EMSL-LV-539-7

NOBLE GAS SAMPLING SYSTEM

by
Monitoring Operations Division
Environmental Monitoring and Support Laboratory

U.S. ENVIRONMENTAL PROTECTION AGENCY
Las Vegas, Nevada 89114

MARCH 1977

This work performed under a Memorandum of
Understanding No. AT(26-1)-539
for the
U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

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ABSTRACT

A system to provide continuous monitoring for atmospheric concentrations of noble gases and tritium has been operated in the Nevada Test Site vicinity since 1972. The field sampling system was designed to utilize the analytical capabilities at the Environmental Protection Agency's Environmental Monitoring and Support Laboratory in Las Vegas. This report describes the noble gas system which provides sample collection and analysis for radiokrypton, radioxenon, and tritium in the form of methane, with detection capabilities, at the time of count, of about 2 picocuries per cubic metre.

INTRODUCTION

Perhaps the most difficult radionuclides to monitor in the environment are those that comprise the nonreactive gases. Releases of particulate and reactive gas radionuclides from testing of nuclear explosives or operation of nuclear power plants and fuel reprocessing facilities are normally well controlled. The nonreactive gases are the most likely to be released to the environment in measurable amounts. This paper describes a system developed by the Environmental Protection Agency's Environmental Monitoring and Support Laboratory in Las Vegas (EMSL-LV) to provide continuous monitoring for those radioactive gases. This system, plus a second system for the collection of atmospheric tritium as water vapor and hydrogen gas, make up the Noble Gas and Tritium Surveillance Network located at stations on and around the Nevada Test Site (NTS). Gases presently monitored by the noble gas sampling system include radiokryptons, radioxenons, and tritium as methane.

SAMPLER DESIGN

An analysis of the problem indicated that the most convenient way to collect the nonreactive gases was to collect a whole-air sample which could be separated for analysis in the laboratory. A laboratory technique had been developed making it possible to do quantitative analysis for the radionuclides of interest with 0.5 to 1 cubic metre (m^3) of air. The method selected to provide the simplest operation in the field was to compress the air sample for ease of transport. Because compressed air samples could not be carried by commercial carriers, they would have to be picked up and carried to the laboratory by members of the off-site radiological safety group. With the large area to be covered, the frequency of collection would have to be weekly. Because of a desire to collect a split sample for backup or duplicate laboratory analysis, it was decided to design for a collection rate of 2 m^3 per week, or about 3.3 cubic centimetres per second at standard temperature and pressure (STP, 0° C, 760 mm Hg). The air was to be collected as two samples of 1 m^3 each. The basic design called for a primary collection tank which would be filled steadily at the design flow rate. The

compressor used to pump air into the high pressure tanks would be periodically activated by pressure switches to remove air from the primary tank.

In the initial design, used for about 2 years, a fish aquarium aerator pump was used to pump air at the low flow rate into the primary tank. When the pressure reached 28 cm H_2O, the compressor was activated and evacuated the air until a vacuum of about 1 cm H_2O was reached.

In order to simplify the system, and remove one component subject to failure, the aerator pump was replaced by a limiting orifice. In the new design the compressor also serves as a vacuum pump. The differential pressure switches activate the compressor until the vacuum reaches about 50 cm Hg. Air bleeds into the primary tank through the limiting orifice, raising the pressure. At about 36 cm Hg vacuum the compressor is again activated. With the compressor now in use, the cycle provides for compressor operation for about 1 minute every 15 minutes.

The current design is shown in Figure 1. Air enters the primary tank through a 0.45-μm pore-size membrane filter and limiting orifice. The pressure differential switch maintains a vacuum between about 36 and 50 cm Hg. At the altitudes of the network stations, the vacuum in the primary tank will provide proper operation of the limiting orifice ($P_2 \leq 0.53\ P_1$; P_2 = absolute pressure in vacuum tank; P_1 = absolute pressure of ambient air). The three-stage Cornelius compressor used was initially built for aircraft use, with a 28-v d.c. motor. It has been modified with a belt drive from a 110-v a.c. motor. The compressor passes some oil, so an oil separator is used to prevent carry-over into the flexible lines and sample bottles. An in-line filter to remove oil, not shown in Figure 1, which has been retained from earlier attempts to solve the problem, follows the oil separator. Since it is not considered essential to the system, it is not included in Figure 1. A check valve prevents high-pressure air from leaking back through the compressor. The manifold provides for simultaneous collection of two tanks of air as a split sample. The sample pressure tanks are fitted with quick-disconnects for ease of connection to the flexible pressure lines from the manifold. Each tank has a volume of about 34.4 litres (2100 cubic inches). If the desired 1 m^3 of air at STP is collected in each bottle, the pressure will be about 3.0 megapascals (MPa) (427 psi). Under the different conditions of air density at the sampling

