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IMPACT OF ACID FOG AND OZONE ON COASTAL RED SPRUCE

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ABSTRACT. High levels of ozone and acidic fog have been measured along the coast of Maine, U.S.A. Some of the ozone and nitrate are a consequence of direct transport from East Coast metropolitan centers, but we discuss evidence supporting N_2O_5, and perhaps ozone, generation at forested sites from NO_x precursors. Fogs with pH below 3.3 occur with greatest frequency in the mid-coast region. Ozone is consistently highest along the southwest coast, but peak hourly averages are often highest in the mid-coast. Historical evidence suggests an increase in fog acidity and nitrate levels, but no change in sulphate within the past 48 years. Red spruce with decline symptoms can be observed from the south-coast to mid-coast region, with the greatest symptom development in the mid-coast. Trees in the mid-coast on soils with high base saturation and Ca/Al ratios develop symptoms of late winter injury (reddening and casting of current year needles in late winter and spring). Trees on poorer soils develop chlorosis on upper surfaces of older needles. Nutrient analysis of soils and foliage reveals differences between stressed and unstressed sites. Scanning electron microscopy of needle surfaces also reveals response differences between pollutant stressed and unstressed trees. Hypotheses and research directions are discussed.

1. Background

Red spruce (*Picea rubens* Sarg.) on the coast of Maine are naturally exposed to varying quantities of ozone and acidic fog. Trees at certain sites are exhibiting symptoms characteristic of forest decline (Jagels, 1986). On some sites the symptoms of needle chlorosis and premature needle loss resemble those found in mid- to upper-elevation Norway spruce forests in Germany (Prinz, 1987), while in nearby stands, the symptoms are reddening of current year needles in late winter, and these mimic those found at high elevations in the Adirondack Mountains (Friedland *et al.*, 1984).

Water, Air, and Soil Pollution **48**: 193–208, 1989.

Our research has been addressing five major questions: (1) What is the pollutant distribution along the coast of Maine? (2) Does the coastal forest contribute to observed pollutant levels? (3) Has fog acidity and chemistry changed historically? (4) Do the observed symptoms represent decline in red spruce and do they reflect pollutant concentrations? (5) Can an integrative hypothesis be developed to explain the variation of symptoms and pollutant levels?

2. Methodology

For back-trajectory analysis, regional weather maps were constructed every 6 hours using data from 20 surface stations, 7 buoys, 13 Coast.Guard stations and 4 automated lighthouses. At three of the weather stations (Chatham, Massachusetts, Portland, Maine, and Sherbourne, Nova Scotia) twice daily radiosonde releases permitted monitoring of low-level, temperature inversion wind-shear normally present in Gulf of Maine fogs. Portions of trajectories beyond the continental shelf were dependent on standard NOAA 6-hour surface maps.

Trajectories were based on an averaging of forward and backward extrapolations. Flow velocity was computed as 80% of geostrophic wind, and flow direction, based on the average drift of the stable marine boundary layer, was considered to be -20° from the geostrophic wind. The model is based on the assumption that the back trajectory of the entire marine layer is the same as that of the surface fog. Under most fog conditions, strong wind shear assures mixing in the marine layer over time, despite the large thermal stability of the layer. Under conditions of local source pollution or low wind shear, the accuracy of the model decreases (Stull, 1988).

Fog collection protocols and procedures for pH determination and chemical analysis are described in Kimball *et al.*, 1988.

Spruce needles used for microscopy or chemical analysis were collected from the upper one-fourth canopy from the SW quadrant. For each procedure, three branches from each of ten trees were collected per site. For scanning electron microscopy, individual needles were immediately mounted on stubs in the field. For the other analyses, needles were refrigerated and returned to the laboratory within a few hours.

Fresh needles collected for terpene analysis were pulverized under liquid nitrogen, and the methylene chloride extracted, steam distillate was analyzed by capillary gas chromatography using an HP5890 gas chromatograph. A separate paper on this analysis is in preparation.

Needles mounted on SEM stubs were dried in a desiccator and sputter coated with gold (16 pulses of 25A° thickness gold, with 30 second cooling intervals to prevent wax degradation). Specimens were examined and photographed in an AMR-1000 SEM.

