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Illinois State Water Survey
at the
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# STUDY OF RAINOUT OF RADIOACTIVITY IN ILLINOIS

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Prepared by

Wayne E. Bradley
and
Pieter J. Feteris

Glenn E. Stout
Project Director

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## INTRODUCTION

During the summer of 1965 network water sampling for radioactivity was continued. In addition, surface air sampling plus airborne water and air sampling were carried out. The project was undertaken in an effort to evaluate changes in radioactivity concentrations in precipitation between cloud-base and the ground, to ascertain the relationship between air and rainwater concentrations, and to seek further information on the source of the initial high concentration of radioactivity frequently observed in the 1962-1964 studies.

An airborne system for sampling atmospheric water vapor has also been developed to collect water vapor for tritium analyses. The aircraft sampling system as well as the laboratory backup system are described, but most of the samples collected have not yet been analyzed for tritium.

## ACKNOWLEDGMENTS

The research on which this report is based was carried out under the general direction of G. E. Stout, Head, Atmospheric Sciences Section, Illinois State Water Survey. F. A. Huff assisted in project planning and in the review and assembly of this report. The radon analyses discussed in the report were made by Harold Danford. Various other staff members aided in the collection and analyses of data used in the research.

## CASE STUDIES

Simultaneous air and water samples were collected on eight different days of rainfall. The samples were taken with the network and instrumentation described by Huff and Bradley (1965). The radioactivity in the rainwater varied in much the same way in time and space as described by Huff (1965a) and will not

be further analyzed. The radioactivity of the air filter samples varied considerably from sample to sample but the overall average of the network usually remained nearly constant throughout the sampling period. Most of the variations, as will be described later, are probably the result of errors in counting the filters. On one day only did the concentration of the radioactivity in the air vary significantly and that was on May 26, 1965. This case along with the case of June 1, 1965, will be described in more detail. The remainder of the cases do not reveal any gross changes in the activity of the air that could be correlated with changes in the water concentration.

It is interesting to compare the amount of radioactivity per unit volume in the precipitation with that in the air. The activities in the air were in the order of a few tenths of a $pc/m^3$. The activities in the water were in the order of hundreds of pc/l or hundreds of thousands per $m^3$ of water. The ratio $\beta_w/\beta_a$ where $\beta_w$ is the gross beta activity in precipitation and $\beta_a$ is the activity in air was usually between $10^5$ and $10^6$. This ratio is an indication of how very effective the precipitation process is in scrubbing the atmosphere. The ratio was determined for 16 showers using the average air and precipitation concentrations obtained from all five network stations for the entire period of each shower. Only the extremely high air and water samples from May 26, 1965, were excluded. The frequency distribution of the ratio $\beta_w/\beta_a$ is given in table 1.

Table 1. Distribution of $\beta_w/\beta_a$ Ratios

| $10^5$ $\beta_w/\beta_a$ | 0-1.9 | 2.0-3.9 | 4.0-5.9 | 6.0-7.9 | 8.0-9.9 | 10.0-11.9 | 12.0-13.9 | 25 |
|---|---|---|---|---|---|---|---|---|
| Frequency | 2 | 3 | 4 | 3 | 0 | 1 | 2 | 1 |

Comparison of Surface and Airborne Precipitation Samples

On several days flights were made to collect airborne precipitation samples above the surface sampling network while samples were being taken below. The problem was to have the network attended, to be able to obtain a pilot, and to have sufficient rain over the network simultaneously. At the same time it was necessary to have sufficient visibility for surface reference and not become involved with too large a storm. All of these conditions were not met frequently. Only on two of these flights were samples taken in sufficient number and under satisfactorily controlled conditions for analysis.

Once the samples are collected and a comparison of the radioactivity concentrations of the airborne and surface samples is sought several more problems arise. These problems are related to determining the rate of fall of the raindrops, sampling the same parcel of rain aloft and at the ground, and the possible variation of concentration with drop size. These difficulties make it impossible to compare precisely the samples, but a good approximation may be obtained if a few assumptions are made.

If the radioactivity concentration varies with the drop size and the duration of a particular shower is short, the activity of the precipitation in the first portion of the surface rainfall will be biased toward the activity of the larger drops with the higher terminal velocity. The precipitation with the lower velocity, that is the smaller drops, will bias the activity toward the end of the shower. However, if the duration of the shower is long compared to the average terminal drop velocity, as is the case in these studies, then this factor will not affect the data.

Another problem inherent in comparing airborne and surface rain samples results from horizontal sorting of drops by the wind when different size drops are acted on by the wind for different lengths of time because of their different fall velocities. Fortunately, in cases discussed here, the surface winds were

light and the drops did not have the opportunity to drift far during their short time of fall.

Because the aircraft must circle while the surface sampler remains stationary, another problem occurs since the activity varies in the horizontal as well as with time. For the purpose of this study, however, it will be assumed that there are no significant horizontal gradients in activity within the turning radius of the aircraft (<one-half mile).

Another problem arises because different sizes of raindrops fall at different terminal velocities and a vertical sorting or mixing of drops occurs between the time the samples are taken aloft and the time they reach the ground. If the radioactivity is varying with time at the cloud base, the time rate of change of the activity at the surface will not be the same as that at cloud base. To compare the airborne with surface samples the drop-size distribution and the rainfall rate of each size of drop must be known to determine what percent of the surface rain is representative of each airborne sample.

In order to compare the airborne water sample radioactivities with surface samples it was assumed that the activity of all the different sizes of drops was the same at the cloud base sampling altitude and that there were no horizontal gradients in the activity. It was also assumed that the rainfall rate was constant for each aircraft sample and was associated with a unique drop-size distribution. Further, it was assumed that there was no coalescence of the raindrops during their fall.

## Storm of May 26, 1965

Before examining the relation between airborne and surface samples in detail, a general description of the overall situation will be given. There were three rains during the day with the second rain being the most intense. Figure 1 shows the gross beta concentration in both air and precipitation for a 7.5-hour

period. The activity in the second rain is illustrated both as smoothed curves and as an enlarged detailed insert.

The radioactivity concentration in the air throughout the morning averaged between 0.3 and 0.4 pc/m$^3$. In the early afternoon, after the rain, the average air concentration, excluding station 12, jumped to between 0.7 and 0.8 pc/m$^3$. Station 12 had a maximum of 15.0 pc/m$^3$ and then slowly decreased. The large variation from sample to sample is believed to be the result of errors in the activity counting.

The first rain did not show any systematic variation in the intensity from station to station. Over the network and throughout the entire rain the activity ranged from a minimum of 120 to a maximum of 900 pc/m$^3$. The second rain is the most interesting. During the first portion of the rain, enlarged in the insert of figure 1, it is seen that the activities at all the stations varied by about an order of magnitude from a maximum of 1000 to a minimum of 100 pc/m$^3$. At approximately 1300 CST, however, the concentrations at several stations increased considerably with a maximum concentration of about 1800 pc/m$^3$ at station no. 6. This relatively large increase in the activity of the precipitation was concurrent with the increase in surface air activity over the entire network, and was associated with the passage of a front. The third rain behind the front had higher activities than the earlier rain. Concentrations were between 700 and 1800 pc/l. Further discussion of the afternoon increase in activity will follow in a later section.

The following method was used to examine the aircraft data collected on May 26. The rainfall rate at aircraft altitude was taken as the average rate measured in the raingage beside the collector for the period of time of the aircraft sample, after correction for the time of fall of the mean drop size from the flight altitude. Then for each minute of surface sampling time the individual rainfall rate for each size drop, along with its terminal velocity and the

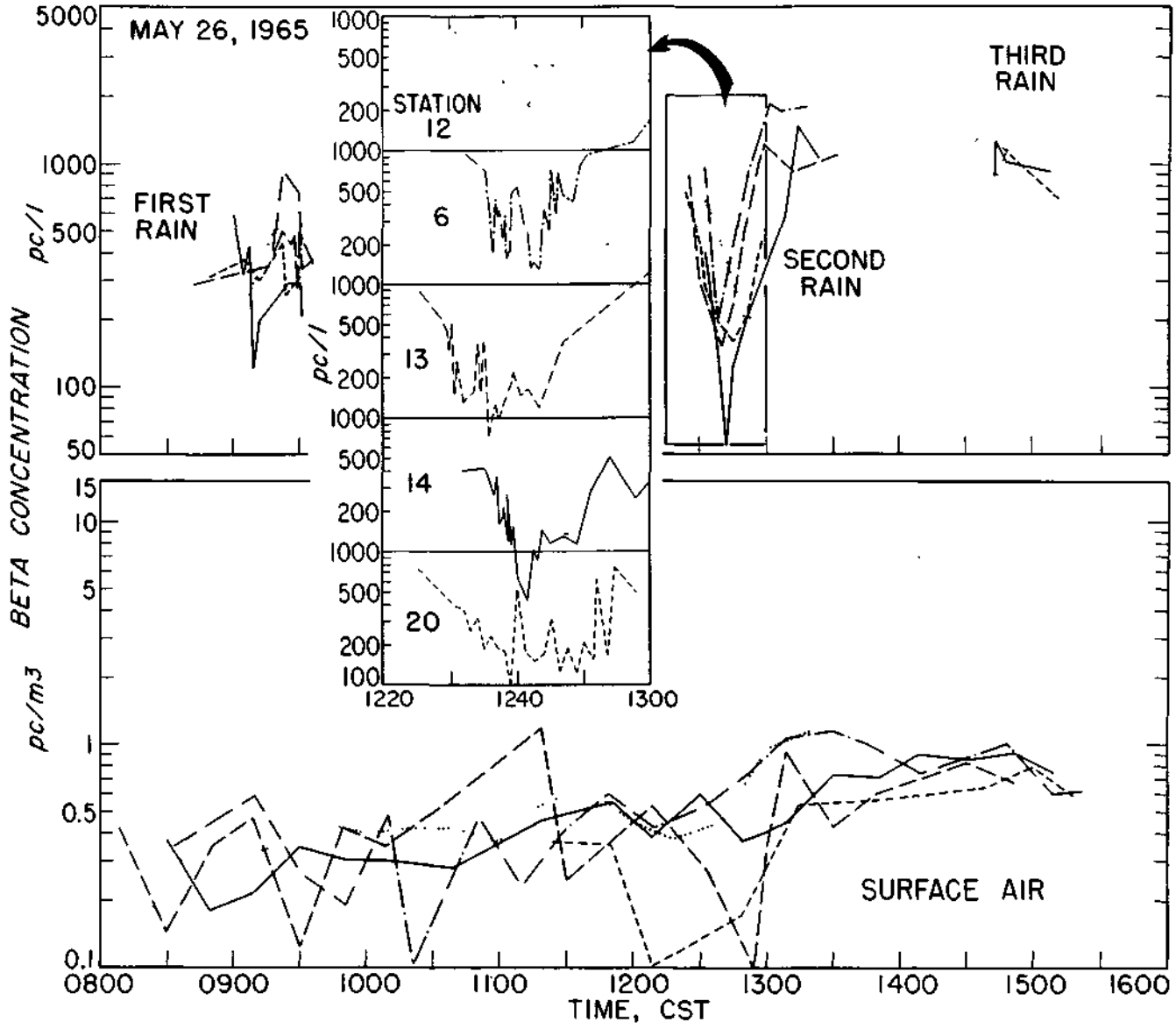


Fig. 1. Gross Beta Concentration of Surface Air and Precipitation Samples on May 26, 1965

aircraft sampling altitude, was used to determine the airborne sample from which each particular drop size originated. For this calculation, drop-size data and rainfall rate for individual drop sizes from Mueller and Sims (1967) were used. Terminal velocity data from Gunn and Kinzer (1949) were also utilized. Then, for each minute, the percentage of the surface precipitation falling from each airborne sample was determined, and the resulting expected surface concentration was calculated.

To facilitate comparing the minute-by-minute calculated surface beta activities with the irregular time period of actual surface samples, a smooth curve was drawn through the centers of the calculated surface activities. This curve is plotted in figure 2. A line was brought down from the center of each of the plotted surface samples to where it intersected the calculated value curve, and the difference was determined by subtracting the calculated from the measured activity. The resulting average surface beta concentration was 337 pc/l. The average of the values calculated for each minute was 198 pc/1. The ratio of, the two, reflecting the apparent increase in concentration during the fall, is 1.7. If the first two very high surface values are excluded, the ratio is 1.6.

