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Monitoring Air Quality in Class I Wilderness Areas of the Northeastern United States Using Lichens and Bryophytes

Alison C. Dibble, James W. Hinds, Ralph Perron, Natalie Cleavitt, Richard L. Poirot, and Linda H. Pardo

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Abstract

To address a need for air quality and lichen monitoring information for the Northeast, we compared bulk chemistry data from 2011-2013 to baseline surveys from 1988 and 1993 in three Class I Wilderness areas of New Hampshire and Vermont. Plots were within the White Mountain National Forest (Presidential Range—Dry River Wilderness and Great Gulf Wilderness, New Hampshire) and the Green Mountain National Forest (Lye Brook Wilderness, Vermont). We sampled epiphyte communities and found 58 macrolichen species and 55 bryophyte species. We also analyzed bulk samples for total N, total S, and 27 additional elements. We detected a decrease in Pb at the level of the National Forest and in a subset of plots. Low lichen richness and poor thallus condition at Lye Brook corresponded to higher N and S levels at these sites. Lichen thallus condition was best where lichen species richness was also high. Highest Hg content, from a limited subset, was on the east slope of Mt. Washington near the head of Great Gulf. Most dominant lichens in good condition were associated with conifer boles or acidic substrates. The status regarding N and S tolerance for many lichens in the northeastern United States is not clear, so the influence of N pollution on community data cannot be fully assessed. Continued monitoring of lichens and bryophytes, especially if integrated with IMPROVE aerosol data, may reveal changes in air quality, climatic conditions, and other potential stressors or stimuli. Lichen health was impacted by low air quality at some of our sites.

Cover Photo

Great Gulf Wilderness, White Mountain National Forest, view from head of Chandler Brook Trail, August 2013. Photo by Alison C. Dibble, used with permission.

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INTRODUCTION

Lichen species on trees are indicative of air quality both in their species composition and their chemical content, and lichens provide a crucial tool in assessing conditions that present ongoing problems for human health. California, Oregon, and Washington have been proving grounds for lichen community composition and chemistry analyses that have improved air quality monitoring and led to development of the National Air Quality and Lichen Monitoring program (Geiser et al. 2010, Jovan 2008). Monitoring in the Northeast could be enhanced by relating lichen sensitivity ratings, N indicator status, and critical loads of N deposition and other metrics in eastern and western forests (e.g., Pardo et al. 2011a, Pardo et al. 2011b). To increase knowledge about lichens and bryophytes and their relevance for monitoring air quality at three Class I Wilderness Areas in two eastern National Forests, we revisited some of the locales at which elemental data had been collected from four lichen species (Wetmore 1989, 1995a, 1995b). While Wetmore did not measure N or Hg, there were numerous elements that we expected to compare.

Biomonitoring has been developed since the mid-1800s in Europe where lichens and bryophytes continue to be widely used for this purpose. These organisms are valued as bioindicators in part because they can concentrate air- and precipitation-borne elements within the lichen thallus or, in the case of mosses, in the plant body. Unlike vascular plants, epiphytic lichens reflect deposition conditions because the soils are not mediating pollution effects; this then makes them more effective as indicators than plants. In the U.S. Forest Service Inventory and Analysis, Forest Health Monitoring Program (FIA/FHM; http://www.fia.fs.fed.us/program-features/indicators/lichen/), described by Geiser et al. (2010), Glavich and Geiser (2008), and Jovan (2008), biomonitoring employs epiphytic lichens but not bryophytes. Bryophytes can dominate the ground layer under conifers and can be abundant epiphytes especially in wetter habitats. Many species associate with bark of either conifer or deciduous trees (Cleavitt et al. 2009). Because of their prevalence in some habitats, bryophytes may be used to monitor air quality, often as a complement to a lichen biomonitoring program. Lichens alone have vast potential to detect chemicals with severe consequences for human health (e.g., Nascimbene et al. 2014). If used together, epiphytic lichens and bryophytes could detect air conditions that affect human health (Augusto et al. 2013) and serve as indicators of ecological responses to complex changes in air quality and climatic conditions.

New efforts have been mounted to standardize methods in lichen biomonitoring so that studies can be more closely related to each other (see Cristofolini et al. 2014). Because some lichens are more tolerant of S and N deposition than others, their presence or absence on boles of forest trees indicates air pollution conditions (Geiser et al. 2010, McCune and Geiser 1997). In this paper, we follow Pardo et al. (2011a) in use of the terms “oligotroph,” “mesotroph,” and “eutroph” (gradient of N-tolerance from least to most), though there are other systems by which to describe this phenomenon. No single set of terms is yet in global use, as goals of studies differ, or because lichen communities offer different opportunities for understanding pollution tolerance depending on the geographical area studied, proximity to pollution sources such as smelters, power plants, and natural gas production areas, and pollutants involved.

Lichen thalli incorporate airborne particulates, where residence time varies according to the element and to some extent, with weather. Lichens of dry habitats may absorb elements, including heavy metals, differently from their counterparts in moist forests. Foliose lichens differ from fruticose species in this regard also. Samples of lichens subjected to mass spectrometry allow comparison of airborne elements across space and time, so monitoring of deposition levels can be effective. Accuracy, accessibility, and relatively low cost compared to air quality instruments are benefits to this type of monitoring. Lichen conservation is an underlying tenet, because the monitoring process uncovers rare and vulnerable species and provides understanding of their habitats. Forest management can reflect knowledge gained through the air quality monitoring activities. It can also favor tree species that harbor certain unusual lichens or favor the lichens themselves that may require forest continuity and/or closed canopies.

Categories of response to various pollutants have been compiled especially for N, S, and O_3 at http://gis.nacse.org/lichenair/?page/sensitivity. Some examples of northeastern epiphytic lichens include *Lobaria pulmonaria*, which has a low to moderate tolerance for N and is sensitive to S, and the widespread *Hypogymnia* *physodes,* which tolerates a wide range of N and ranges from intermediate to tolerant regarding S and acid deposition. *Evernia mesomorpha* has a particular vulnerability to SO_2 while *Ramalina farinacea* has a moderate tolerance for Pb (Alvarez et al. 2012). Thus the use of a wide spectrum elemental analyses, not monitoring of percent N and percent S alone, is typically undertaken because of known toxicity to lichens of some elements.

Sources of pollutants are varied, and multiple types of sources are possible for many elements. Since enactment of the Clean Air Act of 1977 (cited below), major sources have been changing. For example, Pb was mainly from leaded gasoline prior to phase out. The Environmental Protection Agency noted a 94 percent decline in emissions of Pb between 1980 and 1999, but Pb continues to originate from various sources including piston-engine aircraft that use leaded fuel and from ore and metal processing plants (e.g., lead smelters are upwind of the Wilderness Areas we measured). Ni and V are from fossil fuel combustion. SO_2 has largely been from factories and electric utilities. NOx is emitted by both electric utilities and the transportation sector, and ammonia is generated from livestock waste and fertilized soil. Atmospheric Hg in the United States is primarily from coal-fired power plants, and as with many of these other elements, is involved in long-range transport. Fluorine emissions are associated with power plants and with production of Al, Zn, and phosphate ore-reduction. This is a dynamic picture. Recent fracking activities involve ground disturbance, trucking, and leaks at gas wells that could account for increases in methane, butane, propane, and particulates yet to be well documented for the Northeast.

Class I Wilderness Areas, with their few roads, large areas of vegetation and minimal or no soil disturbance, represent air quality conditions that can be compared across regions and over time. As part of the Clean Air Amendments of 1977, Congress established a national goal of "preventing any future, and remedying any existing, impairment of visibility in mandatory class I Federal areas [in] which impairment results from manmade air pollution" (Clean Air Act of 1970, as amended August 1977). This national goal, along with the Federal Land Manager being given "affirmative responsibility to protect air quality related values", including lichens, within Class I Areas (same CAAA source as previous, p. 52), creates a need to understand the existing health of air quality related values, including lichens. Air quality has improved in the United States from 1980 to 2012, due to the 67 percent reduction of aggregate emissions of six common pollutants (EPA n.d.).

Decline in some lichen species has been documented in some areas of the Northeast and there is concern that such decline is ongoing in New England (Hinds and Hinds 2008). This trend is despite reduced acidity of precipitation, e.g., at Hubbard Brook in the White Mountain National Forest (WMNF) where pH of wet deposition in 1965 was about 4.1 and is currently about 5.0. Also, visibility in terms of regional haze has improved at the rate of about 3 percent per year from 2001 to 2008 near Great Gulf, and 3 percent per year from 2001 to 2009 near Lye Brook (Hand et al. 2011)

In a model to establish critical loads for atmospheric N, or loads below which significant harmful effects are expected to not occur, Geiser et al. (2010) related lichen community data for northwestern maritime forests to speciated aerosol measurements collected at 12 sites by the Interagency Monitoring of Visual Protected Environments (IMPROVE) visibility network. IMPROVE monitors are typically sited at ridgetops to measure air quality reflective of broad spatial scales, and it is unknown whether proximity of lichen plots to such aerosol sites can help explain trends in lichen bulk chemistry. IMPROVE data have the benefit of including metals as well as N and S, while sites in the smaller Clean Air Status and Trends Network (CASTNet) include gaseous as well as particulate S and N components and have advantages for dry deposition calculations of those species.

Lichen plots established in three northeastern Class I Wilderness Areas by Wetmore (1989, 1995a, 1995b) present an opportunity to remeasure and possibly detect changes in air quality as evidenced in bulk chemistry of lichen thalli. Approaches of this project were to (1) investigate potential for remeasurement on Wetmore's plots and establish new plots as a baseline using FIA/FHM protocol regarding the P3 lichen indicator, (2) incorporate bryophyte diversity, abundance, and bulk chemistry in the new plots, (3) compare to lichen biomonitoring in the Pacific Northwest and California, and (4) seek to increase utility and repeatability of air quality assessments using lichens.

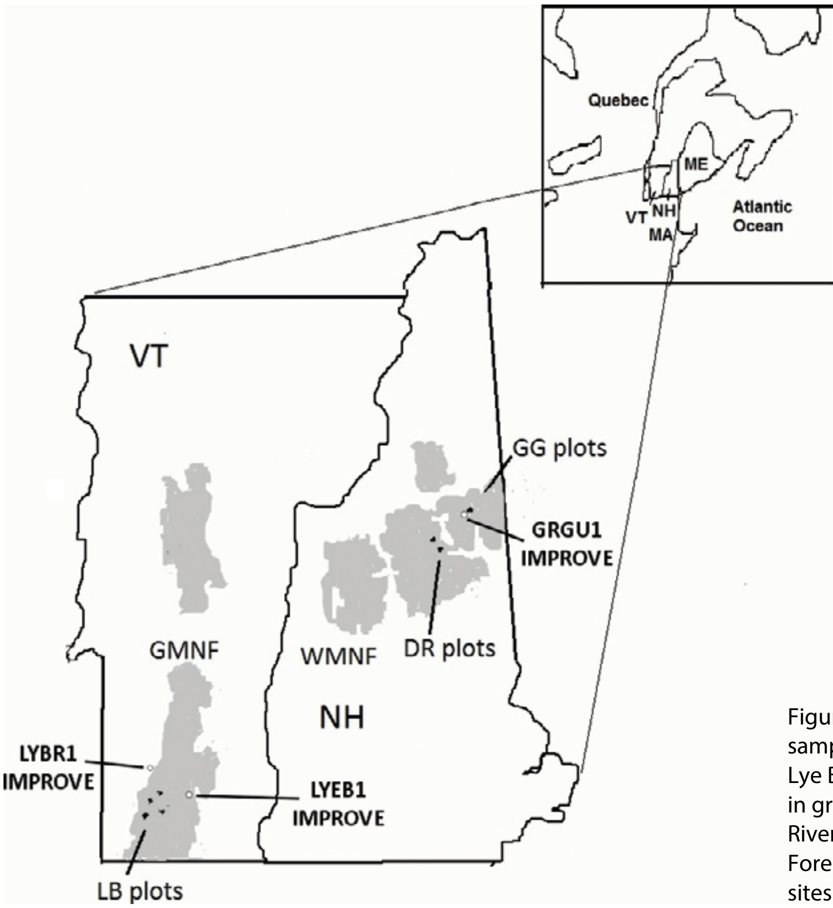


Figure 1.—Location of lichen and bryophyte sample points in three Class I Wilderness Areas, LB= Lye Brook (Green Mountain National Forest shown in gray in Vermont); DR= Presidential Range—Dry River; GG = Great Gulf (White Mountain National Forest shown in gray in New Hampshire). IMPROVE sites labeled as such.

METHODS

Study Locations

We established lichen and bryophyte monitoring sites in 2011-2013 in three Class I Wilderness Areas of the Northeast, attempting to duplicate Wetmore's locations where appropriate for collection purposes (Fig. 1). Two Wilderness Areas are in the WMNF: Presidential Range—Dry River Wilderness and Great Gulf Wilderness. The Lye Brook Wilderness is in the Green Mountain National Forest (GMNF). Four plots were established in Great Gulf and four at Lye Brook, while in Presidential Range—Dry River Wilderness three plots were established within the Wilderness Area and one plot just outside of the Wilderness but still within the National Forest (N=12). Exact locations, plot numbers, and the characteristics at each location are in Tables 1 and 2, and maps are in appendix 1. We were limited to plots that had trees because we were sampling epiphytes.

Timed Plot Surveys

We used a modification of the FIA lichen monitoring protocol as described in McCune et al. (1997). We sought to remeasure some locales sampled by Wetmore (1989, 1995a, 1995b; plot locations in Table 1), so we established a system of 12 circular plots, each 34.7 m in radius. The area of the lichen plot is approximately 3793 m^2 or ≈ 0.379 ha (40,830 ft^2 or ≈ 0.937 acres).

To assess variables that could help explain lichen distribution and elemental load, we collected elevation, slope, and aspect data at each plot and characterized the forest stand. Coordinates were taken for plot center using a handheld global positioning system (GPS). A tree tally was conducted on all boles greater than 10 cm diameter, at 1.37 m above the ground, by tree species, scored alive or dead, in a pie-shaped quarter-section of the full plot (sample area 948.25 m^2) except at plot GR1, where tree tally was on a 7.32 m radius (area 168.33 m^2). Trees were summarized by diameter size classes: 10-12.5, 12.6-25.3, 25.4-36.8, >36.8 cm,

Table 1.—Epiphyte and air quality monitoring plots established in Class I Wilderness Areas 2011-2013. * = on or near lichen plot established by Clifford Wetmore in 1980s-90s in which he collected bulk samples for elemental analysis. + = near plot established by Wetmore but elemental analysis not conducted by him.

Plot name	Wetmore's plot, year(s)	Coordinates in UTM and location notes	Date installed	County, State	Habitat	Bulk samples
DR1*	NE Mt. Crawford 1988, 1993	19 T 0314052, 4889907 White Mountain National Forest, Presidential Range—Dry River, East of Crawford Mtn, along Davis Path	Aug 14, 2012	Coos, NH	Mature montane red spruce forest, among granite ledges	*Evernia mesomorpha, Hypogymnia physodes, Pleurozium schreberi*
DR3*	Rocky Branch Ridge 1993	19T 0317811, 4896845 White Mountain National Forest, Presidential Range—Dry River, Rocky Branch Trail, Engine Hill	Aug 13, 2012	Coos, NH	Mature montane spruce slope forest, balsam fir dominant	*Evernia mesomorpha, Hypogymnia physodes, Pleurozium schreberi*
DR5		19T 0310742, 4898788 White Mountain National Forest, Presidential Range—Dry River, below Mizpah Hut	Aug 15, 2012	Coos, NH	Bench on SE-facing slope, balsam fir dominant	*Hypogymnia physodes, Pleurozium schreberi*
DR6		19T 0310663, 4899744 White Mountain National Forest, Presidential Dry River, Crawford Path, to W of Webster Cliff Trail above Gibbs Brook watershed, Mt. Pierce (Mt. Clinton)	Aug 16, 2012	Coos, NH	Mature balsam fir forest on NW-facing upper slope.	*Hypogymnia physodes, Pleurozium schreberi*
GG1*	Low's (sic) Bald Spot 1993	19T 0320299, 4905656 White Mountain National Forest, Great Gulf Wilderness, Mt. Washington Auto Road, Mile Marker 2, Madison Gulf Trail and Appalachian Trail, Lowe's Bald Knob	Aug 2, 2011	Coos, NH	Mature red spruce-balsam fir with pitch pine on ledge	*Evernia mesomorpha, Hypogymnia physodes*
GG2		19 T 0320019, 4906334 White Mountain National Forest, Great Gulf Wilderness, Mt. Washington Auto Road, Mile Marker 2, Madison Gulf Trail and Appalachian Trail	Aug 15, 2013	Coos, NH	Mature red spruce-balsam fir-yellow birch, with some red maple, with stream	*Evernia mesomorpha, Hypogymnia physodes, Hylocomium splendens*
GG3*	Wamsutta Trail 1993	19T 0317872 4907222 White Mountain National Forest, Great Gulf Wilderness, Mt. Washington Auto Road, Mile Marker 4, Chandler Brook Trail	Aug 16, 2013	Coos, NH	Mature red spruce, balsam fir, mountain paper birch	*Evernia mesomorpha, Pleurozium schreberi, Hylocomium splendens*
GG4		19T 0320717, 4907982 White Mountain National Forest, Great Gulf Wilderness, Osgood Trail ca. 0.1 m W of Jct with Great Gulf Trail; Great Gulf Trail parking lot, proceed ca. 1.8 mi. along Peabody River West Branch	Aug 17, 2013	Coos, NH	Mixed hardwoods, mature yellow birch, eastern hemlock, red spruce, red maple, with striped maple and hobblebush	*Evernia mesomorpha, Hypogymnia physodes, Hylocomium splendens, Pleurozium schreberi*
LB1+		18T 0661520, 4780539 Green Mountain National Forest, Lye Brook Wilderness, off Rootville Trail above Downer Glen Ravine, Manchester	Aug 19, 2013	Bennington, VT	Mature mixed woods in exposed, dry site at top of cliff, red maple, eastern hemlock, yellow birch, some black cherry	*Evernia mesomorpha, Hypogymnia physodes,* no moss
LB2*	Bourn (sic) Pond 1993	18T 0662302, 4774636 Green Mountain National Forest, Lye Brook Wilderness, Bourne Pond, Sunderland	Aug 20, 2013	Bennington, VT	Forested wetland with mid-successional red spruce, balsam fir, red maple	*Evernia mesomorpha, Hypogymnia physodes, Pleurozium schreberi*
LB3+	Lye Brook 1993	18T 0657837, 4775889 Green Mountain National Forest, Lye Brook Wilderness, hill W of Lye Brook, e of and above Rte 7 near summit of ridge above Lye Brook, Manchester	Aug 21, 2013	Bennington, VT	Mature red spruce slope forest, over marble rock	*Evernia mesomorpha, Hypogymnia physodes, Pleurozium schreberi*
LB4*	Kelly Stand 1993	18T 0659626, 4760894 Green Mountain National Forest, Lye Brook Wilderness, Branch Pond Rd (FR 70) near jct Kelley Stand Rd, Sunderland	Aug 22, 2013	Bennington, VT	Low elevation mixed hardwoods dominated by birches, beech, red maple with red spruce	*Evernia mesomorpha, Hypogymnia physodes, Pleurozium schreberi*

Table 2.—Summary of plot characteristics and tree data taken on plots. "Hwd" = deciduous trees (versus conifers). Tree size classes are (1) 12.6-25.3 cm diameter at 1.37 m above the ground, (2) 25.4-36.8, and (3) >36.8.

