

Eutrophic lichens respond to multiple forms of N: implications for critical levels and critical loads research

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Abstract. Epiphytic lichen communities are highly sensitive to excess nitrogen (N), which causes the replacement of native floras by N-tolerant, “weedy” eutrophic species. This shift is commonly used as the indicator of ecosystem “harm” in studies developing empirical critical levels (CLE) for ammonia (NH₃) and critical loads (CLO) for N. To be most effective, empirical CLE and/or CLO must firmly link lichen response to causal pollutant(s), which is difficult to accomplish in field studies in part because the high cost of N measurements limits their use. For this case study we synthesized an unprecedented array of atmospheric N measurements across 22 long-term monitoring sites in the Los Angeles Basin, California, USA: gas concentrations of NH₃, nitric acid (HNO₃), nitrogen dioxide, and ozone ($n = 10$ sites); N deposition in throughfall ($n = 8$ sites); modeled estimates of eight different forms of N ($n = 22$ sites); and nitrate deposition accumulated on oak twigs ($n = 22$ sites). We sampled lichens on black oak (*Quercus kelloggii* Newb.), and scored plots using two indices of eutroph (N tolerant species) abundance to characterize the community-level response to N. Our results contradict two common assertions about the lichen-N response: (1) that eutrophs respond specifically to NH₃ and (2) that the response necessarily depends upon the increased pH of lichen substrates. Eutroph abundance related significantly but weakly to NH₃ ($r^2 = 0.48$). Total N deposition as measured in canopy throughfall was by far the best predictor of eutroph abundance ($r^2 = 0.94$), indicating that eutrophs respond to multiple forms of N. Most N variables had significant correlations to eutroph abundance ($r^2 = 0.36$ – 0.62) as well as to each other ($r^2 = 0.61$ – 0.98), demonstrating the risk of mistaken causality in CLE/CLO field studies that lack sufficient calibration data. Our data furthermore suggest that eutroph abundance is primarily driven by N inputs, not substrate pH, at least at the high-pH values found in the basin (4.8–6.1). Eutroph abundance correlated negatively with trunk bark pH ($r^2 = 0.43$), exactly the opposite of virtually all previous studies of eutroph behavior. This correlation probably results because HNO₃ dominates N deposition in our study region.

Key words: ammonia; bark pH; bioindicators; critical levels of N gas concentrations; critical loads of N deposition; eutrophic lichens; lichens; nitric acid; nitrogen; nitrophytes; southern California, USA; throughfall deposition.

INTRODUCTION

A large body of research has emerged over the past 10 years demonstrating the high sensitivity of epiphytic lichen communities to nitrogen (N) pollution (e.g., van Herk 1999, 2001, Jovan and McCune 2005, Mitchell et al. 2005, Sutton et al. 2009, Vilsholm et al. 2009, Gadson et al. 2010, Geiser et al. 2010). Epiphytic lichens (i.e., growing on woody plants) subsist almost exclusively on atmospheric moisture and nutrients, making them

highly responsive to changes in air quality. Lichens possess an array of chemical, morphological, and physiological adaptations that make species differentially sensitive to N.

Studies of N impacts to lichens of Mediterranean, temperate, and boreal forest ecosystems have shown a remarkably consistent pattern: increasing N inputs cause significant losses of N-sensitive, oligotrophic lichen species, while populations of N-tolerant, eutrophic species increase. At higher N levels the more moderately N-tolerant mesotrophic species are lost as well. From an ecological perspective, this shift entails a major loss of diversity, losses in species utilized by wildlife, and increases in “weedy,” fast-growing lichens of unknown ecological importance.

Researchers have developed a variety of response indices to characterize eutroph abundance within a lichen community (Table 1). These indices increasingly

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TABLE 1. Review of eutroph indices that are calibrated against at least one type of nitrogen estimate or measurement.

Author	Index and classification system†	Site score‡	Calibration data§	Location
Frati et al. (2007)	ILD _n ; NM	summed frequency of nitrophyte species on oak trunks	NH ₃ , pH, bark NH ₄ ⁺	pig stock farm, central Italy
Frati et al. (2007)	ILD _{sn} ; NM	summed frequency of strictly nitrophyte species on oak trunks	NH ₃ , pH, bark NH ₄ ⁺	pig stock farm, central Italy
Wolseley et al. (2009)	L _{an} ; vH	average acidophyle count – average nitrophyte count on tree twigs	NH ₃ , pH	UK-wide
Wolseley et al. (2009)	L _{an} ; vH	average acidophyle frequency – average nitrophyte frequency on tree trunks	NH ₃ , pH	UK-wide
Pinho et al. (2008)	LDV; NM	sum of average nitrophyte frequency from each cardinal direction on oak trunks	NO ₂	1500 km ² , southwest coastal Portugal
Pinho et al. (2011)	LDV _{nitro} ; NM	average sum of nitrophyte frequency on one oak trunk	NH ₃	stock farm, central Portugal
Sparrius (2007)	NIW; vH	same as van Herk 2001?	NH ₃	~150 km ² , southeast Friesland, north The Netherlands
van Herk 1999	NIW; vH	average diversity of nitrophyte on one oak trunk	NH ₃ , NH ₄ ⁺ , SO ₂	about half of The Netherlands
Wolseley et al. (2006)	NIW; vH	average sum of nitrophyte ACs on one oak trunk	NH ₃ , pH	Thetford (63 km ²) and North Wyke (1.5 km ²), UK
Wolseley et al. (2006)	NIW; vH	average sum of nitrophyte ACs on one oak twig	NH ₃ , pH	Thetford (63 km ²) and North Wyke (1.5 km ²), UK
van Herk (2001)	NIW; vH	average sum of nitrophyte ACs on one oak trunk	NH ₃ , pH, NH ₄ ⁺ , SO ₂ , and bark SO ₄ ²⁻ , NO ₃ ⁻ , NH ₄ ⁺	about half of The Netherlands
Gadson et al. (2010)	NIW; vH	average sum of nitrophyte ACs on one oak branch	NH ₃ , pH, NO ₂	Epping Forest (25 km ²), greater London, UK
Geiser et al. (2010)	ordination index; MG	ordination index; highly correlated with eutroph diversity/total diversity on woody substrates	bulk N dep., bulk wet N dep., N in PM _{2.5} aerosols	west of the Cascades crest, Oregon and Washington, USA
Gadson et al. (2010)	PCN; vH	percent cover in nitrophyte on oak branches	NH ₃ , pH, NO ₂	25 km ² , greater London, UK
Jovan and McCune (2005)	PNA; vH	sum of nitrophyte ACs/ sum of all lichen ACs on woody substrates	NH ₃ , NO ₂ , SO ₂ , O ₃	greater Central Valley, California, USA
Fenn et al. (2008)	PNA; vH, J	sum of nitrophyte ACs/ sum of all lichen ACs on woody substrates	N dep. in throughfall	Sierra Nevada Mountain Range, California, USA

Note: The terms eutroph and nitrophyte (nitro.) are synonymous.

† Abbreviations for index names: ILD_n, index of lichen diversity using nitrophytes; ILD_{sn}, index of lichen diversity using strict nitrophytes; L_{an}, lichen atmospheric nitrogen; LDV, lichen diversity value; LDV_{nitro}, lichen diversity value – nitrophytes; NIW, Nitrofiel Indicatie Waarde; PCN, percent cover in nitrophytes; PNA, percentage nitrophyte abundance. Abbreviations for species classification systems: NM, Nimis and Martellos (2008); vH, van Herk (1999, 2001); MG, McCune and Geiser (2009); J, Jovan (2008).

