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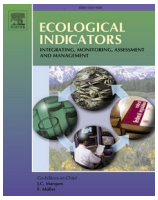

Lichen bioindicators of nitrogen and sulfur deposition in dry forests of Utah and New Mexico, USA

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ABSTRACT

Anthropogenic nitrogen (N) and sulfur (S) deposition can negatively affect ecosystem functions and lichen biomonitors can be a cost-effective way to monitor air pollution exposure across the landscape. Interior dry forests of the southwestern United States face increasing development pressures; however, this region differs from others with well-developed biomonitoring programs in having drier climates and a greater fraction of deposition delivered in dry forms. We measured throughfall N and S deposition at 12 sites in Utah and 10 in New Mexico and co-located collection of 6 lichen species. Throughfall N deposition ranged from 0.76 to 6.96 kg/ha/year and S deposition from 0.57 to 1.44 kg/ha/year with elevated levels near human development that were not predicted by commonly used simulation models. Throughfall N was 4.6 and 1.6 times higher in summer compared with fall-spring in Utah and New Mexico and S deposition was 3.9 and 1.8 times higher in summer. Lichen N and S concentrations ranged from 0.97 to 2.7% and 0.09 to 0.33%. Replicate samples within plots showed high variability in N and S concentrations with within-plot coefficients of variation for N ranging between 5 and 10% and for S between 7 and 15%. In Utah, N and S concentrations in lichen species were correlated with each other in most cases, with R^2 ranging from 0.52 to 0.85. N concentrations in *Melanohalea exasperatula* and *Melanohalea subolivacea* could be correlated with average annual throughfall N deposition in Utah ($R^2 = 0.58$ and 0.31). Those relationships were improved by focusing on deposition in fall-spring prior to lichen sampling in Utah (R^2 for *M. exasperatula, M. subolivacea,* and *X. montana* = 0.59, 0.42, and 0.28). In New Mexico, lichens exhibited greater coefficients of variability within plots than between plots and could not be correlated with throughfall N deposition. In neither study area was S correlated between lichens and throughfall deposition, which may be the result of low S deposition over a narrow deposition range or complex lichen assimilation of S. Lichen biomonitoring for N deposition in the region shows promise, but could potentially be improved by sampling more thalli to reduce within-plot variability, repeated lichen collection synchronized with throughfall changeouts to explore temporal variability, and washing lichen collections to distinguish N and S that has been incorporated by the thalli from dry deposition that may accumulate on lichen surfaces.

1. Introduction

Anthropogenic nitrogen (N) and sulfur (S) deposition have increased compared with historic variability throughout many parts of western North America (Galloway et al. 2004, Clark et al. 2018). Increased N and S deposition affects ecosystems by modifying primary production, herbaceous plant communities (Simkin et al. 2016), forest composition and carbon storage (Thomas et al. 2010, Duarte et al. 2013), and eutrophication or acidification of aquatic systems (Clark et al. 2018). Lichen communities are also affected by N and S deposition, with different suites of species being sensitive to increased N and S deposition (e.g., Geiser et al. 2010, McMurray et al. 2015, Geiser et al. 2019).

Nutrient concentrations in lichens have served as successful bioindicators of deposition in many studies in western North America and

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Europe (Fenn et al. 2008, Jovan et al. 2012, McMurray et al. 2013, Root et al. 2013, Contardo et al. 2020). Lichens are particularly desirable as bioindicators because they lack a cuticle and generally absorb nutrients in proportion to their concentration in the atmosphere (Herzig et al. 1989). As bioindicators, lichens can provide data on a fine spatial scale that signal potential effects on the ecosystem (Geiser et al. 2019) and assist in pinpointing pollution hotspots (Jovan & Carlberg 2007). Lichen biomonitoring, using community composition and elemental concentrations in lichen thalli, has allowed management agencies to identify places where air quality is likely to affect ecosystems and use these maps for planning and management (Pardo and Robin-Abbott 2011, Clark et al. 2018).

Bioindicators are most useful when they can be calibrated with other measurements of deposition. Without calibration, it is unclear what bioindicators are indicating and translating their values to management action becomes challenging. In several studies, lichen N concentrations have been related to instrumented monitoring sites (Root et al. 2015), modeled N from Community Multiscale Air Quality (Byun & Schere, 2006; Geiser et al., 2010), or throughfall dissolved inorganic N (Fenn et al. 2008, McMurray et al. 2013, Root et al. 2013). Throughfall dissolved inorganic N has been particularly well-correlated with lichen N concentrations in seasonally wetter portions of the western United States (Fenn et al. 2007, Jovan et al. 2012, Root et al. 2013), possibly because throughfall integrates multiple kinds of N deposition (Fenn et al. 2009, Jovan et al. 2012). Throughfall ion exchange resin collectors collect wet and dry deposition of N gases and particulates on the surface area of the trees that are washed into collectors during precipitation events (Fenn et al. 2009). This integrated measurement of N deposition may best reflect nutrient conditions of tree-dwelling lichens and other forest organisms. Empirical N deposition measurements from throughfall collectors have been shown to provide a finer-scaled measure of spatiotemporal variability in N deposition compared with broad-scale N deposition models (McMurray et al. 2015, Tulloss & Cadenasso 2015). It is unknown whether the same techniques can be applied in drier coniferous forests of the Interior West Rocky Mountains. Proportions of dry and wet deposition vary significantly by season in the dry coniferous forests of the Interior West (Pardo & Robin-Abbott 2011). Therefore, throughfall and lichens may respond differently to wet and dry deposition in different seasons. Furthermore, the same methods could also be applied to calibrate S deposition (Fenn et al. 2007) but has seldom been tested.

Lichens and other bioindicators are biological organisms, and their accumulation of nutrients from the atmosphere can be complex and variable (Will-Wolf et al. 2017). Lichens accumulate nutrients differently across species and climates (Root et al. 2013, Will-Wolf et al. 2017). Typically, *in situ* lichens are collected to reflect long-term deposition patterns, whereas lichen transplants can help illuminate temporal patterns (Branquinho et al. 2008). When lichen nutrient concentrations vary over time, several potential mechanisms could be at play: 1) changes in deposition, 2) variability in lichen uptake due to metabolic rates, 3) dilution by thallus growth, or 4) wind or precipitation washing nutrients from lichen surfaces. Transplant studies have shown that lichens reflect depositions that are higher or lower than their native habitat within just a few months (Bergamaschi et al. 2007, Branquinho et al. 2010, Contardo et al. 2020). Because N and S are nutrients that lichens actively use, their uptake is likely to vary based on metabolism and thallus needs, which has been shown for N in mosses (Varela et al., 2016). However, deposition held outside the cells can be also leached in precipitation events (Bergamaschi et al. 2007). These temporal dynamics and differences in solubility of nutrients based on their mode of deposition can complicate biomonitoring. For example, near cement industry in a dry climate in Portugal, it was found that lichen accumulation of dust nutrients varied according to the nature of dust as well as the volume and frequency of precipitation (Branquinho et al., 2008).

We focused on the interior southwestern United States, a region that is warmer and drier than other study areas where throughfall and lichen N concentrations have been successfully calibrated (Fig. S1). In this region, biomonitors are needed to evaluate the effects of increasing energy development (Pocewicz et al. 2011) and potential impacts to air quality (McMurray et al. 2013, Edwards et al. 2014, Allen 2016). Our two study areas represent gradients with multiple potential emissions sources, such as area sources from industry and transportation, agriculture, oil and gas development, and proximity to coal-fired power plants. These areas are subject to high levels of particulate matter and ozone that can affect human health (Four Corners Air Quality Task Force, 2007; Baasandorj et al., 2018), as well as N and S emissions that may be deposited in nearby ecosystems. For example, the 2014 emissions inventory in the Uintah Basin of Utah included 13,527 and 40.2 tons/year of NO_x and SO_x associated with oil and gas development (Utah Department of Environmental Quality 2014). Air quality in both study areas is generally poorer in winter in association with inversion events (Four Corners Air Quality Task Force, 2007; Baasandorj et al., 2018) related to elevated levels of NO_x measured at state monitoring stations (Fig. S2). Industrial emissions are largely associated with fossil fuel combustion to run engines on-site as well as venting and flaring (Four Corners Air Quality Task Force 2007, Gorchov Negron et al. 2018).

