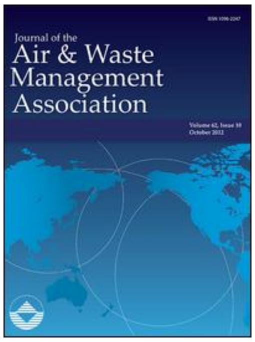

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Estimates of Cloud Water Deposition at Mountain Acid Deposition Program Sites in the Appalachian Mountains

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ABSTRACT

Cloud water deposition was estimated at three high-elevation sites in the Appalachian Mountains of the eastern United States (Whiteface Mountain, NY; Whitetop Mountain, VA; and Clingman's Dome, TN) from 1994 through 1999 as part of the Mountain Acid Deposition Program (MADPro). This paper provides a summary of cloud water chemistry, cloud liquid water content, cloud frequency, estimates of cloud water deposition of sulfur and nitrogen species, and estimates of total deposition of sulfur and nitrogen at these sites. Other cloud studies in the Appalachians and their comparison to MADPro are also summarized. Whiteface Mountain exhibited the lowest mean and median concentrations of sulfur and nitrogen ions in cloud water, while Clingman's Dome exhibited the highest mean and median concentrations. This geographic gradient is partly an effect of the different meteorological conditions experienced at northern versus southern sites in addition to the difference in pollution content of air masses reaching the sites. All sites measured seasonal cloud water deposition rates of SO_4^{2-} greater than 50 kg/ha and NO_3^- rates of greater than 25 kg/ha. These high-elevation sites experienced additional deposition loading of SO_4^{2-} and NO_3^- on the order of 6–20 times greater compared with lower elevation Clean Air Status and Trends Network (CASTNet) sites. Approximately 80–90% of this extra loading is from cloud deposition.

IMPLICATIONS

Clouds with origins in polluted areas of the eastern United States contain high concentrations of acidic ions. These clouds impact the Appalachian Mountains at elevations greater than 800 m. This phenomenon leads to large amounts of acidic deposition on vegetation and into soil. The deposition at these high-elevation areas may be 6–20 times greater than the deposition found at lower-elevation areas. Eighty to ninety percent of this deposition is from clouds. Acid deposition in these areas has been linked to forest damage and changes in soil composition and stream chemistry.

INTRODUCTION

Clouds impact the slopes of the Appalachian Mountains of eastern North America at elevations at or greater than 800 m. This phenomenon has been the subject of a number of studies.[1–5] Cloud water droplets in clouds with origins in polluted areas contain high concentrations of acidic and other dissolved ions.[6–8] A combination of factors, including high frequency of cloud immersion, high wind speeds, orographic enhancement of precipitation, and the large leaf areas of tree species typical of these environments, result in the flux of water and pollutants to these high-elevation ecosystems.[9] Cloud water pollution deposition can be the dominant form of deposition to high-elevation forests, exceeding wet and dry deposition.[10]

Two multisite, multiyear monitoring programs that focused on monitoring cloud chemistry were the Mountain Cloud Chemistry Program (MCCP) and the Chemistry of High Elevation Fog Project (CHEF). The U.S. Environmental Protection Agency's (EPA) MCCP, which collected and analyzed cloud water samples and estimated cloud deposition to the mountains of the eastern United States, operated from 1986 through 1989.[4,8,11–13] Canada's CHEF, which measured cloud chemistry on three mountains in southern Quebec, was in operation from 1985 through 1991.[2,14,15] Data collected by these projects were used by the National Acid Precipitation

Assessment Program (NAPAP) to evaluate the role of airborne chemicals on the changing conditions of forests. The NAPAP Integrated Assessment of 1990 concluded that a limited number of forest ecosystems were at risk from acidic deposition and additional ecosystems would be at risk in the future.[16]

Title IV of the Clean Air Act Amendments of 1990 (CAAA) established the Acid Rain Program, which was designed to reduce the adverse effects of acid deposition. This improvement was to be achieved primarily through reductions of SO_2 and NO_x emissions by electricity producers. The primary goal of the Acid Rain Program is to reduce annual SO_2 emissions from electric utilities to a level that is 10 million tons below the 1980 level. Title IV was created as a two-phased plan to be administered by EPA. Phase I ran from 1995 through 1999, and Phase II, which is more stringent than Phase I, began in 2000. The legislation also required a reduction of 2 million tons of NO_x emissions from utility boilers. Title IX of the CAAA mandated the deployment of a national monitoring network to track the effectiveness of emission reduction programs with respect to deposition, air quality, and changes to affected ecosystems. The Clean Air Status and Trends Network (CASTNet) was implemented by EPA in 1991 in response to this mandate.

The Mountain Acid Deposition Program (MADPro) was initiated in 1993 as part of the research necessary to support the objectives of CASTNet. The two main objectives of MADPro were to develop cloud water measurement systems to be used in a network-monitoring environment and to update the cloud water concentration and deposition data collected in the Appalachian Mountains during MCCP in the 1980s. MADPro measurements were conducted from 1994 through 1999 during the warm season at three permanent mountaintop-sampling stations. These sampling stations were located at Whiteface Mountain, NY, Whitetop Mountain, VA, and Clingman's Dome, TN. A mobile manual sampling station was also operated at two locations in the Catskill Mountains of New York during 1995, 1997, and 1998.

The two main objectives of MADPro have been met. The results of the first objective were presented in Baumgardner et al.[17] and Anderson et al.[18] The results of the second objective are provided herein. This article includes a detailed network description, summary statistics, and analyses of cloud frequency, liquid water content (LWC), cloud chemistry, modeled cloud deposition estimates, and total deposition. A summary of other cloud studies, including MCCP and CHEF, and a comparison to MADPro results are also presented. Additional information about MADPro may be found in the EPA MADPro Summary Report.[19]

EXPERIMENTAL METHODS

Site Descriptions and Measurements

The three permanent MADPro sites were widely dispersed along the Appalachian Mountains. Whiteface Mountain (44 21′58″N, 73 54′10″W) is located in the northeastern Adirondacks in upstate New York. The cloud water sampling research station was above the tree line at 1483 m. The instrumentation was located on the top floor of a four-story observatory at the summit with the cloud water collector, particle volume monitor (PVM), and meteorological sensors mounted on the flat circular roof. Whitetop Mountain (36 38′20″N, 81 36′19″W) is located in the Mount Rogers National Recreation Area of the Jefferson National Forest in southwestern Virginia. It is 6 km southwest of Mount Rogers, the highest peak in Virginia. The MADPro research station was at 1686 m on the main ridgeline of the Appalachian range. Clingman's Dome (35 33′47″N, 83 29′55″W) is the highest mountain in the Great Smoky Mountains National Park. This solar-powered MADPro site was situated at an elevation of 2014 m approximately 50 m southwest of the summit's spiral tourist tower. Electronic instrumentation was housed in a small National Park Service (NPS) building, and the sensors were positioned on top of a 50-ft scaffold tower. Collection at the sites was initiated each spring as soon as local weather conditions would allow and continued through autumn. Collection ran June through September for Whiteface Mountain and May through October for Whitetop Mountain and Clingman's Dome.

The MADPro cloud collection system consisted of an automated cloud water collector for hourly cloud water sampling, a PVM for real-time determination of cloud LWC, and a meteorological station for continuous measurements of wind speed, wind direction, temperature, relative humidity, solar radiation, and precipitation. A data acquisition system was included for collection and storage of electronic information from the various monitors and sensors. The Clingman's Dome and Whitetop Mountain sites also had a filter pack system for dry deposition estimation, while Whitetop Mountain included an additional wet deposition system consisting of a precipitation collector and rain gauge. The filter pack and additional wet deposition systems were not used at Whiteface Mountain because the National Oceanic and Atmospheric Administration (NOAA) and the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) measured these parameters near the cloud water collection site. Likewise, the Clingman's Dome site had access to the data collected by a nearby wet deposition system operated by NPS.

The core of the automated cloud collection system is a passive string collector (also known as the Mohnen collector or the Atmospheric Sciences Research Center

[ASRC] design) previously used in the MCCP study. Collection occurs when ambient winds transport cloud water droplets onto 0.4-mm Teflon wires strung between two circular disks.[1,4] Once impacted, the droplets slide down the strings into a funnel and through Teflon tubing into sample bottles in a refrigerated carousel. The development and design of this system is described in detail in Baumgardner et al.[17]

The PVM-100 by Gerber Scientific measures LWC and effective droplet radius of ambient clouds by directing a narrow laser beam from a 780-nm diode along a 40-cm path.[20] The forward scatter of the cloud droplets in the open air along the path is measured, translated, and expressed as grams of water per cubic meter (g/m^3) of air. This system was programmed so that the collector would be activated and projected out of the protective housing when threshold levels for LWC ($\geq$0.05 g/m^3), wind speed ($>$ 2.5 m/sec), and ambient air temperature ($\geq$2 °C) were reached. In addition, the system was activated only when no precipitation was present. Within the context of MADPro, therefore, a nonprecipitating cloud was defined by a LWC of 0.05 g/m^3 or higher, as measured by the PVM. This threshold was established to have comparability with the MCCP measurements, which were made for the most part with Mallant Optical Cloud Detectors set at a threshold of approximately 0.04 g/m^3.[21] The wind speed threshold of 2.5 m/sec was established because cloud water collection was erratic and inefficient at lower wind speeds. Higher wind speeds were necessary to yield the minimum 30 mL of cloud water required for sample analysis. The temperature limit served to protect against damage from rime ice formation. The absence of rainfall was required because within the objectives of this study, as well as MCCP, only samples from nonprecipitating clouds were collected. If a rain detector was activated, the string collector retracted into the protective case and collection was suspended.

Collection of cloud samples only when these criteria were met did not result in the loss of cloud frequency and cloud duration information because of the continuous data collection by the PVM. All LWC values of 0.05 g/m^3 or greater, independent of the type of cloud (i.e., precipitating or nonprecipitating), were used to calculate cloud frequency and cloud duration information. Cloud deposition estimates presented later may be biased by not sampling for the cloud deposition that occurs during contact with precipitating clouds. Collection of cloud water samples during precipitating cloud events (e.g., with an ETH cloud impactor as deployed by Collett et al.[22]) was not within the scope of this project. However, the bias caused by this lack of sampling during precipitating clouds was minimized because cloud deposition totals were estimated by multiplying the duration-weighted mean chemical fluxes by the cloud-hours for the month. The cloud-hours were calculated as the cloud frequency multiplied by the total hours in the month.