In view of the apparent increase in activity between the cloud base and the surface, the question is whether the increase is due to scavenging of radioactive particles, evaporation, or sampling errors. Dingle and Gatz (1966) have calculated the evaporation of drops for different humidities and rainfall rates for the U. S. Standard Atmosphere, 1962. They express their results in terms of K, the ratio of the liquid water content at a particular level to the surface liquid water content. With rainfall rates greater than 10 mm/hr and a humidity of 90 percent they found K to be less than 1.2 for a fall distance of 800 m. The primary difference between the conditions for Dingle's data and this case is that on the 26th the temperature was about 24°C while Dingle used the U. S.

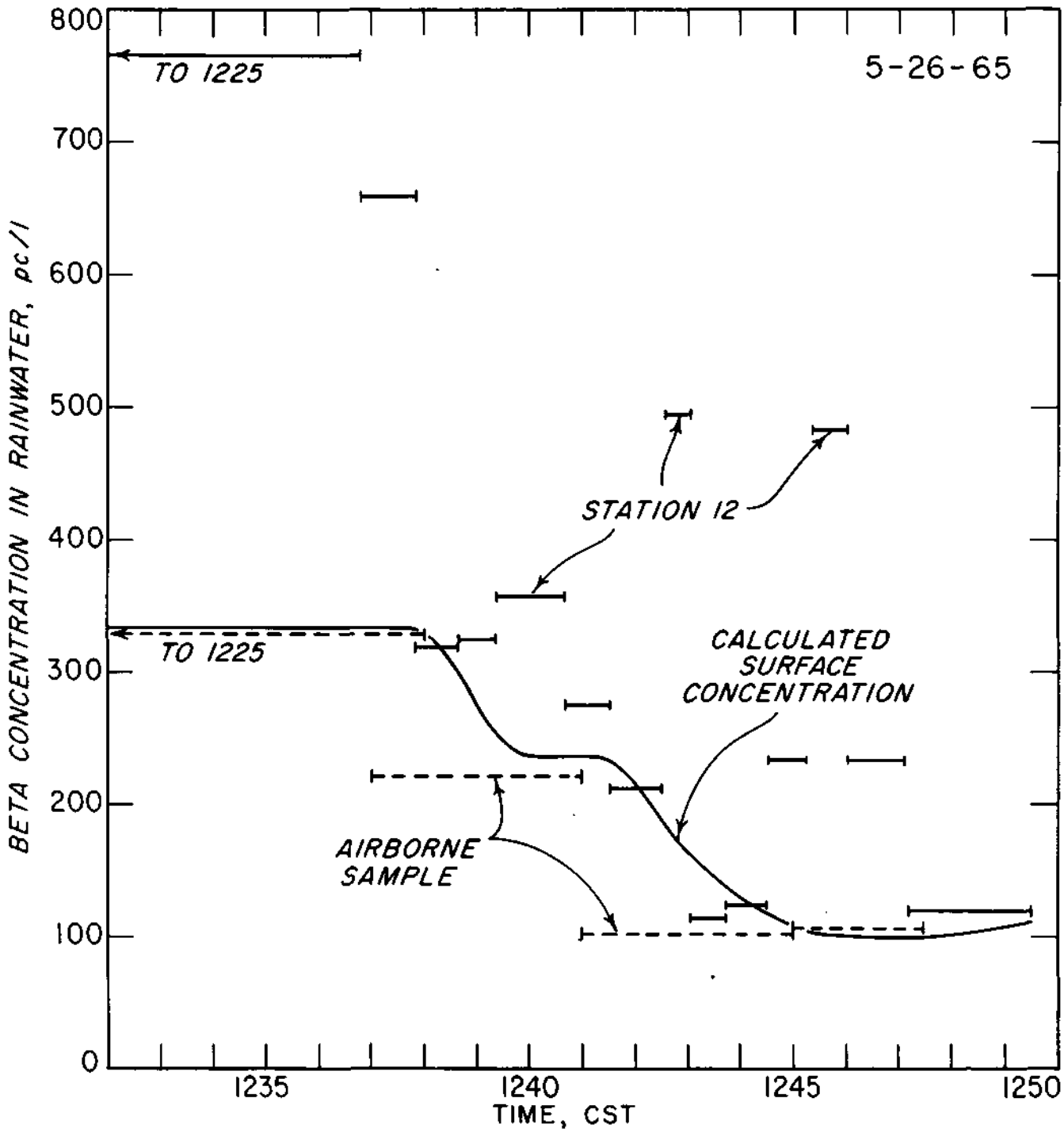


Fig. 2. Comparison of the Gross Beta Concentrations of Airborne, Surface, and Calculated Surface Precipitation Samples on May 26, 1965

Standard Atmosphere with a surface temperature of 15°C. According to Kinzer and Gunn (1951), a difference in temperature of 10°C in the range being considered here will cause a ~40 percent increase in the time rate of change of drop mass from evaporation. In view of this consideration, K should actually be a maximum of 1.3 for the May 26 data because of evaporation. At most, then, evaporation would account for less than one-half of the observed increase in concentration of the beta activity.

Interpretation of this data is difficult because of the assumptions that necessarily had to be made in analyzing the data. Although the data is not conclusive, it is definitely suggestive of a washout process existing between cloud base and the ground.

Storm of June 1, 1965

On June 1, 1965, another airborne water sampling flight was made over the network. Rainfall rates were too light to collect sufficient samples in the first few hundred meters above the surface so sampling was resumed at or near the freezing level. Collection rates at the freezing level are much greater than below because of the lower terminal velocity of the snowflakes as compared to rain, and the resulting larger water content (frozen) per unit volume of air.

The sampling was above a broken layer of cumulus and navigation was by OMNI so that the aircraft position could not be determined as accurately as with visual ground reference, but the aircraft was kept over the network, in general. Because of the high sampling altitude and uncertainty in the aircraft location a direct comparison cannot be made between the surface and airborne measurements but the data are plotted in figure 3 for inspection. The areal variation in the radioactivity concentration of the surface water samplers was not large. The variation with time of the water samples at 3400 m is nearly the inverse of the

surface variation. The magnitude of the concentrations and variations, however, are similar to that at the surface.

If the rain had been as efficient at scavenging the radioactivity on June 1 as it had appeared to be on May 26, the concentration of radioactivity at the surface would have been about five times greater from raindrop scavenging alone than it actually averaged. It may be argued that the samples taken at a relatively high altitude above the entire network were not representative of the surface rain and the samples cannot be compared. This of course is partially true but it is doubtful that the rain sampled by the aircraft was reaching the ground anywhere with a concentration five times the value at aircraft sampling altitude. Although the data from June 1, 1965, is inconclusive it does suggest that very little, if any, radioactivity was being washed out between the cloud base and the ground.

## Potential Increase in Radioactivity

When considering the possibility of precipitation washing out radioactive debris between the cloud base and the ground, it is informative to consider how much radioactivity is in the intervening volume of air swept out by a liter of rain in falling from the cloud base. By assuming a drop-size distribution corresponding to the average rainfall rate during the time of the aircraft sample, corrected for mean time of fall, the individual rainfall rates for each of the drop sizes can be determined with data from Mueller (1967). The individual drop (in 0.1-mm diameter intervals) rainfall rates along with terminal velocity data and height of fall can be used to calculate the total volume of air swept out by the drops that constitute one liter of collected precipitation. Table 2 provides results of such a study for four water samples collected at station no. 12 on May 26, 1965. The volume of air swept out along with the average network air concentration was used to determine the increase in

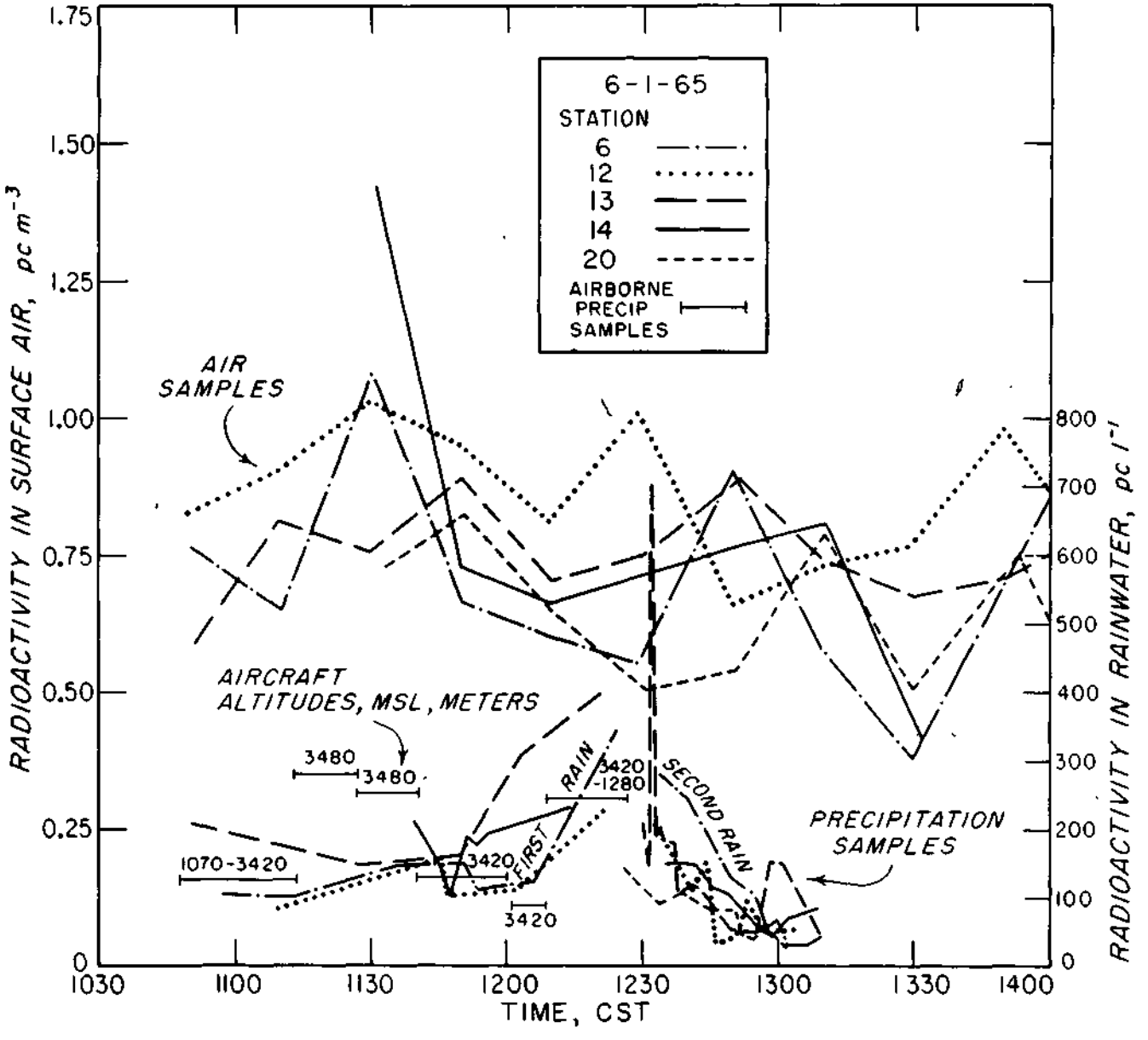


Fig. 3. Gross Beta Concentrations of Surface and Airborne Precipitation Samples Plus Surface Air Samples on June 1, 1965

radioactivity in the water sample that would occur if all the particulates in the paths swept out by the drops were captured, i.e., the potential increase in radioactivity.

Table 2. Determination of Potential Increase in Radioactivity

| Sample No. | Height of Fall (m) | Average Rainfall Rate (mm $hr^{-1}$) | Average Surface Air Concentration (pc $m^{-3}$) | Volume of Air Swept ($m^3$) | Potential $\Delta \beta$ (pc $l^{-1}$) |
|---|---|---|---|---|---|
| 1 | 838 | 8.7 | .45 . | 848 | 382 |
| 2 | 686 | 28 | .45 | 548 | 247 |
| 3 | 686 | 15 | .45 | 645 | 290 |
| 4 | 686 | 2.8 | .45 | 668 | 380 |

The average potential increase in activity for the four airborne samples from May 26 is 325 pc/1. The average increase found for the minute-by-minute calculated surface samples was 139 pc/1. This indicates that there was an apparent increase in the radioactivity of the precipitation equivalent to capturing 43 percent of the activity swept out by the drops. It also means that a volume of air equivalent to the total volume of air between cloud base and the surface was swept out by approximately every millimeter of rainfall.

Sampling Errors

Because of the sampling problems involved the accuracy of the preceding calculations is uncertain. The magnitude of most of the errors introduced cannot be estimated. The error from inaccuracies in the synchronous timing of the surface and airborne samples is less than ±8 percent. Several attempts were made to circle a sampler at very low aircraft elevation to see if the surface and aircraft samples would be in agreement in radioactivity, but the weather never cooperated and insufficient or no precipitation was collected.

