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Subsurface Noble Gas Sampling Manual

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Subsurface Noble Gas Sampling Manual

Capturing Radioactive Noble Gases During a CTBT On-Site Inspection

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Preface

The intent of this document is to provide information about best available approaches for performing subsurface soil gas sampling during an On Site Inspection or OSI. This information is based on field sampling experiments, computer simulations and data from the NA-22 Noble Gas Signature Experiment Test Bed at the Nevada Nuclear Security Site (NNSS). The approaches should optimize the gas concentration from the subsurface cavity or chimney regime while simultaneously minimizing the potential for atmospheric radioxenon and near-surface Argon-37 contamination. Where possible, we quantitatively assess differences in sampling practices for the same sets of environmental conditions. We recognize that all sampling scenarios cannot be addressed. However, if this document helps to inform the intuition of the reader about addressing the challenges resulting from the inevitable deviations from the scenario assumed here, it will have achieved its goal.

Introduction

From field testing and computer-generated gas-migration simulations, we have developed a best practices approach to noble gas sampling at a Comprehensive Test Ban Treaty (CTBT) On-Site-Inspection (OSI) location with the goal of increasing the potential for capturing a noble gas signature of interest, improving noble gas sampling team efficiency and reducing the time devoted to sampling by the OSI inspection team. This best practices project has the additional objective of describing the best available approaches to sampling that minimize the potential for sample contamination by ambient radioxenon atmospheric gases and cosmic-ray-produced argon gases in the soil not associated with any UNE.

In this manual it is assumed that direct venting to the surface has not occurred. That case is relatively straightforward to sample when a vent or fracture at the surface has already been identified. This manual focuses only on the more difficult case of contained UNEs where gases are still able to percolate to the surface through small fracture networks that are inherent to the containment regime.

This manual includes a more detailed discussion of the subsurface sampling sections of the "Noble Gas Concept Of Operation" (Ref. 1). Before starting we mention several assumptions relevant to this discussion.

Main Assumptions/Definitions

An underground nuclear explosion (UNE) is the origin of subsurface noble gases.

As discussed below, other sources of noble gases of interest also exist, but UNEs are assumed to be the most likely origin of gases of interest captured from the subsurface. It is assumed for this document that sites of interest are likely underground explosion sites (e.g., Ref. 2 and Ref, 3, Fig. 1).

Radioactive decay of noble gases of interest creates a "window" for detection by OSI subsurface gas sampling and analysis.

The decay of radioactive noble gases and the fact that they may require some time to arrive at the surface following a UNE means that the possibility of detecting these gases exists within a temporal window of opportunity (e.g., Ref. 3, Fig. 2). In some scenarios, seeps or venting of detonation gases may allow significant quantities to reach the surface and be released into the atmosphere immediately following a UNE. In other release scenarios, days to weeks may be required for gases to reach the surface at detectable levels (e.g., Ref. 2).

Atmospheric and subsurface background concentrations of noble gases of interest may exist potentially contaminating near-surface signatures associated with UNE detonation gases.

An Argon-37 (Ar-37) natural background in the shallow subsurface, resulting from the interaction of cosmic ray neutrons with native calcium (Ca-40) in the rock and soil (Ref. 4), is a potential source of contamination of Ar-37 produced by fast neutrons during the UNE. Additionally, atmospheric contamination and the resulting soil-gas backgrounds of Xenon isotopes may occur owing to releases by nuclear reactors and medical isotope production facilities (Ref. 3, Fig. 10). Fortunately, backgrounds of noble gases of interest tend to be low. Because gas sampling is at or just below the surface where contamination sources may exist, sampling methods should ideally be optimized to minimize contributions from these non-UNE sources.

Summary of Subsurface Transport Processes Affecting Gas Sampling

As mentioned, this manual focuses only on the more difficult case of contained UNEs where gases are still able to percolate to the surface through pre-existing and explosion-enhanced small-aperture fracture networks, which are characteristic of UNE containment regimes. At least two different transport mechanisms or processes determine how radioactive noble gases migrate to the surface to be sampled by the methods outlined in this document. Each process is dominant during different periods of time following a UNE. A simulation illustrating the flux of Xenon-131m at the surface is shown in Figure 1.

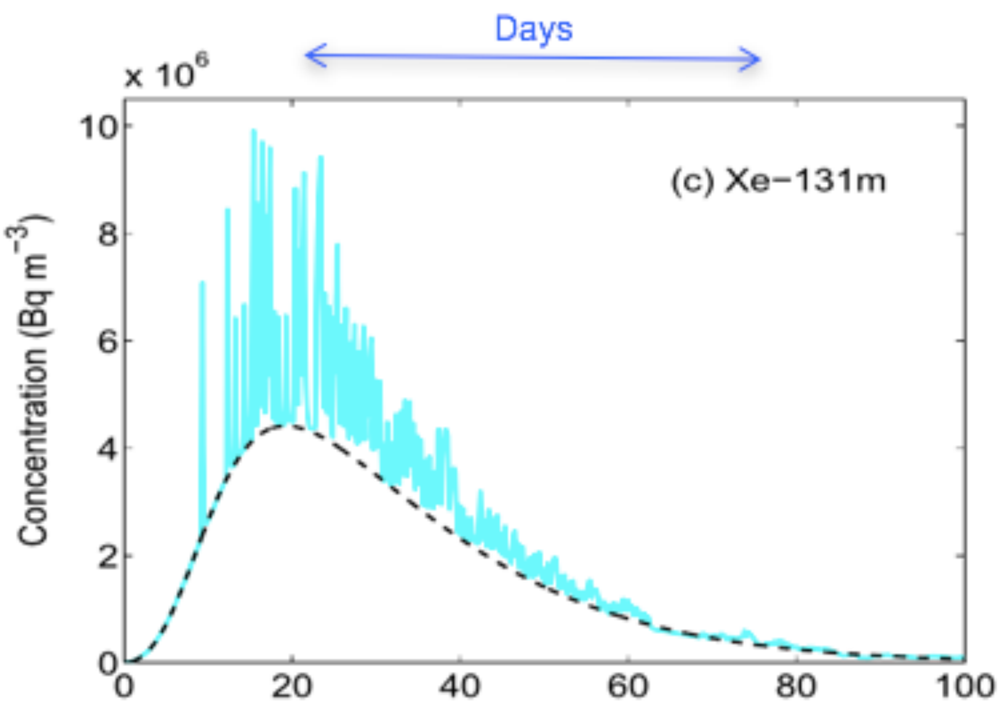


Figure 1. An estimate of the predicted flux of Xenon-131m (blue) at the surface versus time following an underground nuclear explosion. The barometric component of the gas transport is the fluctuating line while the thermally driven component of transport is much smoother dashed curve. Initially only the thermally driven part of migration is responsible for producing a signal at the surface. The barometric part of transport begins contributing significantly approximately 10 days after detonation according to this model.

