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Mapping critical loads of nitrogen deposition for aquatic ecosystems in the Rocky Mountains, USA

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ABSTRACT

Spatially explicit estimates of critical loads of nitrogen (N) deposition (CL_{Ndep}) for nutrient enrichment in aquatic ecosystems were developed for the Rocky Mountains, USA, using a geostatistical approach. The lowest CL_{Ndep} estimates ($<1.5 \pm 1$ kg N ha^{-1} yr^{-1}) occurred in high-elevation basins with steep slopes, sparse vegetation, and abundance of exposed bedrock and talus. These areas often correspond with areas of high N deposition (>3 kg N ha^{-1} yr^{-1}), resulting in CL_{Ndep} exceedances $\geq 1.5 \pm 1$ kg N ha^{-1} yr^{-1}. CL_{Ndep} and CL_{Ndep} exceedances exhibit substantial spatial variability related to basin characteristics and are highly sensitive to the NO_3^- threshold at which ecological effects are thought to occur. Based on an NO_3^- threshold of 0.5 μmol L^{-1}, N deposition exceeds CL_{Ndep} in 21 ± 8% of the study area; thus, broad areas of the Rocky Mountains may be impacted by excess N deposition, with greatest impacts at high elevations.

1. Introduction

High rates of dissolved inorganic nitrogen (N) (inorganic N = nitrate + ammonium) deposition can considerably change the nutrient status and function of vulnerable high-elevation aquatic ecosystems in the western United States (US) (Baron, 2006; Williams et al., 1996). In the Rocky Mountains, inorganic wet N deposition (hereafter, N deposition) has increased over recent years as a result of anthropogenic activities (Ingersoll et al., 2008; Nanus et al., 2008). High-elevation aquatic ecosystems in the Rocky Mountains are particularly sensitive to acidification and nutrient enrichment due to steep topography, thin and rocky soils, sparse vegetation, short growing seasons, and rapid release of pollutants in snowmelt runoff during spring (Baron and Campbell, 1997; Clow and Sueker, 2000; Williams et al., 1996). Previous work has connected increasing rates of N deposition with observed ecosystem effects at select alpine lakes in the Rocky Mountains (Baron et al., 2000; Elser et al., 2009a, 2009b; Nanus et al., 2008; Theobald et al., 2010). These changes include elevated surface-water nitrate concentration (hereafter, surface-water NO_3^-) (Baron et al., 2000; Nanus et al., 2008; Theobald et al., 2010), episodic acidification of surface waters (Williams and Tonnessen, 2000), changes in diatom species indicative of N enrichment (Saros et al., 2010; Wolfe et al., 2001), and shifts in patterns of ecological nutrient limitation from predominantly N-limited to P-limited (Elser et al., 2009a, 2009b). Sensitive ecosystems are common in Class 1 Wilderness Areas in the western US, where air-quality-related values are protected from degradation under the Clean Air Act and Amendments (42 USC 7470[2] and 42 USC 7475[d][2]). US air-quality policy makers and regulators need a standardized approach for assessing ecosystem sensitivity to N deposition, which is a necessary prelude to developing policy to protect sensitive ecosystems from adverse effects (Porter et al., 2005; Porter and Johnson, 2007).

In Europe and Canada, policy makers have adopted the "critical loads" approach, which specifies the amount of deposition of a given pollutant that an ecosystem can receive below which ecological effects are thought not to occur (Jeffries et al., 2010; Ouimet et al., 2006; Porter et al., 2005; UNECE ICP, 2004). In the US, researchers have developed critical loads for terrestrial and aquatic ecosystems using a variety of approaches, including empirical (regression) models and simple mass balance models (Baron, 2006; Baron et al., 2011; Bowman et al., 2006; Fenn et al., 2010, 2008; McNulty et al., 2007; Pardo et al., 2011; Porter and Johnson, 2007; Williams and Tonnessen, 2000). In the western US, ecosystem

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sensitivity can vary considerably between nearby basins due to differences in basin characteristics and N deposition (Clow et al., 2010; Nanus et al., 2009).

Recent studies have predicted surface-water NO_3^- and acid-neutralizing capacity (ANC) in high-elevation basins using empirical models, which relate observed surface-water NO_3^- or ANC to basin characteristics and N or S deposition (Clow et al., 2010; Clow and Sueker, 2000; Nanus et al., 2009; Sickman et al., 2002). The empirical critical loads approach builds on these studies and is based on relating data for ecological indicators, which tend to be limited in availability, to surface-water chemistry from more spatially extensive networks. In the case of nutrient enrichment of aquatic ecosystems, diatom species composition is a useful indicator of ecosystem health, but the spatial extent of diatom data is insufficient for mapping critical loads. Instead, the response of diatoms to variations in surface-water NO_3^- can be established through nutrient addition experiments, and a threshold NO_3^- concentration at which ecological effects occur can be identified.

Several studies in the Rocky Mountains have evaluated responses of lake phytoplankton to in-situ nutrient additions in low-N lakes ($\leq$1.4 μmol NO_3–N L^{-1}). Experiments in the Snowy Range, Wyoming, documented a strong increase in phytoplankton abundance to N additions (71 μmol NO_3–N L^{-1}) (Lafrancois et al., 2004; Nydick et al., 2004). Nutrient additions (163 μmol NO_3–N L^{-1}) to three lakes in Rocky Mountain National Park (RMNP) elicited varying responses, with a low-N lake showing increased phytoplankton biomass (Nydick et al., 2003). The addition of N (18 μmol NO_3–N L^{-1}) to a lake in the Beartooth Mountains, Wyoming, stimulated growth of the diatoms *A. formosa* and *F. crotonensis* (Saros et al., 2005). While these experiments clearly documented that phytoplankton in low-N lakes will respond to the addition of N, they were not designed to identify the minimum NO_3^- at which a biologic response occurs. Defining the $NO_{3(threshold)}$ requires evaluating phytoplankton responses over a concentration gradient.

The objective of this study was to develop spatially explicit estimates of critical loads for nutrient-enrichment effects of N deposition (CL_{Ndep}) in high-elevation areas of the Rocky Mountain region, which extends from northern New Mexico to the Canadian Border (Fig. 1a). This is the first study to map spatial variations in estimated CL_{Ndep}, and explicitly link them to variations in basin characteristics. Our approach was to (1) develop high-resolution gridded estimates of N deposition based on observed N concentrations in precipitation from existing networks, and precipitation amounts obtained from the Parameter-elevation Regressions on Independent Slopes Model (PRISM; Climate Source, 2007), (2) perform a geographic information system (GIS) analysis to quantify basin characteristics and N deposition for basins where surface-water NO_3^- has been measured, (3) develop regression models for surface-water NO_3^-, with basin characteristics and N deposition as explanatory variables, (4) apply the surface-water NO_3^- model to unsampled basins in the study area, (5) identify threshold values of NO_3^- at which ecological effects are likely to occur (hereafter, $NO_{3(threshold)}$), and (6) use the information developed in steps 1–5 to estimate and map CL_{Ndep} and exceedances of CL_{Ndep}.

