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Project Summary

An Assessment of Atmospheric Exposure and Deposition to High Elevation Forests in the Eastern United States

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The spruce-fir forests in the higher elevations of the Appalachian Mountains from North Carolina to Maine are showing visible symptoms of injury and increased mortality. Concern has been raised that exposure to and deposition of atmospheric pollutants might play a role in this decline. The Mountain Cloud Chemistry Project (MCCP) sponsored by the U.S. Environmental Protection Agency (EPA) and the National Acid Precipitation Assessment Program (NAPAP) has studied the exposure and deposition of atmospheric constituents to these forests.

Atmospheric pollution is deposited to the forest in a number of forms, cloud water interception represents a major deposition pattern and may exceed deposition by precipitation and gases. The full report provides estimates of cloud, precipitation and dry deposition to the spruce-fir forests at the MCCP sites.

This Project Summary was developed by EPA's Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

This report is the third in a series of annual summaries of research on the exposure and deposition of airborne chemicals to forest canopies and the forest floor in eastern North America. The report is based on observations and model estimates of atmospheric deposition at the high elevation sites in the eastern United States. The report is produced by the scientists in the Mountain Cloud Chemistry Program, a multi-year study of atmospheric chemistry and physics sponsored by the EPA.

The MCCP has three primary objectives: (1) determine the elevational gradients in wet and dry deposition of pollutants and climate variables; (2) determine the relative significance of various deposition mechanisms to the fluxes of chemical species into and through forest canopies; (3) determine the frequency distributions of chemical, physical and climatic exposure.

This report provides estimations of forest exposure to chemicals in air and cloud water and deposition to the forests from precipitation, wind blown clouds and by dry deposition mechanisms. Measurement methodology used provide data for these estimations, data sets and models used for deposition estimates are discussed in detail. Comparisons of deposition are made between southern and northern MCCP sites. Elevation gradients in exposure and deposition are also discussed.

Two models are used to estimate cloud water and chemical deposition flux. One model was developed by Lovett and modified by Mueller. This model is designed for use with spruce-fir forest canopies. The other model was

developed by Krovetz for use with the deciduous canopy at the Shenandoah MCCP site. The model used to estimate dry deposition is the inferential or "big leaf" model. Since this model was originally developed for flat terrain and the model has not been fully characterized for mountainous regions, deposition estimates reported here reflect these uncertainties.

Technical Approach

Resource and logistical considerations dictate that measurements of inputs to high elevation forests in eastern North America can be performed at only a limited number of sites where proper access and facilities are available. In order to meet the needs of the project, five high elevation sites have been selected from 45°N to 35°N to be representative of the geographic and meteorological variability in this large region. This coverage has been augmented by the addition of a low level site (Howland, ME) to allow evaluation of the impact of elevational gradient forest types and enhance geographical coverage. The research/monitoring sites associated with MCCP are Howland Forest, ME, Mt. Moosilauke, NH, Whiteface Mtn., NY, Shenandoah, VA, Whitetop, VA, and Mt. Mitchell, NC.

Site specific measurements of cloud and rain water, of gaseous sulfur and nitrogen compounds, and of ozone and hydrogen peroxide are sampled hourly or are directly converted into hourly concentration values. In the case of filterpack measurements, samples are integrated over a week's time. These concentration values then represent the primary exposure parameters. The concentration of pollutants and the associated meteorological conditions are needed to provide estimates of deposition by precipitation,clouds, and gases.

Results

Deposition of Pollutant Ions in Precipitation

Wet deposition of pollutants was estimated for the 1987/88 growing season at the MCCP sites using standard National Acid Deposition Program/National Trends Network (NADP/NTN) measurements of rainfall amounts and chemistry. The NADP sites selected to represent MCCP sites are Greenville, ME 09, Whiteface, NY 98, Hubbard Brook, NH 02, Big Meadows, VA 29, Whitetop, VA 28 and Clingmans Peak, NC 45. The growing season is longest at the Shenandoah and Howland sites, extending from early April at both locations to mid-November and early October, respectively. Mt. Mitchell and Whitetop Mountain have the next longest growing seasons, followed by Mt. Moosilauke and Whiteface with the shortest (June to early October).

