LA-7321-MS

Informal Report

Ln. 390

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**Heavy Methanes (Mass-20 and -21) as
Atmospheric Tracers**

A New Research Initiatives Project

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

LA-7321-MS
Informal Report
UC-11
Issued: July 1978

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A New Research Initiatives Project

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HEAVY METHANES (MASS-20 AND -21) AS ATMOSPHERIC TRACERS

A NEW RESEARCH INITIATIVES PROJECT

Compiled by

Paul R. Guth 's and Malcolm M. Fowler

ABSTRACT

Products from the stable isotope production project (ICONS) at the Los Alamos Scientific Laboratory (LASL) have had applications in a number of practical problems. Fully deuterated methanes, using ^{12}C and ^{13}C (mass-20 and -21, respectively), are detectable in the atmosphere at very low concentrations. The detection limits are due to an almost zero background and a distinct mass-spectrometric line. These properties, coupled with long lifetimes in the atmosphere, make the heavy methanes strong candidates for tracers of atmospheric transport and turbulence on regional to global scales.

Three field tests have been conducted to assess the feasibility and to demonstrate the desirable properties of heavy methanes as atmospheric tracers. In the first test in May 1974, heavy methane released at the Idaho National Engineering Laboratory (INEL) was sampled and detected in several midwestern cities up to 2500 km downstream. These results agree with trajectories estimated from observed meteorological data. A second experiment, conducted in December 1975 at the Savannah River Plant, compared two heavy methanes with SF_6 and ^{85}Kr over a transport distance of 100 km. Increased detail in meteorological support data and ground-level sampling offered options of data interpretation unavailable in the first test. Qualitative patterns of tracer concentrations show good agreement among the four tracers and are verified by the meteorological inputs. The third experiment, performed in April 1977 at INEL, built on the experience of the previous two. Detailed ground-level sampling arrays at 3, 50, and 90 km from the release point were supplemented by aircraft and a crude sampling line across the Continental Divide at 300-km distance. Two new perfluorocarbon tracers were included for comparison.

This report covers the activities of several LASL groups in the design, implementation, and interpretation of heavy methane tracer experiments, as well as the development of this tracer system, for application to various atmospheric transport problems.

SUMMARY

The research conducted by H Division (H-8)* and CNC Division (CNC-2, CNC-4, and CNC-11) as part of the New Research Initiatives Program (NRIP) involved the development of experimental field project techniques, experimental apparatus, atmospheric transport and diffusion models, and analytical procedures. These groups participated in designing the field experiments. Attention was given to determination of data required for model proof-testing, how to obtain these data from the experiments, the desired experimental meteorological situations, and experimental apparatus to obtain the desired data. The two most recent field experiments in 1975 and 1977 were useful in pinpointing the required data for model application. Additionally, the planning and execution of these field experiments have identified the weak, as well as the strong, aspects of our approach. We have demonstrated the soundness of the tracer release schemes developed as a part of this research. Prominent in the successes of this research effort has been the excellent analytical sensitivity levels for methane-20 and -21 ($^{12}\text{CD}_4$ and $^{13}\text{CD}_4$) in the atmosphere. The gas mass-spectrometric techniques employed by Norman Daly of the British Atomic Weapons Research Establishment (AWRE) have produced reproducible measurements of $^{12}\text{CD}_4$ as low as two parts in 10^{16} parts of air and one part of $^{13}\text{CD}_4$ in 10^{16} parts of air. Evaluation of these experiences leads one to demand foremost, a greater measure of care to be taken in the storage, handling, and processing of tracer materials and postrelease samples. To attain improved sensitivity levels of these tracers in the atmosphere (a few parts in 10^{17} parts of air) will require even more stringent contamination prevention procedures than those routinely applied in ultra low level radiochemical procedures.

Peripherally related areas of interest investigated as a part of this project include nonmethane atmospheric tracers, analytical instrumentation (real time),

* G-8 as of December 1, 1977.

alternate test areas, and other potential applications for tracers with equal measurement sensitivity and lifetime. Intercomparisons of several potential tracers (SF_6 and perfluorocarbons) have revealed the need to use tracers that complement each other in solution of various experimental and natural constraints.

Among the strongest attributes of the heavy methanes are their high measurement sensitivity in the atmosphere, and relatively low natural abundance (<1 part in 10^{20} parts of air) when compared to other tracers (i.e., SF_6 background, 0.5 - 0.9 parts per trillion in air). They also fulfill the requirement to release small amounts of tracer (grams vs kilograms of other tracer compounds) with the attendant less complicated release mechanisms.

The Los Alamos Scientific Laboratory (LASL) is presently being supported by the Division of Biomedical and Environmental Research (DBER), Department of Energy (DOE), for a continuance and enlargement of the research effort begun with NRIP assistance. This new programmatic support will allow us to build and operate locally a state-of-the-art gas mass spectrometer similar to the AWRE machine. It will also eventually provide support for more detailed research and data application to atmospheric transport and diffusion studies. These studies will provide atmospheric model refinement which will contribute directly to the understanding of environmental impact as a result of energy use and energy technology advances.

I. INTRODUCTION

An increasing number of practical problems require an understanding of atmospheric transport and dispersion of airborne pollutants on regional, continental, or global scales. The development of the requisite knowledge depends on the collection of experimental data in which the transport and diffusion processes can be isolated from source and sink factors. Most of the existing data were derived from effluents from multiple sources subject to deposition, precipitation scavenging and chemical transformation along the travel path. An inert, nondepositing tracer is needed to isolate the effects of long-range transport and diffusion. Heavy methanes, $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$, are two such tracers that possess these desirable characteristics.¹

A suitable tracer for this application should have a residence time in the atmosphere of at least several weeks for local investigations to several years for a global experiment. It should be nontoxic and not harmful to the environment; it should be relatively inexpensive and the cost of sample analysis should not be prohibitive; it must be detectable at very low concentrations and there should be negligible background in the atmosphere. It is desirable, in addition, to have simplicity of tracer release and sample handling, a capability for short-duration sequential sampling, and rapid analytical "turn around" for data reduction.

An important programmatic goal of this effort has been to develop suitable tracer systems, test them under field conditions, and ultimately to conduct tracer experiments to test pollution models on all scales of interest.

An early feasibility experiment^{2,3} was undertaken using a methane-21 ($^{13}\text{CD}_4$) release from the Idaho National Engineering Laboratory (INEL), May 1974. This experiment demonstrated the appropriateness of using this tracer at extended ranges (1500-2500 km) with very small amounts being released (~100 g).

A second experiment⁴ was performed as a part of an ERDA-supported effort to further study atmospheric transport and diffusion. This experiment was conducted at the Savannah River Plant (SRP) in December 1975, with the assistance and cooperation of several participants. Prominent among those groups were personnel from Savannah River Laboratory (SRL), Air Resources Laboratory (ARL) of the National Oceanic and Atmospheric Administration (NOAA), Brookhaven National Laboratory (BNL), and LASL. In addition, groups from Airco, Inc., and EG&G, Inc., of Las Vegas, Nevada, were active participants.

The experimental objectives for the SRP tests were (1) to determine whether comparative results could be obtained from the simultaneous releases of tracers SF_6 , ^{85}Kr , and heavy methanes; (2) to study the transport and diffusion of plant plumes over a 100-km range; and (3) to gain additional field experience for the simple tracer-release schemes, sampling, and tracer-analysis before undertaking more complex experiments. These objectives were, to a large measure, satisfactorily accomplished. Several facets of cooperative experiments were thoroughly tested and this concept of conducting field activities appears to be sound. It is desirable to have LASL management of such projects.

A third tracer-release experiment, one involving a larger number of LASL and non-LASL scientists and technicians, was conducted in April 1977 at INEL. The groups represented at the INEL experiment included NOAA personnel of ARL from Silver Springs, MD, and INEL, as well as those from the Environmental Protection Agency (EPA), Las Vegas, NV; National Weather Service (NWS), Las Vegas, NV; ERDA's Health and Safety Laboratory (HASL) (now the Environmental Measurements Laboratory of the US DOE); Allied Chemical at INEL, Airco, Inc.; BNL, and LASL. The complexity of this experiment has complicated and delayed early availability of experimental data. It is anticipated that this latter work will be reported in more detail when the data, their analysis and evaluation, are complete.

II. EXPERIMENTAL SITES

A. Savannah River Plant - 1975

This experiment was conducted as a part of a larger project. These tracer releases were from the SRP complex, see map of area (Fig. 1). The methane-20 was released through a tube to the top of a 61-m (200-ft) tower near A-site while the $^{13}\text{CD}_4$ was released into a stack port near the top of a 61-m (200-ft) stack at H-area. Fig. 2 locates the release points within the SRP facility. The primary samplers were located along two highways, US Interstate 95 (I-95) and South Carolina State Road 336 (SC-336). The samplers were placed along these roads, a nominal 90 km from the tracer-release points, at approximately 1.25-km intervals. Secondary samplers of a cryogenic type were permanently emplaced at the approximate locations, also shown in Fig. 1.

The operational control center was situated in the office area occupied by members of Todd Crawford's staff at SRL. On-line SRL meteorological tower and Facsimile Readouts were used by operational control personnel. The sampler apparatus preparation, special meteorological instrumentation, and initial air-sample

processing were accomplished in laboratory space near the control center. These experimental support activities were all accomplished at the main ERDA portion of the SRP complex at A site shown in Fig. 2.

Aircraft real-time measurements of ^{41}Ar were made and recorded at altitudes ranging from 0.24 km to 0.7 km above mean sea level (MSL). Delayed "real-time" SF_6 measurements were made at several predetermined altitudes in the 0.24- to 0.7-km region. Whole-air samples for heavy methane, SF_6 , and ^{85}Kr analysis were collected by the aircraft. A background air sample was collected before release day over SRP. A second air sample was gathered on a southwesterly heading (pass 1) as shown in Fig. 3. The third and fourth aircraft samples (passes 8 and 11) were collected in the vicinity of I-95 as shown in Fig. 4.

The topography of the SRP area can be generally classed as flat plains to gently rolling low wooded hills. Cleared areas are generally dedicated to agricultural activities. The forested regions are covered with typical southern pines and low brush. The lower lying portions along the Savannah River and local streams are large swampy areas covered with deciduous plant life of varying heights.

B. INEL Experiments-1974 and 1977

1. 14 May 1974 Experiment. The methane release site was geographically the same in both experiments at INEL. The first experiment with its long-range emphasis differed primarily from the 1977 experiment in its sampler emplacement (Fig. 5). The sampler network for this early investigation was established as a part of a NOAA/ARL air-sampling program.⁵

2. 19 April 1977 Experiment. The third experimental tracer release was made at INEL. The overall experimental area is shown in Fig. 6. The $^{12}\text{CD}_4$, $^{13}\text{CD}_4$, and the comparison tracers (SF_6 , perfluorocarbons) were released at sites shown in Fig. 7. Various "close-in" samplers and real-time tracer measuring devices were deployed along arcs A, C, and D. The nominal distances to these arcs are 3, 50, and 90 kilometers, respectively. The sampling lines were, in the cases of arcs C and D, generally along existing roads, as shown in Fig. 7. Those sampler locations not accessible by vehicle were serviced by helicopter. The more distant sampler emplacements (225-425 km) are shown in Fig. 6.

Whole air samples and real-time SF_6 data were gathered by aircraft over the various sampling arrays. No SF_6 real-time measurements were made over the remote sampling sites. Samples and data were collected over INEL at selected altitudes

from 2 km to 3.6 km MSL. Aircraft samples were gathered over the Bozeman Route (Billings, MT, to Cody, WY) at selected altitudes ranging from 2.5 km to 3.9 km MSL.

III. IDAHO NATIONAL ENGINEERING LABORATORY EXPERIMENT - 1974

A. Objective

As the result of a desire to develop a useful long-range atmospheric tracer using stable isotopes available from the LASL stable isotopes production program (ICONS). Mass-21 methane ($^{13}\text{CD}_4$) was released in a simple feasibility test.

B. Tracers Released

The $^{13}\text{CD}_4$ used on this initial experimental release was synthesized using ^{13}C from the ICONS program by reacting ^{13}CO with D_2 to produce $^{13}\text{CD}_4 + \text{D}_2\text{O}$. Eighty-four grams of the tracer, at a cost of ~\$5000, were released at INEL.

The tracer was emitted from a 70-m stack at the INEL Allied Chemical Corporation processing plant. The initial dilution with stack air initially produced a ratio of $^{13}\text{CD}_4$ to natural methane-16 of ~0.1. The $^{13}\text{CD}_4$ was, however, released in a continuous manner from 1520 to 1750 MDT on May 14, 1975.

C. Samplers and Sampling Array

The cryogenic samplers were funded for the Defense Advanced Research Projects Agency (DARPA), and operated by the NOAA ARL. These samplers were installed at the selected NWS stations 1500-2500 km from INEL, as shown in Fig. 5. Two 10-h samples were obtained daily for a 4-month period.

This particular sampling technique cryogenically concentrates methanes and krypton as the sample air is passed through the apparatus. A second methane concentration step and krypton separation step was accomplished at Airco, Inc. This process is discussed in more detail in a Section IV. E. Using the above technique, several milliliters of methane are collected and later analyzed.

D. Air Trajectories

The tracer was released on the basis of a forecast, by the NOAA/ARL, that winds would carry the tracer toward the center of the sampling array (Nebraska and Iowa), with about a 2-day travel time. Post-factum analysis using the ARL transport and diffusion model⁶ indicated that the plume actually crossed the northern part of the sampling array, as shown in Fig. 5, in good agreement with the sampling data.

E. Methane-21 Measurement Technique

The concentrated methane samples were sent to Britain and analyzed at Aldermaston.⁷ The technique employed by this group involves the use of a

high-sensitivity gas mass spectrometer. This machine, as applied to this analytical problem, provides reproducible results with a sensitivity of one part $^{13}\text{CD}_4$ to 10^{11} parts natural $^{12}\text{CH}_4$.

F. Results

The ratios of the intensities of $^{13}\text{CD}_4$ to CH_4 for the different samples are presented in Table I. An instrumental background correction of 3.7×10^{-11} has been applied to each measurement to obtain a net $^{13}\text{CD}_4/^{12}\text{CH}_4$ ratio. The standard deviations indicated in the table are estimates based on the counting statistics. Due to a sampler problem at Rochester, the May 15 a.m. and May 16 a.m. samples were not obtained from that site.

G. Conclusions

Even though noise levels at mass-20 in the spectra are about a factor of 10 higher than those at mass-21, it may be more economical to use larger amounts of $^{12}\text{CD}_4$ in place of the more expensive $^{13}\text{CD}_4$. This experiment pointed out the need to conduct additional tracer releases to explore the concurrent release of $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ to verify the validity of internal experimental criteria. Conceivably the application of two long-range tracers with similar but discrete analytical measurements for both would provide a cross-check of results. It was also concluded that more experience in field activities of these types of tracer releases would possibly identify more prudent uses of these tracers, as well as identify more efficient sampling and operational techniques.

IV. SAVANNAH RIVER PLANT EXPERIMENT - 1975

A. Objectives

The test of December 10 had three specific objectives. The primary purpose was to determine whether the SF_6 and methane-tracer systems give comparable results when the tracers are released and sampled simultaneously. This provides an overall test of release, sampling, and analysis procedures, as well as atmospheric behavior of both tracers. The second objective was to study the transport and dispersion of a plume out to 100 km from a point source. The third objective was to gain experience in release, sampling, processing, and analysis of these tracers before proceeding to larger scale experiments.

B. Participants

The test was a cooperative effort on the part of several groups, each with expertise in particular aspects of the program. The planning and conduct of the experiment and data analysis was accomplished by scientists from ARL, NOAA, BNL,

LASL, and SRL. In addition to these groups, personnel from Airco, Inc., and EG&G, Las Vegas, NV, participated in the execution of the tests and in data-sample processing and interpretation. The methane and ^{85}Kr separations and the ^{85}Kr analysis were completed by Airco, Inc. The methane-16, -20, and -21 analyses were performed by personnel at AWRE. See Appendix A for complete participant list.

C. Tracers Released

Three tracers (SF_6 , $^{13}\text{CD}_4$, and $^{12}\text{CD}_4$) were released simultaneously over a 4-h period beginning at 1025 EST on December 10, 1975. Calculations indicated that the programmed amounts would permit delineation of the tracer plumes by the ground-level and airborne samplers at 100 km from the source, with reasonable safety margin for meteorological uncertainties and experimental error. The release rates are shown in Fig. 8. Release rates of the two methane tracers were monitored with precalibrated Matheson Gas Mass Flowmeters. Water displacement and flowmeter calibrations were performed, using normal methane to measure flow for either a given time or a fixed total volume. It was assumed that methane behaved as an ideal gas at the temperatures and pressures used (17°C , 0.76 atm). A correction was not made for the vapor pressure of water (14 mm at 17°C) in the calibration. The sulfur hexafluoride release was monitored with a gas rotometer. The rotometer rate, pressure, and temperature were recorded every 10 min. The total mass of each tracer released was also determined by weighing the gas cylinders before and after release.

The $^{12}\text{CD}_4$ was released at the top of the A-area meteorological tower (Fig. 2). The methane, metered through a mass flowmeter, was mixed into a rapidly flowing stream of N_2 gas which was piped to the top of the 62-m tower through a 1/2-in.-i.d. Tygon tube (Fig. 9). The $^{12}\text{CD}_4$ was contained in two compressed-gas cylinders. The pressure regulator on one of the cylinders leaked 98.1 g of $^{12}\text{CD}_4$ at the base of the tower between 0925 and 1240 EST. The 187.4 g of $^{12}\text{CD}_4$ released at the top of the tower was metered through the mass flowmeter as shown by the top release rate record in Fig. 8. The total amount of $^{12}\text{CD}_4$ released was 285.5 g. The $^{12}\text{CD}_4$ released also contained 3.2 g of $^{13}\text{CD}_4$.

The $^{13}\text{CD}_4$ and SF_6 were released into the top of the 62-m reactor cooling-gas exhaust stack at H-area. Area A is approximately 11 km north-northwest of H-area on the 305° azimuth as shown in Fig. 2. The $^{13}\text{CD}_4$ was metered through a mass flowmeter and the SF_6 was metered through a rotometer. The two gases were mixed at the inlet to the SF_6 rotometer. They were routed to the top of the stack

through a stack gas-sampling line as diagrammed in Fig. 10. Flow in the sampling line was the reverse of that normally used for sampling. The 85.5 g of $^{13}\text{CD}_4$ plus 7.6 g of $^{12}\text{CD}_4$ were released at the rate shown by the mass flowmeter record in Fig. 8. Because of a calibration error, only 154 kg (339 lbs) of the planned 450 kg (1000 lbs) of SF_6 were released at the rate shown by the histogram (Fig. 8) constructed from rotometer readings. Fortunately, the safety margin built into the experimental design was sufficient to tolerate this reduced release of SF_6 .