We established ion exchange resin (IER) deposition collectors, to measure throughfall dissolved inorganic N and S deposition, at 12 sites in Utah and 10 in New Mexico, USA (Fig. 1) near oil and gas energy development in the Uintah and San Juan basins. Collectors were established in the open and under forest canopies and were replaced each fall and spring. We also collected several target lichen species at these sites with three primary hypotheses:

1. N and S in lichens and throughfall will be highest at sites nearest to industrial development (southern sites in Utah and western sites in New Mexico) because N and S will be deposited in greater concentrations proximal to industrial emission sources (McMurray et al. 2013), contrary to broad deposition models (Byun & Schere, 2006; Walker et al., 2019). Unforested open sites will have lower

Fig 1. Overview of locations of 12 plots in Utah and 10 in New Mexico, USA, where lichen collections were co-located with passive IER throughfall collectors (circles). Large cities (pop. > 100,000) within the region (stars) sized relative to population and the two small cities closest to our study areas: Vernal, Utah (pop. = 11,384) and Farmington, NM (pop. = 44,126).

deposition than forest throughfall because they capture less dry deposition accumulated on foliage (Fenn et al. 2013). Deposition will be higher in winter because nearby monitors show higher winter levels of NO_x concentrations in the air associated with inversions (Fig. S2), in contrast to the summer inversions observed in similar studies from California (Fenn & Poth 2004).

2. Lichen samples will have consistent N and S concentrations within sites, allowing differentiation of spatial patterns as has been observed in previous studies (McMurray et al. 2013, Landis et al. 2019). N and S concentrations will be correlated across lichen species (Root et al. 2013, Will-Wolf et al. 2017).
3. Average annual throughfall N deposition will be strongly correlated with lichen N concentrations as has been observed in seasonally wetter parts of western North America (Fenn et al. 2008, Jovan et al. 2012, McMurray et al. 2013, Root et al. 2013). This method will also allow the calibration of lichen S concentrations with throughfall S, which has only been previously tested twice before with mixed results (Fenn et al. 2007, McMurray et al. 2013).

2. Methods

2.1. Study area

Sampling was clustered in two areas in Utah and New Mexico (Fig. 1) that each represented an expected deposition gradient in dry coniferous forests. The 12 Utah sites spanned a north–south gradient from 2,143 to 3,225 m in elevation through the Uinta Mountains north of energy development in the Uintah Basin. Conifer species varied from pinyon-juniper (*Pinus edulis, Juniperus osteosperma, J. scopularum*) woodlands to subalpine Engelmann spruce (*Picea engelmannii*) and subalpine fir (*Abies lasiocarpa*) forests with lodgepole pine (*Pinus contorta*) and Douglas-fir (*Pseudotsuga menziesii*) forests at mid-elevations. Mean annual 30-year average temperatures ranged from −2.8 to 6.9 °C and mean annual precipitation from 350 to 821 mm (Table S1). Monthly precipitation was fairly even across the year with slightly more precipitation in winter and fall (Fig. S1).

The 10 New Mexico sites spanned a shorter elevation gradient from 2,197 and 2,617 m in elevation in north-central New Mexico. Sites formed a gradient from the eastern edge of energy development in the San Juan Basin across the Chama River watershed into the southeastern most part of the San Juan Mountains. New Mexico sites were dominated by Ponderosa pine (*Pinus ponderosa*) with a Gambel oak (*Quercus gambelii*) understory and some pinyon (*Pinus edulis*) and juniper (*Juniperus monosperma*). Mean annual 30-year averages temperatures ranged from 5.1 to 8.3 °C and mean annual precipitation from 350 to 523 mm (Table S1). Monthly precipitation showed a seasonal monsoon pattern with increased precipitation in late summer (Fig. S1).

In Utah, the Bonanza coal plant, in the southeastern part of the study area (Fig. 2) emitted 4578.8 and 813.2 short tons of NO_x and SO_2 in 2018. In New Mexico, the Four Corners and San Juan coal plants are on the western part of the study area (Fig. 3). The Four Corners plant emitted 4322.3 and 1372.4 short tons NO_x and SO_2 in 2018 whereas the San Juan plant emitted 5808.8 and 1130.9 short tons of NO_x and SO_x in 2018 (https://www.epa.gov/egrid/emissions-generation-resource-integrated-database-egrid, 9/1/2020). Oil and gas development require burning natural gas and diesel on-site to run equipment and can be a substantial source of NO_x (Gorchov Negron et al. 2018). Monitors established by the state Department of Environmental Quality measured NO_x in the air throughout the study period (Figs. 2 and 3) and suggest that in both study areas NO_x in the air is elevated in winter (Fig. S2).

2.2. Field sampling

N and S deposition were measured with passive ion exchange resin

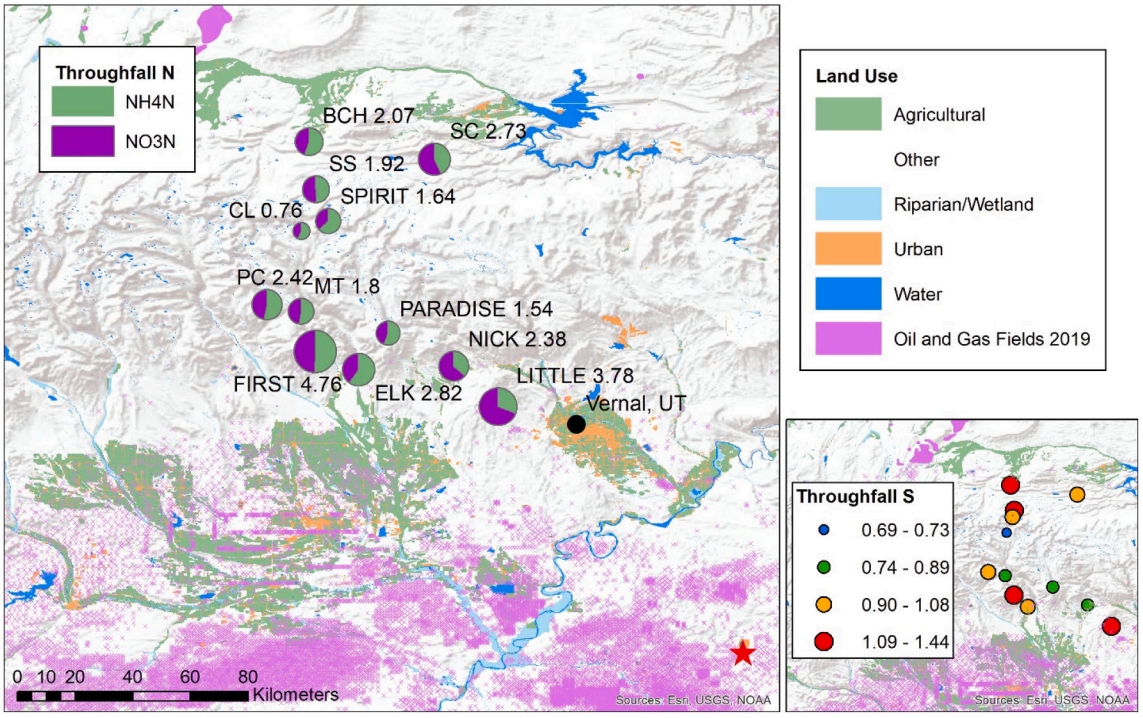

Fig 2. Throughfall N (left) and S (right) deposition (kg/ha/year) at 12 sites in in Utah, USA. Sites are labelled and scaled by total N deposition and divided into the fraction from reduced and oxidized forms. S deposition is scaled and color-coded by total deposition. The large red star in the lower right is the Bonanza coal-fired power plant and the black circle at Vernal is the instrumented monitoring site (Fig. S2). Land use data are from https://gis.utah.gov/data/planning/water-related-land/ (8/27/2020), oil and gas development are from https://gis.utah.gov/data/energy/oil-gas/ (1/19/2020) and https://www.wsgs.wyo.gov/pubs-maps/gis (5/5/2020), and power plant data are from https://www.epa.gov/egrid/emissions-generation-resource-integrated-database-egrid (9/1/2020). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

