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Sources of Ozone and Sulfate in Northeastern United States

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SUMMARY

Measurements of daily concentrations of ^{7}Be, ^{32}P and ozone at Whiteface Mountain, New York are reported for June, July and August of 1977 and 1978. Episodes of high daily ozone are observed to coincide with peak ^{7}Be and ^{32}P concentrations. Since these radionuclides and ozone are produced in the stratosphere, their simultaneous increase is taken to indicate the arrival of stratospheric air. However, ozone is also produced at ground level by photochemical reactions involving nitrogen oxides and olefin hydrocarbons. The amount of photochemically produced ozone is highly variable and difficult to quantify. Therefore, only when the ground level production of ozone is small can a comparison of cosmogenic nuclides and ozone quantitatively yield stratospheric ozone component. Such occasions, although rare, exist. For example, on June 15, 1977 at least 80% of the 50 ppbv (daily average) ozone observed originated in the stratosphere. Since this is a 24-hour average, stratospheric ozone could have been much larger for a given hourly period. Our data indicate substantial stratospheric contributions on numerous other occasions.

We more frequently must apply ^{7}Be and ^{32}P as tracers of stratospheric ozone at times when the stratospheric component is not predominant but substantial. The ^{7}Be and ^{32}P concentration measurements can be used to directly determine the accompanying stratospheric ozone if (1) the ^{7}Be (and/or ^{32}P)/O_3 mixing ratios in the upper atmosphere, and (2) mean residence time of O_3 is known. Since October 1977 we have determined the ^{7}Be concentrations in aerosols samples collected at 10-12 km by NASA's Global Atmospheric Sampling Program. The ^{7}Be concentrations varied from $\leq$ 0.1 to 5.8 pCi/kg air, whereas ozone concentrations ranged between 27 and 527 ppbv. The $^{7}Be/O_3$ mixing ratios, however, were similar in the upper troposphere and in the lower stratosphere. The similarity in the mixing ratios suggest significant exchange between the stratosphere and the troposphere. The subsidence of the upper atmospheric air will bring the stratospheric ^{7}Be and O_3 to ground level. Therefore, if the mean residence times and $^{7}Be/O_3$ ratios are known, ground level ^{7}Be concentration will yield the stratospheric ozone concentrations. For a first approximation we have assumed that ^{7}Be and O_3 have similar residence times. Estimated daily stratospheric ozone component vary from 10-40 ppbv.

Measurements of total syspended particulates (TSP), trace elements and sulfates were also carried out. Since TSP, trace elements and sulfate are produced only at ground level, their daily variations indicate the intensity of pollutant transport from urban/industrial centers to rural Whiteface Mountain. Episodic high concentrations of TSP, trace elements and sulfate were observed during summer months. Comparison of the elevated episodic concentrations with surface air mass trajectories show that the polluted air masses originated in the industrial Midwest.

1. INTRODUCTION

Most of the terrestrial ozone is produced in the stratosphere, the concentration peaking at about 25 km and monotonically decreasing to the tropopause. Transfer from the stratosphere to the troposphere occurs when the tropopause becomes vertical in the core of the jet stream, and then folds beneath the jet core. Reed and Danielsen (1959) showed that the folded structure could be identified by its large values of potential vorticity and used the term "tropopause folding" to describe the process. Danielsen and Mohnen (1977) have experimentally demonstrated that ozone rich stratospheric air is transported into the troposphere with each major cyclonic development. Interactions of cosmic rays with oxygen, nitrogen, and argon in the stratosphere (and to a much smaller degree in the troposphere) produce a variety of radionuclides. During the tropopause folding, along with ozone, these radionuclides are transported to the troposphere. Therefore, these cosmogenic radionuclides can be used as tracers of the stratospheric air.

Data obtained in this laboratory (Husain et al., 1977) and elsewhere (Reiter et al., 1975, 1977; Ludwick et al., 1976) have demonstrated that correlations between ^{7}Be and ozone exist, suggesting that at least at times ozone originated in the stratosphere. However, a considerable amount of ozone is also produced by photochemical reactions at ground level. The resolution of stratospheric O_3 from that produced in the troposphere poses a difficult task. The critical information essential in quantifying the stratospheric ozone at ground level are (1) a knowledge of $^{7}Be/O_3$ mixing ratios in the upper troposphere and the stratosphere, (2) variations of $^{7}Be/O_3$ mixing ratios with latitude and seasons, and (3) differing residence times of O_3 and ^{7}Be, and ^{32}P bearing aerosols. In addition to obtaining the essential data base for correlation of ^{7}Be, ^{32}P and ^{33}P, correlations with O_3, we have also made the measurements of $^{7}Be/O_3$ mixing ratios at 10-12 km and 21°-56° N.

Measurements of trace metals and sulfate, simultaneous with those of O_3, ^{7}Be, and ^{32}P, help identify the tropospheric component of an air mass. Such measurements were also carried out and are included in this report.

The work discussed here was conducted from July 1977 to October 1978 under U.S. Department of Energy Contract No. EE-77-S-02-4501.A000. A reprint and two preprints prepared from the work discussed in this study are attached as appendices.

1. Ozone Transport from Stratosphere to Troposphere, Geophys. Res. Lett. 4, 363-365, 1977 by L. Husain, P.E. Coffey, R.E. Meyers, and R.T. Cederwall.
   A reprint of this paper is attached as Appendix I.
2. Determination of Stratospheric Ozone at Ground Level Using $^{7}Be/O_3$ Mixing Ratios by V.A. Dutkiewicz and L. Husain has been submitted for publication to Geophys. Res. Lett.
   A preprint of this paper is attached as Appendix II.
3. Long Range Transport of Trace Elements by L. Husain and P. Samson has been accepted for publication in J. Geophys. Res.
   A preprint of this paper is attached as Appendix III.

2. EXPERIMENTAL

2.1 Sampling

From June through August 1977 the New York State Department of Environmental Conservation (DEC) collected daily samples of total suspended particulates (TSP) at Whiteface Mountain Field Station (WMFS) and at 3 other New York State sites, Schoharie (SCH), Sugar Hill (SH), and Holland (H) (see map in Figure 1). Particulates were collected on preweighed 25 cm x 20 cm Whatman 41 filter papers of known weight using four calibrated high-volume (Hi-vol) samplers run sequentially at 24-hour intervals (midnight to midnight). Sampled air volumes are calculated from running times and initial and final flow rates. Total air volumes averaged 2,000 standard cubic meters. Uncertainties in air volumes are estimated to be ±10%.

Our primary sampling site is WMFS which is in a remote location in the Adirondack Mountains at an elevation of 1.5 km. The summit weather observatory, shown in Figure 2, is maintained and operated by the Atmospheric Sciences Research Center of the State University of New York at Albany. In addition to standard meteorological data, hourly ozone levels are also recorded with a chemiluminescent analyzer (Coffey and Stasiuk, 1975). During 1977, the Hi-vol samplers were located on the upper floor balcony which can be seen in Figure 2. In 1978, a year-round Hi-vol sampling system was purchased with funds from this U.S. Department of Energy contract and funds from New York State Department of Health (NYSDH). Because of the severe winter weather conditions, the system had to be installed within the observatory building. Air is drawn in at the observatory roof via a 20-cm diameter insulated pipe shown in Figure 3. The incoming air is warmed by heating coils wrapped around the pipe to remove ice crystals which would damage the filters. Four Hi-vol samplers are attached to a sealed stainless steel sampling chamber as shown in Figure 4. By mounting the Hi-vols horizontally, condensed water is allowed to fall to the bottom of the chamber where it is drained to a sealed reservoir. In the early stages, there were problems in maintaining constant flow rates. In June 1978 constant mass flow controllers, bought by NYS Health Department funds, were installed on the Hi-vol samplers. Since then, the samplers have been running smoothly, with approximately 3% uncertainty in the volume of filtered air.

2.2 Analytical Procedures

Procedures used by the NYSDH for analysis of ^{7}Be, ^{32}P, and ^{33}P, and trace metal in particulate samples are described in this section. The general operation of the γ- and β-counting facilities are described in the instrumentation section of this report.

2.2.1 TSP and ^{7}Be

After TSP collection, each preweighed filter was equilibrated for 24 hours at 21°C and 50% relative humidity and reweighed to determine the amount of total suspended particulates (TSP) collected. The individual filters were then folded and analyzed spectrometrically on 7.6 cm x 7.6 cm NaI(Tl) γ-counters connected to 256-channel pulse height analyzers. The γ-spectra were reduced using a least-squares analysis computer code. The counting systems were calibrated by counting a ^{7}Be standard deposited on a Whatman 41 filter in the same geometry as the samples. To assure consistency in the computer fits, the energy calibration of the counting systems were checked daily with source standards. The uncertainties in ^{7}Be activities are generally about ±10%.

2.2.2 ^{32}P and ^{33}P

Three-quarters of each filter was used for radiochemical analysis of ^{32}P and ^{33}P. The filters were ashed, dissolved in concentrated HNO_3 and phosphate carrier added. Ammonium molybdophosphate was precipitated by adding $(NH_4)_2MoO_4$. The precipitate was dissolved in ammonium nitrate at pH 5 and an HCl-magnesia mixture added to precipitate $MgNH_4PO_4$. The precipitate was then dissolved in dilute HCl and run through a cation exchange column. The column effluent was evaporated to 10 ml and the magnesia mixture added to precipitate $MgNH_4PO_4$. The residue was washed and ignited at 950-1000°C to obtain magnesium pyrophosphate which was weighed and mounted on Whatman 42 filter paper. The chemical yield averaged around 80%.

The samples were β-counted several times over at least a four-week period on low-level β-counters. The samples collected in 1977 were counted on Beckman Wide-Beta systems which have backgrounds of 0.6 to 1 cpm. The activity of ^{32}P was obtained from a computer least-squares fit of the decay curve using a 14.4 day half-life after background subtraction and efficiency corrections.

Although ^{33}P is present at levels comparable to those of ^{32}P, the efficiency of the β-detector is significantly less for ^{33}P than for ^{32}P. Thus, ^{33}P contributes little to the total count rate. The decay curve fitting routine accounts for the remainder. Taking into account this possible interference, the uncertainties in the ^{32}P activity level is estimated to be ±15%.

In 1978, ^{32}P and ^{33}P counting was performed with either the Beckman Wide-Beta Counter or, whenever possible, with specially designed low-level proportional counters with background count rates in the range of 0.2 to 0.4 cpm. A description of these counters is given in the instrumentation section of this report. The ^{32}P and ^{33}P fractions were distinguished using total and absorber counting techniques. A 29 mg/cm^2 thick aluminum absorber was used to absorb the ^{33}P betas. With this absorber thickness, the ^{32}P transmission is 75%. ^{32}P standards prepared with the same carrier as the samples were used to establish ^{32}P counting efficiencies. ^{45}C (maximum β-energy of 257 Kev, half-life of 163 days) standards were used to calibrate detection for ^{33}P (maximum β-energy of 250 Kev). Decay curve analysis for ^{32}P and ^{33}P yielded the activities of the two radionuclides at the time of collection. Uncertainties for ^{32}P values >3 pCi are on the average ±10 to 15% whereas for ^{33}P the uncertainties are ±25 to 30%.

2.2.3 Activation Analysis

A 2.5-cm square was cut from each filter paper for neutron activation analysis of trace elements. Each irradiation generally included 14 samples, two blank filters, and three standards. The latter were prepared by evaporating solutions containing known amounts of various elements. Irradiations were performed in the high-flux beam reactor at Brookhaven National Laboratory. Irradiated samples were counted with a 45-cc Ge(Li) detector connected to a 4096-channel analyzer and magnetic tape readout system. The γ-spectra were reduced using the computer program BRUTAL (Gunnink et al., 1967).

The concentrations of individual elements were determined by comparing the activities of appropriate isotopes in the particulate samples with the activities of the same isotopes in the standards. Corrections were made for the average quantity of each element in the filter blanks. Blank concentrations (ng/cm^3) were generally low: Na, 240; K, 40; Sc, 0.0004; Cr, 2.5; Mn, 2.8; Fe, 90; Zn, 4.5; As, 0; Se, 1.6; Sr, 13; Sb, 0.5; Cs, 0; and Hg, 0.4. Thus with four exceptions (Na, Se, Br, and Hg) blank corrections did not significantly contribute to the uncertainties in the concentration measurements.

On the basis of estimated errors in the measurements of radionuclide concentrations, blank corrections, and standard preparations, the average uncertainties are estimated to be Na, 25%; Hg, 20%; K, 15%; and all other trace elements, about 10%.

2.3 Instrumentation

During both 1977 and 1978, three 7.6 cm x 7.6 cm NaI(Tl) counters have been available for γ-analysis of air filters, and counting turn-around time was never more than a few days. During 1977, delays of two weeks or more were encountered in receiving the samples. The delays occurred because the filters were weighed in a controlled environment which was not available at the NYSDH laboratories. Although the delay did not have a significant effect on ^{7}Be measurements, significant ^{32}P decay resulted. This problem was eliminated in July 1978, when a constant-humidity glove box with weighing facilities was acquired by NYSDH.

Two 45-cc GeLi detector systems are presently in operation and are used for γ-spectrometry. The detectors are each connected to 4096-channel multichannel analyzers with magnetic tape output. Background subtraction and peak integration is performed on the NYSDH Burroughs 6700 computer using the computer code BRUTAL.

(Gunnink, 1967). Both GeLi detectors have been calibrated for counting air filters using filters spiked with ^{7}Be standard.

In 1978, development work with USDOE funds was initiated to design and build a low-level β-spectrometer similar to the Geiger-Scintillator coincidence units developed by Tanaka et al. (1963, 1967). In the present design, a thin gas flow proportional counter is used to gate a NE-102 plastic scintillator crystal which is used for β-energy analysis. At the present time, the various components have been tested, but the prototype has not been completed. A prototype is expected to be in operation during the next calendar year.

During 1978, two gas-flow proportional counters operated in anticoincidence with a large plastic scintillator guard counter were built for ^{32}P and ^{33}P counting. The active region is cylindrical with dimensions of 6.4-mm thickness by 28-mm diameter. The counters are operated with P-10 gas and a 0.05-mm stainless steel center wire for charge collection. The background is reduced by placing the counters in one of the low-level counting rooms at the NYSHD. This facility has 23 cm of old steel shielding on all walls and ceiling. Background count rates are 0.2 to 0.4 cpm with muon cancellation rates of around 1.8 cpm. Values for ^{32}P counting efficiency are 34 and 31% while those for ^{33}P are 8 and 8.4%, respectively, for the two counters.

3. CORRELATION OF ^{7}Be, ^{32}P, ^{33}P WITH OZONE

3.1. Summer 1977

The ^{7}Be, ^{32}P, and O_3 concentrations determined daily at WMFS during June 15 through August 15, 1977, are given in Table 1. Daily ^{7}Be and ozone concentrations between June 15 and July 14 are also shown in Figure 5. Large ^{7}Be peaks (in excess of 200 pCi/KSCM (KSCM = 1,000 m^3) are observed on June 15-16, 24-25, 28, July 1, and 3. Ozone concentrations exhibit peaks on June 17-18, 25, 28, July 1 and 4.

Thus, one day delay in ozone peaks is frequently observed, as was reported for the 1975 data (Husain et al., 1977). We shall return to this point later in this report.

Large ^{7}Be concentrations, especially those on June 15-16, 28, and July 3, strongly suggest substantial stratospheric contribution at ground level. Figure 6 shows the daily ^{7}Be, ^{32}P and ozone determinations for July 15 thru August 15. A comparison of ^{7}Be data suggest declining ^{7}Be concentrations from June to July to August. This observation is in agreement with general belief that stratospheric-tropospheric exchange decreases as summer progresses. Peak ^{7}Be concentrations are observed on July 28, August 1 and 13, and possibly July 19. The asterisks in Figure 6 denote substantial precipitation on the given day, which means that the measured ^{7}Be or ^{32}P concentrations may only represent a lower limit as precipitation scavenges aerosols carrying ^{7}Be and ^{32}P. Correlation between ^{7}Be and ^{32}P appears excellent. In each case, when ^{7}Be concentration peaks, so does ^{32}P.

