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Atmospheric Tracer Experiments for Regional Dispersion Studies

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ABSTRACT

Tracer experiments are being conducted to verify atmospheric transport and dispersion calculations at distances from tens to hundreds of km from pollutant sources. In one study, a 2½ year sampling program has been carried out at 13 sites located 30 to 140 km from a source of ^{85}Kr at the Savannah River Plant in South Carolina. Average weekly concentrations as well as twice-daily concentrations were obtained. Sampling data and meteorological data, including surface, tower, and rawinsonde observations are available on magnetic tape for model verification studies. Some verification results for the Air Resources Laboratories Atmospheric Transport and Dispersion Model (ARL-ATAD) are shown for averaging periods from one week to two years.

In addition to tracers of opportunity, such as ^{85}Kr, there is a need for special tracers that could be released on demand, at precisely controlled rates, and measured accurately out to several days travel time. We are developing a perfluorocarbon tracer system, using automated sequential samplers, with a demonstrated capability to measure tracer concentrations to a few parts per 10^{15} of air. This sensitivity should permit tracer measurements to a distance of 1000 km with a release rate of about 10 kg/hr.

Atmospheric transport and dispersion models are being used extensively to simulate the behavior of air pollutants and to estimate regional air concentrations. Increased concern over regional and international aspects of air pollution has created a need for reliable model calculations of concentrations as far as 1000 km from pollutant sources. Experimental verification of these calculations is essential to establish the credibility of the models and environmental assessments based on model simulations.

Attempts to verify models with actual measurements of pollutants are complicated by the presence of multiple sources and imprecise knowledge of emission rates. We therefore chose to take advantage of a relatively unique tracer, ^{85}Kr, routinely emitted from the chemical separations facilities at the Savannah River Plant in South Carolina. ^{85}Kr air concentrations were measured over a 2½ year period at 13 sites located 30 to 140 km from the plant. This experiment provides a large data set where measured air concentrations can be related to a single, well-defined source. Sampling data and meteorological data have been compiled on magnetic tapes which are available to air pollution modellers for model verification studies. A description of the sampling program is presented here with some model verification results to illustrate the use of the data.

The use of ^{85}Kr as a tracer is limited to a very few source locations and processing of ^{85}Kr samples is very costly. There is a need for special non-reactive, non-depositing tracers that could be released at precisely controlled rates and measured at very low concentrations. Sulfur hexafluoride, SF_6, is useful to distances on the order of 100 km but its relatively high and variable background concentration militates against its use over greater distances. Even at shorter distances, a tracer system is needed that would provide automatic sequential sampling and rapid, inexpensive sample analysis. A new atmospheric tracer system, using perfluorocarbons (C_7F_{14} and C_8F_{16}), has been developed to meet this need. The perfluorocarbon tracer system will employ real-time continuous monitors as well as automatic sequential samplers. We plan to demonstrate the capabilities of this system in field experiments this summer and fall.

2. ^{85}Kr Tracer Experiment

The Savannah River Plant (SRP) located near Aiken, South Carolina, routinely releases ^{85}Kr in non-buoyant plumes from two 62 m stacks. Since ^{85}Kr is an inert radioactive gas with a long half-life (10.76 years), it

provides an excellent tracer of opportunity for studying the effects of atmospheric transport and diffusion free of complications introduced by wet and dry deposition or chemical transformations. Cryogenic ^{85}Kr samplers were operated at 13 sites from 30 to 140 km surrounding the SRP as shown in Fig. 1.

^{85}Kr Emissions

Total ^{85}Kr emissions for each month during the experiment are given by Telegadas, et al. [1]. Although the source was only quasi-continuous and emission rates fluctuated, these fluctuations should not seriously affect long-term concentrations. For calculation of seasonal and annual concentrations, the ^{85}Kr release may be assumed to be continuous, at a uniform rate, during each month. Estimates of the actual hourly emission rates may also become available in the near future.

Sampling Data

Weekly average ^{85}Kr concentrations at the 13 sampling sites were measured from March 1975 through September 1977 except for four intensive periods of twice-daily sampling (day and night, with each sample of about 10-hour duration). The intensive periods, one in each season, were each about a month in duration.

Average monthly, seasonal, and annual concentrations were also calculated from the data at each sampling site. A background ^{85}Kr concentration of 14 picocuries/standard cubic meter (pCi/SCM) was subtracted to obtain excess concentrations attributable to SRP plumes. As an example of the concentration distribution over the sampling network, the average excess ^{85}Kr pattern for Spring 1976 is shown in Fig. 2.

Details of the SRP experiment, including all sampling data and information on data tape formats and tape availability are given by Telegadas, et al. [1].

Meteorological Data

Meteorological data for the ^{85}Kr experiment have been stored on magnetic tape for the region within and surrounding the Savannah River Plant. Hourly surface weather observations, twice-daily rawinsonde observations, and hourly meteorological tower observations are combined on a Savannah River Experiment-Meteorological Data Base (SRE-MDB) tape.

Surface weather stations located between 86°W and 77°W; 37°N and 30°N are included in the SRE-MDB. About 60 stations report in this area each

hour. The surface stations within about 200 km of the SRP source are shown in Fig. 1. Parameters included on the SRE-MDB tape are World Meteorological Organization (WMO) block station number, station longitude and latitude, station elevation above mean sea level, wind direction, wind speed, station pressure, dry bulb temperature, dew point depression, and the previous 6-hour precipitation amount. The Pasquill stability category also has been added to each surface observation.