Routine plant operations resulted in a ^{85}Kr release from the H-area stack and from a 62-m stack in F area, about 3 km to the west. Estimates of the combined hourly ^{85}Kr release amounts during the day are given in Table II.

D. Samplers and Emplacement Schemes

Whole-air sampling was accomplished by pumping air into 45-ℓ Saran bags during the predicted plume passage. Thirty sampling sites were selected on, or near, I-95 from mile post 76 southward to the intersection with SC-336 as shown in Fig. 1. Portions of the sample in each bag were later transferred to two cylinders, one for SF_6 analysis and the other for methane and ^{85}Kr analyses. Bag samplers were also activated at two of the sites where there was routine cryogenic sampling of ^{85}Kr .

Whole-air samples of at least 10 (preferably 30) ℓ were required to provide a sufficient sample for processing the heavy methane components. Large plastic barrels of about 40-cm (16-in.) diam and 1-m (39.37 in.) high, were fitted with a Teflon pump (Science Pump Corp., Model D-200) connected with 0.5-cm (1/4-in.) o.d. amber, natural rubber surgical tubing, to a 45-ℓ Saran bag (The Anspec Company, Inc.). Power was provided by a 6-V battery. The sampling from the pump was adjusted to 100 ml/min for the 8-h samplers and 60 ml/min for the 11-h samplers at the cryogenic sites by varying the number of capillary inserts placed between the pump and the bag. At predetermined times after deployment, the sampler pumps were turned on and off manually. A small length of 0.3-mm capillary stainless steel tubing provided the necessary restriction on the pump outlet to give the desired sampling times. A pinch clamp on the short piece of amber tubing provided a leak-tight seal prior to sampling and after collection of the air sample was completed. Before using the bags, they were purged with air containing <0.5 parts per trillion (ppt) SF_6 , rolled flat and closed with the pinch clamp to prevent any possible SF_6 contamination.

1. Bag Samplers

a. Sample Bag and Transfer Cylinder Tests. Before the experiment, the Saran bags were individually checked by BNL for leaks. The bags were checked by filling them with helium and compressing slightly with a weighted plate. Leaks were then sensed with a portable thermal conductivity detector, and about one of five bags was leaking at a seam or fold. The leaking bags were discarded.

Saran bags were filled with different test mixtures of SF_6 in air and analyzed periodically for several days. A portion was then transferred to steel cylinders (as planned for the field samples) and analyzed 18 days later. The concentrations were within a few percent of the starting mixtures. One 45-l bag was purged of the test mixture, filled with zero gas, and analyzed 24 h later. There was no indication of residual SF_6 from the previous mixture. The results of these tests are shown in Table III.

b. Deployment of Bag Samplers. On the morning of the experiment, the samplers were taken to Walterboro (see Fig. 1) and positioned along I-95 and SC-336 in accordance with the latest wind information. Samplers were placed at 3-m intervals from Walterboro north to St. George; 2-m intervals from Walterboro south to Ridgeland, and again at 3-m intervals going west on SC-336. Sampling locations, times, and other pertinent information are given in Table IV.

In general the terrain in which the samplers were located was a forested, gently rolling landscape. The samplers were positioned as close as possible to the edge of the highway right-of-way (15-30 m from the highway).

During the sample-collection period, several of the sampler units were checked, and all but one was functioning normally. One unit (Sample 46) had stopped as a result of a jammed air pump. The pump was cleaned and restarted. The sample collection period selected was 7.5 h with sampler pick-up order the same as the emplacement order.

The position of the plume centerline was determined from the aircraft sampling data and three additional "post plume" samplers were started at 3.75-km intervals near the estimated centerline. These samplers were run for 2 h in an attempt to collect plume material passing the sampling line after the regular sampling had been terminated. All samples were returned to the Savannah River Plant at about 2300 EST, 10 December. Information on these samplers is also shown in Table III.

2. Cryogenic Samplers

a. Sampler Description. The cryogenic samplers were the same units as used on the INEL tracer experiment in 1974 described earlier. These samplers were being used routinely by SRL for ^{85}Kr sampling in a joint project with NOAA/ARL.

b. Deployment of Cryogenic Samplers. The cryogenic samplers measured ^{85}Kr concentrations weekly at 13 sites within about 150 km of the SRP. Three of these sites, St. George, Ridgeland, and Bamberg (Fig. 1), were near our sampling line, and SRL was requested to take special cryogenic samples of shorter duration at these sites. In addition, bag samplers were operated alongside the cryogenic samplers at St. George and Ridgeland.

3. Aircraft Samplers/Instrumentation

a. Bag Samples. The sampler inlet ports on the aircraft were adapted to accommodate gas-sample bags that were provided by LASL. Flow rates were adjusted on the air-sampling ports so that approximately 100 l of air were collected over a 5-min period. The sample bags were produced by LASL from 10-mil polyethylene plastic sheets. They were fitted with a valve stem that allowed them to be filled from the aircraft sampling tube. Aircraft speed and altitudes as hand-recorded are shown in Table V.

b. Portable Gas Chromatographs. Two portable automatic gas chromatographs^{8,9} were developed and operated by BNL on board the EG&G Martin 404 aircraft. Both BNL instruments use a pump to continuously compress the air being passed through a chromatograph column (molecular sieve 5A) into an electron capture detector. Sulfur hexafluoride is eluted first, followed by oxygen, whereupon the column is automatically back-flushed before the next measurement. In general, the longer the chromatograph column, the greater is the length of the continuous SF_6 sampling window. One instrument has a 16-ft column and provides a continuous measurement of SF_6 for approximately 3 min. The increased column length increases the delay time before SF_6 is first eluted, decreases the resolution, and requires longer back-flush times before the next analysis can be made. Total cycle time with this instrument is 12 min and its battery pack will provide up to 8 h continuous operation.

Calibration was performed with a 10-ppt primary standard while the instruments were on the ground, and with a 38-ppt working standard when the instruments were airborne. Linear response with concentrations up to 100 ppt was observed. There was, however, a slight decrease in response with altitude, about 10% at 1.5 km MSL.

The second instrument used in this experiment has a shorter column and provides a 90-s continuous sampling window. This instrument requires only 3 min back-flush time and the total cycle time is about 5 min. It has a rapid response time (for better resolution of the plume) as shown in Fig. 11, by the recorder trace obtained when cycling every 4 s between a 100-ppt and a 10-ppt standard. The limit of detection at a signal-to-noise ratio of 3 is about 0.5 ppt. Both the long- and short-column instruments were mounted in the aircraft with two portable recorders, two spare batteries, and two battery chargers for emergency use.

Data obtained on the recorders were later converted to SF_6 concentration vs crosswind distance or time with the aid of a digitizer-plotter system. Since the two instruments had different delay periods for SF_6 to elute (~160 s for one and ~54 s for the other), it was necessary to find a common point in real time on the scans to compare the results of the two. When the 90-s instrument was turned on and off automatically, an event mark was made on the 3-min instrument recorder. From these marks and the known delay period for each instrument, it was possible to match the outputs of both instruments to within 2 s of real time.

Conversion of the recorder traces to concentration plots was accomplished with a digitizer cursor with cross-hair window that was manually moved along the recorder tracer. Points were either manually or automatically entered in the calculator of the plotter system. Four consecutive points (Y_1, Y_2, Y_3, Y_4) were used to compute the best slope for three straight line segments to be drawn from Y_2 to Y_3 . Thus, as each new location was entered into the plotter calculator, three lines were drawn between the previous two points. At the same time, the area under the curve was summed to obtain the cross-wind integral of plume concentration.

E. Sample Processing and Tracer Measurements

1. Methane-16 and SF_6 Background Measurements. Ambient SF_6 and CH_4 background measurements were made by BNL with a gas chromatograph. The SF_6 background measurements (see Table VI) were made on samples obtained between September 22 and October 1, 1975, from the plant site and along I-26 about 100 km northeast of the plant. These ten "spot" samples (5-10 min each) were taken in 900 cm^3 steel cylinders. Concentrations ranged from 0.4 to 0.9 ppt with an average concentration of 0.4 ppt, however, one sample of 5.9 ppt was later found to be caused by a contaminated cylinder.

Although there is no detectable background concentration of the heavy methanes, the concentration of normal methane (CH_4) in the samples must be known

to calculate the heavy methane concentrations in the samples. Methane concentrations in 23 samples taken in the vicinity of the plant at various times before the tracer experiment varied from 1.1 to 2.2 ppm with an average of 1.64 ppm. (See Table VII). Three spot samples taken between 0900 and 2100 EST on December 10 in the vicinity of the sampling line all had concentrations of 1.70 ppm, which is assumed to be the CH_4 concentration in all samples taken during this experiment.

2. Air-Sample Processing. Immediately upon the return of the samplers to SRL, aliquots were transferred from the sampling bags to cylinders for shipment to BNL for SF_6 analysis, and to Airco, Inc. for krypton and methane separation.

Fifty to one hundred μ l of each aircraft sample were cryogenically condensed into cylinders and processed in the same manner as the ground-level air samples. Processing data on the four samples collected on board the aircraft are given in Table VIII. Processing for the ground samples are shown in Table IX.

a. SF_6 Sample Processing. Approximately 1 μ l of air was transferred from each bag to a pre-evacuated steel cylinder, which was then transported to Brookhaven for SF_6 analysis. Several cylinders were not used in order to determine their integrity during transport. At Brookhaven, these bottles were checked for vacuum, filled with zero gas, and analyzed for SF_6 , which was always below the detectable level (<0.02 ppt).

b. Krypton and Methane Processing and Concentration. After transfer of the SF_6 fraction, the remainder of each sample was transferred to a 1- μ l stainless steel cylinder by condensation with liquid nitrogen. The cylinders were then shipped to Airco's Central Research Laboratory at Murray Hill, New Jersey, for separation of the krypton and methane and measurement of the krypton specific activity. The methane fractions were sent to Norman Daly, Atomic Weapons Research Establishment (AWRE) in Aldermaston, England, for mass spectrometric analysis of the heavy methane content.

Before use of the cylinders, krypton and CH_4 "spikes" had been measured into each cylinder at Airco, Inc. (see Table IX for details of "spike" amounts and recovery percentages). The CH_4 "spike" was necessary because the anticipated 30- μ l ambient-air volumes would not contain sufficient material to physically isolate the CH_4 fraction. A nominal 10 cm^3 of krypton and 0.5 cm^3 of methane were expanded into each cylinder after it had been evacuated to 0.1 mtorr. The volumes were measured from mercury-filled gas burettes readable to 0.025 cm^3 . Essentially "dead" krypton (0.01 dpm/cm^3) from the immediate postwar era was used

as a carrier. The $^{12}\text{CH}_4$ was obtained from Matheson Gas Products as their ultra-pure grade (99.97%).

The volume of each air sample was determined by weighing the full cylinders before processing and the evacuated bottles after processing. The weight measurements were made using a Voland HCE 50 equal-arm balance, Class P stainless steel weights. A stainless bottle without valve was used as a counterpoise. The dew point of the air at the sample collection sites was reported as -1°C . This is equivalent to 0.3% water by weight which could be subtracted from the air weight if results on a dry basis were required. However, relative to other errors, this quantity is insignificant and no moisture correction was made to the reported air weights.

After an initial concentration step by adsorption on charcoal at liquid oxygen temperature, the krypton and methanes were separated by gas chromatography on a mixed-charcoal molecular sieve column using helium as the eluting gas. The krypton and methanes were isolated from the eluting gas by condensing each fraction in separate freezeout traps at the triple point of nitrogen (63 K). This process produces krypton samples in the 10-cm^3 range with less than 100 ppm impurities and 0.5-cm^3 methane fractions with purities of 97-99%. Each methane sample was sealed into a 25-cm^3 Pyrex breakseal bulb for shipment to AWRE.

3. BNL SF_6 Measurement. The low SF_6 levels present in these samples at the 100-km downwind distance (<10 ppt) necessitated a laboratory chromatograph system that consisted basically of a 10-cm-long 13X molecular sieve concentrator trap followed by a 1-m precut column and a 5-m analytical column, each filled with 5A molecular sieve. An air sample from the steel bottles was expanded into the evacuated 40-cm^3 sample loop to a known pressure and then flushed into the concentrator trap at dry ice temperature. After the trap was isolated and heated to 200°C , the SF_6 was flushed into the precut column and into the analytical column. Immediately after the SF_6 eluted into the analytical column, the precut column was backflushed to remove the remaining constituents of the air sample. With this procedure total cycle time for each determination was 10 min with a limit of detection of 0.02 ppt for a 40-cm^3 sample.

Calibration of the chromatograph was accomplished with a prepared gas standard of 10 ppt and a working standard of 1.75 ppt. Linearity of the calibration to less than ambient levels was confirmed by analysis of a 60-fold dilution of the working standard. Intercomparison of samples with the HASL showed very good

agreement with less than 5% relative standard deviation between laboratories. The reproducibility of the measurements is demonstrated by the analyses of background air samples in triplicate, analyzed on two separate days, as shown in Table VI. The concentrations are indicative of the SF_6 background level during the experiment. The precision at these ambient levels could be improved by using double injections, i.e., 80 cm³ total sample size. The laboratory analysis of SF_6 is described by R. Dietz of BNL in Refs. 8 and 9.

4. Methane-16, -20, and -21 Analysis. The $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ ratios to CH_4 were measured at AWRE by the gas mass-spectrometric methods referred to earlier.

5. Krypton-85 Measurement. The separated krypton was loaded into Geiger counters and counted for 1000 min. Each sample was counted twice in two different counters. The second column of Table IX gives the actual weight of the air sample in the cylinder. The third column converts this to sample volume at 25°C and 760 mm Hg. Nine samples had leaked and part of the original air condensed into the bottle and was lost along with the spike material. In each of these cases, when the krypton recovery was below 80%, a check of the cylinder after processing the sample indicated the presence of a leak. The volume of air obtained by weight was corrected for these samples based on the original amount of krypton spike added, with an assumed recovery efficiency of 90%.

The fifth column gives the volume of atmospheric krypton contained in the sample volume given in column 3 based on an assumed natural krypton concentration in air of 1.112 ppm by volume. The volume of krypton recovered from each sample is given in column 6 and the recovery efficiency in column 7 of Table IX. The last four columns of this table give similar information for methane.

F. Meteorological Data

1. Brief Description. Meteorological data obtained at SRP during the tracer tests consist of: (1) wind speeds, directions, and temperatures (including deviations) from the WJBF-TV tower (seven above-ground levels) and the seven-tower system on the SRP (2) hourly surface data for South Carolina, Georgia, North Carolina, Florida (in part), and Alabama (in part); (3) upper-air wind speeds, direction, pressure, and temperatures from Georgia, South Carolina, and North Carolina; (4) wind speeds, directions, temperatures, and relative humidity recordings from the Georgia and South Carolina Forestry Lookout Towers; and (5) hourly pilot-balloon (pibal) wind speed and direction profiles at the SRP site from 0700 to 1900 MST. Good correspondence between the pibal winds and tower measurements

is demonstrated in Fig. 12, which shows the 304-m level on the tower and the mean layer value centered at 327 m from the pibal.

The meteorological tower network consists of seven towers located throughout SRP and the WJBF-TV tower located approximately 20 km northwest of the center of SRP. The TV tower is instrumented at seven levels, from 10 m to 335 m. Wind speeds, wind directions, and temperatures from all eight towers were recorded on magnetic tape every 5.2 s. Values for σ_A (standard deviation of the horizontal wind direction in degrees), σ_E (standard deviation of vertical wind direction in degrees), and mean wind direction and speed from the tower data are calculated as a function of sampling time (normally 60 min).

2. Meteorological Summary. The tracer experiment was conducted in a northwesterly flow behind a well-defined cold front. The day began under clear skies with strong northwest winds at the surface.

Figure 13 shows time-height cross sections of wind direction and speed obtained by visual theodolite tracking of pilot balloons released near the tracer-release point.

A veering tendency of wind direction to northerly appeared at about 1.5 km at 0800 EST and propagated down to 0.5 km by 1000 EST. This was followed by a backing tendency to 270° , which propagated similarly. After 1100 EST this transient feature had passed and the wind field settled into a relatively steady northwest flow at 7-10 m/s in the 1.5-km layer in which the tracer plume was transported. The tracer release began at 1030 EST and so was virtually unaffected by the transient wind disturbance. The subsynoptic feature that produced the wind field disturbance is described in detail in Ref. 10.

Analysis of the vertical temperature structure is important in estimating mixing properties of the boundary layer. The closest upper-air sounding data are from Athens, Georgia, at 1200 GMT (0700 EST). This sounding is shown in Appendix B, along with estimated soundings during the experiment as based on observed hourly surface temperature and advected temperature fields at 850 and 700 mb. These soundings show the initiation of moist convection at 1100 EST reaching a maximum at 1300 and ending at 1500 EST. This coincides very closely with the observed clouds that were first noted at 1030 EST and reached 4/8 sky cover at 1300 but were dissipated completely by 1700 EST. The insight into the vertical structure and convection activity allows us to define the vital elements, the height of the mixed layer (MXHT) and the intensity of mixing in the layer. From these elements, we can estimate the location and dimension of the tracer plume.

Figure 14 shows the estimated mixing depth derived from the Athens sounding by following a dry adiabatic temperature curve from the estimated surface temperature to its intersection with the (evolving) sounding. After the onset of moist convection, the mixed depth is the estimated top of the convection. Fig. 14 shows the growth-curve estimates for the front edge of the tracer cloud and the trailing edge on the absolute time scale. The effective mixing associated with convective turbulence transports tracer material to the top of the mixed layer within a few hours.

Preliminary diffusion estimates can be made using Gaussian plume approximations with suitable parametric inputs. Gifford¹¹ offers a survey with some critical assessment of turbulent diffusion typing schemes. Two points of concern are the functional form of the travel distance-dependent plume growth and the selection of the proper curve from meteorological conditions. There have been very few empirical observations of plume growth beyond 10 km and estimates of plume dimensions at 100 km have typically been made by extrapolation of the Pasquill-Gifford curves. Gifford¹¹ summarizes a more sophisticated extrapolation procedure by Smith.¹²

The selection of the plume growth curve for the December 1975 experiment introduces an inconsistency between the classical Pasquill-Gifford criteria and the suggested equivalent basis of standard deviation of wind direction fluctuations. The wind speed and insolation parameters suggest category D (near neutral), whereas the low-level σ_A , as determined from tower wind measurements, suggest category A (extremely unstable). It is likely that moist convection indicated by extensive cumulus cloud formation generated the turbulence observed at the tower. We have adopted the observed turbulence intensities as the most reliable indicators of the diffusion environment. Under these assumptions, the simple extrapolation of the original Pasquill-Gifford curves gives $\sigma_y = 10$ km at $x = 100$ km. Briggs¹³ attempted to generalize cloud growth curves from elevated sources to longer distances (10-20 km). Extrapolation of these curves to 100 km (against the author's recommendation) gives a slightly smaller $\sigma_y \sim 7$ km.

