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Atmospheric Science Research at Whiteface Mountain, NY: Site Description and History

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ABSTRACT

Whiteface Mountain, with an elevation of 1483 m above sea level, is a relatively low mountain by global standards. At the same time, the summit is some 90 m above the tree line and it is the fifth highest peak in the Adirondack Mountain Range of New York State. Whiteface Mountain is set apart from the other Adirondack High Peaks, providing an ideal location for many types of atmospheric measurements. The geographical location in the northeastern U.S., the lone massif character of the mountain, and the fact that the summit is very often enveloped in cloud has made the observatory an attractive place for scientific research. A four story building specifically for the purposes of scientific research and fire monitoring was built on the summit in 1971. The headquarters of the Whiteface Mountain facility, the Marble Mountain Lodge, is perched on the shoulder of the massif at an elevation of 604 m a.s.l. In some cases the same measurements are made at the two locations to explore the two different but geographically close environments. A summary of past and current measurement activity at both locations is presented in the paper along with selected examples of data sets, analyses, and applications. Important research in the areas of forest ecology, cloud water chemistry, precipitation chemistry, reactive trace gases, and airborne particulate matter are reviewed. Collected data sets for temperature and ozone at the summit are presented, as well as research linking the measured gaseous SO_2 to acid deposition at this location. In addition, an example is given using this long-term data for both gases and particulate matter that helps to establish the accountability of air pollution regulations in the control of sulfur oxides.

Keywords: Mountain sites; Cloud chemistry; Precipitation chemistry; Air chemistry; Particulate matter.

INTRODUCTION

Mountaintop locations offer numerous features and scientific advantages which make them desirable for the purposes of collecting scientific data. While astronomical observatories may first come to mind, there are also a number of iconic weather and atmospheric observation related mountaintop observatories, such as Mauna Loa in Hawaii (Hoffman *et al.*, 2009; Dlugokencky *et al.*, 2014), Mount Fuji in Japan (Watanabe *et al.*, 2006), Jungfraujoch in Switzerland (Li *et al.*, 2005; Leuenberger and Flueckiger, 2008), and others. Many of these locations, including Whiteface, have decades and more of history and scientific observations. The long term data sets produced at these mountain sites are a cultural and scientific resource of great value for many types of research (HEI, 2003; Cooper *et al.*, 2012). For example, there is an increasing interest in climatic and ecological research at the world's mountain sites as the scientific community works to understand the many implications of climate change. As noted in the IPCC AR5 report, mountain regions are highly temperature sensitive, and increasingly vulnerable to high impact extreme events under a warming climate (Cramer *et al.*, 2014). From an experimental scientific perspective, mountain sites are quite often the ideal locations for many types of cloud related research, in particular the connections leading from aerosols to clouds and their effects on radiation and climate (Friedman *et al.*, 2013; Hammer *et al.*, 2015).

For the purposes of atmospheric chemistry and air pollution, mountaintop sites have a number of advantages. Mountaintop observatories can sometimes during the course of a day be disconnected from the planetary boundary layer air, which means there is less local influence on the measurements. This can happen regularly or less frequently,

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depending on the elevation and details of the location. The free troposphere is more regionally representative than locations that sample only the surface boundary layer, so measurements taken at mountaintop sites have greater regional representativeness (Andrews *et al.*, 2011). Along these same lines, mountaintop sites are often ideal receptor sites for source receptor studies (Grosjean and Bytnerowicz, 1993; Zhang *et al.*, 2009; Li *et al.*, 2005).

This paper and its companion papers will describe many aspects of the Whiteface Mountain research station and the observations carried out there. This first paper will present background information on the site, some aspects of the site's history and measurement programs, and examples of specific research results from the location. The second paper (Brandt *et al.*, 2016) will go into more detail on the measurements of trace gases, including ozone, which have been performed for many decades. The third paper (Schwab *et al.*, 2016) will detail measurement activity in the areas of aerosols and particulate matter, and in cloud water and precipitation chemistry. The second and third papers will concentrate mainly, but not entirely on current and ongoing measurements.

SITE LOCATION AND GEOGRAPHY

The Atmospheric Sciences Research Center (ASRC) Whiteface Mountain Field Station is located in the Northeastern quadrant of the Adirondack Park in northern New York State, and shown in Fig. 1. The station operates two environmental monitoring sites at this location which resides in the town of Wilmington, New York. The Adirondack Park in upstate New York covers roughly 2.4 million ha and is the largest publicly protected area in the contiguous United States. Approximately 20% of the park is at elevation above 600 m. Whiteface Mountain is located in the northeastern ("High Peaks") region of the park, and is climatologically downwind of large point sources and metropolitan areas in western New York, the industrial Midwest, and Canada. As there are few large population centers in the immediate region, the location is well-suited to characterize the impacts of transport and aging on air quality and deposition in New York and the northeast U.S.

The Marble Mountain Lodge site is located on the eastern shoulder of the Whiteface Massif at Latitude N 44°23'35" and Longitude W 73°51'33" with an elevation of 604 m above sea level. The site is situated in a heavily wooded area consisting primarily of a mixed Northern Hardwood forest type. The site has an eastern slope aspect and well drained glacial till soil type. The depth to bedrock is approximately 32 m. The bedrock material has been characterized as Metanorthosite bedrock (McLelland and Whitney, 1990). Numerous glacial erratics can be found in the vicinity of the site. The Marble Mountain Lodge site was established in an area that was previously disturbed during the construction of the Marble Mountain Ski center during the late 1940's.

The ASRC Whiteface Mountain Summit Observatory is located on the peak of Whiteface Mountain, Latitude N 44°21'58" and Longitude W 73°54'10" at an elevation of 1,483 m above sea level. Compared to the other Adirondack high peaks, Whiteface is relatively isolated. Except for Esther and Lookout Mountains which are in effect sub peaks of the Whiteface massif, the nearest 1200 m peaks are more than

Fig. 1. Map of the Northeast U.S and portions of Canada showing the location of the Whiteface Mountain Observatory.

16 km to the South. The windswept summit of Whiteface is above tree line and comprised primarily of Anorthosite bedrock with patches of thin soils. These soils are composed mainly of organic material and classified as Histosols with some mineral soils classified as Spodosols. The microclimate at the summit of Whiteface is one of the coldest in the Eastern United States. The short growing season and high winds at the summit limit the type of vegetation that can survive there. The dwarfed alpine vegetation is described as late Pleistocene flora that followed the glaciers northward 11,000 years ago (Holway *et al.*, 1969).

