AN ATMOSPHERIC TRITIUM AND CARBON-14 MONITORING SYSTEM

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PREFACE

This report contains procedures for gaseous tritium and carbon-14 sampling and analysis for the Source Term Measurement Project being performed by Idaho National Engineering Laboratory for the Nuclear Regulatory Commission. This work is being conducted for the Office of Nuclear Regulatory Research in support of requirements of the Effluent Treatment System Branch of the Office of Nuclear Reactor Regulation.

This project is designed to obtain generic source term information at operating light water reactors to update the parameters used in NRC calculational models (GALE codes). Detailed procedures and methods used for collection and analysis of gaseous tritium and carbon-14 are presented. This provides a reference base to supplement a series of reports to be issued by the Source Term Measurements Project which will present data obtained from measurements in specific nuclear power stations. Reference to appropriate parts of these procedures will be made as required.

ABSTRACT

Atmospheric monitoring of gaseous ^3H and ^{14}C in and around nuclear facilities is of interest because these nuclides may contribute to the environmental dose commitment. A sampling system has been designed to measure ^3H and ^{14}C at concentrations on the order of 10^{-6} pCi/cm^3 of air. This system, which may run unattended for periods up to two weeks at a controlled flow of 80 cm^3/min is small, portable, and only requires electrical power. The air sample is pulled through silica gel and molecular sieve adsorbent columns at a rate determined by small orifices. Part of the air stream passes through a heated catalytic oxidizer prior to the adsorbent columns. The adsorbent columns are returned to the laboratory for analysis. Tritium is counted as HTO, and ^{14}C as CO_2, by liquid scintillation techniques. The analysis reveals the concentration of these radionuclides originally present as HTO and CO_2, as well as the concentration in other forms which are oxidizable to these species.

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I. INTRODUCTION

The long term dose commitment due to the release of ^{14}C to the environment is significant. Limited data are available on the quantity of ^{14}C being released from light water reactors, and the data which are available are based on short duration grab samples (Ku 74). Since data on the release of activity from nuclear power plants are sometimes used (Sw 75) to estimate population radiation exposures, it would be desirable to be able to monitor the release over appreciable time spans, thus minimizing the effect of unusual conditions which produce unrepresentative short term concentrations. Also, implementation of the NRC philosophy of "as low as is reasonably achievable" in the nuclear power industry requires the knowledge of the levels of activity being released over long time spans under a wide range of operating conditions.

The measurement of ^{14}C is difficult because it decays with the emission of low energy beta particles (E_{max} = 150 keV) without accompanying gamma radiation. Counting of low activity ^{14}C samples has involved combustion of the sample to CO_2 and using the CO_2 (or converting it to methane or benzene) as the fill gas in a gas proportional counter. This technique has been applied to environmental samples (Dr 65), but the complexity of the sample treatment and expense of low level counting facilities have resulted in the development of other techniques for use in ^{14}C monitoring. Air bubbled through systems containing lithium, cesium, or barium hydroxide may be used to collect CO_2. The resulting carbonate precipitate may be counted as a suspension using liquid scintillation counting (Mo 70, Mo 71), or acidified and the released CO_2 reacted with ethanolamine in a liquid scintillation cocktail (Ca 72). Gases containing carbon compounds other than CO_2 may be passed through a heated catalytic oxidizer, and the resulting CO_2 collected (Go 75).

The desire to have a technique which would allow sampling for extended periods, preferably without operator attention, and the problem associated with the transportation of radioactive liquid samples from the sample site to the laboratory lead to the selection of solid adsorbents as the collection media of choice. Several solid adsorbents are available which will retain CO_2, however, in all cases moisture in the gas stream decreases the effective capacity of the adsorbent. In order to be able to predict the useful life of an adsorbent for CO_2, the sample stream must be moisture free. It is therefore a requirement that a dryer be included in the sampling system. It was decided to make use of this dryer for ^3H sampling.

