



Estimating nitrogen risk to Himalayan forests using thresholds for lichen bioindicators

Christopher J. Ellis^{a,*}, Claudia E. Steadman^b, Massimo Vieno^c, Sudipto Chatterjee^d, Matthew R. Jones^c, Sidharth Negi^e, Bishnu Prasad Pandey^f, Himanshu Rai^g, Dendup Tshering^h, Gothamie Weerakoonⁱ, Pat Wolseleyⁱ, David Reay^b, Subodh Sharma^j, Mark Sutton^c

^a Royal Botanic Garden Edinburgh, Edinburgh, UK

^b School of Geosciences, University of Edinburgh, Edinburgh, UK

^c Centre for Ecology and Hydrology, Bush Estate, Midlothian, UK

^d TERI School of Advanced Studies, Delhi, India

^e Uttarakhand Youth and Rural Development Centre, Ghaziabad, India

^f Department of Chemical Science and Engineering, Kathmandu University, Kathmandu, Nepal

^g Centre of Advanced Study in Botany, Banaras Hindu University, Varanasi, India

^h Sherubtse College, Royal University of Bhutan, Kanglung, Bhutan

ⁱ Natural History Museum, London, UK

^j Department of Environmental Science and Engineering, Kathmandu University, Kathmandu, Nepal

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ABSTRACT

Himalayan forests are biodiverse and support the cultural and economic livelihoods of their human communities. They are bounded to the south by the Indo-Gangetic Plain, which has among the highest concentrations of atmospheric ammonia globally. This source of excess nitrogen pushes northwards into the Himalaya, generating concern that Himalayan forests will be impacted. To estimate the extent to which atmospheric nitrogen is impacting Himalayan forests we focussed on lichen epiphytes, which are a well-established bioindicator for atmospheric nitrogen pollution. First, we reviewed published literature describing nitrogen thresholds (critical levels and loads) at which lichen epiphytes are affected, identifying a mean and confidence intervals based on previous research conducted across a diverse set of biogeographic and ecological settings. Second, we used estimates from previously published atmospheric chemistry models (EMEP-WRF and UKCA-CLASSIC) projected to the Himalaya with contrasting spatial resolution and timescales to characterise model variability. Comparing the lichen epiphyte critical levels and loads with the atmospheric chemistry model projections, we created preliminary estimates of the extent to which Himalayan forests are impacted by excess nitrogen; this equated to c. 80–85% and c. 95–98% with respect to ammonia and total nitrogen deposition, respectively. Recognising that lichens are one of the most sensitive bioindicators for atmospheric nitrogen pollution, our new synthesis of previous studies on this topic generated concern that most Himalayan forests are at risk from excess nitrogen. This is a desk-based study that now requires verification through biological surveillance, for which we provide key recommendations.

1. Introduction

The Himalaya extends longitudinally east-west over c. 2500 km, from western Afghanistan, through Pakistan, northern India, Nepal, and Bhutan. It merges eastwards into the Hengduan mountains, is bound northwards by the Qinghai-Tibet plateau, forming a part of the greater Tibet-Himalaya-Hengduan region. Uplifted during c. 50 million years by

thrust faulting of the Indian and Eurasian tectonic plates, the mountain building process has acted as a catalyst for evolutionary diversification (Manish and Pandit, 2018; Mosbrugger et al., 2018). The mid-altitude Himalaya, up to a treeline of c. 3500–4900 m (west-to-east), is dominated by forest, with longitudinal patterning from endemic *Rhododendron-Quercus* vegetation towards the east (precipitation >4000 mm), through to drought-tolerant Euro-Mediterranean vegetation in the west

* Corresponding author at: Royal Botanic Garden Edinburgh, 20A Inverleith Row, Edinburgh EH5 2PR, UK.

E-mail address: c.ellis@rbge.org.uk (C.J. Ellis).

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(<1000 mm) (Singh and Singh, 1987). Of major importance, these biodiverse forests deliver ecosystem services and goods around which Himalayan communities have built their cultural identities, local economies and associated livelihoods (cf. Joshi and Joshi, 2019; Joshi and Negi, 2011; Måren et al., 2014).

The Himalaya is bounded to the south by the Indo-Gangetic Plain (IGP), an agricultural region that is home to c. 400 million people. Intensification of farming has driven increased inputs of manure, slurry, and urea-based fertiliser (cf. Sigurdarson et al., 2018; Sommer and Hutchings, 2001) such that the IGP now has among the highest global concentrations of atmospheric ammonia (NH₃) (Clarisse et al., 2009; Van Damme et al., 2018; Warner et al., 2017). The release of ammonia leads to a nitrogen-enriched air mass (cf. Asman et al., 1998; Dragosits et al., 2008) that is pushed northwards by monsoon weather patterns to concentrate against the forested south-facing aspects of the Himalaya (Wang et al., 2020). Above certain thresholds an excess of atmospheric nitrogen becomes a pollutant with negative consequences for biodiversity and ecosystem function (Bobbink et al., 2002; Dise et al., 2011; Pardo et al., 2011), generating concern for sustainable delivery of forest ecosystem services and goods. To better understand this pollution risk, we estimated the extent to which Himalayan forests are exposed to excess nitrogen.