Plot	Elev (m)	Aspect (°)	Slope (%)	Basal area (m^2/ha)	Quadratic mean diameter (cm)	Hwd (%)	Size classes of 8 largest trees	Visual gap (%)	Recent gaps (<5yrs)	Tree species in order of importance
DR1	925	70	0	14.25	18.55	4	2	5	Y	*Picea rubens, Abies balsamea*
DR3	959	70	0	33.98	17.50	39	1,2	5	Y	*A. balsamea, P. rubens, Betula* spp.
DR5	1135	80	10	39.60	15.82	36	1,2	12	Y	*A. balsamea, Sorbus* sp., *Betula* spp., *P. rubens*
DR6	1245	70	0	42.97	16.23	1	2	0	Y	*A. balsamea, Sorbus* sp.
GG1	857	150	5	24.92	21.10	9	1,2	40	N	*A. balsamea, Betula cordifolia*
GG2	785	72	30	31.09	21.39	86	2,3	5	N	*A. balsamea, Betula alleghaniensis, Acer* spp., *Betula* spp., *Picea rubens*
GG3	1138	60	30	45.20	19.07	17	2,3	20	Y	*A. balsamea, B. cordifolia, P. rubens*
GG4	594	45	5	37.36	28.64	72	3	15	N	*B. alleghaniensis, A. balsamea, Acer* spp.
LB1	561	20	5	35.24	30.41	34	3	5	N	*Acer rubrum, Tsuga canadensis, Fagus grandifolia*
LB2	852	70	5	21.00	16.04	35	1,2	60	Y	*A. balsamea, P. rubens, B. cordifolia*
LB3	809	290	30	34.46	18.70	2	2,3	2	Y	*P. rubens, B. cordifolia*
LB4	803	170	5	30.94	19.42	85	2,3	5	N	*Fagus grandifolia, P. rubens, Betula* spp., *Acer* spp.

with the eight largest trees indicated as potentially important for assessing old growth. For 12 plots, plot center and at the 7.32 m radius in four cardinal directions, a 10 basal area factor prism was used to obtain additional data on basal area. Over the 12 plots, we measured a total of 1,242 trees for species, d.b.h., and status as live/dead. Percent gap in forest canopy was visually estimated to the nearest 10 percent.

To inventory macrolichens, we conducted two-hour timed surveys in which we documented macrolichen species on woody substrata (live and dead tree boles and shrubs) between 0.5 and 1.5 m from the ground and on recently fallen boles, branches and twigs (McCune et al. 1997, USDA FS 2004). Voucher specimens were taken for each species and abundance ratings were assigned based on the frequency of lichen occurrence (scale of 1 - 4 used by FIA protocol; USDA FS 2004—App. A). A one-hour survey for bryophyte epiphytes was also conducted in the plots during the same visit. On seven of the 12 plots where two collectors were present, the timed lichen and bryophyte surveys were conducted simultaneously totaling 1.5 hours per person for each half of the plot. A companion investigation of stand integrity was conducted alongside this project, mostly off plot, by Steven B. Selva, who inventoried the calicioid lichens or "stubble lichens." These consist of both lichenized and non-lichenized fungi, with use for assessing forest continuity by Selva (2002, 2013, 2014) and others (e.g., Tibell 1992). Selva's results from these Wilderness Areas are reported in Selva (2014).

Species Identification and Richness

Lichen specimens were tentatively identified during field data collection so that abundances could be assigned. Vouchers were collected for all species, and identifications were finalized following the nomenclature of Esslinger (2013) and Hinds and Hinds (2008). Bryophytes likewise were identified to genus in the field and determined under the microscope following the nomenclature of Allen (2005, and unpublished data). Lichen and bryophyte specimens are to be deposited at the University of Maine Herbarium (MAINE). Criteria for designation of lichens as associated with old growth were based on Hinds and Hinds (2008), and old-growth bryophytes were based on Cleavitt et al. (2009). To place our 12 plot surveys in a regional context, we compared our species occurrences to those found on 92 FIA/FHM plots in New England (Hinds and Hinds 2008). Comparison to lichen community data collected by Wetmore (1989, 1995a and b) was not possible because of plot size differences, our focus on epiphytes only, and Wetmore's lists by Wilderness Area over all plots rather than by individual plot.

Figure 2.—Differences in thallus health of the common species, *Parmelia sulcata*. The specimen on the left received a health score of 2 (normal) while the specimen on the right received a health score of 0 (poor condition). Photos were taken at 14 times magnification with the decreased size of the poor specimen being actual size difference. Photos were taken by Patricia Hinds on specimens collected for this project and scored by Jim Hinds. Photos used with permission.

Scoring Lichen Thallus Condition

Macrolichen specimens collected as vouchers were reviewed for scoring of outward appearance of thallus condition, referred to as "health." We used a 4-point scale: 0 = thallus in very poor condition (more than one of the following symptoms: convoluted lobes, bleaching, black speckles, pink blotchy areas) or extensive other discoloration and/or damage; 1 = thallus in poor condition (one or two of the previously listed symptoms, but not as extensive on the thallus surface); 2 = within the normal range for the species; and 3 = robust specimen (Fig. 2). If a specimen was the primary collection in a packet, it was scored for health but secondary specimens in the same packet were not scored. Secondary specimens were not scored. Scores were summed for each species and divided by the number of collections per plot to find the average score for that species on the plot. All scoring was done to avoid variation that could occur among observers. A total of 632 specimens from three Wilderness Areas was evaluated.

Bulk Samples

To increase potential for comparing our data with dry weight chemistry results of Wetmore (1989, 1995a, 1995b), we sought a subset of his sampling areas so that we could compare results for near spots on the ground, and we engaged the same lab he had used for mass spectrometry. Bulk sample collections included both lichen thalli from branches and bark, and moss stems from the ground layer. We sought sufficient material off-plot for *Hypogymnia physodes*, with backup *Evernia mesomorpha*, and for the bryophyte *Hylocomium splendens*, with backup *Pleurozium schreberi*. Vinyl gloves were used to reduce contamination, and air-dried specimens were cleaned of any twigs, spruce/fir needles, or non-target lichens with a time-limit of two hours per sample, and shipped to the Research Analytical Lab at the University of Minnesota, St. Paul, Minnesota (http://ral.cfans.umn.edu/). There they were oven dried, ground, and analyzed simultaneously for total N (LECO FP-528 total N analyzer) and total S (LECO SC-132 S analyzer, manufactured by the LECO Corp., St. Joseph, Michigan), following methods of Geiser (2004). Totals are expressed as percent content of sample dry weight.

We analyzed for 27 elements using inductively coupled plasma atomic emission spectrometry (ICP-AES) (Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Si, Sr, Ti, V, and Zn). Results are expressed in ppm or ppb unless noted otherwise (μg/g = ppm and ng/g = ppb). Dried lichen or plant material of about 500-1000 mg was weighed into a 20 mL high form silica crucible and dry ashed at 485 °C for 10-12 hours. Crucibles were covered during the ashing as a precaution against contamination. Ash was equilibrated with 5 mL of 20 percent HCl at room temperature for 30 minutes. Then 5 mL of deionized water was added, gently swirled, and allowed to settle for 3 hours. The solution was decanted into 15 ml plastic disposable tubes for direct determination by ICP-AES. Simultaneous sampling of total N, total S, and elements was recommended by Blett et al. (2003). For some Areas and samples, data sent from the lab did not include Ba, Be, Co, Li, Rb, Si, Ti, and V. Total Hg was obtained from three plots at Great Gulf and four at Lye Brook (lacking for the Presidential Range—Dry River) for the purpose of comparing Hg across geographic area and to establish a baseline for estimating changes in Hg deposition.

Summarized FIA/FHM Plots

To provide more context for data collected during this project, we compared data from the 12 new plots to a list of lichens found in 92 FIA/FHM P3 lichen indicator plots on National Forests, state lands, or private woodlots across New England (Connecticut, Massachusetts, Maine, New Hampshire, Rhode Island). All plots are part of a system of permanent plots in forested habitats across the United States. These had been analyzed by Hinds and Hinds (2008).

Analytical Approaches for Bulk Chemistry and Thallus Health Scores

To compare new bulk chemistry information to that of Wetmore's earlier reports (1989, 1995a, 1995b), we sought structure in the data for seven remeasured plots using Excel® pivot charts and pivot tables, especially bar graphs for total N, total P, and elements for which a matchup of data were available.

Bulk chemistry samples were also analyzed by organism type (lichens vs. mosses), by National Forest, by Wilderness Area, and within plots over time. Differences between the group of lichens ("lichen thalli": *Hypogymnia physodes, Evernia mesomorpha, Platismatia glauca*) and the group of mosses ("moss stems": *Hylocomium splendens, Pleurozium schreberi*) were greater than within-group differences. Data for Ti and V were lacking for Presidential Range—Dry River and for plot GG1.

For both bulk chemistry and lichen thallus health scores, Welch's test allowing unequal variances was used to accommodate differences in sample size between groups. We then used Tukey's HSD between-pair comparison as a post-hoc test for differentiating significant subsets when the Welch's ANOVA was significant. Analyses were run using JMP Pro 10 (SAS Institute, Cary, NC).

NMS Methods

Species and site data were analyzed using Non-metric Multidimensional Scaling (NMS) in PC-ORD ver. 6.0 (MJM Software, Gleneden, OR). Only species present at three or more plots were included and this left a data set with 12 sites and 60 species (15 mosses, 8 liverworts, 37 macrolichens). The site variables included in the second matrix were elevation, percent of hardwoods, percent conifers, eastness, northness, percent of shrub cover, low shrubs (binary), new gaps (binary), chemistry data from the bulk samples for percent N and S, contents of Al, Cu, Fe, Ni and Pb, epiphyte species richness values, and mean thallus health scores. Aspect was decomposed into variables of eastness as sin (aspect) and northness cos (aspect). For eastness, a value of 1 is due east and a value of -1 is due west. For northness, a value of 1 is due north (0 degrees true) and a value of -1 is due south (180 degrees).

The NMS analysis is an ordination method that uses an iterative search for rankings and placement of the analyzed variables to find the solution that minimizes stress (McCune and Mefford 1999). Because NMS uses ranks, it can be considered a non-parametric form of ordination with relaxed assumptions on data structure that are usually more applicable to ecological data (McCune and Mefford 1999). All NMS analyses were run using Sorensen's distance measure with 100 runs using real data and 50 runs of randomized data. The instability criterion was 0.00001 with 500 as the maximum number of iterations. The final solution was chosen based on the dimensionality with the lowest mean stress from a run comparing randomized to real data (McCune and Mefford 1999).

IMPROVE Aerosol Chemistry Data and Analysis

We obtained elemental (x-ray fluorescence) and ionic (ion chromatography) aerosol data from IMPROVE sites adjacent to the Lye Brook Wilderness (LYBR1, 1991-2012, and LYEB1, 2011-2012) and to the Great Gulf Wilderness (GRGU1, 2001-2012). These data are collected every 3 days, year round. They were acquired from the Federal Land Manager Environmental Database (http://views.cira.colostate.edu/fed/), with the concurrent 2001-2012 data used to assess differences between sites and the longer-term Lye Brook data used to assess long-term trends. More detailed exploration of these regional aerosol data and their relationship to lichen bulk chemistry will be presented in a separate report[1], and they are briefly summarized below in terms of the activity of monitoring using lichens.

RESULTS

Bulk Sample Chemistry

Comparison of present day samples across sites. Epiphytic lichen thalli vs. ground layer moss stems differed significantly for all elements summarized here in ANOVA comparisons by Wilderness Area using Welch's test and involving percent N, percent S, and ppm by sample dry weight for Al, Cu, Fe, Ni, Pb, and Zn (Figs. 3a, b). Lichen thalli contained higher levels of most heavy metals (Al, Fe, Ni, Pb, Zn; Ti and V differed likewise but not shown) while the mosses had higher values for percent N and S and Cu. Subsequently, comparisons among areas were done separately for lichen thalli and moss stems, which decreased our sample size and power to detect among-site differences. For moss samples, only Al was significantly different among sites with higher levels at Presidential Range—Dry River (Fig. 3a).

For lichen samples, percent N was higher at Lye Brook, intermediate at Great Gulf, and lowest at Presidential Range—Dry River, while percent S did not differ significantly among study sites (Fig. 3b). These are not all at the same elevation. Presidential Range—Dry River and Lye Brook tended to be higher for the heavy metals, while Lye Brook was also higher for percent N and S (Figs. 3a and b). Plot DR5 had particularly high levels of heavy metals (Al: 522 µg/g, Fe: 446 µg/g, Ni: 2.46 µg/g, Pb: 30.38, Zn: 125 µg/g), well above the mean for the other plots and exceeding Wetmore's

[1] Cleavitt et al., unpublished data. Available from author N. Cleavitt.

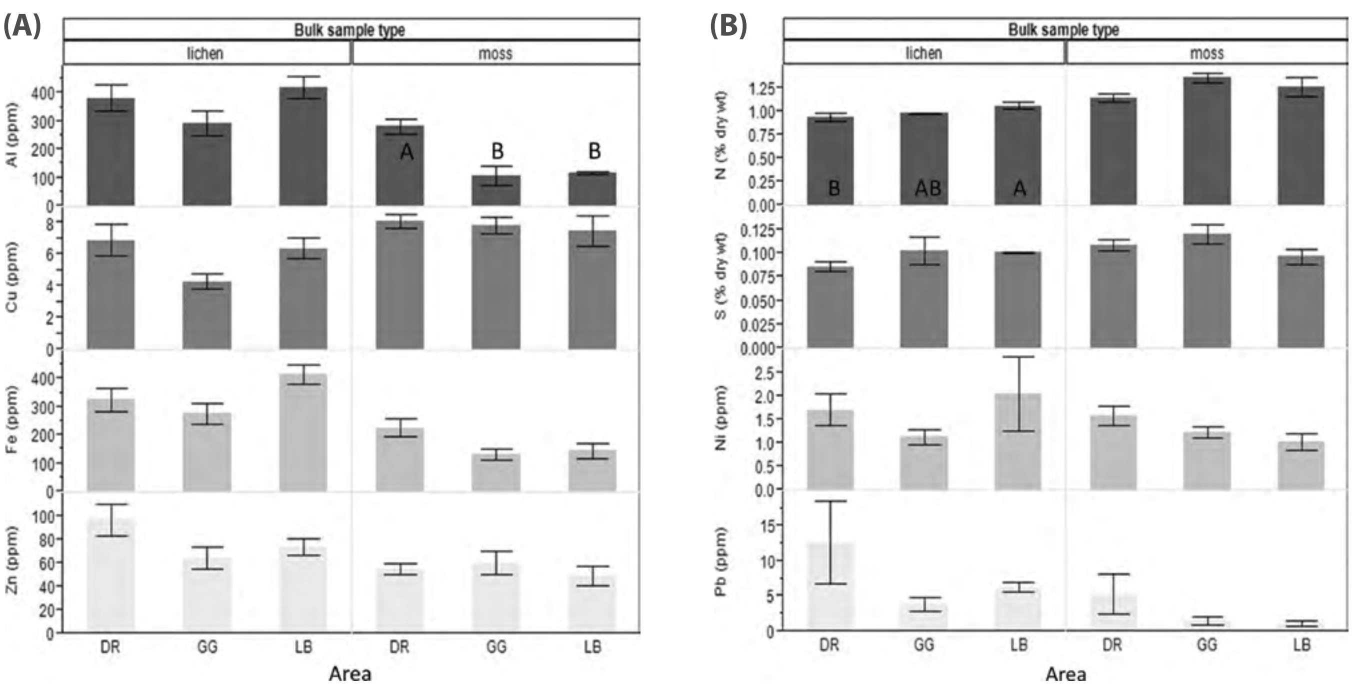


Figure 3.—At the level of Wilderness Area, comparison in 2011-2013 bulk chemistry for lichens and mosses regarding (A) Al, Cu, Fe, and Zn; (B) percent N, percent S, Ni, and Pb. Note that lichen thalli were collected from branches and bark while moss stems were collected from the ground layer. Wilderness Areas codes as for Figure 1. Bars are the mean value with one standard error. Bars with different letters are significantly different at α = 0.05 by Tukey HSD.

maximum value for Cu (DR5: 9.67 µg/g; Wetmore's maximum: 6.70 µg/g; data not shown). Two plots at Lye Brook, LB1 and LB4, had higher heavy metal values. At LB1, the lichen thalli had Ni content of 4.46 µg/g, which was four times the average value (not shown). At LB4, the values for Al (537 µg/g) and Fe (500 µg/g) were both notably high.

Comparison of National Forests across sample times. We pooled plots by National Forest and examined differences in bulk sample chemistry. Four elements have changed between sample times: Pb, total S, Al, and Cu (Figs. 4a-d). We examined three main effects: lichen species (*Hypogymnia physodes* and *Evernia mesomorpha*), year (1988, 1993, or 2011-2013) and National Forest (Green vs. White Mountain National Forests). The lichen species differed significantly in their element contents for all heavy metals (all $p < 0.001$) with the foliose *H. physodes* consistently higher in contents than the fruticose *E. mesomorpha* (Fig. 4).