BRIEF HISTORY OF ATMOSPHERIC MEASUREMENT ACTIVITY

Completion of the Whiteface Mountain Veteran's Memorial Highway in 1935 paved the way for much easier access to the mountain's summit. The Veterans Memorial Highway brings road access to within 92 vertical meters of the summit, and a tunnel through the mountain and an elevator provide personnel and equipment a secure route to the Observatory building on the top of the mountain. Organized atmospheric measurement activity at Whiteface was established in 1937 as a cooperative project between Rensselaer Polytechnic Institute, New York University, the Whiteface Mountain Highway Commission, and the Weather Bureau. These universities, in conjunction with the Weather Bureau (which became the National Weather Service in 1970), continued their activity at Whiteface until 1946, providing the first extensive data record of meteorological parameters at the measurement location. After this activity, there was hiatus of some ten years before renewed observations at the site. This activity is the first entry in Table 1, which lists the timeline of major measurement programs at the two Whiteface Mountain measurement locations.

In 1957 Vincent Schaefer began working with the Whiteface Mountain Authority and the new Whiteface Ski Center to establish a Museum of the Atmosphere program on the mountain. Dr. Schaefer received support from the Whiteface Mountain Authority to purchase and install a wide array of meteorological instrumentation, and with the assistance of colleague Ray Falconer, these instruments were put into service on the mountain. These measurements in the late 1950's and early 1960's were done with significant input from the Whiteface Ski Center, and included the posting of detailed daily weather information at the main Ski Center building.

1961 saw the founding of the ASRC and the establishment of Whiteface Mountain as the pre-eminent field station of the new research center. ASRC, although housed at or near the Albany campus, was established as a system-wide resource for the State University of New York (SUNY). The research station has been open to projects from other SUNY schools, as well as outside universities and organizations. ASRC scientists and staff have maintained a constant presence at Whiteface since its 1961 founding due to a large extent in the early years to the guiding hands of Vince Schaefer and Ray Falconer.

Table 1. Timeline of Major Measurement Activity at Whiteface Mountain.

Dates	Activity	Location	Organization (s)
1937–1946	Temperature Measurements	Summit Observatory	Weather Bureau, RPI, NYU, WMHC
1957–	Meteorological Measurements	Summit Observatory	ASRC
1963–1978	Aerosol Particle Number Concentration	Summit Observatory and MML	ASRC
1973–	Ozone Concentration	Summit Observatory	NYSDEC
1975–	High Volume Aerosol Samples (Total Suspended Particles)	Summit Observatory	NYSDOH
1976–	Cloud Collection (seasonal)	Summit Observatory	ASRC/EPA/ALSC
1976–	Precipitation Collection	MML	ASRC/NYSDEC/NADP
1984–	Ozone and other Pollutant Gases	MML	NYSDEC/ASRC
1988–	Pollutant and Ozone Precursor Gases	Summit Observatory	ASRC
1999–	$PM_{2.5}$ Mass Concentration (Filter Samples and Continuous)	MML	NYSDEC
2001–	$PM_{2.5}$ Chemical Speciation (Filter Samples)	MML	NYSDEC

Notes: when no end date is given, the specified activity continues to present.
Abbreviations used in Table
ALSC - Adirondack Lake Survey Corporation;
ASRC - Atmospheric Sciences Research Center, University at Albany, State University of NY;
EPA - Environmental Protection Agency (United States);
MML - Marble Mountain Lodge;
NADP - National Acid Deposition Program;
NYSDEC - New York State Department of Environmental Conservation;
NYSDOH - New York State Department of Health;
NYU - New York University;
RPI - Rensselaer Polytechnic Institute;
WMHC - Whiteface Mountain Highway Commission.

In 1970 the permanent research observatory building at the summit was completed, replacing the old fire tower on the site. This building was a major advance for the research programs carried out at the summit, and more extensive and long-term measurements became possible. Temperature and meteorological measurements continued throughout this period, as did aerosol measurements. As noted below, the current time series of long-term trace gas measurements began in 1973 with the measurement of O_3 by the New York State Department of Environmental Conservation (NYSDEC). A few of the major research areas since the 1970's are highlighted in the section below.

Whiteface Mountain Ecology

Whiteface Mountain has long been recognized as an ideal location to study the interaction between the mountain environment and the organisms that live there. The harsh climate and short growing season are major factors that determine the species composition and life cycles of these organisms. Many scientists and ecologists have used Whiteface as a living laboratory to study the relationships between the mountain environment and the plants and animals that are able to survive in such harsh conditions (e.g., Reilly, 1963; Scott *et al.*, 1984; Lovett and Kisman, 1990; Schmull *et al.*, 2002).

A relatively small percent of the Whiteface Mountain area has been influenced by man. The greater portion remains in an undisturbed and natural state. These important conditions can be expected to prevail because this land is part of the Forest Preserve of New York State, designated in the state constitution to be preserved in a "forever wild" state for the people and for the future. In 1962 the Whiteface Mountain Site Manager and ASRC Director wrote, "These fortuitous circumstances provide contrasting, yet controlled, conditions for scientific approaches to the phenomena of the mountains. This stability of expected conditions makes possible a wide range of scientific operations, some of which may require years and for which evaluation of successive changes, and those of contrasting influences is desirable" (Falconer and Barry, 1963). The studies described here and in the companion papers (Brandt *et al.*, 2016; Schwab *et al.*, 2016) highlight some of the fruits of the scientific activity made possible in part by conditions referred to above.

The lack of large scale disturbance or land use changes on Whiteface have allowed for several resurveys to be conducted. In a study by Holway *et al.* (1969), 182 forest stands were located by both selective and systematic means and sampled for standard ecological parameters of frequency, density and basal area. Under the systemic stand selection method, elevation and slope aspect were the determining factors in site selection. Selective stand location was used to increase the sample size of under sampled association types. The initial data were collected during three summer field seasons from 1964–1966. These plots were revisited during the late 1980's during the Forest Effects Response Program, conducted at Whiteface during the 1986–1988 field seasons. The Project Manager for this joint US Forest Service/Environmental Protection Agency (USFS/EPA) program was John Battles from the University of Pennsylvania. The primary objective of this program was to investigate the deterioration of Red Spruce at high elevations in the Eastern United States and the possible relation to air pollution. The project conducted forest surveys of Whiteface above 700 m to characterize the vegetation, soils, pathogens and land use history of the mountain. These plots were visited yet again in 2012–2013 by Jay Wason from SUNY ESF, working under the direction of Dr. Martin Dovciak. The report "Climate change impacts in high elevation northeastern boreal forest plant communities" (Wason, 2013) was released in 2013. It is important to note that these long term data sets produced at Whiteface Mountain increase in value over time.