Measurement of ^3H is most commonly accomplished by liquid scintillation counting techniques because it also decays with emission of low energy beta particles (E_{max} = 18 keV) without accompanying gamma radiation.

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HTO vapor may be adsorbed on a desiccant (Au 72), trapped in ethylene glycol, or collected cryogenically (Sh 75). Differentiation between HT and HTO may be achieved by passing part of the dried air stream through a heated catalytic oxidizer, then collecting the water formed by the combustion of the HT (Br 73). The HTO can be added directly to the scintillation cocktail (Os 70). Detailed discussions of ^3H counting techniques are contained in several review articles (Bu 74, Rh 73).

The sampling device described below is therefore capable of monitoring both ^3H and ^{14}C in air streams and is capable of separating the fully oxidized forms (HTO and CO_2) from oxidizable forms. The device makes use of existing technology and combines proven methods to meet the present requirements.

II. SOURCES AND MONITORING FOR ^3H AND ^{14}C AT NUCLEAR FACILITIES

Tritium is produced in the upper atmosphere by cosmic ray interactions, but most of the present atmospheric inventory of ^3H is the result of testing fusion weapons. Tritium concentrations in the environment vary somewhat with nearly all of it present as HTO. The molecular ratio HTO/H_2O for water vapor is about 8×10^{-17} (Bu 74). Consequently, the concentration of ^3H as water vapor is on the order of 10^{-6} pCi/cm^3 of air. Some ^3H is released to the environment from nuclear power plants, but much more is released during fuel reprocessing (Ja 68).

Carbon-14 is produced by the $^{14}N(n,p)^{14}C$ reaction in the upper atmosphere, with the ^{14}C primarily present as CO_2. Prior to 1952, the level of ^{14}C in the biosphere was 14 dpm/g carbon, which corresponds to 10^{-6} pCi/cm^3 of air assuming a CO_2 concentration of 315 ppm by volume. The atmospheric testing of nuclear weapons has increased somewhat the ambient level of atmospheric ^{14}C since 1952 (Dr 65). In light water reactors, ^{14}C is produced by neutron reactions with nitrogen and oxygen in the primary coolant (Ku 74). The ^{14}C may be present in an oxidizable form (CH_4, C_2H_6) or in a fully oxidized form (CO_2).

With some exceptions, air monitoring for ^3H and ^{14}C in and around nuclear facilities has not been as extensively performed as has monitoring for other nuclides or for ^3H as a liquid effluent. Tritium monitoring programs at a number of nuclear facilities are described in reference (Rh 73). The ^3H production facility at Mound Laboratory at Miamiburg, Ohio, has had a monitoring program for airborne ^3H for a number of years. Average values of HTO concentration detected in the vicinity of this laboratory during 1974 ranged from 10^{-5} to 10^{-4} pCi/cm^3 of air (Ca 75). A study of the ^{14}C release from the heavy water production reactors at the Savannah River Plant has been recently published (Ha 77). Several light water power reactors have been studied with respect to the gas phase concentration of ^3H and ^{14}C (Ma 73). The activity levels for both of these nuclides at light water reactors were generally in the range

10^{-4} to 10^{-2} pCi/cm^3 of air, although in some samples the levels ranged as high as several pCi/cm^3 of sample. The major fraction of ^{14}C detected was in the form of CO_2 from BWR's and as CH_4 from PWR's. Similarly, a study of several PWR reactors (Ku 74) indicated that more than 80% of the gaseous ^{14}C was present as CH_4 or C_2H_6. These studies at both the BWR's and PWR's were based on small samples taken over a relatively short time span.

III. SAMPLER DESCRIPTION

The sampling system, designed to operate unattended for periods up to two weeks, draws air through a series of adsorbents which collect the ^{3}H and ^{14}C. The adsorbents are returned to the laboratory for analysis. The system allows differentiation between ^{3}H and ^{14}C present in their fully oxidized forms (HTO, CO_2) or as the other compounds (HT, CO, hydrocarbons) which are oxidizable. The sampling system requires no services other than AC power, and is light, compact, and easy to use. Other radioactive gases present in the sampled atmosphere may be discriminated against during sample collection and/or during sample analysis.