‡ AC stands for abundance code.

§ All compounds are measured air concentrations unless noted otherwise. Parameters with a statistically significant correlation ($P < 0.05$) to the response index are in bold; pH is the measured pH of bark; “dep.” means deposition; “PM_{2.5}” means fine particulates $\leq 2.5 \mu\text{m}$ in size. Modeled data are indicated in italics, while partially modeled data sets are underlined.

influence air quality policy through their use in determining empirical critical levels (CLE) and critical loads (CLO) for N (Cape et al. 2009, Sutton et al. 2009, Bobbink et al. 2010, Fenn et al. 2010, Geiser et al. 2010, Pardo et al. 2011). A CLE or CLO is “a quantitative

estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt 1988). A CLE refers to a gas concentration while a CLO refers to

a depositional load. In the context of lichen-N bioindication, "harm" is commonly defined as a shift to eutroph dominance. Policy makers use CLE and/or CLO as guides for setting emissions limits and target pollutant loads that are expected to preserve ecological function.

Nearly all eutroph indices are adaptations of the indicator-species classification systems developed by van Herk (1999) or Nimis (Nimis and Martellos 2008; Table 1). The former classifies lichen species into acidophytes (syn. oligotrophs; N-sensitive and requiring low substrate pH) and nitrophytes (syn. Eutrophs; "N-loving" and requiring a high-pH substrate) while the Nimis system ranks species on an "eutrophication" scale of 1–5. These systems are similar as to which species are designated "eutrophic." Likewise, response indices based on these systems use similar methodologies for calculating site-level scores (Table 1).

Despite many shared characteristics, eutroph indices exhibit a surprising variety of correlations to different N air pollutants (Table 1). In most cases eutroph indices correlate with NH_3 and the corresponding increase in bark pH caused by NH_3 deposition. The European long-term CLE for NH_3 was lowered from 3 to 1 $\mu\text{g}/\text{m}^3$ (average annual concentration) for lichen and bryophyte-rich ecosystems based in part on these eutroph indices, considered alone and in tandem with oligotroph indices to demonstrate a community shift into eutroph dominance (Cape et al. 2009, Sutton et al. 2009b). While a minority, similar eutroph indices have been convincingly correlated with nitrogen oxides (NO_x), measures of N in particulate aerosols, and N deposition, in the latter case leading to N CLOs for temperate and Mediterranean forest ecosystems (Pardo et al. 2011).

It is hard to reconcile what exactly eutrophs respond to because each study uses a limited diversity of calibration measurements (Table 1), understandable because these measurements are expensive and the required instrumentation and field effort varies widely depending on the pollutant. For instance, only a couple studies mix measurements of reduced N (NH_x) and NO_x (Jovan and McCune 2005, Gadson et al. 2010) and their results conflict (Table 1). Also no studies mix measurements of gases and their deposition products. To be maximally effective at protecting sensitive biota, empirical CLE/CLO must be firmly linked to causal agent(s). Eutroph indices are also used in forest health monitoring activities to provide localized feedback on the success of pollution-abatement strategies, therefore making accurate calibration critical.

The purpose of our present case study was to simultaneously compare eutroph response to all N pollutants used in past studies (Table 1), including measurements of bark pH and potentially confounding environmental variables such as precipitation and temperature. Our main objectives were to: (1) evaluate the widespread assumption that eutrophs respond specifically to NH_3 , and (2) evaluate the assumption

that this response is fully or partly mediated by a rise in bark pH. We used two eutroph indices, one conventional and one derived using nonmetric multidimensional scaling (NMS) ordination, a gradient analysis tool that is not bound to any particular system of indicator-species classification. We studied lichens at air-quality monitoring sites in the South Coast Air Quality Management District (SCAQMD) of California, USA (Fig. 1), which possess an unprecedented variety of N measurements (Table 2). Most prior eutroph work focuses on agricultural N emissions and thus NH_x -dominant deposition regimes. Emissions from the highly urbanized Los Angeles Basin, however, are dominated by NO_x . This is the first study to calibrate lichens against the highly phytotoxic urban gas, nitric acid (HNO_3).

METHODS

Study area and site criteria

The South Coast Air Quality Management District (SCAQMD) of California, USA, covers an area of more than 10 700 square miles (27 713 km^2), spanning from the Pacific Ocean to the east past Joshua Tree National Park in the Mohave Desert. The topography and climate of the basin, together with a population in excess of 17 million people and almost 11 million vehicles create some of the worst air quality in the nation (Cox 2009). This region includes large areas of the San Bernardino, Cleveland, and Angeles National Forests, including several Class I National Wilderness Areas (Fig. 1). We visited a total of 22 sites in the summer of 2008: 11 sites in the San Bernardino National Forest, 6 sites in the Palomar Mountain area of the Cleveland National Forest, and 5 sites in the Sawmill Mountains in the Angeles National Forest. These are long-term monitoring sites originally surveyed for lichens in 1976 by Sigal (1979). Three of her sites were lost to development or fire. Replacement sites were chosen as close to former sites as possible using the sampling criteria described below (in the next several sections and subsections).

All stands were mixed conifer–black oak forest, between 1500 and 2000 m elevation, on north–northeast (NNE)-facing aspects, with 10° to 20° slopes, and with no evidence of recent fires. Mixed conifer forests of southern California typically consist of black oak (*Quercus kelloggii* Newb.), Jeffrey and ponderosa pine (*Pinus jeffreyi* Balfour and *P. ponderosa* C. Lawson), white fir (*Abies concolor* (Gordon et Glendinning) Hildebrand), big cone Douglas fir (*Pseudotsuga macrocarpa* (Vasey) Mayr) and incense cedar (*Calocedrus decurrens* (Torrey) Florin). Tree densities in our plots ranged from 118 to 312 live standing trees per hectare.

Meteorology and air quality data

Throughfall deposition.—Average N deposition in throughfall (in $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$) was measured between 2000 and 2005 at eight of the San Bernardino sites for a different study (Fenn et al. 2008). For best results, however, measurements should be taken in the year