Initially, during the first few days according to the gas-migration simulation, thermal convection and pressure-driven gas flow are responsible for moving gases upward toward the surface (Fig. 1: smooth dashed line). In cases where significant matrix permeability exists in the fracture walls, thermally driven gas migration is responsible for "loading" the porous walls of fractures with detonation-produced noble gases as the gases flow along the network. During the early part of the flux history, the thermal and pressure drive dominate transport and are represented by the rapid and smooth rise of the flux curve as shown in Figure 1. After noble gases are loaded into the matrix, barometric pumping, which can draw gases from both the cavity and the matrix, begins to play a significant role as exemplified by the transition to rapidly fluctuating values of the flux some days after the detonation. The best practice sampling approach during the two periods may be different and will be discussed in more detail.

Summary of Potential Dilution and Contaminant Infiltration Affecting Gas Samples

Dilution

The sampling described here is performed at or very near the interface between the atmospheric and subsurface regimes which adds additional complexity to the sampling process. For gas sampling at or near the surface, it is virtually impossible to capture gases only from the detonation cavity. In the subsurface, gases rising from the cavity will mix with and dilute gases already present in the fracture and rock/soil void space. Besides this dilution in the subsurface, the sampling process at and near the interface may draw gases from the atmosphere into the subsurface and ultimately into gas samples. We have performed simulations of subsurface gas sampling to better illustrate this point. The simulations also illustrate partial solutions to the dilution problem.

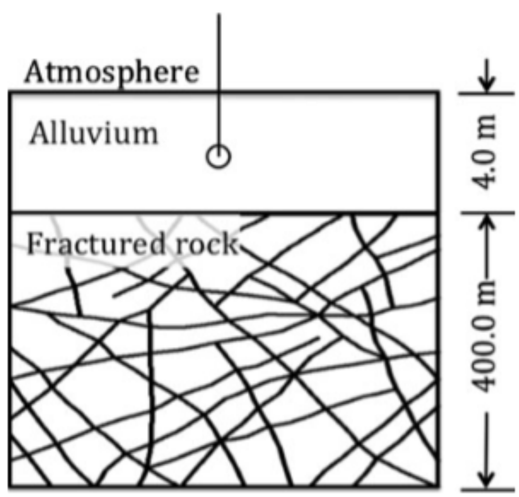


Figure 2. One of several different conceptual models intended to demonstrate the relationship between atmospheric infiltration into alluvium and the flow of gas from underlying fractured bedrock during the extraction of a gas sample from the sampling point indicated by a circle. In this example, alluvium is 4 m thick with a sampling point at a depth of 2 m and the underlying fractured zone is 400 m thick. (The alluvium thickness is exaggerated in the figure to emphasize

emplacement of the sample tube.)

Figure 2 Illustrates a general model of a permeable alluvium layer (4 m thick) overlaying a permeable fractured regime (400 m thick) bounded on the bottom by the water table and at the atmosphere/alluvium interface by a time varying barometric boundary condition. The sampling point is located at a depth of 2 m corresponding to the middle of the alluvium layer. This seems to be a reasonable sampling depth based on augering and direct push attempts, although any sampling model is highly scenario dependent.

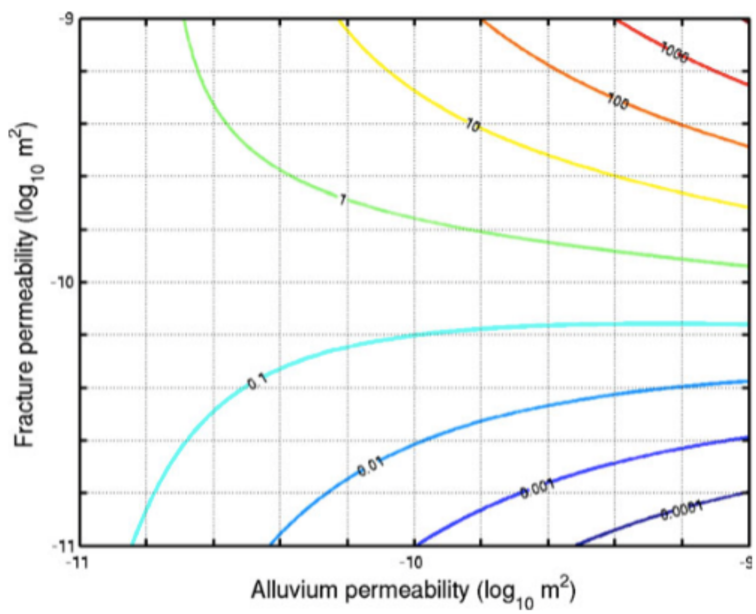


Figure 3. Ratio of gas flow from fractured rock to that from atmosphere captured in a sample after flow in the alluvium layer had reached steady state as a function of the alluvium and underlying fracture permeability

Figure 3 summarizes how dilution of a subsurface sample by atmospheric gases can occur. The figure illustrates that dilution is dependent on both the fracture and soil matrix permeability. That is, dilution of a sample is highly site dependent. We find in Figure 3 that the amount of gas extracted from the fracture zone relative to the amount of gas extracted from the atmosphere during subsurface sampling varies with both the soil and fracture permeability. Thus, for a fracture zone permeability of 8.0×10^{-11} m^2 (80 Darcys) and a soil layer permeability of 1.0×10^{-10} m^2 (100 Darcys), the contribution of gas in a sample volume produced by fractures is only 10 % of the contribution of gas from the atmosphere (light blue line). However, increasing the fracture permeability by an order of magnitude (aperture increase by a factor of 2.15) effectively reverses the situation (yellow line) so that the contribution of gases from the fracture is now ten times greater than the contribution from the atmosphere.

For the particular ranges of fracture and alluvium permeability assumed here, the ratio is far more sensitive to changes in fracture permeability than it is to changes in alluvium permeability. A change of three orders of magnitude in the fracture permeability can change the ratio of the fracture to atmospheric contribution by up to seven orders of magnitude while the same change in the soil permeability changes the ratio by just over three orders of magnitude. We learn from this that atmospheric infiltration is not only determined by the hydrologic character of the alluvium layer but also by the characteristics of the underlying fracture layer, if present.