The sampling system is diagrammed in Figure 1 and shown in Figure 2. The inlet air may be derived from stacks or duct through sampling probes, or drawn in directly from the atmosphere. The air passes through a particulate filter, then through a silica gel column which adsorbs H_2O (and HTO). The air stream then is split, with half going through a molecular sieve column, and half through a catalytic oxidizer. The molecular sieve adsorbs CO_2, CO, and light hydrocarbons, and the catalytic oxidizer causes all oxidizable species containing ^{3}H and ^{14}C to form HTO and $^{14}CO_2$, which are then sequentially adsorbed on silica gel and molecular sieve columns. A unique feature of the sampling apparatus is the use of critical flow orifices to regulate the rate of gas flow through both sections of the sampler. These orifices are made from 15/32-inch diameter discs of 0.008-inch stainless steel, with centered holes approximately 0.004 inches in diameter. The holes are drilled with a laser beam, and the orifices tested to find pairs with similar flow rates. Controlled flow rates around 80 cm^3/min are obtained so long as a vacuum of at least 300 torr is maintained on the pump side of the orifice. Use of these orifices eliminates the need for rotometers and metering valves in the sampler. Rather surprisingly, there has been almost no indication of clogging of the orifices during sampler usage. The adsorbent columns are constructed of 2-inch aluminum pipe, threaded, and capped with bell reducers, and inter-connected with 1/4-inch stainless steel tubing by means of Cajon or Swagelok connectors. Each column contains about 250 g of adsorbent (6-16 mesh silica gel from Davison, or 4 A molecular sieve beads, 8-12 mesh from Davison) confined in a close-packed bed by means of screens and snap rings. The catalytic oxidizer contains a 4-inch bed (about 14 g) of Pt-Pd catalyst on alumina pellets (Engelhard Industries) in a 1/2-inch tube heated by a small tube furnace to about 550°C. Air is drawn through the sampler by a Cenco HyVac vacuum pump.

After the desired amount of air has been processed through the sampler (usually about 2.5 m^3 in a two-week period), the adsorbents are returned to the laboratory for analysis. Usually the silica gel is transferred to plastic bottles and the columns refilled in the field, while the molecular sieve columns are sealed and returned intact. Replicate analysis of aliquots of silica gel taken from the plastic bottles over a six-month period showed that tritium exchange is not a problem.

IV. ANALYSIS DESCRIPTION

Analyses of the molecular sieve columns are performed using the system diagrammed in Figure 3 and shown in Figure 4. The cylinder is heated in a tube furnace for about 4 hours at 350°C with a dry He purge in order to remove the adsorbed CO_2. Exit gases from the cylinder pass through an ice-cooled trap to remove any water vapor, and then through the spiral traps at liquid nitrogen temperature in which the CO_2 is condensed. The CO_2 then is expanded into a bulb of calibrated volume and its pressure measured, thus allowing calculation of the amount of CO_2 collected. A portion of the sample (usually about 150 cm^3 at STP) then is transferred to a glass scintillation vial which is attached to the vacuum line and which contains 3 ml of ethanolamine. Adsorption of the CO_2 into the ethanolamine is indicated by the system pressure drop, and requires only a few minutes. The vial is then removed from the vacuum line, and the sample prepared for counting by adding 7 ml of methanol and 12 ml of scintillation cocktail (Insta-gel from Packard Instrument Company). The molecular sieve column is cooled, filled with He, sealed, and then is ready for reuse. Since molecules other than CO_2 (light hydrocarbons, CO, NH_3, oxides of nitrogen and sulfur) may be adsorbed on the molecular sieve exposed in the sampler to the unheated air, MS-1, some impurities may be present with the CO_2 collected during the bake-out procedure.