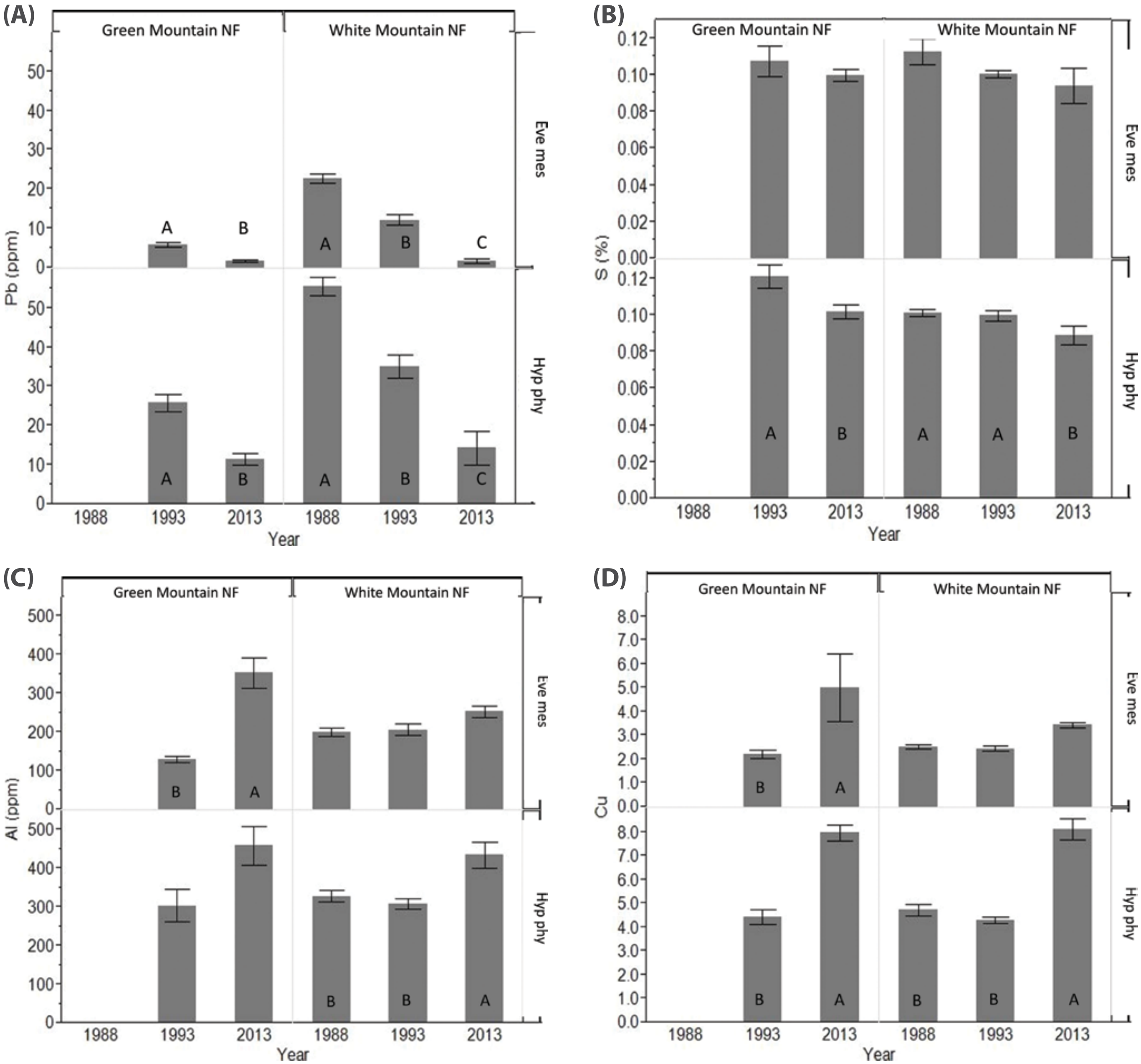


Figure 4.—At the level of two National Forests, comparison between Wetmore's 1993 samples in bulk chemistry and those of 2011-2013 for bulk samples of two lichen species, *Evernia mesomorpha* and *Hypogymnia physodes*. Note that the 1988 sample data from Wetmore for the WMNF is shown but not included in the analysis because of low replication and lack of samples at GMNF. The elements with significant differences were: (A) Pb, (B) S, (C) Al, and (D) Cu. Bars are the mean value with one standard error. Bars with different letters are significantly different at $\alpha = 0.05$ by Tukey HSD.

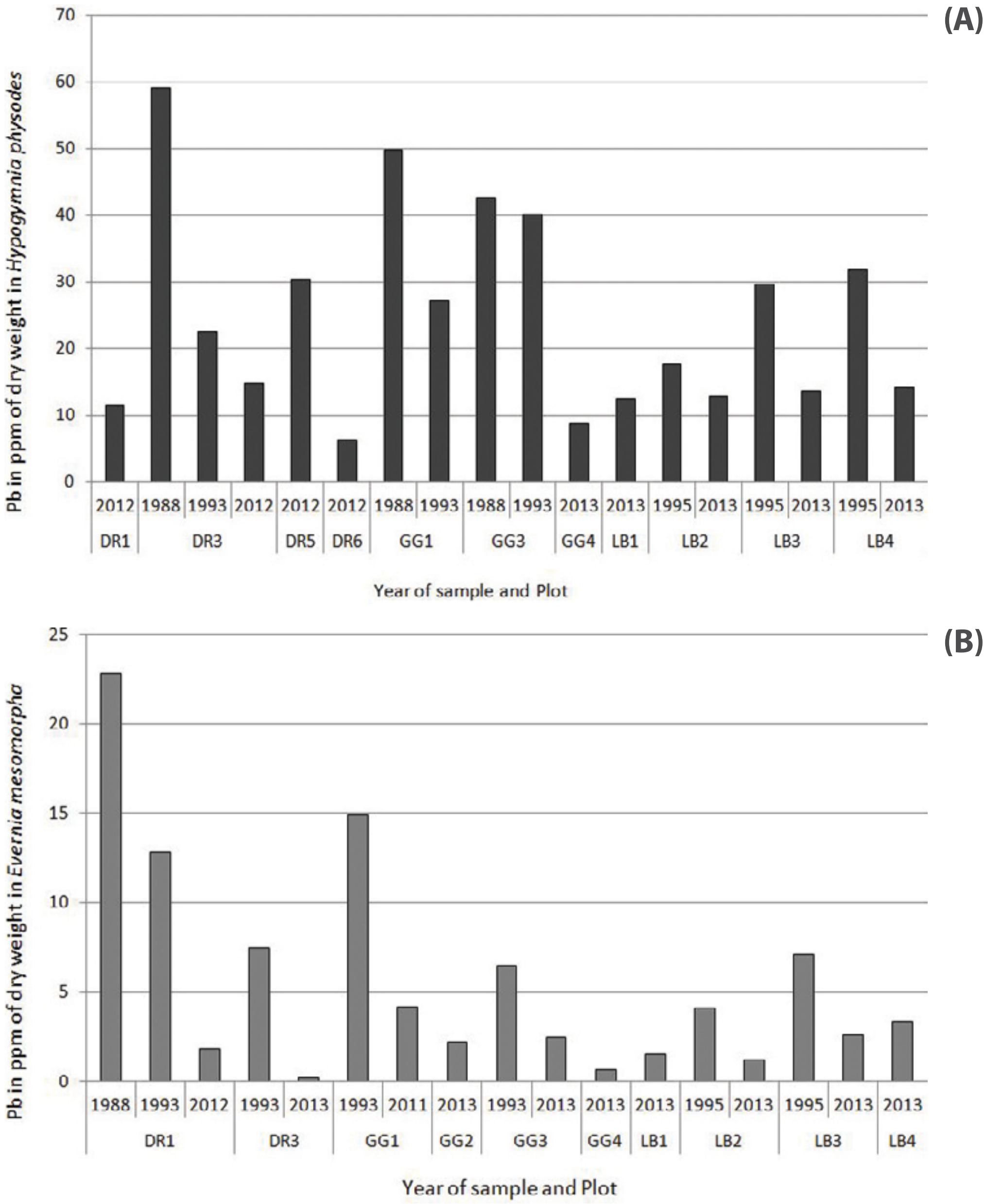


Figure 5.—At the plot level, repeat measurement data for dry weight Pb in ppm within thalli of epiphytic lichens (A) *Hypogymnia physodes* and (B) *Evernia mesomorpha*. Plots for which there are multiple years are those measured initially by Wetmore; 2011-2013 data are from this study.

However, the two lichens show the same patterns in the elements with a decrease in Pb, and an increase in Al and Cu between Wetmore samples and our recent samples (Fig. 3).

Since 1993, both Pb content ($F_{1,67}$ = 41.08; p < 0.0001) and percent of S ($F_{1,67}$ = 5.71; p =0.0197) of the lichen thalli decreased significantly while Al ($F_{1,67}$ = 32.70; p < 0.0001) and Cu ($F_{1,67}$ = 92.46; p < 0.0001) content increased (Fig. 4). In bulk lichen samples at the National Forest level, the Pb content ($F_{1,67}$ = 11.14; p = 0.0014) was higher at WMNF, while percent S ($F_{1,67}$ = 10.76; p = 0.0016) was higher in samples from the GMNF (Fig. 4).

Plot by plot comparisons. Direct comparisons between Wetmore's seven plots that we remeasured in 2011-2013 brought out few unequivocal differences that we consider indicative of change over time. There was a decrease in Pb in two lichens, *Evernia mesomorpha* and *Hypogymnia physodes* (Fig. 5). An exceptionally high level of Mg++ in *Hypogymnia physodes* was found at

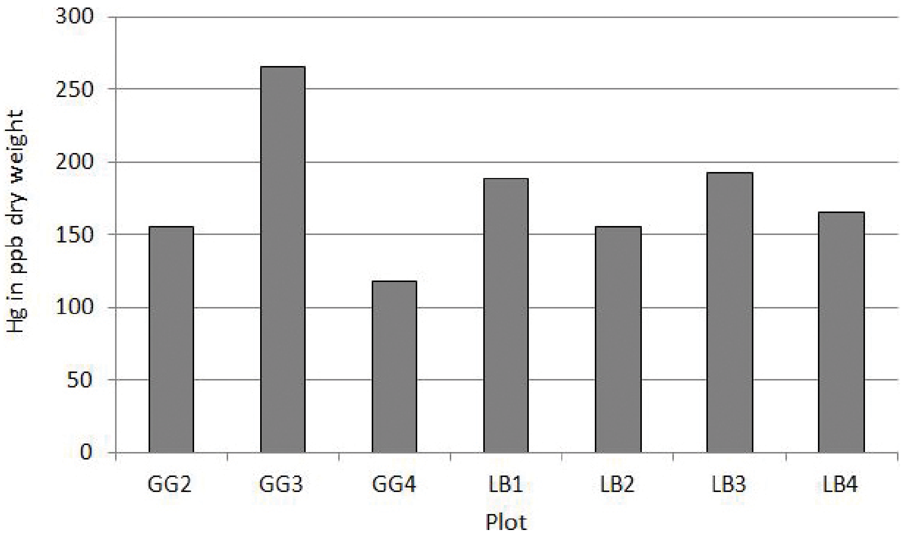


Figure 6.—At the plot level, dry weight of Hg in ppb in samples of epiphytic lichen *Evernia mesomorpha* in seven of 12 plots established in Class I Wilderness Areas in 2011-2013. Wilderness Area codes as in Fig. 1.

LB4 (not shown). It was more than twice as high as any other reading in the seven comparison plots and could be associated with acid rainfall. Otherwise, dry weight results for many elements were inconclusive or showed increase, and because of complications with the greater precision of recent instruments, we interpret that the only clear trend was decrease in Pb in the plot-by-plot comparisons.

The Wilderness Area averages for 2011-2013 samples of Hg content in lichen thalli at Great Gulf (179.4 ± 76 ppb) and Lye Brook (175.7 ± 18 ppb; levels for *Evernia mesomorpha* shown in Fig. 6) were four times that of the moss stems (46.4 ± 6 and 38.8 ± 2 ppb, respectively). Plot GG3 had a much higher Hg level in *Evernia mesomorpha* than any other sample (264.95 ppb). On an individual plot basis, mosses had much lower Hg levels than lichens by as much as five times (not shown).

Multivariate correlations for the elements we tested are provided in appendix 2.

Assessment of Thallus Condition for Common Macrolichens

There were significant differences in condition of lichen thalli, referred to here as "health," among the plots ($F_{11,89} = 12.42$; $p < 0.0001$; Fig. 7a). When plots were lumped by Wilderness Area ($F_{2,167} = 24.99$; $p < 0.0001$), each Area was distinct. Lichens of Presidential Range—Dry River plots were in the best condition, followed by those in Great Gulf and Lye Brook (Fig. 7b). Lichens of DR3 were not significantly different from the Lye Brook plots and had the lowest condition score for Presidential Range—Dry River. Poor condition of lichens in GG4, the lowest in elevation of the Great Gulf plots, approached those of Lye Brook (Fig. 5a). *Cladonia* spp. were less sensitive to difference among areas within the WMNF (Fig. 7c).

Thallus condition differences were also compared by lichen frequency groups (common, infrequent, rare) across the three areas. For the 14 common species (occurring in at least two plots per area: $F_{2,94} = 11.36$; $p < 0.0001$), Lye Brook lichens had significantly worse condition than those of both Presidential Range—Dry River and Great Gulf (subsets not shown). For the remaining less frequent or rare species, thallus health was significantly greater at Presidential Range—Dry River ($F_{2,69} = 14.05$; $p < 0.0001$; subsets not shown). The differences among Wilderness Areas with all the species were a result of poorer condition of both the common and infrequent lichens at Lye Brook and poorer condition of infrequent lichens at Great Gulf.

Species Richness and Among-Plot Patterns

In the timed epiphyte surveys at 12 plots, we found 113 species (39 mosses, 16 liverworts, and 58 macrolichens), of which 58 species of cryptogams were present in sufficient frequency to be included in a multivariate analysis (Tables 3 and 4). For macrolichens, Great Gulf sites had a total of 45 species, but plot GG4 had only 16 species. For the Presidential Range—Dry River plots, there were 41 species, and DR5 had the fewest (19 species). Lye Brook had 33 species in total and both LB1 (13 species) and LB3 (16 species) were particularly depauperate. For the mosses and liverworts, Great Gulf had 39 bryophyte species (28 mosses and 11 liverworts). Presidential Range—Dry River had only 13 bryophyte species recorded across the four plots. Lye Brook had 40 species (26 mosses and 14 liverworts), including many bryophytes that were unique to that Wilderness Area (Tables 3, 4).

The most common lichens, found in at least 10 of our 12 plots, were *Cladonia coniocraea, C. ochrochlora, Hypogymnia physodes, Parmelia saxatilis, P. sulcata, and Usnocetraria oakesiana*. The most common bryophytes were two mosses, *Hypnum pallescens* and *Orthodicranum montanum*, and a liverwort: *Ptilidium pulcherrimum* (Table 4).

Macrolichen species distribution was such that 36.2 percent of species occurred at only one (16 spp.) or two (four spp.) plots (Table 3). The mosses were about twice as likely to be sporadic or rare in occurrence with 61.5 percent of the species found at only one (15 spp.) or two (nine spp.) plots. Fifty percent of liverworts were found at only one (seven spp.) or two (one sp.) plots. Species unique to each site are noted in Table 4.

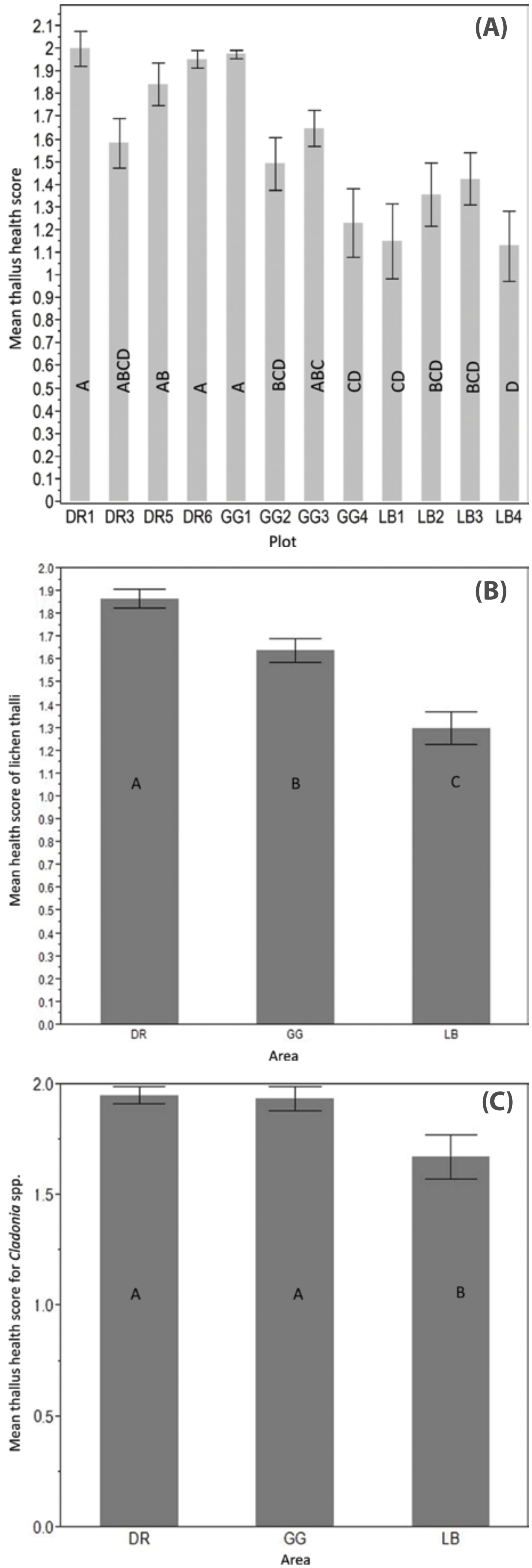


Figure 7.—Visual rating of thallus health on specimens collected during the timed surveys for (A) all 12 plots, (B) all species pooled by Wilderness Area, and (C) *Cladonia* species only, by Wilderness Area. The bars are mean for the plot with one standard error from the mean. Bars with no overlap in letters are significantly different at α = 0.05 by Tukey HSD.

Table 3.—Species richness in a summary of epiphyte surveys on 12 plots located in Class I Wilderness Areas of the National Forest. Old-growth indicators (Cleavitt et al. 2009, Hinds and Hinds 2008) are given as bryophyte species/macrolichen species. Eutroph species status derived in Table 6.

Site/plot	Epiphyte richness	Moss richness	Liverwort richness	Macrolichen richness	Old-growth indicators	Fruticose species	Eutroph species
All sites	113	39	16	58	5/13	7	4
Presidential Range—Dry River							
DR1	35	4	1	30	0/5	5	0
DR3	34	8	2	24	2/2	1	1
DR5	26	6	1	19	0/3		0
DR6	35	3	1	31	0/3	4	0
Great Gulf							
GG1	39	8	2	29	2/2	3	0
GG2	40	10	4	26	2/5	2	1
GG3	44	12	5	27	0/2	4	0
GG4	39	15	8	16	3/1	0	3
Lye Brook							
LB1	26	7	6	13	4/0	0	3
LB2	45	12	9	24	4/2	1	0
LB3	39	11	4	24	2/2	2	1
LB4	44	20	8	16	5/3	1	4

Table 4.—Epiphyte species found in the timed surveys at 12 plots, with presence ("1"), frequency by Wilderness Area, and overall. Plot codes as in Fig. 1. See footnote for list of unique species by Wilderness Area.

