

Wetfall Deposition and Precipitation Chemistry for a Central Appalachian Forest

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ABSTRACT

Although extensive research on acidic deposition has been directed toward spruce-fir forests, less research has been done on the impacts of air pollution on eastern montane hardwood forests. The purpose of this study was to describe precipitation chemistry for several Appalachian hardwood forest sites at or near the Fernow Experimental Forest (FEF) to assess the potential for problems associated with acidic deposition. Emphasis was placed on seasonal patterns of ionic concentrations (H^+ , Ca^{++} , NH_4^+ , NO_3^- , and SO_4^{2-}) and spatial variability of ionic concentrations and deposition among sites. Seasonal patterns of most ions showed highest concentrations during the summer months and deposition of H^+ was especially pronounced during this time. Deposition of all ions was generally greater (related to greater precipitation) at three montane forested sample sites compared to a nonforested riverbottom site. Precipitation chemistry at FEF was similar to other sites throughout the eastern United States and contrasted sharply with mid-western and western sites. Eastern sites, including means for FEF sites, were uniformly 3-4 times higher in H^+ and SO_4^{2-} concentration than the mid-western and western sites. Precipitation at FEF was chronically acidic, more so during the growing season, and highest at higher elevations where environmental stresses can be most severe.

IMPLICATIONS

The results presented in this paper have important implications for individuals interested in air quality, atmospheric deposition, and precipitation chemistry, especially as related to forest exposure. Seasonal patterns for concentrations of acidic ions (H^+ , NO_3^- , and SO_4^{2-}) in wet deposition at this rural, montane site indicate that exposures during the growing season are more severe than those during the non-growing season. Also, higher elevation sites tended to have higher levels of acidic deposition. Thus, we suggest that long-term means of annual concentrations/deposition which do not take seasonal changes and elevation effects into account may underestimate the pollutant conditions to which montane hardwood forests of the eastern United States are exposed.

Furthermore, there were occasional large discrepancies between the low-elevation site and the higher-elevation forested sites for precipitation chemistry and acidic deposition. These results suggest that synoptic-scale (network) data may greatly underestimate the pollutant conditions to which high-elevation forest trees are exposed, since network data rarely take elevation into account and typically are based on annual ionic concentration and deposition means that may be considerably lower than those of the growing season.

INTRODUCTION

Acidic deposition continues to be a concern in the forested Appalachian mountain region. Extensive research has been directed at Appalachian spruce-fir forests because of the observed dramatic decline and public concern over this issue.¹ Much has been learned about these montane ecosystems and their chemical environment,² and a major conclusion cites acidic deposition as a significant contributor to declines in red spruce forests.³ Far less research has investigated the impacts of air pollution on the eastern high-elevation hardwood forests, even though throughout most of the Appalachian region (especially south of Maine) hardwood forests are the predominant forest type, with spruce-fir confined largely to extreme higher elevations.⁴

Hardwood forests differ from conifer forests in several ways, most notably in their deciduous nature, with autumn leaf fall and spring canopy development. Hardwood leaves are most susceptible to pollutant damage during rapid springtime growth;^{5,6} therefore, seasonal patterns of precipitation acidity may be more meaningful than annual averages in assessing the potential impact on hardwood forests. Gilliam⁷ has suggested that the coincidence of seasonally high springtime levels of acidity and ozone at upper elevations indicates the possibility of synergistic damage to higher elevation eastern hardwood forests.

West Virginia, typical of much of the central Appalachians, is a predominantly rural state, with approximately 80% of its land area forested.⁸ Of this forested area, about 90% is hardwood forest, much of which is mountainous.⁸ Because this represents a large and economically significant

resource, the need is great for better understanding the pollution conditions to which these forests are exposed. Although most of West Virginia generally is placed within the zone (isocline) of highest H^+ deposition in the United States,² few long-term empirical data exist for precipitation chemistry for this region. Furthermore, concern has been expressed recently over elevated nitrogen (N) inputs to forest ecosystems; i.e., the N saturation phenomenon.⁹ Indeed, recent studies have suggested that some forests of the central Appalachian region are already N saturated.^{10,11} Such conclusions necessitate a better understanding of the patterns and amounts of N deposition for the region.

The purpose of this paper is to describe and document the chemistry of precipitation and annual elemental wetfall inputs at several Appalachian hardwood forest sites from 1982 to 1989. Emphasis is placed on spatial variability of precipitation chemistry and wetfall inputs among sites and seasonal patterns of ion concentrations in wetfall.