The wet deposition data are shown in Table 1 for SO_4^{2-}, NO_3^- and NH_4^+ ions in precipitation (not cloud). The data indicate that the most westerly sites in the north, Whiteface, NY and Moosilauke, NH, received greater wet deposition via precipitation than did the more northeasterly location in Maine. The southern sites showed slightly higher sulfate and nitrate wet deposition than the northern sites.

Concentration and Deposition of Pollutants in Cloud Water

The 1986-88 results for the five high elevation MCCP sites are summarized in Table 2 as overall chemical composition of cloud water samples from precipitating and non-precipitating clouds. The concentrations in non-precipitating clouds are significantly higher than in precipitating clouds. The differences in cloud water ion concentration between the sites is a result of sample location with regard to cloud base and synoptic weather type. The southern sites frequently are cloudy under the stable, warm sector synoptic type, while the northern sites experience cloudiness associated more frequently with frontal passage. Hence, any north-south differences in cloud water concentrations are likely related to cloud climatology including cloud base height. The importance of sample location in relation to height above cloud base can be demonstrated for Whiteface-1 (1483 m) and Whiteface-2 (1245 m). For simultaneously obtained cloud water samples, Table 3 shows the differences in the mean ion concentration observed for the two vertically separated sites.The substantial vertical gradient in cloud water concentration is mainly explained by increased dilution of precursor substances as liquid water increase with height above cloud base.

In estimating cloud deposition to the forest canopy, MCCP uses an improved version of the 1984 Lovett Cloud Deposition Model (CDM). Cloud impaction events at each MCCP site are classified according to meteorological conditions that prevail during each hour of the event A detailed analysis of cloud chemistry and meteorological variables (wind speed and liquid water content) has demonstrated the usefulness of this technique for uniquely characterizing conditions during events. In addition to synoptic classification, specific air trajectory directions computed within a given event type can further characterize event conditions. This technique is able to explain a major portion of the variance in the chemistry and meteorological data base, thereby allowing more complete growing season estimates of cloud deposition. Deposition estimates for MCCP sites can now be made for periods when data are incomplete as long as cloud frequency and event type can be determined. Event types are (1) pre-warm front, (2) NW sector of cyclone, (3) post-cold front, (4) warm sector of cyclone, (5) stationary front, (6) marine flow off Atlantic, (7) cutoff low in upper atmosphere, and (9) cap cloud. Cloud deposition estimates for each site are made by computing, for each subclass, the mean water deposition flux using the improved CDM and subclass wind speed and liquid water content. Best estimates of canopy structure are then used to calculate the gross (pre-evaporation) cloud water flux to specific forest canopies at each site.

Most of the site-differences in cloud deposition can be explained on the basis of differences in canopy structure. Cloud deposition is found to be highly site specific. Despite an almost 2:1 advantage in cloud frequency, Whiteface Mountain mean deposition estimates for the growing season are generally lower than those for Moosilauke because of lower canopy surface area at the specific Whiteface site. Differences in cloud water deposition between the northern and southern sites are significant and likely caused by differences in such parameters as canopy structure, elevation above cloud base and synoptic meteorology. It is also interesting to note the annual changes in cloud water deposition as a result of changing meteorological conditions from year to year.

Dry Deposition of Gases and Particles

The Atmospheric Turbulence Diffusion Laboratory program (DRY DEPOSITION) used in the MCCP, calculates the deposition velocities of sulfur dioxide, ozone, nitric acid vapor, and sulfate particles from meteorological and site specific biological information. Site information includes: major and minor