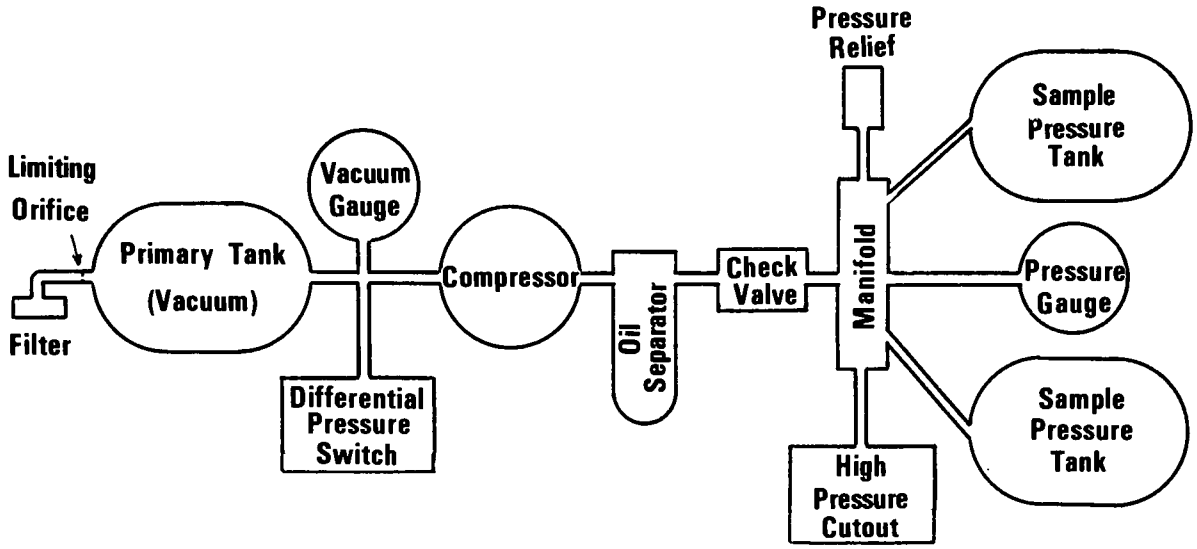

Figure 1. Noble Gas Sampler Schematic

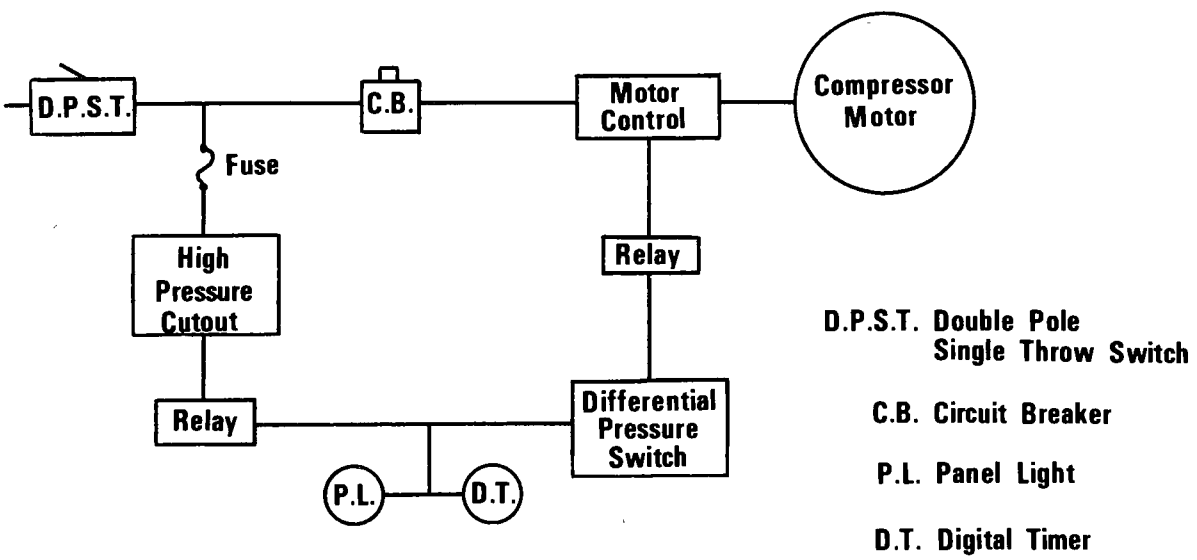

Figure 2. Noble Gas Sampler Electrical Schematic

stations, the actual volumes collected are usually somewhat below 1 standard m^3, with pressures less than 2.8 MPa (400 psi). The pressure tanks are periodically hydrostatically tested to 4.54 MPa (660 psi). In case some defect results in collecting air at too high a rate, a high pressure cutout prevents the pump from operating at pressures above 3.25 MPa (470 psi). As a backup safety factor, a pressure relief valve is set at 3.45 MPa (500 psi). The pressure of 3.25 MPa permits sampling periods longer than 168 hours, if necessary, while still providing an adequate safety factor.

Sample volume is determined from the net weight of air collected. The pressure tanks are evacuated in the laboratory and tare weights are measured. In the laboratory the full tanks are weighed and the net weight is divided by the weight of one m^3 of air (1293 g) to obtain the volume under standard conditions.

The electrical schematic is shown in Figure 2. The digital timer is reset to 0 at the start of each sample collection. It is connected so that if the high pressure cutout is activated, the timer will stop, showing the actual collection time.

SAMPLER OPERATION

Before connecting the sample pressure tanks, the operator starts the sampler to pull a vacuum on the primary tank. He then measures the time required for the vacuum to drop by 2.5 cm (1 inch) Hg. With the 34.4-litre tank, the vacuum drop should take about 6 minutes. A significantly longer time might indicate a partially obstructed limiting orifice. A shorter time may indicate a damaged orifice or a leak in the vacuum lines. Without the pressure tanks connected, the quick-disconnect fittings on the flexible lines retain the pressure generated in the lines during the initial operation of the compressor. While timing the vacuum drop, the operator also observes the pressure gauge reading to see that no decrease in pressure occurs. Any drop would indicate a high pressure leak. After checking the system and correcting any faults, the pressure is bled off the high pressure lines and the collection tanks are connected. A typical station is shown in Figure 3. The small refrigerator at the upper left houses the atmospheric tritium sampler.