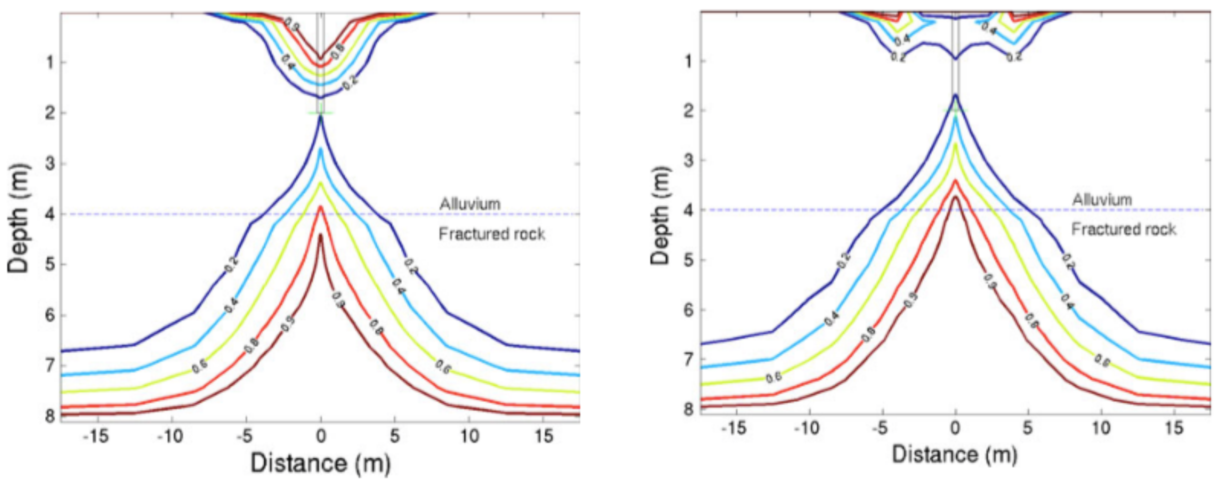


Figure 4. Both figures are based on simulations involving the alluvium-covered fractures shown in Figure 2. The subsurface sampling point is located at a depth of 2 m, which is in the middle of the alluvium layer. Colored lines are concentration contours of either subsurface gas drawn up from below or atmospheric gas drawn down from above. Lowest concentration is the blue color. Figure 4 (left) illustrates contours resulting from extracting gas in the alluvium when no tarp is deployed while Figure 4 (right) shows contours for the same case but with a tarp (3 m) centered on the point were the sample tube reaches the surface. It is clear that the tarp case reduces the concentration of atmospheric gases at the subsurface extraction point.

An approach to reducing infiltration is to use a tarp or plastic sheeting to cover as much surface area around the subsurface sample point as is practical. Simulations (Fig. 4) show the effect of using a 3-m tarp in a sampling arrangement similar to that assumed in Fig. 2. The concentration of atmospheric gas drawn downward from above compared to the concentration of gas drawn upward into the inlet from the underlying fracture regime is clearly affected by using a tarp, and the contribution of atmospheric gas decreases with increasing tarp size in this model.

Contamination

We have only considered the effect of atmospheric infiltration in diluting a soil-gas sample while it is being extracted. In addition, contamination of a soil gas sample by radioxenon in the air can also occur as a result of drawing atmospheric gases directly into the sample. Even with plastic sheeting barriers to prevent atmospheric infiltration during sampling operations, the prevention of atmospheric contamination of a sample cannot be guaranteed without further precautions. Natural gas exchange between the atmosphere and soil caused by barometric fluctuations can also introduce contaminant gases from the atmosphere. During periods when the atmospheric pressure exceeds the pressure of gases in the soil, the atmospheric gas composition can be ‘‘impressed’’ into the shallow soil regime creating a ‘‘memory’’ of the composition of the gases passing over the surface. Atmospheric sources of Xe-133 could conceivably result in the transport of low concentrations of this isotope into the shallow subsurface creating the very low probability of a false Xe-133 detection during an OSI. Besides normal releases from nuclear power reactors or larger accidental releases ([40 Bq m^{-3}) as occurred during the Fukushima nuclear reactor accident (Ref. 5), other possible common sources of radioactive Xenon isotopes are byproducts of the production of radio-pharmaceutical isotopes (Ref. 6).

For UNE confirmation purposes, Argon-37 is highly attractive as a short-lived noble gas isotope. While natural background levels are extremely low, coincidence-counting methods are sufficiently sensitive that low background levels may be detectable in practice. Ar-37 is produced through a spallation interaction with naturally occurring Ca-40 in the soil and fast neutrons bombarding the subsurface created by nuclear explosions. Similarly, the natural background in the near-surface regime results from interaction of Ca-40 in the soil with cosmic-ray neutrons.Riedmann and Purchert (Ref. 4) have argued that the naturally occurring equilibrium Ar-37 concentration in shallow soil gas is a function of an exponentially decreasing production rate from cosmic ray neutrons with increasing soil depth, diffusive transport in the soil air, and radioactive decay. They also show that the highest activities of 100 mBq m^{-3} air are two orders of magnitude larger than in the atmosphere peaking in the 1.0–2.0 m depth range and rapidly decrease with greater depth. It should be noted that the shallow production of Ar-37 near the surface may be “smeared” vertically by barometric pumping of gases in fractures. Additionally, models show that the shallow Ar-37 background is likely to be temporaily variable. Please refer to Ref. 3.

The models presented here are necessarily specific and no attempt has been made to cover all sampling scenarios. It is hoped that this discussion can at least alert the reader to the potential dilution and contamination issues involved in performing subsurface sampling.

Why Timing Subsurface Gas-Sample Acquisition is Important

The results of field experiments and our most complete models of subsurface gas migration indicate that two different timings should be considered when performing noble gas sampling. The first is the time after detonation when first sampling is attempted. At early times, days to weeks, it is likely that the thermally driven component of gas migration is still significant and gases may be captured anytime. However, according to Fig. 1, early-time fluxes at the surface will tend to be quite weak, down orders of magnitude compared to the peak barometric-fluctuation-influenced fluxes occuring at later times. At these later times, weeks to months, it is expected that the best sampling results can be obtained when sampling at times of peak surface flux. This occurs when the barometric pressure is falling. Thus, sampling at later times is likely to yield better results when performed during a period of falling barometric pressure. Figure 5 illustrates the advantages of sampling during a period of falling pressure. In the figure, atmospheric pressure is shown as a function of time with its fluctuations. The green shading superposed over the pressure indicates gas concentrations that exceed minimum levels of detection. In the simulation, the green shading only occurs during periods of falling atmospheric pressure and gradually vanishes as the pressure rises.