	Plots															
Epiphyte species	DR 1	DR 3	DR 5	DR 6	DR freq	GG 1	GG 2	GG 3	GG 4	GG freq	LB 1	LB 2	LB 3	LB 4	LB freq	All freq
Mosses																
Amblystegium serpens					0					0				1	1	1
Anacamptodon splachnoides					0					0				1	1	1
Brachythecium laetum					0			1		1				1	1	2
Brachythecium roteanum					0		1	1		2				1	1	3
Brachythecium rutabulum					0				1	1					0	1
Brotherella recurvans					0		1			1					0	1
Callicladium haldanianum					0				1	1		1	1	1	3	4
Dicranum fuscescens	1	1	1	1	4			1		1					0	5
Dicranum scoparium	1		1		2	1				1		1			1	4
Dicranum viride					0				1	1				1	1	2
Drepanocladus aduncus					0	1				1					0	1
Eurhynchium pulchellum					0				1	1					0	1
Hylocomium splendens					0			1		1					0	1
Hypnum cupressiforme					0					0				1	1	1
Hypnum faurei				1	1					0					0	1
Hypnum imponens					0	1				1					0	1
Hypnum pallescens	1	1	1		3	1	1	1	1	4	1	1	1	1	4	11

continued

Table 4.—continued

Epiphyte species	Plots DR 1	DR 3	DR 5	DR 6	DR freq	GG 1	GG 2	GG 3	GG 4	GG freq	LB 1	LB 2	LB 3	LB 4	LB freq	All freq
Isopterygium elegans					0		1	1	1	3			1		1	4
Leskea polycarpa					0					0				1	1	1
*Neckera pennata**					0				1	1					0	1
Orthodicranum flagellare					0					0			1		1	1
Orthodicranum fulvum					0			1		1					0	1
Orthodicranum montanum	1	1	1		3	1	1	1	1	4	1	1	1	1	4	11
Orthotrichum sordidum		1			1					0				1	1	2
Orthotrichum stellatum					0					0	1			1	2	2
Paraleucobryum longifolium					0	1	1		1	3		1		1	2	5
Plagiomnium ciliare					0				1	1					0	1
Plagiomnium cuspidatum					0			1	1	2					0	2
Plagiothecium laetum		1			1			1		1		1	1		2	4
Platydictya subtile					0					0	1		1		2	2
Platygyrium repens		1	1		2		1		1	2	1	1	1	1	4	8
Pleurozium schreberi					0			1		1		1			1	2
Pylaisiella polyantha					0					0	1			1	2	2
Pylaisiella selwynii					0				1	1		1		1	2	3
Sanionia uncinata				1	1		1	1		2					0	3
Tetraphis pellucida					0				1	1		1		1	2	3
Thuidium delicatulum					0					0			1	1	2	2
*Ulota coarctata**		1			1	1	1			2		1	1	1	3	6
Ulota crispa		1	1		2	1	1		1	3	1	1	1	1	4	9
Liverworts																
Cephaloziella sp.					0					0		1			1	1
Frullania asagrayana					0		1		1	2				1	1	3
*Frullania bolanderi**		1			1	1	1			2	1	1		1	3	6
Frullania eboracensis					0		1		1	2	1	1	1	1	4	6
Frullania inflata					0					0	1				1	1
Geocalyx graveolens					0					0		1			1	1
Jamesoniella autumnalis					0				1	1				1	1	2
Lophocolea heterophylla					0			1	1	2			1	1	2	4
Lophozia ascendens					0			1		1					0	1
Lophozia sp.					0					0		1			1	1
Nowellia curvifolia					0			1	1	2		1	1		2	4
Porella platyphylla					0				1	1	1			1	2	3
Ptilidium ciliare					0					0		1			1	1
Ptilidium pulcherimum	1	1	1	1	4	1	1	1	1	4	1	1	1	1	4	12
Radula complanata					0				1	1	1	1		1	3	4
Tritomaria sp.					0			1		1					0	1
Total bryophyte species	5	10	7	4		10	14	17	23		13	21	15	28		

continued

Table 4.—continued

Epiphyte species	DR 1	DR 3	DR 5	DR 6	DR freq	GG 1	GG 2	GG 3	GG 4	GG freq	LB 1	LB 2	LB 3	LB 4	LB freq	All freq
	Plots															
Macrolichens																
*Ahtiana aurescens**	1				1					0					0	1
Bryoria furcellata	1				1	1				1					0	2
*Bryoria fuscescens**			1		1					0					0	1
Bryoria nadvornikiana	1			1	2			1		1					0	3
Candelaria concolor					0					0				1	1	1
*Cetrelia olivetorum**					0		1			1					0	1
Cladonia caespiticia					0					0	1	1		1	3	3
Cladonia chlorophaea grp	1	1		1	3	1		1	1	3		1	1	1	3	9
Cladonia coniocraea	1	1	1	1	4	1	1	1	1	4		1	1	1	3	11
Cladonia cristatella					0				1	1					0	1
Cladonia decorticata				1	1					0					0	1
Cladonia digitata	1	1		1	3					0					0	3
Cladonia floerkeana	1			1	2	1				1		1	1		2	5
Cladonia macilenta	1	1		1	3	1		1		2		1	1		2	7
Cladonia metacorallifera					0			1		1					0	1
Cladonia ochrochlora	1	1	1	1	4	1	1	1	1	4		1	1	1	3	11
Cladonia ramulosa			1		1					0					0	1
Cladonia squamosa	1		1	1	3	1	1	1	1	4		1	1		2	9
Cladonia sulphurina	1			1	2					0					0	2
Evernia mesomorpha	1	1	1	1	4	1	1	1		3		1	1		2	9
Flavoparmelia caperata					0		1			1	1	1		1	3	4
*Hypenomyce friesii**	1				1					0					0	1
Hypogymnia incurvoides		1		1	2	1	1	1		3					0	5
Hypogymnia krogiae	1	1	1	1	4	1	1	1		3		1	1		2	9
Hypogymnia physodes	1	1	1	1	4	1	1	1		3	1	1	1		3	10
*Hypogymnia tubulosa**	1	1	1	1	4	1	1	1		3			1		1	8
*Hypogymnia vittata**				1	1					0					0	1
Imshaugia aleurites	1	1		1	3	1	1	1		3		1	1		2	8
*Lobaria pulmonaria**					0		1			1					0	1
Melanohalea halei					0	1	1		1	3	1			1	2	5
Melanohalea olivacea					0	1				1					0	1
Melanohalea septentrionalis		1			1	1	1			2					0	3
*Melanelixia fuliginosa**	1				1		1			1		1	1	1	3	5
Melanelixia subaurifera				1	1	1			1	3	1	1	1	1	4	8
Myelochroa aurulenta				1	1				1	1					0	2
Parmelia saxatilis	1	1	1	1	4	1	1	1	1	4	1		1		2	10
Parmelia squarrosa	1	1			2	1		1		3		1	1	1	3	8
Parmelia sulcata	1	1	1	1	4	1	1	1	1	4	1	1	1	1	4	12
Parmeliopsis ambigua					0			1		1					0	1

continued

Table 4.—continued

	Plots															
Epiphyte species	DR 1	DR 3	DR 5	DR 6	DR freq	GG 1	GG 2	GG 3	GG 4	GG freq	LB 1	LB 2	LB 3	LB 4	LB freq	All freq
Parmeliopsis capitata	1				1					0		1	1		2	3
Parmeliopsis hyperopta	1	1	1	1	4	1		1		2			1		1	7
Phaeophyscia pusilloides					0				1	1	1				1	2
Phaeophyscia rubropulchra		1			1		1		1	2	1		1	1	3	6
Physcia millegrana					0				1	1	1				1	2
Platismatia glauca	1	1	1	1	4	1	1	1		3		1	1		2	9
*Pseudevernia cladonia**	1	1	1	1	4	1	1	1		3		1			1	8
Pseudevernia consocians	1	1	1	1	4	1	1	1		3		1			1	8
Punctelia caseana					0		1			1		1	1		2	3
Punctelia rudecta					0		1		1	2	1	1	1	1	4	6
Pyxine sorediata					0				1	1	1			1	2	3
Tuckermannopsis americana					0	1				1					0	1
Tuckermannopsis ciliaris					*0*			*1*		*1*					*0*	*1*
Tuckermannopsis orbata	1	1	1	1	4	1		1		2		1	1		2	8
Usnea filipendula	1	1	1	1	4		1	1		2					0	6
Usnea hirta	1				1					0					0	1
Usnea subfloridana			1	1	2	1		1		2			1	1	2	6
Usnocetraria oakesiana	1	1	1	1	4	1	1	1	1	4	1	1	1	1	4	12
Vulpicidia pinastri	1	1		1	3	1		1		2					0	5
Total macrolichens	31	24	19	31		29	26	27	16		13	24	25	16		
All epiphyte species	36	34	26	35		39	40	44	39		26	45	40	44		
% macro-lichen	86	71	73	89		74	65	61	41		50	53	63	36		

Footnote list:

* = old-growth species, according to Cleavitt et al. (2009) for bryophytes and Hinds and Hinds (2008) for lichens.

Lichen species unique to:

Great Gulf: *Cetrelia olivetorum, Cladonia metacorallifera, Lobaria pulmonaria, Melanohalea olivacea, Parmeliopsis ambigua, Tuckermannopsis americana*, and *Tuckermannopsis ciliaris*;

Presidential Range—Dry River: *Ahtiana aurescens, Bryoria fuscescens, Cladonia cristatella, Cladonia decorticata, Cladonia digitata, Cladonia sulphurina, Hypocenomyce friesii, Hypogymnia vittata*, and *Usnea hirta*;

Lye Brook: *Candelaria concolor* and *Cladonia caespiticia*.

Bryophytes unique to:

Great Gulf: *Brachythecium rutabulum, Brotherella recurvans, Drepanocladus aduncus, Eurhynchium pulchellum, Hylocomium splendens, Hypnum imponens, Neckera pennata, Orthodicranum fulvum, Plagiomnium ciliare, Plagiomnium cuspidatum, Lophozia ascendens*, and *Tritomaria* sp.;

Presidential Range—Dry River: *Hypnum faurei*;

Lye Brook: *Amblystegium serpens, Anacamptodon splachnoides, Hypnum cupressiforme, Leskea polycarpa, Orthodicranum flagellare,Orthotrichum stellatum, Platydictya subtile, Pylaisiella polyantha, Thuidium delicatulum, Cephaloziella sp., Frullania inflata, Geocalyx graveolens*, and *Ptilidium ciliare*.

In a comparison of our plots with lichen data for 92 FIA plots in New England (see Hinds and Hinds 2008), we recorded six species not found in the New England plots (Table 5). By contrast, we did not find some frequent cyanobacteria-containing lichens, *Leptogium cyanescens* and *Lobaria quercizans* (Table 5). We also did not have common eutrophic species, *Xanthomendoza hasseana* or *Physcia aipolia* (Table 5). Of the species for which pollution indicator status was available, our plots had one eutrophic species (*Candellaria concolor* at Lye Brook), four mesotrophic species, and five oligotrophic species (Table 5).

Table 5.—Frequency of macrolichen species in 12 plots established by us in 2011-2013 and in 92 plots summarized by Hinds and Hinds (2008) that were surveyed using the FIA/FHM timed protocol, with notes about N indicator status based on many sources, with some contrasting categorizations from Europe (e.g., Sparrius 2007). Oligo = oligotrophic (compare to acidophyte in Jovan 2008 Table 6); Meso= potential mesotroph (compare to neutrophyte); Eutro = eutrophic (compare to nitrophyte); OG = old-growth indicator species; Cy = stratified cyanolichen; see footnote. For most, tolerance for N is undocumented or unpublished.

Species*	Occurrence in New England	Species notes	Our plots %	New England %
Bryoria trichodes	FREQ	Sensitive to air pollution (Oligo)	0	8
Leptogium cyanescens	FREQ	Cy, should be common on hardwoods	0	9
Lobaria quercizans	FREQ	OG; tripartite with Cy component	0	17
Xanthomendoza hasseana	FREQ	Eutro	0	7
Physcia aipolia	COM	Eutro	0	8
Myelochroa galbina	COM	Hardwood requirement	0	28
Physconia deters	COM	Hardwood requirement	0	13
Physcia stellaris	COM		0	25
Ahtiana aurescens	UC	OG; G3; R3	8	5
Bryoria fuscescens	UC	OG; R3 (Oligo)	8	3
Cladonia decorticata	UC		8	N/A
Cladonia metacorallifera	RARE	7 sites in Meso; R2; (as C. squamosa, Oligo)	8	N/A
Cladonia sulphurina	UC	Northern species	8	N/A
Hypocenomyce friesii	UC	OG	8	0
Hypogymnia vittata	UC	OG	8	0
Cladonia cristatella	COM		8	N/A
Cladonia ramulosa	FREQ		8	N/A
Lobaria pulmonaria	FREQ	tripartite with Cy component, OG; R3; Oligo	8	18
Parmeliopsis ambigua	UC	High elevation; boreal-arctic	8	0
Tuckermannopsis ciliaris	FREQ		8	4
Cetrelia olivetorum	FREQ	OG	8	4
Melanohalea olivacea	UC	Boreal	8	2
Tuckermannopsis americana	COM		8	33
Usnea hirta	COM		8	30
Candelariella concolor	COM	Eutro	8	23
Bryoria furcellata	COM		17	35
Myelochroa aurulenta	COM	OG	17	21
Phaeophyscia pusilloides	COM	Hardwood requirement	17	27
Physcia millegrana	COM	Roadside trees	17	36

continued

Table 5.—continued

Species*	Occurrence in New England	Species notes	Our plots %	New England %
Cladonia digitata	FREQ	Northern	25	N/A
Bryoria nadvornikiana	FREQ	Boreal	25	N/A
Cladonia caespiticia	FREQ	Southern and coastal Meso	25	N/A
Parmeliopsis capitata	FREQ	Northern Meso	25	N/A
Pyxine sorediata	FREQ	Mature forest	25	21
Melanohalea septentrionalis	FREQ	Northern Meso	25	20
Punctelia caseana	FREQ	Temperate	25	11
Hypogymnia incurvoides	UC	Recent segregate, 3 ME	42	0
Vulpicida pinastri	FREQ	Boreal	42	22
Cladonia floerkeana	FREQ		42	N/A
Melanohalea halei	COM	(Meso) for other species in this genus	42	N/A
Usnea filipendula	UC	(Oligo)	42	3
Melanelixia fuliginosa	FREQ	OG	42	5
Usnea subfloridana	FREQ	(Oligo), Rare in VT	42	34
Flavoparmelia caperata	COM		42	7
Parmeliopsis hyperopta	FREQ	Boreal	50	3
Cladonia macilenta	COM		50	N/A
Punctelia rudecta	COM		50	N/A
Phaeophyscia rubropulchra	COM		50	68
Hypogymnia tubulosa	FREQ	OG; boreal	67	4
Imshaugia aleruites	COM		67	21
Pseudevernia cladonia	UC	OG; G3 R3	67	0
Pseudevernia consocians	FREQ		67	0
Tuckermannopsis orbata	FREQ		67	20
Parmelia squarrosa	COM		67	47
Melanelixia subaurifera	COM	(Meso) (or Eutro, Sparrius 2007)	67	53
Hypogymnia krogiae	FREQ	Northern Meso	75	9
Platismatia glauca	COM	(Oligo), Northern Meso	75	11
Cladonia chlorophaea group	COM	(Oligo, Sparrius 2007)	75	N/A
Evernia mesomorpha	COM	(Oligo)	75	N/A
Cladonia squamosa	COM	(Oligo)	83	N/A
Parmelia saxatilis	FREQ	(Meso)	83	N/A
Hypogymnia physodes	COM	(Meso), toxin tolerant, or Oligo, Sparrius 2007)	83	83
Cladonia ochrochlora	COM		92	N/A
Cladonia coniocraea	COM	Northern Meso	92	N/A
Usnocetaria oakesiana	COM		100	34
Parmelia sulcata	COM	(Meso) (or Eutro, Sparrius 2007)	100	88

* In the Northeast, the cyanolichens are *Lobaria scrobiculata* and species within these genera: *Collema, Degelia, Fuscopannaria, Heterodermia, Leptogium, Nephroma, Pannaria, Parmeliella, Parmotrema, Peltigera, Protopannaria, Pseudocyphellaria*, or *Sticta*.

Epiphyte Community Data Related to Lichen Thallus Health and Air Quality

Across all 12 of the new plots, the NMS ordination found Wilderness Area species composition to vary strongly along an elevation ($r^2 = 0.770$) and percent hardwood tree ($r^2 = 0.456$) single gradient (Fig. 8). The first axis explained 91.5 percent of the variation between sites. For the first axis, lichen thallus health ($r^2 = 0.820$) and percent of conifers ($r^2 = 0.679$) were positively related and percent N ($r^2 = 0.274$) and Fe content ($r^2 = 0.255$) of the bulk samples were negatively related to the axis (Fig. 8). Lichen and bryophyte richness separated along the first axis with lichen richness greatest in the conifer-dominated sites and bryophyte richness greatest in the hardwood-dominated sites. A second axis related positively to epiphyte richness ($r^2 = 0.633$) and negatively to bulk sample values for Ni ($r^2 = 0.279$) and Cu ($r^2 = 0.263$) (Fig. 8).The second axis explained an additional 4.7 percent of variability in the data set (total of 96.2 percent of the variation explained). Correlations among plot variables included more big trees in stands with a higher percentage hardwood ($r^2 = 0.546$) (Table 2). The steeper sloped sites faced more south (northness $r^2 = 0.826$). Note that in this study, the percent S of the bulk samples was not related to differences in species composition of the sites. Bryophytes did not figure prominently in the NMS.

