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DETERMINATION OF CLOUD WATER ACIDITY AT A MOUNTAIN OBSERVATORY IN THE ADIRONDACK MOUNTAINS OF NEW YORK STATE

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Determination of Cloud Water Acidity at a Mountain Observatory in the Adirondack Mountains of New York State

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A ground-based, cloud water collection system capable of intercepting significant volumes of water from clouds has been in operation at a mountain field station in the Adirondack Mountains of New York State for three summer seasons. An analysis of cloud water pH for August–September 1977 and 1979 strongly suggests that acid input to the ecosystem due to clouds at Whiteface Mountain may be significantly more important than that due to the much-discussed phenomenon of acid precipitation. The mean pH of nonprecipitating clouds was 3.55 during 1977 and 3.50 during 1979; approximately 90% of all observations were found in the pH interval between 2.66 and 4.66. In the event that precipitation is intercepted concurrently with cloud droplets, a minimum dilution of the cloud water acidity by a factor of 60–80% can be expected. Other aspects of the collection and pH analysis of cloud water at Whiteface Mountain are also discussed.

INTRODUCTION

In 1976, the Atmospheric Sciences Research Center (ASRC) began a routine program of continuous cloud water collection and pH analysis at Whiteface Mountain (44.4°N, 73.9°W) in the northeastern Adirondack Mountains of New York State. The program represented a logical extension of ASRC participation in the Multistate Atmospheric Power Production Pollution Study (MAP3S) at the Whiteface Mountain Field Station [*Battelle Pacific Northwest Laboratories*, 1977], as well as an unusual opportunity to define the magnitude and variation of cloud water acidity in high-elevation ecosystems. In reviewing recent, summer season weather records at Whiteface Mountain, it was clear that the total number of cloud (sometimes referred to as 'high mountain fog') hours was at least four to five times greater than the corresponding number of rain hours above an altitude of 1100 m or so. Numerous studies have already pointed out that a sizeable proportion of the annual water accumulation in mountain regions can be attributed directly to the horizontal interception of cloud moisture by forest vegetation [*Kitredge*, 1948; *Twomey*, 1957; *Vogelmann et al.*, 1968; *Schlesinger and Reinners*, 1974]. Consequently, because orographic cloudiness is a more common occurrence than precipitation on the upper slopes of Whiteface, and in anticipation that acid deposition by clouds could be of major ecological importance in high-elevation ecosystems, we designed a field program to quantify the acidity of clouds which pass the summit of Whiteface Mountain.

Our review of the literature indicates that continuous cloud water collection and chemical analysis is the exception, rather than the rule. Table 1 summarizes the results of numerous cloud water acidity studies. Airborne cloud chemistry programs [cf. *Oddie*, 1962; *Petrenchuk and Drozdova*, 1966; *Bogen*, 1974; *W. D. Scott*, 1978; *Fricke et al.*, 1978; *B. C. Scott and N. S. Laulainen*, 1979] and most ground-based operations [*Houghton*, 1955; *Mrose*, 1966; *Okita*, 1968; *Lazrus et al.*, 1970; *Reinners et al.*, 1977] have depended upon the acquisition of bulk water samples. The ASRC cloud water collection system [*R. E. Falconer and R. Farrell*, 1977] permits us to evaluate cloud water acidity (and such other parameters as conductivity and specific ion content) in real time. Such a system

offers the distinct advantage that pH changes during the course of a cloud event can be evaluated with respect to changes in local meteorology and the physical structure of the clouds themselves. Also, the problems of chemical changes which occur in stored rain (or cloud) water samples, and which could affect acidity [*Penkett et al.*, 1979] are avoided.

This paper describes the results of some of our recent investigations of cloud water acidity during August–September 1977 and 1979. Results from the 1978 summer field program at Whiteface were not extensive enough to include in this report. We view this research as a preliminary step towards understanding the complexities of cloud chemistry in high-altitude ecosystems.