The obvious extent of cumulus convection strongly suggests vigorous mixing within the mixed layer from the surface to 1500-m altitude. A uniform distribution of tracer through this layer is a reasonable assumption at a downwind distance of 100 km.

The total exposure $E(\text{kg s/m}^3)$ from the 4-h release is

$$E = \left(\frac{Q}{\sqrt{2\pi} \sigma_y L u} \right) \exp \left(- \frac{y^2}{2\sigma_y^2} \right) .$$

where Q = source strength in kg
 L = mixing depth in m
 u = wind speed in m/s
 σ_y = diffusion parameter, measure of cloud width in m
 y = crosswind distance in m.

To compare these estimated concentrations with the measurements, we observe that air at a density $\rho_{\text{air}} = 1.3 \text{ kg m}^{-3}$ was pumped for a sampling time $T = 2.7 \times 10^4$, and the measured concentrations are expressed as a normalized molar or volume ratio to air

$$R = \frac{\text{m}^3_{\text{tracer}}}{\text{m}^3_{\text{air}} \text{ kg tracer}} ,$$

thus,
$$R = \left(\frac{E}{Q} \right) \left(\frac{1}{\rho_{\text{air}}} \right) \left(\frac{M_{\text{air}}}{M_{\text{tracer}}} \right) \left(\frac{1}{T} \right),$$

where M_i is the molecular weight of the i^{th} species.

Evaluation of the above equation for the three tracers used at SRL, based on $\sigma_y = 7 \text{ km}$, $L = 1.7 \text{ km}$, and $u = 7 \text{ m/s}$ is

	<u>SF₆</u>	<u>Me-21</u>	<u>Me-20</u>
Q, kg	154	0.089	0.293
M, kg mole ⁻¹	146	21	20
Centerline E, parts/part air by vol.	4.2×10^{-12}	1.7×10^{-14}	5.7×10^{-14}
E _{areas}	5.1×10^{-12}	0.6×10^{-14}	2.5×10^{-14}

G. Results

1. Tracer Concentrations

a. SF₆ Concentrations. The SF₆ concentrations obtained directly from the bag samples by gas chromatography at BNL are shown in Table X. The standard deviation of duplicate determinations on each sample ranged from 0.00 to 0.04 ppt. A plot of the SF₆ concentrations along I-95 are shown in Fig. 15. The main plume crossed the highway between Mile Post 30 and Mile Post 58. Sampling results appear consistent, giving a background concentration of 0.5 to 0.6 ppt and a peak plume concentration of 5.6 ppt. An unexplained elevated level of SF₆ was observed at Mile Posts 22 and 24. Samplers on SC-336, just west of Mile Post 22, showed essentially background concentrations.

Of the two postplume samples obtained after the regular sampling was terminated (see Table IV), sample 54 at Mile Post 34 showed background but sample 39 at Mile Post 40 had 1.6 ppt of SF₆, almost as much as the earlier sample at that position. A question exists as to whether a significant fraction of the release might have arrived at the sampling line after the samplers were turned off.

b. SF₆ Concentrations - Aircraft Instrumentation. The approximate locations of the aircraft sampling passes are shown in Figs. 3 (passes 1-5) and 4 (passes 6-11); more precise positioning may be possible based on the EG&G magnetic tape record of location, time, and SF₆ concentration. All five passes shown in Fig. 3 were flown on flight path labeled A-A. The dashed portion of each pass indicates the segment over which the SF₆ instruments were turned on. The solid portion indicates the segment on which the plume was found while the instruments were active. Arrows indicate the direction of travel on each pass. A plume contact was obtained on four of the five passes along path A-A. As shown in Fig. 4, the two passes made in the vicinity of Allendale along flight path B-B recorded the plume contact, as did three of the four passes along path C-C near I-95.

A typical instrumental response obtained with the BNL instruments is shown in Fig. 16. These profiles were derived from the instrument-recorder plots as described in the section SF₆ chromatographs. Table IV gives the altitude (above terrain), aircraft speed, and time each SF₆ instrument was started on each pass. Also shown in the SF₆ crosswind integral (conc x time) obtained from the concentration plots. These data can be used to estimate the total amount of SF₆ crossing the flight line. The peak SF₆ concentration recorded on each pass is presented in the last column of Table IV.

c. Methane-16, -20, and -21 Concentrations. The results of mass spectrometric analysis for $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ are shown in Table XI for the highway bag samples.

This table indicates errors arising from the mass spectrometric measurements only. Another error is associated with the assumed constancy of the $^{12}\text{CH}_4$ concentration at the experiment site. Although this concentration varies at SRP in the range 1.1 to 2.2 ppm (Table VII) measurements on the day of the experiment indicated a constant value of 1.70 ppm. This value has been used in all the calculations for heavy methane concentrations reported from this experiment.

The reported concentrations of $^{13}\text{CD}_4$ in sample air (21/air) are derived from the abundance ratios of $^{13}\text{CD}_4$ to CH_4 measured by the mass spectrometer, $(21/16)_{\text{meas}}$, from the relationship

$$(21/\text{air}) = (21/16)_{\text{meas}} \left(\frac{V_S}{V_C} + A_{\text{CH}_4} \right),$$

where

V_S = volume of CH_4 spike

V_C = volume of air sample in the cylinder

A_{CH_4} = abundance of CH_4 in air (1.7×10^{-6} by vol).

The volumes V_S and V_C for each sample are given in Table IX. The above equation was also used to determine the $^{12}\text{CD}_4$ concentration in the sample air (20/air) from the measured $^{12}\text{CD}_4$ to CH_4 ratios, $(20/16)_{\text{meas}}$.

To a good approximation, the fractional error in the concentration of $^{13}\text{CD}_4$ in air is the same as the fractional variation in either V_S or V_C . On the other hand, the uncertainty in the value of $(21/16)_{\text{air}}$ arises mainly from the uncertainty of the value of A_{CH_4} .

The credible values of $(21/16)$ as measured by the mass spectrometer ranged from $(-0.17 \pm 0.03) \times 10^{-11}$ to $(37 \pm 0.8) \times 10^{-11}$ as shown in Table X. The peak signal-to-noise ratio in this experiment was thus greater than 100 in the case of $^{13}\text{CD}_4$. The four lowest values can be used to estimate the lower limit of the background of $(21/16)$ in the atmosphere as $(1.2 \pm 1.5) \times 10^{-12}$.

In the case of $^{12}\text{CD}_4$, the four values corresponding to those used for the background values of $^{13}\text{CD}_4$ average $(1.0 \pm 0.17) \times 10^{-11}$. It is not clear whether this is the real background of the atmosphere and/or "spike." In any case, the peak value of the $(20/16)$ mass ratio of $(160 \pm 2) \times 10^{-11}$ observed in this experiment corresponds to a signal-to-noise ratio greater than 100.

The heavy methane concentrations measured along I-95 are plotted in Fig. 17.

Bag samplers, located at miles 73 and 76 on I-95, mile 14 on SC-336, and at the St. George cryo-sampler site, show extremely high 21/16 ratios. They are 15 to 3000 times greater than the peak values of those near the middle of the sampling line. These four samples were kept in the bags at SRL for several days after the experiment in the presence of the cylinders containing the unused methane tracers and probably were contaminated.

Four aircraft bag samples were taken in polyethylene bags as indicated in Table V. Sample AC-1 was obtained before the start of the tracer release, but may have caught some of the $^{12}\text{CD}_4$, which was leaking. Sample AC-2 was collected near the release point and has a $^{13}\text{CD}_4/\text{SF}_6$ ratio of 3.9×10^{-3} , which is very close to the release ratio of 4.2×10^{-3} . Sample AC-3 at 150 m over I-95 was depleted in $^{13}\text{CD}_4$ by about an order of magnitude. Similarly sample AC-4 at 600 m over I-95 was depleted in $^{13}\text{CD}_4$ by a factor of four.

d. Krypton-85 Concentrations. The concentration of ^{85}Kr in air (pCi/SCM) is obtained by first measuring the ^{85}Kr radioactivity in the total krypton isolated from the transfer cylinder (reported in Table XI as dpm/cm³ of ^{85}Kr). This concentration is then adjusted for the volume of the krypton spike in the transfer cylinder and for the abundance of normal krypton in the atmosphere. Thus,

$$C_a = C_{\text{Kr}} \left(\frac{\text{pCi/SCM}}{\text{dpm/cm}^3} \right) \left(\frac{V_s + V_n}{V_n} \right) A_{\text{Kr}},$$

where C_a = concentration of ^{85}Kr in air sample (pCi/SCM),
 C_{Kr} = concentration of ^{85}Kr in krypton from sample (dpm/cm³),
 $\frac{\text{pCi/SCM}}{\text{dpm/cm}^3}$ = conversion factor = 0.45×10^6 ,
 V_s = volume of spike (cm³),
 V_n = volume of normal krypton in cylinder (cm³), and
 A_{Kr} = abundance of normal krypton in air (1.112×10^{-6} by vol),

so that,

$$C_a = 0.5 C_{\text{Kr}} \frac{V_s + V_n}{V_n}.$$

Also,

$$V_n = A_{Kr} V_c ,$$

where V_c = volume of air sample in the cylinder.

For the cylinders that leaked, it was assumed that the air sample and the spike were lost in the same proportion. The ratio of the apparent ^{85}Kr recovery efficiency to the expected efficiency (90%) was used to adjust the sample volume V_c and the volume of normal Kr V_n (see Table IX).

The ^{85}Kr concentrations along I-95 are plotted in Fig. 15. Krypton-85 background concentration has been measured to be ~ 14 pCi/SCM at the sampling sites.

2. Tracer Comparison. The concentration profiles of the two heavy methanes and SF_6 along I-95 appear very similar and all three tracers show the highest concentration in the sample taken at Mile Post 46. However, when the concentrations are normalized to the same release amount, it is seen that the methane concentrations are low by about a factor of four compared to SF_6 . The cause of this discrepancy is not yet understood but some of the possibilities being investigated are discussed in Sec. H.

Two sets of calculations were performed on the measured tracer concentrations to intercompare the cross-wind distributions of the three tracers and to compare them with the simplest plume model (Gaussian).

Table XII shows the results of the gross statistics of the data for each tracer. In this analysis, the SF_6 background has been removed. The width of the distributions in terms of the standard deviations, σ , for SF_6 and $^{13}\text{CD}_4$ were in agreement, whereas that for $^{12}\text{CD}_4$ was slightly larger. The kurtosis (flatness factor) of the $^{12}\text{CD}_4$ distribution is less than the other two tracers, indicating a sharper distribution. The agreement between SF_6 and $^{13}\text{CD}_4$ statistics is not surprising since they were released at the same time and place. The centroids $\bar{X}$ of all three tracers were between Mile Posts 44 and 47.

Table XII summarizes the parameters of a least squares fit of the data to a Gaussian plus a constant function of the form given in the following equation and the results are shown in Fig. 18. In this case, the data points were weighted by their analytical uncertainty,

$$c = (1/\sqrt{2\pi}) (A/\sigma) \exp \left(- \frac{(X-X_0)^2}{2\sigma^2} \right) + B .$$

The peaks of the fitted curves for the three tracers were in good agreement and centered near mile marker 46. This analysis yielded smaller standard deviations than the gross statistics, probably because of, at least in part, the relatively larger uncertainties in the low-concentration values on the "wings." The additive constant B agrees very well with the observed background concentration in the case of SF_6 . Since there is no expected heavy methane background, the B value in these cases probably reflects the noise in the low-level concentration values on the wings of the distribution. The highest observed concentrations (mile marker 46) for all three tracers were 40 to 70% higher than the fitted peaks.

3. Tracer Concentration Ratios. Air concentrations of the four tracers may be directly compared by dividing the concentrations χ , expressed in g/SCM (pCi/SCM for ^{85}Kr) by the respective release amounts Q. The estimated background concentrations of 0.55 ppt for SF_6 and 14 pCi/SCM for ^{85}Kr were subtracted before normalizing the measured concentrations. Release amounts were

SF_6	$1.54 \times 10^5 \text{ g}$
$^{12}\text{CD}_4$	$2.93 \times 10^2 \text{ g}$
$^{13}\text{CD}_4$	$8.86 \times 10^1 \text{ g}$
^{85}Kr	$4.12 \times 10^2 \text{ Ci}$

For this comparison, the ^{85}Kr emitted during the 4-h tracer release (412 Ci) was used, although ^{85}Kr was also released before and after this period.

The normalized concentrations, χ/Q , along I-95 are shown in Fig. 19. Since SF_6 and $^{13}\text{CD}_4$ were released simultaneously through the same stack, their concentration profiles should coincide if there were no flaws in the release, sampling, processing, or analysis procedures, and if the two tracers behaved similarly in the atmosphere. The $^{12}\text{CD}_4$ would be expected to have a somewhat different profile with a slightly lower peak concentration since it was released about 10 km upwind of the SF_6 and $^{13}\text{CD}_4$ release.

All four tracers show a peak at Mile Post 46 with SF_6 and ^{85}Kr in good agreement and the methanes a factor of 3-4 lower. Note that the ^{85}Kr and $^{12}\text{CD}_4$ are high relative to SF_6 on the south side of this peak and low on the north side. Since wind data indicate the plume shifted with time from south to north along the sampling line, this is in qualitative agreement with the decrease in ^{85}Kr release rate during the tracer-release period. The large ^{85}Kr peak to the

south appears to be readily explainable by the ^{85}Kr released before the other tracers (see Table II) as the samplers were turned on early enough to catch this material. The ^{85}Kr peak at the north end of the sampling line may be caused by krypton emitted after termination of the other releases.

In addition to the agreement on the position, a maximum peak at Mile Post 46, all four tracers also show relatively high concentrations at Mile Post 42. However, both methanes show a definite peak at Mile Post 69 that is absent in the SF_6 data, whereas the SF_6 shows smaller peaks at Mile Posts 22 and 24 that are absent in the methane data. No explanation for these differences has been determined.

A major problem revealed in this experiment is the discrepancy between the methanes and SF_6 . Both the peak concentrations and the concentrations summed over the sampling line show the methanes to be low by about a factor of four relative to SF_6 . The SF_6 appears to be in reasonable agreement with ^{85}Kr , and mass balance calculations also tend to confirm the validity of the SF_6 measurements. On the other hand, the two methanes appear to be in good agreement with each other. The ratio of the $^{12}\text{CD}_4/^{13}\text{CD}_4$ concentrations summed over the sampling line is 3.6, in good agreement with the ratio of molecules released; i.e., 3.5.

H. Conclusions

The primary objective of the experiment, a test of the release, sampling, processing, and analysis procedures for the heavy methane tracers simultaneously with SF_6 , was accomplished. The use of $^{12}\text{CD}_4$ as an atmospheric tracer was demonstrated for the first time. The $^{13}\text{CD}_4$ was used successfully in the 1974 experiment with detection out to a range of 2500 km, but had not been checked quantitatively with simultaneous release of another tracer.

Air concentrations of all three tracers were measured at the ground and aloft to 90 km from the release point. The BNL continuous SF_6 analyzers obtained plume profiles or contacts on 9 of the 11 crosswind passes attempted.

The release of >300 g of $^{12}\text{CD}_4$ and >100 g of $^{13}\text{CD}_4$ both gave signal-to-noise ratios of better than 100 to 1 at a distance of about 90 km from the source. If larger samples are collected (on the order of 500 l), making a dilution with a CH_4 "spike" is unnecessary; about 30 g of $^{12}\text{CD}_4$ and 10 g of $^{13}\text{CD}_4$ would be enough to give the same signal to noise levels.

All four tracers showed the peak concentration at the same location and generally similar concentration profiles along the sampling line. Heavy methane concentrations, on average, were low by about a factor of 4 compared to SF_6 after

normalization for the different release amounts. Possible causes for this discrepancy are being investigated but the problem has not been resolved. It is believed that there was probably a flaw somewhere in the release technique, sampling, or processing procedures, rather than a difference in the behavior of the tracers in the atmosphere.

I. Recommendations

Various operational and technically related problems were identified in addition to the unresolved heavy methane/SF₆ ratio issue. All of these suggest improvements for implementation in future atmospheric tracer experiments. Some of the more prominent areas that will require improvement are listed below.

1. Tracer Release Scheme. A leaky pressure regulator resulted in the uncontrolled release of a portion of the ¹²CD₄ tracer. The apparatus for release of SF₆ and ¹³CD₄ functioned satisfactorily; both tracers were released at uniform rates over a 4-h period, a calibration error resulted in the release of only one-third of the planned amount of SF₆. Conservatism in the planned release amount provided enough safety margin to allow for this. These problems are easily corrected.

2. Sampler Deployment. Samplers were properly deployed to intercept the plume in spite of communications difficulties. Commercial telephones provided intermittent communications with the field personnel deploying the sampling array. This was a marginally satisfactory arrangement which would not suffice if experiments were undertaken in less populated areas. Commercial mobile phones also were not very satisfactory because of short range and a lack of good relay stations in the area.

Continuous high-quality communication links between the control point and the sampling teams should be provided in future experiments.

3. Bag Sampler System. The bag samplers were generally successful in sample collection, but improved design and functional features are indicated. The pumps appear to be temperature-sensitive and difficult to adjust to a desired sampling rate. The Saran bags were susceptible to changes in temperature and some surface cracks were observed after exposure to cool temperatures. The mechanical fragility of the bags is an inherent problem demanding extreme care in handling. Leak-testing each bag before the experiment is a laborious, time-consuming, but necessary, effort. A simpler, more rugged sampling system is being sought. A sequential sampler would have obvious advantages in developing a better understanding of plume behavior in the atmosphere.

4. Air-Sample Transfers. The transfer of samples from the bags to steel cylinders to maintain sample integrity until they could be analyzed appears to have been a necessary procedure. Transfer procedures worked very well except for one flaw. The cylinders used for cryogenic transfer of samples for analysis of krypton and the methanes had fittings on both ends. Immersion of one end of the cylinder in liquid nitrogen during the transfer appears to have affected the seal, causing leaks in some of the cylinders. In the future, a cylinder valved at one end only should be used. The other steps in the rather complex sample-processing and -analysis procedure appear to have gone quite well.

5. Aircraft Sampling and Measurements. The aircraft used in this experiment, with its sophisticated navigational equipment, probably represented an "overkill" for our air-sampling requirements. However, the aircraft was available because of its use on other experiments that required this sophistication.

Although good samples were obtained, it is believed that a smaller aircraft flying predetermined sampling tracks could provide more sampling passes in the time it takes the plume to pass a sampling line. Arrangements should be made well in advance to provide for proper installation of sampling equipment and appropriate working space for the sampling team.

It is difficult to obtain complete plume profiles at 100 km from the tracer-release point with instruments that have a very limited sampling window present. In addition, the delay period associated with the BNL semicontinuous instruments, as well as the lengthy backflush time required before a new measurement can be started, reduces the effectiveness of this method. Continuous detection methods for SF_6 or other tracers would significantly improve plume profiling capabilities.