Between June 15 and August 15, 1977, we also measured ^{7}Be concentrations at three sites in the upstate New York. Since stratospheric transport is a long-range process under favorable circumstances, it should be possible to observe simultaneous stratospheric intrusions at several points in a given region. The ^{7}Be measurements at Sugar Hill, Holland, and Schoharie (see Figure 1 for locations) are given in Table 2, and shown in Figure 7 along with the data from WMFS. A notable feature of the data are the peak ^{7}Be concentrations, in excess of approximately 200 pCi/KSCM, observed at all sites on June 15-16, 27-28, July 2-3, 19-20 and 28-29. There are some days when peak ^{7}Be concentration is observed at one site but not the other.

In Figure 7B, an extended ^{7}Be episode is observed at Schoharie and Sugar Hill from July 15 thru 21. Unfortunately, data from Holland and WMFS are not available over part of this period. The high concentrations at all sites on the 20th and 21st indicate that this episode extended over all stations. The unusual feature here is that the episode lasted for seven days clearing out first at the western stations (Holland and Sugar Hill) and a day later at Schoharie and WMFS. On July 28 and 29 (Figure 7B) a ^{7}Be episode is observed at WMFS, and while the other sites also show elevated ^{7}Be concentrations, they are not as pronounced as WMFS. At Schoharie, Holland and Sugar Hill, the episode appears to extend from June 28 through August 1, while at WMFS a pronounced drop in activity is seen on July 29 and 31. On these two days, precipitation of 3.8 and 10.4 mm, respectively, was recorded at WMFS. The ^{7}Be concentrations on these daysc therefore, could have been reduced by precipitation scavenging. From the trends at the other stations, the ^{7}Be concentrations on July 30 should at least be equivalent to the concentration on July 29. A similar situation appears to have occurred on August 3, 4, and 5. ^{7}Be peaks are observed at Schoharie, Holland, and Sugar Hill but not at WMFS. On August 4 and 5 at WMFS, there was 8.1 and 7.9 mm of rain, respectively, thus, a significant amount of aerosol could have been removed. Precipitation data for the other stations have not been compiled at this time.

Some ^{7}Be episodes tend to last for several days, apparently due to the lack of significant precipitation. The foremost feature of the data shown in Figure 7 is the unusually high ^{7}Be concentrations observed on June 15-16. A concentration of around 500 pCi/KSCM is recorded at all sites although the peak is delayed one day at Schoharie and Sugar Hill. Air trajectory analysis for this period would be very informative, but are not presently available. This ^{7}Be episode is the highest recorded thus far at WMFS and is seen to extend over essentially all of upstate New York. In Table 1, ozone levels during this episode

are also observed to be very high. We shall return to a discussion of this episode later. However, these observations suggest that the influence of stratospheric intrusions can be observed at large distances, above and below the mixing layer.

The one-day delay in ozone peak reported earlier is also observed for July 15-August 15 period. One such episode is shown in Figure 8 for July 27-August 1, 1977. Here, 3-hour average ozone concentrations are shown with dark hours shaded. Daily ^{7}Be concentrations are indicated by the dashed lines and right hand scale. A ^{7}Be concentration of 310 pCi/KSCM is recorded on July 28, indicating the arrival of stratospheric air. O_3 concentration remains relatively constant on the 28th at 35 ppbv until around 9 p.m. The lack of a daytime O_3 peak indicates that there was no significant local photochemical O_3 production on this day. On July 29, the ^{7}Be concentration has decreased, but ozone has continued to increase to 80 ppbv around noon. During this period sulfate concentrations, measured on the same filters as ^{7}Be, were steadily increasing from a low of 0.5 $\mu g/m^3$ on July 27 to 10.7 $\mu g/m^3$ on July 29. Since 3.8 mm of rain fell on July 29, both the sulfate and ^{7}Be concentrations measured on that day may be low. Boundary layer (0.4-1.4 km altitude) trajectories calculated by Samson (1978) for this period are shown in Figure 9. On July 27th, ground level air is moving down from Canada. On July 28th and 29th, the trajectories pass through eastern Pennsylvania and New York's southern tier prior to arrival at WMFS. Thus, the air arriving at WMFS on July 29 passes near the New York and New Jersey metropolitan areas where pollutants including O_3 could be accumulated. On July 30, air reaching WMFS around midnight originated almost due south, with the trajectories moving more westward as the day progressed. Looking back to Figure 8, there is a pronounced O_3 peak between midnight and 3 a.m. on July 30 when the strongest influence from metropolitan New York and

New Jersey air is likely. A significant portion of the O_3 at WMFS on July 29 and 30 was probably transported from urban centers to the south (Coffey and Stasiuk, 1975; Cleveland et al., 1976; Hathorn and Walker, 1977; Wolff et al., 1977). As the weather system associated with stratospheric subsidence at WMFS moves through the transport of pollutants from urban centers to the south and southwest appear enhanced, further increasing O_3 concentrations and generating the apparent one-day delay between peak levels of stratospheric O_3 and ^7Be.

3.2 Summer 1978

In an effort to obtain ^7Be, ^{32}P and ^{33}P data during early spring, when maximum stratospheric-tropospheric exchange occurs, sampling for TSP was started in early March. The Hi-vol samplers were installed in the fourth floor of the observatory (see sampling description in section 2). However, because of large variations in line current as well as wind velocities, there were large uncertainties in the volume of the air sample. To circumvent this problem, four mass flow controllers were purchased with NYSDH funds in June and were installed on four Hi-vol samplers. The flow-controllers regulate the air volumes with an estimated error of about 3%. Although the ^7Be, ^{32}P and ^{33}P concentrations are listed in Table 3 beginning with March, we do not consider the concentration measurements as reliable until June.

Measured daily concentrations of O_3, ^7Be, ^{32}P and precipitation at WMFS shown for June, July, and August, 1978, respectively, in Figures 10-12 exhibit general similarities in the daily patterns of ^7Be, ^{32}P and O_3. During June, O_3 episodes occurred on June 11 and 12, 17-19, 21-22, and 26-27, where high ^7Be and ^{32}P concentrations are also observed. Because there were significant amounts of prolongs precipitation on June 19 and 20 (6 amd 16 mm, respectively) the ^7Be, ^{32}P, and O_3 minima on June 19 and 20 may simply be due to

precipitation scavenging. Thus, the episodes on 17-19 and 20-21 probably originated from the same stratospheric air mass. There were 17 days in June where there was at least some precipitation at WMFS. It is difficult to assess the effect precipitation has on ^{7}Be, ^{32}P and O_3 concentrations. For example, on June 13 about 12 mm of precipitation fell and ^{32}P and ^{7}Be show dramatic decreases to detection limit levels. The O_3 concentration also drops dramatically, indicating that the precipitation either destroyed some of the ozone or a new air mass was influencing WMFS on this day. At best, the ^{7}Be and ^{32}P concentrations should be taken as lower limits on days with prolonged precipitation. On July 5-7, 13-16, 19-23, and 25-27, O_3 episodes are observed at WMFS. Each of these episodes are also associated with periods of elevated ^{7}Be and ^{32}P concentration (Figure 11). One particularly large O_3 episode can be seen on July 19-23. On July 20 and 21, daily average ozone concentrations were over 100 ppbv and hourly averages reached peaks of 134 and 143 ppbv, respectively. ^{7}Be activity reached 300 pCi/KSCM on July 19, at least 3 times the normal background levels.

The ^{32}P, ^{7}Be, and O_3 measurements for August 1-15 are displayed in Figure 12. Results for the remainder of August are still pending. Concentrations on August 13 appear to be anomalous, since O_3 and ^{32}P concentrations are high, while the ^{7}Be concentration is near background. In fact, ^{7}Be concentrations are near background throughout the first half of August. Detailed cross checks of the ^{7}Be and ^{32}P data obtained during 1978 have not been completed at this time. These results should, therefore, be taken as preliminary, although major errors either in the ^{7}Be or ^{32}P concentrations are unlikely.

The observed strong correlation between ^{7}Be, ^{32}P and ozone concentrations clearly demonstrates that the downward flow of the upper atmospheric air significantly contributes to the ground level ozone. As pointed out earlier in the introduction,

quantification of the stratospheric ozone from the ground level ^{7}Be and ^{32}P measurements, a knowledge is required of (1) $^{7}Be/O_3$ and $^{32}P/O_3$ mixing ratios, (2) seasonal altitude and latitudinal variations of the mixing ratios, and (3) residence times of ozone, ^{7}Be, and ^{32}P bearing aerosols.

4. MEASUREMENT OF UPPER ATMOSPHERIC $^{7}Be/O_3$ MIXING RATIOS

The separation of ground level and stratospheric ozone can be accomplished using ^{7}Be to determine the stratospheric component. ^{7}Be concentration peaks at about 20 km (Lal and Peters, 1967) where nearly constant concentrations are maintained. Peak O_3 concentration occurs at about 25 km (Hering and Borden, 1967). Thus, for a given latitude the concentration ratio of $^{7}Be/O_3$ should remain relatively constant except for possible seasonal variations. In addition, Fabian and Pruchniewicz (1977) have determined the mean life-time of O_3 in the upper troposphere to be 50 days, essentially the same as the ^{7}Be half life. Thus, in the absence of differential chemical reductions, $^{7}Be/O_3$ mixing ratios should not under go large variations during tropospheric transport, even if the stratospheric air parcel has been significantly diluted with upper tropospheric air.

We have undertaken a study to measure $^{7}Be/O_3$ concentration ratios in the upper atmosphere. Part of the results from the first nine months of this study have recently been submitted for publication in Geophysical Research Letters. A preprint of this paper is attached to this report as Appendix II. In this section, we discuss certain aspects of this study not covered in the publication. The particulate samples for ^{7}Be analysis are collected by the NASA Global Atmospheric Sampling Program (GASP) (Perkins and Gustafsson, 1976; Nastrom, 1977, 1978) on commercial 747 jetliners. Sampling and analyses are discussed in Appendix II. The altitude and latitude distribution of the GASP samples are shown in Figure 13 and the ^{7}Be, O_3 and $^{7}Be/O_3$ concentration ratios as a function of location are summarized in Table 4.

O_3 concentrations range from 32 to 527 ppbv, indicating that both tropospheric and stratospheric air has been sampled. Above 37°N, no systematic difference in $^7Be/O_3$ mixing ratios is observed between samples with high O_3 ($\geq$ 225 ppbv) concentrations (probably stratospheric) and those with low O_3 ($\leq$ 150 ppbv) (probably tropospheric). This suggests that exchange across the tropopause is taking place and significant concentrations of stratospheric ^7Be and O_3 resides in the upper troposphere. Large-scale vertical circulation could bring this enriched air to ground level. Thus, stratospheric ozone may also reach the ground level by a mechanism other than tropopause folding.

For the period from April-June, the upper atmospheric $^7Be/O_3$ mixing ratio in the mid-latitudes varies from 0.75×10^{-2} to 1×10^{-2} pCi/kg ^7Be/ppbv O_3, or an average value which corresponds to 11.05 pCi/KSCM ^7Be/ppbv O_3. In the next section we apply this mixing ratio to 1977 ^7Be data to determine the stratospheric ozone reaching ground level during June-August.

5. DETERMINATION OF STRATOSPHERIC OZONE AT GROUND LEVEL USING UPPER ATMOSPHERIC $^7Be/O_3$ MIXING RATIOS

A paper on this subject has been submitted to Geophys. Res. Lett. and a preprint has been attached as Appendix II. In the discussion below, we have used this technique to deduce the daily stratospheric O_3 contribution at Whiteface Mountain. The daily stratospheric O_3 has been deduced for June 15-July 14, 1977 in the paper by Dutkiewicz and Husain (Appendix II). In particular, detailed discussion is given for the large ^7Be peak (500 pCi/KSCM) on June 15-16. We have carried out similar calculations for the stratospheric ozone at ground level for July 15-August 15, 1977 period.

In Figure 14, the daily variation in ^{7}Be, ^{32}P, and O_3 concentrations is shown for WMFS from July 15 thru August 15, 1977. Daily precipitation is shown as the upper shaded histogram. In Figure 14, ^{32}P and ^{7}Be concentrations show essentially the same general features, with the exception of August 4. In the lower shaded histogram, the stratospheric ozone component has been computed using the measured ^{7}Be concentrations and a $^{7}Be/O_3$ mixing ratio of 0.85×10^{-2}. pCi/kg/ppbv (11.05 pCi/KSCM/ppbv). In light of the earlier discussion of rain scavenging on July 30, stratospheric ozone equivalent to that on July 29 was assumed for July 30. Stratospheric ozone concentrations above 30 ppbv occur during two episodes. Since only lower limits could be determined for ^{7}Be concentrations on August 3 thru 5, the stratospheric O_3 concentrations indicated in Figure 14 for this period are probably low. From the ^{7}Be activity recorded at the other sites, stratospheric ozone of at least 20 ppbv is likely during this period.

The frequency of peak ^{7}Be episodes provides a measure of the impact of stratospheric ozone on ground level concentrations. The fraction of the number of days per month having ^{7}Be concentrations above 220 pCi/KSCM gives an estimate of the probability that the daily average stratospheric ozone component is above 20 ppbv. The results are summarized in Table 5 for all sites using ^{7}Be data for 1977. During June, the probability is higher at Holland and Schoharie, the western-most sites. July and August show about the same probability at all stations. Average values shown in Table 5 decrease from approximately 38% in June to about 6% in August.

Several points must be considered when interpreting these results. The effect of precipitation scavenging on ^{7}Be concentrations could be lowered considerably by

prolonged precipitation scavenging, the estimates of stratospheric ozone given here may only reflect lower limits. At the same time, O_3 concentrations could also be reduced somewhat by precipitation, thus making the results even more difficult to interpret. Daily ^7Be concentrations have been used to estimate the daily average O_3 concentration. Hourly O_3 values could be significantly higher if the upper atmospheric air envelopes the station rapidly. In addition, if the stratospheric air mass subsided into the boundary mixing layer and then underwent significant transport near ground level prior to collection of samples, considerable differential removal of aerosols and O_3 could take place. Thus, the upper atmospheric $^7Be/O_3$ ratios might no longer apply. Backward trajectory analysis for boundary layer air as well as isentropic trajectories should be used to determine where subsidence of stratospheric air mass occurred and when mixing with the boundary layer occurred. This would help determine the applicability of $^7Be/O_3$ concentrations ratios.

6. APPLICATION OF ISENTROPIC TRAJECTORIES

To help understand the relative contributions from the stratosphere and troposphere to ozone concentrations measured in the lower troposphere, certain meteorological analyses are useful in conjunction with measurements of cosmogenic tracers, ozone, TSP, and sulfates. These meteorological analyses involve the calculation of backward trajectories along isentropic surfaces (surfaces of constant potential temperature). The isentropic back trajectories are calculated (Danielsen, 1968), using wind, temperature, and humidity on constant pressure surfaces as analyzed by the National Meteorological Center. In conjunction with the esentropic trajectories, potential vorticities are calculated along the trajectories. Danielsen (1968) found that values of potential vorticity greater than 10^{-8} cm.sec.deg./gm. were indicative of the stratosphere. Although this approach is a rather simplified representation of the atmosphere, considering that

several nonadiabatic effects may be important at times, it does provide reasonable approximations. As an example of using these analyses to interpret measurements at Whiteface Mountain, we can consider a period in mid-July, 1975. Figure 15 shows back trajectories calculated for the 305° K isentropic surface terminating at Whiteface Mountain on the 16th and 17th of July. Table 6 gives the heights and potential vorticities along these trajectories. The back trajectory on the 16th has a large maximum value of potential vorticity at a high altitude (approximately 6 km), compared with that on the 17th where the maximum value of potential vorticity is rather low and occurs at a relatively low altitude (approximately 2 km). Both trajectories terminate at about 2 km, perhaps just above the summer daytime mixing height, below which isentropic trajectories are relatively indeterminate due to good mixing. From these two trajectories one can infer that air sampled on the 16th passed through the stratosphere, while air sampled on the 17th remained in the lower to mid-troposphere. This is in agreement with the ^{7}Be data which show a peak on July 16 and then a decline in ^{7}Be concentration on July 17 (Figure 1 of Husain et al., 1977, Appendix I).

