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Critical loads of atmospheric N deposition for phytoplankton nutrient limitation shifts in western U.S. mountain lakes

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Abstract. In many oligotrophic mountain lakes, anthropogenic atmospheric nitrogen (N) deposition has increased concentrations of N, a key limiting nutrient, and thereby shifted phytoplankton biomass growth from N limitation to P limitation. In the western United States, the critical load N deposition rate for these shifts has not been quantified. We synthesized existing mountain lake chemistry, nutrient limitation bioassay, and N deposition data to estimate N critical loads for shifts from N to P limitation of phytoplankton biomass growth. Data from bioassays in 47 mountain lakes were used to define biological (RR-N/RR-P = 1) and chemical (NO_3, DIN, DIN:TP) thresholds above which biomass P limitation is more likely than N limitation. Logistic regression was used to calculate critical loads as the total N deposition rate with >50% probability of exceeding biological or chemical thresholds, and thus where P limitation is more likely than N limitation. Logistic regression models were developed with N deposition as the only predictor and with both N deposition and watershed characteristics as predictors. Logistic model performance was evaluated by comparing predicted and observed chemical threshold exceedances in 108 mountain lakes. Across models, estimated critical loads ranged from 2.8 to 5.2 kg total $N \cdot ha^{-1} \cdot yr^{-1}$. The best-performing model was a univariate logistic model predicting NO_3 threshold exceedance, with N deposition as the only predictor. This model yielded a critical load of 4.1 kg total $N \cdot ha^{-1} \cdot yr^{-1}$ and accurately predicted NO_3 threshold exceedance in 69% of lakes. We applied this critical load to an independent sample of 385 mountain lakes with NO_3 data to estimate the frequency it would fail to predict a limitation shift—cases where the NO_3 threshold for biomass shifts was exceeded, but the critical load was not. The false-negative rate was 13% across the western United States, but was higher (22%) in the Sierras. Performance analyses suggest a 2.0 kg total $N \cdot ha^{-1} \cdot yr^{-1}$ critical load may avoid false negatives entirely. Critical loads presented here can be used to assess N deposition impacts on western U.S. mountain lakes, and associated performance information can be used to consider if presented critical loads are adequate for specific management applications.

Key words: critical load; mountain lakes; nitrogen deposition; phytoplankton.

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Introduction

Human alteration of the global nitrogen (N) cycle has increased atmospheric deposition of N, a key limiting nutrient, and thereby stimulated eutrophication effects in many ecosystems globally (Galloway et al. 2008, Erisman et al. 2013). Remote mountain lakes are among the most sensitive ecosystems to deposition-induced eutrophication because they are naturally oligotrophic. In the absence of significant anthropogenic N deposition, many remote mountain lakes have low NO_3 concentrations that limit phytoplankton growth (Bergström and Jansson 2006, Elser et al. 2009*a*). Anthropogenic N deposition to a lake and its watershed can increase lake NO_3 (Baron et al. 2011, Hessen 2013) and thereby stimulate phytoplankton changes, including species composition changes, phytoplankton biomass increases, and a shift from N to P limitation of phytoplankton growth (Bergström and Jansson 2006, Elser et al. 2009*a*, Baron et al. 2011). Although phosphorus (P) is often assumed to limit phytoplankton growth in lakes, P limitation is prevalent in regions with elevated N deposition, whereas N limitation is prevalent in regions with low N deposition (Bergström and Jansson 2006, Elser et al. 2009*a*).

Mountain lakes are particularly sensitive to deposition-induced N enrichment because their watersheds typically have limited vegetation cover, steep slopes, or other characteristics that reduce watershed N uptake and promote efficient flux of deposited N to lakes (Sickman et al. 2002, Clow et al. 2010, Nanus et al. 2012). If N deposition increases lake N, many factors mediate the occurrence and characteristics of phytoplankton responses. Within a lake, N enrichment first stimulates species-level responses, followed in some cases by biomass growth responses and nutrient limitation shifts (Hessen 2013). If lake N concentration limits phytoplankton growth, species with low N requirements, such as diatoms *Asterionella formosa*, *Fragilaria crotonensis*, and *Fragilaria tenera*, may increase their growth rates at N concentrations as low as <10 μg/L (Fig. 1; Michel et al. 2006, Arnett et al. 2012, Williams et al. 2016) and thereby increase in abundance relative to species that require greater N concentrations. However, the species observed to respond, and N chemical thresholds for responses, can vary substantially within and across regions because many factors other than N mediate species responses. Additional mediating factors may include phosphorus (P) concentrations or N:P ratios (Bergström 2010), phytoplankton community structure (Williams et al. 2016), lake depth (Spaulding et al. 2015), lake transparency (Williamson et al. 2010), temperature (Bergström et al. 2013), zooplankton grazing (Vinebrooke et al. 2014), and dissolved organic matter (Daggett et al. 2015). As lake N concentration increases, some species may decrease in relative abundance if nitrophilic species outcompete other species. If the net result of all species responses causes an increase in overall phytoplankton biomass, an increase in chlorophyll *a* may be observed. In addition, the element limiting phytoplankton biomass growth may shift from N to P if N enrichment satisfies resource requirements of multiple species driving biomass responses. Lake N chemical thresholds are generally lower for phytoplankton species responses than for phytoplankton biomass responses and

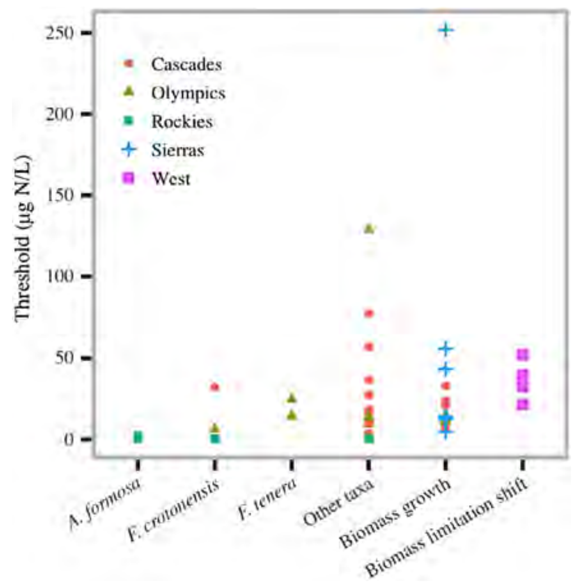

Fig. 1. Nitrogen chemical thresholds for phytoplankton responses to N enrichment in western U.S. mountain lakes. Species growth response thresholds are Monod half-saturation constants from Michel et al. (2006), Arnett et al. (2012), and Williams et al. (2016). Biomass growth response thresholds are from Heard and Sickman (2016) and Williams et al. (2016). Biomass limitation shift thresholds are from this study (Table 2).

limitation shifts, but across lakes there is also large variability and substantial overlap of thresholds within and across biological response levels (Fig. 1). Thus, to assess or manage for deposition-induced phytoplankton changes, one must consider what type and magnitude of phytoplankton response to address, and response variability.

In the western United States, quantifying threshold or critical load (Nilsson and Grennfelt 1988) N deposition rates for chemical and ecological changes in mountain lakes is important for protecting ecosystems from anthropogenic N deposition (Burns et al. 2008). Critical loads for chemical and phytoplankton species changes in mountain lakes (1–1.5 kg wet inorganic $N{\cdot}ha^{-1}{\cdot}yr^{-1}$; Saros et al. 2010, Baron et al. 2011, Sheibley et al. 2014) are lower than those for many other ecological receptors (Pardo et al. 2011). As a result, mountain lakes are an important indicator for U.S. government agencies charged with protecting ecosystems from air pollution. For example, federal and state agencies used a critical load for lake diatom changes to develop a "target" deposition load of 1.5 kg wet inorganic $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ in an air quality agreement intended to protect all types of ecological resources in Rocky Mountain National Park from N deposition (Porter and Johnson 2007). In addition, federal land management agencies compare critical loads to ambient deposition rates to test for critical load exceedances when assessing impacts of proposed new nitrogen emissions sources on Class I lands (USFS et al. 2011), and when developing National Forest Management plans (USFS 2017), among other applications. The U.S. Environmental Protection Agency (USEPA) also has considered critical load and exceedance information, among other data, in periodic reviews of the secondary National Ambient Air Quality Standards for nitrogen oxides and sulfur oxides (USEPA 2009). Among federal agencies, there is a need to assess N deposition effects on western U.S. mountain lakes at the scale of the entire western United States. The National Atmospheric Deposition Program Critical Loads of Atmospheric Deposition Science committee has developed a National Critical Loads Database (NCLD; http://nadp.sws.uiuc.edu/committees/clad/db/) that is used in regional- and national-scale projects assessing deposition effects (Blett et al. 2014). However, the NCLD does not currently include critical loads for eutrophication effects of N deposition in western U.S. mountain lakes.