MATERIALS AND METHODS

Study Site

Precipitation sampling was carried out at four sites located within 15 km of each other on or near the Fernow Experimental Forest (FEF) in northeastern West Virginia (39°3' N, 79°40' W) (Figure 1); all sites will be referred to collectively as FEF. The automatic collectors at Watershed 9 (WS9), South Haddix (SH), and Watershed 4 (WS4) were located in openings on forested watersheds. The Nursery Bottom (NB) site is located in a nonforested river bottom and was established as a National Acid Deposition Program (NADP) site in 1982. All watersheds had generally southeast-facing aspects (Table 1). Collection periods for each site are given in Table 1.

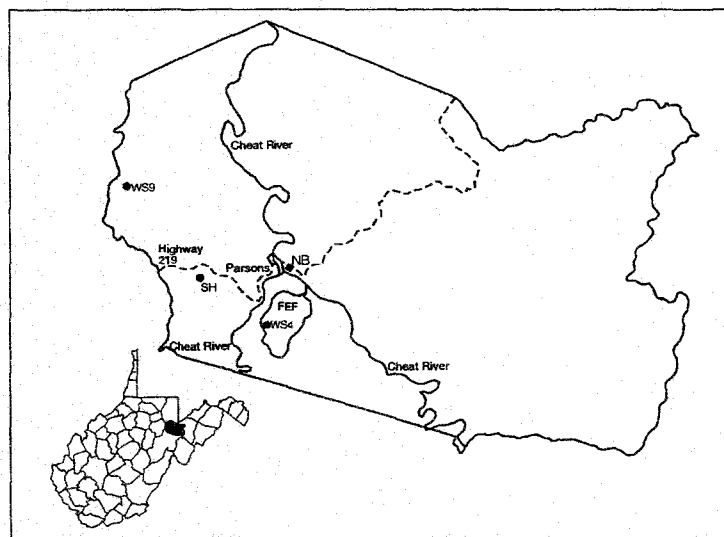


Figure 1. Map of West Virginia showing location of Tucker County (shaded) and detail of Tucker County showing locations of sampling sites. See Table 1 for key to site abbreviations. Shown also are the main drainage (Cheat River), main highway (Highway 219), county seat (Parsons), and location of Fernow Experimental Forest (FEF).

Table 1. Description of precipitation sampling sites on the Fernow Experimental Forest.

Site	Elevation (m)	Aspect	Period of Record for this Study
Nursery Bottom (NB)	505	—	1983-present
Watershed 4 (WS4)	838	SE	1983-present
Clover (WS9)	744	SSE	1983-present
South Haddix (SH)	689	ESE	1983-present

FEF is located in the Allegheny Mountain section of the unglaciated Allegheny Plateau and ranges in elevation from 533 m to 1,112 m, with slopes of 20%-30% common.¹² The climate is typically cool and rainy. Mean annual temperature is about 9 °C and mean precipitation is 1,490 mm; snow is common from December to March. The dominant tree species on FEF are northern hardwood species, including northern red oak (*Quercus rubra* L.), sugar maple (*Acer saccharum* Marsh.), yellow poplar (*Liriodendron tulipifera* L.), and white ash (*Fraxinus americana* L.).¹³

Sampling and Analysis

Precipitation samples were obtained using Aerochem Metric automated wet/dry collectors. Samples were collected weekly and analyzed for pH, Ca^{++} , Mg^{++} , Na^+ , K^+ , NH_4^+ , Cl^- , NO_3^- , and $SO_4^{=}$, according to NADP protocol as described in Stensland et al.¹⁴ Annual (for all months in a year) and monthly (for all weeks in a month) volume-weighted means were calculated for each site and year. Overall mean annual and mean monthly values were calculated arithmetically and compared among sites using analysis of variance and Duncan's multiple range test.¹⁵

RESULTS AND DISCUSSION

Temporal patterns of total annual precipitation (Figure 2A) and seasonal patterns of mean monthly precipitation (Figure 2B) were similar among the four sites. The region-wide drought years of 1987 and 1988 are apparent in Figure 2A. Precipitation generally was greatest during early summer months (Figure 2B), and lowest in January and February. All sites exhibited a large drop in mean monthly precipitation from July to August. Although total annual precipitation did not vary significantly among the three forested sites, WS9 generally received the most precipitation and received significantly more precipitation than the NB site during the non-growing season months