From the data in Table 6, it is observed that the July 16 isentropic trajectory was below 3 km for only one day before reaching WMFS. On the other hand, the July 17 isentropic trajectory remained below 3 km for nine days. Hence, a greater tropospheric contribution on July 17 is to be expected compared to July 16. This conclusion is supported not only by ^{7}Be data, but also in trace element (Husain and Samson, 1978; Appendix III) and sulfate concentrations (Lioy et al., 1977). We must point out, however, that even when the isentropic trajectory originates at very high altitudes, it may still receive substantial tropospheric contribution during subsidence. Therefore, at times. high concentrations of cosmogenic nuclides and pollutants such as trace elements, and sulfate can be observed simultaneously. A discussion of surface trajectories on July 15-19 is given in

Husain and Samson (1978) (Appendix III). It will be particularly interesting to compare the isentropic trajectories for June 15-17, 1977, period when unusually large ^{7}Be peaks were observed. We expect that the isentropic trajectories for this period will be available in the near future.

7. LONG RANGE TRANSPORT OF TRACE ELEMENTS

Concentrations of 13 trace elements and total suspended particulates were measured for 24 consecutive days in July 1975 at WMFS. Elevated episodic concentrations were observed during a five-day period, July 16 through 20. Air trajectory calculations showed that stagnant air masses originating southwest of Whiteface Mountain were the primary sources of these elevated concentrations. During July 1976 the sampling network was extended to the other four stations in the sampling network plus a site in New Jersey. Air trajectory calculations for July 19 and 20, when peak concentrations of particulates and trace metals were observed at all five stations, strongly suggest that the particulates and trace metals were transported from sources southwest of New York State.

The results of this work prepared as a preprint (Appendix III) have been accepted for publication in the Journal of Geophysical Research.

Since this short paper was written to demonstrate the phenomena of long range transport for trace elements much of the data and detailed trajectories were not included. The conclusions deduced in that paper are strongly supported when the elemental data in Tables 7, 8, and 9 are considered. A summary paper including all of the elemental data and surface trajectory analyses is planned for publication in the near future.

8. SOURCES OF SULFATE

During summers of 1975, 1976, and 1977, daily sulfate concentrations have been measured by NYSDEC. A paper showing the long range transport of sulfate from the midwestern U.S. to the upstate New York has been published by Galvin et al. (1978). For June-August 1978, the NYSDH also measured daily sulfate concentrations using methyl thymol blue method (Lazrus et al., 1966) in TSP samples collected at WMFS (Table 10). The surface trajectory calculations to study the origin of various sulfate episodes for 1978 are not currently available. As discussed by Dutkiewicz and Husain (Appendix II), the sulfate concentration measurements play an important role in identifying tropospheric ozone components.

9. PERSONNEL

Dr. Vincent A. Dutkiewicz, B.S. - State University of New York at Stony Brook, 1971
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Dr. M. Jaffar, B.S. - Punjab University, 1964
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Mr. James B. Cummings, B.S. - Hamilton College, 1977
Miss Nesrin Ansary - Undergraduate at State University of New York at Albany
Miss Cicelia Urizar - Undergraduate at State University of New York at Albany

10. PRESENTATIONS AT SCIENTIFIC MEETINGS

1. L. Husain and P. Samson, Trace Elements and Meteorology: A Study of Trace Element Dynamics at Whiteface Mountain, New York, 4th Joint Conference on Sensing of Environmental Pollutants, New Orleans, LA, November 6-11, 1977.

2. L. Husain, Application of Nuclear Techniques in Atmospheric Transport Studies, 175th National Meeting of the American Chemical Society, Anaheim, March 12-17, 1978.

3. V.A. Dutkiewicz, L. Husain and E. Lezberg, Determination of $^{7}Be/O_3$ Mixing Ratios in the Stratosphere and Upper Troposphere, 176th National Meeting of the American Chemical Society, Miami, FL, September 10-15, 1978.

4. L. Husain, Pollutant Transport in Northeastern U.S., 14th Midwest Regional Meeting, Fayetteville, AR, October 26-27, 1978.

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Table 1 : Daily Measured Values of O_3, ^{7}Be and ^{32}P at Whiteface Mountain, New York - 1977

Date (1977)	Ozone (ppb)	^{7}Be[a] pCi/m^3 (x 10^{-3})	^{32}Pa pCi/m^3 (x 10^{-3})	$\frac{^{7}Be}{^{32}P}$	$\frac{^{7}Be}{O_3}$[b] (x 10^{-3})
June 15	51	502			9.8
16	50	506			10.1
17	71	275			3.9
18	70	99[c]			1.4
19	44	108[c]			2.4
20	41	69[c]			1.7
21	46	136			3.0
22	33	107			3.2
23	37	81			2.2
24	61	204			3.3
25	74	207			2.8
26	66	119			1.8
27	54	186			3.4
28	80	353			4.4
29	45	140			3.1
30	29	130			4.5
July 1	42	207			4.9
2	28	109			3.9
3	44	348			7.9
4	55	261			4.7
5	46	227			4.9
66	46	200[c]			4.3
7	47	80[c]			1.7
8	54	47			0.9
9	42	99			2.4

July	10	33	108			3.3
	11	51				
	12	60				
	13	52				
	14	43				
	15	57				
	16	67				
	17	62				
	18	50				
	19	40	296	5.43	54.5	7.4
	20	70	250	4.0	62.5	3.6
	21	51	271	3.85	70.4	5.3
	22	32	141	1.76	80.0	4.4
	23	32	157	2.87	54.7	4.9
	24	46	210	3.21	65.4	4.6
	25	42	176	2.57	68.5	4.2
	26	27				
	27	30	129	2.75	46.9	4.3
	28	45	323	5.36	60.3	7.2
	29	74	246	4.03	61.0	3.3
	30	57	80	1.31	61.2	1.4
	31	51	196	2.92	67.1	3.8
August	1	57	239	4.84	49.4	4.2
	2	38	172	3.85	44.7	4.5
	3	58	190[c]	2.90[c]	65.5	3.3
	4	71	175[c]	4.90[c]	35.7	2.5
	5	56	93[c]	2.41	38.6	1.7
	6	54	162	1.99	81.4	3.0

August	7	47	163	2.64	61.7	3.5
	8	56	104	2.86	36.4	1.8
	9	38				
	10	47				
	11	62	135[c]	2.75[c]	49.1	2.2
	12	41	170	2.90	58.5	4.1
	13	53	238			4.5
	14	41	127	2.48	51.2	3.1
	15	31	99	1.31	75.6	3.2

[a]Estimated uncertainties, see text.

[b]$^{7}Be/O_3$ units in pCi/m^3 air ppb ozone.

[c]Air volume not accurately known; lower limit value given.

Table 2: Daily measured values of ^{7}Be at Holland, Sugar Hill and Schoharie, New York

Date (1977)	Holland $^{7}Be^{a}$ pCi/m^3 $(x\ 10^{-3})$	Sugar Hill $^{7}Be^{a}$ pCi/m^3 $(x\ 10^{-3})$	Schoharie $^{7}Be^{a}$ pCi/m^3 $(x\ 10^{-3})$
June 15	---	302	271
16	590	419	448
17	512	364	353
18	179	231	222
19	---	309	246
20	336	304	224
21	246	283	239
22	263	276	167
23	---	161	127
24	340	231	210
25	168	244	212
26	241	233	238
27	264	304	232
28	235	327	272
29	231	242	194
30	235	207	160
July 1	148	252	230
2	418	263	322
3	327	260	226
4	363	289	---
5	216	193	211
6	131	185	214

July 7	73	77	321
8	62	20	51
9	117	143	162
10	142	164	175
11	128	198	207
12	---	290	125
13	---	106	197
14	---	240	181
15	---	279	365
16	---	284	252
17	---	301	244
18	---	190	210
19	---	261	346
20	318	275	202
21	210	157	360
22	138	108	92
23	130	165	149
24	179	181	205
25	176	201	232
26	136	187	176
27	120	168	119
28	232	225	153
29	230	219	220
30	248	234	248
31	220	234	248
August 1	150	226	232

August	2	118	164	154
	3	256	237	157
	4	---	288	336
	5	162	189	201
	6	176	115	164
	7	179	128	---
	8	169	167	166
	9	151	141	176
	10	156	158	153
	11	216	185	---
	12	194	178	---
	13	232	249	137
	14	180	---	---
	15	---	---	---

Table 3: Daily Measured Values of O_3, ^{7}Be, ^{32}P and ^{33}P at Whiteface Mountain, New York

Date (1978)	Ozone ppb	^{7}Be[a] pCi/m^3 ($\times 10^{-3}$)	^{32}P[a] pCi/m^3 ($\times 10^{-3}$)	^{33}P[a] pCi/m^3 ($\times 10^{-3}$)	$\frac{^{7}Be}{^{32}P}$	$\frac{^{33}P}{^{32}P}$	$\frac{^{7}Be}{O_3}$[b] ($\times 10^{-3}$)
March 21	51						
22	47	256					5.4
23	47	191					4.1
24	45	262	12.6[c]		21		5.8
28	43	84					1.9
29	41	116					2.8
30	45	226					5.0
31	48	178	6.3[c]		28		3.7
April 18	50	150	3.0[c]		50		3.0
19	57	52	0.9[c]		58		0.91
20	50	10					0.2
21	37	100	2.0[c]		50		2.7
May 9		34	3.3[c]				
10	46	106	1.7	1.2	63	0.71	2.3
11	49	163	3.4[c]		48		3.3
12	83	128	2.7[c]		47		1.5
16	52	123	1.4[c]		88		2.4
17	54	72	2.2		33		1.3
18	62	13	0.7		18		0.21
19	61	26	1.0		26		0.43
22		185	3.4	0.5	54	0.15	
23		193	3.7	3.5	52	0.94	
24		242	4.8	2.9	50	0.60	
25		232	5.2		45		

May 26	63	224	3.3	1.4	68	0.42	3.5
27	70	210	4.1	2.1	51	0.51	3.0
28	76	168	1.1	7.7	153	7.0	2.2
30	69	144					2.1
31	79	145					1.8
June 1	40	37					0.92
2	74	149	2.3	6.2	65	2.7	2.0
3	45						
4	40	203	2.5	3.1	81	1.2	5.1
5	43	203	1.1		184		4.7
6	48	296	3.0	6.3	99	2.1	6.2
7		77	$\leq$0.88				
8	62	43	$\leq$0.24				0.69
9	50						
10	43	258	1.4[c]		184		6.0
11	84	232	4.2		55		2.8
12	77	229	5.0	1.6	46	0.32	3.0
13	39	18	$\leq$0.25				0.46
14	34	110	$\leq$0.20				3.2
15	45	189	1.1		172		4.2
16	44	186	2.4	1.9	77	0.79	4.2
17	80	148	3.5	2.1	42	0.60	1.8
18	74	109	2.8	1.3	39	0.46	1.5
19	58	62					1.1
20	42	178	2.0	2.0	89	1.0	4.2
21	67	229	4.1	5.7	56	1.4	3.4
22	55						

June	23	37	110	3.0		37	3.0
	24	40	234	≤0.71			5.8
	25	45	178	4.1		43	3.9
	26	83	148	2.8	5.1	53	1.8	1.8
	27	68	152	3.0	1.8	51	0.6	2.2
	28	51	119	1.0	3.1	119	3.1	2.3
	29	41	105	≤0.41	0.12		.29	2.6
July	3	59	223	1.9		117		3.8
	6	75	202	3.2[c]		63		2.7
	7	88	184	2.5	1.5	74	0.6	2.1
	8	88	216	3.1		70		2.4
	9	56	43	2.1		20		0.77
	10	49	94	1.2		78		1.9
	11	26	48	≤0.07				1.8
	12	35	197	2.6	0.65	76	0.25	5.6
	13	59	237	4.0	3.2	59	0.8	4.0
	14	71	162	3.6	2.7	45	0.75	2.3
	15	71	146	3.5	1.2	42	0.34	2.0
	16	77	171	3.6		47		2.2
	17	50	124	2.0	3.4	62	1.7	2.5
	18	45	220	3.7	4.1	59	1.1	4.9
	19	73	228	3.1	9.3	73	3.0	3.1
	20	102	301	5.5	2.6	55	0.47	2.9
	21	104	211	4.5		47		2.0
	22	70	88	2.2[c]		40		1.2
	23	63	98	2.2[c]		44		1.5
	24	28	149	≤0.69				5.3

July	25	50	153	2.5		61	3.1
	26	63	174	3.4		51	2.8
	27	64	116	2.8		41	1.8
	28	35	53	$\leq$0.6			1.5
	29	37	9	1.5		6	0.2
	30	26	63	1.2[c]		52	2.4
	31	39	103	$\leq$0.91			2.6
August	1	51	99	1.0	2.7	99	2.7	1.9
	2	54	118	1.8		65	2.2
	3	67	70	2.8		25	1.0
	4	40	98	1.8	1.5	54	0.83	2.4
	5	45	129	1.9	3.9	68	2.0	2.9
	6	63	142	1.2		118	2.2
	7	56	170	2.5		68	3.0
	8	57	65	1.0	1.2	65	1.2	1.1
	9	65	97	1.1	2.4	88	2.2	1.5
	10	30	57	1.3		44	1.9
	11	44	157	2.6	3.1	60	1.2	3.6
	12	66	68	1.7		40	1.0
	13	71	144	6.1		24	2.0
	14	55	136	2.8	5.5	48	2.0	2.5
	15	52	168	3.9		43	3.2

[a]For estimated uncertainties, see text.

[b]$^{7}Be/O_3$ in units of pCi/m^3 air/ppb.

[c]$^{32,33}P$ calculated as ^{32}P.

Table 4: Location, Altitude, ^{7}Be, O_3, and $^{7}Be/O_3$ Mixing Ratios for GASP Samples

Date (1977)	Location: North Latitude (Deg.)	Location: Average Longitude (Deg.)	Altitude (km)	Fraction Total Air	^{7}Be[1] (pCi/kg)	Ozone[2] (ppbv)	Be^{7}/O_3 (pCi/kg/ppb) x 10^{-2}
10-6	48.5-45	145W	10.1	1.0	≤.4		
9	26.2-34.7	59E	11.9	1.0	.78		
12	27.5-18.3	87E	11.3	1.0	≤.4		
15	11.1-17.7	107E	10.1	1.0	≤.4		
18	42.3-50.5	58E	10.1	1.0	4.0		
21	34.1-27.1	130E	10.7	1.0	1.7	136	1.3
24	41.4-48.3	60W	10.7 11.3	.77 .23	5.6	259	2.1
25	43-46	78W	11.3	1.0	4.1		
11-11	38.6-41.8	109W	11.3	1.0	1.1	50	2.2
14	33-27.7	130W	10.7 11.9	.54 .46	1.4	87	1.7
(1978)							
1-6	37-33.5	130W	11.1	1.0	≤.5	59	≤0.8
9	36.8-35	120W	10.1	1.0	4.5	368	1.2
9	51.6-55.1	155W	10.1	1.0	6.2	276	2.2
15	42.1-41.2	80W	11.3	1.0	3.1	189	1.6
18	21.9-25.4	152W	12.2	1.0	≤1	57	≤1.8
21	34.8-40.1	107W	11.3	1.0	3.4	157	2.2
24	23-31.4	148W	10.4 11.6	.69 .31	.84	135	0.7
2-17	41.8-42.7	86W	10.8	1.0	4.5	384[3]	1.2

3-7	36.8-31.9	133W	11.0	1.0	≤.3	29	≤1.0
10	37.0-34.6	132W	11.0	1.0	.83	35[3]	2.3
16	22.0-28.7	144W	10.1 11.3	.30 .70	.60	71[3]	0.8
16	53.7-56.0	17W	10.4	1.0	5.8		
20	25.3-29.9	155W	11.9 9.5	.87 .13	≤.6	97[3]	≤0.5
4 18	41.0-38.3	94W	10.7 11.9	.13 .87	4.5	526	0.9
22	37.0-31.2	133W	11.0	1.0	≤.3	61	≤0.5
23	38.3-42.0	108W	11.3	1.0	2.0	236	0.8
25	22.6-30.6	147W	11.3	1.0	.2	91	0.2
28	36.9-32.0	132W	11.0	1.0	.2	79	0.3
29	34.1-38.4	133W	11.3	1.0	1.7	142[3]	1.2
5-18	36-25	149W	10.1 10.8	.5 .5	.50	32	1.5
18	21-28	143W	11.6	1.0	.44	104	0.4
24	38-42	109W	11.3	1.0	3.7	341	1.1
27	38-42	109W	11.3 11.9	.9 .1	2.2	227	1.0
30	23-21	147W	11.3	1.0	.3	63	0.5
6-8	22-29	144W	11.3	1.0	≤.2	43	≤0.5
11	40-41	86W	11.9	1.0	.53	70	0.8
14	41	86W	10.7 11.9	.5 .5	.73	130	0.6
17	38-35	130W	10.7	1.0	3.8	432	0.9
20	40-41	80W	11.9 9.6	.75 .25	.83	140	0.6

[1]Errors ±10% for activity > 2.0 pCi/kg otherwise ±20%.