The water samples were analyzed at the State Water Survey laboratory in the manner reported by Huff (1965b). Air filter samples were analyzed by TRACERLAB, Inc., Waltham, Massachusetts, under subcontract with the Water Survey. The filter papers were ashed and the residue transferred to a counting planchet. Beta counting was done in an internal flow proportioned counter. An accuracy of ±10 percent was reported.

Two tests were made with three air samplers side by side for a series of samples to determine the normal variation in sampling. A number of samples were also recounted by TRACERLAB. From this data it appears that much of the variation from sample to sample and from station to station is the result of the counting and sampling errors. It was intended to achieve considerably more accuracy in the air samples. Unfortunately, it was determined too late that the counting errors were several times larger than the -10 percent estimated by the subcontractor when the analytical procedure was established.

Tests were also carried out to determine if there was significant deposition of radioactive debris in the surface and airborne intake collector tubes. Samples were collected in a normal manner, after which, the tubes were cleaned with one or two moist filters. The air sample and cleaning filters were both counted. The counting error again was large on the cleaning filter because of low activity levels, but the retention on the tube walls appears to have been less than 10 percent.

## TRITIUM SOUNDINGS

The use of tritium as a tracer for the movement of water and water vapor has enjoyed considerable success since the Castle tests in 1954. Libby (1959) used bomb-produced tritium to estimate the northern hemispheric tritium storage time and deposition rates, as well as the ground water balance of the upper

Mississippi Valley. Eriksson (1958) has also studied groundwater storage with the use of tritium. More recently Eriksson and Oden (not dated) and Smith (1966) have pointed out the use of tritium in studies of atmospheric moisture transport. In addition to bomb-produced tritium, reactor-produced tritium promises to be a useful tool for the tracing of water vapor and groundwater.

The increased use of tritium as a tracer has resulted in a need for further information on the present background of atmospheric tritium, and especially on its vertical distribution, variation with time, and variation between synoptic weather types. As a result of these considerations a tritium sampling program, in cooperation with Dr. Eriksson of the International Meteorological Institute in Sweden, was initiated in the winter of 1966.

During 1966, 13 soundings have been made to measure the vertical distribution of tritium in atmospheric water vapor. Most of the soundings were taken during the months of April, June, and July, and were usually made to an altitude of nearly 5000 m MSL. Appendix A lists the date, altitude, and number of samples taken on the soundings. Dr. Eriksson was responsible for gas counting the tritium samples. Various delays in the analyses in Sweden, however, have resulted in only 8 samples being analyzed at the date of this writing. Interpretation of the data, therefore, remains to be done.

## Instrumentation

Water vapor samples for tritium analysis are collected with a twin-engine Beechcraft. The water is absorbed in traps containing 400-500 grams of Lindy molecular sieve No. 4AXW. Air is sampled through a forward facing nozzle and drawn through the sieve traps with a Lieman positive displacement blower, belt-driven by a l-hp motor. The sampling rate is estimated to be about 0.5 $m^3$ $min^{-1}$.

A block diagram of the laboratory vacuum system for removing water samples from the traps is shown in figure 4. The system consists of a small mechanical vacuum pump followed by a liquid nitrogen cooled vacuum trap (LN trap) and an 8-liter-per-second oil diffusion pump. The diffusion pump in turn is followed by an LN trap and the system manifold. The traps on either side of the oil diffusion pump isolate the vacuum system from pump oil vapor and protect the mechanical pump from water vapor.

The system manifold is fitted with both thermocouple and ion vacuum gages. Connected in parallel to the manifold are four LN traps to collect water vapor from the molecular sieve traps to which they are connected. The four sieve traps are heated in an electric furnace to aid in removing the water. All tubing is large-diameter pyrex and connections are with greased pyrex ball-and-socket joints. Several stop-cocks facilitate isolating various portions of the system. A thermocouple temperature regulator maintains the oven temperature at approximately 560°C. Initially, the sieve traps were baked for seven hours at maximum temperature, but with the addition of an automatic temperature regulator overnight bakes of 18 hours or more have been possible.

## ISENTROPIC TRAJECTORY ANALYSES

### Objectives of Study

The objective of this study was to investigate the feasibility of isentropic trajectory analyses for tracing radioactive air with respect to precipitating systems. During the course of the study the airflow associated with frontal precipitation on May 26, 1965 was investigated. A part of the showers which moved over the sampling network on that day were associated with a relatively high concentration of radioactivity in both the air and rainwater (Huff

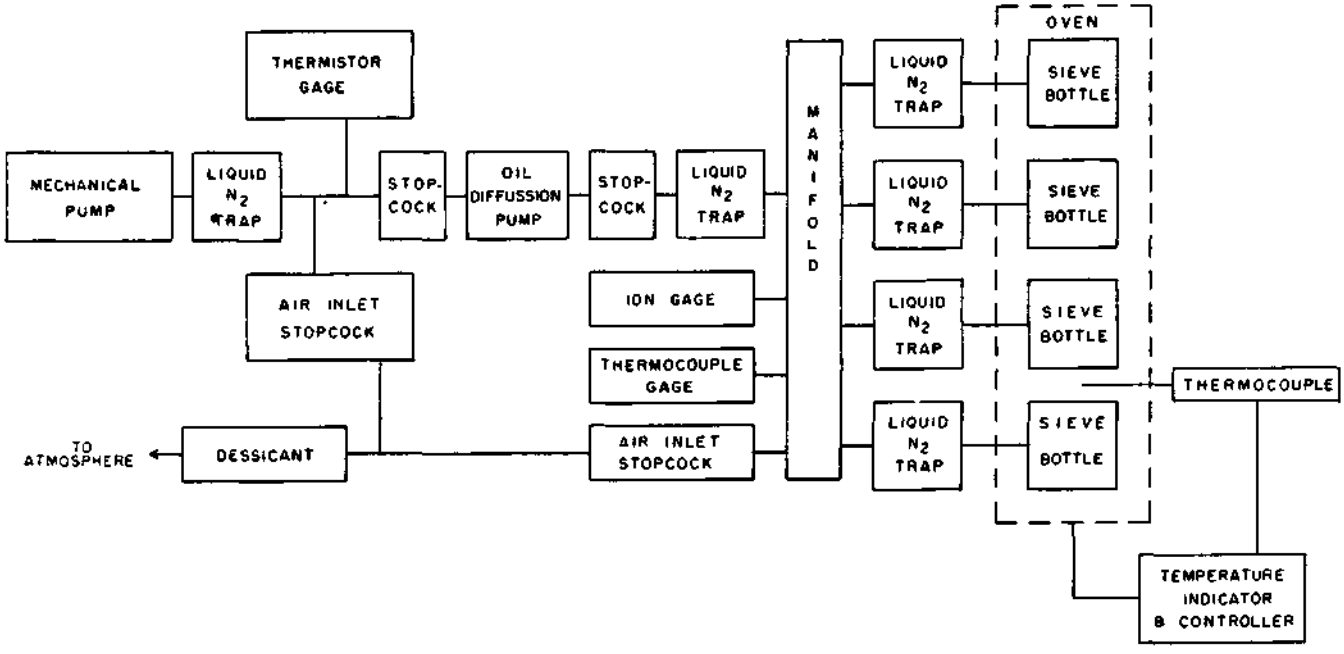


Fig. 4. Vacuum-furnace System for Removing Water Vapor from Molecular Sieves

and Bradley, 1965). This case offered an opportunity to seek an answer to the question whether the radioactivity in air and in precipitation, which are occasionally observed in showers or thunderstorms, is entrained directly from the stratosphere or whether the precipitating clouds are fed by contaminated air of tropospheric origin.

In the initial phase of the project the development of fast methods of trajectory computation was a main objective. These rather simple techniques would have to eliminate a large amount of labor without unduly sacrificing accuracy.

Using the techniques developed, the trajectories of air which arrived at different levels above the precipitation area and its environment were followed backward, over a period of 12 hours, and the average vertical motion of the air computed.

The next objective was to investigate the feasibility of relating the radioactivity at the surface to that aloft. For this purpose, air samples were taken by B-47 and B-57 aircraft at different levels close to the rain area. The measurements of wind and temperature by the airplanes as well as the locations and radioactive content of the air samples were displayed on a vertical cross-section. Then, this mesoscale cross-section was compared with synoptic-scale cross-sections from conventional upper air data to ascertain the presence of disturbances in the flow and variations in radioactivity.

Finally, a feasibility study was made to determine whether the air in which the airplane samples were made could be traced back to approximately the same source as the air which was thought to be involved in the burst of radioactivity over the surface network.

## Analysis Techniques

Three separate analyses are required for three-dimensional trajectory computations on an isentropic surface, namely streamline, isotach, and pressure.

These analyses must be mutually consistent in all areas. The analysis technique which achieves this is described in detail by Feteris (1965).

In the analyses, all short wavelength oscillations are smoothed out, except those associated with troughs and ridges which exhibit time-continuity. Where data is sparse, cross-sections help to locate wind speed maxima and minima on isentropic charts. It has been shown (Feteris, 1965) that this time-consuming analysis still leaves room for ambiguities of at least 50 miles in the horizontal and a few thousand feet in the vertical in the location of the boundaries of different air masses. This is due to the wide spacing between the radiosonde and wind observations over the United States. Trajectory analysis tends to increase this uncertainty even if it is carried out rigorously according to the equations of conservation of energy (Danielsen, 1966). The excessive labor involved in such an analysis, in spite of its rigour, was not justified by the frequency and the density of the data from the radiosonde network and methods were sought to expedite the trajectory computations.

In development of a simpler technique, it was assumed that the total energy was conserved. The total energy is defined as:

$$E = \left(\psi + \frac{V_H^2}{2} + Lq\right) \qquad (1)$$

in which $\psi$ is the Montgomery streamfunction, $V_H$ the horizontal velocity of the air, L the latent heat of condensation and q the humidity mixing ratio. The kinetic energy of the vertical motion of the air is neglected.

It was then assumed that there was no diabatic heating or cooling by radiation and that the energy E of the air parcels was conserved or that:

$$\frac{d}{dt}\left(\psi + \frac{V_H^2}{2} + Lq\right) = 0 \qquad (2)$$

Since in sinking air or in rising dry air the humidity mixing ratio is constant, the term Lq in (1) does not change over the trajectory and can be discarded.

The remaining terms $\psi + \frac{V_H^2}{2}$ form the specific energy $E_{sp}$ which can be computed rapidly by converting units of wind speed to units of energy on the isotach charts and adding $\frac{V_H^2}{2}$ graphically to $\psi$. The assumption that $\frac{d\,E_{sp}}{dt} = 0$ introduces errors since, according to Danielsen (1966),

$$\frac{d\,E_{sp}}{dt} = \frac{d}{dt}\left(\psi + \frac{V_H^2}{2}\right) = \left(\frac{\partial \psi}{\partial t}\right)_\theta + \frac{\partial \psi}{\partial \theta}\frac{d\theta}{dt} \qquad (3)$$

$\frac{d\theta}{dt}$ can change due to radiational heating or cooling and is probably small if the air is dry. $\left(\frac{\partial \psi}{\partial t}\right)_\theta = \left(a\,\frac{\partial p}{\partial t}\right)_\theta$ and depends on the changes in the thermal gradient. This term can be large in areas where cyclonic development takes place. By considering trajectories which did not extend over a period longer than 12 hours, the error introduced by omitting $\left(\frac{\partial \psi}{\partial t}\right)\theta$ was mimimized as much as possible.

Under the above assumptions, $E_{sp}$ can be considered conserved for motions in the isentropic plane only. E can also be conserved for upward motions through that plane. Therefore, where condensation of water interferes in rising air, the air will move to levels with higher potential temperatures, and the total energy will not be constant at the end-points of a trajectory in the isentropic plane. Inspection of the total energy at lower levels near the initial position of the air parcel may indicate qualitatively whether air from lower isentropic levels was involved in the rising motions.

Trajectory analysis was performed for a rectangular cluster of points which was found to be distorted considerably after having passed through the time-dependent wind field on the isentropic plane. The trajectory analysis had to fulfill the following requirements:

1. The air had to travel over a distance which is consistent with the average wind speed over the trajectory during the 12-hour period.

2. The specific energy at the end point of the trajectory had to be the same as that at the starting point.
3. The areas enclosed by a set of four points had to remain the same (the air does not move through the isentropic surface if it is sinking or far from saturation).
4. When the air decelerated, the trajectories had to cross the streamlines towards higher $\psi$ values; the reverse is true for accelerating air.