Hourly samples of cloud water were retrieved within 24 hr of each cloud event, and the time, date, and volume of each sample were recorded on a report form. The site operator also measured pH and conductivity and decanted samples into 250-mL bottles for shipment. A rinse/sample blank was also included with each shipment. According to MADPro protocols, the cloud collector was rinsed with deionized water after each cloud event until the conductivity of the rinsate measured $<$10 μS (micro-Siemens). The rinse/sampler blank consisted of a portion of this clean rinsate.

Filter packs were prepared and shipped to the field on a weekly basis and exchanged at the Clingman's Dome and Whitetop Mountain sites every Tuesday. For a description of the filter pack setup, types of filters used, and the fraction collected on each filter, refer to the CASTNet Quality Assurance Project Plan (CASTNet QAPP)[23] or the CASTNet Deposition Summary Report.[24] A discussion on filter pack sampling artifacts can be found in Anlauf et al.[25]

Filter pack flow was maintained at 1.5 L/min with a mass flow controller (MFC) through the middle of the 1998 season, at which time the flow was increased to 3 L/min for the duration of the project. During 1994 and 1995, a continuous flow was drawn through the filter pack. In 1996, the flow was programmed to shut off during a cloud or rain event to allow for determination of truly dry deposition. Because of this, the total hours of flow and, hence, volume were substantially reduced depending on the weekly weather conditions. The increase in flow to 3 L/min was necessary to increase the volume of flow through the filter pack enabling better detection of lower concentrations of analytes.

Wet deposition samples from Whitetop Mountain were also collected on a weekly basis (according to NADP/NTN protocols) in pre-cleaned polyethylene buckets using an Anderson Model precipitation sampler. Buckets were placed on the sampler on Tuesday and removed, whether or not rainfall had occurred, the following Tuesday. Buckets were weighed in the field, decanted to a 1-L polyethylene bottle, sealed, and shipped to Harding ESE for chemical analysis. In addition, precipitation amount (depth) was monitored at Whitetop Mountain with a Belfort rain gauge.

Chemical Analysis and Quality Assurance

Cloud water and wet deposition samples were analyzed for pH, conductivity, Na^+, K^+, NH_4^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_2^-, NO_3^-, and SO_4^{2-} in the Harding ESE laboratory. Wet deposition samples were filtered before analysis and analyzed for acidity. During the first two years of the project, all cloud water samples were analyzed for pH and conductivity at the central laboratory. Starting in 1996, every 10th sample was analyzed in the Harding ESE laboratory for these two parameters to reduce redundancy but still provide quality control (QC) data.

Cloud water and wet deposition samples were stored at 4 °C until analysis. All analyses were performed within 30 days of sample receipt at the laboratory. The effects of storage on wet deposition samples and event versus weekly sampling have been addressed in NAPAP Report #6.[26] That discussion can be applied to cloud water samples as well. A number of studies were summarized in the report comparing event sampling and weekly sampling. Although variations in sampling procedures, locations, time period, and analytical procedures make it difficult to define specific differences, event and weekly sampling are known to produce different values for all the major analytes.

Concentrations of the four anions were determined by micromembrane-suppressed ion chromatography (IC). Analysis of Na^+, Mg^{2+}, and Ca^{2+} was performed with a Perkin-Elmer P-2 inductively coupled argon plasma (ICAP) atomic emission spectrometer, whereas K^+ was analyzed via atomic emission in 1994 and subsequently via ICAP. NH_4^+ concentrations were determined by the automated indophenol method using a Technicon II or Technicon Random Access Automated Chemistry System (TRAACS)-800 autoanalyzer system. Titration to a pH of approximately 8.3 was used for acidity measurements.

Filter pack samples were loaded, shipped, received, extracted, and analyzed by Harding ESE. For specific extraction procedures refer to Anlauf et al.[25] and the CASTNet QAPP.[27] Filter packs contained three filter types in sequence: a Teflon filter for collection of aerosols, a nylon filter for collection of HNO_3, and dual $K_2CO_3^-$-impregnated cellulose filters for collection of SO_2. Following receipt from the field, exposed filters and blanks were extracted and analyzed for anions and NH_4^+, as described above for cloud water and wet deposition samples.

Precision and Accuracy

Accuracy of field measurements was determined by challenging instruments, with the exception of the automated cloud sampler and Gerber PVM, with standards that were traceable to the National Institute for Standards and Technology (NIST). Continuing accuracy was verified by end-of-season calibrations. No certified standards are currently available to determine the accuracy of the cloud sampler and the PVM on a routine basis. Overall, precision of field measurements was determined by collocating instruments and assessing the difference between simultaneous measurements. Although collocated sampling was not conducted at MADPro sites, it did occur at 11 CASTNet sites over the history of the network. Because the meteorological instrumentation at MADPro sites were identical to those used at CASTNet sites, precision of these instruments can be inferred from the CASTNet Deposition Summary Report.[24] The CASTNet 2000 Annual Report may be referenced for precision and accuracy results for 1994 through 1999 and can be found at http://www.epa.gov/castnet/library.html.[27]

Accuracy of laboratory measurements was determined by analyzing an independently prepared reference sample in each batch and calculating the percent recovery relative to the target value. When possible, the references were traceable to NIST or obtained directly from NIST. On occasion, references were ordered from other laboratories. Analytical precision within sample batches was assessed by calculating the relative percent difference (RPD) and percent recovery of continuing verification samples (CVS) run within that batch. CVS are independently produced standards and approximate the midpoint of the analytical range for an analyte. The standards are run after every 10th environmental sample. Precision within a batch was assessed by replicating 5% of the samples within a run. Replicated samples were selected randomly.

Samples were accepted and used for all subsequent analyses if they met an acceptance criteria based on the cation-to-anion ratio. Samples were eliminated if both the anion sum and the cation sum were $\leq$100 microequivalents per liter (μeq/L) and the absolute value of the RPD was $>$100%, or if either the anion sum or the cation sum was $>$100 μeq/L and the absolute value of the RPD was $>$25%. The RPD was calculated from the following: RPD = $200 \times (\text{cations} - \text{anions})/(\text{cations} + \text{anions})$. Applying these acceptance criteria eliminated 694 out of 6186 samples, or 11.8%.

The Energy Center for the Netherlands (ECN) was contracted to verify the accuracy of the PVMs used in MADPro. The cloud chamber at ECN had the capability to generate clouds of known LWC. The facility, described in detail in Gerber et al.,[28] was used to calibrate the Mallant cloud detector used during MCCP. The fogs were characterized in the test section (or chamber) by measuring the LWC with filters and the droplet spectra with a forward scattering spectrometer probe (FSSP-100).

Two filter systems were run side by side as a check of the measurement procedure. The filters consisted of hydrophobic Pall filters placed in housings that face the flow. Air was drawn through the filters isokinetically at a flow rate of 2.15 m/sec. The filters were conditioned in the operating chamber for at least 2 hr before LWC was measured by weighing the filter both before and after a specified time interval, generally 1 hr. The constant high relative humidity (RH) in the chamber and filter-sampling

Table 1. Results from the testing of cloud samples at ECN facilities in the Netherlands.

Run	ECN Pall Filters (mg/m³)	ECN PVM-100 (mg/m³)	RPD[a]	EPA-1 PVM-100 (mg/m³)	RPD	EPA-2 PVM-100 (mg/m³)	RPD	EPA Valente (mg/m³)	RPD
1	883	808	−8.49	880	−0.34	904	2.38	1170	32.50
2	882	832	−5.67	896	1.59	930	5.44	1110	25.85
3	140	163	16.43	184	31.43	176	25.71	75	−46.43
3a	94	110	17.02	125	32.98	117	24.47	112	19.15
4	393	392	−0.25	423	7.63	439	11.70		
4a	412	412	0.00	446	8.25	462	12.14	510	23.79
5	345	361	4.64	388	12.46	402	16.52		
5a	352	369	4.83	395	12.22	410	16.48	472	34.09
6	360	360	0.00	386	7.22	401	11.39	477	32.50
7	687	686	−0.15	731	6.40	784	14.12	932	35.66
8	105	107	1.90	104	−0.95	110	4.76		
8a	97	94	−3.09	94	−3.09	98	1.03		
9	88	85	−3.41	86	−2.27	88	0.00	72	−18.18
9a	689	695	0.87	700	1.60	739	7.26		
10	675	681	0.89	687	1.78	725	7.41	823	21.93
10a	437	427	−2.29	433	−0.92	449	2.75		
10b	441	431	−2.27	436	−1.13	454	2.95	517	17.23
11	438	429	−2.05	435	−0.68	451	2.97	491	12.10
12	687	689	0.29	691	0.58	732	6.55	841	22.42
13	862	857	−0.58	836	−3.02	904	4.87	1034	19.95
13a	128	116	−9.38	118	−7.81	120	−6.25		
14	97	88	−9.28	88	−9.28	97	0.00	114	17.53
14a	388	406	4.64	409	5.41	426	9.79		
14b	383	401	4.70	404	5.48	419	9.40	482	25.85
14c	388	406	4.64	409	5.41	425	9.54	487	25.52
15	384	402	4.69	405	5.47	421	9.64	488	27.08
16a	647	680	5.10	676	4.48	716	10.66	837	29.37
16	114	120	5.26	123	7.89	126	10.53	121	6.14
Mean			1.04		4.60		8.36		18.20

[a]Relative percent difference (RPD) was calculated using the Pall filter concentrations as references; Source: ECN.

procedures minimized LWC measurement errors caused by evaporation or growth of droplets collected on the filter. Such errors may be important when the filter method is used under ambient conditions where RH can be subsaturated or supersaturated.[29]

The estimated accuracy of filter measurements of LWC in the chamber was 10%. The FSSP-100 was located near the filters in the test section. The MADPro quality assurance testing was conducted in February 2000 and included two EPA PVMs and the Valente instrument used in MCCP. The tests involved a series of cloud LWC from 0.1 to 0.9 g/m^3 at cloud droplet sizes of 10 and 25 mass-median-aerodynamic diameter (MmaD). The test concentrations were replicated on different test days and different runs.

A summary of the results is provided in Table 1. The two EPA PVMs measured within +5 to +8.4% of the calibration standard over the entire test range of LWC and droplet sizes. The Valente instrument measured within +18.2% of the Pall filters. Based on the ECN results and results obtained from field-testing, the precision between the EPA PVMs is within 5%. This precision value also agrees with the results found by Pahl and Winkler[30] in a German intercomparison study of PVM-100s.