V. IDAHO NUCLEAR ENGINEERING LABORATORY EXPERIMENT-1977

A third comprehensive atmospheric tracer experiment was conducted at INEL in April 1977. The actual tracer releases were begun at 1240 MST 19 April and were completed at 1540 MST.

A. Objectives

This was a cooperative effort among several groups and organizations with a number of objectives. For this report we will limit ourselves to those objectives considered important to the fulfillment of LASL programmatic requirements. These objectives were:

1. Investigate improved experimental techniques to resolve the discontinuity in the tracer measurement results found in the SRP-1975 experiment (heavy methanes

vs SF_6). An important activity associated with this objective was to (a) perform preanalysis sample handling and processing with LASL personnel, (b) follow these samples through their non-LASL processing steps and observe closely the procedures of Airco, and (c) to observe the gas mass spectrometry of several samples at AWRE.

2. Implement improved and innovative sampling apparatus for collections at an intermediate range (between 100 km and 1500-2500 km, in particular, 300 km).

3. Apply more comprehensive meteorological data to the conduct of the experiment as well as use these data in experimental interpretation.

4. Use a test site where more persistent meteorological stability would prevail, which in turn would provide the experimentalists with a higher degree of confidence in prerelease meteorology predictions.

5. To fly a less complex, more responsive air platform for the collection of samples and data.

B. Participants

This experience was similar to the SRP experiment in that a rather large complement of scientists and technicians was assembled to conduct and observe the various aspects of the experiment. In addition to the LASL participants, personnel from several NOAA groups at ARL, Silver Spring, MD; ARL/FRO at INEL; and the NWS at Las Vegas, NV, and Salt Lake City, UT, provided scientific management, local field and logistic support; on-site meteorological parameter measurements; and invaluable meteorological interpretation and data processing. ARL/FRO was the experimental-site host and as such was extremely helpful in all aspects of the field activities; without their splendid work the field portions of this experiment would not have been successful. The US Environmental Protection Agency (EPA) at Las Vegas provided two people and a Turbo Beech sampling aircraft, Fig. 20. These people and those from the NWS, Las Vegas, were permitted to assist in this experiment by the ERDA, Las Vegas Operations Office, their funding support organization.

Other technical and scientific personnel were at INEL to conduct special operations of the experiment. These personnel were from HASL, New York City; BNL; Allied Chemical, INEL; and LASL. Contractor personnel from EG&G/INEL, EG&G/Albuquerque, Red Baron Flying Service, and James Lovelock's group from Britain were employed in supporting roles. Certain off-site functions were handled by volunteers not formally associated with any of the official or active participants. A complete list of all participants has been compiled and is shown in Appendix C.

C. Tracers Released

A stated objective of this experiment was the resolution of an observed discrepancy in the mass balance determinations between SF_6 and the heavy methanes as a result of the 1975 experiment. Additionally, it was a prime objective of other non-LASL experiment sponsors to test two potential new tracers, perfluorodimethylcyclohexane (C_8F_{16} ; PDCH) and perfluorodimethylcyclobutane (C_6F_{12} ; PDCB), against the SF_6 and methane tracers.

The heavy methanes used for this experiment were synthesized from carbon-13 produced by the ICONS program (LASL Group CNC-4). The perfluorocarbons (PDCH and PDCB) and SF_6 were procured by ARL from commercial sources. Another potentially useful tracing substance (^{85}Kr) was also measured. The ^{85}Kr used for this measurement is an atmospheric constituent resulting from INEL fuel-element dissolving procedures. Table XIII summarizes the properties of the tracers and the quantities released.

1. SF_6 and $^{13}\text{CD}_4$. The SF_6 , 274 kg (604 lbs), and the $^{13}\text{CD}_4$, 75.31 g, were premixed as low-pressure gases (prerelease pressure 198.8 psia) in a 4-m³ (1000-gal) propane tank. One of the two new experimental tracers (PDCB) was released into the baffled manifold that was used to dispense the $\text{SF}_6/^{13}\text{CD}_4$ mixture. Fig. 21 shows the tracer-release apparatus. Analysis of the tracer-release manifold samples by BNL (Table XIV) shows the $\text{SF}_6 - ^{13}\text{CD}_4$ release ratio to be 448. Analysis also determined the tracer-release ratios $\text{SF}_6/\text{PDCB} = 2.203$, $\text{SF}_6/\text{PDCH} = 3.162$, and $\text{SF}_6/\text{PMCH} = 28.9$. The PMCH (perfluoromonomethylcyclohexane) is a production contaminant found in PDCH.

2. PDCB and PDCH. The PDCB and PDCH, the two new experimental tracers, were released to study their appropriateness as long-range atmospheric tracers when compared with the heavy methanes and SF_6 . The 255 kg (563 lbs) of PDCB were released with the $\text{SF}_6/^{13}\text{CD}_4$ mixture and 260 kg (573 lbs) of PDCH + PMCH were atomized and sprayed into the air near the release point of the $\text{SF}_6/^{13}\text{CD}_4$ mixture.

3. Krypton-85 (^{85}Kr). Fuel element reprocessing was in progress by Allied Chemical at INEL during the April tracer experiment. As a result of this process ^{85}Kr was being released from a 70-m stack at their facility on the site. An estimated ~150 Ci of ^{85}Kr was released at a more or less constant rate during the 3 h of the controlled tracer releases. The fuel reprocessing cycle began at 0001, 19 April and continued until 1500 the same day. An analysis of the H_2 released during this cycle was used to estimate the ^{85}Kr release.¹⁴

4. $^{12}\text{CD}_4$. The $^{12}\text{CD}_4$ was released at a cross-wind site 8 km from the primary release area. This tracer was released at ground level directly from its transportation container. A total 261.60 g of $^{12}\text{CD}_4$ was released from this site and the primary tracer-release site. See Table XIII for data relevant to this release.

D. Samplers and Instrumentation

1. Bag Samplers

a. Surface. An improved version of the barrel-bag type ARL Sampler was the standard whole-air collector used on this experiment. The improvements to the basic unit included an electronically controlled flow rate. The same type Saran bags as those used at SRP were used as collection containers in these samplers. One of the undesirable features of the older units was the somewhat cumbersome flow-rate control scheme. The new electronic control, however, appeared to lose its calibrated flow rate when delivered to the sampling sites. Considerable variation was observed in the total volume of air collected. Tables XV-XVIII show the relevant data for the ground-level samples and arcs A, C, and D.

b. Airborne. The whole-air sample bags fabricated for use on the Turbo Beech (Fig. 20) were made of laminated polyethylene Mylar plastic sheet and were designed to collect a sample of ~100 l. The polyethylene surface was on the insides of these bags. Mechanically these bags and the air inlet apparatus functioned quite well.

2. Cryogenic Samplers. Two new Airco cryogenic samplers were employed on this experiment. Allied Chemical, INEL, had recently obtained these units for other programmatic applications. They were willing to provide them for this experiment to obtain data of mutual interest. These two samplers were operated by two Allied Chemical technical personnel at the airports at Dubois, ID and Billings, MT.

3. Evacuated Cylinder Samplers. Two distinct engineering approaches were applied to two experimental evacuated samplers. Pertinent data for these samples is shown in Table XIX.

a. Propane Tanks. Standard propane tanks (25 gal) were used as experimental evacuated samplers. The tanks were fitted with calibrated leak orifices to allow a sample to be collected for predetermined periods (~8 h). It was calculated and demonstrated that a total volume of 50 l of air could be collected in each cylinder with the carefully selected laser-drilled orifices. The inlet valves were manually opened and closed. The field activation of these

systems was accomplished upon notification or prearranged signal. This task was accomplished by certain designated individuals, some of whom were non-LASL participants engaged specifically to service these samplers; see list of participants Appendix B.

b. Reclaimed Breathing Cylinders. Four reclaimed breathing O_2 cylinders (~295 l) were obtained and outfitted with adjustable low-flow-rate controllers. These controllers were preset to collect 200-l (~70% of container volume) samples in a 12-h period. Operational activation of these experimental samplers was accomplished by both LASL and casual non-LASL participants. (See Appendix B.)

4. Other Samplers and Instrumentation. Several other innovative types or modifications of older sampling techniques were also used by non-LASL participants. Included among these were (1) BNL sequential bag samplers (1 l per sample), (2) Lovelock automated sequential perfluorocarbon samplers designed for NOAA/ARL, and (3) two real-time perfluorocarbon measuring devices designed by Lovelock. One of the real-time instruments designed as an airborne measuring device did not function properly. The other two perfluorocarbon samplers performed properly.

b. BNL used two versions of their on-line SF_6 detection and measuring devices. One of these was designed to provide a 90-s real-time measurement and the other a 3-min real-time measurement of SF_6 (See Refs. 7 and 8). A third BNL instrument to measure continuous on-line total perfluorocarbons and SF_6 was brought to the site but developed difficulties and was not used during the experiment. All three of these instruments were designed to be used as airborne monitoring devices. The two SF_6 devices were flown on the EPA Turbo Beech aircraft (Fig. 20). Late-developing minor instrumentation problems precluded use of the 3-min device during the flights.

Two on-line real-time perfluorocarbon measuring devices were designed by Lovelock in Britain. These instruments were designed for ARL and have been tested by HASL. One of them was to be used as an airborne measuring device, but it now appears it did not function properly during the INEL flights. The ground-based Lovelock instrument, attended by Peter Simmonds of Lovelock, appears to have functioned properly.

E. Sampler Deployment

The air samplers used on this tracer experiment were generally deployed on concentric arcs beginning at 3.2 km (2 mi) and going out to ~300 km (180 mi)

from the release sites. See Fig. 6 for schematic drawings of these emplacement arcs. The inner arc, A, was laid out on a permanent circular arc with no concern for terrain obstacles while the other two arcs were made to conform to acceptable surface access routes wherever possible. The outer fringes of both the C and D arcs were serviced via helicopter.

1. A-Arc. A-arc at 3.2 km (2 mi) consisted of ARL bag samplers placed at 4° intervals beginning on a line running north from the release point for 80° of arc to the southeast.

2. C-Arc. Bag-type samplers were emplaced on a line running from a point due north of the tracer-release site on a line generally following an arc southeast for 80° of arc. This arc was approximately 50 km (30 mi) from the release point. Samplers near the center of this arc were placed at 1.6 km (1 mi) intervals with those on the outer fringes of the arc at intervals ranging from 3.2 km (2 mi) to 8 km (5 mi). The samplers emplaced along roads and highways were deployed and serviced by personnel in cars. The inaccessible sites were serviced by a helicopter-transported team.

Special samplers and the on-line Lovelock perfluorocarbon measuring instruments were located on C-arc at sampling site 321, sequential perfluorocarbon samplers were at positions 317, 319, 323, and 325. This area was selected based on observed and predicted trajectories in the tracer release.

3. D-Arc. The bag samplers on D-Arc were emplaced at a nominal 3.2-km (2-mi) interval near the center of the arc. Near the southern edge these samplers were set ~6.4 km (4 mi) apart. The first sampler at the northern end of the arc was implaced ~13 km (8 mi) east of a line extending due north of the release point. The southernmost sampler was emplaced on a line running at ~80° (magnetic) from the release point. At the Dubois Airport (Fig. 6) the LASL sequential bag samples were collected to determine plume passage times at that location. Also located at that site were one of the two cryo-samplers and one of the 110-l (25-gal) evacuated cylinders. At site 420 another sample was collected in one of the 110-l (25-gal) containers in addition to the normal bag sample.

4. Outlying Locations. Five sampling sites were set up in Montana and Wyoming at distances ranging from 419 km (260 mi) to 270 km (168 mi). These sites were manned and serviced by Allied Chemical at Billings, MT; LASL at Lodge Grass, MT; Bozeman Cooperative Station in Bozeman, MT; and volunteers in Sheridan and Riverton, WY.

The Bozeman, Billings, Lodge Grass, and Sheridan sites had evacuated samplers fabricated from the reclaimed breathing O_2 containers. In addition, the Billings site was equipped with the second Airco cryo-sampler. The Riverton site was provided with two of the 110-l evacuated propane cylinders in parallel with a single inlet. Table XIX lists pertinent sampling data for these units.

F. Sample Processing and Tracer Measurements

1. Methane-16 and SF_6 Background Measurements. No new measurements of ambient CH_4 have been made from samples of air collected in the INEL area. An assumed value of 1.7×10^{-6} v/v has been used in all calculations involving CH_4 and $^{13}CD_4$ - $^{12}CD_4$ ratios. A SF_6 background of 0.5 ppt has been applied to all the SF_6 data furnished by BNL.

2. Air-Sample Processing. Field processing of air samples collected during the INEL, 1977, experiment was limited to bag samples from the ground and aircraft. These samples were collected in Saran or polyethylene-Mylar laminated plastic bags. These processes are described below.

a. Aliquots were taken from the bag samples for subsequent analysis of SF_6 , PDCH, and PDCB by BNL. These aliquots were placed in ~2-l Saran bags, and in a number of cases, a second (backup) 1-l aliquot was transferred to an evacuated steel cylinder.

b. Samples for heavy methane and ^{85}Kr analysis were transferred from their collection bags by immersing aluminum (Al) or stainless steel (SS) transport cylinders in liquid nitrogen and condensing the air from the bags into cylinders. This was basically the same technique used in the air-sample transfer at SRP in 1975. The 3.5-l Al cylinders had a shorter transfer time than the SS cylinders because of better heat transfer characteristics and a larger cooling surface area. Several SS cylinders were used because not enough aluminum cylinders were available to make all the sample transfers. Experience gained during the SRP sample transfers proved invaluable in promoting a more expeditious sample transfer at INEL. See Table XX for sample processing data.

3. Cryo-sample Processing. The samples collected by the two Airco, Inc.-INEL cryogenic devices were removed from the collection apparatus and sent directly to Airco, Inc., for final concentration and separation of krypton and methane. The concentration and purification steps were the same as those used for cryo-samples on the two previous experiments.

4. Special Samplers. Samples gathered in the Lovelock cassette samplers were returned (see V.D. 4) to HASL for tracer analysis. The real-time perfluorocarbon sampler, operated at Site 321, provided a read-out of concentration at 5-min intervals during the experiment.

Evacuated cylinder samplers were returned to LASL where their contents were removed by liquid helium transfer. The cryogenic transfers were accomplished using standard gas and vacuum lines routinely employed in similar tasks by members of LASL Group CNC-11. These transfers were made into the same type Al or SS cylinders used to transport the bag samples to Airco, Inc., for sample processing; they will be analyzed in the same manner as the other samples processed by Airco, Inc.

5. SF₆, PDCB, and PDCH Measurements. Measurements and analysis for SF₆, were conducted by ARL/FRO and BNL. Measurements of the PDCB and PDCH were accomplished by HASL and BNL. The measurement techniques used by these groups are presented below.

a. ARL/FRO removed small amounts from each bag sample for a preliminary measurement of SF₆. The samples were analyzed on a gas chromatograph and the results served as a criteria used by the other participants for their sample selection for detailed tracer analysis. It had been hoped that these early SF₆ measurements would identify the location of plume-crossing on the three arcs. It appears that, due to instrumentation sensitivity levels, SF₆ concentrations below the 6-ppt level were not detectable by ARL/FRO. The BNL results were limited only by the natural background (~0.5 ppt). See Table XXV.

b. BNL made SF₆, PDCB and PDCH measurements on a newly designed laboratory gas chromatograph. Their analytical apparatus and technique are described in Ref. 8.

c. The cassette samples collected by the Lovelock sequential sampler were returned, as collected, to HASL for analysis in special chromatographic apparatus designed by Lovelock. The sample collection cassette and the automatic analyzing system are coupled together in the analysis phase of the perfluorocarbons. The collection tubes of molecular sieve are automatically heated, in sequence, desorbing the sample into the analyzer. The tracer gases are flowed into a chromatographic column and into an electron capture detector.

d. On-line real-time (90-s) measurements of SF₆ were made by BNL on board the aircraft. A typical instrument response is shown in Fig. 22.

channeling of the gradient flow and diurnal topographic influence maintained the requisite southwesterly wind until the tracer plume had passed the grid.

3. Estimates of Diffusion Parameters. The best available wind data for estimating turbulence inputs to plume growth for the experiment were on strip chart recordings from the TRA site, located close to the release point. The estimates were derived from wind direction fluctuations measured at 15.2 m above ground. It is desirable to have information on two major scales of turbulence, the small, mechanically driven eddies that account for local plume growth and the larger scale, quasi-horizontal meander. To do this, we estimated the standard deviation of azimuth wind fluctuations, σ_A over 5-min average. This gave the high-frequency or small-scale component. The low-frequency standard deviation was calculated directly from the set of 5-min means in the longer record, in this case 1 h. The 5-min σ_{A5} values were estimated from the full range of the direction trace using the ratio suggested by Marke¹⁵ and others,

$\sigma_{A5} = \frac{\text{Range}}{6}$. The total 1-h σ_{A60} is given by

$$\sigma_{A60} = (\sigma_{A5}^2 + \sigma_{A(60-5)}^2)^{1/2}.$$

Table XXI gives wind speed, wind direction, and σ_{A60} obtained from the TRA site 15-m strip charts. The average of the σ_{A60} values during the 3 h of tracer release was 24° , which is consistent with the most unstable meteorological conditions (e.g., Pasquill-Gifford Category A).

The other vital parameter for estimating air concentration is the mixing depth. This was estimated from the hourly radiosondes using the height of the first elevated inversion. Using the inversion heights between 1400-1900 MST from Table XXII gives an effective mixing height for the cloud of 2.5 to 3 km.

The observed plume width diffusion parameter σ_y was estimated by two methods for comparison with the meteorological projection from the extrapolated Pasquill-Gifford model, as shown in Table XXIII. One estimate comes from the second moment of a least squares fitted Gaussian curve through the observed crosswind SF_6 concentration data. The other estimate is the square root of a directly calculated second moment of the SF_6 crosswind concentration distribution at the three sampling arcs.

Loading the observed meteorological parameters into the simple Gaussian plume model, we obtained estimates of average air concentration during cloud

passage. The estimates are compared with observed average SF_6 and methane concentrations in Table XXIV. The agreement between measured and calculated centerline concentration is very good, typically within about 20%. The success of such a simple model suggests that under the conditions of the experiment, the atmospheric structure was characterized quite well by the simple parameters outlined above.

H. Results

1. Tracer Concentrations

a. SF_6 Concentrations. The concentration of SF_6 in nearly all of the available samples collected during the experiment has been measured. Many of the samples were analyzed at INEL by gas chromatography immediately following the experiment and those results were to serve as an indication of which samples were to be analyzed by BNL and LASL. Unfortunately the SF_6 sensitivity of the instrument at INEL was limited to ~ 6 ppt, and therefore some of detail in the SF_6 distributions on the various sampling arcs was not verified in these analyses (i.e., no SF_6 was detectable in the D-arc samples with this instrument).

