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An Assessment of Atmospheric Exposure and Deposition to High Elevation Forests in the Eastern United States

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AN ASSESSMENT OF ATMOSPHERIC EXPOSURE AND DEPOSITION TO HIGH ELEVATION FORESTS IN THE EASTERN UNITED STATES

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FOREWORD

This report represents an assessment of exposure and deposition of air pollutants to forests, and interception of clouds by forest canopies. It summarizes data obtained over the first three years of MCCP, 1986 - 1988, with limited exposure data from 1989.

The report is part of a series of reports to the Environmental Protection Agency produced by the Mountain Cloud Chemistry Project in fullfillment of contractual agreements. These include:

Mohnen et. al., "Exposure of Forests to Gaseous Air Pollutants and Clouds", June, 1987.
Mohnen et. al., "Elevational Gradients in Wet and Dry Deposition of Pollutants", Jan 1988.
Mohnen et. al., "Chemical Deposition to High Elevation Spruce-Fir Forests in the Eastern United States", Feb 1988.
Mohnen et. al., "Exposure of Forests to Air Pollutants, Clouds, Precipitation and Climatic Variables, Sep 1988.
Mohnen et. al., "Mountain Cloud Chemistry Project Wet, Dry and Cloud Water Deposition", Sep 1988.

The main emphasis of MCCP scientists was to publish their results in the peer reviewed literature. Over fifty publications stemming from MCCP research have appeared, or will appear, in peer reviewed journals. Over twenty manuscripts are in various stages of preparation for submission to peer reviewed journals. In addition, numerous papers were presented at scientific meetings which have been published in conference proceedings. Finally, all principal scientists of MCCP have been involved in the preparation of the State of Science (SOS) documents dealing with exposure/deposition. A complete list of all these publications produced by MCCP scientists to date is presented in Appendix D.

Similar efforts to those of the MCCP are carried out in southeastern Canada under the CHEF (Chemistry of High Elevation Fog) Program. It will therefore be possible in the future to jointly assess the impact of air pollutants, clouds and precipitation on forests throughout eastern North America.

For a complete discussion of results from the MCCP, the reader is referred to the scientific journals. This report highlights some major results and summaries of MCCP achievements.

SECTION 1

INTRODUCTION

This report is the third in a series of annual summaries of research on the exposure and deposition of airborne chemicals to forest canopies and the forest floor in eastern North America. It presents results from the 1986-88 field seasons, with limited exposure data from 1989. The report is produced by the scientists in the Mountain Cloud Chemistry Program, a multi-year study of atmospheric chemistry and physics sponsored by the United States Environmental Protection Agency.

The Mountain Cloud Chemistry Program (MCCP) is a principal source of atmospheric chemical and physical information available to evaluate the hypothesis that acidic and other airborne chemicals contribute to the observed decline in spruce-fir forests at high elevations in the eastern USA. The objective of the MCCP is to characterize the exposure of montane forested ecosystems (largely spruce fir) to atmospheric inputs (chemical, physical and climatic). To meet this objective, MCCP conducted atmospheric research to determine:

- elevational gradients in wet and dry deposition;
- the relative significance of various deposition mechanisms to the fluxes of chemical materials into and through forest canopies; and
- frequency distributions of chemical, physical, and climatic exposure.

Resource and logistical considerations dictate where measurements can be performed. Five high-elevation sites and one low elevation site were selected from 45°N to 35°N to be as representative as possible of the geographic and meteorological variability in this large region.

During the past two decades, high elevation red spruce and fir forests in large areas from Maine to North Carolina have shown visible symptoms of injury , decreased radial growth, and increased mortality (McLaughlin, 1985; Hornbeck and Smith, 1985; Johnson and Siccama, 1983). Field studies by Falconer and Falconer (1980), Waldman et al. (1982), and Dollard et al. (1983) demonstrated that cloud water can be much more acidic than precipitation, and that substantial fluxes of sulfur and nitrogen can result from droplet interception to forest canopies. Unsworth (1984) calculated that evaporation of intercepted cloud water from forest canopies can lead to chemical concentrations on leaf surfaces that are substantially larger than concentrations measured in the drops themselves.

Because chemicals in cloud water may contribute to the stress experienced by high-elevation forests in eastern and western North America (Fowler et al., 1988; Waldman and Hoffman, 1988; Turner et al., 1988), networks of monitoring sites have been started to characterize the concentration and deposition of atmospheric pollutants in these forests. The principal monitoring network in the USA is the MCCP, which was implemented by the Forest Response Program of the National Acid Precipitation Assessment Program (NAPAP). The data collected by the MCCP is being used in the NAPAP Integrated Assessment to evaluate the role of airborne chemicals in the changing condition of forests.

This report documents typical values, extremes, and sources of variability in chemical concentrations and deposition from clouds that contact the forest canopy. Because the data are limited in coverage, it is difficult to generalize about spatial patterns. Processes controlling the variability in cloud water concentration and deposition are addressed to define limits on extrapolation. Both cloud water chemical concentration and deposition are considered because foliar-mediated processes may reflect

the concentrations in cloud water while soil-mediated processes reflect cloud water deposition (McCune and Lauver, 1986; Kohlmaier, et al., 1986; Schaefer, et al. 1989; Reuss et al., 1987).

Numerous investigators have reported data on cloud acidity and, to varying degrees depending on the focus of their study, on cloud water chemistry. Table 1-1 shows examples of cloud acidity measurements over several decades and locations. The cloud acidities reported by these authors vary greatly, indicating that no "average" cloud pH exists. Each pH measurement reflects the chemical composition not only of the condensed phase, but also of the gaseous phase. The condensed phase contains a mixture of dissolved gases, aqueous solutions, and solids, which may vary with air mass origin, cloud type, and height above cloud base.

Characterizing the chemical composition of cloud droplets depends on the objectives of the investigation. A complete characterization requires, at a minimum, determination of the partitioning of each species of interest among the various phases (Morgan, 1982), and as a function of size in the condensed phase (Noone et al., 1988). Considering the difficulties associated with sampling clouds, it is not surprising that such a complete characterization has not yet been performed. The present understanding of cloud chemistry is therefore based on bulk cloud water samples that represent a weighted average droplet composition.

TABLE 1-1
A Historical Overview of Cloud Acidity Measurements

Cloud water reported from low elevations

Reference	Year of Study	Location	Range of pH
Houghton (1955)	1954	Northeast USA	4.5 - 7.2
Mrose (1966)	1957	Germany	3.8 - 5.1
Lazrus et al. (1970)	1967	Puerto Rico	4.9 - 5.4
Bressan & Larson (1979)	1975	Nova Scotia, Canada	3.0 - 6.9
Waldman et al. (1982)	1981	California, USA	2.2 - 4.0
Fuzzi et al. (1983)	1980-82	Italy	2.5 - 7.0
Munger et al. (1983)	1981-82	California, USA	2.2 - 5.8
Fuzzi et al. (1984)	1982	New York, USA	4.3 - 6.4
Jacob et al. (1985)	1982-83	California, USA	2.2 - 6.2
Jacob et al. (1986)	1983-84	California, USA	2.7 - 7.2
Muir et al. (1986)	1985-86	Midwestern USA	2.9 - 4.1
Weathers et al. (1986)	1984	Eastern USA	2.1 - 3.0

Cloud collected water by aircraft

Reference	Year of Study	Location	Range of pH
Oddie (1962)	1960	United Kingdom	4.4 - 7.2
Petrenchuk & Drozdova (1966)	1961-64	USSR	3.4 - 5.9
Scott (1978)	1976	Australia	4.6 - 7.5
Scott & Laulainen (1979)	1977	Michigan, USA	3.7 - 4.0
Lazrus et al. (1983)	1980	Eastern USA	3.6 - 4.4
Daum et al. (1984)	1981-83	Eastern USA	3.1 - 6.1

Cloud water collected at montane sites

Reference	Year of Study	Location	Range of pH
Okita (1968)	1963	Japan	3.5 - 6.5
Castillo (1979)	1976	Whiteface Mtn, NY USA	3.4 - 4.2
Falconer & Falconer (1980)	1977-79	Whiteface Mtn NY USA	2.7 - 4.7
Weathers et al. (1986)	1984	Eastern USA	2.8 - 3.1
Hering et al. (1987)	1983	California, USA	3.1
Mohnen & Kadlecek (1989)	1982-87	Whiteface Mtn, NY USA	2.5 - 4.8

SECTION 2

CONCLUSIONS

This section presents a summary of the key findings and conclusions of the MCCP during its first three field seasons. A more detailed presentation of these findings is presented in Section 4 of this report. A description of the methods and protocols used to derive these findings can be found in Section 6.

CLOUD FREQUENCY

Regional cloud climatology studies indicate that cloud base height was typically between 800 m and 1200 m in the Appalachian Mountains. The northern Appalachians were cloudier than the southern. MCCP data for five high-elevation sites indicated that the Appalachian peaks (> 1400 m) were in cloud 30% to 40% of the time during the growing season (April-October), and, for one site with year-round data (Whitetop, VA), more frequently during the non-growing than the growing season. Elevations of 1000 m and higher experienced cloud at least 10% of all hours. Clouds were present at some time on at least one out of three days for lower elevations (1000 m to 1400 m), while higher sites (> 1400 m) had clouds up to 75% of days during the growing season. The higher elevation mountains had a greater frequency of cloud at night, in contrast to a predominance in the afternoon over surrounding lower terrain. Approximately 25% of the mountain cloud events were precipitating.

CLOUD AND PRECIPITATION CHEMISTRY

In high elevation spruce-fir forests, observed cloud water pH was approximately 0.6 pH units lower than precipitation pH. Concentrations of the major ions (NH_4^+, H^+, SO_4^{2-}, and NO_3+) were substantially higher in cloud water than in precipitation by factors ranging from 5 to 20, depending on location within the Appalachians. H^+, SO_4^{2-}, and NO_3^- concentrations greater than 100 μeq/L occurred infrequently in precipitation but routinely in cloud water (> 40% of cloud hours) at the five MCCP monitoring sites. Thus, locations below cloud experienced both lower mean concentrations and fewer extreme concentrations than high-elevation sites in the eastern USA. There was a significant altitudinal gradient within clouds at Whiteface Mountain, NY, for NH_4^+, H^+, SO_4^{2-}, and NO_3^+ ions, with higher concentrations at or near cloud base. Thus, in the Appalachian Mountains, a step increase in the concentrations of wet deposited ions was observed as elevation increased from below cloud to cloud base (approximately 1000 m); further increases in elevation (within cloud) should result in a gradual decrease in the concentrations to which a forest canopy is exposed.

LIQUID WATER CONTENT

LWC values averaged approximately 0.20 g/m^3 at all sites, excluding Whiteface Mountain. The exception was Whiteface summit, where the average value was 0.46 g/m^3. A small amount of data from Whiteface Mountain, NY, suggest that the liquid water content of a cloud was lower at cloud base than at higher elevations. Therefore, observed LWC values at any site were most likely a function of site elevation relative to cloud base height as well as synoptic meteorology.

WET, DRY AND CLOUD DEPOSITION

One of the principal findings is that deposition of SO_4^{2-}, H^+, NH_4^+, and NO_3^- in cloud water (cloud interception) represented a significant input to montane forest canopies. In estimating cloud deposition to the forest canopy, MCCP used an improved version of the 1984 Lovett model (CDM). Cloud impaction events at each MCCP site were classified according to meteorological conditions that prevail during each hour of the event. A detailed analysis of cloud chemistry and meteorological variables (wind speed and liquid water content) has demonstrated the usefulness of this technique for uniquely characterizing conditions during events. In addition to synoptic classification, specific air trajectory directions computed within a given event type could further characterize event conditions. This technique was able to explain a major portion of the variance in the chemistry and meteorological data base, thereby allowing more complete growing season estimates of cloud deposition. Deposition estimates for MCCP sites could then be made for periods when data were incomplete as long as cloud frequency and event type would be determined. Event types were (1) pre-warm front, (2) NW sector of cyclone, (3) post-cold front, (4) warm sector of cyclone, (5) stationary front, (6) marine flow off Atlantic, (7) cutoff low in upper atmosphere, and (9) cap cloud.

Cloud deposition estimates for each site were made by computing, for each subclass, the mean water deposition flux using the improved CDM and subclass wind speed and liquid water content. Best estimates of canopy structure were then used to calculate the gross (pre-evaporation) cloud water flux to specific forest canopies at each site. Cloud water deposition can exceed the flux from wet (precipitation) deposition during the growing season at the mountain sites which are frequently exposed to cloud. Most of the site-differences in cloud deposition could be explained on the basis of differences in canopy structure. Cloud deposition was found to be highly site specific. Despite an almost 2:1 advantage in cloud frequency, Whiteface Mountain mean deposition estimates for the growing season were generally lower than those for Moosilauke because of lower canopy surface area at the specific Whiteface site. Differences in cloud water deposition between the northern and southern sites were significant and they were likely caused by differences in such parameters as canopy structure, elevation above cloud base and synoptic meteorology. It was also interesting to observe the annual changes in cloud water deposition as a result of changing metrological conditions from year to year. Whiteface Mountain, NY and Mt. Moosilauke, NH had estimated 1987-88 warm-season sulfate cloud droplet deposition that was of the same order of magnitude as measured precipitation chemical flux. Mt. Mitchell, NC had an estimated cloud water sulfate deposition that was about a factor of two higher than precipitation deposition. Estimated cloud sulfate deposition was up tp 4 times greater than that in precipitation for the Whitetop Mt. MCCP site.

The Atmospheric Turbulence Diffusion Laboratory deposition model (DRY DEP) was used by the MCCP to calculate the deposition velocities of sulfur dioxide, ozone, nitric acid vapor, and sulfate particles from meteorological and site specific biological information. Site input included: major and minor plant species type, leaf area index for plant species, and site location. All available meteorological data including canopy wetness and rainfall were also used. The estimates that have been made suggest that dry deposition of sulfate and nitrate is of greater relative importance at the Howland, ME and Shenandoah, VA sites than at the other high elevation sites.

It is possible to estimate the importance of cloud deposition to the overall flux of pollutants received by the forest canopy from the model results. Although cloud interception is highly dependent upon canopy structure and location above cloud base, it can be nevertheless concluded, that cloud deposition can deliver to the canopy from one to four times the amount of pollutants as precipitation does.

The estimated cloud-to-wet deposition flux ratios for 1987-88 growing season at four MCCP sites are:

Site	Sulfate	Nitrate
Moosilauke	0.8	1.2
Whiteface-2	1.1	0.8
Whitetop	3.5	4.4
Mt. Mitchell	2.0	2.2

At the cloud free Howland site, dry deposition appears to account for less than one third of the total acidic substances deposited. The Shenandoah site has a very low cloud impaction frequency due to its relatively low elevation and dry deposition appears to be of greater relative importance (deciduous trees). The estimates for Whiteface suggest that dry sulfur deposition accounts for less than a quarter of the total sulfur deposition flux.

Based on these research results, it can be concluded that any assessment of forest damage at high elevations must take all delivery mechanisms into account, and in particular, cloud deposition. However, substantial uncertainties may be associated with the deposition estimation procedures available to the MCCP for estimation of both dry deposition and cloud water interception to mountain forests and many of these uncertainties are as yet undefined.

OZONE

The long-term ozone exposure indices indicate considerable variability among the high elevation sites and at a given site from one year to the next. For example, the sum of season dose (total of hourly concentrations exceeding 70 ppb, expressed in ppm-h) for the 1988 growing season at the highest elevation subsite at each of the MCCP locations ranged from 12.51 ppm-h at Mt. Moosilauke to 45.17 ppm-h at Mt. Mitchell. Temporal variability is well illustrated at Mt. Mitchell, where the same dose statistic was 8.34, 5.14, and 45.17 ppm-h for the 1986, 1987, and 1988 monitoring seasons. The severe drought of 1988 along with the accompanying clear skies and stagnation episodes undoubtedly contributed to the high ozone exposures observed at many sites that year.

Quantifying the spatial and temporal variability in ozone episodes also reveals substantial variability. For example, during 1987 the Mt.Mitchell summit had eight episodes of ozone exceeding 70 ppb for at least eight consecutive hours, whereas during 1988 the figure increased to 33 episodes of this magnitude and duration.

Important differences between the high elevation and low elevation sites are revealed in the data. High elevation sites are often above the ground based temperature inversion at night, therefore the ozone concentrations do not drop off to near zero as they do at valley bottom or low elevation flat terrain sites. Therefore, the diurnal cycles and the exposure patterns are substantially different at mountain versus valley stations, even those in close proximity to one another. The two primary effects of this difference are to produce higher mean ozone concentrations and longer episodes at the high elevation sites compared with nearby valley bottom locations

HYDROGEN PEROXIDE

The mean concentrations of hydrogen peroxide in the gas phase at Whitetop Mountain and Whiteface Mountain during the warm season were between 0.6 ppb and 1 ppb. The maximum concentration was 6.1 ppb. The mean concentrations of hydrogen peroxide in cloud water at Whitetop and Whiteface Mountains during the warm season were between 32 and 36 μmol/l with a maximum concentration of 182 μmol/l.

OXIDES OF SULFUR AND NITROGEN

The concentrations of SO_2 and NO_x measured at the MCCP sites were found to be quite low (1-2 ppbv) most of the time. Occasional excursions above 10 ppbv, presumed to be associated with pollutant plumes, rarely lasted more than a few hours.

METEOROLOGY

Regional surface and upper air climatological measurements indicate that there was a significant contrast in weather conditions relative to long term normals between the Northern and Southern portions of the Appalachians from 1986 to 1988. Drought and warmer than normal conditions dominated the Southern Appalachians throughout the period. In some areas this drought was unprecedented over the last 100-300 years. The Northern Appalachians had relatively unexceptional weather overall in terms of persistent trends.

SECTION 3

PROJECT DESCRIPTION

MONITORING RATIONALE

The Mountain Cloud Chemistry Program (MCCP) has been designated by the EPA as a principal source of atmospheric chemical and physical information available to evaluate the hypothesis that acidic and other airborne chemicals contribute to the observed decline in spruce-fir forests in the eastern USA. The objective of the MCCP is to characterize the exposure of montane forested ecosystems (largely spruce fir) to atmospheric inputs (chemical, physical and climatic). To meet this objective, MCCP conducted atmospheric deposition research to determine:

- elevational gradients in wet and dry deposition;
- the relative significance of various deposition mechanisms to the fluxes of chemical materials into and through forest canopies; and
- frequency distributions of chemical, physical, and climatic exposure.

Resource and logistical considerations dictate where measurements can be performed. Five high-elevation sites and one low elevation site were selected from 45°N to 35°N to be as representative as possible of the geographic and meteorological variability in this large region.

Well-defined methods were not available to measure some inputs to high-elevation forests, and therefore many of these inputs must be estimated. Pollutant concentrations were measured at the boundary of forest canopy, and deposition was calculated using models. Measurements included gaseous and aqueous chemical measurements, cloud frequency, cloud liquid water content, and climatic measurements (deposition model inputs as well as a direct stress).

Few of the physical and chemical measurements required were easily performed. For example, no field measurement technique existed in the commercial sector for some measurements, such as hydrogen peroxide and cloud liquid water content. Even relatively simple procedures (e.g., ozone measurement or collection of particulate samples) required modification when applied to the harsh, wet mountaintop environment. So, the MCCP elected to perform certain experiments at a single site as part of a method evaluation experiment prior to use at all six sites. Therefore, some parameters may be available at some sites and not others.

The meteorological monitoring, conducted concurrently with the chemical exposure measurements, was designed to provide frequency of cloud exposure, deposition model inputs, and climatic information. Frequency of cloud immersion is critical in defining the extent, both in time and space, of the cloud water deposition process and in identifying opportunities for chemical contributions to, and subsequent impacts on, montane ecosystems.

The measurement approach taken by the MCCP and its contribution to the three MCCP objectives are:

- Primary Exposure Parameters: Site-specific measurements of cloud and rain water, gaseous sulfur and nitrogen compounds, and oxidants are made every hour or are directly converted into hourly concentration values for use in exposure estimates.

- Derived Deposition Parameters: Wet deposition of pollutants by precipitation can be directly calculated from hourly and weekly totals and, in general, provides a yardstick for comparison of other deposition processes. Cloud water deposition is estimated via indirect physical and chemical measurements. Process-oriented models are needed to estimate dry deposition and the material fluxes into and through the forest canopy.
- Inter-site Characterization: To enhance understanding of exposure patterns at individual sites, some inter-site comparison was performed to determine if any distinguishing regional characteristics could be observed in the data (e.g., meteorological, elevational, or latitudinal differences in cloud water chemical composition).

GEOGRAPHICAL REPRESENTATION

The Mountain Cloud Chemistry Program (MCCP) consists of six sampling sites in the northeastern and southeastern USA. Five sites are located at high elevations that experience frequent cloudiness: Whiteface Mountain, NY; Mt. Moosilauke, NH; Shenandoah, VA; Whitetop Mountain, VA; and Mt. Mitchell, NC. One low-elevation MCCP site in Howland Forest, ME, does not experience substantial cloudiness, but was included for comparison of total deposition and forest condition. The location of the MCCP sites are depicted in Figure 3-1.

At Whiteface Mt., Whitetop Mt. and Mt. Mitchell, the main sites are located on the summit of mountains, while the main sites at Shenandoah and Moosilauke are along a ridgeline. Sub-sites are located on the slopes of Whiteface, Shenandoah, and Mt. Mitchell. At the Shenandoah and Mitchell sub-sites, instruments are mounted on scaffolding towers; at Whiteface, a mobile trailer is equipped with a collector attached to a pole that is hoisted up and down on an event basis.

The Howland Forest site (45°13'N, 68°43'W) is located at 65 m elevation between Howland and Edinburg, Maine, 35 miles north of Bangor. The forest is spruce with lesser numbers of balsam fir, hemlock, and white pine. A second site is located 2 km west of the first at an elevation of 60 m in an extended spruce-fir stand. This site has uniform terrain and extensive fetch ideally suited to classical micrometeorological measurements.

Mt. Moosilauke, New Hampshire (43°59'N, 71°48'W), is one of the most southern peaks of the White Mountains. It is located about 50 km southwest of Mt. Washington and about 10 km northeast of the USFS Hubbard Brook Experimental Forest and Watershed. The site at Moosilauke is at 1000 m and is partially shielded from the prevailing westerly winds. Consistent with its mid-elevation location, the site does not experience the high frequency of orographic clouds that can be seen on the summit (1465 m). The forest composition ranges from mixed hardwoods at lower elevations to spruce-fir (about 10% spruce) at mid-elevations, and pure balsam fir at high elevations.

Whiteface Mountain (44°23'N, 73°59'W) is located in the northeastern Adirondack Mountains in New York, at an elevation of 1483 m. The summit is above tree line, providing access to regional air flow (sub-site 1). A four-story building houses instrumentation and is the primary site for cloud water collection. The Whiteface Mountain-Lake Placid Turn sampling site (sub-site 2) is located at 1245 m adjacent to a balsam fir canopy. A meteorological tower has been installed for routine monitoring, but cloud water collections are made from a mobile tower along the road. The Integrated Forest Study (IFS) sampling and monitoring activities take place at 1025 m (sub-site 3), an elevation frequently near or below cloud base. The forest canopy on the lower slopes consists of balsam fir, white birch, and red spruce. Sampling for three precipitation networks (MAP3S, NTN, and MODES) and U.S. EPA and NOAA dry deposition monitoring are conducted at the base support area (sub-site 4; 620 m).

The Shenandoah, VA site (38°7'N, 78°20'W) is in the Shaver Hollow Watershed, located in the north-central sector of the Shenandoah National Park. Slopes in the watershed average 30% to 50%, facing generally northwest. Meteorological towers are erected at three sites in the watershed: sub-site 1 at 1015 m, sub-site 2 at 716 m, and sub-site 3 at 524 m. The tower locations are representative of the surrounding deciduous forest canopy (primarily, northern red oak, chestnut oak, and red maple, with several northern hardwoods and eastern hemlock in wetter coves).

Whitetop Mountain (36°38'N, 81°36'W) is located in the Mt. Rogers National Recreation Area of the Jefferson National Forest in southwestern Virginia, 6 km southwest of Mt. Rogers, the highest peak in Virginia. The Whitetop Mountain summit research station (at 1689 m) straddles the main ridgeline of the Appalachian range, strategically located to intercept air from several directions. Most measurements are made 5 to 8 meters above ground on a platform over a trailer located in a clearing.

The southernmost MCCP site is located in Mt. Mitchell State Park, North Carolina (35°44'N, 82°16'W), one mile southwest of Mt. Mitchell, which is the highest peak in the eastern USA (2038 m). The summit is covered with Fraser fir, and the region from 1500 m to 1800 m is mixed fir and spruce. A 16.5 m meteorological tower is installed on Mt. Gibbs at 1950 m (sub-site 1); additional gas and meteorological measurements are performed at 1775 m (sub-site 2).

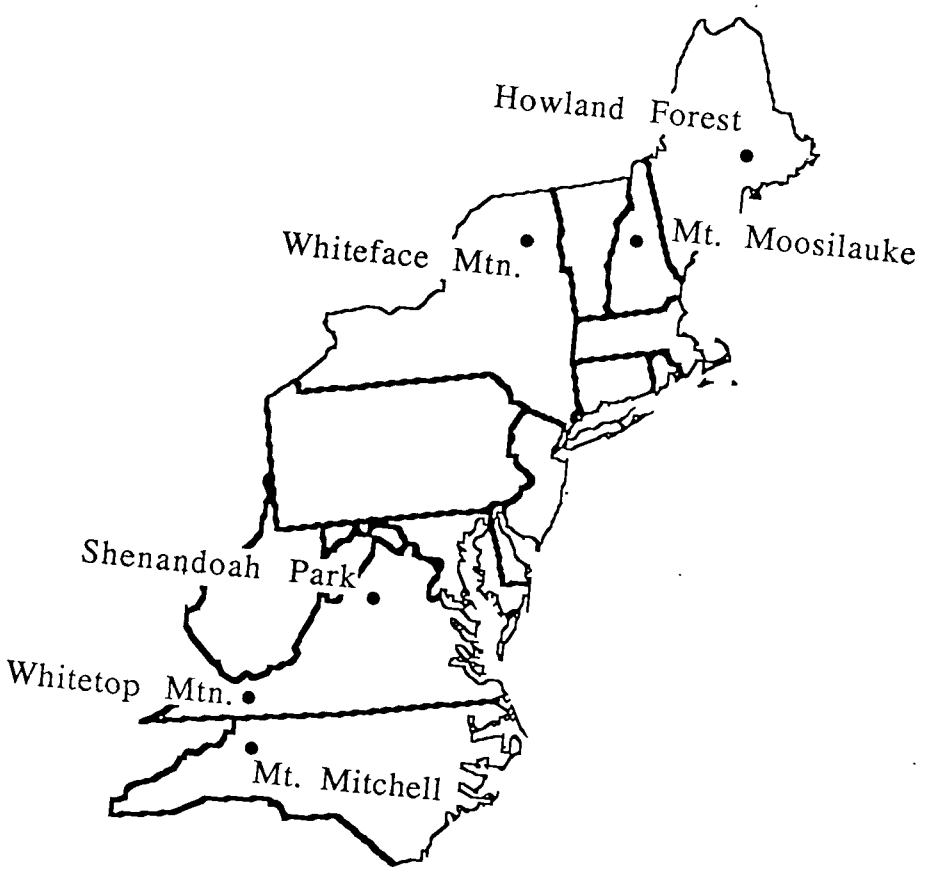


Figure 3-1: The location of the MCCP monitoring sites

SECTION 4

ASSESSMENT

The process of estimating exposure and deposition of atmospheric pollutants involves the assessment of a number of important factors. Measurements of cloud water chemistry, gas concentration, frequency of cloud, cloud liquid water content, other meteorology, and throughfall are factors that must be considered to adequately characterize exposure and deposition. In this section, each of these factors is considered in some detail.

CLOUD WATER CHEMISTRY

This section presents results for cloud water chemical composition for all sites. The 1986-88 growing season mean values for NO_3^-, NH_4^+, H^+, and SO_4^{2-} are presented along with their frequency distributions for non-precipitating clouds. Summer and winter concentrations are compared, as well as differences with elevation at one well-studied site, Whiteface Mountain, NY. Because concentrations of pollutants in cloud water are highly variable, possible meteorological controlling factors are also investigated. H_2O_2 concentrations in cloud water are presented.

Unless otherwise stated, the cloud water chemistry data represent unweighted concentrations. Although the concentrations are expected to vary with liquid water content, few analyses have shown a significant correlation between cloud water concentrations and liquid water content (LWC). One of the reasons for the lack of demonstrated correlation is the relative absence of accurate time-resolved LWC data; although MCCP measured LWC during 1987-88 there are enough missing LWC data (about 30%) for that period to make any LWC-weighing scheme problematic. Mohnen (1988c), however, compared LWC weighted and unweighted methods for the 1987 MCCP data and did not reach different conclusions for different weighing schemes.

We have characterized central tendencies in the MCCP cloud water chemistry data using mean values. Schemenauer (1986) compared mean, median, and daily median statistics for summarizing cloud water data and reached similar conclusions, but sometimes achieved a shift in emphasis, for different summary statistics. To avoid problems associated with skewed data distributions, we either display the data distributions (for qualitative comparisons), transform the data (for statistical analyses), calculate median values, or utilize both mean and maximum values (to characterize exposures).

Seasonal Variation

Although the MCCP only collects cloud water samples during April-October, previous cloud water chemistry data from the Whiteface Mountain, NY summit site provide insight into seasonal variability in ion concentrations. The median and maximum concentrations, the ratio of sulfate to nitrate ion, and the distribution of concentrations in precipitation and cloud water were compared for summer and winter sampling periods at Whiteface Mountain by Kadlecek et al. (1985; 1988). The nearby MAP3S event-based precipitation chemistry data distributions were compared to cloud water chemistry distributions to demonstrate the elevational dependence (in-cloud versus below-cloud) of exposure.

During summer at Whiteface, under non-plume conditions, SO_2(gas) lifetimes are less than one hour if clouds are present (McLaren et al., 1984). Once an air parcel has been entrained into a cloud, ambient SO_2 is rapidly oxidized by H_2O_2 in the aqueous phase to sulfuric acid; thus, future sulfur

transport is governed by cloud droplet, rain, and/or aerosol dynamics. Also, direct deposition of sulfur to the high-elevation canopy by cloud water impaction is substantially greater than it would be if the SO_2 had remained in the gas phase.

Summertime chemical processes do not apply to wintertime conditions because of differences between summer and winter temperatures, solar flux, absolute humidity, and meteorology. To examine the consequences of these differences, cloud water was collected in winter to create a data base similar to that obtained during summer. Table 4-1 summarizes the concentrations of the four major cloud ions observed during January and February of the four winters sampled at Whiteface Mountain, NY; for comparison, the same ions are shown for June-August. Samples taken during cloud-only conditions have been included.

TABLE 4-1

Comparison of Winter and Summer Cloud Ion Concentrations at Whiteface Mountain, NY (from Mohnen and Kadlecek, 1989).

January-February (μeq/L)

Year	H^+ median	H^+ max	SO_4^{2-} median	SO_4^{2-} max	NH_4^+ median	NH_4^+ max	NO_3^- median	NO_3^- max	# of samples	days with data
1983	129	589	100	554	54	205	71	258	107	8
1984	204	1148	160	1632	95	680	140	838	156	11
1985	324	776	366	1100	218	458	250	631	150	11
1986	191	1585	168	972	73	522	86	1131	247	22

June-August (μeq/L)

Year	H^+ median	H^+ max	SO_4^{2-} median	SO_4^{2-} max	NH_4^+ median	NH_4^+ max	NO_3^- median	NO_3^- max	# of samples	days with data
1981	-	-	154	3562	68	732	60	816	289	32
1982	141	1660	242	1224	134	788	74	590	171	33
1983	166	3162	224	3050	130	855	69	1338	304	29
1984	251	977	316	1202	147	612	83	285	124	21
1986	145	1413	142	1318	64	770	46	752	162	23
1987	148	1778	194	2224	113	920	68	1344	140	25

Ion concentrations varied as much among winters as they did between winter and summer. Variability within individual seasons was large, with standard deviations typically 50% to more than 100% of the seasonal medians (the concentrations are not normally distributed). Seasonal medians of different years varied greatly, often by a factor of two to three, and particularly in winter. One reason for large differences in winter concentrations is that large-scale meteorological conditions, once established, tend to persist, but they do not necessarily repeat each winter. Also, samples were taken in only part of each winter. For example, if an event is relatively clean, most samples will have concentrations of a few tens of μeq/L; if the event is relatively polluted, the concentrations will generally

be a few hundreds of μeq/L. If the block of samples obtained tends to emphasize a different set of transport conditions than the unmonitored cloudy periods, the seasonal statistics will be influenced.

Sulfate concentrations were only slightly greater in summer than in winter. While this may seem inconsistent with the much lower rate of SO_2 transformation in winter, the few available measurements indicate that the lower liquid water content of winter clouds (Kadlecek et al., 1988) tends to concentrate the available species in about half the water, suggesting an average wintertime atmospheric clear air sulfate loading equal to about half that of summer.

Since the rapid oxidation of ambient SO_2 characteristic of summer clouds is not available during winter, and since atmospheric loadings of nitrate are similar (summer and winter), a shift in the sulfate to nitrate ratio was expected. This shift is apparent in Table 4-2.

TABLE 4-2

Comparison of Median Sulfate to Nitrate Ratios at Whiteface Mountain, NY.

Year	Summer	Winter
1981	1.3	-
1982	1.6	-
1983	1.6	0.7
1984	1.9	0.6
1985	-	0.7
1986	1.5	1.0
1987	1.4	-

Shown in Figures 4-1 and 4-2 are the summer and winter concentration distributions of the dominant anions in cloud water and precipitation from Whiteface Mt. Across most seasons, the cloud water concentrations of the dominant ions ranged over three orders of magnitude. However, individual cloud events or substantial periods of events lasting many hours had relatively constant ion concentrations. The distributions in Figures 4-1 and 4-2 resulted from the mixing of many different events, each with its own relatively constrained concentration range.

These summer and winter cloud data can also be compared to MAP3S (event-sampled) precipitation data from Whiteface Mountain in Figures 4-1 and 4-2. Nearly all of the precipitation samples in each season had anion concentrations less than 200 μeq/L. During summer for most years, both precipitation distributions had similar shapes, peaking in the 40 to 100 μeq/L range for SO_4^{2-}. During winter, NO_3^- resembled the summer distribution, although it shifted slightly to higher concentrations. The precipitation SO_4^{2-} distribution was shifted substantially to lower concentrations; more than 80% of the winter samples had less than 40 μeq/L, consistent with the reduced oxidation rate of SO_2 in the winter atmosphere.

In addition to cloud liquid water content, differences between seasonal trajectory patterns suggest that the winter inter-annual differences probably result in large part from differences in meteorological flow, emphasizing that average meteorological conditions do not repeat each year (Kadlecek et al., 1988). For example, trajectories ending at Whiteface Mountain, NY, during January, 1985 (the month in which most winter data were taken), showed the flow to be unusually restricted to southwestern sectors, resulting in relatively more samples with higher ion concentrations. Conversely, during 1983, more trajectories traversed areas with low emission sources, such as the Atlantic Ocean or regions to the

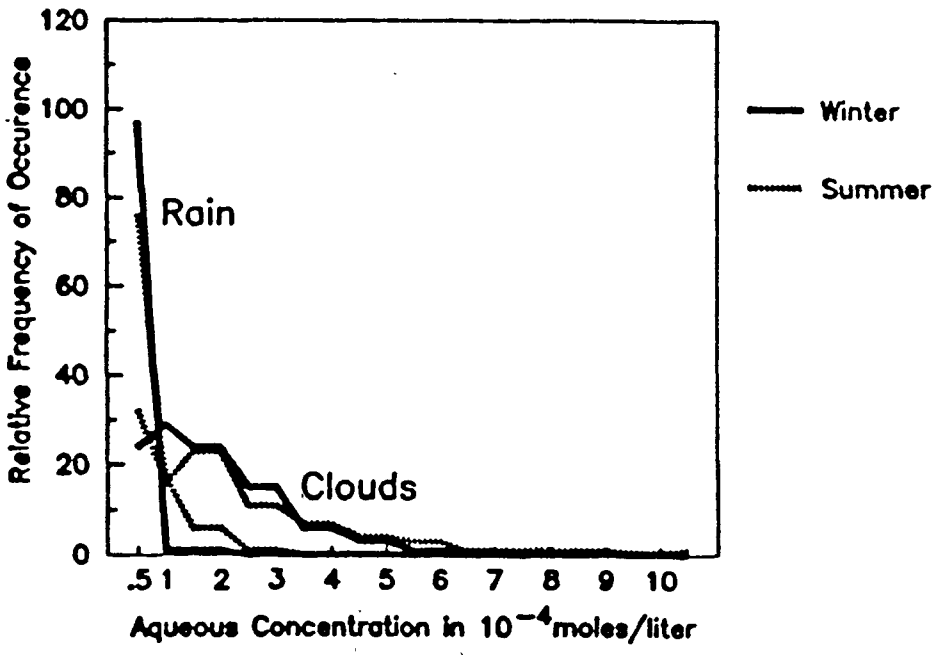


Figure 4-1 Comparison of both summer (June-Aug) and winter (Jan-Feb) SO_4^{2-} distributions for both cloud and rain water. Values shown are percent of cloud samples at the Whiteface Mountain, NY, summit MCCP site and percent of precipitation samples at the Whiteface Mountain, NY, MAP3S site.

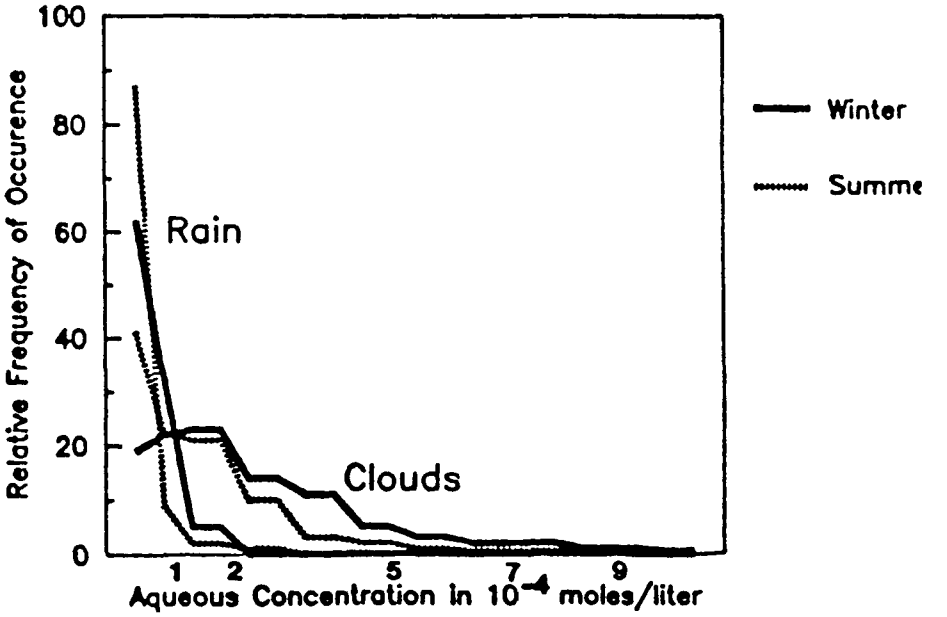


Figure 4-2 Comparison of both summer (June-Aug) and winter (Jan-Feb) NO_3^- distributions for both cloud and rain water. Values shown are percent of cloud samples at the Whiteface Mountain, NY, summit MCCP site and percent of precipitation samples at the Whiteface Mountain, NY, MAP3S site.

northwest. The mean concentrations of sulfate and nitrate during 1985 were more than twice their 1983 values. This provides some insight into the inherent seasonal variability due to meteorological conditions such as cloud and precipitation extent, air mass path and speed, and mixed layer depth (Pratt et al., 1986). The winter seasonal averages were based on individual cloud events, each of which had concentrations identified with particular meteorological conditions, but which did not respond to short-term changes in SO_2 or NO_y levels since neither has a rapid oxidation rate.

Figures 4-1 and 4-2 depict differences between winter and summer cloud water SO_4^{2-} and NO_3^- but these seasonal differences are small relative to differences between precipitation and cloud water SO_4^{2-} and NO_3^-. This suggests that at Whiteface Mountain, NY, the MCCP strategy of sampling only growing-season cloud water does not introduce uncertainties for the chemical concentrations as large as uncertainties for the LWC in winter clouds, where and how frequently cloud immersion occurs for both winter and summer, and prediction of cloud droplet deposition.

Elevational Variation

The height of cloud water sample collection above cloud base is expected to be an important source of variability in both cloud water chemistry and liquid water content. Nucleation of pre-existing aerosol, condensation of diffusing water vapor, and droplet growth occur with vertical motion of clouds overrunning mountaintops. The Junge (1963) formulation for nucleation scavenging implies that the concentration of cloud water solute varies with the inverse of liquid water content (LWC). Therefore, if LWC increases with height above cloud base, the concentrations of any solute with a substantial aerosol phase precursor should decrease with height above cloud base.

From aircraft cloud water sampling, it has been observed that liquid water content of clouds has been found to typically increase to a maximum at some level above cloud base and eventually decline toward the top of clouds, while the chemical concentrations of pollutants dissolved in cloud water usually are highest near cloud base and fall off with increasing altitude (Petrenchuk and Drozdova, 1966; Daum et al., 1984; Romer et al., 1985; Lazrus et al., 1983; ten Brink et al., 1987; Scott and Laulainen, 1979).

The Mountain Cloud Chemistry Project (MCCP) extended the time-restricted aircraft measurements through observations of entire events at two Whiteface Mountain, NY sites: Whiteface-1 (summit, 1483 m) and Whiteface-2 (Lake Placid turn site, 1245 m). The concentrations of SO_4^{2-}, NO_3^-, H^+, and NH_4^+ in 108 coincident-paired hourly cloud water samples have been compared to determine whether height above cloud base is an important determinant of cloud water acidity. Table 4-3 summarizes the data for SO_4^{2-}, NO_3^-, H^+, and NH_4^+ concentrations for all coincident-paired samples collected during 1987-88. As can be seen in the table, the mean concentrations for all four ions at the lower site (WF2) are nearly twice those recorded at the summit (WF1). To statistically compare the ion concentrations at WF1 and WF2, the ion concentration data were log transformed. It is observed that the log of each of the ion concentrations approximates a normal distribution, so a t-test can be applied to the paired differences. Table 4-3 also presents the calculated results for the paired differences of the transformed ion concentrations. For each ion, the t value is well above the critical value for rejecting the null hypothesisof equal means with a confidence level of 0.05. Reisinger (1987) analyzed two cloud events in 1986 and 1987 at Whitetop Mt. for vertical varibility of ion concentrations in cloud water with similar results.

TABLE 4-3

Ion Concentrations Statistics for Coincident-Paired Cloud Water Measurements. Results are for 1987-88 precipitating and non-precipitating clouds.

Ion	WF1 Mean µmol/l	WF2 Mean µmol/l	Number of Obs	Ave. of Paired Differences log(WF2)-log(WF1)	t Statistic
H^+	122	218	107	+0.300	-7.0
SO_4^{2-}	79	151	108	+0.470	-9.8
NO_3^-	48	85	108	+0.478	-14.7
Cl^-	3	7	108	+0.745	-9.6
NH_4^+	74	142	108	+0.593	-11.4

Figure 4-3 presents a histogram of the differences in SO_4^{2-} concentration for the simultaneous samples from two Whiteface sites (calculated as $[SO_4]$ at WF2 - $[SO_4]$ at WF1). By converting the original concentrations to differences between the two sites, substantial covariation is removed from the results, thereby allowing the elevational effect to emerge. During 92% of the sampled hours, the lower elevation site receives cloud water with higher concentrations of SO_4^{2-} than the summit (similar results were obtained for NO_3^- and H^+ differences). The analysis presented in Figure 4-3 confirmed that both sites tend to experience relatively "clean" events or tend to experience relatively "dirty" events at the same time, and that the lower site had consistently higher concentrations.

Histograms for both simultaneous and all other samples collected at the two Whiteface sites (the summit had 8 times more data) suggest that hours with $[SO_4^{2-}]$ greater than 500 µeq/L were more frequent near cloud base than at the summit (see Figure 4-3). Therefore, the occurrence of more frequent extreme events as well as higher mean values distinguished the lower (1250 m) site from the summit.

Few simultaneous LWC measurements are available for the two MCCP Whiteface Mountain sites. A comparison of Whiteface summit (WF1) and Whiteface-2 with available LWC data from the IFS site (1025 m) indicates that fewer than 10% of LWC measurements were greater than 0.45 g/m^3 at the lower (IFS) site, but that 40% of all LWC observations were greater than 0.45 g/m^3 at the summit (Mohnen, 1988c). The factor of two difference in mean LWC and SO_4^{2-} with elevation is mutually consistent and in accordance with Junge's (1963) nucleation scavenging parameterization. In-cloud oxidation of scavenged SO_2 to SO_4^{2-} could offset this effect, but the time for transport from Whiteface-2 to Whiteface-1 is small relative to the conversion rate. Thus, substantial vertical variation in cloud water concentrations at Whiteface Mountain is explained by increased dilution of preexisting aerosol as liquid water increased with height above cloud base. This agrees with the findings of Reisinger and Imhoff (1989) who measured SO_4^{2-} in interstitial air and cloud water for 14 events in 1986 at Whitetop Mt..

However, neither vertically resolved cloud water chemical data nor estimates of hourly cloud base (except Whitetop, VA) are available for the other MCCP sites. Also, a direct comparison of the MCCP Whiteface Mt. sites with the IFS site at a lower elevation was not useful due to an insufficient number of simultaneous samples. A few relatively short-term cloud and rain water data from mountain sites in Europe present mixed indications. Kins et al. (1988) reported significantly higher concentrations near cloud base in stratus and stratocumulus clouds in a study near Deuselbach, FRG. It cannot be stated that the increase in cloud water concentration near cloud base that was demonstrated for

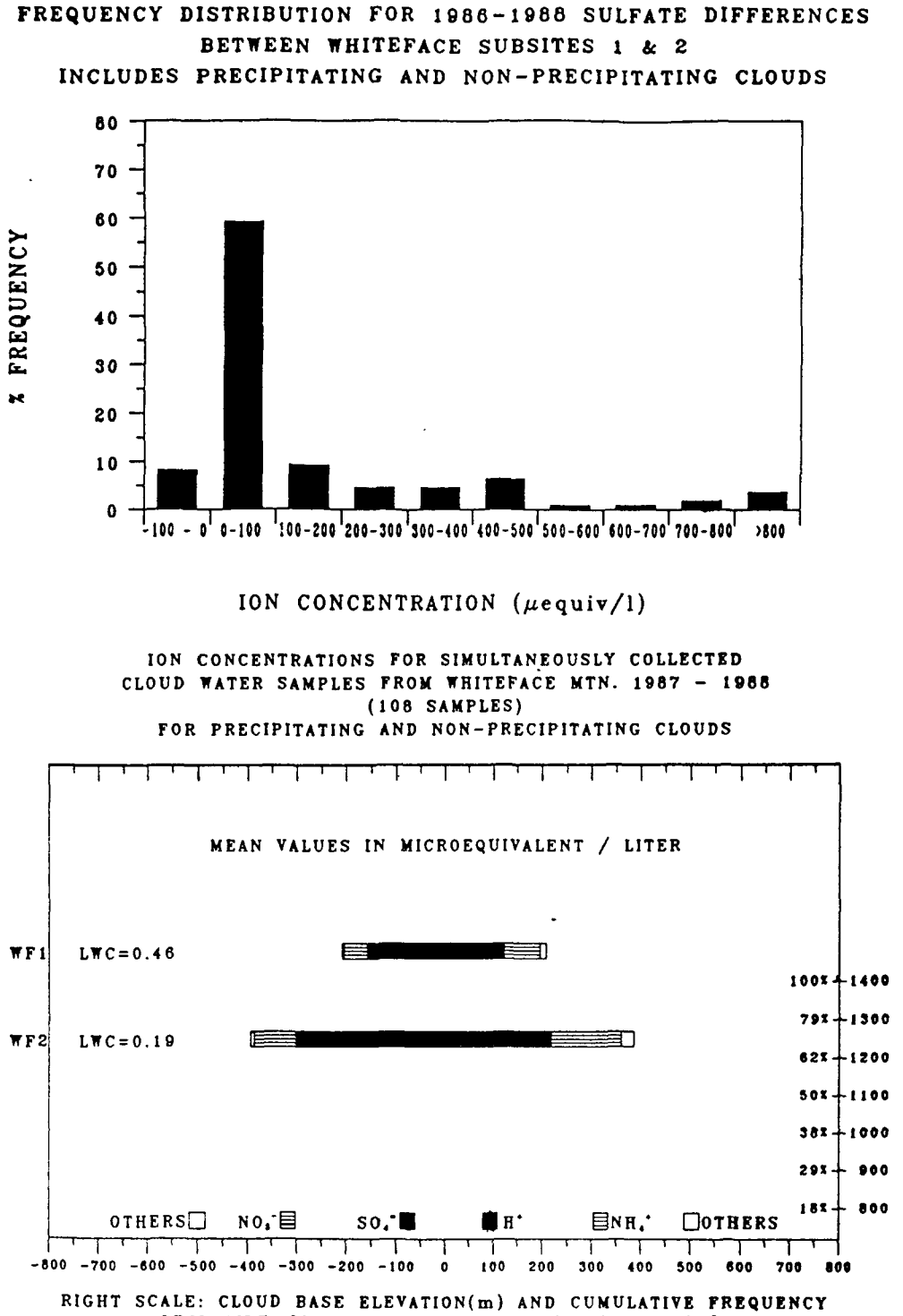


Figure 4-3 Frequency of occurrence for the differences in SO_4^{2-} concentration for the two Whiteface Mountain, NY, sites (calculated as $[SO_4]$ at WF 2 - $[SO_4]$ at WF1) for simultaneous, hourly, samples.

Whiteface Mountain must exist for other mountain sites in the eastern USA. However, it seems likely that it occurs in many locations based on the available vertically resolved aircraft and surface observations of cloud water chemistry, cloud nucleation and scavenging processes.

Data Summary

In this section, the 1986-88 results for the five high elevation MCCP sites are summarized. The overall chemical composition of the cloud water samples is presented, along with differences between precipitating and non-precipitating clouds. Extreme cloud water solute concentrations are described. Statistical modeling indicated a significant ($P < .06$) site-to-site variation in SO_4^{2-} and NO_3^- concentrations; thus the means and their frequency distributions are examined to identify any spatial patterns in the data.

Figure 4-4 presents the unweighted mean concentrations of H^+, SO_4^{2-}, NO_3^-, Cl^- ("other" anion), and NH_4^+, and "other" cations (sum of Ca^{2+}, Mg^{2+}, K^+, and Na^+) for all MCCP cloud samples taken during the 1986-88 growing seasons (generally May to October). The samples were primarily sulfuric acid with substantial amounts of the two nitrogen species but relatively low concentrations of other anions and cations. The samples were in approximate charge balance, suggesting that the major ionic species have been measured on the average.

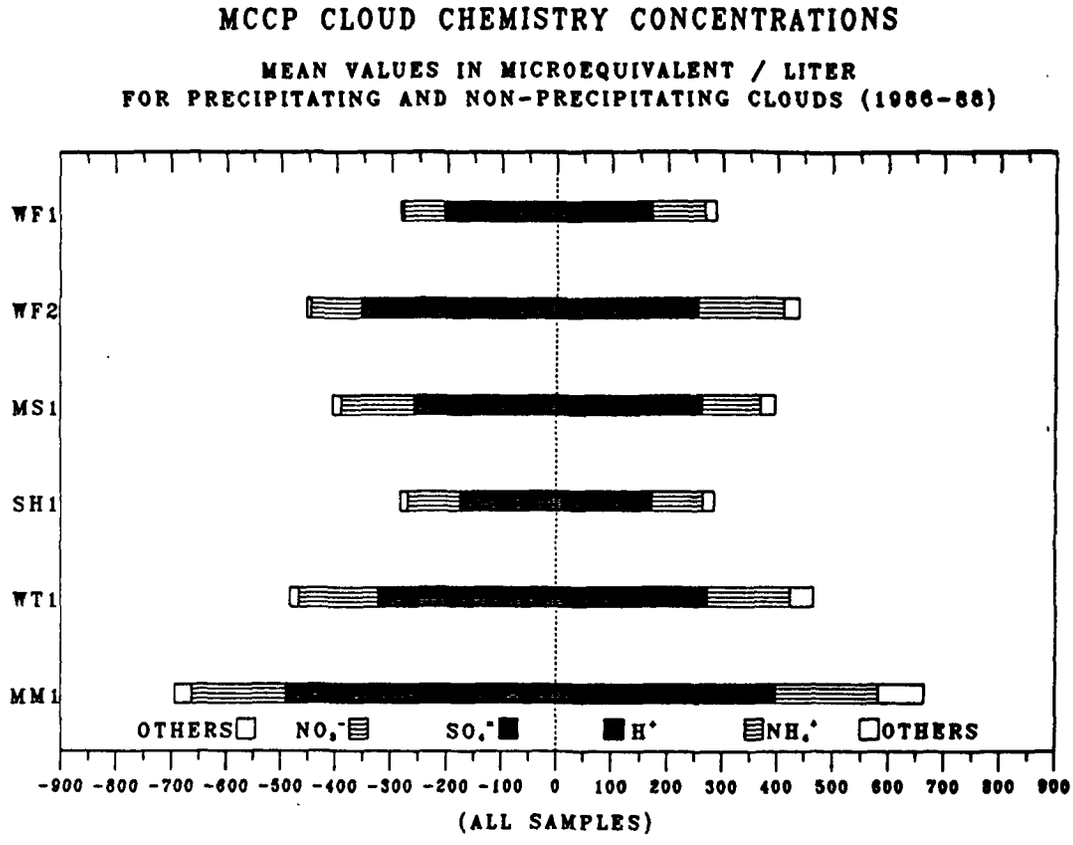
Figure 4-5 classifies the data as precipitating and non- precipitating clouds. The concentrations in non-precipitating clouds were much higher than in precipitating clouds, as was noted by other investigators (Falconer and Falconer, 1980; Schemenauer, 1986; 1988).

It is suggested that MCCP measurements may overestimate liquid water content (LWC) for precipitating clouds. Falconer and Falconer (1980), Schemenauer (1986; 1988), and Mohnen and Kadlecek (1989) suggest that most cloud water samplers do not completely exclude rain drops (especially the ASRC sampler). Therefore, at this time, neither LWC nor cloud water chemical concentrations have been accurately characterized for precipitating clouds.

Non-precipitating cloud data are probably more important than precipitating cloud data for determining forest canopy exposure and cloud water deposition because they represent chemical inputs during a time period when the precipitation is not occurring. However, substantially increased concentrations in precipitating cloud water over precipitation (as collected under the NADP/NTN protocol), coupled with increased water deposition due to cloud interception by the forest canopy, suggest that precipitating clouds contribute to total deposition in as yet undefined quantities. Non-precipitating clouds must be handled separately in cloud water deposition calculations.

Figures 4-6 through 4-8 present the distributions of SO_4^{2-} ion for non-precipitating cloud water at six sites (both Whiteface summit and Whiteface-2 are included). A substantial number of hours at each site (15% to 40%) had cloud water H^+ and SO_4^{2-} concentrations greater than 500 μeq/L, which we define as an "extreme" value. Distributions for H^+, NO_3^-, and NH_4^+ are similar in shape to those for SO_4^{2-}, but are dispersed around different mean values as given in Table 4-4. The "extreme" SO_4^{2-} values were evenly distributed between concentrations of 500 to 1000eq/L. "Extreme" NO_3^- and NH_4^+ are defined as those greater than 250 μeq/L; these values were evenly distributed between concentrations of 250 to 500 μeq/L. Mohnen (1988c) presented the distributions of all ions.

Sulfate is a very important ion in the determination of cloud water acidity. For example, the "extreme" concentrations of SO_4^{2-} alone in non-precipitating clouds (500 μeq/L) would support a pH of 3.3; this type of calculation defines the "potential acidity" that would occur if no other cations or anions were present in the cloud water solution. The near-maximum value of 1000 μeq/L [SO_4] observed at most MCCP sites translates to a potential acidity of pH 3.0. Cloud water SO_4^{2-} concentrations over



SITE	SITE LOCATION	ELEVATION (m)
WF1	Whiteface Mountain, NY	1483
WF2	Whiteface Mountain, NY	1245
MS1	Mount Moosilauke, NH	1000
SH1	Shenandoah, VA	1015
WT1	Whitetop Mountain, VA	1689
MM1	Mount Mitchell, NC	1950

Figure 4-4 Mean concentrations of H^+, SO_4^{2-}, NO_3^-, other anions (Cl^-) NH_4^+, and other cations (sum of Ca^{2+}, Mg^{2+}, K^+, and Na^+) for all MCCP cloud samples for 1986-88.

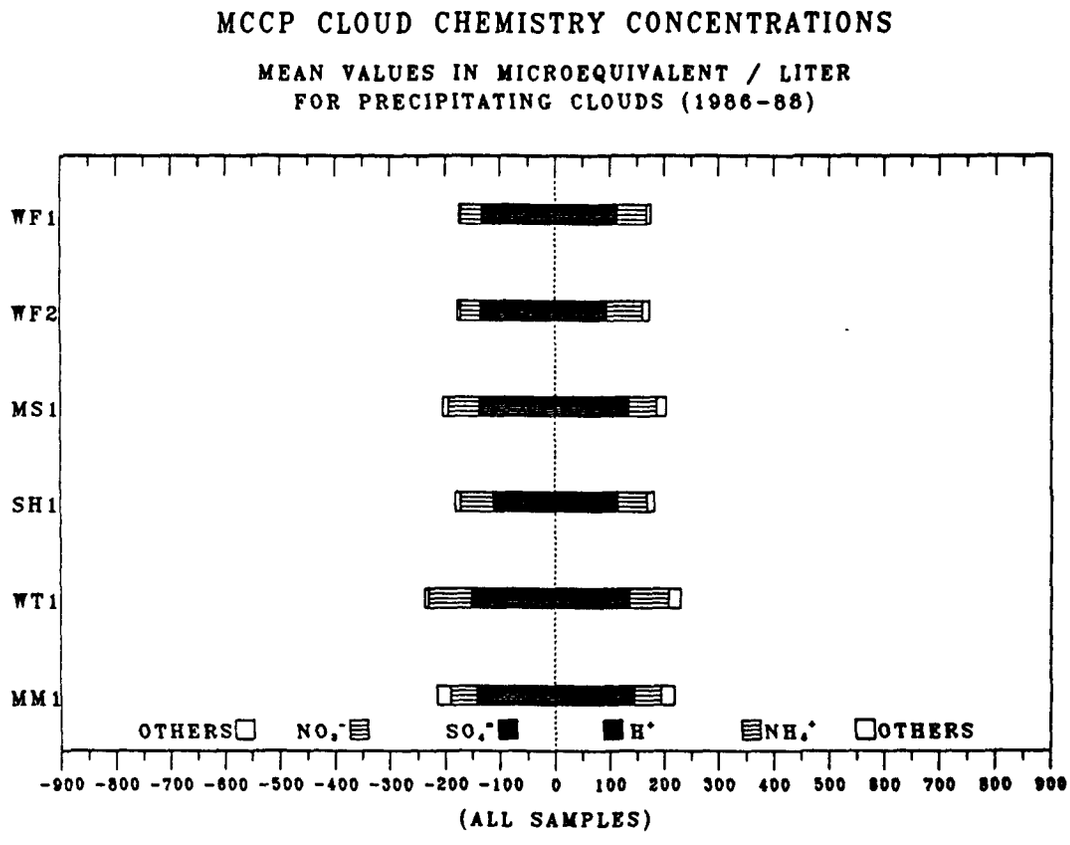
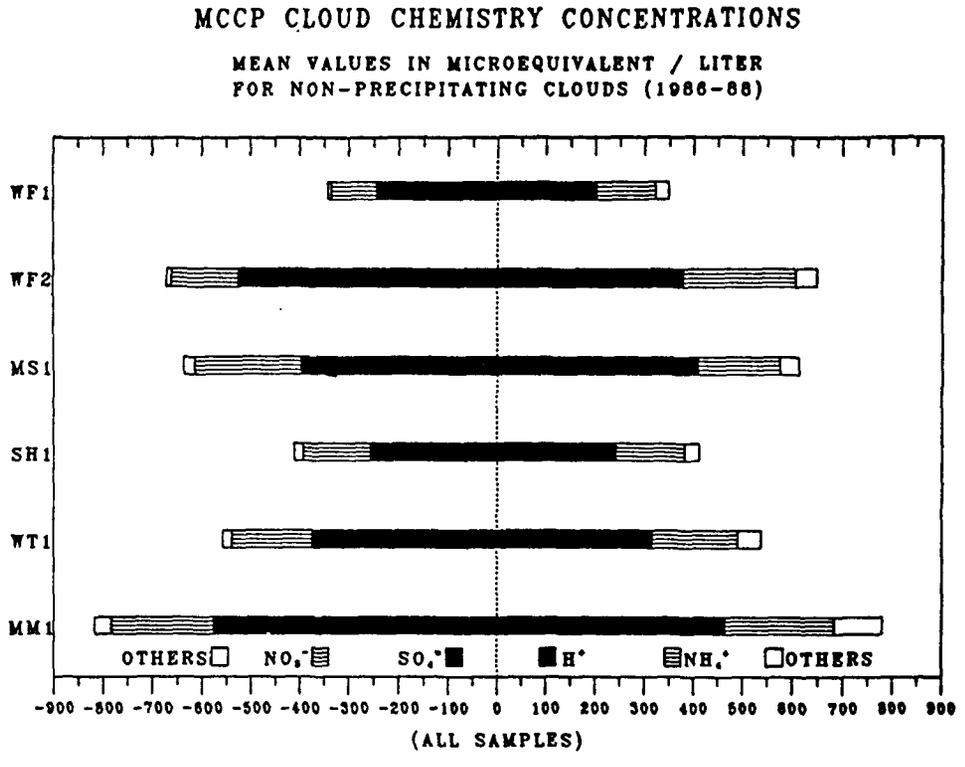
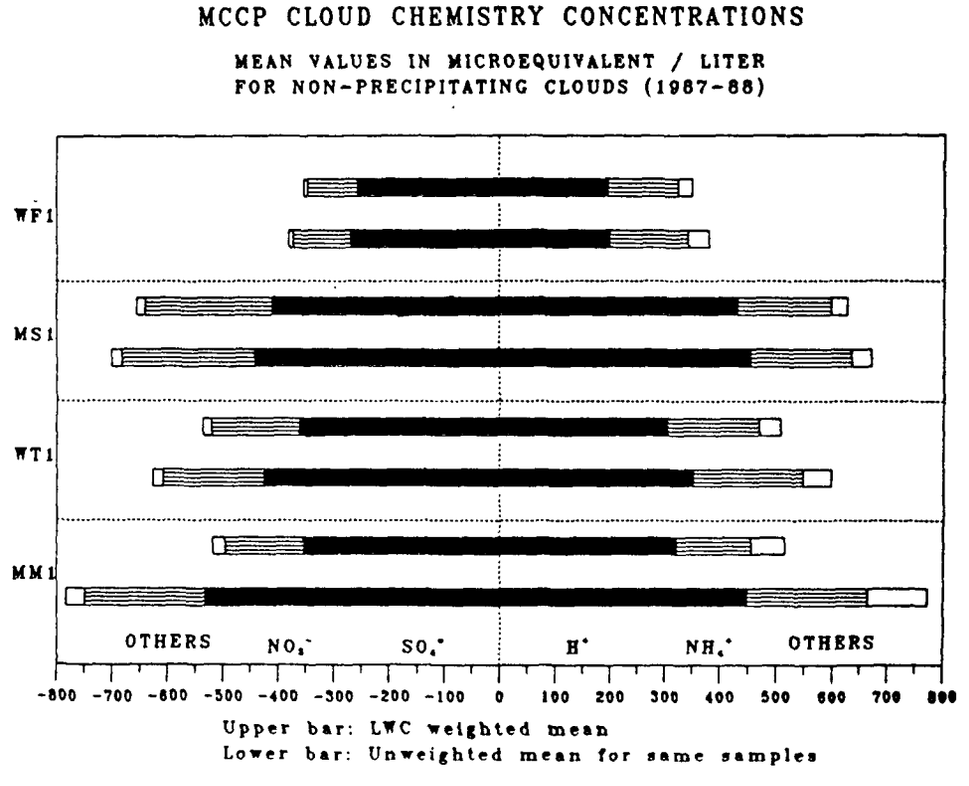
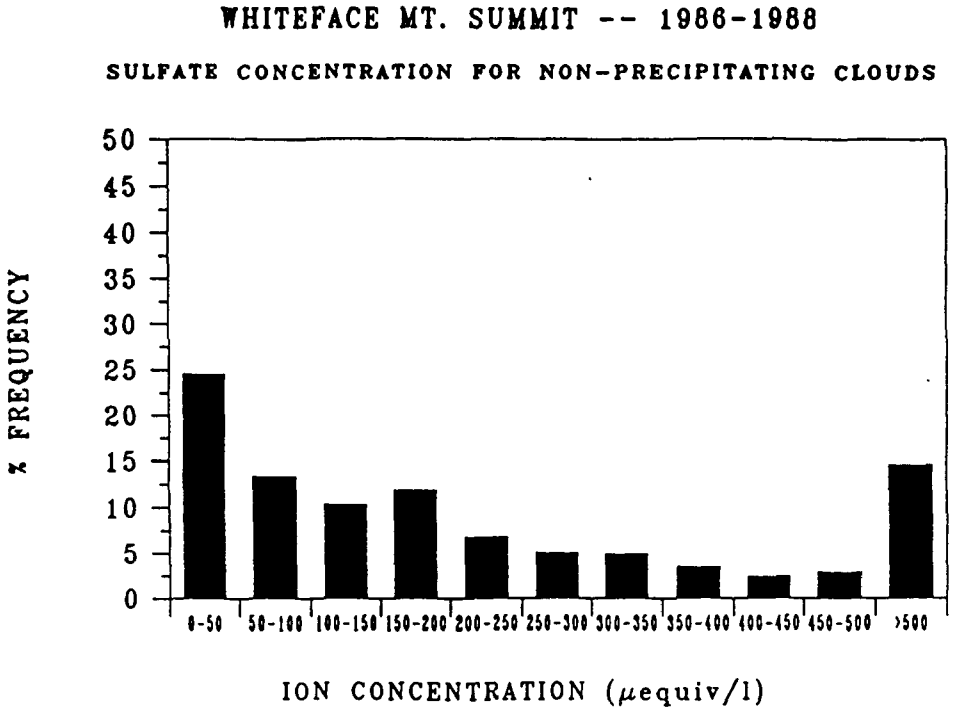


Figure 4-5 Mean concentrations of H^+, SO_4^{2-}, NO_3^-, other anions (Cl^-), NH_4^+, and other cations stratified by precipitating and non-precipitating cloud type, for all MCCP samples, 1986-88.

Figure 4-5 (Continued)







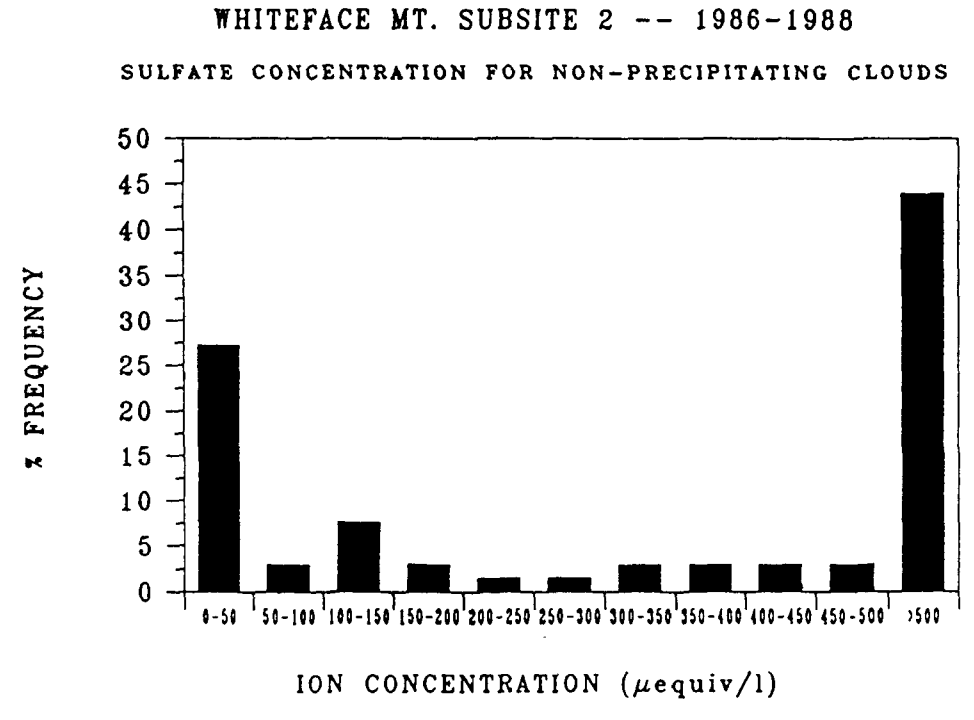
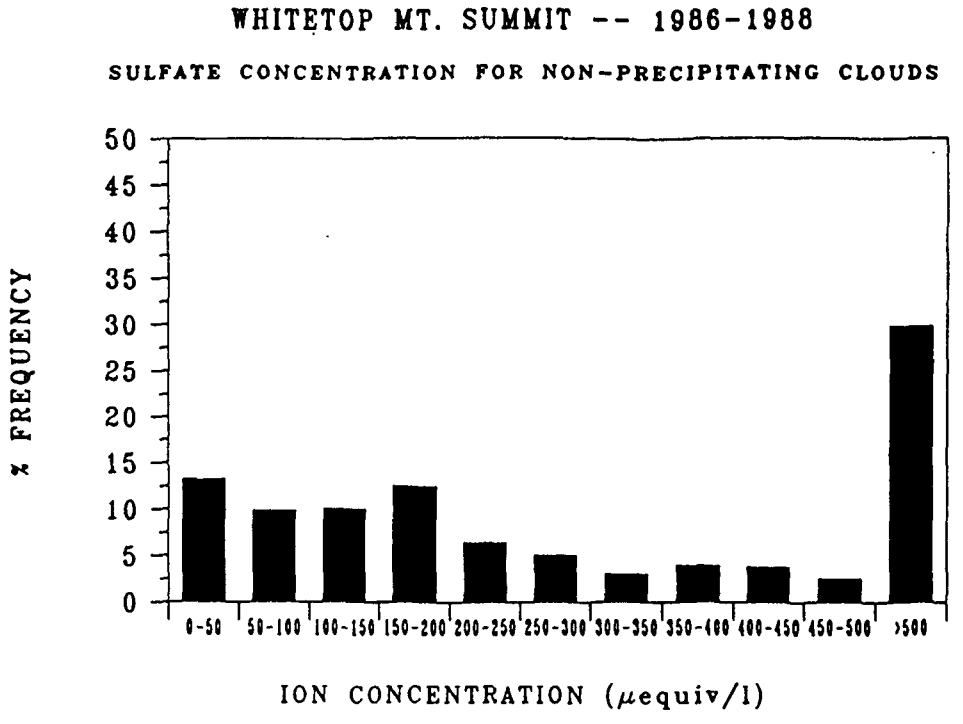


Figure 4-6 Frequency of occurrence for hourly SO_4^{2-} ion in non-precipitating cloud water at Whiteface-1 (summit) and Whiteface-2, NY.