The presence of other molecules does not cause problems because they are partially discarded during movement of the CO_2 from the first spiral trap to the calibrated volume. The CO_2 is moved by cryogenic pumping, that is, liquid nitrogen is placed around the part of the apparatus into which the analyst wishes the CO_2 to be condensed and the initial location of the solid CO_2 is allowed to warm. Prior to starting the movement of the CO_2, the whole vacuum line is evacuated to less than one torr and the vacuum pump is isolated from the system. Other gases do not move at the same rate as the CO_2, and if significant quantities of other species are present, a residual pressure will remain in the system after the bulk of the CO_2 has condensed. This gas is pumped out of the system. Two successive steps are required to move the CO_2 from the first spiral trap to the calibrated volume. In each step some decontamination from other species can occur. In addition, the final transfer into the counting vial depends on the chemical reaction of the gaseous CO_2 with the ethanolamine in the vial. Other species do not react and therefore only CO_2 is counted by this procedure.

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During the movement of the CO_2 in the cryogenic pumping steps a portion of the desired gas may be pumped away along with other species. This ensures that the CO_2 which is finally reacted with the ethanolamine is of the highest purity. The loss of a portion of the CO_2 in the analysis does not jeopardize the results since isotope exchange has been accomplished in the bake-out procedure and the calculation of the activity in the sample is based on the quantity of CO_2 actually reacted in the counting vial.

Three options are available for the analysis of the silica gel. The first option involves the placing of the silica gel in a bake-out apparatus similar to the one used for the molecular sieve columns. The oven need not be operated above 200°C. The released water vapor is condensed and aliquots are counted. This procedure has the greatest sensitivity but is also the most time consuming and therefore the most expensive. The second option has a slightly lower sensitivity than the first and is somehwat less time consuming. An aliquot or the entire sample is washed with low ^3H content water such that exchange between the wash water and the adsorbed water takes place. An aliquot of the wash water can then be counted.

The third alternate and the one which is used on all samples at least initially involves taking replicate 5-g portions from the weighed and well-mixed silica gel sample. These portions are placed in individual glass scintillation counting vials, treated with 3 ml of low ^3H content water, 14 ml of counting cocktail (Ready-solv HP from Beckman Instruments Co.), and counted directly. Glass vials are preferred because under certain circumstances loss of activity from polyethylene vials may occur (Th 77).

This approach of counting the ^3H on the silica gel is the one which has been used in previous studies (Pe 73) and has the advantage of being the least time consuming. It has the disadvantage of being the least sensitive of the three methods for analysis of the ^3H, however, over 90% of the original sample is still available for use in one of the more sensitive methods if required.

The vials containing the prepared samples of ^3H and ^{14}C are counted in a Packard Model 3385 refrigerated liquid scintillation counter which has three independently controlled counting channels. The three channels are set for optimum counting of the ^3H, ^{14}C, and any higher energy beta emitters, respectively. It is important that three channel counting procedures be used. The use of standards allows for the knowledge of the expected ratio between the various channels for each nuclide of interest. If the channels ratios are not as expected the presence of other nuclides is indicated, and the results of the analysis are incorrect and must be discarded.

When a series of samples is being analyzed, blanks are prepared and run along with the samples. The blanks consist of low ^3H content water and CO_2 from a gas cylinder along with the counting cocktails for each

nuclide. In addition, since the counting efficiency in a liquid scintillation system depends on the quenching in the sample, a quench curve is constructed. A quench curve is essentially a working curve which relates the counting efficiency to the amount of quenching in the samples. The Packard Model 3385 counter contains two external sources, one of ^{137}Cs and the other of ^{241}Am; each one is positioned under the sample vial and an external standard ratio (AES) is computed for each sample as it is counted. The AES ratio is a measure of the quenching in the sample. Several standards are prepared, all with the same activity level, in solutions identical to those of the samples. Varying amounts of materials which are known to cause quenching are added to each standard. Methanol is used for ^{3}H and chloroform is used for ^{14}C. These standards are counted and the quench curve constructed (AES vs. efficiency). The measured AES ratio for each sample then is used to determine the counting efficiency for that sample. Typically, ^{3}H and ^{14}C are counted with 22% and 55% efficiency, respectively.