The Electric Power Research Institute/Oak Ridge National Laboratory (EPRI/ORNL) Integrated Forest Study was a four year project conducted from 1985–1988 at Whiteface and 16 other sites representing a range of forest types, geographical and topographical locations as well as proximity to pollution sources (Johnson and Lindberg, 1992). The principal objective of this study was to evaluate the role of atmospheric deposition in producing changes in forest nutrient cycling. Intensive sampling and monitoring sites were established in a Red Spruce/Balsam Fir/White Birch forest type between 970–1100 m in elevation. Deposition of particulate matter, gases, water and ions to the forest canopy were determined by a combination of direct measurements and atmospheric modelling techniques. The movement and storage of water and ions within the forest ecosystem was tracked by continuous monitoring of water passing through the canopy and individual soil horizon.

The ecological studies conducted at Whiteface Mountain have yielded clues about the very complex relationships that exist between the organisms that live on Whiteface and 1) the mountain's climate, 2) the physical environment of the mountain, and 3) human influences such as anthropogenic air pollution. It is important to understand that this process of understanding is not static but evolving over time. Much remains to be learned about the ecology of Whiteface Mountain. The primary focus in the 1980's was acid deposition, while in the 2000's there is greater concern about anthropogenic climate change.

The History of Cloud Water Collection at Whiteface Mountain

In 1976 the ASRC began a routine program of continuous cloud water collection and pH analysis at Whiteface Mountain (Falconer and Falconer, 1980). The ASRC cloud water collector consisted of two polypropylene disks separated and supported by several polypropylene rods. A continuous strand of Teflon FEP-fluorocarbon fiber was strung between the disks at 3 mm intervals along their periphery. A plastic funnel was used to direct the cloud water sample into the summit observatory (Falconer and Kadlecek, 1981).

The Mountain Cloud Chemistry Project (MCCP) was in operation from 1986 to 1989 and was implemented by the Forest Response Program of the National Acid Precipitation Assessment Program (NAPAP) and sponsored by the EPA (Mohnen and Kadlecek, 1989). The principle objective of

the MCCP was to characterize the exposure of montane forested ecosystems to atmospheric inputs. The MCCP was specifically designed to provide reliable information on the chemical, physical and meteorological exposure of Spruce-Fir forests. The data collected by the MCCP have been used in the NAPAP Integrated Assessment to evaluate the role of airborne chemicals in the changing condition of forests (NAPAP, 2005).

The Mountain Acid Deposition Program (MADPro) was a multi-year warm season study of the deposition of air pollution to high elevation forests in the eastern U.S. MADPro was a part of the Clean Air Status and Trends Network (CASTNet), a national monitoring network mandated as part of the Clean Air Act Amendments of 1990 to determine the effectiveness of emission reductions. The EPA established CASTNet to provide data and determine relationships between emissions, air quality, and ecological effects. The two main objectives of MADPro were to develop cloud water measurement systems useful in a network monitoring environment and to update the Appalachian mountain cloud water concentration and deposition data collected by the NAPAP during the 1980's. Cloud water samples were collected at Whiteface Mountain from 1994–2000 under MADPro. Two additional automated cloud water collection sites were operated under MADPro from 1994 to 1999 - Whitetop Mountain, Mt. Rogers National Recreational Area, VA; and Clingman's Dome, Great Smokey Mountain National Park, NC/TN. (CASTNET, 1996). Whiteface is the last of these three original MADPro sites still in operation, albeit in a different incarnation – now sponsored by the NYSDEC and New York State Energy Research and Development Authority (NYSERDA). The Whitetop site closed after the 1999 sampling season, and Clingman's Dome closed after the 2011 sampling season.

Depending on weather conditions, cloud collection at Whiteface Mountain is initiated on or around June first, and runs through the end of September. The Adirondack Lakes Survey Corporation (ALSC) began collecting cloud water samples at Whiteface Mountain in 2001 and continues to operate the cloud collection program at Whiteface Mountain. This work is presented in more detail in Schwab *et al.* (2016). Funding for the cloud collection program is provided primarily by NYSERDA with support from the NYSDEC Division of Air Resources. Summit observatory space and logistical support are provided by ASRC.

Precipitation Collection at the ASRC Whiteface Mountain Field Station

The ASRC Whiteface Mountain Field Station was one of nine rural precipitation chemistry stations established under the Muti-state Power Production Pollution Study (MAP3S) network (Dana, 1977, 1987). Pacific Northwest Laboratory (PNL) was charged with the operation of a research network of separately funded precipitation collection sites in the Northeastern United States. The responsibilities of PNL included site audits, chemical analysis of precipitation event samples collected at the network sites for all species of interest to the Interagency Task Force on Acid Precipitation as well as the reporting of these analyses to MAP3S participants, the MAP3S data base, and the EPA Acid Deposition System data base. Precipitation sampling for the MAP3S project began at the ASRC Whiteface Mountain Field Station on October 11, 1976. The MAP3S study was a precipitation monitoring network that collected samples year round on an event basis. The sampler was located at the Marble Mountain Lodge site and had resulted in the collection of 1000 samples by 9/29/86. Sample collection is ongoing under the aegis of organizations listed in Table 1. Samples have been and are analyzed for H^+, Ca^{2+}, Mg^{2+}, P, PO_4^{3-}, NH_4^+, SO_4^{2-}, NO_2^-, NO_3^-, Cl^-, K^+ and Na^+.

The National Atmospheric Deposition Program (NADP) has monitored precipitation chemistry at predominantly non-urban sites since 1978. The NADP was established to track the impacts of fossil fuel combustion on wet and dry deposition across the US, and is a consortium of governmental (federal, state, local, tribal) and non-governmental organizations, colleges and universities, private research groups, and many volunteer site operators. The NADP's National Trends Network (NTN) collects weekly precipitation samples at more than 260 sites across North America and one site in Argentina. The NTN site at Whiteface Mountain at the Marble Mountain Lodge was established in July 1984 and is currently one of six NTN sites in the Adirondack Park, all of which are between 460–610 m above mean sea level.