One challenge in this study was the lack of regionally consistent, high-resolution input landcover and geology data layers. Previous studies using the empirical approach to map predicted surface-water NO_3^- or ANC in the western US have relied on data available at 1:50 000, or finer, scale (e.g., Clow et al., 2010; Clow and Sueker, 2000; Nanus et al., 2009); however, these types of high-resolution digital landcover and geology maps covering the entire Rocky Mountains do not exist. It was hypothesized that this might limit the predictive power of the regional models and cause them to under represent highly sensitive areas, which would have low CL_{Ndep} (e.g., <1.5 kg ha^{-1}). To test this, a fine-scale analysis was

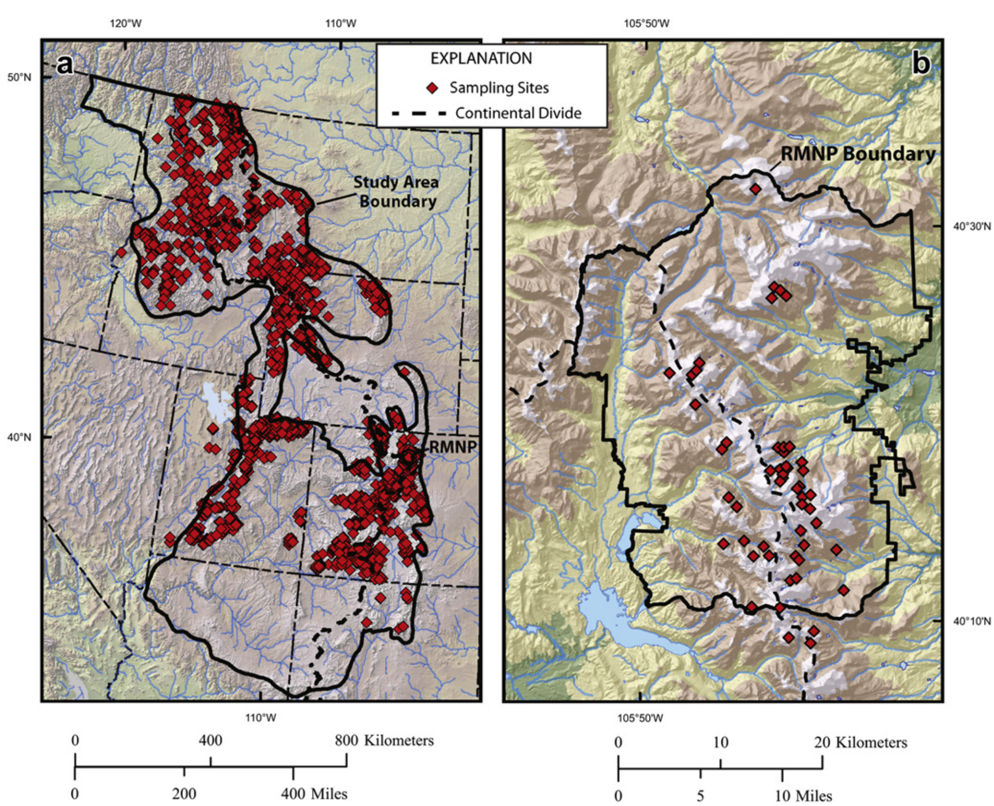

Fig. 1. Sites and study-area boundaries for (a) coarse-scale analysis, and (b) fine-scale analysis.

conducted for RMNP (Fig. 1b), where high-resolution datasets (≤1:50 000) exist for geology, landcover, and topography. Results from the fine-scale analysis were compared to those from the regional (coarse-scale) analysis to check for differences in spatial patterns in sensitivity to N deposition.

2. Methods

2.1. Overview

Previous work indicates that surface-water NO_3^- at high elevations in the Rocky Mountains varies primarily in response to basin characteristics and N deposition (Clow and Sueker, 2000). The relations may be expressed mathematically using a multiple linear regression (MLR) equation of the form:

$$NO_3(\text{observed}) = x(\text{slope}) - y(\%\ \text{forest}) + z(\text{N deposition}) \pm \text{intercept},$$

where x, y, and z are parameter coefficients. The parameters in this equation are examples, and other parameters could be used (e.g., elevation). Appropriate model parameters and their coefficients may be identified using stepwise MLR, in which observed NO_3^- is regressed against potential explanatory variables. The "best" model will include the combination of parameters that provide the lowest standard error and highest amount of variance explained (r^2). After parameterizing the model, one may substitute $NO_{3(\text{threshold})}$ for $NO_{3(\text{observed})}$, and rearrange the equation to solve for CL_{Ndep}:

$$CL_{Ndep} = [NO_3(\text{threshold}) - x(\text{slope}) + y(\%\ \text{forest}) \pm \text{intercept}]/z.$$

$NO_{3(\text{threshold})}$, parameter coefficients, and intercept are constants, so CL_{Ndep} will vary spatially as a function of basin characteristics. In the following section, we provide details on the methods used during each key step in the study.

2.2. GIS analysis

2.2.1. Inorganic nitrogen deposition

CL_{Ndep} may be calculated based on wet deposition, or based on the sum of wet and dry deposition. In this study, wet deposition was used because the spatial distribution of dry deposition is highly uncertain. Wet deposition of N is a function of the amount and inorganic N concentration of precipitation; it was calculated following the method developed by Nanus et al. (2003), which optimizes estimation of NO_3^- deposition by using the most extensive available NO_3^- concentration datasets and spatially explicit estimates of precipitation. Average annual precipitation amount estimates were derived from the 400-m resolution PRISM model (1971–2000) (Climate Source, 2007). Average annual inorganic N concentrations from National Atmospheric Deposition Program/National Trends Network (NADP) wet-deposition sites (http://nadp.sws.uiuc.edu/; accessed 10/14/2011) were combined with those from the U.S. Geological Survey (USGS) Rocky Mountain Snowpack Synoptic program (RMS; http://co.water.usgs.gov/projects/RM_snowpack/; accessed 10/14/2011) to obtain a spatially extensive dataset covering medium to high elevations in the Rocky Mountains. Previous studies indicate that inorganic N concentrations in the two datasets are comparable and can be combined to increase the spatial density of sites (Clow et al., 2002). Sites were screened to include those with at least 5 years of data during 1993–2010, and at least 35 cm yr^{-1} average annual precipitation (to avoid low-elevation desert sites), which yielded 26 NADP and 73 RMS sites for which we determined a long-term average (≥5 years) inorganic N concentration by calculating the geometric mean for each site. The data were spatially interpolated using kriging (ESRI, 2001) to obtain inorganic N concentration estimates, which were then multiplied by 30-year average annual precipitation amounts from PRISM to obtain gridded estimates of average annual N deposition at 400-m resolution. Basin boundaries were overlaid on the N deposition maps to calculate N deposition for each basin.

2.2.2. Site selection and basin characteristics

Basin characteristics were quantified in a GIS framework for areas upstream from 475 high-elevation sites where late-growing-season surface-water NO_3^- was measured during 1983–2007 in the Rocky Mountains. Late-growing-season was defined as mid-August to mid-October, and was chosen because surface-water NO_3^- and hydrologic variability tend to be low and stable at that time of year. Surface-water NO_3^- data were derived primarily from the 1985 Western Lake Survey and synoptic water-quality surveys conducted by the U.S. Department of Agriculture (USDA) Forest Service and USGS (Landers et al., 1987; Mast, 2007). Sites were restricted to elevations >1250-m, where sensitive ecosystems are most likely to occur. For sites with multiple years of late-growing-season data, we calculated an average surface-water NO_3^-. GIS data included soil characteristics from the 1994 State Soil Geographic dataset (STATSGO; 1:250 000; http://soils.usda.gov/survey/geography/statsgo/; accessed 10/14/2011); landcover from the GAP Analysis Program level one dataset (1:100 000; http://gapanalysis.usgs.gov/data/land-cover-data/; accessed 10/14/2011); hydrology from the National Hydrography Dataset (NHD+; 1:100 000; http://nhd.usgs.gov/; accessed 10/14/2011); and hypsography from a 30-m digital elevation model (DEM; http://ned.usgs.gov/; accessed 10/14/2011). A list of potential explanatory variables derived from these GIS layers and used in the regional and fine-scale statistical analysis is provided in Table 1. Selection of potential explanatory variables was guided by current understanding of biogeochemical processes influencing N assimilation in high-elevation ecosystems (Clow et al., 2010; Clow and Sueker, 2000; Sickman et al., 2001, 2002).