The objective of this study was to estimate critical load N deposition rates for phytoplankton biomass nutrient limitation shifts in remote mountain lakes in the western United States. Previous studies estimated N critical loads for western U.S. mountain lake NO_3 increases (Baron et al. 2011, Nanus et al. 2017), and increases in the relative abundance of indicator diatom species for specific regions (Baron 2006, Saros et al. 2010, Nanus et al. 2012, 2017, Sheibley et al. 2014). Although N deposition increases have caused shifts from N limitation to P limitation of phytoplankton growth in lakes throughout the Northern Hemisphere (Bergström and Jansson 2006, Elser et al. 2009*a*), N critical loads for nutrient limitation shifts have not previously been quantified for western U.S. lakes. Phytoplankton N-to-P limitation shifts constitute a fundamental change in factors controlling lake primary production and therefore have the potential to alter biogeochemical processes, trophic dynamics, and species diversity in mountain lake ecosystems (Baron et al. 2011, Hessen 2013).

Here, we use existing western U.S. mountain lake nutrient chemistry data (Williams and Labou 2017), and data from previously published nutrient enrichment experiments from lakes across the western United States (Elser et al. 2009*b*, Slemmons and Saros 2012, Williams et al. 2016) to calculate N critical loads for phytoplankton biomass nutrient limitation shifts. We then discuss results, critical load performance, and uncertainties in the context of how critical loads are typically used in decision making by federal agencies.

Methods

Overview

Nitrogen critical loads for phytoplankton biomass nutrient limitation shifts were developed by applying a multi-step process to existing data. An overview of each step is provided here, and a detailed description is provided in subsequent methods sections. First, we selected a biological measure of nutrient limitation shifts and defined a threshold value of this biological measure. Second, we used logistic regression to define lake water chemical thresholds for nitrate, dissolved inorganic nitrogen (DIN), and DIN-to-total phosphorus (TP) mass ratio DIN:TP associated with 50% and 70% probability of exceeding of the biological threshold. Third, logistic regression models were developed to describe the mathematical relationship between nitrogen deposition and exceedance of biological thresholds (an empirical critical load) or chemical thresholds (a modeled critical load). Critical loads were calculated as the total (wet plus dry) N deposition rate at which there is a 50% probability of exceeding the biological threshold. The critical loads thus represent a break point; below the critical load, N limitation is more likely than P limitation, and above the critical load, P limitation is more likely than N limitation. P limitation is prevalent in regions with elevated deposition, whereas N limitation is prevalent in regions with low deposition (Bergström and Jansson 2006, Elser et al. 2009*a*). When this critical load is exceeded, the probability of N limitation is <50%, and biomass P limitation may become widespread as N deposition increases further.

Critical loads were developed using N deposition as the only predictor variable (univariate binary logistic regression), and using both N deposition and watershed characteristics as predictor variables (multivariate binary logistic regression) to assess whether considering watershed characteristics altered critical load estimates. Regression methods are described in detail below. Lake chemistry data independent of that used to generate critical load estimates were also used to quantify the performance of developed logistic regression models, critical loads, and spatial variability of performance. We quantified the ability of logistic regression models to accurately identify lakes where chemical thresholds are exceeded, and estimated the false-negative rate of our critical load. We defined a critical load false negative as cases where lake chemistry measurements indicate the lake chemical threshold was exceeded, but modeled deposition rates were less than the critical load. In such cases, a critical load may not be protective of nutrient limitation shifts.

Biological measures and thresholds were developed using data from previously published nutrient enrichment bioassays conducted in western U.S. mountain lakes (Michel et al. 2006, Elser et al. 2009*b*, Slemmons and Saros 2012, Arnett et al. 2012, Williams et al. 2016, Table 1). Lake chemistry data used in analyses were obtained from the Georeferenced Lake Nutrient Chemistry (GLNC) database (Williams and Labou 2017). The GLNC database contains lake chemistry data collected by researchers and government agencies for 3602 lakes across the western United States. It includes chemistry data from 25 data sources, including federal agencies that regularly monitor mountain lakes (NPS, USFS), relevant publicly available data sets (WQ Portal, USEPA 2007 and 2012 National Lakes Assessments, USEPA Western Lakes Survey), and data contributed by academic researchers (Williams and Labou 2017). Data in the GLNC database are constrained to data that were not flagged as rejected or invalid by the original data source, where sample date and depth information were available, and where the lake location could be verified through GIS research; see Williams and Labou (2017) for a full description of GLNC database development. All lake chemistry data used for analyses originated within a federally protected land (national park, national forest, or wilderness), and at elevation >1200 m. Lake chemistry data were constrained to samples collected July–October during 2006–2015 that were from the lake surface, epilimnion (if stratified), or a mixed water column. The July–October time window was selected to be consistent with

Table 1. Characteristics of bioassays used to define chemical thresholds and empirical critical loads.

Study	Lakes	Region	Type	Duration (d)	Treatment replicates	N addition (μmol/L)	P addition (μmol/L)	Time period
Elser et al. (2009*b*)	32	Rockies	Laboratory	4	4	7.5	0.5	2006
Slemmons and Saros (2012)	6	Beartooth Mountains	In situ	7	3	8	0.5	2010
Williams et al. (2016)	9	Cascades, Olympics	In situ	7–11	3	8	1	2013–2014

the time window associated with bioassays. Mean nitrate concentrations were calculated in cases where multiple nitrate data points were available across years. Detection limit values were used in cases where results were below detection limits.

For each lake used in analyses, N deposition was estimated as 2006–2011 mean total N deposition (wet plus dry, all species), using output from 12-km grid bias-adjusted Community Multiscale Air Quality (CMAQ) model simulations conducted by the USEPA. This model was selected because Williams et al. (2017) demonstrated it has the best wet inorganic N deposition performance among models available at the national scale required by this study, including other CMAQ models with a smaller grid size. It performed better than other available national-scale models in mountain environments, including at a 1654-m-elevation monitoring site on Mount Rainier (Williams et al. 2017). The deposition data are described in the Appendix S1 and in Williams et al. (2017). All analyses were performed using the R statistical software, version 3.2.1 (R Development Core Team, 2017). Critical load calculations described here are fully reproducible; data and R code necessary to reproduce calculations, and associated metadata are available on FigShare (https://doi.org/10.6084/m9.figshare.4981832).