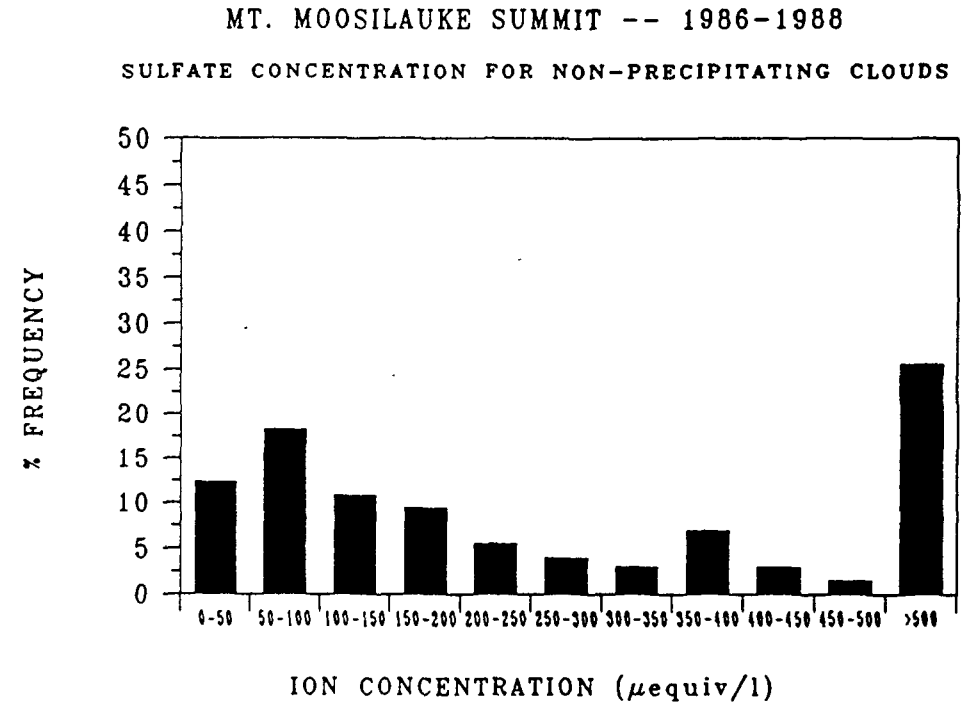


Figure 4-7 Frequency of occurrence for hourly SO_4^{2-} ion in non-precipitating cloud water at Whitetop, VA, and Moosilauke, NH.

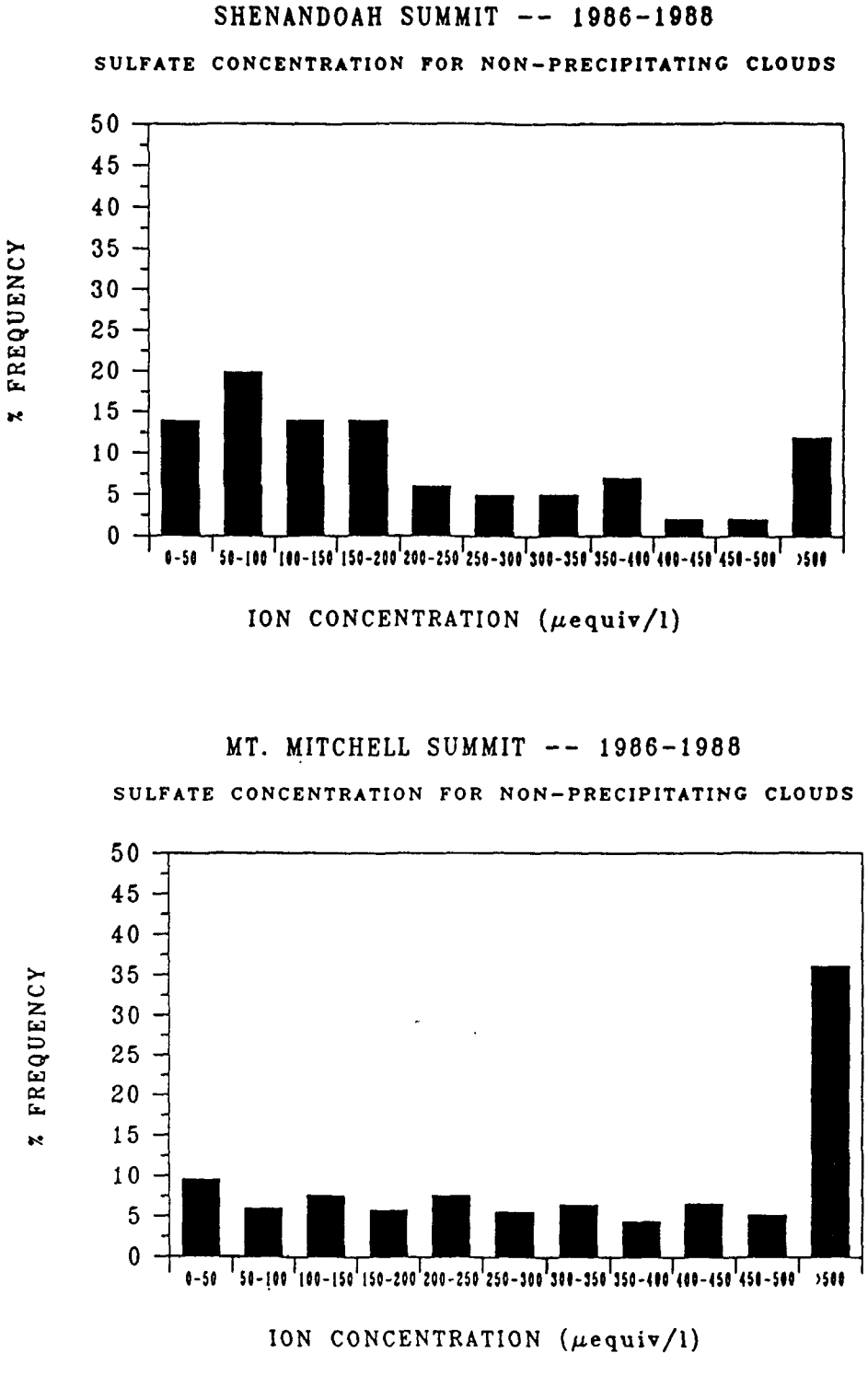


Figure 4-8 Frequency of occurrence for hourly SO_4^{2-} ion in non-precipitating cloud water at Shenandoah, VA and Mt. Mitchell, NC.

2000 μeq/L occur, but rarely. A few hourly cloud water samples of about pH 2.8 were observed at some MCCP sites during most warm season sampling campaigns. Cloud water with pH of 3.0 was routinely sampled at the MCCP sites.

The "extreme" concentrations for H^+, SO_4^{2-}, NO_3^-, and NH_4^+ were observed to occur most frequently at the beginning of a cloud event when LWC was low or at the end of some events where LWC was rapidly decreasing; an increase in the sulfate-to-nitrate ratio sometimes co-occurred with this decrease in LWC. This is consistent with the evaporation of liquid water, concentration of cloud water solutes, and volitization of HNO_3 during the dissipating cloud stage. This qualitatively supports calculations by Unsworth (1984) and Milne et al. (1988) which suggest that evaporation on both the sampler and the forest canopy can lead to concentrations that are enhanced over what is advected to the canopy.

TABLE 4-4

Average Ion Concentrations (μmol/l) for Both Precipitating and Non-Precipitating Clouds for 1986-1988 at Six High-Elevation Locations Monitored by MCCP.

Site	H^+	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	# of hours
Mitchell, NC	398	489	174	31	184	624
Moosilauke, NH*	263	257	132	16	107	328
Shenandoah, VA	171	176	94	13	93	230
Whiteface-1, NY	171	205	73	5	97	987
Whiteface-2, NY*	255	352	92	7	157	120
Whitetop, VA	174	321	144	16	152	656

* Did not collect cloud water during 1986
** See Figures 4-6 through 4-8 for the sulfate ion concentration distributions in non-precipitating clouds

Chloride (presented in Table 4-4) and sodium concentrations (not presented) suggest that the seasalt contributions to the sulfate means are minimal. The MCCP cloud water data are well approximated by considering only the four major ions. The sum of cation concentrations is useful primarily for identifying neutralization of H^+ in individual samples by crustal-like material.

It is difficult to extrapolate from these overall concentration means and distributions to other high elevation forests in the eastern USA. The two southern-most sites, Mt. Mitchell, NC, and Whitetop, VA, have the largest mean H^+, SO_4^{2-}, NO_3^-, and NH_4^+ concentrations and the greatest frequency of extreme values (defined here as > 500 μeq/L) (greater than 30% of hours), followed by Moosilauke, NH, with intermediate values for mean concentrations and 25% of samples representing extreme values. The Whiteface summit (site 1), NY, and Shenandoah, VA had the lowest means and less than 15% of samples constituting extreme H^+ and SO_4^{2-} concentrations.

The Whiteface-1, NY and Shenandoah, VA sites display unusual characteristics when LWC and cloud immersion frequency, respectively, are compared to those of the other MCCP sites. Therefore site-to-site differences among the remaining MCCP sites are considered. When the data for the Whiteface-2, NY (1250 m) site are compared to those for the site at Moosilauke, NH, a picture of cloud water chemistry emerges that is consistent with the results of the Canadian CHEF program (Schemenauer, 1986; 1988). When compared to the northern Appalachian sites, the MCCP data for

Whitetop, VA, and Mt. Mitchell, NC, and IFS data for a site in the Great Smoky Mountain Park, NC (Lindberg et al., 1988) suggest higher mean H^+, SO_4^{2-}, NO_3^-, and NH_4^+ cloud water concentrations for southern Appalachian sites.

The dependence of cloud water concentrations on synoptic weather type and observed differences in the relative frequencies of cap cloud, warm sector, marine flow, and post-cold frontal synoptic types between northern and southern MCCP sites suggest that the north/south differences in cloud water concentrations may be related to cloud climatology. The southern sites frequently are cloudy under the stable, warm-sector synoptic type (high concentrations), while the northern sites experience cloudiness associated more frequently with frontal passage (lower concentrations). Additionally, when air mass trajectories shift from southwest to northeast at the northern sites, the concentrations of H^+, SO_4^{2-}, NO_3^-, and NH_4^+ normally decrease while the southern sites receive high concentrations under northwest flow.

FREQUENCY OF CLOUD

The frequency of cloud immersion, the mass concentration of liquid water (LWC), and its distribution across droplet sizes are important for understanding chemical fluxes to a forest canopy. Collectively, these data describe the presence of liquid water in the atmosphere.

High-elevation forests of the Appalachian Mountains are often immersed in cloud. Previous estimates of cloud impaction frequency on mountain summits in the southern Appalachians range from 30% of all hours in the Great Smokey Mountains to 40% at Mt. Mitchell (USFS, Northeastern Forest Experimental Station, personal communication). Estimates for the northern Appalachians are 50% at Whiteface (Nicholson and Scott, 1969), 55% at Mt. Washington (NOAA, 1975) and 40% at the summit of Mt. Moosilauke (Lovett et al., 1982). Siccama (1974) estimated that the upper slopes of the Green Mountains are in clouds 30% to 50% of all hours. Many of these estimates were derived from visual observations as a biproduct of forest ecosystem research and do not represent actual cloud measurements. The following material addresses recent regional and mountaintop studies dedicated specifically to cloud.

Regional Cloud Studies

Warren et al.(1986) developed global cloud climatology statistics from surface airport observations for the 1971-1981 period on a 5 deg. latitude by 5 deg. longitude grid. Over the eastern U.S., the low cloud types (<2500 m) in order of decreasing average cloud amount are: stratus/stratocumulus, cumulus and cumulonimbus. The combined mean sky cover (Figure 4-9 - Appendix E) for the three low cloud types ranges geographically from 24% to 46%, with the highest values found over the northern portions of the eastern U.S. The stratus/stratocumulus type is strongly dominant, accounting for at least 75% of the overall low cloud amount. The average cloud base height for this class (Figure 4-10 - Appendix E) is generally in the range of 700-900 m. Cumulus clouds have higher average bases, typically between 1000 m and 1200 m; cumulonimbus bases average in the 800-1000 m range.

A 21-year study (1965-1985) of low cloud observations from 12 eastern U.S. airports located near five of the MCCP sites found that the incidence of low cloud (<2134 m) exceeded 40% at all locations with the highest values in the north (Mohnen, 1988). In the vicinities of the southern MCCP sites of Whitetop, Shenandoah, and Mitchell, low clouds were reported 42% to 49% of the time. In the northern Appalachians near Whiteface and Moosilauke, the occurrence ranged from 44% to 63%. Cloud base height at all locations was most frequent in the 900 m to 1200 m range.

The U.S. Air Force Real-Time Nephanalysis (RTNEPH) global cloud database for low clouds (<2.1 km) was examined for the 1985-87 period for a large portion of the Appalachian Mountain region (southwest Virginia north to southeastern Canada). The trend over this period was for decreasing cloud frequencies, consistent with the expanding drought in the southeastern U.S. Figure 4-11 (Appendix E)illustrates this trend and shows the dominance of stratiform cloud types (in agreement with Warren et al., 1986). Cloud base height was found to average between 700 m and 900 m. After taking into account cloud thickness as well as cloud base, the mean probability of low cloud as a function of elevation was found to be greatest between 900 m and 1300 m (Figure 4-12 - Appendix E). Approximately 30% of low clouds were precipitating: 19% in summer, 42% in winter.

An examination of the individual RTNEPH grid cells nearest three MCCP sites indicates local variability. Near Moosilauke (Figure 4-13 - Appendix E), cloud is most likely to occur between 1000 m and 1300 m. Near Whiteface (Figure 4-15 - Appendix E), highest cloud probabilities are within the broad range of 800-1600 m. Near Shenandoah (Figure 4-14 - Appendix E), cloud probability is lower than at either Moosilauke or Whiteface but is relatively strong between 800 m and 1300 m and strongest between 1000 m and 1100 m. It should be kept in mind that these cloud height values apply to relatively large-scale cloud elements in the general vicinity of the referenced mountains and are not necessarily representative of site-specific cloud elevations.

The continuity of cloud heights across the Appalachians is shown in Figure 4-16. This figure presents a cross-section of a west-east transect of cloud probability (according to the RTNEPH database) across the northern Appalachians in summer (June-August, 1985-87 averages) at a latitude of approximately 44.5 degrees N. The cross-section represents a height of 0 to 2 km and a width of 70 deg. W to 78 deg. W longitude. The diagram indicates a zone of maximum cloudiness at a height of 800-1000 m from southern Ontario province across the northern Adirondacks into the Green Mountains of Vermont. The zone lifts to near 1200 m across New Hamphsire and to near 1600 m over southwestern Maine. A second transect (not shown) approximately 85 km south of the first one spanning the central Adirondacks and the Mt. Moosilauke area, exhibits the same trend across New York, Vermont and New Hampshire. This suggests that the preferred zone of cloudiness over the high mountain regions of the northern Appalachians is between 800 m and 1200 m with a tendency for clouds in the lower portion of the range over the Adirondacks and Green Mountains. In the central and southern Appalachians, the results of a single transect analysis across eastern West Virginia through the Shenandoahs of Virginia suggest a slightly higher zone of preferred cloudiness--1000 m to 1400 m--over the high mountains of West Virginia, but a lowering of the zone into the 700-1000 m range east of the Appalachian's major axis.

Site Specific Measurements

Table 4-5 presents the percent frequency of cloud impaction for the combined 1986-88 field seasons for the five MCCP summit sites. The top half of the table indicates that cloud frequencies range from 11% to 37% of all hours during the summer season among the sites. Due to the differences in station elevation, latitude, and local topography, it is not possible to discern spatial trends in these results. Temporally, the general decline in cloudiness from 1986 to 1988 is reflective of the drought conditions which prevailed over the southeastern U.S. and the shift from cool, wet weather in 1986 in the north to drought conditions in 1988. Approximately 25% of the cloud events were precipitating. The bottom half of the chart indicates that, on a daily basis, clouds occur frequently: 32%-42% of the days experienced cloud at the two lowest elevation sites, and 68% to 77% of the days on the three higher mountains.

In an attempt to put the cloudiness of the MCCP field seasons into the perspective of long-term climate, the frequency of low-level (<2100 m) clouds was determined for eight local National Weather

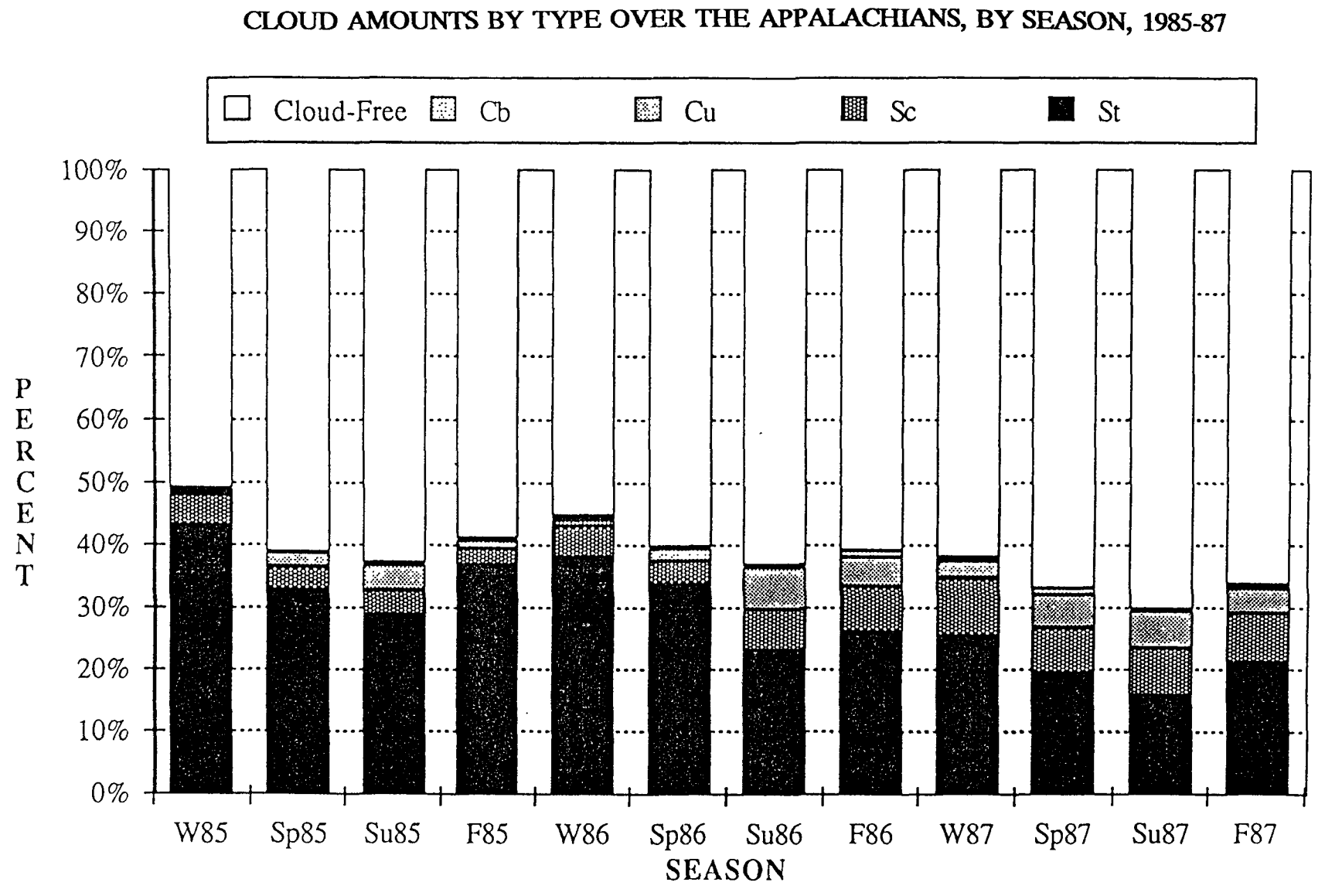
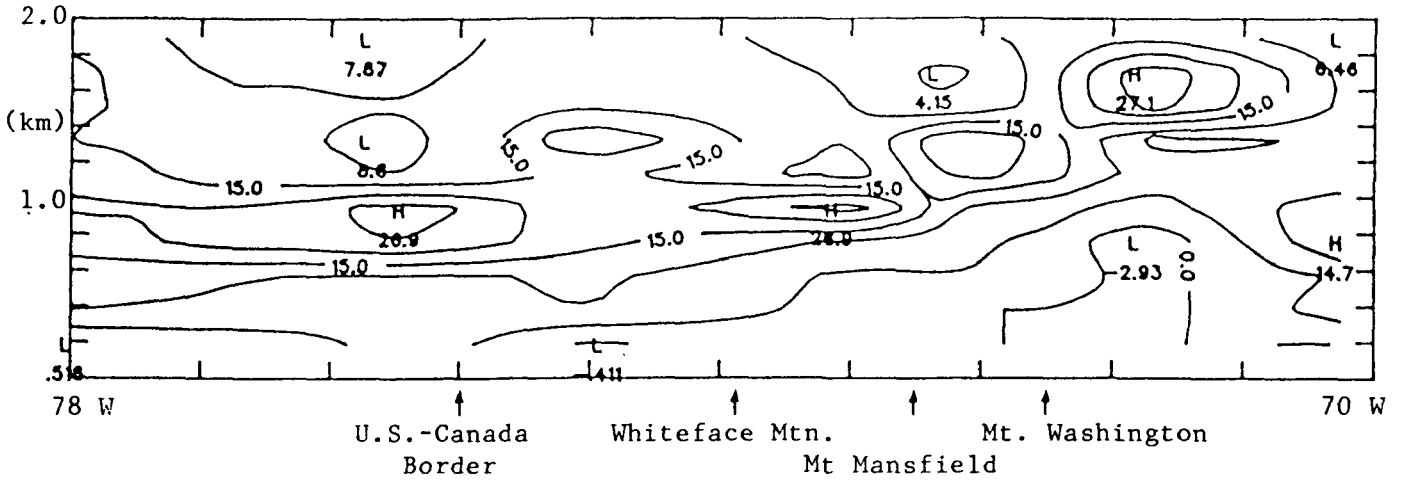

CLOUD AMOUNTS BY TYPE OVER THE APPALACHIANS, BY SEASON, 1985-87
Cloud-Free
Cb
Cu
Sc
St
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
PERCENT
W85
Sp85
Su85
F85
W86
Sp86
Su86
F86
W87
Sp87
Su87
F87
SEASON

Figure 4-11

Summer (1985-87) Probability of Cloud Occurrence (RTNEPH data) for the West-East Transect Across the Northern Appalachians at Approx. 44.5 deg. N Latitude. Cross-section Represents a Height of 0 to 2 km and a Width of 70 deg. W to 78 deg. W.



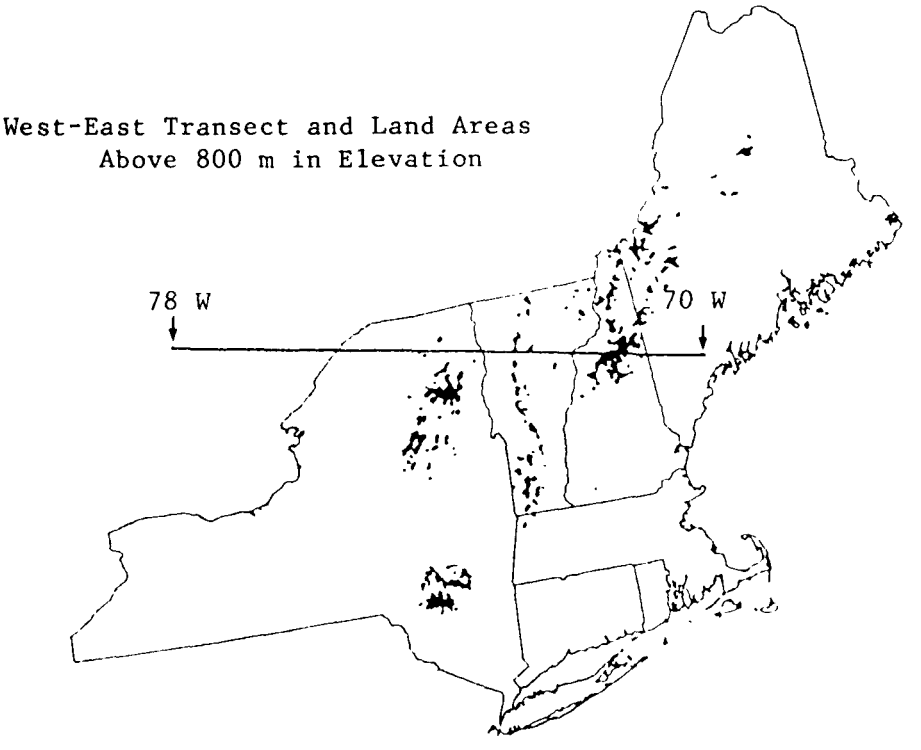


Figure 4-16

Service airports for the same time period and compared with a 21-year climatology (1965-85) previously developed for these airports. The results are presented in Figures 4-17 through 4-19 (Appendix E) which show the percentage departures from normal for each of the three field seasons. In 1986, positive anomalies occurred in the Northeast, near normal conditions existed in the central Appalachians and below normal cloudiness occurred in the southern Appalachians. During the 1987 field season, above normal cloudiness was again observed in the Northeast but values generally were not as high as those in 1986. Below normal cloudiness was observed throughout the central and southern Appalachians. In 1988 the dryness spread into the Northeast where lower than normal cloud amounts were observed, especially during the early portions of the field year. In southern regions, cloud frequencies were once again below normal with the exception of one airport (Asheville, NC) which had a normal frequency. These airport trends are consistent with the 3-year trends observed among the MCCP sites. This finding suggests that the northern MCCP sites experienced above-normal cloudiness in both 1986 and 1987 and below-normal cloudiness in 1988. It also suggests that the southern sites experienced below-normal cloudiness during all three field seasons. What is not known, however, is the impact that large scale trends in cloudiness have on localized, orographically-induced clouds.

TABLE 4-5

Cloud Frequency at MCCP Sites, June to September, 1986-88.

Site	Elevation (m)	1986	1987	1988	Mean
		Cloud frequency (% of hours in cloud)			
Whiteface, NY	1483	45	40	25	37
Moosilauke, NH	1000	25	21	6	19
Shenandoah, VA	1015	18	7	6	11
Whitetop, VA	1689	38	28	26	30
Mitchell, NC	1950	35	28	23	29
		Percentage of total days experiencing some cloud			
Whiteface, NY	1483	79	80	73	77
Moosilauke, NH	1000	52	51	22	42
Shenandoah, VA	1015	46	25	25	32
Whitetop, VA	1689	76	67	62	68
Mitchell, NC	1950	84	75	68	76

Cloud base observations taken at Whiteface and Whitetop (Figures 4-20 and 4-21 - Appendix E) indicate that the frequency of cloud at approximately the 1000 m elevation level is about 10% at Whitetop and 15% at Whiteface. Taken together with the direct station measurements at Shenandoah and Moosilauke which are both at or near the 1000 m level, a north-south trend in cloud frequency becomes apparent: 10-11% frequency at approx. 1000 m in the south and 15-19% frequency in the north.

Diurnal cloud activity indicates a preference for cloud impaction at night (7 pm - 7 am) on mountaintops, as previous MCCP reports have illustrated (Mohnen, 1988c). Some sites experience more than twice as much cloud during the early morning than the afternoon. Moosilauke and Shenandoah did not exhibit such a diurnal trend, probably due to their lower elevations and their relative position downwind from other mountains. In contrast to the mountaintop trends, regional airports and the

RTNEPH database exhibited more cloudiness during daylight hours. This suggests that mountain cloudiness at night is localized and probably linked to orographic mechanisms.

Land Areas Susceptible to Cloudwater Deposition

There is general agreement from regional and site-specific cloud studies in the eastern U.S. that elevations above 800 m are frequently exposed to cloud. Based on this agreement, the location and area of land above 800 m was determined. Figure 4-22 depicts these land areas which total 25 million hectares. Table 4-6 lists total land area for 14 states above five elevations: 800 m, 1000 m, 1200 m, 1400 m and 1600 m.

TABLE 4-6

State Totals for Land Area (hectares x 1000) Above 800 m, In 200 m Increments

State	800-1000m	1000-1200m	1200-1400m	1400-1600m	>1600m	Total
GA	883	149	9	0	0	1041
KY	216	50	1	0	0	267
MA	7	0	0	0	0	7
MD	477	0	0	0	0	477
ME	548	74	9	*	0	631
NC	570	346	151	51	15	1133
NH	718	210	56	20	3	1007
NY	1091	175	33	*	0	1299
PA	255	0	0	0	0	255
SC	75	*	0	0	0	75
TN	1215	698	238	79	56	2285
VA	5670	356	154	33	2	7214
VT	420	30	0	0	0	451
WV	5794	2586	541	1	0	8922

* less than 500 hectares

Land areas based on Defense Mapping Agency digital elevation data. Elevation values for every 30 seconds of latitude and longitude.

Land area calculations by K. Hermann of NSI Technology Services Corporation.

LIQUID WATER CONTENT

Droplet Size

An important parameter in cloud droplet deposition modeling is droplet size because it affects the collection efficiency of the canopy. At any given time, a range of droplet sizes will exist in a cloud. The diameter of a particular droplet depends on its time history. Factors affecting droplet size include the size, composition, and air concentration of cloud condensation nuclei (CCN), the length of time droplets are exposed to supersaturated air, the degree of supersaturation and, in "supercooled" clouds,

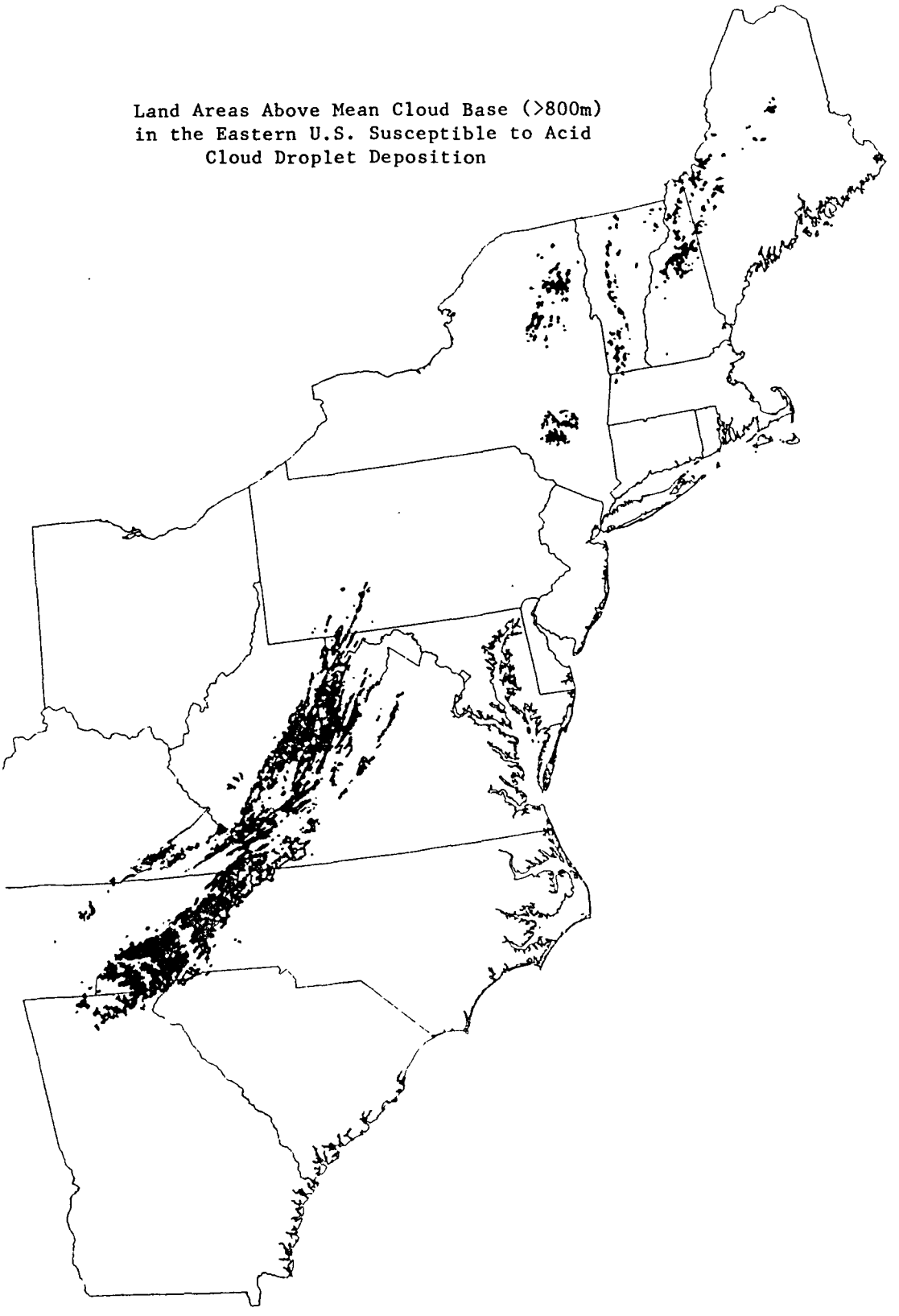

Land Areas Above Mean Cloud Base (>800m)
in the Eastern U.S. Susceptible to Acid
Cloud Droplet Deposition

Figure 4-22

the concentration of ice crystals. Entrainment of dry air can affect droplet sizes because it lowers the saturation ratio and introduces additional unactivated CCN into the cloud. The coalescence of droplets of different sizes is generally not important for non-precipitating clouds overriding mountaintops, although precipitation can reduce cloud droplet concentrations appreciably. These and other processes influencing droplet size are described in detail by Byers (1965), Mason (1971), and Rogers (1976), among others. Droplet size distributions are highly variable in time and space.

In continental clouds, droplet concentrations tend to be higher, the mean droplet diameter tends to be smaller, and the size spectrum tends to be narrower than in marine clouds (Mason, 1971). A survey of cloud droplet size data by Mason (1971) showed that the modal diameter of the droplet size distribution was usually 5 to 15 μm for many non-precipitating continental clouds and 15 to 30 μm in non-precipitating marine clouds, with the difference due mainly to the availability of CCN.

Droplet size spectrum in clouds impacting elevated terrain have been measured using a Forward Scatter Spectrometer Probe (FSSP) at three MCCP sites: Mt. Mitchell, NC (1950 m), Whiteface Mountain, NY (1483 m), and Whitetop Mountain, VA (1689 m). Clouds impacting the sites form in airmasses that are mostly continental in nature and are characterized by relatively high CCN concentrations. Data have been collected in precipitating and non-precipitating frontal and cap clouds. DeFelice (1988) presents cloud droplet spectrum for the Mt. Mitchell, NC, site. Table 4-7 summarizes results of the FSSP measurements at Whitetop Mountain, VA. A description of the FSSP instrument and its measurement uncertainties is given in Section 6 of this report.

TABLE 4-7

Summary of Cloud Droplet Size Distributions at Whitetop Mountain, VA.
Data were collected during five mainly post-frontal cloud events in October, 1987 (Valente et.al., 1989).

	Mean	Std Deviation
Number of observations	903	-
Averaging time (min)	5	-
Liquid water content (g/m^3)[a]	0.21	0.1
Volume mean diameter (μmol/l)	11.9	4.
Number concentration ($\#/cm^3$)	388	221.

[a] Measured with FSSP

A joint intensive effort was made by Rudi Pueschell's group at NOAA-ERL and SUNY-Albany ASRC during the summer of 1982 at Whiteface Mountain, NY to document in detail the aerosol/cloud spectrum prior, during, and after cloud occurrence. LWC was calculated by integrating over individual size distributions (Mohnen, 1987) and size distributions were lumped together into four categories (Figure 4-23 - Appendix E). These were used by MCCP as input for the cloud water deposition model (described later in this report).

For each case, a plot of the number distribution and a statistical summary for each mode of the cloud droplet distribution (gamma distribution, one to three modes) were provided. One goal was to establish a relationship between liquid water content of clouds and cloud droplet size distribution. If cloud droplets below 2 μm radius are disregarded, the remaining cloud spectrum shows a distinct trend with liquid water content: total cloud droplet number concentration increases slightly and the slope of the size distribution towards large particles becomes less steep; that is, it "opens" towards larger cloud

droplets. It therefore appears possible that a set of "standardized cloud spectrum" could be defined that correspond to liquid water content classes, such as < 0.05, 0.05-0.1, 0.1-0.25, 0.25-0.40, 0.4-0.6, and > 0.6 g/m^3.

MCCP's approach was then to tentatively assign a cloud droplet spectrum to a particular range of liquid water content and test and refine this relationship at Whitetop during the field intensives of October 1987 and June 1988. The outcome of these combined efforts was an MCCP-wide recommendation for droplet spectrum to be incorporated in the modified Lovett model (Mueller, 1990) for estimation of cloud interception/deposition. The initial cloud droplet size spectrum with assigned LWC values is presented in Figure 4-23 (Appendix E). The 1982 Whiteface Mountain LWC data were in general agreement with LWC values obtained directly during the 1987-88 field seasons using the MCCP Valente device. The results of this investigation suggest that the MCCP approach should provide satisfactory cloud deposition estimates from a model that inputs measured LWC values and "standardized" cloud droplet spectrum, with cloud droplet spectrum from Whiteface Mountain and Whitetop Mountain field experiments. The sensitivity of these conclusions to the known variability of drop size distribution (Paluch, 1988; Fuzzi et al., 1983; Pruppacher and Klett, 1980) was not calculated.

Information on the liquid water content in clouds impacting the five mountain summits is summarized in Table 4-8 for the growing season (April-October). The MCCP filter technique described by Valente et al. (1989) was used. All data represent hourly sampling intervals with continuous cloud (visibility less than 1 km). There are differences across sites; the difference between Whiteface summit and all other sites is one of the most striking results, and can be explained on the basis of height above cloud base of the sampling site.

TABLE 4-8

MCCP Hourly Averaged Cloud Liquid Water Content Summary for 1987-88.
All values are in g/m^3.

	Whiteface		Mitchell		Whitetop		Moosilauke		Shenandoah	
	1987	1988	1987	1988	1987	1988	1987	1988	1987	1988
Mean	0.39	0.44	0.28	0.30	0.20	0.21	0.18	0.22	0.16	0.22
Std. Deviation	0.22	0.44	0.21	0.21	0.12	0.10	0.13	0.12	0.10	0.16
Median	0.36	0.36	0.22	0.27	0.19	0.22	0.16	0.19	0.13	0.17
# Observations	96	103	77	292	372	245	250	124	17	57

METEOROLOGY

Field Year Summaries

Meteorological data are collected to aid in the interpretation of the cloud water chemical data, as inputs for cloud water and dry deposition models, and to document the climatic conditions at the MCCP sites. The following parameters were recorded on an hourly basis during the 1986-88 field seasons: temperature, relative humidity, precipitation, wind speed and direction, total solar radiation (horizontal surface) and atmospheric station pressure. Summaries of these parameters for each subsite are provided in Appendix C.

Climate Perspective

This section discusses the temperature and precipitation climate of the Appalachian region during the course of the MCCP field program (1986-88) by examining weather records for several low and high elevation weather stations in the proximity of the MCCP sites. Most of the MCCP sites were established in 1986 and therefore cannot be used to address long-term trends. One assumption made is that if a number of regional stations surrounding an MCCP site show similar climatic trends, then that MCCP site likely experienced the same trend but not necessarily of the same magnitude. High correlation coefficients (0.80 - 0.98) derived from daily temperature comparisons between MCCP and regional weather stations for the 1986-88 period support this assumption.

Three separate studies were used to evaluate the representativeness of the 1986-8 field seasons. In one study 18 existing first-order and cooperative observer weather stations (Table 4-9) were used in the analysis. They were divided into nine northern and nine southern stations, together with station elevation above sea level and period of record. Six of these stations were at elevations comparable to the MCCP sites. Approximately 40 years of records are available for most sites.

TABLE 4-9
Selected Long-Term Climate Stations

SITE	ELEVATION(M)	LENGTH OF RECORD
Northern Appalachians		
Bangor,ME	58	1953-88
Jackman,ME	360	1948-88
Orono,ME	18	1948-88
Concord,NH	104	1948-88
Mt. Washington,NH	1908	1948-88
Pinkham Notch,NH	613	1948-88
Burlington,VT	101	1948-88
Mt. Mansfield,VT	1207	1954-88
Lake Placid,NY	573	1948-88
Southern Appalachians		
Big Meadows,VA	1079	1948-88
Burkes Garden,VA	1006	1948-88
Luray,VA	366	1948-88
Elkins,WV	594	1948-88
Bristol,TN	459	1948-88
Asheville,NC	652	1948-88
Banner Elk,NC	1143	1948-88
Celo,NC	817	1948-88
Grandfather Mt.,NC	1615	1955-88

The climate of the 1986-88 period was compared to the long-term climatologies by deriving monthly deviations from the long-term normals for both temperature and precipitation. Tables 4-10 through 4-15 present results in terms of above- and below-normal deviations, defined as follows. A plus(minus)

indicates the month was above(below) the long term mean (LTM). A +A(-B) designates that the month deviated by at least one standard deviation above(below) the LTM and can be described as an above(below) normal month. A +MA(-MB) indicates the month registered at least two standard deviations above(below) the LTM and can be classified as much above(below) normal. An N indicates a normal month (deviation = 0.0). A 1, 2, or 3 following the +/- designation is used to show that the month ranked first, second, or third, respectively, over the length of record (usually the past 40 years). For example, a -MB3 would be used to represent a month whose mean temperature or precipitation was at least two standard deviations below the LTM and ranked as the third most below normal month for that site. The discussion below of each field season's climate incorporates the results from these tables.

In a second study, the climatology of the upper atmosphere as measured by daily National Weather Service rawinsondes (00Z) was determined for six eastern U.S. cities for the 1986-88 field seasons and compared to an available 29 year (1957 - 1985) climatology. Figure 4-24 depicts field season departures from normal for average temperature and pressure height for the 850 mb level which is, at an average height of approximately 1500 m above sea level, comparable to the height of several MCCP summit sites. Temperatures in the northern Appalachians were near or below normal in 1986, near normal in 1987 and above normal in 1988. In the south, seasonal temperatures were above normal for all three years. The 850 mb pressure heights were near or below normal at the northern-most sites throughout the three seasons. In the south, the predominance of a high pressure ridge is reflected by near and above normal height.

A third study, illustrated by Figures 4-25 through 4-27 (Appendix E) shows the mid-summer drought severity throughout the eastern U.S. for the three field seasons according to the Palmer Drought Severity Index (Palmer, 1965). This analysis, generated bimonthly by the NOAA/USDA Joint Agricultural Weather Facility, depicts areas experiencing prolonged and abnormal dryness or wetness. This slowly changing index reflects long-term moisture runoff, recharge, deep percolation and evapotranspiration conditions for relatively low elevation land areas. It is not necessarily representative of high elevations which can experience added moisture inputs from direct cloudwater deposition. Nonetheless, this index is indicative of large-scale weather conditions that influence temperature and moisture conditions at all elevations. The figures are generally reflective of the three field seasons as a whole and are discussed in greater detail below.

Average 850 mb Temperature (°C) and Pressure Height (m) Departures from Normal (1957-1985 period) for the 1986-88 MCCP Field Seasons (June-September) for Six Eastern U.S. Cities.

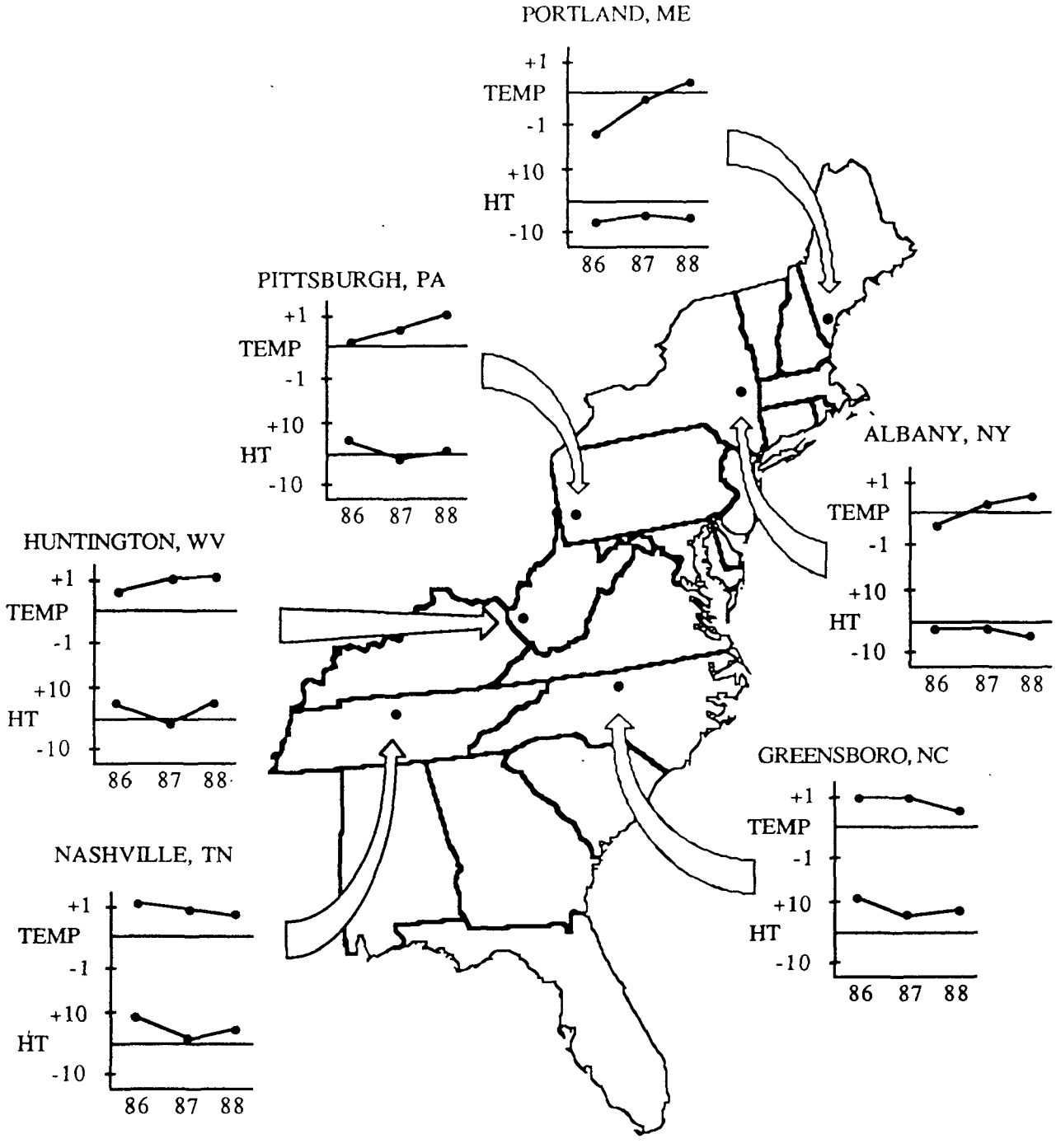


Figure 4-24

TABLE 4-10
1986 Monthly Temperature Deviations from Normal

NORTHERN SITE	APR	MAY	JUN	JUL	AUG	SEP	OCT
Bangor,ME	+MA1	+	+	-	+	-	-
Jackman, ME	+A3	+	-MB2	-B4	-B	-B2	-
Orono,ME	+	-	-B3	-	-	-	-
Concord,NH	+A1	+	-B3	-B	-	-	-
Mt. Washington,NH	+MA1	+	-B	-	-	-	-
Pinkham Notch,NH	+MA1	+	-B	-	-	-B3	-
Burlington,VT	+A2	+	-B	-	-	-	-
Mt. Mansfield,VT	+MA1	+A2	-	-	-	-	-
Lake Placid,NY	+A3	+	-B	-	-B	-	-
SOUTHERN SITE							
Big Meadows,VA	+	-	+	+	-B3	-	+
Burkes Garden,VA	+	+	+A	+A1	-	+A	+
Luray,VA	+A	+	+	+	-B	-	+
Elkins,WV	+	+	-	+A3	-	+	+
Bristol,TN	+	+	+A	+A	-	+	+
Asheville,NC	+	+	+	+A2	-B	+	+
Banner Elk,NC	+A	+	+A3	+A2	-	+A	+A
Celo,NC	+	-	+A	+A2	-	+A	+A
Grandfather Mt.,NC	+	-	+A2	+A1	-B	+	+

TABLE 4-11
1986 Monthly Precipitation Deviations from Normal

NORTHERN SITE	APR	MAY	JUN	JUL	AUG	SEP	OCT
Bangor,ME	+	+	-	+A2	+A	-	-MB1
Jackman,ME	-B2	+A	+	-	+A	+	-
Orono,ME	-	-	+	+A	+A3	-	-B
Concord,NH	-B	-	+A	+A	+A	-	-
Mt. Washington,NH	_	+	+	+A	+A	+	-
Pinkham Notch,NH	_	+	-	+	+A	-	-B2
Burlington,VT	-B2	+	+	+	+A	+A	-
Mt. Mansfield,VT	-	+A	+	+	+A3	+	+
Lake Placid,NY	-B2	+	+MA2	+A3	+	+	-
SOUTHERN SITE							
Big Meadows,VA	-	-B	+	-	+	-	-
Burkes Garden,VA	-	+	-	+	+MA1	+	-
Luray,VA	-B2	-B3	-	-	+	-	-
Elkins,WV	-	-	+A2	+A	+	+	+
Bristol,TN	-MB2	+	-B1	+	N	+	-
Asheville, NC	-B2	-	-B2	-B1	+	-	+
Banner Elk, NC	-B2	+	-B3	-	+A	+	+
Celo, NC	-B2	-	-	-	-	-	+
Grandfather Mt.,NC	-B2	-	-B1	+	+A	-	+

TABLE 4-12
1987 Monthly Temperature Deviations from Normal

NORTHERN SITE	APR	MAY	JUN	JUL	AUG	SEP	OCT
Bangor,ME	+A2	+	+	-	-	-	-
Jackman,ME	+A	-	-	-	-B	-	-
Orono,ME	+A	+	+	-	-	+	-
Concord,NH	+A	+	-	+	-B	-	-
Mt. Washington,NH	+A2	+	+	+	-	-	-
Pinkham Notch,NH	+A3	+	+	-	-B	N	-
Burlington,VT	+A1	+	+	+	-	+	-
Mt. Mansfield,VT	+MA2	+	+	+A	-	-	-
Lake Placid,NY	+A2	+	+	+	-	+	-
SOUTHERN SITE							
Big Meadows,VA	-B3	+	+	+A	-	-	-MB2
Burkes Garden,VA	-B3	+A	+	+	+A	+	-MB2
Luray,VA	-	+	+A	+	+	-	-B3
Elkins,WV	-	+A	+	+A	+A	+	-B2
Bristol,TN	-B	+A2	+	+A	+A2	+	-B2
Asheville,NC	-B	+A3	+	+	+A3	+	-B1
Banner Elk,NC	-B	+A2	+A	+A	+MA2	+	-B
Celo,NC	-B	+A	+	+A	+A1	+	-B1
Grandfather Mt.,NC	-	+A	+	+A	+A	-	-B

TABLE 4-13
1987 Monthly Precipitation Deviations from Normal

NORTHERN SITE	APR	MAY	JUN	JUL	AUG	SEP	OCT
Bangor,ME	-	-	-	-	-	+	-
Jackman,ME	+A	-	+	-	-B1	+	+
Orono,ME	+	-	+	-	-	+A	-
Concord,NH	+A3	-B	+A3	+	-B	+	+
Mt. Washington,NH	+	+	+	-	-	+	+
Pinkham Notch,NH	+MA1	-	+	+	-	+	+
Burlington,VT	-B	-	+	-	-B	+	+
Mt. Mansfield,VT	-	+	+	+	-B	+	+A
Lake Placid,NY	N	-	+	+	-B	-	+
SOUTHERN SITE							
Big Meadows,VA	+MA1	+	+	-	-	+MA2	-
Burkes Garden,VA	+MA1	-	-	-B2	-B	-	-
Luray,VA	+MA1	+	-	-	-	+A	-
Elkins,WV	+	-	-	-B1	+	+A	-
Bristol,TN	+A	-B3	-	-B	-B1	+A	-B
Asheville,NC	+	-B	+MA1	-	-B	+A	-B2
Banner Elk,NC	+MA1	-B	+	-B	-B1	+A	-
Grandfather Mt.,NC	+MA1	-	+	N	-B3	+A	-B3

TABLE 4-14
1988 Monthly Temperature Deviations from Normal

NORTHERN SITE	APR	MAY	JUN	JUL	AUG	SEP	OCT
Bangor,ME	+	+A	-	+A3	+	+	-
Jackman,ME	+	+	-	+	+	-	-B2
Orono,ME	+	+A	+	+A	+A3	+	-
Concord,NH	-	+	-	+A	+A3	-	-B3
Mt. Washington,NH	+	+	-B3	+A	+	-B	-MB1
Pinkham Notch,NH	+	+	-	+A	+A	-	-B3
Burlington,VT	+	+	-	+A	+A	-	-B
Mt. Mansfield,VT	+	+	-	+A2	+A	-	-MB2
Lake Placid,NY	+	+	-B	+A	+A3	-	-B3
SOUTHERN SITE							
Big Meadows,VA	-B	-	-B	-	+	-MB1	-MB1
Burkes Garden,VA	-	-B3	-	+	+A	-	-MB1
Luray,VA	-	-	-	+MA2	+MA2	-	-B
Elkins,WV	-	-	-B	+A	+MA1	+	-B1
Bristol,TN	-	-	-	+	+A	-	-MB1
Asheville,NC	-	-	-	+	+A	+	-B2
Banner Elk,NC	+	-	-	+	+MA1	+	-B2
Celo,NC	N	-B	-	-	+A3	N	-B2
Grandfather Mt.,NC	-	-	+	+	+A3	-	-B1

TABLE 4-15
1988 Monthly Precipitation Deviations from Normal

NORTHERN SITE	APR	MAY	JUN	JUL	AUG	SEP	OCT
Bangor,ME	-	-	+	-	+	-MB1	+
Jackman,ME	-	-B3	-	+	+MA1	-B	-
Orono,ME	-	-	-	+	+	-B1	+
Concord,NH	-	+	-B2	+MA1	+A3	-	-B3
Mt. Washington,NH	+MA1	-	-	-	+A3	+	+
Pinkham Notch,NH	+A	-	-	+	+MA2	-	-
Burlington,VT	-	-	-	-	+	-B	-
Mt. Mansfield,VT	+A2	-	-	-	+A	-	-
Lake Placid,NY	-B	-B3	-B	+	+	-B	+
SOUTHERN SITE							
Big Meadows,VA	-	+MA2	-	-	-	-	-B
Burkes Garden,VA	-	-	-B	-	+	+	+
Luray,VA	-	+A2	-	-	-	-	-
Elkins,WV	-	+	-B1	-	N	+MA1	-
Bristol,TN	-	-	-B2	-	-	+	-
Asheville,NC	+	-B2	-B1	-	-B	-	-
Banner Elk,NC	+	-B	-B2	+	-	-	-
Celo,NC	-	-B	-	-	-	-	-
Grandfather Mt.,NC	+	-	-	+	-	-	-

The following material discusses the regional climate of each year.

1986: The April through October period of 1986 produced contrasting weather conditions between the northern and southern portions of the Appalachians. In the north, it was cool and wet after a warm spring. April ranked as the warmest on record at most stations and had relatively light precipitation. This led to mild to moderate Palmer drought conditions in this region by mid-June. In June and July a series of low pressure systems moved across the Great Lakes and New England, reversing this trend while convective type precipitation dominated the region in August. By late August, and for the rest of the fall, this region was characterized by cool and moist or very moist conditions.

In contrast, the southern zone was warm and dry overall. A warm spring was followed by a hot summer. This was due to a strong upper level ridge which controlled the weather until August. Some synoptic scale systems broke the pattern in August resulting in a cool and wet month. Despite this reprieve, drought conditions in this zone were not alleviated. In April, southern Virginia and western North Carolina were classified to be enduring a severe to extreme drought with northern Virginia in a moderate drought. Some stations in June and July experienced near-record warm temperatures and low precipitation amounts. By late August and onward, the drought was extreme to the south and upgraded to severe in the northern sections of the southern zone.

1987: In the north, the warm spring was followed by a warmer than normal June and July and a cooler than normal August through October. April was one of the warmest on record. Monthly precipitation varied due to its convective nature. Frontal waves began to infiltrate the region beginning in September. Mild to moderate Palmer drought conditions were prevalent in this zone by mid-June due to a dry May, but by early October the region had reversed that trend to moist conditions.

In the south, the drought conditions seen in 1986 continued and in some cases unprecedented weather conditions occurred. The upper level ridge held control over the region until September in the northern part of the zone and until October to the south. This led to warmer and drier than normal conditions for most of the season. The wettest April on record led to moist conditions in Virginia and lessened the drought in North Carolina to moderate levels. By early August, the drought had increased to severe levels in the entire zone. The remnants of a tropical storm in September brought plentiful rains to some portions of the zone. By the end of October, the southern part of the zone was still in a severe drought while the northern portion had a temporary respite. October was also one of the coldest on record.

1988: There was much less of a difference in the climate between the north and south. Both regions experienced a hot July and August and cold October, and generally dry conditions prevailed. An upper level ridge dominated the eastern U.S. until mid-August in the north and late August in the south. July and August were among the hottest on record. After that, cool conditions generally occurred as outbreaks of cool air were able to sweep down from the north. Record and near record cold again occurred in October. This year's weather regime brought drought to the northern zone and prolonged the drought in the southern zone. June was one of the driest on record in the south. By early July, moderate to severe drought levels were attained in the north and extreme levels in the south. With the change in the pattern by September, the northern zone had transformed to a moist region and the southern zone had eased from an extreme drought area to a region of moderate to severe drought.

1986-88 Summary: Two outstanding climate features mark the 1986-88 period: 1) the large contrast in weather conditions between the northern and southern portions of the Appalachians, and 2) the predominance of drought and above normal temperatures in the southern Appalachians throughout the entire period. The southern drought has been cited as an unprecedented event for the last 100-300 years (e.g. Bergman et al., 1986; Cook et al., 1988). Figure 4-28 illustrates a virtually continuous deficit in precipitation over this three year period at Celo, NC. By comparison, the northern Appalachians had

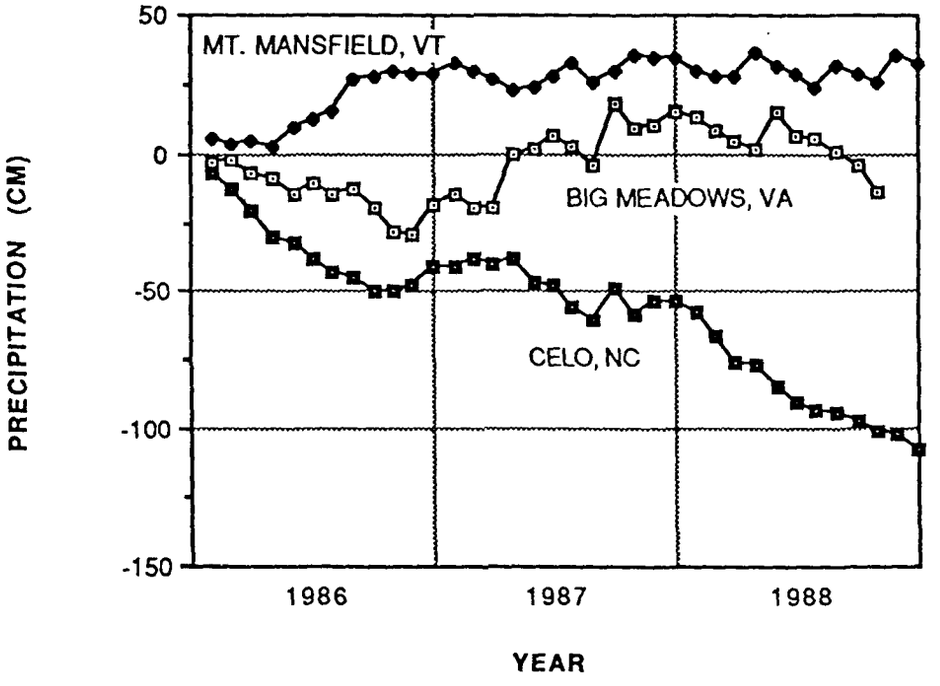


Figure 4-28: Cumulative departure from normal precipitation for three high elevation, long term stations for 1986 through 1988

relatively unexceptional weather overall in terms of persistent trends. Cool and wet weather dominated in 1986 and mixed conditions followed in 1987. It was not until the 1988 field season that, according to the Palmer drought index, moderate to severe drought levels infiltrated the northern Appalachians.

GASES

Most of the nation's long-term ozone, sulfur dioxide and oxides of nitrogen (NO_x) monitoring stations are located in or near population centers. For the most part, the main focus for air quality monitoring has been the determination of concentrations to which large numbers of people are exposed. Only in recent years has there been an interest in documenting ozone levels in rural areas where agriculture and forestry concerns dominate. Of the approximately 1400 ozone sites in the EPA's Aerometric Information and Retrieval System (AIRS), only 33% have been designated as rural or remote. Of the 1831 sulfur dioxide sites in AIRS, approximately 33% are designated as rural or remote. Of the 674 nitrogen dioxide sites in AIRS, only 24% are designated as rural or remote. In the United States, ozone, sulfur dioxide, and oxide of nitrogen concentrations are monitored by local and state governments. In addition to AIRS, other databases containing hourly sulfur dioxide, nitrogen dioxide, and ozone monitoring information exist. In early 1983, the National Park Service established a nation-wide network to collect hourly mean ozone, sulfur dioxide, and NO_x data. Since 1985, the MCCP has monitored ozone, sulfur dioxide, oxides of nitrogen, hydrogen peroxide, and other air quality and cloud chemistry parameters in forested, montane ecosystems across the Appalachian Mountains.

The challenge to identify monitoring data that are characteristic of exposures experienced in forested areas is great. Data derived from the MCCP, as well as EPA's national forest research monitoring program in the early 1980s, provide some assistance in describing the air quality exposures which United States forests receive. However, in truth, there are a small number of air quality monitoring sites located in forests. This is because the majority of commercial and public forest lands appear to be located in areas that are more than 50 kilometers from metropolitan centers and the monitors are mostly located in population centers. Figures 4-29 and 4-30 (Appendix E) represent some forest resources (public, private, and state owned) across the United States, identifying those counties in the U.S. where forest resources dominate.

For perspective, a box plot of trends in annual concentrations is presented in Figures 4-31 through 4-33 (Appendix E). While the absolute numbers may not necessarily reflect the level of forest exposure to these air pollutants, they nevertheless show that no upward trend has occurred over the past decade. The ambient levels of sulfur dioxide even show some statistically significant decline. The temporal trend in O_3, SO_2 and NO_x are consistent with the emission trends for oxides of nitrogen, volatile organic compounds and sulfur dioxide shown in Figures 4-34 and 4-35 (Appendix E).

Ozone

Lefohn and Pinkerton (1988) characterized ozone levels across the United States using hourly-averaged ambient ozone monitoring data for an eight-year period, 1978-1985, for eight forested areas of the United States. The analysis focused on the annual number of occurrences of hourly averaged ozone concentrations greater than or equal to 0.07, 0.08, and 0.10 ppm during the growing season (April-October) as well as during the early (April-June) and late (July-October) portions of the growing season. On the average, the authors reported that elevated ozone concentrations occurred more often in the Piedmont/Mountain/Ridge-Valley and Ohio River Valley areas than in the Pacific Northwest, Upper Great Lakes, and Northern New England/New York areas. In the eastern United States, 1978, 1980, and 1983 were generally the years with the most occurrences of elevated ozone concentrations. In these years, the later part (July-October) of the growing season experienced more elevated concentrations than the earlier part. The area-wide ozone statistics were derived mainly from

urban-oriented ozone monitoring stations, which may overestimate the number of elevated ozone periods which would be observed in the majority of commercial forests in an area.

The selection of rural/remote monitoring sites for this assessment was based in part on the distribution patterns of the hourly ozone concentrations ("ozone diurnal") and also on the location of monitoring sites relative to forest regions. Attempts to gain a coherent picture of ozone exposures across the United States is difficult. Ozone exposures experienced at a specific site in one year may not resemble those that are experienced at the same site the next year. Large year-to-year variation of ozone concentrations measured at sites influenced by urban sources occur (EPA, 1989; Lefohn et al., 1989a). For example, ozone exposures at some urban-influenced sites in the United States were higher in 1983 than in 1984 and 1985 (Lefohn and Pinkerton, 1988; EPA, 1989). In addition, in 1988, ozone exposures experienced at many locations across the United States were higher than in previous years (EPA, 1989). In addition to year-to-year variability, ozone exposures vary from site to site; local meteorological factors influence ozone exposures. In addition, sites that are located at higher elevation experience different exposure profiles than those located near ground level (Lefohn et al., 1989c). Furthermore, because of the importance of the higher concentrations, attempts to average ozone values across a geographic region may lose the identity within a region of those sites that experience the highest exposures. Thus, careful attention is required to adequately describe ozone exposures that occur within geographic regions, as well as across years.

The ozone monitoring sites selected for characterizing forest exposure in the geographic area where forest damage has been observed are listed in Table 4-16. The EPA Mountain Cloud Chemistry Program (MCCP) is a major source of atmospheric chemical and physical information in the northeastern United States for addressing the forest decline hypotheses.

TABLE 4-16
MCCP and AIR Sites

Site Name	AIRS ID #	Site Code	Years	Elevation (m)
Howland Forest		HF	87-88	65
Moosilauke		MS	87-88	1000
Whiteface 1		WF	86-88	1483
Whiteface 3		WF	87	1026
Huntington Co., NY	36031-0005	HU	86-88	500
Hampshire Co., MA	25015-6001	HA	86-88	312
Beaver Co., PA	42007-0003	BE	87-88	1300
Shenandoah 1		SH	87-88	1015
Shenandoah 2		SH	87-88	716
Shenandoah 3		SH	87-88	524
Big Meadows, VA	51113-0003	BM	86-88	1067
Dickey Ridge	51187-0002	DI	86-88	631
Sawmill Run, VA	51015-0004	SM	86-88	453
Whitetop Mtn, VA		WT	86-88	1689
Marion County, VA	51173-0005	MA	86-88	710
Giles Co., TN	47055-0001	GI	86-88	244
Mt. Mitchell 1		MM	86-88	1950
Mt. Mitchell 2		MM	87-88	1775
Mt. Mitchell 3		MM	88	750

There has been considerable discussion in the literature concerning the selection of optimum exposure indices for vegetation effects purposes (Lefohn and Benedict, 1982; 1985; Tingey, 1984; Lefohn and Jones, 1986; EPA, 1986; 1988; Lefohn and Runeckles, 1987; Krupa and Kickert, 1987; Lefohn et al., 1988; Runeckles, 1988; Ashmore, 1988; Parry and Day, 1988; Lee et al., 1988; Hogsett et al., 1988; Lefohn et al., 1989b). The general focus has been on mathematically deriving a "biologically-meaningful" dose surrogate, such as a cumulative index that describes the highest exposure, while not ignoring the lower, biologically less important exposures. Possible approaches for weighting different air pollutant concentrations have been discussed extensively. For example, Lefohn and Runeckles (1987) proposed an exposure index that used the sigmoidal weighting of individual hourly average concentrations of ozone and summing over time. The sigmoidal weighting function was multiplied by each of the hourly average concentrations. The lower, less biologically effective concentrations were included in the integrated exposure summation. They were not eliminated because no threshold was used. An example of two experimental sigmoidal weighting functions (W95 and W126) used by Lefohn et al. (1988) is shown in Figure 4-36 (Appendix E).

In the United States, some exposure indices of ozone that have been used in the past to characterize hourly mean concentrations over monthly and seasonal periods are:

(1) The sum of all hourly mean ozone concentrations using no threshold concentration. This is commonly referred to as "total dose" (SUM0);

(2) The sum of all hourly mean ozone concentrations (W126) where each hourly concentration is weighted by a sigmoidal weighting function;

(3) The sum of all hourly mean ozone concentrations equal to or greater than 0.070 ppm (SUM07);

(4) The number of hourly mean ozone concentrations equal to or greater than 0.070 ppm;

(5) The seasonal means of the average of the daily 7-hour (9:00-15:59 h) concentrations;

(6) The seasonal means of the average of the daily 12-hour (7:00-18:59 h) concentration.

For characterizing ozone episodes, the consecutive numbers of hours when ozone equalled or exceeded 0.070 ppm during any period of the day was determined. These data are presented in Figures 4-37 through 4-54 (Appendix E). For characterizing seasonal ozone exposure, the sum of all hourly mean ozone concentration equal to or greater than 0.070 ppm between the hours of 07:00 and 18:59 was calculated from April 15 through October 15. This analysis was confined to the 12-hour daytime period. In future analysis, it is advisable to include the full 24-h period. Trees may be sensitive to ozone during the nighttime periods. These totals termed "sum of season dose (ppm-hr)" are also shown in Figures 4-37 through 4-54 (Appendix E) as well as presented separately in Table 4-17 and in Figures 4-55 and 4-56 (Appendix E) for 1986-88.

For studying the elevational gradient phenomenon, hourly mean concentrations of ozone data for the three MCCP Whiteface Mountain, Shenandoah National Park sites, and the two Mt. Mitchell sites were used. To supplement the gradient analysis, data from three additional Shenandoah National Park sites (Big Meadows, 1071 m; Dickey Ridge, 631 m; Sawmill Run, 453 m), from one site located 40 miles from Whiteface Mountain (Huntington Forest, 500 m) and from two sites near Whitetop Mountain (Marion, 710 m; Giles, 244 m) were also used. In addition, one site in Massachusetts (Hampshire County, Quabbin Reservoir, 312 m) and one in Pennsylvania (Beaver County, 1300 m) have been selected from the AIRS database in order to bridge the geographic gap between North and South. The results are presented as diurnal ozone concentrations in Figures 4-57 through 4-60. The diurnal pattern is useful for characterizing scavenging efficiency (Lefohn et al., 1989c).