The first key decision was the forest attribute used for estimating when atmospheric nitrogen is in excess, and for this we adopted lichen epiphytes. Lichens have been consistently highlighted in European and North American impact studies as among the most sensitive bio-indicators for atmospheric nitrogen pollution (Bobbink et al., 2002; Cape et al., 2009a; Pardo et al., 2011). We reviewed previous studies that identified nitrogen thresholds at which lichen epiphytes are affected (critical levels for gaseous ammonia concentrations and critical loads for total nitrogen deposition), taking a broad approach across continents and ecological regions to calculate a mean and confidence intervals. A second key decision concerned the nitrogen environment used to recognise exceedance of lichen epiphyte critical levels and loads. We used atmospheric chemistry models specifically relevant to the lower atmosphere (<50 m), and therefore epiphytes, characterising variability by using different models with contrasting spatial resolution and timescales.

Our approach allowed us to cautiously estimate the extent to which Himalayan forests are impacted by atmospheric nitrogen pollution, defined as the area over which the lichen epiphyte critical levels and loads are exceeded. We discuss the potential consequences of our estimates, while recognising a need for direct confirmation. We therefore offer considerations geared towards systematic surveillance of Himalayan lichen epiphytes, as regional bioindicators complementary to their employment across Europe and North America (Asta et al., 2002; Matos et al., 2017).

2. Materials and methods

(a) Critical levels and critical loads

We undertook a review of the published literature that has reported critical levels and critical loads for atmospheric nitrogen pollution using lichens. Throughout 2020, and finally on 1st March 2021, we performed an unrestricted (1900–2020) Boolean search in Web of Science (<https://clarivate.com/products/web-of-science/>) using the topic terms: [lichen AND critical AND (load OR level) AND nitrogen]. We reviewed the abstracts of all papers that were recovered, targeting those that reported on critical levels (concentrations) and loads (deposition amounts/rates) with lichen epiphyte diversity as a response. For these we tabulated: (i) the nitrogen type under investigation (e.g. ammonia concentration, total or partial deposition, etc.), (ii) the critical level or load, (ii) how the nitrogen environment was quantified (e.g. whether modelled, interpolated or measured), (iii) how the lichen response was delimited, and (iv) the citation.

In situations where critical levels or loads had been established for the same nitrogen type by five or more different studies, these values were bootstrapped (10,000 permutations) to generate a central estimated mean with confidence intervals at the 2.5% and 97.5% quantiles. If critical levels or loads were defined by a range, then the median was selected.

(b) Regional mapping

First, we combined a map of national boundaries for Afghanistan, Pakistan, India, Nepal and Bhutan, with percent forest cover (including all types; coniferous, deciduous and mixed) observed by the European Space Agency's Copernicus land cover classification at 100 m grid resolution for the year 2015 (Buchhorn et al., 2020a; Buchhorn et al., 2020b). We then delimited this mapping through sub-selection of internal administrative boundaries, which were merged to focus on the transboundary extent of Himalayan forests.

Second, for this same region, we overlaid three contrasting projections of nitrogen atmospheric chemistry. These projections included two different atmospheric chemistry models, with contrasting spatial resolution and timescales, to provide estimates of nitrogen specifically for the lower atmosphere (<50 m) and relevant to lichen epiphytes. We used the EMEP MSC-W model (Simpson et al., 2012) that was originally developed and tested within Europe and the UK (cf. Fagerli and Aas, 2008; Vieno et al., 2009), though which has recently been successfully applied globally (Ge et al., 2021). Projection was at 0.33° × 0.33° resolution, using emissions data for 2010 (HTAP v. 2 global emissions dataset: Galmarini et al., 2017; Janssens-Maenhout et al., 2015) with 2015 WRF meteorology (Powers et al., 2017; Skamarock et al., 2019), and also at 0.11° × 0.11° resolution, using the same emissions data but with 2018 WRF meteorology. EMEP MSC-W is implemented for discrete years at high spatial resolution and was complemented by UKCA-CLASSIC, which has lower spatial resolution but facilitates averaging of pollution dynamics across multiple years, having been developed and tested globally (Morgenstern et al., 2009; O'Connor et al., 2014). Projection was at 1.25° × 1.875° resolution using emissions data for 2008 (HTAP v. 2 global emissions dataset: Galmarini et al., 2017; Janssens-Maenhout et al., 2015) coupled to the UK Met Office Unified Model (UM) 8.4, with Global Atmosphere 4.0 (GA4.0) (Walters et al., 2014) implemented using sea-surface temperatures and greenhouse gases for a 10-year average climate centred on the year 2000. Inter-comparison of different atmospheric chemistry models is beyond the scope of this work, but we note that the models used here encompass different chemistry schemes, such as in their aerosol deposition velocities, treatment of nitrate etc., as well as their different spatial resolution and timescales.

Third, we compared the bootstrapped mean and confidence intervals for critical levels and loads (see *Critical levels and critical loads*, above), with the three projections of nitrogen atmospheric chemistry, to estimate the extent to which Himalayan forests are impacted by excess nitrogen. However, we also wanted to know how this estimated extent might be sensitive to alternate definitions of forested land, and we made estimates of nitrogen impacts for the total area over which forest was observed, as well as for subsets of 100 m grids with >10%, >30%, >50%, >70% and >90% forest cover.

