

Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States

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Abstract

Human activity in the last century has led to a significant increase in nitrogen (N) emissions and atmospheric deposition. This N deposition has reached a level that has caused or is likely to cause alterations to the structure and function of many ecosystems across the United States. One approach for quantifying the deposition of pollution that would be harmful to ecosystems is the determination of critical loads. A critical load is defined as the input of a pollutant below which no detrimental ecological effects occur over the long-term according to present knowledge.

The objectives of this project were to synthesize current research relating atmospheric N deposition to effects on terrestrial and freshwater ecosystems in the United States, and to estimate associated empirical N critical loads. The receptors considered included freshwater diatoms, mycorrhizal fungi, lichens, bryophytes, herbaceous plants, shrubs, and trees. Ecosystem impacts included: (1) biogeochemical responses and (2) individual species, population, and community responses. Biogeochemical responses included increased N mineralization and nitrification (and N availability for plant and microbial uptake), increased gaseous N losses (ammonia volatilization, nitric and nitrous oxide from nitrification and denitrification), and increased N leaching. Individual species, population, and community responses included increased tissue N, physiological and nutrient imbalances, increased growth, altered root:shoot ratios, increased susceptibility to secondary stresses, altered fire regime, shifts in competitive interactions and community composition, changes in species richness and other measures of biodiversity, and increases in invasive species.

The range of critical loads for nutrient N reported for U.S. ecoregions, inland surface waters, and freshwater wetlands is 1-39 kg N ha⁻¹ y⁻¹, spanning the range of N deposition observed over most of the country. The empirical critical loads for N tend to increase in the following sequence for

different life forms: diatoms, lichens and bryophytes, mycorrhizal fungi, herbaceous plants and shrubs, trees.

The critical load approach is an ecosystem assessment tool with great potential to simplify complex scientific information and communicate effectively with the policy community and the public. This synthesis represents the first comprehensive assessment of empirical critical loads of N for major ecoregions across the United States.

Key words: air pollution, atmospheric N deposition, community shifts, biodiversity, nitrate leaching, nitrogen saturation, plant nitrogen cycling, natural resource protection, vegetation type conversion

1 **1. Introduction**

2 **1.1 Effects of Nitrogen Deposition on Ecosystems**

3 Human activity in the last century has led to a significant increase in nitrogen (N) emissions
4 and deposition (Galloway et al. 2004). Because of past, and, in some regions, continuing
5 increases in emissions (Nilles and Conley 2001, Lehmann et al. 2005), N deposition has reached
6 a level that has caused or is likely to cause alterations in many ecosystems across the United
7 States. In some ecoregions, the impact of N deposition has been severe, altering N cycling and
8 biodiversity. Indicators of altered N cycling include increased N mineralization, nitrification, and
9 nitrate (NO₃⁻) leaching rates, as well as elevated plant tissue N concentration. The eventual
10 outcome of increases in these processes can be N saturation, the series of ecosystem changes that
11 occur as available N exceeds plant and microbial demand (Aber et al. 1989, 1998).

12 As N availability increases there are progressive changes in biotic community structure and
13 composition, including changes in diatom, lichen, mycorrhizal fungal, and terrestrial plant
14 communities. For example, in the Mediterranean California ecoregion native plant species in
15 some ecosystems have been replaced by invasive species more productive under elevated N
16 deposition (Weiss 1999, Yoshida and Allen 2004, Fenn et al. 2010, Rao and Allen 2010, Rao et
17 al. 2010). Such shifts in plant community composition and species richness can lead to overall
18 losses in biodiversity and further impair particular threatened or endangered species (Stevens et
19 al. 2004), as has occurred for the checkerspot butterfly (Weiss 1999).

20

21 **1.2 Critical Loads Definition and Previous Uses**

22 One method for evaluating potential impacts of air pollution on ecosystems is the critical
23 load approach. The critical load is defined as *the deposition of a pollutant below which no*
24 *detrimental ecological effect occurs over the long term according to present knowledge* (UBA

25 2004). The critical load is reported as a flux ($\text{kg ha}^{-1} \text{ yr}^{-1}$). Critical loads have been used broadly
26 in Europe (Posch et al. 1995; Posch et al. 2001) as a tool in the process of negotiating decreases
27 in air pollution. Critical loads have been more widely applied in Canada than in the United
28 States. In Canada, critical loads have been published for upland forests (Ouimet et al. 2006) and
29 lakes (Dupont et al. 2005) in eastern Canada and included in European assessments (Hettelingh
30 et al. 2008). In the United States (US), critical loads have been calculated for specific regions
31 such as the Northeast (NEC/ECP 2003, Dupont et al. 2005), California (Fenn et al. 2003a, b,
32 2008, 2010), Colorado (Williams and Tonnessen 2000, Baron 2006, Bowman et al. 2006), the
33 Pacific Northwest (Geiser et al. 2010) and, at a coarse scale, the conterminous United States
34 (McNulty et al. 2007). Critical loads have been determined most frequently in the United States
35 for effects of acidity (NEG/ECP 2003, Sullivan et al. 2005), but are also being increasingly used
36 in evaluating impacts of N deposition on ecosystems in terms of excess nutrient N availability,
37 also known as eutrophication (Fenn et al. 2008, 2010).

38 Despite relatively limited use in the US, the critical loads approach is being explored at state,
39 federal, and international levels as an ecosystem assessment tool with great potential to simplify
40 complex scientific information and communicate effectively with the policy community and the
41 public (Porter et al. 2005, Burns et al. 2008). The critical loads approach can provide a useful
42 lens through which to assess the results of current policies and programs and to evaluate the
43 potential ecosystem-protection value of proposed policy options. Critical loads are used by
44 policymakers to inform the process of setting emissions standards, for assessing emissions
45 control programs, and by natural resource managers as a tool to evaluate the potential impact of
46 new pollution sources (Burns et al. 2008, Environment Canada 2008, Lovett et al. 2009, Porter et
47 al. 2005, US EPA 2007, US EPA 2008). Policymakers and resource managers have used critical

48 loads to establish benchmarks for resource protection and to communicate the impacts of
49 deposition on natural resource conditions.

50 There are three main approaches for estimating critical loads (Pardo 2010): empirical, steady-
51 state mass balance (UBA 2004), and dynamic modeling (de Vries et al. 2010, Slootweg et al.
52 2007). Empirical critical loads are determined from observations of detrimental responses of an
53 ecosystem or ecosystem component to an observed N deposition input (Pardo 2010). This level
54 of N deposition is set as the critical load and extrapolated to other similar ecosystems. Empirical
55 critical loads for N are based on measurements from gradient studies, field experiments, or
56 observations (Bobbink et al. 1992, 2003, 2010). Steady-state mass balance modeling is based
57 on estimating the net loss or accumulation of N inputs and outputs over the long term under the
58 assumption that the ecosystem is at steady-state with respect to N inputs. Dynamic models also
59 use a mass balance approach, but consider time-dependent processes and require detailed data
60 sets for parameterization and testing (Belyazid et al. 2006, de Vries et al. 2007).

61 The advantage of the empirical approach is that it is based on measurable ecosystem
62 responses to N inputs; however, the method will overestimate the critical load (set it too high) if
63 the system has not reached steady state, i.e., if a similar response would occur at a lower
64 deposition level over a longer period. The advantage of steady-state mass balance approaches is
65 that they are less likely to overestimate the critical load. However, in the United States, the
66 uncertainty associated with steady-state mass balance approaches is high because data are not
67 available to quantify the terms in the mass balance equations accurately. Indeed, our empirical
68 critical loads synthesis may be useful in defining the acceptable critical thresholds for application
69 in steady-state mass balance critical loads calculations.

70 Dynamic models for critical loads of N in the United States have been applied on a limited
71 basis (Wu and Driscoll 2010). For dynamic modeling of nutrient N critical loads, empirical
72 critical loads and other response data are essential: Here, too, current data have not been
73 sufficient to develop, parameterize, and test dynamic models of ecosystem structure and function
74 (including changes in biodiversity). Thus, empirical critical loads currently provide a uniquely
75 valuable approach for assessing the risk of harm to ecosystems in the United States. This
76 synthesis is a first step towards identifying which data are available for key ecosystems and
77 where dynamic modeling could most profitably be applied in the United States after further data
78 collection.

79

80 **1.3 Objectives**

81 Our recent publication (Pardo et al. in press c) synthesized current research relating
82 atmospheric N deposition to effects on terrestrial and freshwater ecosystems in the United States
83 and quantified empirical critical loads for atmospheric N deposition, with one chapter devoted to
84 each of twelve major ecoregions. Our objectives for this paper were to synthesize empirical
85 critical loads for N reported for all the ecoregions of the United States, compare critical loads by
86 life form or ecosystem compartment across all ecoregions, discuss the abiotic and biotic factors
87 that affect the critical loads, and compare critical loads in the United States to those for similar
88 ecoregions/ecosystems in Europe. Finally, we discuss the significance of these findings, and
89 recommend priorities for future research.

90 **2. Approach**

91 For this synthesis, we reviewed studies of responses to N inputs (Pardo et al. in press c) for
92 U.S. ecoregions as defined by the Commission for Environmental Cooperation (CEC) Level I

93 ecoregions map for North America (CEC 1997; Figure 1). We identified the receptor of concern
94 (organism or ecosystem compartment), the response of concern, the critical threshold value for
95 that response, and the criteria for setting the critical load and extrapolating the critical load to
96 other sites or regions. These methods are described in detail in Pardo et al. (in press b) and the
97 Appendix.

98 The receptors evaluated included freshwater diatoms, mycorrhizal fungi, lichenized fungi
99 (henceforth lichens), bryophytes, herbaceous plants, shrubs, and trees. Ecosystem impacts
100 included: (1) biogeochemical responses and (2) individual species, population, and community
101 responses. Biogeochemical responses included increased N mineralization and nitrification (and
102 N availability for plant and microbial uptake), increased gaseous N losses (ammonia
103 volatilization, nitrous oxide from nitrification and denitrification), and increased N leaching.
104 Individual species, population and community responses included increased tissue N
105 concentration, physiological and nutrient imbalances, altered growth, altered root:shoot ratios,
106 increased susceptibility to secondary stresses, altered fire regimes, changes in species abundance,
107 shifts in competitive interactions and community composition (including shifts within and across
108 diatom, bacterial, fungal, or plant taxa groups), changes in species richness and other measures
109 of biodiversity, and increases in invasive species.

110 We considered N addition, N deposition gradient, and long-term monitoring studies in
111 order to evaluate ecosystem response to N deposition inputs. Most of these studies were not
112 designed to quantify critical loads, which presented some challenges. We afforded greater weight
113 to long-term fertilization studies (5-10 years) than to short-term studies. Single-dose forest
114 fertilization studies exceeding 50 kg N ha⁻¹ were generally not considered, although lower dose
115 short-term studies were considered when other observations were limited. When N-addition

116 studies were designed in order to determine critical loads, the studies generally included modest
117 N additions; multiple (3+) treatment levels with smaller increments between the treatment levels;
118 and treatments spanning the critical load. In such cases, estimates of the critical load are made
119 with greater certainty than for other types of N-addition studies. Nitrogen gradient studies
120 implicitly include longer-term exposure to pollutants and therefore are more likely than N
121 manipulation studies to depict conditions that are near steady-state with respect to ambient N
122 inputs. Long-term monitoring studies sometimes offer the opportunity to observe changes over
123 time in response to increasing or elevated N deposition inputs. We estimated critical loads based
124 on data from >3200 sites (Figure 2).

125 In general, we determined the critical load based on the observed response pattern to N
126 inputs. In some cases, there was a clear dose-response relationship where the response changed
127 above a certain threshold. A critical threshold is the value of a response parameter which
128 represents an unacceptable condition. The critical threshold is also referred to as the critical limit
129 (UBA 2004). In other cases, when response to increasing N was more linear, we estimated the
130 “pristine” state of N deposition and the deposition that corresponded to a departure from that
131 state. The criteria for setting critical loads are discussed in detail in Pardo et al. (in press b, c) and
132 in the Appendix.

133

134 **2.1 Deposition**

135 Total N emissions in the U.S. have increased significantly since the 1950s (Galloway 1998,
136 Galloway et al. 2003). As S deposition has declined in response to regulation, the rate of N
137 deposition relative to S deposition has increased since the 1980s (Driscoll et al. 2001, 2003)
138 followed by a general decrease in NO_x emissions from electric utilities since the early 2000s.

139 More recently, the relative proportion of NH_x ($\text{NH}_4^+ + \text{NH}_3$) to NO_x ($\text{NO} + \text{NO}_2$) emissions has
140 also increased for many areas of the United States (Kelly et al. 2005, Lehmann et al. 2005).
141 Nitrogen deposition at sites included in this analysis (Weathers and Lynch in press) was
142 quantified by the Community Multiscale Air Quality (CMAQ) model v.4.3 simulations of wet +
143 dry deposition of oxidized (NO_y) and reduced (NH_x) N species (Figure 2; hereafter CMAQ 2001
144 model; which uses 2001 reported data; Byun and Ching 1999, Byun and Schere 2006). These
145 CMAQ data were used to calculate exceedance. Exceedance of the critical load is defined as the
146 current deposition minus the critical load; when exceedance is greater than zero, the ecosystem is
147 susceptible to harmful ecological effects. Exceedance is useful in communicating the extent of
148 risk to ecosystems under current and future deposition scenarios.