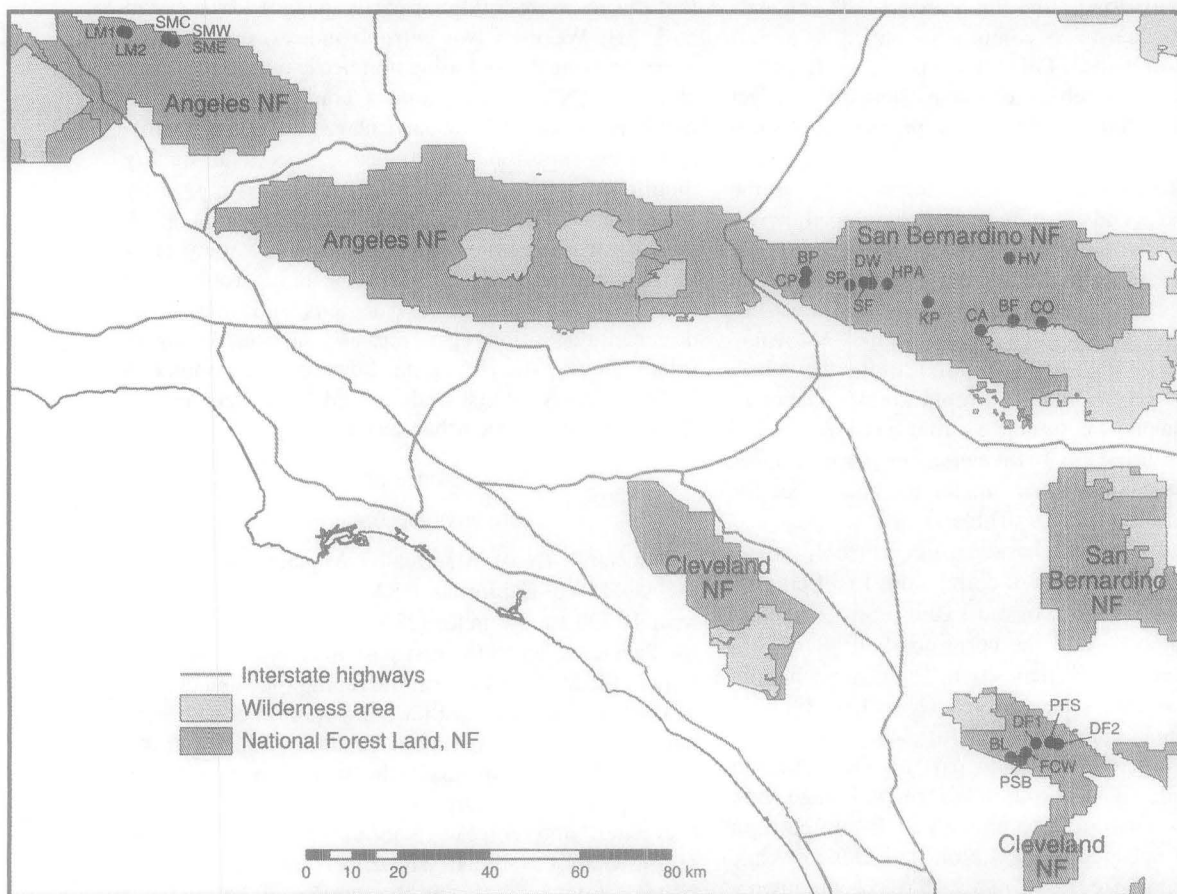


FIG. 1. Map of the study areas with the sites surveyed for epiphytic lichens in 2008. Codes (in alphabetical order) for each site are: BF, Barton Flat; BL, Boucher Lookout no. 1; BP, Breezy Point; CA, Camp Angelus; CO, Camp Osceola; CP, Camp Paivika; DF1, Deer Flat East; DF2, Deer Flat West; DW, Dogwood; FCW, Fry Creek West; HP, Heaps Peak Arboretum; HV, Holcomb Valley; KP, Keller Peak; LM1, Liebre Mt. 1; LM2, Liebre Mt. 2; PFS, Palomar Fire Station; PSB, Palomar State Park Boundary; SF, Sky Forest; SMC, Sawmill Mt. Campground; SME, Sawmill Mt. East; SMW, Sawmill Mt. West; and SP, Strawberry Peak. Essentially all of the land area west and south of the study areas is the greater Los Angeles urban region.

prior to and the same year as the lichen data (in this case, 2007 and 2008). Throughfall is measured year-round with a resin capture system that collects NH_4^+ and NO_3^- in rain water (Fenn and Poth 2004). Funnels are mounted on top of PVC tubes containing mixed-bed ion-exchange resins, and the devices are deployed beneath tree canopies (in this case *Pinus ponderosa* or *P. jeffreyi*) for up to six months. Throughfall data are interpreted as the sum of dry deposition, which collects on foliar surfaces absent rain, and wet deposition. Please see Fenn and Poth (2004) for further discussion of this technique.

N and O_3 gas concentrations.—For atmospheric concentrations of gases we used the passive sampling studies of A. Bytnerowicz (*unpublished data*) from nine sites. In 2007 summer air concentrations of NH_3 , O_3 , and NO_2 were measured using Ogawa filters (Ogawa and Company, Pompano Beach, Florida, USA,) and concentrations of HNO_3 were determined using the passive techniques of Bytnerowicz et al. (2005). Gases were sampled during the dry 6-month

period (May–October) when high levels of leachable NO_3^- and NH_4^+ accumulate in lichen thalli in the study region (Boonpragob et al. 1989). Analyses are reported for average concentrations of NH_3 , NO_2 , O_3 , and HNO_3 .

Modeled N and climate values.—For each site we also had estimates of eight major N pollutants from version 4.4 of the Community Multi-scale Air Quality (CMAQ) model:⁷ wet and dry reduced N (NH_x) deposition, wet and dry oxidized N (NO_x) deposition, and wet and dry atmospheric concentrations of ammonium (NH_4^+) and nitrate (NO_3^-). CMAQ is a photochemical grid model that simulates annual N deposition on a 4-km² resolution grid for much of California based on measured N deposition values from 2002 (Tonnesen et al. 2007). Data for the model were collected from multiple sources, including the National Atmospheric Deposition Program,⁸ the Clean Air Status and Trends

⁷ <http://www.cmaq-model.org/>

⁸ <http://nadp.sws.uiuc.edu/>

TABLE 2. Nitrogen and O₃ measurements for sites in three study areas, including throughfall N (Fenn et al. 2008), summer averages of gaseous pollutants (A. Bytnerowicz, *unpublished data*), modeled site N deposition (Tonnesen et al. 2007), and measurements of twig surface NO₃⁻.

Site	Modeled site N deposition (kg·ha ⁻¹ ·yr ⁻¹)								Throughfall N (kg·ha ⁻¹ ·yr ⁻¹)	Summer averages of gaseous pollutants (μg/m ³)				Twig surface NO ₃ ⁻ (μg/cm ²)
	NH _x		NO _x		Atmospheric NH ₄		Atmospheric NO ₃							
	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet						
Palomar Mt. area in Cleveland National Forest														
Boucher Lookout	0.8	0.2	5.5	0.6	<.1	0.2	<.1	0.6	...	...	...	...	...	7
Deer Flat E	0.9	0.1	5.6	0.4	<.1	0.1	<.1	0.4	...	...	...	...	...	6.9
Deer Flat W	0.9	0.1	5.6	0.4	<.1	0.1	<.1	0.4	...	...	...	...	...	7.8
Fry Creek West	0.7	0.1	5.1	0.5	<.1	0.1	<.1	0.5	...	...	...	...	...	6.3
Palomar Fire Stn.	0.7	0.1	5.1	0.5	<.1	0.1	<.1	0.5	...	3.5	4	5.3	52.7	6.4
Palomar State Park	0.7	0.1	5.1	0.5	<.1	0.1	<.1	0.5	...	...	...	...	...	6.4
San Bernardino National Forest														
Barton Flat	0.4	0.4	2.7	0.8	0.1	0.4	<.1	0.8	8.8	4.4	11.2	6.7	52.1	5.2
Breezy Point	3.5	1.4	9.2	1.5	0.1	1.4	0.1	1.5	...	...	...	...	...	11
Camp Angelus	1.9	0.8	3.9	1	0.1	0.8	<.1	1	12.8	5.8	12.6	8.7	55.8	5.4
Camp Osceola	0.3	0.2	2.2	0.6	0.1	0.2	0.1	0.6	7.5	4.4	11.2	6.7	52.1	2.6
Camp Paivika	3.5	1.4	9.2	1.5	0.1	1.4	0.1	1.5	71.1	8.1	25.4	19	64.0	13.4
Dogwood	1.3	0.6	7.2	0.9	0.1	0.6	<.1	0.9	33.4	8.2	28.9	13.6	59.1	13.4
Heaps Peak	1.2	0.7	6	1	0.1	0.7	<.1	1	36.4	6.9	18.7	14	65.1	10.3
Holcomb Valley	0.5	0.2	3.2	0.5	0.1	0.2	<.1	0.5	6.1	3	5.1	4.5	47.2	2.3
Keller Peak	0.7	0.6	4.4	1	0.1	0.6	<.1	1	...	4.2	8.3	6.4	56.4	4.3
Sky Forest	1.3	0.6	7.2	0.9	0.1	0.6	<.1	0.9	...	...	...	...	...	10.9
Strawberry Peak	1.6	0.8	8	1	0.1	0.8	0.1	1	39.3	8.2	28.9	13.6	59.1	13
Sawmill Mt. area in Angeles National Forest														
Liebre Mt. 1	1.8	0.2	5.4	0.4	0.1	0.2	0.1	0.4	...	...	...	...	...	10.1
Liebre Mt. 2	1.8	0.2	5.4	0.4	0.1	0.2	0.1	0.4	...	...	...	...	...	13.5
Sawmill Mt. Camp.	2.0	0.2	6.4	0.5	0.2	0.2	0.2	0.5	...	...	...	...	...	7.8
Sawmill Mt. East	2.0	0.2	6.5	0.5	0.2	0.2	0.1	0.5	...	...	...	...	...	9.6
Sawmill Mt. West	2.0	0.2	6.4	0.5	0.2	0.2	0.2	0.5	...	...	...	...	...	9.3