3. Results

The initial Web of Science search identified 111 published papers; many were secondary sources that had applied previously established critical levels and loads, while 16 reported either primary research or reviews identifying nitrogen critical levels and loads based on a shift in lichen epiphyte diversity (Table 1). Three papers reported data relevant for critical levels (e.g. gaseous ammonia, particulate nitrogen as PM_{2.5}, concentration of ammonium in wet deposition), eight for critical loads (e.g. total and throughfall nitrogen, or total dry or wet deposited

Table 1

Details relating to 14 primary research papers and two reviews that used lichen epiphytes as bioindicators for atmospheric nitrogen pollution, a sample of which were used here to estimate the bootstrapped mean and 2.5% and 97.5% intervals for the critical level of ammonia ($\mu\text{g m}^{-3}$) and the critical load for total bulk nitrogen deposition ($\text{kg N ha}^{-1} \text{yr}^{-1}$).

Target pollutant				
Impact value	Quantification	Response	Ecological context	Reference
Ammonia air concentration (CLE)				
2.6 $\mu\text{g m}^{-3}$	Measured every two weeks over one year; downwind from point source (cattle barn)	First point of difference (relative to control) for species richness and LDV of all species, oligotrophic, mesophytic or nitrophytic species	Spain: Mediterranean holm-oak forest	Aguillaume et al. (2017)
< 1.9 $\mu\text{g m}^{-3}$	Measured monthly over one year; downwind from point source (cattle barn)	First point of difference (relative to control) for LDV of oligotrophic or nitrophytic species	Portugal; semi-natural evergreen Mediterranean cork-oak woodlands	Pinho et al. (2012)
1 $\mu\text{g m}^{-3}$	Review of eight studies	Synthesis of the no effect concentration from response curves of multiple gradient studies	Pan-European; UK, Italy, Portugal, Switzerland	Cape et al. (2009a)
1 $\mu\text{g m}^{-3}$	Measured (controls), and calculated from modelled data by transfer function (city)	Shifted frequencies of acidophytic indicator species	Finland; boreal forest	Manninen (2018)
0.69 $\mu\text{g m}^{-3}$	measured for three discrete sample periods over the course of a year	First point of difference (relative to control) for species richness and LDV of oligotrophic or nitrophytic species	Portugal; semi-natural evergreen Mediterranean cork-oak woodlands	Pinho et al. (2014)
Total nitrogen deposition (CLO)				
<26 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Modelled for gridded landscape	Last unaffected point (relative to control) for LDV of oligotrophic or nitrophytic species	Portugal; semi-natural evergreen Mediterranean cork-oak woodlands	Pinho et al. (2012)
11–18 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Modelled for gridded landscape	Shifted community composition with respect to stemflow and bark covariables	Britain; oceanic oakwoods	Mitchell et al. (2005)
10–15 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Multiple sources	Literature review; expert assessment of impact on lichens	Pan-European	Bobbink et al. (2002)
10 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Interpolated for gridded landscape	Point at which number of foliose lichen species declines	Southern Ontario	Miller and Watmough (2009)
5 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Modelled for gridded landscape	Point at which percent contribution to species richness of oligotrophic , mesotrophic and/or eutrophic species was impacted (decline by 30–41% compared to control)	North western United States	Geiser et al. (2010)
4.3–5.7 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Modelled for gridded landscape, integrated with monitored wet and dry deposition	Changed community richness and composition, with a focus on species grouped by their nitrogen optima, and functional trait groups, e.g. cyanolichens	North eastern United States	Cleavitt et al. (2015)
^a 3.5 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Modelled for gridded landscape	^a 20% decline in species richness	Continental United States	Geiser et al. (2019)
^b 3.1 $\text{kg N ha}^{-1} \text{yr}^{-1}$		^b 20% decline in oligotrophic species richness		
^c 1.9 $\text{kg N ha}^{-1} \text{yr}^{-1}$		^c 20% decline in forage (hair lichen) abundance		
^d 1.3 $\text{kg N ha}^{-1} \text{yr}^{-1}$		^d 20% decline in cyanolichen abundance		
Throughfall nitrogen deposition (CLO)				
4 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Inferred by transfer function from thallus %N	Changed community structure in terms of diversity (alpha-, beta- and gamma), and composition, interpreted as dominance/abundance of oligotrophic and eutrophic species	Northern Rocky Mountains	McMurray et al. (2015)
3.1 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Measured for 1–5 years, with a control site cross-referenced against thallus %N	Proportional abundance determining point of decline in macrolichen acidophytes , neutrophytes, and nitrophytes; and acidophyte dominance	California	Fenn et al. (2008)
2–3 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Inferred by transfer function from thallus %N	Shifted frequencies of acidophytic indicator species	Finland; boreal forest	Manninen (2018)
2.4 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Measured daily and accumulated to annual value	Last unaffected point (relative to control) for species richness, evenness, percent macrolichens , percent oligotrophic species, and percent oligotrophic-macrolichens	Pan-European; 10 countries	Giordani et al. (2014)
1.54 $\text{kg N ha}^{-1} \text{yr}^{-1}$	Inferred by transfer function from thallus %N	Changed community composition, at the point of transition from oligotrophic to eutrophic species	North western United States	Root et al. (2015)
Particulate nitrogen PM_{2.5} (CLE)				
0.51 $\mu\text{g N m}^{-3}$	Measured nitrate and sulphate (assumed to be balanced by NH_4); every 3 days over a 24 h period for 8 years	Point at which percent contribution to species richness of oligotrophic , mesotrophic and/or eutrophic species was impacted (decline by 30–41% compared to control)	North western United States	Geiser et al. (2010)
0.5 $\mu\text{g N m}^{-3}$	Modelled for gridded landscape, integrated with measured values	Shifted frequencies of acidophytic indicator species	Finland; boreal forest	Manninen (2018)
0.37 $\mu\text{g N m}^{-3}$	Measured nitrate and sulphate (assumed to be balanced by NH_4); every 3 days over a 24 h period for 3 years	Changed community composition, at the point of transition from oligotrophic to eutrophic species	North western United States	Root et al. (2015)
0.26–0.33 $\mu\text{g N m}^{-3}$	Measured nitrate and sulphate (assumed to be balanced by NH_4); every 3 days over a 24 h period for 1 year	Changed community composition, at the point of transition where sensitive species become absent	North western United States	Glavich and Geiser (2008)
Nitrate air concentration (CLE)				
≤ 5 $\mu\text{g N m}^{-3}$		Shifted frequencies of acidophytic indicator species	Finland; boreal forest	