[2]Errors ±5%.

[3]Cabin ozone levels used to estimate outside ozone uncertainties ±20%.

Table 5

Daily Probability of Upper Atmospheric Ozone $\geq$ 20 ppbv[1](%)

	June[2]	July	August[3]
Whiteface Mountain Summit	25	23	0
Schoharie	25	19	10
Holland	46	17	8
Sugar Hill	56	29	8
AVERAGE	38	22	6

[1]Based on 1977 daily average ^{7}Be activity.

[2]Based on 15 days sampling.

[3]Based on $\sim$ 10 days sampling.

Table 6. Heights and Potential Vorticity
Along Isentropic Back Trajectories

Trajectory No. 1[1]			Trajectory No. 2[1]		
Days[2]	Height[3]	Pot. Vort.[3]	Days[2]	Height[3]	Pot. Vort.[3]
0	2225	.43	0	1917	.39
1	2909	.50	1	2238	.42
2	3239	.60	2	2564	.43
3	4095	.56	3	2610	.37
4	4344	.62	4	2554	.33
5	4426	.88	5	2450	.33
6	5033	.85	6	2521	.28
7	4395	.74	7	2623	.32
8	5609	1.52	8	2255	.32
9	6418	2.60	9	2423	.34
10	6078	2.50	10	3431	.48

[1]Trajectories are calculated along the 305° K isentropic surface.

[2]Days are days prior to arrival at Whiteface Mountain, on July 16, 1975, and July 17, 1975, for trajectories No. 1 and 2, respectively.

[3]Units of height are meters and units of potential vorticity are 10^{-8} cm.sec.deg./gm.

Table 7. Elemental Concentrations in Air Particulates at Whiteface Mountain, New York, July 1975

Date	Suspended Particulates ($\mu g/m^3$)	Concentrations (ng/m^3 air)													
		Na	K	Sc	Cr	Mn	Fe	Cu	Zn	As	Se	Br	Sb	Cs	Hg
July 1	34	144	168	0.070	2.5	11.1	310	195	12	N.D.[1]	N.D.	6.9	N.D.	N.D.	1.25
2	51	154	293	0.113	5.8	22	526	75	100	2.7	0.82	15.1	0.42	0.080	0.64
3	38	39	114	0.030	2.9	3.6	111	101	18	0.9	0.24	7.8	0.17	0.033	0.62
4	33	88	92	0.039	2.9	5.9	166	127	24	0.7	0.24	6.1	0.11	0.034	0.93
5	36	102	123	0.039	1.7	10.9	356	291	16	4.1	0.70	14	0.41	0.024	0.79
6	41	100	151	0.059	2.8	8.9	314	257	25	2.3	0.92	26	0.63	0.059	0.39
7	47	153	151	0.062	4.2	7.9	275	350	31	2.5	0.92	34	2.1	0.057	0.42
8	54	188	148	0.090	3.4	10.6	295	217	61	3.8	1.84	21	1.6	0.096	0.34
9	44	52	84	0.057	3.2	8.6	237	33	23	2.6	1.16	12	0.50	0.032	0.48
10	34	53	90	0.039	2.4	5.2	200	284	14	0.6	0.24	8.8	0.12	0.038	1.46
11	44	85	116	0.038	2.8	6.2	162	235	18	1.3	0.70	12	0.27	0.044	0.64
12	42	72	96	0.039	3.37	8.5	206	227	25	1.7	0.70	13	0.52	0.020	0.34
13	33	27	38	0.015	5.47	5.6	164	75	209[2]	N.D.	0.20	7.2	0.30	0.020	N.D.
14	24	15	24	0.0057	3.60	1.3	42	137	7	1.0	0.14	5.3	0.28	0.024	0.27
15	34	37	62	0.021	4.9	7.0	187	220	20	1.1	0.79	13	N.D.	0.048	0.60
16	40	53	111	0.071	2.6	10.6	229	200	24	1.4	1.28	14	1.45	0.139	0.48
17	62	96	229	0.15	8.1	32	627	106	56	5.1	4.6	18	1.38	0.280	0.008
18	81	123	253	0.22	12.9	32	841	146	59	4.1	5.0	15	2.14	0.220	N.D.
19	69	155	250	0.15	19.2	17	591	304	42	3.0	1.86	12	N.D.	0.250	N.D.
20	32	19	56	0.034	2.4	3.1	132	189	8	N.D.	N.D.	6.5	N.D.	0.098	0.075
21	22	15	52	0.020	2.31	2.5	98	20	19	0.5	0.59	2.8	N.D.	0.040	0.79
22	24	101	89	0.024	3.5	3.2	129	180	15	2.8	N.D.	6.5	N.D.	N.D.	N.D.
23	35	71	151	0.049	5.4	9.7	261	321	20	1.6	0.42	13	3.88	0.068	0.17
24	41	58	116	0.093	20.3	16.0	629	120	34	1.7	0.95	9	1.72	0.085	0.12
Average	41	83	127	0.064	5.4	10.4	295	188	37	2.2	1.16	13	1.0	0.081	0.54

[1] Not detected
[2] Contamination suspected

TABLE 8. Simultaneous Measurements of Certain Trace Elements at Five Stations in New York State and New Jersey, July 1976

Site*	Date	Suspended Particulates ($\mu g/m^3$)	Concentrations (ng/m^3)								
			Na	K	Sc	Mn	Fe	Zn	As	Br	Sb
White-face Mountain	July 17	31	24	27	.008	3.87	191	12	1.04	2.8	.16
	18	53	71	104	.028	10.5	315	58	2.67	41	.68
	19	41		104	.032	27.1	352	30	2.07	31	.44
	20	74	354	315	.090	26.4	802	87	3.23	41	1.26
	21	43	470	709	.008	4.67	96	7	0.34	27	.09
	22	45	59	70	.022	7.36	201	15	0.28	0.7	.16
	23	63	154	185	.030	6.80	284	38	0.49	16	.70
	24	49	59	51	.025	3.1	169	11	0.55	15	.31
High Point	July 17	37	56	70	.026	6.16	147	18	1.64	53	1.37
	17	37	39	76	.022	3.8	156	18	1.80	74	1.20
	18	35	58	101	.047	8.4	247	12	0.98	68	.56
	19	29	97	134	.065	13.7	355	52	1.64	62	.79
	20	75	203	254	.157	27.2	464	297	4.17	128	2.44
	21	55	126	144	.078	13.0	429	382	3.22	43	N.O.
	21	55	101	144	.081	14.0	524	376	3.50	64	1.30
	22	41	258	187	.065	22.1	342	62	1.04	125	2.10
	23	29	295	101	.026	5.2	163	27	0.90	130	2.50
	24	38	40	79	.028	7.1	194	24	1.9	55	1.40
	25	31	39	90	.031	4.4	170	7	0.70	77	0.14
Schoharie	July 17	30	77	113	.028	6.8	222	16	1.94	71	.53
	18	28	69	138	.041	10.7	280	13	1.14	120	.39
	19	30	71	127	.042	8.9	277	15	1.14	187	.67
	20	57	87	229	.130	18.7	674	42	2.66	44	.96
	21	56	81	163	.078	18.0	605	53	3.43	39	1.14
	22	32	58	140	.035	16.3	202	18	0.52	33	.64
	23	32	90	107	.034	5.5	202	24	0.85	82	1.50
	24	39	47	105	.030	6.2	218	15	2.00	44	0.99
	25	22	48	118	.036	4.4	213	10	0.40	81	N.O.

Holland	July	17	26	70	118	.036	10.5	293	20	1.32	86	.28
		18	64		207	.076	28.0	716	31	2.23	82	.71
		19	55	123	197	.126	39.0	1079	31	2.30	75	1.56
		20	76	155	339	.210	46.0	1773	80	5.11	74	1.74
		21	46	88	185	.067	16.0	762	34	2.01	135	1.32
		22	46	65	164	.072	13.0	628	20	0.99	189	.93
		23	46	73	133	.052	13.0	514	51	2.50	82	.81
		24	41	55	133	.052	17.0	632	32	1.00	222	.72
		25	29	49	136	.044	9.9	404	17	1.80	365	.51
Albany	July	17	27	121	108	.026	6.6	239	22	1.66	146	1.80
		18	22		229	.042	12.5	371	15	1.22	146	.60
		19	42	193	193	.085	15.0	633	17	1.52	143	2.0
		20	57	134	372	.129	21.9	773	65	2.32	128	2.2
		21	51	118	208	.073	14.9	478	72	2.58	231	2.2
		22	41	152	235	.077	37.0	663	27	N.O.	292	1.4
		23	40	430	237	.078	10.0	551	72	1.20	313	1.7
		24	33	80	111	.029	6.3	248	20	1.30	295	1.0
		25	20	69	87	.038	5.0	274	7	N.O.	182	0.14

*High Point is in New Jersey. The rest are in upstate New York.
N.O. means not observed.

Whiteface Mountain, New York, during June and July, 1977

Date	Concentrations (ng/m^3 air)[1]											
	Na	K	Sc	Cr	Mn	Fe	Zn	As	Se[2]	Br	Sb	Cs
June 16	80	143	0.034	1.67	8.1	237	33	1.15	N.D.	10.7	N.D.	N.D.
17	88	192	0.059	6.9	19.8	450	93	3.36	3.40	35	2.94	0.13
18	156	146	0.043	33	11.6	390	27	2.02	1.03	14	N.D.	0.077
19	43	280	0.038	14.2	57	607	28	0.07	0.32	4.4	N.D.	N.D.
20	21	68	0.010	7.0	77	117	21	0.56	N.D.	2.1		N.D.
21	68	140	0.016	5.4	44	164	29	0.53	0.30	5.0	0.17	1.29
22	194	580	0.010	3.3	42	87	91	2.31	1.19	7.6	0.33	N.D.
23	537	151	0.017	37	12.6	333	60	N.D.	0.30	7.2	0.27	0.03
24	163	327	0.081	9.6	18.3	650	39	2.97	2.53	36	1.35	0.15
25	32	73	0.006	1.88	3.5	70	25	0.97	0.79	8.5	N.D.	0.25
26	36	85	0.005	3.3	13	105	27	1.18	0.75	0.7	N.D.	N.D.
27	94	180	0.018	7.9	18	395	175	2.63	1.54	19	N.D.	N.D.
28	106	238	0.067	6.6	12.6	515	313	3.58	3.10	39	2.50	0.15
29	34	115	0.025	7.1	10.5	298	44	0.98	0.69	13	2.12	0.05
30	29	109	0.019	1.80	12.5	225	96	0.53	0.30	3.7	N.D.	0.30
July 1	91	215	0.048	2.4	13.2	365	40	1.31	1.51	22	N.D.	0.04
19	75	222	0.062	3.1	16	474	41	2.01	2.47	23	2.53	0.13
20	84	137	0.042	1.90	9.7	335	37	N.D.	0.92	18	N.D.	N.D.
21	92	154	0.040	2.4	10.4	380	44	1.84	1.23	21	N.D.	0.07
23	59	80	0.021	0.71	5.5	191	18	1.50	0.21	13	N.D.	N.D.
24	82	207	0.075	4.5	19	548	42	2.52	1.99	45	N.D.	0.21
25	63	99	0.034	12.3	9.6	354	50	2.02	1.20	15	N.D.	0.15
27	49	55	0.011	0.44	3.1	106	12	0.99	0.27	19	N.D.	N.D.
28	66	104	0.029	1.01	6.7	237	25	N.D.	0.42	24	N.D.	0.05
29	171	213	0.062	9.2	22	674	348	1.75	1.60	61	3.16	N.D.
30	114	71	0.004	3.5	20	135	34	1.37	0.98	20	1.59	0.03
Average	101	169	0.034	7.2	19.1	325	47	1.57	1.21	18.7	1.69	0.19

[1]For estimated uncertainties, see text.

[2]N.D. means not detected.

Table 10. Sulfate Concentrations at Whiteface Mountain, New York during June, July, and August, 1978.

Date	SO_4 ($\mu g/m^3$)	Date	SO_4 ($\mu g/m^3$)	Date	SO_4 ($\mu g/m^3$)
June 15	0	July 7	19.2	July 29	.64
16	.32	8	27.2	30	.32
17	11.9	9	3.31	31	1.78
18	10.7	10	7.41	August 1	6.14
19	3.68	11	.66	2	13.7
20	1.30	12	.96	3	16.1
21	7.24	13	3.55	4	.96
22	no sample	14	19.0	5	6.59
23	2.37	15	17.2	6	4.88
24	1.63	16	13.4	7	26.7
25	2.58	17	8.22	8	11.4
26	17.1	18	2.58	9	9.63
27	16.0	19	7.49	10	.99
28	2.92	20	35.1	11	2.30
29	1.75	21	.43	12	10.6
30	no sample	22	11.4	13	12.2
July 1	no sample	23	13.1	14	8.22
2	no sample	24	.33	15	.65
3	1.07	25	3.24	16	20.5
4	no sample	26	.32	17	.33
5	no sample	27	8.84	18	20.3
6	8.30	28	.66	19	.65

Figure Captions

Figure 1: New York State map showing air sampling sites.

Figure 2: The Atmospheric Science Research Center Observatory at Whiteface Mountain Summit. The air inlet port for ozone measurements can be seen near the upper left hand corner of the center third floor window. Prior to 1978 Hi-vol samplers were located on the balcony above third floor. The year-round Hi-vol sampling system installed in 1978 is located in the upper floor observatory with the air inlet mounted through the roof.

Figure 3: The 20-cm diameter heated air inlet for the Hi-vol sampling system used at Whiteface Mountain Summit. The air inlet is located on the roof of the weather observatory pointing southeast.

Figure 4: The year-round Hi-vol air sampling system presently in use at Whiteface Mountain Summit. The air intake is seen coming down from ceiling. It is insulated and heated to remove moisture and ice crystals prior to sampling. Water is collected at the bottom of sampling box and drained to a storage pan which is not shown. Each Hi-vol is equipped with a constant mass flow controller calibrated to maintain the sampling air flow at 40 standard cubic feet per minute.

Figure 5: Daily variations in ^{7}Be and O_3 for June 15-July 15, 1977, at Whiteface Mountain Summit. Numbers in figure indicate selected dates.

Figure 6: Daily ^{7}Be, ^{32}P, and O_3 for the period July 15-August 15, 1977, at Whiteface Mountain Summit. The ^{7}Be and ^{32}P activity on starred days represent lower limits due to precipitation. Numbers in figure indicate selected dates.