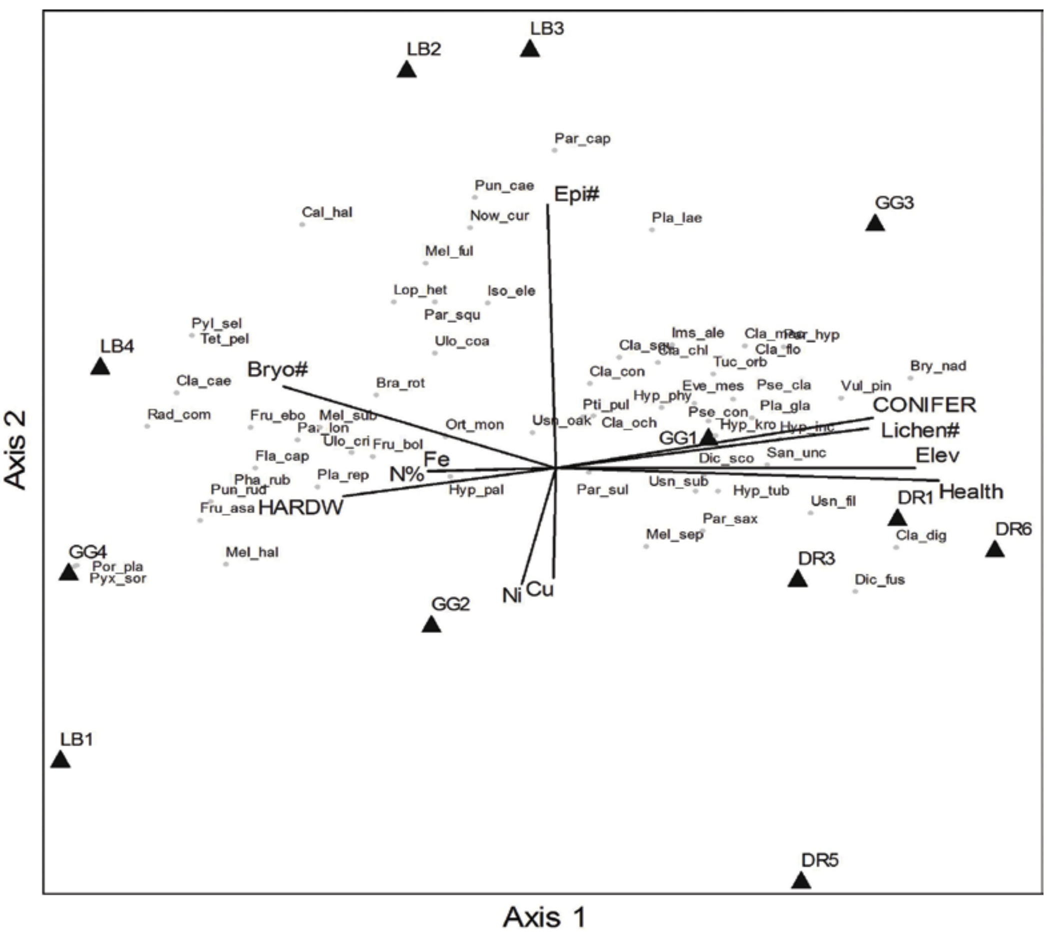
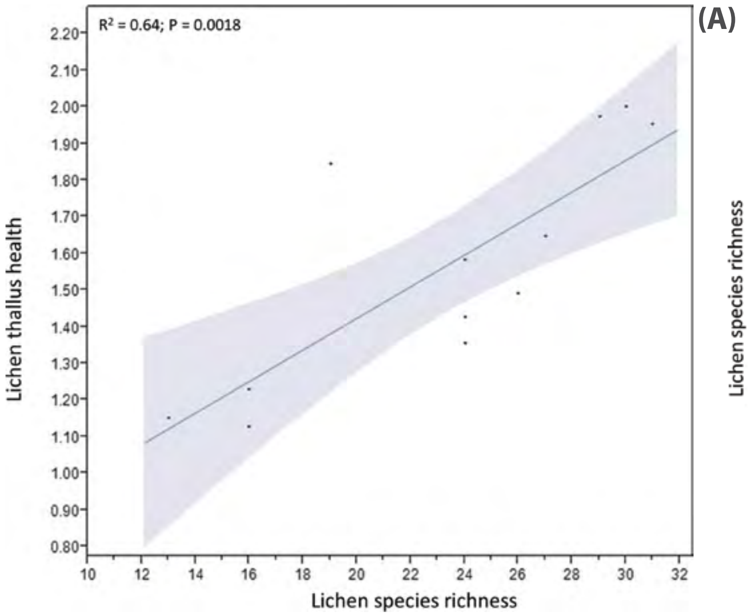


Figure 8.—Non-metric multi-dimensional scaling (NMS) bi-plot of epiphyte species composition and important site variables at 12 collection locations in three Wilderness Areas, with plot codes as in Fig. 1 and with four locations per Area. Epiphyte species are abbreviated by the first three letters of their genus and specific epithet (refer to Table 4). The site variables are in order of the strength of their relation to the ordination axes (r^2): Thallus health rating (0.820); elevation (0.770); conifer (% of tree tally, given in Table 2) (0.679); lichen spp. number (0.667); epiphyte spp. number (0.633); bryo spp. number (0.582); hardwood (% of tree tally, given in Table 2) (0.456); Ni (ppm of lichen dry weight) (0.279); N (% of lichen dry weight) (0.274); Cu (ppm of lichen dry weight) (0.263); and Fe (ppm of lichen dry weight) (0.255).



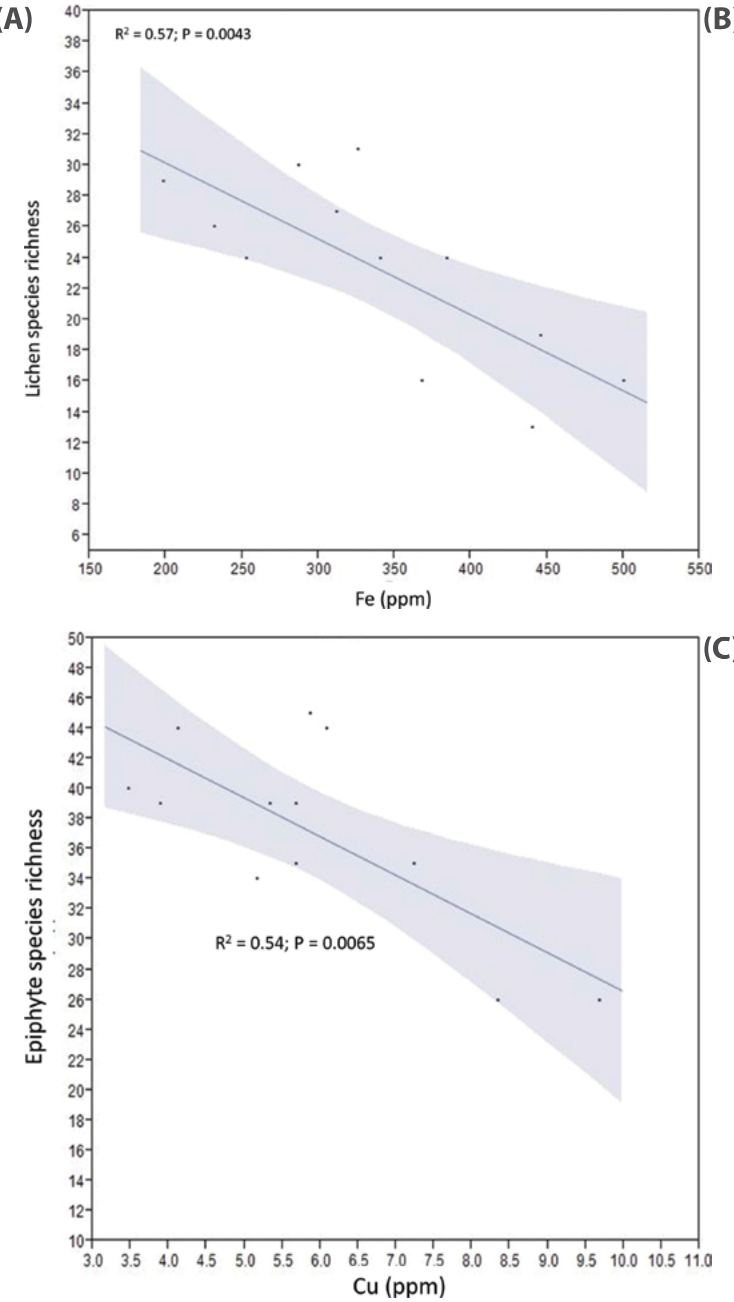


Figure 9.—Regressions between (A) lichen thallus health scores and lichen species richness; (B) lichen species richness and lichen thalli Fe (ppm of lichen dry weight); and (C) epiphyte species richness and lichen thalli Cu (ppm of lichen dry weight).

Relationships With Epiphyte Richness

We examined linear regression for compelling relationships suggested by the NMS bi-plot (Fig. 8). We found a significant positive relationship between health of lichen thalli and lichen species richness (Fig. 9a). Plots that deviated from the line of fit were DR5, which had better health than expected given the number of lichen species, and GG2, LB3 and LB4, which had poorer lichen health than expected given the richness of lichen epiphytes (Fig. 9a). Lichen richness was negatively related to Fe and Cu content of the lichen thalli collected for bulk chemistry (Fig. 9b, 9c). The content of Fe in lichen thalli co-varied strongly with Al ($r^2 = 0.79$; $p = 0.0001$).

Context of Regional Aerosol Data From 1992-2012

IMPROVE data, going back to 1992 at Lye Brook, indicate that there have been improvements in air quality based on decreases of 3 percent per year or more in aerosol SO_4^-, S, Se, Ni, V, Pb, and Zn, with smaller but significant improvements in NO_3^-, Fe, and Cu, and no significant changes in Al, and Cl^-. A comparison of 2001-2012 data from concurrent sample days at the Lye Brook and Great Gulf sites indicates that CL- and V were highest at Great Gulf, while Cu, Pb, Fe, Ti, Se, S, SO_4, and NO_3 were significantly higher at Lye Brook than Great Gulf. The most important influence upon lichens is probably the highly elevated NO_3 levels at Lye Brook, which were more than 50 percent higher than at Great Gulf. A large number of NOx-emitting sources, including urban motor vehicle traffic, industrial plants, and power-generation plants, operate upwind of New England in the region generally closer to Lye Brook than to Great Gulf. [Note: Supplemental tables available from R. Poirot, see appendix 4.]

DISCUSSION

Changes Over Time

A major reason to conduct monitoring is to detect change over time. Air quality improvements can lead to recovery of epiphytic macrolichens on tree boles (based especially on photographic evidence, Hultengren and Gralen 2004), but such an expectation requires care in siting and analyzing bulk samples during remeasurement efforts. Decrease in Pb in lichens at the level of National Forest (Fig. 3a) and on seven individual plots (Figs. 5a and b) indicates benefits to human health. This apparent trend does not explain whether improved air quality has led to improved health of lichens and bryophytes in the Class I Wilderness Areas. For some elements such as S and Pb, the bulk sample comparison clearly shows the expected decrease over time. However, for Al and Cu, sample comparison suggests increases. We think this is likely due to improved chemical extraction and analytical capabilities for those metals. The IMPROVE data we used as a reference confirmed that there have been no significant changes in these elements in the Wilderness Areas over time. There are also no indications of increasing trends of AL or Cu concentrations in any other IMPROVE or EPA Chemical Speciation Network (CSN) sites in the Northeast, neither are there other indications of regional increases in emissions of these metals.

Otherwise, based on lichen and bryophyte elemental analysis alone, it is not clear that the overall air quality influencing lichens in the three Class I Wilderness Areas we sampled is better or worse than it was when Wetmore sampled between 1988 and1994. Incongruity between our findings and those of Wetmore (1989, 1995a, 1995b) became apparent despite our care in remeasurement of a near spot on the ground where Wetmore had sampled and our use of the same analytical lab that Wetmore used. Choice of sample bags is an example of where such discrepancy could arise. We collected bulk chemistry lichens into Ziploc™ Freezer bags while Wetmore used spunbound olefin bags (Wetmore 1989). There may be other seemingly minor differences for which we did not account. Besides differences in methods, there are aspects beyond researcher control, including technological advances in ICP that have brought about more precision in detecting elements in the samples. Between our second and third batches of samples sent to ARL, the ICP instrument used there was updated, and this could have led to incongruity even within our own data set. Wetmore's samples are not available for reanalysis. Our bulk sample results probably represent at least some increases in element levels that cannot be ascribed solely to environmental change but could be due to methodology. While increases are difficult to assess in this situation, any decreases may be underrepresented due to improved precision of ICP instrumentation.

We detected a correlation between adverse factors on lichen health and mineral content. High Hg and Cu are particularly harmful to lichens (Blett et al. 2002). Plots with high levels of heavy metals, high total N and total S, and low species richness tended to also have poor condition of lichen thalli. At Lye Brook Wilderness especially, but also at our highest elevation plot at Great Gulf (GG3), we found correlation among high percent N and percent S, high levels of heavy metals (LB1 and LB4), and poor thallus condition. LB4 had the highest levels of Hg in *Hypogymnia physodes*, and thallus condition overall was poor. The vicinity of LB4 corresponds to a so-called mercury hotspot identified by Evers et al. (2007) at the Deerfield River in southern Vermont based on Hg levels in birds and mammals. GG3 had a high Hg level and lichen thallus condition was poor there also (Fig. 7a). GG3 may be influenced by a Hg hotspot in southern New Hampshire (Evers et al. 2007).

Maturity of forest stand and proportion of conifers to hardwoods were expected to have some influence on our findings. The majority of our 12 plots were in mature forest stands. Each plot had two or more old-growth indicator lichens present (Tables 4 and 5). Continuity of the stands may explain this, but other factors could include air quality, proportion of conifers to hardwoods, and elevation (Table 1). Selva (2014) concluded that there is a likelihood that air quality impacts the algal partner in calicioid lichens; such may apply to the macrolichens in this report also. Despite improvements in air quality over the past few decades, shown in the IMPROVE data, there could be a lag in lichen recovery. Subsequent monitoring at these plots may help uncover such a trend.

Cyanolichens are particularly interesting because of their multi-party symbioses, in which the blue-green alga is likely to be the most vulnerable. Lichen communities we measured lack the cyanolichen species (see footnote in Table 5) with a few minor exceptions; e.g., we found *Lobaria pulmonaria* on one of 12 plots. We expected to find *Leptogium cyanescens*, a cyanolichen that can be common on hardwoods (Cameron and Richardson 2006, Cleavitt et al. 2009). Lack of cyanolichens—a distinctive outcome of our results—could be explained in part by (1) dominance of conifers on many of our plots; (2) the FIA protocol that excludes bole area below 0.5 m where cyanolichens might be found due to accumulation of nutrients through stemflow, increased contact with soil, and a stable moisture environment; (3) lichens not being a part of the community in pre-anthropogenic air conditions, though there is no evidence for this; or (4) bark acidification. Based on our observations across New England, in plots where hardwoods were dominant, cyanolichens should have been present. Unlike S pollution, N deposition tends to have a buffering effect that raises the pH of bark and branches (e.g., Lewis 2012) and would allow some lichens otherwise impacted by acidification to persist. Although buffering of bark is widely thought to explain changing lichen distribution with air quality, Jovan et al. (2012) found multiple sources of N affect lichen distribution, and bark was not the prevailing source. In California and the Pacific Northwest (Washington, Oregon) the SO_2 effects received less notice as being influential upon lichen community composition compared to effects due to NO_2 and O_3 (Jovan 2008). There is potential that the long-term impacts of SO_4^{2-} deposition in the Northeast are high relative to the West. In this regard the Northeast could resemble Europe, though to a lesser extent.

Plots in the Class I Wilderness Areas varied in their lichen richness and heavy metal content. A gradient of air quality degradation was detectable in the lichen data and is consistent with IMPROVE summaries (where Presidential Range—Dry River is lumped with the Great Gulf Wilderness Area due to location of the IMPROVE data collection site associated with these two Wilderness Areas). Great Gulf and Presidential Range—Dry River are each capable of ranking at the upper end of the gradient depending on the aspect of lichen health or the element of interest, while Lye Brook consistently ranked low due to poor conditions listed here. Highlights were the lower lichen species richness and simultaneous highest levels of heavy metals in bulk samples for plots DR5, LB1, and LB4. Although eutrophs were fairly rare in this study, they were more likely to occur at the three sites with lower species richness: GG4, LB1, and LB4. Two of these sites, GG4 and LB1, also had lower lichen richness than expected based on the relationship to Fe content. Lye Brook has higher heavy metal content, higher N, lower species richness, and poor condition of lichen thalli. The gradient detected here is also similar to results of Evers et al. (2007) for Hg hotspots. Note that LB4, though low in lichen richness, had high bryophyte richness (Table 4, footnote). This is explained in part by deciduous trees, dominant at that plot, that support a diverse bryoflora.

Candellaria concolor, found at LB4, is one of the eutrophic lichens recognized by Fenn et al. (2007, expressed as “nitrophilic”) in the Pacific Northwest. They also found *Physcia adscendens* and *Xanthoria polycarpa* in nitrogen-enriched forests. While neither of those species were found in this project, we found *Physcia milligrana* at three plots and think it likely that additional species in the genus *Physcia* might respond as eutrophs also.

Does the deposition at Lye Brook approach the critical load for N in Northern forests, as derived by Pardo et al. (2011a) and consistent with Geiser et al. (2010)? Lichen community composition was not thoroughly explored for indications of detrimental impacts from N deposition, but it will be examined for another report. Most Northeast lichen species lack N indicator status, though we tried to derive this information from many sources (Table 5). Ambiguity can be found between N indicator ratings from one geographic region to another. For example, *Parmelia sulcata* was a common, abundant lichen across most of our plots (Table 4) and was useful for assessing lichen health. The species in Western Washington and Oregon was associated with “acidic, low fertility environments” (Geiser and Neitlich 2007); in The Netherlands, where decreasing ammonia deposition has led to a decrease in some lichens on oak bark, it was categorized as a eutroph (Sparrius 2007); and in Germany, the species increased its area on trees spray-fertilized biweekly for 21 months (Franzen-Reuter and Frahm 2007). *Parmelia sulcata* was not found at all in an environment impacted by high HNO_3 in the Los Angeles Basin, California (Jovan et al. 2012). *Hypogymnia physodes* and *Evernia prunastri,* both frequent in our study, were considered "sub-regional nitrophytic or tolerant species" by Geiser and Neitlich (2007), while Sparrius (2007) classified both as "acidophytes" (compare to oligotroph). In the Austrian Alps where eutrophication is of great concern and lichens are decreasing in diversity, abundance, and thallus size, *Hypogymnia physodes* behaves as a nitrophobe (Mayer et al. 2013; compare to acidophyte or oligotroph) but is not as sensitive to eutrophication as *Bryoria* and *Usnea*. Other examples can be found for common lichens. Variation in lichen response to pollutants such as found in this brief survey is a challenge in developing indicator status information that might be used to estimate critical load for N. Additional empirical studies such as that of Franzen-Reuter and Frahm (2007) could improve the broader reliability of such information.

Comparison to 92 Lichen Indicator Plots Across New England

Eight of the nine lichen species noted as rare in New England by Hinds and Hinds (2008), based on 92 FIA/FHM plots with P3 Lichen Indicator data, were also rare in the 12 new plots (Table 5). On the other hand, *Pseudevernia cladonia* was notably more frequent in conifer-dominated plots than expected. This may be due to mature stands with relative lack of disturbance and to high elevation. Thirteen indicators of old-growth forests were among lichens found in this survey (Table 5). Most eutroph species were rare (*Candelaria concolor* in one plot) or infrequent (21-50 percent of plots), e.g., *Phaeophyscia pusilloides* and *Physcia stellaris* (Table 5).

Context of Regional Aerosol Data From 1992-2013

IMPROVE data, going back to 1992 in summers at Lye Brook, indicated that there has been gradual improvement in most measures of air quality that affect regional haze, some of which are relevant to the lichen bulk chemistry results. First, the two Wilderness Areas (Presidential Range—Dry River is represented with Great Gulf in the IMPROVE data) are highly correlated with each other, likely because pollution sources are distant from the collecting stations, thus the readings for any given day are likely to be similar at Lye Brook and Great Gulf. Among minerals we measured, only Ni, V, and Cl^- tended to influence the IMPROVE sites on different days, were higher at Great Gulf, and are likely to result from residual oil combustion with sources concentrated along the Northeast urban corridor. Concentrations of Cu, Pb, Fe, Ti, Se, S, SO_4, and NO_3 were significantly higher at LYBR than at GRGU. Probably the most important influence upon lichens is the highly elevated NO_3 levels at Lye Brook. At 0.35 µg/m^3, NO_3 was more than 50 percent higher than at Great Gulf. Hundreds of industrial and power-generation plants operate upwind of Lye Brook, with airborne pollutants emanating from industrial areas near the Great Lakes and from coal burning power plants in the Ohio River Valley.