Needles analyzed for total chlorophyll were rinsed in distilled water, air dried and DMSO extracted (Hiscox and Isrealstam, 1979). Extracts were analyzed on a Spectronic 2000 spectrophotometer and total chlorophyll calculated according to the method of Lichtenthaler and Wellburn, 1983.

Concentrations of Ca, K, Mg, P, Al and Na in foliage was determined by inductively-coupled plasma emission spectrometry from needles which had been dry-ashed at 450°C for 8 hours and digested with 1:1 HCl and 70% HNO_3. Nitrogen was determined by micro-Kjeldahl block digestion with H_2SO_4 and H_2O_2 at 400°C.

Soils samples from 6 random cores per site were analyzed for the 01 and 02 horizons. Soil pH was determined from a 4:1 H_2O to soil slurry. Concentrations of Ca, Mg, K, Na were determined by vacuum extraction in 1N NH_4Cl. Hydrogen and Al were determined by titration against 1N KCl; and P was determined by extraction in ammonium acetate at pH3 (Black, 1965; Carpenter, 1953).

For an experiment testing the effect of delayed application or lack of fertilizer, three-year-old red spruce trees which had been grown from seed collected from a single seed source in Nova Scotia were repotted to larger (8") pots in late winter of 1987 and placed on gravel pads under 50% shade cloth or lattice. Trees were fertilized with 10 grams of 3-4 month controlled release 17-6-12 fertilizer with micronutrients (Sierra Chemicals).

Statistical mean separation for leaf and soil analyses are based on the Duncan multiple range test (Miller, 1981). Significance was established at $p \leq 0.05$.

3. Pollutant Distribution

A previous survey of fog and cloudwater collected from ten sites in North America from Alaska to Puerto Rico revealed that fog at a mid-coast site in Maine had the lowest pH with the highest nitrate and sulphate concentrations (Weathers *et al.*, 1988). Our studies of seven coastal sites from New Hampshire to New Brunswick confirm that spruce forests on the coast of Maine are intercepting fogs which are more acidic than those reported for high elevation sites in Maine, and eastern North America, and that the nitrate levels of Maine coast fogs are three to four times more concentrated than at high elevation sites (Kimball *et al.*, 1988; Castillo *et al.*, 1983; Schemenauer, 1986).

On the coast of Maine during the months of June through September, surface wind movement is from the south quadrant over 50% of the time. Pollutants emitted into the stable marine boundary layer spread horizontally (Gossard *et al.* 1985). Ozone and nitrogen oxide urban plumes have been tracked from Boston and New York to coastal Maine, where they retain ozone levels of 130-154 ppb (Spicer *et al.*, 1982; Spicer *et al.*, 1979). Nitrogen oxides can also be transported in these plumes, and are favored over water because of low deposition rates (Spicer, 1982; Hicks, 1984)). Retention of NO_x is enhanced by conversion with H_2O to HONO aerosol (haze, smog, fog) (Singh, 1987), or by production of peroxyacyl nitrates (PAN's). Both of these mechanisms can serve as aerosol reservoirs for NO_x during multiday atmospheric transport (Svensson *et al.*, 1987; Altshuller, 1986). During high daytime concentrations of O_3, N_2O_5 is also generated, which is removed as nitric acid with fog formation; NO_3 cannot regenerate ozone with fog dissipation (Gupta *et al.*, 1987; Singh, 1987).

Back trajectory analyses from our fog collection sites provide confirmatory evidence that fog pH is determined to a large degree by the air mass source region. For example, a calculated back trajectory for Kent Island, New Brunswick (Figure 1, site G) for June 30, 1988 (extending to June 28) showed an air mass path which originated from the tropical tradewind region. This relatively clean air produced a fog pH of 5.6. In contrast, back trajectory of an air mass for July 27 (extending to July 25) traced a path from the Washington, D.C. area which moved in a northerly arc over the Atlantic Ocean, reaching Kent Island within two days. Fog generated in this air mass had a pH of 3.5, recording the passage of more polluted air.