(continued on next page)

Table 1 (continued)

Target pollutant		Response	Ecological context	Reference
Impact value	Quantification			
Wet deposited ammonium ion concentration (CLE) 0.037–0.048 mg L ⁻¹	Modelled for gridded landscape, integrated with measured values Monitored NH ₄ ⁺ concentration in wet deposition, over 4 years	Changed community composition, at the point of transition where sensitive species become absent	North western United States	Manninen (2018) Glavich and Geiser (2008)
Dry Nitrogen Deposition (CLO)* 3.4 kg N ha ⁻¹ yr ⁻¹	Modelled for gridded landscape	Point at which percent contribution to species richness of oligotrophic , mesotrophic and/or eutrophic species was impacted (decline by 30–41% compared to control)	North western United States	Geiser et al. (2010)
Wet nitrogen deposition (CLO)* 1.6–2 kg N ha ⁻¹ yr ⁻¹	Modelled for gridded landscape	Point at which percent contribution to species richness of oligotrophic , mesotrophic and/or eutrophic species was impacted (decline by 30–41% compared to control)	North western United States	Geiser et al. (2010)
Cumulative wet nitrogen deposition (CLO) 60–80 kg N ha ⁻¹	Modelled for gridded landscape, integrated with monitored wet and dry deposition	Changed community richness and composition, with a focus on species grouped by their nitrogen optima, and functional trait groups, e.g. cyanolichens	North eastern United States	Cleavitt et al. (2015)

Notes: CLE = Critical level; CLO = Critical load; LDV = lichen diversity value (cf. Asta et al., 2002); where several response types were tested, the selected (most sensitive) response type can be identified in bold. *Critical loads defined based on maximum levels of dry or wet deposition respectively. Such differences in indicators highlight the importance of indicator reference in the primary studies (Sutton et al., 2003).

inorganic nitrogen), and five reported for both critical levels and loads (Table 1).

Seven of the studies were European and eight were North American, representing various scales and ecological regions, from continental (Geiser et al., 2019; Giordani et al., 2014; Glavich and Geiser, 2008) to regional landscapes from southern European Mediterranean forest (Aguillaume et al., 2017; Pinho et al., 2014; Pinho et al., 2012) to oceanic north-western or boreal Europe (Manninen, 2018; Mitchell et al., 2005). A range of contrasting methods had been used. Two papers derived their results from reviews; one based on narrative review, developing critical loads for total nitrogen deposition (kg N ha⁻¹ yr⁻¹) for the period up to 2002 (Bobbink et al., 2002), and another calculating the 'no effect concentration' for an ammonia critical level (μg m⁻³) by integrating response curves from eight previous studies up to 2009 (Cape et al., 2009a, see also Cape et al., 2009b; Sutton et al., 2009). Where primary data were sampled, the methods varied from the use of point-source pollution gradients to isolate a given factor, such as for ammonia (Aguillaume et al., 2017; Pinho et al., 2012), through to distributed sampling across entire landscapes, which was often accompanied by the analysis of covariables such as for climate or forest structure (Geiser et al., 2010; Geiser et al., 2019; McMurray et al., 2015; Root et al., 2015).

The studies also used different types of lichen response, including representation of a priori lichen bioindicators grouped as oligotrophic/acidophytic or nitrophytic species (Aguillaume et al., 2017; Fenn et al., 2008; Geiser et al., 2010; Pinho et al., 2014; Pinho et al., 2012), through to a more broadly encompassing shift in community structure such as might be interpreted from an ordination diagram (Cleavitt et al., 2015; Mitchell et al., 2005; Root et al., 2015), or using a hybrid approach (Table 1). Taking a scaled perspective on pollution risk, Geiser et al. (2019) used different responses (total species richness, sensitive species richness, 'forage lichen' abundance and cyanolichen abundance), and identified critical loads for each of these, at the points where they declined by 20%, 50% and 80% from a baseline.

