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Chemical climatology of high elevation spruce–fir forests in the southern Appalachian mountains

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The physical and chemical climatology of high elevation (>1500 m) spruce–fir forests in the southern Appalachian mountains was studied by establishing a weather and atmospheric chemical observatory at Mt Mitchell State Park in North Carolina (35° 44' 05" N, 82° 17' 15"W). Data collected during the summer and autumn (May–October) of 1986, 1987, and 1988 are reported. All measurements were made on or near a 16·5 m walk-up tower extending 10 m above the forest canopy on Mt Gibbes (2006 m msl), which is located approximately 2 km SW of Mt Mitchell. The tower was equipped with standard meteorological instrumentation, a passive cloud water collector, and gas pollutant sensors for O_3, SO_2, NO_x. The tower and nearby forest canopy were immersed in clouds 25 to 40% of the time. Non-precipitating clouds were very acidic (pH 2·5–4·5). Precipitating clouds were less acidic (pH 3·5–5·5). The dominant wind directions were WNW and ESE. Clouds from the most common wind direction (WNW) were more acidic (mean pH 3·5) than those from the next most common wind direction (ESE, mean pH 5·5). Cloud water acidity was related to the concentration of SO_4^{2-}, and NO_3^- ions. Mean concentration of H^+, NH_4^+, SO_4^{2-}, and NO_3^- ions in the cloud water varied from 330–340, 150–200, 190–200 and 120–140 μmol litre^{-1} respectively. The average and range of O_3 were 50 (25–100) ppbv (109) in 1986, 51 (26–102) ppbv in 1987, and 66 (30–140) during the 1988 field seasons, respectively. The daily maximum, 1-h average, and 24-h average concentrations were all greatest during June through mid-August, suggesting a correlation with the seasonal temperature and solar intensity. Throughfall collectors near the tower were used to obtain a useful estimate of deposition to the forest canopy. Between 50–60% of the total deposition of SO_4^{2-} was due to cloud impact.

INTRODUCTION

Forests are exposed to a variety of chemical and physical stresses (Bruck *et al.*, 1989; Cowling, 1989; Saxena *et al.*, 1989; Aneja *et al.*, 1990 *a*, *b*; Saxena & Lin, 1990) known to be injurious to some species of trees (Prinz, 1987; Klein & Perkins, 1988). At many high elevation locations in the northern and southern Appalachians, spruce–fir forests have recently shown marked losses of foliar biomass, decreases in growth, and mortality (Johnson, 1983; Johnson & Siccama, 1983; Adams *et al.*, 1985; McLaughlin, 1985; Schütt & Cowling, 1985; Bruck & Robarge, 1988). These changes in forest condition have led to the suspicion that stresses induced by airborne chemicals may be adding to the natural insect, fungal, cold, drought, and nutritional stresses under which these forests grow (Woodman & Cowling, 1987; Bruck, 1989).

Visible changes in forest health are a conspicuous feature of the Fraser fir (*Abies fraseri* (Pursh.) Poir.) forest at the summit of Mt Mitchell and a more subtle

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feature of the lower elevation spruce (*Picea rubens* Sarg.) forests. The balsam woolly adelgid (*Adelges piceae* Ratz) is epidemic on the Fraser firs on Mt Mitchell, but does not affect red spruce. Ice breakage of spruce and fir stems occurs periodically (Nicholas & Zedaker, 1989). The most conspicuous changes in forest conditions occur at the highest elevations where cloud exposure is most frequent. Clouds are now known to be an important pathway of pollutant/nutrient deposition and capture of highly concentrated solution droplets (Bruck *et al.*, 1989; Murthy & Aneja, 1990; Saxena & Lin, 1990). Ozone is also known to occur in phytotoxic concentrations in high elevation forests (Aneja *et al.*, 1991).

Air pollution stress to a forest or vegetative ecosystem occurs whenever forest trees are exposed to toxic concentrations of gases, such as ozone, sulfur dioxide, hydrogen peroxide, fluoride, or when trees are exposed to accumulation of toxic chemicals in soil (Heck *et al.* 1986). Convincing data exist for vegetative injury caused by ozone (Winner *et al.*, 1989). Damage to crops in the United States from ozone alone has been estimated to be $2 to $5 billion annually (Heck *et al.*, 1988).