Figure 7: Daily concentrations of ^{7}Be for the period June 15-July 15 (A) and July 15-August 15 (B) for Whiteface Mountain, Schoharie, Sugar Hill and Holland, New York. (See Figure 1 for site locations.) Numbers in figure indicate selected dates.

Figure 8: Three-hour average O_3 and daily ^{7}Be for July 27-August 1, 1977, at Whiteface Mountain Summit. Dark hours have been shaded.

Figure 9: Boundary layer (0.4-1.4 km) backward trajectories for air arriving at Whiteface Mountain at 2 a.m. (——), 8 a.m. (---), 2 p.m. (-·-), and 8 p.m. (···) on July 27-30, 1977. Each arrow head represents 12 hours.

Figure 10: Daily O_3, ^{7}Be, ^{32}P and precipitation for June, 1978, at Whiteface Mountain Summit.

Figure 11: Same as Figure 10 for July 1978.

Figure 12: Same as Figure 10 for August 1978.

Figure 13: Altitude and latitude distributions of Global Atmospheric Sampling Program samples analyzed for ^{7}Be activity.

Figure 14: Daily precipitation, ^{32}P, ^{7}Be, and O_3 for Whiteface Mountain Summit July 15-August 15, 1977. The lower shaded O_3 histogram shows the inferred stratospheric component as explained in the text. The ^{7}Be and ^{32}P activity on starred days represent lower limits due to precipitation. Numbers in figure indicate selected dates.

Figure 15: Backward isentropic trajectories along 305°K surfaces for air arriving at Whiteface Mountain on July 16 (——) and 17 (---), 1975. Numbers indicate day prior to arrival at Whiteface Mountain. Altitude and potential vorticity for each day is given in Table 6.

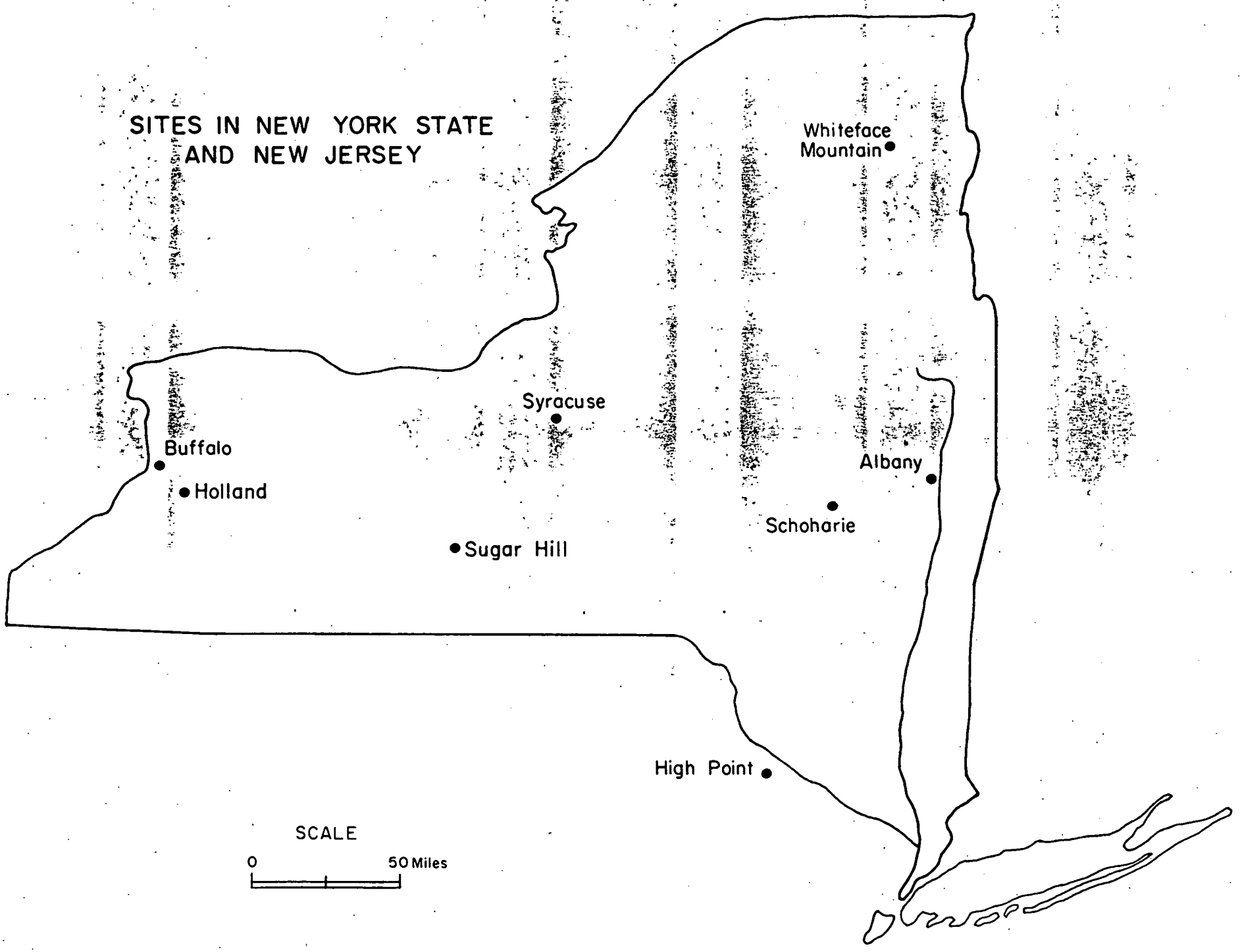

SITES IN NEW YORK STATE
AND NEW JERSEY
Whiteface
Mountain
Syracuse
Buffalo
Holland
Albany
Schoharie
Sugar Hill
High Point
SCALE
0
50 Miles