As the NADP was expanding as a national network in the 1980s, New York State refined its own approaches to address acid rain and its impacts in sensitive ecosystems, including the Adirondack Mountains, Catskill Mountains, and Hudson Highlands. In 1984 the State Acid Deposition Control Act (SADCA) was passed, which required the reduction of SO_2 emissions from existing electric generating facilities and additional NO_x emissions from new facilities. It also established an environmental threshold value for wet SO_4 deposition at 20 kg/ha; at the time, it was assumed that deposition levels below this threshold would not have significant impacts on sensitive ecosystems. After the passage of SADCA, the NYSDEC initiated a complementary wet deposition network consisting of 20 sites in both urban and rural settings, including a sampler less than 10 m from the NTN site at Whiteface Mountain. The NYSDEC network operated until 2012, when it was discontinued to reduce monitoring redundancy and improve coordination and efficiency of deposition measurements across the region.

Air Pollutant and Trace Gases

While atmospheric chemistry measurements took place at Whiteface during the 1960's, the construction of the four story observatory building in 1970 paved the way for more extensive and sustained measurement activity. In cooperation with the NYSDEC, O_3 measurements at the summit began in 1973 (Mohnen *et al.*, 1977). This important measurement continues to this day. The O_3 data set from Whiteface summit has been used in a number of important and influential papers (Logan, 1989; Lefohn *et al.*, 1990; Aneja and Li, 1992; Cooper *et al.*, 2014).

Starting in 1986 and reaching full maturity around 1988 was the program measuring important precursor trace

gases for acid deposition and ozone formation. The first precursor measurements included CO and NO_y, but since the late 1990's year-round measurements NO, NO_2, and SO_2 have been added. These measurements continue to the present day and are highlighted in a companion paper (Brandt *et al.*, 2016). Whiteface Mountain measurement data were also included in two important studies of rural ozone production and transport in the 1990's (Parrish *et al.*, 1993; Trainer *et al.*, 1993). Starting in 2000, measurements of reactive trace gases were begun at the lodge site as well. These began with O_3 and SO_2, but now include CO and oxides of nitrogen. Gaseous atmospheric chemistry measurements remain a major activity and focus of the Whiteface Mountain operation.

Starting in the mid-1990's and continuing for about ten years ASRC also had a robust program for sampling nonpolar hydrocarbons (C_2–C_{10}) at Whiteface (Gong and Demerjian, 1997). Samples were collected into SUMMA polished stainless steel canisters at different times of the day (2–4 times per day depending on the site and season) at the summit during summer months and year round at the lodge site. The sample canisters were sent to ASRC in Albany for analysis, where a GC-FID system identified and quantified as many as 51 target hydrocarbon species.

Particulate Matter

Aerosol research was a major focus of ASRC activity in the 1960's and 1970's, and there were measurements at Whiteface Mountain as well as locations around the world. Some work highlighting the measurement of condensation nuclei, or Aitken nuclei at Whiteface Mountain summit and/or lodge locations include papers by Tester (1964), Hogan (1966), and Hogan (1970). Under the guidance of Ray Falconer, ASRC for a time carried out a project that involved daily monitoring of the condensation nuclei number concentrations at some fourteen locations across New York State, including Whiteface (Falconer, 1971). Collection of filter samples of particulate matter at Whiteface started in 1975 by researchers at NYSDOH as described above. This sampling program of Total Suspended particles (TSP) continues to this day and has produced an invaluable archive of the history of this airshed, including all of the sulfate and selenium measurements presented below.

Using these filter samples, Husain *et al.* (1977) conducted measurements of cosmogenic ^{7}Be (half-life, 53.28 d) in July 1975 at Whiteface Mountain summit as a tracer of stratospheric O_3. The measurements were continued from June 15 through August 31 in 1977. The data were used to estimate stratospheric component of O_3. Subsequently, a new sampling system was installed, and aerosols were collected daily beginning on July 1, 1978 through 1988. From 1989–2005 the sampling duration was changed from 24 to 48 hrs. Currently, the program continues with daily samples. The filters have been analyzed continuously for SO_4^{2-} by ion chromatography (IC), and trace elements using a variety of techniques: instrumental neutron activation analysis (INA), atomic absorption spectrophotometry (AAS), and inductively coupled plasma mass spectrometry (ICPMS). A conscientious effort was made to keep the methodology, i.e., filter media, and analytical methodology same throughout the study as one of the goals was to determine even small changes occurring in concentrations owing to the air pollution control regulations. The studies covered: (1) resolution of stratospheric and tropospheric O_3 Using ^{7}Be as a tracer (Husain *et al.*, 1977); (2) composition of ambient aerosols including SO_4^{2-}, and trace elements (Husain and Dutkewicz, 1990); (3) in-cloud oxidation of SO_2, and NO_2 (Husain *et al.*, 1991), and (4) long term trends in the variations of SO_4^{2-}, SO_2, and black carbon (BC) (Khan *et al.*, 2010).

In 1997 the EPA established an air quality standard for $PM_{2.5}$, defined as those particles with aerodynamic diameter less than 2.5 micrometers that are captured and measured gravimetrically. Shortly after the establishment of this new standard, NYSDEC installed a Federal Reference Method (FRM) filter sampler at the Marble Mountain lodge site. This is the "cleanest" site in their measurement network, and provides a baseline reference for clean background mass concentrations of $PM_{2.5}$. The measurements continue to the present, currently on a 1 in 6 day schedule. The lodge site also hosts filter sampling for chemical speciation, on the same 1 in 6 day sampling schedule as the FRM filters. In addition to the integrated filter samples, continuous $PM_{2.5}$ mass concentration is measured with a TEOM© monitor, and semi-continuous $PM_{2.5}$ sulfate is measured using a sulfate aerosol analyzer. Details of the current PM measurements at Whiteface are described in a companion paper (Schwab *et al.*, 2016).

