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A simple tool for estimating throughfall nitrogen deposition in forests of western North America using lichens

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ABSTRACT

Anthropogenic nitrogen (N) deposition has had substantial impacts on forests of North America. Managers seek to monitor deposition to identify areas of concern and establish critical loads, which define the amount of deposition that can be tolerated by ecosystems without causing substantial harm. We present a new monitoring approach that estimates throughfall inorganic N deposition from N concentration in lichens collected on site. Across 84 study sites in western North America with measured throughfall, a single regression model effectively estimated N deposition from lichen N concentration with an R^2 of 0.58 and could be improved with the addition of climate covariates including precipitation seasonality and temperature in the wettest quarter to an R^2 of 0.74. By restricting the model to the more intensively sampled region including Oregon, Washington, and California, the R^2 increased to 0.77. Because lichens are readily available, analysis is cost-effective, and accuracy is unaffected by mountainous terrain, this method allows development of deposition estimates at sites across broad spatial and topographic scales. Our approach can allow land managers to identify areas at risk of N critical load exceedance, which can be used for planning and management of air pollution impacts.

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1. Introduction

Anthropogenic nitrogen (N) deposition has impacted ecosystems of western North America by changing ecosystem functioning and community composition of some organisms (Fenn et al., 2003a). Excess N deposition has been linked to increased invasion by exotic plants (Fenn et al., 2011; Weiss, 1999) and changes in community composition of lichens (Geiser and Neitlich 2007), ectomycorrhizal fungi (Lilleskov et al., 2008), alpine plants (Bowman et al., 2006), and diatoms (Baron, 2006). N deposition has also impacted ecosystem attributes such as foliar chemistry (Rueth and Baron, 2002), soil chemistry (Baron et al., 2000; Breiner et al., 2007), fine root biomass and NO_3-leaching (Baron et al., 2011; Fenn et al., 2008) and freshwater acidity (Baron et al., 2011; Sullivan et al., 2005). To prevent the decline of forested ecosystems from N deposition impacts, managers and policy makers are increasingly interested in determining N critical loads (Fenn et al., 2010; Pardo et al., 2011), levels of N deposition that can be sustained without adverse effects on communities and ecosystem functioning based on current knowledge (Porter et al., 2005).

Critical loads of N are preferably based on measurements of total N deposition, but are more commonly based on deposition of dissolved inorganic N (DIN) because dissolved organic N is not routinely measured, especially across deposition networks. In practice, critical loads are often based on DIN deposition in throughfall (e.g., Fenn et al., 2008), such as in this study. Throughfall DIN consists of NO_3-N and NH_4-N, both of which are forms of N that are readily biologically available. The establishment and application of critical loads requires accurate measurements of N deposition to calibrate empirical models of critical loads and, subsequently, to monitor status, trends and exceedances. However, cost-effective techniques that measure the major components of N deposition are still in development (Table 1). Individual stations in the western United

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Table 1
Types of N deposition measures available.

Type	Examples	Advantages	Disadvantages
Passive biological monitors	Epiphytic lichens (Herzig et al., 1989) bryophytes (Pitcairn et al., 2003)	Accurate at sites where measured Integrates N deposition and effects Minimal costs	Potential for organisms to accumulate N differently under different conditions
Passive chemical monitors	Passive gas samplers (Bytnerowicz et al., 2002)	Allows for more extensive monitoring than non-passive methods	Data only available for the duration of study Gas samplers provide only time-averaged concentrations
	IER[a] throughfall and bulk deposition collectors (Fenn and Poth, 2004)	Accurate at sites where measured.	IER samplers do not provide ionic concentrations in or pH of precipitation or throughfall solutions
National instrumented networks for dry deposition	CASTNET[b] (Baumgardner et al., 2002) IMPROVE (1995)[c]	Many years of data available	Few sites in western United States Most monitor only some components Fluxes calculated have high uncertainty. Dry deposition is not measured
National instrumented networks for wet deposition	NADP-NTN (2012)[d]	Many years of data available Accurate at sites where measured	
National and regional spatial models	NADP (2012)[e] CMAQ[f] (Appel et al., 2011)	NADP provides multi-year estimates across large areas CMAQ includes all major N pollutants and provides estimates of total N deposition	Interpolations are not as accurate as measured methods May not be available at spatial scale of interest
	ClimCalc (Ollinger et al., 1993)	Large-scale mapping of deposition possible	Simulated deposition is uncertain because of uncertainty in emissions data and simulation model deficiencies

[a] Ion Exchange Resin (IER).
[b] Clean Air Status and Trends Network (CASTNET).
[c] Interagency Monitoring of Protected Visual Environments (IMPROVE).
[d] National Atmospheric Deposition Program – National Trends Network (NADP-NTN).
[e] National Atmospheric Deposition Program (NADP).
[f] Community Multiscale Air Quality (CMAQ).