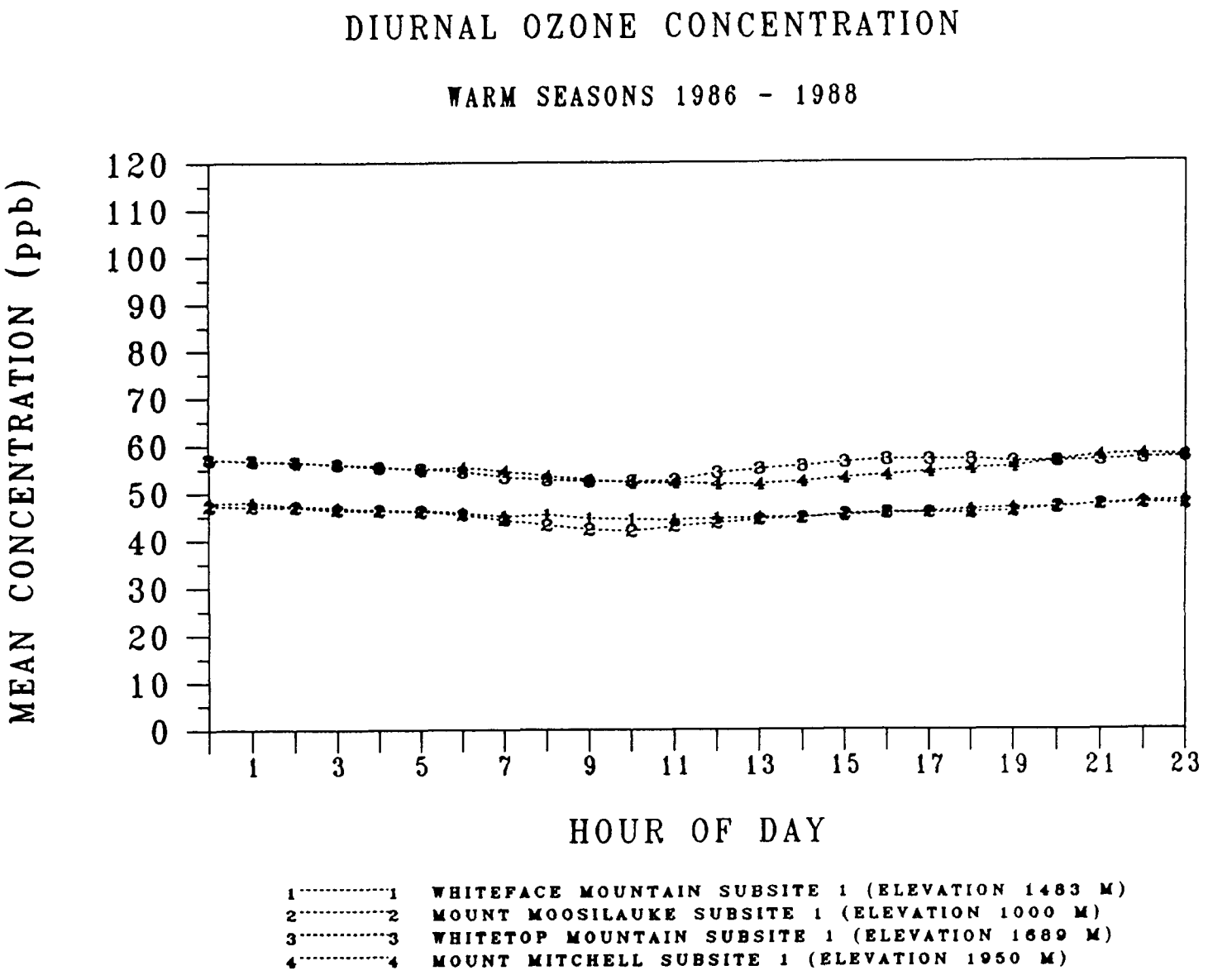

DIURNAL OZONE CONCENTRATION
WARM SEASONS 1986 - 1988
MEAN CONCENTRATION (ppb)
120
110
100
90
80
70
60
50
40
30
20
10
0
1
3
5
7
9
11
13
15
17
19
21
23
HOUR OF DAY
1 WHITEFACE MOUNTAIN SUBSITE 1 (ELEVATION 1483 M)
2 MOUNT MOOSILAUKE SUBSITE 1 (ELEVATION 1000 M)
3 WHITETOP MOUNTAIN SUBSITE 1 (ELEVATION 1689 M)
4 MOUNT MITCHELL SUBSITE 1 (ELEVATION 1950 M)

Figure 4-57

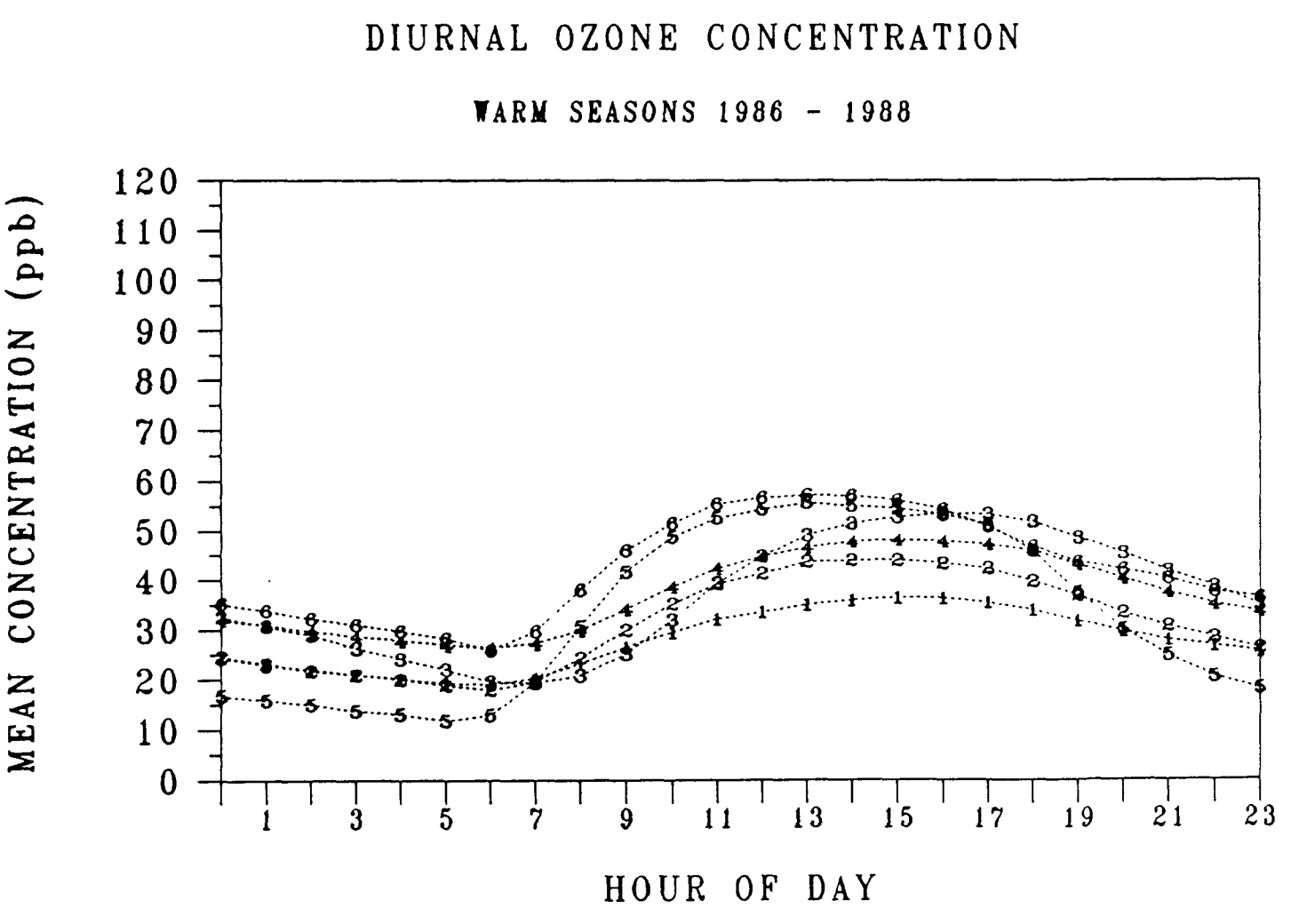

DIURNAL OZONE CONCENTRATION
WARM SEASONS 1986 - 1988
MEAN CONCENTRATION (ppb)
120
110
100
90
80
70
60
50
40
30
20
10
0
1
3
5
7
9
11
13
15
17
19
21
23
HOUR OF DAY
1 HOWLAND FOREST SUBSITE 1 (ELEVATION 65 M)
2 HUNTINGTON, NY (ELEVATION 500 M)
3 BEAVER COUNTY, PA (ELEVATION 1300 M)
4 HAMPSHIRE COUNTY, MA (ELEVATION 312 M)
5 MARION SMYTH COUNTY, VA (ELEVATION 710 M)
6 SAWMILL RUN, VA (ELEVATION 453 M)

Figure 4-58

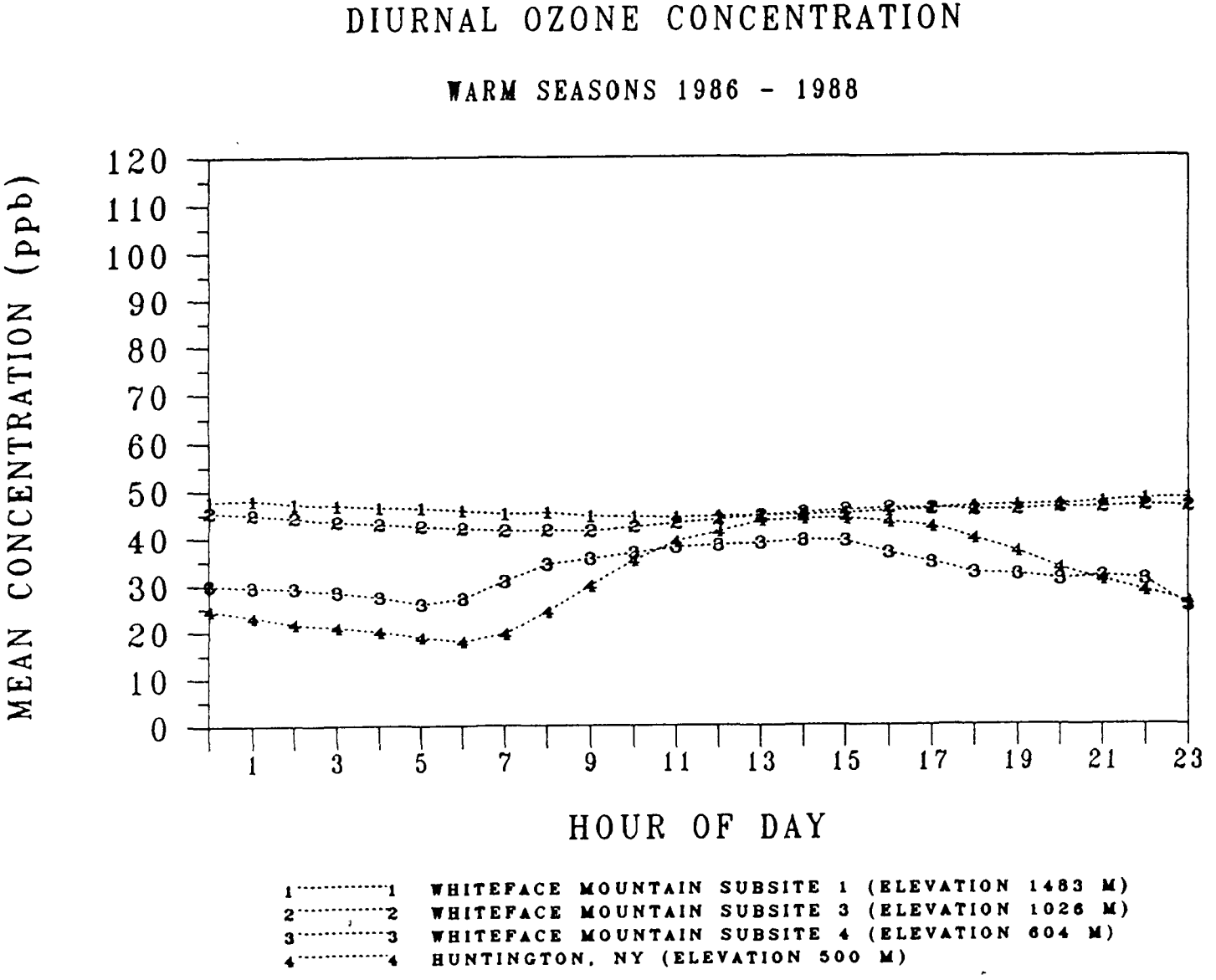

DIURNAL OZONE CONCENTRATION
WARM SEASONS 1986 - 1988
MEAN CONCENTRATION (ppb)
120
110
100
90
80
70
60
50
40
30
20
10
0
1
3
5
7
9
11
13
15
17
19
21
23
HOUR OF DAY
1 WHITEFACE MOUNTAIN SUBSITE 1 (ELEVATION 1483 M)
2 WHITEFACE MOUNTAIN SUBSITE 3 (ELEVATION 1026 M)
3 WHITEFACE MOUNTAIN SUBSITE 4 (ELEVATION 604 M)
4 HUNTINGTON, NY (ELEVATION 500 M)

Figure 4-59

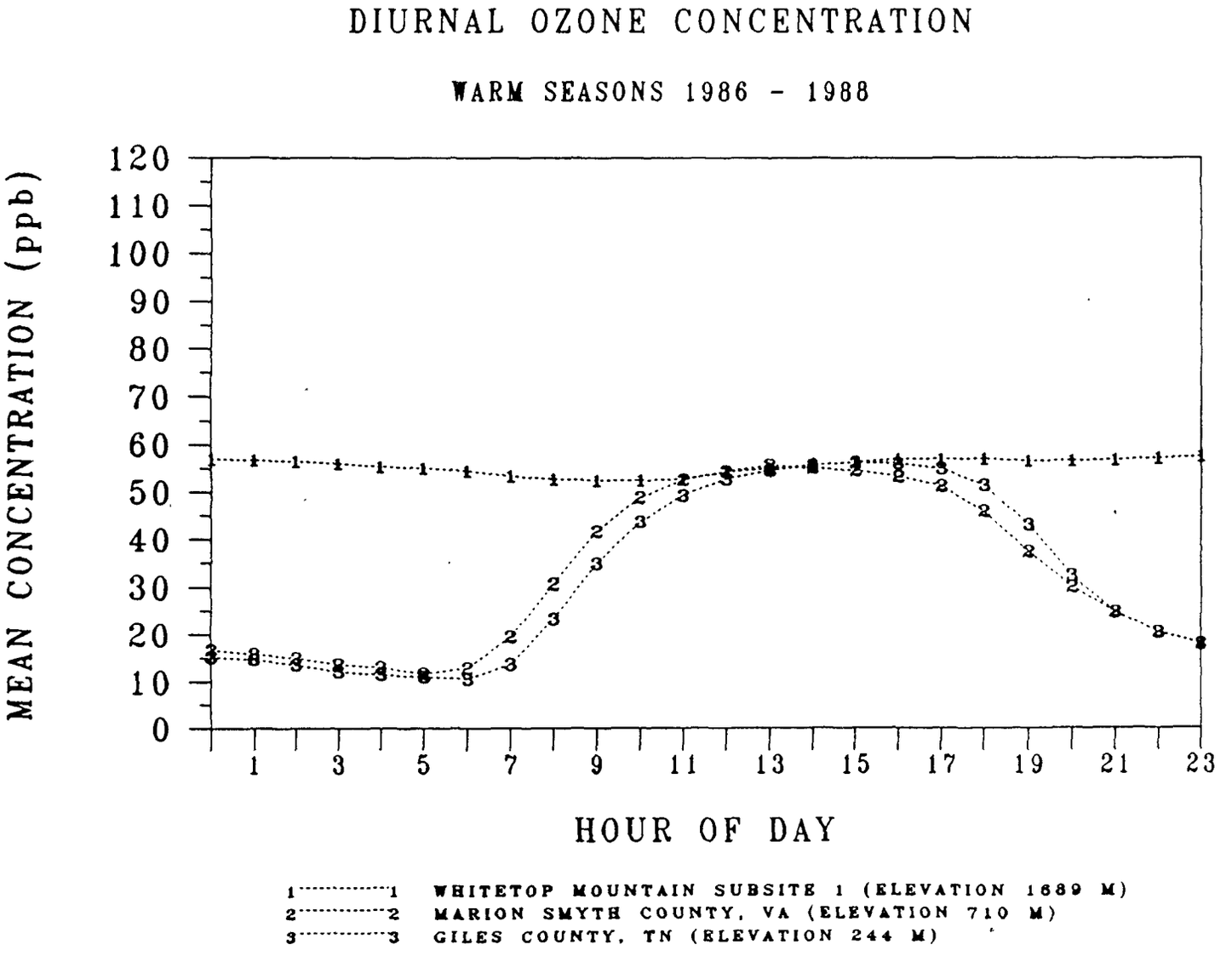

DIURNAL OZONE CONCENTRATION
WARM SEASONS 1986 - 1988
MEAN CONCENTRATION (ppb)
120
110
100
90
80
70
60
50
40
30
20
10
0
1
3
5
7
9
11
13
15
17
19
21
23
HOUR OF DAY
1 1 WHITETOP MOUNTAIN SUBSITE 1 (ELEVATION 1689 M)
2 2 MARION SMYTH COUNTY, VA (ELEVATION 710 M)
3 3 GILES COUNTY, TN (ELEVATION 244 M)

Figure 4-60

The analysis of the data indicates that there is a distinct ozone elevational gradient at Whiteface Mt., and that the MCCP sites in the South experience higher cumulative ozone exposures than sites in the North. Figure 4-61 (Appendix E) also indicates increased mean ozone concentration with elevation for 19 ozone monitoring stations in the eastern U.S.(see Table 4-17 for station codes). The length of time that the MCCP has monitored ozone is too short for investigating the existence of any temporal trend. However, the Whiteface Mountain summit has an uninterrupted monitoring record since 1973. As shown in Figure 4-62 (Appendix E), no trend in annual mean ozone concentration is discernable for this 15 year time period.

TABLE 4-17

Ozone Exposure
Sum of Season Dose
(for daylight hours 7AM - 6PM)

Site Name	Sum of Season Dose (ppm * hr) ≥ 0.07 ppm 1986	1987	1988
Howland Forest (HF)	N.D.	0.82	4.16
Moosilauke (MS)	N.D.	7.81	12.51
Whiteface 1 (WF1)	2.29	9.68	20.51
Whiteface 3 (WF2)	N.D.	9.41	16.50
Whiteface 4 (WF3)	N.D.	3.47	N.D.
Huntington Co., NY (HU)	5.09	5.74	11.36
Hampshire Co., MA (HA)	9.14	9.26	34.93
Beaver Co., PA (BE)	N.D.	13.06	31.70
Shenandoah 1 (SH1)	N.D.	9.49	23.27
Shenandoah 2 (SH2)	N.D.	9.01	39.44
Shenandoah 3 (SH3)	N.D.	6.07	20.88
Big Meadows, VA (BM)	5.56	28.50	31.89
Dickey Ridge, VA (DI)	3.21	31.07	40.25
Sawmill Run, VA (SM)	11.39	26.80	30.16
Whitetop Mtn, VA (WT)	N.D.	38.54	37.68
Marion County, VA (MA)	4.11	9.27	26.92
Giles Co., TN (GI)	16.38	16.73	28.91
Mt. Mitchell 1 (MM1)	8.34	5.14	45.17
Mt. Mitchell 2 (MM2)	N.D.	6.68	19.49

Sulfur Dioxide

Sulfur dioxide concentrations are very low throughout the network as indicated in Table 4-18.

Table 4-18

Sulfur Dioxide Measurements Summary for Selected MCCP Sites

	Max	Mean	# hourly averages
Howland HF	12 ppb	1.07 ppb	1371
Whiteface WF1	20 ppb	1.42 ppb	1990
Whiteface WF3	16 ppb	0.86 ppb	2962
Whitetop WT	47 ppb	2.08 ppb	4310
Mt. Mitchell MM1	12 ppb	1.41 ppb	996

Figures 4-63 through 4-66 (Appendix E) show the frequency distributions derived from continuous SO_2 measurements. Stations operating on a weekly schedule (filterpack) reported similarly low values for the warm season May 1 through October 31, 1987. The northern sites exhibited a strong directional variability on the basis of 36-hour back trajectories. Air masses from the south and west had significantly higher SO_2 concentrations than air masses from any other sector. The southern stations exhibited a more even pattern of SO_2 concentration as function of air mass trajectories. Because SO_2 is the precursor gas to cloud water sulfate, it is not surprising to find similar directional variabilities for both sulfur compounds.

Because of the very low concentration levels of sulfur dioxide during the warm season, it is very difficult to reliably measure an altitudinal gradient. It is equally difficult to discern a geographic gradient from the data set.

Hydrogen Peroxide

Hydrogen peroxide (H_2O_2) plays a key role in the formation of sulfuric acid in the atmosphere (Calvert et al., 1985; Seigneur and Saxena, 1988). The aqueous phase oxidation of dissolved SO_2 by H_2O_2 is acid catalyzed. Thus, the reaction rate can proceed rapidly even at low pH (McArdle and Hoffmann, 1983). Mallant et al. (1985) reported morphological damage, similar to that observed in the Black Forest, when simulated fog containing H_2O_2 was applied to Norway spruce. Joslin et al. (1988) showed that H_2O_2 was effectively removed from cloud water upon contact with red spruce foliage. Cloud water H_2O_2 measurements from the MCCP network can help to quantify the role of this compound as an oxidant and as a possible agent of forest damage.

Olszyna et al. (1988) reported cloud water H_2O_2 measurements for the MCCP site at Whitetop Mountain, VA. The highest concentration measured (246.6 μmol/l) was more than twice any previously reported cloud concentration. Daum et al. (1984) used an aircraft to sample stratiform clouds in the eastern USA and reported H_2O_2 concentrations varying from 0 to 75μmol/l. Romer et al. (1985) used an aircraft to collect cloud samples at various European locations and found that H_2O_2 concentrations varied from 0.05 to 88 μmol/l. Lazrus et al. (1985) reported that in 284 cloud samples at Whiteface Mountain, NY, H_2O_2 concentrations varied from 0 to 112μmol/l. Kelly et al. (1985) reported total peroxide (assumed to be H_2O_2) levels for an additional 190 cloud samples collected at Whiteface Mountain and found similar H_2O_2 levels (0 to 100 μmol/l).

Cloud water H_2O_2 levels for four MCCP sites from 1986-1988 are presented in Table 4-19. Not all cloud water samples were analyzed for H_2O_2. Mean concentrations were similar for all sites except Shenandoah, VA. The low mean concentration reported for this site may be an artifact of the small sample size. The highest H_2O_2 levels were recorded at each site during summer. Spring and fall levels were typically less than half of the summer values. Kadlecek et al. (1985) reported H_2O_2 concentrations in winter clouds at Whiteface Mountain to be always much lower than 1 μmol/l. This seasonal trend is no doubt due to the photochemical production rate of H_2O_2 in the gas phase, which would be expected to exhibit a strong annual trend with maximum production in summer and minimum during winter.

The distribution of H_2O_2 concentrations in cloud water collected at the Whiteface and Whitetop sites is shown in Figure 4-67 (Appendix E). A higher frequency of very high H_2O_2 concentrations was observed at Whitetop Mountain, VA.

TABLE 4-19

Cloud Water H_2O_2 Measurements at MCCP Summit Sites for 1986-88
All concentrations are in μmol/l.

Site	Minimum	Maximum	Mean	# Samples
Whiteface,	1.6	136.4	37.7	54
Shenandoah, VA	0.3	49.0	12.3	22
Whitetop, VA	0.8	246.6	44.1	141
Mitchell, NC	0.3	196.0	41.9	236

THROUGHFALL

The chemistry of throughfall water is a composite of rain, cloud, and dry deposition, tree emissions and absorptions, evaporation from the canopy, and biological activity on the canopy and in the collected sample. Also, physical features (terrain slope and aspect, canopy structure and individual canopy element geometry, and wind velocity) affect where canopy collection of rain and cloud occurs and how the directed flow develops within the canopy. It is difficult to properly attribute the sources and sinks to the correct mechanisms since they are difficult to measure separately and have considerable variations across the canopy and within and between events.

For a rain event, there is an initial period during which the canopy is wetted until the storage capacity is satisfied. The excess accumulation over evaporation drips through the canopy as throughfall or flows down the tree trunk as stemflow. The common method for determining canopy storage capacity involves linearly relating the unperturbed rain amounts (which can be subject to collection problems in high wind environments) to the corresponding average throughfall from many events. The offset from the origin provides the storage capacity, assuming that evaporation is much less than the water held by the canopy.

For a cloud event, the storage capacity may be satisfied only after prolonged exposure to clouds. While the cloud water may be relatively inconsequential to forest hydrology in most areas, it has the potential to contribute to foliar/needle damage (because of the higher concentrations of dissolved species in the cloud water as compared with rain, and the extended period of evaporation that further concentrates the less volatile chemicals). However, measurements of evaporation of intercepted water are rare, further complicating efforts to quantify this process. Most events at high elevation are a mixture of

rain and cloud, but the canopy storage cannot be obtained directly because the cloud water enhances throughfall, without being collected as precipitation. In many cloud events, the throughfall amount slightly exceeds what was measured as rain, providing an estimate of cloud water deposition, if estimates of canopy storage for the appropriate canopy composition can be made.

Seasonal data from the summit of Whitetop Mountain (May 1987 - May 1988) have shown, however, that differences between throughfall volumes and precipitation volumes are generally poor indicators of the volume of cloud deposition due to substantial and unknown evaporation losses, especially during the summer. At this site during the warmer months, monthly deposition volumes under canopies are less than or about equal to those in the open. This pattern occurred despite the fact that other information (computed cloud water flux and total sulfate deposition in throughfall) indicated that cloud deposition occurred during the warmer months as well.

One reason for this summer pattern is the higher frequency of orographic clouds with low liquid water contents and high ion concentrations. These events rarely result in much water reaching the forest floor, while still accounting for a considerable deposition of ions to the canopy. In contrast, during periods when evaporation is less important due to lower temperatures and solar radiation levels, the accumulation of additional water from clouds can be observed. MCCP research is now in progress in which the cloud, precipitation, and throughfall chemistries are monitored. If the dry deposition of SO_4 and Cl are relatively small, their enrichments in the throughfall can be used to further refine the cloud water deposition. The biologically and/or chemically more active species H^+, NO_3^-. and NH_4^+ cannot be used for this purpose since considerable loss of these species is possible.

Within the last two decades, interest in throughfall has broadened from a hydrological focus to include nutrient cycling, particularly fertilization and leaching (Parker, 1983). Data are now being taken with sufficient time resolution to monitor the changing chemical environments on the canopy surfaces. These results will aid simulation studies designed to provide a better understanding of the beneficial and harmful effects from extended periods of canopy wetting.

Initial analysis of Whiteface data indicated that ion depositions have a larger variability than does the deposition of water. The data collected apply only to the site from which they were taken and to sites where the wind regimes and canopy composition and structure are similar. Event-based comparisons of throughfall and rain chemistry were made at the same two sites on Whiteface Mountain, NY. The lower site was affected by rain only, and the upper site had additional cloud water deposition. Throughfall from both sites included whatever dry deposition had occurred since the last wetting, residue after evaporation of the last rain/cloud, and emissions from the tree itself and biology living on the tree surfaces. Table 4-20 summarizes enrichment ratios integrated over the summer season in species deposition, where the uncertainty was obtained using standard error propagation calculations. No estimates of bias were possible.

TABLE 4-20

Deposition Enrichment Ratios (Throughfall/Bulk Precipitation) at Whiteface Mountain, NY

	Under Canopy Upper Site	Open Rain Lower Site
Water (cm)	8.51 ± 0.48	8.31 ± 0.08
H^+	0.95 ± 20%	0.71 ± 13%
NH_4^+	< 0.35	1.11 ± 22%
NO_3^-	0.86 ± 23%	1.65 ± 19%
SO_4^{2-}	1.45 ± 19%	1.16 ± 14%
K^+	9.4 ± 44%	> 13
Cl^-	2.51 ± 27%	> 2.0 ± 25%

Of particular interest is the loss of the first three species at the upper (balsam fir) site, either on the canopy or in the collected sample. Given that this site also had a substantial input of these species from cloud water (current estimates suggest deposition on the same order as from rain), this suggests that about half the H^+ and the NO_3^- and at least 75% of the NH_4^+ that should have been deposited were not present at the time of analysis. Samples were refrigerated immediately after the event and the analysis was completed within a few days. Sulfate and chloride have reasonable enrichments given contributions from dry and from cloud at the upper site. Foliar leaching is responsible for the ten-fold increase in K^+ deposition.

This section does not provide a comprehensive review of throughfall with an investigation of the role of cloud water deposition; rather, it is a characterization of the gross features of the throughfall compared with the precipitation obtained during the same time. For each weekly pair of samples, the ratio of deposition in throughfall to open precipitation was calculated. The median values are presented in Table 4-21.

TABLE 4-21

Median Value of Site Enrichment Ratios (Throughfall/Bulk Precipitation).

Site	SO_4^{2-}	NO_3^-	NH_4^+	H^+
Shenandoah, VA - upper	1.3	1.8	0.9	0.5
Shenandoah, VA - middle	1.3	1.7	1.0	0.2
Shenandoah, VA - lower	1.1	1.2	1.0	0.1
Moosilauke, NH	1.3	1.0	0.8	0.9

Not all sites have the same enrichment behavior because the relative input strengths differ, the fraction of total deposition due to cloud water affects the ratios, and the canopy type helps create its own chemical and biological environment. While dry deposition of various ionic species can be estimated and is in some cases relatively small, foliar exchange can be either a major source or sink. The degree of interaction between the acidic deposition and the canopy is reflected primarily in the loss of H^+ and NH_4^+, and an increase in base cations in the throughfall samples.

Similar weekly integrated throughfall measurements made during 1987 and 1988 at two sites in spruce-fir forests of the Black Mountains showed that while the yearly H^+ average was 24% to 34% of

the total ions in rain, it was only 16% to 23% in throughfall (Robarge, 1989). Similarly, NH_4^+ was 8% to 10% in rain, but only 3% to 5% in throughfall. Similar trends for these ions have been reported in other canopies (Bredemeier, 1988; Joslin et al., 1988; Waldman and Hoffmann, 1988; Olson et al., 1981; Rodenkirchen, 1986) and in controlled experiments (Kaupenjohann et al., 1988; Kelly and Strickland, 1986; Evens 1982; Scherbatskoy and Klein, 1983). Recent data (May 1987 - May 1988) from Whitetop Mountain give further evidence of the importance of the foliar exchange (see Table 4-22).

TABLE 4-22

Actual (keq/ha/yr) and Percent Ion Contribution to Total Deposition at Whitetop, VA
Percentages for cations and anions are treated separately (Joslin et al., 1988).

	H^+	NH_4^+	Ca^{2+}	Mg^{2+}	Na^+	K^+	NO_3^-	SO_4^{2-}	Cl^-
Throughfall & Stemflow	1.27	0.21	1.26	0.27	0.16	0.35	0.72	2.50	0.37
	36%	6%	36%	8%	5%	10%	20%	70%	10%
Precipitation	0.50	0.14	0.07	0.02	0.03	0.01	0.20	0.52	0.04
	66%	18%	9%	3%	4%	1%	26%	69%	5%
Cloud	63%	26%	5%	2%	3%	1%	32%	64%	4%

Precipitation and cloud water had similar proportional contributions to total deposition; throughfall was different from both. Because of the large, unquantifiable amount of foliar exchange, cation deposition in throughfall (with the possible exception of Na^+) was of little use in estimating cloud water or dry deposition. Of the major anions, nitrate appeared slightly depleted (see also Lindberg et al., 1987) and chloride was somewhat enriched (both foliar absorption and leaching of chloride has been observed; Leonardi and Flueckiger, 1987). Sulfate appeared the least affected by foliar exchange. In addition, the concentrations and total quantities of sulfur deposited and passing through these forest canopies was large relative to the size of total foliar pool. The time period for which throughfall sulfate is used to represent total sulfate deposition should be sufficiently long so that differences in canopy residue sulfate are small relative to the amount passing through. The reproducibility and completeness of wash-off, the importance of dry deposition of aerosol sulfate and sulfur dioxide, and the sequence of meteorological systems affecting the site affect the utility of this approach.

DEPOSITION

Cloud Water Droplet Deposition

Estimates of cloud water and chemical deposition to high-elevation forests are now reviewed. Because cloud water is usually collected over hour-to-event time intervals during intensive sampling campaigns, few full measurements exist for seasonal or annual cloud water deposition. Annual cloud deposition estimates that have been published are usually extrapolated from short-duration, growing season (generally April-October) cloud water chemical composition and model or throughfall-based cloud water interception rates. Extrapolation of cloud water or chemical fluxes to annual values is highly uncertain because winter chemical concentrations are generally not known (see section on winter data from Whiteface Mountain, NY) and because cloud water interception rates in winter depend on unknown rime ice collection efficiencies.

Historical Perspective --

Table 4-23 summarizes past estimates of chemical and water deposition due to the interception of wind-driven cloud droplets. The MCCP has been evaluating the cloud deposition model (CDM) and revising the technique used to estimate cloud deposition at its high elevation sites. These estimates are presented later in this section. This historical perspective is presented to give background for the later discussion of deposition at MCCP sites. Cloud water chemical deposition values presented in Table 4-23 have been converted to a consistent set of units (kg/ha/mo) for ease of intercomparison among estimates. Cloud water deposition is reported as an annual estimate (cm/yr, extrapolated from shorter time periods) to facilitate comparison with annual precipitation totals. All deposition estimates presented here are based either on canopy throughfall data or model-derived cloud water fluxes.

Lovett et al. (1982) reported estimates of sulfate and nitrate deposition from clouds on Mt. Moosilauke, NH, using cloud water fluxes computed from a model (Lovett, 1984). He estimated that cloud water deposition contributed about 70% of the total (cloud and precipitation) chemical fluxes for SO_4^{2-} and NO_3^-.

Mueller and Weatherford (1988) computed cloud deposition on Whitetop Mountain, VA, for a 26-day period in the spring of 1986. Using Lovett's model (Lovett, 1984), they computed cloud deposition for every hour during the study period, although some meteorological parameters and ion concentrations had to be estimated to fill in missing data. The cloud water SO_4 flux was between 5.3 and 9.1 kg/ha/mo, while NO_3 flux was between 2.8 and 5.4 kg/ha/mo. The ranges reflect projected variation in unmeasured model input data.

TABLE 4-23

A Review of Chemical Ion and Cloud Water Deposition Via Droplet Interception, Various Locations, Investigators, and Years.

Site[a]	Ref.[b]	Cloud Droplet Deposition H^+	NH_4^+	NO_3^-	SO_4^{2-}	Frequency H_2O	Cloud	H_2O Flux Method[d]	Site Elev	Coll. Per.[e]
			[kg/ha/mo]			[cm/yr]	[%hr]		(m)	
MS,NH	Lovett[1]	.20	1.4	8.5	11.5	68	40	model	1220	80-81
MS,NH	Lovett[1]	-	-	-	-	34-98	40	th.fall	1220	
MS,NH	Lobett[1]	-	-	-	-	153	40	model	1220	
MS,NH	Mohnen[2]	.04	0.24	0.8	1.7	(18)	23	model	1000	87
WF1,NY	Mohnen[2]	.18	2.3	4.8	11.2	(127)	42	model	1483	87
WF,NY	Mohnen[2]	.05	0.8	3.0	7.0	-	27	th.fall	1200	87
WF3,NY	Lindberg[2]	.04	0.5[c]	1[c]	2	13	-	model	1225	86-88
WF3,NY	Lindberg[3]	-	-	1.2	1.3	5-31	5-13	model	1225	86-87
WF,NY	Mohnen[2]	-	-	-	-	112	30	model	1200	86
WF,NY	Mohnen[2]	-	-	-	-	1	10	model	900	86
WT,VA	Mueller[1]	-	-	4.3	8.1	-	-	model	1686	86
WT,VA	Mohnen[2]	.22	2.6	7.7	13.8	-	31	model	1686	87
WT,VA	Mohnen[2]	.20	11.8	10.4	13.1	-	31	th.fall	1686	87
WT,VA	Mohnen[2]	-	-	-	-	25-56	27-41	model	1686	86
MM,NC	Saxena[1]	.17	1.2	3.2	9.8	35-77	25-52	model	2020	86
MM,NC	Stogner[1]	.12	0.8	3.2	7.3	-	-	model	2020	86
CL,NC	Dasch[1]	(.03)	(0.3)	(0.9)	(2.2)	18	62	th.fall	1987	86
MM,NC	Lin[3]	.09	0.6	1.9	5.2	32	28	model	2020	86
MM,NC	Lin[3]	.18	1.3	4.0	8.0	75	41	model	2020	87
MM,NC	Lin[3]	.11	0.7	2.8	5.6	57	30	model	2020	88
MM,NC	Mohnen[2]	.09	0.1	2.6	5.5	-	27	model	2020	87
MM,NC	Mohnen[2]	.13	1.1	2.2	8.9	-	27	th.fall	2000	87
SH,VA	Krovetz[1]	.01	0.13	0.5	0.6	9	7	model	1014	86-87
SH,VA	Sigmon[1]	.02	0.21	0.8	1.2	5	7	th.fall	1014	86-87
SH,VA	Mohnen[2]	.04	0.26	1.3	1.3	-	7	model	1014	87
SM,NC	Lindberg[1]	.14	1.0	2.2	7.2	-	25	model	1740	86
SM,NC	Lindberg[2]	.05	0.5**	1**	4	37	-	model	1740	86-88
SM,NC	Lindberg[3]	-	-	1.1	4	9-70	10-35	model	1740	86-87
RT,QUE	Schemenauer[1]	.11	-	-	-	77	44	assumed	970	85
HN,CA	Waldman[1]	.01	0.03	0.5	0.3	2	-	assumed	780	82-83
GDF,GB	Dollard[1]	.01	-	0.5	0.7	-	7	msmt.	850	82

[a] MS = Moosilauke, NH; WF = Whiteface Mtn, NY; WT = Whitetop, VA; MM = Mt. Mitchell, NC; CL = Clingmans Dome, NC; SH = Shenandoah, VA; SM = Great Smoky Mtns, NC; RT = Roundtop, Quebec

[b] HN = Henninger Flats, CA; GDF = Great Dunn Fell, Great Britain
1 = Peer-reviewed journal article or book chapter; 2 = Technical report subject to EPRI or US EPA review; 3 = Unpublished data summary or submitted manuscript

[c] NO_3^- and NH_4 were estimated from total N cloud deposition and the overall NH_4/NO_3 ratios given by Lovett (in Lindberg & Johnson, 1989)

[d] Annual cloud water flux was determined by: (1) a version of the Lovett model (1984); (2) collecting throughfall under the canopy and correcting for precipitation and evaporation; or (3) direct micrometeorological measurements.

[e] Collection period refers to the years when data were collected. Most data are from short-term, intensive sampling performed during the growing season (April-October). None represent rime ice conditions.

note: Data in parentheses were calculated from information provided in the paper; Mohnen's Whiteface Mt. estimates are based on data from sites at 1483 and 1245m.

Lindberg et al. (1988) used Integrated Forest Study (IFS) data to estimate cloud, precipitation, and dry deposition at a high-elevation site in the Great Smoky Mountains. Cloud water SO_4 and NO_3 deposition were estimated to be 7.2 and 2.2 kg/ha/mo, respectively, both two to five times greater than estimates from a nearby low-elevation site for the period January-April 1986.

More recent deposition estimates from the IFS appear in discussions by Lovett, Knoerr and Conklin, and Ragsdale in a draft summary of the IFS (edited by Lindberg and Johnson 1989). Cloud water H^+ deposition at these two sites was estimated to be 30% to 40% of total H^+ deposition. The IFS scientists concluded that the primary sources of uncertainty in cloud water deposition were immersion time, cloud water amount, and ion concentrations. These IFS results were only presented as a graphical summary with no presentation of methodology, chemical concentrations, or quantitative uncertainties.

Following Lovett's work, Saxena and co-workers estimated cloud water and chemical deposition on Mt. Gibbs (Mt. Mitchell State Park, NC), while assuming canopy structure and collection efficiency for cloud droplets (Saxena et al., 1989a; Lin and Saxena, 1989; Stogner and Saxena, 1988; Saxena et al., 1989b). Table 4-23 illustrates some of the cloud deposition estimates for Mt. Mitchell. There is a wide range of cloud water deposition values from two research institutions (2 to 20 kg of SO_4/ha/mo). Although variation in cloudiness and pollutant concentrations are primary causes of differences between years, the variety of results for one year suggests that the early Mt. Mitchell cloud deposition modeling results are uncertain.

One of the Mt. Mitchell area estimates (Dasch, 1988) from nearby Clingmans Dome has been compared with coincident data collected by Saxena et al. (1989b). The representativeness of the two results remains unknown because of the lack of direct measurements of cloud water deposition. A comparison of cloud frequency versus cloud water flux from all investigators indicates that Dasch's cloud water deposition was anomalously low compared to cloud frequency in other published results (see Table 4-23).

Status of the Cloud Deposition Model --

The cloud deposition model (CDM) used by the MCCP is an improved version of the model originally developed by Lovett (1984). Modifications to Lovett's model are briefly described by Mohnen (1988a). Work related to the CDM has concentrated on examining the following:

* the difference in model performance caused by using a site-specific vertical profile of forest canopy surface area versus the model default profile based on data from Lovett's Mt. Moosilauke forest (Lovett, 1984)

* the difference in model performance caused by using site-specific vertical profiles of wind speed versus the default profile based on Lovett's site

* the difference in model performance caused by changing from the original Lovett droplet collection efficiency scheme, based on individual tree components (twigs and branches), to an experimental scheme designed to consider (crudely) the effect of tree morphology ("bulk collector" versus individual branches and twigs); several experimental schemes were tried, each used a slightly different technique for merging bulk collection efficiency (applicable to the most dense portion of a tree crown) with Lovett's tree component collection efficiencies (Thorne et. al., 1982)

* the importance of using site-specific meteorological data (for model input) versus data from a nearby site

* overall model performance relative to canopy throughfall measurements

Data have been collected from two spruce stands near the summit of Whitetop. These include canopy-top wind speed and related profiles of speed, spatial variation in cloud liquid water, vertical airspeed at canopy top, and canopy throughfall (TF) rates. One TF plot (A) was located about 75 m northeast of the Whitetop summit, while the second (B) was located about 125 m east-northeast of A. A larger TF database exists for plot B because of the construction and operation of an automated TF volume measurement system. Plot A is considered a near-summit site (local slope less than 5%) and its architecture is fairly open (leaf area index=4.8; maximum canopy height=15 m), whereas plot B is a north-facing slope site (local slope almost 20%) that is closed with very little tree mortality (leaf area index=11.0; maximum canopy height=17.5 m). The leaf area index values provided here represent the ratio of the total (full-sided) leaf area to the associated ground area.

Preliminary findings of the CDM evaluation are now summarized:

* model performance was little affected by the surface area (SAI) profile used; this is consistent with expectations from a previous analysis (Mueller 1990) of model sensitivity to this feature (computed cloud water flux is not sensitive to modest uncertainty in the height of the SAI profile maximum)

* model performance was greatly affected by wind speed profile changes--the default profile of Lovett produced lower cloud deposition estimates than actual profiles-- the difference was on the order of a factor of 4 at plot B and a factor of 1.5 at plot A

* several modified (to test different methods for incorporating bulk collection efficiency into the Lovett model framework) droplet collection efficiency schemes were tested--much data are still needed to better understand the relationship between collection efficiency and tree morphology; the scheme used to make subsequent cloud deposition estimates for this report (Mueller, 1990) produced 60-70% lower deposition estimates than Lovett's (1984) original scheme

* site-specific meteorological data (measured canopy-top wind speed and spatially-adjusted liquid water content) were found to result in much better model performance at plot B than data collected at the mountain summit

* overall, the CDM performed better in simulating cloud flux to the closed stand (plot B) as opposed to the open stand (plot A), but the difference was likely due to a greater uncertainty in model input wind data for plot A. Using the most updated version of the model and "best" inputs, flux was overestimated at plot A by 20-50%, depending on which version of the modified collection efficiency scheme was used, with computed flux bias ranging from -20-30% for plot B.

Current Modelling Results --

The version of the CDM used for the spruce-fir sites in this analysis of cloud deposition was configured with the modified version of the droplet collection efficiency scheme that produced the least bias for the Whitetop evaluation. This modified scheme used measured bulk droplet collection efficiency (Joslin et. al., 1990) in place of Lovett's individual tree component efficiencies for those portions of the tree crowns having the highest leaf-to-total surface area ratios (ie. ratios > 0.85). This represents a

major change from the 1988 analysis (Mohnen, 1988a). The uncertainty in deposition calculations caused by using this particular parameterization is unknown. Default SAI profiles (similar to those used in the 1988 analysis) were used for all sites except WT where site-specific profiles were known. Another major change from the 1988 analysis was the use of a = 0.20 in place of a = 0.27 for the wind speed extinction coefficient. The default (Lovett) value of a = 0.27 and the value that appeared to be most representative of conditions at Whitetop a = 0.10 yield considerably different results. The best value for each of the other MCCP sites was not known. A compromise was to use a = 0.20, due to a lack of evidence favoring any specific value over another. Site-specific wind speed profiles were used in all WT CDM runs.

The methodology followed for this analysis was very similar to that used in the 1988 MCCP report (Mohnen, 1988a). Cloud impaction events at each MCCP site were classified according to the synoptic meteorological conditions. A detailed analysis of cloud chemistry and meteorological variables (wind speed and liquid water content) was conducted by Vong et al. (1989). In addition to synoptic typing, Vong et al. found that specific air trajectory directions computed within a given event type can further characterize event conditions. For example, ion concentrations measured during events classified as "warm sector" (with respect to a cyclonic weather system) were sometimes different when segregated according to whether associated backward air trajectories crossed the heavily industrialized Ohio River Valley. This technique was able to explain half of the variance in the chemistry and meteorological variables, and can be used to estimate non-monitored, multiple-hour means for the different synoptic types. Deposition estimates for MCCP sites can now be made for periods when data are incomplete as long as cloud frequency and event type can be determined.

In this analysis, cloud event synoptic type was determined for each hour of each growing season at each site for the 1986-88 period. The results of the synoptic/trajectory (subclass) analysis performed by Vong et al. (1989) were used to develop subclass means of aqueous ion concentrations for non-precipitating clouds, wind speed and liquid water content. These parameters form the basis for estimating cloud deposition to high elevation forests. This approach, by reducing the chemistry and meteorological variance within each subclass, enables subclass means to be used in place of hourly data for computing deposition flux in a manner that removes the bias associated with non-random, part-time cloud sampling. The frequencies of the various synoptic meteorological classes for each site used in this analysis are summarized in Table 4-24. Detailed definitions of these classes are found in the 1988 MCCP deposition report (Mohnen, 1988a). In summary, these classes are:

class 1: pre-warm front
class 2: NW sector of cyclone
class 3: post-cold front
class 4: warm sector of cyclone
class 5: stationary front
class 6: marine flow (off Atlantic)
classes 7&8: cutoff low in upper atmosphere
class 9: cap cloud

Table 4-24

Frequency of Occurrence of Various Synoptic Meteorological Classes by Site and Year for Growing Season

Site	Growing Season	Year	[a] Percent Frequency by Class								[b] Overall Cloud
			1	2	3	4	5	6	7&8	9	
MS	May 15 -	1986	26	3	15	36	0	11	9	0	27
	Oct 15	1987	27	18	8	26	0	19	2	0	22
		1988	31	7	9	51	0	2	1	0	7
WF	Jun 1 -	1986	16	3	33	32	0	6	8	2	44
	Sep 30	1987	27	10	26	20	0	5	0	12	40
		1988	9	1	36	39	1	0	7	6	23
MM	May 1 -	1986	0	3	15	43	3	35	0	1	32
	Oct 15	1987	0	3	13	43	3	21	5	11	29
		1988	0	1	18	39	1	27	3	11	22
WT	May 1 -	1986	0	1	12	57	1	28	0	2	38
	Oct 15	1987	0	5	20	41	2	21	4	6	30
		1988	0	2	16	40	2	21	13	6	26

a. Frequencies include only those cloud hours that could be classified. Frequencies may not sum to 100 due to roundoff.
b. Growing season cloud frequency (%).

Some of the synoptic meteorological classes contained only a few hours per site. Hence, a complete event characterization was not possible for all classes/subclasses at all sites. The subclasses that proved to be of most importance were those involving trajectories crossing or avoiding the Ohio Valley within synoptic class 4, north versus other trajectories in synoptic class 3, and east versus other trajectories in synoptic class 6. Examples of some of the more extreme differences found between subclasses are provided in Table 4-25. All data in the table are for precipitation-free hours only (this avoids the likely contamination of cloud water samples by precipitation).

Table 4-25

Examples of Differences Between Synoptic/Trajectory Subclasses for Selected Sites

Site	Synoptic Class	Subclass	Mean Conditions[a]			
			Speed (m/s)	LWC (g/m^3)	[H] (μeq/L)	[SO4] (μeq/L)
MS	6	East traj.	3.9	0.18	135	101
		Other traj.	3.6	0.16	275	66
WF	3	N traj.	11.3	0.42	30	27
		Other traj.	10.5	0.46	80	116
MM	4	Ohio traj.	11.0	0.16	756	947
		Other traj.	9.0	0.24	383	317
WT	6	East traj.	5.9	0.24	113	130
		Other traj.	5.7	0.23	400	586

a. Average of given parameter over all hourly measurements in subclass. LWC=liquid water content; [H]=cloud hydrogen ion; $[SO_4]$=cloud sulfate ion.

Cloud deposition estimates for each site were made by computing, for each subclass, the mean water deposition flux using the CDM and subclass-mean wind speed and liquid water content. Best estimates of canopy structure (Mohnen, 1988a) and growing season averages of other, less important CDM inputs (i.e., temperature and pressure) were used to estimate the gross (pre-evaporation) cloud water flux to specific forest canopies at each site. However, these deposition estimates are known to be sensitive to canopy structure and the local influences of topography and forest inhomogeneity on meteorology. Thus, the "potential" cloud deposition was also examined to determine site-to-site deposition-related variability not dependent on model performance.

Potential deposition (F_p) for ion X was defined as

$$F_p = u\ W\ [X]$$

where u is canopy top wind speed, W is liquid water content and [X] is the cloud water concentration of X. The deposition of cloud water and, consequently, X, to a forest canopy is known to be highly correlated with the product u W (Mueller and Imhoff, 1989). The mean potential deposition was computed, when possible, for each subclass as

$$F_p = [\{u\ \overline{W\ [X]_{ct}}\ \}/(\bar{u}\ \bar{W})]\ \bar{u}\ \bar{W}^3\ f_{adj} \quad , \qquad (1)$$

where $[X]_{ct}$ was the cube-root transformed cloud water concentration of X and the triple product of $uW[X]_{ct}$ was averaged over all hours in a subclass. The subclass mean solute concentrations [X] used to estimate F_p as in (1) are calculated as water content and wind speed weighted mean values, where the solute concentration is in cube root-transformed units. The concentration is back-transformed to μeq/l

after the weighted mean is determined. Use of the cube-root transform avoids errors associated with skewed data distributions. Weighting [X] by W and u minimizes the influence of any inter-variable correlations on the calculated triple products (uW[X]) for each synoptic class. The factor f_{adj} in (1) is a unique adjustment applied to the computed Fp for each site to correct for placement of the wind speed sensor (canopy top wind speed is most relevant to deposition flux), and the spatial differences, at some sites, between the location where W and [X] were measured and the canopy of interest. The basis for most of these adjustments were previously described in Mohnen (1988a). For Mt. Moosilauke, f_{adj} = 1 because no adjustments were needed. For Whiteface Mt., f_{adj} = 0.33, the product of the adjustment factor (0.33) for converting the summit u to canopy-top u, the adjustment factor (0.357) for converting the summit W to canopy-top W, and the adjustment factor (2.8) for converting the summit [X] to canopy-top [X] assuming that the elevational difference in [X] was due only to dilution (note that the water content and [X] adjustments cancel out). For Mt. Mitchell, the f_{adj} of 0.62 converted u from that measured at 16.5 m above ground to that at canopy-top (about 9 m). For Whitetop Mt. f_{adj} was computed to vary with wind direction (a direction frequency-weighted adjustment was computed for each subclass -- values ranged from 1.14 to 3.41), and the u measured at 7.4 m at the summit was converted to a u value for the nearby canopy top (about 15m).

The results of the F_p analysis are summarized in Table 4-26. Synoptic classes 2, 7 and 8 are rare and, therefore appear together under "other classes" in the table.

Table 4-26

Mean Potential Deposition by Subclass[a] and Site

(a) Sulfate ion

Class/Subclass[b]	Mean Potential Deposition ($\mu eq/m^2s$)			
	MS	MM	WF	WT
1/none	0.40	-	0.48	-
3/N traj.	-	0.39	-	0.20
3/non-N traj.	-	0.97	0.17	0.10
4/OH traj.	0.23	0.44	0.67	1.69
4/non-OH traj.	-	0.54	0.28	0.39
6/E traj.	0.05	0.33	-	0.29
6/non-E traj.	-	-	-	1.63
9/none	-	-	-	0.55
other classes	-	1.20	0.002	0.78

Table 4-26 (continued)

Mean Potential Deposition by Subclass[a] and Site

(b) Nitrate ion

Class/Subclass[b]	Mean Potential Deposition ($\mu eq/m^2 s$)			
	MS	MM	WF	WT
1/none	0.23	-	0.14	-
3/N traj.	-	0.12	-	0.03
3/non-N traj.	-	0.31	0.05	0.10
4/OH traj.	0.14	0.20	0.24	0.50
4/non-OH traj.	-	0.18	0.19	0.11
6/E traj.	0.02	0.16	-	0.20
6/non-E traj.	-	-	-	0.69
9/none	-	-	-	0.23
other classes	-	0.51	0.002	0.33

a. Results shown only for those subclasses having a minimum of 10 hr of observations. At Whitetop (WT), Fp is for plot A (summit).
b. "None" indicates no trajectory subclass considered.

In simple terms, F_p is the canopy-top horizontal cloud flux of ion X. Thus, high values of F_p are associated with windy conditions, dense clouds, and/or high cloud water ion content. Mean values of F_p vary greatly from one subclass to another and from one site to another. If all other conditions affecting cloud deposition were identical at all the sites (this means, among other things, that site-to-site variations in cloud droplet size distribution and canopy structure would be insignificant), then the mean hourly cloud deposition flux would vary according to F_p alone. Values of $F_p > 1$ $\mu eq\ m^{-2}\ s^{-1}$ were found for only two sites. Class-weighted mean values of F_p for each site (Table 4-27) show that the greatest overall cloud deposition potential (using the subclasses shown in Table 4-26 and weighting subclass-mean F_p values by the relative frequencies of observed wind speed for each subclass) for sulfate and nitrate is at Whitetop. This conclusion is valid only for those specific forest canopies considered here. For any given mountain, different forest canopies exposed to different meteorological conditions could have considerably lower or higher deposition potentials.

Table 4-27

Class-weighted Deposition Potential by Site for 1986-88

Site[a]	Class-weighted Deposition Potential (μeq/m2s)	
	Sulfate	Nitrate
MS	0.27	0.16
WF	0.35	0.12
MM	0.54	0.19
WT	0.64	0.24

a. The stand on MS is on a ridge, but not at the highest point on the mountain. The stand on MM is near the mountain summit. The stand on WF is several hundred meters below the summit (no trees are on the WF summit). The WT stand is near the summit.

Another measure of deposition potential--the growing season potential exposure (Pe)--takes into account the frequency of cloud impaction. As defined here, Pe represents the total cloud ion mass passing horizontally at canopy top through a unit area during a growing season. Thus, $Pe=<F_p>t$ where t is the growing season cloud exposure time and $<F_p>$ is the weighted mean value of F_p listed in Table 4-27. Combining the data in Tables 4-24 and 4-27, one finds that the potential growing season exposure to sulfate and nitrate cloud deposition was also a maximum at the Whitetop site for every year of the analysis. If the average cloud collection efficiency was known for a given stand (this is what the CDM is designed to compute) and the total collecting surface area of the stand per unit of ground area (SAI) was known, then the values in Table 4-28 could be used to estimate the growing season cloud deposition total.

Table 4-28

Growing Season Cloud Deposition Potential Exposure (Pe) by Site and Year

Site	Year	Growing Season Potential Exposure to Cloud Deposition[a]	
		Sulfate (eq/m2)	Nitrate (eq/m2)
MS	1986	1.0	0.6
	1987	0.8	0.5
	1988	0.3	0.2
	1986-88	0.7	0.4
WF	1986	1.6	0.6
	1987	1.5	0.5
	1988	0.9	0.3
	1986-88	1.3	0.5
MM	1986	2.5	0.9
	1987	2.3	0.8
	1988	1.7	0.6
	1986-88	2.2	0.8
WT	1986	3.5	1.3
	1987	2.8	1.0
	1988	2.4	0.9
	1986-88	2.9	1.1

[a] The Pe units used here are not to be interpreted as depositable $[H]_{aq}$ equivalents per unit of ground area, but equal instead to the growing season horizontal flux of $[H]_{aq}$ equivalents through a vertical plane at canopy top.

Hourly cloud water, sulfate and nitrate deposition estimates computed for each site using the CDM and class-mean meteorological and chemical variables are listed in Table 4-30 by class/subclass. Deposition was computed for both CDM evaluation stands on Whitetop (plots A and B mentioned earlier in this section) to illustrate the expected differences in deposition at two sites in relatively close proximity on the same mountain. WT Plot A was the open stand of low density and plot B was closed and dense.

Table 4-29

1986-88 Computed Mean Hourly Cloud Deposition by Site[a] and Subclass

(a) Cloud water

Class/subclass	Mean Computed Flux (mm/hr)				
	MS	MM	WF	WT(A)	WT(B)
1/none	0.11	-	0.11	-	-
3/N traj.	-	0.36	-	0.29	0.68
3/other traj.	-	0.31	0.10	0.19	0.21
4/OH traj.	0.15	0.21	0.09	0.49	1.29
4/non-OH traj.	-	0.25	0.09	0.22	0.18
6/E traj.	0.11	0.26	-	0.46	0.26
6/non-E traj.	-	-	-	0.60	1.25
9/none	-	0.09	0.06	0.27	0.55
other classes	-	0.15	0.04	0.29	0.63

(b) Sulfate ion

Class/subclass	Mean Computed Flux (eq/ha/hr)				
	MS	MM	WF	WT(A)	WT(B)
1/none	0.43	-	0.13	-	-
3/N traj.	-	1.15	-	0.59	1.30
3/other traj.	-	1.81	0.12	0.30	0.33
4/OH traj.	0.54	1.96	0.30	2.40	4.94
4/non-OH traj.	-	0.79	0.26	0.56	0.48
6/E traj.	0.11	0.50	-	0.60	0.39
6/non-E traj.	-	-	-	3.53	6.79
9/none	-	0.90	0.32	2.83	5.41
other classes	-	1.07	0.01	0.87	1.65

(c) Nitrate ion

Class/subclass	Mean Computed Flux (eq/ha/hr)				
	MS	MM	WF	WT(A)	WT(B)
1/none	0.24	-	0.06	-	-
3/N traj.	-	0.38	-	0.22	0.49
3/other traj.	-	0.59	0.03	0.16	0.18
4/OH traj.	0.32	0.94	0.04	0.71	1.46
4/non-OH traj.	-	0.28	0.10	0.22	0.19
6/E traj.	0.05	0.21	-	0.29	0.19
6/non-E traj.	-	-	-	1.67	3.22
9/none	-	0.39	0.17	0.96	1.83
other classes	-	0.41	0.003	0.43	0.81

a. Values shown only if hours of data exist for a subclass

The large variations between subclasses in computed water and ion flux was expected because of the large variations in meteorology and chemistry. Uncertainty in these flux estimates is unknown, but is expected to be larger for MS, MM and WF because of the greater uncertainty in the inputs to the model for these sites. It is also expected to be larger for those subclasses for which relatively few hours of data were collected (a mininum of 10 hours was used as a selection criterion). The WT flux estimates may be 20% - 30% higher for both plots, based on results of the CDM testing at these sites. However, some assumptions were necessary (e.g., previously analyzed relationships between summit wind speed at 7 m and canopy top wind speeds at both plots was assumed to hold for the entire 3-year period) to run the CDM for both plots A and B because site-specific wind speed and liquid water content data, representative of canopy top, were not always part of the subclass analysis done by Vong et al. (1989).

One interesting feature of note is the difference in computed cloud deposition between WT plots A and B. Deposition is sometimes greater at one and sometimes greater at the other, depending on subclass. This occurs because of the variation in prevailing wind direction among subclass type. Subclasses with a prevailing direction from southeast through west produce lower water deposition because of the sheltering effect of the mountain--plot B is in the lee of the mountain under these conditions, near the location of the lee eddy that has been observed frequently in videotaped images of the mountain. However, wind speeds are very similar for other directions, and the greater surface area at plot B causes deposition estimates to be much higher during these conditions.

Yearly growing season estimates of cloud deposition were computed using the synoptic class frequency data and the mean hourly flux estimates shown in Table 4-29. For each site, the subclass-mean flux estimates include subclasses that account for more than 75% of the total cloud impaction hours. The mean hourly flux during the unrepresented periods was assumed to equal the computed mean averaged over the represented periods. This was not expected to introduce much bias into the estimated deposition fluxes. However, int the future, other estimation techniques should also be tested. Table 4-30 summarizes the computed growing season deposition totals for each site.

Table 4-30

Computed Growing Season Total Cloud Deposition Flux by Site

Site	Year	Cloud Water	Cation Deposition (eq/ha)/(kg/ha)		
		Flux (cm)	[H]	[NH4]	[K + Mg + Ca + Na]
MS	1986	12.9	486/0.5	189/ 3.4	23/-
	1987	10.1	349/0.3	135/ 2.4	17/-
	1988	3.5	141/0.1	55/ 1.0	7/-
	86-88	8.8	325/0.3	126/ 2.3	16/-
WF	1986	11.6	144/0.1	107/ 1.9	16/-
	1987	10.5	156/0.2	96/ 1.7	17/-
	1988	6.0	81/0.1	64/ 1.2	10/-
	86-88	9.4	127/0.1	89/ 1.6	14/-
MM	1986	33.5	1205/1.2	377/ 6.9	100/-
	1987	27.5	1114/1.1	377/ 6.9	113/-
	1988	21.8	855/0.9	278/ 5.0	83/-
	86-88	27.6	1058/1.2	344/ 6.3	99/-
WT(A)	1986	53.1	1514/1.5	780/14.0	149/-
	1987	39.4	1189/1.2	602/10.8	110/-
	1988	34.3	1047/1.0	525/ 9.5	93/-
	86-88	42.3	1250/1.2	636/11.4	117/-
WT(B)	1986	77.3	2454/2.5	1229/22.1	221/-
	1987	61.2	1983/2.0	985/17.7	170/-
	1988	54.3	1764/1.8	866/15.6	144/-
	86-88	64.3	2067/2.1	1027/18.5	178/-

Table 4-30 (continued)

Site	Year	Anion Deposition (eq/ha)/(kg/ha) [SO4]	[NO3]	[Cl]
MS	1986	435/21	250/16	18/0.6
	1987	313/15	178/11	13/0.5
	1988	127/ 6	73/ 5	5/0.2
	86-88	292/14	167/11	12/0.4
WF	1986	220/11	54/ 3	6/0.2
	1987	202/10	67/ 4	5/0.2
	1988	128/ 6	31/ 2	3/0.1
	86-88	183/ 9	51/ 3	6/0.5
MM	1986	1091/52	406/25	68/2.4
	1987	1052/51	396/25	64/2.3
	1988	791/76	296/18	48/1.7
	86-88	978/60	366/23	60/2.1
WT(A)	1986	1737/83	667/41	87/3.1
	1987	1337/64	522/32	62/2.2
	1988	1173/56	463/29	54/1.9
	86-88	1416/68	551/34	68/2.4
WT(B)	1986	2817/135	1053/65	105/3.7
	1987	2244/108	858/53	82/2.9
	1988	1981/ 95	769/48	74/2.6
	86-88	2347/113	893/55	87/3.1

First note that uncertainties in canopy structure, and hence wind speed profile and droplet collection efficiency could easily make these values biased by a factor of two for all sites but WT. These values indicate changes relative to previous MCCP estimates of cloud deposition. For example, compared to the 1988 estimates for the 1987 growing season, the current 1987 sulfate deposition totals are higher for MS (+81%), lower for WF (-78%), higher for MM (+70%) and lower for WT(A) (-16%). To some extent these differences reflect changes in the CDM, but are mostly due to the more detailed characterization of the forest canopy which became available to MCCP through the efforts of the USDA Forest Service. In addition, improvements in characterizing the meteorology and chemistry associated with the various cloud event synoptic classes also necessitated a recalculation of the 1987 interception values. At WF the differences are also due to different assumptions concerning the wind speed profile, canopy structure and spatial variability of cloud water ion concentrations.

Another interesting finding is the large deposition flux estimated for WT(B) compared to WT(A). This is in spite of the fact that plot B at WT is occasionally sheltered in the lee of the mountain and intercepts less cloud water during such periods. The primary reason that WT(B) estimates are larger than those for WT(A) is the much greater canopy density of the former. CDM performance for WT(B) has been tested (Mueller et al., 1990) and found to be good (within 20% of throughfall rates). At WT, unlike the other sites, different wind directions were considered when computing deposition for the synoptic classes. This was done to maximimze the accuracy of the wind speed

adjustments needed to convert from 7 m speed at the summit to canopy top speed. There is a possibility that the partitioning of wind directions for the synoptic classes (which was done using only 1988 data) was not representative of all years of the analysis. The data indicate that west-northwest is the most prevalent wind direction for most synoptic classes, with south- southeast often being the second most prevalent. Overall, W and WNW winds--which do not place the WT(B) site in the mountain lee--appear to be twice as frequent as SE and SSE winds. Thus, on a long-term basis, the sheltering effect may not be as important as previously believed in its influence on deposition at this site.

The greatest computed growing season cloud deposition fluxes were found for WT and MM which also had the longest growing seasons. The differences between MS and WF may be greater than they appear here because of the uncertainty in model input data for these two sites. The mean monthly computed cloud deposition flux is provided in Table 4-31 to facilitate comparison of deposition rates independent of growing season length. Despite an almost 2:1 advantage in cloud frequency, WF mean monthly deposition estimates were generally lower than those for MS. The primary reason for the difference is the lower canopy surface area at the modeled WF site. Differences between the northern (MS and WF) and southern (MM and WT) sites are so great that they are likely to be real despite uncertainties in the model, and they exist independent of the longer growing season in the south.

Table 4-31

Mean Monthly Computed Cloud Deposition Flux

Site	Year	Computed Water Flux (mm/month)	Computed Ion Flux (kg/ha/month) [H]	[NH4]	[SO4]	[NO3]	[Cl]
MS	1986	25	0.10	0.67	4.1	3.0	0.12
	1987	20	0.07	0.47	2.9	2.2	0.09
	1988	7	0.03	0.19	1.2	0.9	0.04
	86-88	17	0.06	0.44	2.7	2.0	0.08
WF	1986	29	0.04	0.48	2.6	0.8	0.05
	1987	26	0.04	0.42	2.4	1.0	0.05
	1988	15	0.02	0.28	1.5	0.5	0.03
	86-88	23	0.03	0.39	2.2	0.8	0.04
MM	1986	60	0.22	1.21	9.4	4.5	0.43
	1987	49	0.20	1.21	9.0	4.4	0.40
	1988	39	0.15	0.89	6.9	3.3	0.30
	86-88	49	0.19	1.11	8.4	4.0	0.38
WT(A)	1986	95	0.27	2.51	14.9	7.4	0.55
	1987	70	0.21	1.94	11.5	5.8	0.39
	1988	61	0.19	1.69	10.0	5.1	0.35
	86-88	75	0.22	2.04	12.1	6.1	0.43
WT(B)	1986	138	0.44	3.95	24.1	11.7	0.67
	1987	109	0.35	3.17	19.2	9.5	0.52
	1988	97	0.32	2.78	17.0	8.5	0.47
	86-88	115	0.37	3.30	20.1	9.9	0.55

The cloud deposition information contained in this report has been produced using state-of-the-science computational methods. The state of the science is such that considerable uncertainty remains in the deposition estimates despite recent advances in our knowledge concerning cloud deposition to forest canopies. However, the deposition potential--as defined previously--and the computed deposition fluxes both indicate trends toward higher cloud deposition at the southern sites. Differences between northern and southern sites are due to site specific parameters and not latitude. The southern sites have a significantly different canopy structure than the Whiteface Mtn. site. The southern sites are also exposed to a different mix of cloud event types (as defined by synoptic meteorology and computed air trajectories) when compared to the northern sites, and different event types have been shown to be characterized by differences in conditions affecting cloud deposition. More work needs to be done to better define conditions at each site before more accurate deposition estimates can be obtained.

Dry Deposition

Model Data Requirements --

The Atmospheric Turbulence Diffusion Laboratory (ATDL) model used by the MCCP, DRYDEP, calculates the dry deposition velocities of sulfur dioxide, ozone, nitric acid vapor, sulfate, and nitrate from meteorological and site-specific biological information. Site information includes: major and minor plant species type, leaf area index for plant species, and site location. Stomatal resistance parameters include plant species, light response coefficients, minimum stomatal resistance, and optimum, maximum, and minimum temperatures for stomatal function. All available meteorological data such as wind speed, standard deviation of direction, global radiation, air temperature, relative humidity, canopy wetness, and rainfall are also used.

Model Application --

The current big-leaf model does not directly account for nonuniform terrain. Only Howland met the model requirements. Therefore, to estimate dry deposition at the other sites, the model was run with the aerodynamic resistance set to zero in an attempt to estimate the maximum effect of an uneven surface. Table 4-32 shows the warm period and the species for which deposition flux density was inferred at each site.

