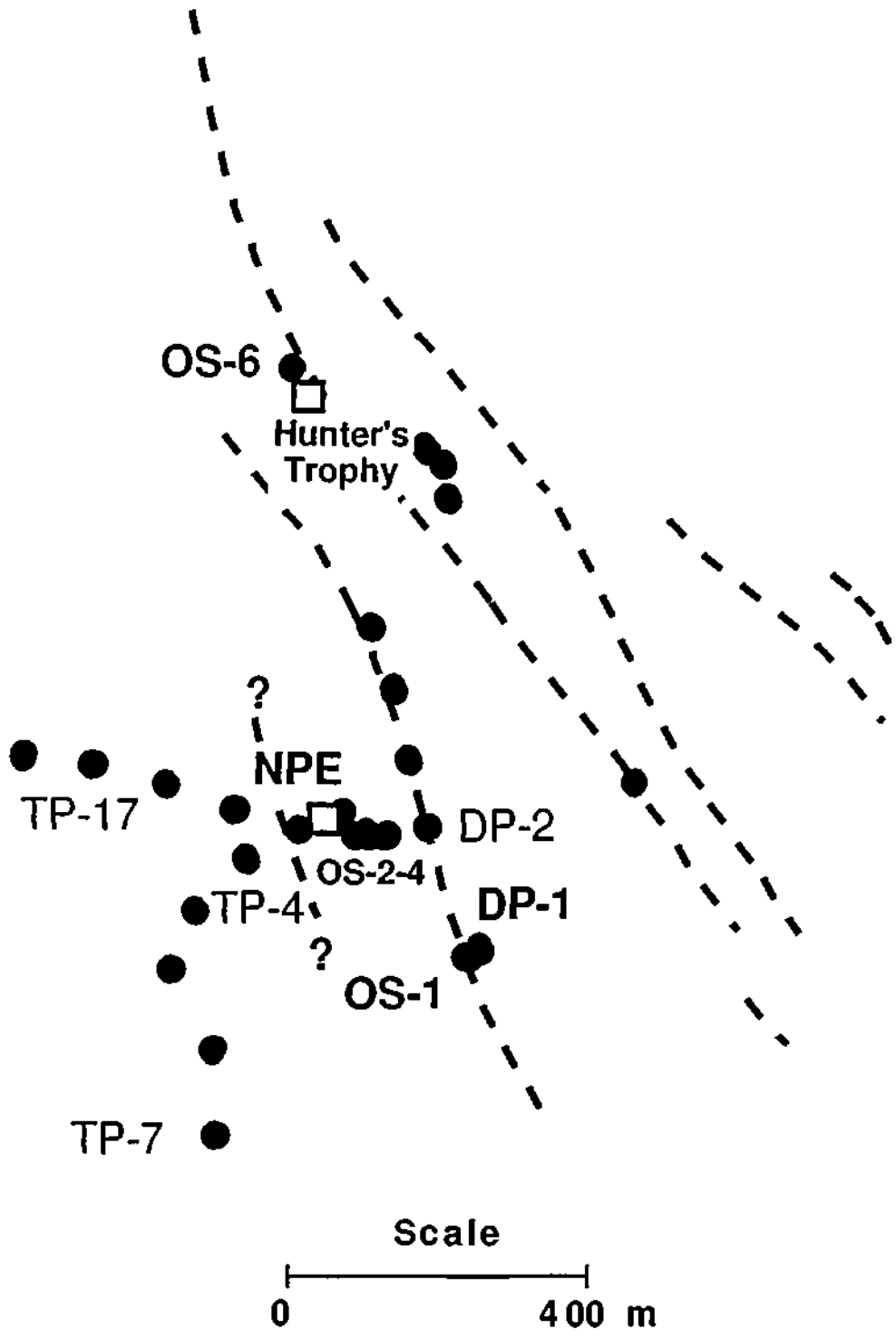

DP-1
OS-1
2200
2100
2000
1900
1800
1700
1600
1500
1400
Elevation (m)
Cavity
Center of Cavity
1852.6 m
0
150
300 m

Fig. 1a



(depth to detonation: 400 m)