Cloud Deposition Model

The model used to estimate cloud deposition was originally described by Lovett.[31] The model (CLOUD) uses an electrical resistance network analogy to simulate the deposition of cloud water to forest canopies. Details of the model are summarized in the Appendix. The model is one-dimensional and assumes vertical mixing of droplet-laden air into the canopy from above. Turbulence mixes the droplets into the canopy space where they cross the boundary layers of canopy tissues by impaction and sedimentation. Sedimentation rates are a function only of droplet size. Impaction efficiencies are a function of the Stokes number, which integrates droplet size, obstacle size, and wind speed.[31] The impaction efficiency was based on wind tunnel measurements by Thorne et al.[32]

Resistance or deposition velocity models represent the important atmospheric dynamics controlling deposition and the properties of the receptor surface and depositing substance. Resistance models are the most common type found in the literature for simulating atmospheric deposition. The forest canopy is modeled as stacked 1-m layers containing specified amounts of various canopy tissues such as leaves, twigs, and trunks. Wind speed at any height within the canopy space is determined based on the above-canopy wind speed and an exponential decline of wind speed as a function of downward-cumulated canopy surface area. The wind speed determines the efficiency of the mixing of air and droplets into the canopy and the efficiency with which droplets impact onto canopy surfaces.

The model is deterministic and assumes a steady state so that for one set of above-canopy conditions it calculates one deposition rate. The model requires the following as input data: Leaf Area Index (LAI) of canopy tissues in each height layer of the canopy, zero-plane displacement height and roughness length of the canopy, wind speed at the canopy top, LWC of the cloud above the canopy, and mode of the droplet diameter distribution in the cloud.

From these input parameters, the model calculates the deposition of cloud water, expressed both as a water flux rate (g/cm^2/min) and as a deposition velocity (flux rate/LWC, in units of cm/sec). Deposition rates of ions are calculated by multiplying the water flux rate by the ion concentration in cloud water above the canopy. In the original version of the model, a calculation of the evaporation rate from the canopy was also included to estimate net deposition of cloud water. For this study, only gross deposition rate was estimated and, therefore, the evaporation routine was not invoked.

Several changes in the original model were made to improve its performance and tailor it to the sites under study. The modifications and a model sensitivity analysis are summarized in the Appendix. A more detailed description of the model may be found in the EPA MADPro Summary Report.[19]

The model was run for all valid cloud samples from the three MADPro sites for which wind speed and LWC data were available. The Whiteface Mountain data were processed to create results for two sites: the summit site and a site at 1350 m. Deposition fluxes were calculated only for the latter. Chemical fluxes were calculated as the product of modeled water fluxes and chemical concentrations for all observations for which chemical data were available. Monthly means were calculated for each site and year and weighted by duration of the sample. These data were merged with monthly data on cloud frequency from each site.

Deposition totals were calculated by site, year, and month by multiplying the duration-weighted mean chemical fluxes by the cloud-hours for the month. The cloud-hours were calculated as the cloud frequency multiplied by the total hours in the month. Deposition fluxes for each sampling season were calculated. To compare equitably across sites, the deposition season was specified as June through September. Data outside that period were almost never available for the Whiteface Mountain site and were uneven at the other sites. Even within the June–September period, the monthly deposition for each site could not always be summed to calculate the seasonal totals because some months had no available data due to equipment failures or data screening. Therefore, the seasonal deposition totals were calculated as the duration-weighted mean chemical fluxes for all observations in the four-month period multiplied by the total cloud-hours for the four-month period. Seasonal deposition totals were not calculated unless at least three months of valid cloud frequency data were available for that season.

RESULTS AND DISCUSSION

Cloud Frequency and Liquid Water Content

Mean monthly cloud frequencies by year for the three sites are shown in Figure 1. Monthly cloud frequencies were determined by calculating the relative percent of all hourly LWC values equal to or greater than 0.05 g/m^3. Any month with less than 70% valid LWC data was not considered representative of the monthly weather conditions and was not used in any analyses. Cloud frequencies vary from month to month, year to year, and site to site. The 6-year mean was 42% for Whiteface Mountain, 30% for Whitetop Mountain, and 38% for Clingman's Dome. These values are comparable to mean cloud frequencies reported at these sites during MCCP.[13,21]

Monthly mean LWC values for each year and site are shown in Figure 2. Mean LWC was calculated by taking the mean of all hourly LWC values equal to or greater than 0.05 g/m^3 during the month. Only those monthly values that passed the 70% completeness criterion were plotted. Whiteface Mountain experienced clouds with significantly higher LWCs than the two southern sites, usually 1.5–2 times greater. The 6-year mean LWC was 0.57 g/m^3 for Whiteface Mountain, 0.32 g/m^3 for Whitetop Mountain, and 0.33 g/m^3 for Clingman's Dome. Because LWC increases vertically from cloud base up through the cloud before declining near the top, height above cloud base could be a significant factor in determining the LWC values at the collection locations. Even though the height above cloud base is not known for the three sites, it is estimated that cloud base is lower at Whiteface Mountain with respect to the two southern sites.[21] MCCP reported hourly mean values for LWC during 1987–1988 of 0.43 g/m^3 at Whiteface Mountain and 0.21 g/m^3 at Whitetop Mountain. Mt. Mitchell, NC, a MCCP site with a similar elevation located approximately 100 mi northeast of Clingman's Dome, measured an averaged hourly LWC of 0.29 g/m^3 during 1987–1988.

Cloud Water Chemistry

The mean annual pH of cloud water samples for 1994–1999 for all three sites is shown in Figure 3. The mean pH was obtained by first calculating the mean of the H^+ concentration and then converting this value to pH units

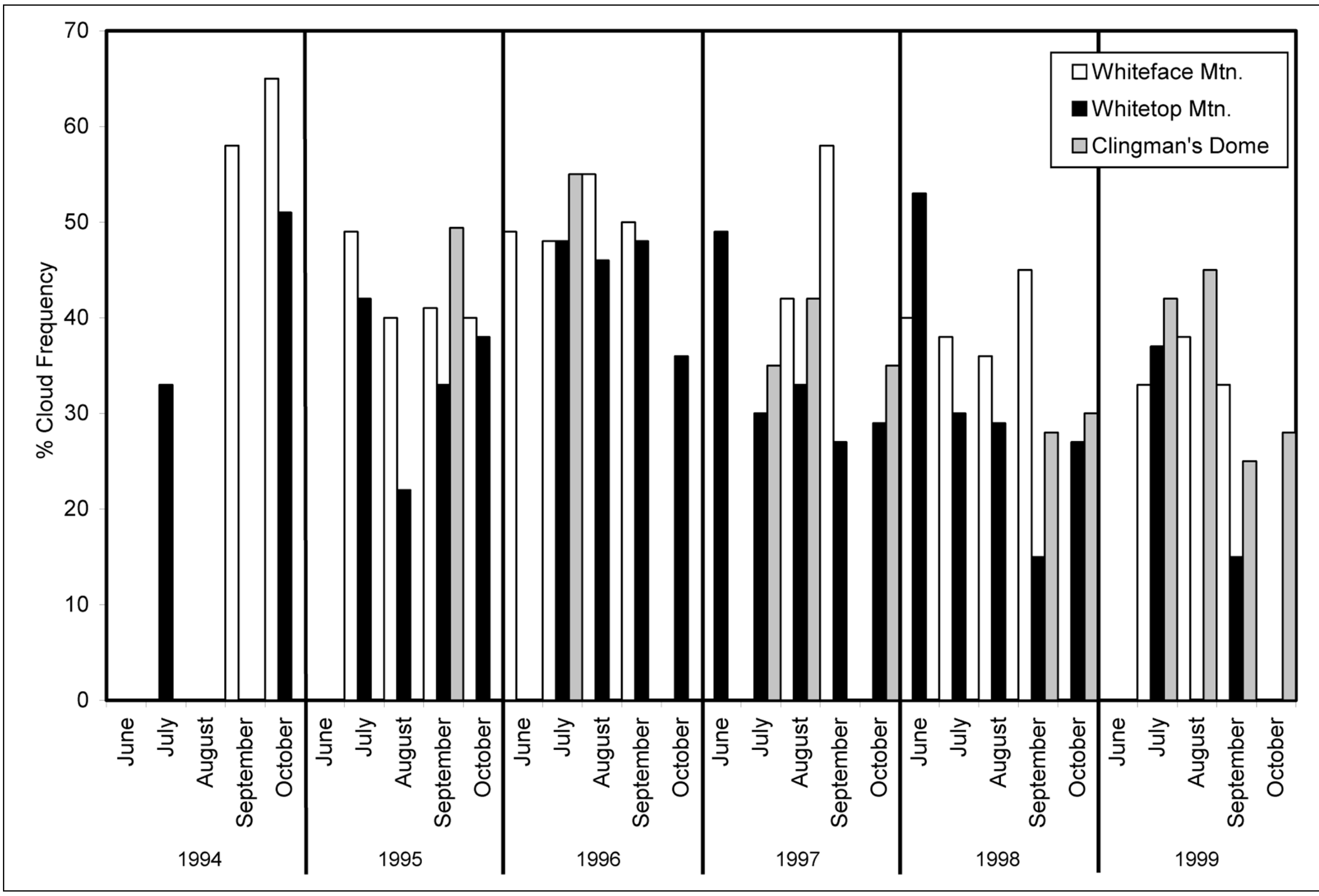

Figure 1. Monthly cloud frequency (1994–1999).

by taking the negative log. The mean annual pH values indicate considerable variability from year to year. All three sites had mean pH values less than 4. The pH values were also analyzed as a frequency distribution over all years showing a greater spread of pH values (3.4–5) for Whiteface Mountain than for either Whitetop Mountain or Clingman's Dome. Whitetop Mountain exhibited a relatively normal distribution with most pH values falling in the 3.6–3.8 range, whereas the majority of pH values for Clingman's Dome were in the 3.4–3.6 range with very few samples above 4.

The seasonal mean cloud water ion concentrations for each of the sites for each year are given in Table 2. Sulfate was the anion with the highest concentration at each site with Clingman's Dome having the highest concentration measured during the study (3687 μeq/L) and the highest mean values (361–443 μeq/L). The anion with the next highest concentrations was NO_3^-. Whiteface Mountain had lowest mean NO_3^- values (87–136 μeq/L) among the three sites, but exhibited the highest maximum value for the study (2251 μeq/L). Chlorine, with mean values of less than 50 μeq/L at all sites for all years, was the only other anion with significant concentration levels.

Hydrogen was the cation with the highest concentrations found in cloud water samples at all sites for all years. Clingman's Dome also had the highest mean values for this analyte (264–447 μeq/L). The cation with the second-highest concentrations was NH_4^+. The mean for NH_4^+ for all sites and all years ranged from 124 to 210 μeq/L at Whiteface Mountain, 106 to 210 μeq/L at Whitetop Mountain, and 106 to 279 μeq/L at Clingman's Dome.