ASRC CLOUD WATER COLLECTION SYSTEM

In keeping with recently recommended design criteria for collectors used in precipitation chemistry measurements programs [cf. *Galloway and Likens*, 1978], the ASRC cloud water collector is constructed entirely of plastic, as shown in Figure 1. The collector consists of two polypropylene discs of diameter, d , equal to 25 cm, separated and supported by several polypropylene rods of diameters 0.9, 1.2, and 2.2 cm, each 1.0 m in length. A continuous strand of Teflon FEP-fluorocarbon fiber ($d = 0.4$ mm) was strung back and forth between the discs, every 3 mm apart, along their outer periphery. A plastic funnel was secured to the base of the collector so as to direct intercepted cloud water through a length of Teflon tubing to the laboratory pH sensor. The interface between the pH probe of the sensor unit and the intake line is an assembly of polypropylene blocks (each 1.6 cm × 7 cm × 12 cm) which, through a succession of predrilled chambers, permit a controlled flow of water from the inflow to the outflow ports, across the pH probe itself. The water flow through the blocks is regulated to maintain approximately 20 ml of fresh sample at any one time at the probe. The output from the pH sensor is recorded on a strip chart.

Prior to any given cloud event, the collector is thoroughly cleansed with distilled water until the conductivity of the rinse water is within 5 $\mu\text{mho cm}^{-1}$ of the distilled water value. An acid prewash with 1% HCl is required only in the event of visible signs of accumulated debris on the strings and rods. As clouds develop, the collector is mounted on a telescoping

TABLE 1. Cloud Water pH Values Reported by Various Investigators

Investigator	Observation Period	Location	Sampling Method	Range of Mean pH
<i>Ground-Based Cloud Water Collection Experiments</i>				
Houghton [1955]	1954	Northeastern U.S.	Cloud water mechanically intercepted on metal screen. Bulk samples.	4.5-7.2
Mrose [1966]	?	German Democratic Republic	Cloud water mechanically intercepted on pervious cloth. Bulk samples.	3.8-5.1
Okita [1968]	1963	(a) Mt. Norikura (b) Mt. Tsukuba, Japan	Cloud water mechanically intercepted on copper screen. Bulk samples.	(a) 3.4-4.0 (b) 5.6-6.5
Lazrus et al. [1970]	1967	Pico del Oeste, Puerto Rico	Cloud water mechanically intercepted on aluminum screen. Bulk samples.	4.9-5.4
Castillo [1979]	1976	Whiteface Mt., New York	Cloud water mechanically intercepted on plastic strings and rods. Bulk samples.	3.4-4.2
<i>Aircraft-Borne Cloud Water Collection Experiments</i>				
Oddie [1962]	1960	United Kingdom	Mechanical interception on glass tube. Bulk samples.	4.4-7.2
Petrenchuk and Drozdova [1966]	1961-1964	European territory of the USSR	Bulk Samples.	3.35-5.90*
W. D. Scott [1978]	1976	Sydney, Australia	(a) Centrifuge system, (b) Mechanical interception. Both bulk samples.	(a) 5.8-7.5 (b) 4.6-6.6
Fricke et al. [1978]	1973	Upper Bavaria, Federal German Republic	Centrifuge system. Bulk samples.	3.6-5.4
B. C. Scott and N. S. Laulainen [1979]	March 5, 1977	Muskegon, Mich. (a) 670 m MSL (b) 1525 m MSL	Super cooled droplet accretion on nylon wand. Bulk samples.	(a) 4.0 (b) 3.7

*Most contaminated cloud water samples.

tower and raised to a height of 3 m above the observatory. At this height, rain splash from the roof is eliminated.

The design of the ASRC cloud water collector is intended to collect, as uniformly as possible, droplets whose size spectrum typifies mountain clouds. The theoretical basis for the collection efficiency for water droplets of varying size moving past cylinders of differing diameters has been developed by Langmuir and Blodgett [1946] and summarized by Mason [1971] and Winters et al. [1979]. The configuration and dimensions of the strings and rods chosen for the cloud water collector are based, in large measure, upon the conclusion that the collection efficiency of a cylinder (such as an individual string or support rod) increases with decreasing diameter and with increasing cloud droplet diameter. For stratiform mountain clouds, whose droplet number density is often uniform for droplet diameters less than 20 μm [cf. Castillo, 1979], the cloud water collector is capable of extracting sufficient water for chemical analysis in most situations. At this time, we are uncertain whether or not the capture efficiency of the entire collector differs from that anticipated for individual rods and strings. Also, the question of how efficient our collector unit remains as wind gustiness increases has not yet been investigated.