Table 1. *Concurrent Growing Season Wet Deposition Via Precipitation*

	kg ion/ha/mo				
Location	NH_4^+	SO_4^{2-}	NO_3^-	Year	Elevation
Greenville, ME(Howland)	0.11	1.28	0.65	1987	322 m
Whiteface, NY	0.33	2.27	0.95	1987	622 m
	1.11	1.18	0.93	1988	
Hubbard Brook, NY	0.28	2.84	1.46	1987	250 m
(Moosilauke)	0.11	2.16	1.10	1988	
Whitetop, VA	0.31	3.20	1.32	1987	1689 m
	0.14	2.84	1.17	1988	
Clingmans Peak	0.46	5.15	2.26	1987	1987 m
(Mt. Mitchell, NC)	0.12	2.69	1.19	1988	
Big Meadows, VA	0.48	2.32	1.43	1987	1074 m
(Shenandoah)	0.92	2.90	1.34	1988	

Table 2. *Average Ion Concentrations in 1986-1988 Cloud Water (μeg/L)*

	H^+	SO_4^{2-}	NO_3^-	NH_4^+	Elevation (m)	Cloud Frequency*
Whiteface-1	171	205	73	97	1483	37%
Whiteface-2	225	352	92	157	1245	
Moosilauke	263	257	132	107	1000	19%
Shenandoah	171	176	94	93	1015	11%
Whitetop	174	321	144	152	1689	30%
Mt. Mitchell	398	489	174	184	1950	29%

*% of hours in cloud.

Table 3. *Mean Ion Concentrations for 108 Simultaneously Collected Cloud Water Samples 1987/88 (μequiv/L)*

	H^+	SO_4^{2-}	NO_3^-	NH_4^+
Whiteface-1	122	79	48	74
Whiteface-2	218	151	85	142

plant species type, leaf area index for plant species, and site location. All available meteorological data including canopy wetness and rainfall are also used.

The growing season dry deposition flux is presented in Table 5 for 1987/88, derived by multiplying weekly deposition velocities (model calculation) by the appropriate weekly averaged concentrations at each site.

As is the case with cloud deposition, the northern sites receive generally less dry deposition than the southern sites due to mainly differences in canopy structure. This is particularly obvious for the Shenandoah site which is the only non-coniferous site within MCCP.

With the exception of ozone, all other pollutants showed very low ambient concentrations. Therefore, only doses for ozone are calculated in MCCP and presented in Table 6. In addition to the MCCP sites, other nearby ozone monitoring stations have been included for the characterization of forest exposure. In order to provide a biologically relevant value, the sum of season dose (ppm/hr) is calculated by summing up all ozone values above 70 ppb occurring during daylight hours (7 am - 6 pm) of the growing season. The exposure data (dose) in Table 6 suggests a significant north-south gradient and an elevational gradient within a region. The data also show a pronounced year to year change in ozone dose for most of the stations with the 1988 exposure higher by about a factor of two. Howland, the most northeasterly site received the lowest ozone dose, while the most southern sites, Mt. Mitchell and Whitetop, experienced highest exposure.

Summary

The average monthly sulfur and acidic nitrogen deposition fluxes determined to date (October 1989) for the MCCP sites and for the 1987-88 growing seasons are summarized in Table 7. From these data it is possible to estimate the importance of cloud deposition to the overall flux of pollutants received by the forest

Table 4. *Calculated Growing Season Cloud Deposition Flux (kg ion/ha)*

		Cloud Water Flux (cm)	SO_4^{2-}	NO_3^-	NH_4^+
Moosilauke	1986	12.9	21	16	3.4
	1987	10.1	15	11	2.4
	1988	3.5	6	5	1.0
Whiteface-2*	1986	11.6	11	3	1.9
	1987	10.5	10	4	1.7
	1988	6.0	6	2	1.2
Whitetop A	1986	53.1	83	41	14.0
	1987	39.4	64	32	10.8
	1988	34.3	56	29	9.5
Whitetop B**	1986	77.3	135	65	22.1
	1987	61.2	108	53	17.7
	1988	54.3	95	48	15.6
Mt. Mitchell	1986	33.5	52	25	6.9
	1987	27.5	51	25	6.9
	1988	21.8	76	18	5.0

**There is no forest on top of Whiteface Summit (WF-1). Therefore, deposition has been calculated on a canopy structure slightly above (WF-2).*
*** Higher canopy density than Whitetop A.*