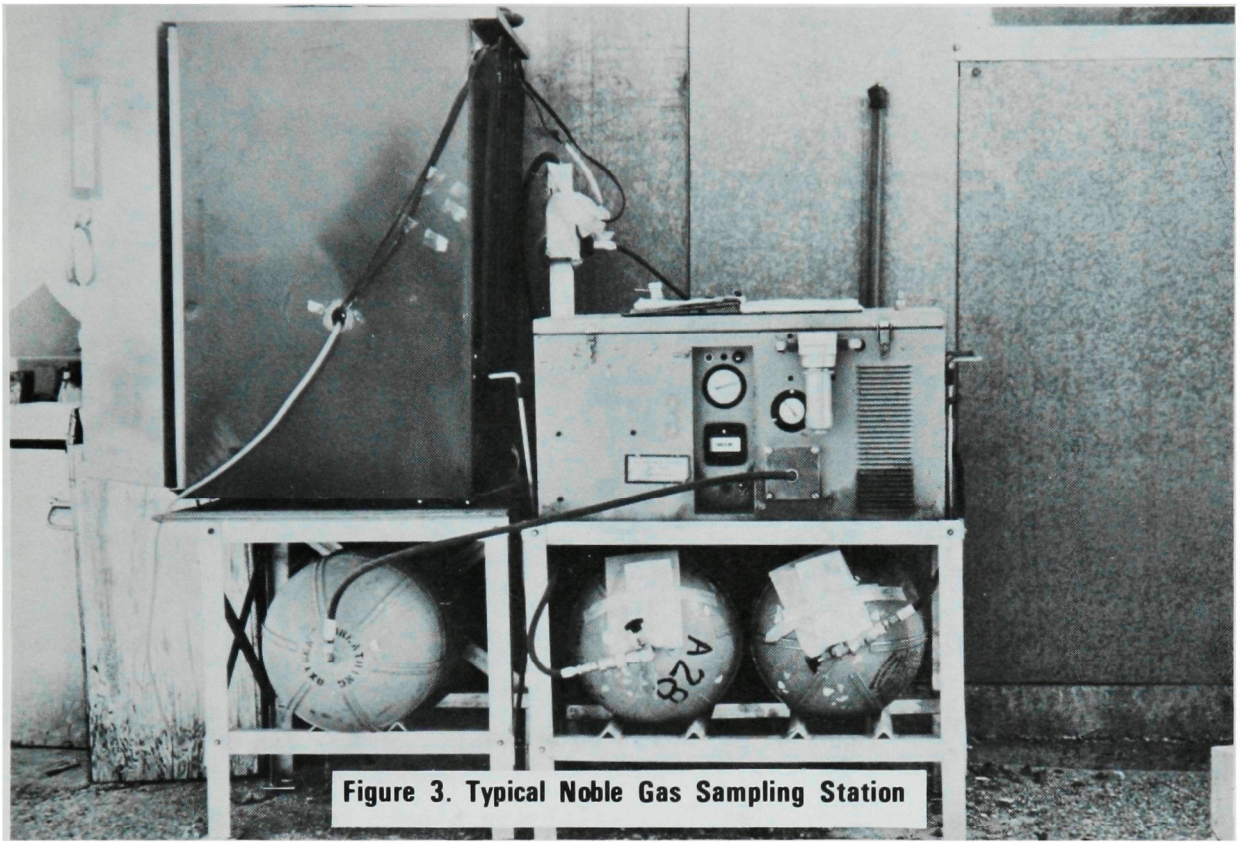

Figure 3. Typical Noble Gas Sampling Station

Beneath the refrigerator is the primary tank for the noble gas sampling system. The compressor and controls are contained in the case at the center and the two sample pressure tanks are located under it. An analysis of the results of the first year of network operation has been reported by Andrews and Wruble.[1]

SAMPLE ANALYSIS

The sample collection system was designed to utilize an analytical system developed at the EMSL-LV. This system (shown in Figure 4) was described in detail at the Noble Gases Symposium.[2]

It uses cryogenic and gas-chromatographic techniques to quantitatively separate the gas fractions of interest. The naturally occurring krypton and methane fractions, about 1.14 and 1 part per million, respectively, serve as sufficient carriers for those gases. One ml of stable xenon added to the pressure tanks before they are sent to the field serves as the carrier for that gas.