When precipitation occurs simultaneously with the interception of cloud water, the rate at which water is delivered to the polypropylene pH block can increase up to a factor of 5. Summit winds typically drive rain horizontally into the collector, where the droplets are preferentially impacted upon the larger rods. A practical scheme by which rainfall would be excluded from the cloud water collector has not been devised. Consequently, the design of the system forces us to collect a mixture of rain and cloud droplets as precipitation falls.

RESULTS

During the August-September measurements program, 824 hourly observations of pH were made during 1977, as compared with 125 such observations during 1979. The discrepancy between the numbers of hourly observations is largely due to a more extensive effort in 1977 to collect cloud water pH data. In 1979, because more diverse field chemistry and meteorological measurements were made during any one cloud collection period [cf. P. D. Falconer and J. A. Kadlecik, 1980] and because more frequent interevent calibration of the field equipment was necessary, we became more restrictive of the number of cloud events we wished to sample in these months.

On many occasions, the collection of cloud water was interrupted by periods of rainfall; such intervals were determined either visually or by reference to the observatory's recording rain gauge. Because, as we have mentioned, the cloud water collection system cannot exclude rain, we are forced to distinguish between the pH of nonprecipitating and precipitating clouds. Observations of rain pH in the absence of clouds along the upper slopes were not included in our field programs.

Frequency histograms for cloud water pH, including and excluding the occurrence of precipitation, are presented in Figures 2 and 3. A time-averaged pH of 3.61 was derived for all cloud water samples, including occasions of rainfall, during both the 1977 and 1979 field season (cf. Figure 2). Computation of these mean values was guided by the customary transformation, $\text{pH}_{\text{mean}} = -\log \left((1/n) \sum [\text{H}^+] \right)$, where $[\text{H}^+]$ is the molar concentration of hydrogen ions associated with each hourly observation of pH, and n is the number of pH observations in the sample. After removing all identifiable oc-

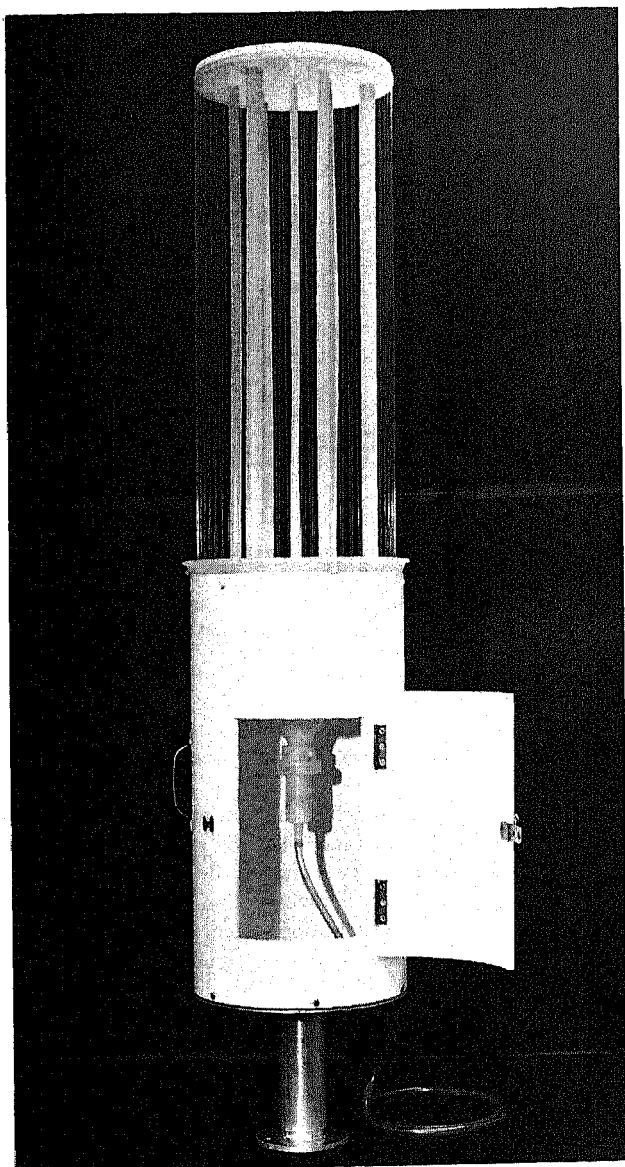


Fig. 1. The ASRC omnidirectional cloud water collector.