Whiteface Mountain was one of the sites chosen by ASRC for its project award under Phase Two of the EPA PM Supersites program (Stanier and Solomon, 2006). The ASRC program was called "Particulate Matter Technology Assessment and Characterization Study – New York" (PMTACS-NY). The study included a month long intensive at the lodge location with upwards of 20 new measurement systems for PM and gaseous species. A number of papers resulted from this very extensive measurement program (Hogrefe *et al.*, 2004; Rattigan *et al.*, 2006; Ren *et al.*, 2006; Schwab *et al.*, 2006; Bae *et al.*, 2010)

EXAMPLE DATA SETS AND ANALYSES

Temperature

Temperature data, presented as the daily high and low temperature, are shown in Fig. 2 for the summit of Whiteface Mountain. As noted above, the data in the late 1930's and 1940's were collected by the Weather Bureau using manual methods, while all of the later data has been recorded by ASRC, mainly using bead thermistors. Unshielded and unaspirated sensors were used since the housings collect rime ice efficiently and make the measurements even more inaccurate than solar heating problems. The most recent two decades' data, using radiation shields, have been electronically recorded and stored – prior to that stripchart recordings were transcribed by hand. The hand-recorded data have been recently entered into data files and stored electronically. We are not able to discern any statistically significant trend in temperature at this location over the measurement period.

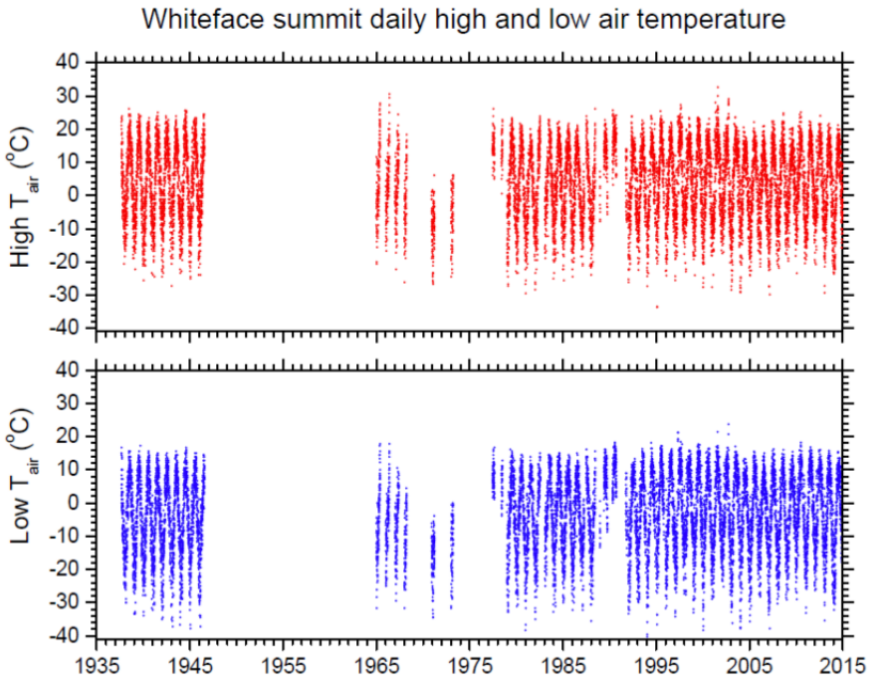

Fig. 2. Daily high and low air temperature at the Whiteface Mountain summit. The largest data gap occurs between old Weather Bureau data and the "modern record". Shorter gaps are due to equipment failure, construction, changes in funding support, etc.

Ozone

Fig. 3 shows the annual O_3 measurements from hourly averaged data at the summit from 1976 through 2014. The earliest measurements were made with an ethylene chemiluminescence detector, operated as a Federal Reference Method and following NYSDEC and USEPA QA/QC procedures. In 1985 the instrument was changed to an ultraviolet absorption instrument, also following NYSDEC and USEPA QA/QC procedures, but as a Federal Equivalence Method. Trace gas measurements will be discussed and presented more fully in a companion paper (Brandt *et al.*, 2016). Fig. 3(a) shows the annual means, medians, and interquartile ranges calculated using all valid hourly averages. Annual means are commonly used to assess trends and compare data sets from locations around the world (Cooper *et al.*, 2014). Fig. 3(b) shows the data displayed in terms of the annual average of the daily maximum 8-hour average ozone. For this calculation, first the hourly data are used to calculate each daily maximum 8-hour average, then the daily averages are combined to monthly, then annual averages. In accordance with USEPA guidelines (USEPA, 1998), 75% data completeness is required at each step, and this requirement is the reason for missing data in Fig 3(b). Perhaps the most impressive aspect of this data is the year-to-year variability, and variation between clusters of high O_3 years (late 1970's, late 1980's and early 2000's) and low O_3 years (early 1980's and the last four years – 2011–2014). While it may be tempting to attribute the recent low O_3 years to a downward trend over the past 10 or even 20 years, the full 39 year record looks to be a cautionary tale against over-interpretation of a trend for this complicated species.

In Situ Oxidation of SO_2 in Clouds

Sulfur dioxide is emitted into the atmosphere in copious amounts in the combustion of coal and other fossil fuels in northeastern US, and Canada. The predominant west to east wind flow results in transport of SO_2 to the Adirondack Mountain region. SO_2 is oxidized to H_2SO_4 in the atmosphere in gas phase at a nominal rate of ~1% per hour. However, the oxidation in cloud drops can be much faster, up to 1000% per hour, when H_2O_2 is present (Seinfeld and Pandis, 2006). In order to determine a quantitative relationship between SO_2 regional emissions and downwind H_2SO_4 deposition, it is important to determine accurately the rate of oxidation of SO_2 by H_2O_2, O_3, and other oxidants, and quantify the contributions from gas and aqueous phase oxidation. In the northeastern US, the low pH of cloud drops (Schwab *et al.*, 2016) results in H_2O_2 being the primary oxidant. Owing to the fact that cloud drops contain H_2SO_4 both from gas phase as well as aqueous phase oxidation, chemical measurements of H_2SO_4 cannot quantitatively resolve the amounts produced from the two processes. Husain (1989) developed a tracer technique to quantify the aqueous phase oxidation. Fig. 4 shows the process of SO_2 oxidation in the atmosphere.

