Chapter 38
A Comparison of Empirical and Modelled Nitrogen Critical Loads for Mediterranean Forests and Shrublands in California

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Abstract Nitrogen (N) deposition is impacting a number of ecosystem types in California. Critical loads (CLs) for N deposition determined for mixed conifer forests and chaparral/oak woodlands in the Sierra Nevada Mountains of California and the San Bernardino Mountains in southern California using empirical and various modelling approaches were compared. Models used included the Simple Mass Balance (SMB) model for nutrient N and acidification (both site-specific and regional approaches) and the Daycent process-based biogeochemical simulation model. Empirical CLs reported herein were based on responses across N deposition gradients of lichen community functional groups and streamwater nitrate (NO_3) leaching. Broad scale CL mapping for the San Bernardino Mountains using the SMB model resulted in nutrient N CL values that were on average approximately 50 % lower than the empirical CL value for NO_3 leaching (17 kg ha^{-1} year^{-1}) in California mixed conifer forests. Over the range of elevations and vegetation types in the San Bernardino Mountains, SMB CL values ranged from 5.1 to 13.0 kg ha^{-1} year^{-1} for nutrient N. For both the empirical NO_3 leaching CL and the SMB estimate, the CL was generally lower for chaparral vegetation than for forests. The estimated CL for NO_3 leaching derived from the Daycent model was equal to the empirical CL (17 kg ha^{-1} year^{-1}), but the severity and frequency of elevated NO_3 leaching was

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underestimated by Daycent. Statewide empirical CL exceedance maps indicate that 3.3 and 4.5 % of the chaparral and forested areas in California are in excess of the NO_3 leaching CL. Likewise, 23.4, 41.2 and 52.9 % of the mixed conifer forest, oak woodland and chaparral areas are in excess of the empirical N CL for epiphytic lichen community effects, respectively. Eutrophication effects in terrestrial ecosystems of California are widespread, while significant acidification effects are limited to the more polluted sites in southern California.

Keywords Biogeochemical models • Critical load exceedance • Epiphytic lichen communities • Nitrate leaching • Soil acidification

38.1 Introduction

Critical loads (CLs) have been developed as a tool for determining the atmospheric deposition below which significant harmful effects do not occur according to present knowledge. Critical loads can be determined for acidification effects of nitrogen (N) and sulphur (S) deposition, and for nutrient or eutrophication effects of N using models of varying complexity or from field observations at sites with varying levels of atmospheric deposition or nutrient addition studies. The latter two are referred to as empirical CLs.

Modelled CLs can be calculated and mapped across the landscape based on soil mineralogy, chemistry, hydrology and nutrient cycling data. Empirical CLs are determined from field observations under real world conditions. Each approach has its own sources of uncertainty and most appropriate applications. Empirical CLs can be used to evaluate whether modelled CLs are realistic. Moreover, empirical CLs are important because sometimes the important factors controlling ecosystem responses to atmospheric deposition and co-occurring stressors are difficult to model or replicate under experimental conditions. Modelled CLs in conjunction with experimental studies can be useful in determining mechanistic processes and controlling factors affecting the CL and for predicting CLs where empirical data are absent. In this study we compare empirical and modelled CLs for forests, oak woodlands and chaparral ecosystems in California. Both acidification and eutrophication CLs were considered.

38.2 Methods

38.2.1 Study Locations

Critical load calculations were performed using soils data for the San Bernardino Mountains which are located inland from the greater Los Angeles, California urban region. Streamwater samples for nitrate analysis were collected from the San Bernardino Mountains and southwestern Sierra Nevada Mountains in central

California. Lichen community data were collected from the mixed conifer zone of the Sierra Nevada Mountains. Soil parent material in the study sites is weathered or decomposed granitic rock. Soils in the San Bernardino Mountains are generally sandy loam in texture, have weakly developed B horizons and become increasingly coarse textured with depth. Percent base saturation ranges from 70 to 100 %, except in the most polluted western San Bernardino Mountain sites.