These conditions were generally met where dry air descended behind a cold front. In areas with cloud and precipitation it was not possible to fulfill the above requirements, which means that the start and end points of the trajectories did not represent air from the same isentropic surface. The analyses were considered satisfactory when the change in area between four adjacent points was less than 10 percent in 12 hours, corresponding to a divergence of less than $2 \times 10^{-6}$ $sec^{-1}$.

Synoptic Conditions in Storm of May 26, 1965

The storm of May 26 included two centers. One with a pressure center of 995 mb over North Dakota and the Canadian border was almost stationary. On the surface map warm air from the Gulf was separated from dry, cold air over the Rockies by a trailing cold front. On this front another low pressure center developed over Western Texas and moved north-northeastward towards Lake Superior while it deepened somewhat. Precipitation was confined to the Northern Rockies and the Northern Plain States where rain and snow showers fell and to a few squall lines which developed Over Kansas and Iowa while moving northeastward into Illinois and Michigan. These smaller scale features dominated the circulation and the weather over the Midwest. Otherwise, no dramatic changes were seen in the general flow pattern on the surface map.

Figures 5 and 6 display the weather maps which pertain to the period in which the field observations were made.

The 500-mb maps in figures 7 and 8 show warm advection from the southwest over the eastern part of the United States and cold advection over the Rockies and the Plain states, which resulted in an intensification of both the thermal gradient and the flow over Kansas, Missouri, Iowa and Illinois. This intensification of the gradients of pressure and temperature is also apparent from the cross-sections of May 26, 1200 GMT, and May 27, 0000 GMT, between Rapid City, North Dakota, and Athens, Georgia (Figs. 9-10). These show a lowering of the jet maximum from 260 to 300 mb and upper level frontogenesis. Cold air descended behind the cold front but apparently did not completely penetrate into the surface boundary layer. There is also some doubt as to whether this air was of stratospheric origin.

A secondary wind maximum was apparent at 1200 GMT over southern Illinois at the 500-mb level. Later, secondary wind maxima also developed at higher levels over Tennessee. The winds in these regions were from a more westerly direction than the main jet, which ran parallel to the thermal gradient across the cold front. The shear between the southwesterly flow at the surface and the westerly winds aloft over Tennessee was only loosely connected with the synoptic-scale thermal gradient in this area.

Considerably thunderstorm activity was reported in the area where the jetstream branched into the two wind maxima. One of these thunderstorms over Illinois was associated with a localized burst of high radioactivity, both in the air and in the rainwater.

Source of Air Sampled on May 26

Air trajectory computations were performed to trace the movements of the air in the vicinity of the precipitation area as well as those of the air