The calculations for the two nuclides are performed in slightly different manners. The ^{3}H results are calculated from the duration of the sample collection, the orifice flow rate, weight of silica gel, weight of aliquot, and the blank corrected disintegration rate per aliquot. If the replicate aliquots do not agree within 10% or within the counting statistics, whichever is higher, new aliquots are taken for the remaining original sample.

The ^{14}C results are based on the actual amount of CO_2 reacted in the counting vial. This is obtained from a volumetric measurement of the gas present in the calibrated volume before and after reaction with the ethanolamine. The calculation is based on the amount in the vial because as mentioned previously a small portion of the total CO_2 may be discarded. Also, the total amount of CO_2 collected in a two-week sample would be far too large to react with 3 ml of ethanolamine. Use of larger amounts of ethanolamine would result in excessive quenching during counting. The blank corrected disintegration rate is obtained per cm^3 of CO_2 in each counting vial, and this value is multiplied by the abundance of CO_2 in air (315 ppm) to obtain the activity per cm^3 of air.

A check on the performance of the system insofar as the ^{14}C portion is concerned is available from an analysis of the results. The incoming air stream is equally split between the two molecular sieve columns. In the analysis of the first adsorber (MS-1) only CO_2 is counted. In the analysis of the second adsorber (MS-2) CO_2 plus an oxidizable species containing ^{14}C is counted. Therefore, the results from MS-2 must be equal to or higher than the result from MS-1 if the system is working correctly.

The sensitivity for both nuclides is on the order of 10^{-6} pCi/cm^3 of air.

V. SYSTEM TESTING

The prooftesting of the design concept has been accomplished in several ways. Initially, air was introduced into the sampler and the CO_2 recovery calculated from the volume of CO_2 recovered and an assumed air concentration of 315 ppm CO_2. Numerous runs gave greater than 90% recovery and indicated that the sample collection and analysis concept was adequate to meet the needs of the monitor system. Methane and air mixtures were then introduced into the sampler to test the oxidizer. The quantity of CO_2 recovered from the MS-2 adsorber indicated complete oxidization of the methane. Other gaseous hydrocarbons were not tested since methane is the most difficult of the hydrocarbons to oxidize.

Radioactive tracers of HTO and $^{14}CO_2$ were introduced into the sampler along with ambient air for periods ranging from two days to fourteen days. The recovery of ^3H averaged $98 \pm 10\%$ and of ^{14}C $98 \pm 6\%$ for 11 runs where the error is one sigma. Laboratory studies were not conducted on HT or tritiated hydrocarbons. Previous investigators (Os 70) have proven the efficiency of noble metal catalysts for oxidation of HT in air, although some difficulty was encountered in removing extremely small amounts of HTO from the adsorption bed. In our sampler, the catalyst is operated at high temperature and the HTO produced is not removed from the adsorbent bed, but counted _in situ_.

VI. SYSTEM LIMITATIONS

Proper operation of the sampling system requires the presence of sufficient oxygen in the sample stream to oxidize any reduced species present. The oxidizer used is a catalytic one and although vastly oversized for any reasonable situation, it does not have any inherent oxidizing power in the absence of oxygen. The system will not operate as designed if oxygen is not present or is not present in sufficient quantity. This restricts the usefulness in certain situations in light water power reactor monitoring. An example would be in PWR's, where waste gas decay tanks are expected to contain high concentrations of both ^3H and ^{14}C and very low concentrations of oxygen. Use of the sample directly would result in low values for the oxidizable portion of the sample for both nuclides. Since waste gas decay tank releases are periodic in nature, long term sampling is not a requirement. Grab samples can be taken in gas sampling cylinders or bombs and returned to the laboratory where the contents can be mixed with oxygen or air and then processed through the sampler. Samples of waste gas decay tanks taken from the point of release are valid in most cases because of the dilution air in the stack or release point.