38.2.2 Empirical Critical Loads

Empirical CLs for streamwater NO_3 losses were determined across N deposition gradients in the Sierra Nevada and San Bernardino Mountains in California (Fenn et al. 2008, 2010). Nitrogen deposition was measured as throughfall with ion exchange resin collectors (Fenn et al. 2009). In addition, to determine areas in exceedance of the NO_3 leaching CL on a statewide basis, N deposition was estimated using the US/EPA CMAQ model. However, because CMAQ underestimated N deposition in pollution-affected montane regions of California, the CMAQ-simulated N deposition estimates were adjusted based on throughfall N deposition data (Fenn et al. 2010). Empirical CLs for lichen community effects in forests were determined from Forest Inventory and Analysis (FIA) lichen community data and throughfall N deposition data in the Sierra Nevada (Fenn et al. 2008; Jovan 2008). Empirical CLs for lichen community impacts in chaparral/oak woodlands were determined from lichen community surveys in the Greater Central Valley of California (Fenn et al. 2011; Jovan and McCune 2005), which includes the central Coast Ranges and Sierra foothills, in combination with CMAQ N deposition estimates.

The empirical CL for soil acidification was determined from a linear regression ($r^2=0.99$) of throughfall N deposition and soil pH (H^+ concentration) from six sites across a N deposition gradient in the San Bernardino Mountains (Breiner et al. 2007). A soil of pH 4.6 was selected as the threshold pH below which the CL was considered exceeded, based on expert judgment and the study of MacDonald et al. (2002).

38.2.3 Modelled Critical Loads

Critical loads for terrestrial acidification and eutrophication across the San Bernardino Mountains were calculated according to the Steady State Simple Mass Balance (SMB) method as described in the Mapping Manual (ICP Modelling and Mapping Manual 2008). The determination of critical loads for acidification requires the consideration of both S and N deposition on the one side, and plant uptake, weathering, and biochemical immobilization processes on the other side. The deposition of acidifying S and N exceeds a critical value if they alter the chemical characteristics of the soil solution. Indicators for the chemical characterization are (a) aluminum concentration, (b) base cation:aluminium ratio, (c) pH value, (d) base saturation of soils, and (e) acid neutralization capacity (ANC).

The simple mass balance (SMB) equations for the maximum critical load for sulphur-based acidity, $CL_{max}(S)$, and the maximum critical load for N-based acidity, $CL_{max}(N)$, are given by the following equations (ICP Modelling and Mapping Manual 2008):

$$CL_{max}(S) = BC^*_{dep} - Cl^*_{dep} + BC_w - BC_u - ANC_{le(crit)} \quad \text{(Eq. 38.1)}$$

$$CL_{max}(N) = N_u + N_{fire} + N_i + N_{de} + CL_{max}(S) \quad \text{(Eq. 38.2)}$$

with

$CL_{max}(S)$	Critical load for sulphur-based acidity
$CL_{max}(N)$	Critical load for nitrogen-based acidity
BC^*_{dep}	Base cation deposition (sea salt corrected)
Cl^*_{dep}	Chloride deposition (sea salt corrected)
BC_w	Base cation weathering derived from soil type and parent material class
BC_u	Base cation uptake and removal by biomass under steady state conditions
N_u	Nitrogen uptake and removal by biomass under steady state conditions
N_{fire}	Nitrogen loss in smoke by fire
N_i	Long-term immobilization of nitrogen
N_{de}	Denitrification rate
$ANC_{le(crit)}$	Acceptable leaching of acid neutralization capacity