The specific sources of pollutants deposited at any given site are largely unknown, but are generally acknowledged to result from the widespread use of coal, oil, and motor fuels in North America. Under a national program entitled Mountain Cloud Chemistry/Forest Exposure Study (MCCP) and funded by the US Environmental Protection Agency (EPA), six high elevation sites are monitoring the chemical and physical climate in the eastern United States. Mt Mitchell is the southernmost site of this network.

In this paper we present an overview of the temporal variability in physical climatology and chemical climatology (gaseous pollutants, cloud water chemistry, canopy deposition) at a high elevation site near Mt Mitchell State Park in North Carolina (35° 44' 05"N, 82° 17' 15" W) during the summer and autumn (May–September) of 1986, 1987 and 1988.

Experimental

A 16·5 m meteorological tower was installed at Mt Gibbs (2006 m msl), just outside the Mt Mitchell State Park, about 2 km southwest of Mt Mitchell. The aluminum walk-up tower was equipped with an electronic weather station capable of measuring temperature, relative humidity, wind speed and direction, solar radiation, and barometric pressure (Table 1). A precipitation sampler was positioned in a clearing near the tower. Meteorological sensors were placed at least 10 m above the forest canopy, which mostly consists of 6– to 7–m Fraser fir trees.

The tower was also equipped with an elevator assembly with an attached instrument carriage on the north face of the tower. The instrument carriage was equipped with two cloud collectors: one passive (ASRC type; Falconer & Falconer, 1980) and one active (Caltech type; Daube *et al.*, 1987), as well a Forward Scattering Spectrometer Probe (FSSP). The cloudwater

Table 1. Measured parameters and instruments used at the Mt Mitchell research station during the observational periods (1986–1988)

Parameter	Instrument	Frequency
Automatic mode		
Wind speed	Propeller anemometer	Continuous
Wind direction	Vane potentiometer	Continuous
Air temperature	Thermister	Continuous
Barometric pressure	Piezoresistive	Continuous
Relative humidity	Capacitor	Continuous
Solar Radiation	Silicon photocell	Continuous
Precipitation	Tipping bucket	Continuous
Cloud water sample	Caltech active collector	Continuous
O_3	UV photometry	Continuous
SO_2	Pulsed fluorescence	Continuous
NO, NO_2, NO_x	Emission spectroscopy	Continuous
Cloud detector	Forward scattering optical device	Continuous
Manual mode		
Cloud water sample	ARSC passive collector	Hourly
Cloud droplet spectrum	Forward Scattering Spectrometer Probe (FSSP)	Event
Liquid water content	Integration of FSSP data, and gravimetric sampler	Hourly
Throughfall	Funnel and buckets	Weekly
Cloud water and throughfall		
pH	Electrode probe	
Conductivity	Electrode probe	
Anions	Ion exchange chromatography	
Metal cations	Atomic absorption spectrophotometer	
Ammonium ion	Spectrophotometer	

collectors were used to collect bulk hourly samples of cloudwater during a cloud event. The immersion of the top of the tower in a cloud for more than 15 min, as noted by an observer and an optical cloud detector (Mallant & Kos, 1990), was defined as the start of a cloud event. Manual cloud water collection then commenced at the top of the tower. Liquid water content (LWC) of the cloud was measured hourly by a gravimetric technique (Table 1).

Continuous, gas-phase measurements of ozone, sulfur dioxide and nitrogen oxides were made near the base of the tower using the instrumentation listed in Table 1.

Throughfall collectors consisted of 27–cm diameter polyethylene funnels placed 1 m above the forest floor, approximately 1 m away from the bole of a tree. Each funnel was connected to an opaque polyethylene covered collection pail (23–litre capacity) by a Tygon tube. Nylon 'wool' was placed at the top of each tube to prevent foreign matter from entering the collection pail. Ten collectors were positioned under the canopy at each site in 1987 and 1988 and two collectors were positioned in an adjacent clearing to collect rainfall.