Note: Where there were no measurements available the data cells contain ellipses.

Network (CASTNET),⁹ and EPA's Aerometric Information Retrieval System (AIRS) data system¹⁰ for archived O₃, NO_x, and CO. N estimates were modeled using algorithms that took meteorology, emissions, and ancillary inputs such as initial and boundary conditions, and photolysis rates into consideration. Output from the CMAQ model has been validated against measurements from all the major ambient air-quality monitoring networks (Tonnesen et al. 2007).

All sites had modeled estimates of dew point, maximum and minimum temperatures and average precipitation that were extracted from interpolated climate maps of 1971–2000 norms (Daly et al. 2001, PRISM Climate Group 2004). While estimates from the CMAQ and PRISM models do not overlap in time with our lichen sample, we expect that they describe current gradients in climate and bulk deposition (i.e., deposition not under a canopy) with reasonable accuracy. Both modeled data sets are commonly used in recent lichen studies from the western United States with strong results (e.g., Geiser et al. 2010, Fenn et al. 2011). Additionally, the suitability of CMAQ estimates of dry NO_x deposition, the dominant form of N in the Basin,

was verified by measurements of NO₃⁻ accumulated on twigs in 2008.

Twig NO₃⁻ assays.—At each sampling site we measured NO₃⁻ accumulated on twig surfaces to serve as a contemporary indicator of dry NO_x inputs. The technique is similar to “leaf wash” methods used to characterize dry-deposition flux (Hanson and Lindberg 1991), although using twigs is more appropriate for deciduous species. Fifteen *Q. kelloggii* twigs (six cm long, 5–7 mm diameter) without lichen cover were collected at each site from healthy branches on upright, living trees, and placed in a plain manila envelope. Twigs were all collected within a 10-day period in August 2008 during which no rainfall was recorded. In the laboratory each twig was capped with melted paraffin wax at each cut end, and soaked in 0.025 mol/L KCl for one hour, periodically shaken, and removed (adapted from Kermit and Gauslaa [2001]). We then measured the pH and NO₃⁻ concentration of sample solutions, and we measured pH with a flathead Hanna HI9026 pH meter and NO₃⁻ with a TRAACS 2000 (Bran-Luebbe, Norderstedt, Germany).

Lichen surveys

At each site we established a transect of 10 mature *Quercus kelloggii* (dbh ≥ 14 cm) with no obvious signs of damage or disease; each tree was at least 10 m from the

⁹ <http://epa.gov/castnet/javaweb/index.html>

¹⁰ <http://www.epa.gov/ttn/airs/airsaqs/>

last sampled tree. Transects ran NNE along the line of the slope. Diameter at breast height (dbh, ~1.4m above uphill ground level) was recorded for each sampled tree, and bark pH was measured on the boles of five oaks per transect, one measurement at breast height in each cardinal direction.

Using Sigal's (1979) protocol, we recorded lichen cover on each *Q. kelloggii* using a 5 × 16 cm wire mesh grid with 0.5-cm² squares (200 squares/grid). We sampled one grid in each cardinal direction per bole at breast height, counting the number of squares filled by each lichen species, including the crusts, down to the half-square. Percentage cover for each species is reported as the percentage of the total sampled area occupied by that species per transect. These data were log₁₀-transformed for all analyses (1 + percentage cover) because values spanned multiple orders of magnitude.

Eutroph index

We designated all macrolichen species as oligotroph, mesotroph, eutroph, or unclassified, using the classification system of Jovan (2008). This system was initially based on the macrolichen classifications used in the Nitrofile Indicatie Waarde (NIW) and Acidofiele Indicatie Waarde (AIW) indices developed by van Herk (1999) for NH₃ bioindication. Then gradient analyses of lichen community data from California were used to classify additional *Physcia* and *Physconia* species as eutrophs (Jovan 2008). Our index is largely in agreement with the ILD_n and the LDV_{nitro} indices used for NH₃ biomonitoring in the Mediterranean forest ecosystems of Europe (Table 1). To build the index we summed the log-transformed percentage cover values for each eutrophic species occurring at a site. Crustose species were excluded from index calculations since affinities for N are less well known and many crusts were only identified to the genus level.

NMS response index: "community air scores"

We used nonmetric multidimensional scaling (NMS) ordination in PC-ORD (McCune and Mefford 1999) to build a second lichen response index that, for the same purpose as the eutroph index, describes how N impacted a lichen community is. A key difference is that an NMS index is not tied to a particular system of indicator species classification. It is inherently more objective for being simply a data-driven, mathematical representation of how similar the communities of plots are to one other. NMS also allows us to see if climate or stand composition variables may be confounding our response index.

NMS is an iterative nonparametric statistical tool commonly used to distill multivariate lichen response data (e.g., a matrix of logged percentage cover values by lichen species and plot) into the strongest gradients in community composition (e.g., Jovan and McCune 2005). We used the Sørensen (Bray-Curtis) distance measure to generate a distance matrix describing how

lichen species composition differs between all possible pairings of plots. Conceptually the Sørensen distance between any two plots is the abundance of species they share relativized by the total abundance of lichens within these two plots. NMS then uses a matrix of these distances to detect community gradients and to score plots relative to one another along each detected gradient. A common outcome for an N-impacted region is an NMS model that segregates the eutroph-rich from the eutroph-poor sites, assigning the former high plot scores and the latter low plot scores (or vice versa; see Jovan and McCune 2005, Jovan 2008). The abundance of oligotrophs and mesotrophs at a plot may also influence plot score, or other differences in species' composition may be important; it just really depends on how the lichen communities vary across the area of investigation. Detected gradients are tested for statistical significance using a randomization test. To reduce analytical noise, rare lichen species with <3 occurrences were removed from response matrices and highly similar species from the same genus were combined if they were difficult to differentiate in the field.