The SMB approach for calculating critical loads for nutrient N assumes steady-state equilibrium of N input, acceptable storage and output. In this case, the N-fixing processes (immobilization, N uptake in the harvested biomass, N loss by fire) and N removal (denitrification, acceptable N leaching) should be in balance with N deposition for steady-state conditions. The mass balance equation to calculate the critical load for nutrient N according to the ICP Modelling and Mapping Manual (2008) is given as:

$$CL_{nut}(N) = N_u + N_{fire} + N_i + N_{le(acc)} + N_{de} \quad \text{(Eq. 38.3)}$$

with

$CL_{nut}(N)$	Critical load for nutrient nitrogen
N_u	Nitrogen uptake and removal by biomass under steady state conditions
N_{fire}	Nitrogen loss in smoke by fire
N_i	Long-term immobilization of nitrogen
$N_{le(acc)}$	Acceptable leaching of nitrogen
N_{de}	Denitrification rate

Nitrogen loss by fire was added to the N uptake term. It was estimated that the annual vegetation loss due to fire at the sites is 0.25 % of the aboveground biomass. Based on estimated plant growth rates and N concentrations in plant tissue, N losses in fire were calculated.

In calculating the acceptable NO_3 leaching for the SMB nutrient N CL [Cl_{nut}(N)], precipitation surplus was determined based on long term streamflow data (Fenn and Poth 1999). The CL for nitrate leaching in mixed conifer forests was also estimated with the Daycent biogeochemical model (Fenn et al. 2008).

38.3 Results and Discussion

38.3.1 Eutrophication Critical Loads: Empirical vs. Modelled

The NO_3 leaching CL derived from the Daycent biogeochemical model simulations (17 kg N ha^{-1} $year^{-1}$) was equal to the empirical CL based on streamwater NO_3 data collected at sites with varying N deposition, notwithstanding the fact that Daycent underestimated NO_3 concentrations in many years (Fenn et al. 2008). Similar to other reports, we found that the steady state SMB CL values for N as a nutrient effects, determined using the approach of the Mapping Methods Manual (ICP Modelling and Mapping Manual 2008), were lower than the empirical CL. In Table 38.1 the values for the calculated SMB parameters for nine sites in the San Bernardino Mountains are given. The SMB CL for the forested sites ranged from 5.6–13.0 kg N ha^{-1} $year^{-1}$ compared to a CL of 5.1 for a chaparral site (Table 38.1).

Nitrogen immobilization (N_i) and denitrification losses (N_{de}) are considered the most uncertain parameters for calculating the SMB CL in the San Bernardino Mountains. The higher calculated denitrification value of 3.7 kg N ha^{-1} $year^{-1}$ at three of the sites (Table 38.1) is due to soil maps indicating alluvial fine textured soils at these locations that are expected to have higher soil moisture content. We consider that these values may be overestimates, particularly at Barton Flats and Holcomb Valley where N deposition is below 10 kg ha^{-1} $year^{-1}$. However in a recent study in a Mediterranean shrubland in southern Italy, high denitrification rates, with dinitrogen as the dominant end product, were reported even in relatively dry soils (Dannenmann et al. 2011). Trace gas losses of N in upland forest and chaparral soils in California occurs primarily as nitric oxide from nitrification. Flux rates from low N deposition sites suggest an ecologically acceptable annual trace gas N loss of 0.5–1.0 kg ha^{-1} $year^{-1}$ (Fenn and Poth 2001).

Both the SMB CL and empirical CLs are known to entail considerable uncertainty, and SMB is probably best used for broad scale mapping of CLs to highlight areas at risk, and when overlain with atmospheric deposition maps, to determine areas of likely CL exceedance. Improved mechanistic understanding and definition of the individual terms used to derive the SMB CL is desired because this will improve the reliability and usefulness of the CLs. In addition, a mechanistic understanding would aid the development of management strategies to mitigate the effects of N excess. Empirical data can be useful in field testing and evaluating the SMB CLs and parameters used to calculate the SMB CL.