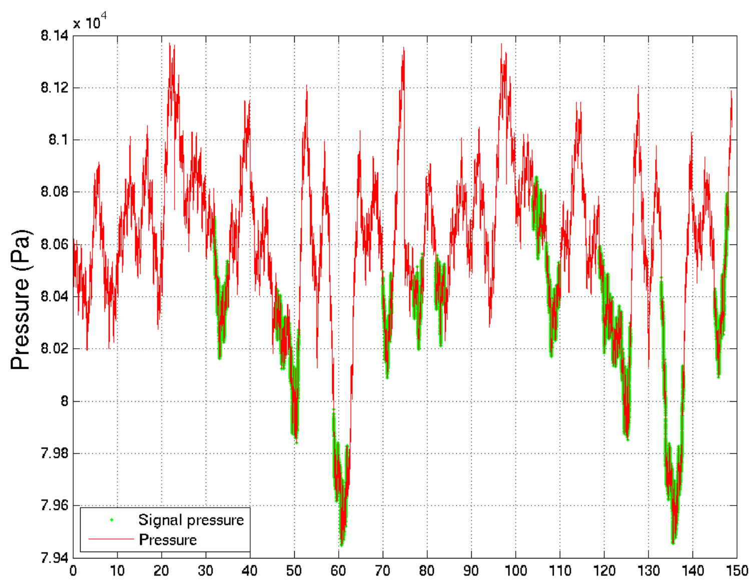


Figure 5. A subsurface gas-migration simulation showing a plot of atmospheric pressure (red line) versus time obtained from a Smart Sampler. Green points are plotted for surface pressures and times when gas concentrations are above minimum detectable levels. Note that sampling during periods of falling barometric pressure produces levels of gas at the sampling site exceeding minimum detectable levels in this simulation. However, even in the early stages of the pressure record, minimum concentrations did not reach detectable levels, as no green is present. At early times, gases are “loaded” into the matrix by the upward transport along fractures during barometric pressure fluctuation. Until the concentration in the pore space has risen to sufficient levels no signal will be detected.

It should be noted that the LLNL Smart Sampler offers two modes of automated sampling that take into account fluctuations in the barometric pressure. The first barometric triggered mode takes a sample during periods when the atmospheric pressure is falling and subsurface gases are most likely to be migrating toward the surface resulting in higher sampled concentrations of the gases of interest. The second atmospheric-pressure-sensitive mode is referred to as a 'continuous barometric' mode and takes samples whether the barometer is falling or rising. However, the sampler separates samples taken during rising pressure from those taken during falling pressure into two different sample containers (e.g., typically 2000 liter sample bags, balloons or bladders). This mode attempts to allow sampling all the time while preserving the potentially much higher concentrations of gases associated with periods of sampling only when the atmospheric pressure is falling.

Objective of Sampling-Station-Site Selection in a Focused Search Area

For subsurface noble gas sampling, the objective of sampling site selection is to use available surface (e.g., fractures, explosion or containment artifacts), near-surface observations (e.g., ground penetrating radar) and geologic considerations to specifically identify the UNE surface-ground-zero as well as nearby sites where subsurface pathways of UNE gas transport terminate at the surface. Ideally, some skill and knowledge relevant to performing the subsurface-gas sampling site selection process is required. In particular, understanding the potential relationship between explosion-produced pathways for gas transport and existing natural pathways is extremely helpful for identifying the most likely sites for the detection of UNE-produced noble gases.

In addition to understanding how the local geology and UNE containment practices may contribute to the existence of gas transport pathways, a similar understanding of how noble gas transport processes (i.e., barometric pumping, cavity pressurization and thermally driven convection) are responsible for noble gas detections obtained from subsurface gas samples is also highly desirable. Because such samples, which typically have large volumes (~2 cu m), tend to be collected very near the ground surface or even at the surface, familiarity with the mechanisms by which atmospheric dilution or contamination of a sample can occur (i.e., atmospheric gas infiltration into soil, leaks in sampling-hole sealant, barometrically driven soil gas memory effect, etc.) is of equal importance. The goal of this understanding is to select optimal sites for sampling that are also amenable to the application of techniques that minimize atmospheric dilution or contamination effects. For example, a fracture detected at the base of a soil layer may be a better candidate for producing a sample that is relatively undiluted by atmospheric infiltration than a readily visible surface fracture providing easy sample point installation and also an easy route for infiltration of the atmosphere to the sample point.

Sampling-Station-Site Selection – When UNE Artifacts Are Present

UNE artifacts that may be directly connected to subsurface detonation point:

- Emplacement casing
- Tunnel portal
- Wells/boreholes
- Cable bundles breaking surface
- Ventilation shafts

Figure 6. Artifacts or objects associated with a potential UNE site that are connected to the subsurface regime are potentially excellent pathways for gases to reach the surface. Photo is of emplacement casing which has produced detections of gases.

Artifacts of the UNE may be discovered that not only represent evidence for the occurrence of a UNE but also function as pathways for gas transport to the surface. Such features as emplacement casing, portals of tunnels, boreholes and cables may be well connected to the subsurface and represent the greatest potential for producing at least forensic levels of noble gas signals (Fig. 6).

Sampling-Station-Site Selection – UNE Induced Pathways

Possible explosion-produced pathways at surface:

- Cratering
- Radial fractures
- Circumferential fractures
- Linear fracture systems
- Down drops induced along local faults

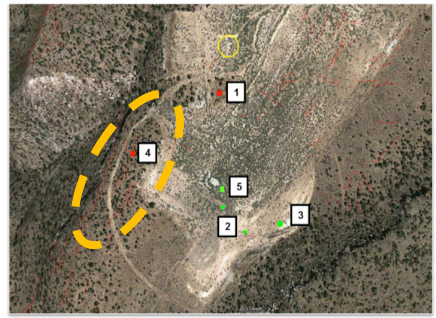


Figure 7. The UNE creates new and enhances existing pathways for subsurface gas transport. Cratering as a result of explosion-cavity collapse as well as fractures having different geometries are common features of a UNE although sufficiently deeply buried events may only exhibit very subtle features or none at all. In the dashed ellipse of the overhead view of a UNE site, a gas-producing fracture, a member of a system of linear fractures, resulted as a post-detonation feature. Numbered locations indicate sites selected for soil-gas sampling.