States monitor wet deposition of inorganic N in precipitation (NADP, 2012) and particulates in aerosols (IMPROVE, 1995) but are limited in their ability to integrate these sources with meteorological patterns and vegetation interactions to provide regional-scale estimates of total deposition affecting forest processes. CASTNET sites (Baumgardner et al., 2002) provide dry deposition estimates for select ions at monitoring sites that are co-located with NADP wet deposition samplers, but there are only 29 CASTNET sites in western North America. Individual researchers have established sites monitoring concentrations of gaseous nitric acid, ammonia, and nitrogen dioxide (Bytnerowicz et al., 2002) and estimates of throughfall N deposition (Fenn et al., 2008). Air quality simulation models such as the Community Multiscale Air Quality model (CMAQ; Appel et al., 2011) provide estimates of total inorganic deposition and various N species across the modeling domain (Fenn et al., 2003b, 2010) but these simulated deposition estimates may not be as accurate as site-specific measurements and are more effective at defining broader spatial scale deposition.

Cost-effective monitors that integrate wet and dry deposition N sources are needed for understanding the distribution and effects of N deposition on forests. Measures of wet or dry deposition alone are typically well-correlated with ecosystem responses (Geiser and Neitlich, 2007; Jovan et al., 2012; Williams et al., 1996; Williams and Tonnessen, 2000) but may underperform compared to more complete measures of deposition. For example, N measured in throughfall explained >30% more variability in lichen community composition than partial measures, most of which were significantly correlated to lichens as well as each other (Jovan et al., 2012).

Throughfall N deposition provides an integrative lower-bound estimate of total inorganic N deposition to forested ecosystems (Lovett and Lindberg, 1993). A passive throughfall monitoring approach relies on ion exchange resins (IER) that absorb inorganic ions from wet deposition as well as dry and cloudwater deposition that washes from canopy surfaces above the monitors (Fenn et al., 2009; Fenn and Poth, 2004). Small-scale studies have found that throughfall N measured with IERs is well-correlated with changes in epiphytic communities and ecosystem attributes (Breiner et al., 2007; Fenn et al., 2007, 2008; Jovan et al., 2012; McMurray et al., 2013).

N concentrations in epiphytic lichens are potentially an alternative approach to passively monitor N deposition in forests. Lacking a cuticle, lichens accumulate N and other water soluble nutrients roughly in proportion to their abundance in the atmosphere (Herzig et al., 1989). Implementation is simple because lichens are readily available throughout forests in the region and require no set-up. Past work shows lichen N concentrations are strongly correlated with the lichen community composition shifts associated with increasing N deposition (Fenn et al., 2008; Geiser and Neitlich, 2007; Geiser et al., 2010), thus linking deposition with biological effects. However, lichen N values have not formerly been calibrated against N deposition measurements across a large region. Our objective was to model the relationship between N concentration in lichen thalli against throughfall N deposition measured across the western United States. If sufficiently accurate, researchers and managers could use the model to estimate throughfall deposition in kilograms per hectare per year for new sites based on N concentrations (% of dry weight) in lichen thalli.

2. Methods

Throughfall IER monitors were established between 2000 and 2011 at 84 sites in western North America (Fig. 1) as part of several separate studies (Table 2). Each site included 9-12 IER funnel collectors attached to IER columns installed under the forest canopy