Logistic regression was used to develop critical loads rather than linear regression for several reasons. First, our goal was to develop chemical thresholds and critical loads for nutrient limitation shifts; nutrient limitation type is a categorical response variable, and thus, assumptions of linearity, homoscedasticity, and error normality required by linear regression are violated. Second, lake water NO_3 responses to N deposition typically show a dogleg rather than linear pattern (Fig. 2; Baron et al. 2011), and some previous studies have not observed a significant linear relationship between NO_3 concentration in surface waters and deposition across mountain lakes (Clow et al. 2010, Nanus et al. 2012). Third, logistic regression helps make variability in phytoplankton responses explicit. The data we present (Fig. 1; Appendix S2: Figs. S1–S3) and previous studies (Elser et al. 2009*a*) demonstrate

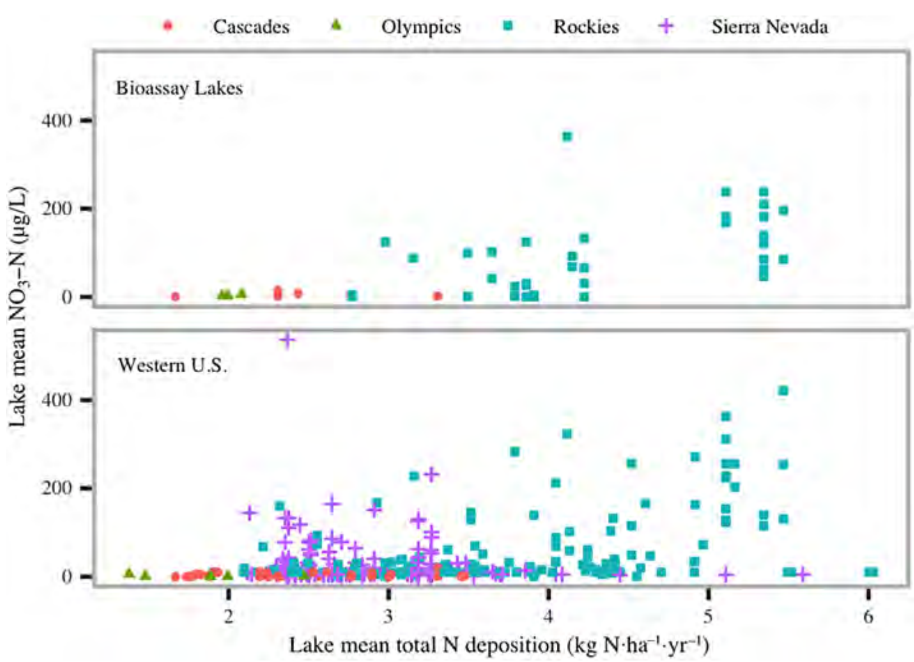

Fig. 2. Relationship between lake mean total N deposition and lake mean nitrate concentration among bioassay lakes (above), and an independent set of 385 lakes in the western United States. The western U.S. data set used in the lower plot is 2006–2015 lake mean nitrate data from the Georeferenced Lake Nutrient Chemistry database, a database of western U.S. mountain lake chemistry developed by Williams and Labou (2017).

there is large variability across lakes in the N concentration required to elicit phytoplankton responses, including biomass nutrient limitation shifts. This variability occurs because many within-lake physical, chemical, and ecological factors mediate phytoplankton responses to N enrichment. By defining our critical load as the N deposition rate where the probability of biomass N limitation is <50%, we account for this variability and make it explicit.

Biological measure and threshold for phytoplankton biomass nutrient limitation shifts

A biological measure for phytoplankton biomass nutrient limitation shifts and an associated threshold value were defined using data from 50 previously published nutrient enrichment bioassays across 47 mountain lakes in the western United States (Fig. 3, Table 1). Data were not used if the lake location could not be verified using GIS, or lake DIN:TP or chlorophyll *a* RR-N/RR-P measurements were not available (see definition below). In each bioassay, lake water was divided among experimental containers to create control, N addition, and P addition treatments. Chlorophyll *a* responses to nutrient additions were then measured after a 4- to 11-d in situ or laboratory incubation period (Table 1). The relative magnitude of biomass growth in response to nitrogen addition (RR-N) and phosphorus addition (RR-P) in nutrient limitation bioassays (RR-N/RR-P, where RR-X = mean chlorophyll *a* treatment X/mean chlorophyll *a* control) was selected as a biological measure of nutrient limitation shifts, and RR-N/RR-P = 1 was selected as the biological threshold.

RR-N/RR-P indicates both the relative magnitude of N and P growth responses in bioassays and the probability of nitrogen or phosphorus limitation. Previous studies have demonstrated its utility as an indicator of N deposition effects (Bergström and Jansson 2006, Elser et al. 2009*a*). As RR-N/RR-P approaches infinity or zero, single-element limitation by nitrogen or phosphorus respectively is more likely. RR-N/RR-P was used as a biological measure rather than the nutrient limitation classifications assigned in the studies because classifications depend on the statistical approach used to define classifications and the range of limitation categories considered, both of

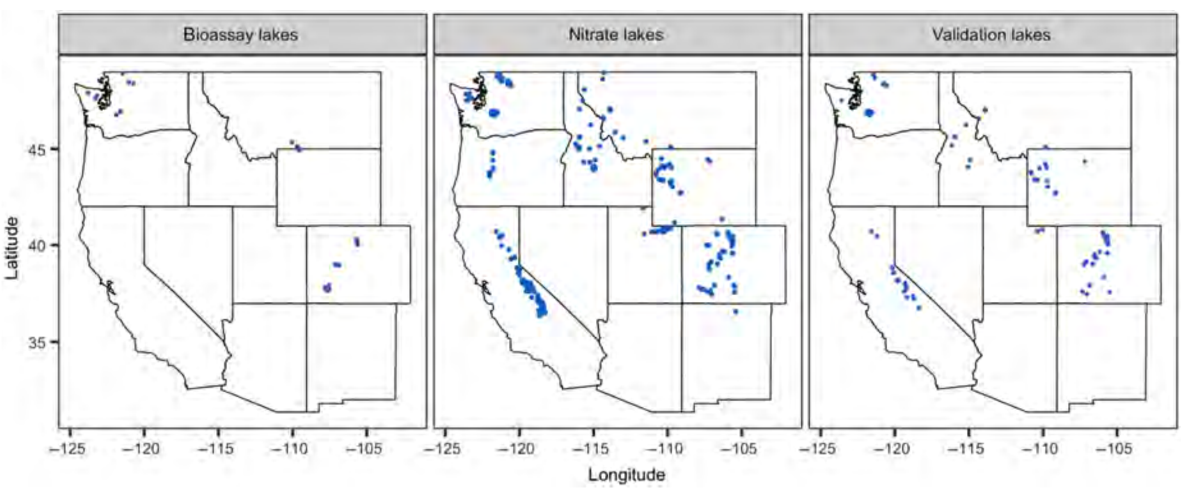

Fig. 3. Location of mountain lakes used in analyses. Bioassay lakes (left, N = 47) are those where nutrient enrichment bioassays have been conducted, and associated data were used to define biological thresholds and chemical criteria for nutrient limitation shifts. Nitrate data lakes (right, N = 385) are mountain lakes where nitrate data are available; these data were used to calculate phytoplankton species critical loads, to plot the relationship between deposition and nitrate (Fig. 2), and to quantify the critical load false-negative rate. Validation lakes (right, N = 108) are mountain lakes, where watersheds were delineated and chemistry and watershed characteristic data were used to evaluate performance of logistic regression models. Bioassay lakes are not included with either the nitrate lakes or validation lakes.

which varied across studies. A threshold of RR-N/RR-P = 1 for limitation shifts is generally consistent with classifications assigned in the studies synthesized (Table 1). N-limitation classifications were only reported where RR-N/RR-P > 1, and P-limitation classifications were only reported where RR-N/RR-P < 1. Other limitation classifications, such as co-limitation, antagonistic growth, or no-limitation, were also assigned by studies in some lakes where RR-N/RR-P > 1 or RR-N/RR-P < 1. This diversity of classifications reflects both variation in classification schemes across studies and that bioassay growth responses and nutrient limitation depend on lake-specific factors such as phytoplankton community structure (Williams et al. 2016), lake transparency (Williamson et al. 2010), temperature (Bergström et al. 2013), and dissolved organic matter (Daggett et al. 2015) in addition to nitrogen and phosphorus.

Chemical thresholds for phytoplankton biomass nutrient limitation shifts

Univariate binary logistic regression was used to calculate threshold concentrations of water NO_3–N, DIN, and a DIN:TP mass ratio associated with RR-N/RR-P = 1 across bioassays. A separate logistic regression model was developed for each parameter, with chemical parameter concentration prior to experimental nutrient addition as the predictor variable, and a binary value (1 or 0) indicating whether lake mean RR-N/RR-P > 1 as the response variable. Predictor variables were natural-log-transformed. The chemical threshold was defined as the parameter value at which there was a 50% probability of RR-N/RR-P > 1 (Table 2). Thresholds for 70% probability of RR-N/RR-P > 1 were also calculated for use in modeled critical load calculations (see *Modeled critical loads for phytoplankton biomass nutrient limitation shifts*; Table 2).