There were five independent estimates for three nitrogen pollutant types: the critical level for ammonia concentration (μg m⁻³), and the critical load for total nitrogen deposition (kg N ha⁻¹ yr⁻¹) and total throughfall nitrogen deposition (kg N ha⁻¹ yr⁻¹). We created bootstrapped estimates for the critical level of ammonia and the critical load for total nitrogen deposition because these matched with the physical models of nitrogen atmospheric chemistry. The mean value of an ammonia critical level was 1.44 μg m⁻³, with the boundaries of the

upper 97.5% and lower 2.5% at 2.08 μg m⁻³ and 0.88 μg m⁻³, respectively, while the mean value for the critical load of total nitrogen deposition (wet and dry, reduced and oxidised) was 8.26 kg N ha⁻¹ yr⁻¹, with the boundaries of the upper 97.5% and lower 2.5% at 13.21 kg N ha⁻¹ yr⁻¹ and 4.24 kg N ha⁻¹ yr⁻¹, respectively.

A projection for the total area of Himalayan forests extended over 35,892,992 ha, declining through to dense forest (>90% forest per 100 m grid cell) which extended over 8,761,285 ha (Table 2; Fig. 1A). Based on the critical levels and loads established for lichen bioindicators, we estimate that most Himalayan forests are currently being impacted by excess atmospheric nitrogen, irrespective of the different nitrogen atmospheric chemistry model used or the alternate definitions of forest cover (Table 2). We highlight the following two points.

First, there is an interaction between the density of forest cover (increasing towards the eastern Himalaya, especially in Bhutan) and the spatial difference in the pattern of ammonia concentration (increasing towards the western-to-central southern Himalaya boundary) compared to total nitrogen deposition (increasing with a pattern of higher rainfall towards the eastern Himalaya). Consequently, while the extent of forest that exceeds a critical level of ammonia declines for land definitions of increasing forest cover, the extent of forest exceeding the critical load of total nitrogen deposition increases for definitions of increasing forest cover. Nevertheless, second, the exceedance of both critical level of ammonia and critical load for total nitrogen deposition is spatially extensive. Adopting a mean value of the ammonia critical level, and comparing among the different atmospheric chemistry models, 80–85% of Himalayan forests exceed this threshold, and 61–77% at dense forest cover. The best-case scenario (adopting the 97.5% confidence interval) provides an estimate of 73–79% of Himalayan forests exceeding the ammonia critical level. Adopting a mean value for the total nitrogen deposition critical load, 95–98% of Himalayan forests exceed this threshold, and >99% of dense forest cover. The best-case scenario (adopting the 97.5% confidence interval) provides an estimate of 82–85% of Himalayan forests exceeding the total nitrogen deposition critical load.

4. Discussion

We have made a first estimate of the area over which Himalayan forests are being impacted by excess atmospheric nitrogen. To do this we reviewed and applied thresholds as critical levels (ammonia concentration: μg m⁻³) and loads (total nitrogen deposition: kg N ha⁻¹ yr⁻¹)

Table 2

Estimated extent over which Himalayan forests are exceeding their critical level for ammonia and critical load for total nitrogen deposition, for alternate definitions of forest cover and considering different models in nitrogen atmospheric chemistry. Estimates are shown as a proportion of the total area from 0 to 1, based on the 2.5% – mean – 97.5% intervals of bootstrapped estimates in critical levels and loads.

Percent forest cover	Extent (ha)	Ammonia Concentration			Total Nitrogen Deposition		
		EMEP-WRF-2015 (0.33 × 0.33)	EMEP-WRF-2018 (0.11 × 0.11)	UKCA-CLASSIC-2000 (1.25 × 1.875)	EMEP-WRF-2015 (0.33 × 0.33)	EMEP-WRF-2018 (0.11 × 0.11)	UKCA-CLASSIC-2000 (1.25 × 1.875)
>90%	8,761,285	0.88–0.735–0.635	0.919–0.77–0.612	0.623–0.617–0.617	0.999–0.993–0.919	0.999–0.991–0.894	0.999–0.998–0.784
>70%	14,898,802	0.9–0.777–0.686	0.928–0.806–0.659	0.693–0.684–0.684	0.999–0.989–0.884	0.999–0.984–0.854	0.999–0.991–0.82
>50%	20,042,603	0.912–0.804–0.717	0.935–0.829–0.69	0.735–0.725–0.725	0.999–0.987–0.87	0.999–0.981–0.837	0.999–0.977–0.833
>30%	24,834,513	0.918–0.82–0.738	0.939–0.841–0.712	0.764–0.751–0.751	0.999–0.985–0.86	0.999–0.977–0.827	0.999–0.963–0.838
>10%	31,659,388	0.924–0.833–0.754	0.942–0.851–0.729	0.794–0.778–0.778	0.999–0.982–0.851	0.999–0.972–0.82	0.999–0.953–0.846
>1%	35,892,992	0.929–0.841–0.764	0.945–0.857–0.739	0.811–0.794–0.794	0.999–0.98–0.853	0.999–0.97–0.822	0.999–0.953–0.857