An analysis of MCCP cloud water data by Mohnen and Vong[13] using air mass back trajectories to determine cloud origin showed that clouds impacting individual sites had different origins and that ion concentrations were dependent on air mass history. Whiteface Mountain experienced the highest ion concentrations in clouds that originated from the southwest. Air mass origin did not produce significant differences in seasonal mean cloud concentrations at Mt. Mitchell and Whitetop Mountain. An analysis of cloud data using back trajectories indicated the same patterns of cloud arrival during the MADPro study.[33] Air mass origin is a possible explanation for the observed difference in cloud water ion concentrations, especially for SO_4^{2-} and H^+.

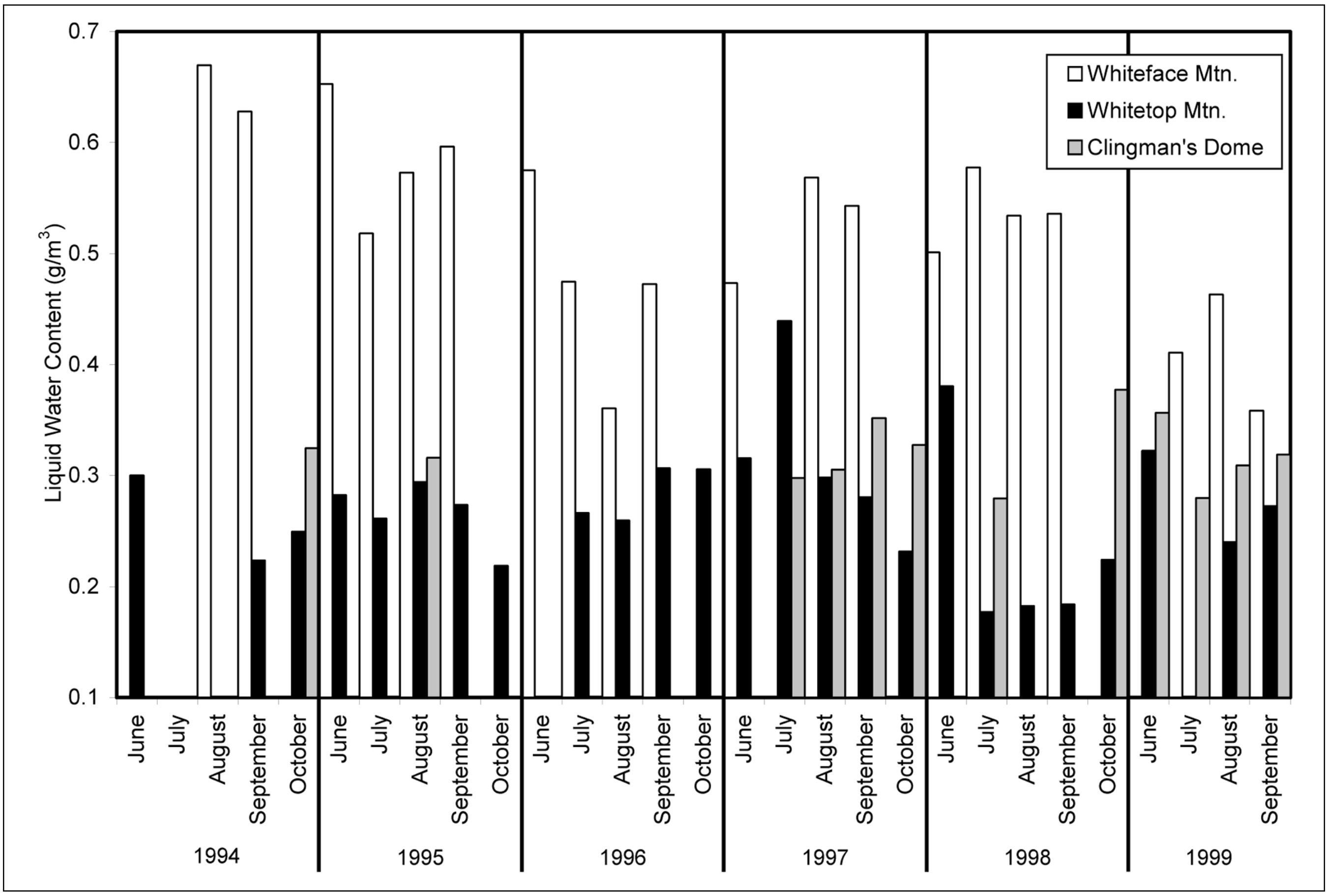

Figure 2. Mean LWC of clouds (1994–1999).

Clingman's Dome exhibited statistically significant upward trends in seasonal concentrations of SO_4^{2-}, NO_3^-, and H^+, and to a lesser extent NH_4^+.[19] The trends at this site may, in part, be caused by an increase in regional summertime SO_4^{2-} concentrations as documented by Mueller.[34] Butler et al.[35] suggest that the lack of improvement and even an increase in concentrations in the vicinity of Clingman's Dome may be attributed to several large coal-fired electric-generating facilities that were not required to reduce SO_2 or NO_x emissions under CAAA. More research and analysis involving back trajectories needs to be conducted to fully understand this pattern at Clingman's Dome.

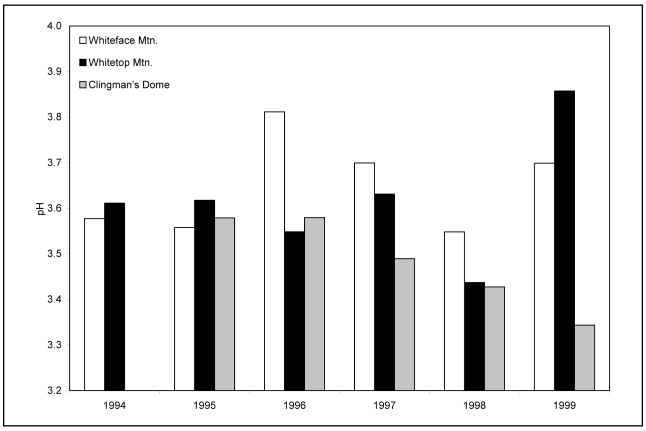

Figure 3. Mean pH of cloud water samples at MADPro sites (1994–1999).

A grand mean for all study years was calculated for each MADPro site for each ion and for pH. This allows for comparisons with other studies, which are shown in Table 3. Cloud water chemistry analyses at Whiteface Mountain, Whitetop Mountain, Mt. Mitchell, and Mt. Tremblant and Roundtop Ridge in southern Quebec (located 120 mi northwest of Whiteface Mountain) are shown in the table. The data show that ion concentrations measured during MADPro were similar to other measurements of cloud water in the Appalachians.

Only a few cloud chemistry projects have been conducted in the eastern United States toward understanding cloud chemical processes and cloud deposition during the winter season, when low altitude supercooled clouds prevail. However, a 3-year wintertime field study at Whiteface Mountain Summit[36] showed that ambient levels of SO_4^{2-} aerosol (the main precursor to cloud SO_4^{2-} in supercooled winter clouds) are significantly lower during

Table 2. Summary statistics for cloud water samples collected at MADPro sites (1994–1999).