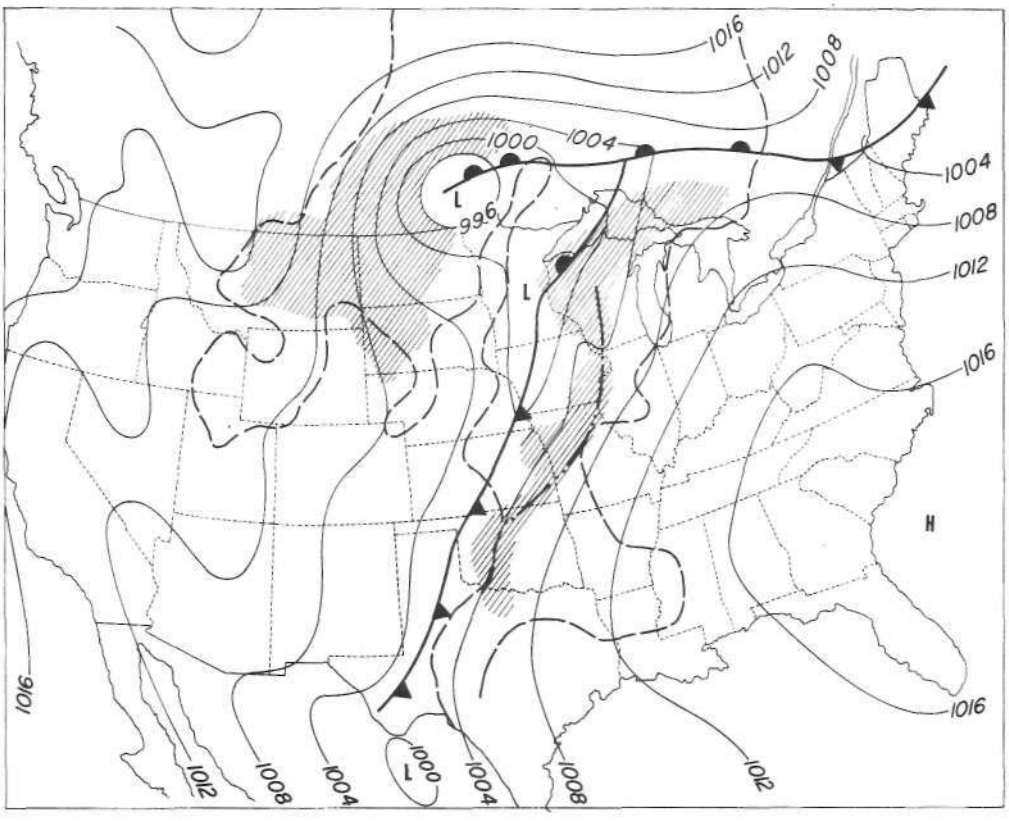


Fig. 5. Surface Analysis, May 26, 1965, 1200 GMT

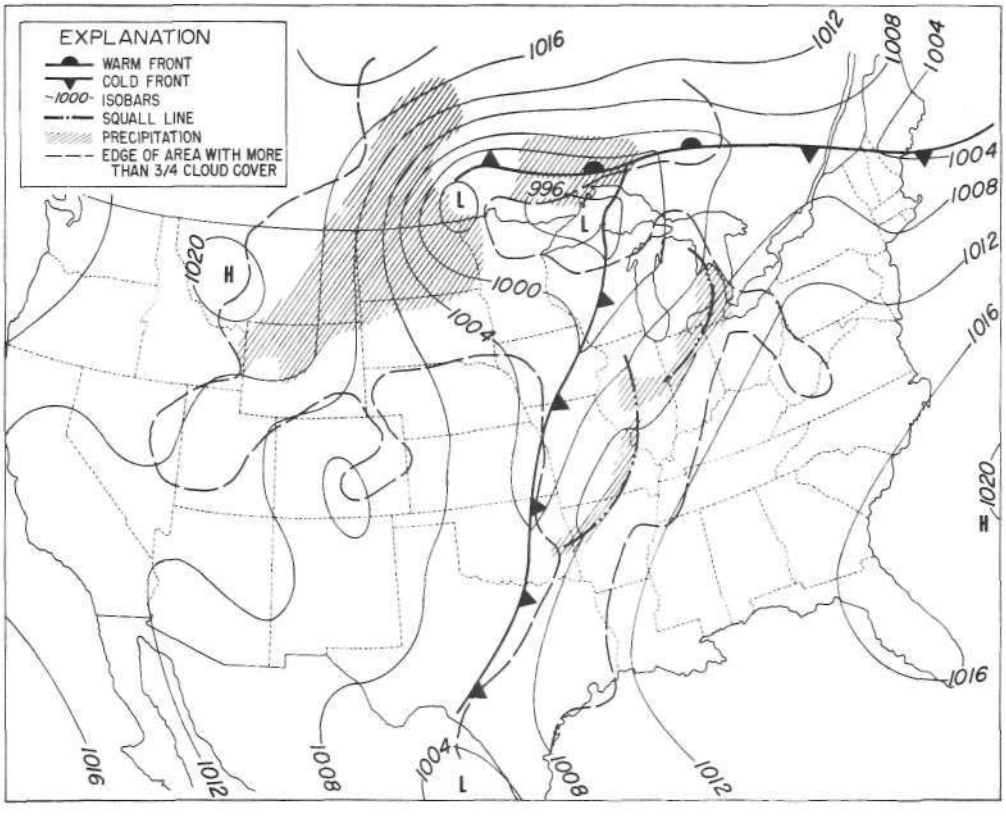


Fig. 6. Surface Analysis, May 26, 1965, 1800 GMT

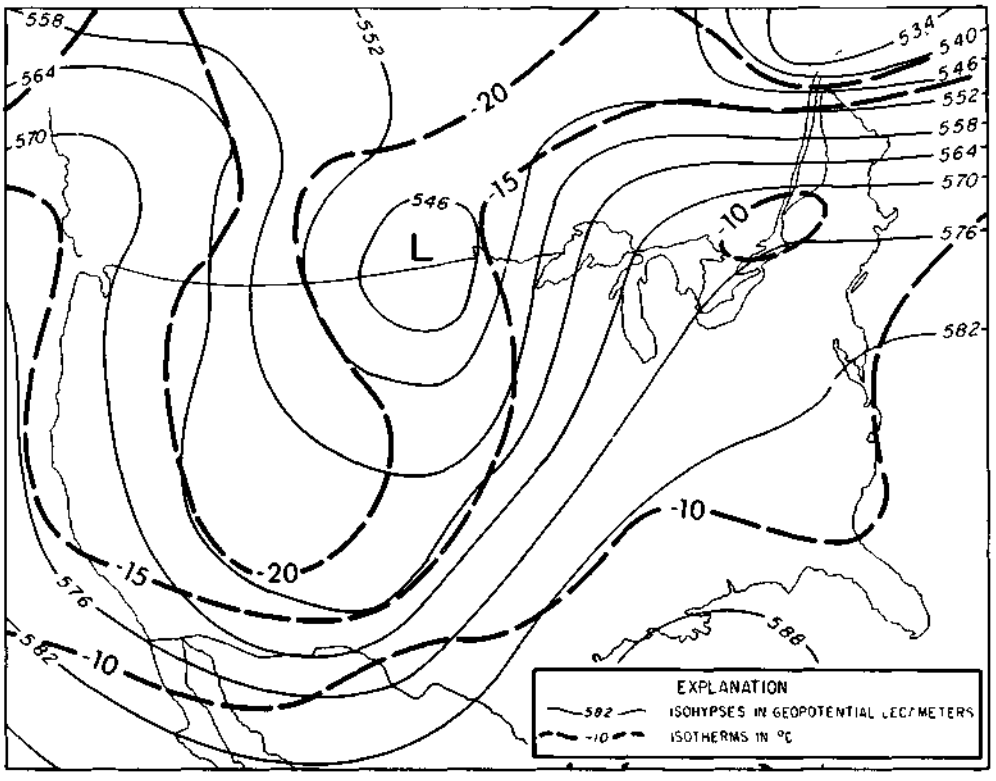


Fig. 7. 500-mb Analysis, May 26, 1965, 1200 GMT

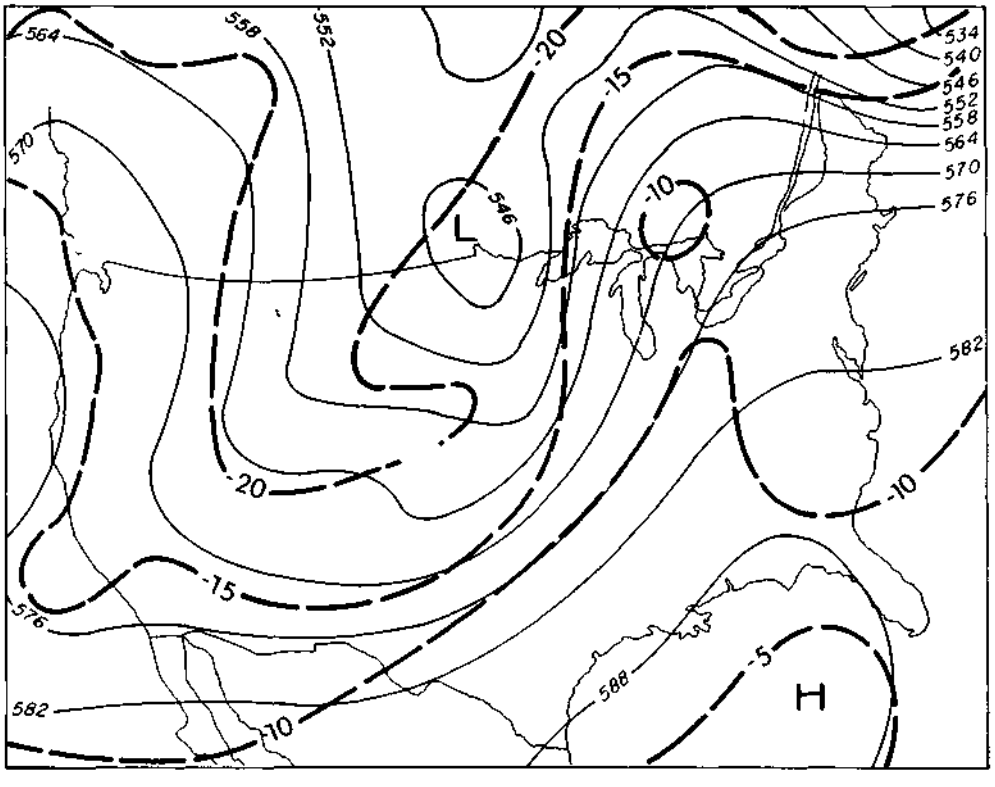


Fig. 8. 500-mb Analysis, May 27, 1965, 0000 GMT

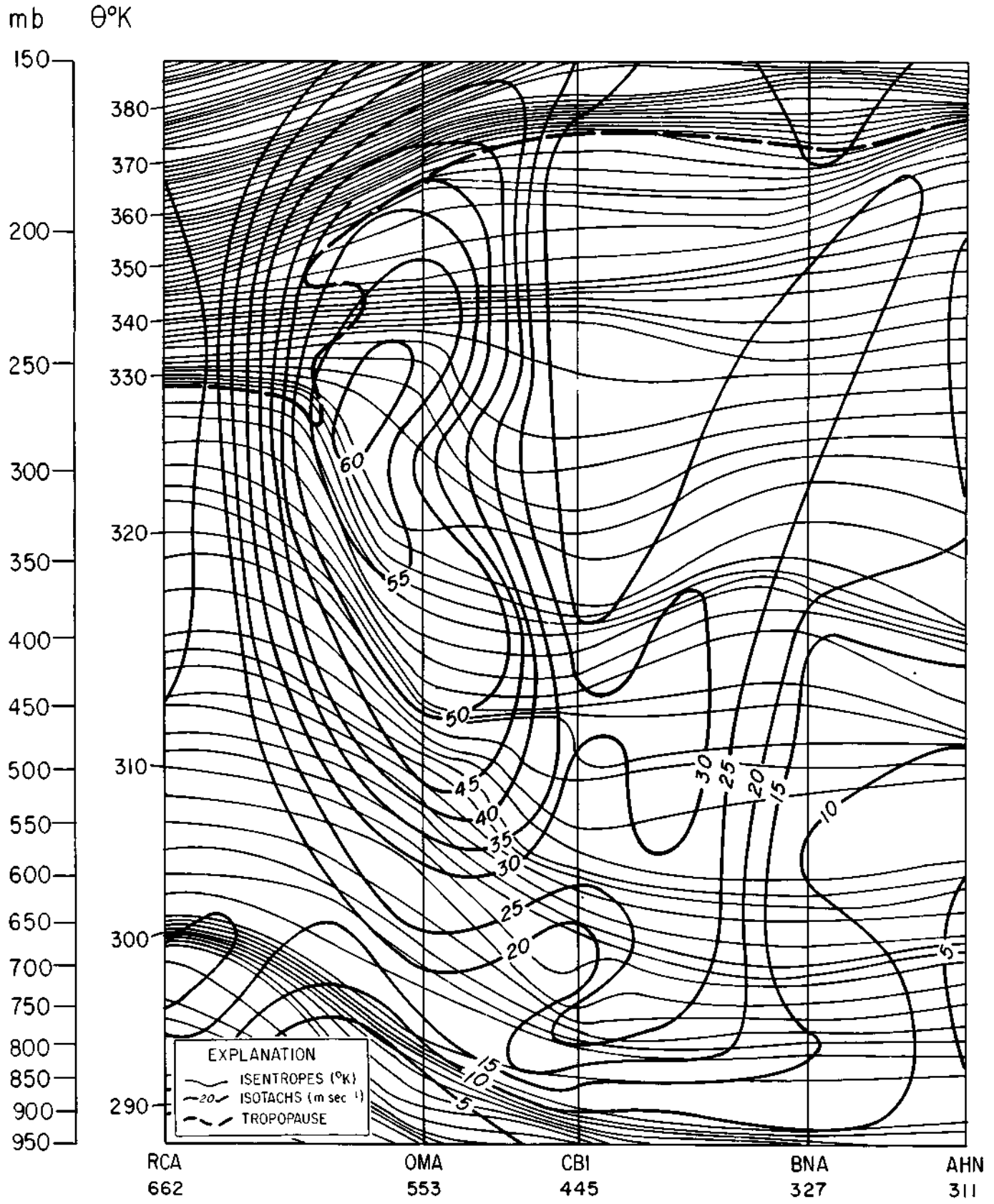


Fig. 9. Atmospheric Cross-section, May 26, 1965, 1200 GMT

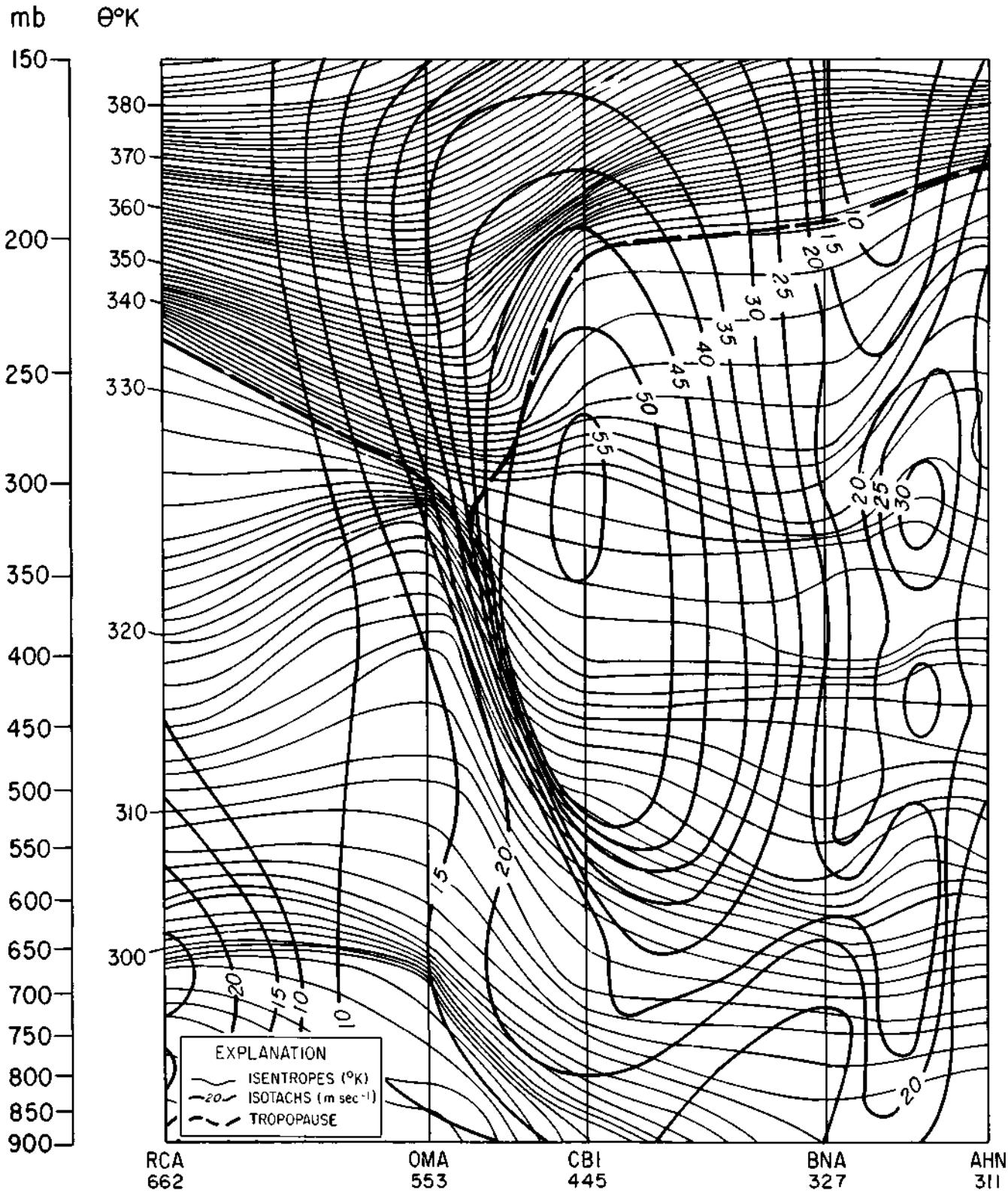


Fig. 10. Atmospheric Cross-section, May 27, 1965, 0000 GMT

which was probed three hours later by B-47 and B-57 aircraft for radioactivity.

Pressure, streamfunction, and isotach analyses were made for seven isentropic levels at five-degree increments from 310k through 340k on May 26, 1200 GMT, and May 27, 0000 GMT. Figures 11 and 12 show the pressure, streamline, and isotach analyses of the levels of 320 and 330k between which most of the air samples were taken. The streamfunction analysis is repeated in the figures to provide a better reference for the isotach and pressure analysis. These maps show qualitatively that the cold advection was accompanied by descending air motions over Kansas, Missouri, Iowa and Illinois, but they provide a poor representation of the real three-dimensional flow, because the air decelerated rapidly over this area. Consequently, the flow was strongly cross-streamline towards higher values of $\psi$ and p. However, ithe pressure analysis is necessary to determine the pressures at the end points of a trajectory.

Trajectory analysis for a rectangular cluster of points is presented for the levels of 320 and 330k in figures 13 and 14. In areas where the vertical velocity changed sign ambiguities appeared in the trajectory analysis due to a very flat specific energy distribution. It seemed possible to trace some part of the air back to two different sources and also satisfy continuity requirements in both cases. One source was consistent with trajectory curvature computations; the other source clearly represented different air which must have moved upwards to a higher isentropic level and a region with higher specific energies. It was assumed that clouds and precipitation were formed in rapidly ascending air along the trajectories from this source.

The edge of the descending air at 1800 GMT, the time of the field observations, is shown in the figures. It almost coincides with the position

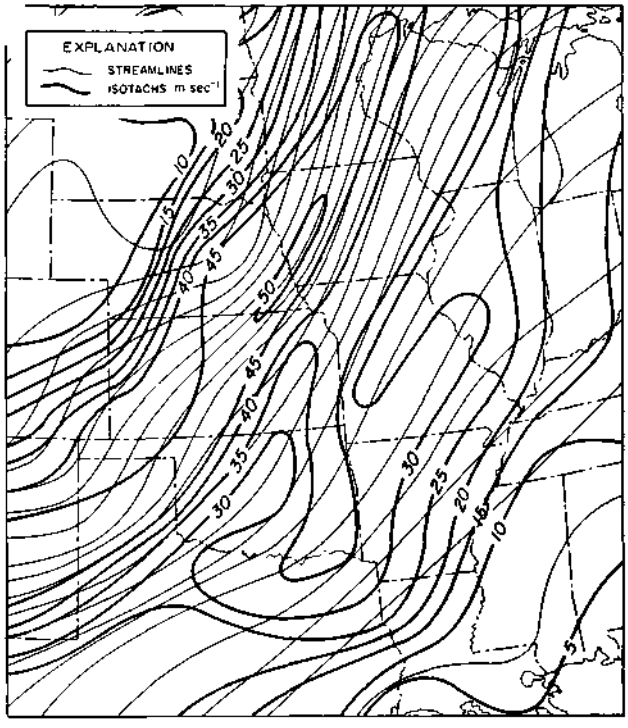


Fig. 11a. 320° K Streamline-Isotach Analysis, May 26, 1965, 1200 GMT

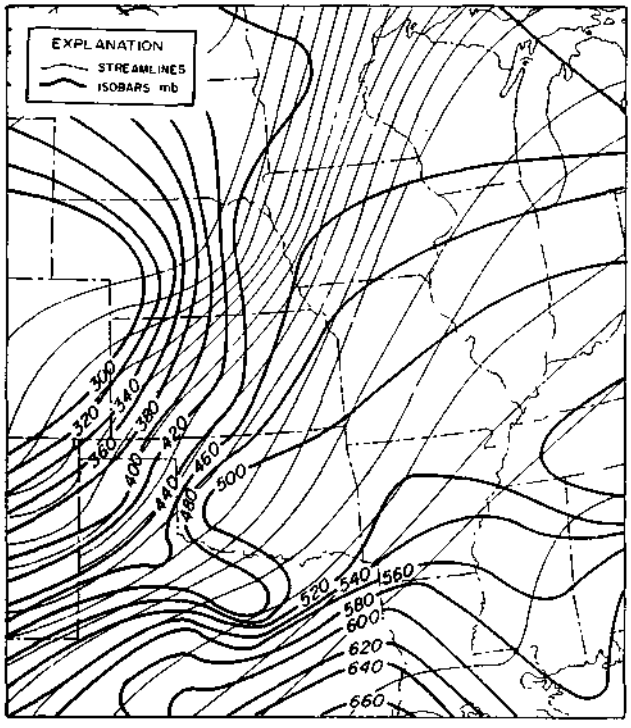


Fig. llb. 320° K Isentropic Pressure-Streamfunction Analysis, May 26, 1965, 1200 GMT