TABLE 4-32

Chemical Species for Which Deposition was Inferred

Site	Period	Species
Howland, ME	4/7 - 10/12	O_3 SO_2 HNO_3 SO_4 NO_3^-
Whiteface, NY	6/1 - 10/6	O_3 SO_2 SO_4
Moosilauke, NH	5/15 - 10/21	O_3
Shenandoah, VA	4/1 - 11/16	O_3 SO_2 HNO_3 SO_4 NO_3
Mitchell, NC	5/1 - 10/21	O_3 SO_2 NO_x
Whitetop, VA	4/26 - 10/18	O_3 SO_2

Model Sensitivity Testing --

The effect of estimating deposition velocities when setting aerodynamic resistance to zero varies by chemical species. For example, the effect for ozone, which has considerable canopy resistance, would be less than the effect for HNO_3, for which canopy resistance is assumed to be zero. Ozone deposition velocities for 1987 Howland data showed a 14% increase when calculated with zero resistance. HNO_3 deposition velocities calculated with and without aerodynamic resistance for Shenandoah showed an offset of 0.36 cm/sec, and an increase of 59% when calculated with zero resistance.

Some uncertainty was included in leaf area index (LAI) values used for most sites; only the values for Mitchell were obtained by direct measurement. Therefore, the sensitivity of deposition velocity to changes in LAI was evaluated using ozone at Howland. The primary and secondary tree species have LAIs of 5.5 and 0.2, respectively. Deposition velocity (Vd) was calculated with primary LAI values from 2.5 to 11.0, with the secondary LAI held constant. For each of seven values of LAI, the resulting Vd values were averaged for the period of record. Paired values of normalized LAI and Vd were determined using the accepted values as the base. The results showed that if estimated LAI values were in error by as much as 25%, Vd would either be overestimated by 12% or underestimated by 15%. This is probably within the uncertainty of the model estimates. The current model also tended to overestimate deposition velocities for chemical species for which canopy resistance is important, such as ozone and SO_2, (Matt and Womack, 1988; Matt et al., 1987).

Model Results --

The warm season total deposition flux density (kg/ha/mo) is a function of deposition velocity, concentration, and season length. Table 4-33 displays the available dry deposition values for 1987 and 1988. The values were derived by multiplying weekly deposition velocities derived from the model by the appropriate weekly concentrations for the warm period at each site. Estimates based on the mean were substituted for missing data. Estimates for 3 to 6 missing weeks yielded reasonable results. However, as in the case of SO_2 and SO_4 for Shenandoah-1, substituting estimates for more than 50% of the warm period led to anomalous results.

During 1987 ozone values for Whiteface seem low due to moderate deposition velocities and the short season. For that year, Moosilauke and Whiteface, if scaled to the Howland warm season, would have ozone deposition values of approximately 6.81 and 9.75 kg/ha/mo, respectively. On the other hand, if Shenandoah was similarly scaled down, ozone deposition would still be the largest with 9.29 kg/ha/mo.

Although ozone exposure was considerably greater in 1988 than in 1987, deposition flux density was approximately the same for both years for three northern sites due to a marked decrease in deposition velocities during 1988. Deposition velocities also decreased somewhat at the southern sites, but this was offset by very much higher average ozone concentrations.

Sulfur dioxide concentrations were also higher during 1988 at all reporting sites. However, SO^2 deposition velocities were uniformly lower, producing only modest increases in deposition flux density at Howland and Whiteface. Only Mt. Mitchell recorded a substantially higher SO^2 flux density during 1988.

In general, for chemical species with sufficient available data, northern sites received less dry deposition than southern sites. However, within each region there was marked variability in seasonal ozone and SO_2 deposition. Also, seasonal deposition totals did not adequately reflect weekly site-specific variability.

TABLE 4-33

MCCP Warm Season Dry Deposition

Site	Elev.(m)	Dry deposition flux density (kg/ha/mo)					
		O_3	SO_2	SO_4	HNO_3	NO_3	NO_2
Howland, ME (1987)	65	5.48	0.22	0.11	0.29	0.002	
(1988)		5.39	0.28	0.15	0.37	0.03	
Moosilauke, NH (1987)	1000	5.96					
(1988)		6.57		0.31	1.61	0.01	
Whiteface, NY (1987)	1483	6.46	0.30	0.36			
(1988)		6.09	0.51	0.28			
Shenandoah1, VA (1987)	1015	14.06	3.06	0.80	0.007		
(1988)		14.83					
Shenandoah3, VA (1987)	524	13.36	1.32	0.19	1.03	0.005	
(1988)		15.11					
Mt. Mitchell1, NC (1987)	1950	7.70	0.39				0.20
(1988)		12.7	1.48		1.64		0.21
Mt. Mitchell2, NC (1987)	1775	7.81					
(1988)		10.56					
Whitetop Mt, VA (1988)	1689	11.95	0.75				

Precipitation Deposition

In this section, wet deposition for the 1987 and 1988 warm seasons at the MCCP sites is estimated using standard NADP/NTN or MAP3S measurements of precipitation amount and chemistry. These estimates were obtained from NADP and MAP3S reports for the network stations closest to the MCCP sites or, in one case, for an NADP-type collector operated at the Shenandoah MCCP site.

To represent wet deposition via precipitation at the Howland, ME, site, Mohnen (1988a) used data from the NADP/NTN site at Greenville, ME, located approximately 100 km west of Howland. Other MCCP sites had NADP/NTN type samplers no more than 25 km distant. Details for these stations are shown in Table 4-34.

For comparison with MCCP cloud water deposition, wet deposition only for the warm season are reported as monthly deposition. The growing season is longest at the Shenandoah and Howland sites, extending from early April at both locations to mid-November and early October, respectively. Mt. Mitchell and Whitetop Mountain have the next longest growing seasons, followed by Mt. Moosilauke and Whiteface with the shortest (June to early October). For consistency the concurrent growing season

(June to October) was used for 1987 data and all available data (until August 23) were used for the same period during 1988.

TABLE 4-34

Sites Used to Estimate Wet Deposition via Precipitation for MCCP Sites.

MCCP Site	Wet Deposition Site	Lat.	Long.	Elevation
Howland Forest, MA	NTN Greenville ME09	45°9'	69°9'	322 m
Whiteface Mtn, NY	NTN Whiteface NY98	44°3'	73°1'	622 m
	MAP3S Whiteface	----------	(co-located)	----------
Moosilauke, NH	NTN Hubbard Brook NO2	43°6'	71°2'	250 m
Shenandoah, VA	NTN Big Meadows VA28	38°1'	78°6'	1074 m
Whitetop, VA	NTN Whitetop VA29	36°8'	81°6'	1689 m
Mt Mitchell, NC	NTN Clingmans Peak NC45	35°44'	82°17'	1987 m

MCCP pollutant deposition and climatological data tend to form two distinctly different groups. Because of physical proximity, and similar influences from synoptic systems, the Howland, MA, Whiteface Mountain, NY, and Mt. Moosilauke, NH, sites are considered together as northern sites and the Shenandoah, VA, Whitetop, VA, and Mt. Mitchell, NC, sites are grouped together as southern sites.

The wet deposition data for the two groups of sites are shown in Table 4-35 [kg/ha/mo] for SO_4^{2-}, NO_3^-, NH_4^+, and H^+ ions in precipitation (not cloud) collected at the NADP/NTN sites. At Whiteface Mountain, NY, both NADP/NTN and MAP3S collected precipitation, but during 1987 the NADP/NTN site was hit by lightning, so only the MAP3S data were used. Rain gauge data were used to calculate precipitation amount. About 98% of the precipitation was analyzed for chemical composition during 1987. Missing samples were replaced by actual precipitation amount for that week and the volume-weighted mean concentrations of growing season data for that site and year. A further discussion of the calculation procedures is presented by Mohnen (1988a).

The data indicate that the most westerly sites in the north, Whiteface, NY, and Moosilauke, NH, received greater wet deposition via precipitation than did the more northeasterly location in Maine. Big Meadows, VA, and nearby Shenandoah had the most NH_4^+ deposition from rain.

These data suggest that the highest elevation site, Mt. Mitchell, NC, had the largest wet flux for SO_4^{2-} and NO_3^-. Hubbard Brook, NH, (a low elevation site in the north), Shenandoah, VA (a medium--elevation site in the south), and Whitetop Mountain, VA, (the second highest site) had similar wet deposition for 1987. The lowest site, Greenville, ME has the least wet deposition via precipitation.

For comparison, available data for the 1988 growing season are presented in Table IV-18 for several MCCP sites. Data for all of the same sites as for 1987 are not currently available.

The 1988 precipitation chemistry data indicate that the southern sites exceeded the northern sites in SO_4^{2-} and NO_3^- deposition, with the differences being greater for $SO_4^=$. Shenandoah had the most NH_4^+ deposition. These values for wet deposition via precipitation are about 20% to 50% of the deposition via cloud water interception during 1987.

TABLE 4-35

Concurrent Warm Season Wet Deposition via Precipitation in the Eastern USA from NADP/NTN or MAP3S Sites Near MCCP Monitoring Locations (for the period 26 May to 6 October, 1987, or 31 May to 23 August, 1988.) [kg ion/ha/mo]. Data are unvalidated and subject to change.

Location	H^+	NH_4^+	SO_4^{2-}	NO_3^-	Year
Greenville, ME	*	0.11	1.28	0.65	1987
Whiteface, NY	*	0.33	2.27	0.95	1987
	.032	1.11	1.18	0.93	1988
Hubbard Brook, NH	*	0.28	2.84	1.46	1987
	.041	0.11	2.16	1.10	1988
Whitetop, VA	*	0.31	3.20	1.32	1987
	.053	0.14	2.84	1.17	1988
Mt. Mitchell, NC	*	0.46	5.15	2.26	1987
	.051	0.12	2.69	1.19	1988
Big Meadows, VA	*	0.48	2.32	1.43	1987
	.016	0.92	2.90	1.34	1988
Shenandoah, VA	*	0.35	3.11	1.25	1987
	.045	1.76	2.80	1.19	1988

* Not reported

Wet, Cloud and Dry Deposition

Cloudwater deposition is the factor which differentiates mountains from lowlands and the relative importance of cloud and rain deposition is discussed here. It is hypothesised that the principal deleterious effects on forests which might be associated with such deposition are direct effects of acids on foliage, mobilization of aluminum because of ions deposited to soils, or excess nitrogen fertilization influence on winter damage. Since most of these effects are more significant for warm season deposition, the analysis intervals were selected for each site to correspond roughly with the periods between local forest bud break and fall dormancy. These periods differed to some extent for the MCCP sites.

However, cloudwater deposition calculated for Whitetop Mountain seems to stand out above the rest. Since this peak is substantially lower than Mount Mitchell which is only about 90 miles southwest of Whitetop, the difference is difficult to explain.

It is apparent from Tables 4-31, 4-33 and 4-35 that cloud deposition based on current models estimate, is comparable to deposition of rain at all summit locations. Dry deposition of sulfur-bearing compounds is reported as sulfur rather than as SO_2 or sulfate anion (Table 4-33).

If the geographic gradients suggested by the data in Table 4-33 continue in future estimates, then some interesting conclusions could be drawn. First, the Shenandoah site seems to experience the greatest dry deposition. In the case of both sulfur and nitrogen budgets, dry deposition is significant in the three Northern sites. This gradient reflects the wet deposition gradient reported in both the National Academy of Sciences findings on acid rain and in the 1989 NAPAP interim assessment. The more easterly sites, Moosilauke and Howland are very much lower in pollutant burden than are the Whiteface and southern sites. With the exception of Moosilauke and Howland, the MCCP site warm season sulfur deposition is 20-40 kg/ha-year. This level is about that reported for annual deposition at the more contaminated low elevation sites in rain.

It is not yet clear whether cold season sulfur deposition should really be factored in to the mountain estimates. There is some evidence of SO_2 loss from snowpack and the excess ions in snowpack melt are of much greater consequence in the acidification of lakes than they might be in determining ion concentrations in frozen soils. In any case, it is clear that mountaintop sulfur deposition is quite significant, approximating or exceeding that of near-source low elevation sites.

For all of the data reported so far, dry deposition sulfate ion in fine particles is very small relative to wet deposition and in many cases relative to that of SO_2 dry deposition as well. Dry deposition of nitrogen is more uncertain because of the lack of dependable data for nitric acid. However, ammonium ion dry deposition accounts for a substantial amount of the total nitrogen deposited; this material appears to be mainly in the form of ammonium sulfate. The season-long values for Shenandoah for nitric acid try deposition may be biased somewhat low of the actual values, because most of the determinations were performed relatively late in the study period. In any case, the nitric acid dry deposition appears to be quite significant for that site, and probably for the other southern sites as well.

Overall growing season fertilization rates are 10-20 kg N/ha-season for the southern sites and lower for the northern ones. This application of nitrogen is driven mainly by wet deposition and the estimates could go up or down as model predictions are improved.

Table 4-36 summarizes the average monthly sulfur and acidic nitrogen (excludes ammonium ion) deposition fluxes determined to-date for the MCCP sites for the 1987-88 growing seasons. At the cloud-free Howland site, dry deposition appears to account for less than one-third of the total acidic sulfate and nitrate ion deposited. The Shenandoah site has a very low cloud impaction frequency due to its low elevation, and dry deposition is probably of greater relative importance there (it could exceed 50% of the total deposition flux) than at any of the other high elevation sites. Estimates for Whiteface suggest that dry sulfur deposition accounts for less than 30% of the total sulfur deposition flux.

The greatest amount of information is available for comparing estimated cloud and wet deposition at the four high-elevation spruce-fir sites (Moosilauke, Whiteface, Whitetop and Mitchell). Cloud-to-wet deposition ratios (Table 4-37) vary from near unity at Moosilauke and Whiteface to near 4 at Whitetop. In terms of acid deposition, clouds seem to be of greater relative importance at the two southern sites (WT and MM) than at the two northern sites.

TABLE 4-36

(a)

Estimated 1987-88 Sulfur Deposition Budgets at MCCP Sites[a]

Site	Year	Growing Season Mean S Deposition (kg S/ha-mo)[b]		
		Wet (Measured)	Cloud (Computed)	Dry (Computed)[c]
Howland, ME[d]	87	0.43	-	0.15
	88	-	-	0.19
	87-88	-	-	0.17
Moosilauke, NH[e]	87	0.95	0.97	-
	88	0.72	0.40	-
	87-88	0.84	0.69	-
Whiteface, NY	87	0.76	0.80	0.27
	88	0.39	0.50	0.35
	87-88	0.58	0.65	0.31
Shenandoah, VA	87	1.04	-	1.80
	88	0.93	-	-
	87-88	0.99	-	-
Whitetop, VA[f]	87	1.07	3.83	-
	88	0.95	3.33	-
	87-88	1.01	3.58	-
Mitchell, NC	87	1.72	3.00	-
	88	0.90	2.30	-
	87-88	1.31	2.65	-

a. Not all data were available in time for inclusion in this report.
b. Average fluxes for combined 1987-88 period were computed by averaging values given separately for the two years.
c. Sulfuf (S) flux computed as the sum of individual computed SO_2 gas and SO_4 aerosol fluxes (approximate maximum rates due to assumption that aerodynamic resistance within forest canopy was zero).
d. Greenville, ME NADP data used for wet deposition fluxes.
e. Hubbard Brook, NH NADP data used for wet deposition fluxes.
f. Near-summit (plot A) site.
g. Nitrogen in ammonium ion not considered.
h. Nitrogen (N) flux computed as the sum of individual computed HNO_3 gas and NO_3 aerosol fluxes (approximate maximum rates due to assumption that aerodynamic resistance within forest canopy was zero).

TABLE 4-36 (continued)

(b)

Estimated 1987-88 Acidic Nitrogen Deposition Budgets at MCCP Sites[a,g]

Site	Year	Growing Season Mean N Deposition (kg S/ha-mo)[b]		
		Wet (Measured)	Cloud (Computed)	Dry (Computed)[h]
Howland, ME[d]	87	0.15	-	0.07
	88	-	-	0.09
	87-88	-	-	0.08
Moosilauke, NH[e]	87	0.33	0.50	-
	88	0.25	0.20	-
	87-88	0.29	0.35	-
Whiteface, NY	87	0.21	0.23	-
	88	0.21	0.11	-
	87-88	0.21	0.17	-
Shenandoah, VA	87	0.28	-	0.23
	88	0.27	-	-
	87-88	0.28	-	-
Whitetop, VA[f]	87	0.30	1.31	-
	88	0.26	1.15	-
	87-88	0.28	1.23	-
Mitchell, NC	87	0.51	0.99	-
	88	0.27	0.75	-
	87-88	0.39	0.87	-

a. Not all data were available in time for inclusion in this report.
b. Average fluxes for combined 1987-88 period were computed by averaging values given separately for the two years.
c. Sulfuf (S) flux computed as the sum of individual computed SO_2 gas and SO_4 aerosol fluxes (approximate maximum rates due to assumption that aerodynamic resistance within forest canopy was zero).
d. Greenville, ME NADP data used for wet deposition fluxes.
e. Hubbard Brook, NH NADP data used for wet deposition fluxes.
f. Near-summit (plot A) site.
g. Nitrogen in ammonium ion not considered.
h. Nitrogen (N) flux computed as the sum of individual computed HNO_3 gas and NO_3 aerosol fluxes (approximate maximum rates due to assumption that aerodynamic resistance within forest canopy was zero).

Table 4-37

Estimated Cloud-to-Wet Deposition Flux Ratios

Site	Year	Sulfate	Nitrate
MS	1987	1.0	1.5
	1988	0.6	0.8
	87-88	0.8	1.2
WF	1987	1.1	1.1
	1988	1.3	0.5
	87-88	1.1	0.8
WT[a]	1987	3.6	4.4
	1988	3.5	4.4
	87-88	3.5	4.4
MM	1987	1.7	1.9
	1988	2.6	2.8
	87-88	2.0	2.2

a. Near-summit plot.

In summarizing the deposition results, MCCP and IFS researchers have estimated that cloudwater deposition provides a substantial fraction of the total chemical deposition to high-elevation eastern USA forests. Lindberg and Johnson (1989) estimated that cloudwater contributes approximately 25% to 50% of total SO_4^{2-}, N, and H^+ deposition at two high-elevation sites on Whiteface Mountain, NY and in the Great Smoky Mountains National park, NC. MCCP results indicate that cloudwater SO_4^{2-}, H^+, NO_3^-, and NH_4^+ deposition exceeded wet deposition via precipitation for three sites located above 1400 m, while two lower elevation sites received cloudwater chemical inputs that were at least one-half of precipitation chemical deposition. Preliminary estimates for dry deposition are considerably smaller than either cloud or precipitation deposition at all MCCP sites except one site located in a deciduous forest at Shenandoah, VA (Mohnen, 1988a; Krovetz et al., 1989). While the current large and undefined uncertainties that are associated with cloud water and dry deposition models make firm estimates for complex terrain impossible, there is a general consensus in the literature that cloud water deposition represents a substantial increment over precipitation and dry inputs to high-elevation forests in the eastern USA.

SECTION 5

METEOROLOGICAL ASSESSMENT OF AIR QUALITY

CASE STUDY ON THE IMPACT OF AIR MASS ORIGIN ON AIR QUALITY

A simple case study was performed at a northern MCCP site to investigate the relationship between synoptic weather patterns and cloudwater ion and gas phase pollutant concentrations. If meaningful results could be obtained, then there would be a basis to extend this type of analysis to other sites and to other synoptic categories.

In this case, ion concentrations during warm sector synoptic conditions were investigated for the 1986-88 field seasons at Whiteface Mountain. Warm sector conditions are defined as the southeast sector of the classical cyclone model or the west side of a high pressure system. Stagnant conditions sometimes occur with semi-permanent "Bermuda High" type situations which often lead to air pollution episodes. Therefore, warm sector conditions were classified as "non-stagnant" and "stagnant", the latter requiring that warm sector, high pressure conditions at or to the south of the site were persisting for at least 72 hours.

Table 5-1 shows the concentrations of selected ions for cloudwater and pollutant concentrations for gases and sulfate aerosol for three categories: non-warm sector (category 0); non-stagnant warm sector (category 1) and stagnant warm sector (category 2) conditions based on hourly samples. For all ions, gases and sulfate aerosol, both warm sector concentrations are greater than other sector concentrations by a factor of 2 to 3, indicative of the relatively warm and humid conditions, together with favorable south to southwesterly trajectories, which favor high pollutant levels. Stagnant conditions exhibited the maximum sample concentrations for all ions, gases and sulfate aerosol.

METEOROLOGICAL INFLUENCES ON CLOUDWATER CHEMISTRY

Air mass back-trajectories during all hours at all MCCP sites originated most frequently from the western quadrant, and least frequently from the southeast. For cloudy periods, back trajectories favored the southwestern quadrant, with a secondary frequency peak in the eastern quadrant, especially for southern sites.

Synoptic scale mechanisms responsible for cloud formation differed for northern and southern portions of the Appalachians. Nearly two-thirds of cloud events at Whiteface Mountain, NY, were associated with pre-warm frontal synoptic conditions. However, at Whitetop Mountain, VA, over two-thirds of all cloud events were associated with warm-sector and marine flow conditions.

The amount of acidic deposition to a mountain site depends on factors such as cloud frequency, windspeed, liquid water content, and the history of the air mass in which the clouds are formed. To relate cloudwater composition with large-scale circulation features, cloud events collected during the 1986-88 field seasons were classified according to nine synoptic features responsible for cloud production. This scheme classifies cloud events according to a mountain's location relative to surface weather map features for hours when cloud was observed at the sites.

TABLE 5-1

CLOUD WATER ION STATISTICS
CATEGORIZED BY SYNOPTIC CONDITIONS
FOR WHITEFACE MT. SUMMIT: 1986-88

HYDROGEN ION (hourly measurements: μmol/l)

Synoptic Cat.	Frequency	Percent	Mean	Maximum	Minimum
0	382	64.9	134	1288	0.3
1	139	23.6	335	1412	0.3
2	68	11.5	320	1778	50.1
1&2	207	35.1	330	1778	0.3
All	589	100	203	1778	0.3

SULFATE ION (hourly measurements: μmol/l)

Synoptic Cat.	Frequency	Percent	Mean	Maximum	Minimum
0	384	65.0	78	547	0.4
1	139	24.5	203	668	0.3
2	68	11.5	214	1112	40.8
1&2	207	35.0	206	1112	0.3
All	591	100	123	1112	0.3

NITRATE ION (hourly measurements: μmol/l)

Synoptic Cat.	Frequency	Percent	Mean	Maximum	Minimum
0	384	65.0	61	752	0.7
1	139	23.5	130	632	0.3
2	68	11.5	191	1344	18.4
1&2	207	35.0	150	1344	0.3
All	591	100	92	1344	0.3

AMMONIUM ION (hourly measurements: μmol/l)

Synoptic Cat.	Frequency	Percent	Mean	Maximum	Minimum
0	384	65.0	80	770	0.7
1	139	23.5	181	652	0.5
2	68	11.5	227	920	43.0
1&2	207	35.0	196	920	0.5
All	591	100	121	920	0.3

Synoptic Categories:

0 - not in warm sector
1 - warm sector, non-stagnant conditions
2 - warm sector, stagnant conditions

TABLE 5-1 (Continued)

GAS CONCENTRATION STATISTICS
CATEGORIZED BY SYNOPTIC CONDITIONS
FOR WHITEFACE MT. SUMMIT: 1986-88

OZONE (hourly measurements: ppbv)

Synoptic Cat.	Frequency	Percent	Mean	Maximum	Minimum
0	6590	78.0	41.9	127.1	11.0
1	1292	15.3	57.3	132.6	23.0
2	565	6.7	69.7	135.3	32.0
1&2	1857	22.0	61.1	135.3	23.0
All	8447	100	46.1	135.3	11.0

SULFUR DIOXIDE (hourly measurements: ppbv)

Synoptic Cat.	Frequency	Percent	Mean	Maximum	Minimum
0	4581	75.6	0.8	13.7	0
1	1025	16.9	2.4	17.7	0
2	456	7.5	2.3	20.0	0
1&2	1481	24.4	2.3	20.0	0
All	6062	100	1.2	20.0	0

HYDROGEN PEROXIDE (hourly measurements: ppbv)

Synoptic Cat.	Frequency	Percent	Mean	Maximum	Minimum
0	594	68.1	0.6	6.0	0.0
1	90	10.3	0.8	2.8	0.1
2	188	21.6	1.5	3.8	0.0
1&2	278	31.9	1.3	3.8	0.0
All	872	100	0.8	6.1	0.0

Synoptic Categories:
- 0 - not in warm sector
- 1 - warm sector, non-stagnant conditions
- 2 - warm sector, stagnant conditions

Four of the nine synoptic types are associated primarily with the main sectors of a cyclone. The nine synoptic types are: a) pre-warm-frontal clouds, b) northwest-sector of cyclone clouds, c) post-cold-frontal clouds, d) warm-sector synoptic types including the southeast sector of the cyclone and stagnant conditions, e) stationary-front cloudiness, f) easterly flow that advects moisture inland from the Atlantic, g) and h) cutoff-low-aloft types (these two categories were created based on the location of the cutoff relative to the site), and i) cap cloud which involves no important synoptic scale features within several hundred km of the site and high cloud bases at nearby airports.

A site-cloud climatology was developed for Whiteface Mountain, NY, Mt. Moosilauke, NH, Whitetop Mountain, VA, Shenandoah, VA, and Mt. Mitchell, NC. As Table 5-2 shows, at the northern sites (NY and NH), the post-cold-frontal type (c) and warm-sector synoptic types (d) comprised most of total cloud hours. At the southern sites (VA and NC), the warm-sector type (d) was most frequent while the second most frequent cloud synoptic type (f) was a result of easterly flow.

Table 5-2

Percentage of Cloud Hours by Synoptic Type for Five MCCP Mountain Sites
(1986 - 88 Field Seasons)

Synoptic Type	Whiteface	Moosilauke	Shenandoah	Whitetop	Mitchell
a. pre-warm front	16.5	25.5	6.5	0.1	-
b. northwest of cyclone	5.8	11.5	4.5	4.2	2.5
c. post cold front	32.4	11.0	23.9	16.0	15.2
d. warm sector	30.7	31.8	11.3	45.4	41.9
e. stationary front	0.3	0.1	3.2	1.5	2.5
f. easterly marine flow	4.0	15.9	43.7	23.1	26.8
g. low aloft (W-flow)	3.4	2.0	1.8	5.0	2.9
h. low aloft (SE-flow)	1.4	2.2	5.1	0.1	-
i. cap cloud	5.5	-	-	4.6	8.2

Two types of statistical models were applied to the data for 1986-88. The first model, partial least-squares regression on latent variables (PLS), was used to identify combinations of meteorological parameters that predicted linear combinations of chemical concentrations (Vong et al., 1988a; Geladi and Kowalski, 1985). The linear combinations were formed from correlated variables.

The original data consisted of hourly cloud water samples and corresponding hourly meteorological parameters of trajectory, synoptic type, cloud type, and local winds. Trajectories were obtained from a mixed-layer model that uses rawinsonde measurements to define the transport field (Kahl and Samson, 1988). The chemical data were square-root transformed before analysis to keep extreme samples from dominating the results. Hourly samples from continuous cloud events were aggregated into sub-events with consistent trajectories.

A six-component PLS model identified significant covariance structure in the meteorological variables that was related to the chemical data. The correlations that were revealed among synoptic types and mixed layer trajectories were physically realistic. For example, the "marine" synoptic type occurred with easterly trajectories and high cloudwater chloride concentrations, the warm-sector synoptic type (d) occurred frequently with SW trajectories and high SO_4^{2-} concentrations, and the post-cold-

frontal synoptic type (c) occurred with NW trajectories and low SO_4^{2-} concentrations. This agreement added confidence to the use of the two types of meteorological variables (synoptic type and mixed-layer trajectory). PLS identified significant covariance in the chemical data also. H^+, NH_4^+, SO_4^{2-}, and H^+ were correlated.

Based on the PLS modeling, the following synoptic-trajectory classes were posed as the most important influences on cloudwater chemistry at the four sites for further analysis:

- Synoptic type (d) = warm-sector, SW trajectory
- Synoptic type (d) = warm-sector, all other trajectories
- Synoptic type (i) = cap-cloud, SW trajectory
- Synoptic type (i) = cap-cloud, all other trajectories
- Synoptic type (c) = post-cold frontal, NW trajectory
- Synoptic type (c) = post-cold-frontal, all other trajectories
- Synoptic type (f) = marine-flow, SE to NE trajectory
- Synoptic type (f) = marine-flow, all other trajectories
- All other synoptic type (a, b, e, g, h) and trajectory combinations

The second model, analysis of variance (ANOVA), was used to test the hypothesis that this smaller number of meteorological variables would describe variability in cloudwater chemistry. Classes 3 and 4 were combined with 1 and 2 because both represented stable atmospheric conditions, and there were few observations of cap cloud compared to synoptic types c, d, and f. The ANOVA model (Box et al., 1978; Vong et al., 1988b) tested for differences in cloudwater chemistry for the seven remaining synoptic/trajectory classes.

This model was highly significant ($p < 0.001$). The three main effects were significant; there were differences between sites, cloud-only (non-precipitating) and mixed rain/cloud (precipitating) types, and synoptic-trajectory classes. Table 5-3 gives mean ion concentrations for all MCCP sites for selected synoptic-trajectory classes and cloud types.

TABLE 5-3
Analysis of Variance Results for Four Ion Concentrations:
Mean Values for all MCCP Sites (1986-88) in μeq/L for Selected Synoptic-Trajectory Classes, All Clouds

	Ion			
Synoptic-Trajectory Class	SO_4^{2-}	H^+	NH_4^+	NO_3^-
Warm sector - Ohio Valley Trajectory	361	304	161	133
Warm sector - Other trajectories	246	190	117	88
Post cold front - NW trajectories	131	90	59	43
Marine flow - E trajectory	134	138	47	61
Mean of all 9 classes (all cloud)	199	170	86	78
Mean: non-precipitating cloud	262	220	116	104
Mean: precipitating cloud	113	101	47	43

The site-class interaction terms confirmed different warm-sector synoptic type dependencies of SO_4^{2-} on trajectory. The highest SO_4^{2-}, H^+, NH_4^+, and NO_3^- concentrations occurred with the

warm-sector synoptic type (d) when the trajectories pointed toward the Ohio Valley. This is consistent with the notion of transport from that high emission area.

Remaining variation within each synoptic-trajectory class is expected to be related to variation in gas- and aqueous-phase chemical processes, scavenging, and meteorology that could not be described in the synoptic-trajectory classification scheme. For example, the height of the monitoring site above cloud base should affect the amount of available liquid water and, therefore, aqueous phase concentrations, but testing that effect for all sites was not possible because cloud base data were not always available. For one site (Whiteface Mountain, NY) there were consistent differences between two collection sites located at different elevations when 106 hours of simultaneous data were compared; the lower site had higher concentrations. The modeling approach described here was able to characterize several factors that influence the composition of mountain cloudwater composition in the eastern USA.

GAS CONCENTRATION VS. AIR MASS TRAJECTORIES

Ozone

Mean ozone concentration is not strongly a function of 36-hour air mass trajectory sector, as shown in Figures 5-1 through 5-6 (Appendix E) for the 1986-88 period. Northern sites have somewhat higher mean values for southeasterly to southwesterly trajectories while southern sites exhibit a fairly uniform distribution. A clearer pattern emerges from high ozone concentrations. Figures 5-7 through 5-17 (Appendix E) focus on trajectory dependency for ozone values greater than 70 ppb and 100 ppb. Howland, Moosilauke and Whiteface favor west-southwesterly trajectories during ozone episodes whereas the southern sites exhibit a broader and more northerly distribution of trajectories. This pattern is consistent with the weaker, more stagnant airflow conditions found over the southeastern states in summer and their proximity to the center of semi-permanent high pressure systems. In the Northeast, ozone episodes are usually associated with backside flow around a stagnant high pressure system (e.g. Bermuda High).

Hydrogen Peroxide

Gas phase measurements for H_2O_2 at Whiteface and Whitetop are shown in Figures 5-18 and 5-19, (Appendix E) respectively. Despite the overall lack of variability in concentrations, mean values at Whiteface are highest for southwesterly trajectories, which is consistent with the pattern found for ozone. Hydrogen peroxide is a secondary product resulting from complex photochemical reactions initiated by ozone. The directional dependence at Whitetop is less clear, but peak H_2O_2 values there are associated with westerly trajectories.

Sulfur Dioxide

In summer, mean SO_2 concentrations are low, averaging under 3 ppb at all sites. As with the other gases, SO_2 values (Figures 5-20 to 5-23 - Appendix E) are higher with west to southwesterly trajectories for sites in the north. Southern sites exhibit higher concentrations for north to northwesterly flow. The differing dependence of gas concentrations on trajectory between northern and southern sites suggests a common air mass source region for relatively high concentrations--the Ohio Valley.

SECTION 6

METHODS AND PROTOCOLS

This section discusses field collection and laboratory analysis methods for cloud water chemistry, liquid water content, and supporting measurements such as meteorology, throughfall, and precipitation. Uncertainties are discussed with each method.

The MCCP collects cloud water only during the growing season, the onset of which varies depending on site location. At most sites, sampling is conducted for intensive campaigns of approximately three to six weeks, during which every cloudy period is sampled. At least three intensive sampling periods have been conducted during the growing seasons (approximately April-October) of 1986 through 1988 except for two sites that started during 1987.

Cloud water collection is begun as soon as possible after the start of a cloud event, which is defined by both visibility and volume. When a stationary object approximately 1 km from the collection site is consistently obscured from view for more than 15 minutes, a cloud event is defined to have started. Collection is discontinued when either the stationary object is no longer obscured from view, or less than 10 ml of sample has been collected over a 20 minute period. Liquid water content and aqueous phase chemistry are monitored during these cloud events. Gases, meteorology, precipitation chemistry, and throughfall chemistry are measured during cloudy and non-cloudy periods throughout each intensive sampling campaign.

CLOUD WATER COLLECTION

Mohnen and Kadlecek (1989) discussed the processes governing cloud collector efficiency and design. Passive cloud water collectors, such as the MCCP designated ASRC design (Falconer and Falconer, 1980), depend on ambient wind speed to impact droplets onto 0.4 mm wires strung between two circular disks. Active cloud water collectors have a blower or fan to provide the velocity difference between the droplet and the collecting string that is necessary for impaction. Factors that govern the overall efficiency for collector accumulation of liquid water include: impaction on the inertial collector surface (based on Langmuir stopping distance); accumulation/coalescence of individual droplets into a film or larger droplet; transport of the water from the collector surface to the storage container via gravitational, centrifugal, or pressure forces; evaporation of deposited cloud water from the collector surface; and resuspension (blow off) of water due to high wind speed at the collector surface. Various collector designs attempt to maximize efficiency for droplets above 2 to 5 μm diameter (a smaller cut-off size would allow collection of unactivated aerosol). However, the reduced collection efficiency below 5 μm diameter allows small droplets to escape collection.

During winter, supercooled cloud water freezes on impact with the collector strings. Collection efficiency is reduced due to the increased surface area of the strings covered by rime, so the accumulated rime is collected manually as often as possible to minimize this effect (Kadlecek et al., 1988). MCCP does not perform winter sampling.

Two types of cloud water collectors are used at MCCP sites, the ASRC passive collector and the Cal-Tech active string collector. Both have been found to collect water with equivalent chemistries over wind speeds ranging from about 3 to 30 m/s (Mohnen and Kadlecek, 1989). At most sites, the collectors are placed on the top platform of towers above the surrounding canopy. The only exceptions to this are Whiteface Mountain, where the collector is on the roof of the summit research lab, and Whitetop Mountain, where it is on a platform built over the research trailer.

Once a cloud event has begun, clean tubing and a collection bottle are attached to a collector and it is deployed. Collection bottles are changed hourly, and a portion of each hourly sample is analyzed in the field for pH. Five to 10 ml is preserved for hydrogen peroxide analysis. Each bottle is refrigerated until it is sent to a laboratory for chemical analysis. At the end of each event, the cloud collectors are detached and rinsed with deionized water until the conductivity of the rinse water is within % 10 μSiemens. Collectors are covered until the next cloud event. When more than 125 ml is collected at the end of one hour, the sample is split into two aliquots; one split is sent to the Central Analytical Lab (CAL) and the other to the site-associated analytical lab. Further details are provided in the MCCP Standard Operating Procedures Manual (1989); Quality Assurance procedures and results are described.

FREQUENCY OF CLOUD

For forest exposure and cloud water deposition estimates, it is necessary to understand when and where liquid water is present, at what mass concentration, and how the water is distributed among different droplet sizes. These data are referred to collectively as describing the presence of liquid water in the atmosphere.

The emphasis of the cloud frequency analyses conducted by the Mountain Cloud Chemistry Project (MCCP) is to estimate cloud impaction at mountain summits and the overall cloud distribution in the Appalachian Mountains. Because routine observations are scarce on mountains, other data sources were necessary to estimate long-term horizontal and vertical cloud distribution. This section describes the methodologies and associated uncertainties in determining regional and site-specific cloud frequency estimates. The data sources considered are National Weather Service (NWS) surface airport observations, US Air Force Real-Time Nephanalysis archives, and site-specific mountain cloud measurements conducted by the MCCP.

Regional Estimates

Airport observations have been analyzed to obtain cloud climatologies for the eastern USA (Warren et al., 1986). Surface airport observations consist of hourly measurements of cloud base height, coverage, and type. Cloud base heights have been measured since the mid 1920s using either a fixed- or rotating-beam ceilometer. This technique has an estimated accuracy of $\pm$ 30 up to 1000 m in elevation, and of $\pm$ 10% above 1000 m (WMO, 1976). Data are archived by the National Climatic Data Center for all reporting airport stations.

A limitation to airport observations is that few stations are located within mountainous areas. Airport-mountain comparisons have been investigated by Bailey and Markus (1987) and Imhoff and Malo (1989). A common finding is that airports are good indicators of mountain cloudiness for large-scale cloud systems but are poor indicators of small-scale cloud systems typically associated with orographic effects (e.g., cap cloud).

Bailey et al. (1989) have examined the temporal and spatial variations of low-level clouds (below 2000 m) in the eastern USA using the Real-Time Nephanalysis (RTNEPH), the global cloud archives produced by the US Air Force. An earlier version, called 3-Dimensional Nephanalysis (3DNEPH) was begun in 1971 (Fye, 1978) and replaced in 1984 by the improved RTNEPH version. Both versions use all conventional surface and rawinsonde data, pilot reports and satellite data to produce three-dimensional cloud information. RTNEPH data are organized according to a horizontal grid system superimposed upon a polar stereographic projection. Grid point spacing is 47.7 km at 60^{o} latitude where projection is true. Each grid point contains the following data for every 3-hour period: type of low, middle and high clouds, present weather, maximum cloud top, minimum cloud base, total percent

sky coverage, and percent coverage for 15 fixed layers. Gordon et al. (1984) have used 3DNEPH analysis of observed monthly mean cloud amounts to calculate radiative fluxes with a cloud model. Hughes and Henderson-Sellers (1985; 1986) have compiled a global cloud climatology for 1979. Schulz and Samson (1988) used 3DNEPH data to determine non-precipitating low cloud frequencies for central North America for 1982.

The uncertainties in the RTNEPH data archives consist of the limitations in each data source that was included. Limitations to the surface airport readings have been discussed previously. The satellite visual data processor cannot distinguish between clouds and bright areas of snow or ice. Therefore, unless accurate snow or ice data were available, the analyses may be in error. In addition, small cloud elements over high variability backgrounds (coastlines and mountainous areas) may not have been detected. The 47.7 km grid spacing precludes resolution at smaller space scales, and thus the RTNEPH analyses are best used for regional analysis. Low-level clouds, particularly for multi-layer cloud conditions, are best detected by surface and rawinsonde observations and least by satellites. In areas where airport station density is low, low-cloud information may not be well represented.

Site-specific Methods

Three different techniques for estimating the frequency of cloud impaction have been used by the MCCP since the 1986 field season. From the project's onset, relative humidity has been used at most sites to estimate periods of cloud presence in lieu of an affordable direct measurement technique. The Whitetop site has always used a reflectometer to determine the presence of cloud, but its cost (in excess of $10,000) precluded its use throughout the MCCP network. Beginning with the 1988 field season, the new Mallant optical cloud detector was installed at most sites. This section describes the three techniques while the next section inter-compares the techniques. A fourth technique (Krovetz et al., 1988) has also been used on an experimental basis, particularly at the Shenandoah site. This technique will not be discussed here because it was not used to derive official presence of cloud estimates.

Relative humidity (RH) is sensed by a Rotronics MP-100 combination temperature/RH probe housed in a naturally aspirated radiation shield. Cloud presence is determined subjectively by analyzing the time series trace during saturation and near-saturation conditions. The tendency for the sensor's response in these conditions to drift upward with time has precluded the use of a wholly objective RH cloud threshold (such as a minimum RH value) to define cloud events. Average RH values are recorded hourly based on 5-second samples. As verified by field observations, the onset of cloud is typically marked by a sharp rise in RH to near 100% followed by a leveling off, and cloud dissipation is marked by an abrupt drop of several percentage points.

The uncertainties inherent in the RH technique are several: the element of subjective interpretation, the sensor's 5% accuracy specification, the use of hourly averaged values, and its slow response time relative to the two optical techniques. Overall, it is suspected that this technique may somewhat overestimate the frequency of cloud due to the fact that near-saturation conditions can occur in the absence of clouds such as during heavy or prolonged rain events.

A backscatter reflectometer (visibility sensor Weathertronics Model 8340) has been used by TVA since 1986 as a cloud detector. According to Valente et al. (1989), "observations at Whitetop have shown that by using a signal of 0.15 (5% of the output range) as the definition of presence of cloud, haze and liquid precipitation are not mistaken for cloud impaction." The instrument has a visibility range of 1000+m to 10 m, corresponding to a signal output of 0 to 100% of scale, respectively. The detection threshold of 5% of scale corresponds to a visibility of about 260 m according to the manufacturer's literature. This instrument has not been used at MCCP sites other than Whitetop.

The uncertainties with the reflectometer are not well defined. However, the method used to obtain hourly signal values will tend to overemphasize the presence of cloud within an hour in which cloud was present less than half the time. This occurs because the signal's scale is logarithmic whereas hourly averages are derived arithmetically (private communication, TVA). Hourly average reflectometer values are based on 5-minute intermediate average values derived from 20-second samples. Given that the first 5% of scale (cloud absent) corresponds to a visibility range of about 740 m while the remaining 95% of scale (cloud present) corresponds to about a 250 m visibility range, arithmetic averaging of percent of scale values within an hour will give greater weight to "cloud present" signals. This can be a source of discrepancy when comparing different measurement techniques during variably cloudy hours.

The Whitetop site has also used a time-lapse video recording system to estimate cloud base elevation. The system is located about 4.5 km northwest of the mountain at an elevation about 760m less than the site elevation of 1689 m. The location provides a clear view of the mountaintop from one direction and is limited to daytime observations. Cloud base heights are derived hourly through manual interpretation of the video record.

The Mallant Optical Cloud Detector is a forward-scattering optical device originally developed by the Energy Research Foundation in The Netherlands (Mallant and Kos, 1989). Following successful laboratory and field tests (Valente et al., 1989), a modified design was built by Associated Weather Services and deployed at all summit sites in 1988. It has an adjustable detection threshold corresponding to liquid water content at an assumed mass median droplet diameter. Hourly recordings indicate the percentage of time with cloud present based on 5-second samples. Each sample produces a binary signal corresponding to cloud presence (=1) and absence (=0).

The Mallant instrument's detection threshold is set at an equivalent LWC value of approximately 0.04 g/m3 for a droplet mass median diameter of about 11 microns. This is roughly equivalent to a visibility of 350 m (Atlas and Bartnoff, 1953). The uncertainty in this threshold is on the order of $\pm$0.02 g/m3 due to the uncertainties in the FSSP-based laboratory calibrations and on the imprecision of the adjustment potentiometers. Under ambient field conditions, the actual detection threshold will also be a function of the cloud's drop size distribution. Therefore, this instrument's response to cloud, especially "thin" clouds, is variable. It has also been observed that this instrument can be susceptible to droplet accumulation on the optical lenses which results in a reduction in the sensitivity to the presence of cloud. The detector does not contain heaters and therefore is suitable only for above-freezing cloud conditions.

Comparison of Methods

This section presents results of several intercomparison tests between the three primary techniques used to estimate presence of cloud.

Mallant vs. Reflectometer--

Several intercomparisons of the Mallant cloud detector and the TVA reflectometer have taken place, beginning with the 1987 liquid water content instrument "shoot-out" at Whitetop Mountain (Valente et al., 1989). This first intercomparison used an original Mallant prototype whereas all later intercomparisons used the modified design. In all intercomparisons the TVA reflectometer was designated an arbitrary "standard" to define periods when cloud was present or absent. The first intercomparison, covering a two-week period in 1987, found that the Mallant prototype had 98% agreement with the reflectometer during cloud events and 93% agreement during non-cloud events.

Four other intercomparisons were conducted during 1988 and 1989 using four different Mallant instruments. These tests comprised over 5700 field hours. The Mallant design agreed with 96% to 99%

of the cloud/no cloud observations taken by the reflectometer. When clouds were indicated by the reflectometer, the Mallants agreed 92% to 95% of the time for three of the instruments, and 80% for the other. The lower agreement of this one instrument was the result of it not responding to cloud for a 24-hour period within one extended cloud event. It is suspected that water droplets accumulated on its lenses, reducing the instrument's sensitivity. When the reflectometer indicated that clouds were absent, agreement with all four Mallants ranged from 98% to 100%.

Relative Humidity vs. Mallant--

Concurrent estimates of hourly cloud presence using the RH and Mallant techniques were made throughout the 1988 field season at four MCCP summit sites: Mitchell, Moosilauke, Shenandoah and Whiteface. A minimum of 2300 hours of simultaneous values were taken at each site. Based on its favorable intercomparison results with the TVA reflectometer, the Mallant detector was designated the standard in the intercomparisons with RH. An hour with cloud as defined by the Mallant required the instrument to be detecting cloud for at least 50% of the hour. Overall agreement between techniques on an hourly basis ranged from 87% to 96%. During cloud conditions as defined by the Mallant, agreement ranged from 81% to 94%. During no-cloud conditions, they were in agreement 88% to 96% of the time.

RH vs. Mallant and Reflectometer--

The early 1989 field season was the first opportunity for the standard MCCP RH sensor to operate together with the Mallant and reflectometer instruments at Whitetop Mountain. Based on over 1100 hours of operation, RH agreed with the reflectometer 91% of the time, and 90% with the Mallant. During cloudy conditions the agreement was 86% and 85%, respectively. During non-cloudy conditions, the agreement was 94% and 91%, respectively.

LIQUID WATER CONTENT

The MCCP initiated network-wide measurements of cloud liquid water content (LWC) in 1987 using the TVA-Valente filter collection method. This section presents the results of a review of LWC and drop size measurement methods and a field comparison of several LWC instruments. Most LWC measurement instruments have been designed for aircraft operation where the speed of the aircraft is an aid to operation. Ground-based instruments are typically modified versions of aircraft systems, and can be classified according to their operating principle as thermal, optical, or inertial impaction methods.

The Johnson-Williams liquid water instrument consists of a heated wire that is cooled by the evaporation of liquid water droplets. An instrument time constant of approximately 1-2 sec may cause underestimation of water contents (Spyer-Duran, 1968). A further limitation is a limited response to droplets larger than 30 μm. The instrument must be calibrated in a cloud tunnel to achieve the expected accuracy.

The NHRL Nimbiometer and the CSIRO (King) Probe are heated-wire instruments. Improvements were made by Merceret and Shricker (1975) by operating the instrument at a constant temperature below 100°C where the power dissipated is proportional to the square of the liquid water content. King et al. (1978) improved the instrument by increasing the cooling by liquid water relative to cooling by dry air. Measurements agree with other instruments to about 5% at LWC of 2 g/m^3. The advantage of this design is that wet calibration is not required. However, at low air speeds typical of ground-based systems, the errors are proportional to $v^{-1/2}$ (King et al., 1978), making it necessary to vary the aspiration speed with wind speed. Particle Measuring Systems, Inc., has developed a King Probe prototype to

measure LWC of up to 1 g/m^3. The probe is oriented using a wind vane. A field test at Whitetop Mountain, VA, indicated that further development and testing are needed because of short circuits during wet, cloudy periods, sensitivity to radio frequency interference, and rain droplet contamination of the inlet (at wind speeds > 3 m/s).

Chylek (1978) investigated an optical technique that uses the extinction of radiation of a wavelength of 11 μm and found that infrared extinction should be directly proportional to liquid water content. Several attempts to use this technique in cloud and fog have been encouraging (Gertler and Steele, 1980; Jiusto and Lala, 1982), but practical problems have limited its full use. Comparison of a laser transmissometer (Jiusto and Lala, 1982) with a droplet spectrometer (FSSP-100) revealed good agreement up to water contents of about 1 g/m^3. The presence of many large droplets (> 28 μm) degrades Chylek's (1978) approximation and contributes to forward scatter errors. At LWC less than 0.5 g/m^3, the transmissometer system was within 10% of filter measurements and 35% of FSSP-100 values. This promising technique is still in the development stage.

Another instrument estimates LWC by measuring scattered intensity in the near forward direction (0.25 to 5^o). Blyth et al. (1984) designed an instrument with a 26 cm path with discrete detectors at five different angles from the scattering axis. Based on theoretical calculations, the signals are weighted to provide a signal proportional to LWC. Comparison with drop spectrometers and infrared transmissometers show excellent agreement. This instrument still requires development to construct a field unit suitable for routine observations.

Gerber (1984) proposed a similar instrument based on the principle that light scattered by the droplets in the near-forward direction is strongly correlated with infrared extinction, and is thus also proportional to LWC according to the Chylek (1978) relationship. Gerber Scientific offers the Particle Volume Monitor (PVM), capable of measuring in situ, and in real time, the integrated volume of particulates suspended in the atmosphere with a stated precision of 0.002 g/m^3. A narrow beam from a 780 nm laser diode irradiates the droplets in the open air along a 40 cm path. Calibration results indicate % 10% agreement with an infrared transmissometer in controlled fogs (Gerber, 1988).

A prototype instrument developed by Scott McLaren of ASRC SUNY-Albany, NY, uses a chilled mirror dewpoint sensor to measure the dewpoint of two differently treated streams of sampled air, and switches streams at five minute intervals. The first stream contains air that has been heated to evaporate the cloud droplets into the air stream, and the second stream is filtered to remove the droplets. By measuring the small difference in dewpoint between these air streams, the LWC can be calculated from psychrometric principles, provided that significant changes in LWC do not occur between the two adjacent averaging periods.

Filters and impactors are the oldest method of measuring liquid water content. Examples are reported by Houghton and Radford (1938) and Wattle et al. (1984). The operating principle is simply to draw a large volume of cloudy air through a filter and then weigh the accumulated water. Using large air flows and large collection areas minimizes the size selection collection problems. With light wind and droplets below precipitation size, this technique comes close to providing an absolute measurement of LWC. One limitation is that during periods with less than 100% relative humidity, evaporation may result in an underestimate of LWC. The method requires constant attention and averages over the one-hour sample period.

Impaction techniques are similar to filtering, but they emphasize droplet sizing instead of mass concentration. A glass slide with a gelatin coating (Jiusto, 1965) or another material sensitive to water drops is used for collection. With gelatin, droplets leave impressions that are twice the diameter of the impinging droplets. The slides are analyzed with an optical microscope to determine drop size distribution and water content. This technique may discriminate against either the large or small end of the

spectrum, depending on the collection method. Baumgardner (1983) estimated a 32% error in measurement.

Optical particle spectrometers measure the size distribution of particles by light scattering. Size is determined by a calibration of the light pulse amplitude from particles of known size. LWC is calculated by integrating the droplet volumes from the measured droplet spectrum and dividing the total mass of liquid water by the sampled air volume. Different optical configurations compensate for the size dependence of the scattering efficiency. Virtually all designs exhibit some uncertainty at sizes of the order of the wavelength of the light source. Perhaps the greatest obstacle to using these instruments in cloud is that the aerosol configuration discriminates against large particles such as cloud droplets (Davies, 1968).

Particle Measuring Systems, Inc., developed the Forward Scatter Spectrometer Probe (FSSP-100) with a unique sample inlet configuration. The laser light source is focused to a 100 μm diameter; scattered light is split to separate the light scattered by particles in the center of the beam from the signal, making it possible to avoid the aerodynamic focusing used in other systems. The signals from particles that pass through edges of the sample volume are eliminated by measuring the time it takes for a particle to cross the beam and comparing this with a running average. This design alleviates inlet problems, but the use of forward scattering with small collection angles has resulted in a scattering response with several multi-valued regions (Pinnick et al., 1981), especially between 0.5 and 2 μm and near 8 μm diameter. FSSP measurement uncertainty under the best conditions is about 34%, and an uncorrected system could have errors from 54% to 105% (Dye and Baumgardner, 1984; Baumgardner, 1983). FSSP-100 sampling efficiency remains open; in low wind conditions the sample should be representative. The probe should be aligned into the wind and, in gusty conditions, correction to the sampling rate may be necessary. To compensate for variable winds, modifications that continuously adjust the fan speed are available.

The TVA-Valente instrument (Valente, 1988) used by the MCCP collects droplets by filtration from a metered air volume in an Ertalyte plastic cartridge filled with eight layers of a high collection efficiency polypropylene mesh. Inlet velocity is matched to the mean wind velocity to minimize non-isokinetic sampling errors. A rain shield minimizes collection of precipitation-size droplets; however, significant precipitation-induced errors are likely at wind speeds above 10 m/s.

Liquid water content (LWC) measurement in the MCCP follows the same collection schedule as that described for cloud water. The LWC collector is deployed near the cloud water collector at the start of an event. The collector cartridge is weighed at the beginning and end of an hour and the hourly LWC is calculated by dividing the weight gain by the sampled air volume. The inlet velocity is adjusted to match the mean wind velocity to minimize anisokinetic sampling errors. A rain shield is used to minimize collection of precipitation-sized droplets. The sampler must also be pointed onto wind and an inlet size is selected depending on wind speed. Detailed operational procedures can be found in the MCCP Standard Operation Procedures (1989).

During 1988, MCCP used optical cloud detectors to record the hours with patchy clouds. The collection mesh and range of inlet velocities were designed to collect cloud droplets from 3 to 200 μm in diameter with greater than 95% efficiency. A rain shield excludes rain (droplet radius > 500 μm) and drizzle (200-500 μm radius) falling at an angle of greater than 15^{o} from horizontal. However, during high winds combined with precipitation and cloud, some precipitation will be sampled. LWC measurements of clouds by the MCCP therefore differentiate between precipitating and non-precipitating events because the technique may overestimate LWC during precipitating events.

Droplet Size Distribution from LWC

Cloud droplet size distributions (or size spectra) are measured using impactors or a forward scattering spectrometer probe (FSSP) that measures the degree of scattering of a narrow laser beam as particles pass through the beam (Knollenberg, 1981). The amount of scattering depends on particle size. The instrument can detect droplet diameters from < 1 μm to 100 μm.

Because median droplet size is important for cloud droplet interception processes but is not routinely monitored by MCCP, it is sometimes estimated from measured liquid water content (LWC). MCCP scientists have investigated the relationship between the median droplet volume diameter (dn) and LWC for stratus and stratocumulus clouds. The parameter dn is defined as the droplet diameter that divides the total cloud liquid water into equal parts by volume (Best, 1951). Stratus and stratocumulus clouds were examined because of their similarity to clouds observed from mountaintop forests. Seventeen data pairs, representing 96 measured droplet size distributions, were used to determine the correlation between dn and LWC. One-half of the variance in dn was explained by variance in LWC, suggesting that LWC and droplet size distribution are related for mountaintop clouds but that considerable size spectra variability is associated with other factors.

LWC from Cloud Sampler Collection rate

An additional size spectra feature having implications for cloud deposition modeling is the large variation in the size spectrum for a given value of LWC. Large fluctuations in droplet number concentration and diameter imply that, despite a general tendency for the size spectrum to vary with LWC, a large amount of noise exists in the data. If droplet-leaf capture efficiency is non-linearly dependent on droplet size, then the large variability in the size spectrum will result in a biasing of cloud deposition estimates.

A data analysis technique for estimating LWC has been examined at several MCCP sites. The collection of cloud water (at a volume collection rate R_c) by a passive ASRC string collector such as used by Falconer and Falconer (1980) is known to depend on the dimensions of the strings, cloud droplet diameter, wind speed (M), and LWC. A given droplet size distribution has a characteristic diameter that is presumed to be representative of the droplet size spectra. This diameter is not known. Assuming a constant linear relationship, several investigators have used regression to determine an empirical relationship between LWC and R_c/M (e.g., Saxena et al., 1989).

At Shenandoah, Krovetz et al. (1989) compared LWC (gravimetric measurement) from the ratio $V^{1.8}/S^{1.5}$, where V is the measured collector cloud water volume and S is the Stokes number (Fuchs, 1964). S was computed using the known string diameter, measured wind speed, and a constant droplet diameter. They reported a correlation ($r^2 = 0.77$) between 11 pairs of measured and estimated LWC values.

These regressions assumed that the effect of droplet size on droplet collection efficiency is constant. However, measured droplet spectra have been found to be highly variable over short time intervals, even when liquid water content was relatively constant (Mueller and Imhof, 1989). Therefore, estimated median droplet size from LWC data or LWC from a passive sampler collection rate are considered to be a crude approximation of the distribution or concentration of liquid water in the atmosphere.

GASES

Gaseous measurements in MCCP are made using a combination of standardized and research techniques. The ultraviolet (UV) photometric method for ozone and the UV fluorescence method for sulfur dioxide are the most well-defined gas measurements, with traceable quality assurance standards. Measurement methods for nitric acid, sulfur dioxide, and hydrogen peroxide lack traceable quality assurance standards, but have been involved in inter-comparison, laboratory, and field studies. Table 6- indicates the measurements that are made at each MCCP site.

Ozone

Ozone measurements at MCCP sites are made with the UV photometric technique described by Bowman and Horak (1972), which is equivalent to the US EPA's reference method (Federal Register, 1980). The TECO Model 49 used in the MCCP has a minimum detectable limit and precision of 2 ppb. Ambient ozone values are 40 to 50 ppb at MCCP sites.

TABLE 6-1
Gas Measurements at MCCP Sites

Site	Gas Measurement
Howland, ME	Ozone, sulfur dioxide, nitrogen oxides
Mitchell, NC	Ozone, sulfur dioxide, nitrogen oxides
Moosilauke, NH	Ozone
Shenandoah, VA	Ozone, sulfur dioxide, nitrogen oxides
Whiteface, NY	Ozone, sulfur dioxide, nitrogen oxides, hydrogen peroxide
Whitetop, VA	Ozone, sulfur dioxide, nitrogen oxides, hydrogen peroxide

Sulfur Dioxide

Sulfur dioxide (SO_2) is measured using continuous and integrated methods. Continuous detection of the characteristic fluorescence by SO_2 when irradiated by ultraviolet light was described by Okabe (1973). These monitors have been designated as equivalent to the US EPA's reference method (Federal Register, 1979). The TECO model 43 used for SO_2 has a minimum detectable limit of 2 ppb with a precision of 5 ppb. The maximum SO_2 concentrations at MCCP sites are in the 20 ppb range with a mean of approximately 2 ppb. This instrumentation thus provides a good estimate of the upper limits of SO_2 at MCCP sites, while lower SO_2 limits are not well defined.

Integrated measurements of SO_2 are made using the filter pack or the annular denuder system. The filterpack system has been characterized by Anlauf (1986) and Sickles (1987). In the filterpack unit, SO_2 is collected on a Whatman 41 filter impregnated with sodium carbonate in glycerol (Huygen, 1963). The minimum detectable limit for SO_2 using the filterpack system is estimated to be 0.5 ppb. Factors influencing the sensitivity of the method include sampling time, analytical sensitivity, and sampling environment. A comparison of the filterpack method and a TECO SO_2 monitor indicated agreement over a range of ambient concentrations (Anlauf, 1986).

The annular denuder system described by Possanzini (1983) is a multi-constituent sampler than can be used to measure a number of atmospheric gases: SO_2, HNO_3, NH_3, and HONO. Diffusion denuders have been used by several investigators to separate gases and aerosols, to collect gases for analysis, or

as preseparators (Forrest, 1984; Shaw, 1982; Ferm, 1972). The overall sensitivity of the technique is comparable to the filterpack method for SO_2.

Nitric Acid

Nitric acid measurements in MCCP are made using either the filterpack or the annular denuder system (ADS). Because of possible biases in HNO_3^- and NO_3^- sampling with the filterpack, a total inorganic nitrate value is obtained from ion chromatographic analysis of the Teflon and nylon filters. Using total inorganic nitrate as a measure of nitric acid could result in overestimation of nitric acid. Nitric acid and nitrous acid are collected on a sodium carbonate coated annular denuder in the ADS system. For long sampling times (greater than 48 hrs), the nitrous acid can be oxidized to nitrate on denuder surface; this gives an overestimation of the nitric acid due to the potential biases listed above.

Hydrogen Peroxide

Gaseous hydrogen peroxide is measured with an automated monitor described by Lazrus (1986). This technique reacts peroxide with p-hydroxyphenylacetic acid in the presence of peroxidase. The minimum detectable limit is estimated to be 0.05 ppb with a precision of 0.01 ppb. Hydrogen peroxide levels range from 0.1 to 3.0 ppb at the MCCP sites. When the monitor was compared with laboratory-generated standards, the two agreed within 10% across a concentration range of 0.06 to 128 ppb (Kleindienst, 1988).

METEOROLOGICAL MEASUREMENTS

The MCCP meteorological measurement program was designed to complement the cloud water collection and analysis as well as to define the climate of high-elevation Appalachian Mountain forests. Each MCCP site measures wind speed, wind direction, temperature, relative humidity, solar radiation, barometric pressure, and precipitation. Beginning with the 1988 field season, cloud presence was measured with an optical cloud detector. All MCCP sites except Whitetop use the same package of sensors and follow the protocols of measurement established by the MCCP Quality Assurance (QA) Plan (1988) and detailed in the MCCP's Standard Operating Procedures (1989) and the Meteorological Site Technician's Handbook. The Whitetop site follows a measurement protocol that is similar but not identical to the MCCP QA Plan.

Wind speed and wind direction are measured with the R.M. Young Wind Monitor (Model # 05103). Wind speed is sensed by a helicoid propeller that rotates a six-pole magnet that in turn generates an AC sine wave with a frequency proportional to wind speed. Wind direction is sensed by a thermoformed plastic vane that transmits its angular position via a coupling to a 10 K ohm potentiometer. The sensor is calibrated at the start and end of each field season, and is subjected to at least one Quality Control (QC) check during the season. Most sites perform the QC check monthly. Wind speed is checked via an 1800 rpm motor and a torque disk for starting threshold, and wind direction is checked versus reference directions.

Temperature and relative humidity are measured with the Rotronic Instrument Model MP-100 combination sensor. A capacitive element senses relative humidity and a resistive device (RTD) senses temperature. Both transducers are housed inside a Gortex cap and wired to a PC board that outputs voltages corresponding to the ambient conditions. The sensor probe is housed in a naturally-aspirated radiation shield. The sensor is calibrated at the start and end of each field season versus a collocated reference. During the field season, weekly QC checks are performed using a psychron.

Solar radiation is measured with a Li-Cor pyranometer (Model # L1200S). The sensor uses a silicon photodiode to measure solar radiation on a horizontal surface. The sensor is calibrated at the start and end of each field season versus a collocated reference.

Barometric pressure is measured with a Met One, Inc. barometer (Model # 090-B). A piezoresistive diaphragm senses the pressure, and its signal is converted by PC circuitry to a voltage. The sensor is calibrated at the start and end of each field season against a collocated reference.

A Weathertronics tipping bucket gauge (Model # 6021-B) is used to measure precipitation. Each 0.1 mm of precipitation results in a tip of the bucket. At the start and end of each field season the sensor is calibrated by applying a known volume of water at a standard rate to the sensor. During the field season, monthly QC checks are performed in the same manner. This parameter has not been measured at the Whiteface Mountain peak since the 1988 field season due to high winds.

Meteorological sensors were installed to best represent weather conditions in the vicinity of the cloud water collector without interfering with the collection process. Most sites locate their cloud water collector on towers or roofs above the local forest canopy top. Thus, most meteorological sensors are similarly exposed adjacent to the cloud water collectors. Precipitation and barometric pressure, however, are measured near the surface. Table 6-2 lists each site's sensor mounting location and height above the local canopy.

The purpose of the meteorological measurement program is to define the weather conditions at each site's cloud water collector station. Therefore, these measurements are site-specific and are not necessarily representative of weather conditions elsewhere. This is especially true for wind measurements, which are strongly influenced by local topography and surface roughness. In addition, the meteorological monitoring program is designed for growing season (April-October) only.

TABLE 6-2

MCCP Site Sensor Mounting Locations and Height Above Canopy

		Height Above Canopy (m)				
Site	Mount Type[a]	Wind Speed/Dir.	Temp/RH	Pressure	Precip.	Cloud Freq.
Howland[b]	R	9.1	9.1	2.0	2.0	N/A
Moosilauke	T	9.0	6.0	-10.0	- 8.0	4.1
Whiteface[b]	R	17.0	16.0	9.0	3.0[c]	15.0
Shenandoah-1	T	5.5	3.6	-10.0	-11.0	1.7
Shenandoah-2	T	8.2	3.7	-16.5	-16.5	N/A
Shenandoah-3	T	8.2	3.7	-21.9	-21.9	N/A
Whitetop	R	2.0	0.0	- 4.5	- 3.0	0.0
Mitchell-1	T	11.9	7.3	- 7.3	- 5.9	5.4
Mitchell-2	T	9.0	6.0	-18.0	-18.0	N/A
Mitchell-3	T	9.4	4.9	-13.2	-15.2	N/A

[a] T - from tower
R - above roof platform
[b] heights are relative to ground level
[c] not measured since 1988

THROUGHFALL

Weekly integrated collections of throughfall and bulk or wet-only precipitation were made during the summer of 1988 at some MCCP sites to compare throughfall inputs to the soil with what would have been deposited had only rain occurred. Ten 16 cm diameter funnels were deployed randomly along the cardinal directions from the center of a 0.1 hectare circular plot. Each collector was mounted approximately 1.5 m above ground level. A nylon wool plug inserted in the neck of the funnels and replaced weekly prevented large material from falling into the collection bottle attached to the funnel. Weekly throughfall data were obtained from a proportional composite of at least 10 collectors, and the precipitation data were from a single collector, either wet-only or bulk. To minimize chemical changes between collection and analysis, about 1 ml of chloroform was added to each bottle at the beginning of the week (occasionally preservative evaporation occurs).

A bulk precipitation (BP) collector identical to a throughfall collector is deployed in an open area. The collection schedule is identical to that of throughfall. The BP is weighed in the field and a 250 ml aliquot is reserved for lab analysis. The wet-only precipitation collector is located adjacent to the BP collector. An Aerochem Metrics collector is used for wet-only (WO) collection and standard NADP procedures for buckets are followed. Collection frequency and sample handling are identical to that for throughfall and bulk precipitation. Laboratory analysis methods and procedures for TF, BP, and WO are identical to those for cloud water analysis.

The measurement of throughfall has involved a variety of collectors, usually insufficient in number given the spatial variability in deposition. Buckets, funnels, rain gauges, and troughs of various sizes have been used (Reigner, 1964; Hamilton and Rowe, 1949; Goodell, 1952). The uniformity of terrain and canopy dictate the effective collection area needed to adequately estimate throughfall to a canopy location. The intention is typically to sample randomly, collecting from canopy features according to their relative densities. For example, Helvey and Patric (1965) determined the number of gauges necessary to sample throughfall to a 5% error depending on rain amount (see Table 6-3).

TABLE 6-3

Mean Number of Throughfall Gauges and Collection Area Needed for 5% Error Under a Uniform Canopy (from Helvey and Patric, 1965).

Throughfall (cm)	# of Gauges	Total Collection Area in m^2
< 0.5	46	0.25
0.5-1.0	18	0.06
1/0-1.5	14	0.04
> 1.5	13	0.04

In a more complex and higher elevation canopy typical of the conditions on Whiteface Mountain, the throughfall variability is larger, requiring more collection area to obtain a 90% confidence level for a 10% coefficient of variation (Kadlecek, 1989), as shown in Table 6-4. At the upper site, 17 buckets (28 cm diameter) were used, and at the lower site 15 buckets were used. The ratio of highest to lowest collected volume during a single event ranged from about 2 to 8, with the higher factor applying primarily to low-volume events. Collectors with larger surface areas, such as troughs, give data with a smaller range, and those with smaller collection areas produce a larger range. The principles are the same for any canopy; the actual values depend on the canopy properties.

TABLE 6-4

Throughfall Bucket Collection Area Required for 10% Relative Standard Deviation in a Complex Canopy on Whiteface Mountain, NY (from Kadlecek, 1989).

	Collection Area in m^2	
Precipitation (cm)	1200 m elevation balsam fir	600 m elevation beech, birch, maple
< 0.25	2.9	8.8
0.25-0.76	2.4	3.3
0.76-1.52	3.0	0.8
> 1.52	1.3	0.6

Note: These averages were obtained from 8 events at the upper site and 13 events at the lower site.

Similar data from the 1000 m elevation (spruce, fir, beech) based on a regular 25-bucket grid (bucket diameter of 28 cm, bucket spacing of 5 m) showed that had only three collectors been used, there was a 90% probability that the three-bucket average would have differed from the 25-bucket average by more than 30%. This is not to say that 25 collectors were enough, only that too few collectors can give biased results because the canopy directs the flow unevenly and, in the case of cloud water, exposed features are the preferred deposition sites. Longer sampling times tend to average out some of these differences because, as the wind vector changes during events, several segments of the canopy are sampled. Sampling at the sub-event level is expected to require more collectors for the same uncertainty discussed here.

Reynolds and Leyton (1963) recommend 20 rectangular troughs with a total collection area of about 10 m^2 to obtain a standard error of 5% to 10%. Eidmann (1959) used trough collectors with an opening of about 1 m^2. One method of calculating how many collectors are necessary is to first determine by experiment the variation in collection over the study area; then, the number of collectors would be equal to the squared ratio of the standard deviations to the desired standard error of the mean (Kittredge, 1948; Helvey and Patric, 1965).

CLOUD INTERCEPTION

Particles greater than a few μm in diameter (including cloud droplets) are deposited onto surface by impaction and sedimentation. Impaction is primarily due to the inability of particles in the airstream to follow rapid changes in air flow; their inertia carries them into surfaces protruding into the airflow. Sedimentation is the result of the acceleration of particles downward under the influence of gravity. After a short time, aerodynamic drag balances gravity for small particles, and the particles fall at a constant velocity. A detailed description of the deposition process is beyond the scope of this section: the interested reader is directed to Fuchs (1964), Chamberlain (1975), Bache (1979a), and Waldman and Hoffmann (1987) for further information.