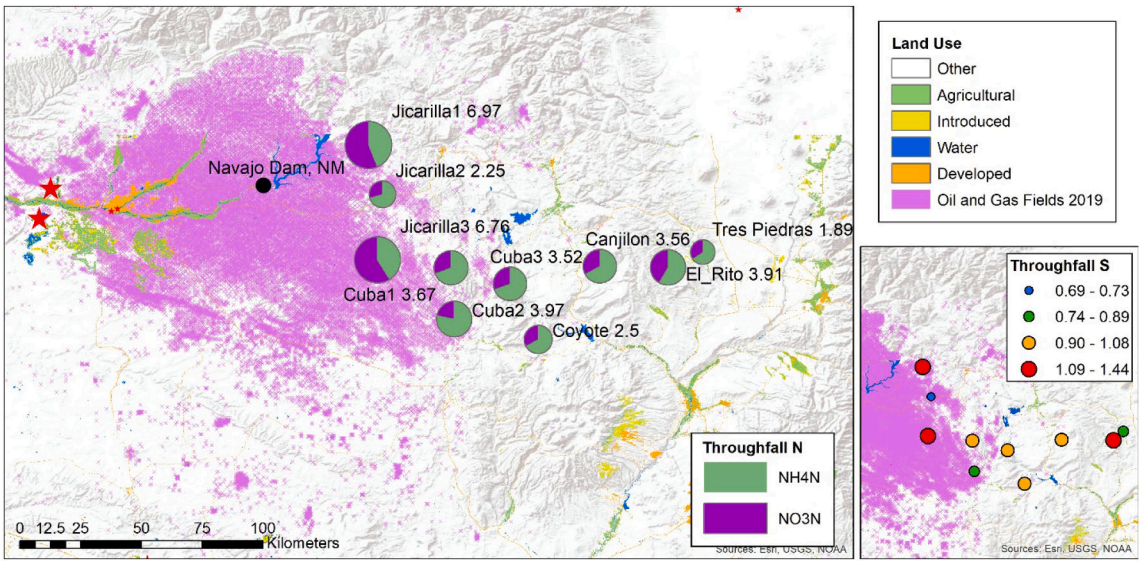

Fig 3. Throughfall N (left) and S (right) deposition (kg/ha/year) at 10 sites in in New Mexico, USA. Sites are labelled and scaled by total N deposition and divided into the fraction from reduced and oxidized forms. S deposition is scaled and color-coded by total deposition. Red stars represent power plants and are displayed proportionally to emissions; the two large red stars on the right are the San Juan and Four Corners coal-fired power plants and the black circle at Navajo Dam is the instrumented monitoring site (Fig. S2). Land use data are downloaded from https://www.usgs.gov/core-science-systems/science-analytics-and-synthesis/gap/science/land-cover-data-download (8/27/2020) and oil and gas development are from http://www.emnrd.state.nm.us/OCD/ocdgis.html (1/19/2020), and power plant data are from https://www.epa.gov/egrid/emissions-generation-resource-integrated-database-egrid (9/1/2020). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(IER) throughfall collectors (Fenn and Poth, 2004; Fenn et al., 2018). In Utah, we deployed 9 replicate throughfall collectors under the dominant conifers for two years beginning in June 2016 with IER column changeouts in spring and fall (Table S2). In New Mexico, we deployed 10 replicate throughfall collectors under *Pinus ponderosa* beginning in October 2014 for three years with IER column changeouts in spring and fall except in the last year (Table S2). At all sites, collectors were positioned approximately halfway between the tree bole and the edge of the canopy. At 5 sites in Utah and 3 in New Mexico, we also installed 6 open-site collectors to measure bulk deposition without the effect of the forest canopy.

The IER columns were filled with Amberlite™IRN 150 Mixed Bed analytical grade ion exchange resin beads pre-rinsed with distilled deionized water. Open or throughfall samples were collected by a polyethylene funnel or snow tube and channeled through the resin column, where ions were retained by the ion exchange resin. The inner diameter of the funnels used for the IER collectors was 21.1 cm and the funnel collectors have a vertical wall 10 cm in height. Snow tubes with an inner diameter of 20.2 cm were inserted into the funnels to allow for snow collection during winter. Snow tubes were 1.0 m in height with the exception of the three lowest-elevation sites in Utah, where snow tubes were 0.5 m in height. Netting was placed on top of the snow tubes at the Utah study sites to minimize squirrels or other animals becoming trapped in the tubes. Bird rings for reducing contamination from bird droppings, modeled after the design of Asman et al. (1982), were installed above the collectors in open areas.

We collected lichen samples for elemental analysis using standard protocols with clean nitrile gloves and ethanol-sterilized titanium tweezers (Will-Wolf et al. 2017), though the small sizes of our lichens dictated smaller samples (target 3 g in the field). Each sample was a composite of at least 10 thalli from across the plot area. In Utah, lichens were collected in June 2017, and target species included *Xanthomendoza montana, Melanohalea exasperatula, Melanohalea subolivacea,* and *Usnea lapponica/hirta*. In New Mexico, *Usnea lapponica/hirta, Xanthomendoza montana,* and *Physcia aipolia/stellaris* were collected in September 2017. At each site, we collected whichever target species were present and at one site in New Mexico, no lichens were collected.

To explore within-site variability and provide more precise estimates, we collected one to three samples of each species per plot, depending on species abundance. Lichen samples were stored in a refrigerator at about 2 °C until they could be processed. They were separated from bark and debris using ethanol-sterilized titanium tweezers under a dissecting scope in ethanol-sterilized polysterene weigh boats (Will-Wolf et al. 2017). We targeted clean, dry weights of 1.0 g; however, some final samples weighed as much as 3 g and one (*U. hirta/lapponica* from CL, Utah) was as low as 0.22 g.

2.3. Laboratory analysis

IER columns were prepared in the laboratory by pouring 25 g of the ion exchange resin beads into PVC tubes (20 cm in length and 1.25 cm I.D.) as an aqueous slurry and then further rinsed with distilled water. After the field exposure periods, IER columns were extracted with 75 ml of 1 N KI, followed by a second extraction with 75 ml of 1 N KI at room temperature. Nitrate and sulfate concentrations in the column extracts were analyzed by ion chromatography (Dionex DX-1600, Sunnyvale, CA) using a procedure modified from Simkin et al. (2004). Ammonium concentrations in the KI extracts were determined colorimetrically (Technicon TRAACS autoanalyser). Atmospheric deposition fluxes were determined by extrapolating from the area of the collector opening and the amounts of inorganic N and S that were extracted from the IER columns (Fenn and Poth, 2004; Fenn et al., 2018). Quality control measures for the IER collectors included a blank IER tube that was capped and deployed on-site for the same length of time, in addition to analysis of laboratory standards and of random duplicate samples (see Fenn et al. 2018 for details). Values of ionic extraction from field blanks were used to blank-correct the sample IER data. For NO_3^-, NH_4^+, and SO_4^{2-}, recovery rates from extracted IER columns were within the range of 94 to 99 percent. Phosphate concentrations in the IER column extracts were also measured using ion chromatography to aid in the detection of samples with potential bird dropping contamination.