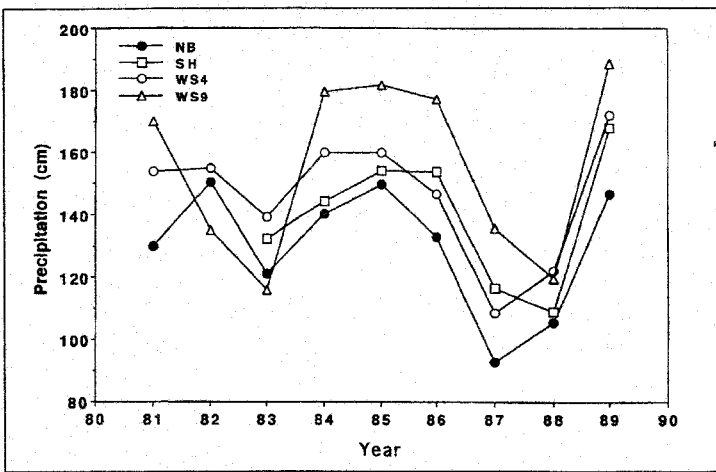


Figure 2A. Total annual precipitation at four sites located on or near FEF, 1982-1989.

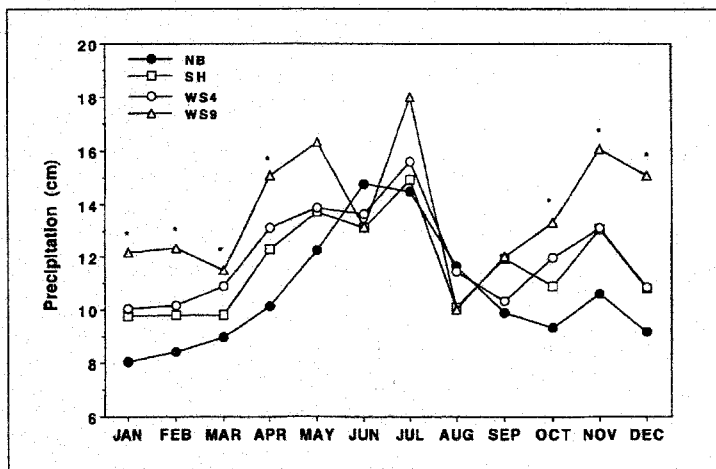


Figure 2B. Mean monthly precipitation at four sites located on or near the FEF, 1982-1989. Significant differences ($p < 0.05$) between WS9 and NB are indicated by asterisks. Lack of an asterisk indicates no significant differences among sites for a given month.

(Figure 2B). For the study period, WS9 received >20% more annual precipitation than NB (Table 2), consistent with other studies that report increased precipitation amounts with higher elevations in eastern U.S. montane forests.²

It is not clear why precipitation was not greatest for WS4 (with highest elevation) among the high-elevation, forested sites. This may be related in part to frontal movements of air masses which produce precipitation. Air masses which originate from the north and west would encounter the site at WS9 before the other sites in this study. Notably, however, northwest air masses would tend to be void of moisture, resulting in minimal precipitation. Indeed, most precipitation in this region arises from moisture originating from the Gulf of Mexico or the Atlantic Ocean. Furthermore, because of the relatively low range of elevations among our sampling sites (<300 m), effects of elevation on amounts and patterns of precipitation are determined more by orientation of local terrain, which may create local rain shadows and rain enhancement zones.

Total cationic equivalents and total anionic equivalents in precipitation were generally within 15% of each other for each site individually and for all sites combined (Table 3). Mean annual wetfall concentrations for most ions were similar between sites at FEF, with no significant differences ($p < 0.05$) for Na^+ , NH_4^+ , H^+ , and Cl^- . The NB site had the lowest concentrations for most ions, including Mg^{++} , K^+ , NO_3^- , and SO_4^{--} (Table 3), which did vary significantly among sites. Concentrations of NO_3^- and SO_4^{--} were greater at the higher elevation forested sites

Table 2. Mean annual precipitation and nutrient input for four sites at Fernow Experimental Forest, 1983-1989. F ratios are the result of analysis of variance for each variable across all sites, along with corresponding probability values ($p > F$). Means followed by the same superscript for a given ion are not significantly different between sites ($\alpha = 0.05$) as determined by Duncan's multiple range test. Site names are as indicated in Table 1.