Table 38.1 Values used to derive the Simple Mass Balance CL for nutrient N [Cl_{nut}(N)] at sites across an air pollution gradient in the San Bernardino Mountains in southern California based on methods of the Mapping and Modelling manual (ICP Modelling and Mapping Manual, 2008). SMB parameters given in the table are: $N_{le(acc)}$, Acceptable N leaching; N_i, N immobilization; N_u, N uptake; N_{de}, Denitrification. Exceedance (Ex) was estimated as the critical load minus deposition. For all parameters units are kg N ha^{-1} year^{-1}

Site	N Dep	Vegetation	$N_{le(acc)}$	N_i	N_u	N_{de}	CL_{nut}(N)	Ex
Strawberry Peak	39	Sierran mixed conifer	1.6	2.0	3.3	1.1	8.0	31.0
Heaps Peak	36	California black oak	1.6	1.4	1.3	1.3	5.6	30.4
Camp Angelus	13	Sierran mixed conifer	1.3	2.5	3.3	3.7	10.8	2.2
Green Valley	9	White fir	1.5	3.1	1.8	1.1	7.5	1.5
Barton Flats	9	White fir	1.1	3.0	1.8	3.7	9.6	−0.6
Converse Flats	6	Interior ponderosa pine	1.1	2.5	4.7	1.9	10.2	−4.2
Holcomb Valley	6	Interior ponderosa pine	1.2	3.4	4.7	3.7	13.0	−7.0
Devil Canyon 3	23	Canyon live oak	1.5	1.0	2.1	1.1	5.7	17.3
Devil Canyon 6	14	Hard chaparral	1.3	2.0	0.5	1.3	5.1	8.9

38.3.2 Critical Loads for Fichen Community Effects

Critical loads for epiphytic lichen community changes in California forests can generally be considered to be based on a eutrophication or nutrient N effect, although lichen responses may be a combined effect of eutrophication and changes in bark pH from ammonia (NH_3) deposition (Sutton et al. 2009). Critical loads for epiphytic lichen responses are not soil-mediated effects, but are based on direct atmospheric deposition to the lichen thallus. In this sense lichen-based CLs are computed from a less complex system, although the community or biodiversity responses can be complex. Lichens are the most sensitive documented terrestrial responders to N in California forests and scrublands, and harmful effects of N to lichen communities are more widespread than other effects of chronic N deposition (Fenn et al. 2010, 2011). The N CL values vary depending on the criterion chosen to make a cut-off:

- initial changes in chemistry and lichen community effects (3.1 kg ha^{-1} year^{-1}),
- shift from acidophyte to nitrophyte lichen dominance (5.2 kg ha^{-1} year^{-1}),
- extirpation of the acidophyte lichen community (10 kg ha^{-1} year^{-1}).

Recent work has demonstrated that the CL for epiphytic lichens is affected by precipitation levels. A simple yet robust model has been developed to determine lichen CLs from N deposition levels and precipitation (Geiser et al. 2010, 2014, Chap. 33: this volume).

Critical levels have been established for lichen community responses to ambient NH_3 air concentrations in some regions (Cape et al. 2009). The critical level of a pollutant gas is defined as 'the concentration in the atmosphere above which direct adverse effects on receptors, such as plants, ecosystems or materials, may occur according to present knowledge' (Cape et al. 2009; Posthumus 1988). Although, NH_3 is a major driver of N deposition (often the most important N compound) in