Lichen samples were prepared by oven drying at 40C for 24 h and ground to < 100 μm mesh (IKA tube-mill, 1 min grinding time for each sample at 15,000 rpm). Lichen samples were analyzed for total N by a

Leco CHN628 combustion analyzer and for total S by Leco TruSpec combustion analyzers calibrated with Leco standards (e.g., Leco Corp, 2009, 2014). Combustion analyzer performance was evaluated by analyzing reference standards interspersed throughout the sample runs. A barley reference standard (Leco 502–277) was used to evaluate total N data analysis quality and a coal reference standard (Leco 502–670) was used to track total S data analysis quality. Mean +/- standard error measured total N values for the barley standard were 1.75 +/- 0.01%, in excellent agreement with the reference value of 1.74% (reference range = 1.66–1.81). Similarly, mean +/- standard error total S values for the coal standard were 0.530 +/- 0.001%, in excellent agreement with the reference value of 0.532% (reference range = 0.506–562).

2.4. Modeled deposition variables

We compared our empirical deposition estimates with those from national spatial models: Community Multiscale Air Quality (CMAQ) model and the Total Deposition Model (TDep). These are commonly used chemical transport models for estimating atmospheric deposition across broad scales (Walker et al., 2019). CMAQ includes three types of modeling components: a meteorology modeling system, emission models for anthropogenic and natural emissions to the atmosphere, and a chemistry-transport modeling system for the simulation of chemical transformation and fate (Byun and Schere 2006). Simulated atmospheric deposition output is on a 12 × 12 km grid for the continental U.S. TDep is a hybrid using modeled dry deposition data from CMAQ supplemented and adjusted with empirical atmospheric pollutant concentration data from national monitoring networks. For TDep, total deposition is determined as the sum of dry deposition and wet deposition data from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) that is extrapolated more broadly by using precipitation data from Parameter-elevation Regression on Independent Slopes Model (PRISM; Schwede and Lear 2014).

2.5. Statistical analyses

To explore the spatial and temporal patterns of deposition measured by IER samplers, we averaged replicates at each site excluding collectors contaminated with bird droppings and extreme outliers. IER collectors were deployed for slightly different numbers of days (Table S2) and we adjusted measurements based on the proportion of the year that they represented to estimate annual throughfall deposition in kg/ha/year. We compared our deposition estimates with modeled data (TDep and CMAQ) and across years of sampling using simple linear regression in the software R (R Core Team 2018).

To explore the variability of lichen samples within and between plots, we calculated the within-plot coefficient of variation for replicate samples in each plot and compared that to the between-plot coefficient of variation. The most reliable bioindicators would have much lower within-plot variability compared with the differences between plots and the ratio of coefficients of variances between versus within plots would be substantially larger than one. We used regression to explore the N and S concentration across lichen species, allowing for relationships to vary between New Mexico and Utah.

We sought to calibrate lichen N and S concentration with deposition measured by throughfall collectors using regression (R Core Team 2018). Lichen N and S concentrations were averaged across within-plot replicates for each species. Because we expected lichens to have integrated N and S over several years, we averaged annual deposition measurements from the throughfall collectors over the 2–3 years that they were deployed. For each species, we regressed lichen N and S concentration on average annual N and S concentration from throughfall collectors. Because we observed a strong seasonal pattern in deposition, we also examined just the season prior (winter 2016–2017) to lichen sampling for the Utah study sites. Unfortunately, lichens in New Mexico were sampled in September 2017 and throughfall IER collectors were not changed out in spring 2017, so examining the relationship to the season just prior to lichen sampling was not possible.

3. Results

3.1. Spatiotemporal patterns in throughfall deposition

Average annual throughfall N deposition ranged from 0.76 to 4.78 kg/ha/year at the Utah study sites and from 1.91 to 6.96 kg/ha/year at the New Mexico sites (Table S5). Average annual throughfall S deposition ranged from 0.73 to 1.44 kg/ha/year at Utah sites and 0.69 to 1.32 kg/ha/year at the New Mexico sites (Table S5). Average throughfall N deposition was highest in Utah at low elevations (LITTLE and FIRST) along the southern slopes of the Uinta Mountains and lowest in the mountains farther from oil and gas and agricultural development (CL, PARADISE, SPIRIT, Fig. 2). Oxidized N was a larger fraction of total N at low-elevation LITTLE, NICK and SC sites. Low-elevation sites ELK, FIRST, and BCH, had a larger fraction of their total N in reduced forms and were also spatially closer to more agricultural land uses (Fig. 2). N deposition at high elevation sites was generally dominated by reduced N. Throughfall S deposition was fairly low throughout the study area, but highest along the north (BCH and SS) and south slopes (FIRST and LITTLE) of the Uinta mountains. S deposition was lowest at more remote sites in the mountains (CL) with a pattern similar to that for oxidized N (Fig. 2).

In New Mexico, throughfall N deposition was highest at the two sites located adjacent to oil and gas development (Jicarilla 1 and Jicarilla 3, Fig. 3), but lower at a nearby site (Jicarilla 2), and lowest at more remote sites (Coyote and Tres Piedras). N deposition was primarily comprised of reduced N throughout most of the sites with the exception of the two sites adjacent to energy development (Jicarilla1 and Jicarilla3; Fig. 3), where oxidized N contributed more than half of throughfall N deposition. Throughfall S deposition was fairly low throughout the study area, but highest at Jicarilla1 and Jicarilla3, adjacent to oil and gas development. Like oxidized N, S deposition was low at Jicarilla2 and at more remote sites.

Empirically-measured N and S were uncorrelated with TDep estimates for both open and throughfall monitors ($p > 0.30$, Table S5) with the exception of throughfall N, which was negatively correlated with TDep estimates (slope est. = -0.75, $p = 0.0095$). A similar pattern was observed for the CMAQ data with no relationships to open or throughfall N or S ($p > 0.2$) except for a marginal negative relationship between throughfall N and CMAQ-modeled N (slope est. = -1.11, $p = 0.073$).

Open deposition was generally lower than throughfall deposition (Fig. S3) with the exception of two sites for N (BCH and CL). Open and throughfall N measured by IER collectors were uncorrelated ($p = 0.99$), but for S there was a weak correlation (slope est. = 0.67, $p = 0.063$).

There was substantial variability across seasons and years in throughfall N and S deposition (Fig. 4) with summers generally experiencing greater deposition of both N and S, particularly in Utah. In Utah and New Mexico, summer N deposition per month were 4.6 and 1.6 times higher than that in the fall-spring. Similarly, S was 3.9 and 1.8 times higher in summer compared with fall-spring in Utah and New Mexico (Table S4). Reduced N (NH_3, NH_4^+) was also substantially higher in summer as compared with winter (Fig. 4). Throughfall oxidized N (NO_x) deposition was also highest in summers (Fig. 4), whereas NO_x concentrations in the air at nearby monitoring sites were higher in winter for both study sites (Fig. S2).