Empirical critical loads for phytoplankton biomass nutrient limitation shifts

Empirical critical loads are calculated by relating deposition rates directly to ecological responses and are typically developed using spatial gradients of deposition, field experiments, or long-term observations (Pardo et al. 2011, de Vries et al. 2015). Empirical critical loads were calculated here based on a N deposition spatial gradient using two approaches. First, a univariate binary logistic regression model was developed with N deposition as the predictor and a binary value (1 or 0) indicating whether or not lake RR-N/RR-P > 1 as the response variable. A critical load was defined as the deposition rate where there was a 50% probability of RR-N/RR-P > 1, and thus represents the deposition rate above which P limitation is more likely than N limitation.

Second, a multivariate logistic regression model was developed with N deposition and lake watershed characteristics as predictor variables, and a binary value (1 or 0) indicating whether lake RR-N/RR-P > 1 was the response variable. Multivariate analyses were performed to assess whether considering watershed characteristics drastically changed critical load estimates. Lake watersheds were delineated using the 30-m digital elevation model and flow direction grids from the National Hydrography Dataset (NHDPlus version 2; Horizon Systems, 2012). Watershed characteristic information evaluated included vegetation cover, hypsometry, bedrock geology, soils, and nitrogen deposition (Appendix S1: Table S1). Data sources and methods for watershed characteristics are described in detail in Appendix S1.

To develop a multivariate logistic regression model, a univariate binary logistic regression model was first developed for each potential explanatory variable. Potential explanatory

Table 2. Results of logistic regressions used to define chemical thresholds.

Predictor	B_0 (SE)	B_1 (SE)	AIC	R^2_N	Classification score	Chemical criteria 50%	Chemical criteria 70%
ln(DIN:TP)	4.1 (1.3)	−1.7 (0.5)	36.3	0.69	43/49	11.4	6.9
ln(NO_3-N)	5.0 (1.7)	−1.4 (0.4)	38.2	0.66	42/49	39.8 μg N/L	21.3
ln(DIN)	7.0 (2.1)	−1.8 (0.5)	37.7	0.67	43/49	52.3 μg N/L	32.4

Notes: Criteria are indicated for 50% and 70% probability of RR-N/RR-P > 1. AIC, Akaike information criterion.

variables used include lake water chemistry, N deposition, and watershed characteristics (Appendix S1). Although including water chemistry variables as predictors limits the applicability of the models to lakes with necessary chemistry data, chemistry variables were included because the chemical threshold analysis in this study and numerous previous studies (Morris and Lewis 1988, Bergström 2010, Williams et al. 2016) demonstrate the importance of lake nutrient chemistry to predicting phytoplankton biomass nutrient limitation. Variables with univariate $P < 0.1$ were used to build a multivariable model (full model). Among water chemistry variables NO_3, NH_4, DIN, TP, and DIN:TP, only DIN:TP was used to build the full model, because its univariate regression was most significant among these, and it integrates the other chemical variables. Similarly, univariate regressions were significant for both wet inorganic and total N deposition, but only total N deposition was used in the full model. The full model was used to develop two additional candidate multivariate models. The manual purposeful model-building approach of Hosmer et al. (2013) was used to develop a reduced candidate model retaining only predictor variables with $P < 0.1$. In addition, a stepwise candidate model was developed using automated forward and backwards stepwise regression in R using the step() function, which recalculates variances at each step after addition of a variable to the model and uses Akaike information criterion (AIC) values to determine which variables to retain in the model. The assumption of a linear relationship between each variable and the logit of the response was assessed as described in Field et al. (2012). Variance inflation factor (VIF) values were used to test for multicollinearity. Variables with VIF > 10 were assumed to indicate multicollinearity (Midi et al. 2010) and were excluded.

The full, reduced, and stepwise candidate models were then compared using several measures of model fit. Nagelkerke's R^2 (R^2_N; Nagelkerke 1991) was used to quantify observed variation explained by the model. The concordance statistic (*c* statistic) was used to summarize model discrimination ability. The c statistic ranges from 0.5 (no discrimination ability) to 1 (perfect discrimination ability) and indicates the probability that for a random pair of observations ($Y = 1$, $Y = 0$), the model correctly predicts that an affirmative ($Y = 1$) observation has a higher probability of $Y = 1$ compared to the $Y = 0$ observation (Austin and Steyerberg 2012). The *c* statistic is equivalent to the area under the curve of a receiver operating characteristic curve (Steyerberg et al. 2010). Model calibration, defined as agreement between observed and predicted outcomes (Steyerberg et al. 2010), was summarized using the Hosmer–Lemeshow (HL) goodness-of-fit test (Hosmer et al. 1997), comparing observations and predictions by decile of predicted probability. A small *P* value resulting from the HL test suggests one should reject the null hypothesis that the model fits. Finally, a model classification score was calculated to describe the proportion of bioassays correctly classified by each model as RR-N/RR-P > 1. A model was considered to correctly classify RR-N/RR-P within a lake if observed RR-N/RR-P > 1, and the logistic regression model yielded >50% probability of RR-N/RR-P > 1 at the mean deposition rate for the bioassay lake. The best-fit model was selected based on the above measures and model parsimony, and is presented in the *Results* section (Table 3). In cases where N deposition was a predictor variable in the best-fit model, a multivariate empirical critical load was calculated as the deposition rate where there was a 50% probability of RR-N/RR-P > 1.

Modeled critical loads for phytoplankton biomass nutrient limitation shifts

Modeled critical loads are calculated by relating N deposition to a chemical indicator of a specified deposition-induced ecological change (Pardo et al. 2011, de Vries et al. 2015). Here, DIN:TP, DIN, and NO_3–N were used as chemical indicators of a biological measure for shifts from N to P limitation of biomass growth (RR-N/RR-P = 1). For each chemical indicator, a critical load was calculated using a univariate and multivariate logistic regression approach for comparison. In the univariate approach, a logistic regression model was developed with N deposition as the only predictor, and the chemical indicator concentration as the response. In the multivariate approach, a best-fit model was developed as described above, with the

Table 3. Logistic regression models and associated critical loads.

Model	Response variable	Coefficients	Estimate (SE)	P	Model R^2_N	Classification score (%)	CL (kg N·ha^{-1}·yr^{-1})
1. Empirical (U)	RR-N/RR-P > 1				0.29	34/50 (68)†	4.1
		Intercept	5.7 (2.0)	0.004			
		ln (N deposition)	−4.0 (1.4)	0.004			
2. Empirical (M)	RR-N/RR-P > 1				0.58	40/50 (80)†	. . .
		Intercept	2.6 (1.3)	0.05			
		DIN:TP	−0.1 (0.03)	0.01			
		PHNSV	0.04 (0.03)	0.1			
		Permeability	−0.2 (0.1)	0.1			
		Temperate forest	0.04 (0.02)	0.1			
3. Modeled (U)	DIN:TP > 6.8				0.60	36/49 (73)‡	4.1
		Intercept	−8.2 (2.6)	0.001			
		N deposition	2.2 (0.7)	<0.001			
4. Modeled (U)	DIN > 32.4				0.60	36/49 (73)‡	4.1
		Intercept	−8.2 (2.6)	0.001			
		N deposition	2.3 (0.7)	<0.001		66/108 (61)§	
5. Modeled (U)	NO_3–N > 21.3				0.60	39/49 (80)‡	4.1
		Intercept	−8.2 (2.6)	0.002			
		N deposition	2.3 (0.7)	<0.001		74/108 (69)§	
6. Modeled (M)	DIN:TP > 6.8				0.73	41/49 (84)‡	2.8–5.0
		Intercept	1.7 (5.0)	0.7			
		N deposition	3.2 (1.7)	0.05			
		Temperate forest	−0.09 (0.04)	0.04			
		Lake elevation	−0.003 (0.002)	0.1			
		Open Water	−0.3 (0.15)	0.04			
7. Modeled (M)	DIN > 32.4				0.73	40/49 (82)‡	2.9–5.2
		Intercept	0.7 (4.7)	0.8			
		N deposition	2.7 (1.4)	0.05			
		Temperate forest	−0.07 (0.04)	0.04			
		Lake elevation	−0.002 (0.002)	0.2		67/108 (62)§	
		Open water	−0.3 (0.2)	0.03			
8. Modeled (M)	NO_3–N > 21.3				0.68	43/49 (87)‡	. . .
		Intercept	4.1 (1.1)	<0.001			
		Temperate forest	−0.08 (0.3)	0.001			
		Open water	−0.50 (0.2)	0.002			

Notes: U, univariate binary logistic regression; M, multivariate binary logistic regression. An ellipsis (. . .) indicates a CL could not be calculated because N deposition was not among the predictor variables in a multivariate model; PHNSV, polar and high-montane nonvascular and sparse vegetation.