149 We rarely had data to distinguish biotic or ecosystem response to reduced forms versus
150 oxidized forms of N. There is some evidence that for some species, reduced forms of N may
151 have more substantial impacts than oxidized forms (Bobbink et al. 2003; Kleijn et al. 2008, Cape
152 et al. 2009, Sutton et al. 2009). Differences in uptake rates and preference for NH_4^+ versus NO_3^-
153 across different plant taxa (Falkengren-Grerup 1995, McKane et al. 2002, Miller and Bowman
154 2002, Nordin et al. 2006) lead to differences in sensitivity to NH_x (Krupa 2003) and NO_y .
155 However, not all species are more sensitive to NH_x than NO_y (Jovan unpub. data); these
156 responses vary by species and functional type. Some species are more sensitive to increases in
157 NO_y , as was demonstrated for boreal forests (Nordin et al. 2006).

158 In order to quantify the critical load, we generally used the deposition reported in the
159 publication or, when that was not available, we used modeled deposition (e.g., CMAQ, ClimCalc
160 (Ollinger et al. 1993), or National Atmospheric Deposition Program (NADP; NADP 2009)). The
161 different forms of N deposition included in this assessment were: wet, bulk, wet+dry,

162 throughfall, and total inorganic N deposition (wet+dry+cloud/fog). Total N deposition was
163 considered the most appropriate value to use in evaluating ecosystem responses; however, in
164 many studies this information is not available. Throughfall N is generally considered a good
165 surrogate for total N deposition (Weathers et al. 2001), because it typically does not
166 underestimate total N inputs as much as wet or bulk deposition. However, because of the
167 potential for canopy uptake of N, throughfall is usually considered as a lower-bound estimate of
168 total N deposition. None of the studies included reported inputs of organic N, so this report
169 focuses on responses to inputs of inorganic N.

170 The accuracy of the atmospheric N deposition estimates clearly influences the accuracy of
171 the critical load and exceedance estimates. Several factors contribute to uncertainty in N
172 deposition estimates: (1) the difficulty of quantifying dry deposition of nitrogenous gases and
173 particles to complex surfaces; and (2) sparse data, particularly for arid, highly heterogeneous
174 terrain (e.g., mountains), and (3) sites with high snowfall or high cloudwater/fog deposition,
175 where N deposition tends to be underestimated. Deposition models cannot account for these
176 kinds of heterogeneity (e.g., Weathers et al. 2006) because the spatial scale (grid size) is
177 typically too coarse to capture topographic and other local influences. These issues are discussed
178 in detail elsewhere (Weathers et al. 2006, Fenn et al. 2009, Weathers and Lynch in press). When
179 more accurate and precise N deposition estimates become available, the data presented in this
180 study may be re-evaluated in order to refine the critical loads estimates.

181

182 **3. Results and Discussion**

183 The range of critical loads for nutrient N reported for the United States ecoregions, inland
184 surface waters, and freshwater wetlands is 1-39 kg N ha⁻¹ y⁻¹ (Table 1). This broad range spans

185 the range of N deposition observed over most of the country (see Weathers and Lynch in press).
186 For coastal wetlands, critical loads are between 50-400 kg N ha⁻¹ y⁻¹. The number of locations for
187 which ecosystem response data were available (Figure 2) for an ecoregion is variable, which
188 impacts the level of certainty of the empirical critical loads estimates. Details on the studies upon
189 which the critical loads values (Tables 1, 2) are based are provided in Pardo et al. (in press c).

190

191 **3.1 Comparison of Critical Load by Receptor across Ecoregions**

192 Because N deposition varies considerably by region and the critical load varies both by
193 region and receptor, we present the critical loads and likely risk of exceedance by receptor.

194 **3.1.1 Mycorrhizal fungi**

195 *Background*

196 Mycorrhizal fungi reside at the interface between host plants and soils, exchanging soil
197 resources, especially nutrients, with host plants in exchange for photosynthates (carbon
198 compound). Due to this important and unique ecological niche, mycorrhizal fungi are at
199 particular risk due to changes in either the soil environment or host carbon allocation.

200 *Response to N*

201 Nitrogen deposition adversely affects mycorrhizal fungi (1) by causing decreased
202 belowground C allocation by hosts and increased N uptake and associated metabolic costs
203 (Wallander 1995) and (2) via soil chemical changes associated with eutrophication and
204 acidification. There are two major groups of mycorrhizal fungi that are evolutionarily and
205 ecologically distinct: arbuscular mycorrhizal fungi (AMF) and ectomycorrhizal fungi (EMF).
206 Under sufficiently high N inputs, the progressive effect of elevated N is an early decline of
207 sporocarp (reproductive structure) production for EMF and spore production for AMF, and

208 subsequent decline in biological diversity and loss of taxa adapted to N-poor environments or
209 sensitive to acidification (Lilleskov 2005). Sporocarp and spore production appears to be
210 especially sensitive to N deposition, often declining before the communities on root tips have
211 been substantially altered, presumably because sporocarps and spores are at the end of the carbon
212 flux pathway from hosts.

213 Of the two plant-fungal symbioses examined here, mycorrhizal fungi appear to be less
214 sensitive to N deposition than lichens (Tables 3 and 4), presumably because the soil environment
215 buffers these soil fungi from some of the immediate impacts of N deposition, to which lichens
216 are directly exposed. Lichens have an advantage as indicators when compared with mycorrhizal
217 fungi because they can be relatively easily inventoried. However, the critical role of mycorrhizal
218 fungi as root symbionts central to plant nutrition and belowground production, and, in forests, as
219 repositories of a large part of the eukaryote diversity, as major components of food webs, and as
220 non-timber forest products of high economic value (edible sporocarps or mushrooms)
221 (Amaranthus 1998) provides sufficient impetus to improve our understanding of their response to
222 N deposition.

223 *Critical loads*

224 We reviewed empirical studies on mycorrhizal fungal response to N inputs as the basis
225 for determining empirical critical loads for the United States (Table 3, Figure 3.a). Despite the
226 sparse data, it is clear that N deposition sufficient to elevate inorganic N, especially NO_3^- ,
227 availability in soils can have measurable effects on mycorrhizal fungi. The data for EMF indicate
228 that N deposition to N-limited conifer forests in the range of 5-10 $\text{kg ha}^{-1} \text{ yr}^{-1}$ can significantly
229 alter community structure and composition and decrease species richness (Dighton et al. 2004,
230 Lilleskov 1999, Lilleskov et al. 2001, 2002, 2008). Similarly, the data for AMF suggest N

231 deposition levels of 7.8-12 kg ha⁻¹ yr⁻¹ can lead to community changes, declines in spore
232 abundance and root colonization, and changes in community function, based on re-analysis of
233 data from Egerton-Warburton et al. (2001) combined with N deposition data, and decreases in
234 fungal abundance (van Diepen et al. 2007, van Diepen 2008) and declines in fungal activity
235 (Egerton-Warburton, unpublished data). The actual threshold for N effects on AMF could be
236 even lower, because high background deposition precludes consideration of sites receiving
237 deposition at or near pre-industrial levels. Therefore, our provisional expert judgment is that
238 critical loads for mycorrhizal diversity for sensitive ecosystem types are 5-10 kg ha⁻¹ yr⁻¹. The
239 uncertainty of this estimate is high, because few studies have been conducted at low N deposition
240 to further refine the critical load. The critical load of N for mycorrhizal fungi, when community
241 change occurs, is often on the order of current N deposition and thus is exceeded across most of
242 the eastern and northern forests and in regions downwind of agricultural and urban emissions in
243 the West (Figure 3.b). The uncertainty associated with the exceedance, like that for the critical
244 load, is high.

245 **3.1.2 Lichens and Bryophytes**

246 *Background*

247 Lichens and bryophytes make substantial contributions to biodiversity. About 4,100
248 lichens and 2,300 bryophytes are known from North America north of Mexico—approximately
249 one fourth of the value for vascular plant diversity, about 26,600 species (USDA, NRCS 2009).

250 *Responses to N*

251 Lichens and bryophytes are among the most sensitive bioindicators of N in terrestrial
252 ecosystems (Blett et al. 2003, Bobbink et al. 2003, Fenn et al. 2003a, 2010, Glavich and Geiser
253 2008). Unlike vascular plants, lichens and bryophytes lack specialized tissues to mediate the

254 entry or loss of water and gases (e.g., waxy epidermis, guard cells, root steele). Thus, they
255 rapidly hydrate and absorb gases, water, and dissolved nutrients during high humidity or
256 precipitation events. However, they dehydrate to a metabolically inactive state quickly as well,
257 making them slow growing and vulnerable to contaminant accumulation. Consequently, the
258 implementation of lichen or bryophyte-derived critical loads may prevent undesired impacts to
259 much of the broader forest ecosystem including biological diversity (McCune et al. 2007).

260 Lichens and bryophytes can play important roles in ecosystems. Species of epiphytic
261 lichens in wet and mesic forests that are most sensitive to N (i.e., the large pendant and foliose
262 species) play important ecological roles that are not duplicated by the nitrophytic (i.e., N
263 tolerant) species that may replace them. Dominant regional oligotrophs (e.g. *Alectoria*, *Bryoria*,
264 *Lobaria*, *Ramalina*, *Usnea*) comprise the bulk of lichen biomass in old-growth forests,
265 contribute to nutrient cycling through N₂ fixation, and are used for nesting material, essential
266 winter forage for rodents and ungulates, and invertebrate habitat (McCune and Geiser, 2009).
267 Storage of water and atmospheric nutrients by these lichen genera and epiphytic bryophytes
268 moderates humidity and provides a slow release system of essential plant nutrients to the soil
269 (Boonpragob et al. 1989, Knops et al. 1991, Pypker 2004, Cornelissen et al. 2007). In the tundra,
270 lichens and bryophytes represent a significant portion of the biomass, and reindeer lichens are a
271 vital link in the short arctic food chain (Kytöviita and Crittenden 2007). Mosses comprise the
272 bulk of the biomass of the extensive boreal peatlands. In the desert, together with other
273 microbiota, lichens and bryophytes form cryptogamic mats important to soil stabilization and
274 fertility.

275 *Critical loads*

276 The critical loads estimated (Pardo et al. in press c) for lichens range from 1-9 kg N ha⁻¹
277 yr⁻¹ (Table 4, Figure 4.a). Although the reported range of critical loads is not as large as the
278 ranges for forests or herbs, the certainty associated with these estimates for lichens varies
279 considerably by ecoregion. This is partially because of differences in sampling scheme and
280 intensity. For example in the Pacific Northwest, lichen communities were assessed intensively
281 across wide environmental gradients spanning low to high N deposition on a fine grid over time,
282 yielding highly reliable critical N load estimates (Geiser and Neitlich 2007, Jovan 2008),
283 whereas assessments in the eastern United States are more problematic due to historical and
284 contemporary S and N deposition. In such cases, where historical information necessary to
285 identify a “pristine” or “clean” state is lacking, it is more difficult to determine the critical load,
286 and the resulting confidence associated with the critical load is low. The critical load of N for
287 lichens, based on the shift in community composition favoring eutrophs over oligotrophs, is on
288 the order of current N deposition and thus is exceeded across most of the Eastern and Northern
289 forest ecoregions and in many areas downwind of agricultural and urban emissions or at high
290 elevation in the West (Figure 4.b). The uncertainty associated with the exceedance, like that for
291 the critical load, is low for the Marine West Coast and Northwest Forested Mountains
292 ecoregions, but high elsewhere.

293 Studies in the Pacific Northwest demonstrate that increasing precipitation allows lichens
294 to tolerate higher N deposition (Geiser and Neitlich 2007, Jovan 2008, Geiser et al. 2010),
295 probably because the concentrations of N compounds to which lichens are exposed are more
296 important than total loading. If such simple models could be tested and confirmed in other
297 regions of the country, the confidence in the critical loads in those regions would improve.

298 **3.1.3 Herbaceous Species and Shrubs**

299 *Background*

300 Herbaceous species and shrubs (Table 5, Figure 5) are found in grasslands, shrublands,
301 forests, deserts, and wetlands and comprise the majority of the roughly the 26,600 vascular plant
302 species found in North America north of Mexico (USDA, NRCS 2009).

303 *Response to N*

304 Herbaceous species and some shrubs appear intermediate between cryptogram and tree
305 species in their sensitivity to N deposition, due to specialized tissues that mediate the entry or
306 loss of water and gases compared with cryptograms, and rapid growth rates, shallow rooting
307 systems, and often shorter lifespan compared with trees. Thus, herbaceous species in a forest
308 understory will likely respond more rapidly to changes in N deposition and to a greater degree
309 than the trees with which they coexist. Herbaceous species in alpine or tundra environments will
310 respond later and to a lesser degree than the cryptograms with which they coexist. Herbaceous
311 plants clearly play an important role in those ecosystems in which they are the dominant primary
312 producers (e.g., grasslands, shrublands). In forests, however, the role of the herbaceous
313 community in ecosystem function has a significance that is disproportionate to its low relative
314 biomass. For example, although they represent only ~0.2 percent of standing above-ground
315 biomass, herbaceous understory species produce >15 percent of forest litter biomass and
316 comprise up to 90 percent of forest plant biodiversity, including endangered or threatened
317 species (Gilliam 2007).