Relationships Between IMPROVE Data and Lichen Data

The data from nearby, remote IMPROVE sites are useful to compare with lichen data. The IMPROVE data indicate broad spatial patterns of pollutant exposures, while the lichens better capture the more complex spatial variability of actual deposition and biological uptake. Lichens are more widely available than IMPROVE data and can be used to fill in some gaps. These two types of data are similar in their associations with N, while they differ in that IMPROVE measures particles only and does not include potentially important gases that may contribute to dry deposition—such as SO_2, O_3, HNO_3, NH_4, and potentially toxic organics. While lichens have more variability associated with the complexity of the thalli (shape, folding, exposure history, etc.), lichen data we present here pointed to high elevation sites receiving more occult (cloud) deposition. Lichens in this report are associated with higher inputs of pollutants. While the lichens are not precise, they are useful indicators that offer finer-scale monitoring than do IMPROVE data.

Some additional limitations can be noted. The IMPROVE sites are intended to measure "regionally representative" atmospheric concentrations, but they are not co-located with our lichen plots, have limited applicability on the landscape, and may not capture the microscale variations in pollutant concentrations to which the lichens are exposed. IMPROVE metals analysis by x-ray fluorescence does not include all the metals analyzed by ICP AES in our lichen samples. A combination of both types of data appears to offer better understanding of air pollution than does just one type of data alone. Lichens were used with IMPROVE data to estimate critical loads by Glavich and Geiser (2008). See Cleavitt et al. (2015) for additional comparisons.

Some Take-home Messages

A lichen species included in 2011-2013 bulk samples analyzed for Hg was *Evernia mesomorpha*, and this consistently held more Hg than either of two mosses that were also analyzed for the element across National Forests (Fig. 2a) and a subset of plots (Fig. 6). Due to cost of sampling for Hg, it appears from these limited data that effectiveness will be increased if epiphytic, not ground layer, lichens and bryophytes are included in Hg surveys (see use of *Orthodicranum montanum* on oak boles in Pennsylvania; Davis et al. 2007). However, bryophytes would be difficult to clean if used as bulk samples. The high level of Hg in *Evernia mesomorpha* at GG3 is consistent with the expectation that a remote forest just below the upper slopes of Mt. Washington would have mercury contamination, as an elevation gradient for atmospheric Hg deposition is well established (e.g., Zhang et al. 2013). In our study, two other high elevation plots in WMNF were not sampled for Hg.

Blett et al. (2002) offered a list of benefits and limitations regarding use of lichens to monitor air quality (Table 1 in Blett et al. 2002). They recommend baseline sampling four to five times in the same area, at the same time of year, using the same lichen species. While we concur that this would enhance a monitoring effort, that intensity of effort was not feasible for this project.

Vouchering specimens not only enables taxonomic revisions and field training for those who will revisit a plot but may be valuable later for an unanticipated use in future research. Our assessment of lichen thallus condition was valuable in understanding air quality conditions that bulk chemistry made apparent.

We offer additional region-specific considerations for improving lichen and bryophyte biomonitoring of air quality in Class I Wilderness Areas of New Hampshire and Vermont:

1. **Locate lichen plots near IMPROVE sites.** The relationship between air quality impacts on lichens and aerosol data might be resolved if proximity to the IMPROVE collectors is taken into account. New lichen plots situated immediately near IMPROVE sites could be especially helpful. Also suggested: add passive NH_3 and HNO_3 samplers at some IMPROVE sites.
2. **Where FIA/FHM plots are used to assess air quality, include lichen thallus condition.** Even though a lichen is present, it might be in poor condition. Slow growth, malformations and necrosis could be due to pollution, and would relate to bulk chemistry results and measurable pollution loads. If thalli are small within the plot, then to reduce impacts on the plot, collections should be taken from off-plot for a subset of species only.
3. **Archive samples for future comparisons.** Technological advances are likely to continue to make comparison between historic and current conditions a mismatch, as we found here. This is because priorities change (e.g., Wetmore did not screen for N, but that is a high priority now), and because precision and reliability can be much greater in newer ICP instruments or potentially in a replacing technology. Monitoring air quality using bulk chemistry in forest organic matter should include the collection of sufficient volume to accommodate at least three measurements: (a) current, (b) near future (e.g., within a decade), and (c) distant future (e.g., two decades or more). The latter two (or more) sets of samples will need to be archived, so infrastructure for secure storage of such material needs to be planned from the outset.
4. **Conduct bulk chemistry analysis of tree bark samples.** At many of our plots, we had difficulty locating sufficient volume of common lichens to run elemental analyses. Given climate change and pollution impacts,

others may find this even more problematic as thalli decline in presence, size, and abundance. While we urge that future monitoring should continue to use the P3 lichen indicator to assess change in lichen communities, and that there is much potential for community analysis in estimation of critical loads, we suggest that bulk chemistry elemental analysis, including Hg, could be done on tree bark samples instead of lichens alone (or epiphytic bryophytes, as suggested above, but cleaning samples is problematic). This would potentially reduce the amount of lichen material to be collected, and bark is likely to continue to be readily available and abundant throughout forested plots. A vast literature on bark chemistry is already in place and will be useful in understanding how results from air quality monitoring applications fit with what is known elsewhere.

5. **Expand pollution tolerance ratings for northeastern lichens.** Several sets of descriptors have been developed to categorize lichen species, or genera, according to their tolerance to pollutants (e.g., Jovan 2008, Lewis 2012). Globally, bryophytes are not much categorized as to N indicator status but there appear to be prospects for developing such a system. For lichens, the northeastern United States has had less attention than some other parts of the world; N indicator status is unknown for many species we found (Table 5). We did not have sufficient sample size to assign pollution tolerance ratings to lichens or bryophytes in this study. To place our results in context of known tolerance ratings, we considered congenerics where species-specific ratings are not available, but given the variability in lichen response to N, that could be ineffective.

   Without this information, the potential for lichen communities to indicate air quality in the Northeast cannot be fully realized. Among common lichens, such patterns may be detected from surveys such as those cited here. For rare lichens, multiple factors probably contribute to their absence: they might always have been rare, their habitat could be altered (e.g., old forest), there may be poor dispersal, or they may have a low rate of reproduction. Pollution might contribute, but pollution alone may not be the factor that explains their absence. Although there is temptation to infer pollution tolerance from their absence, this would be a difficult hypothesis to test.

6. **Continue to test bryophytes as bioindicators.** While some of our data indicate only weak utility for bryophytes (e.g., low influence of bryophytes in the NMS, see Fig. 8), that might be explained in part by our bulk samples, which were of ground layer bryophytes and not epiphytic species. Three species were found at almost all our plots (*Hypnum pallescens, Orthodicranum montanum,* and *Ptilidium pulcherrimum*) and could be present in sufficient quantity that they can be collected for bulk chemistry from off-plot. Their diversity and the conservation of their habitats can be considered along with lichens in the same locales, as indeed they are habitat for and may influence the establishment of some lichen species on branches and boles.

CONCLUSION

While this monitoring project is a start toward integrating data from the Northwest with greater effort in the northeastern United States, there are unresolved areas for further exploration that we think will greatly enhance the ability of the IMPROVE monitoring program to strengthen what is learned from lichen monitoring, and vice versa. Yet to be developed are the pollution sensitivity ratings for lichens of the Northeast. Despite evidence from the IMPROVE data that air quality is better now than in the recent past, we cannot be sure from lichen data alone that there have been substantial changes in air quality since Wetmore's sampling, apart from our result for Pb. If there is to be a lag in lichen recovery, then evidence might be sought from lichen health data and increases in abundance and richness. We found a gradient of air quality conditions that can be followed using lichens (and bryophytes), with implications for human health, improving the environment, and cryptogam conservation. Finally, we identified that air quality at Lye Brook Wilderness Area is in need of improvement for the sake of people, lichens, and all their various habitats.

ACKNOWLEDGMENTS

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LITERATURE CITED

Allen, B. 2005. **Maine mosses: Sphagnaceae to Timmiaceae.** The New York Botanical Garden Press. 420 p.

Alvarez, R.; del Hoyo, A.; Garcia-Breijo, F.; Reig-Arminana, J.; del Campo, E.M.; Guera, A.; Barreno, E.; and Casano, L.M. 2012. **Different strategies to achieve Pb-tolerance by the two Trebouxia algae coexisting in the lichen *Ramalina farinacea*.** Journal of Plant Physiology. 169(18): 1797-1806.

Augusto, S.; Maguas, C.; Branquinho, C. 2013. **Guidelines for biomonitoring persistent organic pollutants (POPs) using lichens and aquatic mosses—a review.** Environmental Pollution. (180): 330-338.

Bennett, J.P.; Wetmore, C.M. 1997. **Chemical element concentrations in four lichens on a transect entering Voyageurs National Park.** Environmental and Experimental Botany. 37: 173-185.

Blett, T.; Geiser, L.H.; Porter, E. 2003. **Air pollution-related lichen monitoring in national parks, forests, and refuges: guidelines for studies intended for regulatory and management purposes.** Tech. Rep. NPS-D2292. Denver, CO: U.S. Department of the Interior, National Park Service, Air Resources Division and U.S. Fish and Wildlife Service, Air Quality Branch: Corvallis, Oregon: U.S. Department of Agriculture, Forest Service. 26 p.

Cameron, R.P.; Richardson, D.H.S. 2006. **Occurrence and abundance of epiphytic cyanolichens in protected areas of Nova Scotia, Canada.** Opuscula Philolichenum. 3: 4-14.

Clean Air Act of 1970, as amended August 1977. Public law 95-95. 42 U.S.C. s/s 7401 et seq., 58.

Cleavitt, N.L.; Dibble, A.C.; Werier, D. 2009. **Influence of tree composition upon epiphytic macrolichens and bryophytes in old forests of Acadia National Park, Maine.** The Bryologist. 112(3): 467-487.

Cleavitt, N.L.; Hinds, J.W.; Poirot, R.L.; Geiser, L.H.; Dibble, A.C.; Leon, B.; Perron, R.; Pardo, L.H. 2015. **Epiphytic macrolichen communities correspond to patterns of sulfur and nitrogen deposition in the Northeastern United States.** The Bryologist. 118(3): 304-324.

Davis, D.D.; McClenahen, J.R.; Hutnik, R.J. 2007. **Use of the moss *Dicranum montanum* to evaluate recent temporal trends of mercury accumulation in oak forests of Pennsylvania.** Northeastern Naturalist. 14: 27-34.

Esslinger, T.L. 2012. **A cumulative checklist for the lichen-forming, lichenicolous and allied fungi of the continental United States and Canada.** North Dakota State University. http://www.ndsu.edu/pubweb/~esslinge/chcklst/chcklst7.htm (accessed 13 December 2012).

Environmental Protection Agency. n.d. http://www.epa.gov/air/airtrends/aqtrends.html#comparison (accessed 27 May 2014).

Evers, D.C.; Y-J Han, C.T.; Driscoll, N.C.; Kamman, M.W.; Goodale, K.F.; Lambert, T.M.; Holsen, C.Y.; Chen, C.Y.; Clair, T.A.; Butler, T. 2007. **Biological mercury hotspots in the Northeastern United States and Southeastern Canada.** BioScience. 57(1): 29-43.

Fenn, M.E.; Geiser, L.H.; Bachman, R.; Blubaugh, T.J.; Bytnerowicz, A. 2007. **Atmospheric deposition inputs and effects on lichen chemistry and indicator species in the Columbia River Gorge, USA.** Environmental Pollution. 146: 77-91.

Franzen-Reuter, I.; Frahm, J-P. 2007. **Effects of nitrogen additions on epiphytic lichen and bryophyte cover on permanent observation plots in Rhineland-Palatinate, Germany. Herzogia.** 20: 61-75.

Geiser, L.H. 2004. **Manual for monitoring air quality using lichens on National Forests of the Pacific Northwest.** Portland, OR: U.S. Department of Agriculture, Forest Service, Pacific Northwest Region. http://gis.nacse.org/lichenair/doc/AQBPMthds.pdf (accessed 14 May 2014).

Geiser, L.H.; Neitlich, P.N. 2007. **Air pollution and climate gradients in Western Oregon and Washington indicated by epiphytic macrolichens.** Environmental Pollution. 145: 203-218.

Geiser, L.H.; Jovan, S.E.; Glavich, D.A.; Porter, M.K. 2010. **Lichen-based critical loads for atmospheric nitrogen deposition in Western Oregon and Washington Forests, USA.** Environmental Pollution. 158: 2412-2421.

Glavich, D.A.; Geiser, L.H. 2008. **Potential approaches to developing lichen-based critical loads and levels for nitrogen, sulfur and metal-containing atmospheric pollutants in North America.** The Bryologist. 111(4): 638-649.

Goward, T.; Arsenault, A. 2002. **Cyanolichen distribution in young unmanaged forests: a dripzone effect?** The Bryologist. 103 (1): 28-37.

Hinds, J.W.; Hinds, P.L. 2008. **Macrolichens of New England.** Bronx, NY: New York Botanical Garden.

Holland, E.A.; Braswell, B.H.; Sulzman, J.; Lamarque, J.-F. 2005. **Nitrogen deposition onto the United States and Europe: synthesis of observations and models.** Ecological Applications. 15: 38-57.

Hultengren, S.; Gralen, H. 2004. **Recovery of the epiphytic lichen flora following air quality improvement in southwest Sweden.** Water, Air, and Soil Pollution. 154: 203-211.

Jovan, S.E. 2008. **Lichen bioindication of biodiversity, air quality, and climate: baseline results from monitoring.** Gen.Tech. Rep. PNW-GTR-737. Portland, OR: U.S. Department of Agriculture, Forest Service, Pacific Northwest Research Station. 124.

Jovan, S.E.; Riddell, J.; Padgett, P.E.; Nash III, T.H. 2012. **Eutrophic lichens respond to multiple forms of N: implications for critical levels and critical loads research.** Ecological Applications. 22(7): 1910-1922.

Lendemer, J.C.; Hodkinson, B.P. 2010. **A new perspective on *Punctelia subrudecta* (Parmeliaceae) in North America: previously rejected morphological characters corroborate molecular phylogenetic evidence and provide insight into an old problem.** Lichenologist. 42: 405-421.

Lewis, J.E.J. 2012. **Bio-monitoring for atmospheric nitrogen pollution using epiphytic lichens and bryophytes.** Ph.D. Dissertation. University of Nottingham, UK. 320 p.

Likens, G.E.; Buso, D.C.; Butler, T.J. 2005. **Long-term relationships between SO_2 and NO_x emissions and SO_4^{2-} and NO_3^- concentration in bulk deposition at the Hubbard Brook Experimental Forest, NH.** Journal of Environmental Monitoring. 2005 October 7(10): 964-8.

Mayer, W.; Pfefferkorn-Dellali, V.; Türk, R.; Dullinger, S.; Mirtl, M.; Dirnböck, T. 2013. **Significant decrease in epiphytic lichen diversity in a remote area in the european Alps, Austria.** Basic and Applied Ecology. 14: 396-403.

McCune, B.; Geiser, L.H. 1997. **Macrolichens of the Pacific Northwest.** Corvallis, OR: Oregon State University Press. 386 p.

McCune, B.; Mefford, M.J. 1999. **PC-ORD. Multivariate analysis of ecological data, version 4.0.** Gleneden Beach, Oregon: MjM Software Design. 237 p.

McCune, B.; Dey, J.; Peck, J.; Cassell, D.; Heiman, K.; Will-Wolf, S.; Neitlich, P. 1997. **Repeatability of community data: species richness versus gradient scores in large-scale lichen studies.** Bryologist. 100: 40-46.

Nascimbene, J.; Tretiach, M.; Corana, F.; Lo Schiavo, F.; Kodnik, D.; Dainese, M.; Mannucci, B. 2014. **Patterns of traffic polycyclic aromatic hydrocarbon pollution in mountain areas can be revealed by lichen biomonitoring: a case study in the Dolomites (Eastern Italian Alps).** Science of the Total Environment. 475: 90-96.

Pardo, L.H.; Robin-Abbott, M.J.; Driscoll, C.T. 2011a. **Assessment of nitrogen deposition effects and empirical critical loads of nitrogen for ecoregions of the United States.** Gen. Tech. Rep. NRS-80. Newtown Square, PA: U.S. Department of Agriculture, Forest Service, Northern Research Station. 291 p.

Pardo, L.H.; Fenn, M.E.; Goodale, C.L.; Geiser, L.H.; Driscoll, C.T.; Allen, E.B.; Baron, J.S.; Bobbink, R.; Bowman, W.D.; Clark, C.M.; Emmett, B.; Gilliam, F.S.; Greaver, T.L.; Hall, S.J.; Lilleskov, E.A.; Liu, L.; Lynch, J.A.; Nadelhoffer, K.J.; Perakis, S.S.; Robin-Abbott, M.J.; Stoddard, J.L.; Weathers, K.C.; Dennis, R.L. 2011b. **Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States.** Ecological Applications. 21 (8): 3049-3082.

Selva, S.B. 2003. **Using calicioid lichens and fungi to assess ecological continuity in the Acadian Forest Ecoregion of the Canadian Maritimes.** The Forestry Chronicle. 79: 550-558.

Selva, S.B. 2013. **The calicioid lichens and fungi of the Acadian Forest Ecoregion of northeastern North America, I. New species and range extensions.** The Bryologist. 116(3): 248-256.

Selva, S.B. 2014a. **The calicioid lichens and fungi of the Acadian Forest ecoregion of northeastern North America, II. The rest of the story.** The Bryologist. 117(4): 336-367. BioOne.

Selva, S.B. 2014b. **Using calicioid lichens and fungi as biomonitors of pollution in five Class I Wilderness Areas in eastern North America.** Report to the U.S. Forest Service. 69 p. On file at White Mountain National Forest, 71 White Mountain Drive, Campton, NH 03223.

Sillett, S.C.; Antoine, M.E. 2004. **Lichens and bryophytes in forest canopies.** In: Lowman, Mary D.; Rinker, H. Bruce, eds. Forest canopies. London, UK: Academic Press. Chapter 8:151-174.

Sparrius, L.B. 2007. **Response of epiphytic lichen communities to decreasing ammonia air concentrations in a moderately polluted area of The Netherlands.** Environmental Pollution. 146: 375-379.

Tibell, L. 1992. **Crustose lichens as indicators of forest continuity in boreal coniferous forests.** Nordic Journal of Botany. 12: 427-450.

U.S. Department of Agriculture, Forest Service [USDA FS]. 2004. Forest Inventory and Analysis national core field guide, vol. 2. Field data collections procedures for phase 3 plots, ver. 2.0. Internal report on file with USDA Forest Service, Forest Inventory and Analysis, 1400 Independence Ave., Washington, D.C.

Wetmore, C.M. 1989. **Lichens and air quality in White Mountain National Forest Wilderness Areas.** Final Report for USDA Forest Service, Contract USDA 40-1484-7-614. http://gis.nacse.org/lichenair/doc/WhiteMt88.pdf (accessed 30 Mar 2016).

Wetmore, C.M. 1995a. **Lichens and air quality in Lye Brook Wilderness of the Green Mountain National Forest.** Final Report for USDA Forest Service, Contract 42-649. https://conservancy.umn.edu/handle/11299/158484 (accessed 30 Mar 2016).