3.2. Lichen N and S concentrations

Individual lichen sample N and S concentrations ranged from 0.97 to 2.7 and 0.09 to 0.33 percent dry weight and exhibited considerable within-plot variation (Fig. 5). *U. hirta/U. lapponica* had the lowest average N and S concentrations and *X. montana* had the highest average concentrations (Fig. 5). For both N and S, most species showed 5–10%

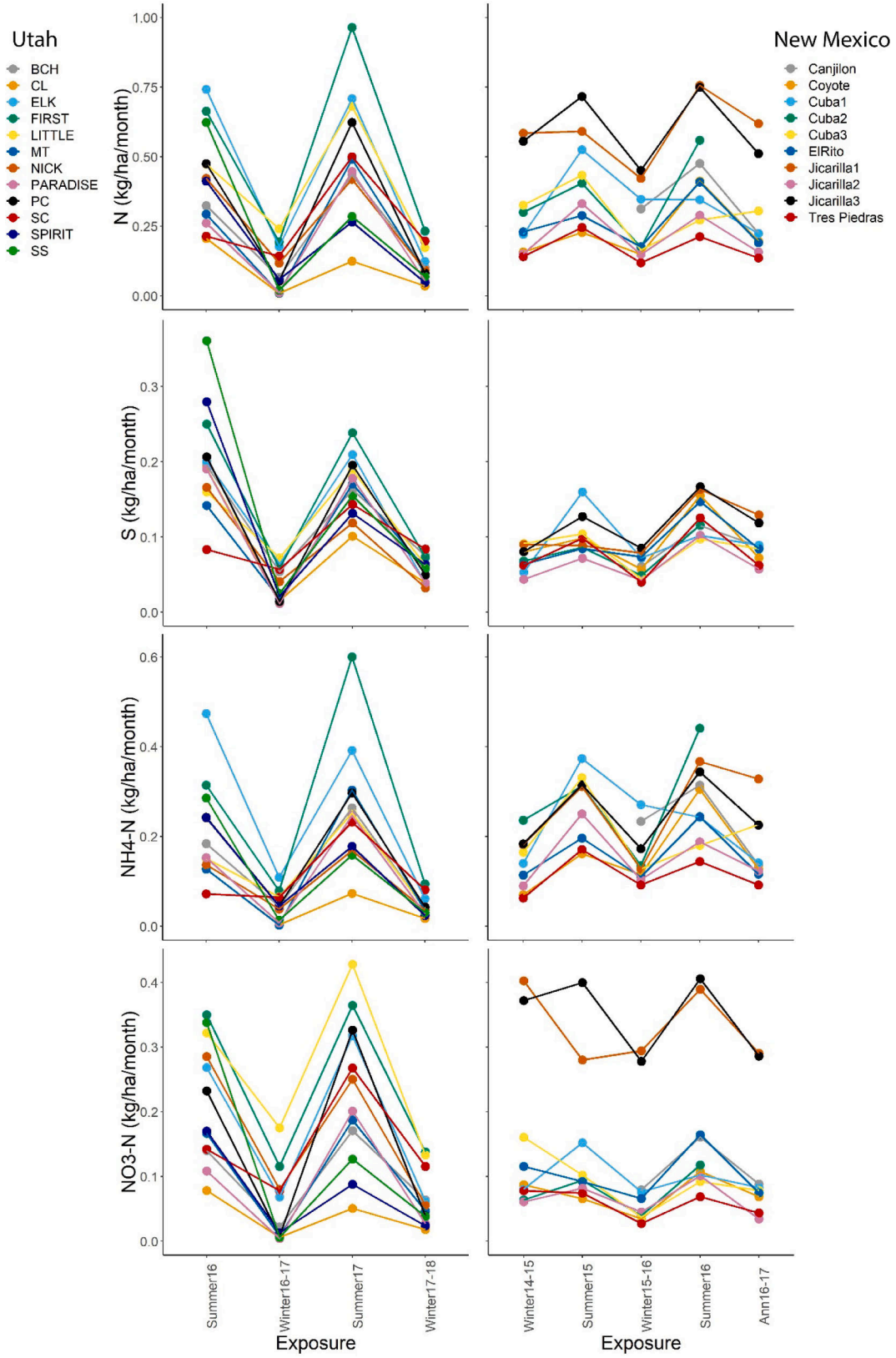

Fig 4. Variability in N and S deposition and reduced and oxidized N deposition measured by passive throughfall monitors across exposure periods at 12 sites in Utah and 10 sites in New Mexico, USA. Exact exposure time frames are in Table S2. Generally, summer exposures were June - October and winter exposures included most of the fall and spring, running from October through the following June. For comparison, deposition for each time period is adjusted to reflect average deposition per month of exposure. In New Mexico, IER columns were not changed out in spring 2017, thus the final measurement represents the annual total.

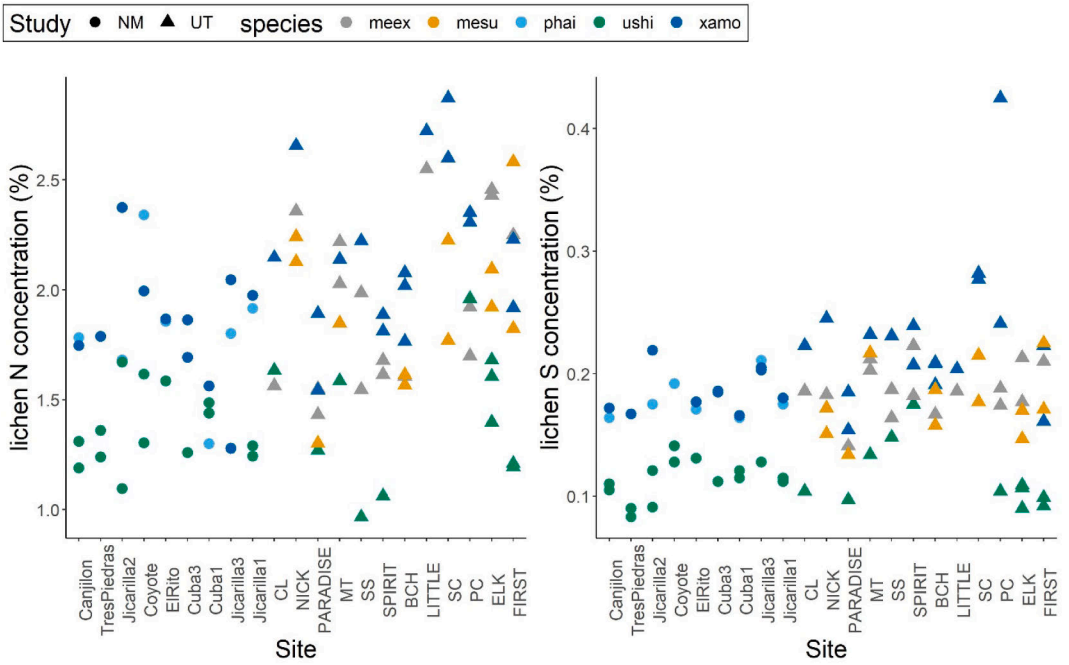

Fig 5. Lichen elemental N and S concentrations (percent dry weight) in 21 plots from New Mexico and Utah for 5 lichen species: *Melanohalea exasperatula* (meex), *Usnea hirta/U. lapponica* (ushi), *Xanthomendoza montana* (xamo), *Melanohalea subolivacea* (mesu), and *Physcia aipolia/P. stellaris* (phai). Plots Canjilon through Jicarilla1 are in New Mexico and CL through FIRST are in Utah.

coefficient of variation within plots (Table 1). In Utah, this was balanced by a>15% coefficient of variation between plots for most species, resulting in a between versus within coefficient of variation greater than one for all species in Utah. However, for New Mexico lichen N concentrations, there was greater coefficient of variation within plots than between plots (Table 1).