The amount of SO_4 formed in cloud drops, (SO_{4in}) can be written by (Husain, 1989) Eq. (1):

$$(SO_{4in}) = (SO_4)_{cw} + \alpha/\beta[(SO_4/Se)_{aa}](Se)_{cw} \quad (1)$$

cw and aa represent concentrations in cloud water and ambient aerosols; α and β are scavenging efficiencies of SO_4 and Se aerosols, respectively, into cloud water. It should be noted that the only source of Se in cloud water is the scavenging from ambient aerosols, whereas SO_4 is both

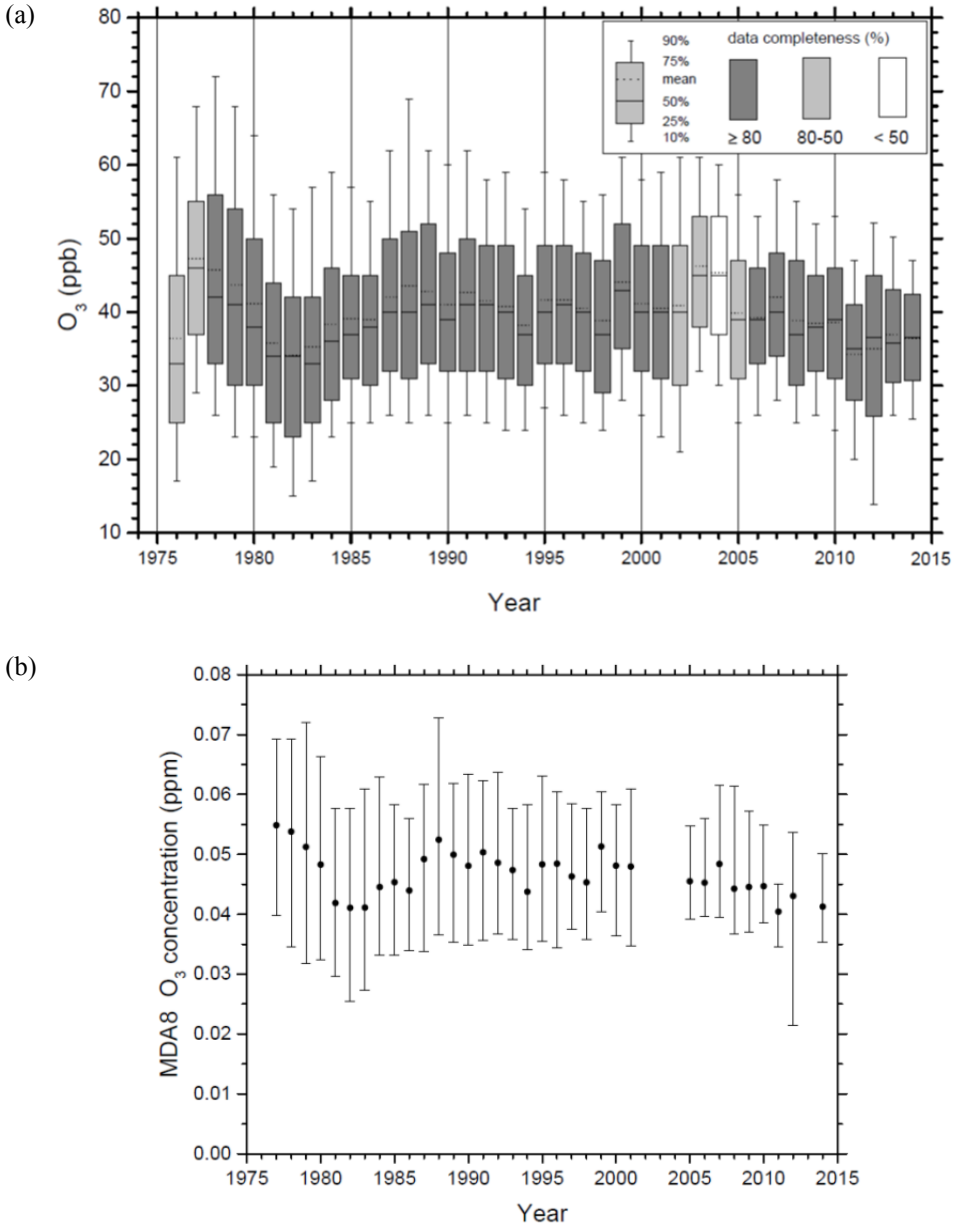

Fig. 3. a) Annual ozone measurements, compiled from hourly averaged data, at the Whiteface Mountain summit starting in 1976. The boxes display the interquartile range, and means and medians are also shown. b) Annual mean of monthly averaged maximum daily 8-hour ozone concentrations. A 75% data completeness guideline was used for all data (daily, monthly, and annual), in accordance with USEPA guidance.

scavenged from aerosols, and produced from oxidation in clouds drops. Subsequently, it has been shown that SO_4, and Se aerosols are scavenged with equal efficiency so that $\alpha/\beta = 1$ (Husain *et al.*, 1991). Burkhard *et al.* (1995) showed that As, and Sb can also be used as tracers.

From a series of studies of summer clouds at Whiteface Mountain, it was shown that ~25% of the aqueous phase SO_4 is produced from in situ oxidation (Husain *et al.*, 1991; Burkhard *et al.*, 1994; Dutkiewicz *et al.*, 1995). This amount could be much higher closer to the emission sources. The technique was extended to the study of SO_2 oxidation as a function of drop size in clouds and ground level fog (Rattigan *et al.*, 2001; Reilly *et al.*, 2001), and also to study oxidation of NO_2 to HNO_3 in clouds (Judd and Husain, 2000).

Acid Rain

Large quantities of fossil fuel have been consumed for many decades in the industrial Midwest which results in copious emissions of SO_2. As noted above, SO_2 is slowly oxidized to H_2SO_4 in gas phase, and much more rapidly oxidized in cloud drops. The mean residence time of SO_2 in the atmosphere is about a week (Seinfeld and Pandis, 2006), so it can travel thousands of kms. Sulfuric acid formed via oxidation quickly condenses on particulate matter (PM), or participates in new particle formation, and its

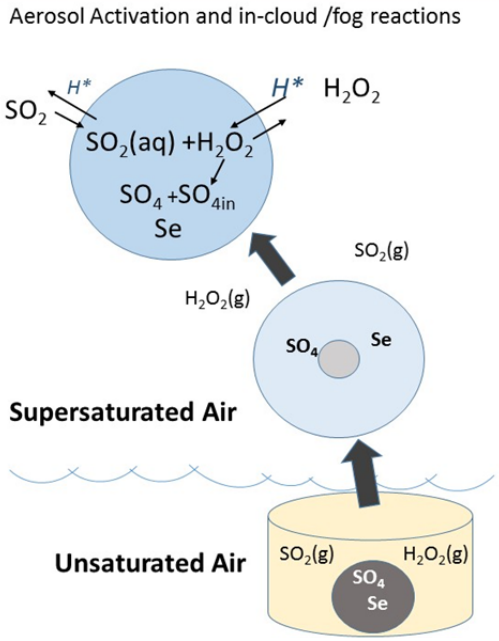

Fig. 4. Process of activation of aerosols in cloud drops, and oxidation of SO_2 by H_2O_2.

subsequent uptake into cloud and rain water is the primary path for acidification of precipitation and surface waters in the eastern US. Long-term data, of the type advocated by Falconer and Berry (1963), are required to establish the relationship between regional SO_2 emissions and downwind acid deposition. The data are needed to develop strategies to minimize acid deposition. Specifically, these data are collectively used to: (1) evaluate temporal and spatial trends of these species across the Northeast over the past 30 years; (2) study the impact of changes in regional emissions on the downwind concentrations; (3) investigate the question of linearity/nonlinearity between regional SO_2 emissions and acid deposition; and (4) develop an empirical model for sulfur transport along the Northeast corridor.