Based partly on the preliminary measurements at INEL, samples were taken in both 2-l Saran bags and in 1-l evacuated steel cylinders for analysis at BNL. The instrument at BNL had a much greater sensitivity than the INEL instrument and measurable SF_6 was found in almost all of the available samples.

The main difficulty associated with BNL analysis came from loss of samples in shipment. The 2-l bag samples sent to BNL were in transit for 3 months. This delay resulted in the loss of many samples, forcing the use of back-up samples in 1-l cylinders which were less desirable for perfluorocarbon analysis.

The SF_6 concentrations measured by BNL in the samples from the various arcs are presented in Table XXV and are plotted in Figs. 27-29. A background of 0.5 ppt has been subtracted from the data supplied by BNL before they were plotted.

Centerline concentrations at each of the downwind arcs were compared with estimates from a simple meteorological model and agreed within 20%. Plume width parameters agreed to within 30% of estimates from the Pasquill-Gifford model curves for the A category.

The samples that have been analyzed for SF_6 from the D-arc are somewhat erratic and it has been proposed that there was some instrument difficulty in addition to possible contamination. The ratio of SF_6 to PDCH appears to be correct for at least eight of the samples analyzed and the distribution shows a broad poorly defined maximum around position 418.

b. Methane-16, -20, and -21 Concentrations. The measured concentrations of $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ at the various sampling positions along arcs A, C, and D are presented in Table XXV. The measured distributions of $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ along the sampling arcs are depicted in Figs. 23-25. The concentrations of $^{13}\text{CD}_4$ and $^{12}\text{CD}_4$ were derived from the mass spectrometrically determined abundance ratios of $^{13}\text{CD}_4/\text{CH}_4$ and $^{12}\text{CD}_4/\text{CH}_4$ in air samples. This procedure has been described earlier (see Sec. IV. G). In lieu of on-site ambient CH_4 measurements, a value of 1.7 ppm was used in the calculations of the $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ concentrations.

Only two of the aircraft samples have been analyzed. Both of them were taken during the dry run on April 15, 1977. No $^{12}\text{CD}_4$ was released and only background concentrations of these tracers would have been expected. The measured concentrations, however, are extremely high (see Table XXV), which confirms that the bags used for these samples were contaminated during fabrication at LASL. These bags were fabricated in the same building that the tracers were stored.

The single measurement at position 112 on arc A gave concentrations close to those expected from the meteorological conditions and release ratios. Furthermore, the measured ratio of SF_6 concentration to that of $^{12}\text{CD}_4$ is very close to that of the mixture released indicating that both tracers are conservative for transport distances of 3.2 km. The observed concentration of $^{12}\text{CD}_4$ is lower than expected from consideration of release quantities alone. The $^{12}\text{CD}_4$ was released some 5 km crosswind from the SF_6 - $^{13}\text{CD}_4$ release point and calculations indicate that little $^{12}\text{CD}_4$ would have diffused laterally into the region of the SF_6 $^{13}\text{CD}_4$ plume after a transit of only 3.2 km.

Only a few positions on the C-arc were sampled for $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ (see Table XXV and Fig. 23). The data show rather large fluctuations and do not track well with either the SF_6 or ^{85}Kr measurements. It now seems that the only plausible explanation for the large variations is that the samples were contaminated during processing after collection.

In the case of the D-arc, Figs. 24 and 25, the data again show large variations, even in adjacent samples, and this also suggests that some of the samples were contaminated during handling. The data, however, show a trend with a broad maximum at approximately position 420. This peak agrees well with that observed for the ^{85}Kr and SF_6 as seen in Figs. 26 and 29.

A crude box model calculation using a mixing depth of 2.8 km, an average wind speed of 25 km/h, and an average D-arc $^{13}\text{CD}_4$ volume fraction (v/v) of 7×10^{-14}

gives an integrated plume mass of ~1600 g. This represents an amount ~20 times that released. This simple calculation again points to probable sample contamination at a level that makes experimental interpretation questionable.

Additional evidence supporting the contamination theory comes from comparison with the Savannah River experiment where similar amounts of $^{13}\text{CD}_4$ were released and a similar transport distance was involved. In the SRP experiment, a peak concentration of 6×10^{-15} v/v was observed, whereas on the D-arc in the INEL experiment, concentrations as high as 2.5×10^{-13} were observed. This large difference seems impossible, especially when one considers that at INEL, the meandering of the wind would be expected to spread the plume and hence, lower its concentration.

Finally, evidence for sample contamination can be derived from comparison with the SF_6 results. In Fig. 30 the concentration of $^{13}\text{CD}_4$ vs that of SF_6 for each sample is plotted. If there were no postrelease fractionation of SF_6 relative to $^{13}\text{CD}_4$, all of the data points would fall on the line indicating the release ratio of SF_6 to $^{13}\text{CD}_4$ (504:1). Here, again, the point for the A-arc is close to that line. The data for the C- and D-arcs show a great variability; however, two observations can be made. First, the D-arc data are generally farther from the expected line than the C-arc data are. The D-arc samples are to be expected to be lower in concentration than those of the C-arc so that, for a given amount of contamination, the ratio of SF_6 to $^{13}\text{CD}_4$ will be perturbed more, and hence, the points would tend to be farther away from the release ratio line than those of the C-arc. Second, most points indicate that the samples are enriched in $^{13}\text{CD}_4$ relative to SF_6 . Both observations support the contamination theory. The lower background and higher detection sensitivity for $^{13}\text{CD}_4$ relative to SF_6 makes it more vulnerable to contamination and, in fact, the SF_6 data are probably also contaminated, but it is not as obvious because of the significant natural SF_6 background.

c. ^{85}Kr Concentrations. The ^{85}Kr activity (dpm/cm³) associated with each sample was determined as in the SRP experiment and is listed in Table XXV. The data are plotted as a function of sampler position in Fig. 26. As expected, comparisons between ^{85}Kr and the other tracers released are not straightforward since ^{85}Kr was being released continuously. This facility was 2.9 km, slightly upwind, from the tracer-release point and it seems reasonable to assume that the ^{85}Kr behaved in a similar fashion to the other tracers.

The data in Fig. 26 show that the ^{85}Kr was transported radially and was deflected by a somewhat meandering wind pattern. Relative to the A- arc, it appears that the plume first veered to a more easterly direction, then back westerly upon reaching the D-arc. The data from the C-arc are too sparse to predict a plume width (σ) but the peak seems to be at about position 327. The ^{85}Kr data for the D-arc indicate a peak around 418 with $\sigma = 21$ km. This is about twice the width of the other tracer plumes on this arc and is probably explainable by the different source configuration of the ^{85}Kr .

Several types of samplers were operated at the Dubois Airport for comparison. Only the 110- ℓ evacuated cylinder and the conventional bag samplers can be compared at this time, and they show an unexplained difference in ^{85}Kr concentration. Sampling site 420 also had both types of samplers, a bag and 110- ℓ evacuated cylinder. The ^{85}Kr analysis of the cylinder sample appears to be about as expected although the corresponding number is not available for the bag sampler.

Only two of the aircraft samples have been analyzed for ^{85}Kr . These samples were collected during the dry run on April 5, 1977. Pertinent data for these two samples are shown in Table XXV. Sample DR-4 was collected at 2042 m above mean sea level (MSL) near the reprocessing facility, and shows an expected high level of ^{85}Kr . The other sample, DR-1, was collected at 2438 m MSL 16 km west of Idaho Falls and shows an activity level near background.

2. Sampler Comparisons. An effort was made to quantitatively compare the conventional ARL/FRO bag sampler with the Airco cryogenic sampler and the two types of evacuated cylinder samplers.

The only directly comparable results come from samplers at the Dubois Airport. The ^{85}Kr data (Table XXV) for a bag sampler and one of the LASL evacuated cylinders at this position show an unexplained, rather poor, agreement with the evacuated cylinder being low. The Airco cryogenic sampler, also at the Dubois Airport, should have provided ^{85}Kr and heavy methane data, but based on the amount of gas collected, the analyzed sample represents a collection over a longer period than the 7-h period used for operation at this site. Furthermore, the ^{85}Kr activity level is at background and the concentration levels of $^{13}\text{CD}_4$ and $^{12}\text{CD}_4$ are at the lower limit of detection. It seems reasonable to assume that this sample was, in fact, a background sample taken at some other time and place. Future analysis

of remaining samples will provide more data for comparison with samples on the D-arc as well as those at Billings, MT (Airco cryogenic sampler) and a 295-l evacuated sampler.

3. Long-Range (~300-km) Samplers. At this time the samples taken in evacuated cylinders at ~300 km downwind have not been analyzed. It is felt that these samples are the least likely to have been contaminated and will therefore provide useful results from analysis of both ^{85}Kr and heavy methane.

4. Aircraft. The aircraft activity, from an operational viewpoint, went smoothly. Two sampling/data flights and dry runs were conducted, see Table XXVI for details of location, time, altitude, etc., of these flights. The dry run was used to "shake out" instrumentation problems, acquaint the air crew with the experimental area, and to collect background samples and data. The two "live run" flights were flown over the primary sampling lines as shown in Fig. 7 and over the secondary distant sampler sites as seen in Fig. 6.

The only real-time instrument which produced useful data was the BNL 90-s SF_6 device. A plot of typical data from this instrument reveals tracer-plume contact as shown in Fig. 22. The other BNL SF_6 monitor and Lovelock perfluorocarbon monitor were not functional.

The air-sample inlet to the aircraft worked well by providing adequate air supplies for sample collection and tracer measurements. The bag sampler scheme was a prudent method for obtaining whole-air samples.

The ^{85}Kr analysis may still provide useful data in the determination of a plume mixing depth. We expect these to be available at a later date.

I. Conclusions

Most of the stated objectives were achieved to a greater or lesser degree. The successful achievement of many of these was overshadowed by the problem of sample contamination, which caused questionable analytical results. The question of SF_6 - $^{13}\text{CD}_4$ material balance at 90-100 km raised during the SRL experiment has not been resolved.

The technique developed for simultaneous release of SF_6 and $^{13}\text{CD}_4$ performed well and gave the desired controlled release as verified by manifold samples taken during the release. New, evacuated tank samplers were field tested for the first time and performed well. Samples were collected by this technique at distances of up to 300 km downwind. Direct comparisons were made between this type sampler, conventional bag samplers, and Airco, Inc., cryogenic samplers.

The Turbo-Beech aircraft performed well during the INEL experiment and, in comparison with the Martin 404 used during the SRP experiment, was more responsive to experiment needs. Additionally, the operating cost and personnel requirements were significantly lower. This type of aircraft should be very useful in future work if instrumentation is developed to take full advantage of its capabilities.

We have found that closely following the progress of the various samples through processing has resulted in their expeditious treatment. Furthermore, there has been much less confusion as to sample identification and processing status than had been experienced earlier.

Finally, it is necessary to conclude that many of the samples from this experiment were inadvertently contaminated. This conclusion applies to all of the tracer systems used, although the methane tracer systems were more sensitive than others to contamination.

J. Recommendations

As in the case of the SRP experiment, a number of operational and technical problems were identified. Many of the needed improvements that were brought to light during the SRP experiment were implemented during the INEL experiment. Problem areas that still need attention are outlined below.

1. Sampler Deployment. Communications were much improved over the SRL experiment, but there is still a problem of properly deploying, monitoring, and retrieving the downwind samplers. Some of the sampling personnel involved in the INEL experiment were not adequately informed as to procedures to be used with respect to record keeping, sampler handling, and troubleshooting. An experiment involving this number of people demands dynamic leadership which will ensure good on-site personal communications with these participants.

2. Tracers. It is now apparent that many of the samples were inadvertently contaminated either before or after the tracer release. In the future much more attention will be directed to the prevention of this situation. All tracers to be used must be stored, at all times, in a location remote from areas in which any sample handling or analyses will be done. This is true both before and after an experimental release of the tracers. Special attention must be given to the transport of tracers to and from an experimental location to prevent possible sample contamination during shipment. Personnel involved with the release of the tracers must not handle or process the collected samples because their clothing, and perhaps even their bodies, will undoubtedly remain contaminated for some time after the release.

There is definite need to monitor continuously the ambient conditions (preferably in real time) during all sample handling and analysis operations to ensure that accidental contamination is not taking place. Furthermore, in the case of heavy methane tracers, the natural ambient background concentration of CH_4 at the experimental area must be measured, preferably, before, during, and after the tracer release. It is necessary to know this natural background for final calculation of the $^{13}\text{CD}_4$ and $^{12}\text{CD}_4$ concentrations in the atmosphere.

The discrepancy of concentrations observed between SF_6 and heavy methanes observed at the SRP experiment raised questions as to whether the heavy methanes are indeed conservative atmospheric tracers. The test at INEL was designed to try to answer that question and because of the aforementioned contamination problems the question remains. Other tests of a less complicated nature on a smaller scale should be designed and executed to explicitly answer this question.

3. Samplers. The bag samplers used at INEL were similar to, but improved versions of those used at SRP. Most of the problems encountered at SRP with these samplers (see Sec. IV) were still in evidence at INEL. It is apparent that to be reliable for routine use, the bag samplers must be further improved to give constant sampling rate, independent of temperature. The Saran bags were again structurally marginal, expensive, and required individual testing. A more acceptable substitute will be sought.

4. Aircraft. All aircraft instrumentation used for measurement of atmospheric tracers must be built to the rigid specifications required of other normal aircraft instrumentation. Acceptance testing of these experimental instruments must be conducted to insure satisfactory operation in the hostile aircraft environment. Interfacing of special instrumentation with airborne platforms must begin well in advance of the desired use period to ascertain compatibility and operability of these special instruments.

VI. PROGRAM RESULTS AND DISCUSSION

A. INEL - 1974

1. $^{13}\text{CD}_4$ released as a tracer demonstrated its usefulness as an atmospheric tracer over a range of ~2500 km.

2. The measured concentrations of $^{13}\text{CD}_4$ at the sampling sites were somewhat lower than originally predicted from calculations based upon the ARL meteorological model.

3. The extremely favorable signal-to-noise ratio demonstrated in the measurement of $^{13}\text{CD}_4$ by the Daly group encouraged experimenters to consider also mass-20 ($^{12}\text{CD}_4$) methane as a long-range tracer. The lower production cost of $^{12}\text{CD}_4$ makes this compound attractive as a complementary tracer with other atmospheric tracers such as $^{13}\text{CD}_4$ and SF_6 .

4. We have used both $^{13}\text{CD}_4$ and $^{12}\text{CD}_4$ on the same experiment and we believe it is worthwhile.

B. SRP - 1975

1. $^{13}\text{CD}_4$ and $^{12}\text{CD}_4$ were both released during this experiment. This first test release of $^{12}\text{CD}_4$ proved to be no more difficult than that previously encountered in $^{13}\text{CD}_4$ releases and gave good comparative results.

2. $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$ were both detected and measured in samples collected at a nominal 100 km. A signal-to-noise level of 160 and 120 was found for both tracers, respectively.

3. The background of $^{13}\text{CD}_4$ was estimated by considering the samples from SRP that were collected far from the center of the plume. In these cases, it was estimated that the $^{13}\text{CD}_4$ concentration was <1 part in 10^{11} relative to $^{12}\text{CH}_4$. This estimate is limited by the instrumentation sensitivity. The calculated statistical background of $^{13}\text{CD}_4$ is ~ 5 parts in 10^{18} of CH_4 , not considering any anthropogenic sources. If the total known releases to date (~ 300 g) are considered, the calculated background is ~ 4 parts per 10^{14} of CH_4 , assuming no destruction of $^{13}\text{CD}_4$.

4. Good qualitative correlation between $^{13}\text{CD}_4$ and SF_6 and fair correlation between plant-process-produced ^{85}Kr was observed in the 100-km samples. The relatively long-term release of the ^{85}Kr was the probable cause of the poorer $^{13}\text{CD}_4$ - ^{85}Kr correlation factor.

5. The tracer dispersion observed in this experiment compares favorably with that predicted by transport and diffusion models.

6. No evidence has been uncovered to support a theory for any separation-induced exchange of the CH_4 and the heavy methanes.

7. A discrepancy was observed in mass-balance comparisons between the heavy methanes and SF_6 . The heavy methanes agree with one another, but are about a factor of 4 low compared to SF_6 . No logical solution for this discrepancy has been suggested.

C. INEL - 1977

1. New potentially useful atmospheric tracers were introduced into this experiment. Two perfluorocarbons, PDCB and PDCH, were released to be compared with the previously tested SF_6 and heavy methanes.
2. Several new air-sampling devices (evacuated cylinders and sequential samplers) were successfully employed at INEL.
3. Real-time, on-line instrumentation to measure SF_6 and the perfluorocarbons was introduced for the first time at INEL. The new airborne instrumentation did not function as designed; however, the ground-base perfluorocarbon measuring devices functioned well. Numerous improvements and field hardening of all these instruments was indicated in these trials.
4. The new premix $\text{SF}_6/^{13}\text{CD}_4$ release scheme tested during the experiment worked very well. Release-mix samples taken from the outlet manifold and subsequently analyzed for $\text{SF}_6/^{13}\text{CD}_4$ agree with the mixture ratios in the storage mixing vessel.
5. The heavy methane analysis was compromised by cross-contamination.

D. Results Summary

1. Heavy methanes have amply demonstrated their utility as long-range atmospheric tracers to 100-2500 km.
2. Analytical sensitivity of the Daly gas mass-spectrometric technique is sufficient to insure suitable signal-to-noise ratios for analysis of heavy methane samples gathered at long ranges.
3. The analysis of low-level heavy methane samples can be accomplished at costs that are not prohibitive.
4. The relatively small amounts of methane tracers required in long-range atmospheric transport and diffusion studies is economically attractive.
5. Release techniques used for $^{13}\text{CD}_4$ and $^{12}\text{CD}_4$ are relatively simple when compared to the high-molecular-weight tracers such as SF_6 and exotic perfluorocarbon compounds.
6. Experiences and observations of the results of these three experimental tracer releases vividly point out the need for complementary use of a number of tracers.
7. The low natural abundance of mass-20 and -21 methanes has provided us with powerful atmospheric tracers. The large dynamic range evidenced in these measurements presents certain hazards to the users, which must be circumvented in the field-use of these tracers. Judicious employment must be exercised in

release of the tracers if any eventual utility is to be realized in the global application of such tracers. The postulated 4- to 7-yr life of these tracers in the atmosphere may limit the useful yearly release of these compounds. It is highly desirable to control the world background levels of these compounds so that a mass-21(-20)/mass-16 ratio of $\sim 1 \times 10^{-13}$ is not exceeded, which means that no more than 400 kg of each tracer should be released each year unless a single, long-term, worldwide experiment is required. Total releases have averaged less than 75 g per year for the past 5 years.

VII. RECOMMENDATIONS

Experience and knowledge gained as a result of this Research Initiatives Project precipitate the following recommendations.