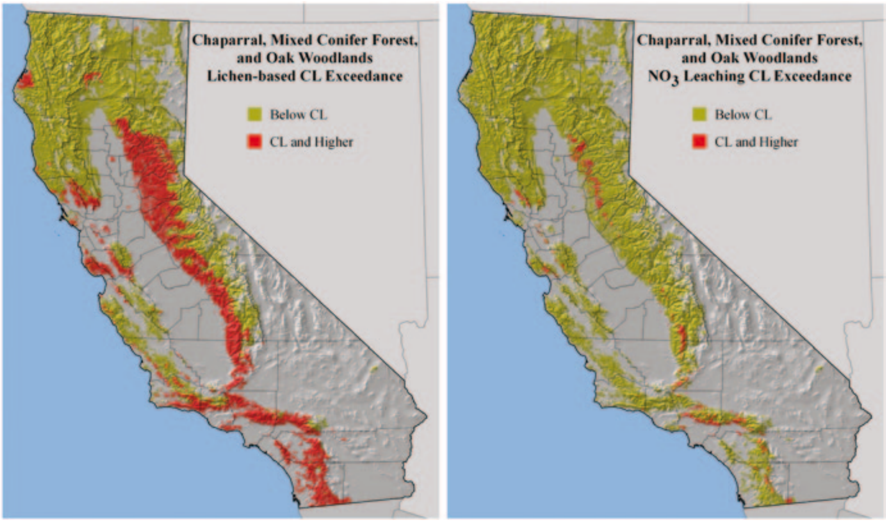

Fig. 38.1 (*Left*) Critical load exceedance map for lichen community changes. Forest CL = 5.2 kg ha^{-1} year^{-1}; chaparral and oak woodlands CL = 5.5 kg ha^{-1} year^{-1}. (*Right*) Critical load exceedance map for NO_3 leaching. Forest CL = 17 kg ha^{-1} year^{-1}; chaparral CL = 14 kg ha^{-1} year^{-1}

much of California, critical levels for NH_3 effects on lichen communities have not been established. Preliminary results in the Sierra Nevada suggest that critical levels for NH_3 effects on lichens may be similar to those reported in Europe (Andrzej Bytnerowicz and Sarah Jovan, unpublished data). Further work is needed however.

38.3.3 Eutrophication Critical Load Exceedance in California Forests and Scrublands

Statewide empirical CL exceedance maps (Fig. 38.1) indicate that 3.3 and 4.5 % of the chaparral and forested areas in California are in excess of the NO_3 leaching CL. By comparison, 23.4, 41.2 and 52.9 % of the mixed conifer forest, oak woodland and chaparral areas are in excess of the empirical N CL for epiphytic lichen community effects, respectively (Fig. 38.1). Ammonia concentration exposure profiles in California indicate that most if not all of the Sierra Nevada from Lake Tahoe southward is in exceedance of the critical level for NH_3 (Andrzej Bytnerowicz, unpublished data). Moreover, the proportional area shown in Fig. 38.1 to be in exceedance of the CL is a conservative estimate. The data used to derive the CL (Fenn et al. 2008) suggest that decreases in the occurrence of sensitive acidophyte species were initiated at N deposition levels below the lower-bound CL of 3.1 kg ha^{-1} year^{-1}, but more data points at low N deposition inputs are needed to better define this relationship. This becomes difficult with throughfall measurements however, because at low N deposition inputs (e.g., 2–4 kg ha^{-1} year^{-1}) forest canopies consume a large fraction of atmospherically-deposited N that is not washed off from tree canopies in precipitation and thus not measured as throughfall; this can result in negative net throughfall values. In these cases calculating N deposition with the inferential method can be an effective alternative approach (Fenn et al. 2009).

A fraction of the N consumed within the canopy is presumably due to N retention by epiphytic lichens. Negative net throughfall is by definition when deposition as bulk precipitation in forest clearings is greater than throughfall deposition measured under tree canopies. The critical level approach avoids this complication of canopy retention of N, because the response is based solely on the relationship between atmospheric concentrations of NH_3 and lichen community changes. However, in California the effects of co-occurring oxidant gases such as nitric acid vapor (HNO_3) and ozone must also be considered when determining the critical level for NH_3 (Riddell et al. 2008). In any case, NH_3 exposure profiles in the Sierra Nevada and unpublished estimates of the critical level for lichen community effects, and CLs for lichen effects (Fenn et al. 2010, 2011), suggest that approximately one-third of the land area of California covered by forests, chaparral and oak woodlands is in exceedance of the critical load for lichen communities.

Because of the capacity of terrestrial watersheds to retain and process significant levels of added N, the CL for NO_3 leaching is much higher than the CL for lichens. Although only an estimated 4.2 % of the combined forest and chaparral areas are projected to be in exceedance of the NO_3 leaching CL, the affected area represents approximately 570,000 ha, including watersheds vital for the water supply to millions of people. The NO_3 leaching CL for forested watersheds is based on a N deposition level of 17 kg ha^{-1} $year^{-1}$. At this deposition level, NO_3 concentrations in runoff from forested or chaparral catchments only begin to exceed background levels. Thus, the significance for areas in exceedance of the CL is that NO_3 levels will be higher than 'normal' for varying periods of the year and that the amount of excess NO_3 will vary.