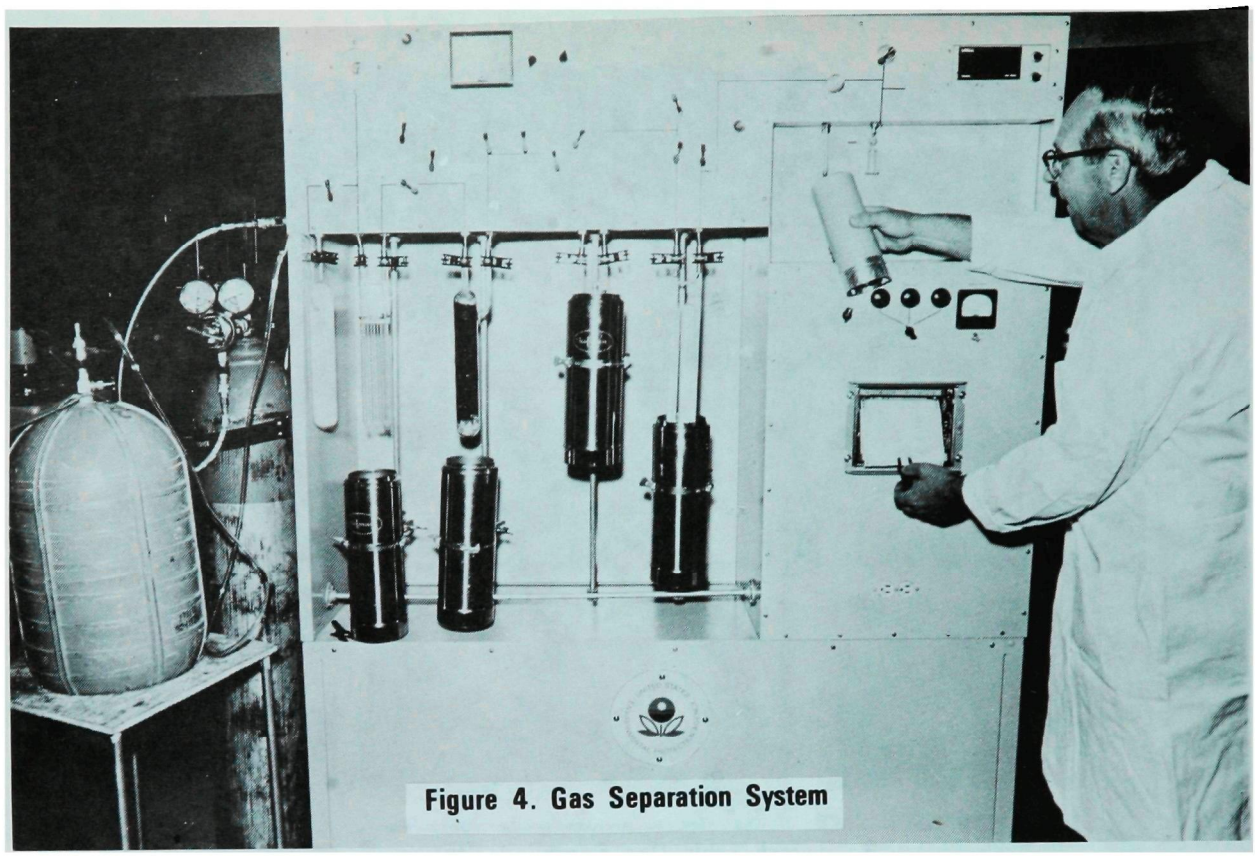

Figure 4. Gas Separation System

Three gas chromatographs in series (Figure 5) form the basic system. Thermistors T-1, T-2, and T-3, following each of the three chromatographic sections, provide indications of the passage of the various air sample components. Water vapor and carbon dioxide are collected in the molecular sieve trap on the inlet. Krypton, xenon, and methane are separated from most of the air sample on the activated charcoal column, C-1. The three gases, plus some other unwanted gases, are then carried to the second chromatographic column, MS-1, using helium carrier gas. Argon, oxygen, and most of the remaining nitrogen are eluted from this column and the sample gases are transferred to the third chromatographic column, MS-2. Krypton, xenon, and methane are separated from MS-2 and collected one at a time on column C-2, filled with activated charcoal. They are then driven from C-2 and

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[1] Andrews, V. E., and D. T. Wruble. "Noble Gas Surveillance Network, April 1972 through March 1973." Proceedings of the Noble Gases Symposium, Las Vegas, Nevada. September 24-28, 1973. CONF-730915. pp 281-289.

[2] Johns, F. B. "Portable Apparatus and Procedure for the Separation of Krypton, Xenon, and Methane in Air." Proceedings of the Noble Gases Symposium, Las Vegas, Nevada. September 24-28, 1973. CONF-730915. pp 225-238.