318 *Critical loads*

319 The range of critical loads for N for herbaceous species and shrubs across all ecoregions
320 is 3-33 kg N ha⁻¹ yr⁻¹ (Table 5, Figure 5). Although this range is broader than those for lichens or

321 mycorrhizal fungi, many of the critical loads for herbaceous species fall into the range of 5-15 kg
322 N ha⁻¹ yr⁻¹. The uncertainty of these estimates is moderate. The shorter lifespan of some
323 herbaceous species can result in a more rapid response to N addition. This is especially relevant
324 for perennials with little N storage or annuals. In grasslands, for example, elevated N deposition
325 often leads to a rapid (1-10 years) increase in herbaceous production and a shift in biomass
326 allocation towards more aboveground tissue. This often decreases light levels at ground surface
327 and decreases the numbers of plant species, primarily of perennials, legumes, and natives (Clark
328 and Tilman 2008, Suding et al. 2004, Tilman 1993).

329 As a result of this relatively rapid response, experimental studies of moderate to long
330 duration (3-10 years) allow determination of the critical load with reasonable certainty. Longer
331 studies (>10 years) would decrease the uncertainty further. In some cases, it can be difficult to
332 determine whether the condition in reference plots or at the low end of a deposition gradient
333 represents a “pristine” condition or whether a site has already been altered by N deposition prior
334 to or at the time of the study. For example, the Watershed Acidification Study at Fernow
335 Experimental Forest, West Virginia added 35 kg N ha⁻¹ yr⁻¹ via aerial application in addition to
336 ambient deposition of 15-20 kg N ha⁻¹ yr⁻¹, which has led to changes in understory species
337 composition (Adams et al. 2006). Recently, similar changes in understory species composition
338 have occurred on the adjacent reference watershed receiving only ambient atmospheric
339 deposition (Gilliam unpublished data, Gilliam et al. 1996) suggesting that the deposition to the
340 reference watershed currently exceeds the critical load. Where deposition rates exceed the
341 critical load, empirical measurement of the rate of change of an ecological metric (e.g. plant
342 abundance, diversity, or community composition) over a range of N inputs provides an approach

343 to estimate the N level at which that metric begins to change further (Bowman et al. 2006), but it
344 is difficult to determine the critical load.

345 The critical load of N for herbaceous species and herbs, when community change occurs
346 (in some cases with invasives replacing native species), is exceeded across much of the Great
347 Plains and in portions of the Southwest and high elevation and high deposition areas of the other
348 ecoregions (Figure 5.b). The uncertainty associated with the exceedance, like that for the critical
349 load, varies.

350 **3.1.4 Trees/Forest Ecosystems**

351 *Background*

352 In this section we discuss the responses of trees and the overall biogeochemical responses
353 of forest ecosystems to N inputs (Table 6), excluding the specific responses of mycorrhizal fungi,
354 lichens, or understory herbaceous plants. Forest ecosystems represent about a third of landcover
355 in the United States (FIA 2001) and are significant in Northern, Eastern, Tropical Wet, and
356 Marine West Coast Forests, Northwestern Forest Mountains, and Mediterranean California
357 ecoregions.

358 *Response to N*

359 In northeastern forests, gradient studies demonstrate that N deposition enhances growth
360 in some fast-growing tree species, including many hardwoods with AMF associations, whereas it
361 slows growth in some EMF species (red spruce, red pine), and has no detectable effect on still
362 other species (Thomas et al. 2010). Similarly, N deposition enhances survivorship in a few
363 species capable of forming AMF associations (black cherry, red maple, paper birch) and
364 decreases survivorship in others, all ectomycorrhizal (Thomas et al. 2010). Survivorship under
365 chronic N deposition, and possibly other co-occurring pollutants such as ozone, is often

366 dependent on interactions with other stressors such as pests, pathogens, climate change, or
367 drought (Grulke et al. 2009, McNulty and Boggs 2010). Over the long-term, these differential
368 effects of N deposition on tree growth and survivorship are likely to shift species composition,
369 possibly to more nitrophilic species, similar to patterns seen for organisms with shorter lifespans.

370 We have few data that show a major structural or functional shift in forest ecosystems
371 because of the long response time of trees and forest soils to changes in N inputs and N
372 availability (Table 6). The relatively large pools of organic N in the forest floor, mineral soil, tree
373 biomass, and detritus contribute to the relatively long lag-time in forest ecosystem response to N
374 inputs. Because of the long lag-time in response to N treatments, it can be difficult to determine
375 the actual critical N load for forest ecosystems based on short-term fertilization studies. If a
376 response is observed over a relatively short period of time (i.e. years), it is nearly certain that the
377 critical load is below the total N input at the treatment site and it can be difficult to further
378 constrain the critical load. It is expected that the more complex and interconnected processes in
379 forests will result in a higher critical load than other ecosystem types, in part because large N
380 storage pools give forest ecosystems a greater capacity to buffer N inputs.

381 *Critical loads*

382 The range of critical loads reported for forest ecosystems is 4-39 kg N ha⁻¹ yr⁻¹ (Table 6,
383 Figure 6.a). The threshold N deposition value which caused increased NO₃⁻ leaching from forest
384 ecosystems into surface water was 8-17 kg N ha⁻¹ yr⁻¹; the lower end of the range representing
385 Northern and Eastern Forests, the upper end representing Mediterranean California mixed
386 conifers (Table 6, Figure 7.a). At 4 kg N ha⁻¹ yr⁻¹ in the Colorado Rockies, increasing [NO₃⁻] was
387 reported in the organic horizon, which suggests incipient N saturation (Rueth and Baron 2002).
388 The highest critical loads were reported for Mediterranean California mixed conifer forests for

389 forest sustainability and for soil acidification caused by increased N deposition. These sites
390 experience some of the highest N deposition reported in the United States, up to approximately
391 70 kg N ha⁻¹ yr⁻¹ (Fenn et al. 2008).

392 The critical load is exceeded across much of the East. The lower end of the critical load
393 range is exceeded for the remaining portions of the eastern forests, as well as portions of the
394 Marine West Coast, Northwest Forested Mountains, and Tropical and Sub-tropical Humid Forest
395 Ecoregions (Figure 6.b).

396

397 **3.1.5 Freshwater and wetland ecosystems**

398 *Background*

399 Freshwater lakes and streams, and wetlands (freshwater and estuarine intertidal) are
400 ecosystem types that occur in most ecoregions in North America. In freshwater lakes and
401 streams, phytoplankton, or algae that live in the water column, are sensitive to the chemical
402 environment in which they reside, and many species can be used as indicators of the levels of
403 nutrients or acidity because of individual species' preference for specific chemical conditions.
404 Diatoms are used in this discussion because there has been more work published on these algae
405 than others, but other types of algae also respond to N deposition (Lafrancois et al. 2004, Michel
406 et al. 2006). Of the wetlands which occur in the conterminous United States, 95 percent are
407 freshwater and 5 percent are estuarine or marine (USDI FWS 2005). The species composition
408 differs between freshwater and intertidal wetlands, although together they support more than
409 4200 native plant species. Despite the high biodiversity, the effects of N loading are studied in
410 just a few plant species.

411 *Response to N*

412 For the analysis of nutrient N effects to freshwater lakes and streams, we relied on papers
413 and studies that linked aquatic biological and ecological response to atmospheric deposition, but
414 the results are consistent with laboratory or *in situ* dose-response studies and even land use
415 change studies. The productivity of minimally disturbed aquatic ecosystems is often limited by
416 the availability of N, and slight increases in available N trigger a rapid biological response that
417 increases productivity and rearranges algal species assemblages (Nydick et al. 2004, Saros et al.
418 2005). The mechanism for change is alteration of N:P ratios, which can increase productivity of
419 some species at the expense of others (Elser et al. 2009). As with the terrestrial systems
420 described above, the nutrient responses of lakes and streams are most evident where land use
421 change and acidic deposition have been limited, thus most evidence of exceedance of critical
422 loads comes from high elevations of the western United States (Baron et al. in press). As with
423 terrestrial plants, some diatoms respond rapidly to an increase in available N. An example that
424 has been observed from a number of different lakes of the Rocky Mountains is dominance of two
425 diatoms (*Asterionella formosa* and *Fragilaria crotonensis*) in lakes with higher N, in contrast to
426 the flora of lakes with lower N deposition where there is a more even distribution, thus high
427 biodiversity, of diatoms. Higher trophic levels (zooplankton, macroinvertebrates) may be
428 secondarily affected by N, but further increases in primary, or autotrophic, production will be
429 limited by other nutrients such as P or silica (Si).

430 Both freshwater and estuarine intertidal wetlands tend to be N-limited ecosystems
431 (LeBauer and Tresseder 2008, U.S. EPA 1993). Known responses to N enrichment are generally
432 derived from nutrient-addition studies in the field and observations along gradients of N
433 deposition. A variety of ecological endpoints are evaluated, such as altered soil biogeochemistry,
434 increased peat accumulation, elevated primary production, changes in plant morphology,

435 changes in plant population dynamics, and altered plant species composition (U.S. EPA 2008).
436 In general, the sensitivity of wetland ecosystems to nitrogen is related to the fraction of rainfall
437 (a proxy for atmospheric N deposition) in its total water budget. Most freshwater wetlands, such
438 as bogs, fens, marshes and swamps, have relatively closed water and N cycles, thus are more
439 sensitive to N deposition than intertidal wetlands, such as salt marshes and eelgrass beds
440 (Greaver et al. in press).

441 *Critical loads*

442 In general, critical loads for freshwater lakes and streams tend to be low, because the target
443 organisms are unicellular algae that respond rapidly to changes in their chemical environment.
444 The range of critical loads for eutrophication and acidity is 2-9 kg N ha⁻¹ y⁻¹ (Baron et al. in
445 press); the range reported for terrestrial ecosystems is much broader (Table 1). Critical loads for
446 NO₃⁻ leaching from terrestrial ecosystems ranged from 4-17 kg N ha⁻¹ y⁻¹ (Figure 7a), but many
447 sensitive freshwaters at high altitudes are found above the tree-line where few watershed
448 buffering mechanisms exist due to sparse vegetation, poorly developed soils, short hydraulic
449 residence time, and steep topography. These factors influence how rapidly a system exhibits
450 elevated N leaching in response to increased N deposition, and how this increased N availability
451 subsequently influences biota. In general, lakes have relatively rapid N turnover times compared
452 to soil N pools and are at least seasonally well-mixed. They would, thus, be expected to have
453 lower critical loads.. Thus responses by terrestrial plants would not be expected to be as rapid as
454 those of freshwater organisms. The critical load for NO₃⁻ leaching is exceeded in portions of the
455 Mediterranean California and the lower end of the critical load range is exceeded for most of the
456 Eastern Forest and part of the Great Plains ecoregions (Figure 7.b).

457 Generally, freshwater wetlands are more sensitive to N deposition than estuarine wetlands,
458 with critical loads for freshwater wetlands that range from 2.7-14 kg N ha⁻¹ y⁻¹ (Greaver et al. in
459 press). The bryophyte genus *Sphagnum* and the carnivorous pitcher plant are the two taxa most
460 commonly studied. The critical loads reported for freshwater wetlands (Greaver et al. in press)
461 fall between those reported for inland surface waters (Baron et al. in press) and those reported for
462 terrestrial ecosystems (Pardo et al. in press a). This pattern may be related to the rate of N
463 released by soils/sediment to the ecosystem. The critical load tends to be higher for intertidal
464 wetlands than other types of ecosystems because they have open nutrient cycles which are often
465 strongly affected by N loading sources other than atmospheric deposition. Based on field
466 observations of N loading to plant growth and species composition on salt marsh and eel grass
467 habitat, the critical load ranges between 50-400 kg N ha⁻¹ y⁻¹.

468 **3.2 Relative sensitivities of different receptors, ecosystem types, and regions**

469 This synthesis demonstrates that empirical critical loads for N differ among lifeforms,
470 tending to increase in the following sequence: diatoms < lichens and bryophytes < mycorrhizal
471 fungi < herbaceous plants and shrubs < trees. This variation likely reflects a variety of factors,
472 including generation time and buffering against N impacts. That is, N deposition more rapidly
473 affects those species that experience the most direct exposure to elevated N levels in the
474 atmosphere (lichens and bryophytes) or receiving waters (diatoms), especially for those
475 organisms that lack protective structures, such as a cuticle, for example. By contrast, the
476 capacity of soil organic matter to accumulate large quantities of N may delay adverse impacts on
477 many herbs, shrubs, and trees. The effects of altered N availability in shifting species
478 composition often appears to occur most rapidly within those communities dominated by species
479 with short lifespans (diatoms) compared to those with long lifespans (trees).

480 Critical loads vary more by receptor and response type than by region. For the same
481 response of a given receptor, the western U.S. has generally similar critical load values to the
482 eastern U.S., with the apparent exception that the critical load for NO_3^- leaching is approximately
483 twice as high in Mediterranean California mixed conifers compared to northeastern forests
484 (Figure 7). In contrast, the critical load for NO_3^- leaching in high elevation catchments in the
485 Colorado Front Range are lowest in the United States, likely attributable to low biological N
486 retention and storage capacity in these steep, rocky catchments (Baron et al. 2000, Williams and
487 Tonnessen 2000, Sickman et al. 2002, Fenn et al. 2003a, b).