Datasets used in the fine-scale analysis for RMNP included vegetation (1:12 000; National Park Service (NPS) geospatial dataset 1043989; https://irma.nps.gov/App/Reference/Welcome; accessed 10/14/2011), geology (1:50 000; Braddock and Cole, 1990; NPS geospatial dataset 1038961; https://irma.nps.gov/App/Reference/Welcome; accessed 10/14/2011), and 30-m DEM (http://ned.usgs.gov/; accessed 10/14/2011). Water-quality data for the fine-scale analysis included 53 sites sampled in RMNP during early-September to early-October 1999 (Clow et al., 2003).

2.3. Development of surface-water nitrate model

Stepwise MLR was used to create a predictive model for surface-water NO_3^- at high elevations of the Rocky Mountains, as in Clow et al. (2010). Explanatory variables entered the model in an iterative fashion, with the variable explaining the most variance in surface-water NO_3^- entering the model first. Variances explained by the remaining variables were recalculated, and the variable that explained the next greatest amount of variance entered the model next. The threshold for inclusion in the MLR was set to $p \le 0.1$. Multicollinearity among explanatory variables was evaluated using the variance inflation factor (Hair et al., 2005), with a threshold for exclusion of 5. The partial regression coefficients in the model represent the

Table 1
Basin Characteristics and Linear (Pearson Product–Moment) Correlations with Surface-Water Nitrate From Coars-Scale Analysis, Grouped by Data Layers.[a]

Variable	Rocky Mountain Region		Rocky Mountain N.P.	
	Correlation	*p*-value	Correlation	*p*-value
Degrees	**Aspect**			
0–45	0.17	*0.000*	0.13	0.077
45–90	−0.04	0.354	−0.01	0.869
90–135	−0.08	0.080	−0.09	0.244
135–180	−0.04	0.323	−0.04	0.590
180–225	−0.17	*0.000*	−0.24	*0.001*
225–270	−0.16	*0.000*	−0.16	*0.038*
270–315	0.05	0.206	0.12	0.105
315–360	0.28	*0.000*	0.21	*0.005*
	Hypsometry			
Basin Area	−0.05	0.212	−0.14	0.058
Min. Elevation	0.21	*0.000*	0.26	*0.000*
Ave. Elevation	0.35	*0.000*	0.38	*0.000*
Max. Elevation	0.39	*0.000*	0.26	*0.000*
Mean Slope	0.51	*0.000*	0.44	*0.000*
Max. Slope	0.30	*0.000*	0.17	*0.027*
Slope > 30°	0.45	*0.000*	0.38	*0.000*
Slope > 30°, North facing	0.45	*0.000*	0.33	*0.000*
Relief	0.21	*0.000*	−0.03	0.696
	Climate			
Average Annual Precipitation	0.11	*0.008*	0.47	*0.000*
	Deposition			
Ammonium Deposition	0.22	*0.000*	0.42	*0.000*
Nitrate Deposition	0.30	*0.000*	0.45	*0.000*
Inorganic Nitrogen Deposition	0.32	*0.000*	0.49	*0.000*
Sulfate Deposition	0.18	*0.000*	0.46	*0.000*
Acid Deposition	0.12	*0.006*	0.43	*0.000*
	Landcover			
Barren	0.60	*0.000*	0.59	*0.000*
Deciduous Forest	−0.11	*0.013*	−0.08	0.274
Mixed Forest	−0.11	*0.008*	−0.13	0.079
Evergreen Forest	−0.31	*0.000*	−0.38	*0.000*
Grassland	−0.21	*0.000*	−0.19	*0.010*
Pasture	0.01	0.775	0.01	0.929
Shrub	−0.25	*0.000*	−0.03	0.650
Open Water, Snow	0.17	*0.000*	−0.13	0.091
Wetland	−0.18	*0.000*	−0.08	0.274
Developed	0.02	0.582	0.08	0.308
	Soil			
Organic Matter	0.40	*0.000*	0.30	*0.000*
Thickness	−0.07	0.130	−0.31	*0.000*
Hydrogroup	−0.15	*0.000*	−0.28	*0.000*
Permeability	0.37	*0.000*	0.31	*0.000*

[a] Statistically significant correlations ($p \le 0.05$) are italicized.

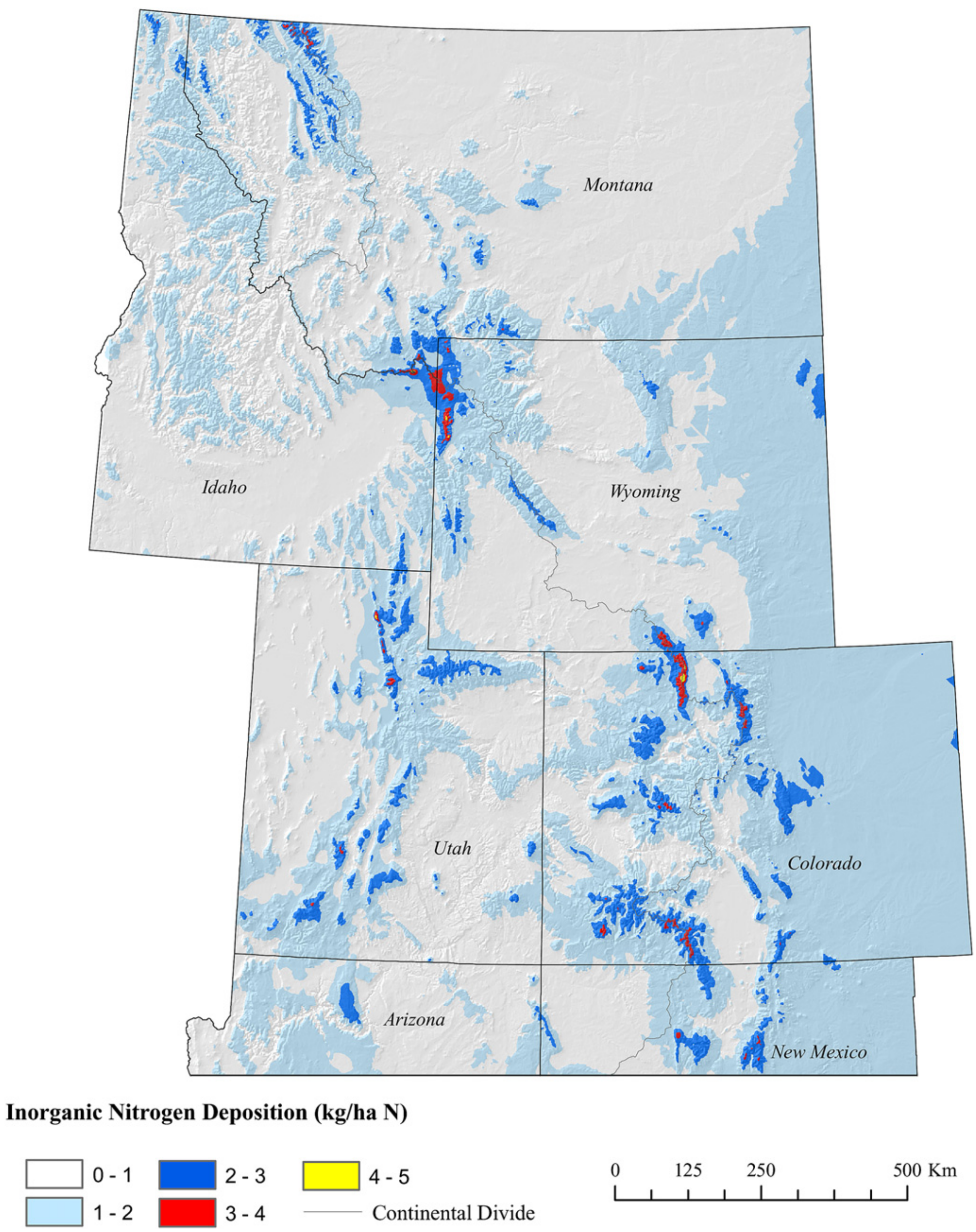

Fig. 2. Estimated annual wet inorganic N deposition in the Rocky Mountain region.