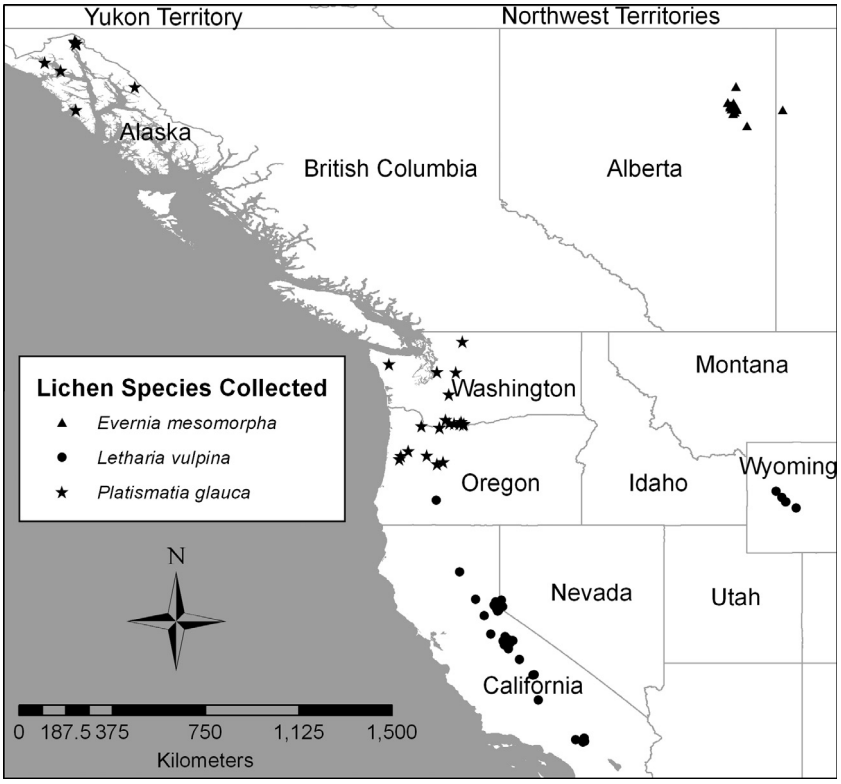

Fig. 1. Locations of sites where epiphytic lichens were sampled and throughfall measured with ion exchange resin (IER) collectors.

Table 2
Data sources used in calibration from western North America, number of sites monitored, years throughfall data were collected, range of total annual precipitation (Precip., cm), temperature during the wettest four months of the year (Wet, °C), precipitation seasonality (Seas., %) and total dissolved inorganic N measured in throughfall (DIN, kg/ha/year). Lichen N concentrations for several studies are available in the Northwest Alliance for Computational Science and Engineering (NACSE, 2012) public online database; however, throughfall deposition measurements are not.

Study	Region	Sites	Years	Precip.	Wet.	Seas.	DIN	Source
Athabasca oil sands	Alberta and Saskatchewan	13	2007–2009	41–47	14.1–16	53–60	1.3–24.8	Unpublished data
Coast to crest	Oregon and Washington	13	2006–2009	97–291	−0.8–5.8	49–70	0.5–11.6	NACSE 2012
Sierra Nevada Mountains	California	11	2000–2003	64–164	−1.2–6.9	66–81	1.2–18.3	Fenn et al., 2008
Yosemite National Park	California	11	2011	78–103	−2.6–6.4	69–80	2.7–4.7	Unpublished data
Southeast Alaska	Alaska	9	2008–2009	82–233	−3.6–6.7	36–47	0.1–2.4	NACSE 2012
Columbia River Gorge	Oregon and Washington	9	2006–2008	68–256	0.6–3.4	70–73	1.2–11.8	Fenn et al., 2007; NACSE 2012
Lake Tahoe	California	9	2011–2012	53–93	−2.7–1.0	61–72	1.3–5.6	Unpublished data
Wind River Range	Wyoming	5	2011	28–39	4.1–10.8	16–31	1.2–1.5	NACSE 2012; McMurray et al. 2013
Southern California	California	4	2000–2005	66–87	0.8–4	71–83	6.1–39.3	Fenn et al., 2008 and Jovan et al., 2012

roughly halfway between tree boles and driplines (Fig. 2A, Fenn and Poth, 2004); funnels were 10 cm in diameter in California sites established prior to 2005. In all the other study sites funnel diameters were 21.1 cm. Both funnel sizes were scaled to kg/ha/year by area represented; the larger funnels were expected to provide less variable estimates but were otherwise comparable. In all studies, some individual resin columns were excluded because they were damaged by wildlife or contaminated by bird droppings; on average, each study excluded 8.1% of the IER resin columns with a minimum of 4.0% and maximum of 13.9%. Resin columns were usually left in the field for approximately six month intervals and exchanged in the spring and fall for one to two years. However, IER columns in the Wind River Range and sometimes in the Sierra Nevada study were exposed for an entire year at a time. Previous studies have demonstrated that IER samplers left in the field for an entire year at forested sites in California give equivalent deposition results as the sum of IER columns left out for shorter intervals and the same results as liquid throughfall samples collected from the same sites (Fenn and Poth, 2004). Southeast Alaska sites were only monitored during summer months because access proved challenging in winter due to snow accumulation. We assumed the same rate of deposition across all months to calculate a yearly total, a likely overestimate because air quality is noticeably affected by marine vessels most active in summer (Graw et al., 2001). Despite this potential bias, deposition values at these sites were among the lowest in our dataset.