Four rawinsonde stations within the subgrid regularly take observations every 12 hours: Waycross and Athens, Georgia; Greenville and Charleston, South Carolina. All reported winds are included and temperatures are linearly interpolated to a wind level when no temperature was reported.

Meteorological tower data collected at seven power plant sites in the area near the Savannah River Plant were provided by local utility companies. Measurements include hourly wind speed, direction and temperature at one to three levels. Tower heights range from 40 to 100 m.

Meteorological data from a television tower, about 21 km from the SRP source, are also included on the SRE-MDB tape. Temperature and wind data at three levels (10 m, 91 m and 243 m), averaged over 15-min periods, are tabulated at one-hour intervals. The off-site towers within 200 km of the SRP are shown in Fig. 1.

Seven on-site towers with wind sensors at 62 m (stack height) are located in pine forests within a 10 km radius of the SRP stacks. The 15-min average wind speed and direction were tabulated at one-hour intervals. Because individual towers usually did not have a continuous record, an average wind from all on-site towers was calculated each hour from the available data and included on the SRE-MDB tape. This average wind is assumed to be representative of wind conditions on the SRP site.

3. Verification of the ATAD Model

Data from the Savannah River experiment are being used in the U.S. for model verification [2-6]. Some recent results on verification of the Air Resources Laboratories' Atmospheric Transport and Dispersion Model (ATAD) will be shown here. The ATAD model [7] is oriented toward practical application for pollution studies. ATAD calculates trajectories of 5 days duration from any number of origins, starting every 6 hours during any selected period (e.g., a day, month, or season), moving either forward or backward in time. Each trajectory is calculated from observed upper-air winds at rawinsonde and pibal stations. Transport winds are averaged in a vertical

layer whose variable depth is determined by the model from temperature soundings along the trajectory.

The main output table in ATAD lists the latitude and longitude of trajectory positions at 6-hour intervals. Another option provides a plot of calculated trajectories on a computer page, using a Mercator map projection with a scale selected by the user (see Fig. 3). Four trajectories (one day) are plotted on each page.

A pollutant plume is represented by a series of puffs, one per hour, where each puff diffuses as it is transported along a trajectory path. Each puff is assumed to have a Gaussian horizontal distribution and to be vertically mixed through the transport layer. Air concentrations contributed by the puffs are averaged (over selected time periods of 6 hrs. or more) at each grid point. ATAD can provide a coded plot of surface air concentrations for selected averaging periods as shown in Fig. 4.

For one origin, one month of individual forward trajectories of 5 days duration with maps of monthly average surface air concentration and deposition amounts can be run on an IBM 360/195 in about 2 minutes. Each additional origin adds about 20 seconds to the run.

The first two years (March 1975 to February 1977) of weekly ^{85}Kr measurements were used in a verification study of the standard ATAD model compared to simplified versions using transport winds at a single level and a constant mixing layer. About 1200 ratios (R) of calculated (C) to observed (O) excess concentration (R = C/O) were compared over the two-year period. The ratio mean, $\bar{R}$, and the ratio root mean square error, RMSE, about the 1 to 1 correpondence line were determined as follows:

$$\bar{R} = \exp\left(\frac{1}{n}\Sigma \ln R\right)$$

$$\text{RMSE} = \exp\left(\left[\frac{1}{n}\Sigma(\ln R)^2\right]^{\frac{1}{2}}\right)$$

Figure 5 shows RMS errors for the weekly excess concentration ratios and for longer averaging periods out to 2 years. The solid curve is for the standard version of ATAD. The dashed curve, ATAD (950), is for calculations where 610 m transport winds (about 950 mb) and a constant mixing depth of 1000 m were substituted for the variable transport and mixing layer. The dotted curve, ATAD (850), is for 850 mb transport winds and 1000 m mixing depth. The ATAD (850) calculations gave significantly larger RMS errors at all averaging periods than either ATAD or ATAD (950). Since the climatological average transport layer depth over the sampling area is

about 1000 m, 850 mb winds (about 1500 m) are generally above the transport layer and do not adequately reflect pollutant movement. It should be noted that 850 mb winds are often the only transport winds used in many regional-scale models. Use of the 610 m winds in ATAD (950), winds closer to mid-transport level, shows significant improvement over ATAD (850). Thus, about 68% (percent of ratio values within the RMSE) of the weekly average concentrations calculated by ATAD were within a factor of 4 of the observed values. Similarly, about 68% of the calculated yearly averages were within a factor of 1.5 of the observed values, i.e., 9 of the 13 stations had a calculated yearly average concentration within 50% of the observed concentration.

The ratio mean, $\overline{R}$, is a measure of model bias. Values of $\overline{R}$ were .84 for ATAD and .78 for ATAD (950) indicating a tendency toward underprediction. $\overline{R}$ for ATAD (850) was .41, a substantially larger underprediction.