currences of rain from each cloud water collection period, the time-averaged pH_{mean} for nonprecipitating clouds was 3.55 during August–September 1977 and 3.50 for these same months in 1979. There is a further indication (cf. Figure 3) that the pH of nonprecipitating clouds is lower during times when the dew point temperature exceeds approximately $15^{\circ}C$; we will return to this point in a subsequent section. Table 2 summarizes the average August–September levels of cloud water acidity at Whiteface Mountain during our two years of study.

Changes in acidity for precipitating and nonprecipitating stratiform cloudiness at the summit are readily identified in the original strip chart records. Ordinarily the transition to rain is followed by an immediate increase in the pH number, as dramatically emphasized by Figure 4. As rain ceases, an equally abrupt pH decrease will frequently occur. A unit increase or decrease of the pH number is an extreme example of such a change, and corresponds to a tenfold difference in the hydrogen ion concentration between precipitating and nonprecipitating clouds. Our informal survey of the 1977 and 1979 strip chart records suggests that the most abrupt changes tend to occur during brief periods of air mass shower rainfall rather than during intervals of frontal rainfall. There are also indications that the greatest changes in pH which we observe in our collection system occur when rain begins shortly after cloud formation at the summit. These observations are, at present, tentative pending further field investigations.

The Wilcoxon test has been used to evaluate the significance of differences in sample mean pH 's for nonprecipitating clouds (pH_{np}) and precipitating clouds (pH_{pc}) which occur during the transition between these two conditions. The Wilcoxon test is particularly suitable when an exact statement of the true population distributions of pH_{np} and pH_{pc} cannot be made, and when sample size is small. In its application to this problem, because we suspect that there exists a difference of at least x pH units in the sample means, we introduce a general expression for the null hypothesis, H_0 , stated as follows:

$$H_0: pH_{pc} - x = pH_{np}$$

where pH_{pc} and pH_{np} are the 15-min (linear) averages prior to,

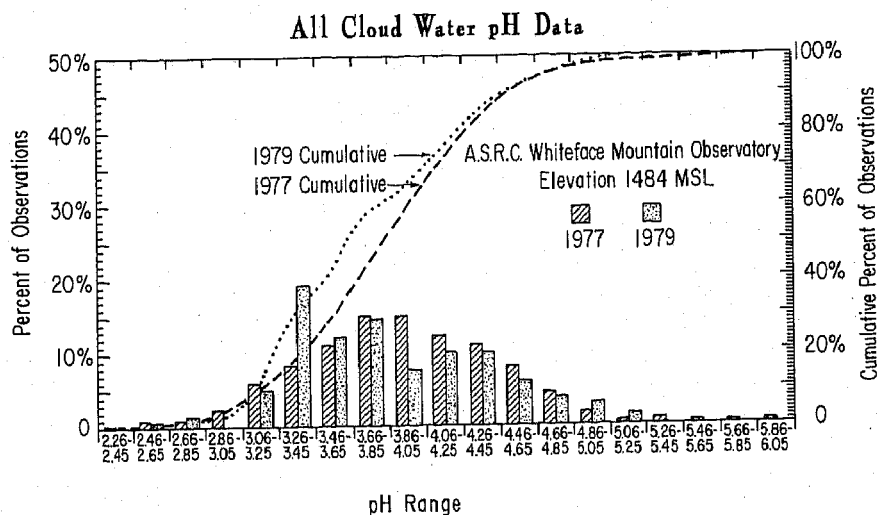


Fig. 2. Frequency histograms and the corresponding cumulative frequency distributions of hourly pH values for August–September obtained from all precipitating and nonprecipitating cloud events at the Whiteface Mountain Summit Observatory (1484 m) during 1977 and 1979.