We visualized the NMS gradients as axes in a scatter plot and overlaid all 33 climatic, topographic, air quality, and environmental site variables to examine correlations. We rotated the NMS solution to maximize loading of air-quality variables along the axis that was our most likely candidate for an air-quality response. Plot scores along this axis (hereafter referred to as "community air scores") thus serve as a lichen response index for N. We ran Pearson's correlations in SAS (version 9.1.3; SAS Institute 2002), using the community air scores as a response variable, to determine which environmental variables are the best predictors of community composition. We examined how closely community air scores related to the conventional eutroph index, to see to what extent these scores describe how *eutrophic* a lichen community is.

RESULTS

Lichen community gradients

Most macrolichen species (20 out of 26) encountered in the field surveys are widely considered eutrophic (Table 3), indicating a highly N-rich environment. Lichen community composition was well-described by a statistically significant nonmetric multidimensional scaling (NMS) ordination with three gradients that explained a total of 87.9% of the variability among lichen communities at our 22 sites (Fig. 2). The strongest gradient (axis 1; $r^2 = 0.483$) was clearly related to air quality as described by N measurements and estimates. Axis two ($r^2 = 0.236$) did not have any strong associated vectors, and axis three ($r^2 = 0.160$) was roughly climate driven. As our focus is on air quality, we did not investigate the second two axes further.

The NMS-based lichen response index, *community air scores*, shows a strong positive linear relationship to the conventional eutroph index ($r^2 = 0.71$). These indices

TABLE 3. Species list for lichens found on *Quercus kelloggii*.

Species	N indicator group
<i>Buellia dakotensis</i> (H. Magn.) Bungartz	
<i>Caloplaca</i> sp. (with apothecia)†	
<i>Caloplaca</i> sp. (sorediate)	
<i>Candelaria concolor</i> (Dicks.) B. Stein	E
<i>Candelariella vitellina</i> (Hoffm.) Müll. Arg.	
<i>Collema furfuraceum</i> (Arnold) Du Rietz	U
<i>Hypogymnia imshaugii</i> Krog	O
<i>Lecanora subfusca</i> group	
<i>Lecidea</i> sp.	
<i>Letharia vulpina</i> (L.) Hue	O
<i>Megaspora verrucosa</i> var. <i>mutabilis</i> (Ach.) Nimis & Cl. Roux	
<i>Melanelixia glabra</i> (Schaer.) O. Blanco et al.	M
<i>Melanohalea elegantula</i> (Zahlbr.) O. Blanco et al.	M
<i>Melanohalea subolivacea</i> (Nyl.) O. Blanco et al.	M
<i>Ochrolechia</i> sp. A. Massal.	
<i>Parmelia quercina</i> (Willd.) Hale	M
<i>Phaeophyscia orbicularis</i> (Necker) Moberg	E
<i>Phaeophyscia</i> sp. Moberg	E
<i>Physcia aipolia</i> (Humb.) Fűrnröhr	E
<i>P. biziana</i> (A. Massal.) Zahlbr.	E
<i>Physcia</i> sp.	E
<i>Physcia</i> complex: <i>P. adscendens</i> (Fr.) H. Olivier and <i>P. tenella</i> (Scop.) DC.	E
<i>Physcia tribacia</i> (Ach.) Nyl.	E
<i>Physconia americana</i> Essl.	E
<i>P. enteroxantha</i> (Nyl.) Poelt	E
<i>P. fallax</i> Essl.	E
<i>P. isidiigera</i> (Zahlbr.) Essl.	E
<i>P. perisidiosa</i> (Erichsen) Moberg	E
<i>Rinodina</i> sp. (gray thallus)	
<i>Waynea californica</i> Moberg	
<i>Xanthomendoza fallax</i> (Hepp ex Arnold) Söchting, Kärnefelt & S. Kondratyuk	E
<i>X. fulva</i> (Hoffm.) Söchting, Kärnefelt & S. Kondratyuk	E
<i>X. galericulata</i> L. Lindblom	E
<i>X. hasseana</i> (Räsänen) Söchting	E
<i>X. oregana</i> (Gyeln.) Söchting, Kärnefelt & S. Kondratyuk	E
<i>Xanthoria candelaria</i> (L.) Th. Fr.	E
<i>X. polycarpa</i> (Hoffm.) Th. Fr. ex Rieber	E

Notes: Rare species (designated by daggers [†]) and crustose species were not used in the nonmetric multidimensional scaling (NMS) analysis. Macrolichen species are designated as eutroph (E), mesotroph (M), oligotroph (O), or unclassified (U) if sensitivity is unknown. Designations follow the classification system of Jovan (2008).

† Rare species.

correlated similarly to pollution variables although correlations of the NMS-derived index were stronger in most cases (Table 4). By far the best predictor of eutroph abundance was (1) throughfall N under *Pinus* spp. (statistics reported for community air scores; $r^2 = 0.94$), a measure of the wet and dry deposition of NH_4^+ and NO_3^- to the forest canopy and its flux to the forest floor. The next best descriptors among all 33 environmental variables were: (2) total dry N deposition estimates from CMAQ ($r^2 = 0.62$); (3) dry NO_x deposition estimates from CMAQ ($r^2 = 0.60$); (4) air concentrations of HNO_3 ($r^2 = 0.59$); and (5) twig NO_3^- ($r^2 = 0.58$).

Other N measurements and O_3 concentrations were correlated with both response indices though not as

closely. Fig. 3 shows the correlation structure between select pairings of lichen response indices and N measurements. All relationships can be characterized as approximately linear, the most noteworthy exception being the slight logarithmic curvature of relationships between throughfall and average concentrations of N gases. Overall, N measurements exhibited a high degree of inter-correlation.

The pH of trunks was consistently high, varying from 4.8 to 6.1. Both lichen response indices correlated negatively, suggesting that eutroph cover on *Quercus kelloggii* in the SCAQMD decreases as pH increases (Table 4). Neither index correlated with the pH of oak twigs, which varied little across sites (pH range, 4.2–4.7).

DISCUSSION

Quercus kelloggii stands in Southern California (USA)'s South Coast Air Quality Management District (SCAQMD) currently support a fairly simple macrolichen flora comprised almost entirely of eutrophs, a pattern indicative of a highly polluted environment (Table 3). Lichen communities are subject to N inputs ranging from moderate to severe (Table 2), which corresponds to an increase in eutroph dominance as demonstrated by both the eutroph index and the NMS-derived response index, community air scores (Fig. 3). If we could find a pristine situation in the hot, dry SCAQMD, we would expect to find many of the eutrophic species we observed on oaks (Table 3), just not at their current richness and abundance. Lichenological records from the 1900s (Hasse 1913) and 1976 (Sigal and Nash 1983) suggest we should find more N-sensitive species in the mix, such as *Ramalina menziesii* Taylor at lower elevations, cyanolichens (*Pseudocyphellaria* spp., *Leptogium* spp., *Collema* spp.), and drought-tolerant *Usnea* spp. in the montane regions.

Throughfall deposition

Among the many N measurements we considered, throughfall N measured under *Pinus* spp. emerged as an exceptionally good predictor of eutroph abundance (Table 4, Fig. 3) despite being a few years out of synchronization with the lichen surveys. The only other study comparing a eutroph index vs. throughfall N concerns the less N-impacted, conifer-dominated stands of the Sierra Nevada Range in central California (Fenn et al. 2008). Throughfall was their only calibration measurement (Table 1) although it correlated well with their eutroph index when hardwood basal area was also accounted for ($r^2 = 0.77$). Lichen data were collected using the standard methodology of the Forest Inventory and Analysis Program Lichen Communities Indicator (Jovan 2008), which does not discriminate among substrate tree species. Eutrophs are primarily hardwood-dwelling species.