4. Perfluorocarbon Tracer System

Investigations by J.E. Lovelock, in England, suggested that a perfluorocarbon tracer system could be developed that would be ideal for atmospheric dispersion studies. The perfluorocarbons are extremely stable nontoxic compounds, measurable at very low concentrations by electron-capture gas chromatography. At present we are working with two perfluorocarbons, perfluoromethylcyclohexane (PMCH; C_7F_{14}) and perfluorodimethylcyclohexane (PDCH; C_8F_{16}). Comparative data on SF_6, PDCH and PMCH are shown in Table I. The atmospheric background concentration of PDCH is about 0.03 parts per trillion by volume (3×10^{-14}), about 1/20 of the SF_6 background. Background of PMCH is another order of magnitude lower. The amount of tracer released in any experiment must be sufficient to distinguish the plume from background at the maximum sampling distance. The required release rate for PDCH is about 10% of that for SF_6; for PMCH it is about 1% of the SF_6 rate. Taking the higher price of the perfluorocarbons into account, the PDCH required for an experiment would cost about the same as SF_6; the cost of PMCH would be less than 10% of the SF_6 cost.

The NOAA Air Resources Laboratories contracted with Lovelock to design and build prototypes of three different perfluorocarbon samplers. One instrument, an automatic sequential sampler, incorporates 24 sampling tubes containing molecular sieve material to trap the PDCH tracer. Air is pumped through each tube on a preset schedule. When sampling is completed, the unit is returned to the laboratory and inserted into an analyzer unit which

automatically heats each tube in turn, to desorb the sample into a catalytic reactor which destroys most unwanted components. The sample then flows through a chromatograph column which separates the perfluorocarbon tracer from other surviving compounds and then tracer concentrations are determined with an electron-capture detector.

Another prototype instrument, the "Two-Trap" sampler, combines the sampling and analysis functions into a single unit. The unit contains two sampling tubes which are automatically cycled so that one tube samples while the other is being analyzed. This instrument provides readout of tracer concentrations every five minutes at the sampling site.

The third instrument is a real-time continuous monitor intended primarily for use in an aircraft to obtain crosswind concentration profiles. Ambient air is drawn through a catalytic reactor that reduces the O_2 and other electron-absorbers, leaving the perfluorocarbons and nitrogen. This is passed directly to an electron-capture detector providing continuous concentration readout with a 3-second delay.

The prototype instruments were delivered by Lovelock in 1976. Since that time the NOAA Air Resources Laboratories, the Department of Energy, Environmental Measurements Laboratory, and the Brookhaven National Laboratory have been engaged in a cooperative effort to develop a practical perfluorocarbon tracer system.

Field Tests of Perfluorocarbon System

Perfluorocarbon tracer release, sampling, and analysis techniques were tested in a field experiment in April 1977 [8]. PDCH and PMCH were released simultaneously with SF_6 for three hours and whole-air samples were collected along three arcs out to 90 km from the release point.

Five Lovelock sequential samplers were deployed on the 50 km arc. These samplers performed well; about 60 half-hour samples were taken and all data were consistent as shown in Fig. 6. As the plume passed the arc, PDCH concentrations were measured from the background of about 0.03 ppt to a peak near 10 ppt and down to background again. These measurements agreed well with SF_6 concentrations in whole-air samples taken at the same locations.

The "Two-Trap" sampler, operated alongside one of the sequential samplers, provided in situ readout of 5-min concentrations. Results from these two samplers were in good agreement as shown in Fig 7.

Problems were encountered with the Lovelock continuous monitor which apparently was adversely affected by pressure changes and vibration in the aircraft. Since the concept appears to be sound, and our enthusiasm for a real-time monitoring capability remains undiminished, NOAA has contracted with Lovelock for further development work.

Improved Samplers

Efforts are in progress to improve the operation and sensitivity of all three sampling instruments.

Sequential sampler: The molecular sieve traps in the Lovelock sampler collect PDCH, but not PMCH. Investigations by R. Dietz at Brookhaven showed that charcoal-type adsorbents can be used to sample and recover both PDCH and PMCH with measurement capability down to 0.001 ppt as demonstrated by the chromatogram of a 40-liter background air sample shown in Fig. 8. Dietz developed a conceptual design for an improved sequential sampler dubbed the Brookhaven Atmospheric Tracer Sampler (BATS) and NOAA has contracted with Gilian Instrument Corp., Wayne, New Jersey, for production of 60 units to be delivered in May 1980. The new sampler has 23 charcoal traps which can be sequenced on command from an internal digital clock. The BATS contains an internal pump with selectable flow rates and digital printer for recording the time and air volumes sampled. Features include automatic start at a preselected time, selectable number of samples and duration of sampling (1 min to 1 week per trap), interchangeable lids for field replacement of a fresh set of 23 traps, and provision for thermal desorption of the samples. Internal batteries provide sufficient power for unattended operation for up to a month. The sampler measures approximately 35x25x17 cm, weighs about 9 kg and is currently priced under $4000.

Two-Trap Sampler: This sampler is also being converted to charcoal traps which is expected to increase its sensitivity to 0.1 ppt for PDCH and PMCH.

Real-time Continuous Monitor: A new prototype, designed to alleviate the problems encountered with the original prototype, was delivered in February. This instrument is presently undergoing laboratory tests and we plan to flight-test it this summer. It is expected to provide a continuous in-flight record of tracer concentrations down to 0.1 ppt or better.