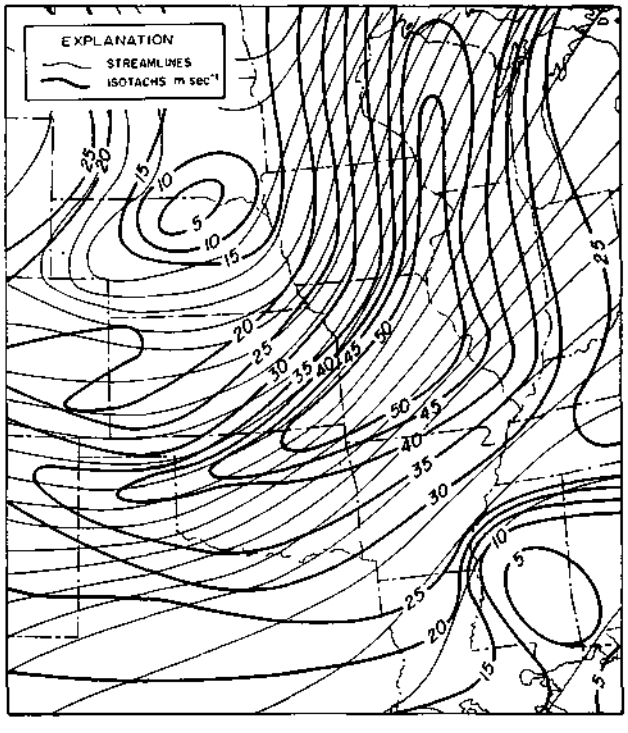


Fig. llc. 320° K Isentropic Streamline-Isotach Analysis, May 27, 1965, 0000 GMT

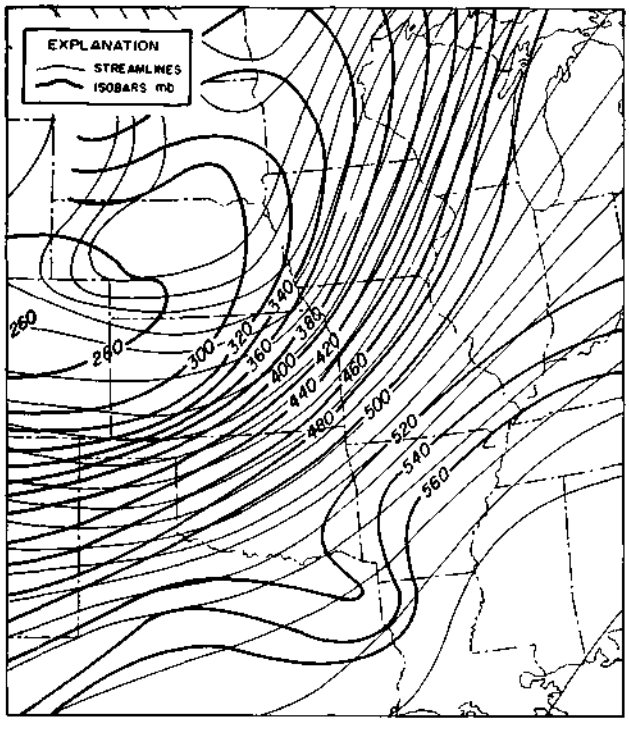


Fig. lld. 320° K Isentropic Pressure-Streamline Analysis, May 27, 1965, 0000 GMT

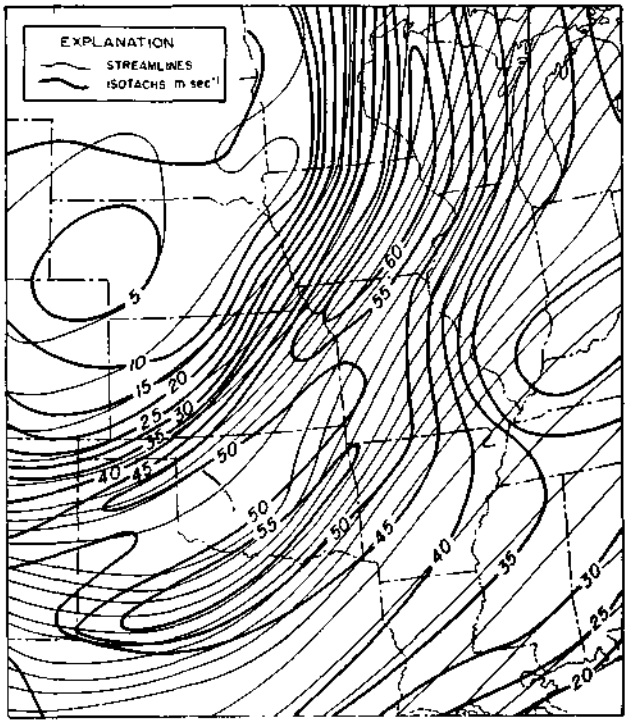


Fig. 12a. 330° K Streamline-Isotach Analysis, May 26, 1965, 0000 GMT

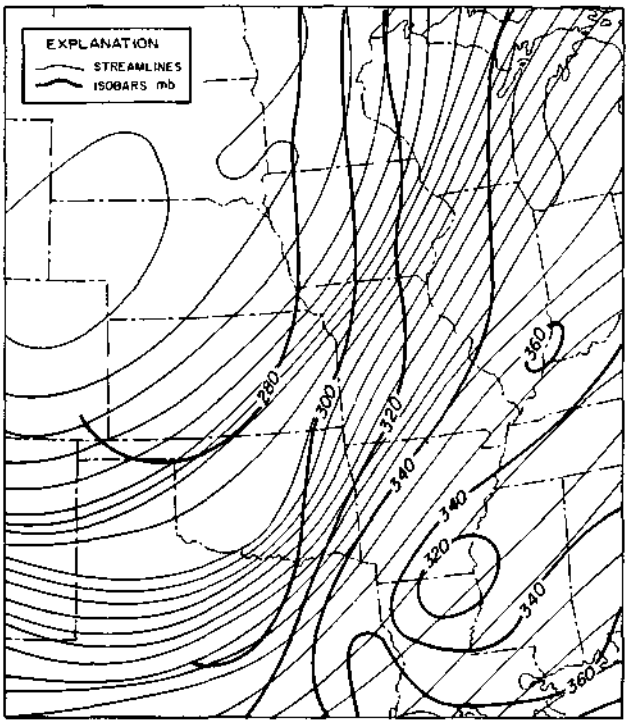


Fig. 12b. 330° K Pressure-Streamline Analysis, May 26, 1965, 0000 GMT

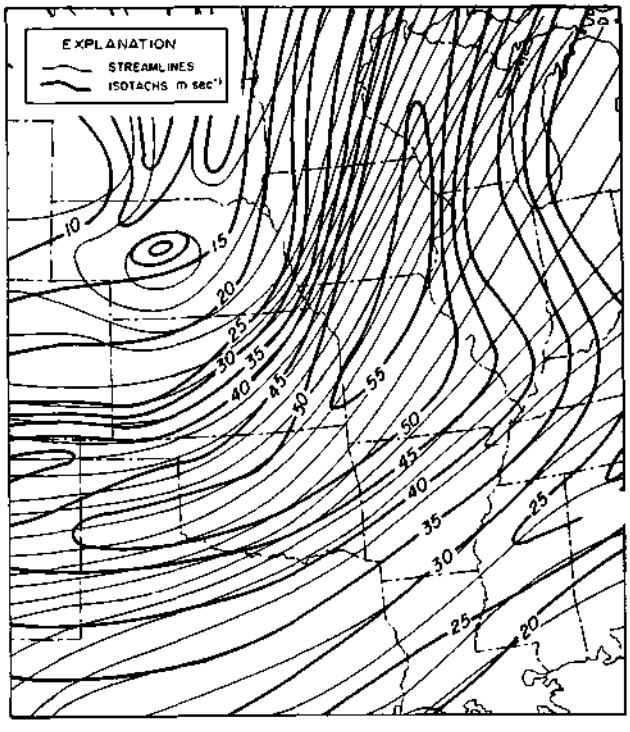


Fig. 12c. 330° K Streamline-Isotach Analysis, May 27, 1965, 0000 GMT

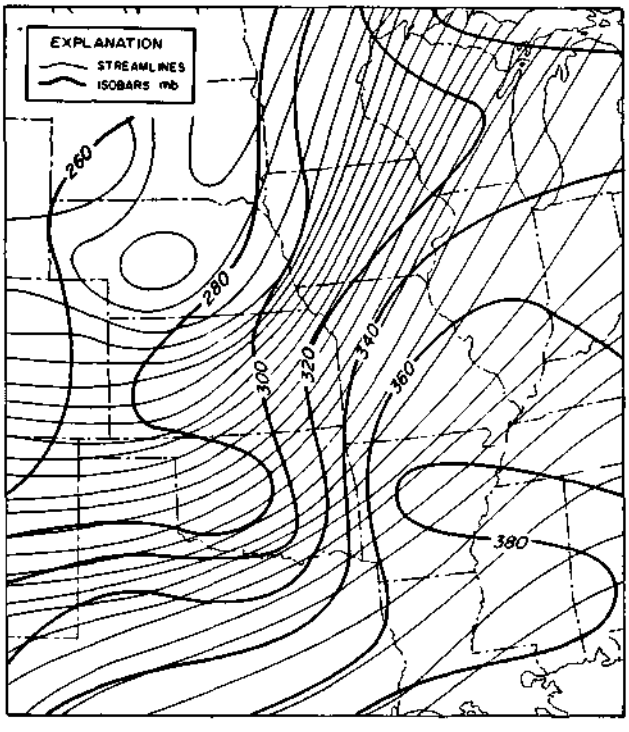


Fig. 12d. 330° K Pressure-Streamline Analysis, May 27, 1965, 0000 GMT

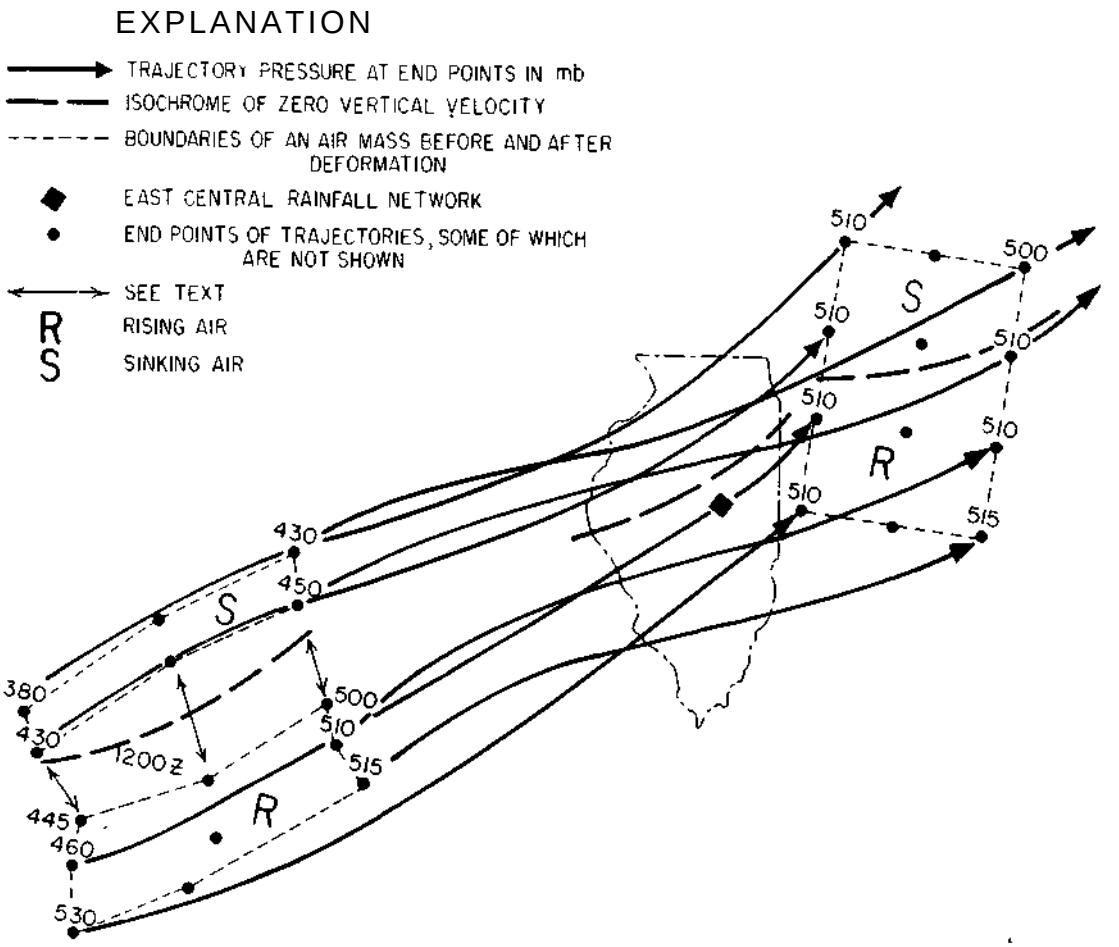


Fig. 13. Trajectories and Isochrones of Zero Vertical Velocity at 320 K on May 26, 1965. Note ambiguity in the location of air parcels near the zero vertical velocity isochrone, the position of which is interpolated between the two boxes at 1200Z.