Fig. 1b

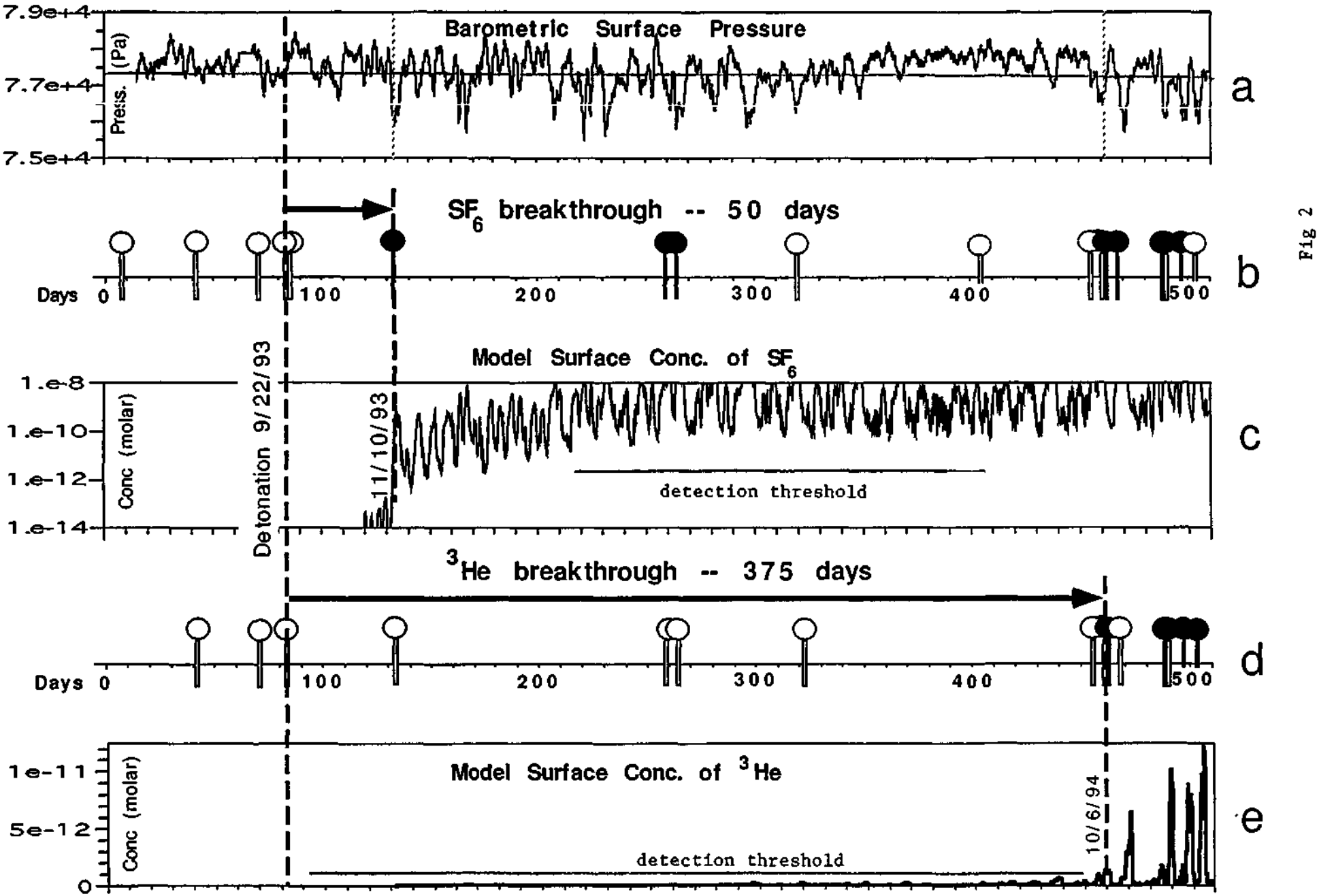

7.9e+4
7.7e+4
7.5e+4
Press. (Pa)
Barometric Surface Pressure
a
SF6 breakthrough -- 50 days
Days 0
100
200
300
400
500
b
Model Surface Conc. of SF6
1.e-8
1.e-10
1.e-12
1.e-14
Conc (molar)
Detonation 9/22/93
11/10/93
detection threshold
c
3He breakthrough -- 375 days
Days 0
100
200
300
400
500
d
1e-11
5e-12
0
Conc (molar)
Model Surface Conc. of 3He
10/6/94
detection threshold
e