488 In setting critical loads, ideally one would identify an indicator that would allow prediction
489 of future deterioration in ecosystem structure or function before it occurs—an early indicator of
490 ecosystem change. We are not yet able to definitively determine which early responses to N
491 deposition are the best indicators of ecological harm, the central criterion for setting a critical
492 load. In some cases, alteration of community composition for a given taxa group (e.g., lichens),
493 may signal the beginning of a cascade of changes in ecosystem N cycling, which may
494 dramatically alter the structure or function of the ecosystem as a whole. In many cases, changes
495 in a single taxa group may have implications beyond that taxa group. In other cases, alterations
496 within the community of a given taxa group may have little impact on the overall structure and
497 function of the ecosystem. It can be difficult to know, at the outset, whether the ultimate
498 consequences of changes indicated by alterations to a given taxa group will be large or small for
499 the overall ecosystem over the long term.

500 However, understanding of the progressive series of changes that occurs during N saturation
501 should inform this process, along with recognition of the role of N in increasing vulnerability to
502 other stressors such as insects, drought, freezing, and other pollutants. For example, elevated N

503 inputs may lead to plant nutrient imbalances, which then increase plant susceptibility to stressors
504 such as cold, drought, or pests (Bobbink et al. 1998, Schaberg et al. 2002). These responses have
505 been observed in a southern Vermont montane red spruce stand, where low-level N additions led
506 to increased foliar N concentration, decreased foliar membrane-associated calcium and cold
507 tolerance and increased winter injury (Schaberg et al. 2002). Another key indicator is increased
508 soil NO₃⁻ leaching, especially during episodic acidification of surface waters, harming fish
509 species (Baker et al. 1996). Thus changes in foliar nutrient status or increased nitrification rates
510 are prime candidates for early biogeochemical thresholds that can be very useful for setting
511 critical loads.

512

513 **3.3 Factors that Affect the Critical Load**

514 One of the objectives of this assessment was to lay the groundwork for further refining and
515 improving estimates of critical loads. Multiple abiotic and biotic factors affect where the value of
516 the critical load falls within the reported range (Table 2). Abiotic influences include a range of
517 climatic, hydrologic, and soil factors that can affect the timing and magnitude of N delivery to
518 sensitive receptors. Climatic factors include temperature, precipitation amount and distribution,
519 and the extent and rate of climate change. For example, increased precipitation increases the
520 critical load for lichens (Geiser et al. 2010, Appendix). Hydrologic factors include catchment
521 size, topographic relief, and flowpath , which affect the processing and delivery of NO₃⁻ to
522 surface waters. Soil factors include soil type, age, depth, coverage, and parent material, all of
523 which can influence soil capacity to store or remove N, and increase a site's critical load.
524 Disturbance may also play a substantial role, for example, N removal by fire or forest cutting

525 may increase the critical load for nutrient N. Past agricultural land uses may either increase or
526 decrease the critical load depending on their impact on soil nutrients and biotic communities.

527 Biological factors likely to contribute to lower N critical loads include particularly sensitive
528 species (diatoms, lichens, mycorrhizal fungi, certain plants), single species versus community
529 responses, low biomass and low productivity ecosystems, short lifespan of receptor of concern,
530 presence of invasive species, and presence of ozone-sensitive species (Fenn et al. 2008, Grulke
531 and Balduman 1999, Grulke et al. 1998, 2009). For example, low-biomass ecosystems (e.g.,
532 grasslands, coastal sage scrub, desert) are more sensitive to N-enhanced growth of invasive
533 species, if invasive pressure occurs. These low-biomass ecosystem types sometimes occur
534 because of warm and dry climatic conditions. Because warmer temperatures often correspond to
535 greater metabolic rates, longer periods of biological activity, greater biomass, and more rapid N
536 cycling, one might expect that the critical load would increase with increasing temperature as has
537 been suggested in Europe (Bobbink et al. 2003). We do not observe such a pattern across U.S.
538 ecoregions in the critical loads reported in this synthesis, but Europe does not have warm and dry
539 deserts with low critical loads as does the U.S. Note, however, that the uncertainty of the critical
540 load estimates varies and is often fairly high, which may make it difficult to discern patterns in
541 critical load values across regions. Moreover, a temperature pattern may be confounded by
542 gradients in deposition form and quantity, moisture and elevation.

543 The factors discussed above provide general guidance in applying critical loads. In order to
544 set a critical load for a given site, the first step would be to determine whether the site of concern
545 is similar to the site/or sites on which the critical load for that ecosystem type is based. Details on
546 the estimation of critical loads are described by ecoregions in Pardo et al. (in press c). If the site
547 differs from the sites upon which the critical load is based, Table 2 lists ecoregion-specific

548 factors affecting the critical load that can be useful in adjusting the estimated critical load for a
549 given site.

550 Note that the magnitude or type of ecosystem change that is unacceptable may vary
551 according to resource management goals or ecosystem services that are valued by a particular
552 stake holder. In a conservation area, for example, any alteration in N cycling may be considered
553 unacceptable, whereas for other land areas, changes of a certain magnitude or scope may be
554 considered acceptable or desirable based on resource use (such as timber harvesting) or other
555 factors. For example, some responses to low levels of elevated N deposition, such as increased
556 plant growth and increased C-sequestration by trees (Thomas et al. 2010) may be considered
557 beneficial where forests are managed for tree growth.

558 The more we are able to identify and quantify the factors that affect the critical load, the
559 more we move towards a mechanistic understanding of the responses, and the better we are able
560 to extrapolate observations across and within ecoregions. In some cases, it may be possible to
561 develop simple empirical relationships as a function of one or several variables that allow us to
562 refine our critical loads estimates. For example, for lichens, Geiser et al. (2010) developed
563 simple regression relationships which included precipitation that explain much of the variability
564 in lichen community composition in response to N deposition, because decreasing precipitation
565 corresponds to exposure to higher concentrations of N. These regression models can be used to
566 estimate critical loads in other regions and also can provide an estimate of the uncertainty
567 associated with the critical load. Such models, strongly tied to empirical observations, will prove
568 invaluable in the development of dynamic models for nutrient N critical loads.

569 **3.4 Uncertainty in Critical Loads Estimates**

570 There are several sources of uncertainty in our assessment of empirical critical loads beyond

571 those associated with atmospheric deposition (see section 2.1). These include data gaps, time
572 lags, and effects of multiple stressors.

573 **3.4.1 Data Gaps**

574 In general, there is a dearth of observations on ecosystem response to N inputs near the
575 critical load. Without extensive, spatially stratified observations, it is not possible to know
576 whether a study site is more or less sensitive than other sites in the ecoregion. The threshold
577 value is best defined by a large number of studies which demonstrate the range of responses
578 observed.

579 **3.4.2 Time lags in the response to N deposition**

580 Time lags are often observed in N addition studies, with the magnitude of the time lag a
581 function of the N residence time and the organisms considered. This time lag increases with
582 lifespan and size of organism; a tree will respond more slowly than an herbaceous annual, for
583 example. Time lags are also a function of the rate of N input and the system's capacity for N
584 storage, with lower rates of input typically leading to longer time lags before an initial response
585 (Clark and Tilman 2008).

586 **3.4.3 Effects of multiple stressors and other confounding variables**

587 Uncertainty also arises from the confounding effects of multiple stressors and other
588 covariates, especially in N gradient studies, where it can be difficult to sort out the impact of
589 other factors that vary along the gradient, such as climate, weather, soils, vegetation,
590 disturbances, land-use history, and exposure to other pollutants. Multivariate statistics or other
591 approaches can sometimes tease apart effects of N from other factors, but sometimes correlated
592 stressors can be difficult to separate. In these cases impacts attributed to N could actually be the
593 result of a correlated variable or the interaction between the two. On the other hand, because

594 these represent “real-world” conditions—in most locations, multiple stressors co-occur—the
595 critical loads estimated in the presence of these stressors might better protect the ecosystems
596 under the current conditions (Fenn et al. 2008).

597

598 **3.5 Comparison to Critical Loads in Europe**

599 With a few exceptions, the critical loads for N deposition we report for the United States
600 (Pardo et al. in press a) are lower than those reported for Europe (Bobbink and Hettelingh 2011;
601 Figure 8). There are several potential reasons including: greater availability of pristine baselines
602 in the U.S., more intensive land use in Europe; greater dominance of N deposition by reduced
603 forms of N in Europe, and different threshold criteria.

604 *Availability of pristine baselines.* Because of high historic deposition levels, many European
605 systems lack pristine baseline ecosystems as a reference to compare to those experiencing
606 elevated N deposition. For example, European critical loads for lichens have been much higher
607 than those in the US (Bobbink et al. 2003). These loads were influenced by study sites in
608 Scotland experiencing a deposition gradient from 10-22 kg N ha⁻¹ y⁻¹ from which critical loads
609 were set at 11-18 kg N ha⁻¹ y⁻¹ (Mitchell et al. 2005). However, no oligotrophic species were
610 observed, presumably because they were eliminated prior to the initial studies.

611 *Land Use.* A larger fraction of the forested landscape in Europe is heavily managed
612 (harvested and planted) relative to the U.S. High rates of harvest removals of N in biomass,
613 creating greater N demand and storage during re-establishment of the forest stand could
614 contribute to higher critical loads in Europe than the U.S.

615 *Forms and mode of measurement of N inputs.* NH₄⁺ inputs tend to be higher and represent a
616 greater proportion of total N inputs in Europe, particularly in past decades; this is changing in the

617 U.S. Some receptor species can be more sensitive to reduced than oxidized forms of N inputs,
618 and nitrification of NH_4^+ inputs can accelerate ecosystem acidification relative to inputs of NO_3^- .

619 *Threshold criteria.* Another possible explanation for the higher critical loads is that the
620 response thresholds utilized in Europe are sometimes higher. For example, choosing a threshold
621 of a shift in lichen community composition will produce a much lower critical load than a
622 threshold of near extirpation of lichen species as used in earlier European work (Bobbink et al.
623 2003). As a second example, choosing a threshold of initial changes in N biogeochemistry in the
624 Colorado Front Range interpreted as incipient responses of N saturation, led to a critical load < 4
625 $\text{kg N ha}^{-1} \text{ y}^{-1}$ (Rueth et al. 2003). This is a subtle initial N enrichment response when compared to
626 the magnitude of change (a later stage of N saturation) for the critical loads thresholds in Europe
627 (10-15 kg/ha/yr).

628

629 **4. Conclusions**

630 The most significant changes that we are currently observing in the United States in
631 response to elevated N deposition are changes in species composition: losses of N-sensitive
632 species, shifts in dominance, and losses of native species in favor of exotic, invasive species.
633 Shifts in diatom and lichen community composition away from N-intolerant (oligotrophic)
634 species are observed across the country. Alterations in herbaceous species are broadly observed,
635 but are not always clearly documentable because of the long-term pollution inputs and other
636 disturbances (including land-use change) that caused changes prior to the initiation of careful
637 observations.

638 Numerous examples illustrate the significance of these species- and community-level
639 effects. In serpentine grasslands in California, it was clearly demonstrated that unless N inputs

640 are decreased or N is removed in biomass, a larval host plant and numerous nectar source plants
641 utilized by a threatened and endangered butterfly will decrease to levels unable to sustain the
642 checkerspot butterfly population (Weiss 1999, Fenn et al. 2010). In Joshua Tree National Park in
643 southern California, N deposition favors the production of sufficient invasive grass biomass to
644 sustain fires that threaten the survival of the namesake species (Fenn et al. 2010, Rao et al.
645 2010). Other sensitive ecosystems include alpine meadows, where relatively low levels of N
646 deposition have already changed species composition (Bowman et al. 2006). Changes in
647 historical diatom community composition from N-limited to N-tolerant species have been
648 observed in lake sediment cores at many locations in the western United States, providing early
649 evidence of freshwater ecosystem eutrophication (Wolfe et al. 2001, 2003).

650 Changes in ecosystem structure are linked to changes in ecosystem function. For example,
651 extirpation of lichens can alter food webs by reducing the availability of nesting material for
652 birds, invertebrate habitat, and critical winter forage for mammals, and can also affect nutrient
653 cycling (Cornelissen et al. 2007). In some arid low-biomass California ecosystems, N-enhanced
654 growth of invasive species results in increased fire risk, even in areas where fire is normally
655 infrequent (Allen et al. 2009, Rao et al. 2010, Fenn et al. 2010).

656 There is also evidence of N deposition contributing to multiple stress complexes, resulting
657 in reduced forest sustainability (Grulke et al. 2009, McNulty and Boggs 2010). In North
658 Carolina, elevated N deposition predisposed a pine ecosystem to a pest outbreak following a
659 drought (McNulty and Boggs 2010). These types of complex interactions may be difficult to
660 predict, but may intensify the impact of elevated N deposition in concert with other stressors,
661 including climate change. (Wu and Driscoll 2010). Further examples of changes in ecosystem
662 structure and function are observed in coastal areas, where increased N export has led to toxic

663 algal blooms (Rabalais 2002). As an example of N deposition effects on trace gas chemistry and
664 climate change, N loading to ecosystems results in increased emissions of N trace gases, such as
665 NO (nitric oxide, an ozone precursor), N₂O (nitrous oxide, a long-lived and powerful greenhouse
666 gas); as well as declines in soil uptake of CH₄ (methane, another long lived and powerful
667 greenhouse gas) (e.g., Liu and Greaver 2009).

668 This synthesis demonstrates that elevated N deposition has altered ecosystem structure and
669 function across the United States. Empirical critical loads for N provide a valuable approach for
670 evaluating the risk of harm to ecosystems. This approach has been used broadly in Europe
671 (Bobbink et al. 2003, UBA 2004) and has the advantage of being scientifically based on
672 observed responses. This link to actual ecosystem responses is especially beneficial in resource
673 management and policy contexts. This document and Pardo et al. (in press c) provide the first
674 comprehensive assessment of empirical critical loads of N for ecoregions across the United
675 States. They represent an important step toward providing policymakers and resource managers
676 with a tool for ecosystem protection, as was suggested by the National Research Council (NRC
677 2004).