1. Continue work leading to the development of improved tracer sampling technique and apparatus, i.e., low-cost dependable evacuated cylinders and evacuated cryogenic samplers.
2. Pursue a diligent effort to insure good quality control to prevent minute cross-contamination of tracers, samples, and analytical apparatus.
3. Investigate the basic properties and reaction behavior of CH_4 , $^{12}\text{CD}_4$, and $^{13}\text{CD}_4$ as influenced during atmospheric residences.
4. Develop an adequate data bank of ambient CH_4 concentrations to promote an understanding of its sources, sinks, and natural fluctuations.
5. Continue the development of gas mass spectrometry to provide better sensitivity levels that may allow us to analyze small methane samples.
6. Investigate the development of mobile heavy methane dilution instrumentation that can be employed on airborne platforms. This instrumentation should be capable of real-time, in situ, readout in order to obtain, at least, early time vertical and horizontal plume development information.
7. Develop an in-house capability for the rapid separation of methanes in preparation for mass-spectrometric analysis.
8. Undertake an active pursuit of programmatic support to continue this research and its practical application to the solution of atmospheric transport and diffusion questions.

ACKNOWLEDGMENTS

Scientists and technicians from several DOE laboratories, EPA, NOAA, and their contractors participated in the work reported here (see Appendix A). They are all to be commended for their splendid efforts and support of these activities. Special note is made of the following groups or individuals. Gilbert Ferber (NOAA/ARL) was largely responsible for bringing together those people with common interests and required expertise which made these experiments possible. Anthony Turkevich, University of Chicago, provided invaluable technical consultation in all phases of these experiments. Nicholas Matwiyoff, Mohammed Alei, Jr., Maxwell Goldblatt, and Thomas Whaley (LASL) were instrumental in the production of ^{13}C and the synthesis of the heavy methane tracers. Robert and Helen Bench (Airco, Inc.) and their technicians provided the essential methane- ^{85}Kr separation and ^{85}Kr analysis for all the samples as designated. The personnel of the Norman Daly group (AWRE), in particular Nan Campbell, were responsible for all of the heavy methane analysis. Russell Dietz and his group at BNL provided real-time SF_6 measurements, as well as detailed analysis and interpretation of samples gathered at both SRP and INEL. The two host groups for the three field experiments described here were the Todd Crawford group (SRP) and the Ray Dickson group (ARL/FRO) at INEL. The National Weather Service personnel, under the direction of Harold Mueller at the Nevada Test Site, were generously loaned to the 1977 INEL experiment by the Nevada Operations Office at Las Vegas, NV. The continued support from the Director's New Research Initiatives Program at LASL has enabled us to become a vital part of these atmospheric-tracer efforts with sponsorship now transferred to DOE/DBER.

Without the combined secretarial assistance of Alice Staritzky, Etheleen Willow, and Jean Holmes, all of CNC-11, this report could not have been published. We extend our thanks to them for their very competent help in putting this report together and preparing it in its final form. The editorial supervision and assistance provided in LASL's group ISD-6 was invaluable in the preparation of this report.

TABLE I
RATIOS OF $^{13}\text{CD}_4$ TO $^{12}\text{CH}_4$ IN AIR SAMPLES
(INEL 1974)
 $^{13}\text{CD}_{14}/^{12}\text{CH}_4$
(units of 10^{-11})

Day	Minneapolis, MN			Rochester, MN			Waterloo, IA			Detroit, MI		
	Obs	Net	Calc	Obs	Net	Calc	Obs	Net	Calc	Obs	Net	Calc
May 15 PM	3.7 ± 1.7	0.0 ± 1.7	0	---	---	0	---	---	0	---	---	0
May 16 AM	7.0 ± 1.3	3.3 ± 1.3	2.4	---	---	0.25	---	---	0	---	---	0
May 16 PM	6.0 ± 1.0	2.3 ± 1.0	21	8.4 ± 1.3	4.7 ± 1.3	20	4.1 ± 0.7	0.4 ± 0.8	6.0	---	---	0
May 17 AM	5.6 ± 0.7	1.9 ± 0.8	4.8	5.2 ± 0.7	1.5 ± 0.8	16	5.5 ± 0.7	1.8 ± 0.8	18	---	---	0.04
May 17 PM	4.7 ± 0.8	1.0 ± 0.9	10	5.5 ± 0.9	1.8 ± 1.0	12	5.0 ± 0.8	1.3 ± 0.9	7.5	3.8 ± 0.9	0.1 ± 1.0	0.7
May 18 AM	5.0 ± 0.9	1.3 ± 1.0	3.3	3.7 ± 0.8	0.0 ± 0.9	1.8	---	---	0.6	5.1 ± 0.8	1.4 ± 0.9	0.3
May 18 PM	5.1 ± 0.8	1.4 ± 0.9	4.4	3.2 ± 0.7	-0.5 ± 0.8	4.3	4.0 ± 0.8	0.3 ± 0.9	1.8	5.4 ± 1.1	1.7 ± 1.1	0.02
May 19 AM	3.4 ± 0.7	-0.3 ± 0.8	2.9	4.1 ± 0.8	0.4 ± 0.9	1.5	---	---	0.4	3.1 ± 0.5	-0.4 ± 0.6	0
May 19 PM	---	---	0	---	---	0	---	---	0	3.5 ± 0.9	-0.2 ± 1.3	0

TABLE II
TOTAL HOURLY RELEASES OF Kr-85
10 DECEMBER 1975
(SRP)

Time (EST)	^{85}Kr Release ^a (Ci)	Time (EST)	^{85}Kr Release (Ci)
0100	454	1000	152
0200	408	1100	127
0300	352	1200	110
0400	257	1300	94
0500	138	1400	81
0600	72	1500	70
0700	45	1600	64
0800	38	1700	56
0900	108	1800	24

^aEstimated amount released from 30 min before to 30 min after the indicated time.

TABLE III
SAMPLE BAG AND CYLINDER TESTS
(SRP)

<u>Sample Container^a</u>	<u>SF₆ Test Gas (ppt)</u>	<u>Days Elapsed After Filling</u>	<u>Meas. SF₆ Conc. (ppt)</u>
45-1	Zero gas	1	0.05
		6	0.04
45-2	1.90	0	2.02
		5	1.87
		5	1.96
45-3	38.0	0	41.1
		5	38.9
		5	40.7
45-3	Zero gas	0	0.03
		1	0.03
34	from 45-1	18	0.04
29	from 45-2	18	2.01
		18	1.97
33	from 45-3	18	38.0
		18	38.7

^a 45-1, -2, -3 Saran bags, 45-ℓ.
29, 33, 34 steel cylinders, 1-ℓ.

TABLE IV
GROUND-LEVEL SAMPLING DATA
(SRP)

Sample No. ^a	Sampler Position ^b	Collection Time (EST)		Collection Duration (h)
		Start	Stop	
30	I-95, Mile 76 ^c	11:49	19:12	7.38
18	73	11:46	19:02	7.27
25	70	11:39	18:53	7.23
31	67	11:27	18:46	7.32
6	64	11:15	18:36	7.35
59	61	11:04	18:30	7.43
63	58	10:55	18:20	7.42
2	56	10:40	18:10	7:50
57	54	10:51	18:20	7.48
65	52	10:59	18:30	7.52
40	50	11:08	18:38	7.50
60	48	11:15	18:45	7.50
48	46	11:22	18:55	7.55
4	44	11:31	19:05	7.57
8	42	11:46	19:16	7.50
52	40	11:55	19:24	7:48
46	38	12:05	19:35	7.50
51	36	12:14	19:44	7.50
44	34	12:23	19:53	7.50
10	32	11:19	18:27	7.13
5	30	11:26	18:33	7.12
64	28	11:33	18:41	7.13
3	26	11:41	18:47	7.10
55	24	11:49	18:53	7.07
49	U.S. 17 22 ^d	11:56	19:00	7.07
53	Route 336 ^e	12:08	10:07	6.98
35	Route 336	12:17	19:18	7.02
47	Route 336	12:26	19:27	7.02
61	(336) ^f	12:34	19:36	7.03
56	(336)	12:42	19:44	7.03
1 ^g	I-95, Mile 46	18:57		2.12
39	40	19:26		1.90
54	34	19:55		1.67

^aThe sample no. is the number of the bag in the sampler.

^bSamplers were positioned off of the highway at the mile markers.

^cMiles 64-76 were on an unopened section of I-95 and no mile markers were present. In these cases odometer readings were used.

^dSampler 49 was placed on US 17 running parallel to I-95 at a position corresponding to the 22-mile marker on I-95.

^eThere were no mile markers on SC-336; odometer readings were used for placement.

^fThe road west from Tillman, SC, has no number but is a natural extension of SC-336; odometer readings were used.

^gPost-plume samples.

TABLE V
AIRCRAFT SAMPLING DATA
(SRP)

Plume Profiles

<u>Pass No.</u>	<u>Downwind Distance (km)</u>	<u>Altitude Above Ground (m)</u>	<u>Aircraft Speed (m/s)</u>	<u>Start of 3-min Profile (EST)</u>	<u>3-min SF₆ Integral (ppt-s)</u>	<u>Start of 90-s Profile (EST)</u>	<u>90-s SF₆ Integral (ppt-s)</u>	<u>Peak SF₆ Conc. (ppt)</u>
1	16	150	77	1131	339	---	---	9.5
2	16	150	77	1151	4940	1151	6700	220
3	16	150	77	1212	4250	1213	199	124
4	16	300	77	1228	2400	1229	2650	139
5	16	450	77	1254	--- ^a	1254	--- ^a	--- ^a
6	42	300	93	1521	1740	1524	--- ^a	35
7	42	300	93	1550	1340	1552	--- ^a	22
8	92	150	93	1618	445	1621	1045	15
9	92	300	93	1650	677	1654	526	12
10	92	300	82	---	---	1720	--- ^a	16
11	92	600	93	1746	--- ^a	1749	--- ^a	--- ^a

Bag Samples

<u>Pass</u>	<u>Sample No.</u>	<u>SF₆ (ppt)</u>	<u>Kr-85 (pCi/m³)</u>	<u>(21/air) (cm³/cm³) x 10¹⁶</u>	<u>(20/air) (cm³/cm³) x 10¹⁶</u>
0	A/C-1	0.52	986	--	--
1	A/C-2	2.30	364	--	--
8	A/C-3	6.10	113	21.5	145
11	A/C-4	1.28	21.5	--	--

^aBackground.

TABLE VI
BACKGROUND SF₆ MEASUREMENTS FROM FIELD SAMPLES
DECEMBER 10, 1975

Sample No.	(SRP)	
	SF ₆ Concentration $\pm$ σ (ppt) ^a	
	14 April 70	16 April 76
18	0.50 $\pm$ 0.40	0.49 $\pm$ 0.01
22 ^b	0.50 $\pm$ 0.01	0.49 $\pm$ 0.03
56	0.55 $\pm$ 0.02	0.56 $\pm$ 0.01
30	0.50 $\pm$ 0.04	0.49 $\pm$ 0.03

^aThe standard deviation is for triplicate determinations and does not include that for the working standard (1.75 $\pm$ 0.03 ppt).

^bSample 22 was collected at the St. George cryogenic site.

TABLE VII
AMBIENT METHANE CONCENTRATIONS NEAR THE
SRP, DECEMBER 1975

Sample	Location ^a	Methane Conc. ^b (ppm)
1	H-AREA SRP	2.13
2	I-26, Mile 146	2.2
3	I-26, Mile 168	1.97
4	I-95, Mile 57	1.70
5	S.C.336, 1.5 mi from Savannah River	1.70
6	I-95, Mile 46	1.70
7	1	1.87
8	2	1.85
9	3	1.88
10	4	1.68
11	5	1.70
12	6	2.00
13	7	1.72
14	8	1.42
15	9	1.47
16	10	1.42
17	11	1.38
18	12	1.13
19	13	1.30
20	14	1.38
21	15	1.42
22	16	1.68
23	17	1.50
24	18	1.50
25	19	1.57
26	20	1.60

^aLocation, where indicated by number only, is indicated on the SRL Area Map (Fig. 1).

^bMeasured by gas chromatography.

TABLE VIII

AIRCRAFT BAG SAMPLE PROCESSING DATA
(SRP)

Sample No.	A/C-1	A/C-2	A/C-3	A/C-4
Cylinder No.	W4929	W4785	W4780	W4754
Pass No.	0	1	8	11
Time (EST)	0918	1131	1618	1746
Sample Started				
Altitude (ft above ground)	500	500	500	2000
Aircraft Speed (Knots)	150	150	180	180
Sample				
Weight (g)	59.97	113.6	105.7	98.54
Volume (ℓ)	50.6	95.9	89.2	83.2
Krypton:				
Spike (cm ³) ₃	8.99	8.99	8.75	8.63
Natural (cm ³) ₃	0.057	0.11	0.10	0.093
Recovered (cm ³)	8.1	8.3	8.2	8.1
Efficiency (%)	90	91	93	93
Methane:				
Spike (cm ³) ₃	0.72	0.63	0.55	0.62
Natural (cm ³) ₃	0.086	0.16	0.15	0.14
Recovered (cm ³)	0.75	0.66	0.50	0.70
Efficiency (%)	93	84	71	92

TABLE IX
GROUND-LEVEL BAG SAMPLE PROCESSING DATA
(SRP)

Air Sample			Krypton				Methane			
No.	Weight (g)	V _c ^a (ℓ)	V _s (cm) ³	Natural ^b (cm) ³	Recovered (cm) ³	Recovery Efficiency (%)	Spike (cm) ³	Natural ^c (cm) ³	Recovered (cm) ³	Recovery Efficiency (%)
30	16.99	14.3	9.12	0.016	7.5	82	0.58	0.024	0.59	98
18	22.84	19.3	9.24	0.022	7.9	85	0.61	0.033	0.46	72
25	46.30	39.1	9.50	0.044	8.7	91	0.66	0.066	0.62	86
31	13.32	11.2	7.81	0.013	6.6	84	0.64	0.019	0.60	91
6	55.28	46.6	8.69	0.052	7.5	86	0.58	0.060	0.56	86
59	42.08	35.5	9.12	0.040	8.3	91	0.59	0.060	0.56	86
63	19.40	47.5	9.27	0.053	2.9	31	0.45	0.081	0.16	30
2	19.06	33.2*	8.83	0.037	3.9	44	0.72	0.056	0.41	53
57 ^d	38.52	32.5	9.20	0.036	8.0	87	0.48	0.055	0.46	86
65	39.63	33.4	8.89	0.038	7.9	89	0.66	0.057	0.65	91
40	16.50	49.1	8.66	0.055	2.3	26	0.57	0.083	0.20	31
60	38.51	32.5	8.81	0.036	7.9	89	0.59	0.055	0.60	93
48	47.85	40.4	9.04	0.045	8.3	91	0.56	0.069	0.60	95
4	31.37	33.1*	9.00	0.037	6.5	72	0.72	0.056	0.52	67
8	12.91	34.9*	8.87	0.039	2.5	28	0.51	0.059	0.20	35
52	25.79	21.8	8.90	0.024	7.8	88	0.47	0.037	0.46	91
46	12.97	14.0*	8.94	0.016	6.3	70	0.62	0.024	0.47	73
51	33.50	31.6*	9.05	0.035	7.3	80	0.59	0.054	0.50	78
44	31.72	26.7	8.90	0.030	7.9	89	0.62	0.045	0.66	99
10	42.43	35.8	8.88	0.040	8.1	91	0.50	0.061	0.50	89
5	21.84	38.6*	8.71	0.043	3.4	43	0.69	0.066	0.35	46
64	48.62	41.0	8.67	0.046	8.0	92	0.74	0.070	0.78	96
3	28.06	23.7	8.84	0.027	7.8	88	0.65	0.040	0.64	93
55	36.49	30.8	8.96	0.034	7.3	81	0.74	0.052	0.72	91
49	37.71	31.8	8.86	0.036	7.9	89	0.57	0.054	0.46	74
53	32.06	27.1	8.97	0.030	8.0	89	0.48	0.046	0.44	84
35	27.22	23.0	8.67	0.026	7.7	89	0.57	0.039	0.50	82
47 ^e	7.84	6.6	8.64				0.65			
61	29.77	59.5* ^f	8.91	0.067	3.4	38	0.59	0.100	0.21	30
56	20.11	17.0	9.25	0.019	7.9	85	0.60	0.029	0.66	105
23	33.93	28.6	8.87	0.032	7.2	81	0.64	0.049	0.57	83
22	14.97	12.6	9.16	0.014	7.5	82	0.64	0.021	0.62	94

*Cylinder leaked; air volume corrected assuming 90% recovery efficiency for krypton.

^aAll gas volumes corrected to 25°C; 760 mm Hg (air density = 1.185 g/liter).

^bAbundance of atmospheric krypton = 1.112 ppm (v/v).

^cAbundance of methane at SRP = 1.70 ppm (v/v).

^dKr lost in counting.

^eAbundance of methane at SRP = 1.70 ppm (v/v).

^fBad sample; maximum capacity of bag was about 48 liters.

TABLE X
HEAVY METHANE CONCENTRATIONS FROM HIGHWAY BAG SAMPLES
(SRP)

Position I-95 Mile No.	(20/16) gas (cm ³ /cm ³) x 10 ¹¹	(20/air) (cm ³ /cm ³) x 10 ¹⁶	(21/16) gas (cm ³ /cm ³) x 10 ¹¹	(21/air) (cm ³ /cm ³) x 10 ¹⁶
76a	935.0 + 6.0	935.0 + 25.3	98.5 + 2.0	414.5 + 8.4
73a	260000.0 + 300.0	866000.0 + 999.2	10200.0 + 20.0	34000.0 + 66.7
70	1.02 + 0.30	1.9 + 0.56	0.10 + 0.30	0.186 + 0.557
67	3.18 + 0.38	18.71 + 2.24	1.36 + 0.34	8.00 + 2.00
64	0.99 + 0.33	1.40 + 0.47	0.54 + 0.33	0.764 + 0.467
61	0.64 + 0.35	1.17 + 0.64	-0.17 + 0.30	-0.311 + 0.550
58	4.47 + 0.72	4.99 + 0.80	0.83 + 0.52	0.927 + 0.581
56a	11.4 + 0.60	52.9 + 2.78	5.40 + 0.40	25.0 + 1.85
54	13.8 + 0.60	22.73 + 0.99	9.43 + 0.49	15.53 + 1.81
52	16.9 + 0.60	36.27 + 1.29	7.70 + 0.47	16.52 + 1.01
50	49.4 + 1.20	65.75 + 1.60	19.00 + 0.80	25.29 + 1.06
48	64.0 + 1.00	127.0 + 2.0	20.80 + 0.60	41.29 + 1.19
46	160.0 + 2.0	249.0 + 3.1	37.0 + 0.80	57.57 + 1.24
44	37.8 + 0.90	88.64 + 2.11	9.88 + 0.49	23.17 + 1.15
42	45.2 + 1.1	73.72 + 1.79	11.80 + 1.60	19.24 + 0.98
40a	20.8 + 0.6	48.4 + 1.40	6.73 + 0.42	15.6 + 0.97
38a	15.0 + 4.0	6.9 + 1.84	0.44 + 0.32	2.0 + 1.45
36	27.8 + 0.90	56.63 + 1.83	1.86 + 0.37	3.79 + 0.75
34	17.6 + 0.60	43.86 + 1.50	0.74 + 0.32	1.84 + 0.59
32	40.4 + 0.90	59.1 + 1.32	1.86 + 0.36	2.92 + 0.57
30	9.75 + 0.50	38.2 + 1.96	0.70 + 0.35	2.79 + 1.40
28	5.27 + 0.39	10.42 + 0.77	0.57 + 0.30	1.13 + 0.59
26	1.80 + 0.20	5.24 + 0.58	0.50 + 0.30	1.46 + 0.88
24	0.90 + 0.28	2.32 + 0.72	0.27 + 0.31	0.695 + 0.798
22a	1.20 + 0.40	2.35 + 0.78	0.70 + 0.30	1.37 + 0.59
2a	1.50 + 0.40	2.91 + 0.78	0.60 + 0.30	1.16 + 0.58
5a	1.92 + 0.33	5.08 + 0.87	-0.04 + 0.26	-0.11 + 0.72
8				
11a	2.64 + 0.40	66.5 + 10.07	0.94 + 0.34	23.7 + 8.57
14a	536.0 + 4.0	1980.0 + 14.78	166.5 + 2.0	616.0 + 7.40
ST. Geo. Cryo-Bag ^a	48.5 + 0.09	255.0 + 0.47	7.9 + 0.46	41.5 + 2.42

^aMethane standard was 40% higher than on previous run (data are not normalized).