FIGURE 1

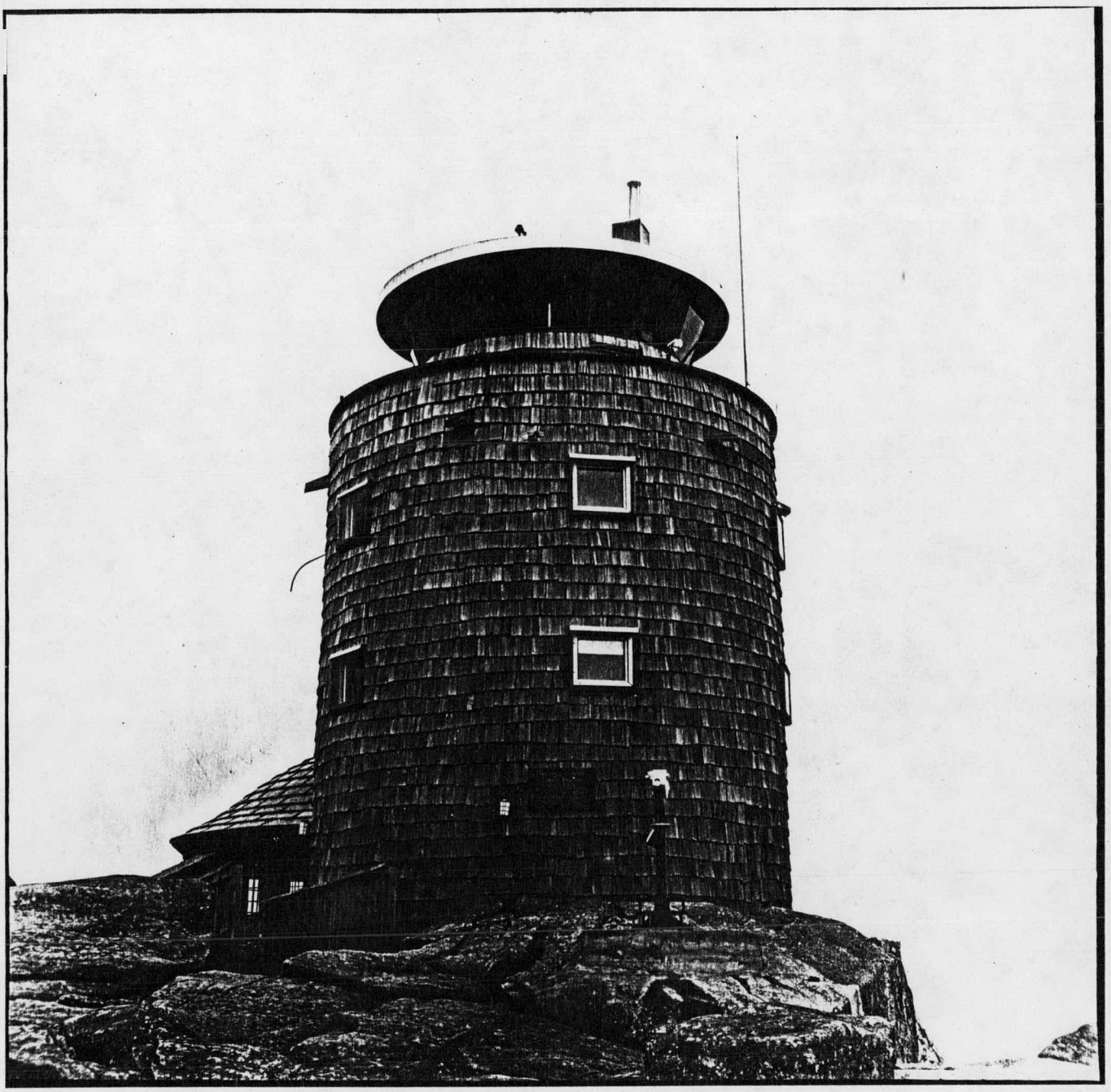


FIGURE 2

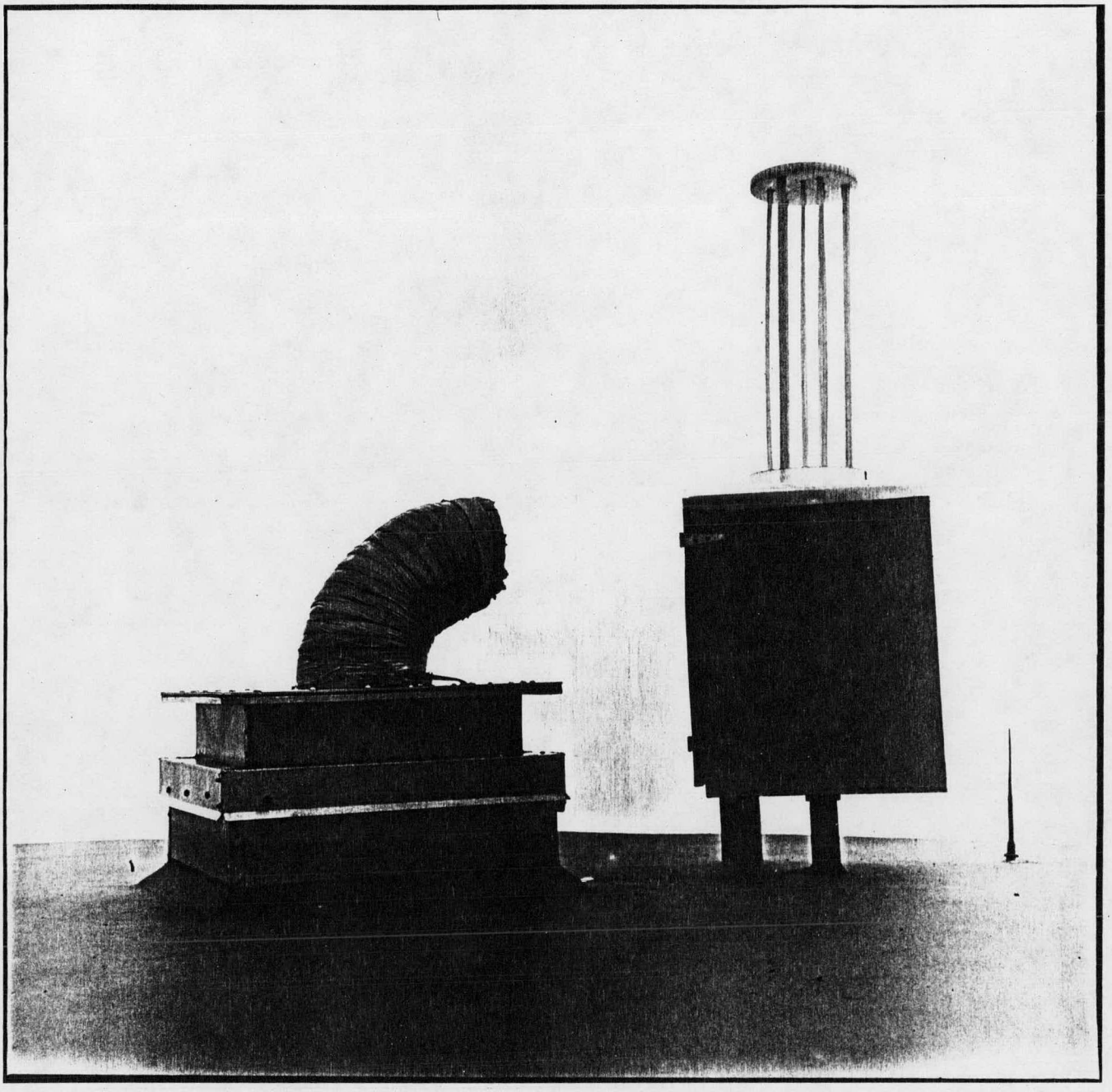


FIGURE 3

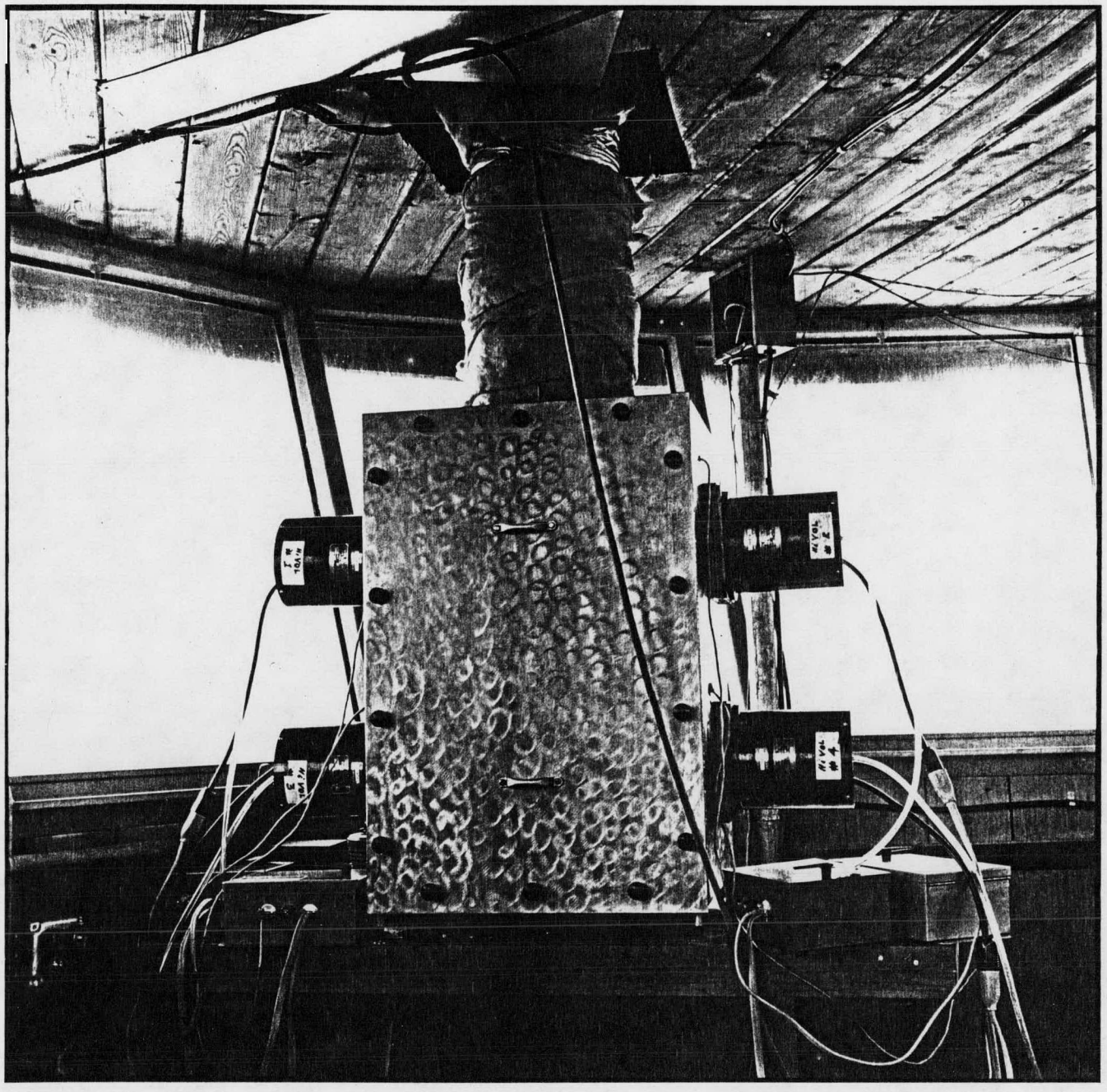


FIGURE 4

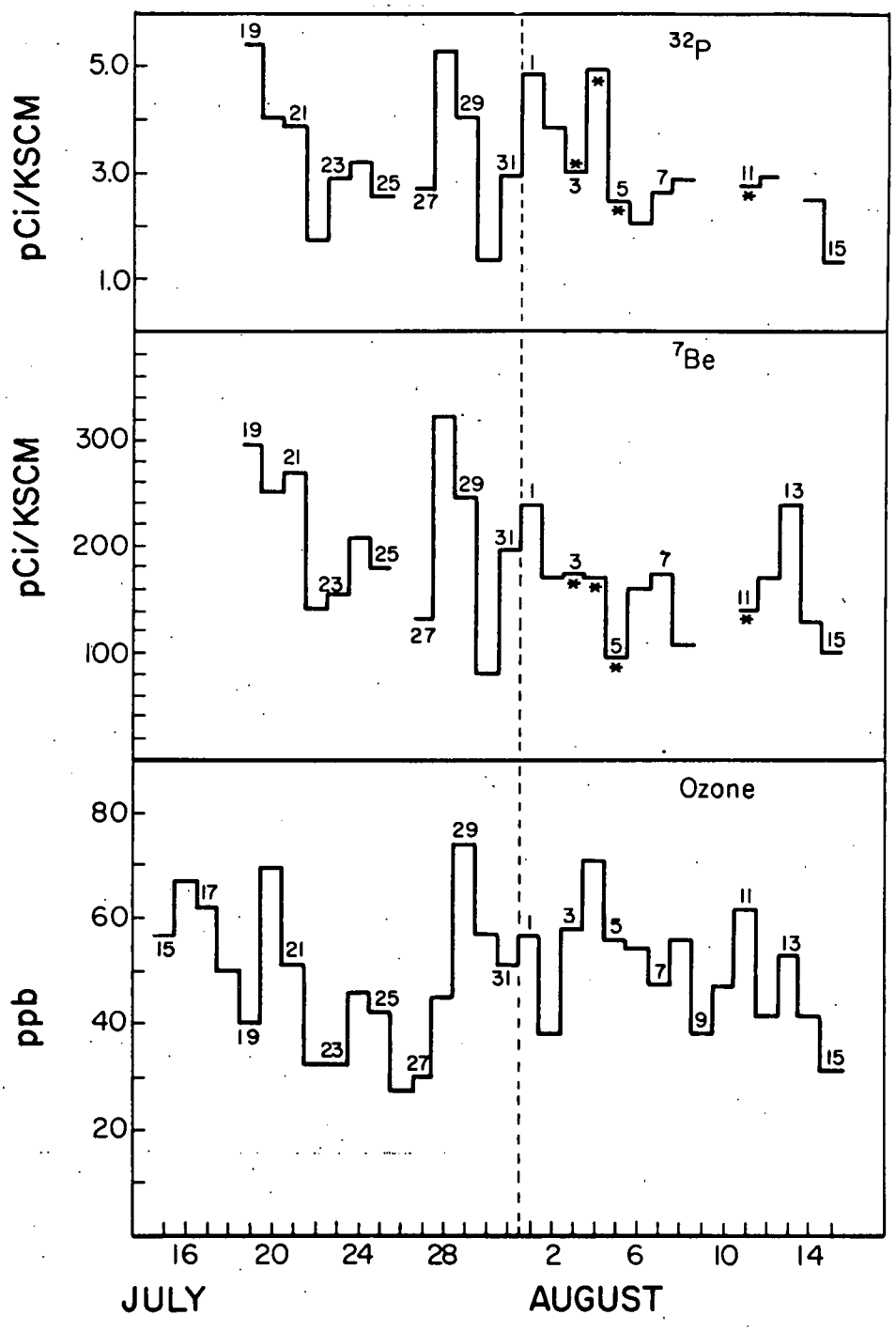

32P
pCi/KSCM
5.0
3.0
1.0
7Be
pCi/KSCM
300
200
100
Ozone
ppb
80
60
40
20
16
20
24
28
2
6
10
14
JULY
AUGUST