† Classification score calculated as proportion of bioassays correctly classified as RR-N/RR-P > 1.

‡ Classification score calculated as proportion of bioassays where chemical threshold exceedance was correctly predicted.

§ Classification score calculated as proportion of lakes where chemical threshold exceedance was correctly predicted in 108 independent lakes (validation lakes; Fig. 3).

chemical indicator concentration as the response variable. In both cases, a critical load was defined as the deposition rate at a >49% probability of exceeding the biological threshold (RR-N/RR-P = 1). The critical load was calculated using the deposition rate for a 70% probability of exceeding the chemical threshold, and the chemical concentration for a 70% probability of RR-N/RR-P > 1 (70% × 70% = 49%). Model selection details for multivariate models are described in Appendix S3.

Modeled critical load performance

The GLNC database was used to select 108 lakes to use in performance analysis of logistic regression models (Table 3). If logistic regressions used to calculate modeled critical loads can correctly predict when N concentrations exceed chemical thresholds (Table 2) in a larger independent sample of lakes, this would increase confidence that modeled CL equations could be applied to similar mountain lakes without chemistry or bioassay data and yield reliable results.

Lakes were filtered from the GLNC database using the same location and sampling criteria applied above, with the additional requirements that lakes have NH_4 data in addition to NO_3 data, and the criteria that we were able to delineate the lake watershed, given project scope constraints and challenges of accurately delineating lakes with very small watersheds in complex terrain. Locations of these 108 lakes are displayed in Fig. 3 (validation lakes). For each lake, the watershed was delineated, and watershed characteristics were quantified as described above to enable evaluation of both univariate and multivariate modeled CLs. The number and percentage of lakes where chemical concentrations were correctly predicted to be above or below thresholds was calculated and used to compare performance among models.

Results

Chemical thresholds for phytoplankton biomass nutrient limitation shifts

Across bioassays, lake NO_3–N was positively related to N deposition, a pattern consistent with mountain lakes throughout the western United States (Fig. 2). Lake NO_3–N was a primary driver of relative responses in bioassays; the magnitude of biomass growth responses to N addition was negatively related to lake NO_3–N prior to nutrient addition (Fig. 4). Lake NO_3–N, DIN, and DIN:TP were all statistically significant predictors of RR-N/RR-P response category, and explained similar amounts of variation in response outcomes (R^2_N = 0.66–0.68; Table 2). There was a 50% probability of RR-N/RR-P > 1 (N limitation) at

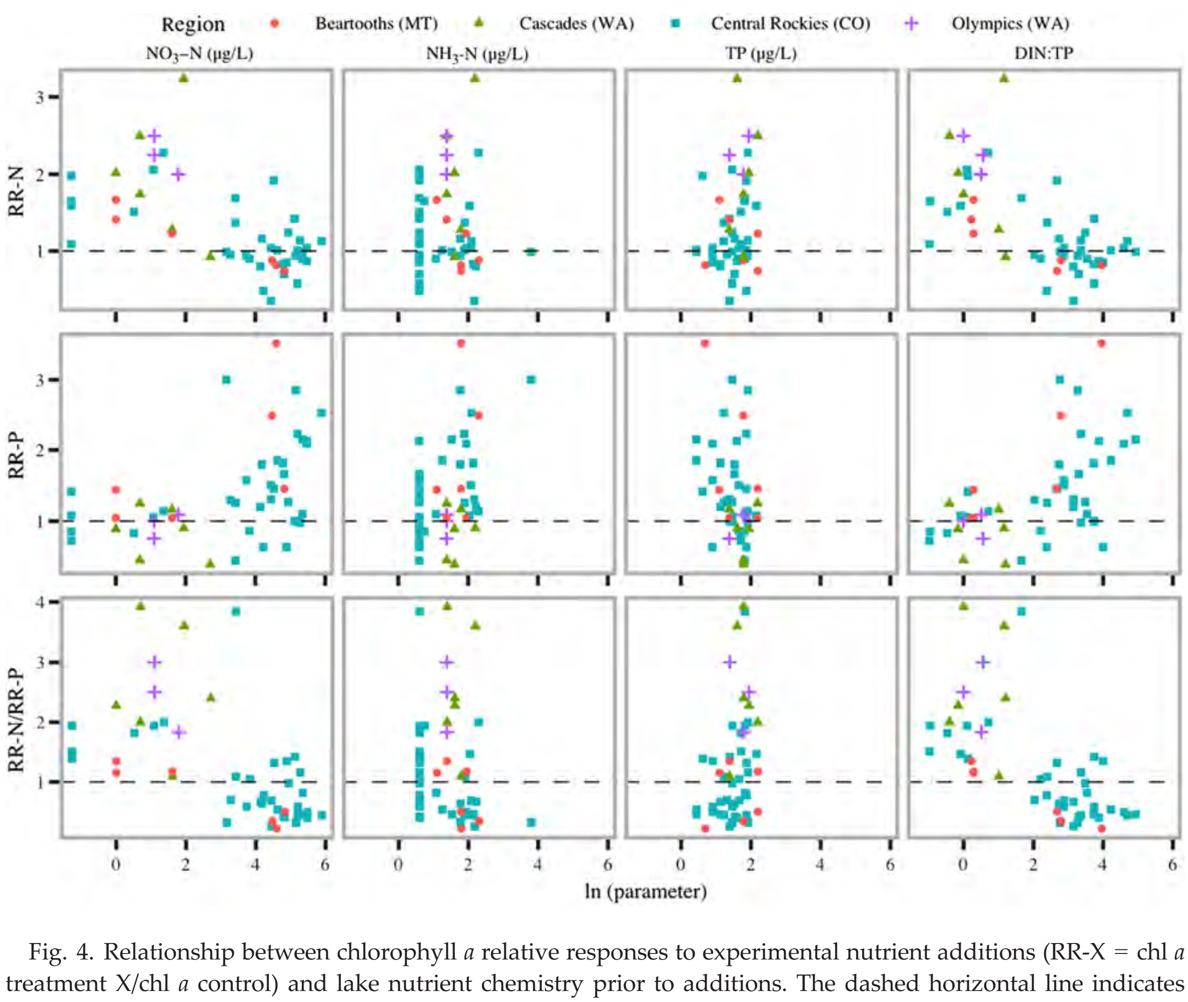

Fig. 4. Relationship between chlorophyll *a* relative responses to experimental nutrient additions (RR-X = chl *a* treatment X/chl *a* control) and lake nutrient chemistry prior to additions. The dashed horizontal line indicates RR = 1 (top, middle) or RR-N/RR-P = 1 (bottom).

39.8 μg NO_3–N/L, 52.3 μg DIN/L, and 11.4 DIN:TP, and a 70% probability of RR-N/RR-P > 1 at 21.3 μg NO_3–N/L, 32.4 μg DIN/L, and 6.9 DIN:TP (Table 2). The probability of biological threshold exceedance is plotted against concentration for each parameter in Appendix S2: Figs. S1–S3.

Empirical critical loads for phytoplankton biomass nutrient limitation shifts

The univariate binary logistic regression model with N deposition as the only predictor was statistically significant, and yielded an empirical critical load value of 4.1 kg total $N \cdot ha^{-1} \cdot yr^{-1}$ for a 50% probability of RR-N/RR-P > 1 (Table 3, model 1). A multivariate empirical CL could not be calculated because N deposition was not among the variables that best predicted RR-N/RR-P > 1 (Table 3, model 2).