Within the throughfall IER columns (Fig. 2), precipitation was funneled into a tube filled with Amberlite IRN-150 analytical grade mixed bed (anion + cation) exchange resin beads. After the field exposure the IER columns were shipped to the Pacific Southwest Research Station in Riverside, CA and extracted with 1 *N* KI. Concentrations of NO_3^-, SO_4 and PO_4 were measured using a Dionex high performance ion chromatograph (Thermo Scientific, Sunnyvale, CA, USA) and NH_4 was measured with a TRAACS 800 Autoanalyzer (Tarrytown, NY, USA). Quality control measures included a blank IER tube that was capped and deployed with other tubes on-site for the same length of time, in addition to laboratory standards and analysis of random duplicate samples. Phosphate concentrations were measured to aid in the detection of bird dropping contamination.

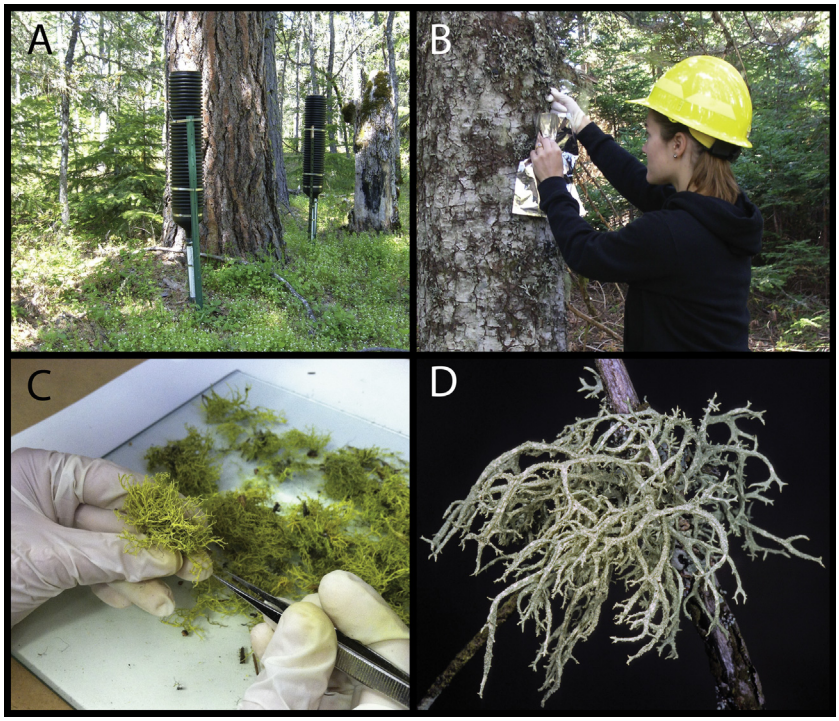

Fig. 2. Photos of throughfall collectors and lichens sampled. (A): Throughfall collectors under *Pinus ponderosa* in the Columbia River Gorge, Oregon (Linda Geiser). (B): *Platismatia glauca* field collection in western Oregon (Linda Geiser). (C): Removing debris from *Letharia vulpina* lichen samples for chemical analysis (Joe DiMeglio). (D): *Evernia mesomorpha* (Stephen Sharnoff).

Lichens were typically collected in the immediate vicinity of throughfall monitors during the summer that IER monitors were removed. However, eight Sierra (Table 2) lichen samples were linked to throughfall monitors post-hoc and were as far as 4 km from the IER monitors and a year before or after IER removal. Lichens from three plots in the Coast to Crest study sites were sampled a year following the throughfall monitoring time period. At sites where lichen and throughfall timing were not synchronous, we expected time periods to reflect fairly similar conditions. At seven sites in Alaska and three in Oregon, two years of throughfall deposition were collected along with two lichen visits; these revisits were averaged across the two years to provide the best estimate for the site.

Lichens were collected at each site across at least a 0.4-ha area, integrating a larger area than the throughfall monitors (Geiser, 2004). Healthy lichen individuals were targeted from tree boles and branches more than 35 m from a road. Whenever lichens were handled, care was used to avoid contamination by wearing gloves and refraining from touching other objects (Fig. 2). Each collection consisted of a single air-tight polyester bag containing individuals of the same species from at least eight trees with a target field (air dry) weight of 20 g. Typically, two to four field replicate bags of a target lichen were collected at each site. Averaging several field replicates allowed the best representation of the population mean for the plot. The target lichens were *Platismatia glauca*, *Letharia vulpina*, and *Evernia mesomorpha*, which grow on conifers and are widely-distributed in wet temperate, dry temperate, and boreal forests, respectively.

Lichens were spread on paper to air dry and carefully hand-cleaned of debris including bark, other lichen species, and necrotic tissue resulting in a final sample weighing approximately 10 g. At the University of Minnesota, St. Paul Research Analytical Laboratory, samples were pulverized, homogenized, dried to constant weight and percent N dry weight was estimated using a LECO FP-528 Nitrogen Analyzer (St. Joseph, MI, USA) except for lichens from southern California, which were sent to University of California, Riverside and analyzed using a Costech elemental analyzer (Valencia, CA, USA). Every tenth lichen sample was split into two lab replicates used to estimate the consistency of lab analyses. For final plot-level estimates, lab replicates were averaged, then field replicates. Both labs analyzed lichen samples with a variety of standards that suggested consistent readings among batches.