FIGURE 6

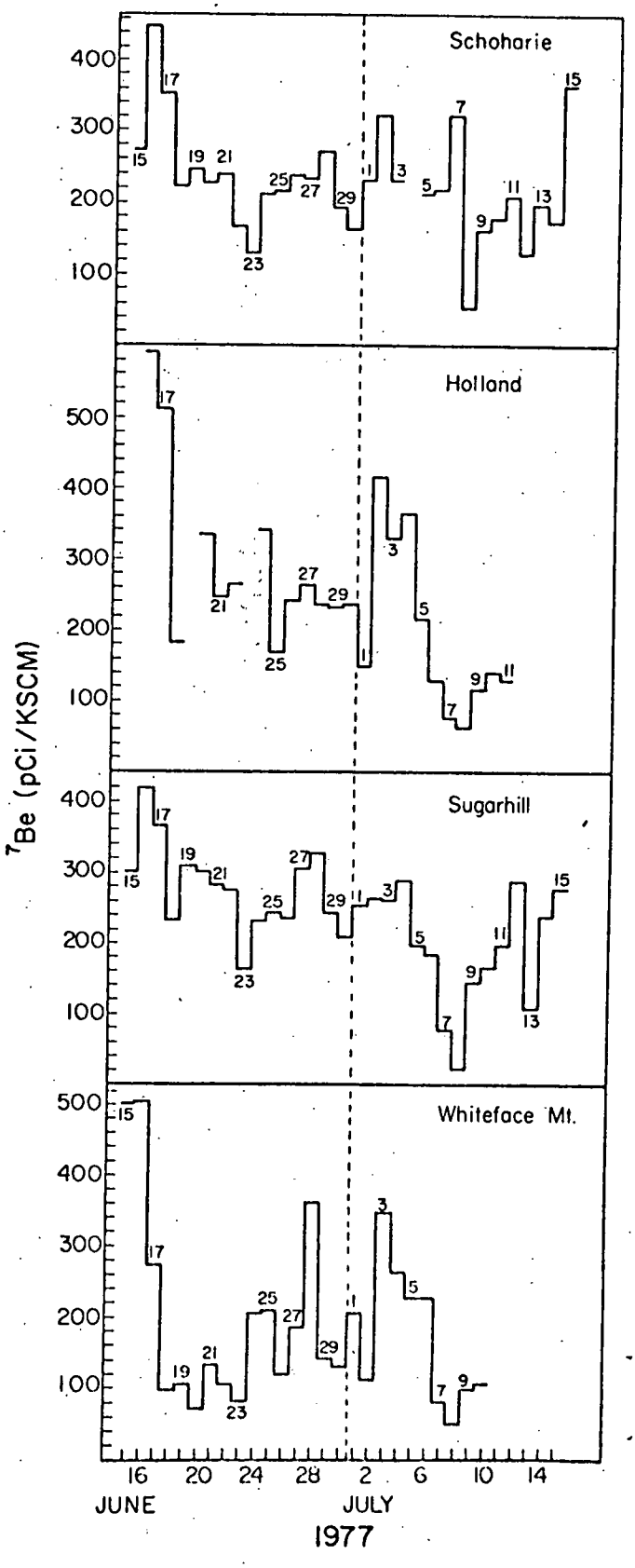


FIGURE 7A

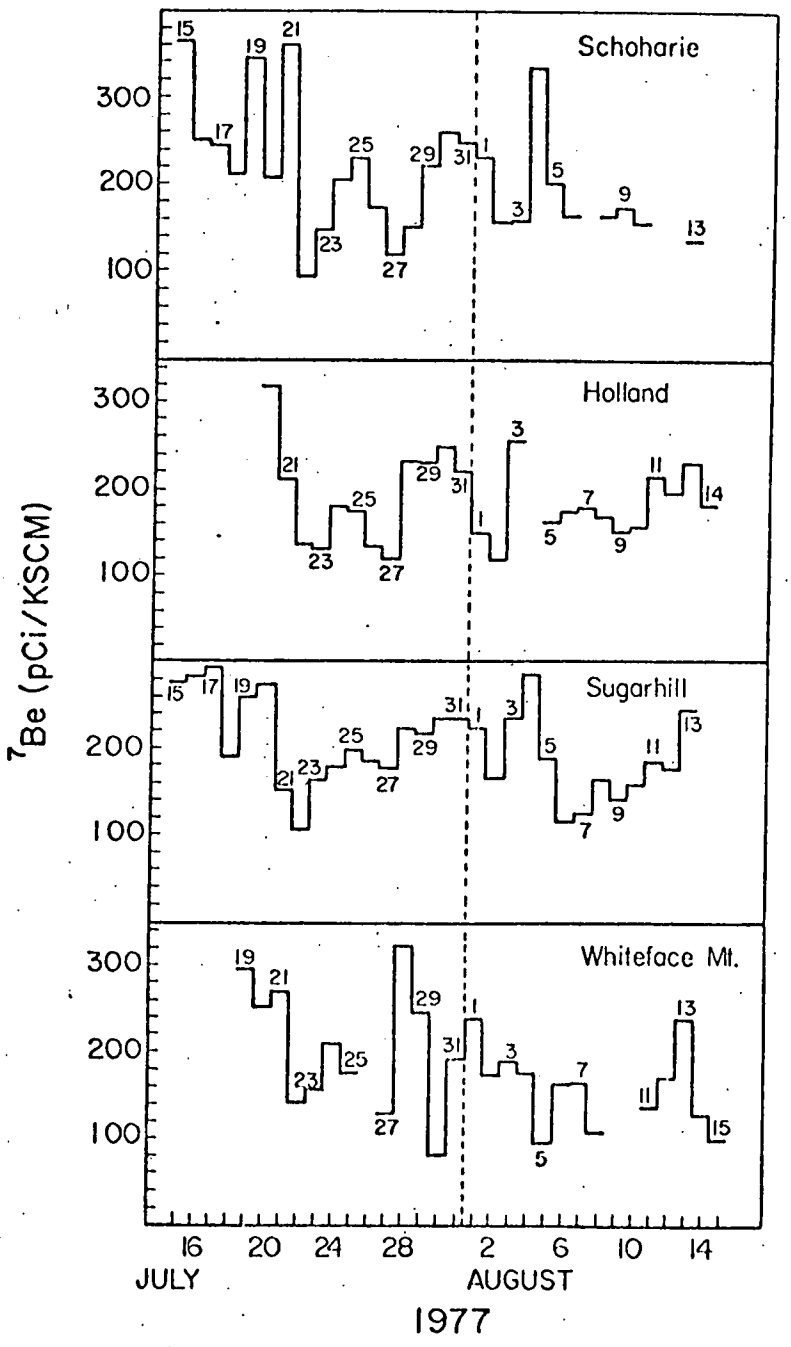


FIGURE 7B

FIGURE 7

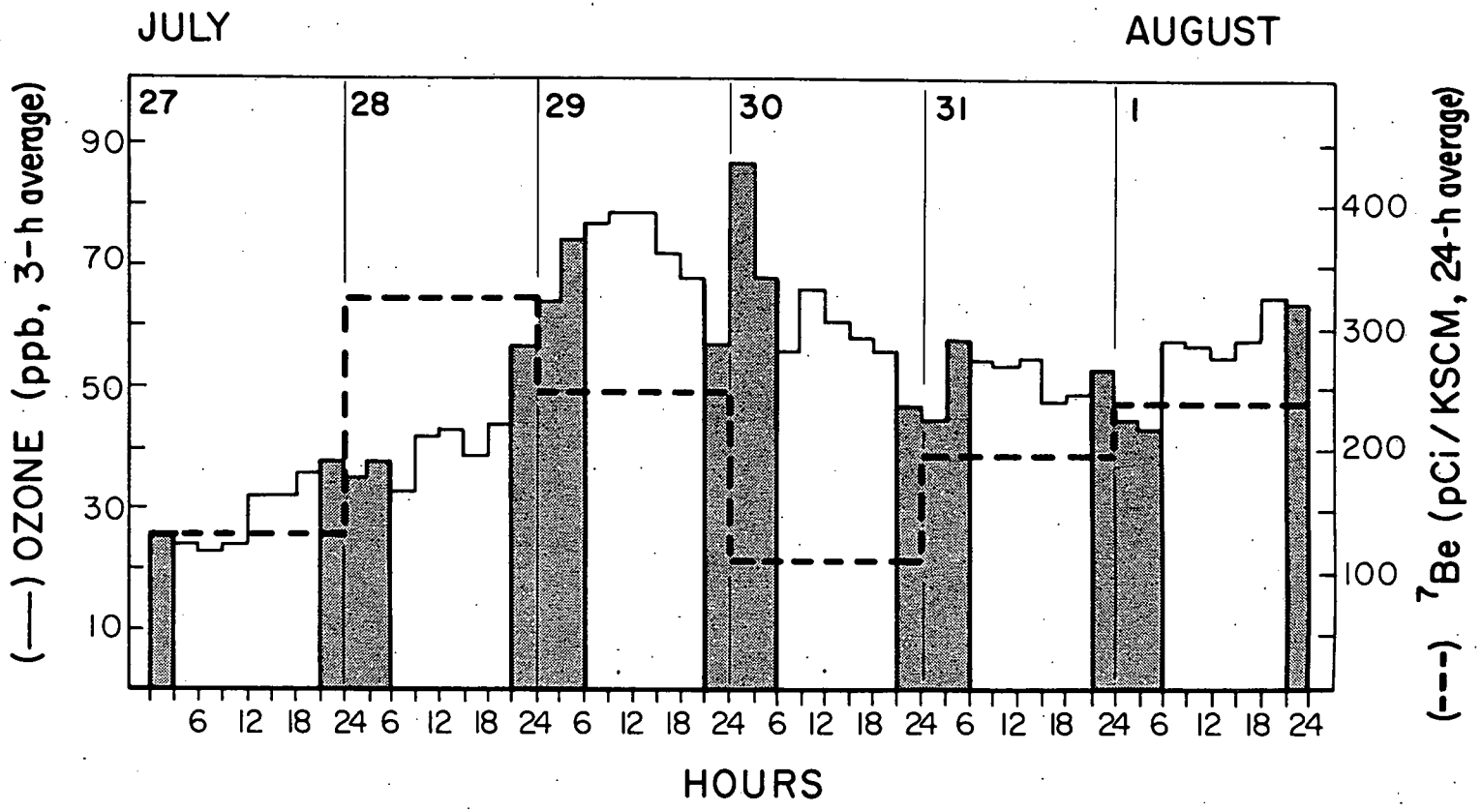
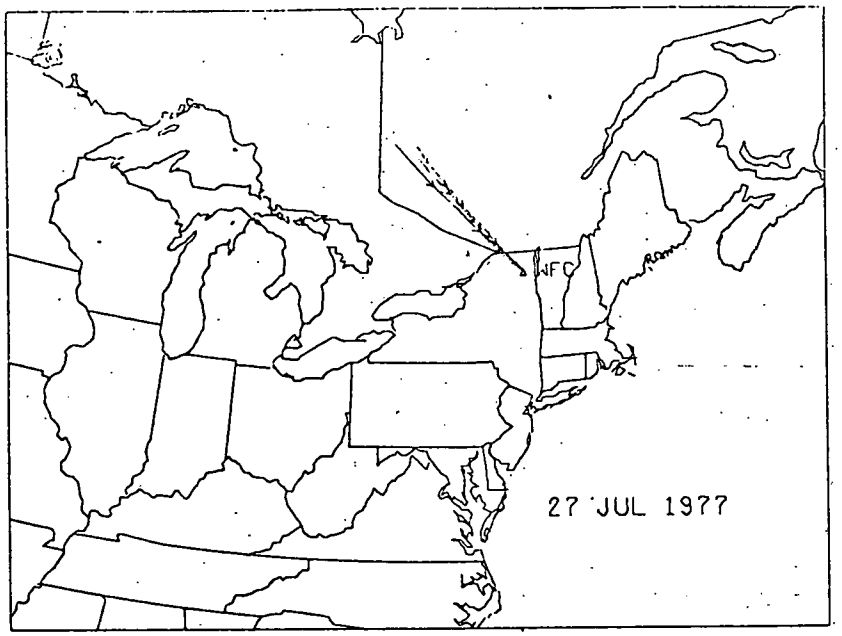
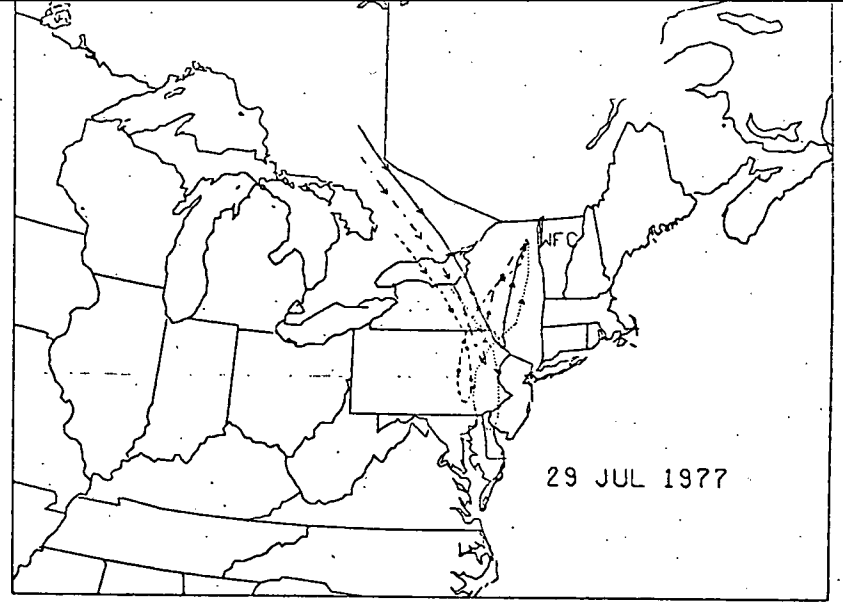
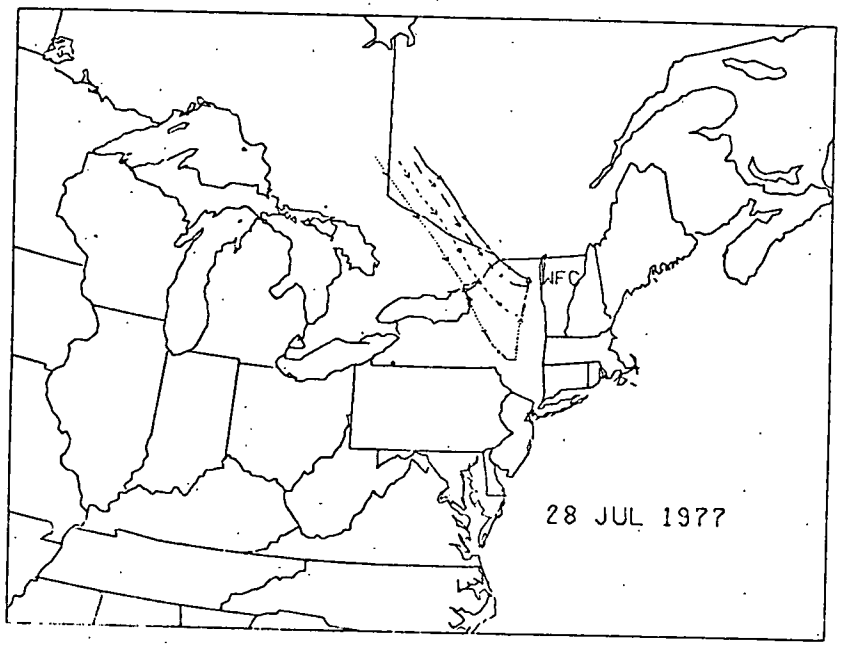

JULY
AUGUST
27
28
29
30
31
1
90
70
50
30
10
400
300
200
100
(—) OZONE (ppb, 3-h average)
(---) ^{7}Be (pCi / KSCM, 24-h average)
6 12 18 24 6 12 18 24 6 12 18 24 6 12 18 24 6 12 18 24 6 12 18 24
HOURS

FIGURE 8





——— 2 a.m.
----- 8 a.m.
-·-·- 2 p.m.
····· 8 p.m.



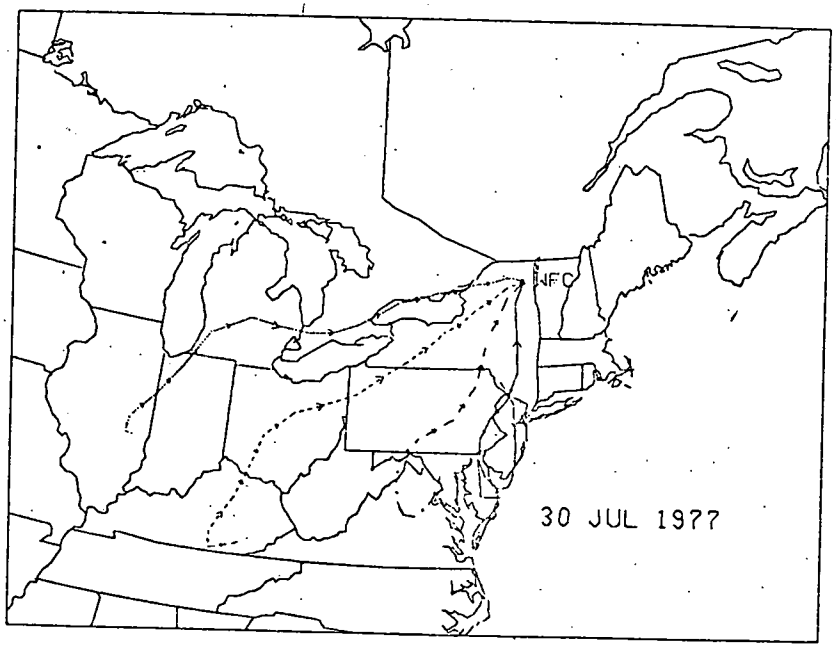


FIGURE 9

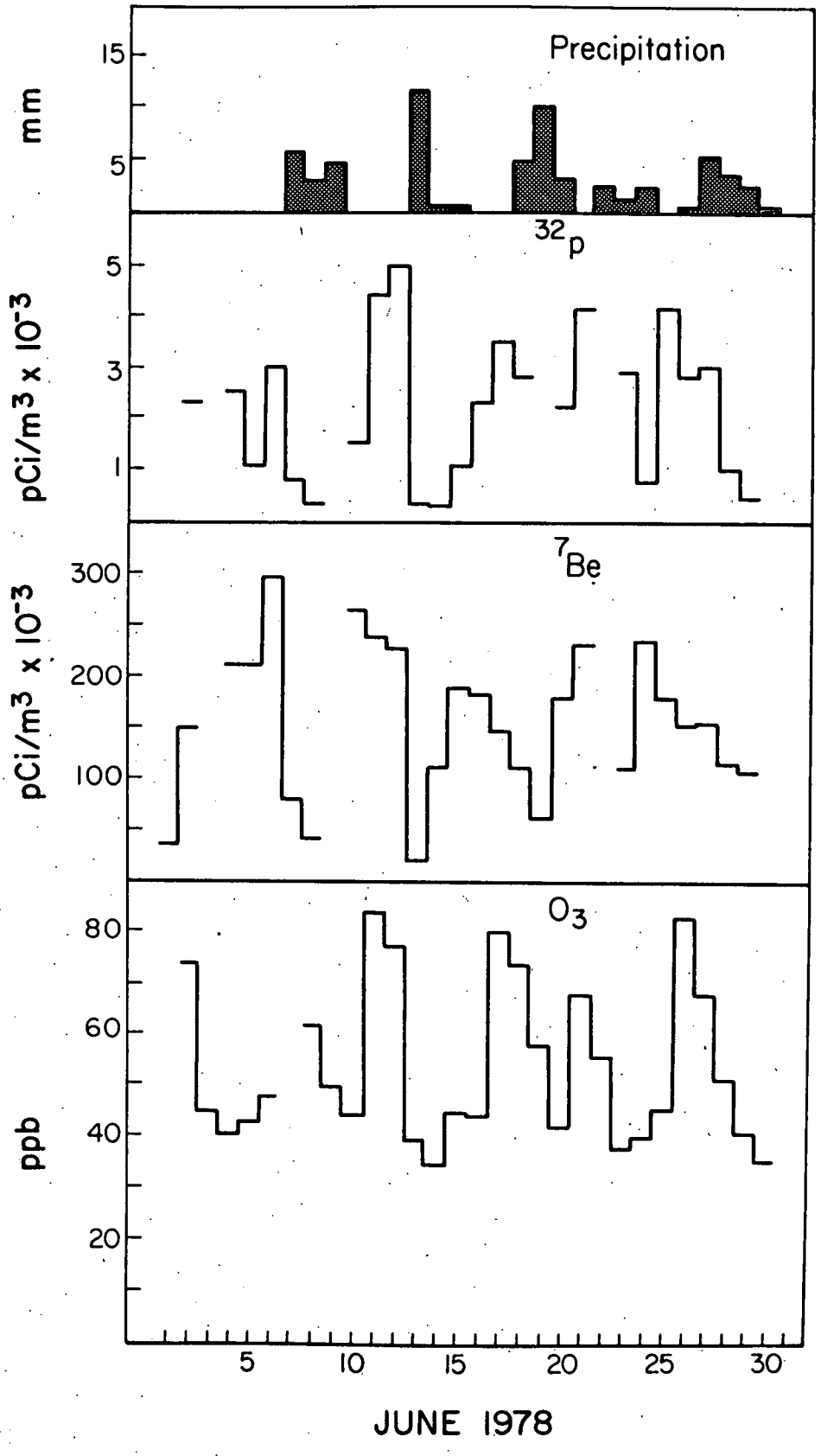

Precipitation
mm
15
5
^{32}P
pCi/m^3 x 10^{-3}
5
3
1
^{7}Be
pCi/m^3 x 10^{-3}
300
200
100
O_3
ppb
80
60
40
20
5
10
15
20
25
30
JUNE 1978

FIGURE 10

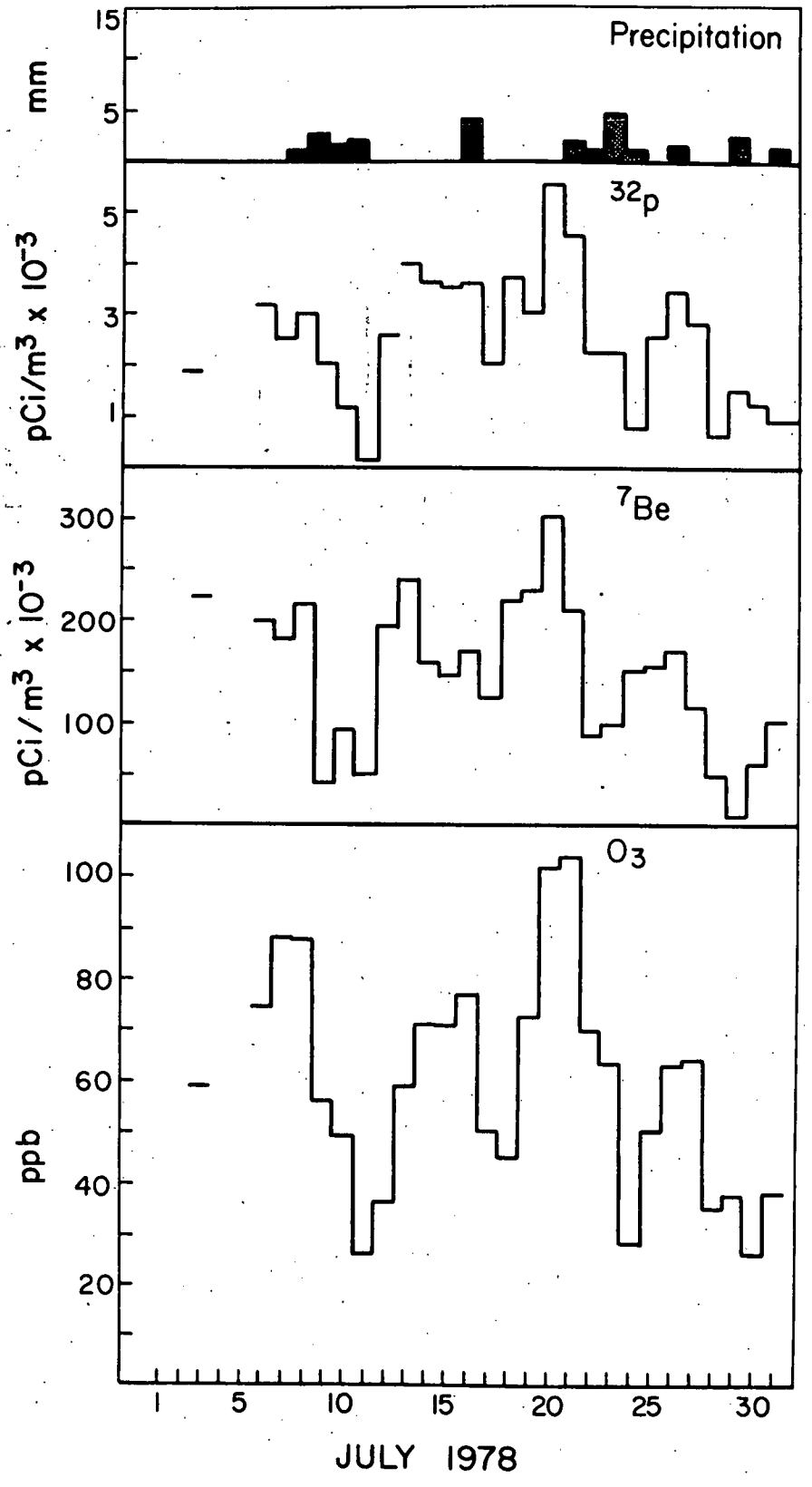

Precipitation
mm
15
5
^{32}P
pCi/m^3 x 10^{-3}
5
3
1
^{7}Be
pCi/m^3 x 10^{-3}
300
200
100
O_3
ppb
100
80
60
40
20
1
5
10
15
20
25
30
JULY 1978