Satellite images using visible and infra-red bands (NOAA-GOES) have been used in our study to distinguish fog banks from clouds. Typically these images indicate a clear "hole" in the northeast coast fog bank over the southwest coast of Maine (perhaps because air masses crossing Cape Cod and south Boston are warmed sufficiently to dissipate the fog). Reports from Maine Coast Guard stations confirm the greater incidence of fog from the mid-coast to the Bay of Fundy.

Since 1985 we have monitored fog and rain acidity and chemistry on the coast of Maine using active fog collectors (Kimball *et al.*, 1988). Rain pH and chemistry are similar between sites and typical for New England; the following discussion concentrates on fog. Table I summarizes the pH and chemistry of fogs collected in 1987 (see Figure 1 for map locations of sites). These data generally show a declining gradient in acidity, nitrate and sulphate from SW to NE along the coast. Fog pH and chemistry for previous collection years (Kimball *et al.*, 1988) show the same pattern with the exception of sulfate values for Cape Elizabeth and Damariscove Island which were unusually high in 1987. The gradient is consistent with the hypothesis that as SO_2, ozone, NO_x and anthropogenic hydrocarbons from metropolitan east coast centers are injected into air masses in southern Maine, aerosol SO_4 and NO_3 provide nuclei for fog droplet growth, particularly in marine aerosols (Meinert and Winchester, 1977; Hitchcock *et al.*, 1980; Martens *et al.*, 1973; Clegg and Brimblecome, 1988).

As air masses travel northeast over coastal waters, the acidity and ionic concentration of fog droplets is reduced by dilution through droplet growth (as fog passes over colder water) and gravitational separation by conversion to drizzle.

Table I also shows that fog collections increase from SW to NE with a particularly sharp increase at mid-coast. This is consistent with the typical fog bank pattern discussed previously.

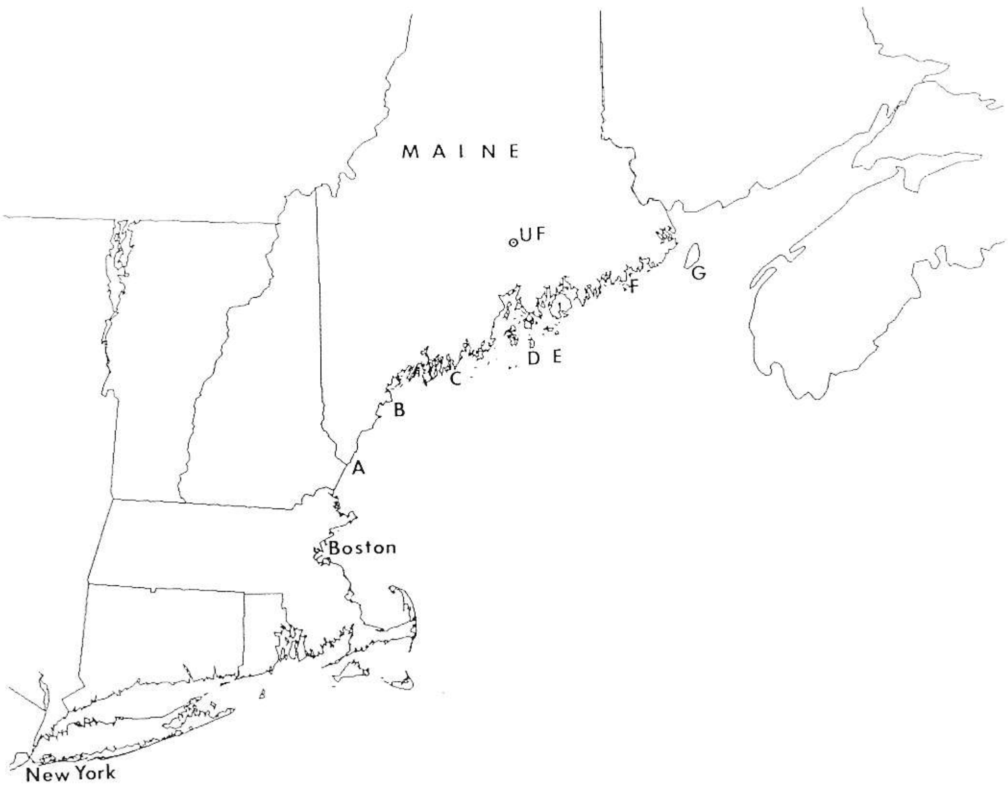

Fig. 1. Map of research area showing location of fog monitoring sites (A,B,C,D,E,F,G) and University Forest site (UF).