We focused on three widespread lichens, *Letharia vulpina*, *Platismatia glauca* and *Evernia mesomorpha* (Fig. 2). Because different species may accumulate different concentrations of N in their thalli under the same deposition conditions we related the other two species to *P. glauca* using regression. Drawing on collections from 47 sites in Oregon and Washington where *L. vulpina* and *P. glauca* were collected, we modeled their relationship using simple linear regression in the software R (R Development Core Team, 2012; Table 3). Only four sites were available to link N concentrations between *E. mesomorpha* and *P. glauca*; however, the regression spanned much of the range of N concentration in *P. glauca* observed in the dataset (0.72–1.2% N) and showed a strong relationship. To ensure that all lichen N concentrations were on a comparable scale, we used these regressions to predict N concentrations in *P. glauca* from those in *L. vulpina* or *E. mesomorpha* where *P. glauca* was not present. Despite the small number of sites available to link *E. mesomorpha* and *P. glauca,* we included those sites in the final model because the relationship between lichen

Table 3
Regression models used to predict N concentration in *Platismatia glauca* from that in other lichen species.

Model	R_{adj}^2	*n*	*p*-Value
N in *P. glauca* = 0.1227 + 0.763 × N in *Letharia vulpina*	0.78	47	<0.001
N in *P. glauca* = 0.320 + 0.496 × N in *Evernia mesomorpha*	0.88	4	0.04

N concentration and throughfall N measurements was consistent with the model developed excluding those sites.

We developed a simple linear regression between lichen N concentration and total inorganic N in throughfall measured by IER collectors at all 84 sites using the function lm in the software R (R Development Core Team, 2012). We graphically explored relationships among variables to determine appropriate transformations. Throughfall variables were considered responses and lichen concentrations predictors because the objective of the calibration regression was to estimate throughfall from lichen N concentration and because preliminary analyses suggested that the within-plot coefficient of variation was greater for throughfall than for lichen measurements.

Climatic 50-year normals (1950–2000) for 19 synthetic bioclimatic indicators were extracted from WorldClim 30-s resolution raster data (Hijmans et al., 2005) for each throughfall monitoring site using ArcMAP v. 10 (ESRI, Redlands, CA, USA). These included mean temperature of the wettest quarter (°C*10) and precipitation seasonality, calculated as the standard deviation of monthly precipitation (mm) divided by the average monthly precipitation (mm) and multiplied by 100. We then used the package leaps in R (R Development Core Team, 2012) to identify the best predictive model for throughfall N.

We calculated throughfall estimates, confidence intervals and prediction intervals for various levels of lichen N using the function predict in the software R (R Development Core Team, 2012). Confidence intervals reflect certainty about the mean throughfall across the study sites at a given level of lichen N concentration. Prediction intervals are wider and reflect the confidence a manager/researcher can have in prediction of throughfall N at a single new site based on a future measurement of lichen N concentration.

We further explored this overall regression by dividing dissolved inorganic N into NO_3-N and NH_4-N. In addition to the inclusive model, we developed a model for the subset of sites in Oregon, Washington, and California, which were more intensively sampled. We also developed models specifically for *Platismatia glauca* and *Letharia vulpina* for this region.

3. Results

Lichen N concentration (% dry weight) varied between 0.44 and 2.1 and throughfall total inorganic N deposition spanned a range from 0.14 to 39 kg/ha/year (Table 2). The ratio of N from NO_3-N versus NH_4-N varied from 0.004 to 1.9 and all the studies incorporated plots with a range of NO_3-N to NH_4-N ratios. The highest deposition estimates were from southern California, the Sierra Nevada, the Athabasca Oil Sands and Oregon and Washington. Southeast Alaska, the Wind River Range, Lake Tahoe and Yosemite spanned shorter deposition gradients with low to moderate deposition levels.

Throughfall total dissolved inorganic N was strongly related to lichen N concentrations (Table 4) on a log–log scale (Fig. 3). Among the 20 potential predictors, lichen N was the best single predictor for throughfall dissolved inorganic N. This model was improved by incorporating the mean temperature of the wettest quarter and precipitation seasonality (Tables 4 and 5). Our model predicted that each 50% increase in the concentration of N in lichen thalli was associated with a 2.25-fold increase in throughfall N deposition.