Modeled critical loads for phytoplankton nutrient limitation shifts

Univariate regressions predicting DIN:TP, DIN, and NO_3–N response category all yielded a modeled critical load of 4.1 total kg total $N \cdot ha^{-1} \cdot yr^{-1}$ and had similar model fit (Table 3, models 3–5). Best-fit multivariate regression models had greater R^2_N values and classification scores (calculated using the same bioassay lakes used to construct the model) than corresponding best-fit univariate models (Table 3). In best-fit multivariate regression models predicting DIN (Table 3, model 7) and DIN:TP (Table 3, model 6) response categories, N deposition was among variables that best predicted response category, and a modeled critical load could be calculated. Across the bioassay lakes, modeled critical loads averaged 3.8 kg total $N \cdot ha^{-1} \cdot yr^{-1}$ (range 2.8–5.0 kg total $N \cdot ha^{-1} \cdot yr^{-1}$) when based on DIN:TP response category, and 3.8 kg total $N \cdot ha^{-1} \cdot yr^{-1}$ (range: 2.9–5.2 kg total $N \cdot ha^{-1} \cdot yr^{-1}$) when based on DIN response category. For NO_3–N, N deposition was not among predictor variables in the model with the best classification score (43/49; Table 3, model 8); the full, reduced, and stepwise models all had similar R^2_N, c statistic, and H–L value, so the best-fit model was identified based on classification score and parsimony. Variables in the best-fit model included percent watershed cover of temperate forest and open water (Table 3, model 8). N deposition was among predictor variables in a model predicting NO_3–N response category that had a slightly lower classification score (42/49), but two additional variables, and so was not selected as the final model based on parsimony and is not shown in Table 3. This model included N deposition, percent watershed temperate forest cover, lake elevation, and percent watershed open water as predictors. When applied across bioassays to calculate critical loads, this model estimated a mean critical load of 3.7 kg total $N \cdot ha^{-1} \cdot yr^{-1}$ (range: 2.7–5.2).

Modeled critical load performance

When applied to an independent sample of 108 lakes, logistic regression equations used to calculate modeled critical loads correctly predicted whether or not chemical thresholds were exceeded in 61% of lakes using the DIN univariate model (model 4; Table 3), 62% of lakes using the DIN multivariate model (model 7; Table 3), and 69% of lakes using the NO_3-N univariate model (model 5; Table 3). Prediction accuracy was positively related to lake mean N deposition rate (Fig. 5). A performance evaluation was not conducted for models predicting DIN:TP exceedance because of a limited number of lakes with DIN:TP data, and because relative response plots (Fig. 4) and thresholds (Table 2) suggest lake N was the primary driver of RR-N/RR-P > 1.

The univariate binary regression model predicting lake NO_3 threshold exceedance (Table 3, model 5; 4.1 kg total $N \cdot ha^{-1} \cdot yr^{-1}$) had the best performance and was the most parsimonious model among those from which a critical load could be calculated, so we evaluated this critical load's regional false-negative rate. We defined a critical load false negative as cases where the lake chemistry threshold for 50% probability of RR-N/RR-P = 1 (39.8 μg NO_3–N/L) was exceeded, but the critical load was not. In such cases, exceedance of the chemical threshold, and thus potentially also a biomass nutrient limitation shift, occurs when the deposition rate is lower than the critical load, and thus, the critical load may not be protective. This analysis assumes this chemical threshold is representative of mountain lakes throughout the western United States. To estimate the false-negative rate, we identified 385 mountain lakes with nitrate data from the GLNC database that met data criteria described in the *Methods* section (nitrate lakes; Fig. 3). Among the 385 lakes, there were a total of 49 false negatives

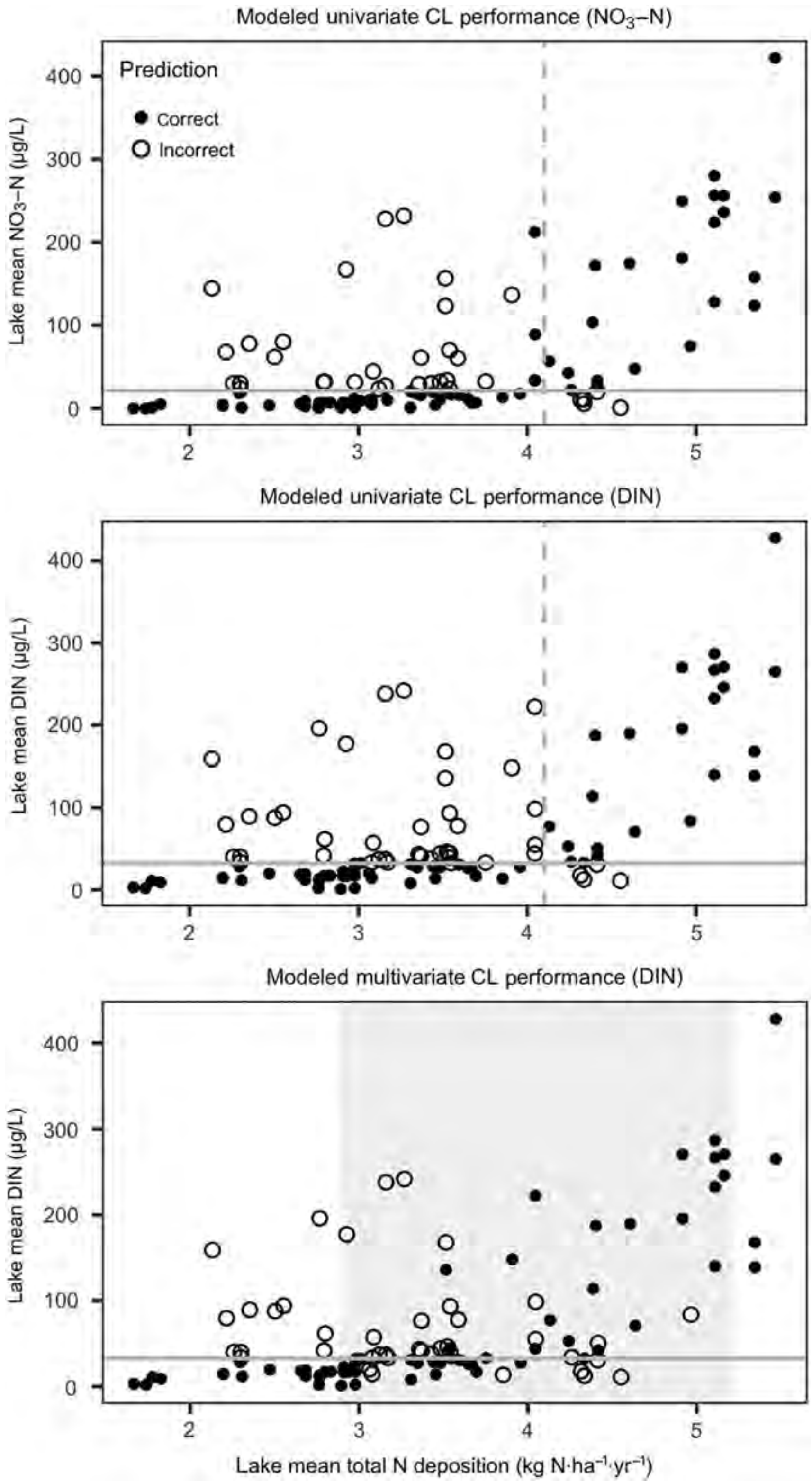

Fig. 5. Ability of logistic regression equations used to develop modeled critical loads (Table 3) to predict

(Fig. 5. *Continued*)
chemical threshold exceedance when applied to an independent data set of 108 mountain lakes. Solid horizontal lines indicate chemical thresholds (Table 2). Dashed vertical lines or shaded area indicates critical loads predicted based on bioassay lakes data (Table 3). Open and closed circles indicate lakes where exceedance category (i.e., above or below threshold) was predicted incorrectly and correctly, respectively.

(13%), where the NO_3 threshold was exceeded, but the critical load was not. No false negatives were observed in the Cascades or Olympics, and multiple false negatives were observed in the Rockies and Sierra Nevada (Fig. 6). False-negative rates were 10% (17/175) in the Rockies and 22% (32/146) in the Sierras. Among lakes where the NO_3 threshold was exceeded, 35% (17/48) in the Rockies and 100% (32/32) in the Sierra Nevada had lake mean deposition rates below 4.1 kg total $N \cdot ha^{-1} \cdot yr^{-1}$. These false-negative rates would be reduced to near zero at a critical load of 2.0 kg total $N \cdot ha^{-1} \cdot yr^{-1}$ (Fig. 6).