Besides manmade artifacts such as emplacement shafts, portals and cable bundles, UNE produced artifacts (e.g., fractures, reactivation of geologic faults) may produce excellent paths for gas transport to the surface. In rocky materials extensive subsurface fracturing tends to occur. Depending on the geology of the containment regime, fracturing may sometimes be enhanced. For example, the overhead view of the UNE site shown in Fig. 7 shows an ellipse surrounding a set of long, parallel fractures. The fractures occur near the edge of a terrace or down-drop that could not fully contain the outward motion of the underground detonation. As a result, underlying jointed columns of volcanic rock were slightly displaced horizontally creating this series of open parallel fractures that have produced strong gas signals during tracer experiments performed at the UNE site.

Sampling-Station-Site Selection – Natural Pathways

Local natural pathways interconnected with explosion-induced fracture network:

- Natural fracturing at surface
- Nearby regional faults (< 500-600 m from ground zero)
- Tree roots especially in thin alluvium over bedrock
- Other features indicating subsurface fractures

Figure 8. The subsurface tends to be an imperfect container for UNEs as illustrated. Geologic pathways for gas flow occur in the form of cooling fractures in rock, local and tectonic faults and even sedimentary layering of sand or gravel. Such natural pathways form networks that may become interconnected with the explosion-produced fracture regime allowing transport of noble gases to the surface.

UNEs that are particularly deep for their level of yield may produce few fractures at the surface. However, the resulting explosively produced fracture systems may well interconnect with existing natural pathways allowing noble gases to be transported to the surface. Sampling along natural faults and fractures has produced detectable levels of gas transported from deep detonation points. Faults that may be reactivated locally by the explosion may form small down drops or offsets that represent ideal indicators of sites worth sampling using subsurface techniques.

Sampling-Station-Site Selection – Featureless Site

When a suspected site is featureless on surface or does not fit any of the criteria 1-3:

- Sample near/over suspected SGZ
- Other features indicating possible subsurface fractures
- Tree roots especially in thin alluvium over bedrock

Figure 9. Guidance in selecting sampling sites may be obtained from relational considerations (e.g., proximity to suspected surface ground zero as evidenced by portals or tunnels as shown in circles) and indirect indicators of pathways in shallow soil such as tree roots, especially if indicating damage from ground motion. In the case of evident portals and tunnels, gas sampling should also be performed if accessible.

The most difficult case for siting sampling stations occurs when the ground surface is essentially featureless except maybe for the presence of trees and other flora. In such cases, inspectors may only have the possibility of using the proximity to other more distant features (e.g., portals or tunnels below potential sampling areas as shown in Figure 9.) as indicators for establishing possible sampling stations. In some cases the distribution of flora (e.g., trees and bushes) could indicate the presence of a sharp subsurface division such as a fault that may serve as a pathway to the surface. Random sampling has been found to be far less effective for detecting signals than sampling associated with UNE artifacts or the geology of the site (e.g., faults).

Sampling-Station – Subsurface Gas Acquisition Design

Following sampling-site selection, a decision must be made concerning the type of subsurface sampling to be performed at each of the selected sites in a particular area of interest. Two sampling designs have been previously employed in tracer experiments. They can potentially produce detections and each has its advantages and disadvantages.

The first method involves the insertion of a sample tube into the subsurface while the second method, broad-area tarping, utilizes tarps or plastic sheeting placed on the surface over perforated or irrigation tubing through which gases reaching the surface are withdrawn (Figure 10.). A major advantage of the sample tube approach is that samples are drawn directly from depth and the potential for atmospheric infiltration is reduced. The prevention of infiltration is particularly good when small, liter-sized samples are acquired but is somewhat less optimal with continuous sampling or when very large samples (e.g., 2 cu m) are needed. To some degree, the atmospheric infiltration problem can be mitigated by the use of a tarp or plastic sheet laid on the surface and centered on the sampling tube that helps to minimize atmospheric gases from infiltrating through the soil near the sample tube to the gas-extraction sample point when large volumes are needed (Fig. 4 and associated discussion). A disadvantage of the single-point sampling tube method is the subsurface sampling point may not be in the near-vicinity of a gas-migration pathway or fracture. Ideally, the tube should draw gases from a zone that intersects multiple fractures acting as migration pathways.

Finally, multi-point or distributed sampling of a feature (e.g., inserting multiple sample tubes spaced along a surface fracture) can reduce infiltration effects associated with large volume extractions by distributing the extracted volume among the sample points connected in common. The Smart Sampler allows up to 5 separate inputs, each having flow-rate control, for drawing a common sample simultaneously from multiple tubes. The multiple tube approach may also help to minimize any signal depletion effects that might result from excessive gas withdraw from a single sampling tube.

The second gas-acquisition method (broad-area tarping) involves extraction of gases from beneath a tarp or plastic sheet placed on the surface over a feature or suspected feature of interest. Perforated tubing used in garden irrigation is looped beneath the tarp to extract gases coming to the surface. Sand or other material is typically placed along the edges to hold the tarp down (Fig 10.).

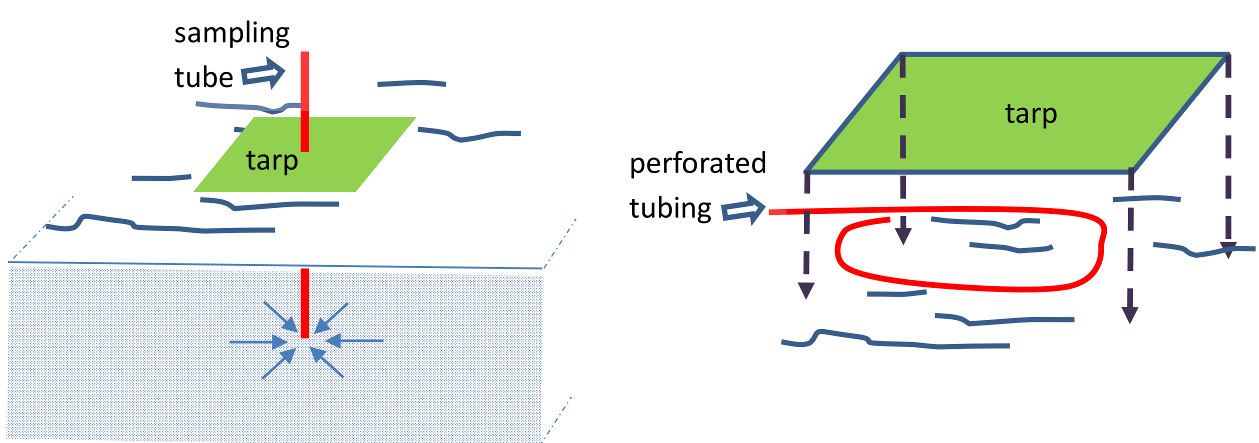


Figure 10. Two subsurface soil gas sampling methods are illustrated. The first (left) involves inserting a samping tube to several meters depth and drawing gases into sampling bags. The second, broad-area tarping (right) captures gases crossing the surface under a tarp. This is achieved by laying a perforated tube, such as the type used for garden irrigation, under a large

tarp and extracting gases through the tubing. Loose dirt or sand is mounded along the edges of the tarp to reduce infiltration effects.