independent contributions of each explanatory variable (Kachigan, 1986). Residuals plots and normal probability plots were used to screen outliers and to check for violation of assumptions of normality, linearity, and homoscedasticity (Kachigan, 1986). The MLR model was applied to each NHD+ stream reach ($n = 254\,892$) to predict surface-water NO_3^- at high elevations throughout the Rocky Mountains. Similar methods were used in the fine-scale analysis to develop and apply a separate MLR model for surface-water NO_3^- in RMNP.

2.4. *Identifying threshold nitrate concentrations*

2.4.1. *Nutrient addition experiments*

To define a $NO_{3(\text{threshold})}$, we quantified the response of the diatom *A. formosa* to changes in inorganic N concentrations during nutrient addition experiments. This species of diatom was selected because of widespread changes in its relative abundance across high-elevation lakes of the western US (Saros et al., 2010, 2005; Wolfe et al., 2001), and previous experimental manipulations in the central Rocky Mountains that demonstrated N is the nutrient that stimulates growth of this species (Saros et al., 2005).

Batch culture experiments were used to assess growth rates of *A. formosa* along a NO_3^- gradient. For these experiments, whole lake water was used from Fossil Lake, Beartooth Mountains, Wyoming. This lake had *A. formosa* present and also had very low initial concentrations of N (<0.07 μM L^{-1}). Lake water was collected at 3-m depth using a van Dorn bottle, and pre-screened through 100-μm mesh to remove zooplankton grazers. Three 50-mL aliquots of this initial water were preserved with Lugol's iodine solution and used to determine initial cell densities. Six 500-mL aliquots of this water were added to 500-mL bottles that had been acid washed (20% HCl) and rinsed thoroughly with deionized water. All bottles received phosphorus (1 μM NaH_2PO_4) and silica (100 μM Na_2SiO_3) amendments so that the response to N additions could be isolated. The bottles then received the following amounts of N in the form of $NaNO_3$: 0, 0.005, 0.01, 0.05, 0.1, and 0.5 μM. After mixing, the amended water was distributed into three flat, polystyrene culture flasks. The 18 flasks were placed between two pieces of 3.2-mm Lucite plastic sheeting held together with 2.5-cm PVC pipe, as in Michel et al. (2006), and incubated in the lake at 3-m depth for 6 days. Lake water transparency data are provided in Rose et al. (2009), and reveal that only 1% of the surface ultraviolet-B radiation remained at the incubation depth, while the 1% attenuation depth for photosynthetically active radiation was 15.6-m.

At the end of the experiment, Lugol's iodine was added to each flask to preserve cells. The density of *A. formosa* was determined in each flask and the three initial samples by settling in an Utermöhl-style chamber and examining through a Nikon

Table 2
Basin Characteristics and Linear (Pearson Product–Moment) Correlations with Surface-Water Nitrate from Fine-scale Analysis on Rocky Mountain National Park, Grouped by Data Layers.[a]

Variable	Correlation	*p*-value
	Hypsometry	
Median Slope	0.44	*0.001*
% Slopes > 30°	0.50	*0.000*
	Geology Layer	
Colluvium	−0.25	0.075
Talus	0.11	0.428
Rock Glacier	−0.01	0.967
Pleistocene Till	−0.26	0.056
Open Water	0.00	0.992
Schist	0.13	0.337
Granite	0.09	0.530
Snow and Ice	0.56	*0.000*
Holocene Till	0.15	0.276
Young Debris	0.15	0.292
Old Debris	−0.33	*0.016*
	Landcover	
Tundra	−0.57	*0.000*
Bedrock	0.57	*0.000*
Open Water	−0.05	0.741
Snow and Ice	0.43	*0.001*
Forest	−0.33	*0.017*
Wetland	−0.13	0.341

[a] Statistically significant correlations ($p \leq 0.05$) are italicized.

TS-100 inverted microscope at 400× magnification. Cell densities were used to calculate growth rates and Monod growth kinetics was calculated using the program JMP (SAS Institute Inc., Cary, NC, USA), as described in Michel et al. (2006). Assuming that the growth of *A. formosa* along the NO_3^- gradient follows standard Monod growth kinetics as described in Michel et al. (2006), the NO_3^- concentration at which *A. formosa* first achieved maximum growth rates was used as the $NO_{3(\text{threshold})}$.

3. Results and discussion

3.1. Atmospheric deposition of inorganic nitrogen

Precipitation patterns in the Rocky Mountains are complex, but generally, average annual precipitation amount increases as a function of elevation and latitude (Barry, 2008). Inorganic N concentrations in precipitation showed the opposite pattern, with higher inorganic N concentrations at lower elevations. Inorganic N concentrations showed less variance than precipitation amounts. Thus, variation in precipitation amount tended to drive patterns in N deposition. N deposition was greatest at high elevations, exceeding 3 kg N ha^{-1} yr^{-1} in several locations including the Front Range, Colorado, and the Wasatch Front, Utah, which are adjacent to large urban areas (Fig. 2). Elevated levels of N deposition (>3 kg N ha^{-1} yr^{-1}) were also found at high elevations in the Park Range and San Juan Mountains, Colorado, the Greater Yellowstone Ecosystem, Wyoming, and southern Wyoming (Fig. 2). Median N deposition estimated for the region was 1.1 ± 0.5 kg N ha^{-1} yr^{-1}. Study area N deposition values <1 kg N ha^{-1} yr^{-1} were generally found at lower elevations (<3000-m), far from large emissions sources (Fig. 2).

3.2. Controls on surface-water nitrate concentrations

Basin characteristics may be considered surrogates for processes or lack of processes that tend to occur or be more active in certain environments; thus, correlations between basin characteristics and surface-water NO_3^- may provide useful information about processes influencing surface-water NO_3^- (Clow et al., 2010). Surface-water NO_3^-, for example, was positively correlated with north-facing aspects (Table 1), probably due to short growing seasons and low N assimilation rates in these cold, shaded environments. In contrast, surface-water NO_3^- was negatively correlated with southwest-facing aspects (180–225°) (Table 1), which tend to be warm and have longer growing seasons than most other aspects. Surface-water NO_3^- was positively correlated with elevation (Table 1), which may be explained by a decrease in soil, vegetation, air temperature, and growing season length as elevation increases. Surface-water NO_3^- was positively correlated with slope, probably because steep slopes tend to have short hydrologic residence times and minimal soil and vegetation (again limiting N uptake). Surface-water NO_3^- was positively correlated with N deposition (Table 1), reflecting the importance of N deposition as a source of N in Rocky Mountain surface waters (Nanus et al., 2008). Landcover exerted substantial control on surface-water NO_3^-, with barren land (bedrock, talus, colluvium) having the strongest positive linear correlation to NO_3^- among all basin characteristics (Pearson Product–Moment correlation coefficient = 0.60; Table 1).