Site	Ppt (cm)	H ⁺	Ca ⁺⁺	Mg ⁺⁺	K ⁺	Na ⁺	NH ₄ ⁺	NO ₃ ⁻	SO ₄ ⁻⁻	Cl ⁻
					kg/ha/yr					
WS9	164 ^a	1.15 ^a	2.0 ^a	0.2 ^b	0.5 ^b	0.7 ^a	5.3 ^a	6.2 ^a	48.6 ^a	0.8 ^a
WS4	141 ^{ab}	0.90 ^{ab}	2.5 ^a	0.3 ^{ab}	0.8 ^{ab}	0.8 ^a	4.7 ^a	5.0 ^{bc}	38.7 ^{ab}	0.7 ^a
SH	140 ^{ab}	0.94 ^{ab}	2.2 ^a	0.3 ^a	1.4 ^a	0.9 ^a	5.1 ^a	5.2 ^b	41.4 ^{ab}	0.6 ^a
NB	130 ^b	0.85 ^b	2.0 ^a	0.2 ^b	0.7 ^{ab}	0.7 ^a	3.1 ^a	4.1 ^c	32.9 ^b	0.4 ^a
F ratio	2.48	2.48	1.46	3.94	2.50	0.27	1.69	7.35	4.12	0.37
p>F	0.092	0.087	0.252	0.021	0.085	0.844	0.203	0.001	0.018	0.776
Mean	143	0.95	2.2	0.3	0.9	0.8	4.7	5.1	39.9	0.6

Table 3. Volume-weighted mean ion concentrations for four sites at Fernow Experimental Forest, 1983-1989. F ratios are the result of analysis of variance for each variable across all sites, along with corresponding probability values ($p > F$). Means followed by the same superscript for a given ion are not significantly different between sites ($\alpha = 0.05$) as determined by Duncan's multiple range test. Site names are as indicated in Table 1. Also given are ratios of total cations (C^+) to total anions (A^-) for each site and for all sites combined.

Site	pH	H^+	Ca^{++}	Mg^{++}	K^+	Na^+	NH_4^+	NO_3^-	$SO_4^{=}$	Cl^-	C^+/A^-
	----- $\mu eq/L$ -----										
WS9	4.16 ^a	69.0 ^a	6.4 ^b	1.1 ^b	1.1 ^b	1.9 ^a	22.6 ^a	26.5 ^a	61.0 ^a	1.2 ^a	1.15
WS4	4.21 ^a	62.2 ^a	8.9 ^a	1.7 ^a	1.4 ^{ab}	2.5 ^a	20.0 ^a	25.1 ^{ab}	56.6 ^a	1.2 ^a	1.17
SH	4.18 ^a	66.9 ^a	8.2 ^{ab}	2.1 ^a	2.0 ^a	2.8 ^a	22.2 ^a	26.7 ^a	61.5 ^a	1.3 ^a	1.16
NB	4.19 ^a	65.3 ^a	7.0 ^{ab}	1.0 ^b	1.1 ^b	2.1 ^a	16.6 ^a	22.7 ^b	54.3 ^b	0.9 ^a	1.20
F ratio	1.27	1.21	2.54	9.73	3.10	0.99	1.21	2.92	1.91	0.18	
$p > F$	0.307	0.326	0.080	0.0002	0.046	0.415	0.328	0.055	0.156	0.907	
Mean	4.18	65.9	7.6	1.5	1.4	2.3	20.3	25.2	58.3	1.2	1.17

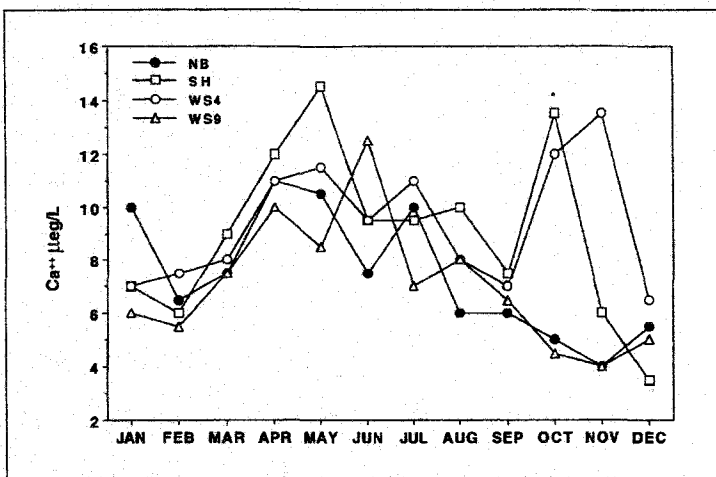


Figure 3A. Mean monthly concentrations of Ca^{++} at four sampling sites at FEF. Significant differences ($p < 0.05$) between SH/WS4 and WS9/NB are indicated by asterisks. Lack of an asterisk indicates no significant differences among sites for a given month.