This sampling technique is of particular value when sampling an extended pathway regime or when some uncertainty exists concerning the location of a pathway (near-surface buried fractures). The technique also has value for continuous low-flow sampling of subsurface gases as signal depletion is not likely to be a problem as it potentially may be for the focused sampling associated with buried sample points.

A disadvantage of the broad-area tarping technique is the possibility of drawing atmosphere through the soil/sand piled at the edges of the tarp into the sampling zone beneath the tarp. This possibility must always be confronted during continuous sampling operations. However, sampling during a period of falling pressure may allow for samples to be acquired that have only a minimal atmospheric component. During falling atmospheric pressure, gases are coming to the surface beneath the tarp. As the pressure continues to fall, gases will tend to build up beneath the tarp eventually flowing outward through the sand at the edges of the tarp. During periods when this occurs, only gases from the subsurface are expected to be present beneath the tarp, providing an ideal situation for extracting soil gas samples.

Which Sampling Method is better?

It is wrong to claim that one sampling approach will always yield better results than the other as there are so many possible sampling scenarios where one method may perform better than the other. Indeed, there are possible UNE containment regimes where one method will produce a better outcome than the other. This is why it is very beneficial to have some understanding of subsurface gas-migration processes as they relate to a specific sampling approach in designing a sampling station.

To gain a general understanding, we compare simulations of sampling using a subsurface tube with broad-area sampling beneath a tarp. The model we use is similar to Figure 2 except the gas of interest producing the “signal” of Figure 11 has risen to within 8 m of the surface, presumably due to previous thermal and barometric transport. As in Figure 2, the sample tube is inserted to a depth of 2 m while the tarp is placed on the surface. Figure 11 allows comparision of the “signal” to “noise” that will be encountered for three different sampling cases where the noise is non-signal gas that is already present in pore and fracture volumes as well as drawn or infiltrated into the sample from the atmosphere. In the simulation, sampling is performed over an hour during a period of barometric low pressure, which is preferred for sampling regardless of the technique used. The plots with sampling by inserted tube, withdrawing subsurface gas at about 3 liters/min (Fig. 11, left), show the time-dependent, instantaneous signal and noise during the one-hour sampling period. These plots show that, in this case, there is little difference between sampling with and without small infiltration tarps around the tube to reduce infiltration. This is yet another indication of how dependent the outcome of the sampling technique relies on the characteristics of gas migration in the UNE containment regime and sampling arrangement. The initial conditions assumed here are consistent with sampling just before the signal of interest reaches the surface and significant ambient pore and fracture gas is drawn into the sample. If gas has already diffused into the alluvium as will occur at later times after detonation, the signal-to-noise ratio of fluxes would be expected to be higher than shown in a) and c).

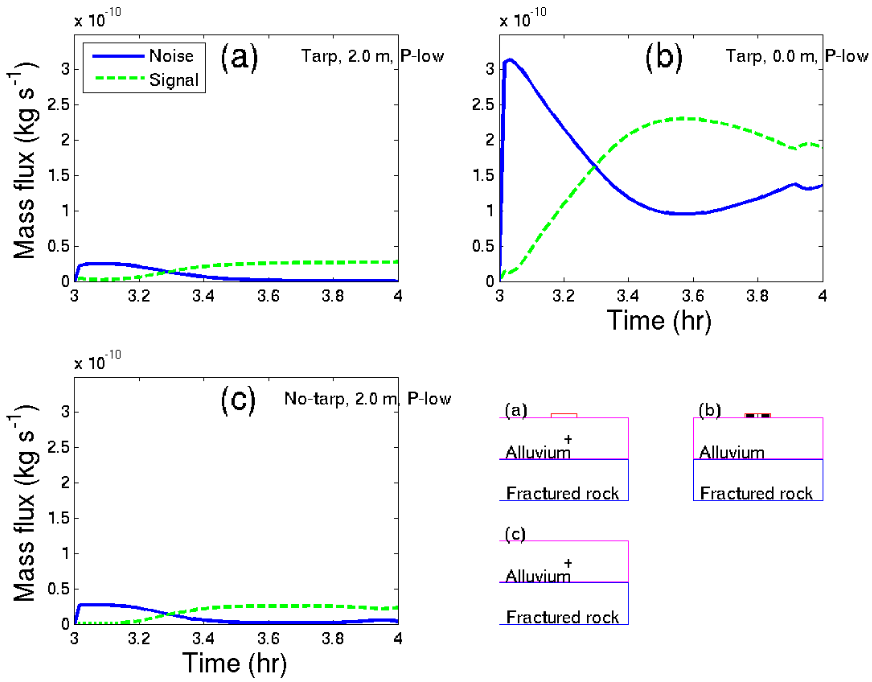


Figure 11. The plots a) – c) compare the trace-gas levels that are of interest and therefore called "signal" to ambient atmosphere, resident pore and fracture gas levels that only dilute or contaminate the trace gases which are the "noise" in these examples. Case a) assumes a small tarp (3 m x 3 m) covering the surface centered n the sampling tube. Case c) assumes no tarp is present. Case b) is for a large tarp placed over a perforated extraction tube as shown in Fig. 10 (right). Cases a) and c) are very similar with the "noise" (blue line) initially dominating any "signal". For this particular case, later samples will have a better signal-to-noise than early samples. Sampling using a tarp with underlying tube (b) produces a very different result. Noise still dominates at early times but ultimately the signal dominates later. The values of noise and signal are much larger for this case than for the inserted-tube withdrawal case. Gases are extracted at the rate of 3 liters/min in all cases.