678

679 **5. Future Research Priorities**

680 The objective of future research should be better integration of improved atmospheric
681 deposition models, empirical and dynamic critical load models in order to develop critical load
682 and exceedance maps at scales useful for regulatory, policy making, land management, and
683 resource protection purposes. A key step in refining critical loads estimates and laying the
684 groundwork for more complex dynamic models is identifying mechanisms that control plant and
685 ecosystem responses to N deposition. We recommend the following research priorities:

- 686 • Improved quantification of total N deposition (wet, dry and cloud/fog), including the
687 measurement of reduced (NH_x) and organic N.
- 688 • An expanded network of long-term, multi-level, low-N fertilization studies, and adequate
689 N deposition gradient studies across a greater diversity of ecosystem types, and extending
690 to regions of low N deposition. Such a network would allow development of dose-
691 response curves for the receptors discussed here that better define the critical load and
692 associated uncertainty.
- 693 • Evaluation of the environmental and ecological factors that influence critical loads for
694 ecoregions and quantifying how the critical load varies as key factors change across
695 ecoregions. In the United States, the sparsest datasets on N deposition effects are in the
696 tundra, taiga, tropical and subtropical forest, and desert ecoregions.
- 697 • Evaluation of the differential response to reduced versus oxidized N inputs. Because
698 some plants are particularly sensitive to NH_x (Krupa 2003) while others are more
699 sensitive to NO_y (Nordin et al. 2006), assembling comprehensive data about species-
700 specific responses would allow more accurate assessment of potential risks to ecosystems
701 in relation to the major N emissions sources.
- 702 • Use of methods that can account for effects on longer-lived organisms, and lack of
703 pristine baselines caused by historical N deposition, other pollutants, or habitat alteration,
704 e.g., dendrochronology, paleolimnology .
- 705 • Quantification of effects of N deposition on forest growth and susceptibility to secondary
706 stressors. Insufficient data are available to determine critical loads for the effects of
707 increasing N inputs on pest outbreaks, drought, cold tolerance, tree vigor, and other
708 multiple stress complexes.

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717

718

719

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Table 1 – Summary of critical loads of nutrient N for North American ecoregions

reliable; # fairly reliable; (#) expert judgment

Ecoregion	Ecosystem Component	CL for N deposition <i>kg N ha⁻¹ yr⁻¹</i>	Reliability	Response	Comments	Study
Tundra	Prostrate dwarf shrubs	1-3	##	Changes in CO ₂ exchange, cover, foliar N, and community composition of vascular plants	N addition study, Greenland high arctic, P enhanced N effects.	Arens et al. 2008 ^a
Tundra	Lichens	1-3	(#)	Changes in lichen pigment production and ultrastructure, changes in lichen and bryophyte cover	N addition studies, high and low arctic, P enhanced or moderated N effects.	Arens et al. 2008 ^a , Hyvärinen et al. 2003 ^b , Makkonen et al. 2007 ^b
Taiga	Forest	1-3	#	Changes in alga, bryophyte, and lichen community composition, cover, tissue N or growth rates.		Berryman et al. 2004 ^c , Berryman and Straker 2008 ^c , Geiser et al. 2010, Moore et al. 2004 ^c , Poikolainen et al. 1998 ^b , Strengbom et al. 2003 ^d , Vitt et al. 2003 ^c ,
Taiga	Spruce forests	5-7	(#)	Ectomycorrhizal fungi, change in community structure	Expert judgment extrapolated from Marine West coast spruce and northern spruce-fir forest	Lilleskov 1999; Lilleskov et al. 2001, 2002, 2008

Taiga	Shrublands	6	##	Shrub and grass cover, increased parasitism of shrubs	Long term, low N addition study: shrub cover decreased, grass cover increased	Nordin et al. 2005 ^d , Strengbom et al. 2003 ^d
Northern Forests	Hardwood and Coniferous Forests	>3	#	Tree growth and mortality	Decreased growth of red pine, and decreased survivorship of yellow birch, scarlet and chestnut oak, quaking aspen, and basswood	Thomas et al. 2010
Northern Forests	Lichens	4-6	(#)	Epiphytic lichen community change	Loss of oligotrophic species. Synergistic/confounding effects of acidic deposition not considered; assumes response threshold similar to Marine West Coast Forest	Geiser et al 2010.
Northern Forests	Ectomycorrhizal fungi	5-7	#	Change in fungal community structure		Lilleskov et al. 2008
Northern Forests	Herbaceous cover species	>7 and <21	#	Loss of prominent species	Response observed in low-level fertilization experiment	Hurd et al. 1998
Northern Forests	Hardwood and Coniferous Forests	8	##	Increased surface water NO ₃ ⁻ leaching		Aber et al. 2003
Northern Forests	Old-growth montane red spruce	>10 and <26	#	Decreased growth and/or induced mortality	Response observed in low-level fertilization experiment	McNulty et al. 2005

Northern Forests	Arbuscular mycorrhizal fungi	<12	(#)	biomass decline and community composition change		van Diepen 2008, van Diepen et al. 2007
Northwest Forested Mountains	Alpine lakes	1.5	##	Diatom assemblages	As wet deposition only	Baron 2006
Northwest Forested Mountains	Lichens	1.2-3.7	(#)	Epiphytic lichen community change in mixed-conifer forests, Alaska	Application of western Oregon and Washington model	Geiser et al. 2010
Northwest Forested Mountains	Lichens	2.5-7.1	##	Epiphytic lichen community change, thallus N enrichment in mixed-conifer forests, non-Alaska		Fenn et al. 2008, Geiser et al. 2010
Northwest Forested Mountains	Sub-alpine forest	4	##	Increase in organic horizon N, foliar N, potential net N mineralization, and soil solution N, initial increases in N leaching below the organic layer		Rueth and Baron 2002, Baron et al. 1994
Northwest Forested Mountains	Alpine lakes	4.0	#	Episodic freshwater acidification		Williams and Tonnesson 2000
Northwest Forested Mountains	Alpine grassland	4-10	##	Plant species composition		Bowman et al. 2006
Northwest Forested Mountains	Ectomycorrhizal fungi	5-10	(#)	Ectomycorrhizal fungi community structure in white, black, and Engelmann spruce forests	Expert judgment extrapolated from Marine West Coast spruce and northern spruce-fir forest	Lilleskov 1999; Lilleskov et al. 2001, 2002, 2008

Northwest Forested Mountains	Mixed conifer forest	17	## #	NO ₃ ⁻ leaching, reduced fine root biomass		Fenn et al. 2008
Marine West Coast Forest	Western OR and WA forests	2.7-9.2	##	Epiphytic lichen community change	Loss of oligotrophic species, enhancement of eutrophic species. CL increases with regional range in mean annual precipitation from 45-450 cm	Geiser et al. 2010
Marine West Coast Forest	SE Alaska forests	5	(#)	Fungal community change; declines in ectomycorrhizal fungal diversity		Lilleskov 1999; Lilleskov et al. 2001, 2002 ; Whytemare et al. 1997
Eastern Temperate Forest	Eastern Hardwood Forest	>3	#	Decreased growth of red pine, and decreased survivorship of yellow birch, scarlet and chestnut oak, quaking aspen, and basswood		Thomas et al. 2010
Eastern Temperate Forest	Lichens	4-8	(#)	Epiphytic lichen community change	Loss of oligotrophic species. Synergistic/confounding effects of acidic deposition not considered; based on application of model and estimated response threshold	Geiser et al. 2010
Eastern Temperate Forest	Southeast Coastal Plain	5-10	(#)	Ectomycorrhizal fungi community response		Dighton et al. 2004; Lilleskov et al. 2001, 2002, 2008

Eastern Temperate Forest	Eastern Hardwood Forests	8	##	Increased surface water loading of NO ₃ ⁻		Aber et al. 2003
Eastern Temperate Forest	Michigan deposition gradient	<12	(#)	Arbuscular mycorrhizal fungal biomass decline and community composition change		van Diepen 2008, van Diepen et al. 2007
Eastern Temperate Forest	Herbaceous species	<17.5	(#)	Increases in nitrophilic species, declines in species-rich genera (e.g., <i>Viola</i>)		Gilliam 2006, 2007; Gilliam et al. 2006
Great Plains	Tall-grass prairie	5-15	#	Biogeochemical N cycling, plant and insect community shifts		Clark et al. 2009, Clark and Tilman 2008; Tilman 1993, 1987; Wedin and Tilman 1996
Great Plains	Mixed-grass prairie	10-25	#	Soil NO ₃ ⁻ pools, leaching, plant community shifts		Clark et al. 2003, 2005; Jorgenson et al. 2005
Great Plains	Short-grass prairie	10-25	(#)		Inferred from mixed grass	Epstein 2001, Barret and Burke 2002
Great Plains	Mycorrhizal fungi	12	(#)	Decline in arbuscular mycorrhizal fungal activity		Egerton-Warburton, unpub. data
North American Desert	lichens	3	(#)	Lichen community shifts, thallus N concentration	Uncertainty regarding modeled estimates	Geiser et al. 2008, Porter et al. 2007
North American Desert	shrubland, woodland, desert grassland	3-8.4	#	Vegetation response, vascular plant community change		Allen et al. 2009; Baez et al. 2007; Inouye 2006; Rao et al. 2010

Mediterranean California	Coastal Sage Scrub	7.8-10	#	Invasive grass cover, native forb richness, arbuscular mycorrhizal fungi richness	Modeled and inferential N deposition estimates and published data for mycorrhizae, unpublished data for vegetation survey.	Fenn et al.,2010, Egerton-Warburton and Allen 2000, Tonnesen et al. 2007
Mediterranean California	Chaparral; Lichens	3-6	#	Epiphytic lichen community change	Lichen critical load is from modeled N deposition data and published data for lichens.	Geiser et al. 2010; Jovan 2008; Jovan and McCune 2005;
Mediterranean California	Chaparral, Oak Woodlands, Central Valley	10-14	#	NO ₃ ⁻ leaching; stimulated N cycling	Critical load for NO ₃ ⁻ leaching of 10 kg N ha ⁻¹ yr ⁻¹ is based on one year of throughfall data in Chamise Creek and an additional year of throughfall data from adjacent Ash Mountain, both in Sequoia National Park.	Fenn et al. 2003a, b, c; Fenn and Poth 1999; Meixner and Fenn 2004
Mediterranean California	Mixed conifer forest; Lichens	3.1-5.2	##	Lichen chemistry and community changes	The lowest critical load is based on lichen tissue chemistry above the clean site threshold.	Fenn et al. 2008, 2010
Mediterranean California	Mixed conifer forest; plant physiology	17	#	Reduced fine root biomass		Fenn et al. 2008, 2010; Grulke et al. 1998
Mediterranean California	Mixed conifer forest; soil processes	17-25.9	#	NO ₃ ⁻ leaching; soil acidification		Breiner et al. 2007, Fenn et al. 2008, 2010

Mediterranean California	Mixed conifer forest; forest sustainability	24-39	(#)	Understory biodiversity; forest sustainability	N deposition from Fenn et al. 2008	Allen et al. 2007; Grulke and Balduman 1999; Grulke et al. 1998, 2009; Jones et al. 2004
Mediterranean California	Serpentine grassland	6	##	Annual grass invasion, replacing native herbs	Critical load based on a local roadside gradient; Serpentine grassland site is actually west of the Central Valley.	Weiss 1999; Fenn et al. 2010
Temperate Sierras	Lichens	4-7	(#)	Epiphytic lichen community change	Increase in proportion of eutrophic species. Estimated from MWCF model, response threshold allows ~60% eutrophs due to dry, hot climate, hardwood influence	Geiser et al. 2010
Temperate Sierras	Las Cruces and Chichinautzin Ranges S/SW of Mexico City	15	#	Elevated NO ₃ ⁻ in stream and spring waters	Data are from <i>Pinus hartwegii</i> sites in the Desierto de los Leones National Park and Ajusco	Fenn et al. 1999, 2002
Tropical and Subtropical humid Forests	N-rich forests	<5-10	(#)	NO ₃ ⁻ leaching, N trace gas emissions	CL for N-rich forests should be lower than for N-poor forests based on possibility of N losses.	ND
Tropical and Subtropical Humid Forests	N-poor forests	5-10	(#)	Changes in community composition; NO ₃ ⁻ leaching, N trace gas emissions	CL for N-poor forests based on estimates for Southeastern Coastal Plain forests.	ND

Wetlands	Freshwater wetlands	2.7-13	#	Peat accumulation and NPP	CL for a wetlands in the northeastern U.S. and southeastern Canada	Aldous 2002 ^c , Moore et al. 2004 ^c , Rochefort et al. 1990 ^c , Vitt et al 2003 ^c
Wetlands	Freshwater wetlands	6.8-14	(#)	Pitcher plant community change	CL based on northeastern populations	Gotelli and Ellison 2002, 2006
Wetlands	Intertidal wetlands	50-100	##	Loss of eelgrass		Latimer and Rego 2010
Wetlands	Intertidal salt marshes	63-400	(#)	Salt marsh community structure, microbial activity and biogeochemistry		Caffrey et al. 2007, Wigand et al. 2003
Aquatic	Western Lakes	2	##	Freshwater eutrophication		Baron 2006
Aquatic	Eastern Lakes	8	#	NO ₃ ⁻ leaching		Aber et al. 2003

a– based on data from Greenland; b – based on data from Finland; c – based on data from Canada; d – based on data from Sweden