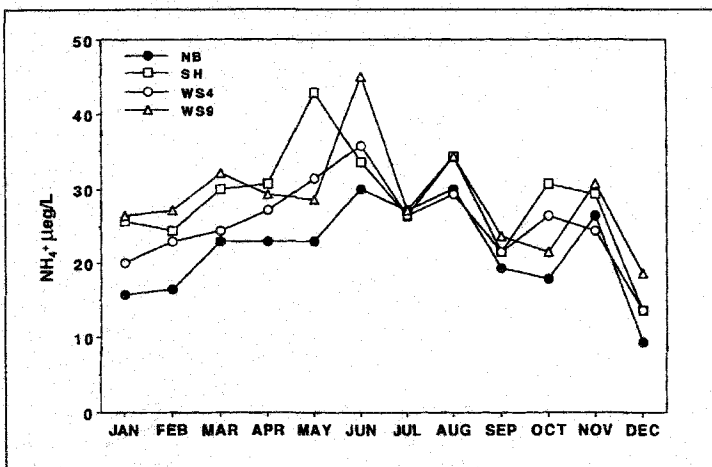


Figure 3B. Mean monthly concentration of NH_4^+ at four sampling sites at FEF. Lack of an asterisk indicates no significant differences among sites for a given month.

than at the NB site; this difference was significant ($p < 0.05$) for WS9 and SH (Table 3). The pattern of greater NO_3^- and $SO_4^{=}$ concentrations at higher elevations at FEF does not agree with theoretical expectations based on higher precipitation amounts at these higher elevation forested sites (i.e., a "dilution effect"). Thus, it is possible that orographic precipitation processes may serve to concentrate, rather than dilute, anthropogenic pollutants at FEF.

Figures 3A-E present monthly concentration means for the dominant ions in wetfall at FEF (Ca^{++} , NH_4^+ , H^+ , NO_3^- , and $SO_4^{=}$). Because of great variability between years of the study period, there were few significant differences between sites for mean monthly concentrations of any ion. Concentrations of Ca^{++} exhibited the least consistent seasonal pattern of all the ions and were significantly higher at SH/WS4 than at NB/WS9 for October (Figure 3A). Concentrations of NH_4^+ also showed great month-to-month variability, but little seasonal pattern, and were not different between sites for any month (Figure 3B). Concentrations of H^+ were remarkably similar between sites for all months. Furthermore, H^+ exhibited a pronounced seasonal pattern, with concentrations increasing throughout the growing season to a maximum in August, followed by a sharp drop (Figure 3C). Concentrations of NO_3^- lacked the seasonal pattern of H^+ , but showed the same autumn decrease; seasonal patterns of NO_3^- were generally similar across sites, except for December, when NO_3^- was significantly higher at WS4/WS9 than at NB for December (Figure 3D). The

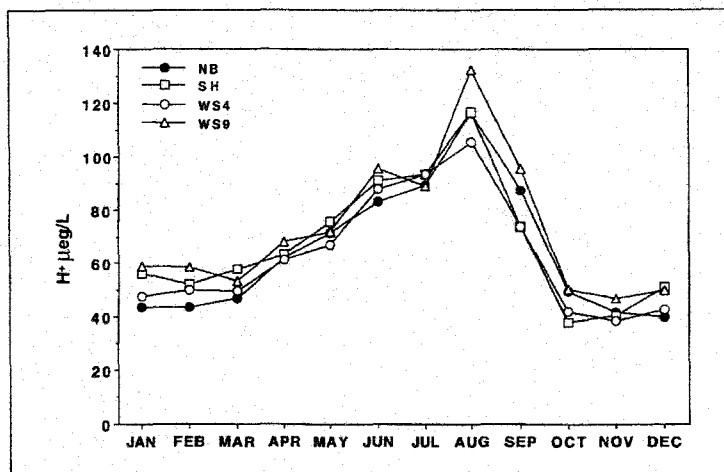


Figure 3C. Mean monthly concentration of H⁺ at four sampling sites at FEF. Lack of an asterisk indicates no significant differences among sites for a given month.