	Whiteface Mountain					Whitetop Mountain					Clingman's Dome				
Year	**n**	**Mean**	**SD**	**Min**	**Max**	**n**	**Mean**	**SD**	**Min**	**Max**	**n**	**Mean**	**SD**	**Min**	**Max**
1994	**Total Records Accepted = 235**					**Total Records Accepted = 141**					**Total Records Accepted = 9**				
LWC	213	0.64	0.34	0.01	1.55	89	0.31	0.16	0.01	0.71	8	0.30	0.20	0.05	0.56
pH	223	3.97	0.69	2.73	5.71	141	3.80	0.45	2.81	5.36	9	4.36	0.71	3.82	6.14
Cond	221	166.35	182.97	4.00	1072.30	141	149.04	129.15	12.80	779.00	9	62.53	37.52	17.10	144.00
H^+	223	264.61	317.98	1.95	1862.09	141	244.72	240.07	4.37	1548.82	9	73.93	48.18	0.72	151.36
NH_4^+	235	210.26	256.10	0.56	1421.67	141	184.63	213.46	5.07	1349.99	9	126.36	125.30	5.93	422.14
SO_4^{2-}	235	379.37	471.04	2.29	2624.45	141	316.72	296.22	24.38	1606.25	9	169.28	153.67	55.21	556.25
NO_3^-	235	133.22	164.74	1.94	1078.95	141	143.66	130.35	11.07	828.58	9	61.21	35.13	20.71	110.00
NO_2^-	235	0.63	0.70	0.22	4.78	141	0.92	1.23	0.29	7.29	9	0.48	0.23	0.29	0.86
Ca^{2+}	235	24.82	41.21	0.00	227.54	141	20.16	22.10	0.15	124.85	9	29.21	30.89	6.64	100.45
Mg^{2+}	235	6.63	9.62	0.82	58.44	141	8.26	9.92	0.74	80.66	9	16.67	17.58	2.80	51.03
Na^+	235	3.29	4.89	0.87	51.76	141	16.46	36.76	0.78	323.01	9	50.54	65.26	1.35	184.99
K^+	235	2.37	3.27	0.26	39.13	141	6.15	25.19	0.43	296.68	9	3.79	4.43	0.97	15.35
Cl^-	235	10.29	13.07	0.56	94.50	141	27.39	38.37	1.55	301.83	9	43.28	55.41	2.48	172.07
1995	**Total Records Accepted = 525**					**Total Records Accepted = 550**					**Total Records Accepted = 136**				
LWC	502	0.61	0.27	0.05	1.02	519	0.30	0.13	0.00	0.66	123	0.30	0.15	0.00	0.61
pH	523	3.92	0.63	2.57	6.57	550	3.84	0.48	2.59	5.23	136	3.87	0.63	2.67	6.27
Cond	524	137.90	159.24	1.91	1142.00	550	151.22	158.65	3.28	1590.00	136	156.46	159.37	4.50	1060.00
H^+	523	276.73	361.90	0.27	2691.53	550	241.28	259.82	5.89	2570.40	136	263.57	285.87	0.54	2137.96
NH_4^+	517	191.63	223.59	0.71	1314.29	550	200.06	221.74	0.71	1521.43	135	182.08	212.35	3.50	1571.43
SO_4^{2-}	519	334.29	429.31	1.81	3416.67	550	325.67	337.52	3.17	2395.83	135	361.34	440.63	4.73	3270.83
NO_3^-	519	136.08	162.00	0.71	1078.57	550	166.76	194.65	2.14	1992.86	135	132.33	131.15	2.00	850.00
NO_2^-	519	1.11	1.40	0.21	9.93	550	1.27	1.44	0.29	9.86	135	1.27	2.05	0.29	15.07
Ca^{2+}	516	24.39	41.29	0.25	292.70	548	24.64	33.36	0.18	336.41	133	27.68	53.19	0.36	349.09
Mg^{2+}	516	6.76	10.98	0.25	81.36	548	10.32	13.63	0.25	146.09	133	11.63	11.63	0.40	64.43
Na^+	516	5.10	13.58	0.13	235.85	548	20.81	39.29	0.23	278.54	133	28.73	33.58	0.69	264.15
K^+	516	2.34	3.17	0.08	18.28	548	7.54	22.16	0.53	375.78	133	2.83	2.77	0.21	17.87
Cl^-	519	11.59	16.27	0.08	134.84	550	33.46	46.95	0.79	420.31	135	38.17	35.62	0.56	252.19
1996	**Total Records Accepted = 597**					**Total Records Accepted = 194**					**Total Records Accepted = 105**				
LWC	549	0.54	0.26	0.01	1.02	191	0.36	0.14	0.06	0.74	29	0.21	0.10	0.06	0.42
pH	569	4.09	0.56	2.81	5.89	181	3.79	0.54	2.82	5.49	103	3.73	0.41	3.06	4.85
Cond	568	104.59	115.26	1.80	927.00	170	169.92	190.64	4.00	935.00	105	221.50	133.12	19.00	621.00
H^+	569	154.25	184.44	1.29	1548.82	181	282.79	292.70	3.24	1513.56	103	263.38	197.15	14.13	870.96
NH_4^+	557	124.34	159.10	0.71	1178.57	194	174.29	182.70	2.79	871.43	105	219.81	166.11	1.93	750.00
SO_4^{2-}	557	202.47	273.43	1.35	2479.17	194	342.84	388.19	6.27	2354.17	105	394.30	286.71	13.40	1189.58
NO_3^-	557	87.68	106.71	1.29	764.29	194	147.29	144.19	3.71	757.14	105	152.78	95.86	8.71	417.86
NO_2^-	557	0.93	1.16	0.29	12.21	194	1.46	1.83	0.29	10.86	105	1.82	1.67	0.29	9.21
Ca^{2+}	556	14.88	30.02	0.15	349.68	194	19.82	27.05	0.61	174.55	105	40.42	52.56	0.55	257.45
Mg^{2+}	556	3.76	6.00	0.25	47.55	194	7.35	9.20	0.47	52.05	105	12.97	15.30	0.25	85.10
Na^+	556	3.07	6.95	0.22	104.34	194	11.46	21.42	0.22	162.33	105	16.71	30.57	0.22	124.55
K^+	556	1.60	2.58	0.13	40.52	194	3.56	4.31	0.28	46.93	105	3.13	2.87	0.13	21.17
Cl^-	557	7.22	10.49	0.56	77.01	194	23.58	28.52	0.73	201.41	105	27.36	26.53	0.56	120.45
1997	**Total Records Accepted = 448**					**Total Records Accepted = 501**					**Total Records Accepted = 324**				
LWC	335	0.57	0.26	0.09	1.02	464	0.39	0.17	0.06	1.01	283	0.38	0.19	0.01	0.98
pH	393	4.08	0.69	2.87	6.10	501	3.92	0.55	2.80	6.06	318	3.76	0.61	2.80	5.88
Cond	378	138.54	158.84	1.90	978.00	464	153.29	150.10	4.10	880.00	180	187.75	173.04	2.60	1080.00
H^+	393	199.73	236.27	0.79	1348.96	501	233.73	260.02	0.87	1584.89	318	323.89	296.52	1.32	1584.89
NH_4^+	382	184.38	261.86	0.71	1721.43	501	210.09	256.23	0.71	1721.43	318	245.71	246.39	1.07	1650.00
SO_4^{2-}	381	304.28	398.22	1.58	2916.67	501	331.41	360.97	1.83	2375.00	318	432.77	409.84	3.67	2791.67
NO_3^-	381	147.54	216.86	0.64	1900.00	501	157.58	182.21	2.86	1135.71	318	183.38	169.15	2.57	1007.14
NO_2^-	381	0.95	1.17	0.29	8.00	501	1.33	1.59	0.29	11.00	318	1.40	1.50	0.29	13.86
Ca^{2+}	376	36.44	75.38	0.15	739.77	501	20.77	31.56	0.21	303.49	318	41.65	59.72	0.15	512.48

Table 2. (cont.)

	Whiteface Mountain					Whitetop Mountain					Clingman's Dome				
Year	**n**	**Mean**	**SD**	**Min**	**Max**	**n**	**Mean**	**SD**	**Min**	**Max**	**n**	**Mean**	**SD**	**Min**	**Max**
Mg^{2+}	376	10.33	20.43	0.25	222.77	501	7.82	11.41	0.25	114.40	318	10.68	12.66	0.25	77.37
Na^{+}	376	8.04	22.18	0.28	231.63	501	14.50	35.69	0.22	419.56	318	16.32	27.61	0.22	244.89
K^{+}	376	2.52	3.48	0.13	35.88	501	2.95	5.31	0.13	69.92	318	2.89	3.12	0.13	20.74
Cl^{-}	381	11.28	16.40	0.56	110.01	501	26.29	47.46	0.56	547.25	318	24.41	27.32	0.56	204.51
1998	**Total Records Accepted = 387**					**Total Records Accepted = 276**					**Total Records Accepted = 269**				
LWC	362	0.59	0.26	0.07	1.02	265	0.28	0.17	0.01	0.95	223	0.36	0.17	0.03	0.78
pH	387	4.01	0.71	2.56	5.80	271	3.65	0.48	2.71	5.28	268	3.57	0.39	2.82	5.82
Cond	387	155.30	213.44	3.82	1353.00	271	198.79	164.60	19.00	908.00	268	201.89	139.92	7.10	793.00
H^{+}	387	282.98	406.05	1.58	2754.23	271	365.46	332.57	5.25	1949.84	268	373.82	302.60	1.51	1513.56
NH_4^{+}	387	198.50	280.13	1.86	1528.57	276	233.81	237.13	1.07	1457.14	267	279.24	202.93	3.93	1300.00
SO_4^{2-}	386	337.97	526.62	2.17	3499.27	276	467.18	418.92	7.83	2124.56	269	443.58	320.10	15.48	2030.83
NO_3^{-}	386	133.13	188.88	1.22	1329.48	276	189.74	170.47	4.79	1093.61	269	208.50	138.38	10.08	829.14
NO_2^{-}	386	0.68	0.57	0.29	6.07	276	1.30	1.35	0.29	8.00	269	0.83	0.88	0.29	7.43
Ca^{2+}	387	23.35	45.89	0.15	380.85	276	39.10	51.64	0.62	465.09	269	46.95	42.38	1.30	324.50
Mg^{2+}	387	6.89	12.20	0.25	122.52	276	13.62	19.42	0.46	202.28	269	13.27	10.47	0.43	75.42
Na^{+}	387	4.79	11.93	0.22	108.87	276	18.73	32.73	0.22	210.17	269	23.59	24.55	0.44	104.59
K^{+}	387	2.17	2.81	0.13	21.48	276	3.95	4.92	0.13	34.03	269	3.80	3.08	0.13	18.32
Cl^{-}	386	9.46	15.02	0.56	128.63	276	31.63	40.48	0.56	423.15	269	30.40	26.37	1.64	135.40
1999	**Total Records Accepted = 473**					**Total Records Accepted = 143**					**Total Records Accepted = 174**				
LWC	395	0.50	0.22	0.05	1.00	141	0.38	0.12	0.09	0.67	171	0.41	0.26	0.02	1.02
pH	473	4.11	0.65	2.57	6.15	143	4.18	0.55	2.74	5.36	173	3.68	0.64	2.68	5.48
Cond	473	107.15	150.36	3.20	1230.00	143	89.79	121.36	0.13	978.00	173	211.12	213.90	3.70	1081.00
H^{+}	473	199.95	310.03	0.71	2691.53	143	138.85	218.52	4.37	1819.70	173	447.69	472.84	3.31	2089.30
NH_4^{+}	473	127.53	180.36	1.43	1600.00	143	106.63	119.09	1.43	632.86	174	203.36	219.44	1.43	1528.57
SO_4^{2-}	473	233.75	342.00	2.10	3020.35	143	201.17	244.58	4.42	1533.09	174	487.01	545.14	3.54	3686.91
NO_3^{-}	473	125.51	250.21	0.57	2251.54	143	96.59	144.52	2.72	1279.45	174	198.20	206.49	1.64	964.95
NO_2^{-}	473	1.00	0.77	0.71	7.64	143	1.03	0.59	0.71	4.14	174	0.90	0.56	0.71	5.00
Ca^{2+}	473	32.56	99.09	0.15	1277.44	143	16.02	22.99	0.20	103.79	174	72.89	119.92	0.15	793.41
Mg^{2+}	473	10.93	29.97	0.25	353.89	143	5.41	6.91	0.33	29.87	174	16.64	26.79	0.25	216.45
Na^{+}	473	10.41	28.78	0.22	276.23	143	4.29	9.78	0.22	56.55	174	9.17	15.33	0.22	127.02
K^{+}	473	2.32	3.94	0.13	35.54	143	1.92	2.12	0.13	10.82	174	3.57	5.15	0.13	42.19
Cl^{-}	473	10.48	22.97	0.56	184.78	143	9.88	15.86	0.56	131.18	174	14.34	17.81	0.56	128.36

Note: All units are μeq/L except for LWC (g/m^3), pH (unitless), and conductivity (Φmhos/cm); Max = maximum; Mean = arithmetic average; Min = minimum; n = sample size used in calculations; SD = standard deviation.

the winter than during the summer and sometimes below the detection limit of about 0.003 μmole/m^3. Consequently, the SO_4^{2-} concentrations in winter clouds and precipitation are also lower. Gas-phase HNO_3 (the main precursor to cloud NO_3^{-} in supercooled winter clouds) also exhibits an annual pattern with lower values in winter, but the decline in concentration during winter is less than for SO_4^{2-} aerosol. Therefore, most of the samples during the warm season have SO_4^{2-} concentrations that are greater than the NO_3^{-} concentrations, but the winter samples have NO_3^{-} concentrations that are greater than the SO_4^{2-} concentrations.

The relative concentrations of H^{+} and NH_4^{+} are similar in summer and winter, but both have higher concentrations in summer. More than 75% of all winter cloud samples showed concentrations less than 300 μM/L for SO_4^{2-}, NO_3^{-}, and NH_4^{+}.[37,38] One essential parameter required for calculating cloud deposition fluxes is the LWC of clouds, which was not systematically measured in any of the field studies. A few measurements from Whiteface Mountain indicated lower LWC in winter clouds by at least a factor of 2. On the basis of previous results for cloud water concentrations sampled during freezing conditions shown in Kadlecek et al.[36] and assuming a 50% lower LWC, it can be estimated that cloud deposition fluxes during the winter season are substantially lower for all major ions than during the warm season.