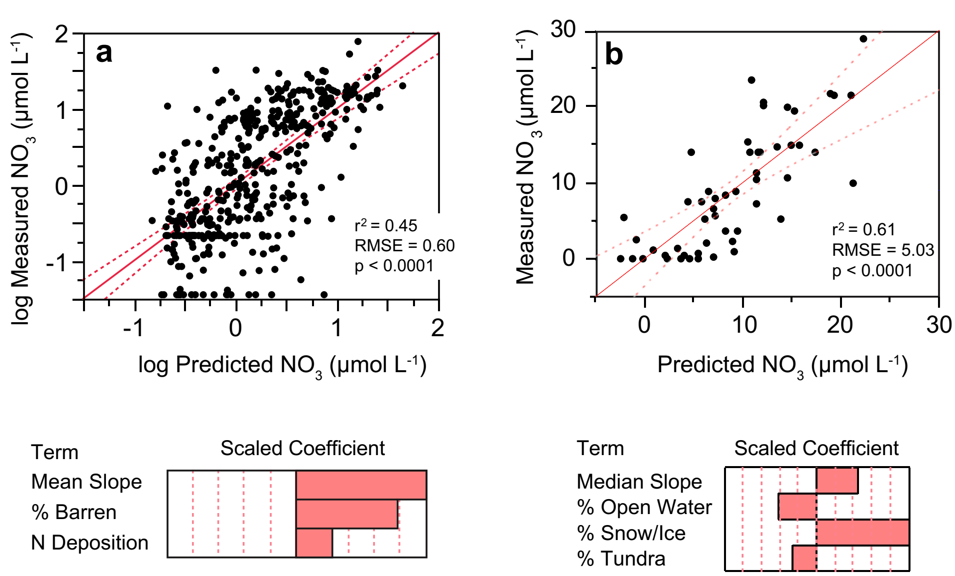

Fig. 3. Predicted versus measured log NO_3^- for surface-water samples used in (a) coarse-scale analysis, and (b) fine-scale analysis. Solid line in upper panels represents line of fit; dashed lines represent 95% confidence intervals. Scaled coefficients are beta coefficients centered by mean, scaled by range divided by 2, and show the relative influence of factors in the regression equation. RMSE is root mean square error in microequivalents per liter.

Surface-water NO_3^- was negatively correlated with forest, grassland, shrub, and wetlands, reflecting uptake of NO_3^- in those environments. Soil organic matter and permeability were positively correlated with surface-water NO_3^- (Table 1).

The correlations observed for the Rocky Mountains tended to hold for RMNP (for consistency, all correlations presented in Table 1 were calculated using the coarse-scale GIS datasets). They are also consistent with correlations observed in previous studies using fine-scale datasets in National Parks of the western US (Clow et al., 2010; Clow and Sueker, 2000; Nanus et al., 2009), and with results of the fine-scale analysis performed in this study using the 1999 RMNP water-quality dataset (Table 2). The observed correlations between surface-water NO_3^- and N deposition are consistent with those documented in the western US by Sickman et al. (2002), the Swiss Alps by Lepori et al. (2003), and in the northern Hemisphere by Bergström and Jansson (2006).

In the regional stepwise MLR analysis, the variables in the "best" model for predicting surface-water NO_3^- included mean slope, percent barren land, and N deposition, in decreasing order of importance (Fig. 3a). The model had only modest predictive power, explaining 45% of the variance in observed surface-water NO_3^-, and tended to over-predict surface-water NO_3^- at the low end of the range (Fig. 3a). Uncertainty in predicted surface-water NO_3^-, expressed as estimate ± 95% confidence interval (CI), was generally 20–30%, and was greatest at the upper and lower ends of the range of values (Fig. 3a).

The MLR equation from the regional analysis was applied to all of the NHD+ stream reaches in the study area to predict surface-water NO_3^- in the Rocky Mountains. Predicted surface-water NO_3^- was less than 3 ± 0.6 μmol L^{-1} in over 95% of the study area, but it exceeded 5 ± 1.6 μmol L^{-1} in most high-elevation areas (≥3000 m) of the Southern Rockies, including most of RMNP (Fig. 4).

For the fine-scale analysis in RMNP, the MLR model explained 61% of the variance in surface-water NO_3^-, and included percent steep slopes (slopes > 30°), percent open water, percent snow/ice, and percent tundra (Fig. 3b). Snow/ice and steep slopes were

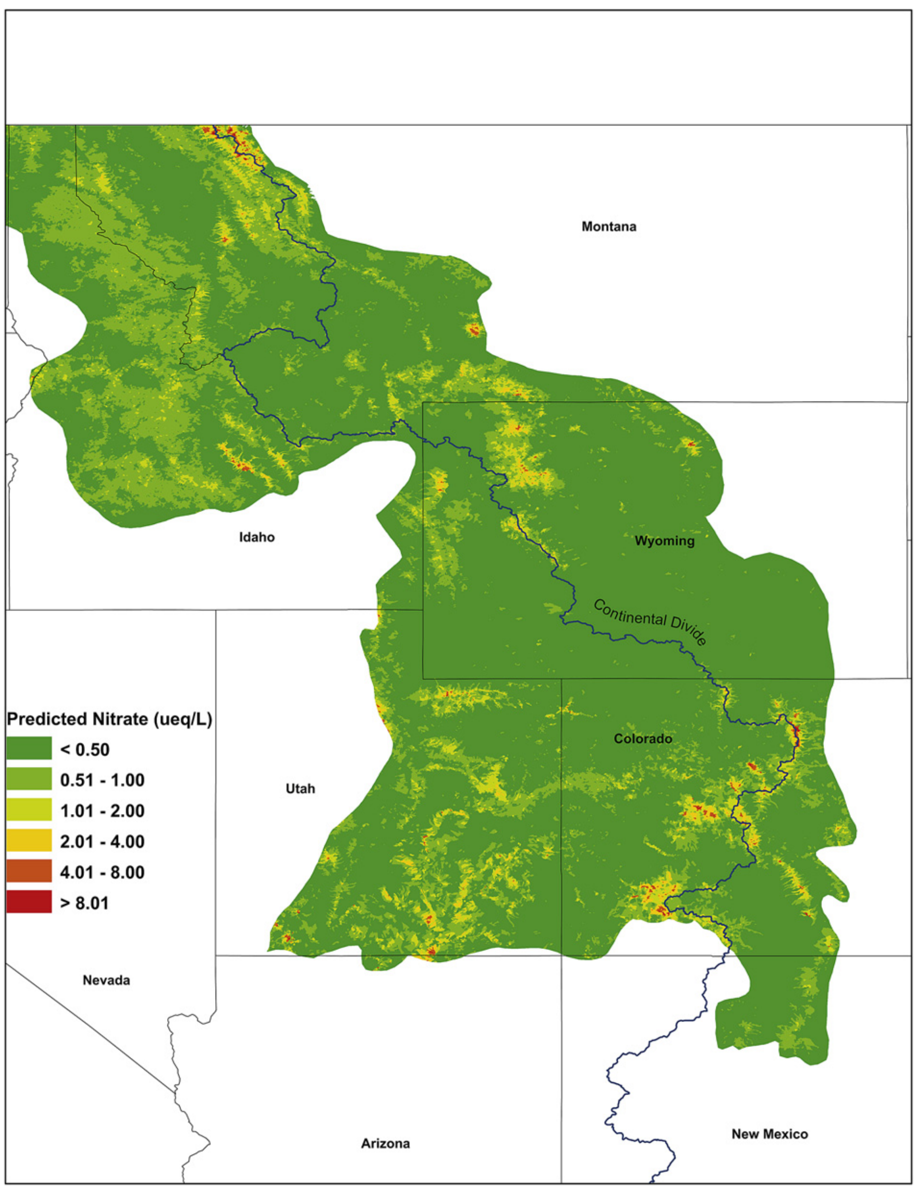

Fig. 4. Predicted late-growing season (mid-August to mid-October) surface-water NO_3^- concentrations from coarse-scale analysis. Results pertain to streams in each subbasin identified in the NHD+ dataset in the Rocky Mountain region.