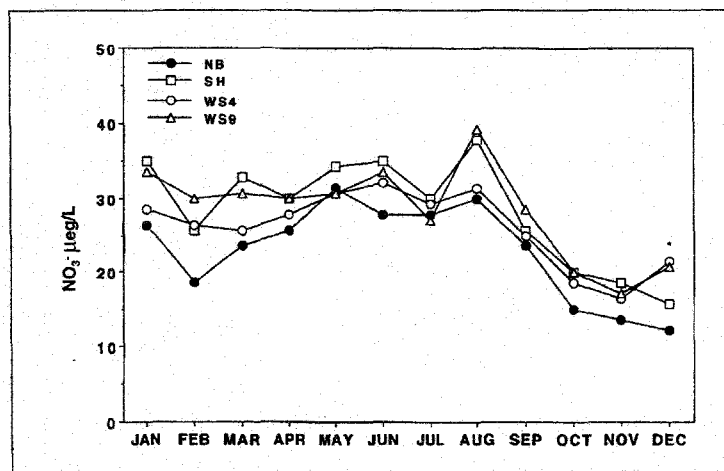


Figure 3D. Mean monthly concentration of NO₃⁻ at four sampling sites at FEF. Significant differences ($p < 0.05$) between WS4/WS9 and NB are indicated by asterisks. Lack of an asterisk indicates no significant differences among sites for a given month.

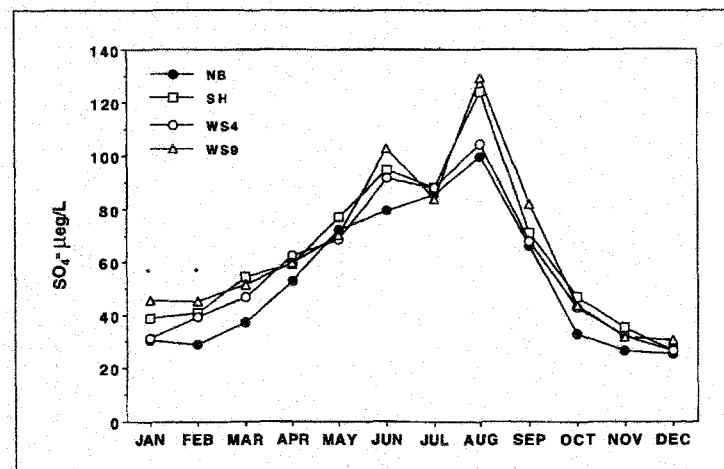


Figure 3E. Mean monthly concentration of SO₄⁼ at four sampling sites at FEF. Significant differences ($p < 0.05$) between WS9 and NB are indicated by asterisks. Lack of an asterisk indicates no significant differences among sites for a given month.

seasonal pattern for SO₄⁼ was nearly identical to that of H⁺, with the exception of January and February, when SO₄⁼ was significantly higher ($p < 0.05$) at WS9 than at NB (Figure 3E).

Although H⁺ concentrations in wetfall at FEF were significantly correlated with both NO₃⁻ and SO₄⁼ concentrations ($r = 0.67$ and 0.88 for H: NO₃⁻ and H: SO₄⁼, respectively, $p < 0.001$), acidity for FEF appears to be most closely associated with SO₄⁼. Both H⁺ and SO₄⁼ had similar magnitudes and seasonal patterns of change in monthly concentrations, whereas NO₃⁻ concentrations were relatively constant through the year (Figure 4).