Discussion

Many remote mountain lakes in the western United States are located within federally protected lands, such as National Parks, National Forests, or Wilderness Areas, where federal laws and policies of federal land management agencies require ecosystems to be protected from adverse effects of pollution, including atmospheric N deposition (Porter et al. 2005, USFS et al. 2011, Cummings et al. 2014). This study estimated chemical thresholds and critical loads for phytoplankton biomass nutrient limitation shifts resource managers could use to help assess or prevent eutrophication effects of N deposition. This was the first study to estimate critical loads for biomass nutrient limitation shifts in the western United States, and the first to estimate mountain lake phytoplankton critical loads at the scale of the western United States.

Resource managers in the western United States typically use critical loads to test for critical load exceedances through a deterministic approach. A Eulerian atmospheric model is typically used to estimate N deposition rates over a region of interest, and then, a critical load value is subtracted from modeled deposition rates within each deposition model grid cell, assuming no uncertainty in either the deposition value or critical load value (Williams et al. 2017). An exceedance occurs if ambient deposition is greater than a critical load. Each exceedance calculation may have a different objective, depending on the spatial scale, relevant legal mandates for resource

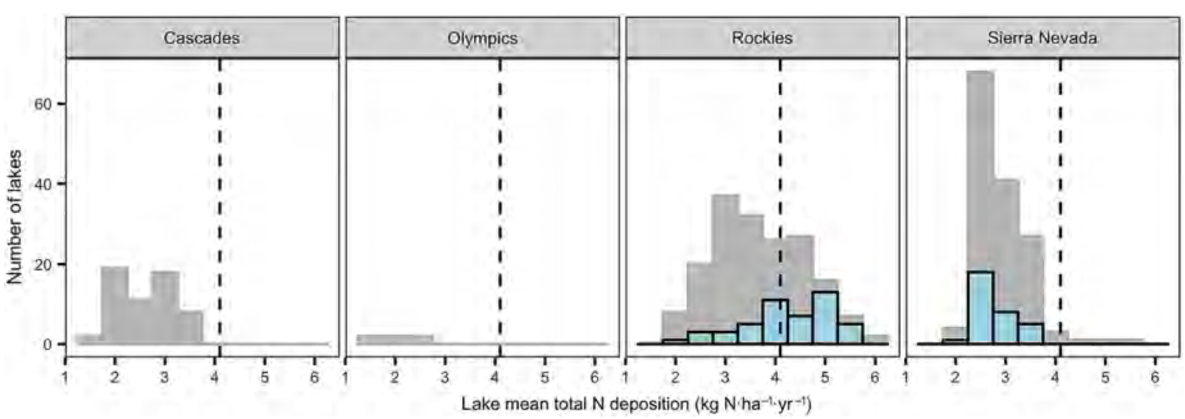

Fig. 6. Distribution of N deposition rates for all lakes among 385 western U.S. mountain lakes with 2006–2015 NO_3 data (gray bars), and for lakes where the NO_3 threshold for 50% probability of RR-N/RR-P = 1 is exceeded (blue bars). The dashed horizontal line is the critical load (4.1 kg $N \cdot ha^{-1} \cdot yr^{-1}$). Blue bars with deposition rates less than the critical load represent lakes where the NO_3 threshold is exceeded, but the critical load is not (a false negative).

protection, and decisions calculations are intended to inform. For our recommended critical load for biomass nutrient limitation shifts (4.1 kg total N·ha^{-1}·yr^{-1}; Table 3, model 5), we estimate from our performance analyses that in 13% of western U.S. mountain lakes, and a higher percentage of Rockies and Sierras lakes (Fig. 6), this critical load may not be fully protective of nutrient limitation shifts, because our estimated chemical threshold for limitation shifts will be exceeded, but the critical load will not be exceeded. If a resource manager seeks to prevent false negatives entirely, our performance analyses suggest a critical load of 2.0 kg total N·ha^{-1}·yr^{-1} would be needed to be fully protective of lake chemical threshold exceedances, and thus nutrient limitation shifts (Fig. 6).

Three major sources of uncertainty affect our critical load estimates. First, bias (modeled–observed) of N deposition models can be large relative to, or even exceed 100% of critical load values for sensitive receptors such as phytoplankton (Williams et al. 2017). We minimized this source of uncertainty by using published wet inorganic N deposition bias (modeled–observed) values (Williams et al. 2017) to select the best-performing model among those available at the national scale needed for this study. Our selected model had a mean bias of 0.05 kg wet inorganic N·ha^{-1}·yr^{-1} (range: −1.1 to 1.4) across National Trends Network monitoring sites in the Pacific Northwest, and a mean bias of −0.48 kg wet inorganic N·ha^{-1}·yr^{-1} (range: −2.2 to 0.16) at a high-elevation monitoring site on Mount Rainier (Williams et al. 2017). These wet inorganic N deposition bias values range from −53% to 34% of our 4.1 kg total N·ha^{-1}·yr^{-1} critical load, but only represent one component of deposition model bias; dry deposition bias may also be large relative to critical loads, but is currently not easily quantifiable due to limitations of available measurement data. While bias of available deposition models is potentially large relative our critical values and those published for other sensitive receptors, by selecting a model for which wet inorganic N deposition bias has been quantified, we made the potential magnitude of this bias explicit to critical load users.

Second, our critical load estimates may be affected by GIS layers used here to quantify watershed characteristics. The land cover (1:100,000), soil (1:250,000), and geology (1:500,000) GIS layers used here are relatively coarse, because fine-scale maps are not available for the entire western United States. Multivariate models in Table 3, which include watershed characteristics as predictors, had greater R^2_{N} and classification scores than corresponding univariate models with N deposition as the only predictor variable. However, when models were applied to an independent data set of 108 lakes, the classification score of the multivariate DIN model (62%) was only slightly better than that of the univariate DIN model (61%). Our univariate model with N deposition as the only predictor performed best when applied to an independent data set (69%), but a multivariate model including watershed characteristics may have performed better if finer-resolution GIS layers were available at the national scale. Nanus et al. (2012) developed linear regression models predicting lake NO_3 in the Rockies using several of the same coarse GIS layers used here and found that watershed mean slope, percent barren area, and N deposition best predicted lake NO_3. In contrast, using fine-scale layers available only for Rocky Mountain National Park yielded different predictor variables and higher predicted NO_3 concentrations compared to the model developed using coarser-scale GIS layers (Nanus et al. 2012). Thus, it is possible using finer-scale GIS layers for watershed characteristics would have yielded different multivariate models with better performance and perhaps lower critical load values.

The third and perhaps largest source of uncertainty is variability in the lake N concentration required to elicit a nutrient limitation shift across lakes. Our critical loads are defined as the total (wet plus dry) N deposition rate at which there is a 50% probability of phytoplankton biomass P limitation. While P limitation of biomass growth is more prevalent in lakes with elevated N concentrations and N deposition rates, previous syntheses of nutrient enrichment bioassays conducted across the Northern Hemisphere (Elser et al. 2009*a*), as well as bioassay data used here (Appendix S2: Figs S1–S3), demonstrate P limitation may occur at a wide range of lake N concentrations, because many other within-lake physical, chemical, and ecological factors mediate phytoplankton responses to lake N enrichment. We used logistic rather than linear regression because logistic regression is a probabilistic approach that accounts for this uncertainty and makes it explicit. Previous studies highlighted the disproportionate effect of chemical threshold values on phytoplankton critical load estimates. Nanus et al. (2012) used multivariate linear regression to calculate critical load for growth responses of the indicator diatom *A. formosa* in the Rockies and reported that their critical load values were highly sensitive to chemical threshold values and that uncertainty in critical loads and exceedances attributable to chemical thresholds "may be much larger than that attributable to other sources of error, such as MLR [multiple linear regression] modeling (p. 133)." Fig. 1 highlights that published species-level chemical thresholds are highly variable. We believe our chemical thresholds for biomass nutrient limitation shifts are relatively well constrained because they are based on data from 47 mountain lakes across the western United States. However, chemical thresholds for phytoplankton responses to N enrichment need to be quantified in additional western U.S. mountain lakes to verify thresholds estimated here and reduce uncertainty in critical loads for phytoplankton responses.