Total volume for each collector was measured weekly. Subsamples from each collector were then combined in proportion, based on the total volume collected, to form a composite sample. These composite samples were stored at 4°C. The collectors were rinsed with de-ionized water after each collection or at least once every seven days during periods of no measurable rainfall or throughfall. Throughfall samples were collected on an event basis in 1986 using a different number of collectors. Results for these samples are presented elsewhere (Bruck *et al.*, 1989).

Chemical analysis consisted of pH, soluble Cl^-, NO_3^-, and SO_4^{2-}, and total dissolved Ca^{2+}, Mg^{2+}, K^+, Na^+, and NH_4^+ (Table 1). The pH of the cloud water samples was measured in the field within 15 min of collection and again in the laboratory. Internal and external audits were conducted during the sampling period at each field location and at the analytical laboratory. Chemical analyses were monitored using external quality control samples and by submitting separate cloud water subsamples to the Central Analytical Laboratory of the National Atmospheric Deposition Program. In addition, internal and external EPA audits were performed by US EPA personnel at the field site and in the chemical laboratory throughout each sampling season.

RESULTS AND DISCUSSION

Physical climatology

Meteorological parameters including temperature, relative humidity, precipitation, wind speed and direction were monitored continuously during the three field seasons. These parameters can be used as both input to deposition models for estimating pollutant fluxes into the forest ecosystem and references for understanding the dynamic roles of meteorological forces in determining the characteristics of montane clouds (Aneja *et al.*, 1990*a*). The ecosystem receives airborne chemicals in both dry form as gases and aerosols, and wet form as cloud water, rain, or snow (Dasch, 1988). Extremes of high wind speed, low temperature and drought; heavy loads of freezing rain and snow; and low humidity, provide periodic episodes that cause stress in montane forests (Nicholas & Zedaker, 1989).

The average and/or extreme values of meteorological parameters observed during the 1986 through 1988 field seasons are summarized in Table 2 as an aid to the interpretation of the cloud water chemical data and to describe the climatic conditions at the site.

Abnormally dry conditions prevailed in the southern Appalachians during the 1986 field season and continued into the summer of 1988. Part of the reason for the dry conditions during the 1986 field season was due to a persistent storm track, for the months of June and July, that extended from the Midwest into the Northeast. This storm pattern resulted in the summer of 1986 in the northern Appalachians being slightly cooler and wetter than normal. In contrast, this same storm

Table 2. Summary of meteorological data during the field seasons at Mt Mitchell site (1986–1988)

	Month				
Year	May	Jun	Jul	Aug	Sep
Average Temperature (°C)					
1986	11·6	13·3	14·0	12·2	11·6
1987	12·8	13·3	15·1	14·6	10·3
1988	10·8	13·6	14·8	15·2	12
Maximum	20·5	22·1	23·5	23·5	19·9
Minimum	2·0	0·4	6·1	−0·6	2·2
Mean	11·7	13·4	14·6	14·0	11·3
Average Relative Humidity (%)					
1986	—	79·7	82·3	90·0	87·9
1987	90·2	84·4	84·4	88·0	90·8
1988	71·4	73·3	84·2	88·1	88·5
Mean	80·8	79·1	83·6	88·7	89·1
Total Precipitation (mm)					
1986	57·2	2·3	0·0	4·3	22·9
1987	79·2	145·3	51·3	47·2	306·6
1988	26·9	36·8	96·8	105·2	145·3
Mean	54·4	61·5	49·4	52·2	158·3
Average Wind Speed ($m\ s^{-1}$)					
1986	6·7	6·1	8·3	6·4	6·4
1987	4·2	6·5	5·3	6·5	6·6
1988	5·6	5·8	5·5	5·6	6·5
Mean	5·5	6·1	6·4	6·2	6·5
Average Wind Direction (degrees)					
1986	272	293	296	299	285
1987	267	290	293	350	83
1988	285	293	271	245	266
Mean	275	292	287	298	211

pattern tended to leave the entire Southeast in a relatively hot and dry air mass, especially in July. The highest monthly average temperature on Mt Mitchell site 1 was 14°C in July (Table 2). The maximum wind speed was 37·2 m s^{-1}, which was also observed in July (Table 2).