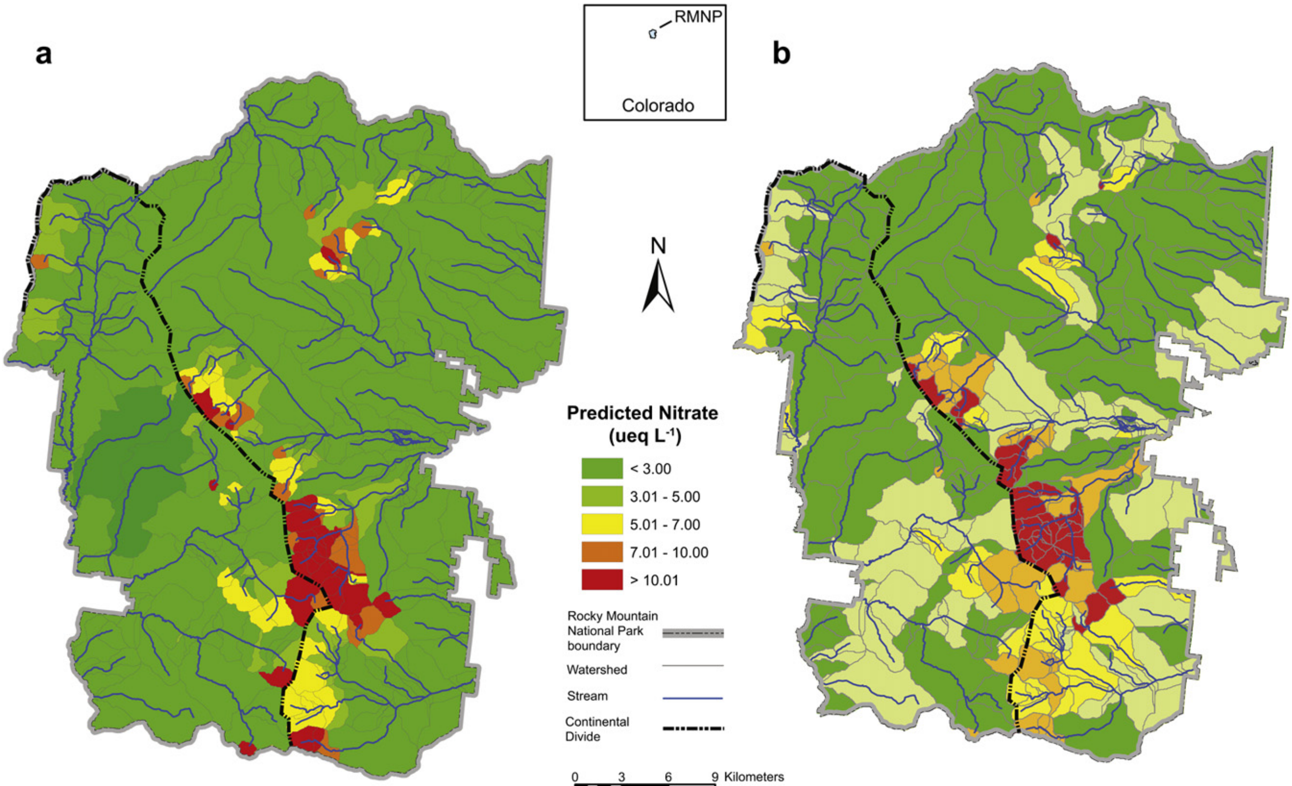

Fig. 5. Predicted late-growing season (mid-August to mid-October) surface-water NO_3^- concentrations from (a) coarse-scale analysis, and (b) fine-scale analysis. Results pertain to streams in each subbasin identified in the NHD+ dataset in Rocky Mountain National Park.

positive influences on surface-water NO_3^-, and tundra and open water were negative influences. Surface-water NO_3^- predicted for the NHD+ stream reaches in RMNP using the fine-scale MLR model were highly variable, with low concentrations in forested, moderate-elevation areas, and high concentrations in steep, high-elevation cirques near the Continental Divide.

In general, surface-water NO_3^- predictions in RMNP were lower in the coarse-scale analysis than the fine-scale analysis (Fig. 5). This is probably due, in part, to the coarse resolution of GIS data available for the regional analysis, which did not capture important fine-scale landscape features, such as small water-bodies, snow fields, and steep slopes. Another reason for differences in results pertains to interannual variability in observed surface-water NO_3^-. The fine-scale analysis used data from a synoptic water-quality survey conducted in 1999, whereas the regional analysis used data from multiple years. Antecedent weather affects hydrologic conditions (particularly flushing rates), and thus, can cause interannual variations in surface-water NO_3^-, which contributes to error in the MLR analysis (Sickman et al., 2001). To test this, an MLR analysis was conducted using fine-scale GIS datasets for RMNP and all available late-growing-season surface-water NO_3^-, including the 1999 survey data. The "best" MLR model from this analysis had an r^2 of 0.35 versus 0.61 for the MLR using just the 1999 synoptic survey data.

3.3. Nitrate threshold concentrations

Results of the growth kinetic experiments with the diatom *A. formosa* indicated a maximum growth rate of 0.2 ± 0.04 day^{-1} at 0.5 μM (Fig. 6). This may be considered the surface-water NO_3^- that elicits a major change in this species, and thus, can be used as the $NO_{3(threshold)}$. This value is considered a conservative estimate given that if the maximum growth curve occurred between 0.1 μM and 0.5 μM it would lower the $NO_{3(threshold)}$ and increase our estimate of areas that exceed CL_{Ndep}. Based on a simple comparison of lake NO_3^- concentrations to estimated N deposition for high elevations in RMNP, Theobald et al. (2010) estimated an $NO_{3(threshold)}$ of 1.6 μmol L^{-1}. No other $NO_{3(threshold)}$ estimates for N-enrichment effects of atmospheric N on aquatic biota have been made for the Rocky Mountains. Given this range of $NO_{3(threshold)}$, we chose to calculate CL_{Ndep} using four different $NO_{3(threshold)}$ estimates spanning a range of plausible values, including 0.5, 1.0, 1.6 and 2.0 μmol L^{-1}. Our discussion focuses on CL_{Ndep} results based on a $NO_{3(threshold)}$ of 0.5 μmol L^{-1}; maps of critical loads and exceedances calculated using the $NO_{3(threshold)}$ values are presented in the Supplementary Figures.

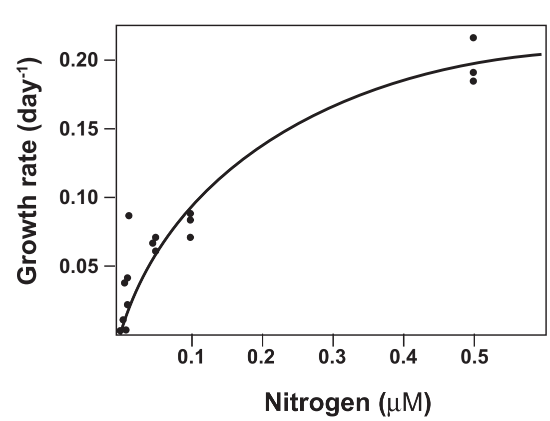

Fig. 6. *Asterionella formosa* growth rate curve.