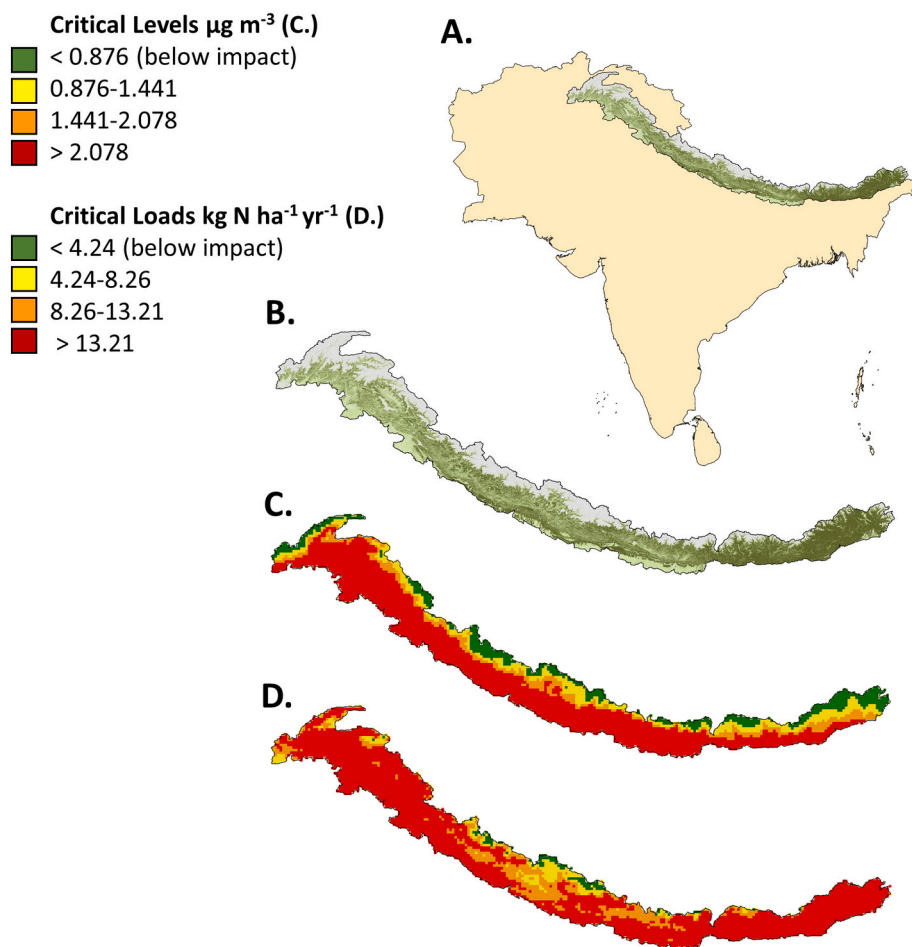


Fig. 1. A. The Himalayan forest zone, highlighted within the wider context of south Asia (Afghanistan, Pakistan, India, Nepal, Bhutan, Bangladesh and Sri Lanka), and B. Showing the density of forest cover at a 100 m grid scale for 2015, with example projections for C. The region exceeding critical levels of ammonia, and D. exceeding critical loads of total bulk nitrogen deposition, both based on the EMEP-WRF model projection at 0.11×0.11 resolution, using 2010 emissions and 2018 meteorology.

calculated for lichen epiphytes, which are among the most sensitive bioindicators for nitrogen pollution (Bobbink et al., 2002; Cape et al., 2009a; Pardo et al., 2011). We maximised the potential transferability of these thresholds by calculating from studies across scales in Europe and North America, including different climatic zones and forest types, and for regions that have been affected to a greater or lesser degree by cultural legacies of human activity (Table 1). This broad scope yielded mean critical levels and loads with confidence intervals that are arguably appropriate to a Himalayan region with a diverse climate, vegetation structure, and complex cultural history. Using lichen bioindicators as a benchmark for excess atmospheric nitrogen, an estimated 80–85% of Himalayan forests are impacted by gaseous ammonia, and 95–98% by total nitrogen deposition.

(a) Critical levels and critical loads

Recent reviews have summarised a multifaceted response of lichens to atmospheric nitrogen in its different forms (Carter et al., 2017; Hauck, 2010; Zarabska-Bozejewicz, 2020), and, consequently, the previously published literature had recognised critical levels and loads affecting lichens over a broad range of nitrogen pollutant types (Table 1). However, in practice, tackling real-world covariation to isolate the unique effect of a given nitrogen pollutant can be extremely challenging at field-scale (Will-Wolf et al., 2015). The response to compound variables, such as throughfall nitrogen deposition, integrating the wet and dry deposition of different nitrogen gases and particulates, may instead provide a relevant overall explanation of lichen epiphyte response (Jovan et al., 2012). Furthermore, throughfall nitrogen deposition might also directly

reflect conditions in the epiphytic habitat; since throughfall deposition tends to be lower than total deposition, because of canopy exchange (Lindberg et al., 1986; Lovett and Lindberg, 1993), this can explain why critical loads estimated for total deposition tend to be higher than for throughfall deposition (Table 1).