During the 1987 field season, the northeastern states were primarily influenced by synoptic scale cloud and precipitation systems. As a result, the southern Appalachians again experienced another hot and relatively dry summer. Above-normal temperatures and below-normal precipitation were observed in the period July through August. The hottest period was during the middle of July, and the driest month was August. Average temperatures in July and August at the site were 15·1°C and 14·6°C, respectively (Table 2). Total precipitation in July was 51 mm on the site and in August 47 mm (Table 2). In contrast, precipitation in June and September was relatively high at 145 mm and 306 mm respectively.

In the 1988 field season, very high temperatures and low precipitation were experienced, especially during June and early July. The period April–June was very dry due to a strong upper level ridge of air over the central portion of the United States. More normal precipitation returned in July and August when the ridge weakened. Monthly maximum temperatures were higher in 1988 at 14·8°C and 15·2°C in July and August respectively, than during the previous two years (Table 2). Precipitation in June was low at 20·6 mm, but precipitation in July and August was relatively high at 98·9 mm and 97 mm respectively. The prevailing and secondary wind direction at the site during 3 years of observation were west-northwest (26·7%) and northwest (15·7%), respectively. Average wind speed was the highest in the prevailing wind direction, WNW. The most acidic clouds were also generally associated with the prevailing wind direction.

Relative humidity readings greater than 97% were highly correlated with the presence of clouds as determined by an observer during the 1986 field season. The period of time when relative humidity exceeded 97%, therefore, was taken as a measure of the frequency of cloud occurrence for each of the field seasons. The frequency of relative humidity readings greater than 97% during the 1986 field season was 42%. In 1987 and 1988, these values were 33% and 29%, respectively.

Chemical climatology

Gaseous pollutants

Beginning in 1986, hourly averaged ozone, and beginning in 1987, NO, NO_x, and SO_2 concentrations, were monitored continuously from May–September. Sulfur dioxide concentrations ranged from less than 0·6 ppbv (10^9) to as high as ~25 ppbv (observed in May, 1988). Levels of NO ranged from less than 2 ppbv to nearly 7 ppbv (June, 1987). Total oxides of nitrogen ranged from less than 2 ppbv to ~11 ppbv (observed in May, 1988). Ozone ranged from 10 to ~150 ppbv. Mean ozone concentrations were 50, 51, and 61 ppbv for the 1986, 1987, and 1988 field season, respectively.

The highest ozone concentrations were observed during the early portion of summer. The lowest concentrations were observed during September and October. The highest monthly mean ozone concentration (at the 99% confidence level) was observed during June for all the three years (~75 ppbv, ~60 ppbv, ~83 ppbv during 1986, 1987 and 1988 respectively). These maxima coincided with the maxima in temperature and solar radiation; suggesting an enhanced photochemical activity during this time period. The seasonal trend in the ozone concentration observed at Mt Mitchell agrees with that noted by Meagher and co-workers (1987) and Logan (1985), who reported summer maxima in ozone over most of the United States for latitudes ranging between 35° and 45° N.

A reverse diurnal variation in ozone concentration (with night time maxima) was noted for each of the three field seasons (Aneja *et al.*, 1990*b*). The average daytime value for the entire field season was 49 ppbv in 1986 compared to an average night-time value of 50 ppbv (~2% higher). The average daytime value was 49 ppbv in 1987 compared to an average night-time value of 53 ppbv (8% higher). Similarly, in 1988 the average daytime value was 65 ppbv, compared to the average night-time value of 68 ppbv (~5% higher). The differences between the daytime and night-time average concentrations were found to be statistically significant (*t*-statistic) at >95% confidence level for 1986, 1987, and 1988. When daytime and night-time values were compared on a month-by-month basis, the difference was statistically significant at the 99% confidence level. Generally, the ozone maximum occurred between 2000 and 2100 EST while the minimum was observed during midday. The nocturnal ozone maximum was thought to be due to the location of the site above the nocturnal inversion layer, which isolated the site from the effects of surface deposition (Aneja *et al.*, 1991).

Cloud water chemistry

Cloud events sampled during the 3 field seasons were segregated into three categories: long (> 8 h in duration), short (< 8 h in duration), and mixed (rainfall detected during cloud event). Long cloud events were mainly associated with frontal clouds (cumulus, stratocumulus), while short cloud events were either orographic clouds (mainly stratus) or fogs induced by radiative cooling (Saxena *et al.*, 1989).