Throughfall is the most integrative N measure considered in this study: it captures both wet and dry

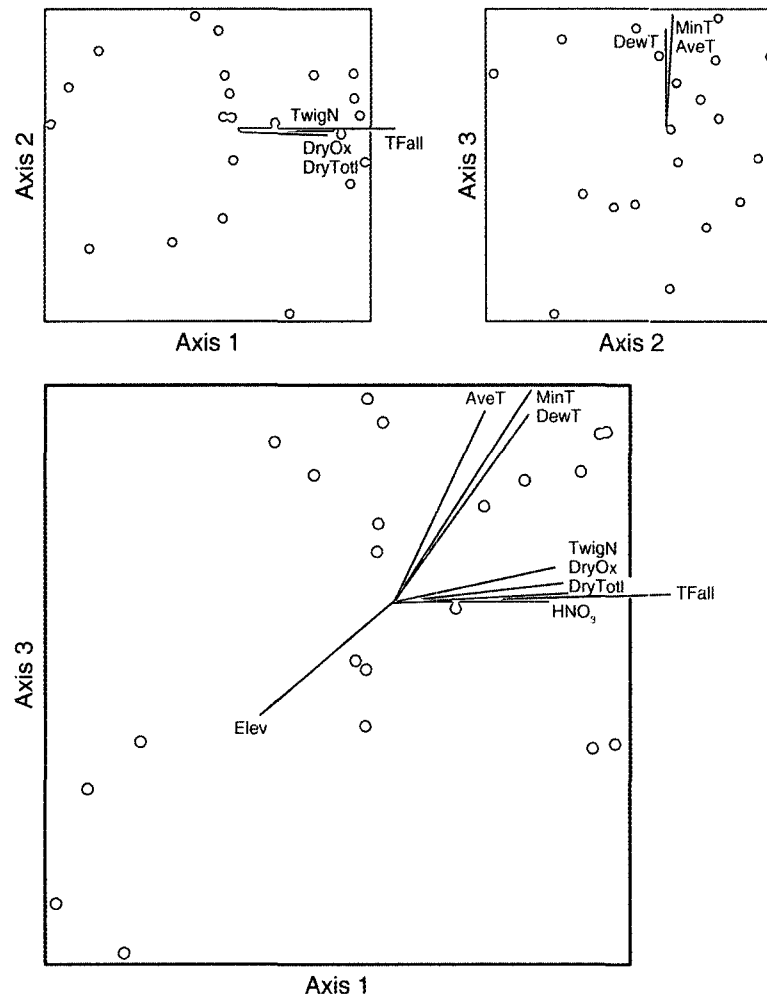


FIG. 2. Ordination diagram showing community air scores along three axes, each representing an independent gradient in the composition of lichen communities. Environmental variables with a strong correlation to one or more axes are shown as vectors scaled according to Pearson's correlations (Table 4). Nitrogen variables are loaded along axis 1, and climate variables are loosely associated with axis 3. Abbreviations: Tfall, throughfall N; HNO_3 , nitric acid; DryOx, estimated dry NO_x deposition from community multi-scale air quality (CMAQ) model; DryTotl, estimated total dry N deposition from CMAQ; AveT, average temperature; MinT, minimum temperature; DewT, dew temperature; Elev, elevation; TwigN, twig NO_3 surface deposition.

deposition of all N gases and particulates. Throughfall under *Pinus* spp. explained 45% more variability in lichen communities than our best estimates of bulk N deposition from the Community Multi-scale Air Quality (CMAQ) model (see "Total N (wet + dry)" in Table 2). It is unknown to what degree the lesser performance of CMAQ estimates reflects their separation in time from the lichen surveys (2002 vs. 2008, respectively). However the exceptional performance of throughfall is intuitive, especially for Mediterranean and desert systems where dry deposition is the primary mode of N loading. Tree canopies modify N regime in several important ways. The high surface area of leaves and stems provides abundant surfaces for collection of dry deposition (Fenn and Bytnerowicz 1997). The canopy is known to absorb or retain some deposited N (Gaige et al. 2007), therefore reducing N inputs reaching lichens. Canopies may leach organic forms of N (Cape et al. 2010) that lichens can

utilize (Dahlman et al. 2004). All of these processes affect N availability during the wet season and periodic moist events when lichens are metabolically active. Surprisingly few analyses of epiphytic lichen or bryophyte communities consider throughfall or the related N flux measurement, stemflow (but see Mitchell et al. 2005, Zechmeister et al. 2007, Fenn et al. 2008, Leith et al. 2008).

We used measurements under *Pinus* spp. to characterize the effect of throughfall on lichen communities of *Quercus kelloggii* because those were the measurements available to us. It was previously demonstrated at one of our study sites, Barton Flats, that N flux is highest under white fir (*Abies concolor*), intermediate under *Pinus* spp., and lowest under *Quercus kelloggii* (Fenn and Bytnerowicz 1997). This is largely because the high surface area of needles in a conifer crown intercepts more dry deposition than a deciduous oak crown. The strong

TABLE 4. Pearson correlations between all environmental variables and both lichen response indices, with a minus sign added to denote negative correlations.

Environmental variables	Community air scores		Eutroph index	
	r^2	P	r^2	P
Deposition measurements				
Throughfall N	0.94	<0.0001	0.77	0.0044
Twig NO_3^-	0.58	<0.0001	0.42	0.0011
Deposition estimates (CMAQ)†				
Total dry N	0.62	<0.0001	0.34	0.0043
Dry NO_x	0.60	<0.0001	0.35	0.004
Total N (wet + dry)	0.49	0.0003	0.22	0.0291
Dry NH_3	0.48	0.0004	0.24	0.0222
Dry atmospheric NH_4^+	0.38	0.002	0.27	0.0134
Dry atmospheric NO_3^-	0.36	0.003	0.21	0.0307
All atmospheric N	0.06	0.25	0.00	0.87
Wet NH_3	0.05	0.31	0.00	0.84
Wet atm. NH_4^+	0.05	0.31	0.00	0.84
Total wet N	0.03	0.45	0.01	0.64
Wet NO_x	0.01	0.65	0.03	0.45
Wet atmospheric NO_3^-	0.01	0.65	0.02	0.45
Air concentration measurement				
HNO_3	0.59	0.006	0.27	0.099
Total N average‡	0.50	0.02	0.23	0.14
O_3	0.52	0.01	0.33	0.08
NH_3	0.47	0.02	0.27	0.099
NO_2	0.42	0.03	0.19	0.18
Site environmental variables				
Minimum temperature	0.49	0.0003	0.63	0.0000
Dew point	0.49	0.0003	0.53	0.0001
Elevation	-0.48	0.0004	-0.40	0.0015
pH of oak bole bark	-0.43	0.001	-0.24	0.018
Average temperature	0.33	0.01	0.45	0.0006
Hardwood basal area	0.15	0.52	0.37	0.0025
Hardwood cover (%)	0.14	0.35	0.38	0.0023
Softwood cover (%)	0.07	0.08	0.008	0.69
Total basal area	0.04	0.07	0.039	0.38
Maximum temperature	0.03	0.43	0.073	0.22
Tree density (trees/ha)	0.02	0.50	0.00	0.74
Conifer basal area	0.02	0.13	0.11	0.12
Twig surface pH	0.01	0.60	0.00	0.96
Precipitation	0.00	0.86	0.021	0.52

† CMAQ is the Community Multi-scale Air Quality modeling system.