The deposition process for cloudwater is a function of the free stream velocity of the droplets, the concentration of droplets (liquid water content of the cloud), the droplet size distribution, and the size, shape and distribution of elements protruding into the flow. While there are several empirical descriptions of particle deposition to vegetative canopies (e.g., Bache, 1979b; Thorne et al., 1982; Grant, 1983), the actual deposition process is not well understood. One attempt at relating impaction efficiency (deposition) to these basic physical parameters is (Bache, 1979a):

$$E.I. = St^2/(St + 0.6)^2 \qquad (1)$$

where:

E.I. = efficiency of impaction
St = Stokes number, defined as:

$$St = (Dp - Da) \cdot u \cdot c \cdot dp^2/9 \cdot v \cdot dc \qquad (2)$$

where:

Dp = density of the particles
Da = density of air
u = velocity
c = Cunningham slip correction factor
dp = diameter of the particles
v = dynamic viscosity
dc = characteristic length scale of the collecting surface

Stokes calculations have previously been applied to deposition processes by Davidson and Friedlander (1978). Unfortunately, there are few field data with which to evaluate these relationships. Turbulent transport over and among non-homogeneous surfaces, the driving mechanism for cloudwater deposition, requires more research before the process will be fully understood and viable relationships identified.

Shuttleworth (1977) described a simple, steady-state computational model of fog/cloudwater deposition to, and evaporation from, a uniform vegetation canopy. The model is based on the analogy of electrical resistance in a direct-current circuit similar to the "big leaf" concept first defined by Monteith (1965) and subsequently applied to deposition by Wesely and Hicks (1977), Unsworth (1981), and Hicks et al. (1987), among others.

Lovett (1984) adapted the Shuttleworth model to a balsam fir forest canopy on Mt. Moosilauke, NH. Lovett included the effects of vertical variation in canopy structure through the construction of a multiple-layer model in which the vertical turbulent transport of droplets is controlled by the

aerodynamic resistances between model layers and between the top layer and the air above the canopy. The droplet size distribution in the model is based on a distribution function described by Best (1951), with all droplet diameters occurring in the range of 0 to 30 m. While research carried out in forests has raised questions about the applicability of aerodynamic theory for fluxes over forests (e.g., Stewart and Thom, 1973; Thom et al., 1975; Knoerr and Mowry, 1977), the model has been applied to estimate the flux of both water and ions in cloudwater to forests (Lovett and Reiners, 1986).

The Lovett model forms the basis of two cloud deposition models used to estimate the contribution of cloudwater deposition to the total deposition budget at the MCCP sites (Mueller 1989; Krovetz et al., 1989). Modifications to the model have been made for the Whitetop Mountain site and the Shenandoah site, and manuscripts describing this work are currently in review. The modeling work at Whitetop has been directed towards spruce forests, while the work at Shenandoah has been directed towards deciduous forests.

The canopy structure portion of the model had to be generalized to enable computations at locations other than the original Mt. Moosilauke study site. The spruce version of the model (CDM-S) allows the input of projected (silhouette) leaf area index which is used to calculate the full-sided leaf area index. Unless other input data are provided, the spruce model uses the vertical profile of total surface area index (sum of leaf area index and total non-leaf surface area index) and the distribution of surface area by canopy component type determined by Lovett for his balsam fir canopy. Canopies shorter than the Moosilauke canopy studied by Lovett (10.6 m) are assumed to have the full crown of the Moosilauke canopy with shortened boles. Conversely, the boles are lengthened for canopies of more than 10.6 m in height, producing a smaller crown-to-total height ratio (a modification consistent with Whittaker et al., 1974).

The spruce model also allows modifications to both the wind speed profiles and the eddy diffusivity profiles within the canopy. These modifications are not based on experimental data, but they can be used for computational comparisons of model output. The spruce model also allows the user to select the cloud droplet size distribution relevant to the cloud type being modeled if the original distribution (Best, 1951), is considered inappropriate, and to input a measured cloud liquid water content if available.

The deciduous version of this model (CDM-D), developed at the Shenandoah site, is a hybrid of the original Lovett (1984) model, the model described by Bache (1979b), and recent research on deciduous forest canopies. The distribution of both leaf and twig/branch surface area are determined using the grid technique of Aber (1979). The deciduous model uses the Best (1951) droplet size distribution with additional droplet size categories and a mean droplet diameter of 20 m. Liquid water content can either be entered directly or calculated from an empirical formula relating capture efficiency of the cloud water collector to liquid water content. Droplet capture efficiency was changed to correspond to the morphology of deciduous leaves; it is computed for planar objects having characteristic sizes determined from field sampling of fallen leaves (see Bache, 1979a). Wind speed profiles above and through the canopy are based on work in a deciduous forests by Sigmon et al. (1984), in complex terrain by Mowry (1980), and basic micrometeorological theory. Exponential decay of wind speed is used above the canopy, while linear decay is used within the canopy.

The differences in structure between the spruce and deciduous versions of the model result in different outputs for identical meteorological conditions. While differences in deposition between the two types of canopies would be expected from theories concerning the capture efficiency of leaves versus needles (as well as the morphology of coniferous vs. deciduous canopies), separating actual differences in deposition from model errors is difficult. No comparison of the models has yet been performed, and very few validation data are available. A priority for future work is to validate and compare the models to identify differences in deposition between the canopy types.

Cloud Deposition Model Application

Lovett tested his original model against a limited amount of data collected on Mt. Moosilauke, NH. The CDM-S has undergone limited testing at Whitetop Mountain, VA; for six cloud events during 1987, mean water flux (0.33 mm/hr) appeared to agree with the mean measured throughfall rate but only 36 percent of the variance was described. Another model test in a new spruce stand resulted in model-predicted water flux of 0.4 mm/hr compared to a measured throughfall rate of 0.3 mm/hr. Cloudwater collection rate explained 84 percent of the variance in throughfall rate. These results are not conclusive because of uncertainties in the inputs (such as forest stand characteristics, liquid water content, and windspeed) (Mueller and Imhoff, 1989). Due to limited testing, the uncertainties in cloud droplet interception modeling are unknown at this time.

The response of the original Lovett cloudwater deposition model to varying meteorological and canopy structure inputs was summarized by Lovett (1984) and Lovett and Reiners (1986). Some key results and sensitivities are:

- Computed cloudwater flux showed a near-linear dependence on input wind speed.
- Cloudwater deposition was proportional to liquid water content (LWC); this proportionality defined a deposition velocity generally in the range of 10 to 70 cm/sec for windspeeds of 2 to 20
- Cloudwater flux, as simulated by the model, was a complex function of the distribution of surface area among various canopy component types (e.g., the relative amounts of needle and twig sizes).
- The modeled contribution of droplet sedimentation compared to impaction decreased rapidly as wind speed increased above 2 m/sec.
- Computed cloudwater deposition was sensitive to droplet size distribution.
- Cloudwater flux increased locally downwind of a forest edge or gap; edge effects could raise droplet deposition velocities from about 40 to 200 cm/sec.
- Cloudwater flux was insensitive to the vertical droplet eddy diffusivity profile.

The sensitivity of the CDM-S model (used by MCCP) to changes in model parameters has been examined by Mueller (1989) (see Table II-5). He calculated that the cloudwater flux parameter was most sensitive to lack of canopy uniformity as represented by a simulated forest edge. Cloudwater deposition at the edge was computed to be four to five times greater than in a closed forest.

The second most important model parameters, in terms of output sensitivity, were cloud liquid water content (LWC) and drop size distribution. As with Lovett's original model, the CDM-S and CDM-D produced computed fluxes that vary linearly with LWC. When the droplet size spectrum is assumed to co-vary with LWC, model response became non-linear; different forms of the drop size and LWC relationship result in computed flux differences by a factor of two. The model is sensitive to the concentration of larger droplets because of collection efficiency and fall speed dependencies on droplet size.

Droplet capture efficiency (computed from the Stokes equation 2, above) was the third most sensitive parameter in the CDM-S version of the cloudwater deposition model (it was second in the CDM-D). Mueller (1989) tested the CDM-S model sensitivity to capture efficiency by varying it across the range of possible values derived from the uncertainty in the experimental data (Thorne et al., 1986) and found that deposition varied by 100 percent. Differences between CDM-S and CDM-D result from different capture efficiency related to morphological differences between conifer needles and deciduous leaves.

Other parameters had less influence on cloudwater flux calculations. Evaporation was important for computed net cloudwater flux when testing model simulations against canopy throughfall data. The effect of net radiation data on model performance is not known because net radiation was not measured by MCCP (Mueller, 1989).

The relative insensitivity of the CDM-S to vertical in-canopy profiles of surface area, wind speed, and eddy diffusivity is fortunate because these parameters are poorly characterized for the MCCP sites and nearby forests. Of these three parameters, the surface area profile appears most important because it determines the computed wind speed profile in the CDM-S and it affects estimates of the vertical turbulent transport rate. Unlike deciduous canopies that tend to have surface area more evenly distributed in the vertical, spruce-fir canopies tend to have the surface area concentrated in a shallow crown space. Differences of 10 to 20 percent in the vertical location of the canopy surface area maximum are relatively unimportant for predicting cloud water deposition with CDM-S. Lovett (1988) calculated that simultaneously increasing three key model input parameters (cloud liquid water content, cloud frequency, and wind speed) by 25 percent resulted in an increase from 154 cm/yr to 300 cm/yr cloud water deposition using the Lovett (1984) cloud droplet interception model. Decreasing these three parameters by 25 percent reduced the predicted cloud water flux from 154 to 65 cm/yr. Therefore, the cloudwater deposition model errors for Lovett's sensitivity analysis were linearly related to the product of the windspeed, liquid water content, and cloud frequency errors. (Uncertainty in these three measurements and in cloudwater chemical composition are discussed in Sos/T chapter 6). Lovett concluded that uncertainty in the input data in addition to extreme variability in time and space make cloudwater deposition modeling impractical for the estimation of cloud water deposition over hectare spatial scales or seasonal temporal scales.

DRY DEPOSITION

Dry deposition is more difficult to measure directly than wet deposition, and the corresponding data set is therefore meager. Methods to measure dry deposition include the use of surrogate surfaces, mass balances, and micrometeorological techniques.

The use of surrogate surfaces has been reviewed by Stevens (1985). Uncertainties in this method result from unknown collection efficiencies, unknown relationships of surrogate to natural surfaces, and exposure of the surrogate surfaces to wet forms of deposition. Mass balance approaches have been used by Eaton et al. (1978) and Galloway and Whelpdale (1980), among others. In this approach, dry deposition results as the residual in a mass balance calculation. Therefore, at best, dry deposition estimates can be as good as the combined variability of the measured parameters. Micrometeorological techniques include at least five methods (Hicks et al., 1980): (1) the eddy correlation method (the only direct method for determining mass flux density); (2) the variance method; (3) the eddy accumulation method; (4) the gradient method; and (5) the modified Bowen ratio method. Micrometeorological techniques produce more reliable results than surrogate surfaces or mass balance methods, but routine implementation is difficult and expensive. In the MCCP, models requiring readily available data sets have been used to estimate dry deposition.

The Inferential Model - Development and Description

Models used to estimate dry deposition take two general forms, both using the concept of electrical resistance. The more complex form, which considers the vertical distribution of canopy structure and air transport structure, has been used in models by Murphy et al. (1977), Shreffler (1978), Bache (1979a & b, 1984), Slinn (1982), and Davidson et al. (1982). The simpler form has often been referred to as the "big-leaf" concept, first introduced by Monteith (1965).

The MCCP uses the best available simple modeling tool to convert dry species concentrations to dry deposition flux densities in adopting the inferential big-leaf model under development by the ATDD/NOAA. This model conceptualizes the system as an Ohm's law analogy of mass flowing to and from a surface through a sequence of series and parallel resistances. These include an aerodynamic resistance that is a function of the turbulent exchange properties of the atmosphere, a quasi-laminar boundary layer resistance that accounts for the role molecular diffusivity plays adjacent to leaf surfaces, and a surface canopy resistance that accounts for leaf transfer processes involving stomatal, cuticular, and mesophyll resistances as well as soil resistances. Simplified models of this type, which simulate canopy transfer processes as if the canopy was a single or "big leaf", show exchanges driven by the concentration gradient of the species in question (ozone, sulfur dioxide, particles, etc.) limited by the appropriate set of resistances in the diffusion pathway. Measured data or submodels must provide estimates of the resistances and the source or sink strengths. Errors may occur for environments having complex terrain or patchy surfaces.

When using big-leaf models for water vapor and sensible heat exchange (e.g., Stewart and Thom, 1973; Jarvis et al., 1976), internal leaf resistance is negligible. For other gases, such as carbon dioxide and air pollutants, leaf sinks or sources are taken into account by defining a virtual resistance to a zero sink or by using a relationship between the environment and the source or sink strength. Such methods have been used to predict carbon dioxide exchange (Waggoner, 1969; Sinclair et al., 1976). For most atmospheric pollutants, the zero-sink case is appropriate, and a deposition velocity is defined as the inverse of the sum of the resistances (Hicks et al., 1987). Such models have been used to predict pollutant flux (Matt et al., 1987; Hosker and Lindberg, 1982; Unsworth, 1980; Wesely and Hicks, 1977).

Currently, estimations of resistance come from climatological and physiological data. Hicks et al. (1985) propose estimating aerodynamic resistance from the standard deviation of the horizontal wind direction and mean horizontal wind speed. Based on work by Brutsaert (1975) and Garratt and Hicks (1973), they also propose the quasi-laminar boundary layer resistance as a function of the Schmidt and Prandl numbers. Canopy surface resistance is the most difficult to estimate where the primary transfer resistance is through the stomata but may also include cuticular and mesophyll resistances. An additional complication arises when resistances vary through the canopy depth and a weighted average value for the big leaf must be derived. In some experimental work, the canopy surface resistance is evaluated as a residual by measuring the flux, the total resistance, the aerodynamic resistance, and the quasi-laminar boundary layer resistance (Baldacchi, 1987). In the model, gross stomatal resistance is combined in series with gross mesophyll resistance, while cuticular resistance is combined in parallel with these, and the gross canopy resistance is determined by weighing with the leaf area index (Hicks, 1985). A review of stomatal resistance concepts is presented by Jarvis (1971). Gross stomatal resistance can be modeled as a function of PAR (Baldacchi et al., 1987; Burroughs and Milthorpe, 1976) as mediated by the effects of water stress, humidity, and temperature (Rodriguez and Davies, 1982; Fisher et al., 1981; Jarvis, 1976). Mesophyll and cuticular resistances, although measured experimentally, have yet to be adequately modeled.

SECTION 7

DESCRIPTION OF DATA BASE AND QUALITY ASSURANCE PLAN

DESCRIPTION OF DATA BASE

The Data Management Center (DMC) of the Fleming Group in Albany, NY, is the central collection, processing, analysis, and distribution point for data collected at MCCP sites and laboratories. DMC activities include data acquisition and entry, storage and documentation of data files, data validation, reduction, and certification, report generation and distribution, data archiving and analysis, and model testing and evaluation. Types of data handled are:

- physical measurements (e.g., presence of cloud, precipitation, cloud LWC)
- meteorological measurements (e.g., temperature, wind speed and direction)
- aqueous phase measurements (e.g., cloud, precipitation, and throughfall chemistry)
- gas phase measurements (e.g., ozone, sulfur dioxide, hydrogen peroxide)
- ancillary data (e.g., site latitude, longitude, and elevation).

These data are stored in an ORACLE relational database. The computer hardware is a DEC VAX 11/750 with the VMS version 4.1 operating system. The database is implemented using ORACLE version 5.0.20. All programming applications are written in C using the Digital version 2.1 compiler.

MCCP data are available from the Data Management Center by written request to The Fleming Group, 55 Colvin Ave., Albany, NY 12206. Timeseries data sets have been developed for the analysis of multiple parameters in time sequences. The MCCP data are organized into hourly records containing all measurements available for that hour, including meteorological, gas, physical, and chemical data. The data are further organized by site, month, and year, and are available on floppy disk or magnetic tape. These data sets are available for 1986 to 1988 data.

INTERNAL QUALITY ASSURANCE

Internal quality assurance/quality control (QA/QC) for the MCCP is coordinated and managed by the Fleming Group, in Albany, NY. QA consists of:

- development of standard operating procedures (SOPs) and quality assurance plans;
- preparation of training sessions and site technician certification;
- writing annual QA/QC summaries;
- conducting system audits; and
- data validation and certification.

Standard quality control practices for the field (i.e., methods of tracking performance) include training (by on-site personnel), station checks, station logs, zero/span checks, precision measurements, and calibrations. All station checks, routine measurements, and record keeping follow the MCCP SOP and QA plan. Table III-1 summarizes the data quality objectives for the MCCP.

Additional quality control is provided by a site QA coordinator who verifies that monitoring procedures are followed, checks all instrument calibration records, and confirms any effect of problems on the final data. The network QA officer informs the project manager of problems at each site.

The QA office also monitors laboratory quality control procedures for both the on-site laboratories and a central analytical laboratory (CAL), the Illinois State Water Survey (ISWS), that analyzes samples from all the sites. The latter is the same laboratory that analyzes the NADP/NTN rain samples. The CAL established the QA/QC protocols for the laboratories and prepares QC check solutions.

Performance of site labs with respect to CAL are evaluated by split sample comparisons. A split is a sample that has been divided into two aliquots in the field. The results from the analyses of these split samples are used to determine the precision between the two labs. Table III-2 summarizes the splits analysis results for 1986, 1987, and 1988.

An internal systems audit for each site is performed annually by the QA Office. Inspection includes evaluation of all aspects relevant to the goals of the program and adherence to the Project QA Plan. Upon completion, a report is written to describe the results of the audit and recommending actions for improvement.

Quality Assurance reports to management have been prepared on an annual basis. Copies of these reports may be obtained from The Fleming Group or the EPA.

APPENDIX A

EUROPEAN MONITORING

We present this review European cloud water chemistry to provide a context for viewing USA and Canadian data. Few true "networks" exist; site specific data for Sweden and Italy are included for comparison with FRG and North American data.

GERMANY

Since 1916, several researchers in the Federal Republic of Germany have collected fog/cloud water using string collectors (Linke, 1916; Rubner, 1931; 1935; Grunow, 1953; 1957; 1958; Baumgartner, 1958a; 1958b). The interest in fog/cloud chemistry has grown in recent years primarily in response to an increase in forest damage. It has been hypothesized that chemical species in fog/cloud water intercepted by forest canopies contribute to that damage. Hence, several institutions developed a variety of fog/cloud collectors. To ensure data compatibility and quality, in 1986 the Federal Ministry for Research and Technology (BMWFT) funded a comparison of collectors at the Center for Environmental Research, University of Frankfurt (Enderle and Jaeschke, 1988).

Although there is currently no cloud monitoring network in Germany, several groups have initiated limited field experiments. Of special interest are cloud chemistry data obtained over an extended period of time, such as reported by the University of Frankfurt group (Schmitt, 1989), the German Weather Service in Hamburg (Kroll and Winkler, 1989), and the University of Bayreuth (Trautner, 1988).

The Frankfurt group obtained cloud samples from Little Feldberg Mountain, a region close to urban centers and heavily industrialized complexes, including chemical factories. These results are shown in Table A1-1. The Weather Service group operated five mountain sampling sites during an 18-month period from October 1986 through May 1988: Hohenpeissenberg (HP), Kahler Asten (KA), Wasserkuppe (WK), Grosser Arber (GA), and Schauinsland (SL). Each fog event contributed one sample for chemical analysis. These values, presented in Table A1-2, were standardized to a uniform liquid water content (LWC) of 0.2 g/m^3 to avoid additional variability due to different LWCs.

TABLE A1-1

Arithmetic mean, maximum, and minimum concentrations in 410 cloud/fogwater samples for the falls of 1983 - 1986 in Kleiner Feldberg/Taunus, 15 km north of Frankfurt. Values are in µeq/L.

	Mean	Maximum	Minimum
SO_4^{2-}	831	5620	87.4
NO_3^-	778	7067	28.6
Cl^-	243	2257	17
NH_4^+	1197	5210	55.4
H^+	158	5011	0.0126

TABLE A1-2

German Weather Service Mountain Sites (Active impactor-type collector)
All values are normalized to LWC of 0.1 g/m^3.

	Station				
	HP	KA	WK	SL	GA
Ion Species (μeq/L)					
H^+	151	200	43	85	148
SO_4^{2-}	110	219	108	73	260
NO_3^-	102	229	82	47	223
NO_4^+	172	321	139	133	310
# of events	381	126	20	22	195
LWC (g/m^3)	0.025	0.057	0.055	0.042	0.127

The Bayreuth group (Trautner, 1988) collected a total of 25 weekly samples at a mountain site in the Bavarian Fichtelgebirge (Wuelfersreuth). Each fog event yielded one sample, and no attempt was made to differentiate precipitating and non-precipitating events. The mean ion concentrations are listed in Table A1-3.

TABLE A1-3

Weekly Fog/Cloud Water Samples Obtained with Passive String Collector at Wuelfersreuth (Fichtelgebirge) September 1984 - March 1986 (Fall/Spring only).

	Mean	Standard Deviation	Maximum	Minimum
pH	3.9	4.0	3.4	6.5
LWC (g/m^3			0.137	0.003
Ion Species (μeq/L)				
SO_4^{2-}	336	191	892	65
NO_3^-	334	192	936	68
NH_4^+	253	149	569	44

Appreciable regional difference sin the chemical composition of cloud water have been observed for these mountain measurements. The mean concentrations varied greatly not only among different stations but also within stations. Since each investigator adhered to his/her own standard operating procedures for cloud/fog collection and analysis, it is difficult to prepare a quantitative summary from the results to date in Germany. It is clear, however, that cloud/fog water was enriched compared with precipitation water for all major ions (including those not summarized in Tables A1-1 through A1-3). The results show that high-elevation forests were exposed to substantial concentrations of chemicals in clouds/fogs.

ITALY

A field test to investigate microphysical and chemical characteristics of fog was organized by FISBAT (Istituto Perlo Studio dei Fenomeni Fisici e Chimici della Bassa e Alta Atmosphera) in the eastern Po Valley during February and November of 1984 (Fuzzi et al., 1988). Twelve American and European institutes participated (Atmospheric Sciences Research Center, Albany, NY (USA); Atlanta University, Atlanta, GA (USA); Italian National Electricity Board (Italy); Lawrence Berkeley Laboratory, Berkeley, CA (USA); Technical University Vienna (Austria); University of Bologna (Italy); University of Frankfurt (FRG); University of Padova (Italy) and University of Vienna (Austria). Rotating string collectors collected fog on an hourly basis. Twenty samples were obtained during February 1984, and 87 samples from six events were obtained during November 1984. The mean, maximum, and minimum concentrations for the major chemical components in the Po Valley fog are presented in Table A1-4. The average anion to cation ratio was 0.96, an indication that no important constituents were missed. Four ions accounted for most of the ionic strength of the fog water solutions: H^+, NH_4^+, NO_3^-, and SO_4^{2-}. The average liquid water content (LWC) was 0.08 g/m^3 (maximum 0.29 g/m^3) measured by a high-volume filter technique.

TABLE A1-4

Hourly Fog Samples Obtained with Rotating String Collectors in Po Valley, Italy February and November, 1984.

	Mean		Maximum		Minimum	
	Feb	Nov	Feb	Nov	Feb	Nov
H^+	200	160	3600	1200	10	1
SO_4^{2-}	1000	900	6300	2100	350	150
NO_3^-	1100	900	8200	3300	290	80
NH_4^+	2100	1400	8100	4200	700	300
H_2O_2	< 0.1					
LWC (g/m^3)	0.08		0.29			

SWEDEN

A ground-based passive string collector sampled water from stratiform clouds for approximately five weeks during the summers of 1983 and 1984 in the mountains of central Sweden (Areskutan, 1250 m asl; Ogren and Rodhe, 1986). These measurements were considered representative to air arriving over central Scandinavia. Cloud liquid water content was measured only in 1984, using a heated-rod impactor King probe (King et al., 1978). A total of 179 cloud water samples (resolution less than one hour) were obtained and a wide range of concentrations was encountered; for example, sulfate concentrations ranged from 1 to 1600 μeq/L. Table A1-5 shows the mean ion concentrations classified into four air mass arrival sectors. Transport of air from the North Atlantic Ocean (NW) was associated with low concentrations, while high concentrations occurred with transport from industrial regions in Europe (S). Long-range transport appears to be the primary factor controlling the chemical composition of cloud water at the sampling site in central Scandinavia.

TABLE A1-5

Mean Ion Concentrations in Hourly Cloud Water Samples from Four Trajectories for Areskutan, Sweden, 1250 m asl
A Passive String Collector Sampled Precipitating and Non-precipitating Clouds in the Summers of 1983 and 1984.

	Trajectory			
Ion Species (μeq/L)	NE	NW	W	S
H^+	33	13	38	370
SO_4^{2-}	34 max 150	6 max 19	30 max 220	700 max 930
NO_3^-	9	2	10	68
NH_4^+	17	0.5	1	NA
LWC	Averaged over all directions = 0.16 g/m^3			
No. of Samples	47	41	33	4

CANADIAN MONITORING

There are areas of forest at elevations above 600 m scattered throughout southern Quebec, along the Gaspe Peninsula and in northern New Brunswick. Because higher elevation forests show unexplained damage symptoms in central Europe and the eastern United States, the Chemistry of High Elevation Fog (CHEF) program was started in late 1985 to measure wet and dry deposition as well as the meteorological conditions at several higher altitude sites. The objectives of the CHEF program are similar to those of the Mountain Cloud Chemistry Project (MCCP) in the USA (Schemenauer, 1986; Schemenauer et al., 1988; Schemenauer and Winston, 1988). The CHEF program operates 12 months a year on two mountains (Mt. Tremblant and Roundtop Mountain) and for four months on a third mountain (Mt. Epaule). Paired fog/cloud water samples are collected with a passive string collector that is very similar to the MCCP cloud water collector. The program is designed to sample every fog event on each day of the year. The sampling periods can vary from 1 to 24 hours depending on operator availability. Because an operator is at the field station only eight hours per day, when fog is expected overnight a pair of collectors are exposed at the end of the afternoon and the samples collected the next morning.

The CHEF program is still in the initial stages of data analysis and interpretation. Cloud/fog samples at Mt. Morency were collected between 25 June and 24 September 1986. The data are presented as sample means in Table A1-6. The concentration of all ions was 10 to 20 times higher in non-precipitating cloud water than in precipitation. The precipitating cloud/fog samples had intermediate values but more closely resembled the precipitation data. The dominant cations were H^+ and NH_4^+ and the dominant anions SO_4^{2-} and NO_3^-. Fog/cloud samples from Roundtop collected between 1 May and 25 September 1986 are also shown in Table A1-6. Note that the small number of

samples precludes any comparison of precipitating and non-precipitating clouds without more recent CHEF work.

The principal CHEF sampling locations are at altitudes near the typical cloud base heights for southern Quebec (845 to 970 m). Thus, the main depth of the cloud is usually above the sampling location and the likelihood of precipitating being mixed with cloud events is high. Any indication of precipitation at the field sites during the exposure of the passive fog collector results in the sample being classified as a "precipitation fog/cloud" event. The CHEF data are in agreement with those reported for northeastern MCCP sites. Schemenauer (1986) reported that the CHEF monitoring sites in Quebec are in cloud approximately 44% of the year, with lower elevations (about 500 m) experiencing clouds for about 23% of observations.

Schemenauer et al. (1988) collected paired samples to screen the data for possible contamination. These results describe the combined experimental precision related to uncertainties in cloud water collection (ASRC-type passive sampler), chemical analysis, and handling procedures. About 3% of the CHEF samples were eliminated due to a "significant" difference in concentration between the sample pairs. The remaining 17 pairs of samples had a median difference of 4 μeq/L for sulfate (mean total concentration was 199 μeq/L) and 1.9 μeq/L for nitrate (mean concentration was 65 μeq/L); the median difference was about 2% of observed concentrations.

Except for the length of the sampling period the CHEF and Mountain Cloud Chemistry Project (MCCP) protocols are similar. Samples from MCCP and CHEF have been exchanged for comparisons of analyte concentrations. It is anticipated that for non-precipitating cloud water that the CHEF and MCCP cloud water concentration data are directly comparable.

TABLE A1-6 .

Mean Ion Concentrations at Two Canadian CHEF Sites in 1986 (from Schemenauer and Winston, 1988).

	Morency (970 m)		Roundtop (970 m)		Roundtop (845 m)	
	NP[a]	P	NP	P	NP	P
# samples	7	20	3	57	17	81
pH	3.42	3.66	4.52	3.65	3.89	3.71
Ion species (μeq/L)						
H^+	383	218	29.9	224	129	197
SO_4^{2-}	522	301	44	244	227	248
NO_3^-	87.5	51.1	21.9	104	72.9	104
NH_4^+	215	141	52.4	116	149	125

[a] NP = non-precipitating clouds
N = precipitating clouds

APPENDIX B

DATA REPORTS

This section lists the principal data reports and publications from MCCP, ISF, and CHEF cloud water routine monitoring activities. A brief summary of each is provided.

Mohnen and Kadlecek (1989) presented typical winter and summer events to contrast time-dependent histories of chemical concentrations for the two seasons. During summer, sufficient hydrogen peroxide (H_2O_2) existed to oxidize available SO_2 to SO_4, temporarily depleting the H_2O_2. In winter, H_2O_2 and oxidation rates were low; thus, sulfate concentrations in cloudwater were generally independent of SO_2. A chemical cloud climatology for Whiteface Mountain was presented for data from 1982-1987. Summer mean hydrogen ion concentrations ranged from a low of 174 μeq/L in 1987 to a high of 331 μeq/L in 1984. For sulfate, summer means ranged from 113 μeq/L in 1987 to 198 μeq/L in 1984. Summer means for ammonium ranged from 110 μeq/L in 1986 and 1987 to 173 μeq/L in 1983, and for nitrate ranged from 49 μeq/L in 1982 to 82 μeq/L in 1986. Standard deviations were typically of the same magnitude as mean concentrations for these four ions. All summer sampling seasons recorded values of 3.1 or less, with three of five recording hourly cloudwater pHs of less than 2.8. The minimum pH for five summers was 2.5 during 1983.

Canadian CHEF data from spring 1986 were presented and compared to the fall 1985 data by Schemenauer (1986; 1988). Daily median and mean values both led to the same conclusions. Precipitation in the valley and on the summit had a similar median pH of 4.5, compared to pH 4.0 for cloudwater on the Tremblant summit during the spring of 1986. The CHEF ground-based cloudwater concentrations were similar to aircraft measurements made 400 km west (North Bay, Ontario), published by Issac and Daum (1986). Cloud water deposition (77 cm/yr) was calculated to be similar to average annual precipitation in Quebec. Cloud water H^+ deposition (135 $\mu eq/m^2$) was about three times that from precipitation at nearby low-elevation sites (Schemenauer, 1986; 1988).

Saxena et al. (1989) presented cloudwater chemistry results from Mt. Mitchell, NC, for the summer of 1986. Precipitation had an average pH of 4.4, while cloud events averaged pH 3.4 (events ranged from pH 2.2 to 5.4). Cloud liquid water content (LWC) was highly variable. Short-duration events (2 to 8 hr) were more acidic and more frequent (58% of total events) than longer events associated with frontal passage. Easterly surface winds were associated with less acidic cloudwater than westerly winds. The pH level was usually lower during the onset and dissipation stages of a cloud event than during the middle of a long-duration event. Clouds occurred during 40% to 50% of the hours at the Mt. Mitchell summit (1950 m) during June-August 1986, but only 5% of the same period at a lower elevation on Mt. Mitchell (1740 m). Stogner and Saxena (1988) presented much of the same Mt. Mitchell data.

Olszyna et al. (1988) measured aqueous and gas phase H_2O_2 and aqueous ions in cloud and rainwater at Whitetop Mountain, VA, during summer and fall of 1986. Gas phase concentrations of H_2O_2 were higher in summer than in fall (0.8 versus 0.15 ppb) and were strongly correlated with ozone, temperature, and dew point. Daytime H_2O_2 exceeded nighttime values by 26%. Cloudwater acidity data are not presented here. During spring, summer, and fall of 1986 at Whitetop Mountain, the mean cloudwater [H_2O_2] was 26 μmol/l (for 100 samples), while the mean rainwater H_2O_2 was 10 μmol/l (28 samples). A maximum aqueous H_2O_2 of 247 μmol/l was observed, the highest value reported to date in the literature; 13% of summer cloud samples showed greater than 120 μmol/l H_2O_2. Comparison of rain and cloud H_2O_2 during periods of poor vertical mixing in the atmosphere suggested that H_2O_2 levels aloft exceed those at the ground.

Mueller and Weatherford (1988) computed cloud deposition on Whitetop Mountain, VA for 26 cays in the spring of 1986. Using Lovett's model (Lovett, 1984), the computed cloud deposition for the study period. The cloudwater SO_4 flux was between 5.3 and 9.1 kg/ha/mo, while NO_3 flux was between 2.8 and 5.4 kg/ha/mo. The ranges reflect projected variation in unmeasured model input data.

Lindberg et al. (1988) used Integrated Forest Study (IFS) data to estimate cloud, precipitation, and dry deposition at a high-elevation site in the Great Smoky Mountains. Cloudwater SO_4^{2-} and NO_3^- deposition were estimated to be 7.2 and 2.2 kg/ha/mo, respectively, both two to five times greater than estimates from a nearby low-elevation site for January-April 1986. This is the only published paper from the IFS cloud water studies.

More recent deposition estimates from IFS appear in discussions by Lovett, Knoerr and Conklin, and Ragsdale in a draft summary of the IFS that was edited by Lindberg and Johnson (1989). This report does not present cloud water concentrations and presents cloud deposition only in the form of graphs. Cloudwater H^+ deposition at these two sites was estimated to be 30% to 40% of total H^+ deposition. The IFS scientists concluded that the primary sources of uncertainty in cloudwater deposition were immersion time, cloudwater amount, and ion concentrations.

Other than Lovett's original work at Mt. Moosilauke, the best documented estimates of cloud water chemical exposure and deposition are from the Mountain Cloud Chemistry Program (MCCP) by Mohnen and co-workers (1988a; 1988b; 1988c). The CDM-S and CDM-D models (Mueller, 1989; Krovetz et al., 1989) were used to predict deposition to the spruce-fir forests on five mountains in the eastern USA. Sigmon et al. (1989) and Joslin et al. (1988) estimated deposition at Shenandoah and Whitetop Mountain, VA, from canopy throughfall measurements.

APPENDIX C

SUMMARY OF METEOROLOGICAL DATA

FOR THE MOUNTAIN CLOUD CHEMISTRY PROJECT SITES

1986 - 1988

SUMMARY OF METEOROLOGICAL DATA
SITE: HOWLAND FOREST

Periods of Record: 1987: Apr 11 - Nov 11
1988: Mar 25 - Nov 15

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1987	8.4	12.1	16.9	18.8	17.9	13.5	7.5
1988	5.4	13.2	17.0	21.1*	20.0	13.7	6.4
Max	25.6	31.1	35.9	33.0	34.2	28.7	26.9
Min	-5.4	-2.8	1.6	4.9	3.4	-2.5	-5.0
Mean	6.6	12.7	17.0	19.7	18.9	13.6	7.0

Average Relative Humidity (%):

1987	59.4	57.3	67.8	73.6	68.1	74.4	72.2
1988	68.9	62.0	66.2	77.5*	74.1	74.3	73.6
Mean	65.1	59.6	67.0	75.1	71.0	74.3	72.9

Total Precipitation (mm)

1987	55.1	62.7	54.9	53.6	25.4	146.3	56.1
1988	48.5	13.0	47.2	68.8*	121.9	41.1	84.3
Mean	51.1	37.8	51.0	59.6	73.6	93.7	70.2

Average Wind Speed (m/s):

1987	2.8	3.1	2.7	2.4	2.5	2.6	2.6
1988	3.2	3.1	2.8	1.8*	2.0	2.6	2.6
Max Hr.	8.9	10.9	8.4	6.7	9.0	7.6	8.8
Mean	3.1	3.1	2.7	2.2	2.2	2.6	2.6

Resultant Wind Direction (degrees):

1987	30.5	258.1	241.4	220.6	257.0	264.2	233.9
1988	6.3	243.8	290.4	243.9*	234.4	264.5	244.0
Mean	15.8	250.9	265.9	230.0	245.7	264.4	239.0

Average Solar Radiation (W/m2):

1987	333.1*	237.5	220.5	197.6	213.9	140.3	99.8
1988	242.2*	299.0*	340.7*	182.0*	175.5	203.9*	110.2*
Max	869.0	906.3	956.8	954.3	880.5	801.7	658.5
Mean	277.4	263.3	270.3	191.4	195.2	167.2	104.2

Average Pressure (mB):

1987	1013.8	1012.9	1007.8	1007.2	1007.7	1008.3	1010.2
1988	1003.8	1010.8	1004.5	1009.5*	1009.4	1010.5	1011.4
Max	1025.2	1026.2	1019.6	1017.7	1020.3	1026.0	1028.6
Min	973.4	994.2	987.2	994.8	995.8	984.3	988.1
Mean	1007.8	1011.9	1006.2	1008.1	1008.6	1009.4	1010.8

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SUBSITE: MT. MITCHELL 1

Periods of Record: 1986: May 12 - Nov 20
1987: May 14 - Oct 22
1988: May 10 - Oct 1

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	--	11.6*	13.3	14.0*	12.2	11.6	7.3
1987	--	12.8	13.3	15.1	14.6	10.3	4.8
1988	--	10.8	13.6	14.8	15.2*	12.0*	--
Max	--	20.5	22.1	23.5	23.5	19.9	17.2
Min	--	2.0	0.4	6.1	-0.6	2.2	-10.2
Mean	--	11.7	13.4	14.8	13.9	11.3	6.2

Average Relative Humidity (%):

1986	--	--	79.7*	82.3*	90.0	87.9*	77.9
1987	--	90.2	84.4	84.4	88.0	90.8	60.7
1988	--	71.4	73.3	84.2	88.1*	88.5*	--
Mean	--	79.6	79.1	83.9	88.7	89.2	70.5

Total Precipitation (mm):

1986	--	57.2	2.3	0.0*	4.3	22.9	186.9
1987	--	79.2	145.3	51.3	47.2	306.6	3.3
1988	--	26.9	36.8	96.8	105.2*	145.3*	--
Mean	--	52.4	61.6	58.5	49.4	161.0	123.8

Average Wind Speed (m/s):

1986	--	6.7	6.1	8.3*	6.4	6.4	7.7
1987	--	4.2	6.5	5.3	6.5	6.6	7.4
1988	--	5.6	5.8	5.5	5.6*	6.5*	--
Max Hr.	--	21.0	19.2	21.6	20.0	18.9	19.9
Mean	--	5.6	6.1	6.0	6.2	6.5	7.6

Resultant Wind Direction (degrees):

1986	--	271.9	293.3	296.2*	298.9	284.8	295.7
1987	--	266.5	289.7	293.4	349.8	83.4	297.1
1988	--	284.6	292.6	270.7	245.4*	265.5*	--
Mean	--	275.0	291.9	285.1	300.8	206.9	296.2

Average Solar Radiation (W/m2):

1986	--	165.3	220.6	206.1*	140.9	150.4	135.5
1987	--	162.6	231.5#	--	--	--	--
1988	--	301.1*	355.5	316.6*	285.9*	269.5*	--
Max	--	1014.8	1075.9	971.3	946.6	910.7	880.3
Mean	--	226.8	272.1	266.7	192.0	193.2	135.1

Average Pressure (mB):

1986	--	807.0	808.5	809.4*	812.8	815.8	811.3
1987	--	810.4	808.5	810.9	810.2	807.3	804.6
1988	--	806.1	808.5	811.3	810.3*	808.9*	--
Max	--	817.3	818.7	817.1	823.6	828.1	821.4
Min	--	798.4	796.9	801.5	802.2	794.7	793.0
Mean	--	807.7	808.5	810.7	811.1	810.8	808.6

-- No Data, * Data Recovery 50% to 85%, # Data Recover < 50%

SUMMARY OF METEOROLOGICAL DATA
SUBSITE: MT. MITCHELL 2

Periods of Record: 1986: Jun 28 - Nov 20
1987: May 18 - Oct 23
1988: May 12 - Sep 30

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	--	--	17.5	17.2#	--	14.5*	8.7
1987	--	15.1	15.6	17.6	17.0	12.7	3.3#
1988	--	12.3	15.6	17.0	17.5	13.4	--
Max	--	19.7	24.6	25.0	26.3	22.1	21.0
Min	--	5.0	2.2	8.8	10.1	3.9	-7.9
Mean	--	13.4	15.7	17.3	17.3	13.4	8.0

Average Relative Humidity (%):

1986	--	--	73.8	71.1#	--	81.3	--
1987	--	80.9	83.2	77.6	83.2	87.3	56.6#
1988	--	66.4	67.7	78.1	83.0	84.5	
Mean	--	72.3	75.1	77.2	83.1	67.2	56.6

Total Precipitation (mm):

1986	--	--	11.4	200.4	--	23.0*	164.6
1987	--	40.6	121.9	54.1	58.9	309.6	0.0#
1988	--	19.3	40.6	95.8	122.2	126.5	--
Mean	--	28.0	78.7	116.4	90.5	170.5	142.4

Average Wind Speed (m/s):

1986	--	--	3.3	3.8	3.1#	3.9	4.2
1987	--	2.8	3.9	3.6	3.7	4.1	5.3#
1988	--	4.3	3.3	3.9	3.2	3.6	--
Max Hr.	--	12.4	13.0	16.6	11.6	13.9	12.1
Mean	--	3.7	3.6	3.8	3.4	3.9	4.3

Resultant Wind Direction (degrees):

1986	--	--	287.9	284.9	290.1#	264.8	266.5
1987	--	257.5	277.9	281.0	283.4	251.3	286.6#
1988	--	261.0	305.6	260.8	260.1	255.8	--
Mean	--	259.6	291.6	275.5	274.8	257.1	269.2

Average Solar Radiation (W/m2):

1986	--	--	187.7	221.8	155.0#	170.2	153.0
1987	--	159.5	213.9	201.8	177.7	157.5	204.1#
1988	--	229.2	252.9	208.2	193.2	152.4	--
Max	--	1027.5	1027.5	1052.1	979.6	954.2	861.9
Mean	--	200.6	231.8	210.8	180.3	159.9	159.9

Average Pressure (mB):

1986	--	--	828.6	831.8	831.5#	832.5	829.3
1987	--	824.9	822.6	825.0	824.5	821.7	817.7#
1988	--	820.1	823.1	826.3	825.6	824.5	--
Max	--	839.3	832.8	838.6	835.5	837.5	809.6
Min	--	812.1	812.2	815.6	817.6	809.6	838.8
Mean	--	822.1	823.1	827.7	826.1	826.1	827.7

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SUBSITE: MT. MITCHELL 3

Periods of Record: 1988: May 14 - Sep 29

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1988	--	17.2	19.7	21.6	22.0	18.1	--
Max	--	28.2	31.9	33.0	33.8	28.7	--
Min	--	2.3	4.3	9.2	12.3	6.9	--

Average Relative Humidity (%):

1988	--	68.4	71.4	80.2	84.1	85.9	--

Total Precipitation (mm):

1988	--	22.1	64.3	105.9	71.4	117.3	--

Average Wind Speed (m/s):

1988	--	1.4	1.2	1.1	1.0	1.1	--
Max Hr.	--	6.1	4.3	5.4	4.2	3.7	--

Resultant Wind Direction (degrees):

1988	--	193.8	222.9	198.7	210.5	211.8	--

Average Solar Radiation (W/m2):

1988	--	245.4	258.9	222.5	204.6	159.6	--
Max		1060.7	1015.8	987.2	957.9	892.4	--

Average Pressure (mB):

1988	--	927.3	929.5	932.0	930.6	930.9	--
Max	--	934.9	940.3	941.0	942.9	940.3	--
Min	--	920.3	917.8	923.7	921.7	917.3	--

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SITE: MT. MOOSILAUKE

Periods of Record: 1986: Jul 12 - Oct 22
1987: Apr 9 - Oct 15
1988: Jun 2 - Oct 18

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	--	--	--	15.9	13.5*	9.8	4.7
1987	6.5	9.9	14.0	18.0*	14.7	10.9	4.8
1988	--	--	14.1*	18.9	17.1	10.8	4.2
Max	21.7	25.7	28.6	28.7	28.7	20.8	19.4
Min	-4.5	-3.1	2.4	4.5	1.2	-0.1	-5.9
Mean	6.5	9.9	14.0	17.8	15.3	10.5	4.6

Average Relative Humidity (%):

1986	--	--	--	85.4	82.1*	79.8	76.8
1987	65.4	66.2	77.4	80.9*	77.3	84.6	76.7
1988	--	--	61.6*	74.9	82.6	79.3	86.0
Mean	65.4	66.2	70.5	79.8	80.6	81.3	79.8

Total Precipitation (mm):

1986	--	--	--	83.3	101.6	82.3*	23.9
1987	24.6	30.2	163.8	--	128.5#	141.0	46.7
1988	--	--	26.4	68.8*	139.7	51.8	9.1
Mean	24.6	30.2	100.9	74.4	125.2	81.4	25.2

Average Wind Speed (m/s):

1986	--	--	--	3.2	4.1*	4.4*	4.5
1987	4.6	4.6	4.7	3.4*	4.1	4.7	5.3
1988	--	--	4.9*	3.3	4.0	5.0	4.6
Max Hr.	13.3	13.3	17.8	13.0	15.4	15.4	21.3
Mean	4.6	4.6	4.8	3.3	4.1	4.7	4.8

Resultant Wind Direction (degrees):

1986	--	--	--	312.0	265.0*	--	311.6
1987	53.5	307.7	306.5	312.2*	305.9	313.4	327.9
1988	--	--	307.5*	303.4	288.6	296.1	273.7
Mean	53.5	307.7	306.9	308.8	294.8	304.8	303.4

Average Solar Radiation (W/m2):

1986	--	--	--	176.9	183.6*	144.1	102.0
1987	313.1*	227.3	197.1	209.7*	208.5	125.6	110.0
1988	--	--	259.5*	226.1	225.3*	151.7	65.9
Max	890.3	923.1	966.0	968.9	994.4	816.8	703.5
Mean	313.1	227.3	224.3	207.3	206.8	140.3	92.2

Average Pressure (mB):

1986	--	--	--	907.8	908.1*	908.4	906.0
1987	905.5	908.0	905.1	908.8*	908.4	907.3	904.7
1988	--	--	904.3*	909.7	909.2	908.8	907.7
Max	915.2	918.3	915.8	919.8	918.3	921.7	921.0
Min	880.5	892.8	887.3	894.4	892.0	887.1	886.3
Mean	905.5	908.0	904.8	908.9	908.6	908.2	906.2

-- No Data, * Data Recovery 50% to 85%, # Data recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SUBSITE: SHENANDOAH 1

Periods of Record: 1986: Apr 24 - Nov 24
1987: Apr 20 - Nov 15
1988: Apr 14 - Nov 3

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	15.1	14.0	18.1#	20.2#	17.0	15.2	10.1
1987	9.6	15.0	19.4	21.8*	20.3	16.7	8.2
1988	6.7	13.7	17.4	21.6	21.0	14.9	6.4
Max	23.6	27.0	29.8	33.9	32.5	26.4	25.4
Min	-1.9	-0.4	4.2	6.5	4.0	1.5	-4.0
Mean	9.2	14.2	18.3	21.5	19.4	15.6	8.2

Average Relative Humidity (%):

1986	48.9	68.4	61.8#	84.3#	85.5	80.7	69.3
1987	64.4	64.6	68.2	69.9*	69.4	75.2*	59.3
1988	55.4	72.0	66.2	69.1	72.7	79.3	64.3
Mean	57.0	68.5	66.2	71.4	76.0	78.5	64.2

Total Precipitation (mm):

1986	2.3	123.7	--	6.9#	184.4	21.1	30.7
1987	19.3	154.2	91.4	49.0	40.6	265.9	30.5
1988	10.9	217.9	41.1	66.5	74.9	66.0	5.8
Mean	11.9	165.3	66.3	52.4	100.6	114.3	24.3

Average Wind Speed (m/s):

1986	5.7	5.0	4.5*	4.1#	4.2	4.2	4.8
1987	5.5	4.0	4.0	3.7	4.2	4.4	5.3
1988	6.4	4.2	4.3	3.6	4.0	3.9	4.8
Max Hr.	15.2	14.1	12.7	10.3	13.6	12.1	13.5
Mean	6.0	4.4	4.3	3.7	4.1	4.2	5.0

Resultant Wind Direction (degrees):

1986	301.1	265.0	283.1*	291.8#	195.1	262.0	288.4
1987	12.1	259.0	294.5	295.4	205.2	209.5	290.6
1988	294.1	214.6	300.6	273.1	236.5	257.5	284.9
Mean	208.1	246.2	293.3	284.4	212.3	243.8	288.0

Average Solar Radiation (W/m2):

1986	288.0	243.7	286.3*	250.3#	201.3	173.6	142.1
1987	255.1*	230.4	258.6	240.3	228.2	155.2	167.1
1988	219.5	220.1	264.6	233.9	219.0	180.7	145.1
Max	979.3	1038.0	1113.0	984.0	1037.0	915.1	834.8
Mean	242.7	231.4	268.8	238.4	216.0	170.2	151.4

Average Pressure (mB):

1986	902.0	901.9	903.6*	903.0#	905.6	907.3	905.8
1987	901.0	906.1	904.2	906.1	906.1	904.7	905.1
1988	895.5	903.2	904.6	907.7	907.3	907.7	904.4
Max	908.3	911.9	916.8	916.0	914.5	917.2	919.3
Min	884.9	892.2	896.3	897.1	898.3	890.1	887.2
Mean	898.4	903.7	904.2	906.5	906.3	906.6	905.1

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SUBSITE: SHENANDOAH 2

Periods of Record: 1986: May 17 - Nov 22
1987: Apr 22 - Nov 17
1988: Apr 16 - Nov 2

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	--	18.6	21.6	23.6	20.2	18.7	13.7
1987	11.3	17.9	22.5	23.5*	22.8	19.1	10.8
1988	9.7	16.8	20.4	24.5	23.9	17.8	9.6
Max	25.5	29.8	32.3	36.1	34.9	29.3	27.8
Min	2.1	3.7	7.2	9.7	8.4	6.1	-0.6
Mean	10.3	17.6	21.5	23.9	22.3	18.5	11.4

Average Relative Humidity (%):

1986	--	68.0	60.6	68.6	72.4	69.6	60.1
1987	59.4	64.4	64.7	66.1*	66.2	73.0	54.0
1988	46.9	61.7	58.0	61.7	64.3	70.5	55.2
Mean	51.5	64.0	61.1	65.4	67.7	71.0	56.4

Total Precipitation (mm):

1986	--	98.0	94.5	1.5	--	13.0	24.6
1987	16.3	125.2	89.2	19.1*	30.2	218.9	32.3
1988	10.2	201.2	38.6	86.4	79.0	76.5	31.8
Mean	12.4	149.7	74.1	47.6	38.4	100.7	29.6

Average Wind Speed (m/s):

1986	--	2.1	2.6	2.6	2.5	2.4	2.3
1987	3.6	2.2	2.3	2.4*	2.8	2.5	3.0
1988	4.1	2.9	2.7	2.2	2.6	2.4	3.0
Max Hr.	12.6	11.5	8.2	10.8	9.3	9.0	9.9
Mean	3.9	2.5	2.5	2.4	2.6	2.4	2.8

Resultant Wind Direction (degrees):

1986	--	192.6	253.4	256.4	182.8	195.1	267.0
1987	276.3	206.4	246.2	183.4*	176.0	191.0	274.1
1988	311.4	165.8	285.7	195.1	175.2	191.7	287.3
Mean	298.5	187.9	261.8	216.3	178.0	192.6	276.1

Average Solar Radiation (W/m2):

1986	--	217.0	272.5	240.5	192.1	169.5	132.5
1987	218.6	228.2	257.2	248.2*	223.7	158.7	151.1
1988	184.3	216.6	242.7	227.6	212.1	169.6	136.0
Max	965.6	1030.5	1075.4	970.8	973.3	880.1	808.0
Mean	196.5	221.5	257.5	237.2	209.0	166.0	139.9

Pressure (mB):

1986	--	935.1	937.0	937.9	939.3	941.4	940.7
1987	936.1	940.6	937.6	939.7*	939.8	938.5	940.1
1988	934.8	942.2*	--	938.4*	938.4	940.1	937.9
Max	975.6	969.0	945.3	947.8	949.9	950.5	955.9
Min	922.1	930.1	929.0	927.4	929.2	922.3	921.4
Mean	935.3	939.8	937.3	938.5	939.2	940.0	939.6

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SITE: WHITEFACE

Periods of Record: 1986: Jun 19 - Oct 28
1987: May 29 - Oct 13
1988: May 26 - Oct 17

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	--	--	8.0*	11.9	9.7	6.4	0.7
1987	--	15.8	11.0	13.3	11.5	6.2*	-0.5
1988	--	11.7	7.3*	14.6*	12.8	6.6	-0.2
Max	--	20.5	23.9	31.3	27.4	17.7	13.3
Min	--	2.9	-2.9	0.5	-4.2	-4.5	-10.9
Mean	--	13.0	9.1	13.2	11.3	6.4	0.2

Average Relative Humidity (%):

1986	--	--	76.4*	88.1	92.2	89.0	88.7*
1987	--	86.1	80.8	85.5	80.0	89.8*	82.9
1988	--	67.5	72.0*	81.6*	89.1	83.1*	91.7
Mean	--	83.3	76.8	85.3	87.5	87.1	88.2

Total Precipitation (mm):

1986	--	--	3.3*	54.4	47.2	62.5	24.9
1987	--	0.0	40.9	66.8	23.4#	3.0#	1.0#
1988	--	--	--	--	--	--	--
Mean	--	--	32.8	60.7	39.8	34.9	20.5

Average Wind Speed (m/s):

1986	--	--	10.3*	8.4	8.7	9.2	9.4
1987	--	12.8	9.3	8.2	7.6	10.4*	8.9*
1988	--	9.9	9.7*	7.6	8.9	10.7	7.8
Max Hr.	--	21.9	22.7	23.6	25.3	24.2	30.0
Mean	--	10.8	9.6	8.1	8.4	10.1	8.9

Resultant Wind Direction (degrees):

1986	--	--	275.9*	283.8	274.9	266.3	256.8
1987	--	277.0	297.4	77.6	97.3	271.5*	257.1*
1988	--	276.0	286.8*	260.6*	263.9	263.7	231.9
Mean	--	276.3	290.4	200.6	216.2	267.0	250.6

Average Solar Radiation (W/m2):

1986	--	--	225.5*	190.3	154.0	110.1	77.1
1987	--	190.1	189.6	203.3	184.0	100.9*	92.0
1988	--	247.4	248.0*	174.6*	159.0	129.5	46.7
Max	--	934.7	1018.5	955.9	927.9	774.6	669.2
Mean	--	229.6	217.9	191.7	165.0	114.0	71.7

Average Pressure (mB):

1986	--	--	847.7*	848.9	849.2	848.7	845.2
1987	--	849.8	846.4	848.6	852.6	853.3*	849.1
1988	--	848.1	843.5	848.8	846.9	845.1	842.0
Max	--	852.3	856.8	859.2	863.7	865.7	859.4
Min	--	843.5	831.0	833.4	832.6	822.6	821.4
Mean	--	848.7	845.2	848.8	849.4	848.9	845.1

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SITE: WHITETOP

Periods of Record: 1986: Apr 1 - Oct 31
1987: Apr 1 - Oct 31
1988: Apr 1 - Oct 31

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	7.0#	9.8	14.9	16.7	13.9	12.8	7.1
1987	1.0*	12.5	14.4	16.5	15.9	13.4#	4.7*
1988	4.9	9.9	14.1	16.6	16.8	12.5	2.9
Max	20.4	21.4	24.0	25.3	25.3	19.5	18.0
Min	-11.9	-4.7	1.3	5.0	1.2	3.0	-8.6
Mean	4.3	10.7	14.5	16.6	15.6	12.8	4.9

Average Relative Humidity (%):

1986	62.6#	83.4	84.6	86.9	91.3	91.6	86.2
1987	77.6*	81.2	85.2	82.3	85.6	87.3#	65.2*
1988	70.6	67.8	69.1	75.9	81.1	91.2	71.4*
Mean	70.8	77.5	79.8	81.6	85.9	90.6	75.4

Total Precipitation (mm):

1986	48.0	193.0	74.2	90.4	172.7	141.7	95.0
1987	220.2	93.0	152.9	66.0	57.7	85.1	23.6*
1988	121.7	78.2	63.5	103.4	95.8	143.3	83.1
Mean	128.7	121.4	97.6	86.6	108.2	123.4	71.9

Average Wind Speed (m/s):

1986	2.4#	2.5	2.4	1.9	2.3#	2.7#	3.7
1987	3.9*	2.3	2.5	1.9	3.0	3.6#	3.0
1988	3.7	2.8	2.2	2.5	2.9	3.3	--
Max Hr.	14.6	10.2	8.1	7.9	9.5	10.9	13.8
Mean	3.5	2.5	2.4	2.1	2.8	3.2	3.4

Resultant Wind Direction (degrees):

1986	328.4#	207.9	200.2	229.3	191.7#	191.7#	191.7
1987	129.2*	243.6	267.4	283.6	232.7	158.8#	97.1*
1988	177.2	240.7	121.3	241.9	272.2	2.1	188.0
Mean	195.2	230.9	198.0	252.0	241.6	85.3	163.9

Average Solar Radiation (W/m2):

1986	232.8#	176.3	235.5	230.8	179.6	141.2	137.4*
1987	172.8	225.9	228.2	235.3	193.7	151.3	174.6
1988	197.1	247.0	292.1	230.7	216.7	147.7	148.7
Max	1068.7	1083.7	1090.7	1083.8	1013.6	996.0	871.8
Mean	194.1	216.8	251.2	232.3	197.0	146.7	155.1

Average Pressure (mB):

1986	828.8#	--	834.1	835.6	839.9#	838.1	835.4*
1987	826.0	836.3	835.8	838.0	837.7	836.0#	824.3
1988	826.8	833.1	836.2	839.0	838.3	837.4	830.7
Max	839.1	842.2	846.3	845.1	844.1	844.8	846.6
Min	810.3	822.5	824.0	827.0	830.1	822.6	820.0
Mean	826.8	711.0	835.4	837.6	838.1	837.4	829.8

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

SUMMARY OF METEOROLOGICAL DATA
SUBSITE: SHENANDOAH 3

Periods of Record: 1986: May 21 - Nov 23
1987: Apr 24 - Nov 18
1988: Apr 15 - Nov 1

Average Temperature (C):

	APR	MAY	JUN	JUL	AUG	SEP	OCT
1986	--	18.5	22.4	24.0#	20.7	19.4	14.1
1987	11.4	18.3	21.8	23.6	22.1	18.3	9.6*
1988	11.4	17.6	21.4	25.0	25.0	19.0	10.4
Max	25.8	31.2	34.9	38.4	37.5	31.5	30.5
Min	2.6	3.6	6.6	9.2	7.1	6.0	0.5
Mean	11.4	18.0	21.9	24.3	22.6	18.9	11.6

Average Relative Humidity (%):

1986	--	68.3	61.9	74.9#	75.9	70.5	60.4
1987	52.5	64.7	62.3	63.9	63.7	71.1	52.5*
1988	44.9	64.8	59.6	65.9	67.9	71.1	57.0
Mean	47.3	65.3	61.3	66.5	69.3	70.9	57.2

Total Precipitation (mm):

1986	--	--	--	--	180.8*	29.0	22.4
1987	0.0	123.7	101.6	45.0	31.5	225.0	33.3*
1988	10.9	204.0	35.3	85.1	75.9	80.3	34.5
Mean	7.5	163.9	69.0	65.1	93.3	110.7	29.6

Average Wind Speed (m/s):

1986	--	1.3	1.6	1.6#	1.6	1.5	1.5
1987	2.1	1.6	1.5	1.4	1.7	1.5	1.9*
1988	2.4	1.8	1.7	1.4	1.5	1.5	1.8
Max Hr.	6.3	5.7	5.7	4.9	5.5	5.3	5.5
Mean	2.3	1.6	1.6	1.4	1.6	1.5	1.7

Resultant Wind Direction (degrees):

1986	--	187.4	170.3	181.2#	182.4	179.3	170.8
1987	359.6	179.8	157.3	166.4	166.6	165.6	91.3*
1988	13.1	169.8	159.2	168.0	166.8	166.1	126.4
Mean	119.9	176.6	162.3	169.5	172.1	170.6	135.2

Average Solar Radiation (W/m2):

1986	--	234.7	269.6	343.4#	224.6	170.5	136.2
1987	209.6	223.8	255.3	243.0	220.0	159.9	142.3*
1988	205.1	227.4	262.3	233.0	221.7	177.4	140.4
Max	957.3	1041.1	1054.4	982.1	973.2	911.3	835.3
Mean	206.5	226.9	262.4	250.4	222.0	169.1	139.2

Average Pressure (mB):

1986	--	958.5	959.8	961.2#	962.6	964.8	964.4
1987	959.2	964.1	961.6	963.7	963.7	963.1	963.5*
1988	954.6	961.8	963.2	965.6	965.0	966.1	965.4
Max	968.1	970.7	975.5	974.7	973.2	977.9	980.2
Min	942.1	950.4	951.0	953.4	954.6	948.1	947.9
Mean	956.0	962.3	961.5	964.1	963.7	964.6	964.6

-- No Data, * Data Recovery 50% to 85%, # Data Recovery < 50%

APPENDIX D

MCCP Publications

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APPENDIX E

ADDITIONAL FIGURES

Average Annual Low (<2500 m) Cloud Amounts (%) in Eastern North America, For Land Areas Only, 1971 - 1981, (from Warren et al., 1986). The Time-Averaged Cloud Amount is the Product of Frequency of Occurrence and Amount-When-Present. Values Represent Stratus/ Stratocumulus, Cumulus and Cumulonimbus Cloud Types.