Table I. Fog pH and Chemistry - 1987

Site		Map Location	No. of Samples	Mean pH (range); derived from H+ concentration	Mean NO_3 (μeq/ℓ)	Mean SO_4 marine corrected (μeq/ℓ)
Coastal						
SW	Appledore Island	(A)	4	3.45 (3.09-6.49)	305	467
↓	Cape Elizabeth	(B)	3	2.84 (2.56-3.62)	907	1200
	Damariscove Island	(C)	5	2.73 (2.38-3.69)	703	1127
	Isle Au Haut	(D)	11	3.24 (2.70-3.97)	467	251
	Roque Island	(F)	5	3.50 (3.34-3.73)	150	323
NE	Kent Island	(G)	15	3.54 (3.02-5.76)	129	203
Montane						
	Sugarloaf		19	3.52 (2.93-5.80)	150	236
Historical						
1939, Brooklin, ME*			11	4.7 (3.5-6.3)	---	316

*see Houghton, 1955

Ozone has been monitored by the Department of Environmental Protection, state of Maine, since 1979. Coastal ozone levels exceed those found at any inland sites in Maine (Emery, 1987). In general a gradient of decreasing ozone concentration can be found from southwest to northeast along the coast. In cooperation with the State, we have added additional ozone monitors at some of our fog collecting sites in 1987 and 1988. Some of the 1988 data is discussed in the next section.

4. Contribution of Forest to Pollution Levels

Fog pH and nitrate chemistry on Appledore Island (site A in Figure 1, and Table I) are inconsistent with the simple gradient hypothesis, but we initially considered these data to be an aberration (particularly since we have so few fog collections from Appledore). But we also have observed another anomaly to the simple gradient hypothesis. Ozone levels are often as high or higher at mid-coast sites than they are at sites closer to Boston. For instance, during the summer of 1988, we measured an hourly average ozone level of 200 ppb at Isle Au Haut in mid-June. Other south coast sites exceeded the federal standard during the period but were lower than Isle Au Haut (150-180 ppb for peak hourly averages). These anomalies suggest that perhaps some of the ozone and nitric acid are being generated from NO_x precursors reacting with biogenic hydrocarbons (terpenes) at forested sites (Abelson, 1988; Chameides *et al.*, 1988). This might explain why treeless Appledore Island has lower fog nitrate levels than Cape Elizabeth, Damariscove or Isle Au Haut (all of which are forested).

To examine these anomalies further we re-assessed fog data collected in 1985. During that summer we collected fog at an additional island site, Mt. Desert Rock (site E, Figure 1). The collector was placed at the same elevation and aspect as for Isle Au Haut. Table II compares the pH and chemistry of these two mid-coast islands for 1985. The simple gradient hypothesis would suggest similar anthropogenic hydrocarbon concentrations, fog acidities and nitrate levels at both sites. The nearly fourfold higher nitrate level at Isle Au Haut cannot be explained by fog droplet size differences since mean temperature during fog collections was similar (actually slightly warmer at Mt. Desert Rock). The most notable difference between these two sites is the presence or absence of trees.

Table II. Evidence for Contribution of Biogenic Hydrocarbons to N_2O_5 Formation.

	TWO COASTAL MAINE ISLANDS	
	Isle Au Haut	**Mt. Desert Rock**
Location	mid coast near other islands	40Km ESE of IAH 22Km from any land mass
Approximate size	2,400 hectares	1.6 hectares
Vegetation	spruce & some hardwoods	a few patches of herbaceous vegetation
Mid-range summer air temperatures	18°C	16°C
Mean temperature during fog collection	14°C	16°C
	n=7	n=11
Mean fog pH and (range)	3.21 (3.75-2.90)	3.79 (5.14-3.28)
Mean Nitrate in fog and (range)	335 $\mu eq \ell^{-1}$ (525-144)	86 $\mu eq \ell^{-1}$ (201-26)

We have recently analyzed red spruce needles from coastal sites and have isolated and identified 21 different mono- and sesquiterpenes. Total terpene concentrations can exceed 5 mg/g fresh weight,suggesting that spruce needles should be a source of volatile cyclic olefins. Monoterpene analysis of Norway spruce needles yields similar results (Saint-Guily, 1988).