‡ Average ($\text{HNO}_3 + \text{NH}_3 + \text{NO}_2$).

correlations we found between the lichen response indices and *Pinus* spp. throughfall suggest that the ratio of N flux under *Pinus* vs. *Quercus kelloggii* canopies is consistent across our study sites.

Deposition vs. gas concentrations

Dry deposition of oxidized forms of N dominates the N-loading processes in Southern California (Bytnerowicz and Fenn 1996, Padgett et al. 1999). Due to vehicle emissions from the Los Angeles Basin, concentrations of oxidized N gases exceed NH_3 by a factor of 3 to 5 at all sites (Table 2)—so it is unsurprising that both lichen response indices and throughfall measurements are closely linked to deposition measures such as total dry N, dry NO_x , and twig NO_3^- (Table 4, Fig. 3).

Concentrations of atmospheric gases were comparatively weak predictors of eutroph abundance as described by both response indices. Timing of the gas measurements (2007) was well aligned with the lichen community surveys (2008) and so relatively weak correlations are not the result of temporal disconnect. The strongest correlation was between community air scores and HNO_3 ($r^2 = 0.59$), a highly reactive vapor for which lichen sensitivity was only recently demonstrated experimentally using the oligotrophic species, *Ramalina menziesii* (Riddell et al. 2008; J. Riddell, P. Padgett, and T. H. Nash III, unpublished manuscript). Of all the N species of gas, HNO_3 has the highest deposition velocity by at least an order of magnitude, and so atmospheric concentrations and deposition are closely correlated. Nitric acid is the dominant pollutant contributing to throughfall N ($r^2 = 0.95$; Fig. 3) even though the less reactive gas, NO_2 , occurs in higher concentrations (Table 2).

In European studies, eutroph response indices correlate well with concentrations of NH_3 gas (e.g., Cape et al. 2009, Sutton et al. 2009b; Table 1) and are used to negotiate critical levels (CLE) for NH_3 in Western Europe (Cape et al. 2009, Hallsworth et al. 2010). Using linear regression, we tried various data transformations on eutroph cover, percentage eutroph cover, eutroph frequency, and average eutroph frequency vs. NH_3 , but did not find any statistically significant relationships between them even when oak bole pH and twig pH were included as covariates (data not shown). The strongest relationship to NH_3 is shown in Table 4 for the eutroph index, which is based on $\log_{10}$ transformed percentage cover measurements ($r^2 = 0.27$, $P = 0.099$). Removal of an outlier in scatterplots between the eutroph index and N gas concentrations improves these correlations by about 30% although throughfall would still remain the best predictor. Removal of the outlier in the scatterplot of the eutroph index vs. throughfall results in $r^2 = 0.94$. We did not think it was justified to throw out any of these data, however, because these outliers did not systematically appear in other bivariate relationships.

These results raise the important question whether European eutroph indices specifically indicate NH_3 or whether high correlations result because NH_3 dominates N deposition in regions where the indices were calibrated (Table 1); nearly all studies, including Jovan and McCune's (2005) work in the highly agricultural Central Valley, focus on agriculturally influenced landscapes or point sources. The work of Gadson et al. (2010) seems to support the latter hypothesis. They calibrated a eutroph index (percentage cover in nitrophytes) in a forest influenced by urban and industrial emissions from the London metropolitan area and found correlations to NO_2 . Interestingly, they also tested the NIW (Mitrofile Indicatie Waarde, a widely used eutroph index developed in the Netherlands for NH_3 biomonitoring), which did not correlate significantly with either NO_2 or NH_3 (Table 1).

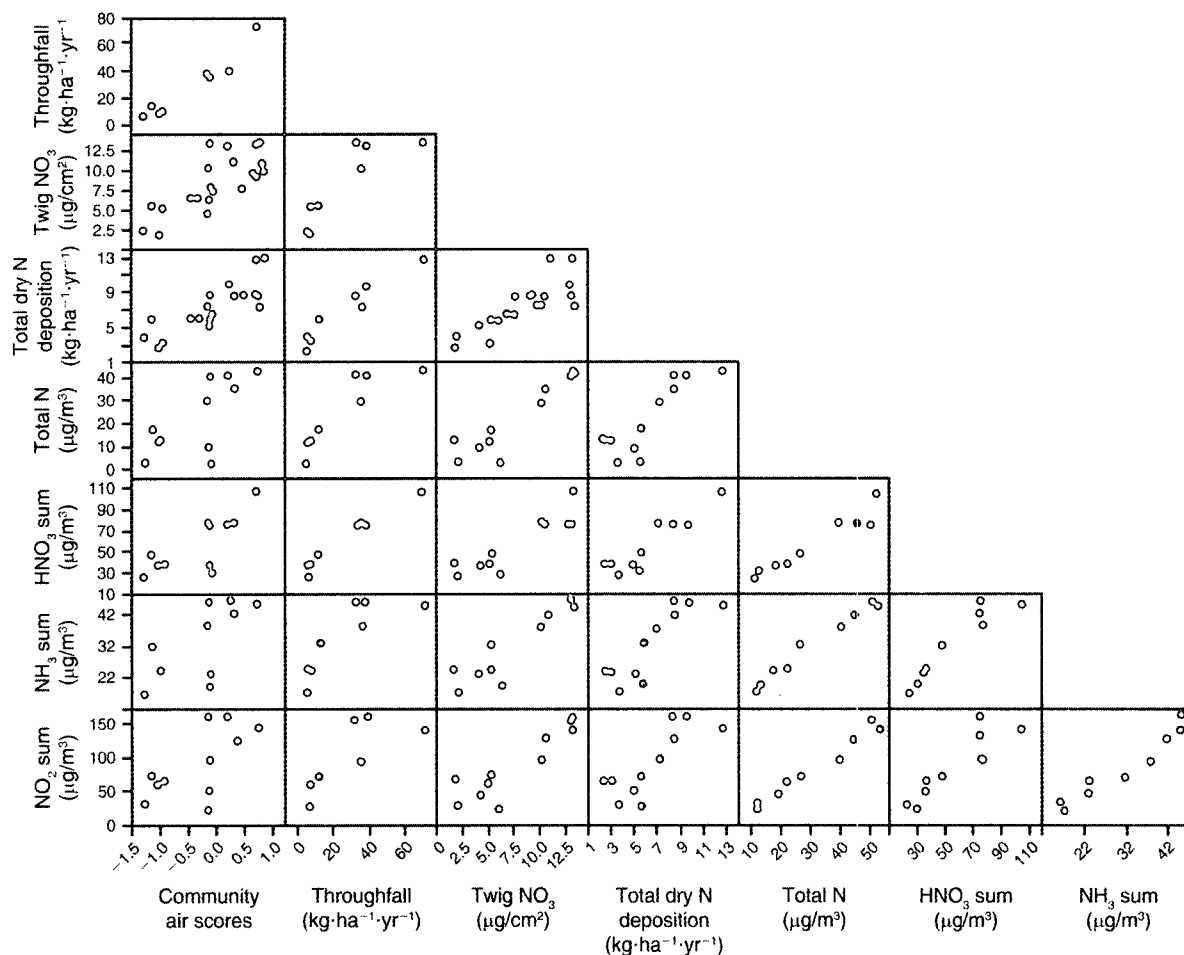


FIG. 3. Bivariate scatterplots showing the correlation structure between community air scores ($n = 22$ sites) and a selection of N measures (throughfall, $n = 8$ sites; twig NO_3 , $n = 22$ sites; total dry N deposition, $n = 22$ sites; total N, $n = 11$ sites; HNO_3 sum, $n = 11$ sites; NH_3 sum, $n = 11$ sites; NO_2 sum, $n = 11$ sites). The r^2 values are indicated in the lower right-hand corner of each scatterplot. All relationships are statistically significant except for the four relationships between the eutroph index and N gas concentrations.