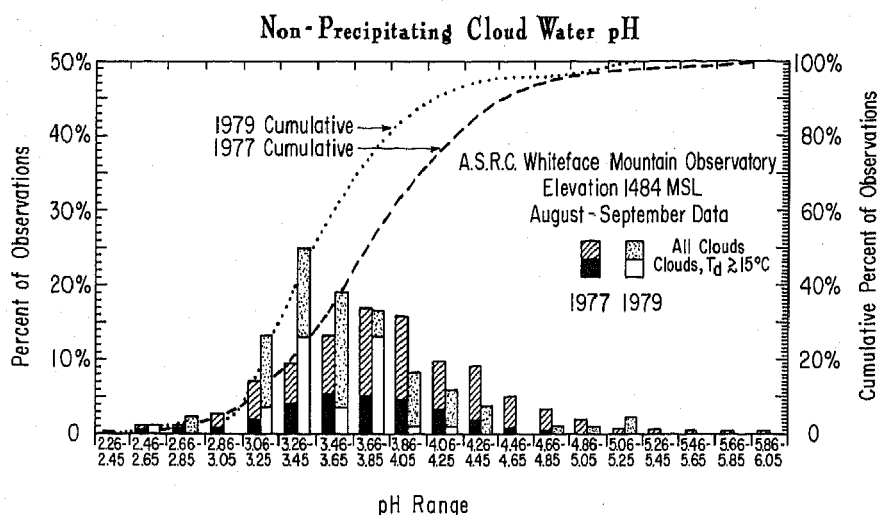


Fig. 3. Frequency histograms and the corresponding cumulative frequency distributions of hourly pH values for August-September obtained exclusively from nonprecipitating cloud events at the Whiteface Mountain Summit Observatory during 1977 and 1979. For each interval of pH along the abscissa, the percentage of nonprecipitating cloud water pH values when dew point temperatures (T_d) exceeded approximately 15°C (60°F) is also shown. T_d was registered at the ASRC Whiteface Mountain Field Station (610 MSL).

and following the transition. The transition is ordinarily complete within 20 min. The alternate hypothesis, H_A , is

$$H_A: (pH_{pc} - x) > pH_{np}$$

Whether or not we can confidently reject the null hypothesis and state that the pH differences which occur at the onset, or cessation, of precipitation will be at least x units depends upon the critical region we establish. For this study, a 5% level of significance has been used. By testing a sequence of values for x , we can establish a (one-sided) 95% confidence interval for the minimum pH change to be expected. For the transition from nonprecipitating to precipitating clouds, a minimum increase of 0.25 pH units (the value x in the null hypothesis) is indicated at the 95% confidence level; this is equivalent to an acidity decrease of at least 80%. The corresponding minimum

change of pH for the transition from rain back to clouds is 0.20 unit, roughly equivalent to an increase in acidity of at least 60%.

We may satisfy ourselves that dilution mixing of rain droplets with cloud droplets on the collection surface can account for nearly all of the abrupt pH changes described above. Depending upon the intensity of precipitation and the strength of the prevailing winds, the collection system will typically intercept two to four times more rain than cloud water. Using representative, late summer pH values for rain ($4.0 \leq pH_r \leq 4.5$) and for cloud droplets ($3.25 \leq pH_c \leq 3.75$) at Whiteface, and assuming that the rain-to-cloud interception ratios vary, as noted, between 2 and 4, we can account for pH differences ranging from 0.15 to 0.61 pH units by the simple process of dilution mixing.

TABLE 2. Mean Hydrogen Ion Concentration $[H^+]$ and pH for Various Cloud Water Conditions at Whiteface Mountain (1484 MSL) During 1977 and 1979

Data	Number of Hourly Observations	$[H^+]_{\text{mean}}, \mu\text{Ml}^{-1}$	pH_{mean}
1977			
All occurrences of cloud water	824	246	3.61
Precipitating cloud water	171	111	3.96
Nonprecipitating cloud water	653	282	3.55
Nonprecipitating cloud water when $T_d \geq 15^\circ\text{C}$	192	478	3.32
Nonprecipitating cloud water when $T_d < 15^\circ\text{C}$	461	200	3.70
1979			
All occurrences of cloud water	125	243	3.61
Precipitating cloud water	43	98	4.01
Nonprecipitating cloud water	82	320	3.50
Nonprecipitating cloud water when $T_d \geq 15^\circ\text{C}$	31	358	3.45
Nonprecipitating cloud water when $T_d < 15^\circ\text{C}$	51	296	3.53

Dew point temperatures (T_d) were obtained at the ASRC Whiteface Mountain Field Station (610 MSL).