TABLE XI
CONCENTRATION OF ^{85}Kr AND SF_6 IN HIGHWAY BAG SAMPLES (SRP)

Sample No.	Position I-95	^{85}Kr (dpm/cm ³) (pCi/SCM)		SF_6 (ppt by vol.)
30	~76	0.60	172	0.50
18	~73	0.99	212	0.50
25	~70	0.24	26	0.61
31	~67	0.07	23	0.58
6	~64	0.21	18 ^d	0.57
59	61	--d	--d	0.61
63	58	0.23	21	0.56
2a	56	0.23	21	1.20
57	54	--d	--d	1.58
65	52	0.26	32	1.94
40 ^a	50	0.45	36	2.81
60	48	0.47	57	4.04
48	46	1.00	100	5.57
4 ^a	44	0.66	79	3.17
8 ^a	42	0.87	100	3.06
52	40	0.32	58	1.88
46 ^{a,b}	38	0.06	17	0.58
51 ^a	36	0.52	66	1.24
44	34	0.46	68	0.82
10	32	1.35	150	0.77
5 ^a	30	2.03	205	0.60
64	28	2.25	213	0.63
3	26	1.03	171	0.57
55	24	0.66	86	1.12
49	~22	0.35	43	1.02
53	SC-336: 2	0.16	23	0.67
35	5	0.10	17	0.54
47	8	--d	--d	0.59
61 ^a	11	5.00	334 ^c	12.3 ^c
56	14	0.20	49 ^e	0.63
39	I-95: 40	--e	--e	1.64 ^f
54	34	--e	--e	0.55 ^f

^aTransfer cylinder (for Kr) leaked.

^bSampler malfunctioned-cleaned pump, restarted, bag contained ~1 h of sample when stopped.

^cBad data, contaminated cylinder.

^dKr lost in counting.

^eSample too small to process for Kr.

^f"Post-plume" SF_6 Samples.

TABLE XII

STATISTICAL PARAMETERS FOR THE CROSS-WIND SURFACE CONCENTRATION AT 100 km
(SRP)

A. Gross Statistical Parameters

<u>Tracer</u>	<u>$\bar{x}^{(a)}$</u>	<u>σ(km)</u>	<u>Kurtosis</u>
Me-20	44.2	12.7	3.7
Me-21	47.3	11.6	5.5
SF ₆	45.0	11.6	6.0

B. Parameters from Fitted Gaussian + Constant^b

<u>Tracer</u>	<u>$\bar{x}^{(a)}$</u>	<u>σ (km)</u>	<u>Peak (m³/m³ Air)</u>	<u>B (m³/m³ Air)</u>
Me-20	46.0 ± 0.5	5.5 ± 0.9	(144 ± 32) × 10 ⁻¹⁶	(6.9 ± 3.5) × 10 ⁻¹⁶
Me-21	46.8 ± 0.4	6.6 ± 0.7	(40.9 ± 6.0) × 10 ⁻¹⁶	(1.3 ± 1.0) × 10 ⁻¹⁶
SF ₆	45.9 ± 0.4	7.4 ± 0.6	(3.4 ± 0.4) × 10 ⁻¹²	(0.64 ± 0.04) × 10 ⁻¹²

^a $\bar{x}$ = Peak position in terms of I-95 mile markers.

$${}^b C(x) = \left(\frac{A}{\sqrt{2\pi}\sigma} \right) \exp \left(- \frac{(x-x_0)^2}{2\sigma^2} \right) + B .$$

TABLE XIII
TRACER-RELEASE DATA
(INEL, 1977)

Tracer	mol wt	bp, °C	<u>Weight Released</u>		<u>Release Volumes</u>	
			kg	g mol	Relative to SF ₆	Reciprocal
SF ₆	146	-63.8(Sub)	274 ± 2	1877	1.000	1.000
C ₅ F ₁₂	300	44.4	255	850	0.454	2.203
C ₈ F ₁₆ (PDCH)	400 ^a	102	237	592	0.316	3.166
C ₇ F ₁₄ (PMCH)	350 ^a	76.3	22.6	64.6	0.034	29.03
¹² CD ₄ (Total)	20	~-180	0.2616	13.8	6.96 x 10 ⁻³	143.6
(Alt. Rel.)			1.2559	12.80	6.18 x 10 ⁻³	146.8
(With SF ₆)			0.0057	0.28	1.51 x 10 ⁻⁴	6625.4
¹³ CD ₄ (Total)	21	~-180	0.0782	3.72	1.98 x 10 ⁻³	504.0
(Alt. Rel.)			0.0029	0.14	2.34 x 10 ⁻⁵	4.27 x 10 ⁺⁴
(With SF ₆)			0.0753	3.59	1.91 x 10 ⁻³	523.0

^aThe PCDH was 9.83 ± 0.10 % (mole fraction) PMCH. The reported release amounts reflect this.

TABLE XIV
ANALYSIS OF TRACER-RELEASE MANIFOLD SAMPLES*
(INEL, 1977)

Bottle	Concentrations (vol %) by TC ^a				by FID ^b
	SF ₆	N ₂	¹³ CD ₄	PDCB	¹³ CD ₄
1	66.7 ± 0.1	1.55 ± 0.03	0.11 ± 0.02	31.6 ± 0.1	--- ^c
2	68.1 ± 0.5	1.53 ± 0.27	---	30.3 ± 0.3	0.155 ± 0.004
3	68.8 ± 0.3	1.01 ± 0.03	0.11 ± 0.05	30.1 ± 0.3	0.150 ± 0.002
Average (vol %)	67.9 ± 0.4			30.7 ± 0.3	0.152 ± 0.004
Average (wt %)	51.8 ± 0.3			48.1 ± 0.5	0.0166 ± 0.0004
Calc. wt. Released ^d	604 lb.			561 lb. ± 9	88.0 g. ± 2.3
Meas. wt. Released	604 lb.			563 lb.	81.0 g.

* Data Supplied by Russell Dietz, BNL.

^a Thermal conductivity gas chromatography (gc).

^b Flame ionization gc.

^c Bottle sample inadvertently pumped away.

^d Calculated from gc determinations assuming 604 lb SF₆.

TABLE XV
GROUND-LEVEL SAMPLING DATA FOR ARC A (3.2 km)
(INEL, 1977)

<u>Position^a</u>	<u>Barrel Number^b</u>	<u>Time (MST)</u>		<u>Remarks</u>
		<u>Start</u>	<u>Stop</u>	
102	36	1213	1840	
103	98	1214	1841	Rate Low
104	106	1215	1842	
105	62 (82?)	1216	1843	
106	06	1217	1844	Rate Low
107	39	1218	1845	
108	79	1219	1846	
109	11	1221	1847	
110	91	1222	1848	Off at 1315 with little air - started again
111	---	1224	1850	Overfilled
112	17	1225	1851	
113	87	1226	1852	
114	77	1227	1853	
115	70	1228	1854	
116	73	1245	1855	Wouldn't Start
117	30	1231	1856	
118	71	1232	1857	
119	27	1234	1858	
120	06	1237	1900	Hose found twisted at 1340
121	98	1239	1903	Overfilled

^aSamplers were placed at 4° intervals on an arc that was 3.2 km from the tracer-release point. Position 101 was north of the release site and increasing position numbers indicate positions to the east (clockwise) from the north.

^bSamplers consisted of 50-ℓ Saran bags in plastic barrels. The bags were inflated with a small battery-powered pump.

TABLE XVI
GROUND-LEVEL SAMPLING DATA FOR ARC C (47 km)
(INEL, 1977)

<u>Position^a</u>	<u>Barrel Number^b</u>	<u>Time (MST)</u>		<u>Remarks</u>
		<u>Start</u>	<u>Stop</u>	
305	57	1350	1949	
307	104	1346	1945	
308	22	1343	1942	
309	72	1340	1940	
310	352	1338	1937	
311	--	1335	1935	
312	320	1333	1932	
313	814	1330	1930	
314	348	1327	1928	
315	306	1324	1926	
316	302	1322	1924	
317	324	1316	1845	
318	340	1314	1920	
319	348	1305	1900	
320	350	1303	1914	
321-1	324	1300	1940	Sequential sampler at this location
321-2	102	1300	1940	
322	328	1305	1936	
323	305	1306	1931	
324	347	1314	1946	
325	304	1317	1900	
326	316	1323	1903	
327	338	1301	1906	
328	327	1306	1913	
329	337	1310	1918	
330	342	1314	1923	
331	331	1320	1812	
332	68	1324	1758	
333	19	1327	1045	Off next morning
335	32	1330	1938	
337	101	1335	1922	
339	85	1339	1918	
341	3	1342	1914	

^aSamplers were placed at intervals of 2° per position on an arc that was 47 km from the tracer-release point. Position 301 was north of the release point and increasing position numbers indicate positions to the east (clockwise) from the north.

^bSamplers consisted of 50-ℓ Saran bags in plastic barrels. The bags were inflated with a small battery-powered pump.

TABLE XVII
GROUND-LEVEL SAMPLING DATA FOR ARC D (90 km)
(INEL, 1977)

Position ^a	Barrel Number ^b	Time (MST)		Remarks
		Start	Stop	
409	023	1445	2107	
410	340	1453	2122	
411	349	1500	2132	
412	325	1507	2142	
413	351	1513	2152	
414	307	1512	2105	
415	315	1505	2111	
416	321	1458	2130	
417	345	1446	2137	
418	336	1440	2145	
419	339	1439	2110	
420	329	1448	2116	
421	344	1454	2123	
422	313	1501	2129	
Dubois Airport	317	1542	2110	
423	300	1506	2134	
424	318	1512	2139	
425	312	1519	2147	
426	--	1530	0825	Turned off next day
427	10	1535	0830	Turned off next day
428	334	1539	0839	Turned off next day
429	353	1543	0844	
430	335	1546	0848	Bag ruptured
431	333	1552	0916	
432	330	1555	0921	
433	332	1601	0930	Failed to operate
435	311	1604	0936	
437	308	1610	0719	Bag ruptured
439	310	1615	0711	
441	309	1623	0705	

^aThe samplers were placed at intervals of 2° per position on an arc that was ~90 km from the tracer-release point. Position 401 was north of the tracer-release point and increasing position numbers indicate positions to the east (clockwise) from the north.

^bThe samplers consisted of 50- μ l Saran bags in plastic barrels. The bags were inflated with a small battery-powered pump.

TABLE XVIII
 SEQUENTIAL SAMPLES COLLECTED AT THE DUBOIS AIRPORT^a
 (INEL, 1977)

<u>Sample Number^b</u>	<u>Time MST</u>	
	<u>Start</u>	<u>Stop</u>
R-0	1430	1455
R-1	1500	1525
R-2	1530	1555
R-3	1600	1625
R-4	1630	1655
R-5	1700	1725
R-6	1730	1755
R-7	1800	1825
R-8	1830	1855
R-9	1900	1925
R-10	1930	1955
R-11	2000	2025
R-12	2030	2055
R-13	2100	2125
R-14	2130	2155
R-15	2200	2225

^aNear Sample Position 422.

^bSamples were collected in 50-ℓ Saran bags at a rate of 1.5 ℓ/min for 25 min each.

TABLE XIX
SAMPLING TANK DATA
(INEL, 1977)

Tank No.	Location	Time On 4/19/77	Time Off 4/20/77	Tank Vacuum(In Hg)		Filled(%)	Vol. Sampled (L)	Spiked Cylinders	BNL Cylinders
				Initial	Final				
1	Bozeman	1750	0553	25.5	6.5	74	144	X 773	
2	Sheridan	2000	0801	24.75	7.0	72	212	X 3567	W 28
3	Lodge Grass	1952	0822	25.8	2.0	92	271	X 3458	
4	Billings	1945	0745	26.1	5.2	80	236	X 684	W 27
249	Riverton	2000	0800					X 1228	W 26
and									
W-313406									
W-313358	Dubois, Airport	1430	2110				31.25	W 4757	W 24
11369 P100n	D-Arc 420	1448	2116				31.10	W 4913	W 25

TABLE XX
AIR-SAMPLE PROCESSING DATA
(INEL, 1977)

Position	Air Sample			Methane		Krypton					
	Cyl #	Wt (g)	Vol (L)	Spike (cm ³)	Recovered (cm ³)	Spike (cm ³)	Natural (cm ³)	Recovered (cm ³)	Measured (dpm)	Std. deviation (%)	Sample activity (dpm/cm ³)
1112	W4929	34.833	29.39	0.63	0.6	9.14	0.033	8.2	1.446	2.0	405.4
318	X3548	37.915	32.00	0.53	0.5	8.88	0.36	7.2	0.492	3.4	123.2
319	X3505	23.910	20.18	0.56	0.3	9.06	0.023	8.1	0.374	4.0	151.3
321-2	W4950	37.671	31.79	0.59	0.6	10.00 ^d	0.035	9.0	0.532	2.8	150.8
323	W5542	43.048	36.33	0.56	0.7	9.56 ^d	0.040	8.6	0.735	2.4	178.9
325	W4947	33.383	28.17	0.58	0.5	9.33 ^d	0.031	8.4	0.594	3.0	177.5
327	X0773	27.275	23.02	0.57	0.6	9.15	0.026	7.0	0.570	5.1	204.3
328	W4771	32.907	27.77	0.52	0.7	10.80 ^d	0.031	9.8	0.508	4.7	178.0
330	W4781	45.121	38.08	0.60	0.6	9.78 ^d	0.042	8.8	0.631	2.7	146.3
409	W4782	27.374	23.10	0.52	0.7	10.30 ^d	0.025	9.3	0.078	10.7	31.5
410	W5538	21.016	17.74	0.52	0.6	9.33 ^d	0.020	8.4	0.111	9.2	52.8
411	W5539	17.812	15.03	0.53	0.6	9.78	0.017	8.8	0.077	12.0	45.4
412											
413	W4780	15.908	13.42	0.53	0.5	8.77	0.015	6.2	0.120	28.6	70.6
414	X3549	16.116	13.60	0.52	0.5	9.05	0.015	7.8	0.085	11.6	50.9
415	W4954	10.842	9.15	0.54	0.6	10.20 ^d	0.010	9.2	0.104	12.3	104.3
416	W4916	10.174	8.59	0.59	0.7	10.10 ^d	0.010	9.1	0.063	15.0	66.7
417	W4918	10.405	8.78	0.52	0.6	10.20 ^d	0.010	9.2	0.104	10.0	109.2
418	W4964	9.081	7.66	0.59	0.6	10.00 ^d	0.009	9.0	0.093	9.7	109.1
419	X1655										
420	X3547										
420a	W4913	36.851	31.10	0.72	0.7	9.07	0.035	7.9	0.379	11.7	99.8
421	W4779	12.731	10.74	0.50	0.6	9.89 ^d	0.012	8.9	0.076	13.0	62.9
422	X3517	22.685	19.14	0.51	0.4	9.08	0.021	6.8	0.209	15.9	89.3
422b	W4943	13.050	11.01	0.50	0.5	10.50 ^d	0.012	9.5	0.094	9.9	80.6
422a,b	W4757	37.035	31.25	0.59	0.7	9.21	0.035	8.0	0.206	6.8	54.8
423	W5537	23.963	20.22	0.54	0.7	10.40 ^d	0.023	9.4	0.161	6.2	74.3
424	X3531										
425	X3622	41.989	35.43	0.53	0.6	9.01	0.039	7.50	0.380	5.9	87.3
426	W4784	11.134	9.40	0.63	0.7	9.44 ^d	0.011	8.50	0.088	11.8	80.0
427	W4783	40.506	34.18	0.73	1.0	10.40 ^d	0.038	9.40	0.209	5.2	57.3
428	W4959	44.883	37.88	0.47	0.6	8.32	0.042	6.60	0.262	6.4	52.0
429	W4958	31.963	26.97	0.55	0.7	8.88	0.030	9.20	0.168	6.7	49.7
430	W4942	10.301	8.69	0.66	0.9	8.95	0.010	9.30	0.051	19.0	47.2
431	W4785	35.634	30.07	0.57	0.7	9.25	0.034	8.80	0.209	5.9	58.1
432	W5536	34.178	28.84	0.84	1.0	8.99	0.032	9.10	0.197	6.3	55.4
433											
434											
435	X3492										
439	W4924	45.601	38.48	0.74	0.9	8.42	0.043	8.80	0.176	6.9	34.7
DR-1 ^c	W4754	38.401	32.41	0.68	0.9	9.20	0.036	8.80	0.142	7.5	36.2
DR-4 ^c	W4927	35.197	29.70	0.59	0.8	8.83	0.033	8.80	2.240	1.5	600.6

^a110-L evacuated cyl inder.

^bLocated at Dubois Airport (approximately position 422).

^cDry run (Prerelease) samples.

^dError in spike volume measurement, value estimated assuming a 90% recovery of Kr.

TABLE XXI
OBSERVED WINDS DURING TRACER RELEASE
(15.2-M TOWER AT TRA SITE)
(INEL)

<u>Hour (MST)</u>	<u>Average Speed (M/s)</u>	<u>Average Direction (deg)</u>	<u>σ_{A60} (deg)</u>
13-14	5.5	210	28.6
14-15	5.5	211	23.2
15-16	5.0	200	19.4
3-h mean	5.3	207	24

TABLE XXII
ESTIMATED MIXING DEPTH, 19 APRIL
(INEL, 1977)

<u>Time^a (MST)</u>	<u>Mixing Depth^b (Meters)</u>
0659	122
0930	823
1046	1555
1300	2165
1342	2075
1445	2378
1553	1982
1710	2957
1913	2896
2000	2774

^aRadar observed balloon release time.