Fig 2

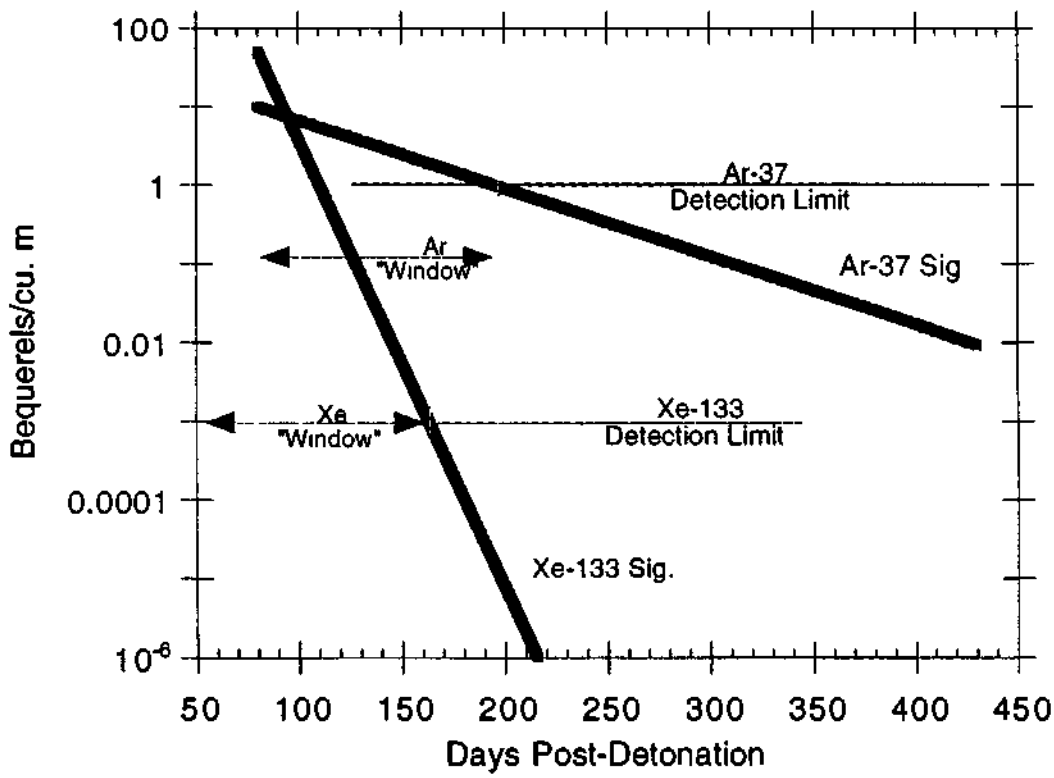

100
1
0.01
0.0001
10^{-6}
Bequerels/cu. m
Ar-37
Detection Limit
Ar
"Window"
Ar-37 Sig
Xe
"Window"
Xe-133
Detection Limit
Xe-133 Sig.
50
100
150
200
250
300
350
400
450
Days Post-Detonation

Fig 3

Effect of atmospheric infiltration on gas sample concentration in fractured and uniform porous formations