Long-Range Tracer Experiment

An atmospheric tracer experiment is planned for July 1980 as a proof-test and demonstration of the perfluorocarbon tracer system. The two

perfluorocarbons (PMCH and PDCH) will be released in Oklahoma over a 3-hr period and the new sequential samplers will be operated on arcs about 100 km and 600 km from the release point. Sufficient SF_6 will be released simultaneously to allow us to compare tracer concentrations at the 100 km arc. About 20 samplers will be placed at 4 km intervals on the inner arc and 40 samplers at about 20 km intervals on the outer arc (see Fig. 9).

Deployment of samplers over the long distances involved in this type of experiment could be very costly and present difficult logistics problems. Therefore, we have secured the cooperation of the NOAA National Weather Service (NWS) to allow us to use their substation network as a fixed sampling array. This network is comprised of 13,000 locations in the U.S. where cooperative observers, mostly volunteers, gather weather data for NWS. We will deliver the samplers, in advance, to the cooperative observers at selected sites and notify them, by telephone, when sampling is to start. They need only set the specified time into the instrument and it will automatically run through its pre-set sampling routine.

The ATAD model was used to simulate a 3-hr release of 50 kg/hr of PMCH in this experiment. Tracer concentrations were calculated for successive 6-hour sampling periods beginning with the start of the release. Contours are shown in Fig. 9 for calculated PMCH concentrations corresponding to twice the ambient background (0.005 ppt) and 100 times background (0.2 ppt). In this simulation, the plume is discernible from background at 7 sampling sites on the 600 km arc. Other model calculations suggest that releases on the order of 10 kg/hr would be sufficient for plume measurements out to 1000 km.

Measurements of the different tracers will be compared to establish the reliability and precision of release, sampling and analysis techniques. The real-time continuous monitor will be flown to measure the vertical distribution of tracer and crosswind concentration profiles. Data will be used to estimate diffusion parameters and to test model calculations of plume transport and dispersion out to 600 km.

5. Summary

A 2½ year ^{85}Kr sampling program at 13 sites surrounding the Savannah River Plant provides extensive data for regional atmospheric transport and dispersion model verification. Weekly and twice-daily sampling data are available on magnetic tapes along with meteorological data covering the area of interest. Researchers desiring to obtain copies of the tapes

should consult the final project report [1] for a detailed description of the data and information on where tapes may be obtained.

When tested against the ^{85}Kr sampling data, the Air Resources Laboratories ATAD model shows an RMS error of a factor of 4 for weekly calculated-to-observed concentration ratios. The RMS error is reduced to 50% for calculations of mean annual concentrations.

The perfluorocarbon tracer system should soon provide a capability to perform long-range atmospheric transport and dispersion experiments, at reasonable cost, for verification and improvement of air pollution models. This system provides dual-tracer capability (simultaneous sampling of PMCH and PDCH), automated sequential sampling, and rapid, inexpensive, automatic analysis of large numbers of samples. The detection limit with the BATS sequential sampler is about .001 ppt. Detection limit of the real-time continuous monitor is expected to be better than 0.1 ppt. This new system should prove useful in many applications from mesoscale studies of atmospheric transport and dispersion over complex terrain, to continental-scale studies of atmospheric dispersion processes. Model calculations indicate that tracer concentrations would be measurable with the BATS sampler at a distance of 1000 km from a release of about 10 kg/hr of PMCH.

Acknowledgments

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The ^{85}Kr sampling program was a joint effort with the Savannah River Laboratory, E.I. DuPont and Company, Aiken, SC.

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Table 1. Comparative Data on SF_6 and Perfluorocarbons

Tracer	Sulfur-Hexa fluoride	Perfluoro-Dimethyl-cyclohexane (PDCH)	Perfluoro-Methyl-cyclohexane (PMCH)
Formula	SF_6	C_8F_{16}	C_7F_{14}
Mol. Wt.	146	400	350
Background (pptv)	0.6	0.03	0.0025
Cost/kg	$11	$90	$90
Relative Release Rate (by wt.)	100	14	1.0
Relative Cost/ Release	1.0	1.1	0.08