Nationally, SO_2 emissions have decreased 20% between 1990 and 2000 with the largest decrease in 1994–1995 because of the implementation of Phase I of the

Table 3. Comparison of mean cloud water concentrations and pH (MADPro, MCCP, CHEF, and earlier studies).

Cloud Water Monitoring	Year	Site	H^+ (μeq/L)	SO_4^{2-} (μeq/L)	NO_3^- (μeq/L)	NH_4^+ (μeq/L)	pH (μeq/L)
Falconer and Falconer (1980)[1]	1979	Whiteface Mtn.					3.45
Weathers et al. (1988)[3]	1984–1985	Whiteface Mtn.	23–676 (Range)	27–673 (Range)	53–230 (Range)		3.52
MCCP[a] Mohnen and Vong (1993)[13]	1986–1989	Whiteface Mtn.	171	205	73	97	3.70
		Whitetop Mtn.	174	321	144	152	3.49
		Mt. Mitchell	398	489	174	184	3.32
CHEF[b] Schemenauer et al. (1995)[15]	1986–1991	Mt. Tremblant	181	339	170	239	3.74
		Roundtop Ridge	116	221	90	158	3.93
MADPro	1994–1999	Whiteface Mtn.	230	298	127	172	4.01
		Whitetop Mtn.	257	331	150	184	3.86
		Clingman's Dome	270	424	175	290	3.71

[a]MCCP = Mountain Cloud Chemistry Program; [b]CHEF = Chemistry of High Elevation Fog.

greater than 25 kg/ha. Compared with Whitetop Mountain, Whiteface Mountain typically had higher seasonal deposition of SO_4^{2-} and NO_3^-.

Previous estimates of cloud water deposition at MADPro sites are discussed in papers by Mohnen et al.,[21] Miller et al.,[43] Miller et al.,[44] and Lindberg.[45] Table 5 provides MADPro/CLOUD and previous model calculations for the three MADPro sites and for a site at Mt. Moosilauke, NH, which is included because of similarities in location and forest canopy structure. Results for the MCCP site at Mt. Mitchell are also presented. The table provides information on each site, the name of the simulation model, name of principal investigator or study, study years, cloud ion deposition rates (kg/ha/month), water deposition rate, and cloud frequency. All of the models listed in the table were based on the original Lovett model[31,46] with variations and additions. MCCP used the cloud deposition model (CDM) variation as described by Mueller,[47] as did Saxena and Lin.[7] Miller's model, entitled M-CLOUD, incorporated some of the CDM changes as well as additional modifications.

Mohnen's estimates for Whiteface Mountain during MCCP reflect a canopy structure near the summit (1483 m) of krummholtz and mixed alpine tundra, as compared with the Balsam fir canopy assumed in CLOUD. Miller's estimates of cloud water deposition on Whiteface Mountain at an elevation of 1350 m in a predominately balsam fir stand are more comparable to MADPro. The MADPro water deposition rates are significantly higher than those of Miller et al.,[43] but Miller's group simulated the entire year, including the winter season, which they suggested had water deposition rates of less than half those of the warm season. Miller et al.[43] reported a mean warm season deposition velocity (45.3 cm/sec), which is higher than the MADPro result of 37.2 cm/sec. Miller et al.[43] also reported lower chemical concentrations in cloud water (132 μeq/L of SO_4^{2-} vs. 347 μeq/L of SO_4^{2-}).

For the site at Mt. Moosilauke, Lovett et al.[46] estimated higher cloud water deposition than estimated by Mohnen et al.[21] However, model calculations were made for different elevations (1000 m and 1250 m) in the two studies.

Mohnen et al.[21] estimated cloud water deposition at two sites on Whitetop Mountain: an open stand with low

CAAA emission reduction program. Emissions of SO_2 have been approximately level between 1996 and 1999.[39] Large decreases of SO_4^{2-} in precipitation and ambient SO_2 and aerosol SO_4^{2-} have occurred during the 10-year period starting in 1990.[40–42] Corresponding decreases of SO_4^{2-} in cloud water have not been observed.[9,19] Nitrogen oxide (NO_2 and NO) emissions increased steadily from 1985 to 1995 but have since decreased slightly.[39] Concentrations of NO_3^- in precipitation and ambient concentrations of NO_3^- (HNO_3 + NO_3^- particles) have not decreased over the last 10 years in most areas of the United States.[27] NO_3^- concentrations in cloud water have not shown a decreasing or an increasing trend. The MADPro results are consistent with these observations.

Estimates of Cloud Water Deposition

Estimates of seasonal cloud water deposition for the three MADPro sites are provided in Table 4. All sites produced seasonal cloud water deposition rates for SO_4^{2-} greater than 50 kg/ha and NO_3^- deposition rates

Table 4. Seasonal deposition estimates produced with the CLOUD model (kg/ha).

Site	Year	H^+	SO_4^{2-}	NO_3^-	NH_4^+
Whiteface Mtn.	1995	2.01	98.49	59.04	24.04
	1996	1.06	74.55	40.00	18.58
	1997	1.42	92.67	55.10	19.75
	1998	1.44	77.40	43.01	18.52
Whitetop Mtn.	1995	0.68	43.05	27.33	9.09
	1996	1.81	105.57	57.09	18.62
	1997	1.15	78.51	29.72	16.78
	1998	1.05	60.87	31.98	11.30
Clingman's Dome	1997	0.85	52.55	26.35	10.20

Note: Three of the four months were required to calculate seasonal deposition. Three-month deposition rates were multiplied by 4/3 to obtain seasonal depositions. A season is defined as June through September.

Table 5. Cloud water chemical deposition via droplet interception for the eastern United States.

Author (Reference)	Site	Year	Elevation (m)	Cloud Deposition Model	Cloud Droplet Deposition (kg/ha/mo) H^+	SO_4^{2-}	NO_2^-	NH_4^+	H_2O (cm/yr)	Frequency of Cloud (%)
Mohnen et al. (1990)[21]	Whiteface Mtn., NY	1986–88	1483	CDM	0.03	2.20	0.80	0.39		24.0
Miller et al. (1993a)[43]	Whiteface Mtn., NY	1985–90	1350	M-CLOUD	0.15	7.80	4.80	1.60	153.5	35.9
MADPro	Whiteface Mtn., NY	1994–99	1350	CLOUD	0.35	20.70	11.90	3.90	229.0	42.0
Lovett et al. (1982, 1983)[32,46]	Mt. Moosilauke, NH	1980	1250	Lovett	0.20	11.50	8.50	1.40	68.0	40.0
Mohnen et al. (1990)[21]	Mt. Moosilauke, NH	1986–88	1000	CDM	0.06	2.70	2.00	0.44	20.0	19.0
Mohnen et al. (1990)[21]	Whitetop Mtn., VA	1986–88	1686	CDM	0.22	12.10	6.10	2.00	90.0	30.0
MADPro	Whitetop Mtn., VA	1994–99	1686	CLOUD	0.22	13.20	7.50	2.80	100.0	29.0
Mohnen et al. (1990)[21]	Mt. Mitchell, NC	1986–88	2000	CDM	0.19	8.40	4.00	1.10	59.0	29.0
Lindberg et al. (1988)[45]	Clingman's Dome, TN	1988	1740	Lovett	0.14	7.20	2.20	1.00		25.0
MADPro	Clingman's Dome, TN	1994–99	2014	CLOUD	0.23	14.30	7.70	3.00	96.0	38.0

tree density and a closed stand with high density. Stand characteristics at the closed strand are similar to those used in model calculations for MADPro. Cloud water deposition estimates are not available from MCCP (Mohnen et al.[21]) for Clingman's Dome and, therefore, the MCCP site at Mt. Mitchell is used for the comparison. Mohnen et al.[21] estimated a 3-year mean cloud water deposition of 8.4 kg/ha/month for SO_4^{2-} at Mt. Mitchell. Saxena and Lin[7] estimated a mean cloud deposition at Mt. Mitchell of 6 and 9.8 kg/ha/month for SO_4^{2-} for 1986 and 1987, respectively. Lindberg[45] estimated a cloud deposition of 7.2 kg/ha/month for SO_4^{2-} for 1986 and a 3-year mean (1986–1988) of 4 kg/ha/month at a site in the Great Smoky Mountain National Park close to Clingman's Dome but at a slightly lower elevation.

Total Deposition

Total deposition is typically defined as the sum of dry and wet (or precipitation) deposition. In the eastern United States, frequent cloud exposure occurs at elevations above 800 m.[21] Total deposition in such areas of frequent cloud exposure is the sum of cloud, wet, and dry deposition. In this analysis, total sulfur is the sum of dry (SO_2 + SO_4^{2-}), wet (SO_4^{2-}), and cloud (SO_4^{2-}) deposition, as sulfur. Total nitrogen is the sum of dry (HNO_3 + NO_3^-), wet (NO_3^-), and cloud (NO_3^-) deposition, as nitrogen. NH_4^+, provided as nitrogen, and H^+ are presented separately. The analysis of total deposition provides the contribution of each fraction (i.e., cloud, wet, and dry) to the total pollution loading at a location. Total deposition from high-elevation sites is compared with locations where total pollution loading results from dry and wet deposition only.

Cloud, wet, and dry deposition estimates were calculated on a monthly basis from 1994 through 1998 for the months of June through September at Whiteface and Whitetop Mountains. Cloud deposition values at the sites for 1994–1998 were estimated by the CLOUD model. The multilayer model (MLM)[48,49] was utilized to estimate dry depositions from filter pack concentration data for Whitetop Mountain. The MLM is used to simulate dry deposition at CASTNet sites.[27] The dry deposition values for Whiteface Mountain were obtained from NOAA's Atmospheric Integrated Monitoring Network (AIRMoN) site at Whiteface Mountain and were estimated using a different version of the MLM. Total deposition was not estimated at Clingman's Dome because of low data completeness.

The MLM calculations are considered reasonable for Whitetop Mountain and Whiteface Mountain because onsite meteorological measurements were used directly in the model. Data from Meyers et al.[48] show little overall bias with an uncertainty of ±50% for estimates of the dry deposition of SO_2 and HNO_3 calculated using the MLM. Data from Finkelstein et al.[49] suggest the MLM underestimates deposition velocities for SO_2 for complex, forested sites. The differences are expected to be lower for longer averaging times (i.e., monthly and seasonal periods). Dry and wet deposition values for Whitetop Mountain were calculated from data collected by instrumentation collocated with the cloud sampler site.