Table 2 – Assessment and interpretation of empirical critical loads of nutrient N for North American ecoregions

Ecoregion	Factors affecting the range of CL ^a	Comparison within Ecoregion ^b
Tundra	<ol style="list-style-type: none"> 1) moisture 2) competition between vascular plants and cryptogams 3) P-limitation 4) temperature 5) pH 	<p>The critical load is higher in wet and P-limited tundra; acidic tundra may be more sensitive to N deposition than non-acidic tundra. Increased N deposition may be more detrimental to lichens in the presence of graminoids and shrubs in the low and mid arctic than to lichens with less competition in the high arctic. Response time increases with latitude due to colder temperatures, less light, and poorer N and P mobilization.</p>
Taiga	<ol style="list-style-type: none"> 1) soil depth 2) vegetation type and species composition 3) latitude 	<p>Morphological damage to lichens has been observed at a lower deposition in forests and woodlands than in shrublands or bogs and fens; cryptogam dominated mats on thin soils become N saturated faster than forest islands.</p>
Northern Forest	<ol style="list-style-type: none"> 1) receptor 2) tree species 3) stand age 4) site history 5) pre-existing N status 	<p>CLs for lichen are generally lowest, followed by CLs for ectomycorrhizal fungi and NO₃⁻ leaching. CLs for herbaceous species and forests are generally higher than for other responses.</p>
Northwest Forested Mountains	<ol style="list-style-type: none"> 1) biotic receptor 2) accumulated load of N 3) ecosystem 4) region 	<p>In alpine regions, diatom changes in lakes are seen at lowest the CL. Changes in individual plants are seen next, followed by vegetation community change, then soil responses. In subalpine forests, the CL of 4 kg ha⁻¹ yr⁻¹ for foliar and soil chemistry changes is similar to the lichen CL of 3.1 – 5.2 for lichen community change.</p>
Marine West Coast Forest	<ol style="list-style-type: none"> 1) background N status 2) soil type 3) species composition 4) fire history 5) climate 	<p>The midrange of responses reported for lichens (2.7 – 9.2 kg ha⁻¹ yr⁻¹) is broadly comparable to that for plant, soil, and mycorrhizal responses (5 kg ha⁻¹ yr⁻¹), despite limited studies for non-lichen responses.</p>

Eastern Forests	<ol style="list-style-type: none"> 1) precipitation 2) soil cation fertility and weathering 3) biotic receptors 	<p>The CL for NO_3^- leaching, lichen community change, and ectomycorrhizal fungal response are within the same range. Arbuscular mycorrhizal fungal and herbaceous CLs are higher.</p>
Great Plains	<ol style="list-style-type: none"> 1) N status 2) receptor 3) precipitation 	<p>CLs are lower in the tall grass prairie than in the mixed- and short-grass prairies. CL in tall- and mixed-grass prairie is lower on N poor sites and sites with very N responsive plant species. CL in the short-grass prairie is likely lower in wet years than in dry years.</p>
North American Deserts	<ol style="list-style-type: none"> 1) receptor 2) interaction of annual grasses with native forb cover 3) precipitation 	<p>The lichen CL is lowest, at $3 \text{ kg N ha}^{-1} \text{ yr}^{-1}$; vegetation CL varies from $3\text{-}8.4 \text{ kg N ha}^{-1} \text{ yr}^{-1}$</p>
Mediterranean California	<ol style="list-style-type: none"> 1) presence of invasive exotic annual grasses interacting with a highly diverse native forb community 2) N-sensitivity of mycorrhizal fungi 3) N-sensitivity of lichens 4) N retention capacity of catchments, catchment size 5) co-occurrence of ozone and ozone-sensitive tree species. 	<p>The lowest CLs in Mediterranean California are for sensitive lichen in chaparral and oak woodlands and mixed conifer forests. The CL for plant and mycorrhizal fungal community change in coastal sage scrub is higher, at $7.8 \text{ to } 10 \text{ kg ha}^{-1} \text{ yr}^{-1}$. CL for NO_3^- leaching is lower in chaparral and oak woodlands ($10\text{-}14 \text{ kg ha}^{-1} \text{ yr}^{-1}$) than in mixed conifer forests ($17 \text{ kg ha}^{-1} \text{ yr}^{-1}$). CLs are highest for mixed conifer forest plant community change and sustainability. Fine root biomass in ponderosa pine is reduced by both ozone and elevated soil nitrogen.</p>
Wetlands	<ol style="list-style-type: none"> 1) vegetation species 2) the fraction of rainfall in the total water budget 3) the degree of openness of N cycling 	<p>CL is much higher for intertidal wetlands ($50\text{-}400 \text{ kg ha}^{-1} \text{ y}^{-1}$) than for freshwater wetlands ($2.7\text{-}14 \text{ kg ha}^{-1} \text{ y}^{-1}$), which have relatively close water and N cycles.</p>
Freshwater s	<ol style="list-style-type: none"> 1) extent of upstream vegetation development 2) topographic relief 3) land use/deposition history 	<p>CLs are lower in Western mountain lakes/streams with poorly vegetated watersheds and steep catchments. CLs are greater in eastern lakes with prior land use and decades of acidic deposition.</p>

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- ^a - This explains what factors cause the critical load (CL) to be at the low or high end of the range reported.
 - ^b - Comparison of values and causes for differences if multiple critical loads are reported for an ecoregion.

Table 3 – Empirical critical loads of nutrient N for mycorrhizal fungi in U.S. ecoregions

reliable; # fairly reliable; (#) expert judgment

Ecoregion	Ecosystem (Site)	CL for N deposition <i>kg ha⁻¹ yr⁻¹</i>	Reliability	Response of	Comments	Study
Taiga	Spruce forests	5-7	(#)	Ectomycorrhizal fungi, change in community structure	Expert judgment extrapolated from Marine West coast spruce and northern spruce-fir forest	Lilleskov 1999; Lilleskov et al. 2001, 2002, 2008
Northern Forests	Spruce-fir forest (Northeastern U.S. deposition gradient)	5-7	#	Ectomycorrhizal fungi, change in morphotype community structure	Wet deposition estimated from Ollinger et al (1993) model	Lilleskov et al. 2008
Northern Forests	Northern hardwood forests; Sugar maple dominated (Michigan gradient)	<12	(#)	Arbuscular mycorrhizal fungi, decrease in abundance in roots, soil, community change	N fertilization experiment	Van Diepen et al. 2007 van Diepen 2008
Northwest Forested Mountains	Engelmann spruce forests	5-10	(#)	Ectomycorrhizal fungi, change in community structure	Expert judgment extrapolated from Marine West coast spruce and northern spruce-fir forest	Lilleskov 1999; Lilleskov et al. 2001, 2002, 2008
Marine West Coast	White spruce forest (Kenai Peninsula, Alaska)	5	(#)	Ectomycorrhizal fungi, change in community structure, decrease in species richness	Bulk deposition. Historic N deposition was higher but unquantified. CL estimated from regression	Lilleskov 1999, Lilleskov et al. 2001, 2002, Whytemare et al. 1997
Eastern Temperate Forests	Southeast Coastal Plain	5-10	(#)	Ectomycorrhizal fungi, change in community structure	From one study in pine barrens plus extrapolation from other oligotrophic conifer forests	Dighton et al. 2004; Lilleskov et al. 2001, 2002, 2008
Eastern Temperate	Pine Barrens (New Jersey;	<8	(#)	Ectomycorrhizal fungal morphotype	Bulk deposition. Gradient study	Dighton et al. 2004

Ecoregion	Ecosystem (Site)	CL for N deposition <i>kg ha⁻¹ yr⁻¹</i>	Reliability	Response of	Comments	Study
Forests	Southeast Coastal Plain)			community change	with three sample points	
Eastern Temperate Forests	Eastern Hardwoods; sugar maple dominated (Michigan gradient)	<12	(#)	Arbuscular mycorrhizal fungi; decrease in abundance in roots, soil, community change	Long term (12 yr) N fertilization experiment in sugar maple	Van Diepen et al. 2007, van Diepen 2008
Great Plains	Chicago Grassland	12		Arbuscular mycorrhizal fungi; decrease in % colonization, spore density	CL estimated from logarithmic curve of soil N vs. AMF activity. No low N baseline, so CL may be lower.	Egerton-Warburton, unpub. data
Mediterranean California	Coastal sage scrub (southern California)	7.8-9.2	#	Arbuscular mycorrhizal fungi, decrease in % colonization, spore density, spore richness	CL estimated from logarithmic curve fitted to data from this study compared to modeled and inferential N deposition data	Egerton-Warburton and Allen 2000, Tonnesen et al. 2007

Table 4 – Empirical critical loads of nutrient N for lichens in U.S. ecoregions

reliable; # fairly reliable; (#) expert judgment

Ecoregion	Ecosystem (Site)	CL for N deposition <i>kg ha⁻¹ yr⁻¹</i>	Reliability	Response of	Comments	Study
Tundra	Tundra	1-3	(#)	Changes in lichen pigment production and ultrastructure, changes in lichen and bryophyte cover	N addition studies, high and low arctic, P enhanced or moderated N effects.	Arens et al. 2008 ^a , Hyvärinen et al. 2003 ^b , Makkonen et al. 2007 ^b
Taiga	Taiga	1-3	#	Changes in alga, bryophyte, and lichen community composition, cover, tissue N or growth rates.		Berryman et al. 2004 ^c , Berryman and Straker 2008 ^c , Geiser et al. 2010, Moore et al. 2004 ^c , Poikolainen et al. 1998 ^b , Strengbom et al. 2003 ^d , Vitt et al. 2003 ^c
Northern Forests	Northern Forests	4-6	(#)	Changes in lichen physiology and community structure	CL proposed based on values in the Taiga and Northwest Forested Mountains.	Geiser et al. 2010
Northwest Forested Mountains	Coniferous forests, Alaska	1.2-3.7	(#)	Lichen community composition	Application of western Oregon and Washington model	Geiser et al. 2010
Northwest Forested Mountains	Coniferous Forests, non- Alaska	2.5-7.1	##	Lichen community composition	Application of western Oregon and Washington model	Geiser et al. 2010
Northwest Forested Mountains	Central Southern Sierras	3.1-5.2	##	Shifts in epiphytic lichen communities favoring eutrophs	CL based on exceedance of a N concentration threshold in the	Fenn et al. 2008, 2010

Ecoregion	Ecosystem (Site)	CL for N deposition <i>kg ha⁻¹ yr⁻¹</i>	Reliability	Response of	Comments	Study
Marine West Coast Forests	Western OR and WA forests	2.7-9.2	##	Shifts in epiphytic lichen communities favoring eutrophs	lichen <i>Letharia vulpina</i> CL increases with increasing mean annual precipitation from 40 to 240 cm.	Geiser et al. 2010
Eastern Forests	Eastern hardwoods and Southeast Coastal Plain	4-8 4-6	(#) (#)	Shifts in epiphytic lichen communities favoring eutrophs		Geiser et al. 2010
North American Deserts	Cold desert (Hells Canyon National Resource Area)	3	(#)	Increased cover and abundance of nitrophilous lichens on tall shrubs, increased parasitism of lichens.	CL estimated from overlay of course grid (36 km) CMAQ N; local N deposition from NH ₃ was likely higher.	Geiser et al. 2008, Porter et al. 2007
Mediterranean California	Oak woodlands and chaparral (Central Valley: Sacramento Valley, Coast Ranges and Sierra foothills)	3-6	#	Shifts in epiphytic lichen communities favoring eutrophs	FHM lichen survey of 118 forested sites. Eutrophs dominated communities when CMAQ 4 km N deposition estimates were >5.5 kg ha ⁻¹ yr ⁻¹	Geiser et al. 2010, Jovan 2008, Jovan and McCune 2005
Mediterranean California	Mixed Conifer forest (Sierra Nevada)	3.1-5.2	##	Shifts in epiphytic lichen communities favoring eutrophs	Extrapolated from Northwestern Forested Mountains Sierra Nevada study.	Fenn et al. 2008
Temperate Sierras	Lichens	4-7	(#)	Shifts in epiphytic lichen communities favoring eutrophs	Increase in proportion of eutrophic species.	Geiser et al. 2010

a – based on data from Greenland; b – based on data from Finland; c – based on data from Canada; d – based on data from Sweden