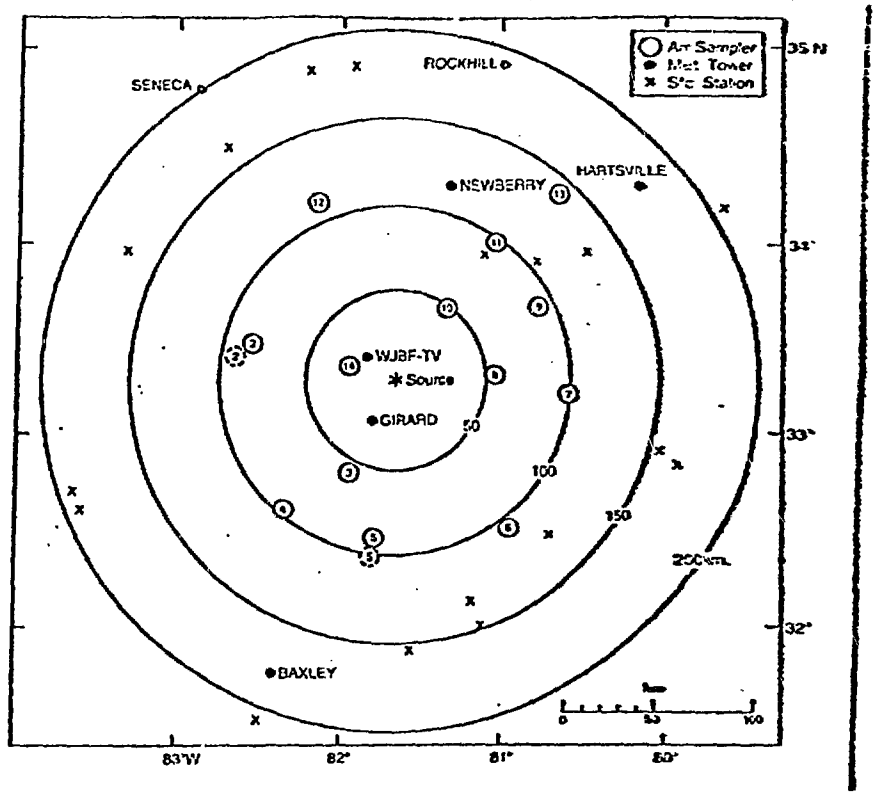

Figure 1. Air sampling stations, meteorological towers and surface weather stations within 200 km of the SRP.

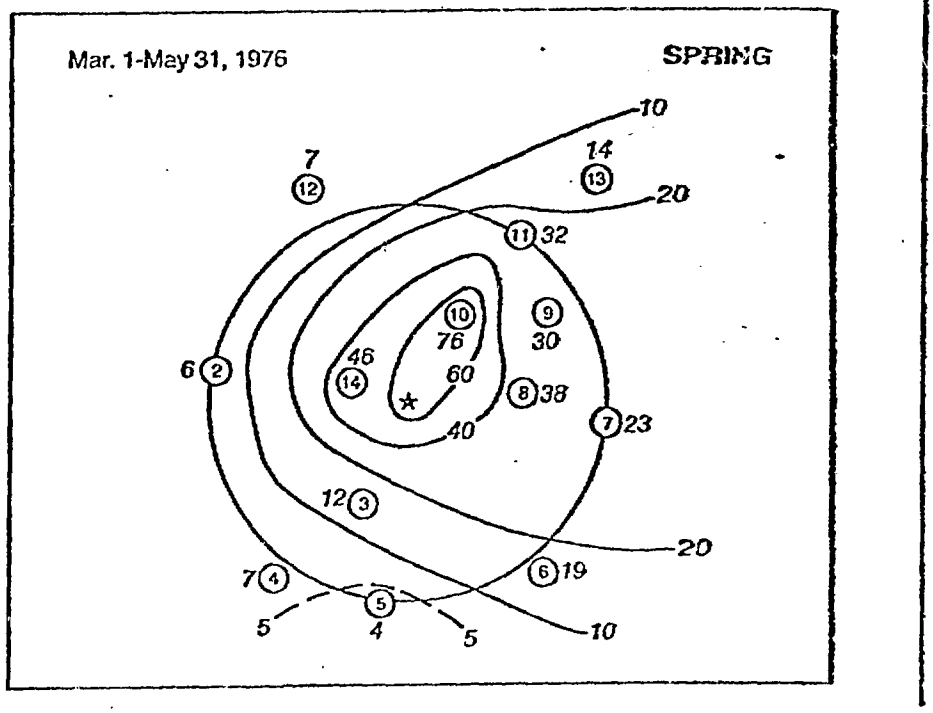

Figure 2. Observed excess ^{85}Kr concentrations (pCi/SCM) for Spring 1976. Star indicates source location and a 100-km radius circle about the source is shown.