Fig 4

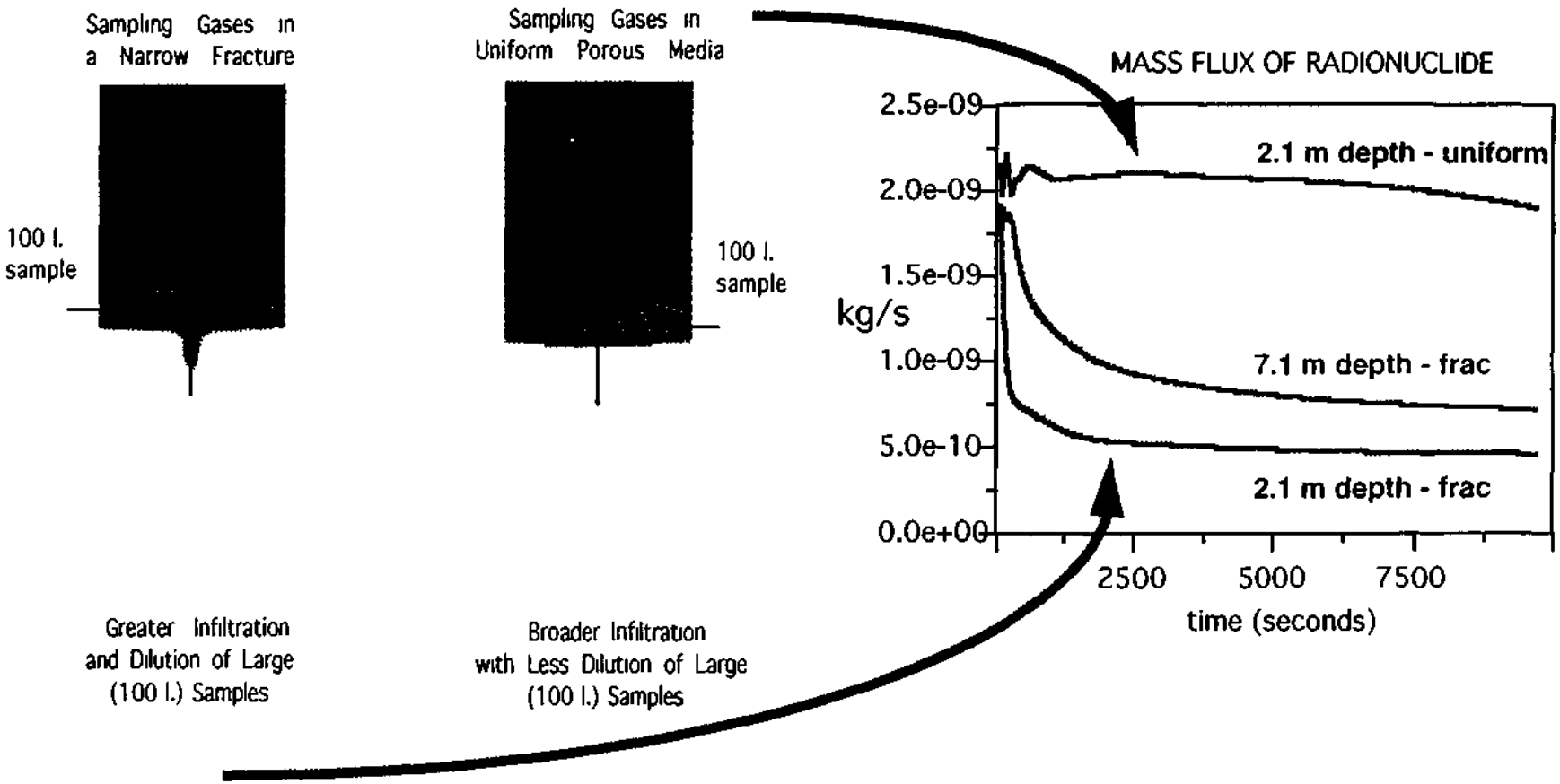


TABLE 1 Summary of Rainier Mesa Gas Sampling Observations

Sample Suite	Date	No. Sites F=fracture/fault S=soil	Total Detects (Total Samples)	Locations of SF-6 Detection	Detected SF6 Concentration pptv	Locations of He-3 Detection	Detected He-3 Concentration pptv	Barometric Pressure* millibars
1	7/8/93	3F	0 (3)		ND		--	772.1
2	8/11/93	3F	0 (5)		ND		ND	775.2
3	9/9/93	3F	0 (6)		ND		ND	777.6
4	9/22/93	Tunnel Portal	0 (2)		ND		ND	772.5
5	9/24/93	3F	0 (3)		ND		--	778.2
6	11/10/93	3F	1 (6)	OS-6	340		ND	764.8
7	3/17/94	3F	3 (7)	OS-1, OS-6**	540, 580, 280		ND	771.2
8	3/23/94	3F	3 (6)	OS-1, OS-2, OS-3	580, 450, 400		ND	765.3
9	5/17/94	2F	0 (2)		ND		--	765.2
10	5/19/94	3F	0 (3)		--		ND	770.7
11	8/11/94	1F	0 (1)		ND		--	779.1
12	9/29/94	5F	0 (9)		ND		ND	771.6
13	10/4/94	5F	0 (11)		ND		ND	766.8
14	10/6/94	6F	2 (11)	OS-1	13	OS-6	8.42	771.2
15	10/12/94	6F	1 (12)	OS-3	18		ND	769.8
16	11/2/94	6F	3 (15)	DP-1, OS-6	45, 45	DP-1	9.22	760.9
17	11/3/94	6F	0 (15)		ND		ND	764.5
18	11/10/94	13S, 6F	3 (41)	DP-1, TP-4(S)	18, 9	DP-1	21.4	760.8
19	11/16/94	18S, 6F	1 (40)		ND	DP-1	14.7	762.8

Multiple gas samples were obtained from sites on Rainier Mesa at the Nevada Test Site on at least 19 separate occasions starting before the underground detonation and continuing for over 400 days after it. Both diagnostic modeling[5, 10] and some previous field work by R.H. Nilson and his colleagues (unpublished manuscript, 1996) strongly indicated that deeper, barometric lows, which are associated with storms, enhanced the probability of detecting gases at the surface. Thus, our data tends to be taken during such lows. (Owing to the poor accessibility of Rainier Mesa during storms, sampling by a two-man team tended to be limited by the onset of snowfall.) Sites on faults and fractures were also preferred in view of the previous work. Here a site is classified as being either on a fracture/fault (F in column 3) or on tarped soil (S). Only one tarped soil site, TP-4, not obviously associated with a fault or fracture, yielded a value of SF_6 above the background. In the fourth column the total number of detections is indicated along with the total number of samples taken on a given day. The locations where detections occurred are given in the fifth and seventh columns along with the respective concentrations of SF_6 and ^3He (sixth and eighth columns) that were obtained from those sites with the symbol "ND" indicating that excess concentrations were not detected (a dash indicating that no analyses were performed). An average barometric pressure is given for each sampling event in the last column. For comparison, the average barometric pressure for the entire pressure history is 773 millibars.

** Two samples were obtained from OS-6 separated by several hours: the first yielding a concentration of 580 pptv followed by another concentration of 280 pptv.

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