Table 5 – Empirical critical loads of nutrient N for herbaceous plants and shrubs in U.S. ecoregions

reliable; # fairly reliable; (#) expert judgment

Ecoregion	Ecosystem (Site)	CL for N $kg\ ha^{-1}\ yr^{-1}$	Reliability	Response	Comments	Study
Tundra	Prostrate dwarf shrub	1-3	##	Changes in CO ₂ exchange, cover, foliar N, and community composition of vascular plants	N addition study, Greenland high arctic, P enhanced N effects.	Arens et al. 2008 ^a
Taiga	Shrublands	6	##	Change in shrub and grass cover, increased parasitism of shrubs	Long term, low N addition study: shrub cover decreased, grass cover increased	Nordin et al. 2005 ^b , Strengbom et al 2003 ^b
Northern forests	Northern hardwood forests (Adirondacks)	> 7 and <21	#	Alteration of herbaceous understory		Hurd et al. 1998
Northwestern Forested Mountains	Alpine grasslands	4-10	##	Plant species composition change	Based on long-term experiment	Bowman et al. 2006
Eastern Forests	Eastern hardwood forests (Fernow Experimental Forest, WV)	<17.5	(#)	Increases in nitrophilic species, declines in species-rich genera (e.g., <i>Viola</i>)		Gilliam 2006, Gilliam 2007, Gilliam et al. 2006
Great Plains	Tall-grass prairie	5-15	#	Biogeochemical N cycling, plant and insect community shifts	Long-term, low N addition study that also added other nutrients.	Clark et al. 2009; Clark and Tilman 2008; Tilman 1993, 1987; Wedin and Tilman 1996
Great Plains	Mixed-grass prairie	10-25	#	Soil NO ₃ ⁻ pools, leaching, plant community shifts	Short-term, low N addition study.	Jorgenson et al. 2005, Clark et al 2003, 2005
Great Plains	Short-grass prairie	10-25	(#)		Inferred from mixed grass.	Epstein 2001, Barrett and Burke 2002
North American Desert	Warm desert (Joshua Tree National Park, Mojave Desert)	3-8.4	#	Increased biomass of invasive grasses; decrease of native forbs		Allen et al. 2009, Rao et al. 2010

Ecoregion	Ecosystem (Site)	CL for N <i>kg ha⁻¹ yr⁻¹</i>	Reliability	Response	Comments	Study
Mediterranean California	Serpentine grassland	6	##	Annual grass invasion, replacing native herbs	Critical load based on a local roadside gradient; Serpentine grassland site is actually west of the Central Valley.	Weiss 1999; Fenn et al. 2010
Mediterranean California	Coastal Sage Scrub	7.8-10	#	Invasive grass cover, native forb richness	Modeled and inferential N deposition estimates and unpublished data for vegetation survey.	Fenn et al. in press, 2010, Egerton-Warburton et al. 2001, Tonnesen et al. 2007
Mediterranean California	Mixed conifer forests (San Bernardino mountains)	24-33	(#)	Biodiversity of understory: percent cover and no. of species/3 ha	Based on plant surveys in 1970s and 2003.	Allen et al. 2007 N deposition data: Fenn, unpublished data ; Fenn et al. 2008
Wetlands	Freshwater wetlands	6.8-14	(#)	Pitcher plant community change	CL based on northeastern populations	Gotelli and Ellison 2002, 2006
Wetlands	Intertidal wetlands	50-100	##	Loss of eelgrass		Latimer and Rego 2010
Wetlands	Intertidal salt marsh	63-400	(#)	Salt marsh community structure, microbial activity and biogeochemistry		Caffrey et al. 2007, Wigand et al. 2003

a - based on data from Greenland; b – based on data from Sweden

Table 6 – Empirical critical loads of nutrient N for forest ecosystems in U.S. ecoregions

reliable; # fairly reliable; (#) expert judgment

Ecoregion	Ecosystem (Site)	CL for N $kg\ ha^{-1}\ yr^{-1}$	Reliability	Response	Comments	Study
Northern forests	Northeastern gradient	>3	#	Decline in survivorship of sensitive species	Based on study of gradient of N deposition from 3-11 kg N ha ⁻¹ yr ⁻¹	Thomas et al. 2010
Northern Forests	Hardwood and coniferous forests	8	##	Increased surface water and NO ₃ ⁻ leaching		Aber et al. 2003
Northern forests	Montane spruce fir (Mt. Ascutney, VT)	>10 and <26	#	Declines in growth and increased mortality		McNulty et al. 2005
Northwestern Forested Mountains	sub-alpine forest	4	##	soil organic horizon and foliar N enrichment and higher potential net N mineralization rates		Baron et al. 1994, Rueth and Baron 2002
Northwestern Forested Mountains	Mixed conifer forest	17	## #	NO ₃ ⁻ leaching, reduced fine root biomass	Co-occurring ozone also affects fine root biomass in ponderosa pine	Fenn et al. 2008
Marine West Coast Forests	Coastal white spruce forest (South-Central Alaska)	5	(#)	declines in tree health; changes in understory composition; foliar nutritional imbalances; elevated NO ₃ ⁻ in forest floor and mineral soil		Lilleskov 1999, Lilleskov et al. 2001, 2002; Whytemare et al. 1997
Eastern Forests	Eastern hardwood forests	>3	#	Decline in survivorship of sensitive species	Based on study of gradient of N deposition from 3-11 kg N ha ⁻¹ yr ⁻¹	Thomas et al. 2010
Eastern Forests	Eastern Hardwood Forests	8	##	Increased surface water loading of NO ₃ ⁻		Aber et al. 2003

Ecoregion	Ecosystem (Site)	CL for N <i>kg ha⁻¹ yr⁻¹</i>	Reliability	Response	Comments	Study
Mediterranean California	mixed conifer forests (San Bernardino mountains and southern Sierra Nevada range)	17	##	Streamwater [NO ₃ ⁻] > 14 µM	Based on regression of throughfall vs. peak streamwater NO ₃ ⁻ concentrations. Daycent simulations gave similar results.	Fenn et al. 2008
Mediterranean California	Mixed conifer forests (San Bernardino mountains)	17	#	Reduced fine root biomass	Based on regression of throughfall N deposition and fine root biomass in ponderosa pine (also affected by co-occurring ozone).	Fenn et al. 2008; Grulke et al., 1998
Mediterranean California	Mixed conifer forests (San Bernardino mountains)	25.9	#	Soil acidification; pH <= 4.6	Based on regression of throughfall N deposition and mineral soil H ⁺ .	Breiner et al. 2007
Mediterranean California	Mixed conifer forests (San Bernardino mountains)	39	(#)	Forest sustainability	Based on shifts in plant phenology and C allocation. Caused by combined effects of ozone and N deposition. Leads to increased bark beetle mortality and wildfire risk.	Grulke and Balduman 1999; Grulke et al. 1998, 2009; Jones et al. 2004 N deposition data from Fenn et al. 2008.
Tropical and Subtropical Humid Forests	N-poor tropical and subtropical forests	5-10	(#)	ND	CL for N-poor forests based on estimates for Southeastern Coastal Plain forests.	ND
Tropical and Subtropical Humid Forests	N-rich tropical and subtropical forests	<5-10	(#)	ND	CL for N-rich forests should be lower than for N-poor forests based on possibility of N losses.	ND

Figure legends

Figure 1-- Ecological Regions of North America, Level I

From the Commission for Environmental Cooperation (1997)

Figure 2 – Locations of the more than 3200 locations for which we report ecological responses to N deposition labeled with estimates of wet + dry nitrogen (N) deposition in $\text{kg ha}^{-1} \text{ yr}^{-1}$ (includes wet ammonium and nitrate, dry nitric acid, particulate nitrate and ammonium, and gaseous ammonia, but not organic forms) generated by the CMAQ 2001 model. In some areas of elevated N deposition CMAQ at this grid scale (36 km) likely underestimates total N deposition. This is certainly the case over much of California (Fenn et al. 2010).

Figure 3 – Map of (a) critical loads and (b) exceedances for mycorrhizal fungi by ecoregion in the United States

The range of critical loads reported for mycorrhizal fungi is shown for each ecoregion. The hatch marks indicate increasing level of uncertainty: no hatch marks for the most certain "reliable" category, single hatching for the "fairly reliable" category, and double hatching for the "expert judgment" category. The color sequence moves from red toward blue and violet as the critical load increases. As the range of the critical load gets broader, the saturation of the color decreases.

Exceedance (critical load – deposition) is shown for several categories: (1) No exceedance (Below CL), when deposition is lower than the CL range, (2) At CL, when deposition is within +/-1 of the CL range, (3) Above CL_{min} , when deposition is above the lower end of the CL range, but lower than the upper end of the range, (4) Above CL_{max} , when deposition is above the upper

end of the CL range. CMAQ deposition data were not available for Alaska, so we were not able to calculate exceedance for Alaska.

Figure 4 - Map of (a) critical loads and (b) exceedances for lichens by ecoregion in the United States

The range of critical loads reported for lichens is shown for each ecoregion. The hatch marks indicate increasing level of uncertainty: no hatch marks for the most certain "reliable" category, single hatching for the "fairly reliable" category, and double hatching for the "expert judgment" category. The color sequence moves from red toward blue and violet as the critical load increases. As the range of the critical load gets broader, the saturation of the color decreases.

Exceedance (critical load – deposition) is shown for several categories: (1) No exceedance (Below CL), when deposition is lower than the CL range, (2) At CL, when deposition is within +/-1 of the CL range, (3) Above CL_{min} , when deposition is above the lower end of the CL range, but lower than the upper end of the range, (4) Above CL_{max} , when deposition is above the upper end of the CL range. CMAQ deposition data were not available for Alaska, so we were not able to calculate exceedance for Alaska.

Figure 5 – Map of (a) critical loads and (b) exceedances for herbaceous plants and shrubs by ecoregion in the United States.

The range of critical loads reported for herbaceous plants and shrubs is shown for each ecoregion. The hatch marks indicate increasing level of uncertainty: no hatch marks for the most certain "reliable" category, single hatching for the "fairly reliable" category, and double hatching

for the “expert judgment” category. The color sequence moves from red toward blue and violet as the critical load increases. As the range of the critical load gets broader, the saturation of the color decreases.

Exceedance (critical load – deposition) is shown for several categories: (1) No exceedance (Below CL), when deposition is lower than the CL range, (2) At CL, when deposition is within +/-1 of the CL range, (3) Above CL_{min} , when deposition is above the lower end of the CL range, but lower than the upper end of the range, (4) Above CL_{max} , when deposition is above the upper end of the CL range. CMAQ deposition data were not available for Alaska, so we were not able to calculate exceedance for Alaska.

Figure 6 – Map of (a) critical loads and (b) exceedances for forest ecosystems by ecoregion in the United States.

The range of critical loads reported for forest ecosystems is shown for each ecoregion; this map does not include the responses of mycorrhizal fungi, lichens, or understory herbaceous plants already represented. The hatch marks indicate increasing level of uncertainty: no hatch marks for the most certain "reliable" category, single hatching for the "fairly reliable" category, and double hatching for the “expert judgment” category. The color sequence moves from red toward blue and violet as the critical load increases. As the range of the critical load gets broader, the saturation of the color decreases.

Exceedance (critical load – deposition) is shown for several categories: (1) No exceedance (Below CL), when deposition is lower than the CL range, (2) At CL, when deposition is within +/-1 of the CL range, (3) Above CL_{min} , when deposition is above the lower end of the CL range, but lower than the upper end of the range, (4) Above CL_{max} , when deposition is above the upper

end of the CL range. CMAQ deposition data were not available for Alaska, so we were not able to calculate exceedance for Alaska.

Figure 7 – Map of (a) critical loads and (b) exceedances based on increased nitrate leaching by ecoregion in the United States.

The range of critical loads based on increased nitrate leaching for each ecoregion. The hatch marks indicate increasing level of uncertainty: no hatch marks for the most certain "reliable" category, single hatching for the "fairly reliable" category, and double hatching for the "expert judgment" category. The color sequence moves from red toward blue and violet as the critical load increases. As the range of the critical load gets broader, the saturation of the color decreases.

Exceedance (critical load – deposition) is shown for several categories: (1) No exceedance (Below CL), when deposition is lower than the CL range, (2) At CL, when deposition is within +/-1 of the CL range, (3) Above CL_{min} , when deposition is above the lower end of the CL range, but lower than the upper end of the range, (4) Above CL_{max} , when deposition is above the upper end of the CL range. CMAQ deposition data were not available for Alaska, so we were not able to calculate exceedance for Alaska.

Figure 8-Comparison of European and US empirical critical loads for nutrient nitrogen

Critical loads for comparable European ecosystems, based on Bobbink and Hettelingh (2011), are typically higher than those reported for US ecoregions. For a number of receptors, the Bobbink and Hettelingh (2011) values are lower than previous empirical critical loads for Europe (Bobbink et al. 2003).