NH_4-N and NO_3-N were also strongly related to lichen N concentrations; each 50% increase in lichen N was associated with a 2.21-fold increase in NH_4^+ deposition and a 5.48-fold increase in NO_3^-. Neither of these relationships was as strong as the prediction for inorganic N. Restricting the geographic scope to Oregon, Washington, and California allowed development of a better model and climate variables were not significant (Table 4). Confidence and prediction intervals for throughfall were narrowest in models restricted to Oregon, Washington and California (Table 5). Changes in deposition were best measured by lichens when deposition estimates were below approximately 10 kg/ha/year (Fig. 3B). Above this range large increases in deposition were associated with small increases in lichen N concentration and confidence intervals became quite wide (Table 5).

Table 4
Regressions predicting deposition of throughfall N components (kg/ha/year as dissolved inorganic N (DIN), NH_4-N and NO_3-N) from lichen N concentrations at all study sites and in Oregon, Washington and California. *Platismatia glauca* N concentrations include estimates based on N measurements in other lichen species where marked with (*). *P*-values for all slopes are less than 0.001; natural log transformations are indicated by ln.

All study sites*	R_{adj}^2	*n*
ln(DIN) = 1.137 + 2.433 × ln(N in *P. glauca*)	0.58	84
ln(NH_4-N) = 0.664 + 1.951 × ln(N in *P. glauca*)	0.51	84
ln(NO_3-N) = −0.117 + 4.195 × ln(N in *P. glauca*)	0.56	84
All study sites incorporating mean temperature of wettest quarter (Wet °C*10) and precipitation seasonality (Seas%)		
ln(DIN) = −0.768 + 2.000 × ln(N in *P. glauca*)+0.006 × Wet + 0.026 × Seas	0.74	84
Oregon, Washington and California*		
ln(DIN) = 1.193 + 2.300 × ln(N in *P. glauca*)	0.77	57
Oregon and Washington where *P. glauca* was measured		
ln(DIN) = 1.338 + 2.591 × ln(N in *P. glauca*)	0.71	21
Oregon and California where *Letharia vulpina* was measured		
ln(DIN) = 0.886 + 1.968 × ln(N in *L. vulpina*)	0.74	36

4. Discussion

Lichen N concentration can be used to estimate throughfall N deposition, which is defined as the hydrologic flux of N from tree canopies to the forest floor. Such throughfall N deposition estimates can be used to identify potential areas of critical load exceedance. Our study suggests that lichen N concentration is particularly useful for detecting deposition inputs less than 10 kg/ha/year, which coincides with N critical load estimates for many ecosystem components in western North America (Pardo et al., 2011). Reliable estimates of critical loads for inorganic N in western North America vary from 1.5 kg/ha/year wet deposition affecting diatom assemblages in Rocky Mountain alpine lakes (Baron, 2006) to 17.0 kg/ha/year of throughfall deposition causing elevated nitrate leaching in stream water in forested watersheds (Fenn et al., 2008).

Prediction intervals represent our level of confidence in predictions of throughfall DIN at new sites where researchers in the future may measure lichen N concentration. These intervals are conservative, indicating considerable uncertainty in predicted throughfall deposition values. Other biomonitoring studies seldom calculate prediction intervals, making it difficult to compare the uncertainty in our method to others. However, the high R^2 and tight confidence intervals, particularly at low deposition, suggest that the model can be used to reliably identify areas of critical load exceedance. Confidence in prediction at new sites could be improved by taking the average of multiple replicates at each site.

The strong relationship between throughfall N deposition and lichen N concentration is consistent with previous individual studies (Fenn et al., 2007, 2008; Jovan et al., 2012; McMurray et al., 2013) and allows application of the relationship to sites throughout western North America. Our models suggest that lichens passively accumulate both nitrate and ammonium, which parallels Jovan et al.'s (2012) finding that lichen communities in southern California are indifferent to N form. The weaker relationship between lichen N concentration and throughfall N for smaller-scale datasets, such as Yosemite National Park and the Wind River Range, suggests that subtle patterns could be difficult to observe