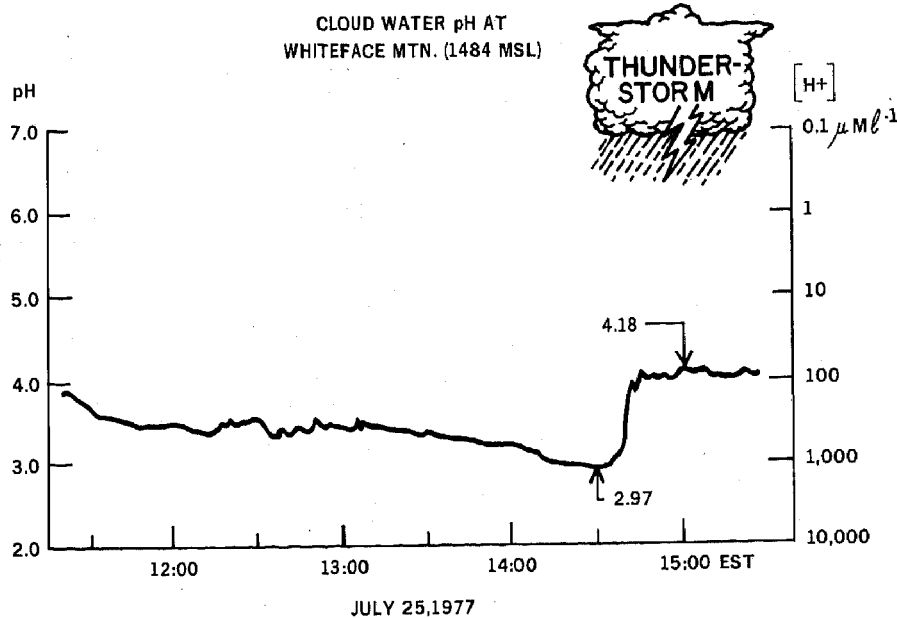


Fig. 4. Continuous record of cloud water pH at the summit of Whiteface Mountain on the afternoon of July 25, 1977. The summit was covered by nonprecipitating clouds from shortly before noon until the onset of a thundershower at about 1430.

Although dilution due to condensational growth of cloud droplets into rain droplets (a process which would add water volume but no additional acidity) would mathematically account for the pH changes observed, it can be shown that this process proceeds too slowly to significantly contribute to the formation of rain [cf. Mason, 1971; Fletcher, 1966]. For instance, in stratiform clouds composed of droplets in the 5- to 20- μm diameter range, the time required for condensational growth of these droplets to precipitation-sized particles of diameters 250–500 μm would be on the order of 1 day. Coalescence, on the other hand, is a much faster means of increasing the size of cloud droplets. For the same set of circumstances noted above, the likely growth of cloud droplets to rain droplets would be 3 to 4 hours [Mason, 1971]. However, the coalescence of droplets of similar pH would not result in the acid dilution of a final droplet population. We can find no compelling meteorological reason to believe that the abrupt cloud water pH increases which we observe whenever rain begins to mix into the collector can be produced by the condensation process.

Figure 4 shows an unmistakable, unit decrease of cloud water pH for the 3-hour collection period prior to the passage of a thunderstorm in the early afternoon of July 25, 1977. Although the collector was exposed at 1119 EST, clouds had already formed at the summit nearly 7 hours earlier. No specific measurements of the physical structure of the cloud deck were available for this weather system. However, both the duty observer's records and the relative humidity and solar radiation traces indicate that the stratiform overcast was gradually lifting and dissipating throughout the morning hours prior to rainfall. We have frequently observed that the pH of nonprecipitating clouds decreases during the final hours of cloud cover [Castillo, 1979; P. D. Falconer and J. A. Kadlecik, 1980] and believe the result to be consistent with acidification due to the evaporation of cloud droplets, rather than to any increase in the acid input to the cloud system. In the simplest example of the evaporative effect upon cloud water pH consider any