Notwithstanding the relevance of both total and throughfall deposition, within the context of the Indo-Gangetic Plain (IGP) atmospheric ammonia is the source pollutant of major concern, because of the extremely high levels compared with other regions globally (Clarisse et al., 2009; Van Damme et al., 2018; Warner et al., 2017). It has also been argued that gaseous ammonia may have a stronger effect on lichens than nitrogen deposition (cf. Wolsey et al., 2006; Sutton et al., 2020). Ammonia will generate multifaceted and reinforcing effects on lichens, including through its toxicity (Munzi et al., 2014; Paoli et al., 2015; Paoli et al., 2010), accumulation in lichen tissues to cause nutrient imbalance (Frati et al., 2011; Frati et al., 2007), and an effect on microhabitat bark pH (De Bakker, 1989; Van Herk, 1999, 2001). We are therefore confident that by using both critical level of ammonia and critical load for total nitrogen deposition we provide context relevant and compound thresholds, respectively, which support the use of lichen bioindicators in estimating the impact of excess atmospheric nitrogen for Himalayan forests. However, we include a proviso that our estimates could be considered conservative. This is because certain studies included in our calculations set their baselines within traditionally managed, semi-natural landscapes that may have been impacted by low levels of atmospheric nitrogen pollution over long periods of time (cf. Aguilhaume et al., 2017; Pinho et al., 2012). Such studies potentially recognise higher critical levels and loads than if a baseline is established for a 'pristine' environment. This may nevertheless be appropriate to the cultural landscapes of the Himalaya (see above), while the calculation of confidence intervals accounts for this baseline variability and makes it possible to query estimates at higher or lower thresholds, depending on landscape context.

(b) Ecosystem consequences

Lichen bioindicators are a subset of one component (epiphytes) among the various attributes of forest biodiversity and its associated ecosystem functions and services. Different forest attributes have contrasting sensitivities and thresholds at which they become affected by excess nitrogen (Bobbink et al., 2002; Pardo et al., 2011). Nevertheless, our assessment based on lichen bioindicators does raise concern for both direct and wider effects of atmospheric nitrogen in Himalayan forests. First, directly, there is growing appreciation of the importance of cryptogam diversity (including lichens) in delivering global ecosystem function and services (Elbert et al., 2012; Porada et al., 2014). Accordingly, lichen epiphytes contribute – in general – significant levels of biodiversity to forest ecosystems (Komposch and Hafellner, 2000; Vondrák et al., 2019), provide regulating services that mediate the water (Pypker et al., 2006a, 2006b; Pypker et al., 2017) and nutrient cycles (Antoine, 2004; Reiners and Olson, 1984; Van Stan and Pypker, 2015), and, specifically within the Himalaya, they provide subsistence-level provisioning (food, medicines) and cultural services (e.g. for ritual and aesthetic purposes) (Devkota et al., 2017a; Upreti et al., 2005) in addition to their capital value in local economies when harvested for sale into national and international markets (Devkota et al., 2017b; Upreti et al., 2005). The exceedance of critical levels and loads puts these regulating, provisioning and cultural services, plus capital gain, at risk.

Second, considered more widely, the exceedance of thresholds for lichen bioindicators can be viewed as a red flag, signalling the potential for a much broader suite of impacts across various forest attributes. For example, survival rates for approximately 45% of North American tree species appear affected by atmospheric nitrogen deposition (Horn et al., 2018), with survival declining from a threshold $<10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ and comparable therefore to the mean critical load applied here for lichen bioindicators. In contrast, other forest attributes may be more resistant

to impact, with nitrate leaching and soil acidification occurring at loads of $17\text{--}26 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ for drought tolerant North American forest (Pardo et al., 2011). Overall, our estimates based on lichen bioindicators point to a multiplicity of extant or potential consequences, including simplification of vegetation structure through plant toxicity, soil acidification, eutrophication of excess nitrogen, weakened resistance to pests/diseases, and lowered resistance to other environmental pressures such as climate change (Dise et al., 2011; Krupa, 2003; Pardo et al., 2011), all of which can undermine the sustainable supply of ecosystem services and goods from Himalayan forests to human communities (Joshi and Joshi, 2019; Joshi and Negi, 2011; Måren et al., 2014).

(c) Confirmation and future surveillance

Our results raise clear concern over excess atmospheric nitrogen, threatening lichen epiphyte diversity, and signalling the potential for wider consequences across most Himalayan forests. However, our estimates were derived from a desk-based study that matched published critical levels and loads with projections from atmospheric chemistry models. Our results need to be confirmed through field observation, and we offer three key considerations for the future development of lichen bioindicators in the Himalayan region: (i) develop spatially transferable metrics, that (ii) are unconfounded by and/or account for covariables, and (iii) take account of cultural landscape context.

First, confirmation of atmospheric nitrogen impact through direct monitoring will need to categorise Himalayan lichen epiphyte species into bioindicator 'trophic groups', such as oligotrophic/acidophytic or nitrophytic. Such categorisation is at an early stage in south Asia (Gupta et al., 2013; Thakur and Chander, 2018), though necessary because contrasting species pools for European, North American, and Himalayan lichens (Galloway, 2008) preclude the direct transfer of European or North American bioindicator species, to the Himalaya. Furthermore, alternative metrics with greater apparent potential for spatial transferability, such as overall species richness of lichen epiphytes, appear to be weaker indicators of atmospheric nitrogen impact than patterns of replacement among species representing different trophic groups (Aguilhaume et al., 2017; Giordani et al., 2014; Pinho et al., 2014; Will-Wolf et al., 2015), i.e. the decline of oligotrophic/acidophytic and increase in nitrophytic species. Transferability may be partly addressed by extrapolating at higher-levels of taxonomic resolution, such as assuming Teloschistaceae as being nitrophytic (Crittenden et al., 2015; Smith et al., 2020) or focussing on sensitive functional components including 'forage lichens' and nitrogen-fixing cyanolichens (Cleavitt et al., 2015; Geiser et al., 2019), though this raises the second consideration of environmental covariables.