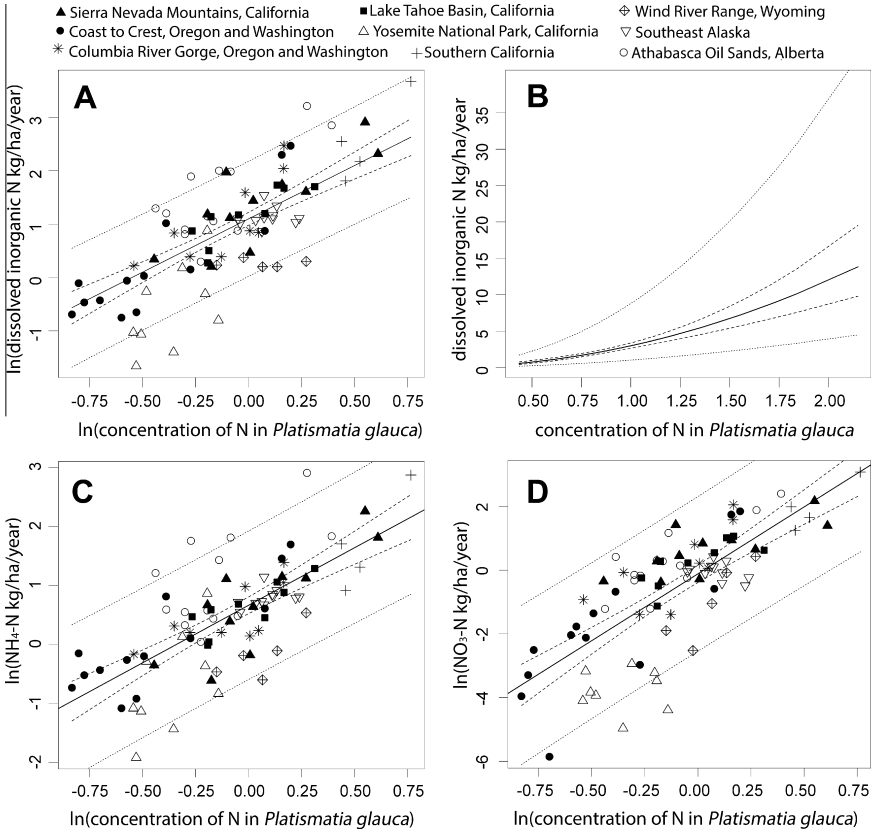

Fig. 3. Regressions between throughfall nitrogen measures and N concentration in *Platismatia glauca* at all study sites. Dashed lines show confidence intervals and dotted lines prediction intervals. (A): Natural log of N concentration in *P. glauca* vs. natural log of dissolved inorganic throughfall N at the mean level of climate covariates. (B): The same relationship as shown in A except transformed back to the original units (with 95% confidence intervals). (C): Natural log of N concentration in *P. glauca* vs. natural log of throughfall NH_4-N. (D): Natural log of N concentration in *P. glauca* vs. natural log of throughfall NO_3-N.

Table 5
Estimates (Est), confidence intervals (CI) and prediction intervals (PI) for throughfall inorganic N deposition (kg/ha/year) at selected levels of N concentration in *Platismatia glauca*. Estimates are based on models in Table 3 across all study sites and restricted to Oregon, Washington, and California; the intervals for all study sites are calculated without the inclusion of covariates, which would decrease their width.

P. glauca N	All study sites			Oregon, Washington, California		
	Est.	CI	PI	Est.	CI	PI
0.6	0.9	0.7–1.1	0.2–3.6	1.0	0.8–1.2	0.4–2.5
1.0	3.1	2.7–3.6	0.8–12.1	3.3	2.9–3.7	1.3–8.1
2.0	8.4	6.4–11.0	2.1–33.1	8.4	6.9–10.1	3.4–20.8
2.5	16.8	11.5–24.6	4.1–68.5	16.2	12.3–21.3	6.4–41.1

using our broad-scale model. Deposition in these areas may be best-estimated using local models.

We present relationships between lichen N concentrations and throughfall N because we presume that the latter is the most integrative estimate of the N to which forest-dwelling organisms are exposed. Most of our sites also included IER collectors in forest openings, which we did not analyze in-depth. However, comparison of bulk deposition in forest openings with throughfall deposition under canopies in low pollution sites in the Pacific Northwest (southeast Alaska, Oregon, Washington, southern Idaho) reveals what appears to be a near universal strong preferential uptake of NO_3-N by the canopy in this region. This typically results in a 75–90% reduction in NO_3-N deposition under canopies compared to bulk deposition or wet deposition in relatively low-deposition sites (e.g., less than 3–4 kg/ha/year; Edmonds et al., 1995; Klopatek et al., 2006; Lovett and Lindberg, 1993; Mark Fenn, unpublished data). For sites where throughfall collectors detected less N deposition than those in the open, we experimented with applying a correction factor to estimate the NO_3^- retained by the canopy (Mark Fenn, unpublished data). Using this correction factor weakened regressions, suggesting that lichens experience the lower levels of NO_3^- where it is absorbed by canopies in low-deposition sites.

Approximately 30–50% of the variation in throughfall N deposition was not represented by our models. Potential sources of remaining variation may include: estimation of throughfall deposition using the IERs; estimation of lichen N concentration; or the biology of how N is accumulated in lichen thalli, especially across different landscapes. Understanding these sources of additional variability may allow future refinement of the model.