Lichen N and S concentrations were correlated across most species in Utah but not in New Mexico (Figs. S4 & S5). In Utah, N concentration in *M. exasperatula* was significantly correlated with N concentration in *X. montana* (slope est. = 0.916, R^2 = 0.62, p = 0.007, Fig. S4) and *M. subolivacea* (slope est. = 0.817, R^2 = 0.85, p = 0.009, Fig. S45). Also in Utah, N concentration in *M. subolivacea* was weakly correlated with *X. montana* (slope est. = 0.638, R^2 = 0.52, p = 0.103, Fig. S4). *M. subolivacea* and *M. exasperatula* in Utah were strongly correlated in their S concentrations (slope est. = 1.000, R^2 = 0.67, p = 0.046, Fig. S5). In neither study area was *U. hirta/lapponica* N or S concentrations correlated with those in other lichen species.

3.3. Relationships between throughfall deposition and lichen concentrations

Lichen N concentrations in Utah were best calibrated with throughfall deposition in the season prior to lichen collection (R^2 for *M. exasperatula, M. subolivacea, X. montana* = 0.59, 0.42, and 0.28; Table 2, Fig. 6). Average annual throughfall N deposition was also

Table 2
Relationships between lichen N concentrations and throughfall N deposition for lichen species *M. exasperatula* (Meex), *M. subolivacea* (Mesu), *X. montana* (Xamo) at 12 study sites in Utah. Only statistically significant relationships are displayed. Average annual throughfall is measured in kg/ha/year and fall-spring throughfall is measured in kg/ha/month and includes October – May (change-out dates in Table S2).

Lichen species	Predictor	Estimates (intercept, slope)	R^2	p
Meex	Average annual throughfall N	1.30, 0.266	0.58	0.0004
Mesu	Average annual throughfall N	1.43, 0.171	0.31	0.062
Meex	Fall-Spring 2016–2017 throughfall N	1.645, 3.817	0.59	0.0003
Mesu	Fall-Spring 2016–2017 throughfall N	1.520, 3.436	0.42	0.0227
Xamo	Fall-Spring 2016–2017 throughfall N	1.950, 2.682	0.28	0.0238

Table 1
Coefficients of variation (*cv*) for lichen N and S levels at 9 sites in New Mexico (NM) and 12 in Utah (UT), USA. For each species, *cv* is calculated within plots, between plots, and the ratio of between versus within. Ratios greater than one indicate greater variance between plots than within them. All species included at least 5 plots per study with within-plot replication except *X. montana* in New Mexico, which had only two plots from which to calculate within-plot *cv* (marked with ~), and *P. aipolia/stellaris* in New Mexico, which had no within-plot replication.

		N			S		
Study	Species	Between *cv*	Within *cv*	Ratio	Between *cv*	within *cv*	Ratio
NM	*P. aipolia/stellaris*	0.171			0.096		
NM	*U. hirta/lapponica*	0.091	0.105	0.864	0.127	0.070	1.806
NM	*X. montana*	0.127	~0.197	~0.644	0.101	~0.004	~25.853
UT	*M. exasperatula*	0.198	0.070	2.843	0.099	0.086	1.156
UT	*M. subolivacea*	0.175	0.105	1.670	0.161	0.129	1.250
UT	*U. hirta/lapponica*	0.239	0.051	4.661	0.243	0.077	3.158
UT	*X. montana*	0.156	0.075	2.094	0.194	0.152	1.274

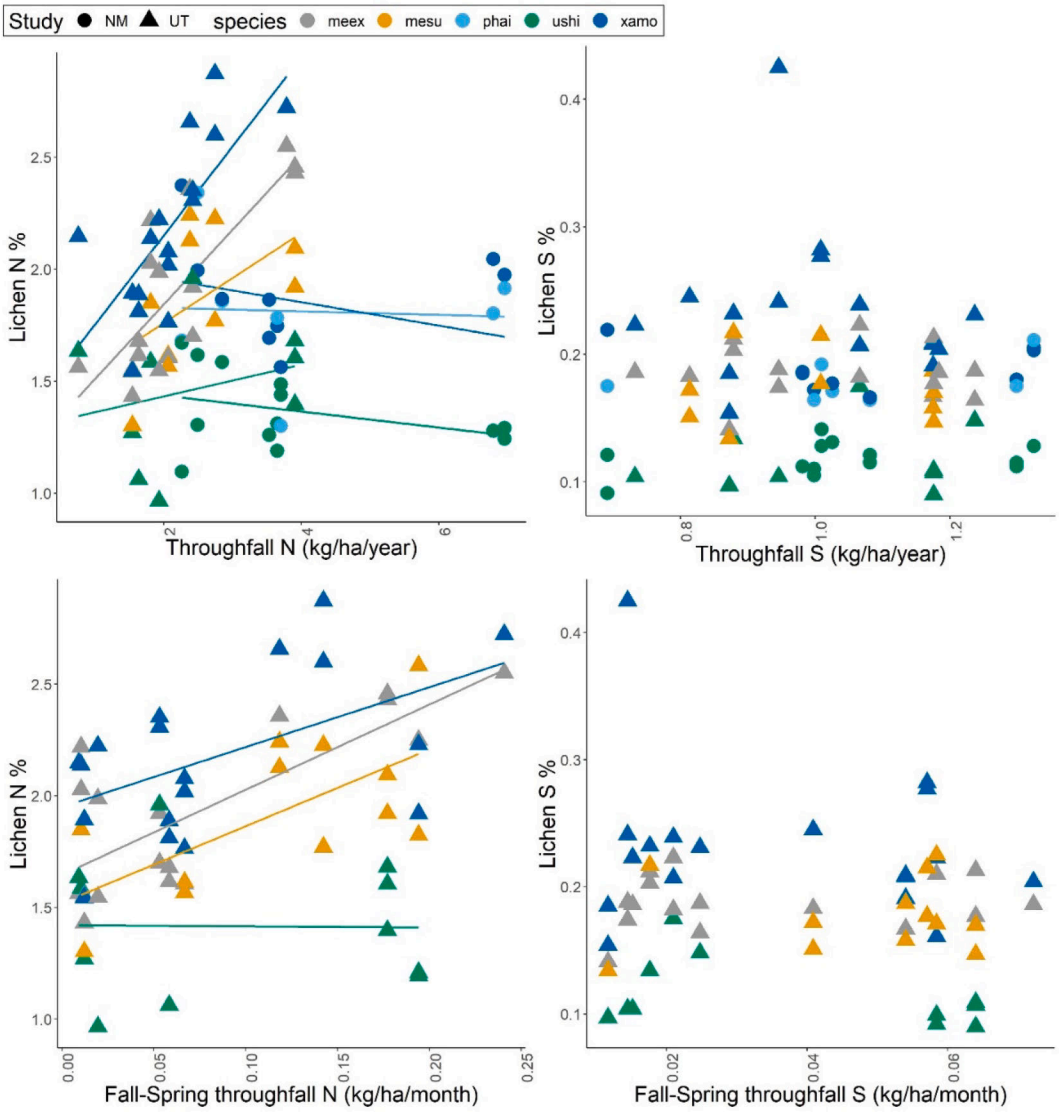

Fig 6. Lichen N and S concentrations (%) related to throughfall deposition by lichen species at 21 plots in Utah and New Mexico. Top panels display annual throughfall measurements. Bottom panels display only the fall-spring preceding lichen sampling in Utah. Regression lines display relationships where some lichen species concentrations have significant correlations with throughfall IER deposition. Relationships that are statistically significant ($p < 0.05$) are described in Table 2.

correlated with two lichen species' N concentrations (R^2 for *M. exasperatula* and *M. subolivacea* 0.58 and 0.31; Table 2, Fig. 6). In New Mexico, none of the lichen N or S concentrations were strongly related to average annual throughfall deposition ($p > 0.10$). Focusing on the year prior to lichen collection (2016–2017) did not substantively strengthen relationships nor did focusing on throughfall measurements in summer 2016. Because New Mexico IER columns were not changed out in spring 2017, measurements for summer 2017 preceding lichen sampling in September 2017 were not available. None of the relationships between lichen S concentrations and throughfall S deposition were statistically significant in either study area (Fig. 6, $p > 0.10$). Log transformations or the exclusion of the unusually high *X. montana* S concentration at the Utah PC site did not qualitatively change the lack of correlation.