Cyclic olefins (terpenes) are considerably more efficient than the anthropogenic straight chain hydrocarbons in converting NO to NO_2, which, in turn, increases oxidant levels (Stevens and Scott, 1962; Abelson, 1988). Experiments by Franzen *et al.* (1988) provide indirect evidence of possible monoterpene-NO_x reactions leading to "montane yellowing" in Norway spruce. The NO_x for the reactions at rural forested sites is likely anthropogenic in origin in view of the low emissions from natural sources (Robinson and Homolya, 1983). Transport mechanisms probably involve aerosol HONO and PAN's which would be enhanced in fogs, as previously noted.

Summarizing for coastal pollutants, the highest fog acidities are found in the south coast of Maine, but the mid-coast receives the greatest loading of fog with minimum pH levels consistently below 3.3. Moderately high ozone levels are found consistently along the south coast but very high peaks (up to 200 ppb) are observed in the mid-coast region.

5. Historical Reconstruction of Fog Acidity and Chemistry

Houghton (1955) and his co-workers collected fog during the late 1930's on nickel or stainless steel screens and analyzed the collected water for pH and sulphate. We have determined by laboratory experiments with one of the nickel screens used in that study, and from side-by-side comparisons with teflon and stainless collectors in the field that the nickel screen contaminates the fog water with nickel oxide, and as a result, yields artificially high pH values during fog collection. Side-by-side stainless and teflon collectors yield comparable pH values (chemical analysis has not yet been completed). The last line in Table I contains the pH and sulphate data from fog collected on the only stainless steel screen used along the coast in 1939 (Houghton, 1955). Brooklin, Maine is a forested site in the same mid-coast area as Isle Au Haut. Comparison of contemporary fog from Isle Au Haut with 1939 fog from Brooklin reveal roughly comparable sulphate levels but more than a ten-fold increase in acidity in 48 years. If these are valid comparisons, one would conclude that the change in pH over time was a consequence of increasing nitrate levels which would parallel increased NO_x emissions.

6. Foliar Symptoms in Coastal Red Spruce

The major symptom observed on coastal red spruce is upper surface chlorosis of needles older than current year. The symptoms in these trees (described more thoroughly in Jagels, 1986) mimic the European *neuartige Waldschaden*, type I (Hauhs and Wright, 1986). Trees displaying needle chlorosis and associated symptoms are generally found in more open, rough canopy stands on the poorer soils in the south to mid-coast region. Symptoms reach a peak in the mid-coast region and disappear to the northeast.

On better soils in the mid-coast region, trees display a different set of symptoms. Current needles turn red-brown in late winter and are shed by July, similar to type II decline in Europe (Hauhs and Wright, 1986) except that the symptoms are confined to current growth. Trees are quite variable in response, some displaying no symptoms, others displaying the symptoms for one year but not the next, and some displaying the symptoms every year for the three years of observation. Significant twig death occurs following multiple-year needle loss, and a few trees appear to be approaching senescence. These symptoms mimic those reported for high elevation red spruce in the Northeastern U.S.A. (Friedland *et al.*, 1985; Evans, 1986).

Two sites on the south end of Isle Au Haut, separated by a narrow harbor, have been studied intensively for three years, and these, in turn, have been compared to a less polluted island site near the New Brunswick border (Roque Island, site F, Figure 1; see Table I for fog chemistry), and to a reference, University Forest site, at Orono, Maine (Figure 1). The latter is outside the coastal fog/ozone belt. Soil characteristics for the four sites are summarized in Table III. All sites have relatively thin organic soils over rock or hardpan. The sites are nearly indistinguishable based on soil pH but percent base saturation and Ca/Al ratio are significantly higher at the Eastern Head site. This mid-coast site contains trees without type I decline symptoms but with type II symptoms, while the nearby Head Harbor site contains trees with type I symptoms but not type II. Roque Island and University Forest trees are basically asymptomatic.