Wet deposition data for Whiteface Mountain were obtained from NADP/NTN. The location of the NADP/NTN wet/dry bucket and the AIRMoN filter pack at Whiteface Mountain is approximately 750 m lower in elevation from the location of the cloud deposition estimates. Higher wind speeds and greater LAI values at the higher elevation site tend to increase dry deposition of HNO_3.[43] The orographic increase in precipitation above 750 m is significant (approximately 20%). It is estimated, therefore, that wet and dry deposition values may be underestimated significantly at the NADP/NTN/AIRMoN site relative to expected deposition values at the 1350-m cloud-modeling site. For comparison, CASTNet estimates of dry deposition fluxes for two high-elevation sites, a valley site and a ridge site, near Coweeta, NC, show differences up to a factor of 4.4 for weekly means.[24] Again, differences in seasonal means are expected to be lower.

Monthly total deposition values for June through September were calculated by summing the monthly cloud, wet, and dry deposition values when available or when data completeness permitted for 1994–1999. Monthly total deposition amounts were calculated only if all three fractions for a month were available. For Whiteface Mountain and Whitetop Mountain, the percent that each fraction contributed to the total deposition was calculated for those months when data for all three fractions were available. An overall mean percent value was then calculated from the monthly percentages of each fraction. For Whiteface and Whitetop Mountains, overall mean percentages were calculated for S, N, and H^+. NH_4^+ dry deposition estimates were not available from the AIRMoN site at Whiteface Mountain, but overall mean percentages for NH_4^+ were calculated for Whitetop Mountain. The percent composition results are presented in Table 6.

Table 6 shows that clouds are, by far, the largest source of deposition of pollutants to high-elevation ecosystems. Between 80 and 90% of S deposition at the sites occurs via cloud exposure as does 70–87% of the total H^+ loading. Cloud deposition is also responsible for 90–95% of NH_4^+ deposition at Whitetop Mountain. Dry deposition is a minor contributor to the total S and NH_4^+ loading but contributes between 22 and 28% of N deposition and approximately 15–16% of H^+ deposition at the southern site.

Seasonal dry, wet, and total deposition estimates for the major ions at Whiteface Mountain and two nearby CASTNet sites are presented in Table 7a. The same information is presented in Table 7b for Whitetop Mountain and two nearby CASTNet sites. Total deposition values from the MADPro sites were approximately 6–20 times greater for S, N, and NH_4^+ (NH_4^+ results refer to Whitetop Mountain only) when compared with the CASTNet sites. Whiteface Mountain H^+ deposition was 1.3–2.4 times greater than the CASTNet H^+ deposition. Whitetop Mountain H^+ deposition values for 1996 were 6.5–10 times greater than H^+ deposition amounts at the CASTNet sites. Dry deposition values at Whiteface Mountain were comparable to dry deposition values for the two CASTNet sites. Even though total seasonal deposition estimates could be calculated only for 1996 for Whitetop Mountain, dry and wet deposition estimates were similar to the closest CASTNet sites. Total deposition rates at the closest CASTNet sites were much lower than rates for Whitetop Mountain.

Table 6. Percent composition of total deposition at two MADPro sites.

Site	Deposition Type	H^+	S	N	NH_4^+
Whiteface Mtn.	Cloud Deposition	87.61	90.95	83.67	NA
	Wet Deposition	7.90	7.58	8.17	NA
	Dry Deposition	4.49	1.47	8.15	NA
Whitetop Mtn.	Cloud Deposition	80.87	89.27	68.77	95.37
	Wet Deposition	3.94	3.96	2.96	1.80
	Dry Deposition	15.19	6.78	28.27	2.84

Note: Sulfur deposition includes SO_2 and/or SO_4^{2-}; Nitrogen deposition includes HNO_3 and/or NO_3^-; NH_4^+ deposition is presented in terms of nitrogen (N); NA = not available.

MCCP researchers[21] have estimated that cloud water deposition provides a substantial fraction of the total chemical deposition to the forests that they studied in the eastern United States. MCCP results indicate that cloud SO_4^{2-}, NO_3^-, H^+, and NH_4^+ deposition exceeded wet deposition in precipitation for three sites located above 1400 m. Two sites located near 1000-m elevation received cloud water chemical inputs that added at least 50% above precipitation chemical deposition.[8] Lindberg and Johnson[5] estimated that cloud water contributes approximately 25–50% of total (cloud + rain + dry) SO_4^{2-}, N, and H^+ deposition at sites on Whiteface Mountain and in Great Smoky Mountains National Park.

Based on a decade of observations from 1986 through 1996 (using data from MCCP, MADPro, and EPRI's Integrated Forest Study [IFS]), Miller and Friedland[9] estimated the atmospheric deposition at the Whiteface Mountain 1050-m site. Total nitrogen averaged 17.2 kg/ha/yr (37% deposited as NH_4^+ and 63% as NO_3^-) and sulfur deposition averaged 18.3 kg/ha/yr. Precipitation and cloud water deposition contributed nearly equally to total SO_4^{2-} deposition.

In a separate study, Miller et al.[43] determined that cloud water contributed 2, 39, and 73% of total annual SO_4^{2-} deposition at elevations of 600, 1025, and 1350 m, respectively, on Whiteface Mountain as well as 1, 28, and 59% of total annual NO_3^- deposition at the same elevations. This shift in the fraction of total deposition contributed by cloud water at high elevations is confirmed by the results from MADPro. The MADPro results from the 1350-m elevation site at Whiteface Mountain show that approximately 91 and 84% of SO_4^{2-} and NO_3^- deposition, respectively, were from cloud water interception. Again, the dry and wet deposition estimates from Whiteface Mountain are likely to be underestimated because of the elevation difference in sampling locations. Scaling the dry and wet deposition estimates to the 1350-m cloud-modeling site would reduce the percent composition of

Table 7a. Dry, wet, and total seasonal depositions (June–September) for Whiteface Mountain and two nearby CASTNet sites for 1995–1998 (kg/ha).

		H^+			S			N		
Deposition Type	**Year**	**Whiteface Mtn.**	**WST109**	**CTH110**	**Whiteface Mtn.**	**WST109**	**CTH110**	**Whiteface Mtn.**	**WST109**	**CTH110**
Dry	1995	0.080	0.013	0.116	0.349	0.136	1.202	1.033	0.155	1.235
	1996	0.057	0.011	0.095	0.321	0.116	1.074	0.713	0.121	0.971
	1997	0.147	0.013	0.113	0.706	0.162	1.402	1.837	0.143	1.086
	1998	0.101	0.009	0.108	0.641	0.119	1.608	1.265	0.091	0.930
Wet	1995	0.125	0.198	0.203	1.945	2.762	3.325	1.001	1.250	1.594
	1996	0.103	0.188	0.231	1.489	2.822	3.765	0.796	1.234	1.615
	1997	0.143	0.198	0.154	2.328	2.996	2.857	0.919	1.335	1.249
	1998	0.203	0.189	0.200	3.688	2.944	3.398	1.421	1.297	1.490
Total	1995	2.230	0.942	1.490	35.180	2.898	4.527	15.380	1.405	2.828
	1996	1.230	0.840	1.484	26.770	2.938	2.840	10.540	1.355	2.686
	1997	1.720	0.903	1.229	34.060	3.168	4.259	15.210	1.478	2.335
	1998	1.750	0.932	1.301	29.990	3.063	5.006	12.390	1.387	2.420

Note: WST109 is the designation for the CASTNet site at Woodstock, NH (182 km from Whiteface Mtn.); CTH110 is the designation for the CASTNet site at Connecticut Hill, NY (312 km from Whiteface Mtn.).

total deposition from cloud deposition to approximately 84% for SO_4^{2-} and 71% for NO_3^-. This provides better agreement with the results of Miller et al.[43]

The variation in cloud water deposition with cloud frequency produces an elevational gradient in total chemical deposition.[43] A summary by Lovett and Kinsman[10] suggests that eastern United States sites at elevations below 1000 m receive less than 20% of total SO_4^{2-} deposition via cloud water, but sites above 1500 m receive 45–80% from cloud water. These estimates are in agreement with independent summaries by NAPAP[16,50] and with the MADPro results presented here.

Table 7b. Dry, wet, and total seasonal depositions (June–September) for Whitetop Mountain and two nearby CASTNet sites for 1996 (kg/ha).

Deposition Type	Analyte	Whitetop Mtn.	PNF126	VPI120
Dry	H^+	0.227	0.085	0.122
	S	1.020	0.980	1.319
	N	2.908	0.942	1.312
	NH_4^+	0.187	0.201	0.196
Wet	H^+	0.071	0.124	0.204
	S	0.971	1.772	3.475
	N	0.409	0.655	1.218
	NH_4^+	0.040	0.567	0.799
Total	H^+	2.130	0.209	0.326
	S	37.250	2.752	4.794
	N	16.220	1.597	2.530
	NH_4^+	14.720	0.768	0.995

Note: PNF126 is the designation for the CASTNet site at Pisgah National Forest, NC (71 km from Whitetop Mtn.); VPI120 is the designation for the CASTNet site at Horton Station, VA (120 km from Whitetop Mtn.).

CONCLUSIONS AND RECOMMENDATIONS

Since its beginning in 1994, MADPro has produced a 6-year data set that is comparable to data produced by past networks. The field standard operating procedures, quality assurance program, and laboratory analytical methods for MADPro were designed, when possible, with this objective of comparability in mind. Differences in equipment with respect to past networks consisted mainly of the use of an automated cloud water sampler and a continuous LWC measurement system (PVM-100). These changes were implemented as refinements identified as necessary from past experience. Two of the three permanent MADPro sites were located at sites operated in past studies, and all three sites collected samples during the warm season, which also mirrored past networks. More than 5300 valid hourly samples were collected over the 6-year period of operation. Therefore, MADPro has successfully achieved its two main objectives: to develop and implement a cloud water measurement system to be used in a network-monitoring environment and to update the cloud water and deposition data collected in the Appalachian Mountains during MCCP. MADPro has produced one of the largest cloud water data sets in the world (based on the number of samples collected from 1994 to 1999).

For the major ions, Whiteface Mountain exhibited the lowest mean and median concentrations, and Clingman's Dome exhibited the highest mean and median concentrations. This is indicative of the north–south concentration gradient noted during MCCP.[6,21] This gradient is partly an effect of the different meteorological conditions experienced at northern versus southern sites as well as a difference in the back trajectories of air masses reaching the sites. Air mass origin may also be an important factor in the upward trends in seasonal SO_4^{2-}, NO_3^- and H^+ observed at Clingman's Dome during the study period. The MADPro sites experienced additional pollution loading on the order of 6–20 times for SO_4^{2-} and NO_3^- compared with lower elevation CASTNet sites. Approximately 80–90% of this extra loading was from cloud deposition.