The pattern and duration of cloud sampling differed between the three field seasons. In 1986, cloud sampling began on June 4th and was continued through August 30th. An attempt was made to sample every cloud event during this period. In 1987, a decision was made by MCCP to limit sampling to 3 three-week periods; one in May, one in July, and one in September. All cloud events were sampled during each of the three-

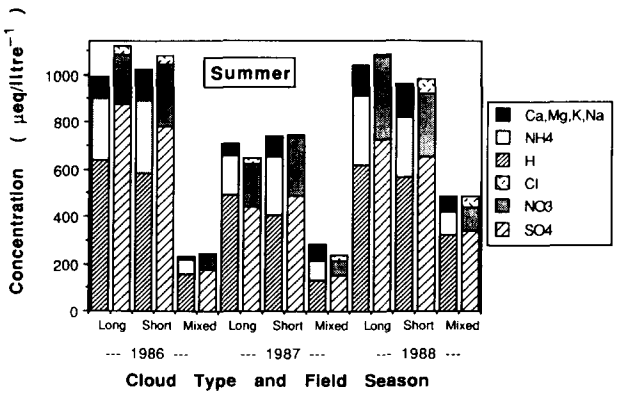

Fig. 1. Summary of chemical composition of clouds collected during the summer of the 1986, 1987, and 1988 field seasons.

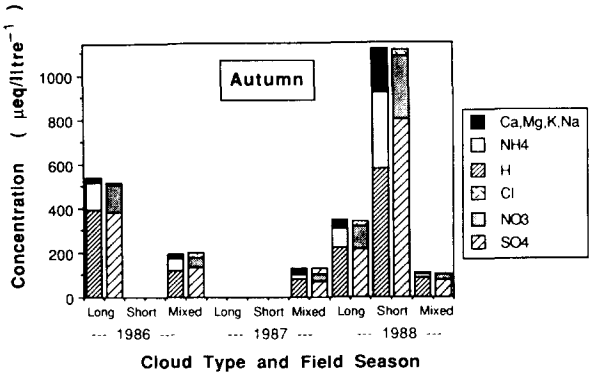

Fig. 2. Summary of chemical composition of clouds collected during the autumn of the 1986, 1987, and 1988 field seasons.

week periods. In 1988, cloud sampling began on May 10th and continued until September 30th. Every cloud event that occurred during this period was sampled.

Because of the decision to use relatively short sampling periods during the 1987 field season, the total number of samples collected (98) was considerably less than in 1986 (246) or 1988 (187). Comparisons of chemical composition between sampling seasons, therefore, may not be entirely valid because the database for 1987 was derived from a relatively small number of events. This difference in sampling frequency may account for the apparent lower acidity of clouds collected during the summer of 1987, as compared to the summer of 1986 or 1988 (Fig. 1). It was also probably the reason for the lack of results for long and short cloud events during the autumn of 1987 (Fig. 2). However, certain general comparisons in chemical composition for clouds from each field season were possible.

The ionic composition of the cloud water from each field season was dominated by H^+, NH_4^+, NO_3^-, and SO_4^{2-} ions (Figs 1 & 2), which agrees with similar observations in the eastern United States (Weathers *et al.*, 1988; Reisinger & Imhoff, 1989, Sigmon *et al.*, 1989). The concentration of SO_4^{2-} was highly correlated with H^+ concentration for all events sampled (r^2 = 0·89). Similar correlations were noted for SO_4^{2-} and NH_4^+ (r^2 = 0·83), and for NO_3^- and NH_4^+ (r^2 = 0·84). Sulfate was the dominant anion, while H^+ was the dominant cation (Figs 1 and 2).

Cloud water samples collected during long and short term events were more acidic than samples collected during mixed cloud events, which was due in part to differences in liquid water content (Table 3) (Reisinger & Imhoff, 1989). This trend was consistent for each of the 3 field seasons and between summer and autumn. Cloud samples from short-term events collected during the summer consistently had a higher percentage of NH_4^+ ions than samples from long-term events (Fig. 1). Overall the mixed cloud and rain samples had less of their acidity neutralized by NH_4^+ than either the long or short-term events (Figs 1 & 2), which demonstrated the ability of non-precipitating clouds to completely scavenge gaseous ammonia from the surrounding atmosphere (Saxena *et al.*, 1989).