Wetmore, C.M. 1995b. 1**993 Elemental analysis of lichens of the White Mountain National Forest Wilderness Areas.** Final Report for USDA Forest Service and Northeast Area State and Private Forestry, Forest Health Protection, Contract 42-649. http://conservancy.umn.edu/handle/11299/164285 (accessed 30 Mar 2015).

Will-Wolf, S.; Geiser, L.H.; Neitlich, P.N.; Reis, A.H. 2006. **Forest lichen communities and environment—How consistent are relationships across scales?** Journal of Vegetation Science. 17: 171-184.

Wetmore, C.M. 1995b. **1993 Elemental Analysis of Lichens of the White Mountain National Forest Wilderness Areas.** Final Report for USDA Forest Service and Northeast Area State and Private Forestry, Forest Health Protection. Contract 42-649.

Zhang, H.; Yin, R-S.; Feng, X-b; Sommar, J.; Anderson, C.W.N.; Sapkota, A.; Fu, X-w; Larssen, T. 2013. **Atmospheric mercury inputs in montane soils increase with elevation: evidence from mercury isotope signatures.** Scientific Reports. 3: art. 3322.

CONTRIBUTIONS OF THE AUTHORS

A.C. Dibble designed the project, led all aspects of writing this report, assisted in location of field sites, assisted with field work organization, collected lichens and bryophytes on all 12 plots, cleaned bulk samples, compiled data, helped develop the lichen thallus condition ratings, and took photographs.

J.L. Hinds examined lichen specimens and prepared final identifications for all lichen vouchers, helped develop and carried out lichen thallus condition ratings, helped develop the manuscript, and contributed to report revisions.

R.L. Perron conceived and initiated the project, led the proposal writing and field logistics, located and visited all 12 plots, led measurement of trees on plots, cleaned bulk samples, and helped with revisions on this report.

N.L. Cleavitt assisted in designing project, collected lichens and bryophytes at 7 of the 12 plots, prepared final identifications for all bryophyte specimens, oversaw and took part in most of the bulk specimen cleaning, helped develop the lichen thallus condition ratings, did most of the statistical analyses, wrote the initial results section, and contributed to report revisions.

L.H. Pardo contributed to project planning and oversight, and contributed to report revisions.

R.L. Poirot provided air quality data, provided insight on deposition data relevant to interpretation of our results, and contributed to report revisions.

L. Geiser contributed to project planning and to report revisions.

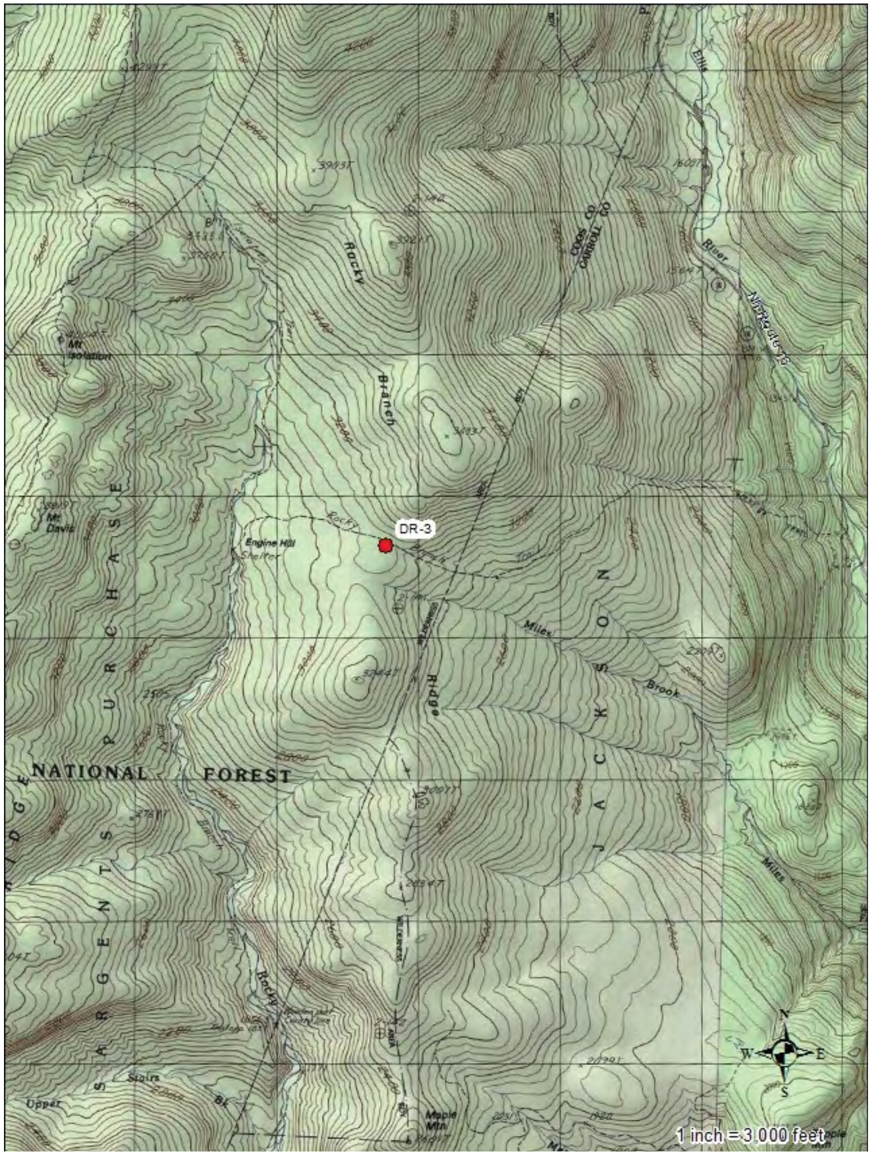
APPENDIX 1

Maps of study areas in White Mountain National Forest, NH, and Green Mountain National Forest, VT

Lichen plot locations at White Mountain National Forest, NH, by Christine Perron, used with permission. "GG" are plots in Great Gulf, and "DR" are those in the Presidential Range—Dry River Area.

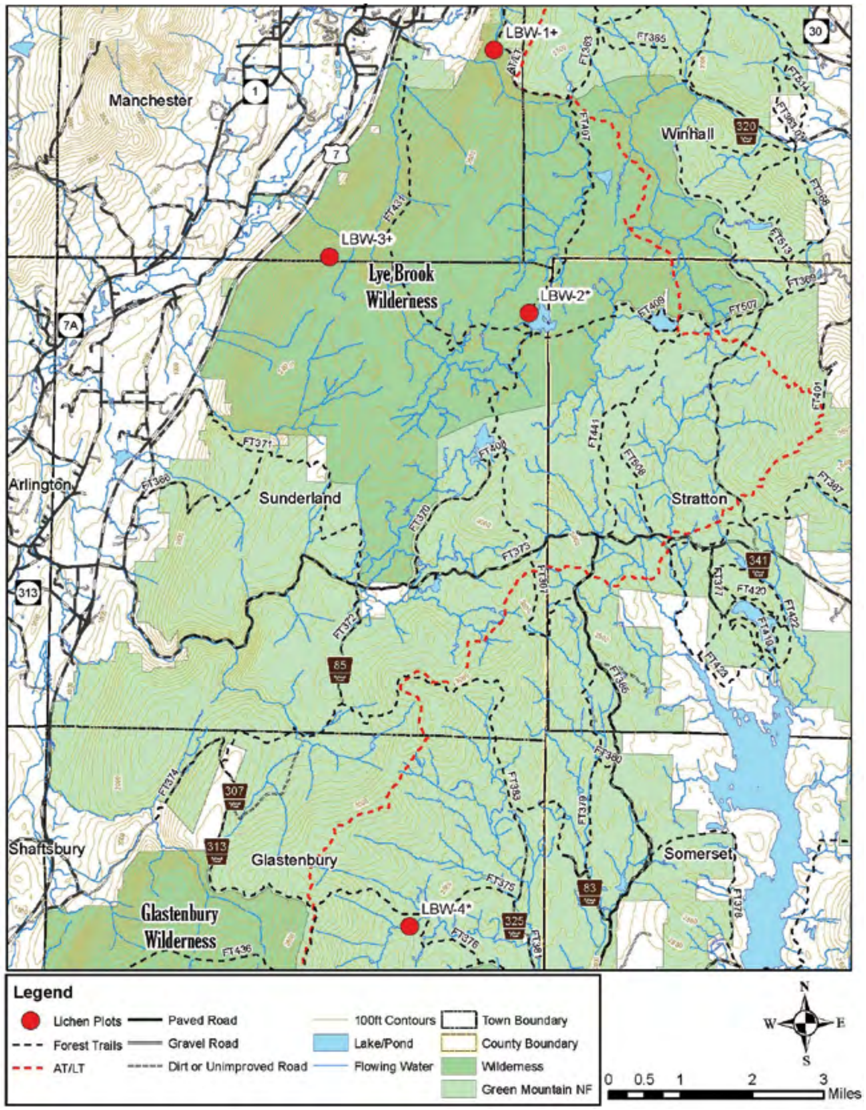
Lichen plot locations at Presidential Range—Dry River Area, NH, by Christine Perron, used with permission.

Lichen plot locations at Great Gulf Wilderness Area, NH, by Christine Perron, used with permission.

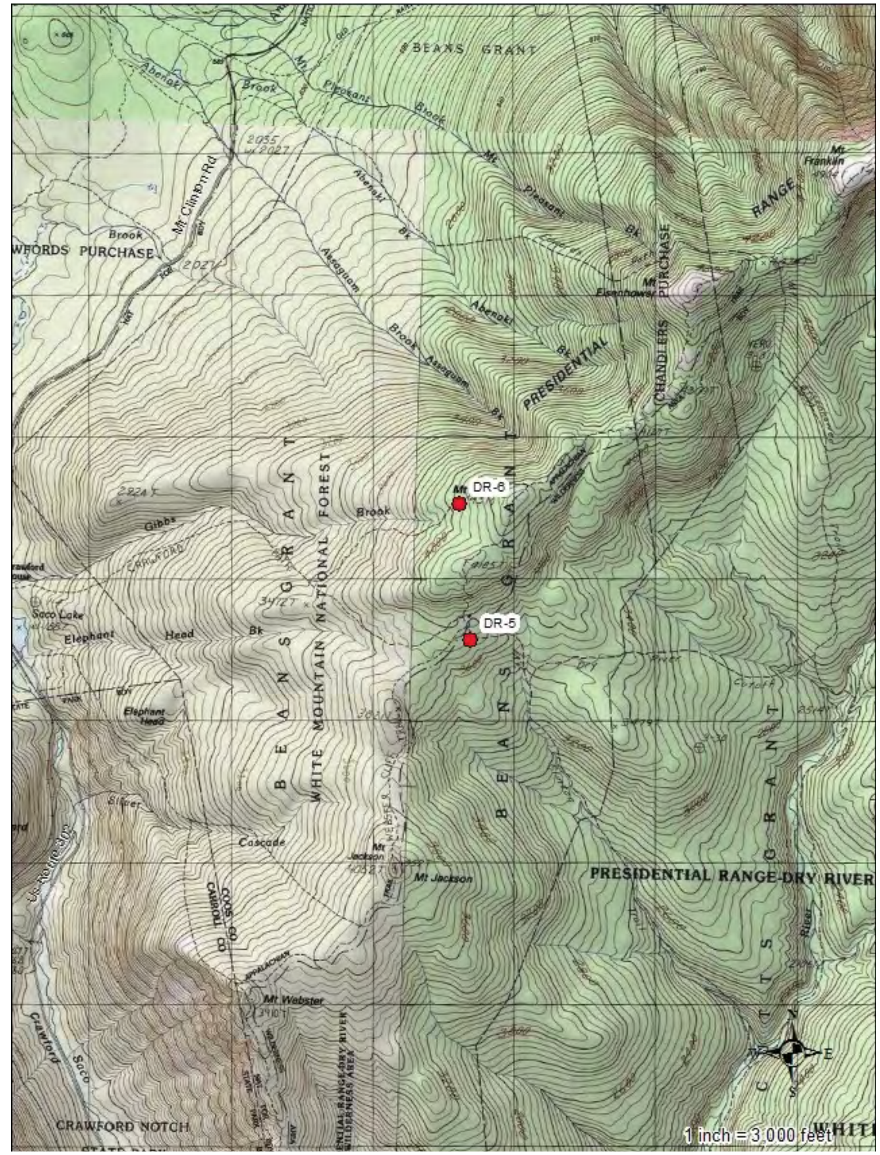


Presidential Range—Dry River Wilderness, detail for lichen plot DR-3, by Christine Perron, used with permission.

Presidential River—Dry Range Wilderness, detail for lichen plot DR-1, by Christine Perron, used with permission.



Locations of lichen plots lin Lye Brook Wilderness, Green Mountain National Forest, VT, by Thomas M. Tenyah, used with permission. "LBW" corresponds to plots labeled "LB" in the text.



Presidential Range—Dry River Wilderness, detail for lichen plots DR-5 and -6, by Christine Perron, used with permission.

APPENDIX 2

Multivariate Correlations

Multivariate correlations for S and N (as percent) plus 16 additional elements. There are 22 missing values. The correlations are estimated by Pairwise method.

	S percent	N percent	Al	B	Ca	Cd	Cr	Cu	Fe
S percent	1.0000	0.5087	-0.2307	0.1846	-0.1707	-0.2878	-0.0142	0.2471	-0.1480
N percent	0.5087	1.0000	-0.5753	-0.1968	-0.1063	-0.1224	0.1241	0.5590	-0.3921
Al	-0.2307	-0.5753	1.0000	0.1407	0.5739	0.4148	0.1083	-0.0630	0.9392
B	0.1846	-0.1968	0.1407	1.0000	0.0617	0.1121	-0.3169	0.1902	-0.0327
Ca	-0.1707	-0.1063	0.5739	0.0617	1.0000	0.7612	0.3117	0.4176	0.6526
Cd	-0.2878	-0.1224	0.4148	0.1121	0.7612	1.0000	0.2327	0.5377	0.4594
Cr	-0.0142	0.1241	0.1083	-0.3169	0.3117	0.2327	1.0000	0.1966	0.3327
Cu	0.2471	0.5590	-0.0630	0.1902	0.4176	0.5377	0.1966	1.0000	0.0639
Fe	-0.1480	-0.3921	0.9392	-0.0327	0.6526	0.4594	0.3327	0.0639	1.0000
K	0.2798	0.8363	-0.7520	-0.1764	-0.2005	-0.0980	-0.0899	0.5452	-0.6509
Mg	0.3000	0.8351	-0.5644	-0.1180	0.0599	0.1040	-0.0744	0.5587	-0.4443
Mn	-0.1199	0.2145	0.1920	0.2646	0.6606	0.6688	0.4059	0.6804	0.2816
Na	-0.2164	-0.1971	-0.0809	0.2525	-0.1408	-0.1311	-0.1001	-0.1366	-0.1608
Ni	0.0812	0.0593	0.1582	0.0206	0.1069	0.3703	0.6320	0.3761	0.3190
P	0.3939	0.8274	-0.6154	-0.0415	-0.0627	-0.0132	-0.1011	0.5656	-0.5233
Pb	-0.0785	-0.2437	0.6263	0.4439	0.6637	0.6174	0.0854	0.3980	0.6077
Zn	-0.2096	-0.0208	0.5092	0.1614	0.7907	0.7900	-0.0268	0.4492	0.5477
Hg	-0.2932	-0.6914	0.8308	-0.4727	-0.4674	-0.0487	0.2652	-0.6156	0.8503

Continued

	K	Mg	Mn	Na	Ni	P	Pb	Zn	Hg
S percent	0.2798	0.3000	-0.1199	-0.2164	0.0812	0.3939	-0.0785	-0.2096	-0.2932
N percent	0.8363	0.8351	0.2145	-0.1971	0.0593	0.8274	-0.2437	-0.0208	-0.6914
Al	-0.7520	-0.5644	0.1920	-0.0809	0.1582	-0.6154	0.6263	0.5092	0.8308
B	-0.1764	-0.1180	0.2646	0.2525	0.0206	-0.0415	0.4439	0.1614	-0.4727
Ca	-0.2005	0.0599	0.6606	-0.1408	0.1069	-0.0627	0.6637	0.7907	-0.4674
Cd	-0.0980	0.1040	0.6688	-0.1311	0.3703	-0.0132	0.6174	0.7900	-0.0487
Cr	-0.0899	-0.0744	0.4059	-0.1001	0.6320	-0.1011	0.0854	-0.0268	0.2652
Cu	0.5452	0.5587	0.6804	-0.1366	0.3761	0.5656	0.3980	0.4492	-0.6156
Fe	-0.6509	-0.4443	0.2816	-0.1608	0.3190	-0.5233	0.6077	0.5477	0.8503
K	1.0000	0.8673	0.1874	-0.0468	-0.1609	0.8543	-0.3220	-0.1255	-0.8813
Mg	0.8673	1.0000	0.2497	-0.0968	-0.1092	0.8529	-0.1630	0.1901	-0.7774
Mn	0.1874	0.2497	1.0000	-0.0631	0.2530	0.1532	0.6580	0.5378	-0.6235
Na	-0.0468	-0.0968	-0.0631	1.0000	-0.0907	-0.0625	-0.1497	-0.1203	-0.2202
Ni	-0.1609	-0.1092	0.2530	-0.0907	1.0000	-0.0913	0.1783	0.1233	0.2740
P	0.8543	0.8529	0.1532	-0.0625	-0.0913	1.0000	-0.1698	0.0454	-0.6727
Pb	-0.3220	-0.1630	0.6580	-0.1497	0.1783	-0.1698	1.0000	0.6309	0.6692
Zn	-0.1255	0.1901	0.5378	-0.1203	0.1233	0.0454	0.6309	1.0000	-0.2273
Hg	-0.8813	-0.7774	-0.6235	-0.2202	0.2740	-0.6727	0.6692	-0.2273	1.0000

APPENDIX 3

Project Photos

Plot GG-3, view to N from plot center, Aug. 16, 2013. Photo by A.C. Dibble, used with permission.

Plot GG-4, view toward plot center, Aug. 17, 2013. Photo by A.C. Dibble, used with permission.

PD-3, view to W from plot center, Aug. 13, 2012. Photo by A.C. Dibble, used with permission.

PD-5, view from plot center (below Mizpah Hut), Aug. 15, 2012. Photo by A.C. Dibble, used with permission.

PD-6, view from plot center, Aug. 16, 2013. Photo by A.C. Dibble, used with permission.

LB-3, hill W of Lye Brook, Aug. 21, 2013. Photo by A.C. Dibble, used with permission.

LB-4, view to S from plot center, above Downer Glen Ravine, Aug. 19, 2013. Photo by A.C. Dibble, used with permission.

Cladonia floerkeana, PD-6, near Crawford Path, Aug. 16, 2012. Photo by A.C. Dibble, used with permission.

Imshaugia aleurites, lichen of conifer forests, PD-1, Aug. 14, 2012. Photo by A.C. Dibble, used with permission.

Pseudevernia cladonia, ghost antler lichen, at PD-1, Aug. 14, 2012. Photo by A.C. Dibble, used with permission.