We expect estimates of throughfall deposition to vary within a stand depending on tree species composition, leaf area index, and edge effects (Weathers et al., 2001). Collector locations under dominant tree species away from the edge of the canopy make measurements more repeatable. However, integration of spatial variability in canopy conditions may also be desirable because it can allow for a better estimate of stand-level deposition to which forest-dwelling organisms are exposed and may correlate better with lichen N concentrations. A greater number of IER funnels would likely provide better throughfall estimates but would be more costly to establish. Perhaps this investment would be more worthwhile at sites expected to have higher deposition because they show greater variability in deposition estimates.

Variation in estimates of lichen N concentration also likely contributed to the unexplained variability in our model. The necessity to estimate N concentration in *P. glauca* from N measured in other lichen species was unavoidable because no single lichen species

was present at every location. Focusing on the three species used here and further developing the number of co-collected locations, particularly for *Evernia mesomorpha* and *P. glauca,* will improve our ability to relate lichen concentrations among species. Unpublished pilot analyses using *P. glauca* collected from Mt. Hood National Forest (NACSE, 2012) suggested that lichens collected from tree boles incorporate slightly less N than those collected from branches and that tree species can have minor effects on N concentration. Furthermore, we would expect variation related to canopy edge effects, especially in open stands. These sources of variability can be minimized in future studies by collecting large field samples and more field replicates to better represent the population mean N concentration at the plot, particularly where deposition is expected to be high.

By combining thalli across a large plot, lichen collections represented the spatial variability within sites better than the IER funnel collectors; however, lichens were typically collected only once per year whereas throughfall IER measurements monitored deposition continuously for an entire year. The differing temporal integration of deposition may also account for some of the variation not explained by our model.

We found that geographically distant sites (southeast Alaska, Athabasca Oil Sands Region in northern Alberta, and Wind River Range in Wyoming) did not fit the overall model as well as the majority of sites in Oregon, Washington and California; regressions restricting the scope to the latter region performed better than incorporation of the entire study area. Precipitation seasonality and temperature during the wettest quarter of the year were related to this pattern, but our data do not allow a definitive explanation of the cause of this relationship. It could be an artifact of our sampling locations or related to seasonal variability in lichen N incorporation associated with climate characteristics. We could better understand the spatial and climate patterns by additional sampling in the interior west of the United States and Canada. Our sample sites span the range of deposition and climate in western North America fairly well with the exception of the interior southwest; however, inference could be improved with more even sampling of climate conditions present in the region.

The inclusion of precipitation seasonality and temperature during the wettest quarter of the year may also be related seasonal patterns of N assimilation in lichens. Boonpragob et al. (1989) found that *Ramalina menziesii* from southern California leached NO_3^- and NH_4^+ when rinsed with water and that ionic concentrations were highest in summer. Similarly, four of seven sites in Oregon and Washington showed significant intra-annual variability of N concentration in lichen thalli (NACSE, 2012). More detailed measures of seasonal precipitation, temperature, deposition, and lichen N concentration may allow better understanding of this pattern. Because western Oregon, Washington, and California experience most of their precipitation in winter months, seasonal patterns for lichens collected in summer may be most comparable within this subset of the data.

Lichen N concentration biomonitoring offers several advantages as a tool to estimate total N deposition for management purposes (Table 1). It is strongly correlated with passive throughfall data, lichens are readily available at nearly all sites, and lichens can be collected at any time with minimal resource investment. These benefits allow lichens to be monitored at a finer spatial scale and across a broader geographic area than most other methods allow. For example, McMurray et al. (2013) found that lichens were able to detect smaller differences in deposition at a finer spatial scale than that modeled by application of CMAQ (Appel et al., 2011) to 4-km grid cells. Our results suggest that concentration of N in lichens predicts throughfall N deposition within a level of confidence that can be useful for management applications and detection of critical load exceedances.

5. Conclusions and management implications

Lichen N concentration can be used to predict throughfall N deposition in forested ecosystems of western North America. This relationship is strong enough to allow identification of areas that may be exceeding critical loads for sensitive ecosystem components and is especially useful when deposition is less than 10 kg/ha/year, which coincides with estimates of critical loads for many organisms and ecosystem processes (Fenn et al., 2010; Pardo et al., 2011). The lichens used to establish our model are easily recognized, widespread and abundant across western North America and their analysis costs are reasonable. In western North America, this tool is of particular utility to federal land managers who can use deposition estimates to select areas for more intensive monitoring, to influence air quality in their units through planning processes and by providing input in the review of potential new sources or modification of existing sources of air pollution.

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