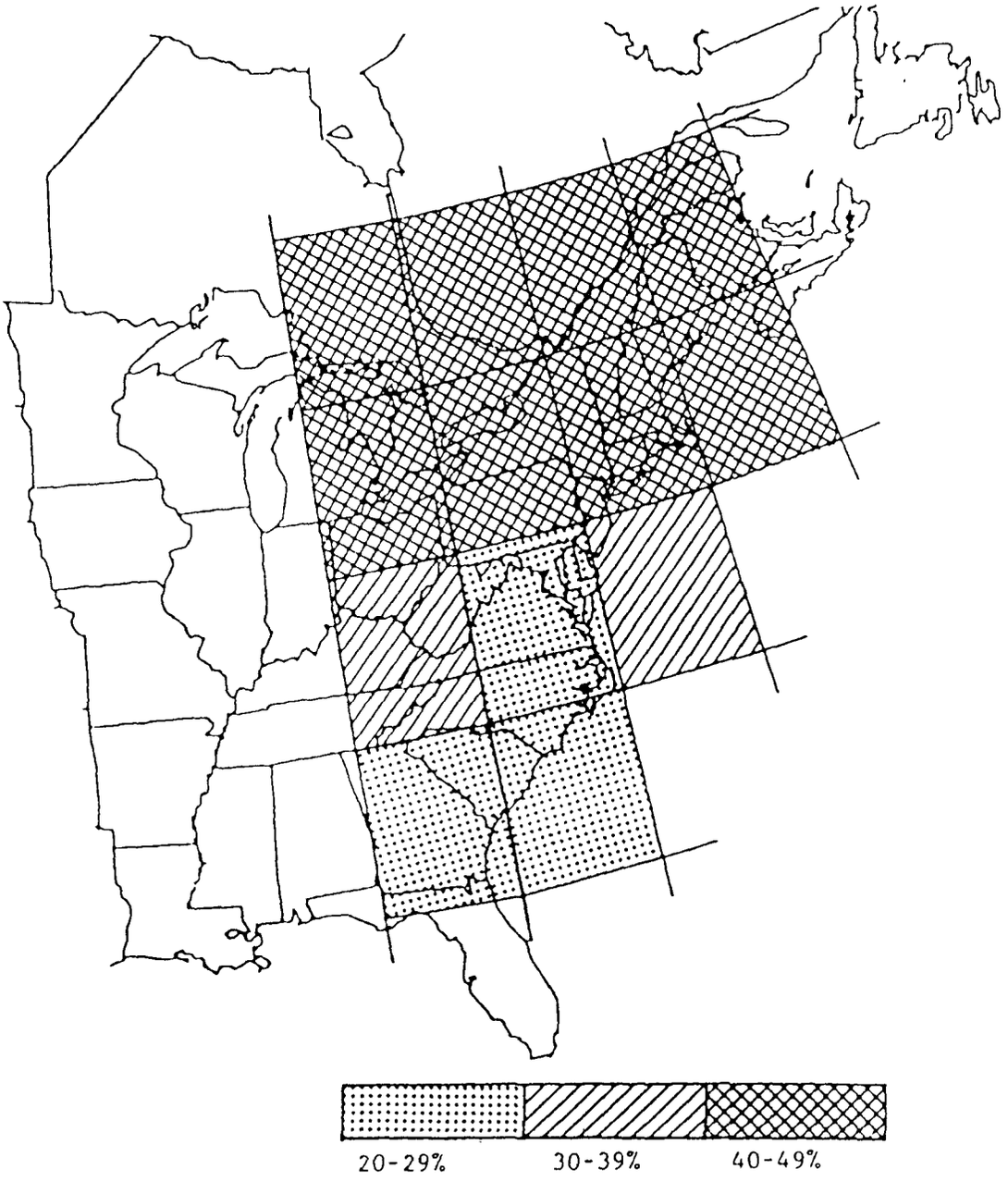


Figure 4-9

Average Cloud Base Height (m) in Eastern North America for Stratus/Stratocumulus Clouds, the Dominant Low Cloud Type, For Land Areas Only, 1971-81 (From Warren et al., 1986)

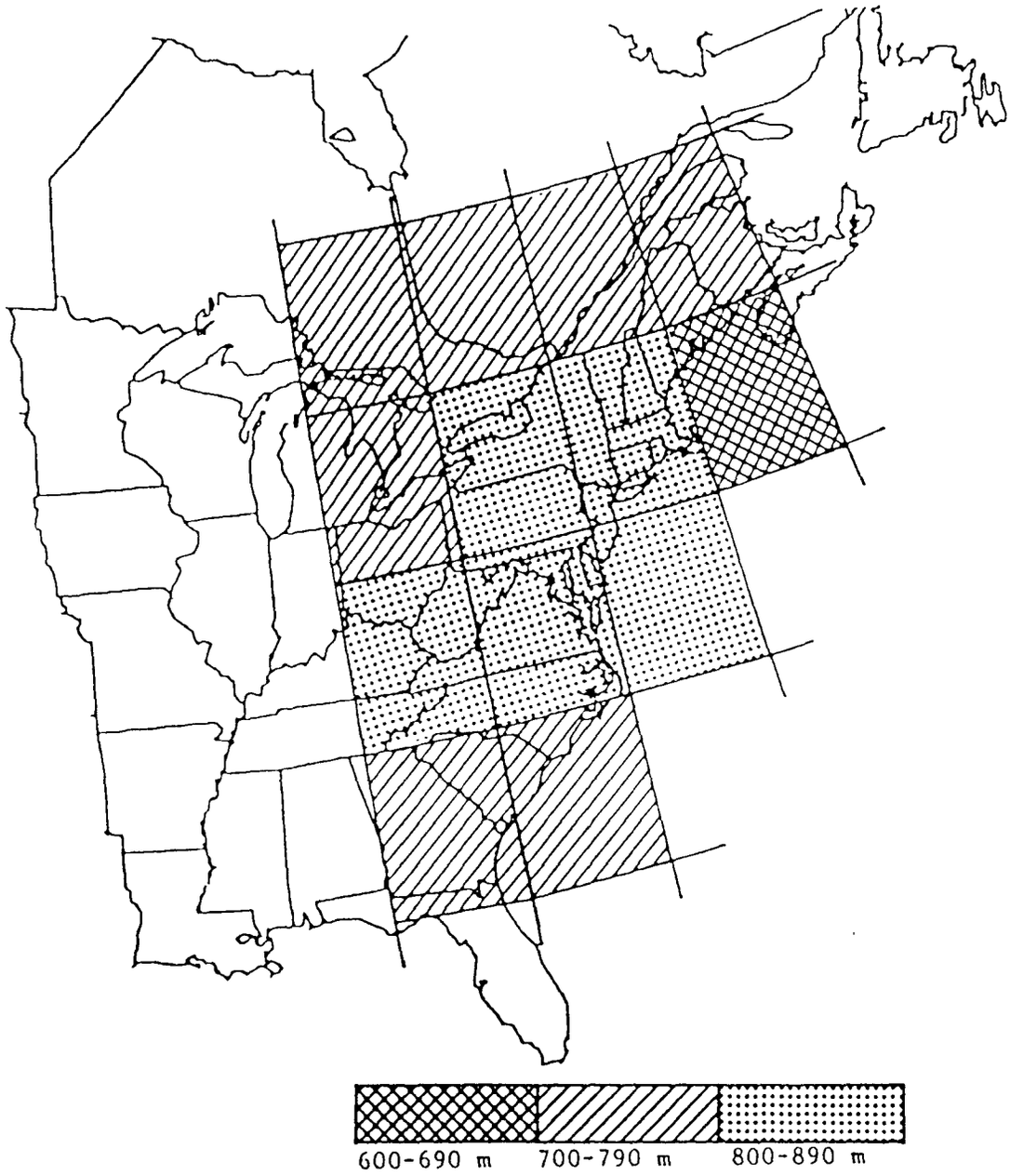


Figure 4-10

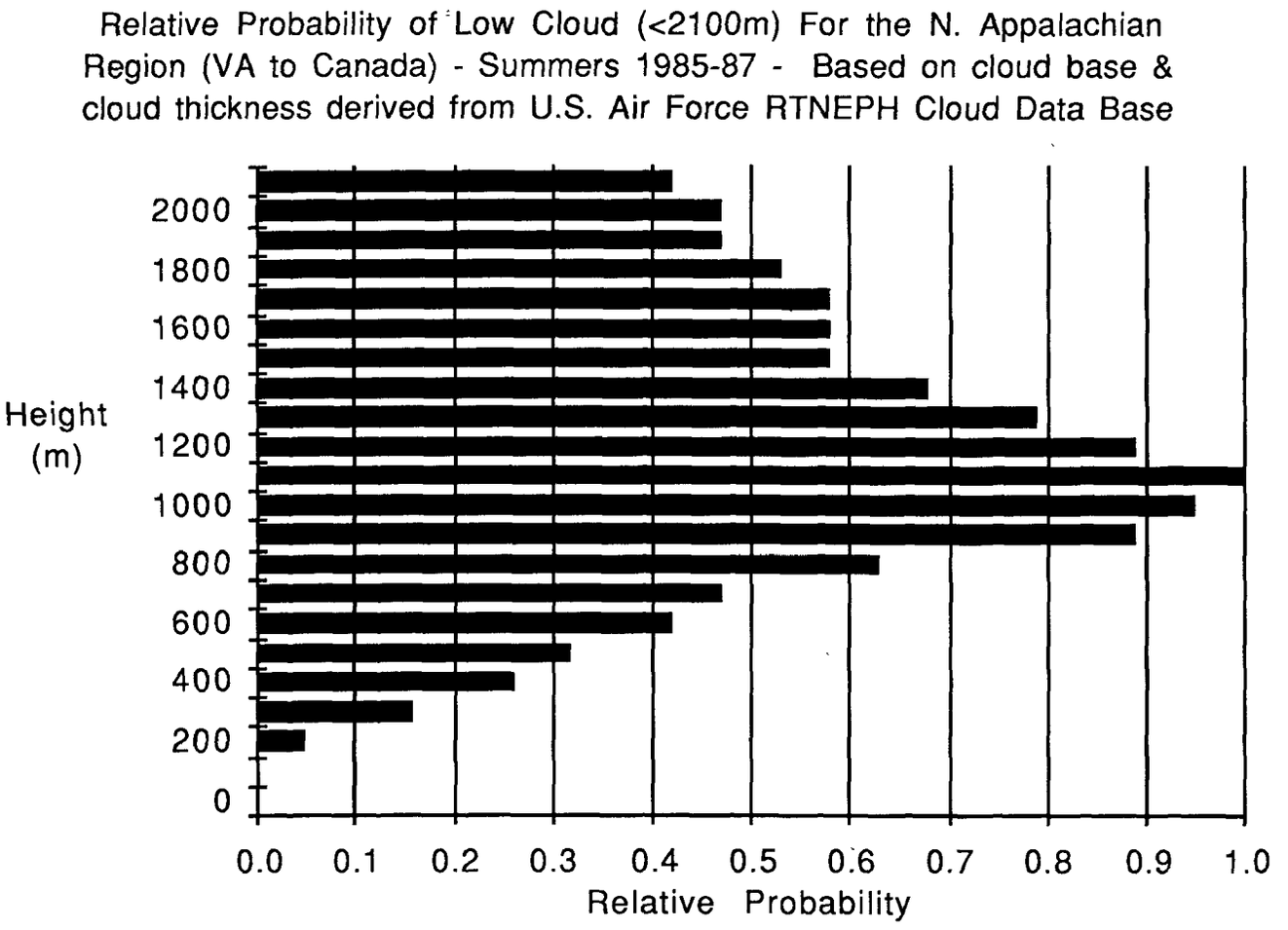

Relative Probability of Low Cloud (<2100m) For the N. Appalachian Region (VA to Canada) - Summers 1985-87 - Based on cloud base & cloud thickness derived from U.S. Air Force RTNEPH Cloud Data Base
Height (m)
2000
1800
1600
1400
1200
1000
800
600
400
200
0
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
Relative Probability

Figure 4-12

Probability of Cloud by Height and Season
at Moosilauke, NH, (1985-1987)

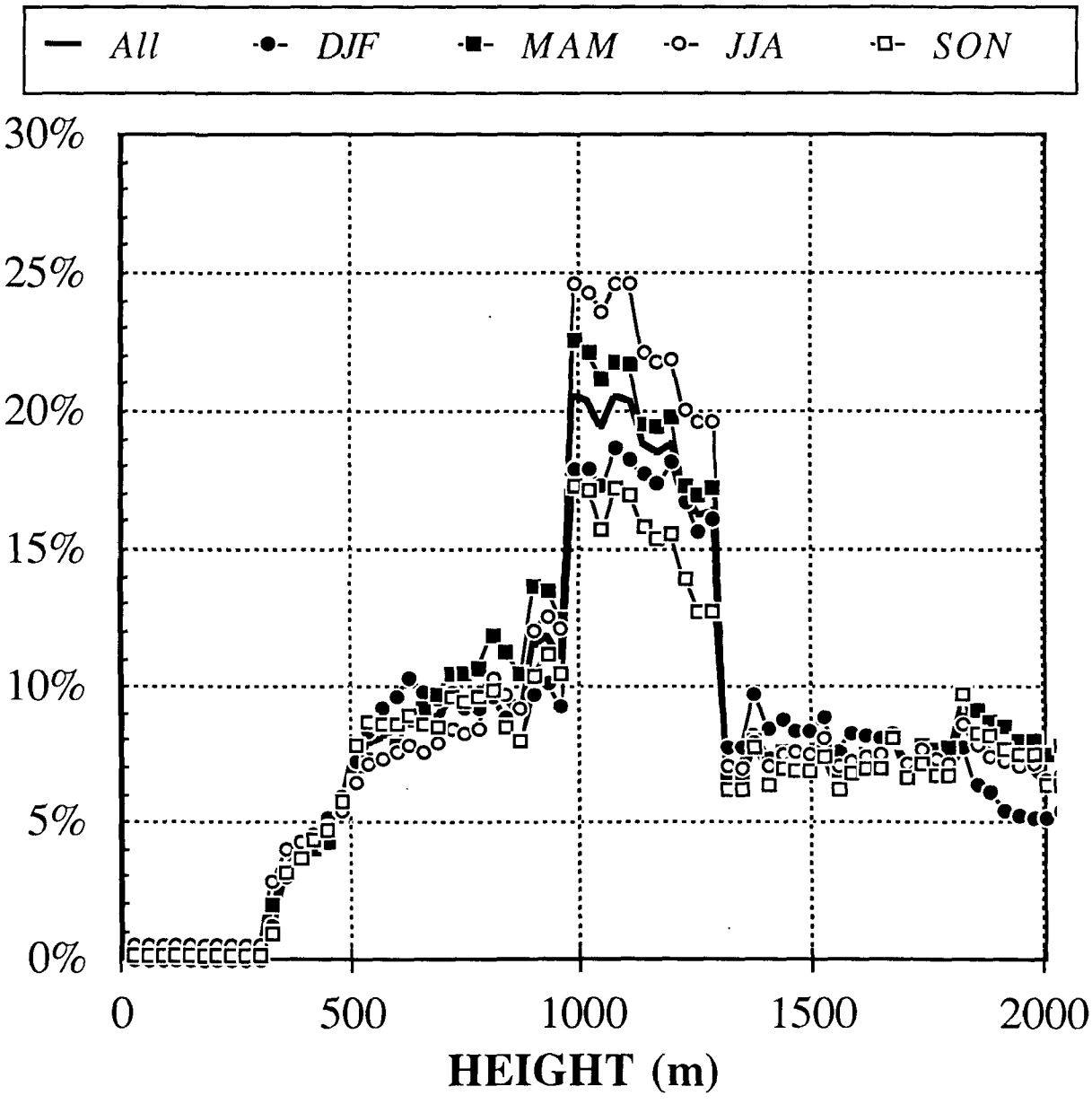


Figure 4-13

Probability of Cloud by Height and Season
at Shenendoah, VA, (1985-1987)

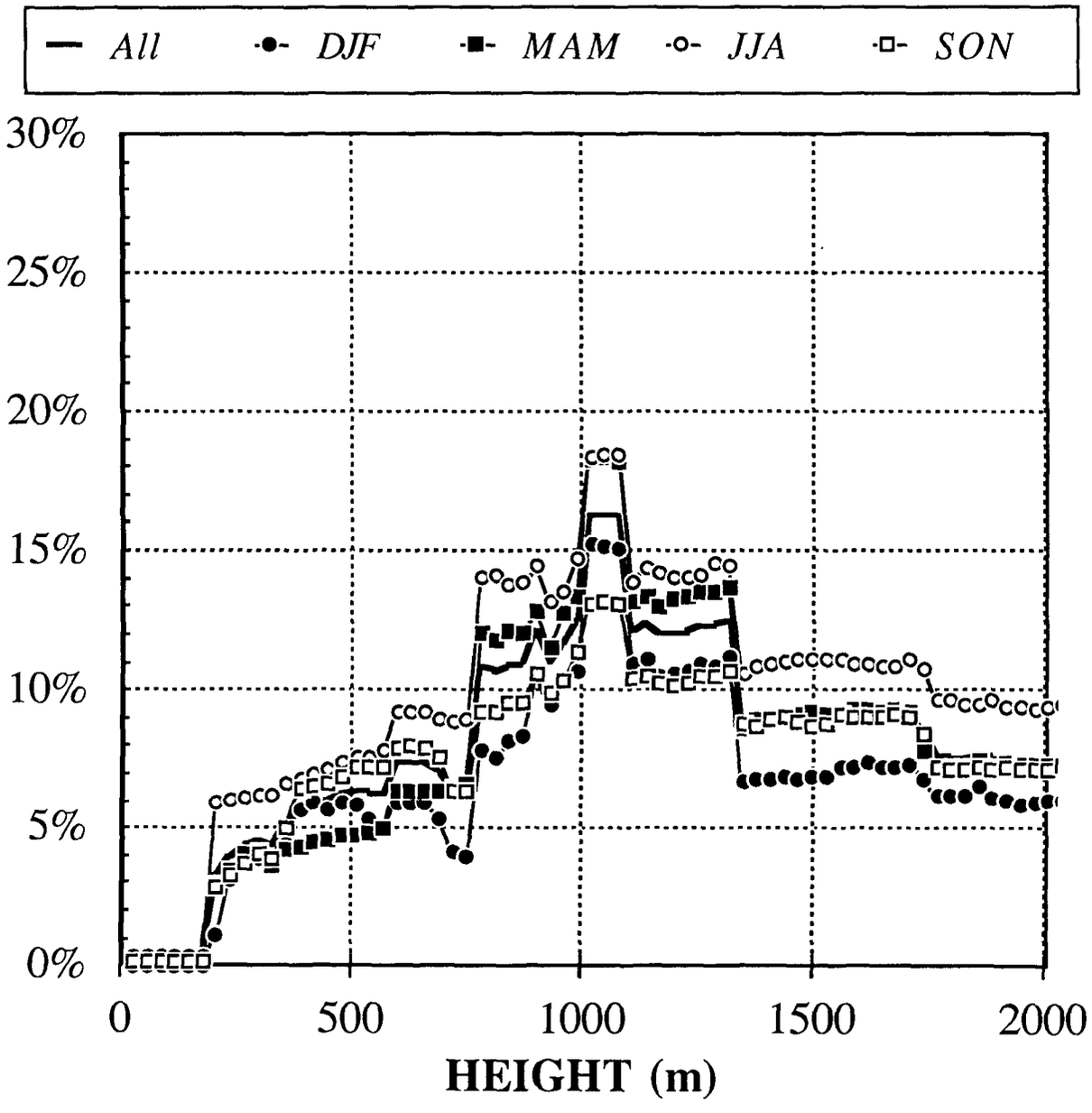


Figure 4-14

Probability of Cloud by Height and Season
at Whiteface Mountain, NY, (1985-1987)

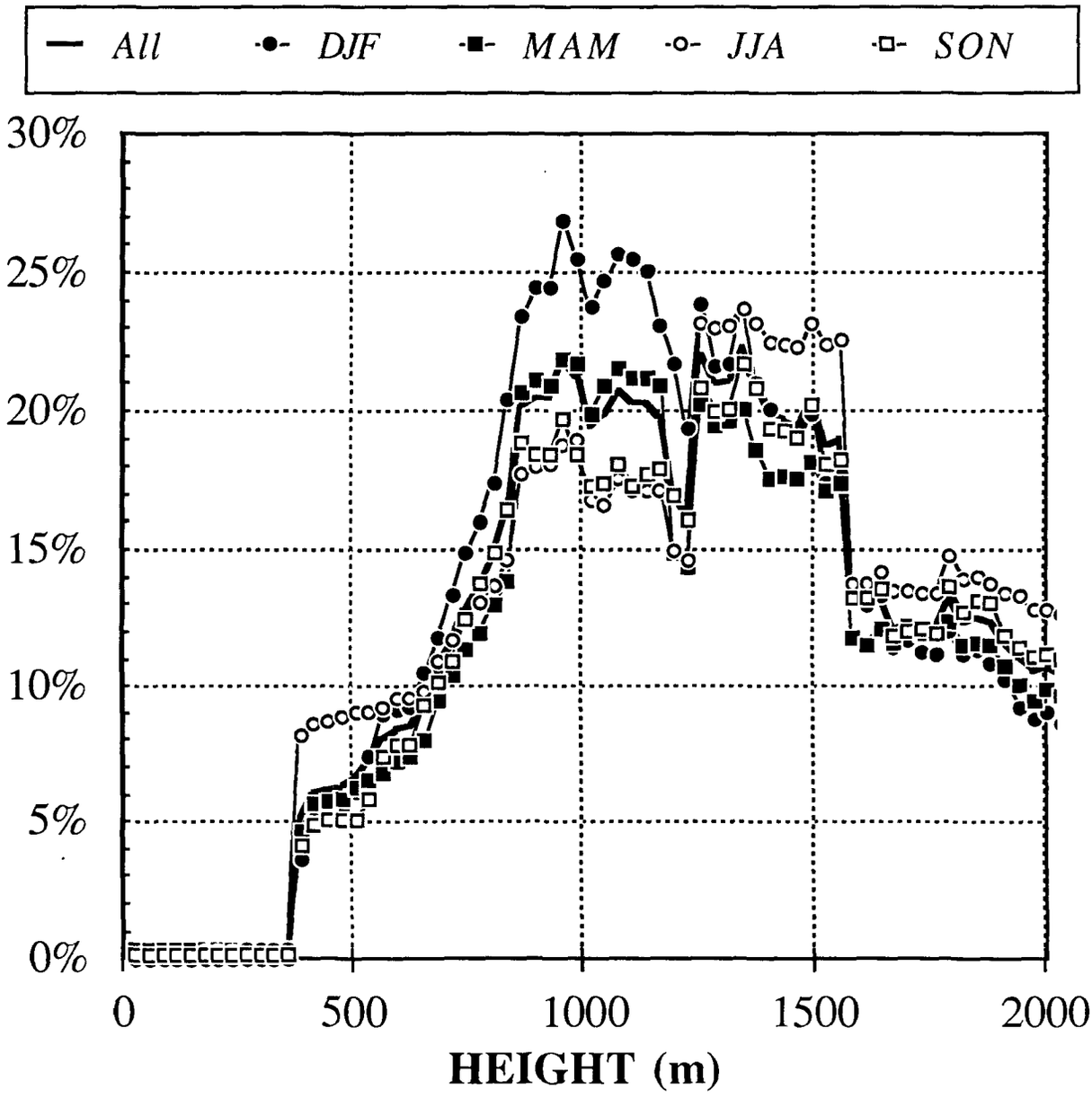


Figure 4-15

Low-level Cloud Frequency Departure (%) From
1965-85 Normals For 8 NWS Airports

1986 Field Season

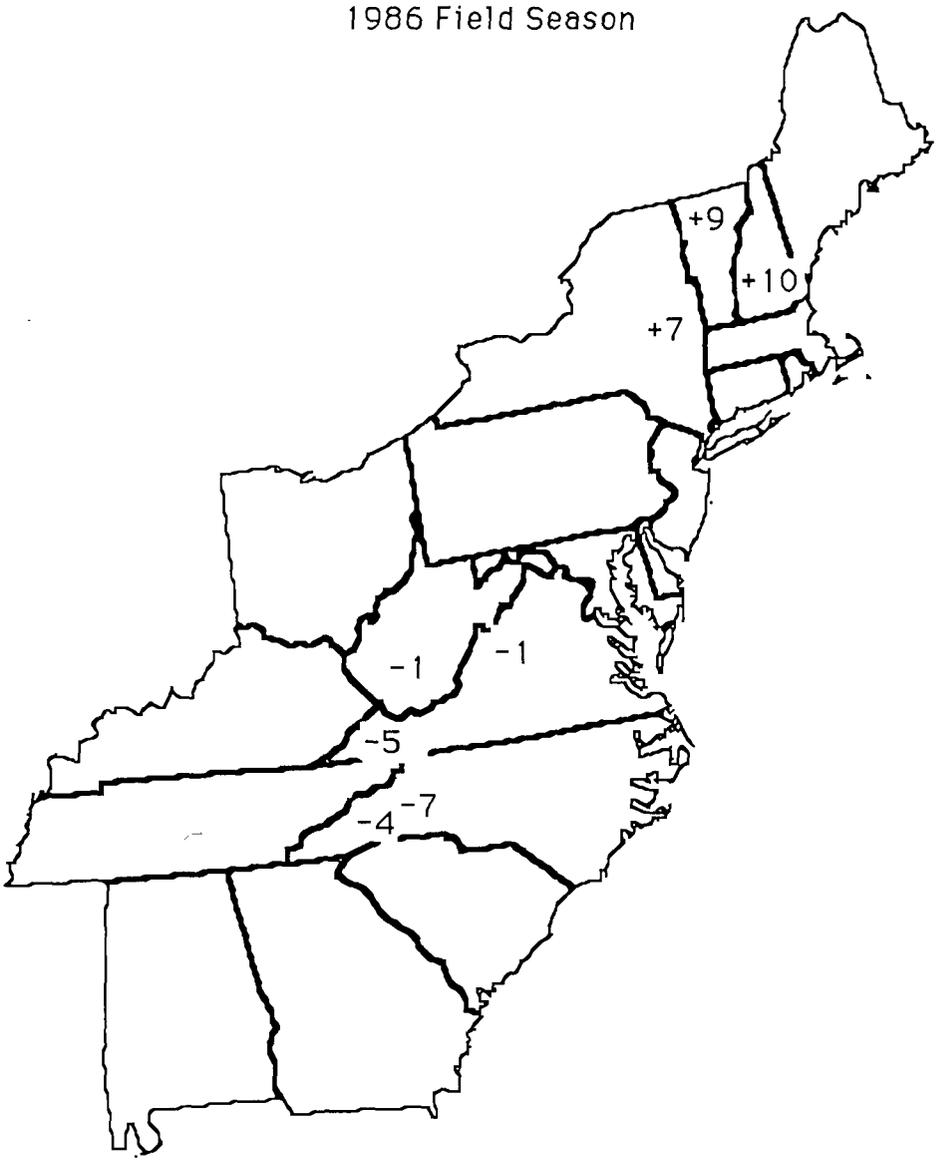


Figure 4-17

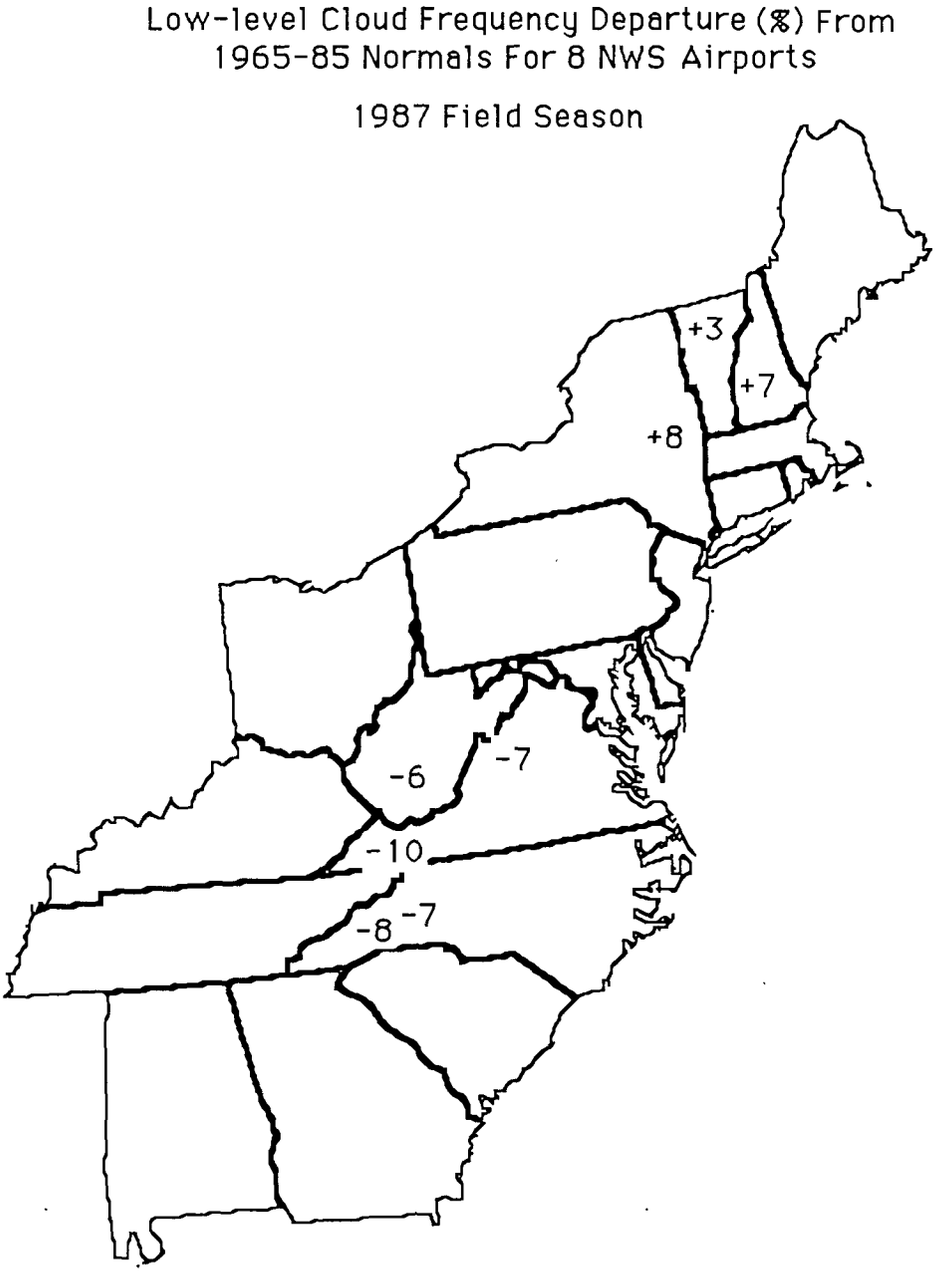
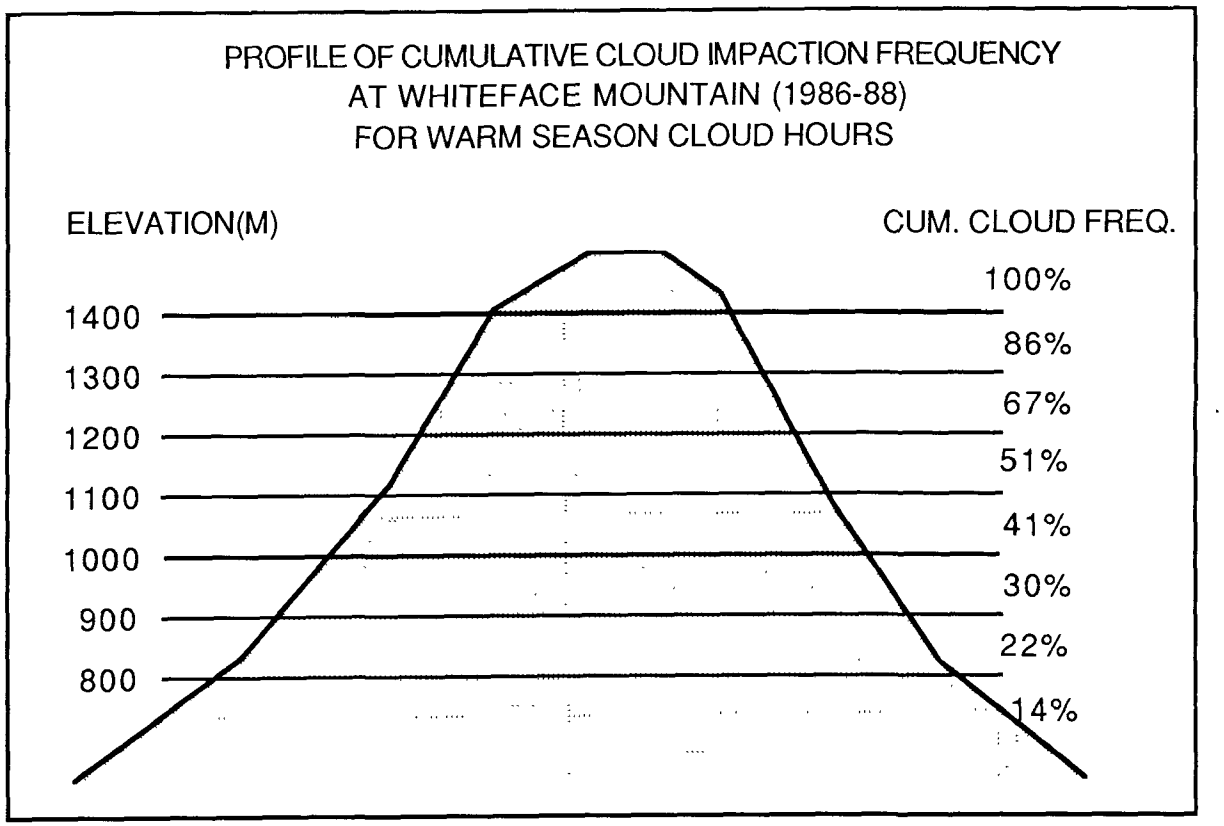

Low-level Cloud Frequency Departure (%) From
1965-85 Normals For 8 NWS Airports
1987 Field Season
+3
+7
+8
-6
-7
-10
-8
-7

Figure 4-18

Low-level Cloud Frequency Departure (%) From
1965-85 Normals For 8 NWS Airports

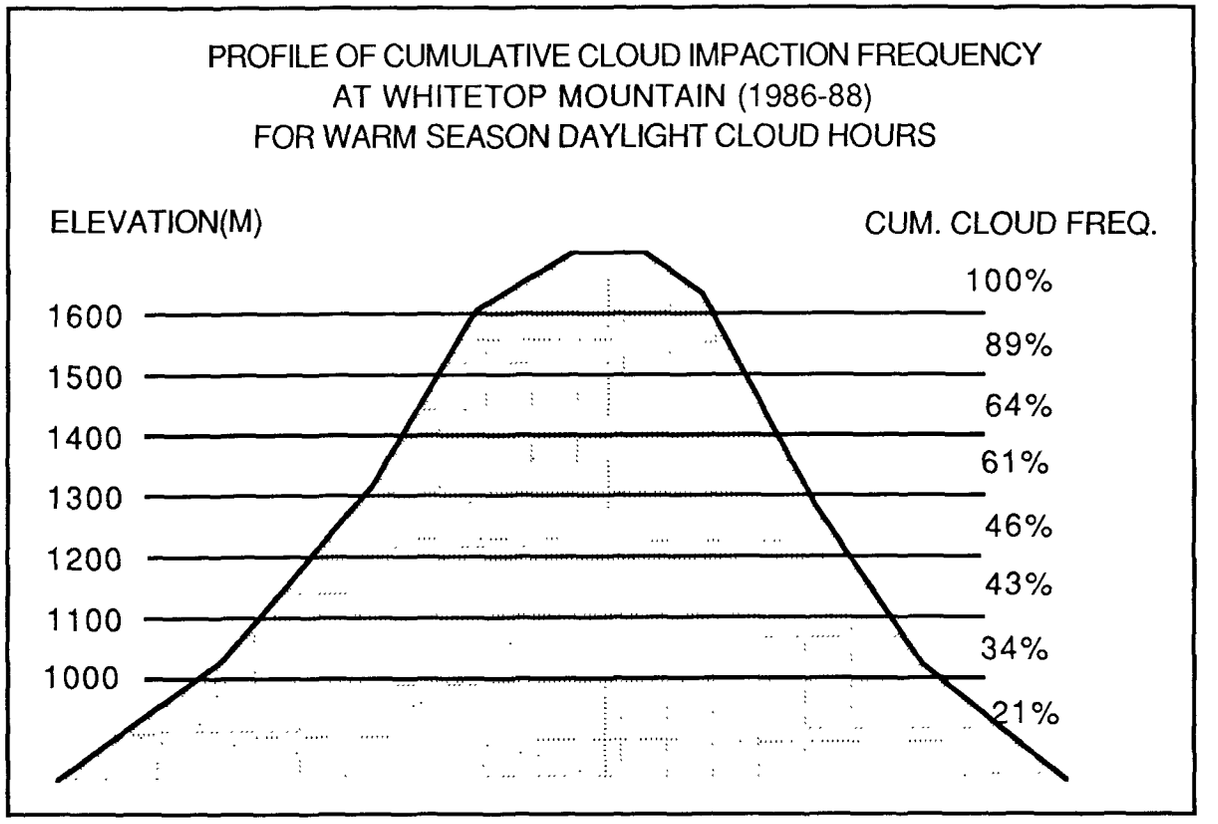
1988 Field Season

Figure 4-19



Total cloud impaction frequency at the summit=37% of all hours

Figure 4-20



Total cloud impaction frequency at the summit=28% of all hours

Figure 4-21

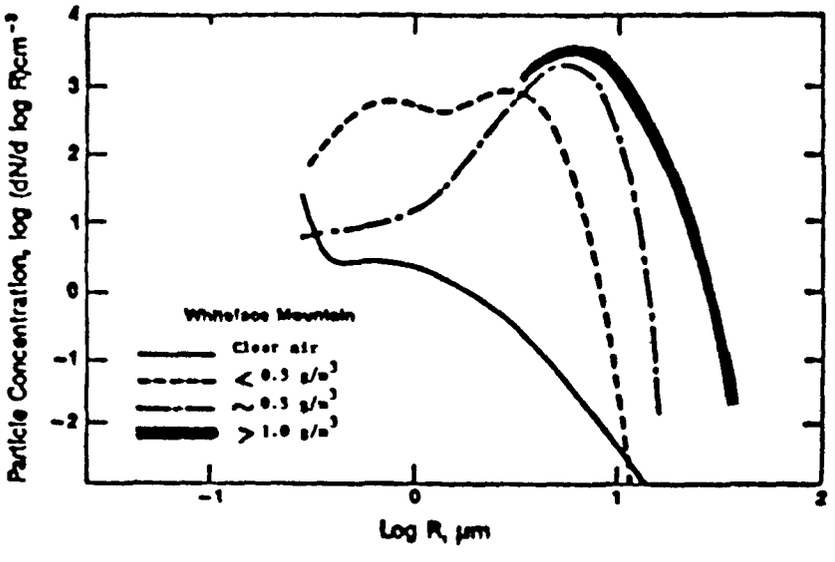
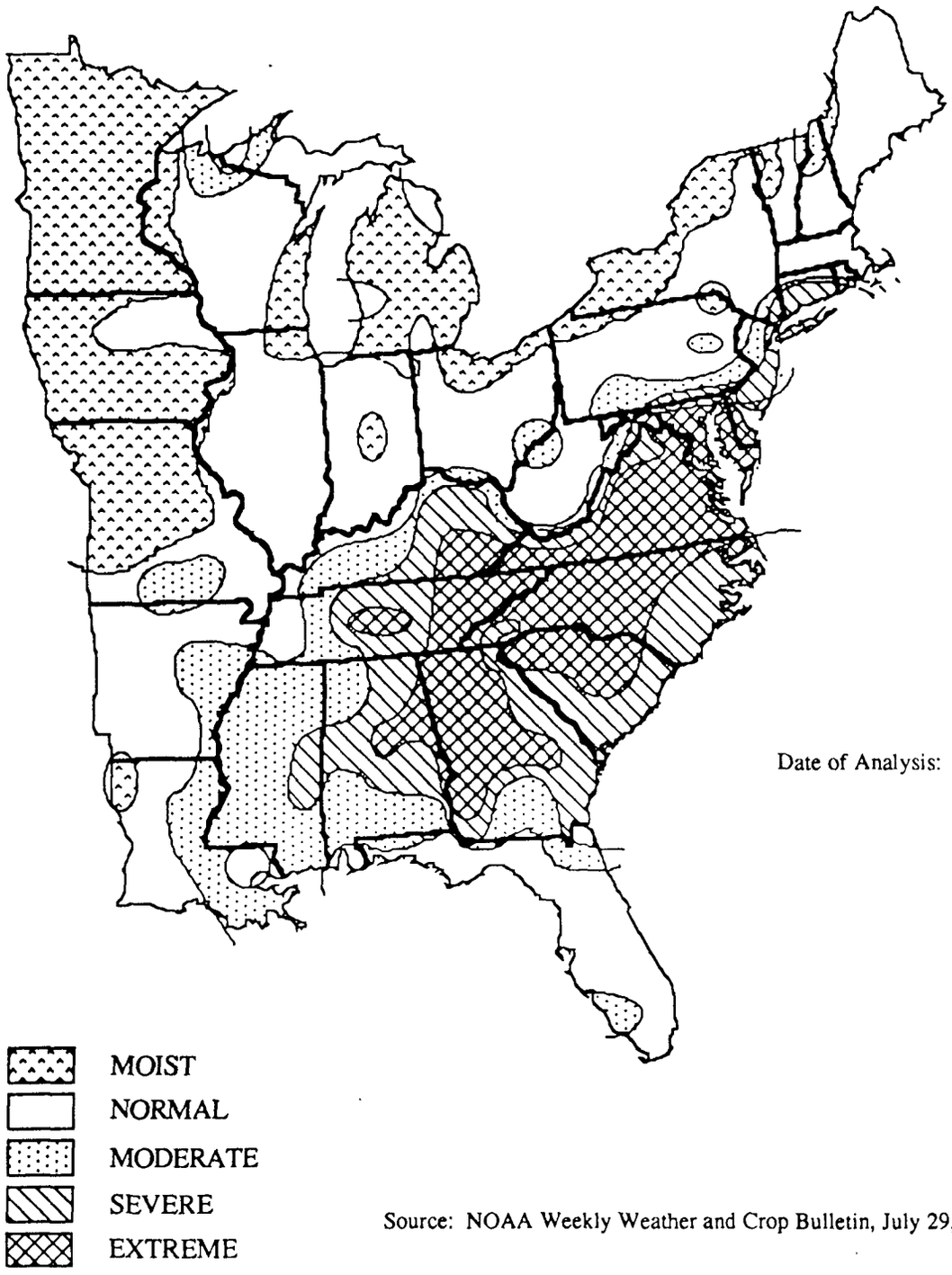


Figure 4-23 Cloud droplet size distributions from Whiteface Mountain, NY calculated by integrating over individual size distributions and grouping the distributions into three categories by liquid water content (LWC < 0.5 g/m^3, 0.5 < LWC < 1.0 g/m^3, and LWC > 1.0 g/m^3), and clear air (Mohnen, 1987).

DROUGHT SEVERITY (Long Term Palmer) OVER THE EASTERN U.S.

MID-SUMMER 1986



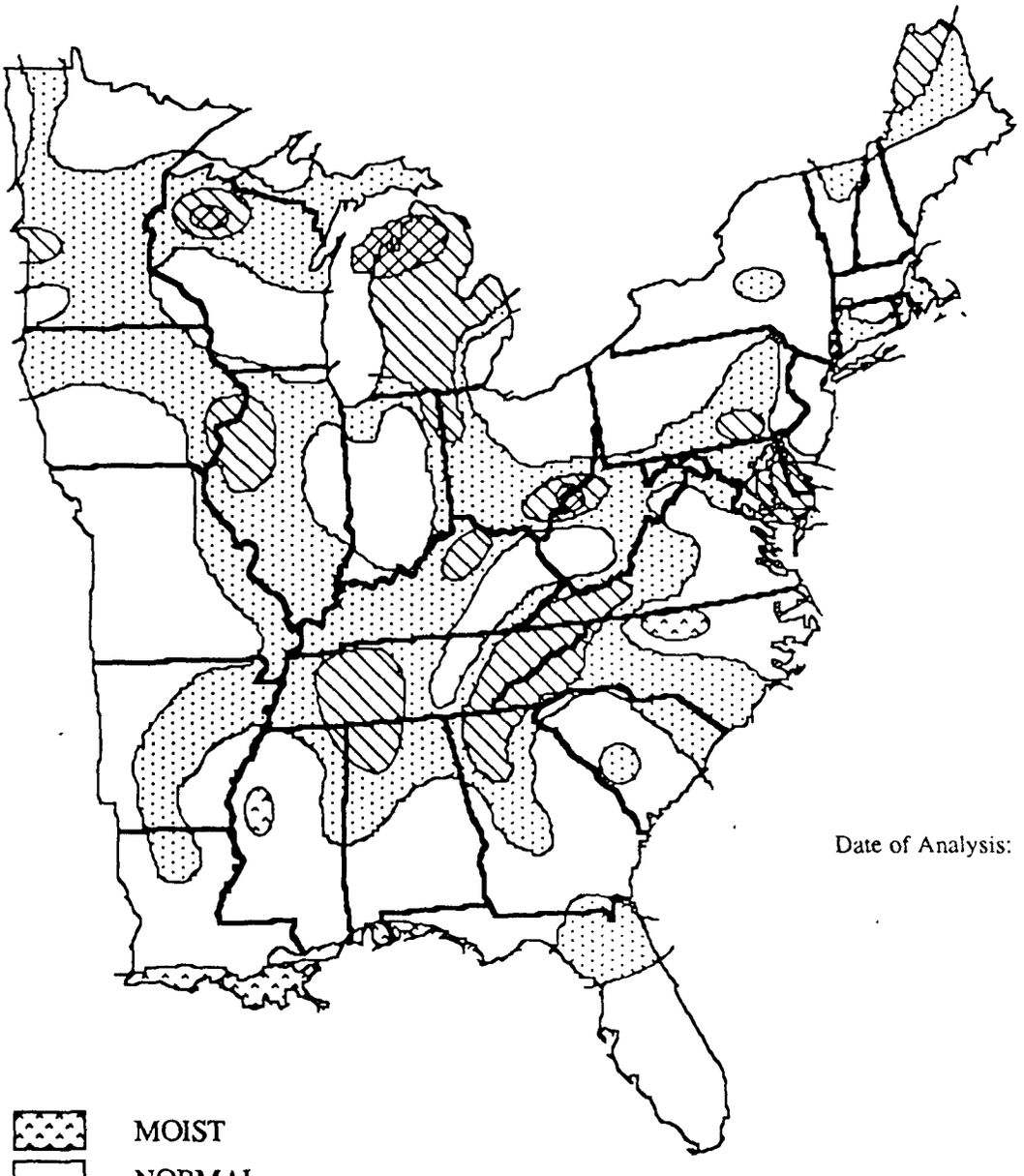
Date of Analysis: July 26

Source: NOAA Weekly Weather and Crop Bulletin, July 29, 1986.

Figure 4-25

DROUGHT SEVERITY (Long Term Palmer) OVER THE EASTERN U.S.

MID-SUMMER 1987



Date of Analysis: August 1

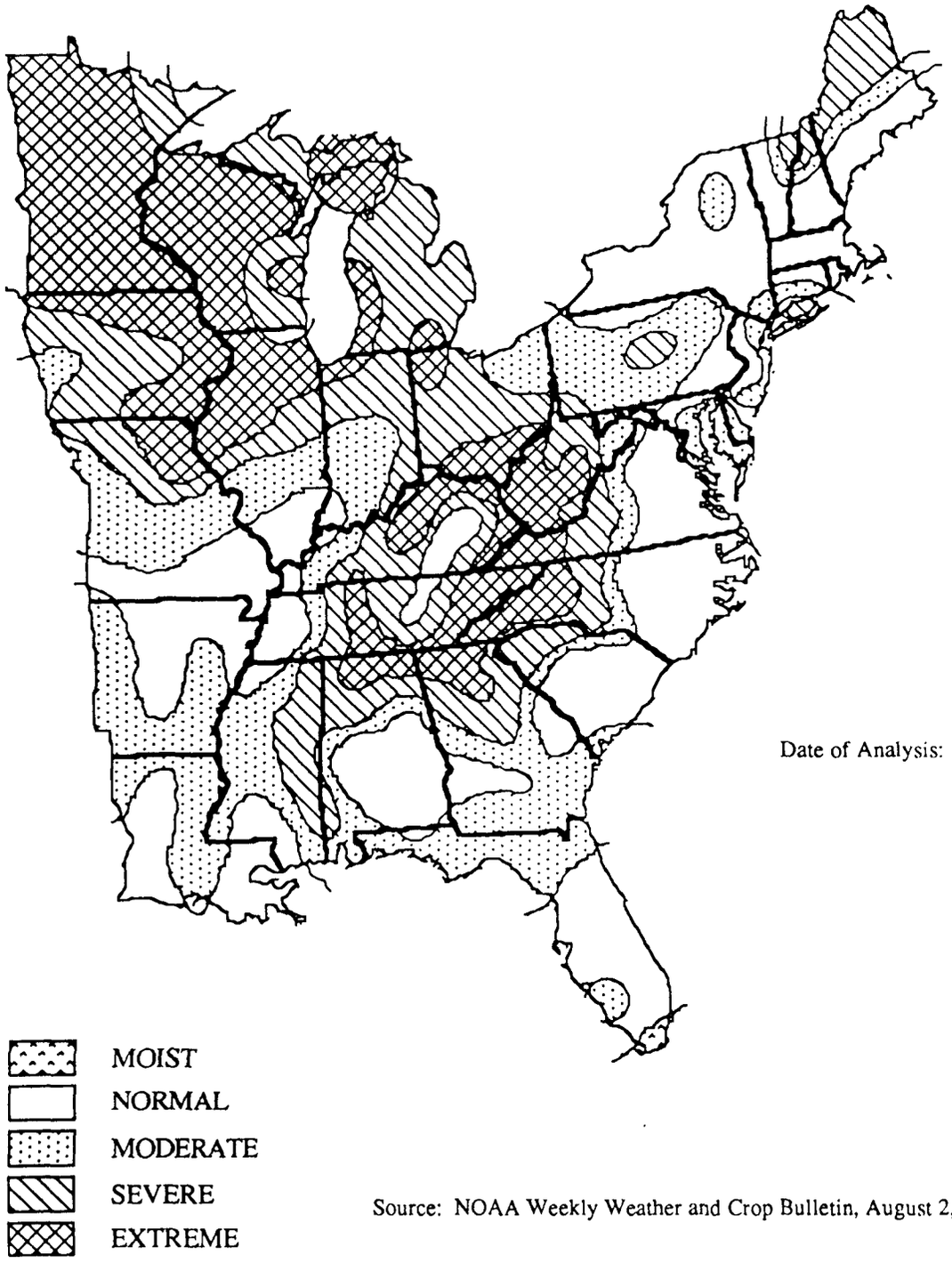
MOIST
NORMAL
MODERATE
SEVERE
EXTREME

Source: NOAA Weekly Weather and Crop Bulletin, August 4, 1987.

Figure 4-26

DROUGHT SEVERITY (Long Term Palmer) OVER THE EASTERN U.S.

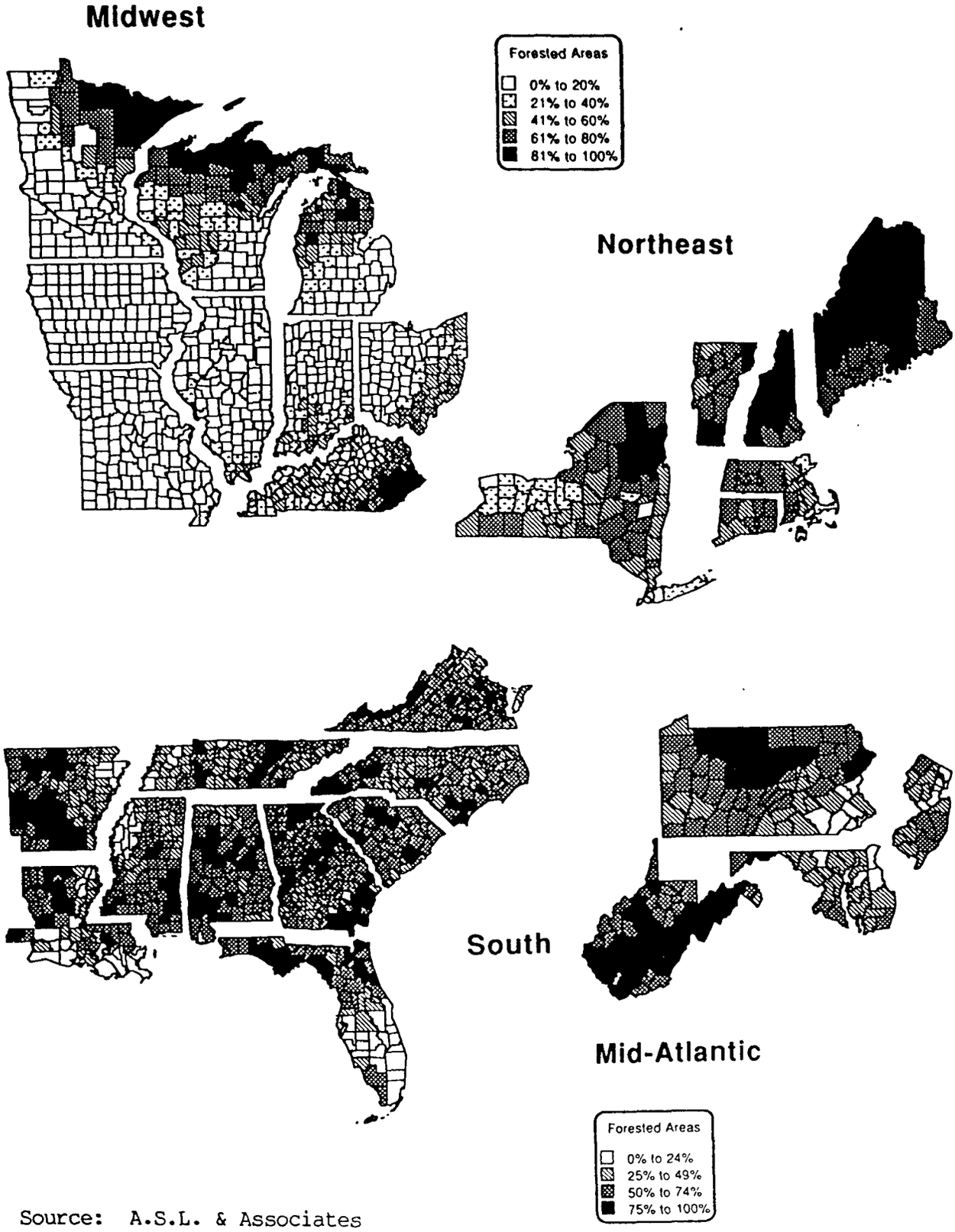
MID-SUMMER 1988



Date of Analysis: July 30

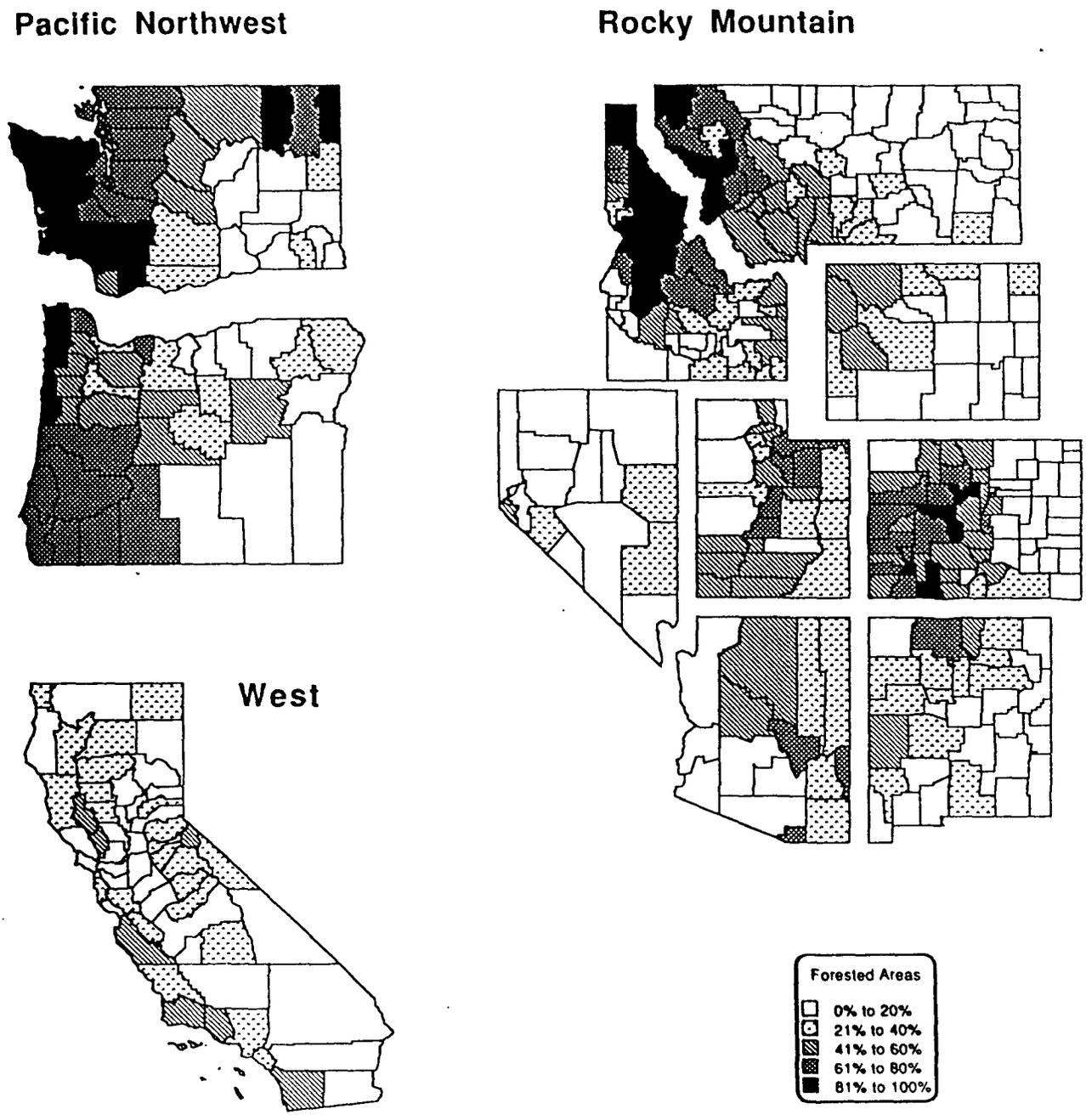
Source: NOAA Weekly Weather and Crop Bulletin, August 2, 1988.

Figure 4-27



Source: A.S.L. & Associates
Helena, MT

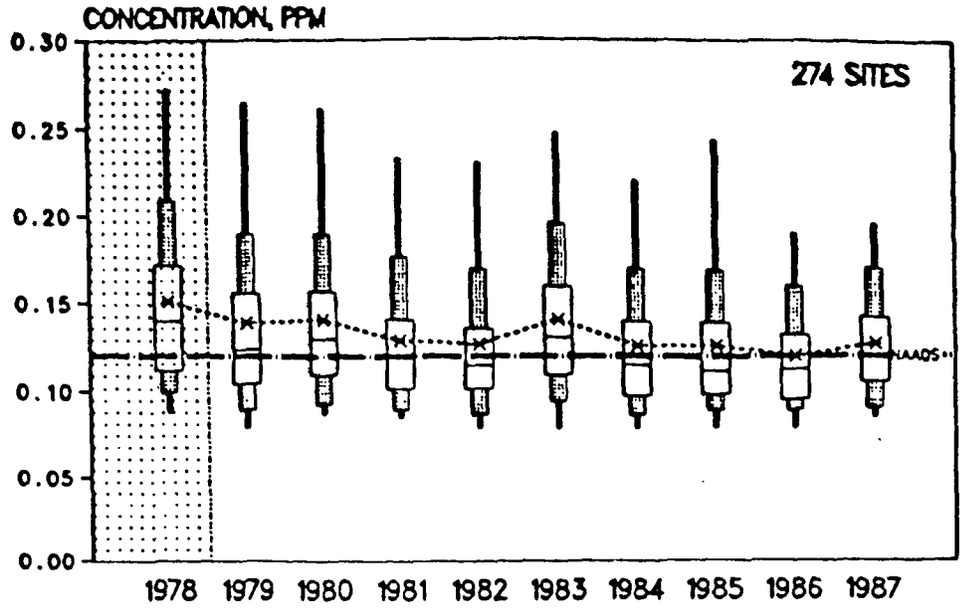
Figure 4-29 Percent forest cover by county (Regions: Midwest, Northeast, South, and Mid-Atlantic).



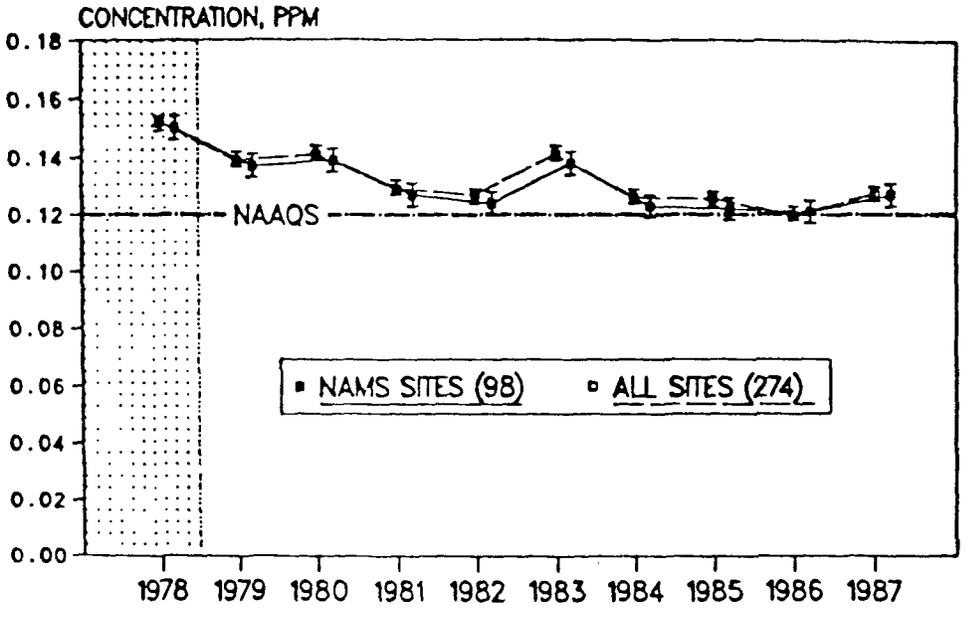
Source: A.S.L. & Associates, Helena, MT

Figure 4-30 Percent forest cover by county (Regions: Pacific Northwest, Rocky Mountain, and West).

Source: EPA (1989)



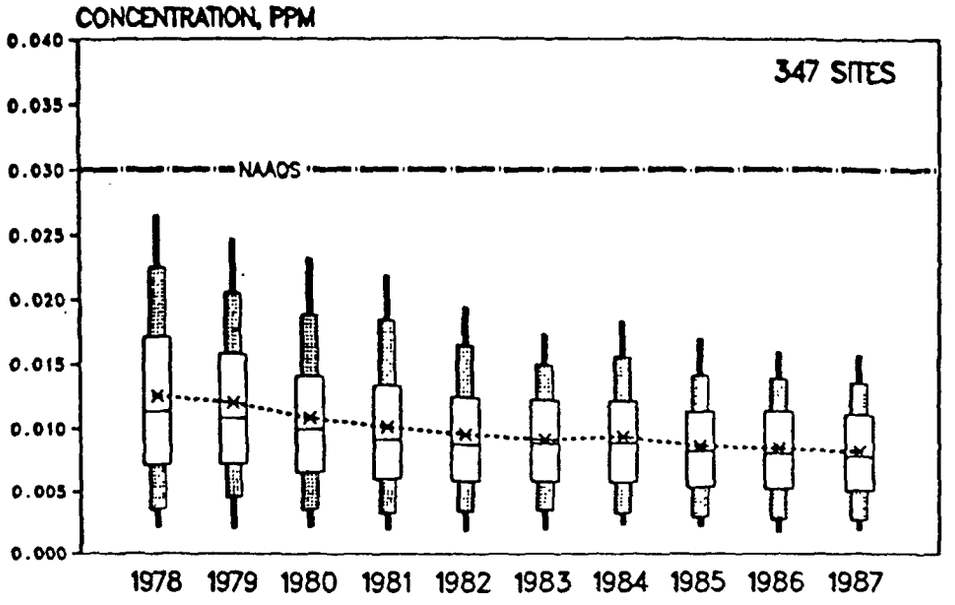
Boxplot comparisons of trends in annual second highest daily maximum 1-hour ozone concentration at 274 sites, 1978-1987.



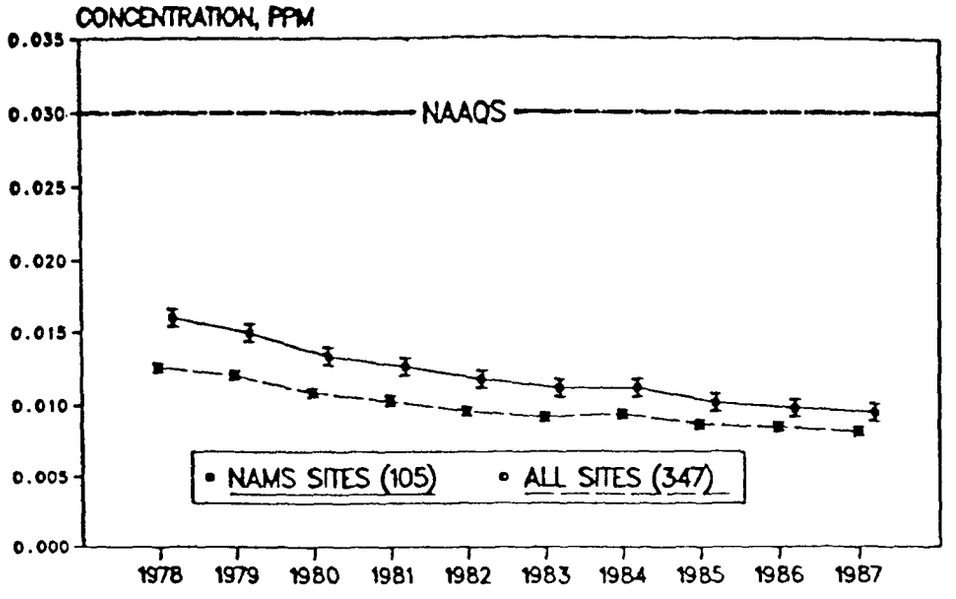
National trend in the composite average of the second highest maximum 1-hour ozone concentration at both NAMS and all sites with 95 percent confidence intervals, 1978-1987.

Figure 4-31

Source: EPA (1989)



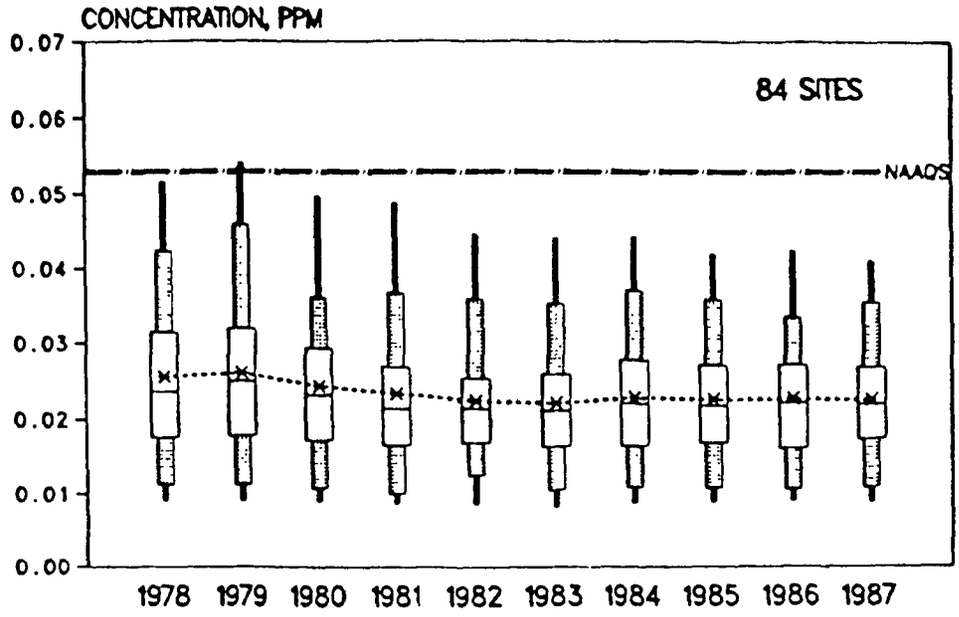
Boxplot comparisons of trends in annual mean sulfur dioxide concentrations at 347 sites, 1978-1987.



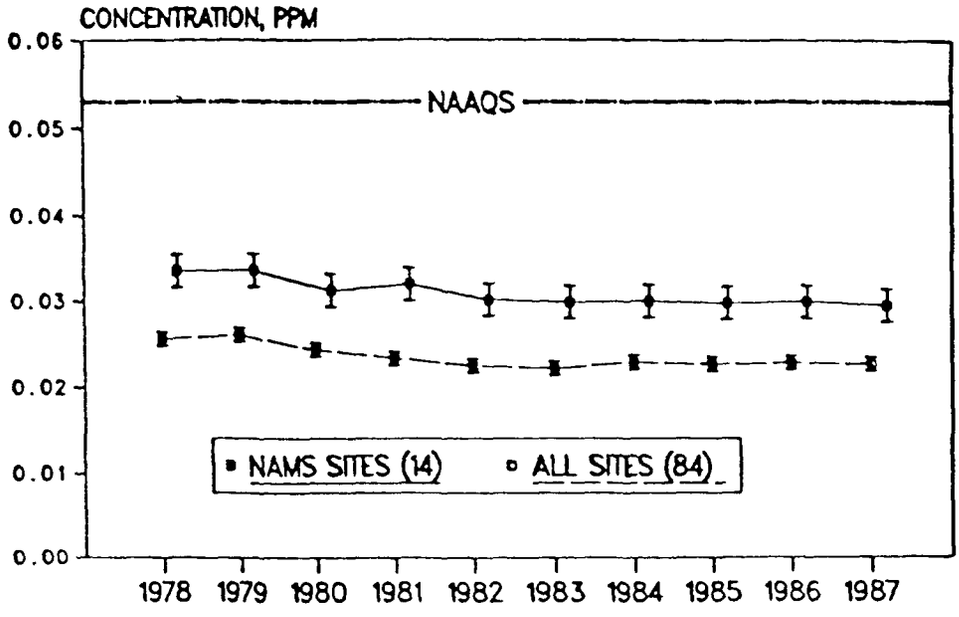
National trend in the composite average of the annual average sulfur dioxide concentration at both NAMS and all sites with 95 percent confidence intervals, 1978-1987.

Figure 4-32

Source: EPA (1989)



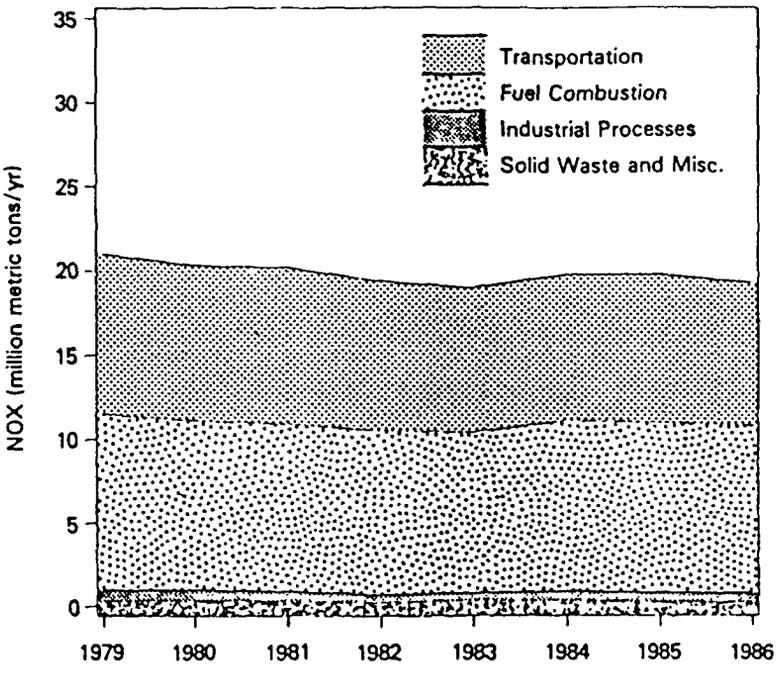
Boxplot comparisons of trends in annual mean nitrogen dioxide concentrations at 84 sites, 1978-1987.



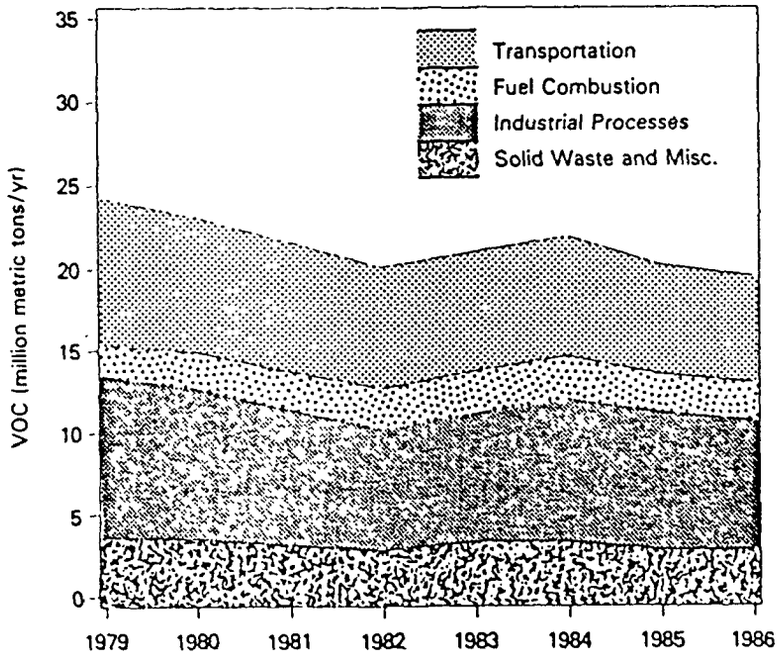
National trend in the composite average of nitrogen dioxide concentration at both NAMS and all sites with 95 percent confidence intervals, 1978-1987.

Figure 4-33

Source: EPA (1989)



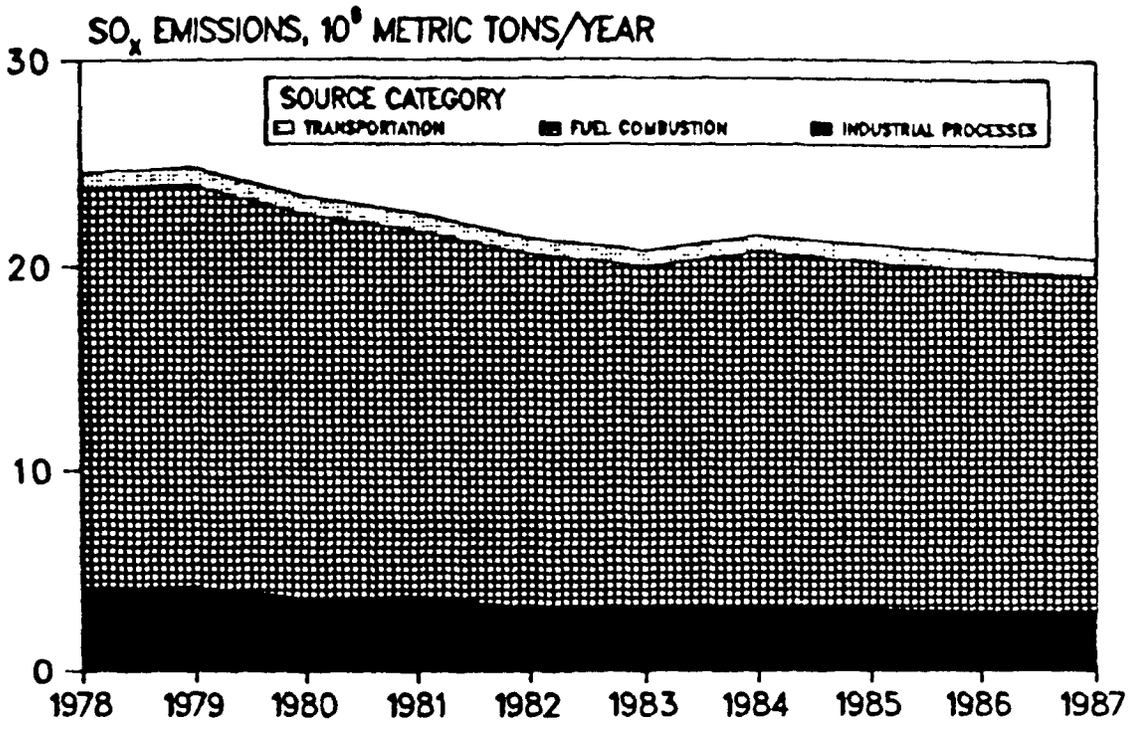
National Trend in Nitrogen Oxide Emissions, 1979-1986.



National Trend in Volatile Organic Compound Emissions, 1979-1986.

Figure 4-34

Source: EPA (1989)



National trend in sulfur oxide emissions, 1978-1987.