The pronounced seasonal patterns for H⁺ and SO₄⁼ (Figure 4) may be strictly a result of a meteorological phenomenon; i.e., higher concentrations of ions in wetfall could arise from lower monthly precipitation amounts (a "concentration effect"). However, precipitation increased during the growing season (Figure 2B), not decreased, as this hypothesis would predict. Moody and Samson¹⁶ also found seasonal differences in SO₄⁼ concentrations in precipitation, with warm season (April-September) values 45%-60% higher than cold season (October-March) values for two sites in Kentucky and Indiana. Using back trajectory analysis, they concluded that these seasonal differences resulted from stagnant wind flow conditions during the warm season. Furthermore, Eder¹⁷ analyzed data for SO₄⁼ concentrations in wetfall for much of the eastern United States and found a somewhat consistent seasonality throughout most of the region, a pattern similar to our results (Figure 3E). He found a strong relationship of high SO₄⁼ with major SO_x emissions. Thus, we suggest that the pattern of much higher concentrations of H⁺ and SO₄⁼ (Figure 4) and higher deposition of H⁺ (Figure 5) during summer months may have resulted from anthropogenic pollutant loading into the atmosphere and from higher summertime rates of transformation from SO₂ to SO₄⁼, rather than resulting as an artifact of the concentrating effect of lower precipitation amounts.

Seasonal patterns of dominant ions in precipitation are also likely to be related to seasonal changes in meteorological processes that bring about precipitation events. These systems would tend to be of a stratified nature during the winter and more convective during the summer months. Based on conclusions of Moody and Samson¹⁶ and Mohnen² for other eastern United States sites, we suggest that the

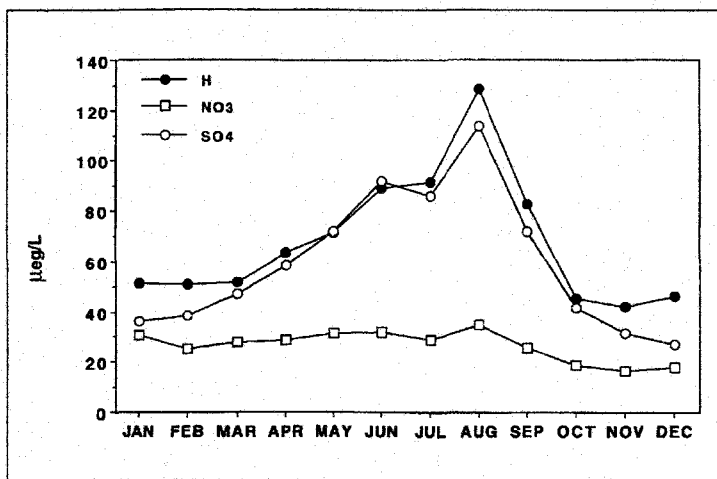


Figure 4. Mean monthly H⁺, NO₃⁻, and SO₄⁻ concentrations at FEF for all sites combined.

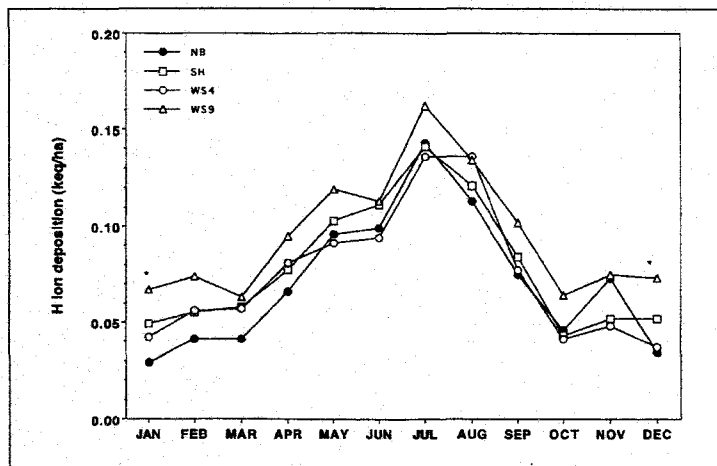


Figure 5. Mean monthly inputs of H⁺ at four sampling sites at FEF. Significant differences ($p < 0.05$) between WS9 and NB are indicated by asterisks. Lack of an asterisk indicates no significant differences among sites for a given month.

convective storms of the summer would produce greater precipitation volumes per event. These "rainout" events are reflective of cloud chemistry (as opposed to below-cloud atmospheric chemistry) and may be enriched in SO₄⁻ and H⁺ as a result of rapid warm-season conversion of SO₂ to SO₄⁻, coupled closely with the higher in-cloud scavenging of SO₄⁻ aerosols, which is typical of rainout processes.^{18,19}