Table 3. Mean, minimum and maximum pH values and mean liquid water content (LWC) for long, short and mixed cloud events sampled during the 1986, 1987, and 1988 field seasons

		Summer					Autumn				
Year	Cloud type	Number of samples	Mean (pH)	Min (pH)	Max (pH)	LWC (g m^{-3})	Number of samples	Mean (pH)	Min (pH)	Max (pH)	LWC (g m^{-3})
1986	Long	57	3·19	2·67	4·81	—	20	3·40	3·07	3·65	—
	Short	57	3·23	2·47	5·92	—	—	—	—	—	
	Mixed	132	3·81	2·73	5·35	—	15	3·92	3·33	4·16	—
1987	Long	35	3·30	2·83	3·80	0·29	5	4·51	4·02	6·70	0·17
	Short	24	3·30	2·82	4·24	0·21	—	—	—	—	
	Mixed	39	3·98	3·51	4·88	0·33	13	4·10	4·35	6·75	—
1988	Long	62	3·17	2·62	4·38	0·29	29	3·46	2·85	3·85	0·41
	Short	72	3·09	2·24	4·34	0·21	25	3·11	2·53	4·29	0·15
	Mixed	53	3·42	2·79	4·52	0·33	46	3·89	3·06	5·25	0·45

Table 4. Volume-weighted mean concentrations for rain and throughfall collected in 1987 and 1988

Collector	Year	pH	H^+ (μeq litre^{-1})	NH_4^+ (μeq litre^{-1})	NO_3^- (μeq litre^{-1})	SO_4^{2-} (μeq litre^{-1})	Other[a] (μeq litre^{-1})	Sum (μeq litre^{-1})
Rain	1987	4·43	37	14	13	44	30	138
	1988	4·34	46	11	13	48	18	136
Throughfall	1987	4·01	98	23	44	166	155	486
	1988	3·94	116	13	52	170	153	504

[a] Other = Cl^-, Na^+, K^+, Ca^{2+}, and Mg^{2+}.

Canopy deposition

Indirect estimates of the deposition of H^+, NO_3^-, NH_4^+ and SO_4^{2-} to the forest canopy were obtained using throughfall collectors. The volume-weighted mean concentrations for rain and throughfall samples collected during 1987 and 1988 are given in Table 4. The throughfall samples are more acidic than rain and are higher in ionic strength. The increase in ionic strength can result from the evaporation of deposited rain or cloud water from the needle surfaces, as demonstrated by Joslin and co-workers (1988) and Adams & Hutchinson (1987), but the increase in ionic strength is several times larger than that expected from evaporation during a rain or cloud event (Bredemeier, 1988). In fact, more water was collected via throughfall than rainfall (Table 5). The increase in ionic strength therefore, represents input from dry deposition, deposition via cloud impact, and the interaction of acidic deposition from all three sources with the canopy foliage. The degree of interaction with the canopy is reflected in the difference in volume-weighted mean concentrations between the rain and throughfall samples for Cl^-, Na^+, K^+, Mg^{2+}, and Ca^{2+} (labelled as Other in Table 4). Similar trends for these ions have been reported in natural canopies (Olson *et al.*, 1981; Bredemeier, 1988; Joslin *et al.*, 1988; Waldman & Hoffmann, 1988).

The interaction of acidic deposition with the forest canopy limits the usefulness of flux estimates for H^+, NH_4^+ and NO_3^- by dry deposition and cloud impact by calculating net throughfall (Table 5; Garten *et al.*, 1988; Lindberg & Garten, 1988). The net throughfall flux for H^+, NH_4^+,and NO_3^- in Table 5 only represents the net input of these ions into the forest floor due to dry deposition and cloud impact. Estimates of the deposition of SO_4^{2-} from these two sources can be obtained from net throughfall, however, if it is assumed that SO_4^{2-} is conserved within the forest canopy (Lindberg & Garten, 1988). Based on this assumption, deposition of SO_4^{2-} at our site was higher during the 1987 field season with 80% due to dry deposition and cloud impact. During 1988 there was a decrease in overall deposition of SO_4^{2-}, but the relative proportion from dry deposition and cloud impact (73%) remained essentially constant.