The sampler has failed to operate as designed when used to sample a stack which contains appreciable quantities of NO_x (500 ppm). The NO_x competes for adsorption sites on the molecular sieve with the CO_2.

To compound the problem this particular stack contains tens of thousands of ppm CO_2 rather than the 300 ppm design level. It has not been established if the system will not function in such a stream or if the useful life is too short to be effective. This illustrates the problem that if a sample stream contains grossly higher than the design level of CO_2 or other species which are strongly adsorbed by the molecular sieve an evaluation of the useful sampling duration must be made.

A final example of operation outside the design concept has occurred on one occasion. The sampler is designed to retain the ^3H species on the silica gel and the ^{14}C species on the molecular sieve. A sample was taken from a PWR waste gas decay tank which contained sufficient oxygen to oxidize the hydrogen present. Prior to the analysis in the laboratory significant amounts of ^{133}Xe were observed on the SG-1 adsorbent and lesser amounts on the SG-2 adsorbent. In this case an aliquot of the silica gel was placed in a boiling flask fitted with a reflux condenser and low ^3H water was added. The mixture was heated to boiling while a stream of He was passed through the colution to sparge out the Xe. Counting of an aliquot of the water indicated the ^{14}C was present in the silica gel.

The presence of ^{14}C was interpreted to mean that a portion of some organic species had been retained by the silica gel adsorber. There was no ^{14}C on the SG-2 media. The species did not even reach the oxidizer. The species was inferred to be of high molecular weight because of two facts. First, light carbon compounds such as CO_2 and CH_4 were proved not to be adsorbed by the silica gel and second, light hydrocarbons would have been sparged from the boiling flask during the Xe decontamination.

In an attempt to verify the performance of the sampler with heavy hydrocarbons, ^{14}C was introduced as labeled toluene. Direct analysis of the silica gel using in situ counting methods previously described indicated that there was ^{14}C on the first silica gel column (SG-1). Standards containing ^{14}C on silica gel were prepared and quench curves were constructed. Using the three channel counting method, channels ratios were calculated for pure ^3H and ^{14}C. Solving simultaneous equations allowed for the calculation of the ^{14}C present on the silica gel independently of any ^3H which was present. The recovery of the ^{14}C-toluene was 100%.

The sampling system was designed to oxidize hydrocarbons to CO_2 and H_2O, and does function that way for light hydrocarbons. For heavy hydrocarbons which do not reach the oxidizer but are adsorbed on the silica gel, analysis is still possible when the silica gel is counted directly.
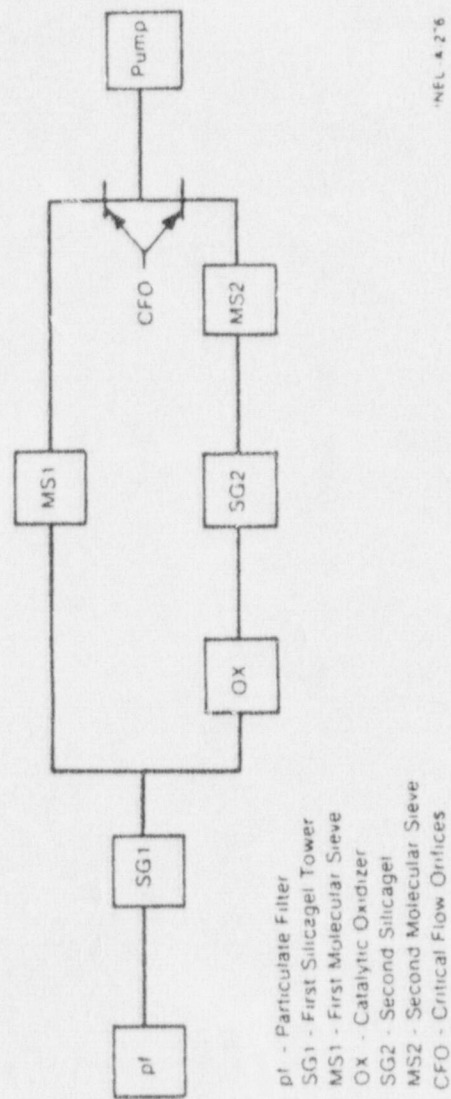
pf - Particulate Filter
SG1 - First Silicagel Tower
MS1 - First Molecular Sieve
Ox - Catalytic Oxidizer
SG2 - Second Silicagel
MS2 - Second Molecular Sieve
CFO - Critical Flow Orifices