In many chaparral and forested catchments surrounding Los Angeles and adjoining urban centers, NO_3 concentrations are elevated year-round, even during base flow conditions, indicating that groundwater NO_3 is also elevated. Ecological effects of N deposition occur at thresholds of at least an order of magnitude lower than for drinking water effects. However in highly-polluted catchments (e.g., N deposition of 25–70 kg ha^{-1} $year^{-1}$), elevated NO_3 concentrations can also reduce water quality, particularly when water from forests are relied upon as relatively pristine water sources, often blended with lower quality sources before delivery to domestic water supplies.

38.3.4 Acidification Critical Loads: Empirical vs. Modelled

Contrary to eutrophication CLs, the empirical N CL for soil acidification (26 kg ha^{-1} $year^{-1}$; see Breiner et al. 2007) was lower than the SMB N CL values (23–168 kg ha^{-1} $year^{-1}$), although it was within the lower end of the range of the

Table 38.2 Comparison of eutrophication (kg N ha^{-1} $year^{-1}$) and acidification (kg S ha^{-1} $year^{-1}$ or kg N ha^{-1} $year^{-1}$ [meq m^{-2} $year^{-1}$]) critical loads for California mixed conifer forests

	Eutrophication Critical Loads (kg N ha^{-1} $year^{-1}$)				Acidification Critical Loads		
	SMB Mapping	Daycent Model[1]	Empirical NO_3 Leaching[1]	Lichen Comm. Response[1]	SMB Mapping	SMB Site-Specific[2]	Empirical[3]
CL_N	5–13	17	17	3–10	23–168	BF: 32 (228) CF: 37 (266) CP: 46 (328)	26
CL_S					26–59	BF: 29 (183) CF: 35 (217) CP: 44 (273)	n.d.

[1] Daycent model and empirical NO_3 leaching CLs and the lichen CLs are from Fenn et al. 2008
[2] Critical loads for soil acidification $CL_{max}(N)$ and $CL_{max}(S)$ (maximum critical load of nitrogen and sulphur). $CL_{max}(S)$ and $CL_{max}(N)$ were calculated using site-specific mineralogy and soil weathering rates estimated with the PROFILE model to protect ponderosa pine (Bc:Al = 2.0). The three sites in the San Bernardino Mountains are: Barton Flats (BF), Converse Flats (CF), and Camp Paivika (CP)
[3] The empirical CL for soil acidification is from Breiner et al. 2007. The CL was determined by linear regression of throughfall N deposition vs. soil pH at sites across an N deposition gradient in the San Bernardino Mountains in southern California

SMB mapping CL values (Table 38.2). High calculated CLs for acidity are largely due to high soil weathering rates (89–145 meq m^{-2} $year^{-1}$). Historical soil pH data and spatial trends in soil pH across the N deposition gradient in the San Bernardino Mountains demonstrate that soil acidification is occurring in the most polluted sites (Breiner et al. 2007). Over an 18 year period from 1975 to 1993, soil pH decreased from 4.8 to 3.1 at Camp Paivika, a site in the western San Bernardino Mountains (Wood et al. 2007) where current average N deposition is 70 kg ha^{-1} $year^{-1}$ (Fenn et al. 2008). The range of SMB and empirical CLs for nutrient N effects (3–17 kg N ha^{-1} $year^{-1}$) are lower than those reported for acidification effects (23–168 kg N ha^{-1} $year^{-1}$), which highlights the far greater risk of eutrophication effects in California terrestrial ecosystems compared to soil acidification in this semiarid climate. Likewise, in Europe CL exceedance for nutrient N effects are more prevalent than for soil acidification effects (Hettelingh et al. 2008).

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