Broad-area tarping of the surface over an underlying perforated extraction tube (Fig. 11 b) produces a very different result both noise and signal are very much larger than in the subsurface tube cases. As in the subsurface tube cases, the noise signal is much larger at first and falls off as pumping proceeds. However, the signal becomes larger within 20-30 minutes following the start of pumping. The much larger signal in the surface tarp case b) is due to the broad areal coverage.

When designing a sampling station, it is difficult to anticipate all the geohydrologic and containment details that will influence the outcome of sampling. *It is therefore recommended that, if possible, both subsurface tubes and broad-area surface tarps covering perforated extraction tubes be set up at a site. Alternatively, a hybrid technique combining both methods (surface and subsurface extraction) at the same location might be considered.*

Sampling-Station – Shallow Hole Sampling

Sample points can be deployed at a feature of interest or site where direct push/augering is possible:

- At least one sample point should be considered for identified subsurface pathway
- Depth of sampling is greater than 2 m *unless* high-flow layer or fracture is shallower
- A plastic sheet should be used to minimize infiltration.
- Multiple sample points may be installed along fractures/cracks with infiltration tarps
- Sampling tube installation is time and effort intensive which should be considered in context of resources relative to broad-area surface tarps

Figure 12. Sampling tubes are most applicable when a feature of interest can be identified.

Sampling tubes are appropriate in situations where visible features can be identified such as cracks or depressions indicating cracks in the ground surface. The goal of sampling tube insertion is to emplace the sampling point as deeply as possible into the feature and usually more than 2m unless a known high-permeability layer or fracture is shallower in which case the sample point should be emplaced in the vicinity of such subsurface features. Attempting to penetrate to depths greater than 2 m is necessary to minimize the contributions from the natural Ar-37 background which is anticipated to be at its maximum value at approximately this depth. To minimize the possibility of depleting a signal during a large volume sampling operation, it is advisable to insert multiple sampling tubes along a feature if possible. This procedure also increases the possibility of capturing a signal in the case where all sample points along a feature of interest are not equally productive.

Deploying Subsurface Sampling Tube

Photos courtesy of Dudley Emer

Figure 13. Creating a subsurface sampling point involves several steps. Photographs here illustrate how a subsurface sampling point may be produced. Initially, a hole, several meters deep, was created with a manual Geoprobe® insertion tool requiring two people to hammer a hollow steel shaft with expendable penetrating point into the subsurface (left). Following insertion of the hollow tube into the subsurface, the series of steps for completing the subsurface gas sampling point are shown on the right (a-f). a) A section of Geoprobe® pre-packed screen is attached to stainless steel tubing. Screen section allows subsurface gases to be withdrawn from the formation through a sandpack, which will be added between the screen and hole wall. Before inserting the screen a small amount of coarse sand is first dropped into the hollow shaft allowing the screen to rest upon the sand pack. b) The screen connected to the stainless tubing is inserted to the bottom of the hollow shaft. c) More coarse sand is now added through a funnel while moving the tube and screen up and down to prevent bridging of the sand and allow the sand to fill the zone between the hole and screen. Sand is added until the sandpack extends at least a dfew centimeters above the screen. A tool to estimate sandpack thickness, such as a small diameter rod, may be required. As more sand is slowly added through the shaft hole, the shaft is slowly withdrawn from the hole to allow the sand to fill-in completely between the screen and hole wall. d) Once sandpack is complete, Bentonite clay is

funneled into the hole. e) As necessary, the stainless tubing is moved up and down to prevent bridging of the clay and help it settle without air pockets. f) Left-over dirt is used to fill in any depression and the site is tamped to ensure a good seal between the sampling screen and surface.

Several methods can be used to deploy a subsurface sampling tube. The method described in Figure 13 has been used and seems to be a quick and reasonable approach. The objective of deployment is to locate the screened section ads near as possible to any identified gas migration pathway. Ideally, the depth of the screened section should clear the near-surface zone of maximum natural Ar-37 production in the soil, which has a typical depth of 2 m. A more permanent and time consuming sample point completion approach is provided by Geoprobe (Ref. 7). If no potential pathways are identified, it is probably better to use the broad area surface tarping approach to sampling.

Sampling-Station –Broad Area Tarping

Plastic sheeting can be deployed at a feature of interest or site where desired if sample points are not deemed appropriate:

- Random tarp placements tend to be unsuccessful (e.g., unrelated to features)
- Water saturated soil is inappropriate for tarp sampling and potentially for any sampling
- After covering the multi-hole gas extraction tubing (irrigation) with sheeting, dry soil is heaped covering edges of plastic sheeting
- Sand, rocks placed on tarp prevent billowing
- Tarping may become time intensive if bushes must be cleared

Figure 14. Broad area tarping is appropriate when a tube cannot be easily inserted into a visible surface feature or there is some uncertainty about the location of a feature or pathway.

Broad area tarping has yielded good results when some evidence for a feature exists. Alternatively, random tarping can be attempted if an area is thought to contain potentially high-permeability pathways that are not visible at the surface and other possibilities for inserting sample points into or tarping over visible features are minimal. Soils with high levels of water saturation are generally poor hosts for extracting soil gases whether using sample tubes or tarps.

Sample Extraction Method

Tracer-gas experiments show that a period of falling pressure is ideal for capturing gases of interest. When the barometric pressure is falling, the arrival at the surface of gases from depth is at a maximum and sampling that is timed to occur during a falling barometer has generally been found to produce the best results in terms of signal detection and the concentration of the gas of interest. Two general approaches to extracting subsurface gases exist and may be considered for extracting from sampling stations. *With either approach, the operator should carefully consider what appropriate soil gas extraction rate to use. Very high rates may yield a sample quickly, but likely at the cost of significant dilution and possible contamination of the sample from natural and man-made sources of noble gases.* The range of hydrogeologic scenarios is great and no one extraction rate can be considered optimal. Some recent simulations involving only a few realistic sampling scenarios suggest that 50 liters/hour total extraction rate at a site represents a reasonable and practical extraction rate and the operator should consider the necessity of using extraction rates much greater than this.

The first approach involves simple manual sampling using a gas pump and a collection bag. This is performed by inspectors possibly as part of a quick, early-stage survey. In general, randomly timed grab samples have been found to produce results that are somewhat inferior to timing sample extractions by the onset of barometric lows, although very soon after a UNE, detectable signatures of interest may nevertheless be present at all times depending on the level of containment and persistence of subsurface transport mechanisms such as thermal convection resulting from the heat of detonation. However, consistent manual sampling during optimal periods may be difficult or impossible because the onset of a significant barometric low is often associated with a storm and it may not be possible for inspectors to be present in the IA during such times (Figure 15). Besides poor weather conditions, constraints placed by the Inspected State Party on access to sampling sites or the general remoteness of sampling stations may not permit inspectors to be present for sample acquisition during the best periods for sampling.