Daily aerosol collection at Whiteface Mountain summit observatory was initiated in 1978 (Parekh and Husain, 1981) and the program is ongoing. The aerosol samples collected at these sites have been archived and carefully stored in our laboratory. The concentration of SO_4^{2-} is measured using ion chromatography. The sampling and analytical measurement methodology has been diligently kept the same throughout to enable accurate determination of even small variations in SO_4^{2-} concentrations, and hence in SO_2 upwind emissions (Husain and Dutkiewicz, 1990; Dutkiewicz *et al.*, 2000; Khan *et al.*, 2010). SO_4^{2-} aerosol mass concentrations can be converted to effective mixing ratios, then combined with SO_2 measurements (see Brandt *et al.*, 2016) to obtain total sulfur (TS).

Total Sulfur (TS) and SO_2 Data

Since aerosol SO_4^{2-} is the secondary product of SO_2 oxidation, for the purpose of relating emissions of SO_2 with acid deposition, it is also necessary to track TS. The range for TS values is limited by the availability of appropriate SO_2 measurements. SO_2 was measured with TECO model 43s instruments operated by the NYSDEC at locations near each of the sites. For Mayville, a site in the western most corner of New York State near Lake Erie, data was used from a site in Westfield, NY that is only 9.6 km west northwest of Mayville. The area in between the sites is open farmland. SO_2 measurements commenced at Westfield in 1991. For Whiteface Mountain, data collected at the Marble Mountain Lodge site was used.

Regional SO_2 Emissions

Based on various analysis of backward in time air trajectories reaching the site (Parekh and Husain, 1982), air masses primarily arrive at Whiteface Mountain from the southwestern and northwestern quadrants so they must pass through the high SO_2 emission regions in the Midwestern US and Ontario Canada. The changes in the annual mean SO_4^{2-} at Whiteface Mountain and Mayville, NY were linearly related to the SO_2 emissions for the Industrial Midwest states (WI, IL, IN, MI, KY, TN, WV, and western PA) plus the emissions for Ontario Canada (MW + ONT) as demonstrated in Figs. 5 and 6 (Husain *et al.*, 1998).

In Fig. 5, SO_4^{2-} at WFM decreased sharply from 1979 to 1981 but then only varied slightly through 1991. The mean concentration for the 1992 to 1994 period is 23% lower, compared to the 1981 to 91 period. An additional 26% decrease was observed for the 1995 to 1997 period.

Although not shown, summit filters have been analyzed for sulfate concentrations through 2008 and work is proceeding on continuing the trend out to 2013. Sulfate concentrations from 1997 through 2005 exhibit no discernable trend and decreased slowly thereafter. The average sulfate mass concentration in 2008 was 1.41 μg m^{-3}. The scatter plots for sulfate and total sulfur at Whiteface Mountain versus the MW + ONT emissions in Fig. 6 show the linear relations between sulfur species and upwind SO_2 emissions.

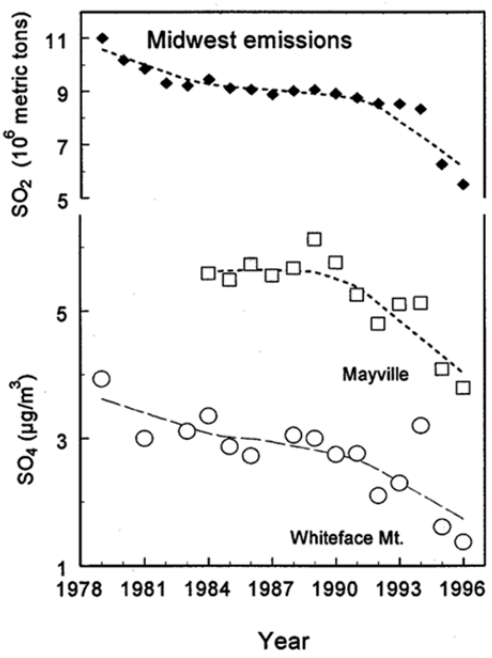

Fig. 5. Annual mean SO_4^{2-} conc. at Whiteface Mt. and Mayville, NY along with annual mean SO_2 emissions from Ont. Canada and the 8 US states upwind to NY State (PA, OH, IN, IL, WI, MI, KY, WV). Lines are a multiple regression smoothing added to aid the eye (Husain *et al.*, 1998).

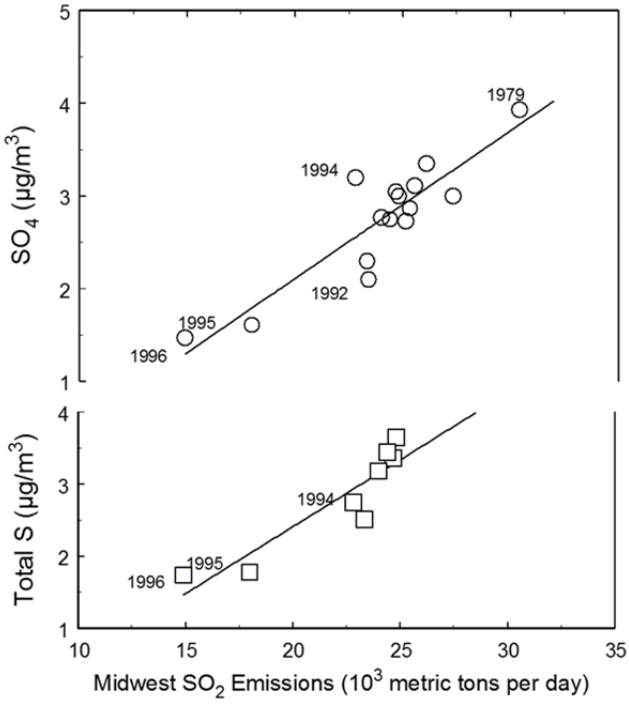

Fig. 6. Scatter plots showing annual mean SO_4^{2-} and TS at MAY and WFM versus the annual region's SO_2 emissions. The solid lines are linear regression fits to the data passing through zeros as explained in the text (Husain *et al.*, 1998).