4. Discussion

Our study was the first to attempt to calibrate lichen N and S concentrations with throughfall N and S deposition in interior dry forest ecosystems of the southwestern United States, a necessary step for developing lichen biomonitors for management. We found strong spatial and seasonal patterns in throughfall N and S deposition that differed from widely-used deposition models, substantial variability in lichen N and S concentrations within plots and across species, and had the most success calibrating lichen N concentrations with throughfall N using throughfall deposition in the season prior to sampling in Utah. Our N and S gradients (N and S up to 6.97 and 1.44 kg/ha/year), were less pronounced than those in many other parts of North America, such as California, the Pacific Northwest, and eastern forests (Fenn & Poth 2004, Fenn et al. 2007, Root et al. 2013, Geiser et al. 2019), but exceeded estimated N critical loads above which lichen communities are expected to be negatively affected by air quality (Pardo and Robin-Abbott, 2011; McMurray et al., 2013)

4.1. Spatiotemporal patterns in throughfall N and S deposition

Our results suggested throughfall N and S deposition were highest near energy and agricultural development at low elevation in these regions, which was consistent with our first hypothesis. This spatial pattern is similar to that observed for N deposition near energy development in Wyoming, USA (McMurray et al. 2013). Our empirical throughfall N deposition measurements, which were generally higher at

low elevation sites, suggested the opposite pattern from modeled TDep and CMAQ N deposition, which generally modeled more deposition at higher elevations. In forested regions with nearby oil and gas emissions, it may be particularly difficult for deposition models to simulate effects from these dynamic and variable emissions sources. Empirical throughfall measurements can give better spatial resolution of what these ecosystems experience in deposition.

The differences that we observed between open and throughfall monitors are consistent with our hypotheses and previous work, suggesting that throughfall deposition reflects a balance between canopy interception and uptake of deposition and pollutants that are washed from the canopy by precipitation (Fenn et al. 2013). Tree species are likely to intercept and uptake dry deposition differently because they vary in surface area and nutrient needs. This may complicate interpretation of spatial patterns, particularly in Utah where tree distributions necessitated placing throughfall collectors under a variety of conifer species. Lower-elevation sites had smaller pinyon and juniper trees that exhibited lower surface areas as compared with taller spruces and firs with dense canopies at higher elevations. Where we observed higher N and S deposition at low-elevation pinyon-juniper sites (LITTLE, NICK and ELK), it is presumably because of significantly greater air pollution exposure. ELK and FIRST were geographically close to each other but had substantially different forest types; the former was dominated by pinyon and juniper and the latter by fir and Douglas-fir (Table S1). The taller conifers of FIRST likely had greater canopy surface area, which may be responsible for higher throughfall N and S deposition compared with ELK.

Oxidized and reduced N showed different spatial patterns, suggesting differing sources. Oil and gas development typically require diesel or natural gas combustion to operate drilling rigs, hydraulic fracturing equipment, and on-site engines for lifts, dehydrators, compressors, and other equipment (Gorchov Negron et al. 2018). This development contributes to oxidized N and SO_2 deposition (Islam et al. 2020) and is estimated to contribute more than half the NO_x pollution in the Uintah Basin near the Utah sites (Edwards et al. 2014). NO_x can be a precursor for problematic ozone and particulate matter levels during winter inversions in the Uintah Basin and in some places, such as the Wasatch Front in northern Utah, ammonium nitrate can comprise the majority of fine particulate matter (Baasandorj et al. 2018). In Utah, both types of N were elevated in the southwestern part of the study area, close to agricultural and energy development sources; however, the southeastern plots, close to energy development but with less agricultural land use, were elevated in oxidized N but not reduced N. In New Mexico, oxidized N was elevated only at the two sites adjacent to energy development, whereas reduced N was elevated across most of the study area and highest at Cuba2, which is adjacent to ranch lands used for livestock grazing.

Our study showed clear seasonal patterns with greater throughfall deposition of S, oxidized N and reduced N in summer, which was contrary to our expectations. Agriculture and livestock are generally primary sources of NH_4^+ emissions (Fenn et al., 2003), which we may expect to increase in spring, summer, and fall because those are the main growing seasons in both study areas. Furthermore, increased temperatures can increase NH_3 volatilization from soils, fertilizers, and livestock waste (Fan et al., 2011; Laubach et al., 2012). This NH_3 from agricultural sources may contribute to the formation of fine particulate matter ($PM_{2.5}$) containing ammonium nitrate, ammonium sulfate or ammonium chloride (Plautz 2018), which can be transported long distances and contribute to throughfall deposition. Indeed, winter fine particulate matter ($PM_{2.5}$) in urban northern Utah near Salt Lake City was observed to be 57% NH_4^+ or NO_3^- and 8.7% SO_4^{2-} (Baasandorj et al., 2018); this dry deposition may contribute to N and S concentrations in lichens and throughfall. The seasonal synchronicity across study sites and nearby instrumented monitors showing higher NO_x concentrations in winter (Fig. S2) suggest that increased summer throughfall deposition is the result of how these nutrients are deposited rather than emission seasonality.

Weather patterns that affect chemical reactions and dispersion can strongly affect deposition (Pleijel et al. 2016). Deposition may be most likely to reach forests in summer due to transport patterns or because warmer weather facilitates chemical reactions that lead to deposition. When dry deposition sits on the canopy for a longer time before being washed off, there is more opportunity for absorption by foliage (Pleijel et al. 2016). In the New Mexico study area, the pronounced monsoonal pattern with flashy thunderstorms in late summer (Fig. S1) may allow dry deposition to sit in the canopy in spring and early summer, then wash off rapidly such that less of the deposition can be absorbed by understory lichens. However, thunderstorms likely foster greater ion washoff from the canopies and collection by the throughfall samplers. In Utah, the more uniform precipitation throughout the year (Fig. S1) may allow for more consistent absorption by both lichens and throughfall samplers.

4.2. Lichen N and S concentrations

Lichen accumulation of N and S was quite variable within plots but could still be correlated across species in Utah but not New Mexico. Differences in the size and structure of lichen thalli might contribute to variable accumulation across species. In particular, where lichen thallus structure and surface area differ, they may accumulate dry and wet deposition differently. Though we would expect lichens with larger surface areas, such as *Usnea*, to accumulate more nutrients, other studies have also found that in some situations, fruticose lichens accumulate less N or S than foliose species (St. Clair et al. 2002, Will-Wolf et al. 2017, Will-Wolf et al. 2020).

Differential accumulation of dry deposition may also account for the large within-plot variability that we observed because position on the outer vs. inner branches of trees, in more open or closed parts of the forest, or at different heights in the forest canopy, may be associated with more or less dry deposition. The within-plot coefficient of variations for the New Mexico study area were likely too large for lichen N and S concentrations to reliably show subtle differences among plots. Within-plot variability could potentially be reduced to allow distinction among plots by sampling a larger number of lichen thalli to better average across the microhabitats within plots or with more rigorous cleaning protocols (Will-Wolf et al. 2017). Our within-plot coefficients between versus within-plot coefficient of variation ratio for our strongest indicator species (*M. exasperatula*) was 2.8 as compared with 6.9 and 4.0 in nearby Wyoming (McMurray et al. 2013) and>7 in the Athabasca Oil Sands region in northern Alberta (Landis et al. 2019).