INEL A 276

Figure 1. Schematic for ^{3}H and ^{14}C Sampler
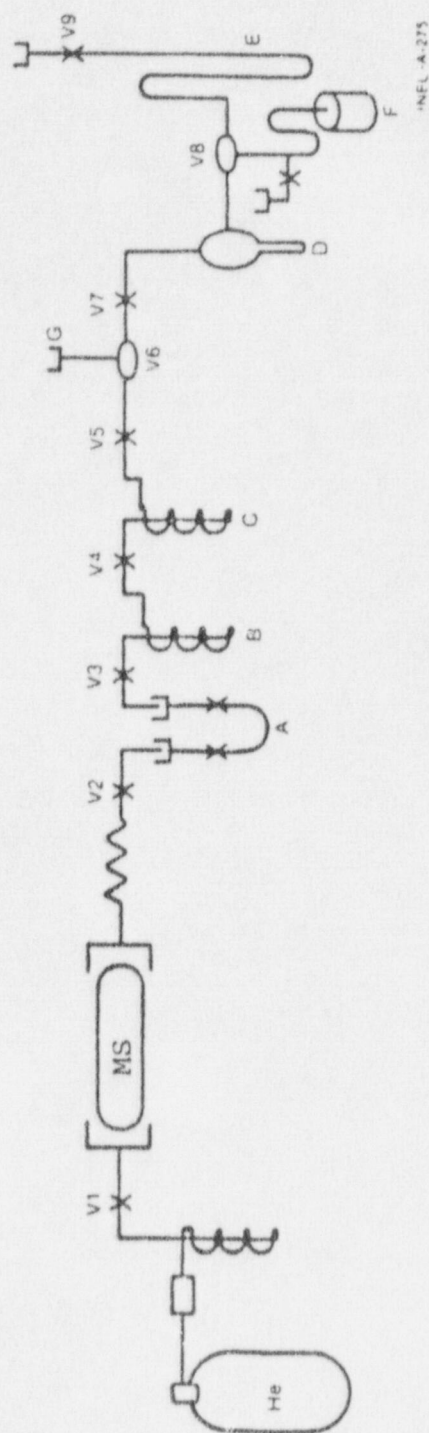
Figure 2. Sampler System