Accordingly, second, a sufficient precision may only be achieved for lichen epiphyte bioindicators when controlling for covariables, with the lichen nitrogen response being sensitive to and potentially confounded by spatial patterns in climate and landscape/forest structure (Fenn et al., 2008; Geiser et al., 2010; Giordani et al., 2014). Climate has been included as an important covariable when isolating the atmospheric nitrogen pollution response (McMurray et al., 2015; Root et al., 2015; Smith et al., 2020; Will-Wolf et al., 2015), and operates in several ways, such as in altering the nitrogen concentration that lichen epiphytes are exposed to (Geiser et al., 2010; Glavich and Geiser, 2008) or moderating their physiological response to pollutants (Riddell et al., 2008). More subtly, nitrophytic species may be co-adapted to warmer and drier environments that have lower precipitation and higher temperatures (McMurray et al., 2015; Root et al., 2015; Smith et al., 2020). Lichen species that can achieve higher photosynthetic capacity, up-regulating the production of carbon skeletons, may be able to neutralise nitrogen toxicity by converting excess nitrogen to amino acids (Hauck, 2010). Nitrogen tolerance therefore invokes a regulatory increase in photobiont concentrations and photosynthesis (Dahlman et al., 2003; Gaio-Oliveira et al., 2005; Palmqvist and Dahlman, 2006), explaining covariation between adaptation to high light (and warmth) and tolerance of excess

atmospheric nitrogen (Hauck, 2010; Hauck and Wirth, 2010), while intra-cellular osmotic pressure as an adaptation to drier environments may also protect xerophytic lichens against higher concentrations of nitrogen salts (Frahm, 2013). Similarly, the trophic status of forests can be important. Studies in oligotrophic habitats such as boreal/coniferous systems (Manninen, 2018) are likely to detect nitrogen impacts at lower thresholds than for naturally eutrophic environments (e.g. broadleaf forests on fertile soils) in which component species may be adapted and environmentally filtered to tolerate higher nitrogen (Cleavitt et al., 2015; Giordani et al., 2014).

Third, these spatial considerations extend to cultural history, which – as already noted – can determine the baseline against which impacts are assessed. If species assemblages are already skewed to nitrogen tolerance then the identification of thresholds will reflect this ‘shifted baseline’ (cf. Ellis et al., 2011, 2018), occurring at higher values than for ‘pristine’ environments. Retrospective studies suggested that in North America, for example, a critical load for atmospheric nitrogen deposition had been exceeded over 46% of the continent by 1855 (Clark et al., 2018), and Cleavitt et al. (2015) proposed that there was stronger effect on lichens of this cumulative deposition, as opposed to annual deposition totals (Cleavitt et al., 2015). Likewise, it is possible that atmospheric nitrogen in the Himalaya has started to achieve cumulative effects over relatively long periods of time (Galloway and Cowling, 2002; Galloway et al., 2004), and it is therefore unclear the extent to which there may be opportunity to establish ‘pristine’ baselines against which future monitoring can be interpreted.

Alongside this future research geared towards bioindication, certain key lichen genera are already known to have a disproportionate cultural or economic relevance for Himalayan communities, including for example *Everniastrum*, *Heterodermia*, *Parmotrema*, *Ramalina*, and *Usnea* spp. (Chatterjee et al., 2011; Devkota et al., 2017a). Considering the urgency with which emerging nitrogen impacts may need to be addressed, a complementary shorter-term approach might target these genera (or component species), sampling along atmospheric nitrogen gradients to determine patterns of physiological damage, investigating their response as presence-absence or abundance, and working towards a sustainable harvest rate that protects biodiversity, and associated ecosystem services and goods, for Himalayan forests with excess nitrogen.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.biocon.2021.109401>.

Data statement

Two sets of data used in this article are accessible in scientific archives and available for use: (i) lichen critical levels and loads accessed via Web of Science and presented in Table 1, and (ii) forest cover publicly available via GMES/Copernicus on the principle of full, open and free access as established by policy regulation (EU) #1159/2013 (<http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32013R1159>).

CRedit authorship contribution statement

Christopher J. Ellis: Conceptualization, Methodology, Formal analysis, Writing - original draft, review & editing. **Claudia E. Steadman:** Data curation, Formal analysis, Validation, Writing - review & editing. **Massimo Vieno:** Data curation, Formal analysis, Validation, Writing - review & editing. **Sudipto Chatterjee:** Conceptualization, Methodology, Writing - review & editing. **Matthew Jones:** Conceptualization, Methodology, Writing - review & editing. **Sidharth Negi:** Writing - review & editing. **Bishnu Prasad Pandey:** Conceptualization, Writing - review & editing. **Himanshu Rai:** Writing - review & editing. **Dendup Tshering:** Conceptualization, Writing - review & editing. **Gothamie Weerakoon:** Writing - review & editing. **Pat Wolseley:** Writing - review & editing. **David Reay:** Project administration.

Subodh Sharma: Project administration. **Mark Sutton:** Funding acquisition, Project administration, Resources, Writing - review & editing.

Declaration of competing interest

- The work is original research carried out by the authors.
- All authors agree with the content of the manuscript and submission to the Journal.
- No part of the research has been published in any form elsewhere.
- The manuscript is not being considered for publication elsewhere.
- There are no financial benefits resulting from publication.
- All appropriate ethics and approvals have been obtained.

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