Figure 4-35

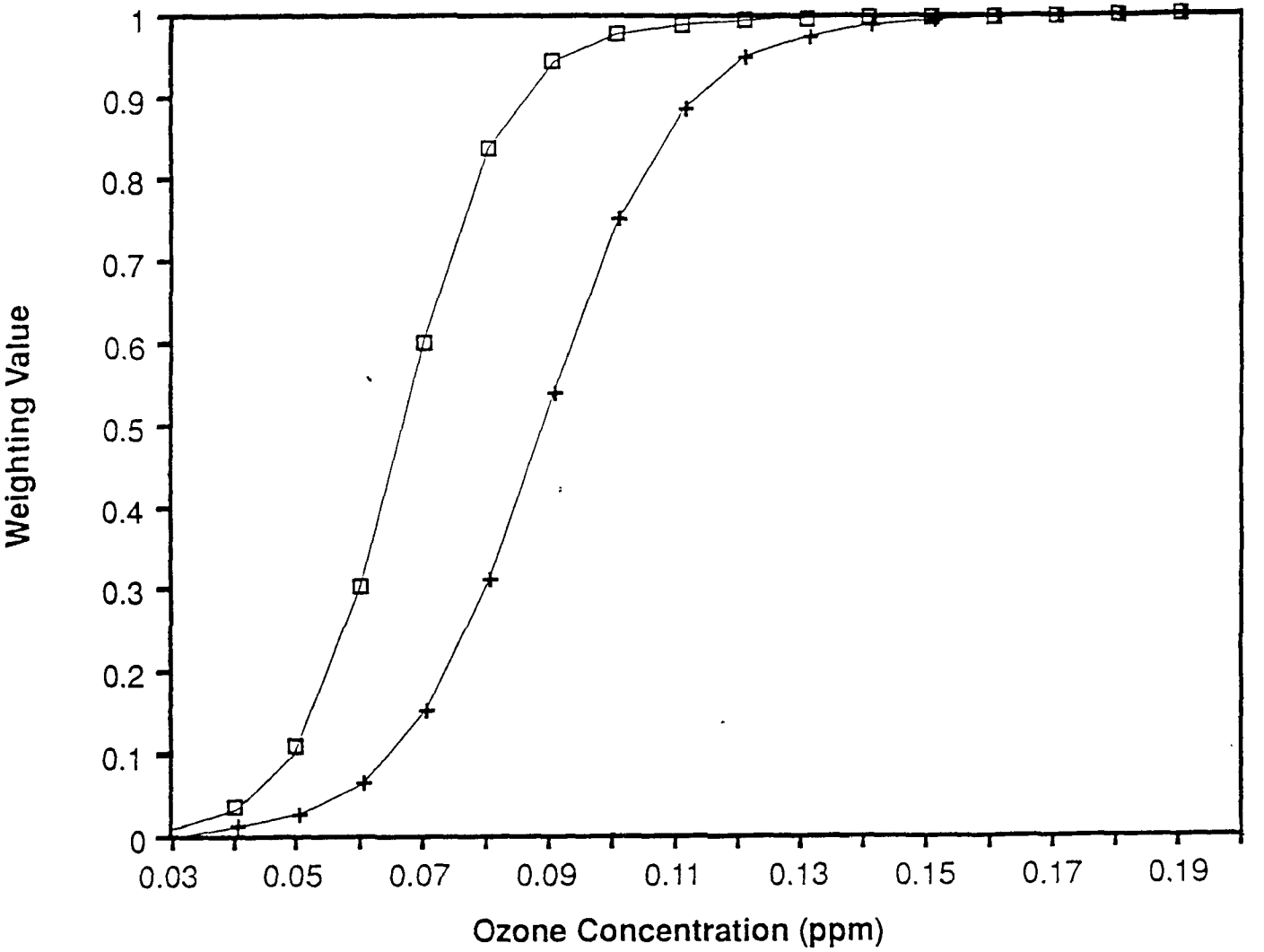


Figure 4-36 Examples of two sigmoidal weighting functions. The W95 (+) weighting focuses on the Higher hourly average concentrations, while the W126 ( ) weighting focuses on both the higher and lower hourly ozone concentrations. The W126 weighting was used in this analysis to calculate the sigmoidal integrated exposure index.
Source: Lefohn et al., 1988

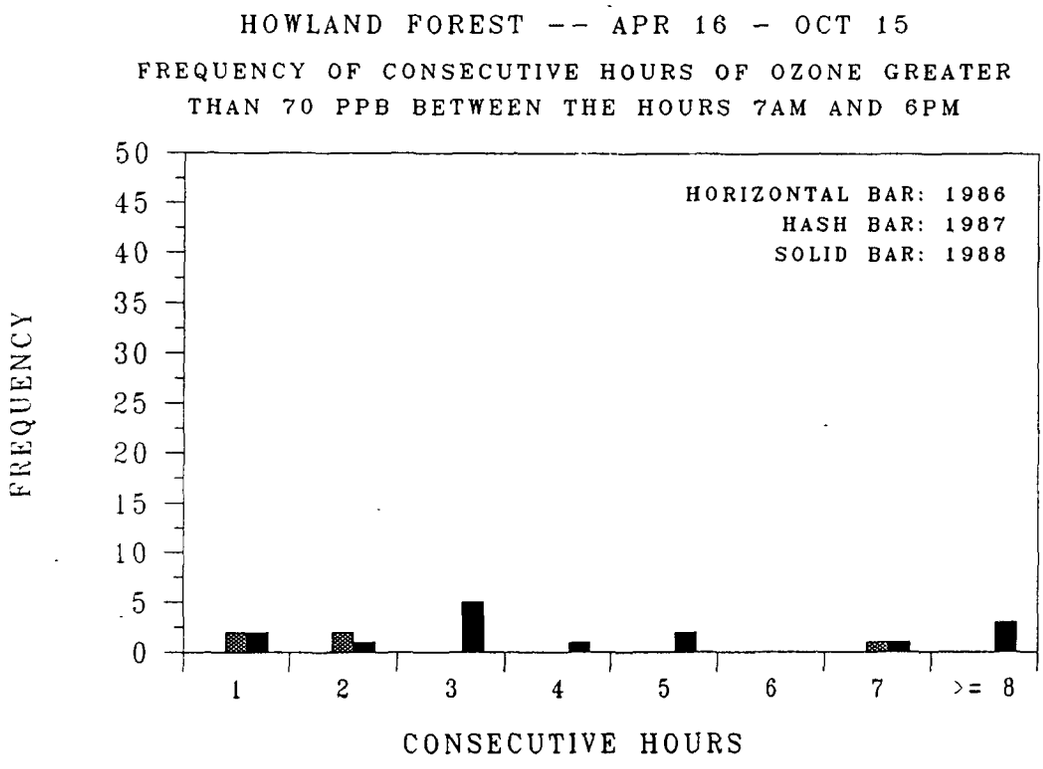


Figure 4-37

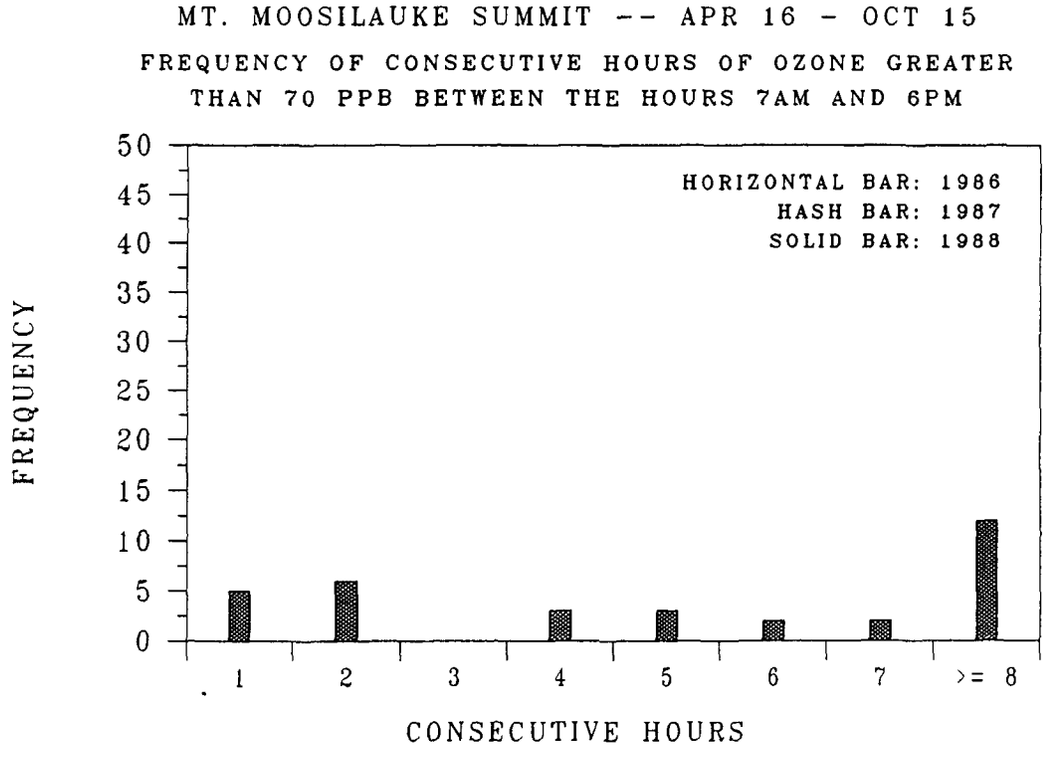


Figure 4-38

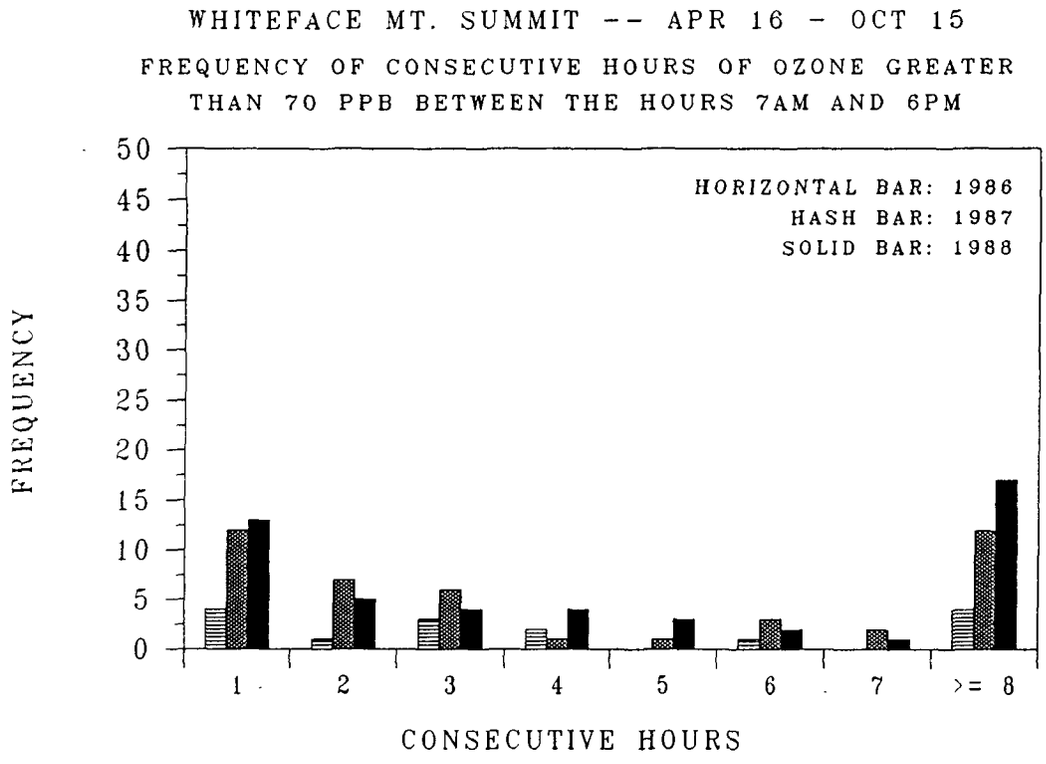


Figure 4-39

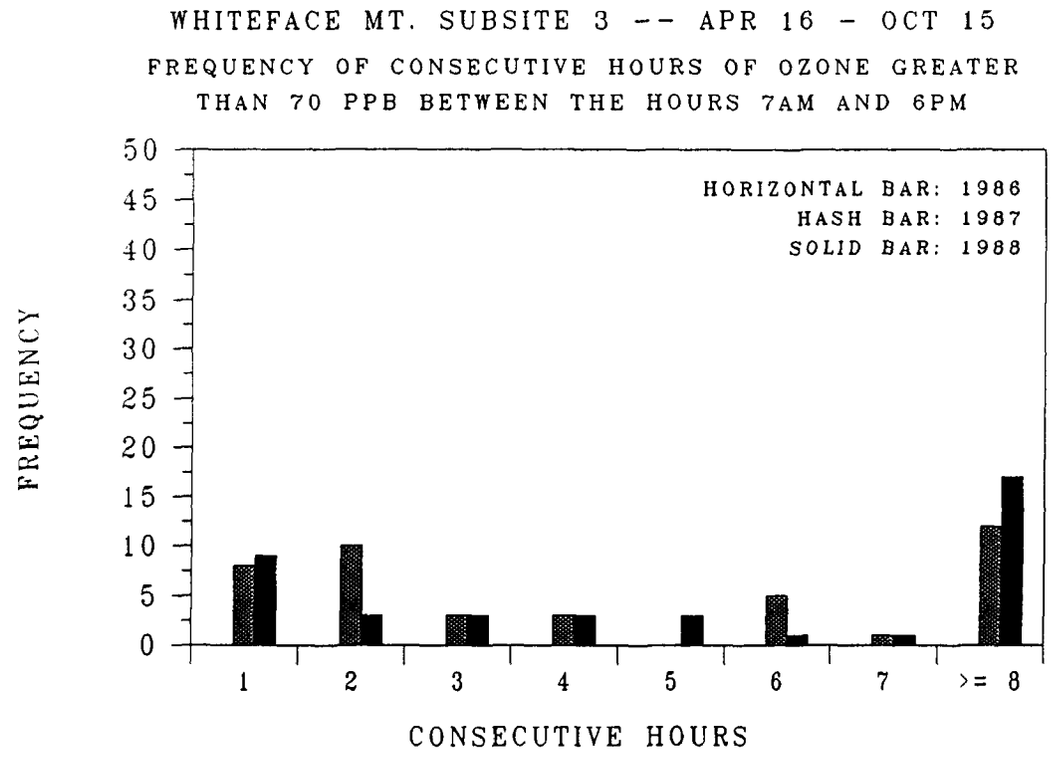


Figure 4-40

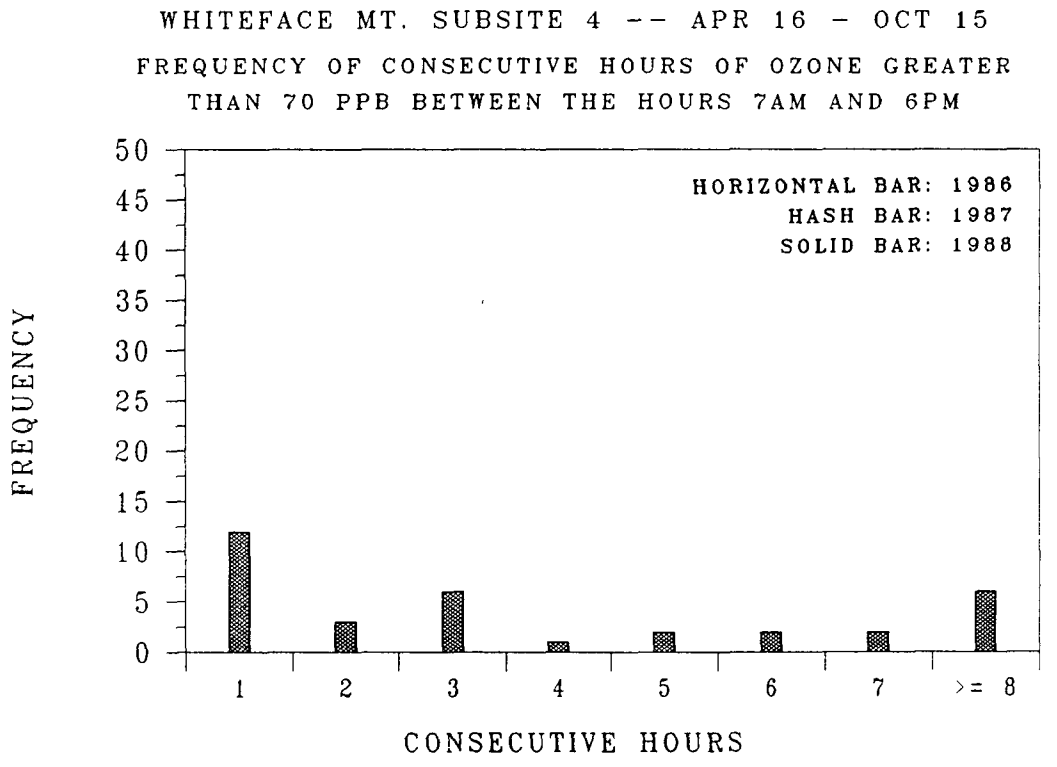


Figure 4-41

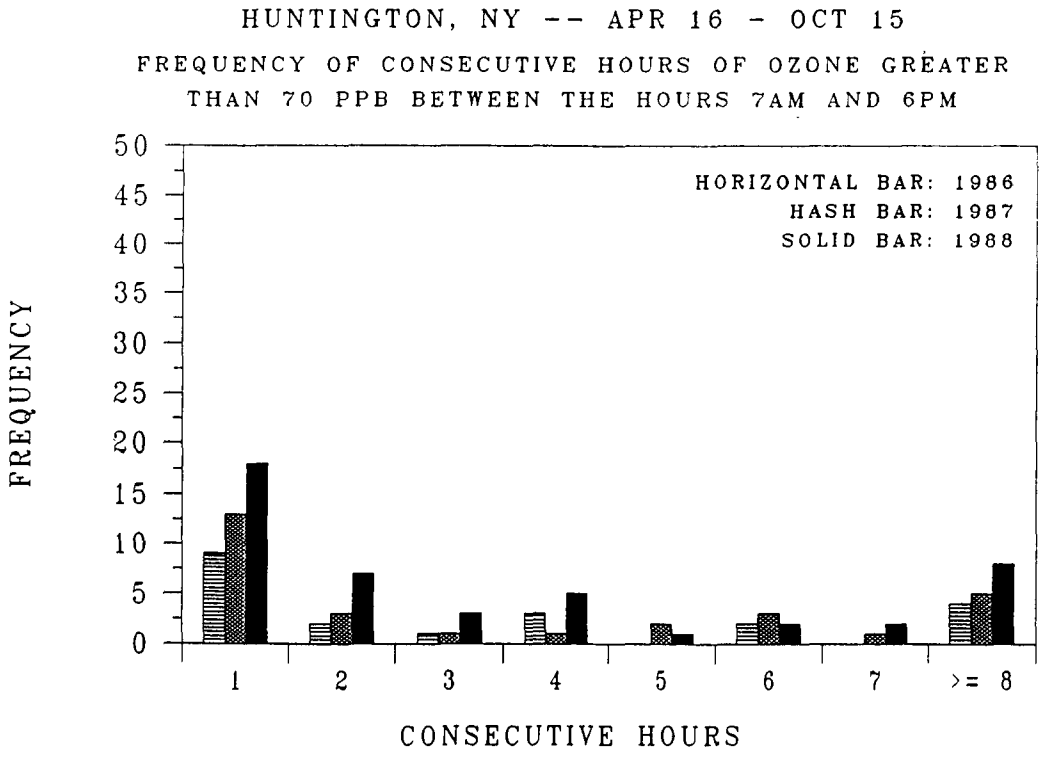


Figure 4-42

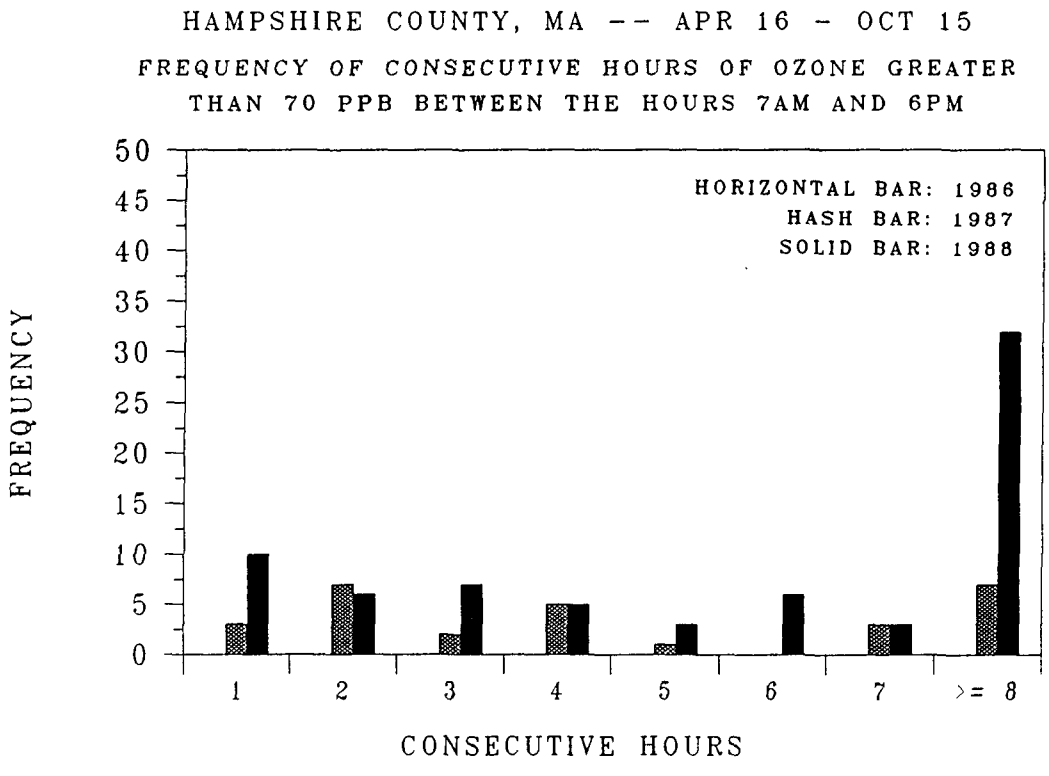


Figure 4-43

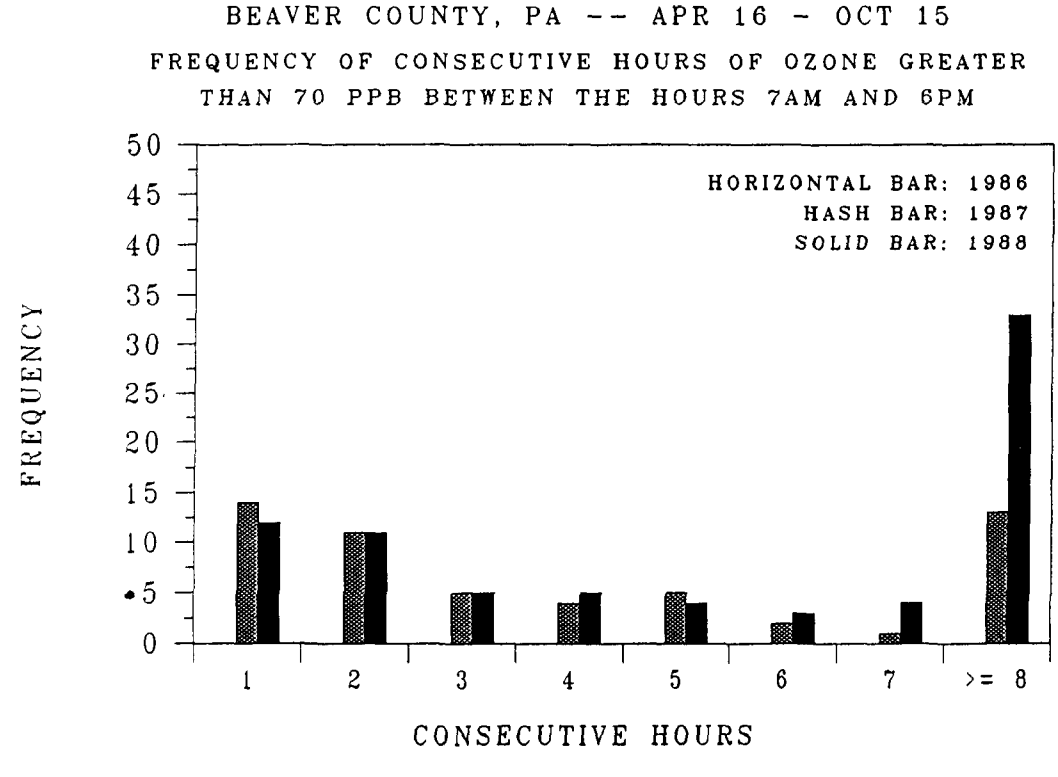


Figure 4-44

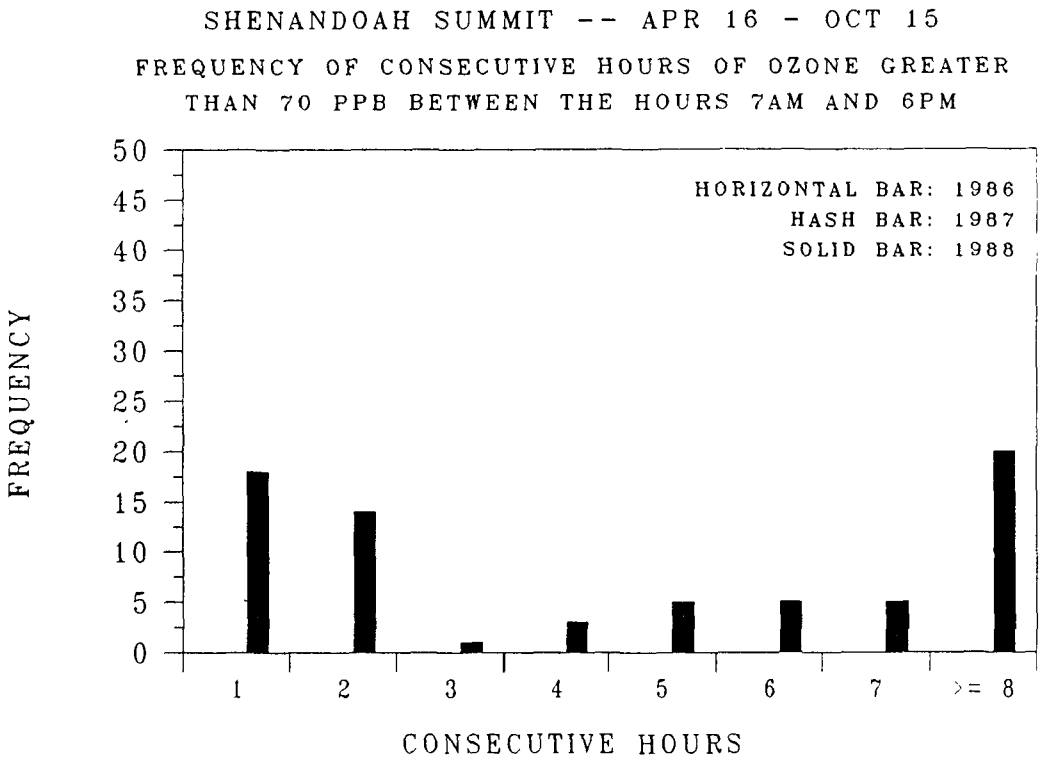


Figure 4-45

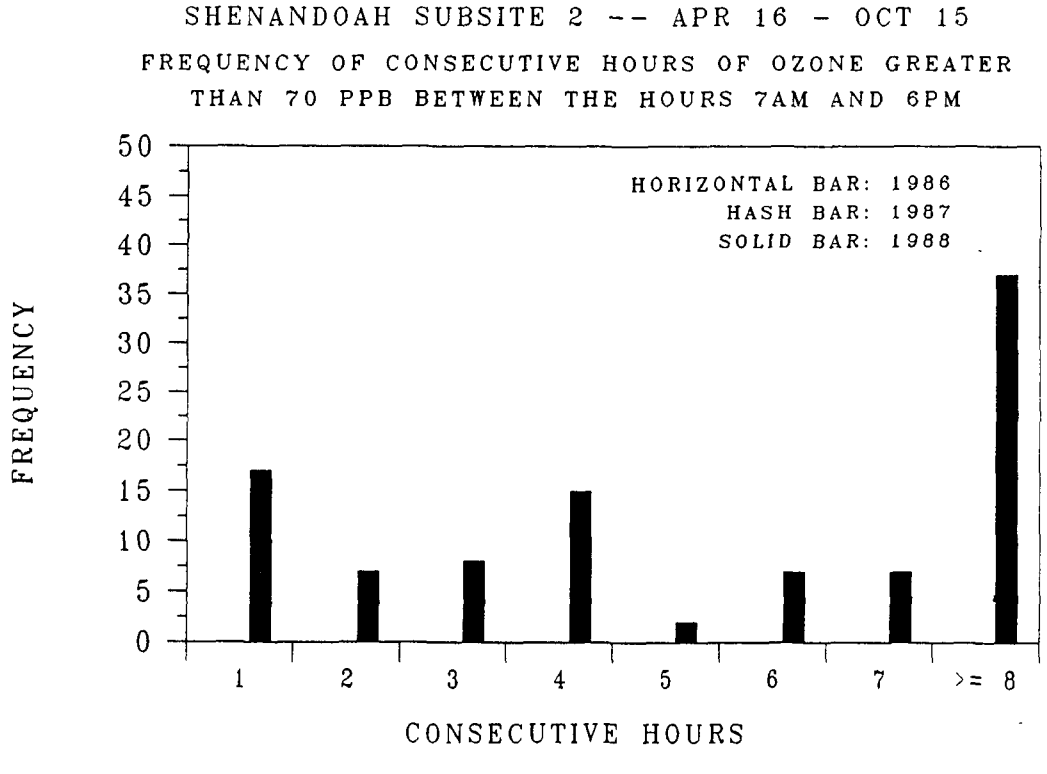


Figure 4-46

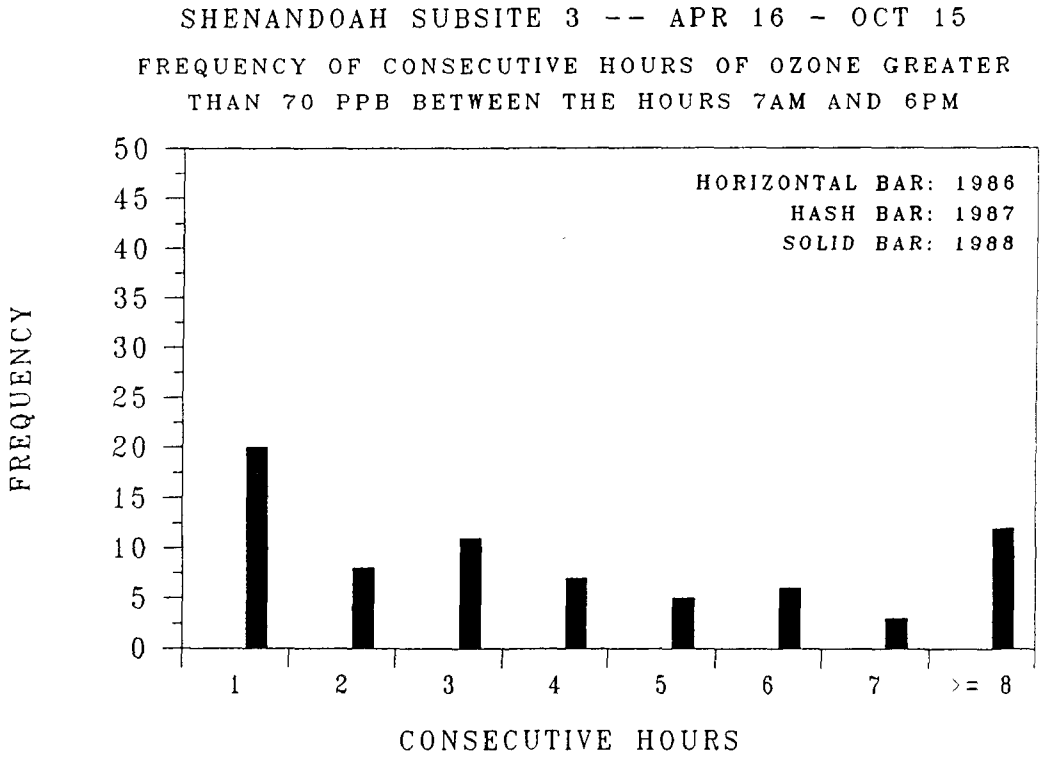


Figure 4-47

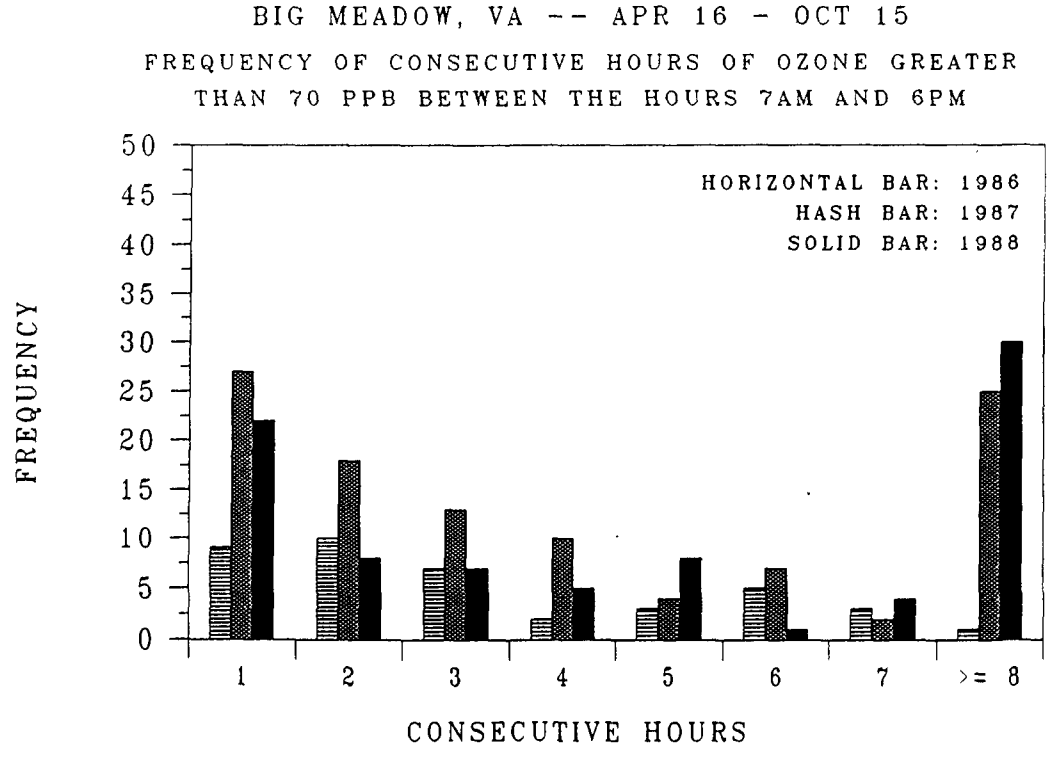


Figure 4-48

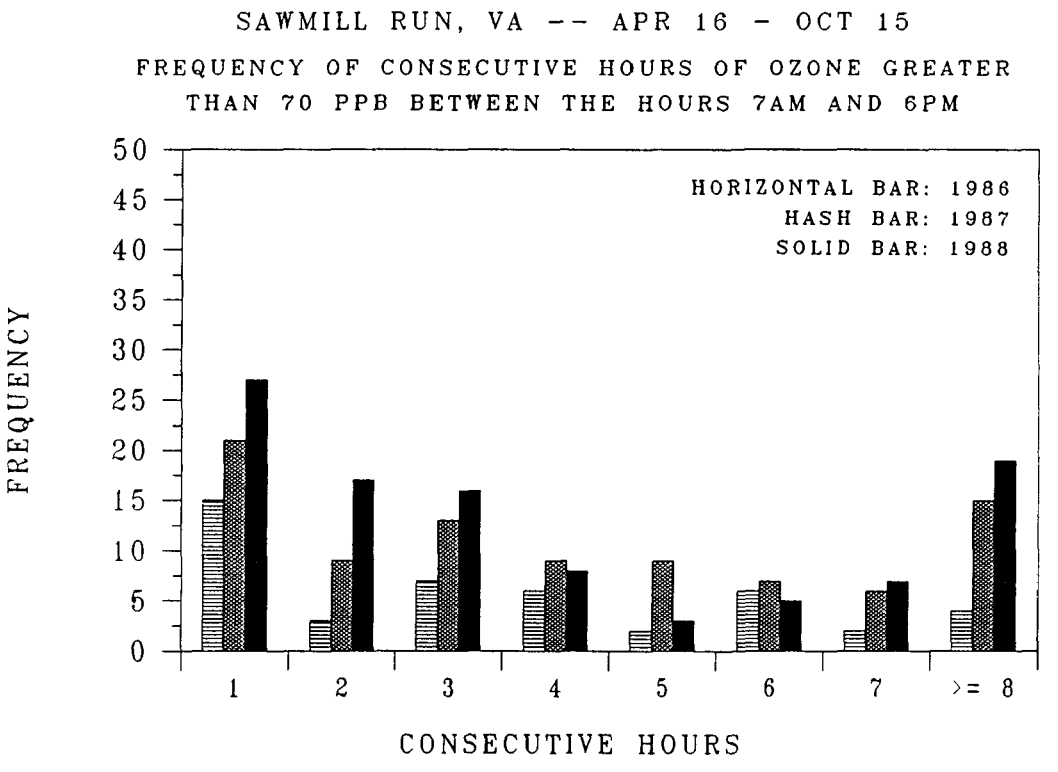


Figure 4-49

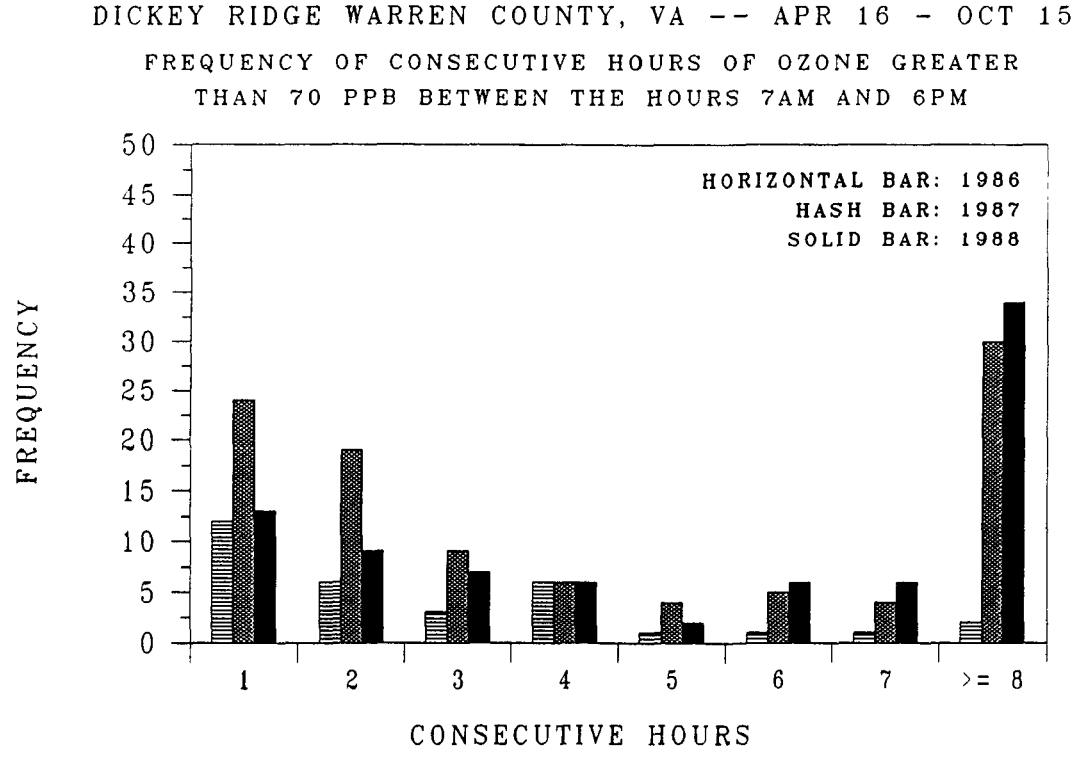


Figure 4-50

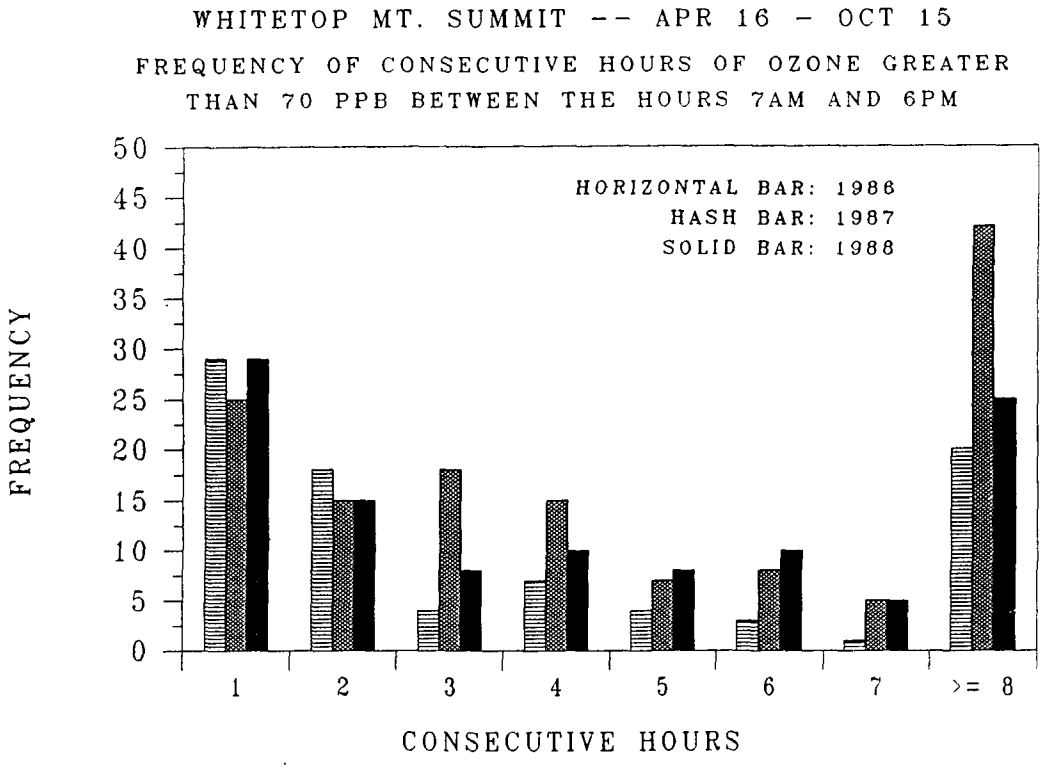


Figure 4-51

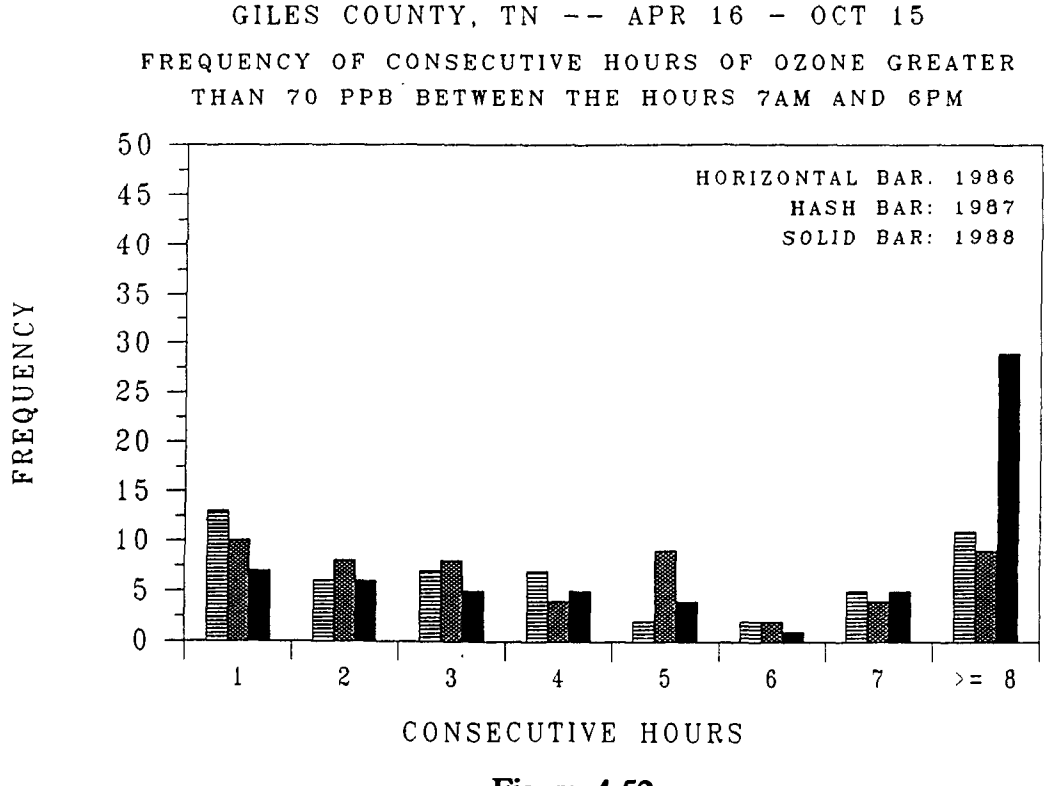


Figure 4-52

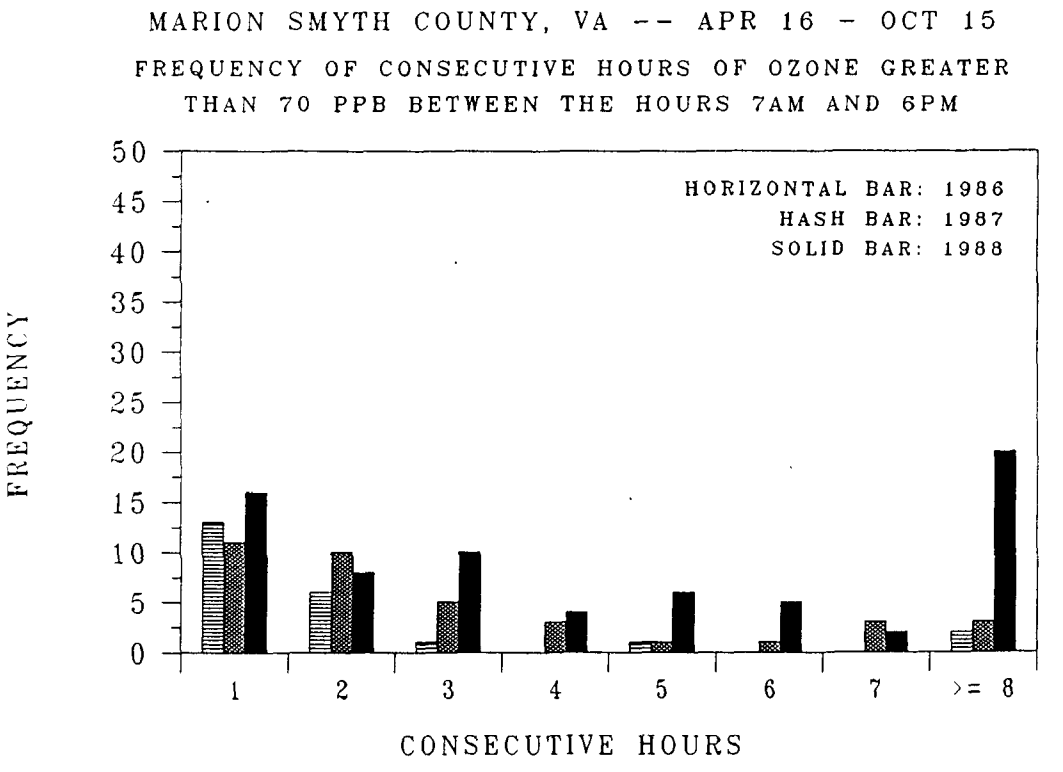


Figure 4-53

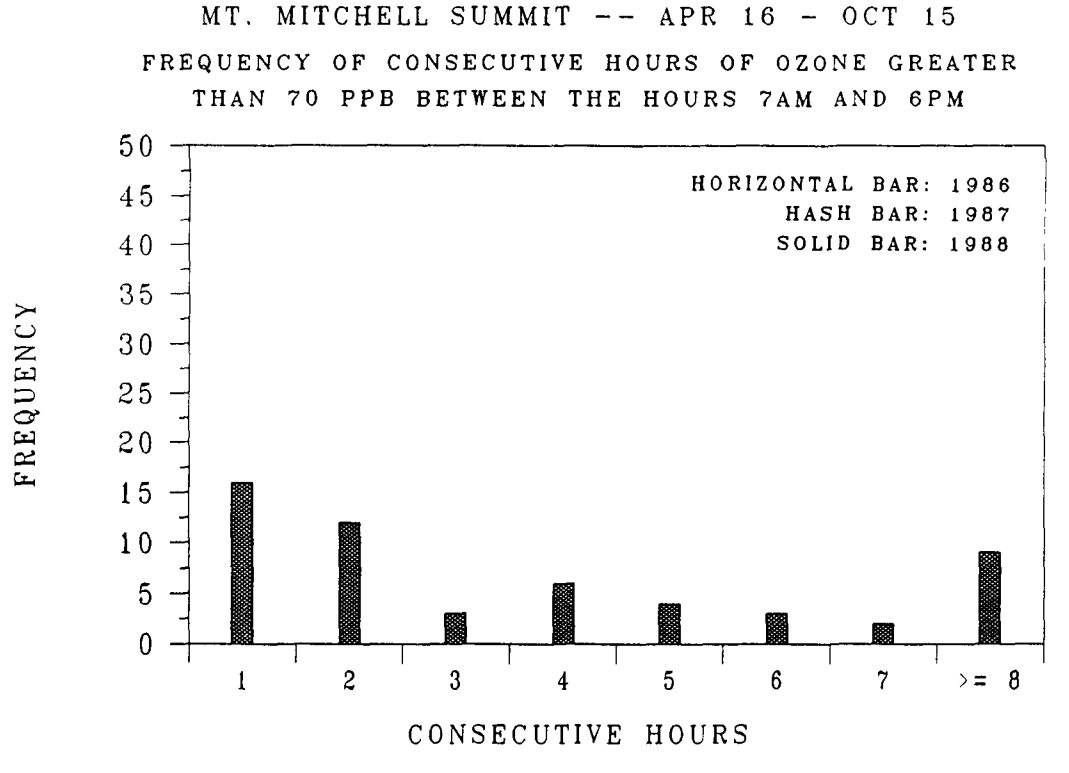


Figure 4-54

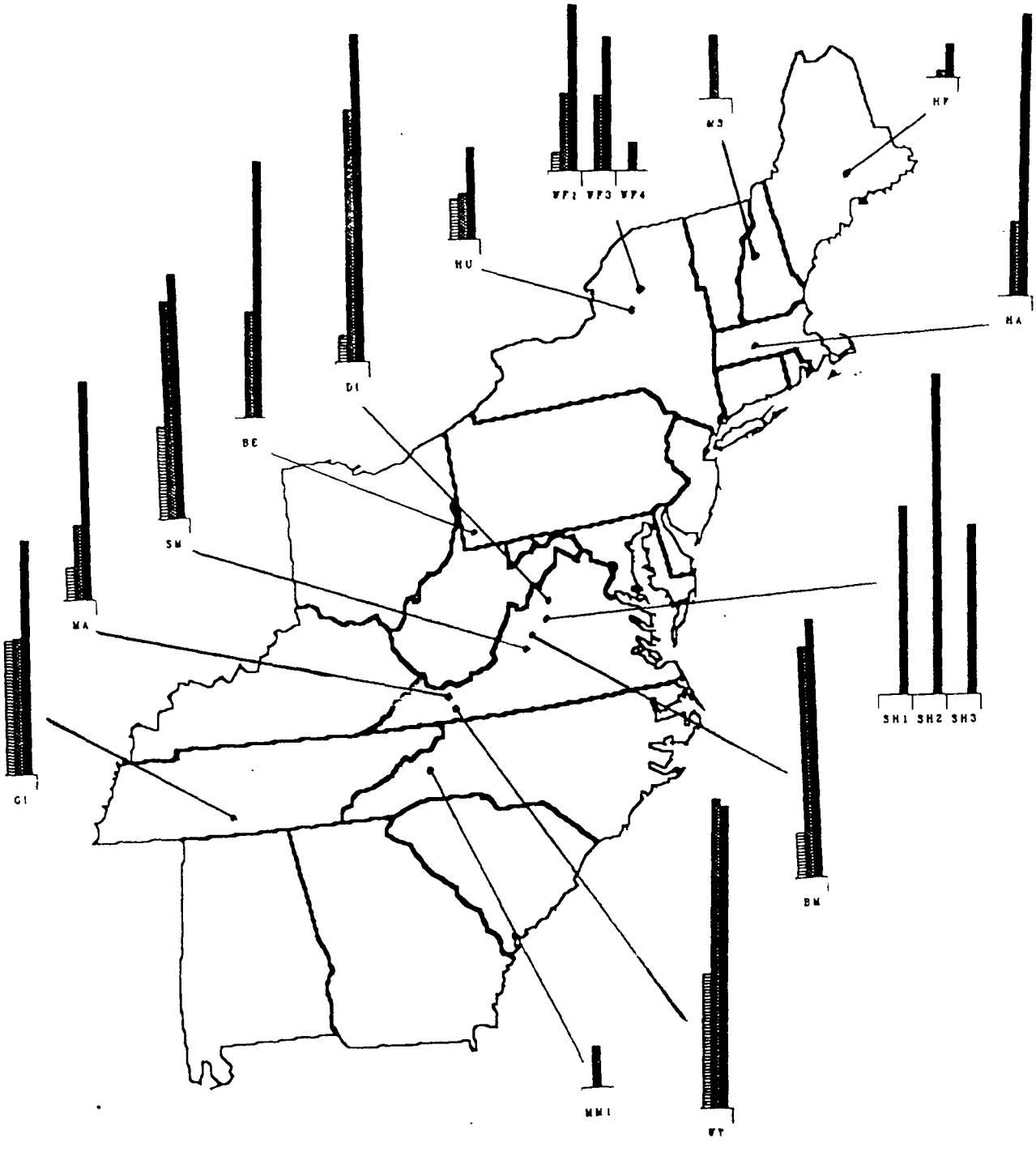


Figure 4-55: Ozone exposure - total season, Apr 15 - Oct 15, (Daylight hours 7AM - 6PM)

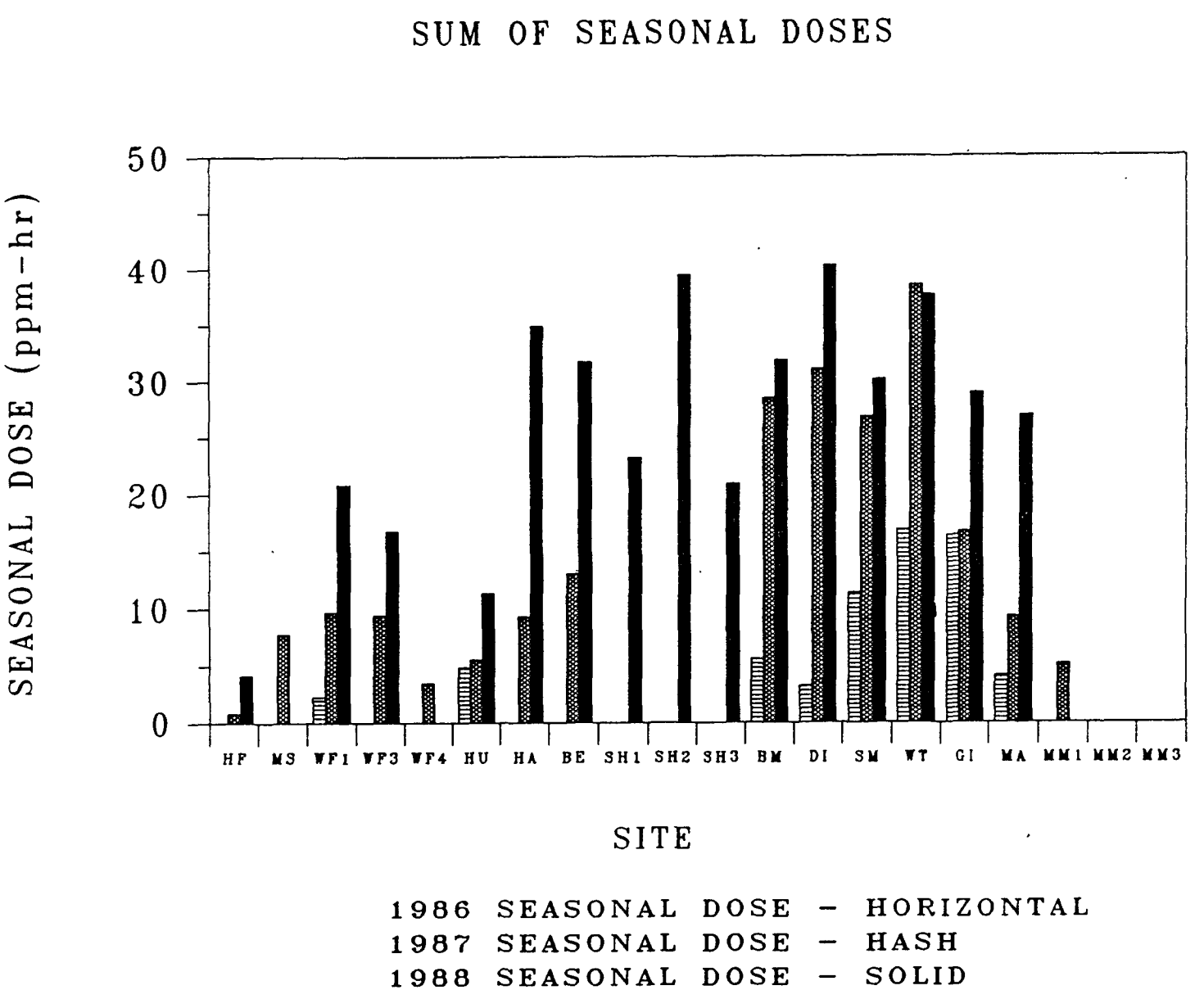


Figure 4-56 Ozone exposure - total season April 15-October 15 (Daylight hours 7 AM-6 PM).

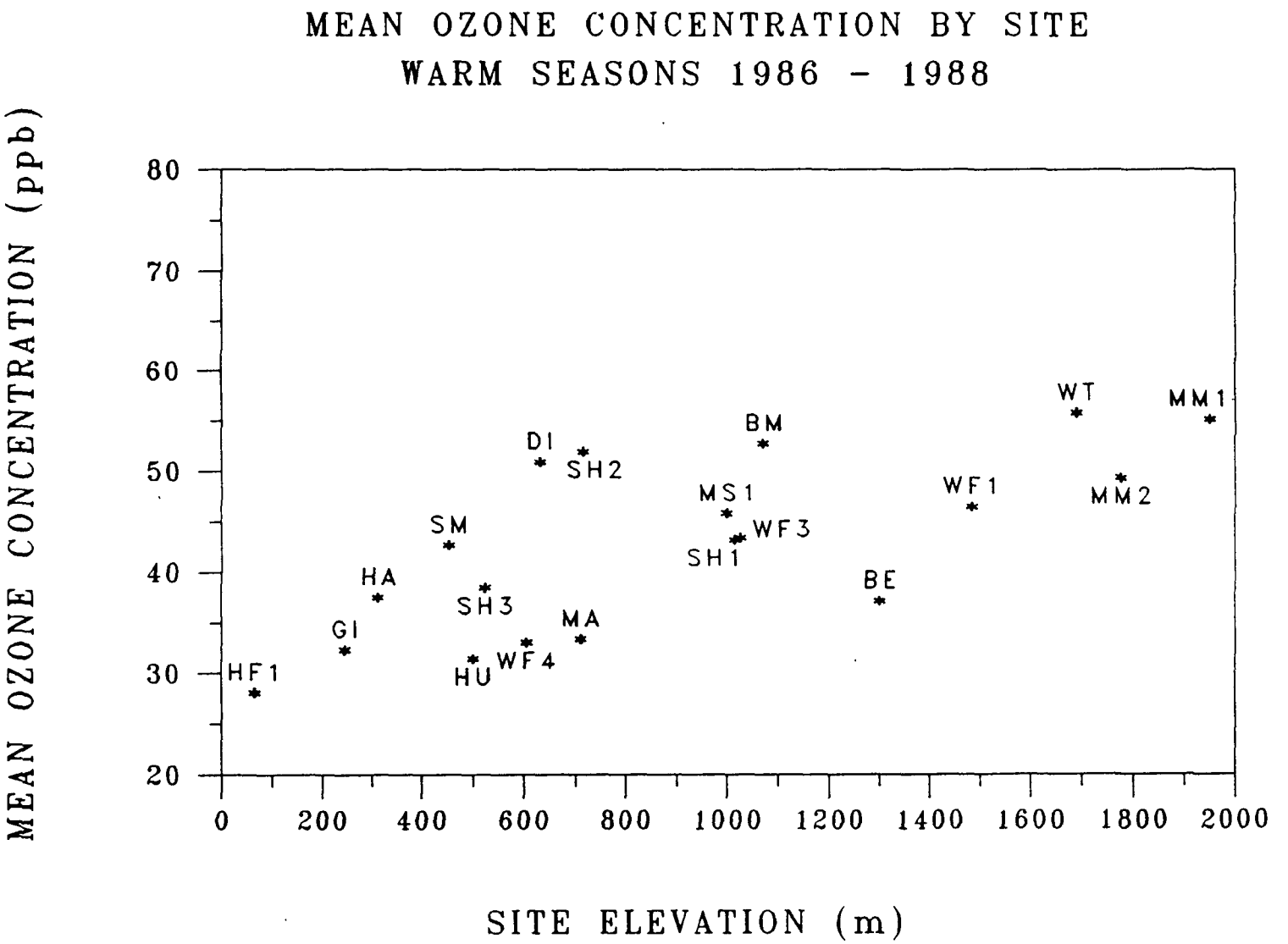

MEAN OZONE CONCENTRATION BY SITE
WARM SEASONS 1986 - 1988
MEAN OZONE CONCENTRATION (ppb)
80
70
60
50
40
30
20
0
200
400
600
800
1000
1200
1400
1600
1800
2000
SITE ELEVATION (m)
HF1
GI
HA
SM
DI
SH2
SH3
MA
HU
WF4
BM
MS1
WF3
SH1
BE
WF1
WT
MM2
MM1

Figure 4-61

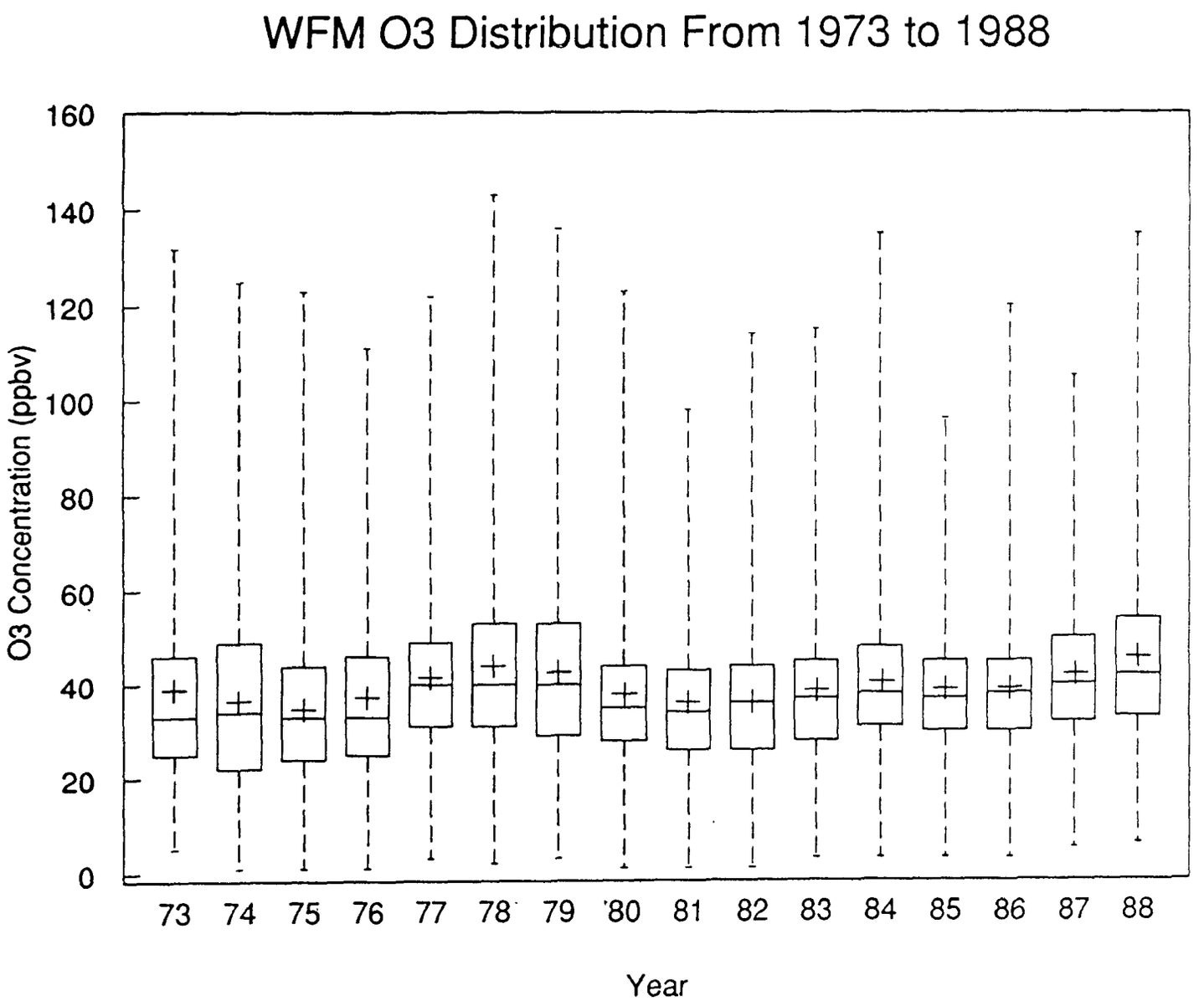

WFM O3 Distribution From 1973 to 1988
O3 Concentration (ppbv)
160
140
120
100
80
60
40
20
0
73
74
75
76
77
78
79
'80
81
82
83
84
85
86
87
88
Year

Figure 4-62

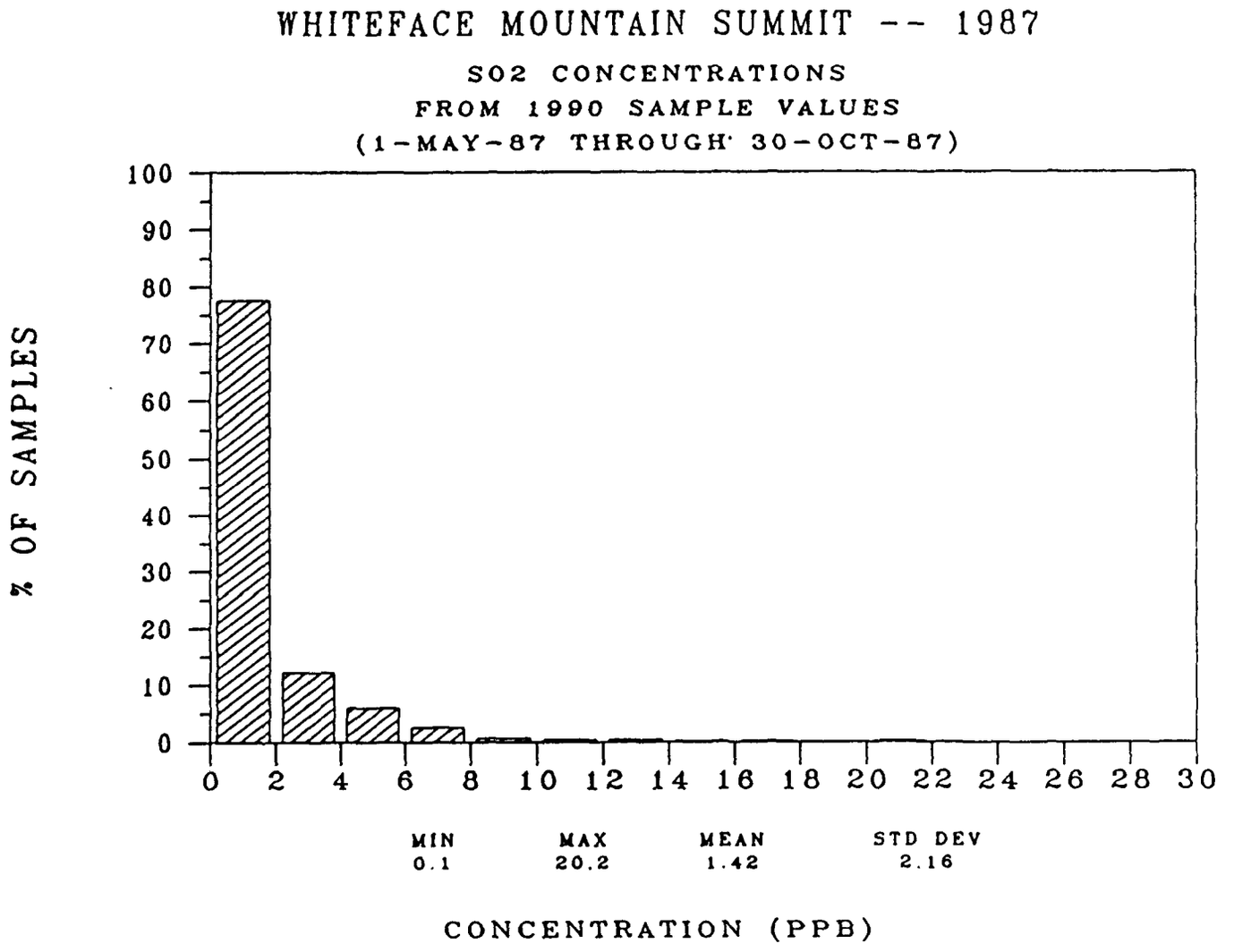

WHITEFACE MOUNTAIN SUMMIT -- 1987
SO2 CONCENTRATIONS
FROM 1990 SAMPLE VALUES
(1-MAY-87 THROUGH 30-OCT-87)
% OF SAMPLES
100
90
80
70
60
50
40
30
20
10
0
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30
MIN 0.1
MAX 20.2
MEAN 1.42
STD DEV 2.16
CONCENTRATION (PPB)