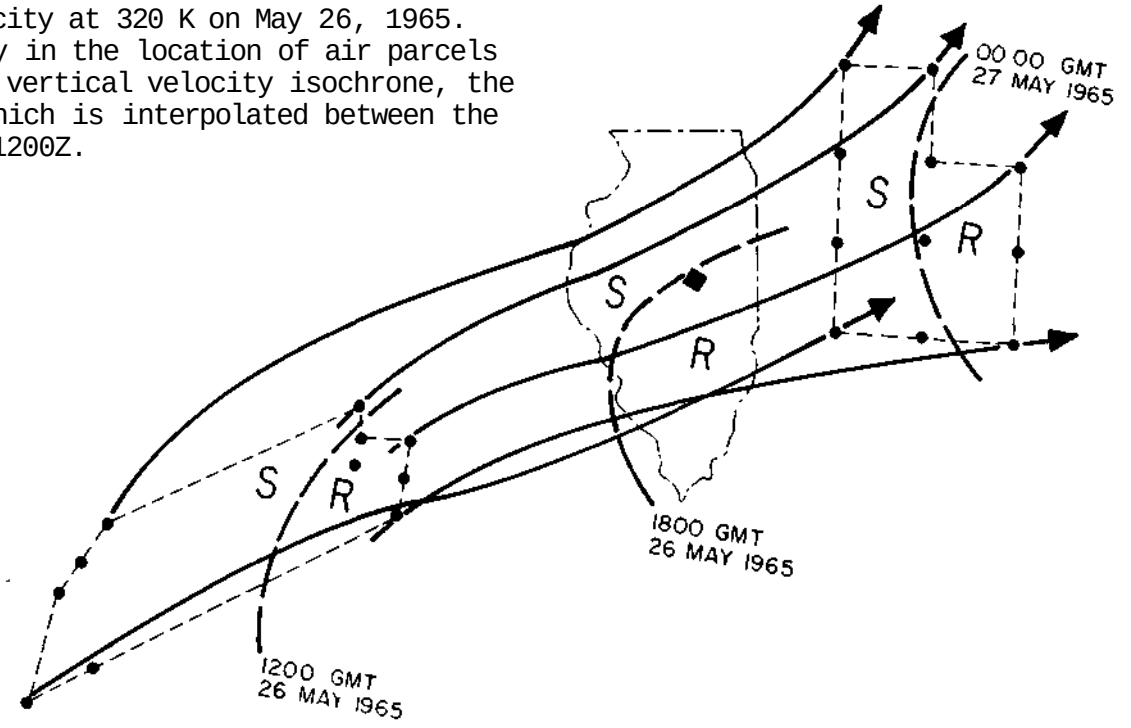


Fig. 14. Trajectories and Isochrones of Zero Vertical Velocity at 330° K on May 26, 1965

of the squall lines in figure 6, so that it seems likely that some of this air was introduced in a local downdraft which produced the observed high concentrations over the rain sampling network. No downward motions were found below levels of approximately 550 mb. A study of the situation on the local scale revealed that the rise in radioactivity at the western edge of the sampling network preceded the increase in radioactivity in the rain-water by at least 20 minutes. The wind, which had been almost calm during the morning, increased from the west to 6 mph when the radioactivity increased. The showers which followed moved in from the south-southwest and passed mainly to the east of the network. This makes it unlikely that they were associated with the contaminated air which only affected the westernmost edge of the network. A possible source might have been the outflow from a strong, sharply defined echo which passed 10 miles west of the network, 45 minutes before the radioactive air arrived. Lack of wind observations in the vicinity of this echo precludes any further evidence as to the source of the contamination, but it can be concluded that it was very local and that rain which fell through it collected a considerable amount of radioactive debris.

It was pointed out by Huff and Bradley (1965) that much of this debris was from a Chinese bomb test which was made 12 days earlier. The distribution of radioactivity in the air samples taken by the B-47 and B-57 airplanes suggests the same.

Figure 15 shows a cross-section constructed from the aircraft observations over Illinois between 1900 and 2100 GMT. A comparison was made with the cross-sections from Dodge City to Burrwood on May 26, 1200 GMT, and from International Falls to Pittsburgh on May 27, 0000 GMT, which were constructed from radiosonde data (Figs. 16-17). The comparison shows meso-scale features not detected in the synoptic-scale cross-sections. Although the pressures and

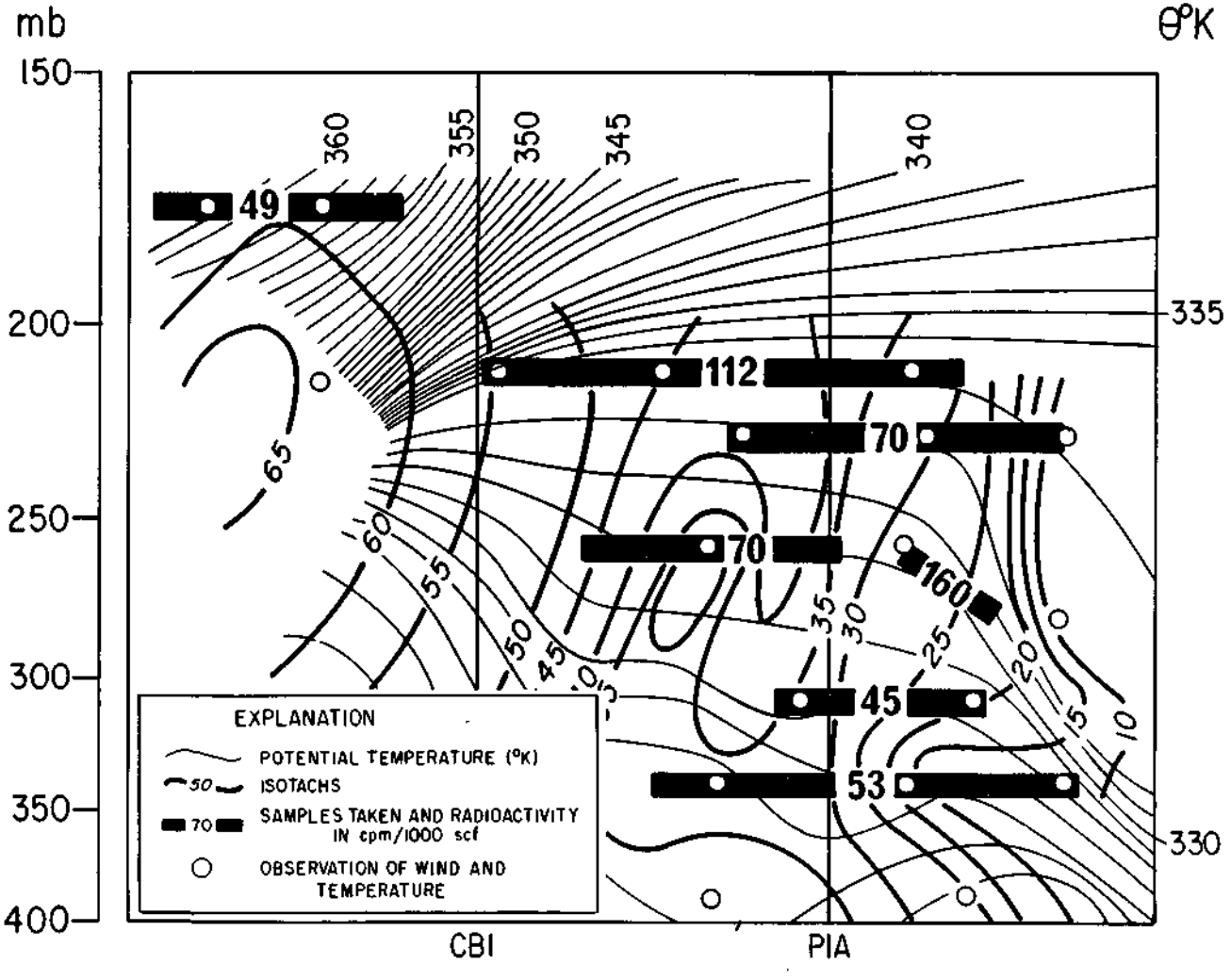


Fig. 15. Cross-section Composed from Aircraft Observations of Wind and Temperature, 2030-2220 GMT