The dry and wet deposition fraction estimates from the MADPro sites were relatively comparable to dry and wet deposition amounts at nearby CASTNet sites. A clear north–south pattern in total deposition amounts is not evident from the data presented here. However, data completeness for deposition values for the southern sites must be improved before further conclusions can be made. A review of the available monthly cloud and total deposition values from 1994 through 1999 does show that, unlike cloud water concentrations, Whiteface Mountain experiences equal, if not greater, amounts of cloud and total deposition as the southern sites. The higher deposition rates at Whiteface Mountain relative to cloud water concentrations are in large part caused by higher wind speeds and LWC experienced at this site.

To further increase regional representativeness of the cloud deposition values, canopy structure must be ascertained accurately and very specifically throughout areas of the eastern United States that are frequently impacted by clouds. Use of satellite sensors for this purpose is recommended. This information will enable a more refined estimate of deposition values as compared with the homogenized deposition estimates provided in this paper. To establish an east–west gradient, additional sites in the Green or White Mountains of New England are recommended. An additional site in West Virginia is also recommended because of its proximity to Ohio River Valley pollutant sources.

Based on the findings of this study that locations above 1500 m can experience pollution loadings up to 20 times greater than lower elevation sites and that areas such as Clingman's Dome are experiencing statistically significant increases in ions such as NO_3^-, it is extremely important to continue monitoring cloud water concentrations and depositions at such locations and to assess ecosystem impacts.

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APPENDIX

Model Parameterization

Comparing cloud deposition rates across sites is difficult because canopy structure (e.g., height and LAI) can vary between locations even within a single forest. To focus on meteorological and chemical controls on the deposition process, a "standard" canopy structure was used for all CLOUD model runs at all sites. The structure chosen was described by Lovett.[31] It is a monospecific balsam fir canopy with the following parameters: maximum structure height of 1061 cm, zero-plane displacement of 837 cm, roughness length of 97 cm, and LAI of 7.6 m^2/m^2. While this canopy parameterization was originally developed for the subalpine forests of New Hampshire, it is similar to forests at high elevations on Whiteface Mountain.[43] Forests near the summits of Whitetop Mountain and Clingman's Dome are similar in structure but may be somewhat taller.[47] Furthermore, the deposition model calculations represented the deposition expected to a forest stand with a homogeneous canopy structure. If that stand were at a forest edge, the deposition would be greater.[51] However, no information was available on the gap and edge distribution in the forests at the three MADPro sites. Consequently, deposition estimates were made only for homogeneous canopies.

The model calculates the droplet size distribution associated with each measured LWC according to the formula of Best.[52] The calculation requires specifying the mode of the distribution. Because droplet size information was not collected at the sites, the droplet size at all sites was assumed to be a function of LWC as reported for the Whitetop Mountain site by Joslin et al.[53] Best[52] provides a method for calculating the droplet size distribution when the modal droplet diameter is known. Data from Whitetop Mountain were used to estimate the mode of the size distribution as a function of LWC. The Best function was then used to calculate the full droplet size distributions.

In the original model, three droplet diameter classes were used: 0–10, 10–20, and 20–30 μm. Miller et al.[43] suggested that as many as 500 droplet size classes may be necessary to simulate the deposition rate to within 0.1%.

Simulations were run with different numbers of droplet size classes, and it was found that the deposition velocity using 20 classes was within <1% of the deposition velocity using 500 classes. Accuracy of <1% was considered sufficient, given the error inherent in calculating or measuring a droplet size distribution, and 20 droplet size classes (in the range of 0–100 μm) were used for all simulations.

The model uses canopy-top wind speed as an input parameter. However, in practice, wind speed is usually measured some distance above the canopy to avoid shadowing by surrounding tree crowns. Assuming a logarithmic wind profile above the canopy, the canopy-top wind speed (u_h) can be calculated from the wind speed at any height z (u_z). The anemometers at Clingman's Dome and Whitetop Mountain were 3.2 m above the canopy, and that information was used to calculate canopy-top wind speed. The cloud chemistry, LWC, and wind speed measurements at Whiteface Mountain were made on the roof of the summit observatory building at an elevation of 1483 m. Miller et al.[43] reported an elevational gradient of canopy structure on Whiteface Mountain, and their highest site, at an elevation of 1350 m, closely resembled the height and species composition of the forest used in the deposition calculations. Therefore, 1350 m was used as the elevation for the model application at Whiteface Mountain. Wind speed, LWC, and cloud chemistry all change with elevation, and, therefore, the measured values at the canopy top were scaled for modeling at lower elevations as described by Miller et al.[43]

Based on data from the summit and a site at 1050 m on Whiteface Mountain, Miller et al.[43] estimated that cloud LWC decreases linearly with elevation on Whiteface Mountain from a growing-season mean of 0.5 g/m^3 at the summit to 0.39 g/m^3 at the 1050-m site. This represents a 22% decrease in LWC over a 433-m elevation drop. All LWC values reported for Whiteface Mountain were adjusted to calculate the LWC at the modeled site.

Model Sensitivity

The sensitivity of the model to various input parameters, including wind speed, droplet size, and canopy structure, has been reported in several publications.[31,43,54,55] A sensitivity analysis provides a framework for assessing the potential for bias and extent of uncertainty in MADPro cloud water deposition estimates. For this purpose, a semi-independent "QA Model"[43] was used that employs alternative parameterizations and some additional model components beyond those offered by the "primary" model.[56] The MADPro standard canopy description, as presented earlier, was used with the QA Model. The general sensitivity analysis suggests that ±15% of the uncertainty in deposition estimates can be related to parameters such as canopy species composition, height, LAI, and vertical distribution of leaf area. There is a likely range of –10 to +20% uncertainty in deposition estimates because of uncertainty in key input data (LWC, wind speed, cloud immersion frequency, chemical concentrations) in some site-year data sets. Estimates at all three sites for all years are subject to a constant –8% bias resulting from numerical error introduced into the models by the choice of a 1-m model layer thickness. There is some potential for an unknown but possibly significant uncertainty in the deposition estimates at all sites arising from limited representation of meteorological and chemical conditions during cloud periods.

Specific results of the sensitivity analysis and relevant observations are discussed, by parameter.

- Cloud LWC and wind speed exert the dominant influences on cloud water deposition for a given forest canopy. Observations at Whiteface Mountain have shown that LWC varies linearly with altitude (1000–1400 m) as does wind speed at 1 m above the forest canopy. Cloud water fluxes to the standard MADPro canopy for 1 < wind speed < 20 m/sec showed variations of 0.50–1.90 mm H_2O/hr for LWC = 0.83 g/m^3; 0.15–1.10 mm H_2O/hr for LWC = 0.42 g/m^3; and 0.05–0.75 mm H_2O/hr for LWC = 0.26 g/m^3.
- The variation in cloud water ion concentrations between 1050 m elevation and the Whiteface summit appear to be a function of LWC, implying that air concentrations and scavenging efficiencies are more or less constant with elevation. Therefore, cloud water ion concentrations have been estimated at lower elevations by converting the summit cloud chemistry observations to air concentrations and calculating the ion concentrations at the elevation of interest using the estimated LWC and a constant scavenging efficiency.[44]
- Cloud immersion frequency is also strongly dependent on elevation in the northeastern United States. Miller et al.[43] reported an exponential increase in immersion frequency with elevation at Whiteface Mountain. This relationship has been used to scale the summit cloud frequency data to other elevations on the mountain.
- For a canopy with an assumed LAI of 10 m^2/m^2, fairly wide variations in canopy height (10–17 m) and LAI (6–12 m^2/m^2) individually produced deviations in the QA Model response of –10 to +3% for 6 < LAI < 12 m^2/m^2 and 0.26 < LWC < 0.83 g/m^3.
- Broad leaf versus needle leaf vegetation has only a minor effect on deposition rates. For example, a

forest mixture of 16% broad leaf and 84% balsam fir yields a deviation from the standard MADPro canopy of +2.5 to –2% for 4 < wind speed < 12 m/sec and 0.21 < LWC < 0.83 g/m^3.

- For a canopy with an assumed LAI of 10 m^2/m^2, decreasing the canopy height from 17 to 10 m decreased cloud water deposition by at most 3.5% for 0.21 < LWC < 0.83 g/m^3.
- At a constant canopy height of 10 m, variation in LAI from the MADPro canopy value of 10 m^2/m^2 resulted in a mixed response, depending on the combination of LWC (0.26–0.83 g/m^3) and wind speed (3.6–12.3 m/sec). Generally, deviations were in the range of +2 to –4%. However, conditions of low wind speed combined with medium and high LWC produced deviations of –6.5 and 9.8%, respectively, at 6 m^2/m^2 LAI.
- Variations of the droplet size distribution suggest a complicated pattern of QA Model response to different combinations of LWC and wind speed. Using the two different Whiteface Mountain and Whitetop Mountain drop size distributions results in a difference in droplet distribution of 1–24% for all LWC at wind speeds below 10 m/sec. At wind speeds greater than 10 m/sec, use of the Whitetop Mountain distribution increased deposition rates from 1 to 6% over the results obtained with the Whiteface Mountain distribution. This model response tended to average out over the broad range of combinations of wind speed and LWC encountered at Whiteface Mountain. Seasonal mean cloud water fluxes (mm H_2O) differed by approximately 3% using the Whitetop Mountain distribution for the Whiteface Mountain 1997 data set.

In conclusion, uncertainty estimates for cloud water deposition are nearly directly proportional to uncertainty in wind speed and LWC measurements, both of which are thought to be small for most site-year data sets. Deposition estimates are directly proportional to cloud frequency, a parameter that is also well known at each site for the majority of the months of study. The response of the cloud water model appears to be least sensitive to the least constrained parameters, such as canopy species composition, height, LAI, and vertical distribution of leaf area (±12% or less for 10–40% changes in these parameters).

The model does not depend on temperature, RH, solar radiation, or evaporation. Two other parameters, although not used in the calculation of instantaneous deposition rates in the model, are important in the final calculation of cloud deposition values. These include cloud chemistry and cloud frequency (the percent of hours that the site is immersed in cloud). Calculated cloud deposition totals are directly proportional to both of these factors. For example, if other parameters are held constant, doubling the cloud SO_4^{2-} concentration will double the calculated cloud SO_4^{2-} deposition, and doubling the cloud frequency will double the calculated deposition of all ions.

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