Chemical thresholds

Lake N chemical thresholds defined here for biomass nutrient limitation shifts overlap with those previously defined in western U.S. mountain lakes for different phytoplankton responses (Fig. 1). Thresholds for stimulation of phytoplankton biomass growth in Pacific Northwest mountain lakes (13–25 μg DIN/L; Williams et al. 2016) and in Sierra Nevada mountain lakes (0.3–4 μmol N/L, 5–56 μg N/L; Heard and Sickman 2016) overlap with nutrient limitation shift thresholds defined here (21–53 μg N/L; Table 2). Monod K_s values for the most sensitive species are an order of magnitude or more lower for than our thresholds for limitation shifts (Fig. 1). However, there can be large variability in Monod constants across lakes and regions for some indicator diatoms such as *Fragilaria* species, and some less-sensitive species have a K_s comparable to biomass limitation shift thresholds defined here (Williams et al. 2016). While published threshold concentrations are generally lowest at the species level and are higher for stimulation of phytoplankton biomass growth and nutrient limitation shifts, the variability and overlap of thresholds within and across studies (Fig. 1) highlight that phytoplankton community composition and other within-lake variables can strongly affect chemical thresholds and lake sensitivity to N deposition.

Bergström (2010) previously estimated lake DIN:TP mass ratio thresholds for a 50% (2.2) and 70% probability (3.4) of biomass P limitation using an independent data set. Our corresponding DIN:TP thresholds for 50% (11.4) and 75% probability of P limitation (21.6, based on 25% probability of N limitation, RR-N/RR-P > 1) are higher. Although both studies used binary logistic regression to estimate thresholds, our analyses predicted RR-N/RR-P = 1 as a biological threshold, whereas Bergström (2010) used nutrient limitation categories reported by bioassay studies. Our thresholds are also specific to western U.S. mountain lakes, whereas Bergström (2010) used bioassay data from a few mountain lakes in the Colorado Rocky Mountains and a large number of European lakes. The binary logistic regression model developed in this study explained a similar amount of variance in our western U.S. mountain lake population (R^2_N = 0.68) as that reported by Bergström (2010; R^2_N = 0.72). Compared to the lake population used by Bergström (2010), the population used here had lower N deposition rates, but similar DIN and DIN:TP. Other within-lake factors discussed above can also modify phytoplankton responses to nutrient enrichment and may also contribute to interregional differences.

Critical loads

To our knowledge, only one previous study estimated a N critical load for phytoplankton biomass nutrient limitation shifts. Bergström and Jansson (2006) compiled chlorophyll *a*, nutrient chemistry, and N deposition data for lakes throughout the Northern Hemisphere. They plotted response of regional average chlorophyll *a*:TP ratio to regional average wet inorganic N deposition, which revealed that the increase in lake chlorophyll *a* per unit TP leveled off as N deposition increased (Bergström and Jansson 2006). Based on this plot, the authors estimated N limitation in regions with less than 2.5 kg wet inorganic $N \cdot ha^{-1} \cdot yr^{-1}$, and P limitation in regions with greater than 5 kg wet inorganic

N·ha^{-1}·yr^{-1} (Bergström and Jansson 2006). In this study, critical loads were defined using total (wet plus dry) N deposition estimates, so our estimated critical load of 4.1 kg total N·ha^{-1}·yr^{-1} for a shift to P limitation is lower.

Several previous studies estimated a critical load for NO_3 increases in high-elevation lakes by plotting lake NO_3 across a spatial gradient of N deposition, as in Fig. 2, and inferring a critical load as the deposition rate where elevated lake NO_3 first appears (Baron et al. 2011, Nanus et al. 2017). Using this approach, a critical load of 2.0 kg total N·ha^{-1}·yr^{-1} could be inferred from Fig. 2, which is the same as our minimum critical load necessary to prevent false negatives and is similar to lake NO_3 critical loads estimated by Baron et al. (2011) for the Rockies (3.0 kg total N·ha^{-1}·yr^{-1}) and Sierras (2.0 kg total N·ha^{-1}·yr^{-1}) and by Nanus et al. (2017) for the Greater Yellowstone Area (3.0 kg total N·ha^{-1}·yr^{-1}). Considering these independent estimates are similar and are similar to critical loads for phytoplankton responses quantified here, 2–3 kg total N·ha^{-1}·yr^{-1} may be a robust critical load estimate for the onset of deposition-induced lake nitrate increases in the most sensitive western U.S. mountain lakes. However, as noted above, such critical loads may be prone to type I error because deposition model bias may be large relative to these critical load values.

Many studies have estimated critical loads for growth stimulation of indicator diatom species, with critical load estimates ranging from 1.0 to >10 kg wet inorganic N·ha^{-1}·yr^{-1} (Baron 2006, Saros et al. 2010, Nanus et al. 2012, Sheibley et al. 2014) and <1.5 to >4.0 kg total N·ha^{-1}·yr^{-1} (Nanus et al. 2017). Considering dry deposition ranges from 30% to 90% of total deposition across mountainous regions of the western United States (NADP 2017), our 4.1 kg total N ha^{-1} yr^{-1} overlaps with critical loads previously estimated for indicator diatoms. To compare diatom and biomass nutrient limitation critical loads, we also calculated a range of potential critical loads for *A. formosa* growth responses. We calculated the range of modeled total N deposition rates observed among 385 lakes with nitrate data (nitrate lakes; Fig. 3) where measured lake nitrate exceeds the lowest published *A. formosa* Monod half-saturation constant (K_s; 2.5 μg N/L; Arnett et al. 2012). This analysis is described in detail in Appendix S4. Total N deposition rates in lakes with NO_3–N >2.5 μg N/L ranged from 1.4 to 6.0 kg total N·ha^{-1}·yr^{-1} (mean = 3.3, median = 3.2). Ranges were 1.8–3.3 kg total N·ha^{-1}·yr^{-1} in the Cascades (mean = 2.5), 1.9–6.0 kg total N·ha^{-1}·yr^{-1} in the Rockies (mean = 3.6), and 2.1–5.6 kg total N·ha^{-1}·yr^{-1} in the Sierras (mean = 2.8). Considering that our biomass nutrient limitation shift critical load of 4.1 kg total N·ha^{-1}·yr^{-1} falls within the range of diatom critical load values estimated through this analysis and in previously published studies and that chemical thresholds of phytoplankton species are highly variable (Fig. 1), our biomass nutrient limitation shift critical load may also be protective of indicator diatom changes in some lakes.

Conclusions

This was the first study to estimate critical loads for phytoplankton biomass nutrient limitation shifts in the western United States, and the first to estimate mountain lake phytoplankton critical loads at the scale of the western United States. We estimated N critical loads for phytoplankton biomass nutrient limitation shifts (4.1 kg total N·ha^{-1}·yr^{-1}, range: 2.8–5.2 kg total N·ha^{-1}·yr^{-1}) in mountain lakes across the western United States. Performance analyses predicted that using a critical load of 4.1 kg total N ha^{-1} yr^{-1} in critical load exceedance calculations may yield false-negative prediction of nutrient limitation shifts in 13% of western U.S. mountain lakes. A critical load of 2.0 kg total N ha^{-1} yr^{-1} would likely reduce the occurrence of false positives to near zero, but may be more prone to type I error because deposition model bias is typically large relative to low critical load values. Presented critical loads overlap with those estimated for mountain lakes in previous studies, but also have substantial uncertainties related to accuracy of N deposition models and GIS data sources used, and variability across lakes in the N concentration required to elicit a nutrient limitation shift that critical load users should consider. Results provide critical load values managers can use to assess N deposition impacts on western U.S. mountain lakes, and associated performance information managers can use to consider if presented critical loads are adequate for specific management applications.

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Supporting Information

Additional Supporting Information may be found online at: http://onlinelibrary.wiley.com/doi/10.1002/ecs2.1955/full