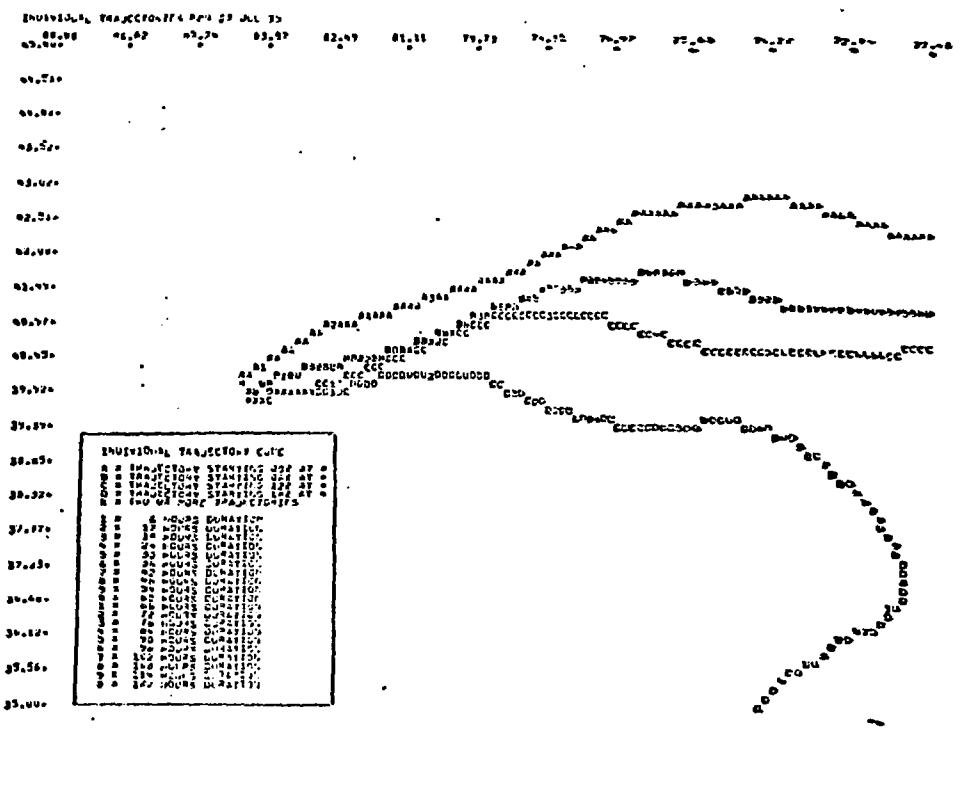

Figure 3. Optional ATAD output of individual trajectory plots.

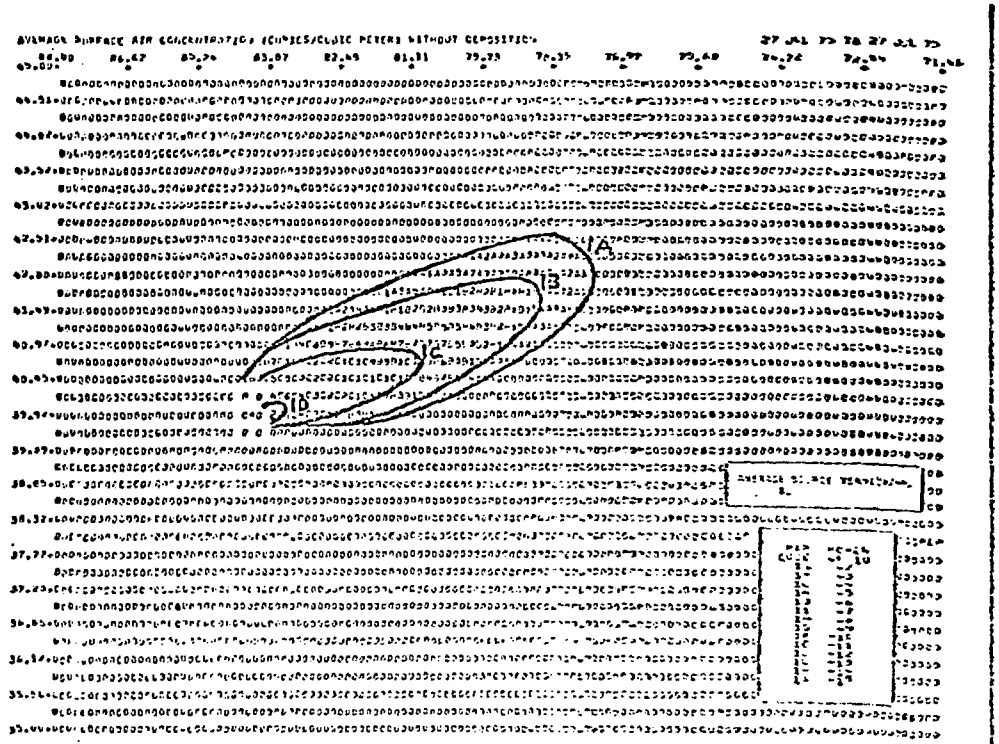

Figure 4. ATAD output of average surface air concentration coded values. The contours are hand-drawn.

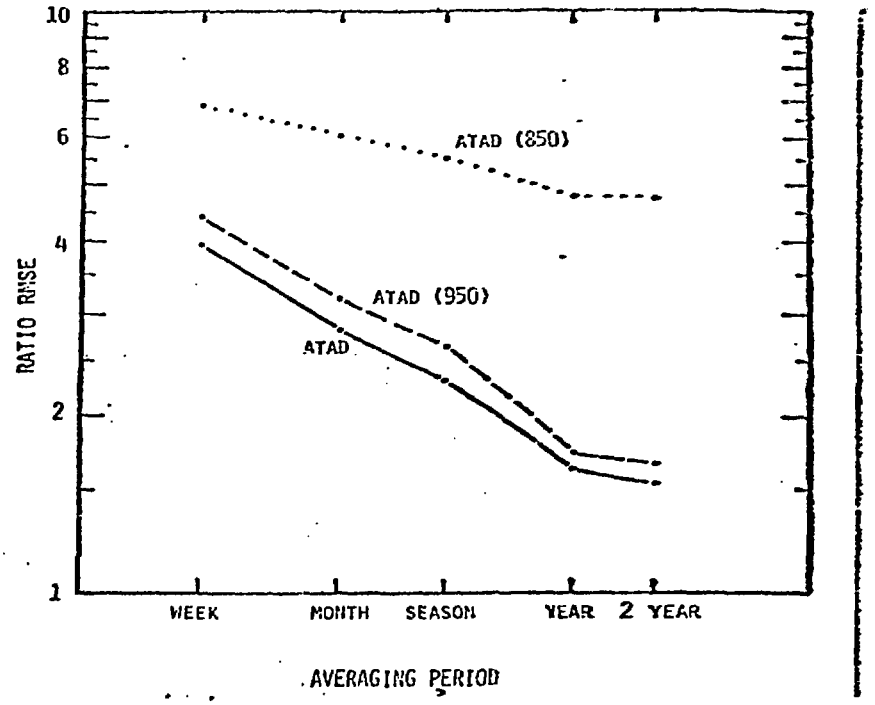

Figure 5. Root mean square error (RMSE) of the ratio of calculated ^{85}Kr concentration to the observed concentration for various averaging periods.