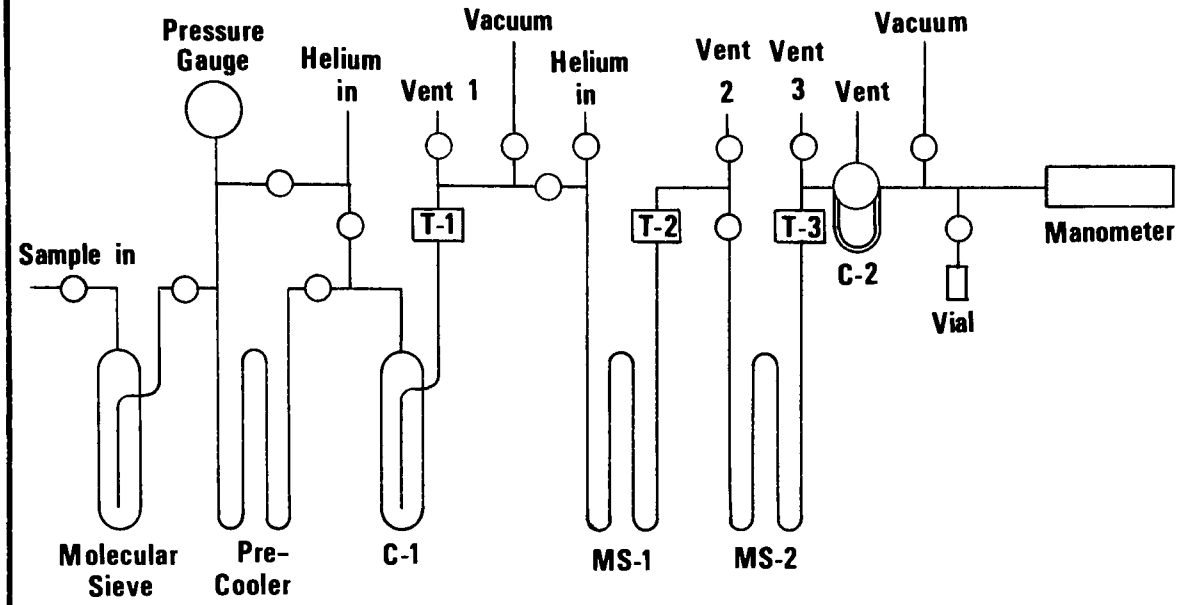

Figure 5. Gas Separation System Schematic

collected in evacuated liquid scintillation vials. The volume of gas recovered is calculated from the measured pressure, temperature, and known volume of the vial. Fraction recovered is determined from the volume recovered and the theoretical volume of krypton and methane in the measured air volume or known volume of xenon carrier added.

Degassed, toluene-based liquid scintillation cocktail is added to the vials and they are counted for radioactivity. Repeated counts are obtained to determine decay rate. This is used to confirm nuclide identification and, in case of a mixture of krypton or xenon radioisotopes, is used to quantitate the different isotopes by means of simultaneous equations based on individual half-lives.

Recoveries by this technique are about 85 percent for krypton, 70 percent for xenon, and 40 percent for methane. Detection limits, defined as that activity which gives a 2-sigma counting error of ±100 percent, are about 2 picocuries per cubic metre (pCi/m^3) for each gas.

OPERATING EXPERIENCE

Four years of operation have shown the utility of the total system. During 1975, the total success rate was greater than 95 percent. The reasons for failure to collect an adequate sample generally relate to the limiting orifice, compressor faults, or high pressure fitting leaks. Continual checking of performance in the field and performance of indicated maintenance have resulted in a steadily increasing success rate.

Limiting orifice problems have been caused by quality control in manufacture and by plugging. All orifices are now tested for compliance with the desired nominal diameter of 0.15 mm (0.006 inch), then in a measured flow test. Occasional plugging has occurred, even though filters have always been used. Whatman 41 cellulose filters were used previously, but seem to shed fibers which have been found in the orifices. Only membrane filters of 0.45-μm pore size are now used.

The compressor used in the sampler is a three-stage Cornelius compressor with a capability of 20.7 MPa (3000 psi). The pistons depend on close fits to the cylinder walls and lubricating oil for a seal. Using the first stage as a vacuum pump causes the introduction of some of the lubricating oil into the pressure lines. Pumping inefficiency caused by wear in the final two stages results in some sample loss. When a noticeable drop in sample pressure occurs which cannot be explained by low flow into the primary tank or by pressure fitting leaks, the sampler is changed and the compressor is submitted for replacement or repair.