Air Pollution Accountability

Another way to look at the transformation of SO_2 emissions to atmospheric concentrations of SO_2 and to deposition of fully oxidized SO_4^{2-} is shown in the three panels of Fig. 7. In contrast to Fig. 6, this figure (panel c) uses the SO_4^{2-} measured in precipitation chemistry at the Marble Mountain site as the deposition endpoint. These two methods of tracing the effects of lower emissions on the atmospheric endpoints of lower SO_4^{2-} in aerosols and precipitation are complementary and both are necessary to tell the complete story. The reduction in air pollution endpoint – reduction of acidic SO_4^{2-} deposition in this case

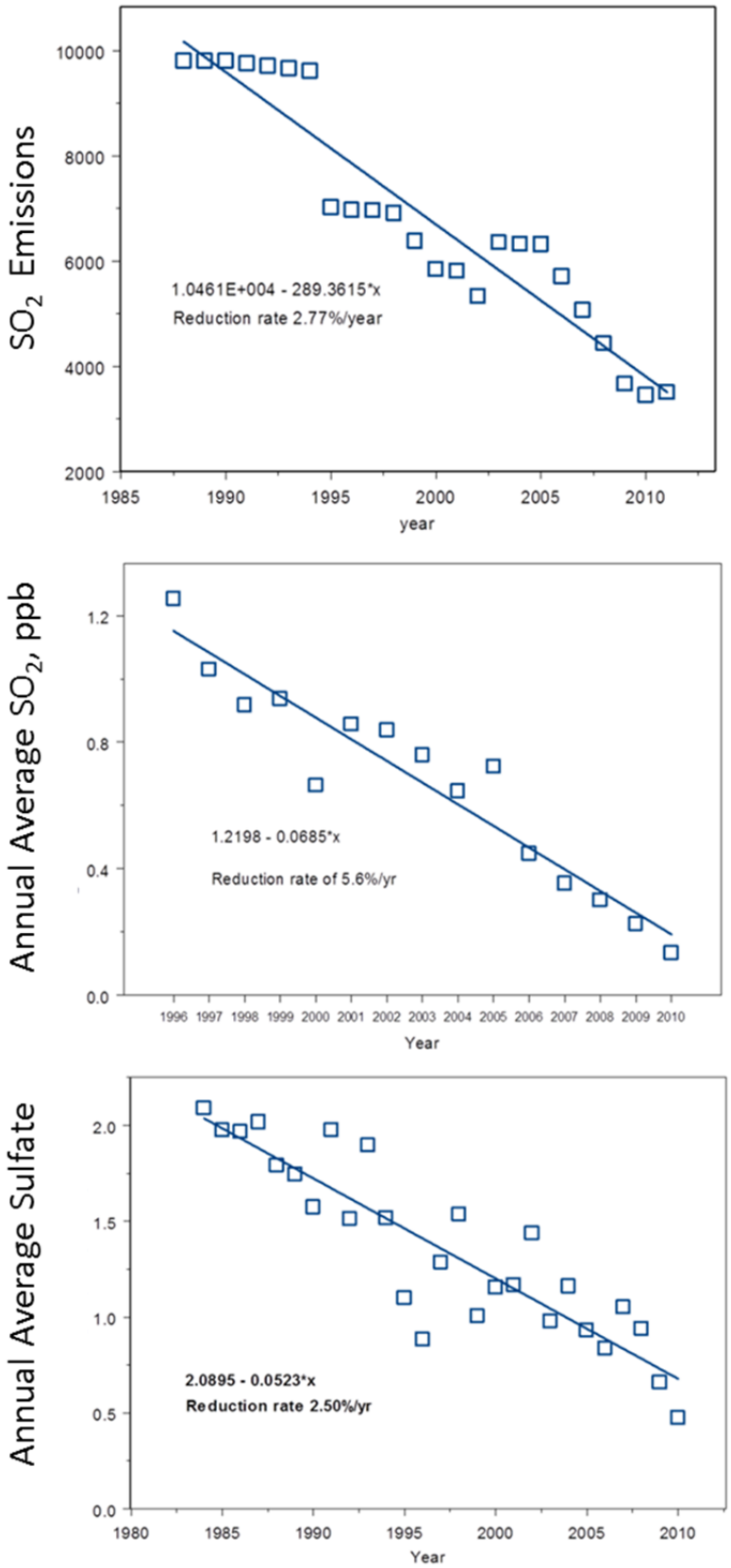

Fig. 7. Three steps along the accountability chain for air pollution due to atmospheric sulfur in the northeastern U.S. Panel a) shows the industrial emission of SO_2 in the Mid-Atlantic Northeast region in units of 1000 tons; panel b) shows the annually averaged gaseous SO_2 measured at the Whiteface summit observatory; and panel c) shows the volume weighted aqueous SO_4^{2-} measured in precipitation collected at the Marble Mountain Lodge site.

– provide the accountability check on the efficacy of air pollution regulation mandated by the Clean Air Act and subsequent Amendments.

SUMMARY

Whiteface Mountain and its lower elevation associated site, Marble Mountain Lodge, have proven to be unique sites for atmospheric measurements of cloud water, precipitation and air pollutants since the 1970's. These long term measurements are crucial in understanding the impact of distant pollutant emissions on the sensitive ecosystem of the Adirondacks. The data provide a direct link between acidic species and SO_2 emissions, a key factor in the accountability process. Measurements include various trace gas species, initiated at the summit in the 1980's, and currently carried out at both the summit and Marble Mountain Lodge sites. Numerous cloud water sampling intensives have occurred at the summit on in-cloud SO_2 oxidation, producing an important record of cloud water chemistry in the Adirondack region. Daily measurements of Total Suspended Particle (TSP) at the summit observatory started in 1978. The TSP filter samples have provided key information linking downward trends in particle SO_4^{2-} with mid-west SO_2 emissions.

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