Total chlorophyll content (pooled for three collection periods; summer, fall, spring) is significantly lower in trees at Head Harbor with type I symptoms ($p \leq 0.05$). Separate analyses of chlorophyll a and b give similar pooled seasonal results. However, on an individual season basis, chlorophyll a is significantly lower at Head Harbor only in spring, while chlorophyll b is lower only in the summer. In the fall (October), total chlorophyll and chlorophyll a and b are not statistically different among the sites.

Elemental analysis of soils at the four sites (Table III) reveals low levels of calcium in soils at Head Harbor and University Forest and low levels of phosphorous at Head Harbor, Roque Island and University Forest. Foliar analysis (Table IV) of current and 1-year-old needles reveals that non-metabolized elements (Al and Na) and the major cell wall element (Ca) show an accumulative increase with needle age, while the mobile elements of K, Mg and P show a distinct decrease with needle age. On the poorer soils (University Forest and Head Harbor) apparent transfer of mobile elements from older to younger needles and conversely the increase in aluminum from younger to older needles is enhanced.

Table III. Chemistry of Forest Floor -- Experimental Red Spruce Sites*

	Sites and Horizons							
	Head Harbor		**Eastern Head**		**Roque Island**		**Univ. Forest**	
	01	02	01	02	01	02	01	02
Exchangeable ions (meq/100g)								
Ca	5.15	4.26	13.84	12.02	11.69	5.85	8.08	2.39
Mg	6.65	7.69	7.75	9.27	4.89	2.0	2.34	1.04
K	1.21	0.71	1.15	.87	0.92	0.33	1.38	0.55
P	3.16	2.45	7.70	6.67	4.65	2.91	5.46	2.72
Al	6.86	14.28	2.39	3.29	9.78	20.16	11.57	22.6
Na	2.07	2.08	3.54	4.07	0.77	0.61	0.29	0.27
Sum Base Cations	15.1	14.2	26.3	26.2	18.6	8.78	12.1	4.25
Cation Exchange Capacity	30.9	41.2	34.8	39.2	36.5	33.21	30.8	34.7
Percent Base Saturation	53.9	37.6	75.0	65.4	52.9	26.3	39.7	12.7
Ca/Al ratio	0.45		4.53		0.30		0.60	
Soil pH	4.15		4.05		4.10		3.80	
Soil Depth (cm)	22		14		28		14	

*Values presented are means of 6 soil cores.

Table IV. Foliar Chemistry of Current and One Year Needles* (µg/g dry tissue; except N = %)

	Head Harbor, IAH			Eastern Head, IAH			Roque Island			Univeristy Forest		
	current	1y old	% change**	current	1y old	% change**	current	1y old	% change**	current	1y old	% change**
Ca	1866	2653	42↑	2516	3973	58↑	2454	3240	32↑	2534	4123	63↑
K	5071	4119	19↓	4819	4818	--	5357	4846	10↓	5876	4616	21↓
Mg	985	823	16↓	947	879	7↓	1032	921	11↓	1140	896	21↓
P	951	722	21↓	1490	1321	11↓	1456	1263	13↓	1480	1158	22↓
Al	55	92	67↑	35	51	46↑	53	75	41↑	47	77	64↑
Na	162	278	72↑	137	184	34↑	58	72	24↑	73	118	62↑↓
N	1.00	0.95	5↓	1.20	1.07	11↓	1.13	1.01	11↓	1.16	1.04	10↓

*Years 1986 and 1987, collected in October, combined for each needle age class. Values presented are means (n=20 for each site).
** ↑ = increase; ↓ = decrease.

On the best soil (Eastern Head) these differences are minimized. These effects seem to be essentially soil regulated. However, the absolute amounts of foliar elements cannot be predicted simply from soil comparisons. In particular Head Harbor has significantly lower Ca (current year needles), and P (current and 1st-year needles) than University Forest ($p \leq 0.05$), while Eastern Head, which would be expected to have the highest levels, has approximately the same levels as University Forest. These observations suggest that some other mechanism such as pollutant induced foliar leaching is resulting in foliar nutrient levels which are lower than one would predict from soil comparisons.