Pressure fitting leaks sometime occur due to vibration-induced loosening of threaded connectors. The connectors have now been replaced with aircraft type fittings and such loosening is no longer a real problem. The pressure tanks are connected by means of flexible lines and quick-disconnect fittings. Compressor oil in the lines caused two problems. The oil tended to collect sand during sand storms which would work into the fitting when it was removed and replaced, causing leaks or plugging. The rubber O-rings which provide the seal in the quick-disconnects were found to be susceptible to damage by the oil. They occasionally worked loose and plugged the pressure tank or caused leakage. Installation of an oil separator and use of oil-resistant O-rings have eliminated those problems.

Sample losses in the laboratory are infrequent, but do occur due to unexpected response of the gas separation system or operator error. The process requires close attention for extended periods of time. Losses due to operator errors have generally been eliminated through experience on the part of the laboratory personnel.

Close cooperation between analytical, field, maintenance, and data reporting personnel has resulted in a reliable system capable of detecting and quantitating very low concentrations of gaseous radioactive fission products. The gradual increase of average ^{85}Kr concentrations in the atmosphere has been documented, along with a definition of the variations which occur in those concentrations.

At the current ^{85}Kr level of about 17 pCi/m^3, the 2-sigma counting error is about 1 pCi/m^3. Analysis of split samples has shown that laboratory analytical errors are of about the same value. The ambient concentration of radioxenon is essentially 0. Most measured concentrations of radioxenon are well above the detection limit, starting at about 10 pCi/m^3. The tritiated form of methane is rarely detected and may then be due to statistical variations in counting.

The system, as operated at and around the NTS, requires about 2 man-years of support. The extended network coverage requires 3 full days each week for collection. Analysis of 13 samples each week (11 stations, plus one duplicate sample and one random split sample for quality assurance) requires about 4 days The remaining 0.6 man-year of effort is spent in maintenance and data analysis and reporting.

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