Figure 4-63

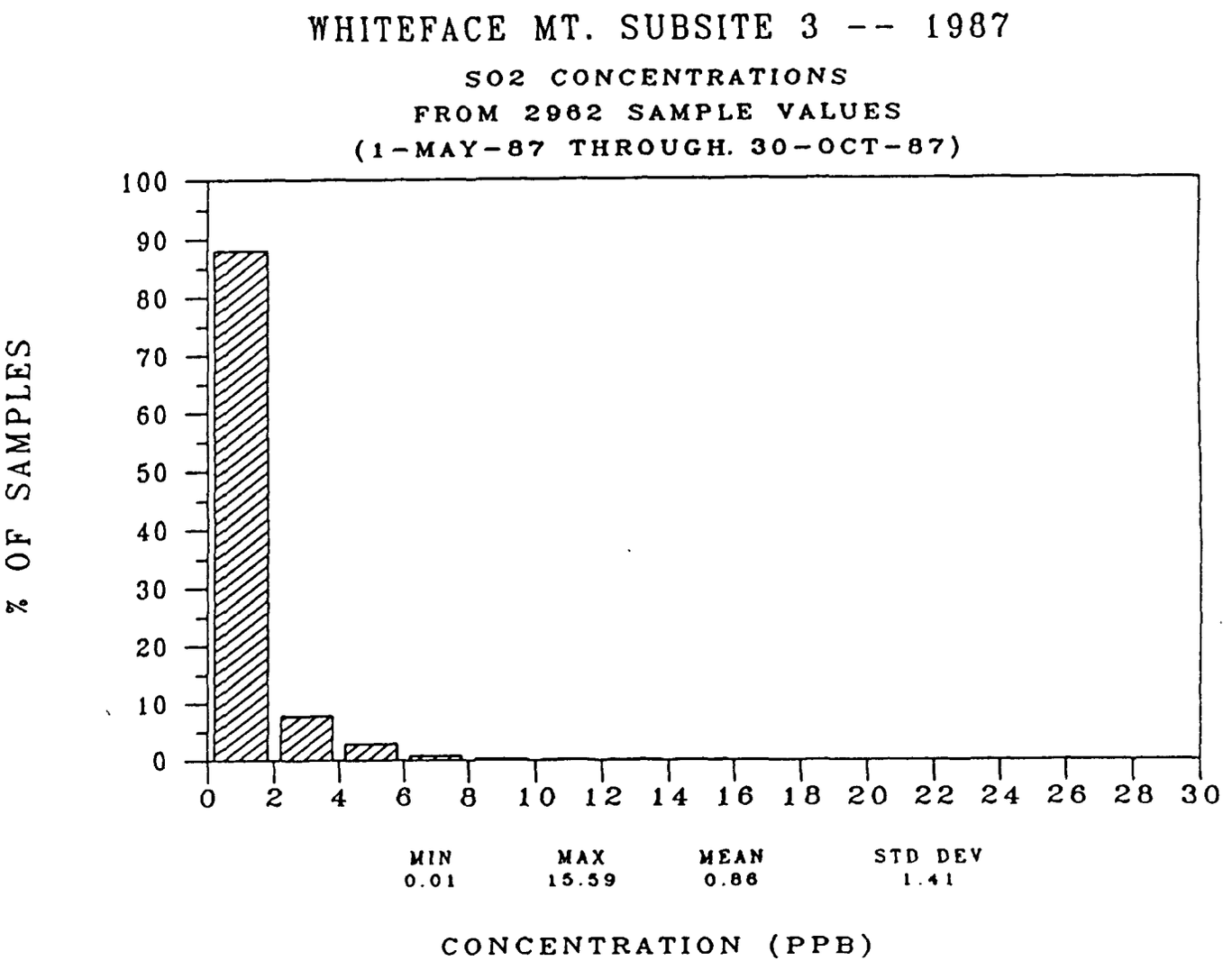

WHITEFACE MT. SUBSITE 3 -- 1987
SO2 CONCENTRATIONS
FROM 2962 SAMPLE VALUES
(1-MAY-87 THROUGH. 30-OCT-87)
% OF SAMPLES
100
90
80
70
60
50
40
30
20
10
0
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30
MIN 0.01
MAX 15.59
MEAN 0.86
STD DEV 1.41
CONCENTRATION (PPB)

Figure 4-64

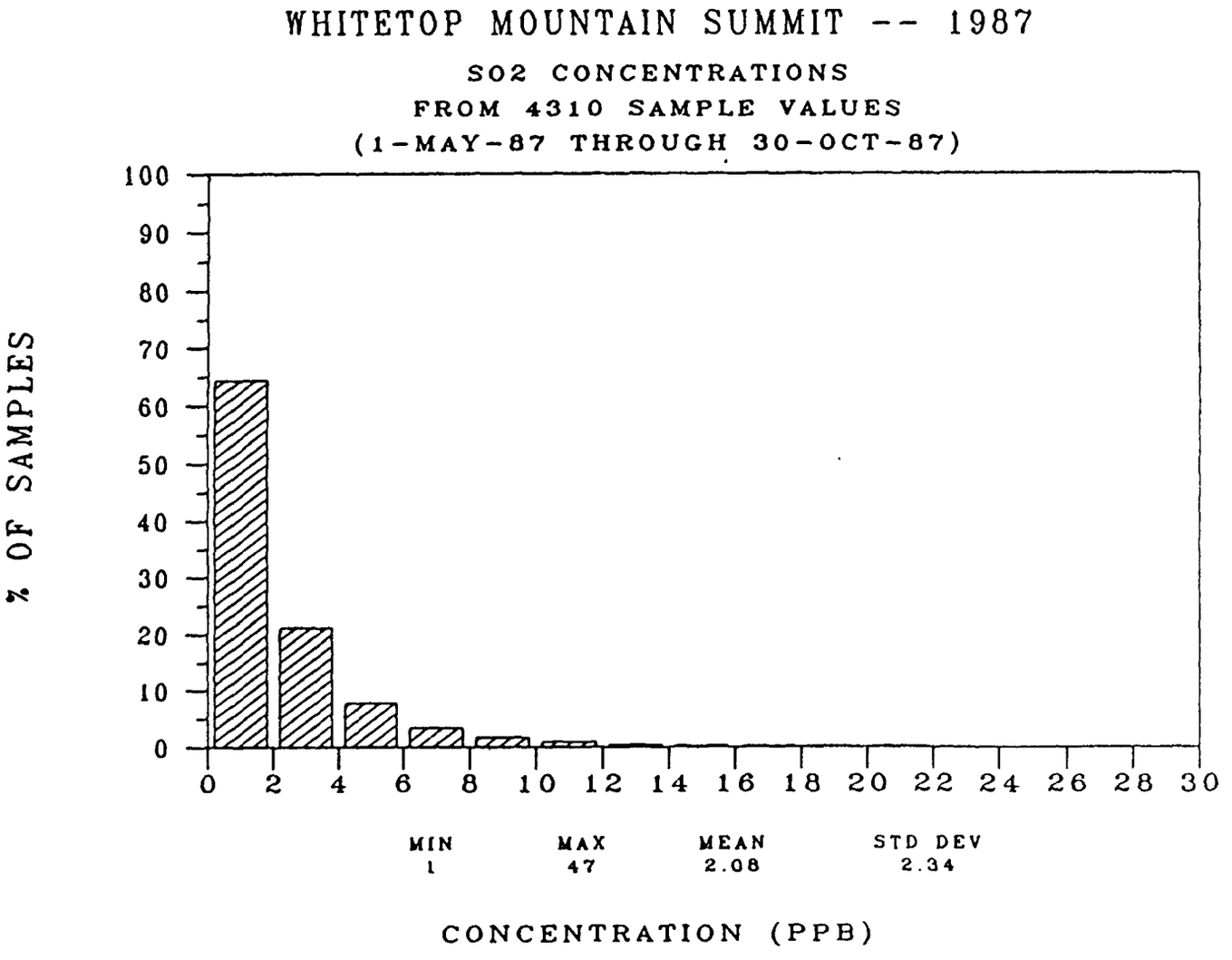

WHITETOP MOUNTAIN SUMMIT -- 1987
SO2 CONCENTRATIONS
FROM 4310 SAMPLE VALUES
(1-MAY-87 THROUGH 30-OCT-87)
% OF SAMPLES
100
90
80
70
60
50
40
30
20
10
0
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30
MIN
1
MAX
47
MEAN
2.08
STD DEV
2.34
CONCENTRATION (PPB)

Figure 4-65

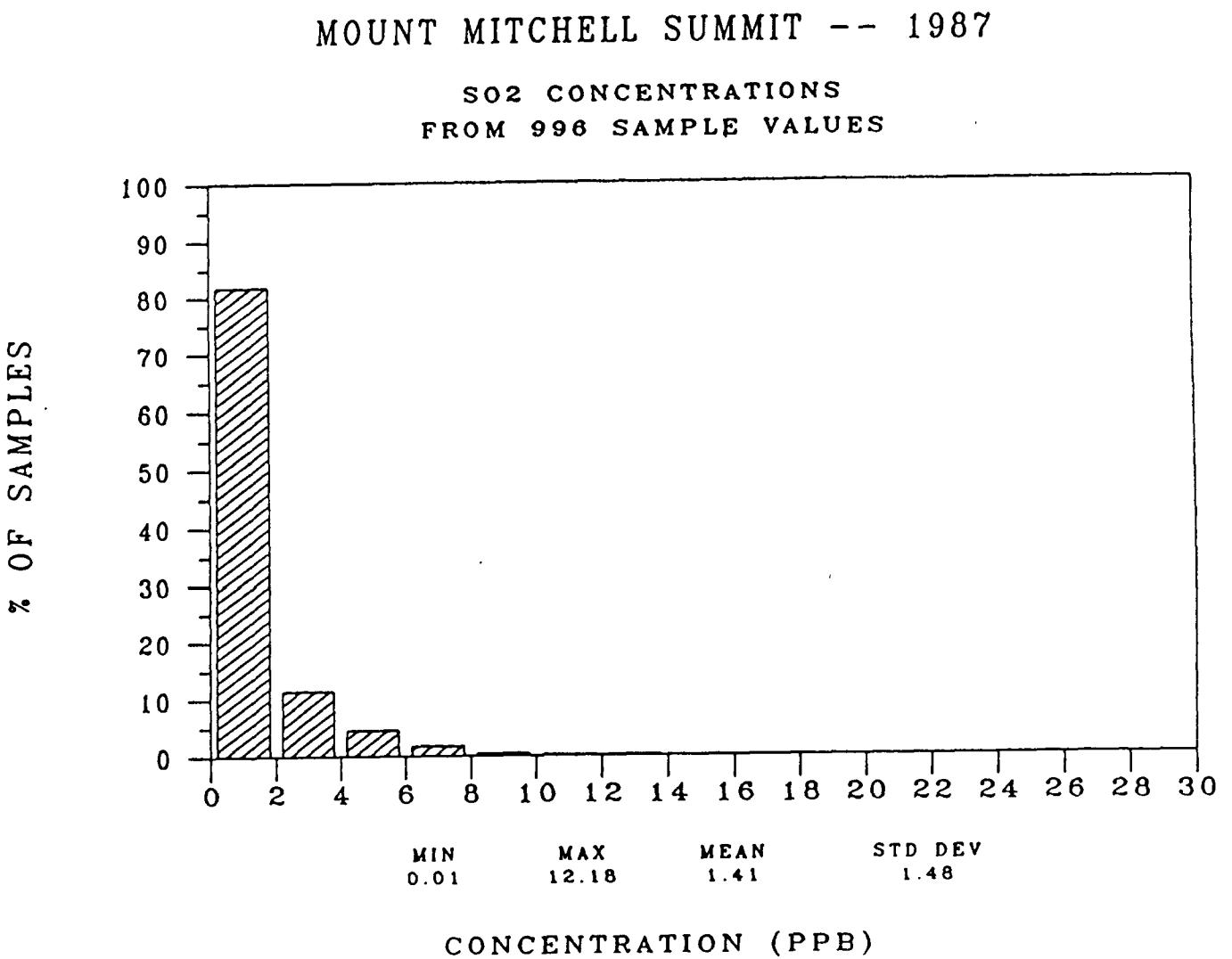

MOUNT MITCHELL SUMMIT -- 1987
SO2 CONCENTRATIONS
FROM 996 SAMPLE VALUES
% OF SAMPLES
100
90
80
70
60
50
40
30
20
10
0
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30
MIN
0.01
MAX
12.18
MEAN
1.41
STD DEV
1.48
CONCENTRATION (PPB)

Figure 4-66

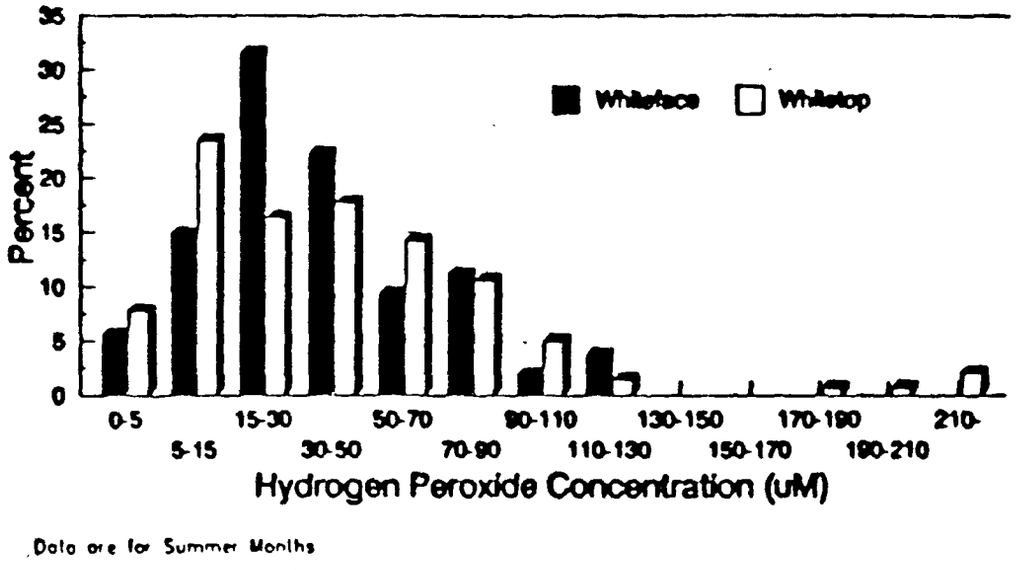


Figure 4-67 Frequency of occurrence for H_2O_2 concentrations in cloud water collected at the Whiteface, NY and Whitetop, VA MCCP sites.

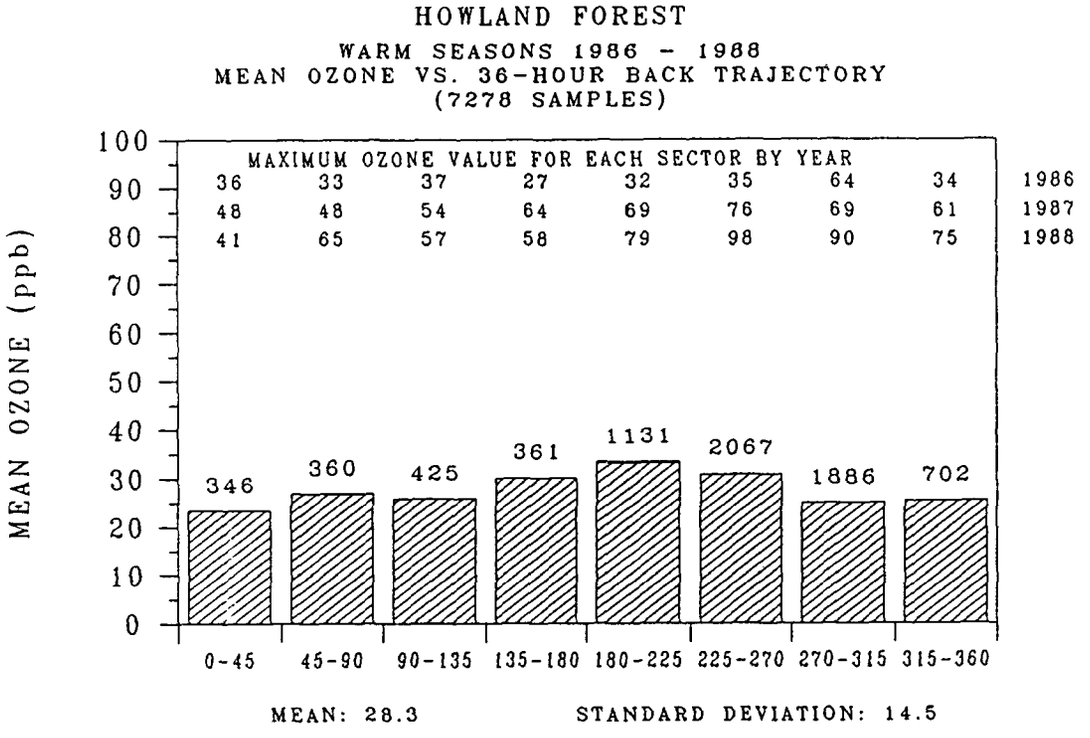


Figure 5-1

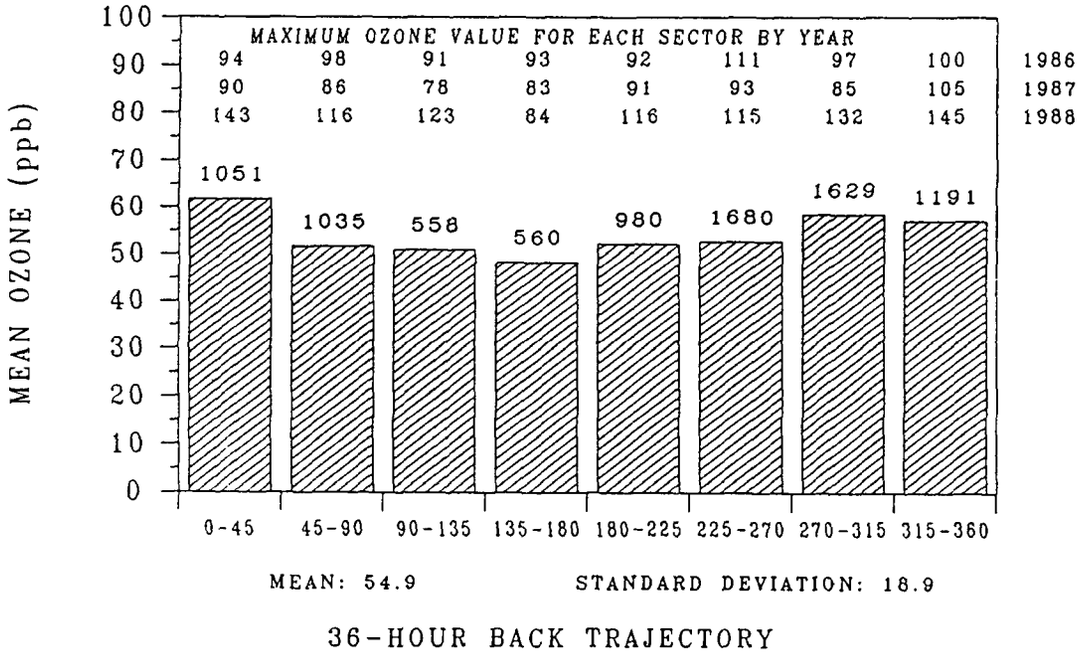


Figure 5-2

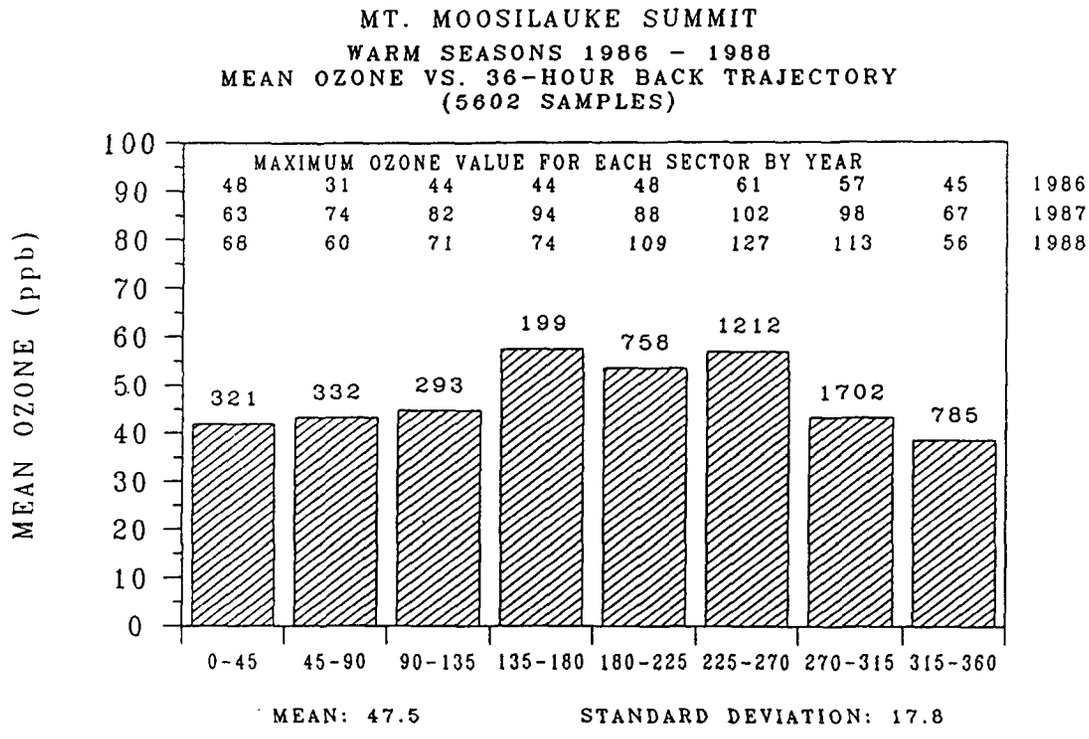


Figure 5-3

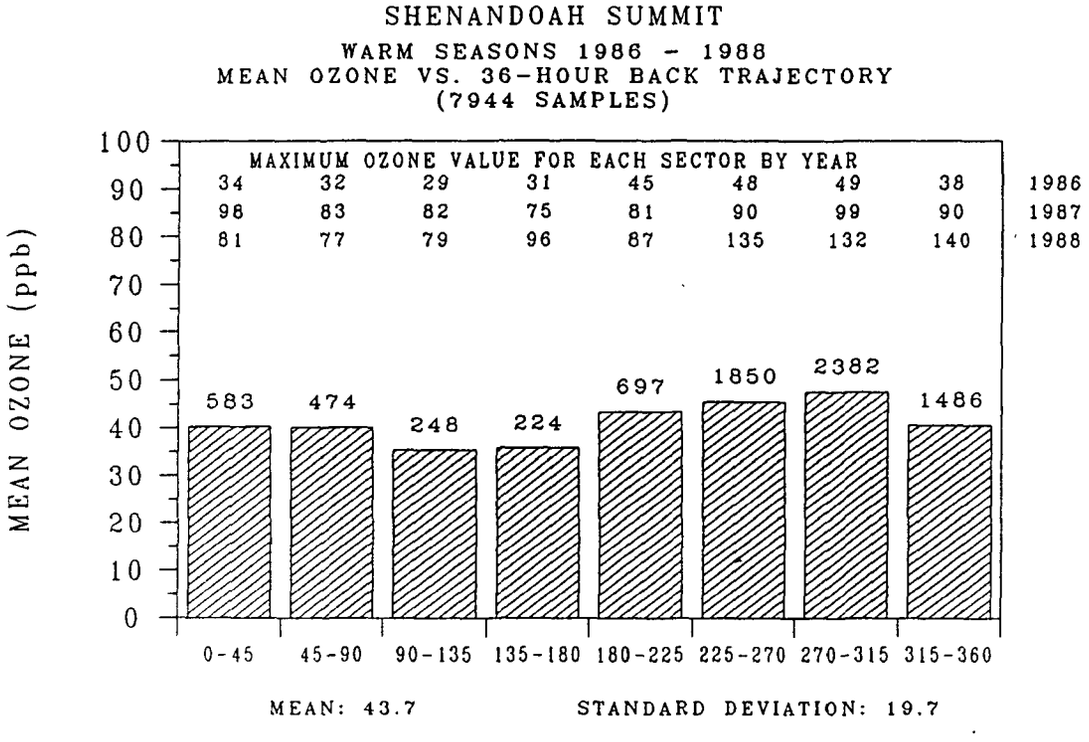


Figure 5-4

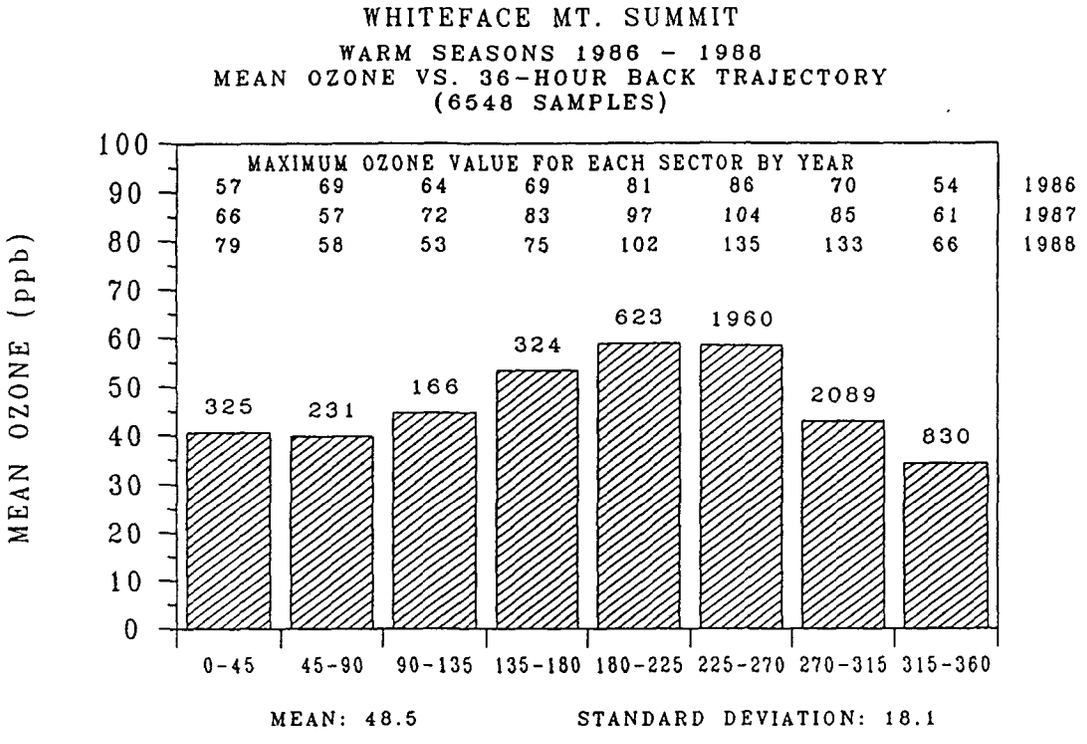


Figure 5-5

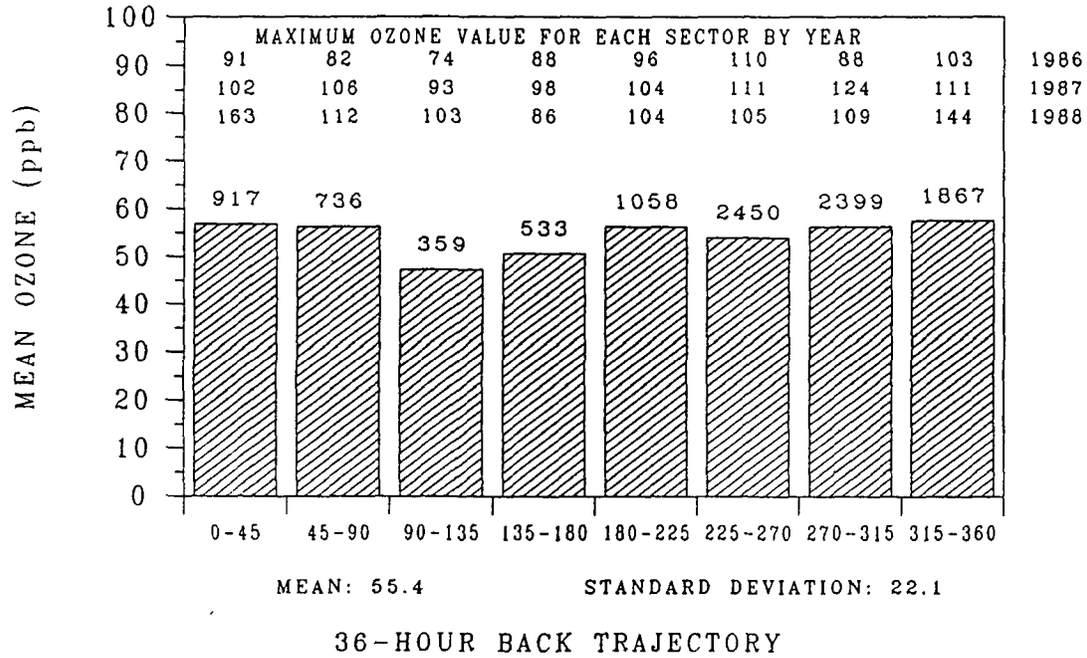


Figure 5-6

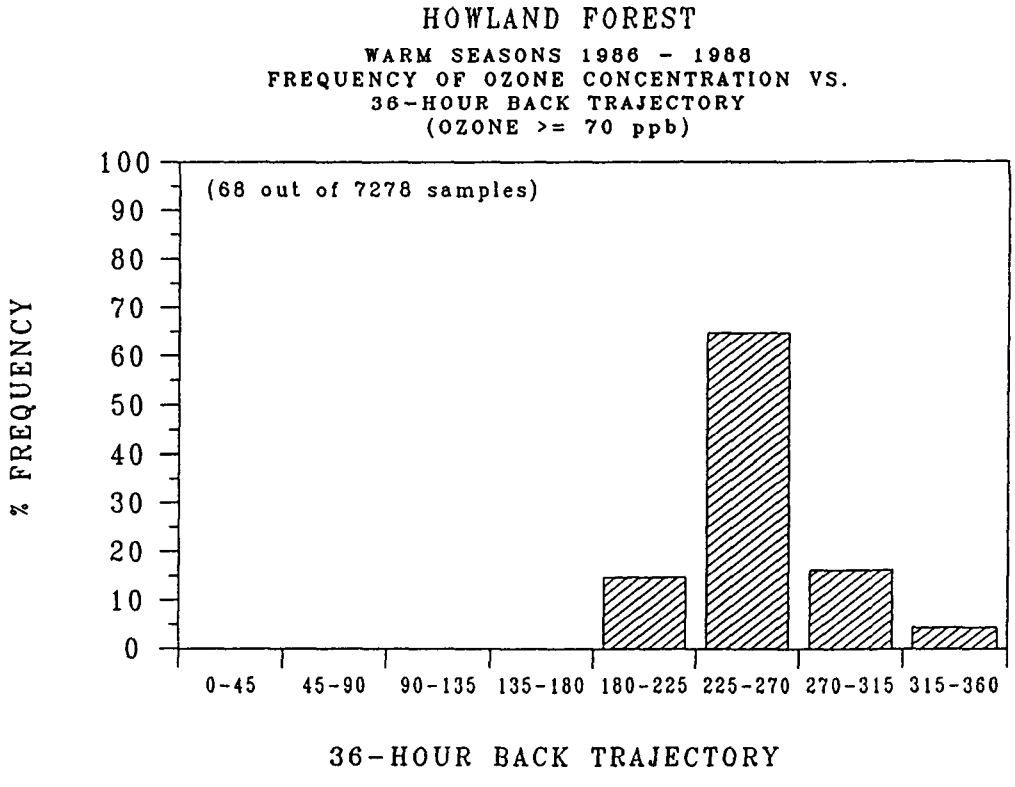


Figure 5-7

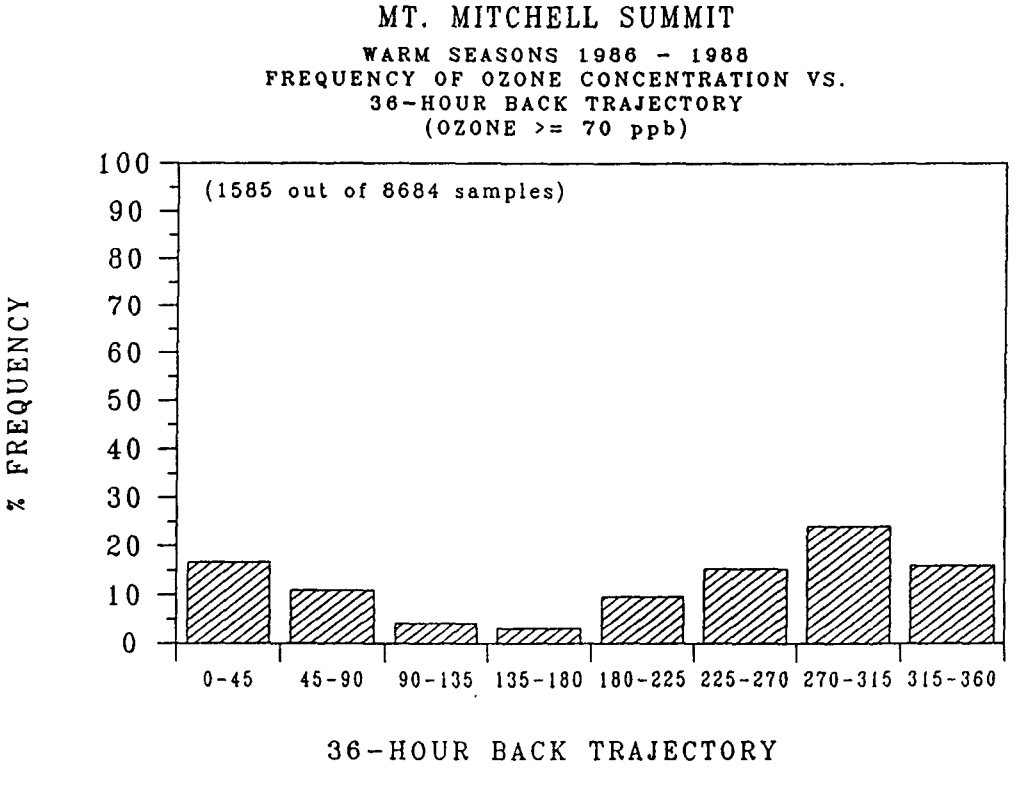


Figure 5-8

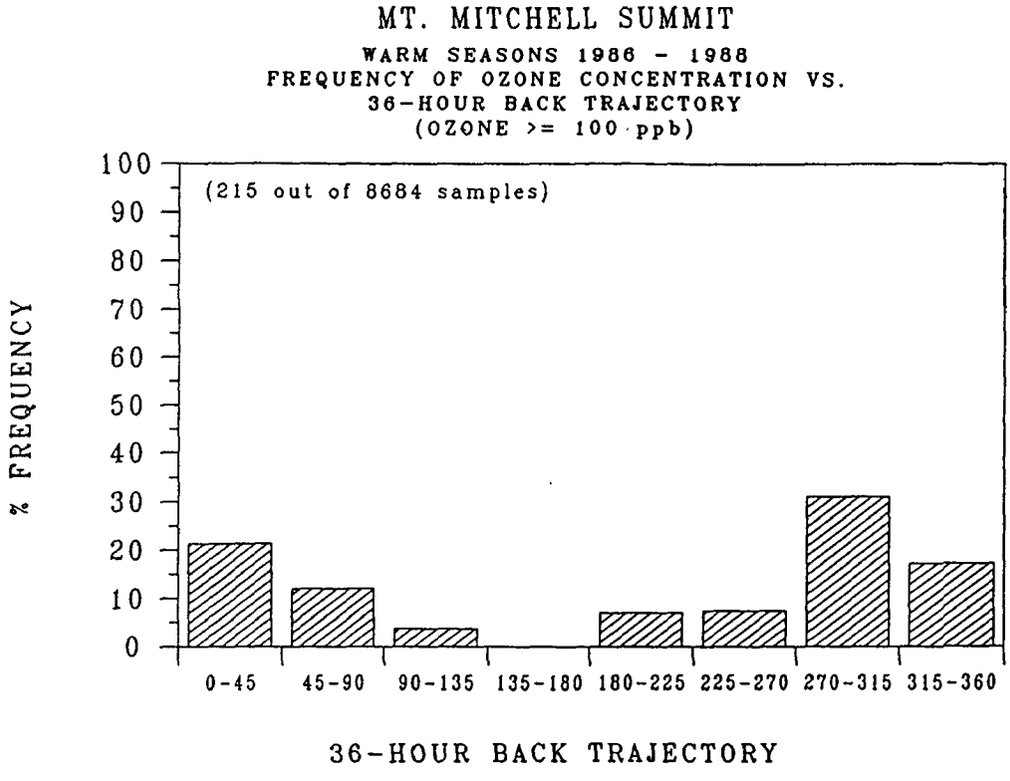


Figure 5-9

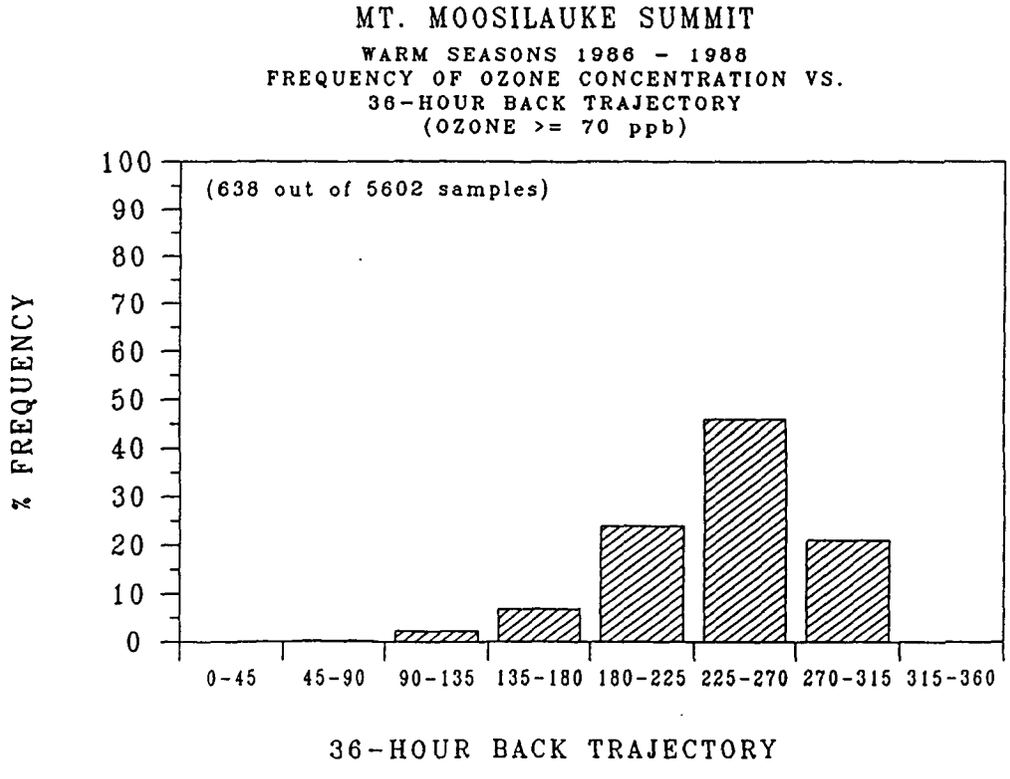


Figure 5-10

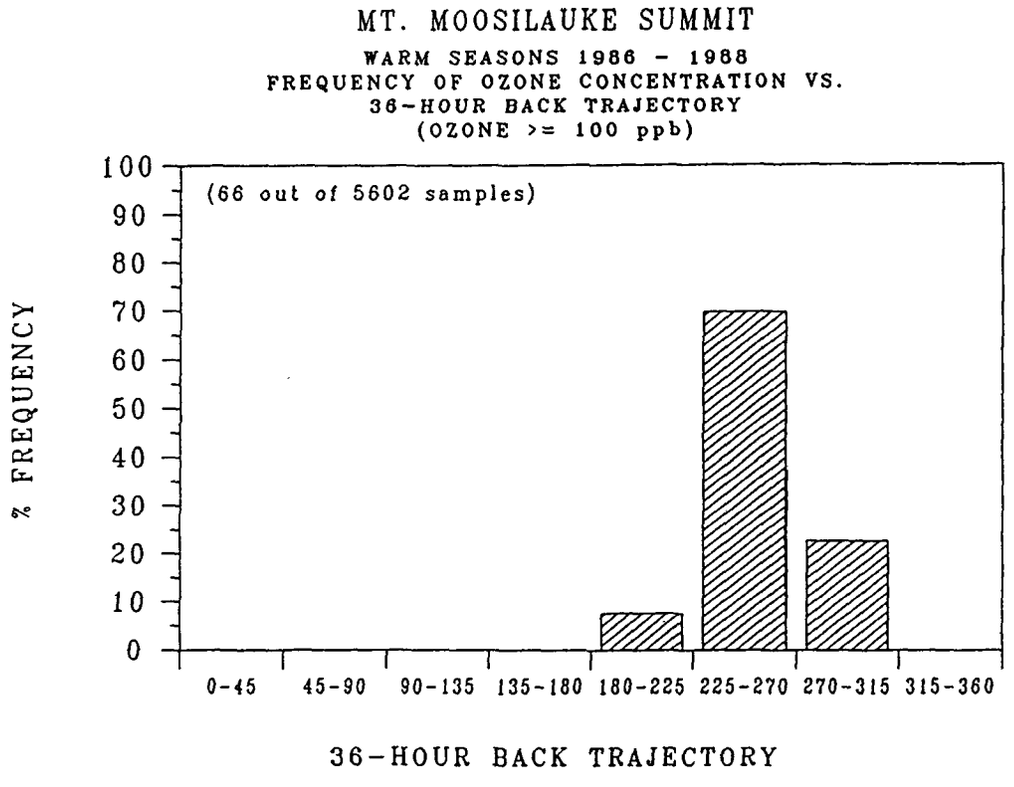


Figure 5-11

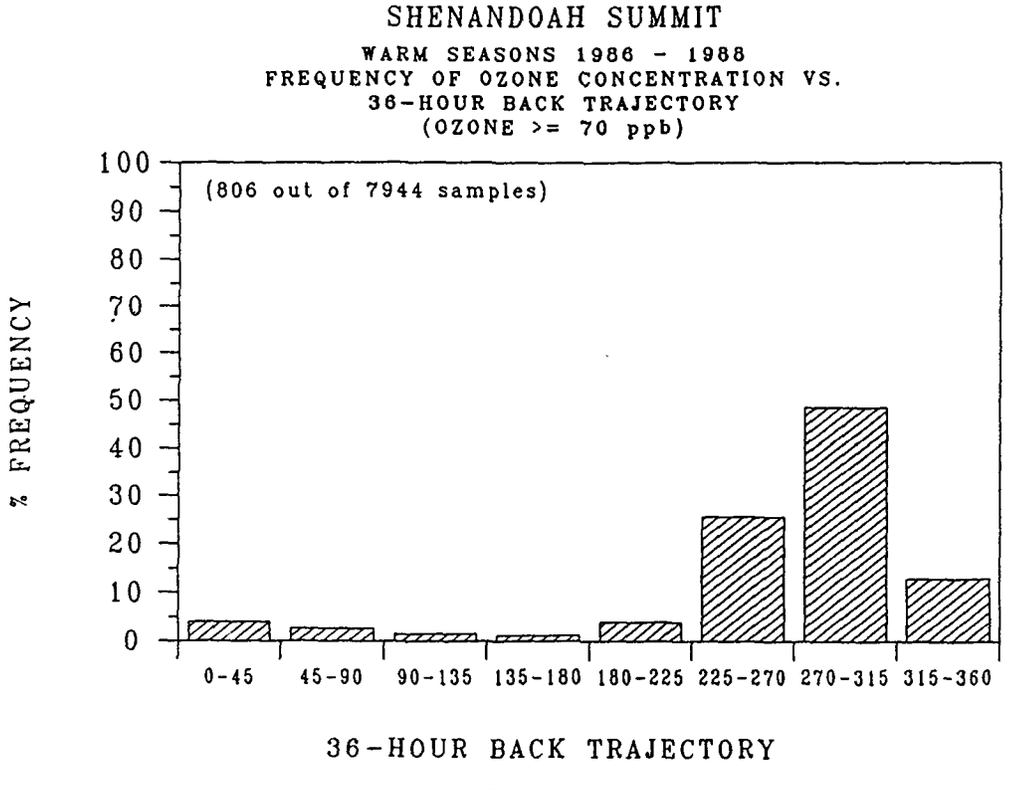


Figure 5-12

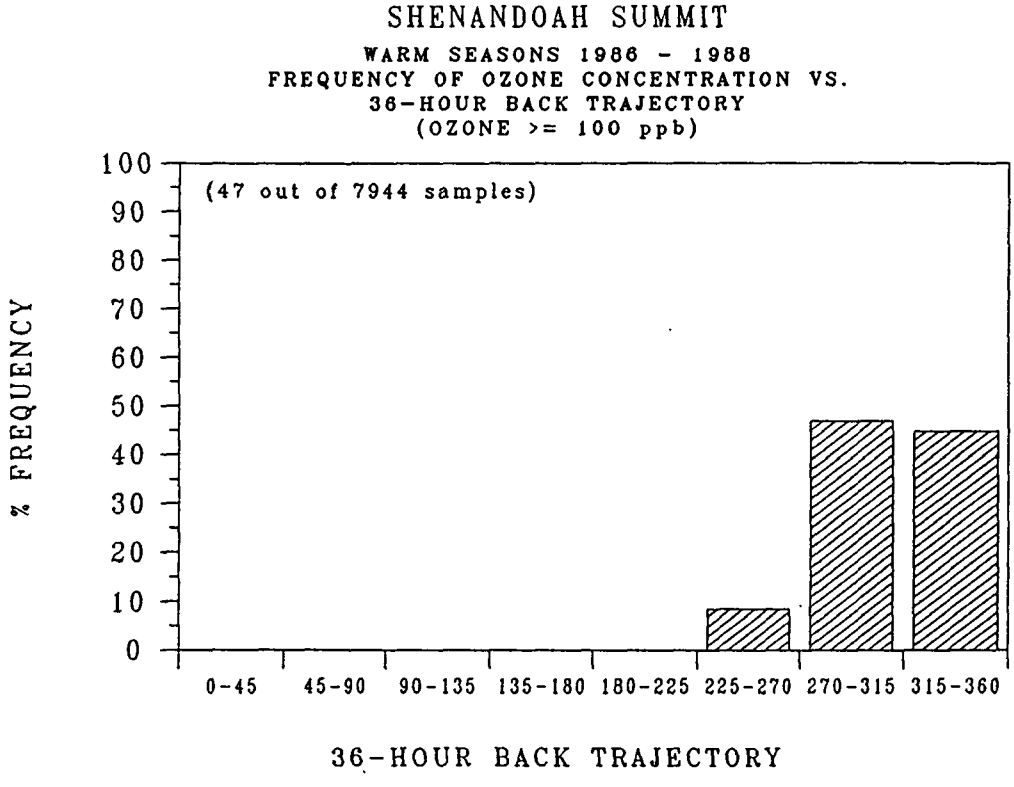


Figure 5-13

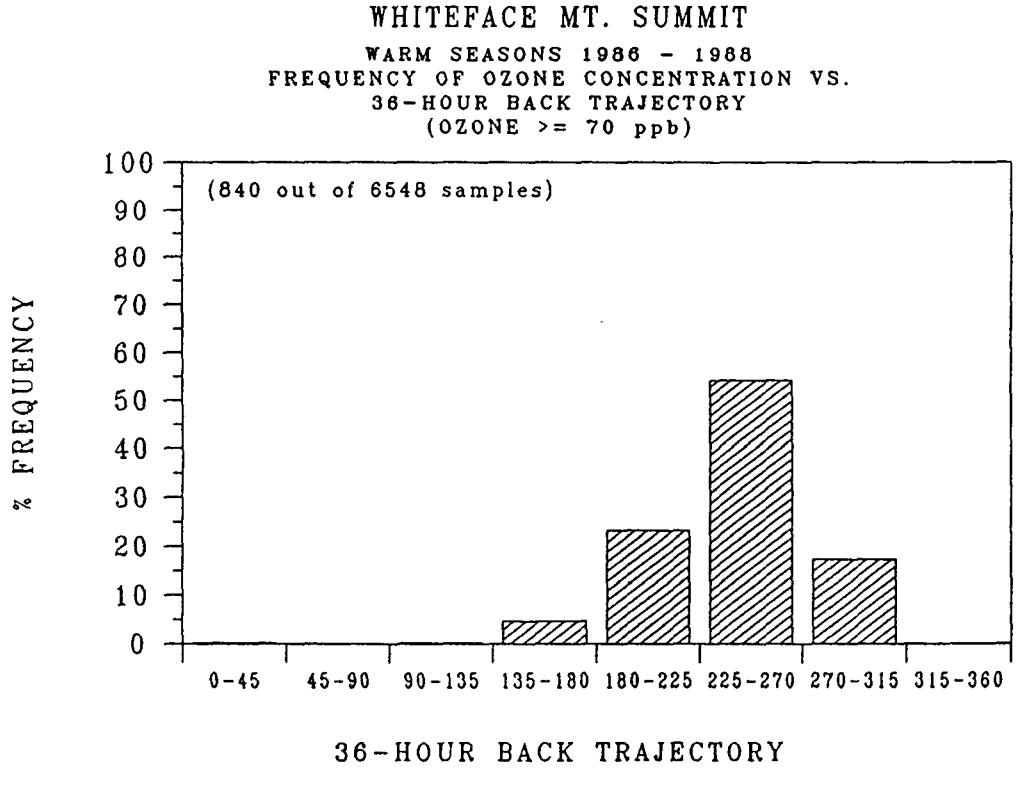


Figure 5-14

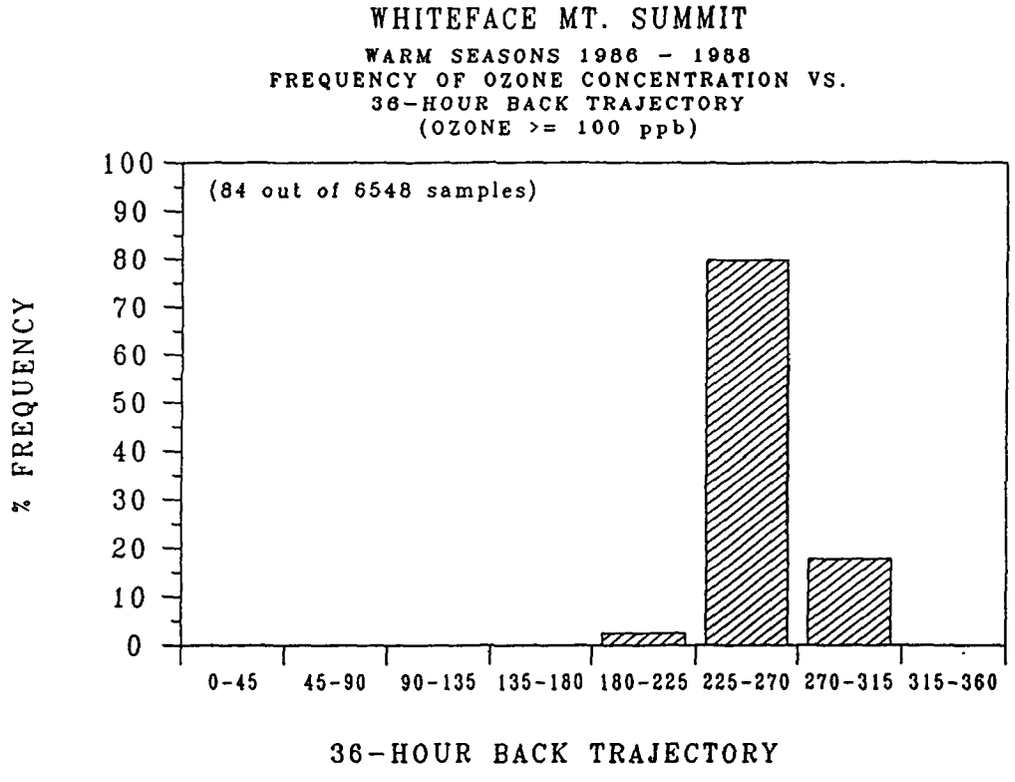


Figure 5-15

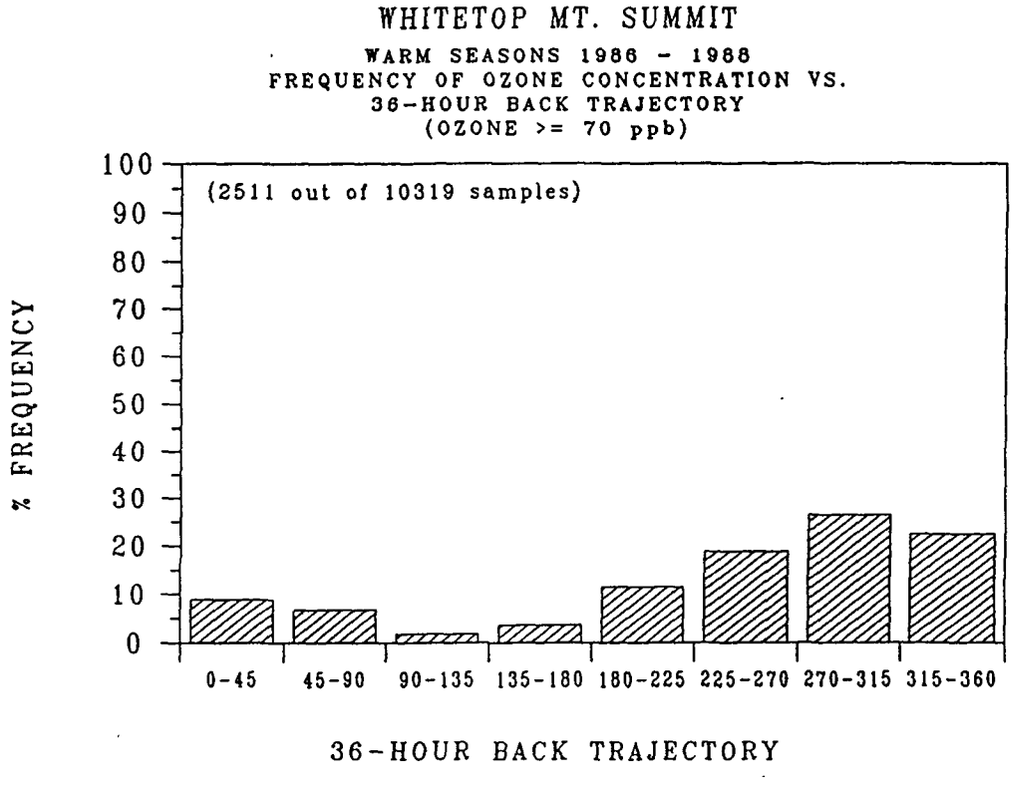


Figure 5-16

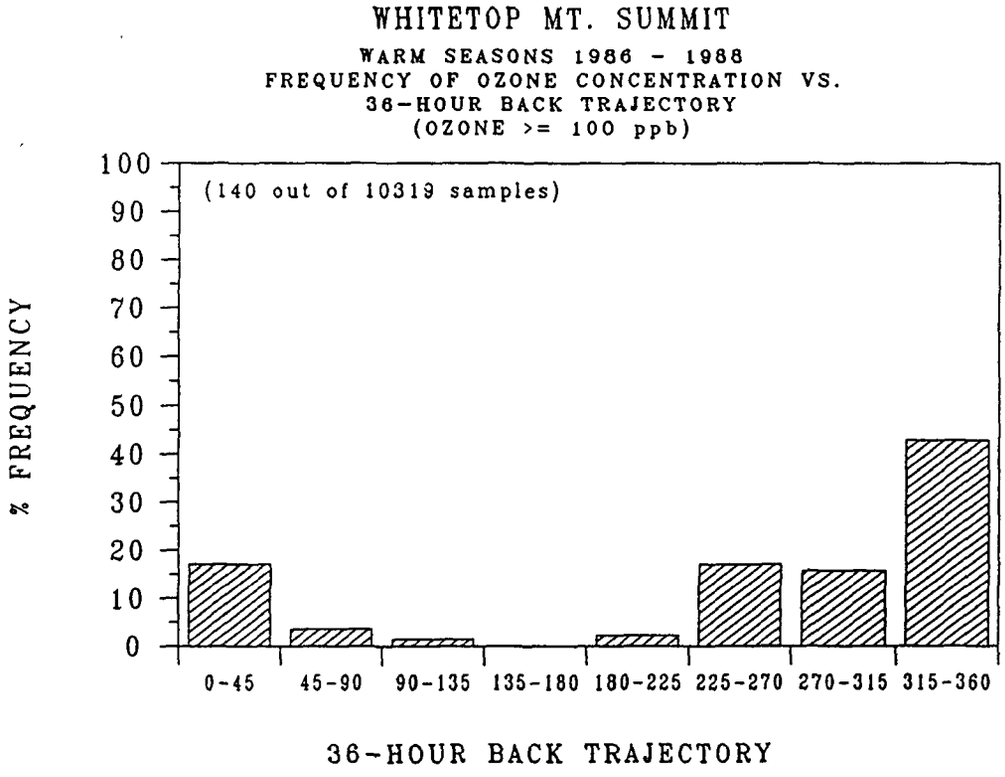

WHITETOP MT. SUMMIT
WARM SEASONS 1986 - 1988
FREQUENCY OF OZONE CONCENTRATION VS.
36-HOUR BACK TRAJECTORY
(OZONE >= 100 ppb)
(140 out of 10319 samples)
% FREQUENCY
100
90
80
70
60
50
40
30
20
10
0
0-45
45-90
90-135
135-180
180-225
225-270
270-315
315-360
36-HOUR BACK TRAJECTORY

Figure 5-17

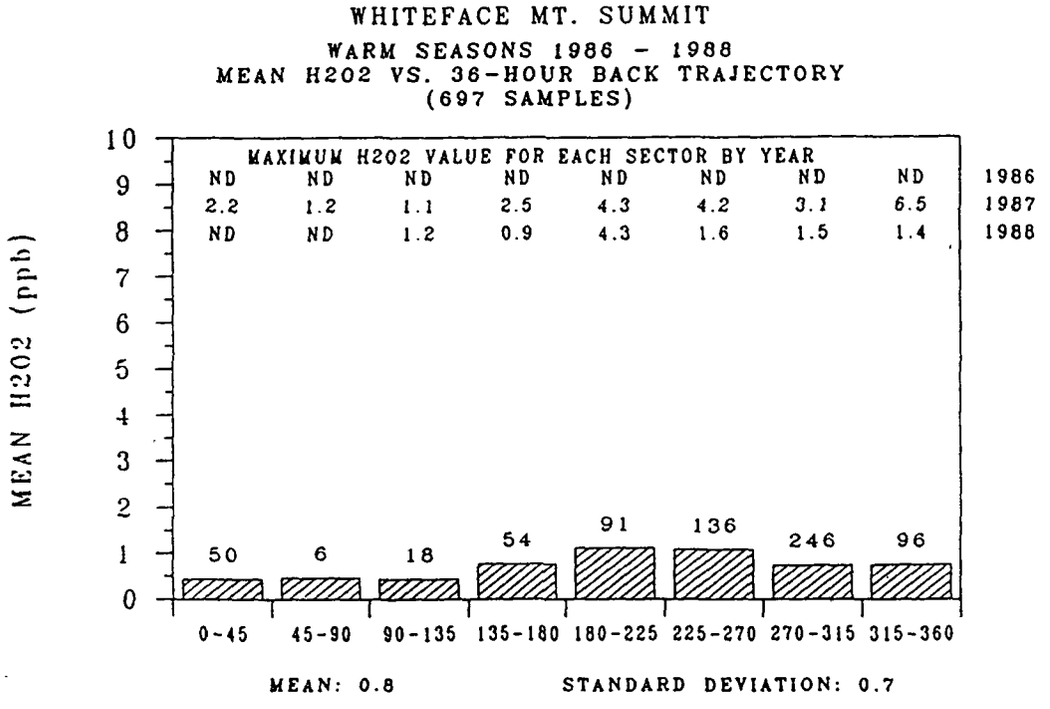


Figure 5-18

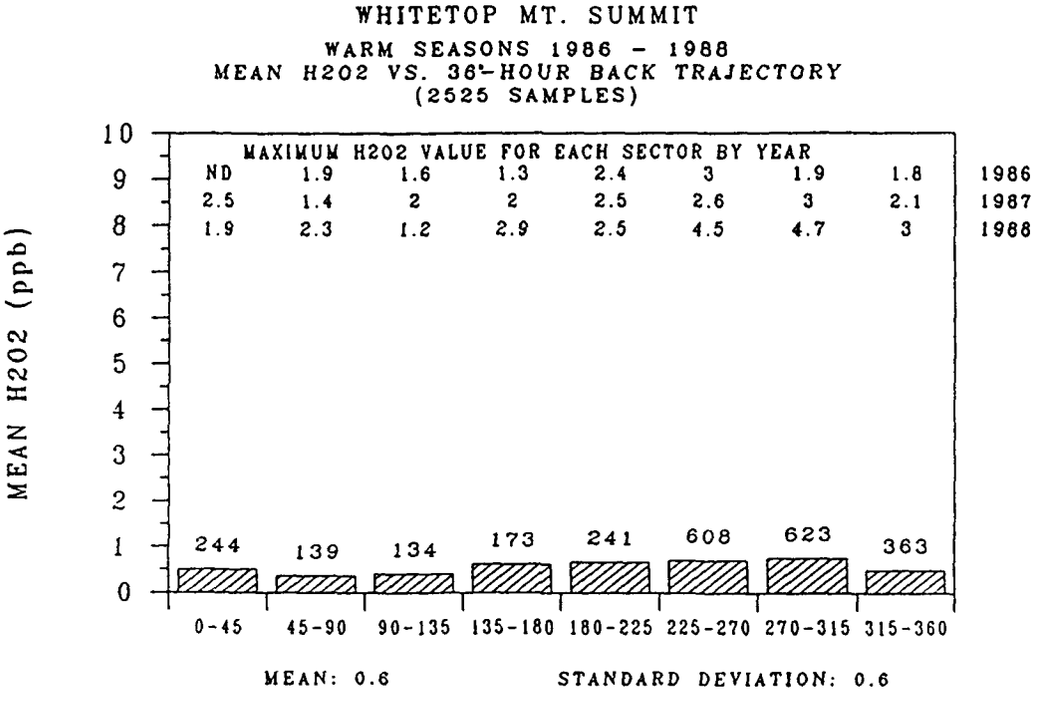


Figure 5-19

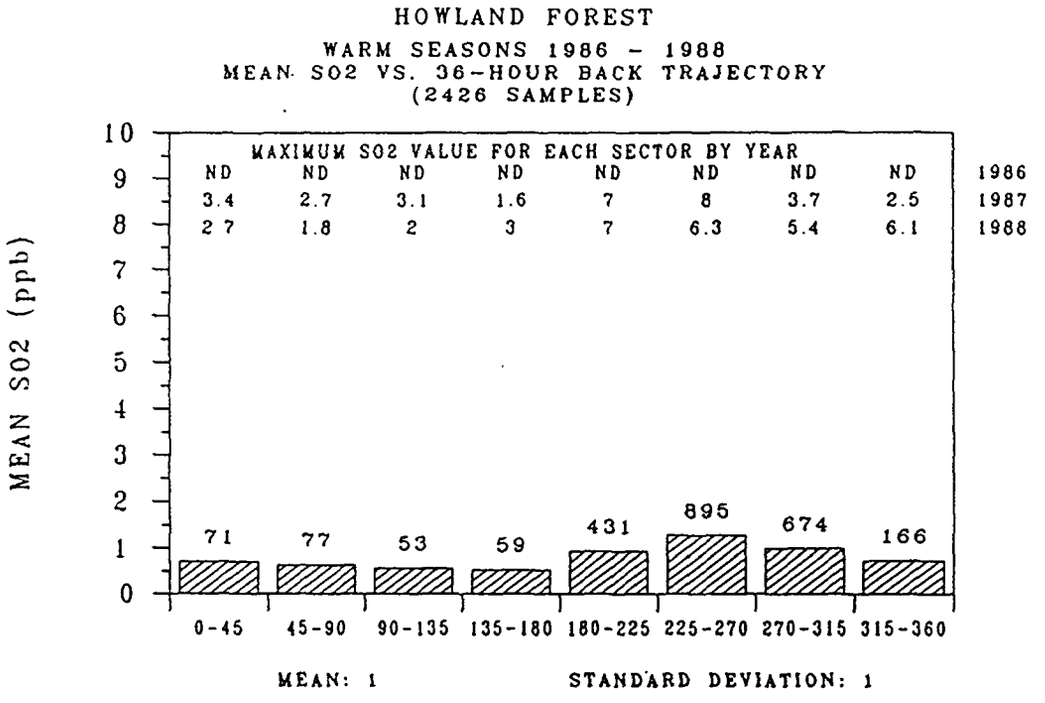


Figure 5-20

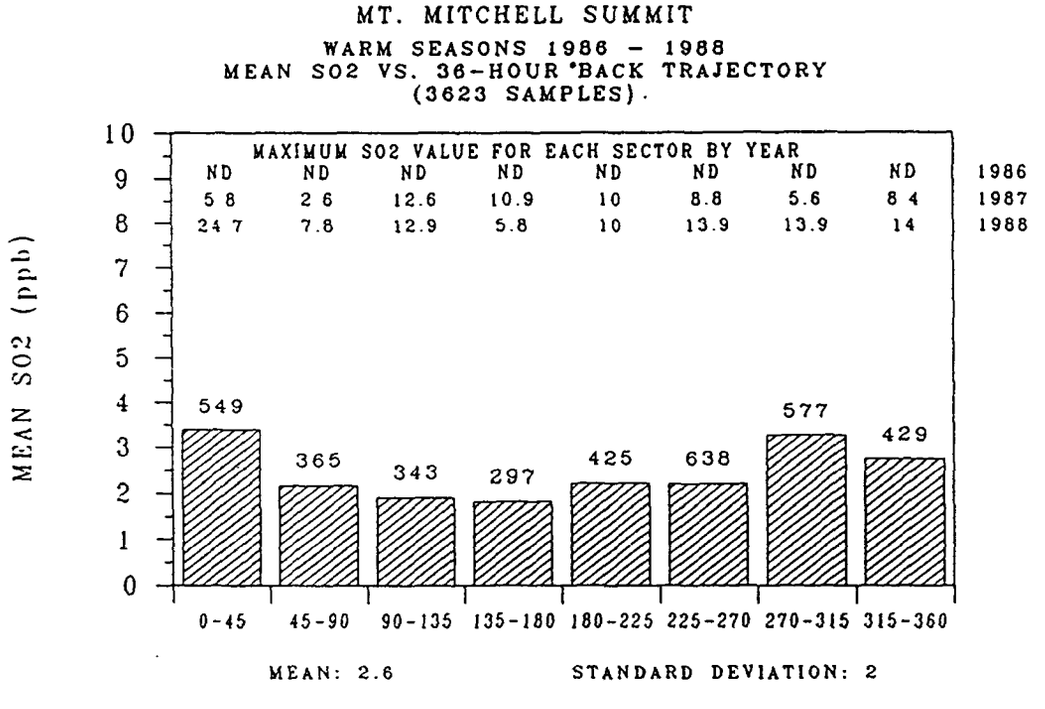


Figure 5-21

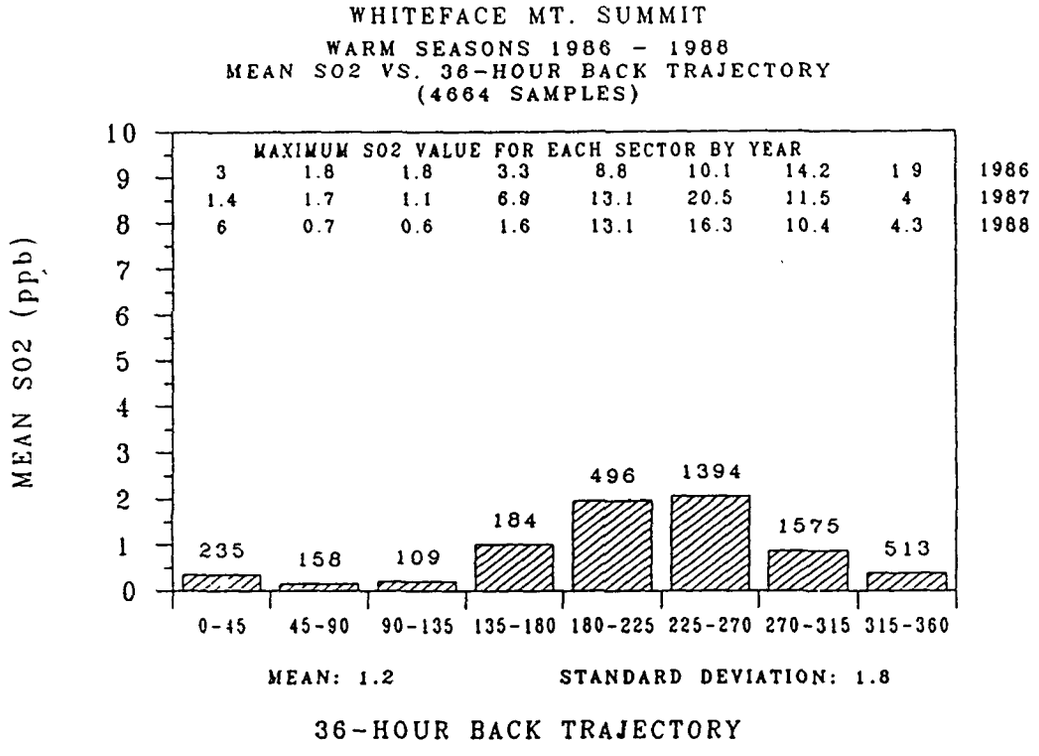


Figure 5-22

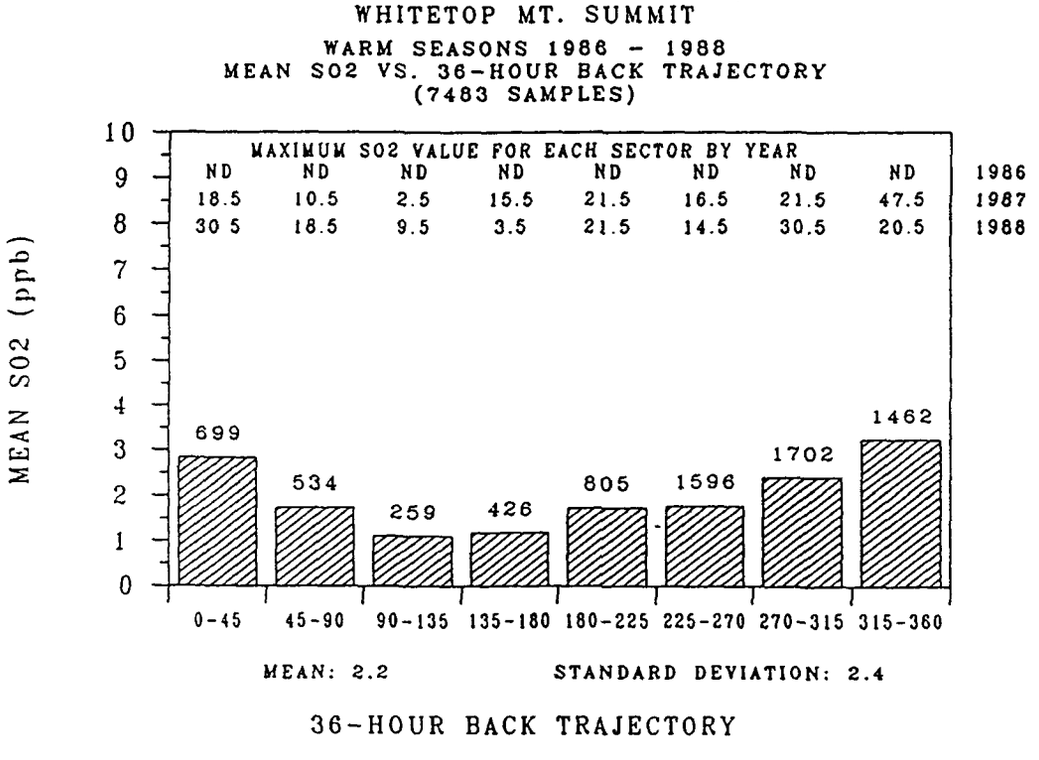


Figure 5-23

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