meteorological process, say, the entrainment of drier air into the cloud deck or a decrease in the supersaturation ratio, which gradually decreases the median volume droplet radius by 50%. Evaporative loss of water from the droplets would produce a nearly tenfold increase in the acidity which, in turn, represents a unit decrease in the pH number. Condensational growth of a nascent droplet would, by similar reasoning, result in a reduction of acidity. Reasonable rates of condensational growth and evaporative dissipation of a population of cloud droplets can be used to explain most aspects of the time-dependent pH variations of nonprecipitating clouds which we measure at Whiteface.

In addition to the differences in pH which we observe between precipitating and nonprecipitating clouds at Whiteface Mountain, the acidity of cloud water is also a function of air mass source. During nearly half of all summer months, air whose origin would be classified as maritime tropical enters northern New York State from the south [Bryson, 1966]. During these warm weather conditions, visibilities gradually deteriorate as the concentrations of water vapor, suspended particulates and oxidants increase. The dew point temperature, although not normally used to discriminate between air mass types, can be used with little ambiguity to identify maritime tropical air provided that the dew point temperature which we measure at the ASRC Field Station (610 MSL) exceeds approximately 15°C (60°F). Our information observation that cloud forms at the summit are most acidic when dewpoint temperatures are high, that is to say, when maritime tropical air prevails throughout the Adirondack Mountain region, is supported by the data presented in Figure 3 and Table 2.

DISCUSSION

The routine cloud water collection and pH analysis program at Whiteface Mountain appears to offer an important perspective on the issue of acid precipitation. Nearly 90% of all (nonprecipitating) cloud water pH values lie in the range of 2.66–4.66. By comparison, the pH of even the most acidic rain

collected at Whiteface is seldom less than about 4.0. Given the much greater proportion of cloud hours to rain hours, and the ability of exposed vegetative surfaces to intercept appreciable quantities of cloud moisture, we suggest that acid deposition along the upper slopes of Whiteface Mountain, and along other mountain ranges in the Northeast, will be contributed primarily by clouds. We have presented data which further suggests that the physical structure of the cloud itself, that is, whether it is growing or dissipating, is an important determinant of cloud water acidity.

The ASRC cloud water collector has proven to be a reliable means of extracting a sufficient volume of water from clouds passing the summit. Moreover, the ability to monitor and record real time variations in cloud water pH represents a distinct advantage over bulk sampling methods because the relationship between meteorological processes and cloud chemistry can be studied in greater detail as the cloud event develops. The prime disadvantage of our collection system is that precipitation cannot be easily eliminated from cloud water samples. A standard rain water collection system would be required for a true discrimination between the chemistry of rain and clouds.

CONCLUSIONS

Our field study of cloud water collection and pH analysis permits us to draw the following conclusions:

1. The time-averaged pH of late summer clouds, both precipitating and nonprecipitating, was 3.61 in both 1977 and 1979. The average pH levels for nonprecipitating clouds were 3.55 and 3.50 during 1977 and 1979, respectively. In instances where precipitation was collected concurrently with clouds, the average pH values were 3.96 and 4.01 for these same years.
2. The acidity of nonprecipitating cloud water is greater for an air mass classified as maritime tropical than for air masses of other origins. Using dew point temperatures in excess of approximately 15°C as an unambiguous indication of maritime tropical air at Whiteface, the corresponding mean pH of nonprecipitating clouds is 3.32 in 1977 and 3.45 in 1979. Otherwise, for lower dew point temperatures, the mean pH values are 3.70 and 3.53, respectively.
3. At the onset and cessation of rain, a rapid change in cloud water pH frequently occurs. A (one-sided) 95% confidence interval indicates a pH increase of at least 0.25 pH units as precipitation begins, and a decrease of at least 0.20 units as precipitation ceases. For typical cloud and rain water acidities and collection rates, simple dilution mixing appears to explain the reduced acidity of precipitating cloud water.
4. The evolving, physical structure of mountain clouds is probably a major determinant of cloud water acidity. Condensational growth or evaporation of a cloud droplet can reasonably explain the time dependence of nonprecipitating cloud water pH through the collection period.

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