Snow and lightning with brush fire can occur during barometric lows

Figure 15. Barometric lows tend to often be associated with the onset of storms. Automated sampling may be the only possibility for dealing with the requirement of sampling during poor weather.

To date, the best results for capturing signals have been achieved with an automated sampling approach that does not require human involvement. Several different automated sampling modes exist in the case of the LLNL Smart Sampling system. A continuous mode allows sampling all the time at some pre-set sampling interval and flow extraction rate. A barometric sampling mode can be selected that turns on the Smart Sampler to a pre-set flow extraction rate only during a period of falling atmospheric pressure. However, if concern exists that obtaining large-volume samples only during falling atmospheric pressure might cause a critical sample to be missed when the sampler pump is not operating, an alternative sampling mode allows sampling during both falling and rising barometric pressure. Gases sampled during a falling pressure are stored in a separate sample bag from gases sampled during a period of rising pressure.

While sampling continuously may seem to have the advantage of capturing any gases of interest that might be present in the subsurface, gases of interest that have an evanescent nature will tend to be highly diluted by soil gases causing a reduction of any signature of interest potentially below meaningful levels. The barometric mode of sampling should produce a larger signal potentially resulting in detection of the gases at an earlier time during an OSI. Another advantage of sampling in the barometric mode is the minimization of atmospheric infiltration as a falling barometer produces outflow from the ground surface and not inflow.

Sample Extraction-Manual

Manual sampling is appropriate for capturing gases from subsurface anytime a grab sample is desired for site/signature evaluation:

- Atmospheric or subsurface gas grab samples may be obtained
- Adjustable extraction rate up to 20 liters/min to match extraction requirements in field
- Volume totalizer to determine total sample size acquired

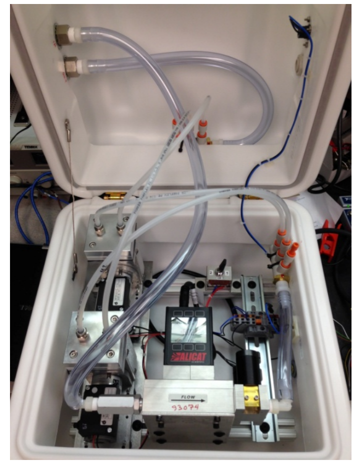
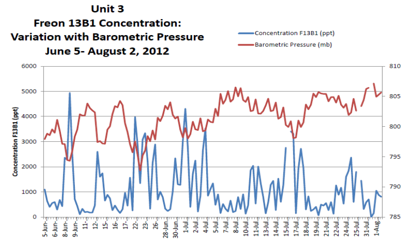


Figure 16. Manual sampler is highly portable being readily transported by one inspector. At least one 12-V battery is required to power this system.

When inspectors can be present to sample at a site during optimal times for sampling or simply when a grab sample is desired, a manual sampler as illustrated in Figure 16 should meet some of the field requirements. The basic unit can extract gases and determine the extraction rate as well as the total volume extracted. However, the unit has no capability to monitor and record environmental parameters (e.g., atmospheric pressure, radon level, subsurface pressure, etc.) and the state of the sampling station for documenting the quality of a particular subsurface gas extraction. *Given the potentially contentious nature of interpreting subsurface gas analyses, it is important to document sample quality with any available information obtainable from environmental and state-of-health information obtained either manually or automatically recorded by the Smart Sampling system.*

Sample Extraction-Automated/Triggered



Triggered sampling is appropriate for capturing gases from subsurface using either sample tubes or tarps:

- Barometric triggering used to sample during falling barometer to obtain maximized NG signal
- Continuous sampling/Barometric switching samples continuously switching between storage bladders as barometer rises and falls
- Radon monitoring is experimental but may have application for triggering and monitoring for leaks
- Pressure-drop measurements allow monitoring for changes in extraction process indicating potential quality issues

Figure 17. The automated smart sampling system shown is capable of operating in a number of extraction modes as indicated. It also allows environmental and potential diagnostic information to be recorded during the sampling process which may be useful for evaluating the quality of a sample, for example, whether or not atmospheric leaks were significant. The plot (top right) shows the strong anti-correlation between gas concentration reaching the surface (blue line) and atmospheric pressure (red line). Note: gas concentration history at a former underground nuclear test site was obtained using the tarp-and-tube surface extraction technique.

The automated smart sampling system that is currently in use is a suit-case-sized unit that is still portable and can be carried over uneven ground easily by two people (Figure 17.). The unit has five rather than the manual sampler's one sampling input readily allowing distributed subsurface sampling to be performed. Each of the five input sampling lines can be separately set for flow rate and individually monitored for flow rate, total volume extracted per line and flow pressure. In addition, the radon level of the total stream can be monitored with time. *Both the radon levels and pressure measurements can be used to infer the state of health of the sampling site during the sampling process. For example, a rapid decline in radon levels may indicate the existence of a leak or a sudden increase in atmospheric infiltration during the sampling process.* If a pressure sensor also shows a sudden decrease in line pressure required for a given flow rate, this supports the interpretation of a leak in one of the sampling lines or in the sealing of a sample point itself. Because the pressure is monitored in each of the smart sampler's five lines, it is then possible to identify the line or sample point in which the leak may be occurring. The smart sampler periodically records its location using an internal GPS system and also monitors for physical shocks as well as attempts to open it – two features pertinent to maintaining its physical security.

The smart sampler is intended to be straightforward to program from a touch-screen monitor. Connecting the touch screen externally allows programming or resetting of sampling parameters to be performed without opening the sampler unit, which is desirable to avoid during extreme weather. Besides the gas samples taken, sampler and sampling station data can be downloaded externally to a laptop or to a memory stick.

While the smart sampler can also be used to obtain grab samples, it was designed to function unattended at a site of interest during the course of a sampling campaign. It can be powered by car batteries, a small portable generator or by solar panels. Recent upgrades in Smart Sampler capability allow full monitoring and control of the Smart Sampling system usihg either satellite telemetry or 4G LTE cell phone technology.The addition of telemetry allows inspectors to monitor Smart Sampler state-of-health, environmental parameters, sample status and location from the Base of Operations.

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