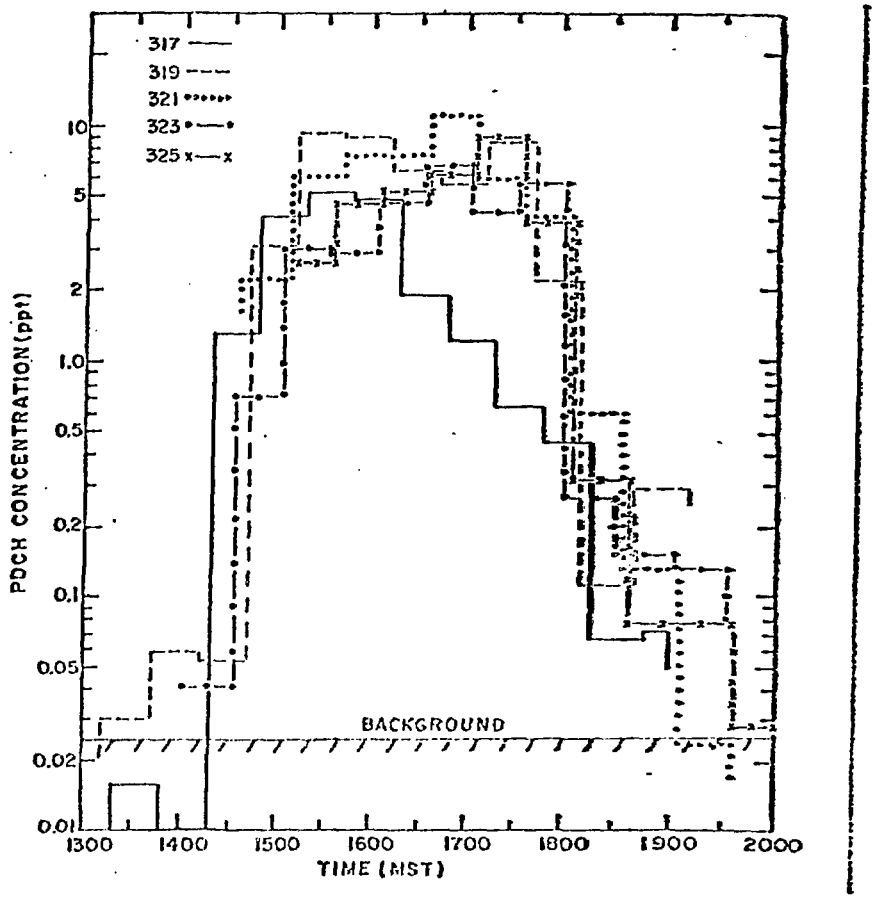

Figure 6. Perfluorocarbon (PDCH) concentrations measured in ½-hr sequential samples at five locations on an arc 50-km from the tracer release site.