The role of sodium in the expression of foliar symptoms in coastal red spruce cannot be overlooked. Typically, excessive salt spray, following events such as hurricanes, results in tip burn similar to the type II symptoms described. In this study, however, Na levels in needles are highest at Head Harbor where no tip burn is observed, and somewhat lower at Eastern Head where winter tip burn is seen (Table IV). These observations are inconsistent with a direct salt-spray injury phenomenon. However, soil analysis (Table III) shows the highest Na levels at Eastern Head, thus we cannot rule out a possible role of sodium in symptom development. The potential physiological role of Na is further complicated by the presence of sodium chloride crystals trapped in the leaf cuticle (Figure 2), presumably from salt spray. This sodium would not be part of internal leaf metabolism.

Examination of epicuticular waxes on needles with SEM reveals a pattern of destruction similar to but somewhat different from that reported for declining trees of Norway spruce (Sauter and Voss, 1986). Current year needles for all trees studied contain a network of tubular wax crystalloids in epistomatal chambers (Figure 3). In some current year needles from Head Harbor trees and in 1- and 2-year-old needles on trees from the other sites, a thin wax covering with pores develops over the tubular network (Figure 4). In declining trees from Head Harbor, the tubular wax network breaks down in 1- or 2-year-old needles and fuses into irregular block-like segments which results in large (up to 10μ) fissures into the epistomatal cavity (Figure 5). To what degree this may enhance pollutant uptake or nutrient leaching is open to speculation.

A preliminary trial experiment was conducted with 3-year-old potted spruce to test whether field symptoms of declining spruce could be simulated either by soil nutrient withdrawal or by delayed application of nutrients, in the absence of acid fog or high ozone levels. The trees were all grown from seed collected from a single stand in Nova Scotia. One group of trees (n=38) received no added fertilizer in 1987, one group (n=240) received a 3-4 month controlled release 17-6-12 fertilizer with micronutrients (Sierra Chemicals) applied in late April and one group (n-376) received the same fertilizer applied in mid-July. Trees which received the April fertilization showed good growth, green foliage and no winter damage the following year. The trees which received the July fertilization also showed good growth and green needles, but 51% displayed late winter injury (type II symptoms) the following spring. All needles on the non-fertilized trees turned yellow-green (uniform on ad-and abaxial leaf surfaces) by late summer, had very limited growth, but showed no winter injury the following year. Treatments will be repeated and followed through subsequent seasons.

7. Hypotheses and Research Directions

Prinz (1987) has presented a hypothesis for the probable causes of *neuartige Waldschaden* in Germany, for which needle chlorosis is the major symptom. He cites the direct impacts of ozone and acid fog or rain as being the inciting factors leading to metabolic changes, membrane weakening and enhanced leaching of nutrients, in particular, magnesium.

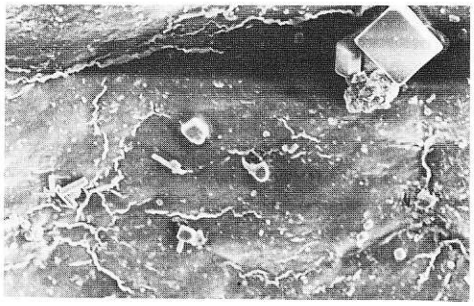

Fig. 2. Salt crystals, some partially embedded in cuticular wax, on the surface of a red spruce needle.

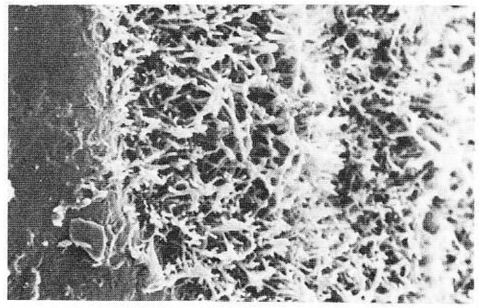

Fig. 3. Tubular, crystalloid wax in epistomatal chamber of a current year red spruce needle.