3.4. Critical loads and exceedances

Estimated CL_{Ndep} exhibited substantial spatial variability, reflecting differences in basin characteristics across the study area as shown in Fig. 7 for a $NO_{3(threshold)}$ of 1.0 μmol L^{-1}. The lowest CL_{Ndep} values (<1.5 ± 1 kg N ha^{-1} yr^{-1}) were in high-elevation basins with steep slopes, sparse vegetation, and an abundance of exposed bedrock and talus (barren terrain). These areas represent the most sensitive environments to N deposition and are prevalent throughout the Rocky Mountains. Lower elevation areas with forest and grassland tended to have much higher estimated CL_{Ndep} values (>10 ± 1 kg N ha^{-1} yr^{-1}), reflecting relatively high N assimilation rates in those areas. The uncertainty in predicted CL_{Ndep} that was attributable to MLR modeling errors was approximately 1 kg N ha^{-1} yr^{-1}.

CL_{Ndep} estimates were very sensitive to estimates in $NO_{3(threshold)}$, as expected given their direct, linear relation (Supplementary Fig. 1). Estimated CL_{Ndep} for Wall Lake (39.9500, −107.2409), Colorado, for example, were 1.5, 4.9, 7.3, and 8.4 kg N ha^{-1} yr^{-1}, based on assumed $NO_{3(threshold)}$ values of 0.5, 1.0, 1.6 and 2.0 μmol L^{-1}. Thus, uncertainty in CL_{Ndep} (and exceedances) attributable to uncertainty in the $NO_{3(threshold)}$ may be much

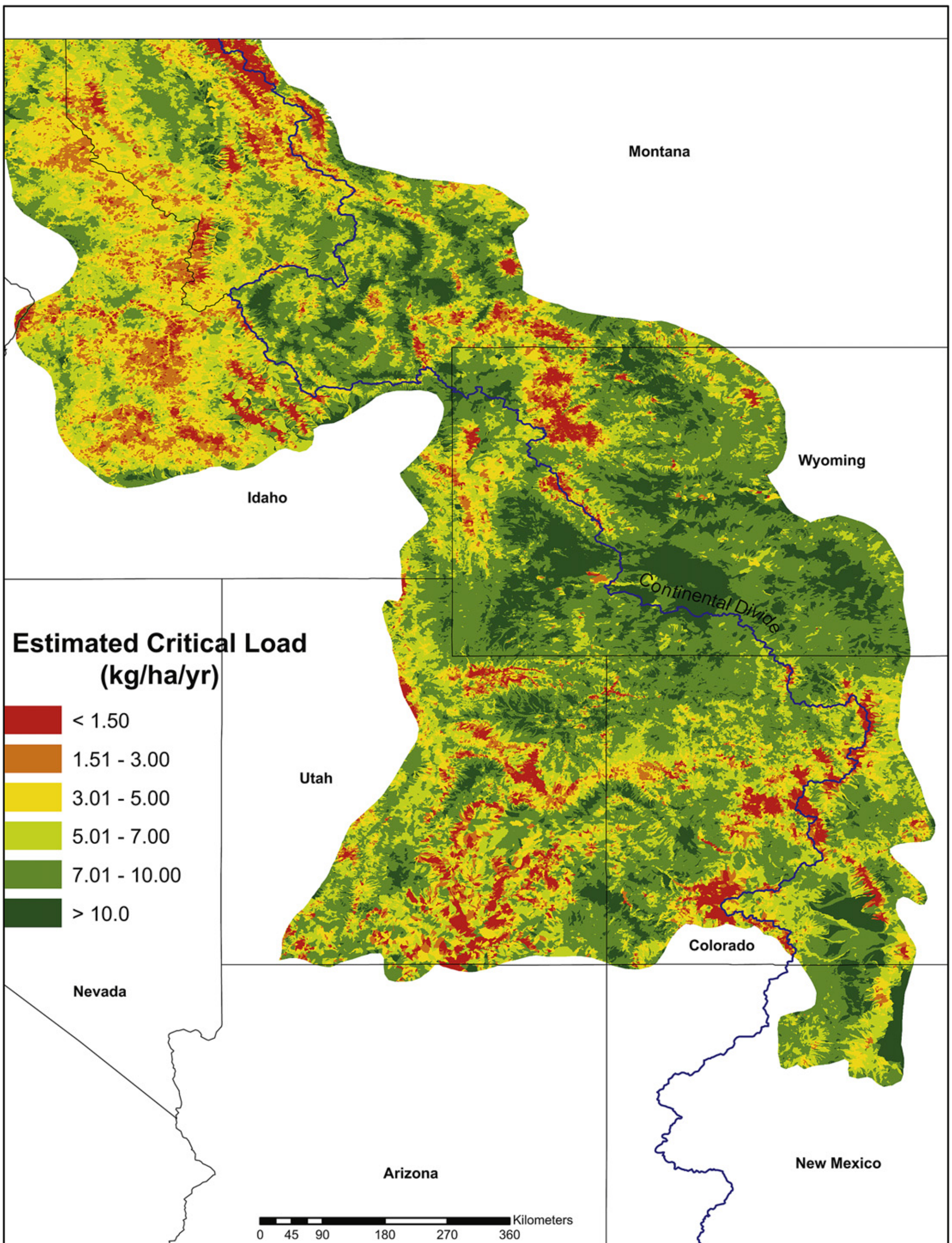

Fig. 7. Estimated critical loads of N deposition (CL_{Ndep}) for nutrient enrichment in aquatic ecosystems of the Rocky Mountain region.

greater than that attributable to other sources of error, such as MLR modeling.

Several other studies have estimated CL_{Ndep} for nutrient enrichment and episodic acidification in parts of the Rocky Mountains. Baron et al. (2011) estimated CL_{Ndep} for nutrient enrichment in sensitive lakes in the western US ranges from 1.0 to 3.0 kg N ha^{-1} yr^{-1}, and the CL_{Ndep} for episodic acidification in low-ANC lakes is 4 kg N ha^{-1} yr^{-1}. Baron (2006) estimated a CL_{Ndep} of 1.5 kg N ha^{-1} yr^{-1} for Loch Vale in RMNP, based on changes in diatom species assemblages and hindcasted NO_x emissions for the Colorado Front Range. Our estimated CL_{Ndep} for Loch Vale was $\leq 1.5 \pm 1$ kg N ha^{-1} yr^{-1} (Fig. 7), consistent with Baron (2006). Other relevant CL_{Ndep} estimates for nutrient-enrichment effects include 2 kg N ha^{-1} yr^{-1} for selected sites in the western US (Pardo et al., 2011), and 2.5 kg N ha^{-1} yr^{-1} for the US (Bergström and Jansson, 2006). Each of these studies presented CL_{Ndep} estimates for "sensitive," usually specific, aquatic ecosystems.