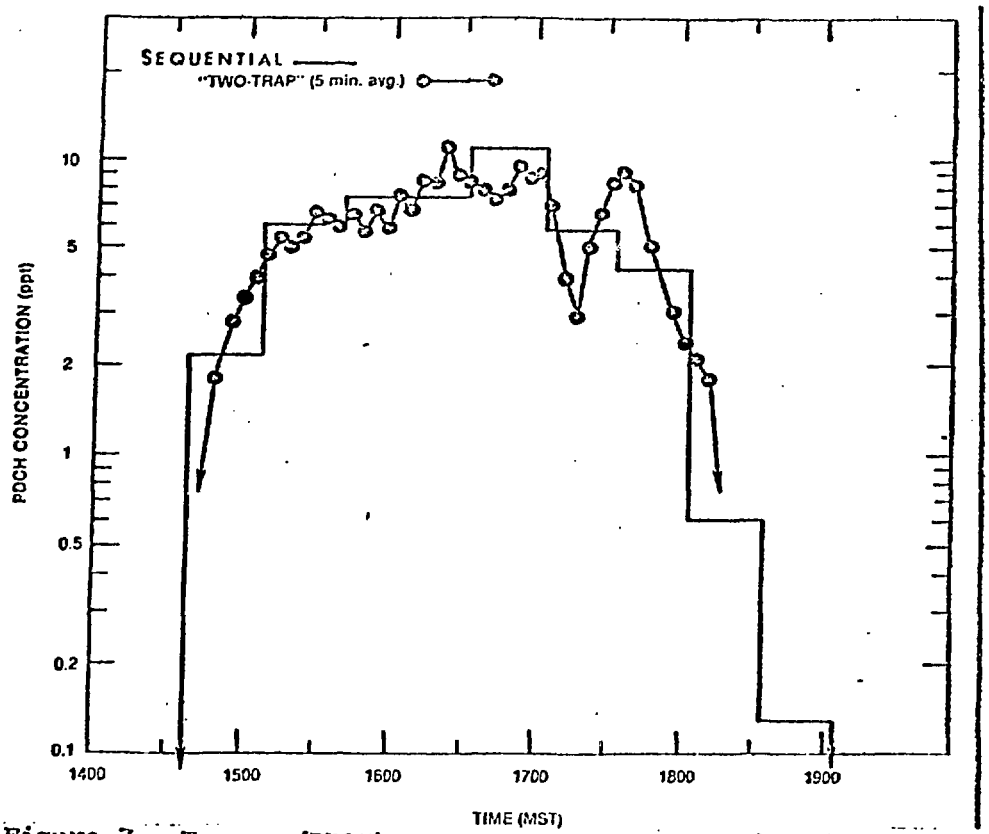

Figure 7. Tracer (PDCH) concentrations measured with a sequential sampler (½-hr intervals) and the "Two-Trap" sampler (5-min intervals).

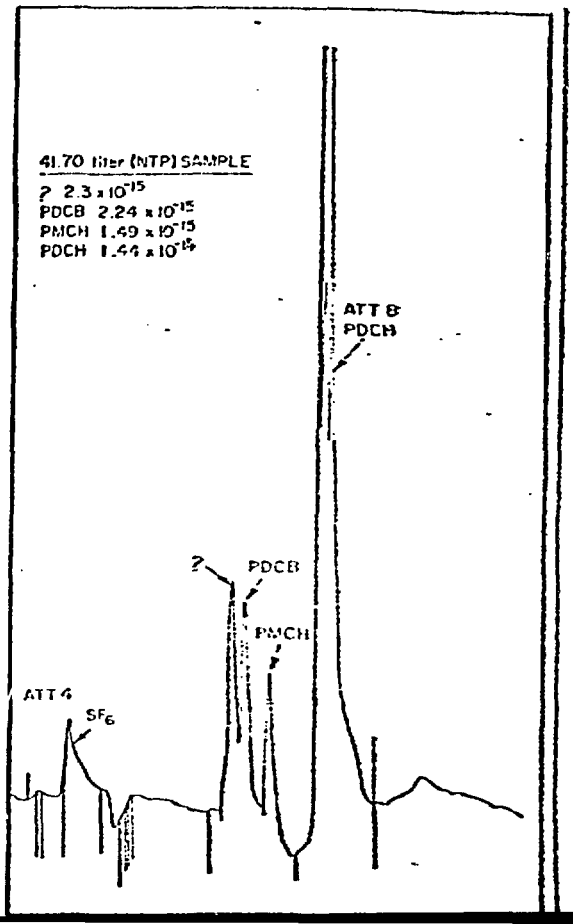

Figure 8. Chromatogram showing simultaneous measurement of background concentrations of PDCH and PMCH in ambient air.

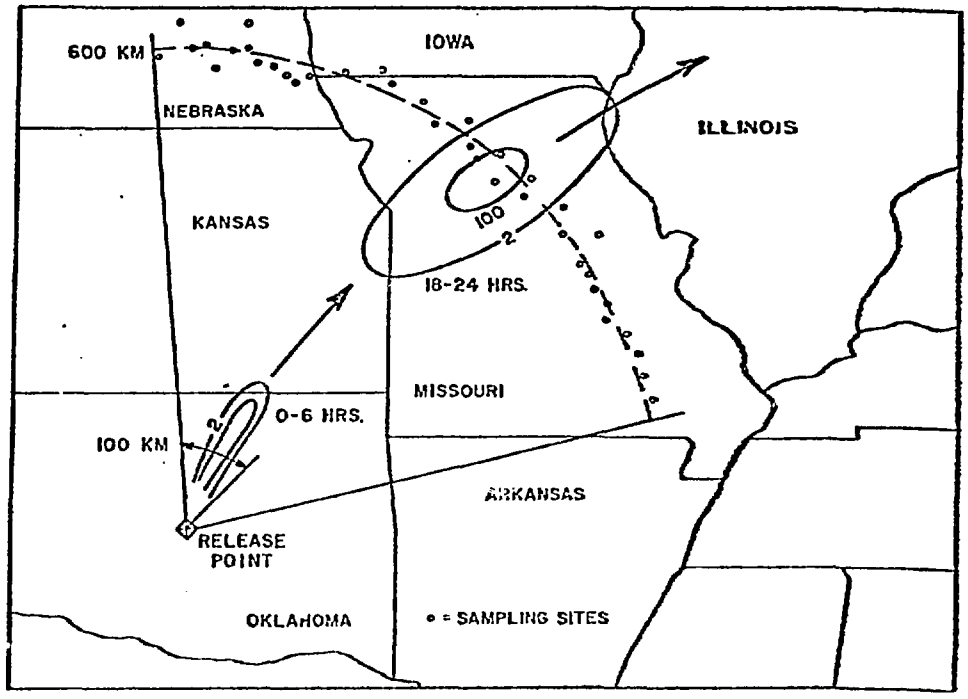

Figure 9. Experimental sampling array and model-simulated 6-hr sample concentrations after a 3-hr release of 50 kg/hr of PMCH. Contours are drawn at twice the background value and 100 times background.