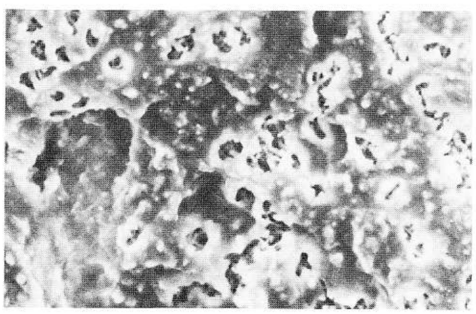

Fig. 4. Amorphous wax layer, with pores overlaying tubular crystalloids epistomatal chamber 3-year-old needle of red spruce from University Forest.

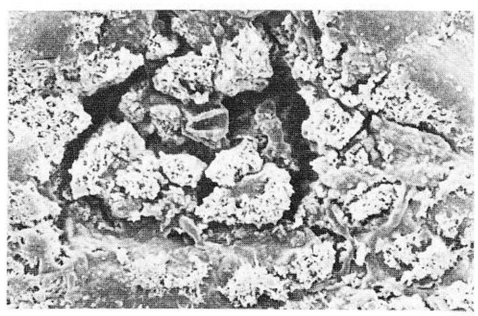

Fig. 5. Epistomatal chamber of 3-year-old needle of red in spruce from Head Harbor showing destruction of wax structure resulting in large openings into chamber.

Data from our studies would, in general, support this hypothesis, with some modifications and suggestions for new research approaches. Specifically:

1) Preliminary results from our field sites and potted tree experiments suggests that reduced levels of soil nutrients without pollutant stress produces either no symptoms (University Forest site) or with potted trees not fertilized leads to symptoms (uniform yellowing in all needles and on all needle surfaces) which are different from those observed on pollutant stressed trees. Foliar leaching seems to be essential for *Waldschaden* symptom development.

2) Under similar pollutant loading, symptom development may be quite different on different soils and/or canopy structure. Where base saturation and Ca/Al ratio in soils are low and canopy structure is rough and open, type I symptoms appear to be favored and this seems to preclude the development of type II symptoms. Where soil base saturation and Ca/Al ratio are high and canopy closure is more complete, type I symptoms are absent and type II develop. This differs, somewhat, from observations by Hauhs and Wright (1986). Experiments with potted trees suggest that fertilization applied late in the growing season may initiate type II symptoms in the absence of pollutants.

3) Since sulphate pollution has probably not increased as much as nitrate during the past four decades on the coast of Maine, high ozone and/or fog acidity, enhanced considerably by nitrate, are likely pollutant stresses associated with the observed red spruce canopy alterations. Nitrate in fog may be an indicator that HONO and/or PAN's are present as aerosols. During fog dissipation in a coniferous forest, these could be released to react with terpenes and generate more ozone and peroxy radicals (which could also be deleterious to spruce foliage). Chemical analyses of polluted fogs should include nitrous as well as nitric acid quantification. Chamber experiments should be designed to expose trees to mixtures of O_3, NO_2, and NO in atmospheres of different relative humidities. Mixed gas exposures have been shown to enhance visible symptoms in peas (Mehlhorn and Wellburn, 1987). Such experiments might help to resolve the anomaly of "no response" to pure ozone exposures in some chamber experiments. Open-top chambers would probably not permit sufficient reaction times for these kinds of experiments.

4) Consistent with other research findings (Mehlhorn and Wellburn, 1987) intermittent, elevated levels of ozone in the mid-coast region are correlated with greater symptom development than the more consistent, moderately high ozone levels of the south coast. Greater incidence of acid fog in the mid-coast is likely also enhancing symptom development. Sodium levels in soil may also be a predisposing factor. Chamber experiments should be designed to investigate the potential response differential in red spruce to intermittent versus consistent ozone exposures with and without acidified aerosol injection.

Acknowledgements

The authors wish to thank Dr. Barbara Cole and Dr. Michael Bentley for providing chemical analysis of terpenes in red spruce needles, Dr. Christopher Cronan for analysis of soil samples and T. Perkins and J. Emery, Air Bureau, Maine Dept. of Environmental Protection for assistance with ozone monitoring. This research was supported by funding from the Andrew W. Mellon Foundation and a U.S.D.A. Competitive Grant through C.S.R.S. This is Maine Agricultural Experiment Station Report No. 1386.

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(Received May 30, 1989; revised July 25, 1989)