According to Cape et al. (2009:1034), a critical level (CLE) of gas concentration can be defined when “a direct adverse effect on the receptor of [the N gas] can be demonstrated as distinct from any contribution of [the N gas] to N deposition and soil-mediated effects.” Our findings suggest that (1) eutroph indices should not be used in the development of NH_3 CLEs if these guidelines are to be strictly followed and (2) eutroph indices are not suitable for mapping NH_3 unless NH_3 is the only major N pollutant in the vicinity. It is definitely possible we are underestimating the significance of what appear to be minor differences among eutroph indices; maybe by construction some indices are more tuned to NH_3 . Our methods are not an exact match with any previous CLE studies. Also, several species-focused studies highlight the individuality of species’ responses to N pollutants (e.g., Davies et al. 2007, Larsen et al. 2007, McCune and Geiser 2009) and so potentially different eutrophs differ in their tolerances to different N pollutants. Moreover, it may well be that the NH_3 -specificity of a eutroph index is conditional, arising when available substrates are

acidic (see *Oak bole pH*, below). We recommend that our results be confirmed using other eutroph indices and data sets from regions influenced by different climatic conditions and historical pollution regimes.

The critical level (CLE) for NH_3 in Europe as well as published critical loads (CLO) for N are not wholly dependent upon eutroph indices. Most proposed CLE/CLO values are supported by two or more measures of biological response; many studies on lichen response to N also include indices of oligotroph and/or mesotroph abundance, which are often leveraged against a eutroph index to quantify N level associated with a shift into eutroph dominance (the threshold defining “harm” to the lichen community). Oligotroph and mesotroph sensitivities cannot be commented on here directly because *Quercus kelloggii* in the Los Angeles Basin hosted very few of these sensitive species (Table 3). The CLO for oak woodlands in California, $5.5 \text{ kg N} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$, is far exceeded at most of our sites (Fenn et al. 2010). One should not assume, though, that oligotrophs and mesotrophs are *not* differentially

sensitive to different N compounds; several experimental studies in the laboratory suggest ammonium (NH_4^+) is taken up more readily than nitrate (NO_3^-) (e.g., Dahlman et al. 2004, Gaio-Olivera et al. 2005, Palmqvist and Dahlman 2006). On the other hand, a recent experiment using two oligotrophic species growing under natural conditions did not find a significant difference in the uptake of these two ions (*Platismatia glauca* (L.) W.L. Culb. and C.F. Culb. and *Alectoria sarmentosa* (Ach.) Ach.; Johansson et al. 2010). Fenn et al. (2008) did find throughfall N was a strong predictor of mesotroph and oligotroph indices (r^2 ranged from 0.58 to 0.70) in the Sierra Nevada range although throughfall was the only calibration measurement they studied.

Synthesizing an all-inclusive array of N calibration measurements is impractical to expect for every CLE and/or CLO study. However, we do need a few comprehensive studies to lay the foundation for isolating responses of oligotrophs, mesotrophs, and eutrophs, to particular N compounds. The high multicollinearity of N measurements in the SCAQMD airshed demonstrates the risk of mistaking correlation for causality in field studies (Fig. 3). Had we lacked deposition measurements, we could have easily overemphasized the moderately strong correlations between eutrophs and any one of the N gases, all of which were statistically significant for the community air scores index (see Table 4: "Air concentration measurements").

As such, we recommend that lichen CLE/CLO studies utilize measurements of both NH_x and NO_x within the same study. We also recommend these studies look at both gas concentrations and deposition values. Likewise, it is essential that indices developed from studying lichen response around a pollution point source undergo additional calibration before being applied across a broader geographic region with a more complex deposition regime. Our throughfall results should be experimentally confirmed; technically we cannot be 100% certain that eutroph abundance in our sample is caused by throughfall N since this study lacked experimental manipulation. While our calibration data cover major N species, they are not all inclusive (e.g., we lacked measurements of peroxyacetylnitrate (PAN), which Sigal and Taylor (1979) demonstrated reduces photosynthesis in Southern California species that were low (*Hypogymnia imshaugii*) or absent (*Collema nigrescens* (Huds.) DC., *Parmelia sulcata* Taylor, *Peltigera rufusces* (Weiss) Humb.) at our study sites).

Oak bole pH

Our results give very strong evidence that eutroph abundance is primarily dependent on N inputs, not bark pH, in *Quercus kelloggii* stands of the Los Angeles Basin. Unexpectedly, both community air scores and the conventional eutroph index correlated negatively with bole pH. This means eutroph abundance declines as bole pH increases across our study area, which conflicts with

several dozen studies demonstrating the opposite relation (e.g., Barkman 1958, van Herk 2001, Frati et al. 2008). Our correlation is most likely just a correlation, resulting because acidifying pollutants (HNO_3 and secondarily HNO_2 , which are both strong acids) dominate deposition in the Basin, leading to more N-impacted sites having more acidic oak boles.

Discourse on the relative importance of substrate pH vs. N inputs is anything but clear; some studies suggest pH is the primary driver of eutroph colonization and abundance (e.g., 2008; Gilbert 1976, van Dobben and ter Braak 1998, Frati et al. 2007) while many others attribute this to both NH_3 and pH (e.g., van Herk 2001, Wolseley et al. 2006, 2009). The agricultural- NH_3 bias in lichen biomonitoring studies has helped to obscure the potential independence of these two factors because NH_3 deposition raises bark pH. Moreover, some European forests are recovering from the high SO_2 of the 1970s, which left a legacy of acidified bark surfaces that appears to limit eutroph colonization (van Dobben and ter Braak 1998). This legacy is a major argument for studying lichens on younger and more easily standardized surfaces, like twigs (Wolseley et al. 2006).

We hypothesize that oak bole pH is high enough in the basin (4.8–6.1) to not constrain eutroph establishment and proliferation. The possibility for a eutroph–pH threshold was recently posited by Spier et al. (2010). They worked with lichen communities on deciduous tree trunks, all with pH at or above 5.0, and showed that tree species is a more important determinant of eutroph and oligotroph abundance than bole pH. In the SCAQMD airshed, high bole pH is not necessarily connected to NH_3 . The dust from a dry, hot Mediterranean climate substantially raises the bark pH of *Quercus* species (Loppi and De Dominicis 1996). Likewise bark in the SCAQMD was never exposed to SO_2 concentrations as extreme as urban areas in the United Kingdom and eastern United States. Where tree substrates are more acidic, either naturally or due to pollution regime, it is very possible that eutroph abundance may depend on NH_3 deposition until bark surfaces are made sufficiently alkaline.

Conclusions

The distribution of N pollutants in the SCAQMD airshed was substantially inter-correlated and many individual pollutants, including both gas concentrations and N deposition products, correlated significantly with eutroph abundance. We found strong evidence that eutrophic lichen species distributions in this arid landscape are driven by the combined effect of multiple N pollutants, represented best by the N flux in canopy throughfall. The pH of oak boles was high, which hypothetically releases eutrophs from a dependence on NH_3 or other alkalizing agents (like calcareous dust) to establish. Future work should include a head-to-head

comparison of how various N measurements correlate to oligotroph and mesotroph response indices.

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