FIGURE 11

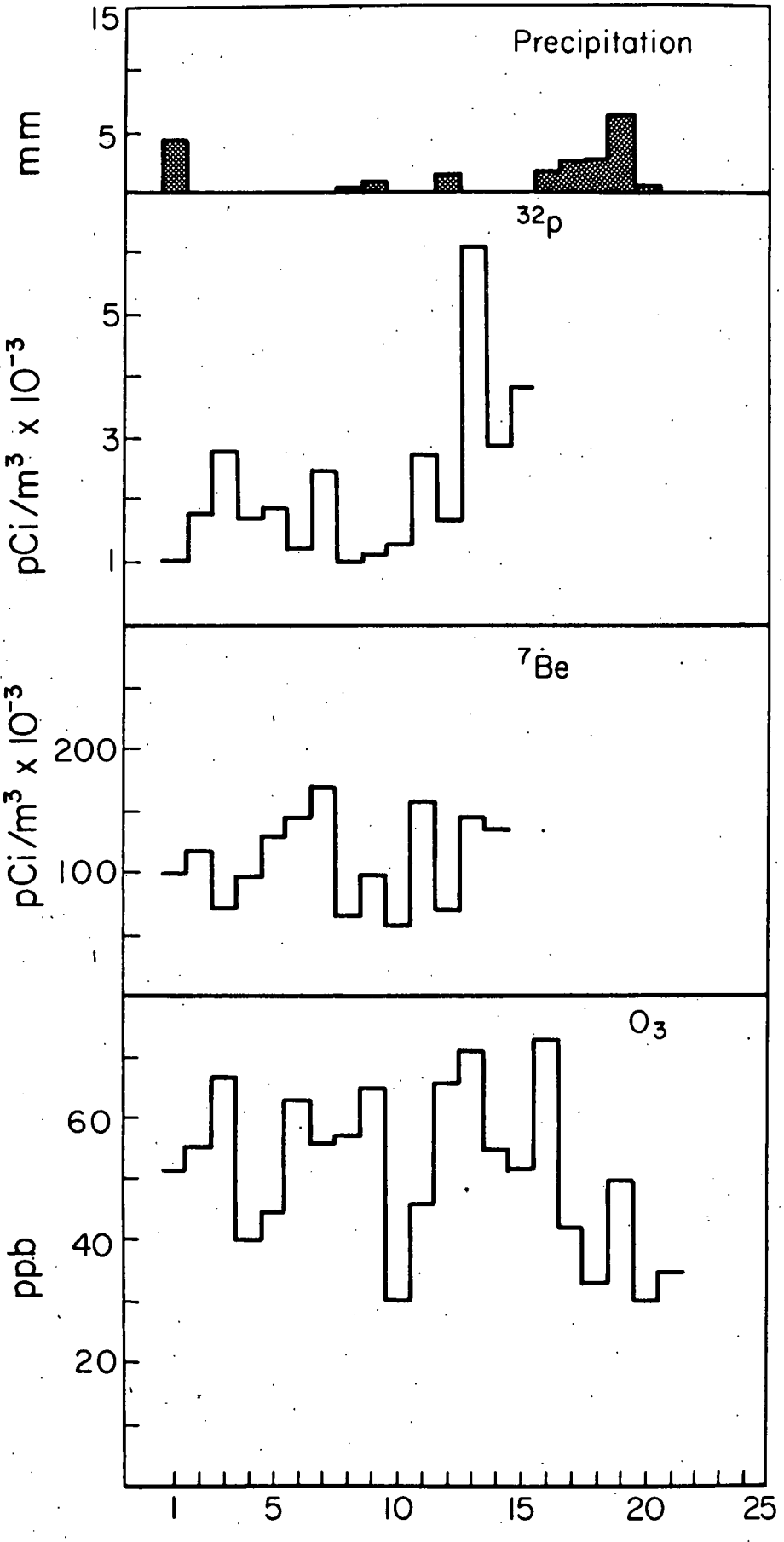

Precipitation
mm
15
5
^{32}P
pCi/m^3 x 10^{-3}
5
3
1
^{7}Be
pCi/m^3 x 10^{-3}
200
100
O_3
ppb
60
40
20
1
5
10
15
20
25
AUGUST 1978

FIGURE 12

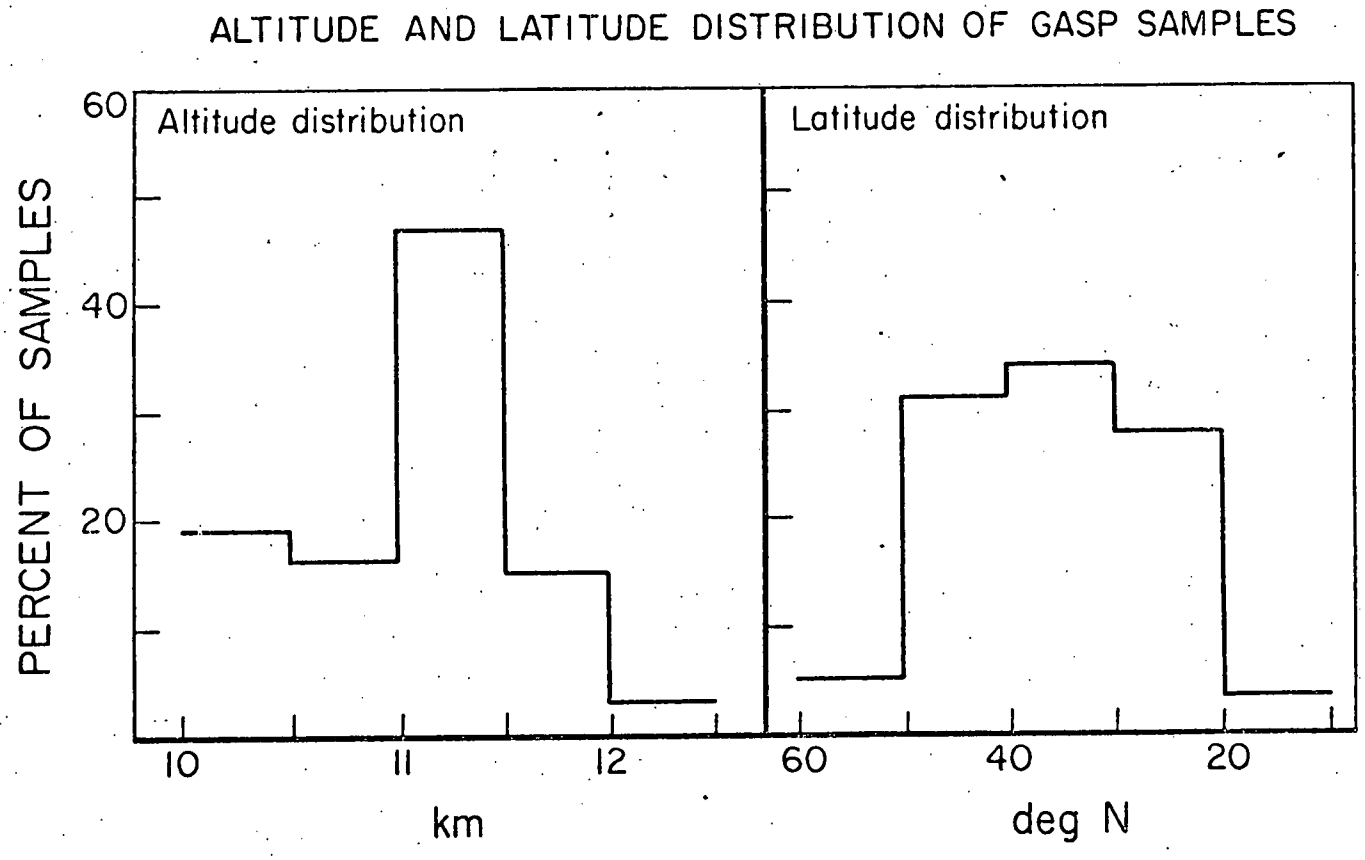
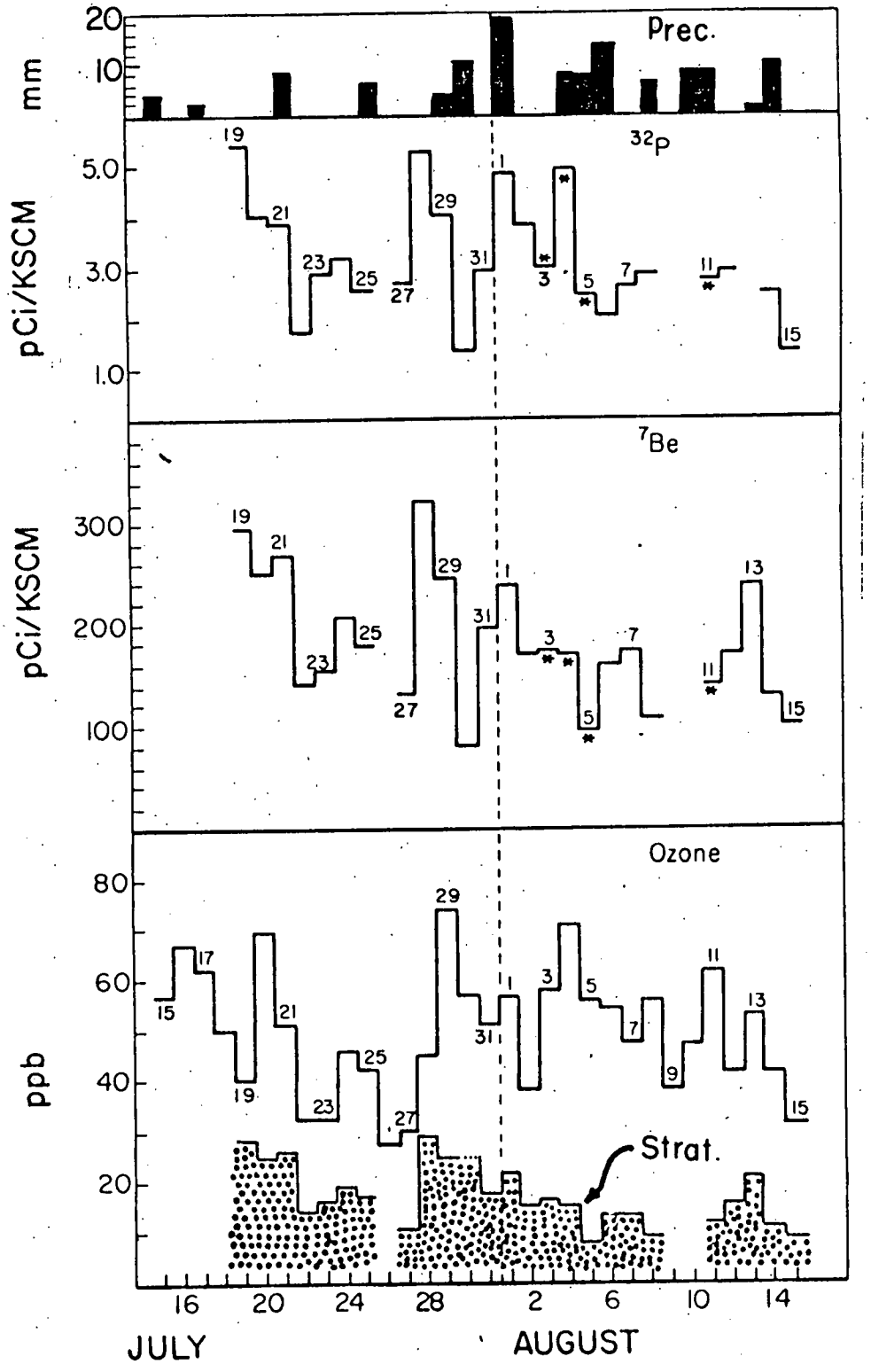

ALTITUDE AND LATITUDE DISTRIBUTION OF GASP SAMPLES
Altitude distribution
Latitude distribution
PERCENT OF SAMPLES
60
40
20
10
11
12
km
60
40
20
deg N

FIGURE 13


Prec.
mm
20
10
^{32}P
pCi/KSCM
5.0
3.0
1.0
^{7}Be
pCi/KSCM
300
200
100
Ozone
ppb
80
60
40
20
Strat.
16
20
24
28
2
6
10
14
JULY
AUGUST

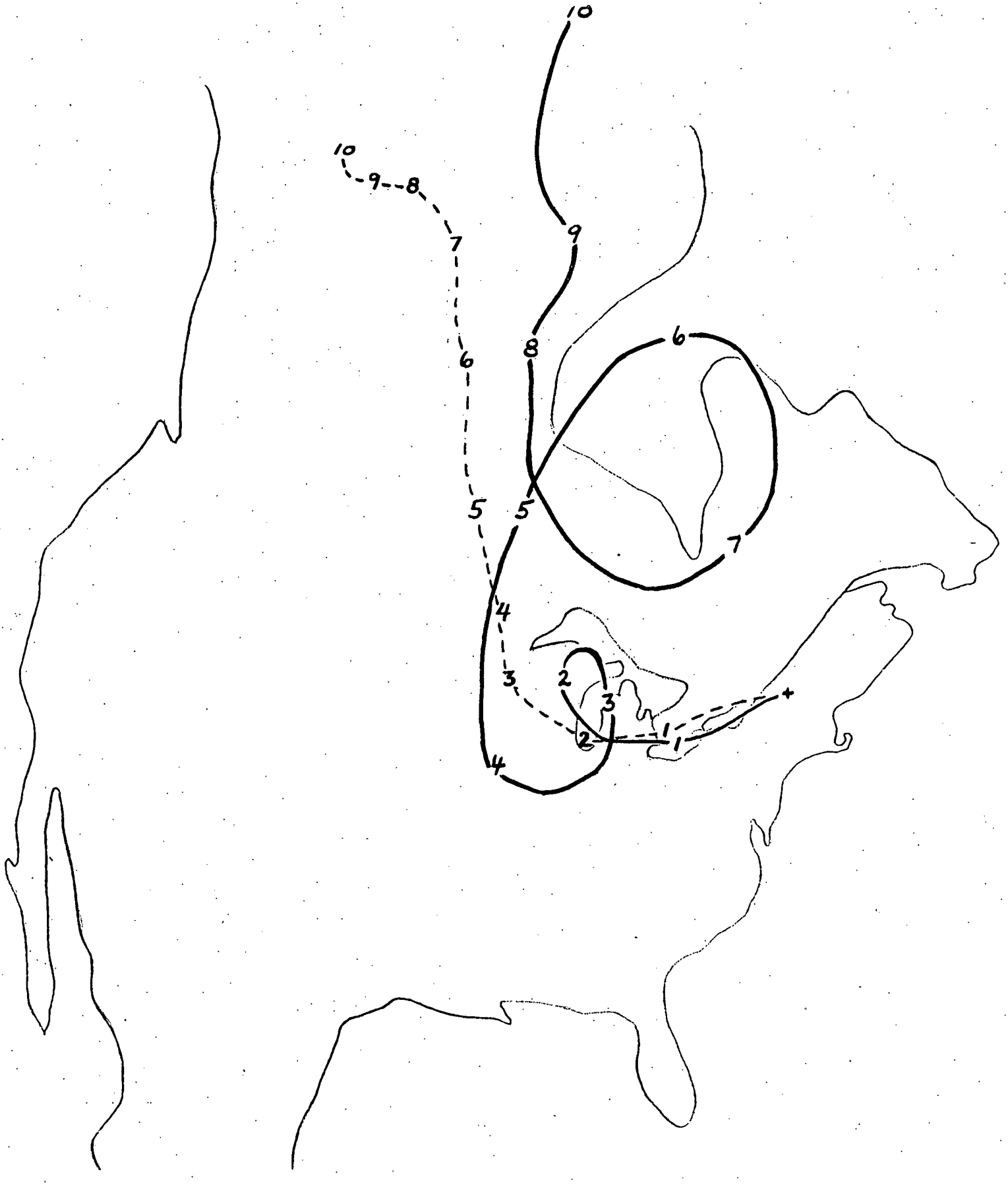


FIGURE 15 Backward isentropic trajectories along the 305° surface; numbers are days prior to arrival at Whiteface Mountain on July 16, 1975 (——) or July 17, 1975 (– – –).

APPENDIX I

Ozone Transport from Stratosphere to Troposphere
Husain et al.

Reprints Removed