The low variability among plots may reflect a short deposition gradient; all sites in both study areas have fairly low S deposition and low to moderate N deposition. Oil and gas development generally have substantially lower SO_x emissions compared with NO_x emissions (Utah Department of Environmental Quality 2014, Baasandorj et al. 2018). With such subtle differences between sites, variability among sites due to different tree canopies may partially obscure the subtle pollution gradient in the Utah study area. In the New Mexico study area, lichen sampling occurred during a season punctuated by flashy thunderstorms that may have washed dry deposition from lichen surfaces. These heavy precipitation events during the sampling period may have contributed to additional variance in lichen nutrient concentrations among plots that was unrelated to total deposition.

4.3. Relationships between throughfall deposition and lichen concentrations

Our results were most successful at calibrating lichen N concentrations with throughfall N deposition observed in the winter prior to sampling in Utah. *M. exasperatula* and *M. subolivacea* were the most successful lichen bioindicators, and their medium-sized foliose growth form is consistent with successful bioindicators elsewhere (Will-Wolf

et al. 2017). This calibration, accounting for 28–59% of lichen N concentration (depending on lichen species), suggests that lichen biomonitoring of N deposition has promise in the region. Other studies in the western United States have found the correlations between lichen and throughfall N with R^2 values from 0.33 to 0.96 (Fenn et al., 2007; McMurray et al., 2013; Root et al., 2013). Combined analysis of throughfall and lichen N concentrations across several studies in western North America suggested that this relationship is modified by climate variables, with different relationships for temperate coastal sites compared with more interior sites, such as the Athabasca Oil Sands region in northern Alberta and Wind River Range in Wyoming (Root et al. 2013). The Wind River sites were climatically most similar to our sites (Fig. S1) but exhibited a stronger correlation between lichen and throughfall N (McMurray et al. 2013), which may be due to the higher summer precipitation in our study areas.

Our calibrations were unsuccessful in New Mexico, which was unexpected based on previous successful application of this technique at other sites across western North America (Fenn & Poth 2004, Fenn et al. 2008, Jovan et al. 2012, McMurray et al. 2013, Root et al. 2013). Because rainfall in the New Mexico study area arrives as summer monsoons, timing of collections, or multiple collections with synchronized throughfall collections may be necessary. In these regions where dry deposition dominates, we may be able to improve biomonitoring by better understanding temporal absorption and leaching dynamics of dust. Washing samples to remove particles held on the surface may allow differentiation between surface dust and nutrients absorbed by the lichens (Bergamaschi et al. 2007). In these inland ecosystems, dry deposition is often dominant (Pardo & Robin-Abbott 2011) and surface dust may represent local or distant soils or particulate matter derived from human development emissions.

Contrary to our expectations, calibrations were unsuccessful for S in both study areas. S deposition has been well-correlated with lichen community composition across western North America with low-risk ecological effects observed when S deposition exceeded 2.3 kg/ha/year (Geiser et al. 2019). Despite the industrial development in our region, S deposition was still lower than this critical load and the lack of effect is likely due to low S deposition throughout the study areas. This is not surprising considering the low levels of SO_2 emissions from oil and gas well activity (Four Corners Air Quality Task Force, Utah Department of Environmental Quality 2014). The nearby Wind River study also did not successfully calibrate lichen S with throughfall S (McMurray et al. 2013). Lichen S and throughfall S were successfully calibrated in the Columbia River Gorge, Oregon, USA, where S deposition included values as high as 6.7 kg/ha/year, (Fenn et al. 2007), suggesting that this relationship can be observed when higher levels of S deposition are in the study area.

Compared with more temperate regions where the majority of lichen biomonitoring work has focused, arid and semi-arid regions may have different dynamics of precipitation that allow lichens to metabolically absorb nutrients, or sporadic rain events may leach surface nutrients (Branquinho et al. 2008). Furthermore, the form of deposition may also affect nutrient uptake. The interior southwestern part of the United States generally receives more of its deposition in dry form compared with the more temperate parts of the country (Pardo and Robin-Abbott 2011) and coarse particles may have less easily-acquired nutrients than fine particles (Bergamaschi et al. 2007, Tretiach et al. 2011). Our finding that lichen N concentration is most strongly correlated with the winter prior to sampling and that most deposition occurs in summer, suggests that the dynamics of rainfall and dry deposition in summer are complicating lichen bioaccumulation. While local dust could be variable from site to site depending on proximity to roads, vegetation cover, and soil composition; long-distance dust is also part of the regional air quality gradient that we want to detect, and dust is known to bring a different suite of nutrients than those found in the native soils (Munroe 2014). Dust can include ammonium nitrate and sulfates in particulate matter (Baasandorj et al. 2018) as well as N and S in fine soil dust (Neff et al. 2008) and thus could contribute to both throughfall and lichen N and S. Our sampling design did not allow us to distinguish the potentially different accumulation of this dust in throughfall collectors or species of lichen. However, the increased deposition in most throughfall collectors compared with open collectors suggests that much of the deposition is in a dry form that is captured by the forest canopy. Furthermore, N and S emissions are sufficiently high in the study regions to result in accumulation of dry-deposited pollutants on the canopy that are subsequently washed off during precipitation or snow melt events in amounts over and above the fraction of these pollutants that are retained by the canopy (Fenn et al., 2018). The differences between deposition measured at our collectors and those modeled for our area may suggest small-scale spatial variability in deposition that is poorly modeled (Tulloss & Cadenasso 2015). We suspect that some of the variance not explained in our models is due to differences in dust accumulation and absorption through the year.

5. Conclusions

We found higher levels of N and S throughfall deposition proximal to industrial development, in contrast to TDep and CMAQ model estimates (Byun & Schere, 2006; Walker et al., 2019). Whereas these two regions experience poor air quality during winter inversions (Four Corners Air Quality Task Force 2007, Baasandorj 2018), we observed deposition to be highest in summer. Lichen N and S concentrations were more variable within plots than had previously been reported (McMurray et al. 2013, Landis et al. 2019). The predominance of dry deposition in the study areas and the timing of rainfall may complicate lichen absorption of nutrients. Increasing within-plot sampling, washing samples, and synchronizing repeated lichen and throughfall collections may lead to stronger relationships between throughfall and lichen nutrient concentrations and more successful development of bioindicators. Despite high within-plot variation, N concentrations could be successfully calibrated with winter or annual throughfall N deposition for three lichen species in Utah. Lichen S concentrations could not be calibrated with throughfall S deposition, which may be the result of high within-plot variability, complexity in lichen nutrient absorption, or a short gradient in S deposition.

Author contributions

HTR, SJ, MF, and JS conceived of the study, developed the design, and secured funding. JS and JH took the lead on establishing field sites in Utah and New Mexico. HTR led fieldwork in Utah. JH and SJ led fieldwork in New Mexico. MF supervised the design and laboratory analysis of IER collectors. MF and MA compiled and quality control checked lab results. HTR led statistical analyses with input from SJ, MA and MF. HTR led writing the manuscripts with input and revisions from all other co-authors.

CRediT authorship contribution statement

HTR: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Project administration, Writing - original draft, Writing - review & editing. **SJ:** Conceptualization, Funding acquisition, Investigation, Methodology, Writing - review & editing. **MF:** Data curation, Methods, Writing - review & editing. **MA:** Data curation, Methods, Writing - review & editing. **JH:** Investigation, Writing - review & editing. **JDS:** Conceptualization, Funding acquisition, Investigation, Project administration, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecolind.2021.107727.

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