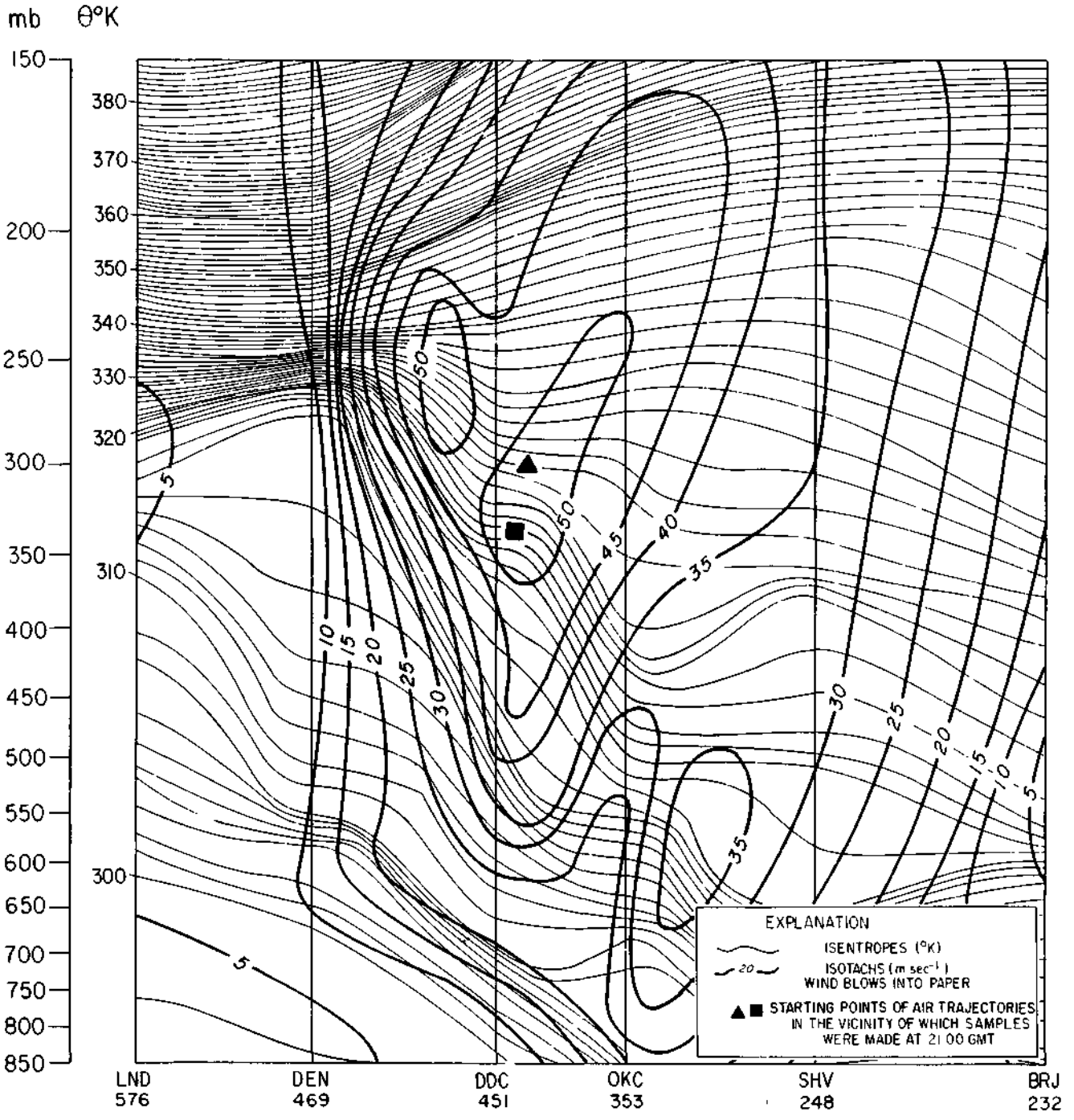


Fig. 16. Atmospheric Cross-section, May 26, 1965, 1200 GMT

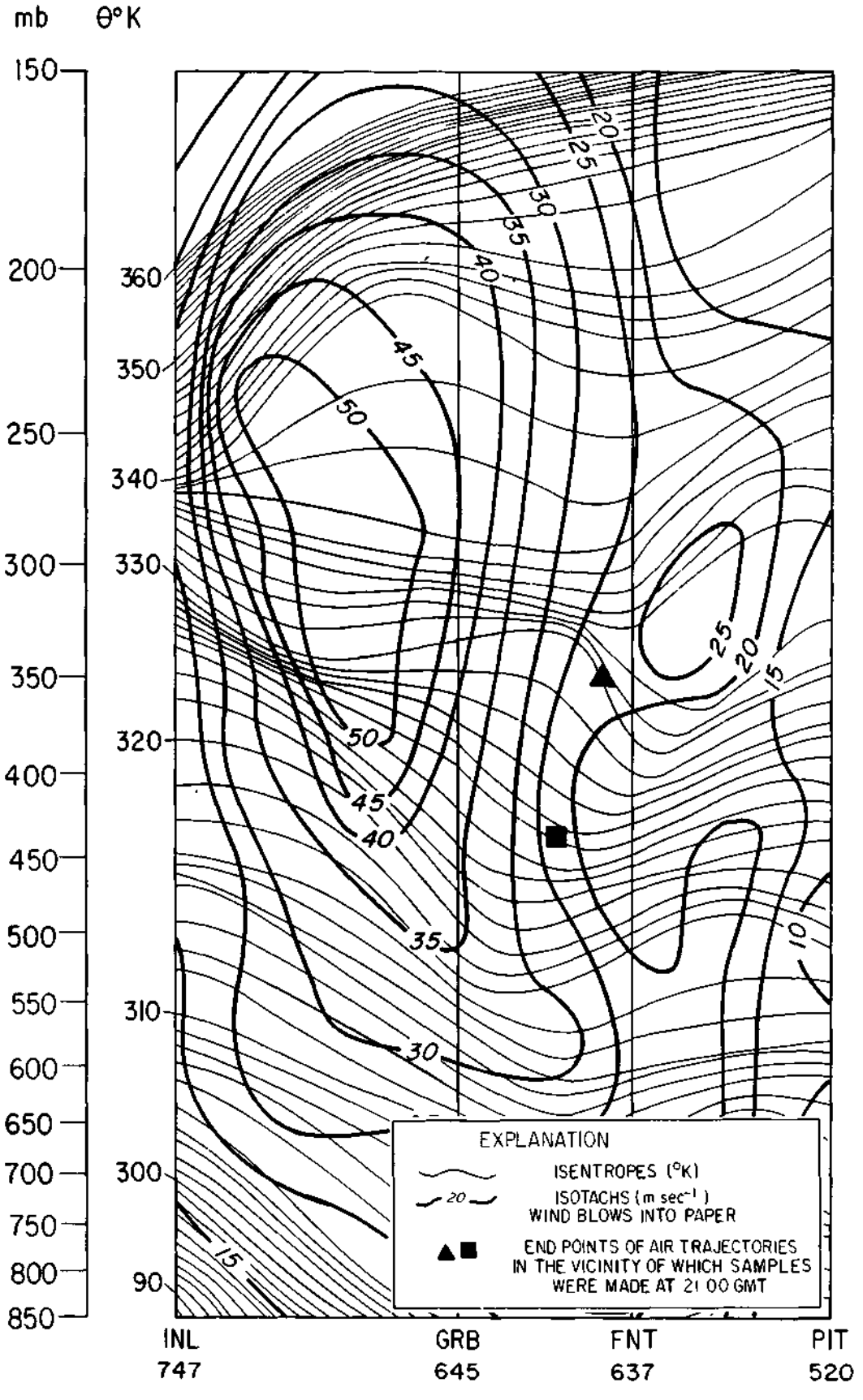


Fig. 17. Atmospheric Cross-section, May 27, 1965, 0000 GMT

temperatures on these figures depart several degrees from those on figure 15, due to differences in the computations of heights, temperature corrections, etc., the set of aircraft data is internally fairly consistent. The distribution of the γ counts in the sampling region is not consistent with that which would be expected on the basis ,of Danielsen's model (Danielsen, 1964) and lack of data from higher and lower levels make it difficult to distinguish between stratospheric air and tropospheric air on the basis of radioactivity alone. This radioactivity may be partly from the Chinese atomic explosion. Since the Chinese test was a low yield detonation, it seems unlikely that it penetrated the stratosphere. The 12-hour trajectories of air in the sampling region also give inconclusive evidence of a stratospheric extrusion. At 330k the average descent of the air was about 45 mb in 12 hours or 3 cm/sec. Stronger descending motions took place at 320k, where the vertical velocities were of the order of 90 mb in 12 hours. None of this air came directly from the stratosphere. The reasons for not tracing this air farther backwards are the scarcity of data near the Mexican border, where it probably entered the United States, and, secondly, the inaccuracy of the fast analysis technique.

## AERIAL SAMPLING OF RADON CONCENTRATION IN THE LOWER TROPOSPHERE

The vertical distribution of radon decay products was measured in the lower troposphere through the use of DC-6 aircraft belonging to the Research Flight Facility of ESSA. The observations were made on research flights in the general vicinity of Oklahoma City and were planned by the National Severe Storms Laboratory under the direction of Dr. T. Fujita. These flights were conducted on 12 separate days during the period April 27 through June 2, 1965. Samples were taken in both CP and MT air masses in weather conditions ranging from fair weather to the intense convection associated with a squall line.

Flight altitudes varied from 3700 to 13,800 feet. In conjunction with the sampling operation, sky conditions were photographed simultaneously from both sides of the aircraft at 5-second intervals.

Since all sampling operations occurred at or above 3700 feet, secular equilibrium was assumed between radon and its daughter products. Beta radiation was recorded and counted with equipment on loan to the project from N.R.L. Radon concentration was calculated using a method prescribed by Charles R. Hosier of ESSA. This involved 20-minute collections of radon daughter activity on IPC 1478 paper in the I-2A sampler. The radon concentration was defined as follows:

$$\text{Radon concentration } (P_c m^3) = \frac{\text{Total observed count}}{\text{Theoretical count from unit source concentration at equilibrium}} \times \frac{\text{Filter efficiency correction}}{\text{Air flow}}$$

Prominent features which were observed during the course of the operation are listed below: -

1. There was a decrease in concentration with an increase in altitude. On three separate days, flights were conducted at altitudes of 4000, 5000, and 6000 feet in both CP and MT air masses. All three days were characterized by large-scale vertical and horizontal movement of air. A plot of radon concentration vs. altitude clearly points out that at any given time concentration decreases with altitude. Exceptions are indicated below.
2. Preliminary investigation of large deviations, which were not due to changes in altitude indicated they were closely associated with change's in sky condition. A relatively smooth curve on the plot of radon concentration vs. altitude

showed an abrupt increase of great magnitude when the aircraft was flown into the immediate vicinity of cumulus cloud types.

3. Time of day was the largest single influence on the variation of concentration. At all altitudes sampled, a marked decrease in concentration occurred near 1300 LST and it reached a minimum at approximately 1500 LST. In all cases where data were available, a sharp increase in concentration occurred at approximately 1630 LST and a maximum was observed near 1800 LST. Here again sky condition photographs seemed to confirm the observation that large deviations from the expected distribution of radon concentration with height were associated with the atmospheric dynamics and composition which produce clouds.

4. Some large-scale deviations in concentration were observed which could not be explained by a significant change in altitude, the time of day, or a particular sky condition. A noteworthy situation occurred on a day which involved sampling on both sides of a weak ill-defined cold front. In a particular location radon was measured at 1245 LST and again at 1740 LST. Concentration after a 5-hour interval was only 13 percent of its original value. This probable exchange of air indicates the value of radon concentration as a natural tracer for vertical diffusion of the atmosphere.

5. Radon distribution on a clear day was measured by two separate aircraft simultaneously while flying at the same altitude about three miles apart. Since the operating environment was the same for both aircraft, the beta count from radon daughter products should have been the same. This was not the case, however; one aircraft consistently experienced a greater concentration than the other,.

More detailed analysis of these data is currently in progress. However, additional air sampling flights are required to confirm preliminary assumptions. Simultaneous air samples at several levels using identical measuring equipment would provide information for a profitable investigation of the natural dynamics of the atmosphere.

## CONCLUSIONS

The ratio of the gross beta radioactivity concentration in precipitation to that in an equal volume of air varied from approximately $10^5$ to $10^6$ in 16 showers investigated. This ratio is an indication of how very effective the precipitation process is in scrubbing the atmosphere.

The ratio of beta activity concentration in surface precipitation to that collected at cloud base by aircraft was 1.7 on May 26, 1965. This represents a scavenging of 40 percent of the radioactivity estimated to fall within the paths swept out by the drops. Evaporation accounts for less than half of the increase. However, samples taken on June 1, 1965, although under less controlled conditions, do not show an increase in the radioactivity of the rainwater between the cloud base and the ground. The data suggest that washout of radioactivity between the cloud base and ground may be considerable on some occasions and inconsequential in other cases.

A satisfactory system was developed for airborne sampling of water vapor for tritium analysis. Tritium analysis of most of the water samples remains to be done.

Flight samples of radioactivity were made in air that descended at a rate of 45 to 90 mb per 12 hours between 200 and 400 mb. The variability of the radioactivity in this air precluded determination of its sources. Some of the debris was from a low-yield Chinese explosion on May 14, which probably

did not penetrate into the stratosphere. The flight data also show more details in the flow and in the distribution of potential temperatures than the conventional widely-spaced radiosonde data, but due to differences in the evaluation of the various parameters, a comparison between the two can only be qualitative. The difference in time and location of the cross-sections in a time-dependent flow also makes this comparison more difficult. Furthermore, the flights did not extend to levels low enough to bring out any differences between the radioactive air and any other air which could be incorporated in these showers which were accompanied by relatively high concentrations of radioactivity in the rainwater. However, there is evidence that some air from the upper troposphere descended far enough to be incorporated in a local downdraft which extended to the surface. This rather small and sharp-edged parcel of radioactive air skirted the rain sampling network, and rain which subsequently fell through this air, apparently collected high concentrations of debris.

The problems of obtaining conclusive evidence about the air motions which transfer radioactive debris from the stratosphere into precipitating clouds and from there to the surface clearly lie in the incapability of achieving an appropriate location of the flight observations in space and time with respect to the events over a relatively small sampling network. Other problems are the cumbersome trajectory computations and the sparsity of surface and upper air data. Future analyses may be expedited to a considerable extent by computer programs which have recently become available. Serial radiosonde observations around the experiment area during the field project are, however, essential for this kind of study.

Preliminary results of the analyses of radon concentrations in the lower atmosphere on 12 separate days indicate a trend for the concentration

to decrease with height, for a diurnal minimum to occur in early afternoon followed by a peak in late afternoon, and for large variations in the vertical distribution to be associated with sky conditions, particularly in the presence of cumulus cloud types.

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# APPENDIX A

## LOG OF TRITIUM SAMPLING FLIGHTS

| Date | Max. Alt. Sampled (meters MSL) | Number of Samples Collected |
|---|---|---|
| 1-17-66 | 1520 (5 K ft.) | 1 |
| 4-15-66 | 3660 (12 " ) | 4 |
| 4-21-66 | 5180 (17 " ) | 4 |
| 4-29-66 | 3960 (13 " ) | 3 |
| 6-8-66 | 4880 (16 " ) | 3 |
| 6-16-66 | 5180 (17 " ) | 4 |
| 6-23-66 | 4570 (15 " ) | 4 |
| 6-30-66 | 5180 (17 " ) | 4 |
| 7-11-66 | 4270 (14 " ) | 4 |
| 7-26-66 | 2440 ( 8 " ) | 4 |
| 7-26-66 | 2440 ( 8 " ) | 4 |
| 7-26-66 | 2440 ( 8 " ) | 4 |
| 10-6-66 | 5490 (18 " ) | 4 |