^bBased on base of first elevated inversion.

TABLE XXIII
OBSERVED AND PREDICTED PLUME GROWTH PARAMETERS
FOR 19 APRIL

<u>Arc</u>	<u>Distance (km)</u>	<u>σ_y (km)</u>		
		<u>Gaussian Fit</u>	<u>Gross Statistics</u>	<u>PG-A</u>
A	3.2	0.78	0.71	0.64
C	47.0	10.4	7.0	6.4
D	88.0	12.3	12.8	10.0

TABLE XXIV
OBSERVED AND PREDICTED SF₆ CENTERLINE CONCENTRATIONS
(INEL, 1977)

<u>Arc</u>	<u>Sampler</u>	<u>Centerline Concentration</u>			
		<u>SF₆ (ppt)</u>		<u>Me-21 x 10¹⁴ (cm³/cm³)</u>	
		<u>Measured</u>	<u>Calculated</u>	<u>Measured</u>	<u>Estimated</u>
A	110	100	78	22.6 ^a	15.0
C	322	11.5	9.9	1.6	2.0
D	417	5.4	5.8	9.4	1.1

^aEstimated from adjacent sample.

TABLE XXV
DATA FOR ME-20, ME-21, SF₆, ⁸⁵Kr
(INEL, 1977)

A <u>D-Arc</u>					<u>Remarks</u>
<u>Position</u>	<u>Me-20^a</u>	<u>Me-21^a</u>	<u>SF₆^{b,c}</u>	<u>Kr-85^a</u>	
409	0.876	1.547	0.34 ^f	31.5	
410	3.008	1.430	0.427 ^c	52.8	
411	5.914	2.440	0.680 ^c	45.4 ^f	
412	3.3 ^f	2.4 ^f	3.82 ^d	55.0 ^f	
413	2.03	2.38	3.63 ^d	70.6	
414	1.92	14.37	3.5 ^f	50.9	
415	34.0	2.489	3.43 ^d	104.3	
416	1.197	10.628	4.01 ^d	66.7	
417	14.561	9.322	5.48 ^e	109.2	6.08 ^c , 4.88 ^d
418	7.35	9.37	4.97 ^d	109.1	
419	5.7 ^f	6.8 ^f	4.0 ^f	104.0 ^f	
420	4.4 ^f	4.8 ^f	3.24 ^d		
420E				99.8	Evac cyl
421	3.28	3.42	2.63 ^c	62.9	
422	4.53	23.98	4.0 ^f	89.3	
422A	0.1979	0.2167	4.09 ^c	80.6	Dubois bag
422C	0.0518	0.0117		36.18	Dubois cryo 1
422C	0.0612	0.0121		36.18	Dubois cryo 2
422E				54.8	Dubois evac cyl
423	4.15	1.61	0.66 ^c	74.3	
424	2.30 ^f	3.00 ^f	1.00 ^f	80.0 ^f	
425	1.43	5.50	1.66 ^c	87.3	
426	2.54	26.52	0.50 ^f	80.0	
427	0.419	0.869	0.163 ^c	57.3	
428	1.778	14.39	0.475 ^c	52.0	
429	0.619	1.573	0.63 ^f	49.7	
430	0.932	1.840	0.77 ^c	47.2	
431	0.5019	0.3635	0.77 ^f	58.1	
432	0.709 ^f	1.171 ^f	0.77 ^f	55.4	
433	0.65 ^f	1.14 ^f		43.0 ^f	
434	0.60 ^f	1.13 ^f		42.0 ^f	
435	0.56 ^f	1.12 ^f		40.0 ^f	
436	0.52 ^f	1.11 ^f		39.0 ^f	
437	0.48 ^f	1.10 ^f		37.5 ^f	
438	0.48 ^f	1.10 ^f		37.5 ^f	
439	0.4186	1.0884		34.7	
440	0.40 ^f	1.08 ^f		34.0 ^f	

TABLE XXV (cont)

B C-Arc

<u>Position</u>	<u>Me-20</u>	<u>Me-21</u>	<u>SF₆</u>	<u>Kr-85</u>	<u>Remarks</u>
305			5.76 ^e		5.89 ^c , 5.63 ^d
307			0.73 ^e		0.87 ^c , 0.58 ^d
315			5.21 ^c		
316			5.98 ^c		
317			34.7 ^e		8.05 ^c , 61.4
318	2.54	29.04	43.0 ^c	123.2	
319	1.17	4.68	10.05 ^e	151.3	10.0 ^c , 10.1 ^d
320			10.3 ^c		
321-2	0.547	1.416	14.75 ^e	150.8	7.80 ^c , 21.7 ^d
322			11.3 ^c		
323	1.15	1.78	9.12 ^e	178.9	9.40 ^c , 8.84 ^d
324			10.3 ^e		10.5 ^c , 10.1 ^d
325	0.851	0.259	9.32 ^c	177.5	
326			6.37 ^c		
327	6.40	43.66	7.67 ^c	204.3	
328	4.19	0.952	9.70 ^e	178.0	9.70 ^c , 9.70 ^d
329			5.39 ^c		
330	8.82	1.85	3.12 ^c	146.3	

C A-Arc

112	13.86	17.03	73.7 ^e	405.4	77.1 ^b , 70.3 ^d
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D Dry Runs

DR-1	22.43	22.16		36.2	
DR-4	97.04	97.04		600.6	

^aMe-20 and Me-21 are cm³/cm³ times 10¹⁴, SF₆ in ppt, Kr-85 in DPM/cm³, 1.7 ppm assumed for the ambient methane concentration.

^bA background of 0.5 ppt has been deducted from the SF₆ data and concentrations are those in the samples for all tracers.

^cBag data.

^d1-l cylinder data.

^eAverage of bottle and bag data.

^fEstimated from the adjacent data.

TABLE XXVI
AIRCRAFT SAMPLING DATA
(INEL, 1977)

Date	Pass/Sample No.	Time (MST)	Altitude (Ft, MSL)	Airspeed (Knots)	Temp. (OAT)	Location (Approx.)	Remarks
4/15/77	/DR 1	1250	8000	170		West of Idaho Falls	Background Sample
	/Dr 2	1255	8000			West of Idaho Falls	Background Sample
	1	1329	5100			Vicinity of A-arc	SF ₆ Observed
	2	1331	5700			See Fig. 7	SF ₆ Observed
	3	1332	5700				SF ₆ Observed
	4	1337	6700				SF ₆ Observed
	5	1340	6700				SF ₆ Observed
	6	1345	7700				No SF ₆ Observed
	7	1348	7700				No SF ₆ Observed
	8 /DR 3	1352	6700				No SF ₆ Observed
4/19/77	9 /DR 4	1354	6700				SF ₆ Observed
	10 /DR 5	1358	6700				SF ₆ Observed
	1	1412	5500	145	5° C	B-Arc	No SF ₆ Observed
	2	1420	6500	145		B-Arc	No SF ₆ Observed
	3	1425	6000	145		B-Arc	No SF ₆ Observed
	4	1433	6000	140		B-Arc	SF ₆ Observed
	5	1437	6500	155		B-Arc	SF ₆ Observed
	6	1445	8000			B-Arc	No SF ₆ Observed
	7	1513	11000	140	-8	C-Arc	No SF ₆ Observed
	8	1519	10000	150	-6	C-Arc	No SF ₆ Observed
	9	1528	6000	145		C-Arc	
	10 / 1	1533	6000	145		C-Arc	
	11	1536	7000	145		C-Arc	
	12 / 2	1542	7000	155		C-Arc	Sample at Position 324
	13	1550	8000	150		C-Arc	SF ₆ Observed
	14 / 3	1557		155	+1	C-Arc	Sample at Position 321
	15	1610	10000	155		C-Arc	SF ₆ Observed
	16	1617	11000			C-Arc	SF ₆ Observed
	17	1625	11000	145		C-Arc	SF ₆ Observed
	18	1630	12000			C-Arc	No SF ₆ Observed
	19 / 4	1635	12000	145		C-Arc	No SF ₆ Observed

TABLE XXVI (cont)

Date	Pass/Sample No.	Time (MST)	Altitude (Ft, MSL)	Airspeed (Knots)	Temp. (OAT)	Location (Approx.)	Remarks
		1645	7000			C-Arc	SF ₆ Observed
	21	1705	11000			D-Arc	No SF ₆ Observed
	22	1708				D-Arc	
	23 / 5	1719		150		D-Arc	Sample at Position 419
	24	1722	10000			D-Arc	No SF ₆ Observed
	25 / 6	1734	10000	150		D-Arc	Sample at Position 419
	26	1742	9000			D-Arc	SF ₆ Observed 416
	27 / 7	1753	9000	150	-2	D-Arc	Sample at Position 416
	28	1756	8000			D-Arc	SF ₆ Observed 416-419
	29 / 8	1803	8000	150		D-Arc	Sample at Position 416
	30	1809	7000			D-Arc	SF ₆ Observed 414-421
	31 / 9	1817	7000	150		D-Arc	Sample at Position 422
	32	1826	6500			D-Arc	SF ₆ Observed 423
	33 / 10	1838	6500	150		D-Arc	Sample at Airport
	34 / 11	2145	10000	170	-10	South of Billings	Southbound
	34 / 12	2153				South of Billings	Southbound
	34 / 13	2201				South of Billings	Southbound
	35 / 14	2215	8500	172	-4	North of Cody	Northbound
	35 / 15	2224	8500	172	-4	North of Cody	Northbound
	36 / 16	2233	11000	165	-12	South of Billings	Southbound
	36 / 17	2241	11000	170	-12	South of Billings	Southbound
	37 / 18	2302	13000	140	-15	SW of Billings	NW bound
	38 / 19	2310		170	-16	West of Billings	NW bound
	38 / 20	2338		165	-19	Bozeman	Southbound

Fig. 1.
Savannah River experimental area.

Fig. 2.
Savannah River Plant and nearby area.

Fig. 3.
Aircraft sampling pass No. 1 (SRP, 1975).

Fig. 4.
Aircraft sampling passes at 42 and 92 km from tracer release (SRP, 1975).

Fig. 5.
Cryogenic sampler location, INEL, 1974.

Fig. 6.
Map of experimental area, INEL, 1977.

Fig. 7.
Close-in experimental area map, INEL, 1977.

Fig. 8.
Tracer-release rates, SRP, 1975

Fig. 9.
Diagram of $^{12}\text{CD}_4$ release system, SRP, 1975

Fig. 10.
Diagram of SF_6 and $^{13}\text{CD}_4$ release system, SRP, 1975

Fig. 11.
Response of the BNL 90-s SF_6
instrument when cycling at 4-s
intervals between a 100-ppt and
a 10-ppt SF_6 standard.

Fig. 12.
Wind speed and direction
from pibal and tower data
(SRP, 1975).

Fig. 13.
Time-height wind speed and
direction data (SRP, 1975).

Fig. 14.
Estimated mixing depth
derived from soundings
(SRP, 1975).

Fig. 15.
Measured distribution of tracers along I-95
(ground-level bag samplers, SRP, 1975).

Fig. 16.
Typical response of the BNL 180-s SF_6 instrument
during traverse of the tracer plume.

Fig. 17.
Distribution of ^{85}Kr along I-95.

Fig. 18.
Gaussian least squares fit to tracer data
from the December 1975, SRL experiment.

Fig. 19.
Normalized tracer concentrations
along I-95 (SRP, 1975).

Fig. 20.
Turbo-Beech aircraft used at INEL, 1977.

Fig. 21.
Schematic diagram of release apparatus
for SF_6 and ${}^{13}\text{CD}_4$ (INEL, 1977).

Fig. 22.
Typical response of the BNL 90-s SF_6 instrument
during traverse of the tracer plume (INEL, 1977).

Fig. 23.
Distribution of $^{12}\text{CD}_4$ and $^{13}\text{CD}_4$
on the A- and C-arcs (INEL, 1977).

Fig. 24.
Distribution of $^{13}\text{CD}_4$ on the D-arc
(INEL, 1977).

Fig. 25.
Distribution of $^{12}\text{CD}_4$ on D-arc
(INEL, 1977).

Fig. 26.
Distribution of ^{85}Kr on the A-,
C-, and D-arcs (INEL, 1977).

Fig. 27.
Distribution of SF_6 on the A-arc (INEL, 1977).

Fig. 28.
Distribution of SF_6 on the C-arc (INEL, 1977).

Fig. 29.
Distribution of SF_6 on the D-arc
(INEL, 1977).

Fig. 30.
Comparison $\text{SF}_6/^{13}\text{CD}_4$.

APPENDIX A

ACTIVE PARTICIPANTS IN SAVANNAH RIVER PLANT (SRP) ATMOSPHERIC TRACER EXPERIMENT IN DECEMBER 1975.

		<u>Assignment</u>
SRL/SRP	C. Kern	Experimental Direction
	M. Pendergast	Meteorology Evaluation
	P. Long	Meteorology Evaluation
	D. G. Coats	Experimental Engineering
	B. Hyde	Field Support
	A. Boni	Sample Preparation
	J. Schubert	Experiment Planning
	L. Tilley	
	D. Pepper	Meteorology Evaluation
	G. Ferber	Program Coordination
NOAA/ARL	W. Elliot	Program Coordination
	N. Maisel	Aircraft Coordinator
BNL	R. Dietz	Samplers and Instrumentation
	E. Cote	Aircraft Instrumentation
EG&G	P. Boynes	Aircraft Coordinator
	G. Ragland	Aircraft Pilot
	E. Schultz	Aircrew
	B. Vernhyden	Aircraft Support
	T. Heis	Aircraft Support
	G. Menkel	Aircraft Support
	B. Mazurkewiz	Aircraft Support
	J. Cates	Aircraft Support
	R. Bench	Sample Separations
	B. Weltmer	Sample Separations
Airco, Inc.	S. Barr	Program Coordination
	W. Sedlacek	Tracer Release
LASL	M. Fowler	Field Samplers
	J. Archuleta	Field Samplers
	W. Clements	Meteorology Evaluation
	P. Guthrie	Program Coordination

APPENDIX B

TURBULENCE PARAMETERS AS PREDICTORS OF PLUME DIFFUSION

The selection of lateral and vertical diffusion rates based on gross turbulence statistics (e.g., σ_A) has conventionally been based on values measured close to the ground as predictors even though the dispersing plume may occupy a very deep layer. The value of such simplifications depend on the systematic variation of the governing parameters. Micrometeorologists have studied lateral and vertical turbulent velocity fluctuations, their interrelationships and their dependence on height, wind speed, and stability. Cramer et al.¹⁶ describe a consistent ratio of $\sigma_E/\sigma_A = 0.3$ in the lowest 50 m above ground. Lumley and Panofsky¹⁷ show empirical results where the lateral turbulence component $\sigma_v = \bar{u} \sigma_A$ essentially independent of height up to 100 m.

Figure C-1 shows the vertical profiles of σ_A and σ_E up to 340 m from the instrumented WJBF-TV tower at SRP. Except for 2 h, from 1000 to 1200 EST, when the upper levels of the tower show very large σ_A values (probably associated with the subsynoptic perturbation described in Appendix A), the profiles demonstrate a systematic decrease in σ_A . The 10-m values vary from 14° in the morning up to 30° through the most unstable period of the day and decrease again in the evening. The vertical fluctuations σ_E are generally smaller than σ_A . The invariance of σ_v with height is tested in Fig. C-2, which shows σ_v essentially constant with altitude, at least to 340 m.

The relations between the boundary layer turbulence parameters, the height, and stability appear to be quite consistent during the tracer experiment of 10 December 1975. Hence, it is expected that the conditions are favorable for good performance of relatively simple diffusion predictors such as ground level σ_A .

Fig. B-1. Vertical profiles of turbulent standard deviations of lateral (σ_A) and vertical (σ_E) angular fluctuations. (Labeled times are eastern standard)

Fig. B-2. Standard deviation of cross-wind velocity fluctuations at several elevations normalized by the 10-m value

APPENDIX C

ACTIVE FIELD PARTICIPANTS IN IDAHO NATIONAL ENGINEERING LABORATORY (INEL) ATMOSPHERIC TRACER EXPERIMENT IN APRIL 1977.

		<u>Assignment</u>
Nuclear Laboratory (INEL):		
NOAA/ARL-FRO	R. Dickson	Control
	G. Start	Control
	R. Ackermann	Control
	L. Thorngren	Control Secretary
	D. Forsyth	Tracer Release
	N. Ricks	Radar/Aircraft
	L. Klingler	Radar/Aircraft
	G. White	Radar/Aircraft
	G. Palmer	Radar/Tetroon
	J. Cate	Radar/Tetroon
	J. Sagendorf	Helicopter
	V. Drake	Helicopter
	N. Jeffery	C-Arc Instrumentation
	T. Sorenson	D-Arc Samplers
	D. Harker	D-Arc Samplers
	J. Thorpe	D-Arc Samplers
	S. Rinie	D-Arc Samplers
	N. Shaw	A-Arc Samplers
NOAA/ARL	G. Ferber	Control/Radar-Aircraft
	W. Hoecker	Tetroon Launch
	J. K. Angell	Radar/Tetroon
	R. Draxler	C-Arc Instrumentation
NWS/SLC	T. Adler	GMD Soundings
NOAA/Las Vegas	D. Randerson	Control
	R. Page	GMD Soundings
	K. Dustin	Tetroon Launch
	G. Gilbert	Pibal
	D. Sudweeks	Pibal
	S. Heinrichs	Pibal

APPENDIX C (cont)

HASL(EML)	P. Krey	C-Arc Instrumentation	
	C. Gogolak	C-Arc Instrumentation	
	F. Wilson	C-Arc Instrumentation	
	R. Lagomarsino	Aircraft Instrumentation	
EAA(LAS)	V. Andrews	Aircraft Instrumentation	
	D. Rockwell	Aircraft Pilot	
BNL	R. Goodrich	Aircraft Instrumentation	
	E. Cote	Release Manifold Sampling	
Allied Chemical	J. Keller	D-Arc Cryo-sampler	
	W. Motes	Billings Cryo-sampler	
LASL	S. Barr	Control	
	W. Clements	Control/Tracer Release	
	W. Sedlacek	Tracer Release	
	M. Fowler	Tracer Release	
	S. K. Wilson	Montana Samplers	
	W. D. Auble	Montana Samplers	
	P. Guthals	Aircraft Sampler and Instrumentation	
	J. J. Breslin	D-Arc Samplers	
	J. Archuleta	D-Arc Samplers	
	Lovelock Group Volunteers	P. Simmonds	C-Arc Instrumentation
		E. Sedlacek	Wyoming Samplers
J. Loew		Bozeman Samplers	
D. Miller		Wyoming Samplers	

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