Table 5. *Monthly Mean Growing Season Dry Deposition (kg species/ha/mo)*

		SO_2	SO_4^{2-}	HNO_3	O_3
Howland	1987	0.22	0.11	0.29	5.48
	1988	0.28	0.15	0.37	5.39
Moosilauke	1987				5.96
	1988		0.31	1.61	6.57
Whiteface	1987	0.30	0.36		6.46
	1988	0.51	0.28		6.09
Shenandoah	1987	3.06	0.80		14.06
	1988				14.83
Whitetop	1988	0.75			11.95
Mt. Mitchell	1987	0.39		1.64	7.7
	1988	1.48			12.7

canopy.Although it must be kept in mind that cloud interception is highly dependent upon canopy structure and location above cloud base, it can be nevertheless concluded, that cloud deposition can deliver to the canopy the same and up to four times the amount of pollutants as precipitation does (see Table 8).At the cloud free Howland site, dry deposition appears to account for less than one third of the total acidic substances deposited. The Shenandoah site has a very low cloud impaction frequency due to its relatively low elevation and dry deposition appears to be of greater relative importance (deciduous trees). The estimates for Whiteface suggest that dry sulfurdeposition accounts for less than a quarter of the total sulfur deposition flux.

Based on these research results, it can be concluded that any assessment of forest damage at high elevations must take all delivery mechanisms into account, in particular, cloud deposition.

Table 6. *Ozone Exposure - Total Season April 15 - October 15 (Daylight Hours 7 AM - 6 PM)*

Site Name/State	Sum of Season Dose (ppm/hr) ≥ 0.07 ppm 1986	1987	1988	Elevation (m)
Howland Forest, ME	N.D.	0.82	4.16	250
Moosilauke, NH	N.D.	7.81	12.51	1000
Whiteface Mountain-1, NY	2.29	9.68	20.85	1483
Whiteface Mountain-3, NY	N.D.	9.41	16.5	1026
Whiteface Mountain-4, NY	N.D.	3.47	N.D.	604
Huntington, NY	5.09	5.74	11.36	500
Hampshire Co., MA	9.14	9.26	34.93	312
Beaver Co., PA	N.D.	13.06	31.70	1300
Shenandoah-1, VA	N.D.	9.49	23.27	1015
Shenandoah-2, VA	N.D.	9.01	39.44	716
Shenandoah-3, VA	N.D.	6.07	20.88	524
Big Meadow, VA	5.56	28.50	31.89	1071
Dickey Ridge, VA	3.21	31.07	40.25	631
Sawmill Run, VA	11.39	26.80	30.16	453
Whitetop, TN	N.D.	38.54	37.68	1689
Giles Co., TN	16.38	16.73	28.91	244
Marion, VA	4.11	9.27	26.92	710
Mt. Mitchell-1, NC	8.34	5.14	45.17	1950
Mt. Mitchell-2, NC	N.D.	6.68	19.49	1750

Table 7. *Estimated 1987-88 Deposition Budgets at MCCP Sites Growing Season Mean Sulfur and Nitrogen Deposition (kg S or N/ha-mo)*

	Wet		Cloud		Dry	
	S	N	S	N	S	N
Howland	0.43	0.15	no clouds		0.17	0.08
Moosilauke	0.84	0.29	0.69	0.35		
Whiteface-2	0.58	0.21	0.65	0.17	0.31	
Shenandoah	0.99	0.28	insufficient data		1.80	0.23
Whitetop	1.01	0.28	3.58	1.23		
Mt. Mitchell	1.31	0.39	2.65	0.87		

Table 8. *Estimated Cloud-to-Wet Depsition Flux Ratios for 1987-88 Growing Season*

	Sulfate	Nitrate
Moosilauke	0.8	1.2
Whiteface-2	1.1	0.8
Whitetop	3.5	4.4
Mt. Mitchell	2.0	2.2

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The complete report, entitled "An Assessment of Atmospheric Exposure and Deposition to High Elevation Forests in the Eastern United States," (Order No. PB 91-100 164/AS; Cost: $31.00 subject to change) will be available only from:
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