Deposition of SO_4^{2-} via cloud impact was undoubtedly the dominant source of SO_4^{2-} collected by the throughfall collectors (Table 5). Mueller & Weatherford (1988) estimated that dry deposition of SO_4^{2-} ha^{-1} per field season was in the range 6–9 kg ha^{-1} for a spruce–fir canopy located at Whitetop Mountain in Virginia. Taking their upper limit of 9 kg SO_4^{2-} ha^{-1} per field season for dry deposition would mean that between 50–60% of the total deposition of SO_4^{2-} at our site during the 1987 and 1988 field seasons was due to cloud impact.

CONCLUSION

The high elevation spruce–fir forests in the southeastern United States are exposed to natural stresses, such as severe ice storms (Nicholas & Zedaker, 1989) and insect attacks (balsam woolly adelgid on Fraser fir) that can cause relatively rapid and visually distinctive signs of forest decline and death (Prinz, 1987; Klein & Perkins, 1988; Andersson, 1989). The observations we report here, together with data from similar research

Table 5. Deposition of H^+, NH_4^+, NO_3^-, and SO_4^{2-} via rain and throughfall for the 1987 and 1988 field seasons

Collector	Year	Volume (cm^3)	H^+ (kg ha^{-1})	NH_4^+ (kg ha^{-1})	NO_3^- (kg ha^{-1})	SO_4^{2-} (kg ha^{-1})
Rain	1987	47·3	0·18	1·22	4·0	10·1
	1988	45·7	0·21	0·92	3·6	10·7
Throughfall	1987	64·1	0·63	2·66	17·6	51·0
	1988	49·3	0·57	1·13	15·8	40·3
Net Throughfall[a]	1987	16·8	0·45	1·44	13·6	40·9
	1988	3·6	0·36	0·21	12·2	29·6

[a] Net Throughfall = Throughfall—Rain.

sites in the southeastern United States (Reisinger & Imhoff, 1989; Sigmon *et al.*, 1989; Winner *et al.*, 1989) documents the presence of chemical pollutants that can exert a more subtle stress on these high elevation ecosystems (Bormann, 1988); in particular from ozone and acid deposition (predominantly as acid cloud water).

The mean and range of ozone concentrations we observed are within the range of ozone concentrations known to result in visible foliar damage to sensitive plant species (Garner *et al.*, 1989). Visible foliar damage from ozone has been documented along an elevation gradient in the Shenandoah National Park, Virginia (Winner *et al.*, 1989). Lack of a pronounced diurnal pattern in ozone concentrations means that plant species in these high elevation ecosystems receive a continuous dosage of ozone throughout the growing season. Continuous exposure to ozone at the concentrations we observed can result in physiological damage of internal structures without any indication of visible damage on the outer surfaces (Prinz, 1987; Reich *et al.*, 1987).

The frequency of exposure, ion composition, and range of acidity of cloud water that we observed are also important factors in regards to damage to plant foliage. Controlled experiments with red spruce seedlings have shown that intermittent exposures to acid mists with long drying periods in between causes greater amounts of foliar injury than continuous exposures (Jacobson *et al.*, 1990). Visible foliar symptoms of injury are also less or absent during such exposures using combined sulfuric–nitric acid mists. Intermittent exposures of acid mist at the mean pH of cloud water (3·4) that we observed can damage the epicuticular wax plugs in the epistomatal chamber of stoma on red spruce (Bruck *et al.*, 1989) and enhance foliar leaching of base cations (Joslin *et al.*, 1988). This in turn may have a detrimental effect on the waterholding capacity of foliage, especially under drought stress (Mengel *et al.*, 1989). These effects may be further aggravated by the presence of ozone, and there is a growing consensus that the continuous exposure to both ozone and acid cloud water that exists in these high elevation ecosystems can increase the sensitivity of trees to drought and freezing (Eamus *et al.*, 1990). As we have noted, the years 1986–1988 in the southeastern United States were below average in rainfall with substantial periods of drought, especially in the fall of 1986 and early spring of 1988. This added water stress combined with the levels of pollutant exposure we have documented will undoubtedly contribute to the continued long-term decline of the ecosystem.

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