APPENDIX 4

Aerosol Data

Aerosol measurements from sites in the Interagency Monitoring of Protected Visual Environments (IMPROVE) network (Eldred et al. 1990, Hand et al. 2011) can provide an indication of relative concentrations of, and exposures to, various air pollutants in and around the Great Gulf National Wilderness (GRGU1 site) and the Lye Brook National Wilderness (LYBR1 and LYEB1 sites). The GRGU1 IMPROVE site (44.308-71.218, 454 m elev.) was established in 1995 and began year-round operation in 2001 (sampling in earlier years was summer only). The LYBR1 IMPROVE site (43.148-73.127, 1015 m elev.) was established in 1991 and began year-round operation in 1992. The LYEB1 site (42.953-72.910, 943 m elev.) was established in 2012, to replace the LYBR1 site, which was terminated in September 2012 due to recurring power supply problems.

IMPROVE aerosol sites collected 24-hour filter samples twice a week (Wednesdays and Saturdays) prior to 9/1/2000, after which the network switched to an every 3rd day schedule. Four samples are collected each sample day, including a coarse particle (PM_{10}) sample on a Teflon filter and three fine particle ($PM_{2.5}$) samples collected on Teflon, nylon, and quartz filters respectively. The PM_{10} and $PM_{2.5}$ Teflon filters are analyzed for mass (gravimetrically). The Teflon $PM_{2.5}$ filters are also analyzed for light absorption (by laser integrating sphere) and for elemental composition by proton induced x-ray emission (PIXE – used for lighter elements, Na through Mn, prior to 12/1/2001) and x-ray florescence (XRF – used for heavier elements prior to 12/1/2001 and for all elements after that date). Nylon $PM_{2.5}$ filters are analyzed for major inorganic ions (Cl^-, NO_3^-, and $SO_4^=$) by ion chromatography (IC), and quartz $PM_{2.5}$ filters are analyzed for elemental and organic carbon by thermal optical reflectance (TOR). IMPROVE aerosol data are stored in the Federal Land Manager Environmental Database (FED).

IMPROVE data for the GRGU1, LYBR1 and LYEB1 sites were obtained using the FED Data Query Wizard (http://views.cira.colostate.edu/fed/DataWizard/Default.aspx). The $PM_{2.5}$ elemental and ionic data are most relevant to interpreting the measurements of similar species in lichens and mosses. The 2001 to 2012 period with concurrent results from both sites can be used to compare "current" concentrations of pollutants between sites, while the 1992 to 2012 Lye Brook data are more appropriate for exploring changes over time. The raw IMPROVE XRF and IC data were adjusted by substituting values equal to half the detection limit for species where the attempted measurement was below the minimum detection limit. For the 2012 Lye Brook samples, data from the long-term LYBR1 site were used when available, and LYEB1 samples were used for days when LYBR1 data were missing. For comparing data between sites, the data were also constrained to include only sample dates where valid elemental and ionic samples were collected and analyzed at both GRGU1 and LYBR1 (or LYEB1) sites, resulting in 1,132 paired XRF and IC samples from both sites over the 2001 to 2012 period.

Comparative results for selected species are displayed in Table 6, including means and standard errors, with bold font indicating species and sites for which concentrations were significantly higher at one site than the other. Because the data for all species were highly skewed, a Wilcoxon Signed Rank test was applied to identify significant differences in species between the sites, and the significance levels (p) are reported. Table 6 also shows the percentage differences between the means and the correlations (R) between the sites for the paired samples for each species.

The daily concentrations are moderately well correlated ($0.50 < R < 0.90$) between the two sites for most species in Table 6, with the exceptions of Ni ($R= 0.34$), V ($R = 0.37$) and Cl^- ($R = 0.32$). This is consistent with the remote nature of these IMPROVE sites and suggests that regional concentrations of most species are influenced by relatively distant sources that are likely to influence both sites on the same sample days. Species more heavily influenced by nearby sources such as sea salt (Cl^-) and residual oil combustion (Ni and V from Northeast urban corridor power plants) would be more likely to influence the two IMPROVE sites on different days, and so tend to be less correlated between the sites.

Table 6.—Summary of selected $PM_{2.5}$ species for Lye Brook and Gt. Gulf IMPROVE Sites, 2001-2012. Means of species that were significantly higher at one site than the other are in bold. CI = confidence interval.

	Lye Brook 2001-2012	Great Gulf 2001-2012			
	Mean concentration (μg/m^3) ± 95% CI	Mean concentration (μg/m^3) ± 95% CI	p value (different means)	% difference in 12-year means (1-GG/LB)*100	LYBR:GRGU Correlation (R)
Cu	**0.00047 ± 1.55e-05**	0.00039 ± 1.13e-05	<0.001	18%	0.51
Pb	**0.00130 ± 3.68e-05**	0.00112 ± 2.96e-05	<0.001	14%	0.60
Zn	0.00339 ± 0.00011	0.00333 ± 0.00010	0.247	2%	0.52
Fe	**0.01633 ± 0.00076**	0.01427 ± 0.00062	<0.001	13%	0.81
Ti	**0.00174 ± 0.00014**	0.00163 ± 0.00012	0.004	6%	0.76
Al	0.01449 ± 0.00108	0.01331 ± 0.00093	0.055	8%	0.59
Al′	**0.02386 ± 0.00197**	0.02219 ± 0.00172	0.002	7%	0.88
Ni	0.00029 ± 1.68e-05	0.00034 ± 2.55e-05	0.967	-14%	0.34
V	0.00061 ± 3.52e-05	**0.00089 ± 7.48e-05**	<0.001	-44%	0.37
Se	**0.00039 ± 1.45e-05**	0.00026 ± 4.26e-06	<0.001	35%	0.80
S	**0.64151 ± 0.03429**	0.54885 ± 0.02608	<0.001	14%	0.84
$SO_4^=$	**1.79018 ± 0.10123**	1.54045 ± 0.07817	<0.001	14%	0.85
NO_3^-	**0.35000 ± 0.01925**	0.15392 ± 0.00842	<0.001	56%	0.63
Cl^-	0.02929 ± 0.00357	**0.03902 ± 0.00461**	<0.001	-33%	0.32

Concentrations of Cu, Pb, Fe, Ti, Se, S, $SO_4^=$, and NO_3^- were significantly higher at LYBR than at GRGU. No significant differences were observed for Zn and Ni. Concentrations of Cl- and V were significantly higher at GRGU than at LYBR. Ni was higher at GRGU, but not significantly so. The initial comparison for Al showed no significant difference between sites. However, Al had the largest fraction of below-detects of these reported species (45 percent were below MDL, while more than 75 percent or more of samples were above MDL for all other species in Table 6). A problem has also been identified with miscalculated MDLs in historical IMPROVE Al data (Poirot et al. 2001, White 2006). To eliminate the possible influence of MDL errors on the Al comparison, a second test (Al′) was run on a much smaller subset of sample days (n = 471) when Al was above MDL at both sites. The above-MDL Al′ data were slightly but significantly higher at LYBR, and the inter-site Al correlation (R) increased from 0.59 when below-detects were included to 0.88 when only paired above-detects were considered.

The somewhat higher concentrations at LYBR than GRGU for most species are consistent with influence from relatively distant sources to the West and Southwest of both sites, but closer to LYBR – for example industrial sources along the Great Lakes and coal burning power plants along the Ohio River Valley. NO_3^- (56 percent higher at LYBR) and Se (35 percent higher at LYBR) were the species that showed the greatest increases in mean concentrations at LYBR compared to GRGU. Fine particle nitrate concentrations at remote IMPROVE sites are predominantly in the form of ammonium nitrate, formed from atmospheric reactions of nitric acid and ammonia. Previous studies have implicated ammonia emissions from midwestern agricultural regions as an important source region for aerosol nitrate concentrations at IMPROVE sites throughout the eastern United States (Pitchford et al. 2009, Poirot 2009). This distant westerly regional origin and relative absence of sources of either ammonia or nitric acid between LYBR and GRGU are consistent with the much higher LYBR NO_3^- concentrations. Se has been demonstrated to be a good tracer for midwestern coal combustion at IMPROVE sites throughout the eastern United States (Hopke et al. 2006, Poirot 2009), and this relatively distant southwesterly origin and absence of coal sources between LYBR and GRGU is consistent with higher Se concentrations at LYBR. The higher concentrations of Cl^-, V, and Ni at GRGU and the lower inter-site correlations for these species are consistent with influences from relatively nearby emissions in closer proximity to GRGU, including marine sea salt (Cl^-) and northeast urban corridor residual oil combustion (V and Ni).

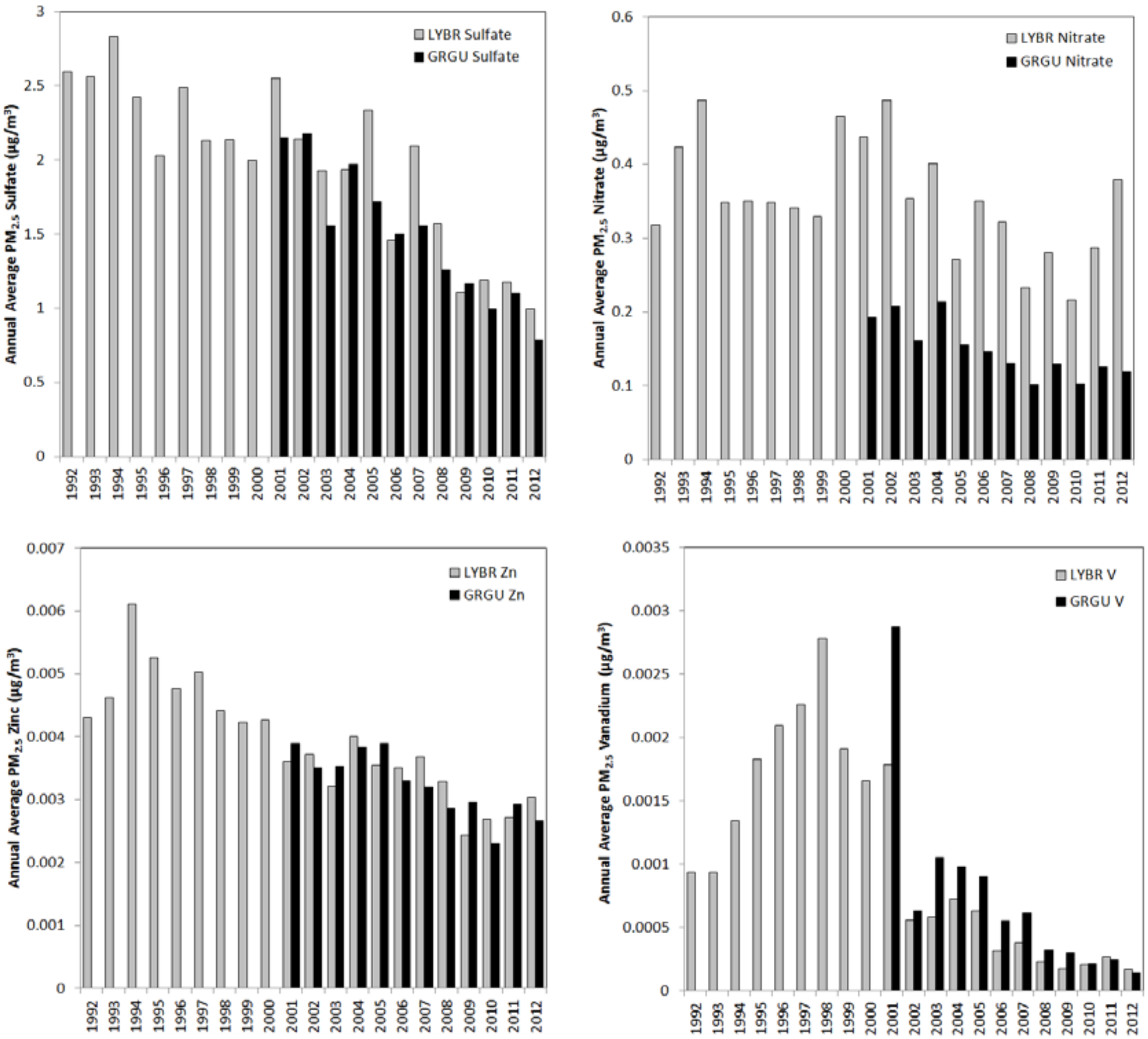


Figure 10.—Annual average concentrations of selected aerosol species at the Lye Brook and Gt. Gulf IMPROVE Sites.

Longer-term (1992-2012) trends in annual average concentrations of selected species at LYBR are plotted in Figure 10, along with more recent 2001-2012 data from GRGU. All of these species exhibit downward trends in the longer-term LYBR data. Sulfate and nitrate concentrations were highest in LYBR in most years, with nitrate showing weakest long-term trend but the largest between-site difference. Downward trends are also evident for Zn (no significant between-site difference) and V (steep downward trend and highest at GRGU.

To compare trends among species and characterize rates of change over time, the 1992-2012 annual average concentrations ay LYBR were "normalized" by dividing annual average concentrations by the 1992 concentrations of that species, and expressing each year's concentration as a percent of 1992 concentrations. A linear trend line was fit to the normalized annual concentrations, with the resulting slope expressed as "percentage change per year." The correlations, slopes (if significant), F values, and significance levels of the regressions of average annual aerosol species concentrations vs. time are summarized in Table 7. Elemental Ti, included in the Table 6 comparison between sites for 2001-2012, is not included here, as a significant positive bias has been noted in the IMPRPOVE (PIXE) Ti measurements prior to January 1, 2001.

No significant trends were indicated for Al and Cl^-, as might be expected for species resulting primarily from natural sources, such as wind-blown soil (Al) and sea salt (Cl^-). Limiting the Al data to only above-detects resulted in a similarly insignificant change over time. All other species summarized in Table 7 show significant improvements averaging at least 2.5 percent/yr, with the largest improvements for Pb (-3.8 percent/yr), Ni (-4.1 percent/yr), Se (-5.1 percent/yr), and V (-6.7 percent/yr).

Table 7.—Trends in annual means of selected $PM_{2.5}$ species at Lye Brook, 1992-2012

$PM_{2.5}$ Species	Correlation vs. date (R^2)	Slope (% / yr)	Regression F	Significance of F
Cu	0.54	-2.9	21.9	<0.001
Pb	0.62	-3.8	30.8	<0.001
Zn	0.73	-3.1	52.6	<0.001
Fe	0.45	-2.6	15.3	<0.001
Al	0.00	NS	0.02	0.886
Ni	0.56	-4.1	24.4	<0.001
V	0.61	-6.7	29.5	<0.001
Se	0.33	-5.1	9.2	0.007
S	0.71	-3	47.5	<0.001
SO4=	0.84	-3.2	100.5	<0.001
NO3-	0.64	-2.5	13.2	0.002
Cl-	0.02	NS	0.3	0.588

The 1992-2012 reductions in Pb occured after the phase-out of leaded gasoline had been completed and are consistent with more recent controls on various other Pb source categories, including mining, smelting, and metal working activities (U.S. EPA 2013). Reductions in Se, Ni, and V are consistent with emission controls on coal and oil-fired power plants resulting from the acid rain provisions of the 1990 Clean Air Act Amendments, EPA's Clean Air Interstate Rule, and other regional emissions control programs.

LITERATURE CITED

Eldred, R.A.; Cahill, T.A.; Wilkinson, L.K.; Feeney, P.J.; Chow, J.C.; Malm, W.C. 1990. **Measurement of fine particles and their chemical components in the NPS/IMPROVE Networks**. In: Mathai, C.V., ed. Transactions: Visibility and Fine Particles. A&WMA. Pittsburgh, PA.

Hand, J.L.; Copeland, S.A.; Day, D.E.; Dillner, A.M.; Indresand, H.; Malm, W.C.; McDade, C.E.; Moore, C.T.; Pitchford, M.L.; Schichtel, B.A.; Watson, J.G. 2011. **Spatial and seasonal patterns and temporal variability of haze and its constituents in the United States: IMPROVE Report V**. http://vista.cira.colostate.edu/improve/Publications/Reports/2011/2011.htm.

Hopke, P.K.; Zhou, L.; Poirot, R.L. 2005. **Reconciling trajectory ensemble receptor model results with emissions.** Environmental Science and Technology. 39: 7980-7983.

Pitchford, M.; White, W.; Poirot, R.; Schichtel, B.; Malm, W. 2009. **Characterization of the winter midwestern particulate nitrate bulge.** Journal of Air & Waste Management Association. 59: 1061-1069.

Poirot, R.L.; Wishinski, P.R.; Hopke, P.K.; Polissar, A.V. 2001. **Comparative application of multiple receptor methods to identify aerosol sources in northern Vermont.** Environmental Science & Technology. 35: 4622-4636.

Poirot, R.L. **Use of trace metals as source fingerprints.** In: Legge, A.H., ed. Air quality and ecological impacts: relating sources to effects. 2009. Oxford, UK: Elsevier Ltd.

US EPA. 2013. **Integrated science assessment for lead.** EPA/600/R-10/075F.

White, W.H. 2006. **Elemental concentrations above the MDL can go undetected.** IMPROVE Data Advisory, Doc #: da0010.

White, W.H. 2008. **Positive interference in PIXE titanium determinations.** IMPROVE Data Advisory, Doc #: da0018.

Dibble, Alison C.; Hinds, James W.; Perron, Ralph; Cleavitt, Natalie; Poirot, Richard L.; Pardo, Linda H. 2016. **Monitoring air quality in class I wilderness areas of the northeastern United States using lichens and bryophytes.** Gen. Tech. Rep. NRS-165. Newtown Square, PA: U.S. Department of Agriculture, Forest Service, Northern Research Station. 44 p.

To address a need for air quality and lichen monitoring information for the Northeast, we compared bulk chemistry data from 2011-2013 to baseline surveys from 1988 and 1993 in three Class I Wilderness areas of New Hampshire and Vermont. Plots were within the White Mountain National Forest (Presidential Range—Dry River Wilderness and Great Gulf Wilderness, New Hampshire) and the Green Mountain National Forest (Lye Brook Wilderness, Vermont). We sampled epiphyte communities and found 58 macrolichen species and 55 bryophyte species. We also analyzed bulk samples for total N, total S, and 27 additional elements. We detected a decrease in Pb at the level of the National Forest and in a subset of plots. Low lichen richness and poor thallus condition at Lye Brook corresponded to higher N and S levels at these sites. Lichen thallus condition was best where lichen species richness was also high. Highest Hg content, from a limited subset, was on the east slope of Mt. Washington near the head of Great Gulf. Most dominant lichens in good condition were associated with conifer boles or acidic substrates. The status regarding N and S tolerance for many lichens in the northeastern United States is not clear, so the influence of N pollution on community data cannot be fully assessed. Continued monitoring of lichens and bryophytes, especially if integrated with IMPROVE aerosol data, may reveal changes in air quality, climatic conditions, and other potential stressors or stimuli. Lichen health was impacted by low air quality at some of our sites.

KEY WORDS: lichens, bryophytes, air quality, monitoring, lead, mercury, nitrogen, thallus condition, IMPROVE, National Forest, Wilderness Area

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