This is the first study that explores how CL_{Ndep} relates to basin characteristics that vary spatially across the Rocky Mountains. Past research has shown that N saturation is occurring in high-elevation

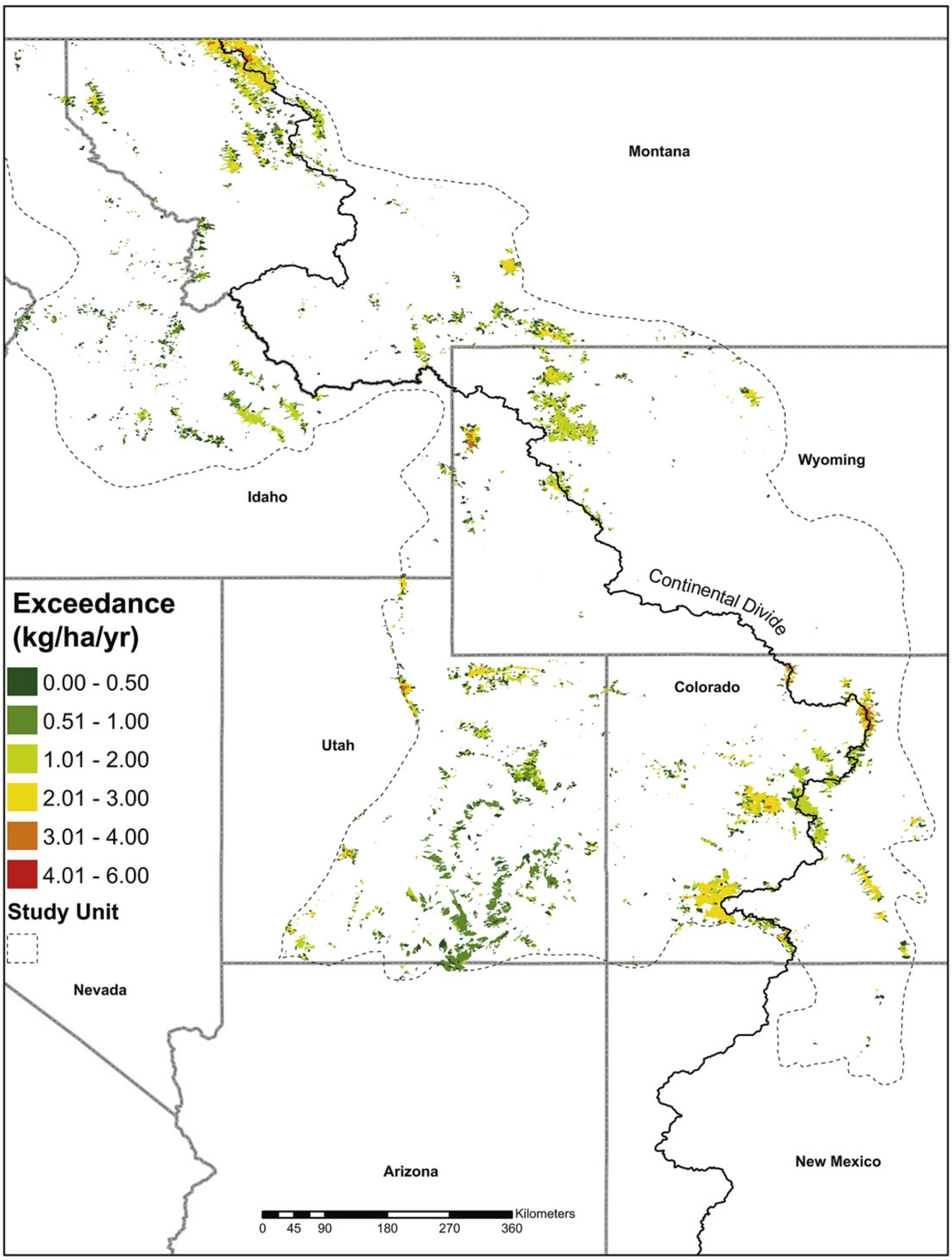

Fig. 8. Estimated exceedances of critical loads of N deposition (CL_{Ndep}) for nutrient enrichment in aquatic ecosystems of the Rocky Mountain region. Exceedances were calculated by subtracting CL_{Ndep} from estimated inorganic N deposition for each NHD+ subbasin in the Rocky Mountain region.

basins in Colorado that receive N deposition in excess of combined plant and microbial demand (Williams et al., 1996; Baron, 2006). This excess N can result in acidification and nutrient enrichment, increasing primary productivity in high-elevation lakes and streams, and altering diatom community structures that form the base of the food web (Wolfe et al., 2001). We show that basins with the lowest CL_{Ndep} ($<1.5 \pm 1$ kg N ha^{-1} yr^{-1}) have steep slopes, high % barren land, and high rates of N deposition. These results support the hypothesis that these basins are most at risk to current or future N saturation due to high rates of N deposition combined with barren land and steep slopes, which have short hydrologic residence times and minimal soil and vegetation which limits N uptake.

Exceedances, which occur where N deposition exceeds CL_{Ndep}, showed substantial spatial variability (Fig. 8), as expected given the variability in N deposition and CL_{Ndep} estimates. Exceedances were $>2 \pm 1$ kg N ha^{-1} yr^{-1} at high elevations, particularly near the Continental Divide, where high N deposition coincided with low CL_{Ndep}. These areas are most at risk to current or future N enrichment effects from N deposition.

As with CL_{Ndep} estimates, exceedances were very sensitive to $NO_{3(threshold)}$ estimates. The effect of $NO_{3(threshold)}$ on CL_{Ndep} exceedances was evaluated by comparing the percentage of the study area currently in exceedance under different $NO_{3(threshold)}$ scenarios. Based on an $NO_{3(threshold)}$ of 0.5 μmol L^{-1}, 21 ± 8% of the study area (>1250-m elevation) was in exceedance of the CL_{Ndep}. The 21 ± 8% CL_{Ndep} exceedance value is reasonable, considering the widespread changes in *A. formosa* at high elevations in the southern and central Rockies (Saros et al., 2010, 2005; Wolfe et al., 2001). Other $NO_{3(threshold)}$ values and their corresponding exceedances were 1 μmol L^{-1} (4.3 ± 1%), 1.6 μmol L^{-1} (1.6 ± 0.5%), and 2 μmol L^{-1} (1.1 ± 0.2%).

In addition to the sources of uncertainty in CL_{Ndep} previously mentioned, there are several other sources of error worth considering. CL_{Ndep} estimates rely on inorganic N concentrations and precipitation amounts interpolated from widely distributed measurement sites, which are sparse at high elevations. GIS input datasets used to build and apply the regression models depend on accurate mapping. The accuracy of the regression models may be expected to decrease as the resolution of GIS decreases; this is particularly important in mountainous areas with high relief, where landcover changes dramatically over short distances (McDonnell et al., 2010). These errors are difficult to quantify and are worthy of additional research but are likely to be smaller than those associated with uncertainty in the $NO_{3(threshold)}$.

4. Conclusions

This study provided some of the first spatially explicit estimates of critical loads for nutrient-enrichment effects of N deposition in the western US. Nutrient-enrichment effects occur at lower levels of N deposition than those that would cause acidification effects (Fenn et al., 2003). Therefore, development of critical loads of N based on nutrient-enrichment effects should also provide protection from acidification. Estimated CL_{Ndep} in the Rocky Mountains ranges from $<1.5 \pm 1$ kg N ha^{-1} yr^{-1} to ≥ -10 kg N ha^{-1} yr^{-1}. The lowest estimated CL_{Ndep} values apply to high-elevation basins with steep slopes, sparse vegetation, and an abundance of exposed bedrock and talus. These areas often correspond with high N deposition (>3 kg N ha^{-1} yr^{-1}), resulting in exceedances greater than 2 kg N ha^{-1} yr^{-1}. Critical loads and exceedances exhibit substantial spatial variability related to basin characteristics and are highly sensitive to estimates of the $NO_{3(threshold)}$ at which ecological effects are thought to occur. Based on a $NO_{3(threshold)}$ of 0.5 μmol L^{-1}, N deposition exceeds CL_{Ndep} in 21 ± 8% of the study area (>1250 m). Thus, broad areas of the Rocky Mountains may be impacted by excess N deposition, and impacts are likely greatest at high elevations. The empirical approach used in this study is readily adaptable to other indicators (e.g., soil chemistry or vegetation chemistry) or ecoregions, and the estimated critical loads can be refined as additional information becomes available.

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Any use of trade names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

Appendix. Supplementary data

Supplementary data related to this article can be found online at doi:10.1016/j.envpol.2012.03.019.

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