Elevated H⁺ and SO₄⁻ concentrations in precipitation appear to be widespread throughout the eastern United States.⁷ This is supported by comparing concentrations of dominant ions in wetfall of eastern U.S. sites to sites in the mid-western and western United States (Table 4). Wetfall concentrations of NH₄⁺ are variable at both eastern and mid-western/western site types, possibly reflecting regional and local heterogeneity in soil pH, wind-blown soil erosion, and agricultural practices.¹⁸ Concentrations of NO₃⁻ varied greatly among the mid-western and western sites, but were very similar among eastern sites at ~24 meq/L. The greatest differences between

sites were for H⁺ and SO₄⁻. Not only were H⁺ and SO₄⁻ concentrations 3-4 times higher at eastern sites, but these concentrations were consistently high over the range of sites in the eastern U.S. (Table 4). It is further notable that H⁺ and SO₄⁻ concentrations at FEF are among some of the highest in the U.S.

Since ion loading (inputs) in wetfall is a function of both ionic concentrations in precipitation and precipitation amounts, it might be expected that differences in ionic inputs among sites at FEF would closely match those of precipitation (Table 2). Although mean annual inputs of many ions did not vary among sites, all acidic species (H⁺, NO₃⁻, and SO₄⁻) showed significant among-site differences. These all were highest at WS9 and lowest at the low-elevation NB site (Table 2), with 35%, 51%, and 48% greater inputs for H⁺, NO₃⁻, and SO₄⁻, respectively, at WS9 relative to NB. Although they worked at a quite different site (Colorado Rocky Mountain region), Lewis, et al.²⁰ also found greater strong-acid ion (H⁺, NO₃⁻, and SO₄⁻) deposition at higher elevations. They attributed this relationship to the gaseous nature of strong acid precursors (NO_x and SO₂). It is of particular importance to note that our NB site is a National Acidic Deposition Program (NADP) network sampling site and that regional patterns (isoclines) published in summaries of NADP data and other similar networks are based on values from such a site. Our findings suggest that these data may underestimate actual acid deposition conditions for high-elevation forested sites.

At a comparable elevation (~500-1000 m) in Shenandoah National Park, Virginia, Gilliam et al.²¹ found that deciduous forest canopy development can be initiated as late as early May and not be complete until early June. Thus, precipitation acidity at Fernow increases at a time when the canopy leaves are growing most rapidly (Figure 4). This translates to higher monthly H⁺ loading during this time (Figure 5), when the forest is metabolically the most active and when leaves are most susceptible to pollutant damage.⁵ Damage can result from several factors, particularly the incomplete development of a protective waxy cuticle. The long-term effects on forest health and productivity of prolonged exposure to these conditions (e.g., dose-response relationships) remain unresolved.

CONCLUSION

In conclusion, data reported here suggest the potential for problems with acid deposition at this remote deciduous forest site. Not only is precipitation chronically quite acidic, but highest levels of acidity occur during a time when the forest

Table 4. Ionic concentrations ($\mu\text{eq/L}$) in wetfall for various sites in the United States.

Site	NH_4^+	H^+	NO_3^-	SO_4^{2-}
Santa Ynez Mountains, CA ²²	1	20	2	13
Sequoia National Park, CA ²³	13	8	12	13
lodgepole pine forest, WY ²⁴	14	20	20	36
Rocky Mountain region, CO ²⁰	—	6	26	39
Isle Royale National Park, MI ²⁵	9	22	18	15
Konza Prairie, KS ¹⁸	22	15	23	32
Oak Ridge, TN ²⁶	17	70	24	64
Hubbard Brook, NH ²⁷	9	57	24	43
Whiteface Mt., NY ²⁷	14	46	21	43
Shenandoah National Park, VA ⁷	12	46	22	55
Fernow Experimental Forest, WV	20	66	25	58

is most susceptible to damage, beginning with and continuing through the growing season. Furthermore, at a similar high elevation hardwood forest site, Gilliam et al.²¹ found that ozone concentrations increased similarly during this same seasonal period, suggesting the possibility for synergistic acidity/ozone effects on the forest.⁷ Future research is needed to more closely examine what real effects, if any, these levels of acidic deposition and other forms of pollution have on tree growth and productivity in high-elevation hardwood forests.

Our data do not in any way call into question the validity of network precipitation chemistry data, such as those of the NADP. We suggest, however, that conclusions based on such data concerning the chemical environment of eastern United States high-elevation forests be made with caution. Long-term annual concentration and deposition means, which take into account neither higher growing season concentrations and deposition nor elevation effects, may substantially underestimate the pollutant conditions to which these forests are exposed.

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