Figure 3. Removal System for Eluting ^{14}C from Molecular Sieve Towers
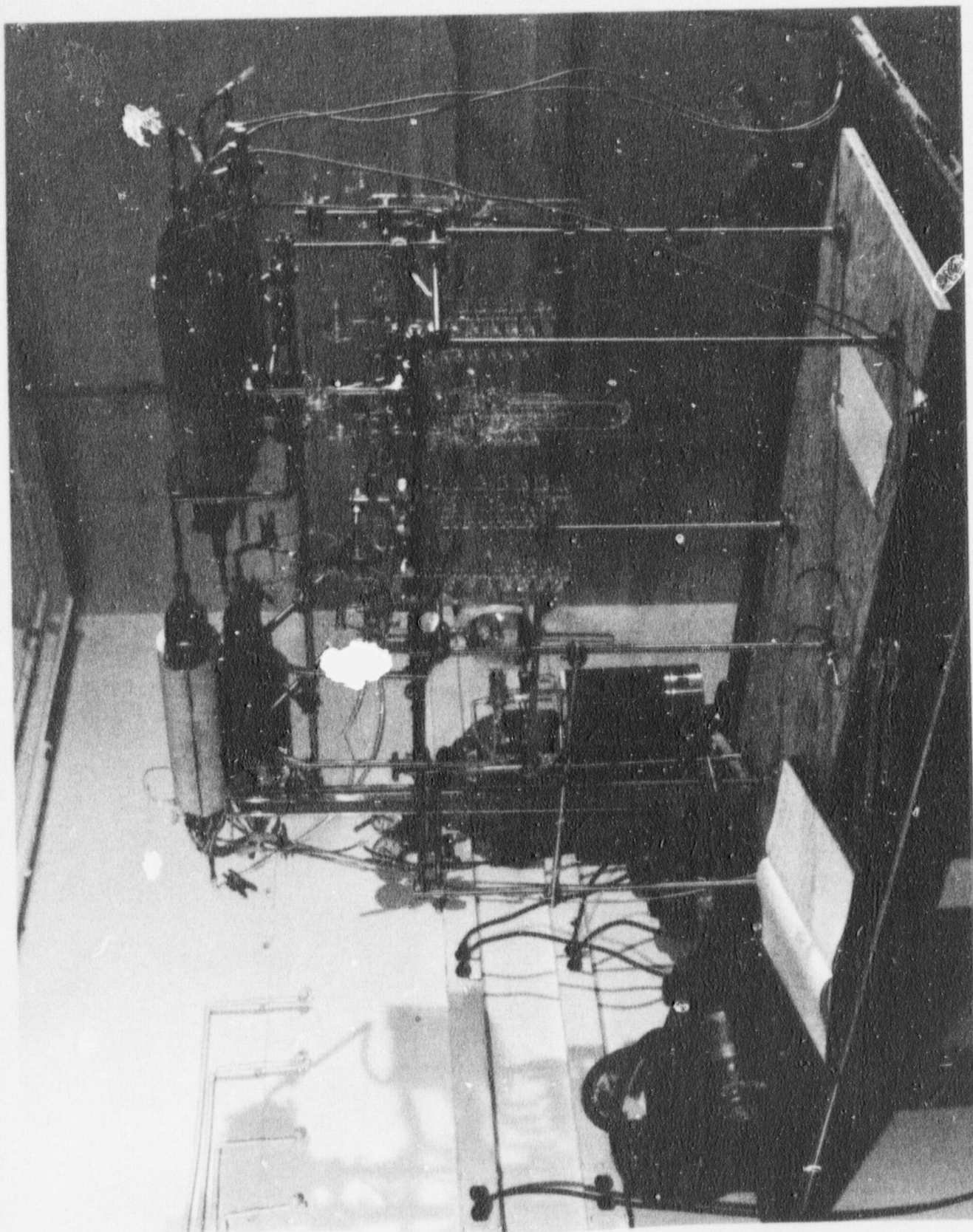
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Figure 4. Analysis Apparatus

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Atmospheric monitoring of gaseous ^3H and ^{14}C in and around nuclear facilities is of interest because these nuclides may contribute to the environmental dose commitment. A sampling system has been designed to measure ^3H and ^{14}C at concentrations on the order of 10^{-6} pCi/cm^3 of air. This system, which may run unattended for periods up to two weeks at a controlled flow of 80 cm^3/min is small, portable, and only requires electrical power. The air sample is pulled through silica gel and molecular sieve adsorbent columns at a rate determined by small orifices. Part of the air stream passes through a heated catalytic oxidizer prior to the adsorbent columns. The adsorbent columns are returned to the laboratory for analysis. Tritium is counted at HTO, and ^{14}C as CO_2, by liquid scintillation techniques. The analysis reveals the concentration of these radionuclides originally present as HTO and CO_2, as well as the concentration in other forms which are oxidizable to these species.

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