Figure 1

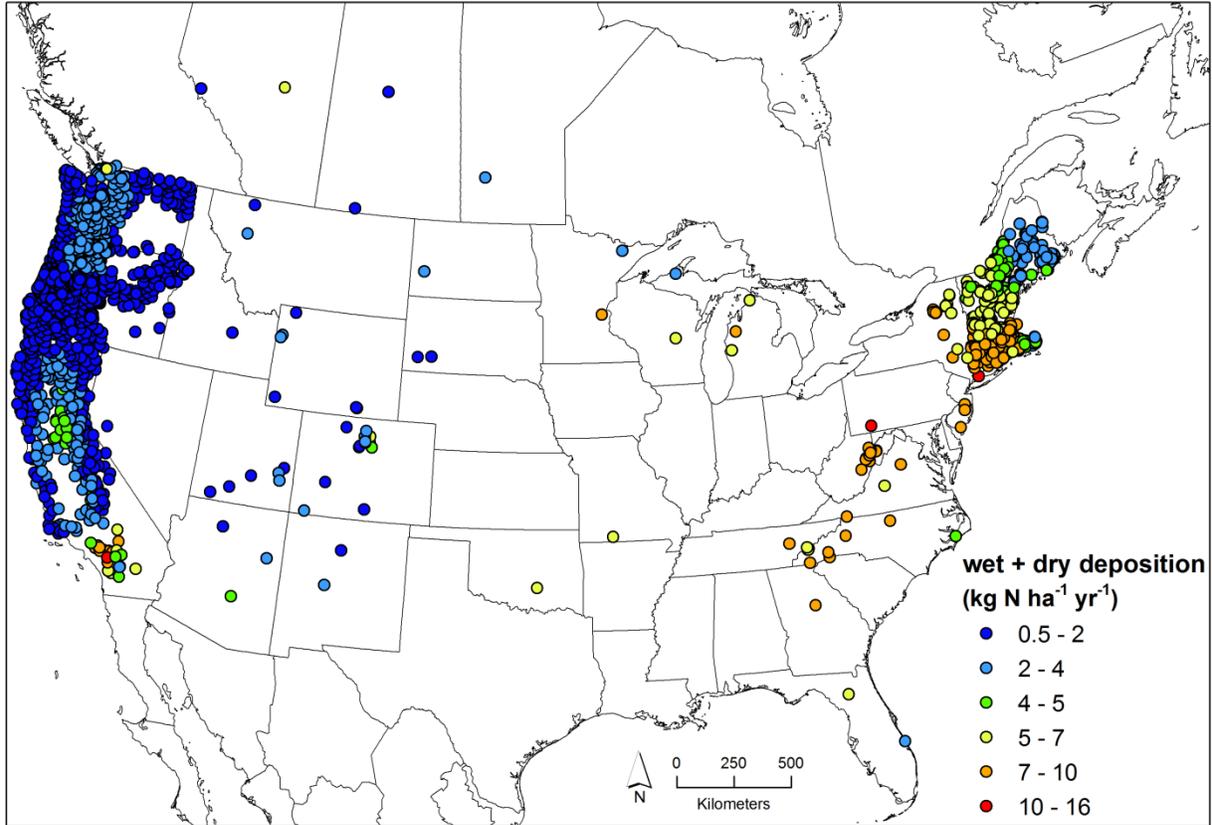
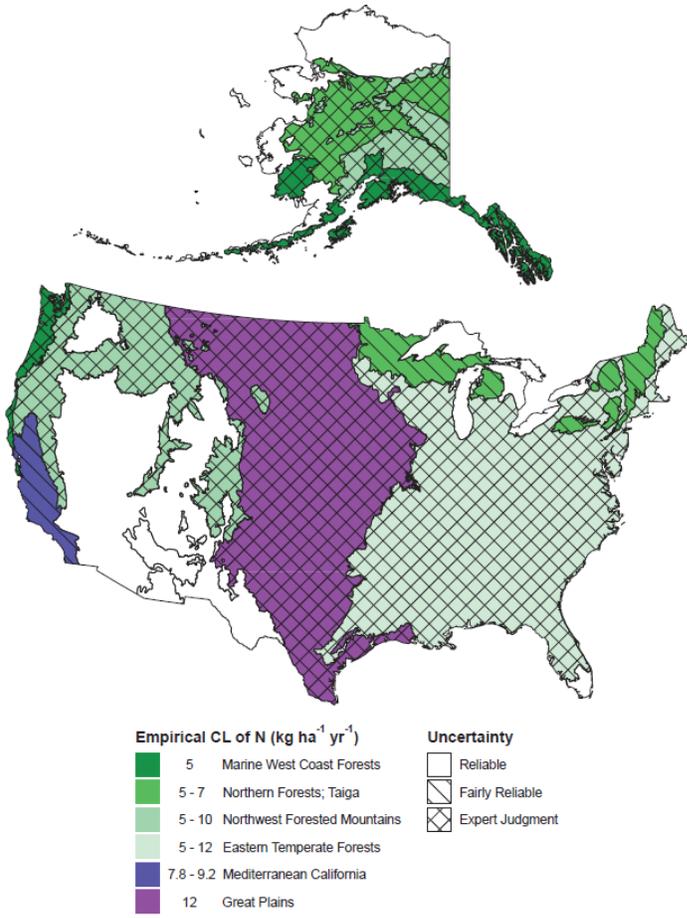


Figure 2

a.



b.

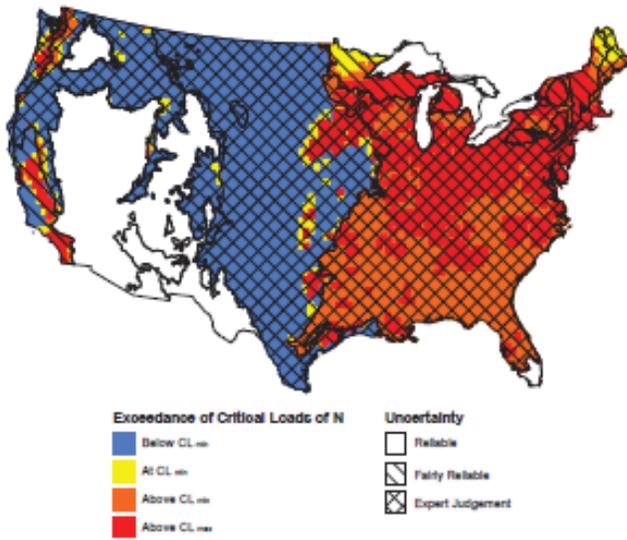
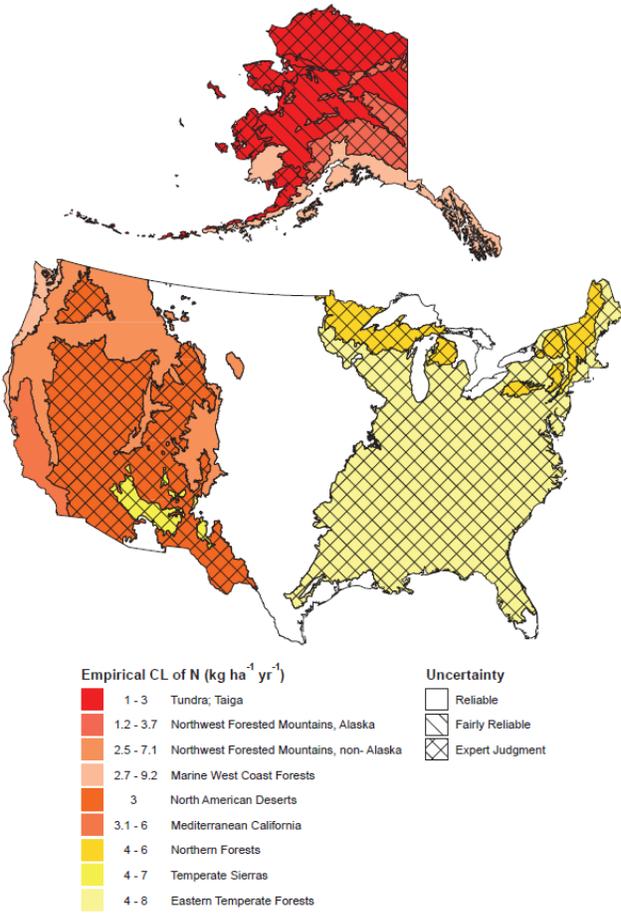


Figure 3

a.



b.

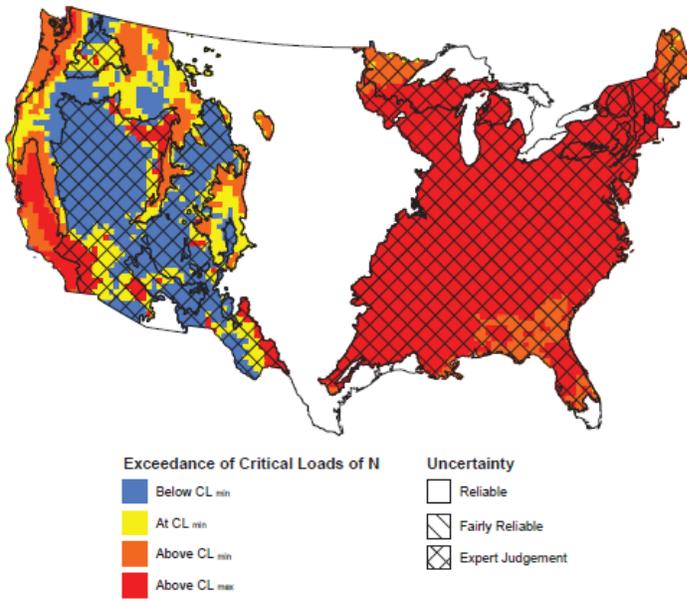
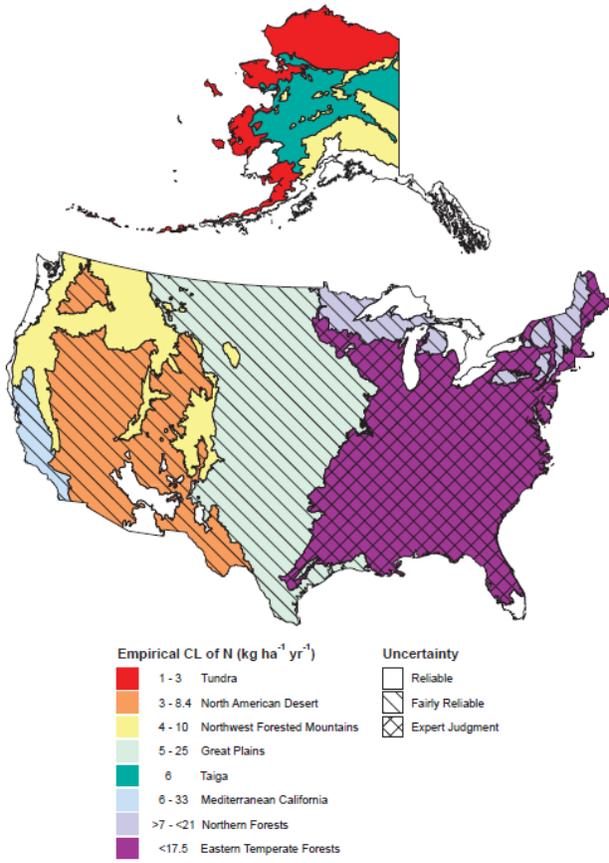


Figure 4

a.



b.

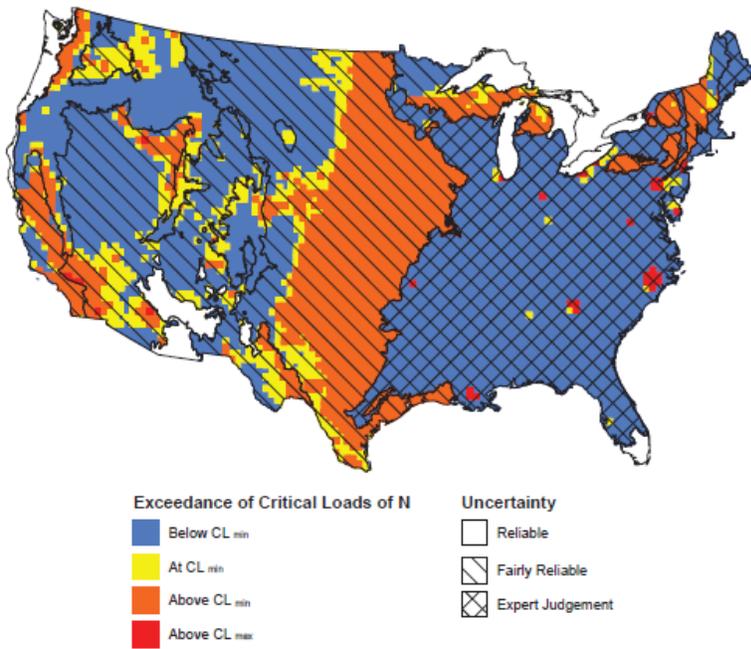
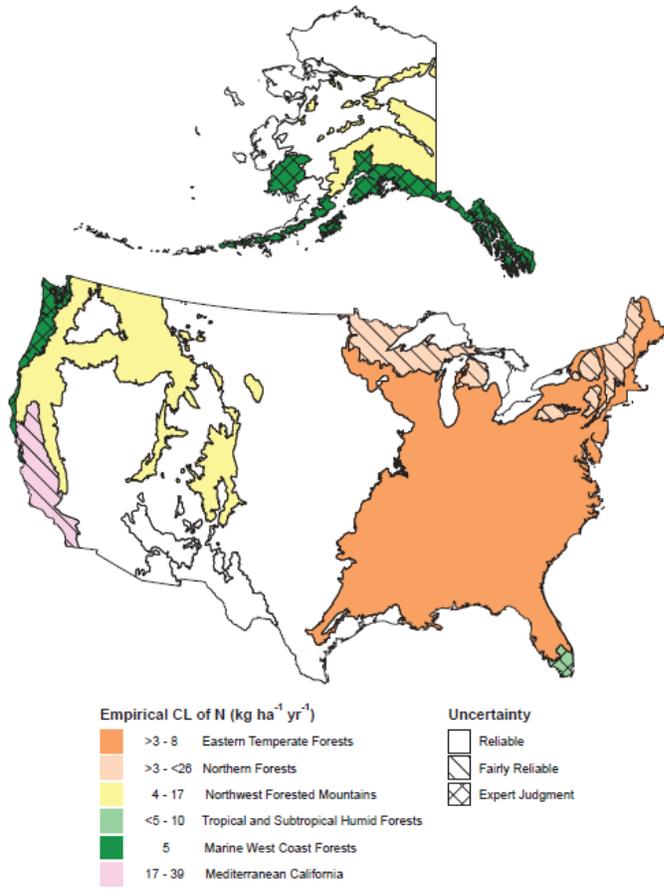


Figure 5

a.



b.

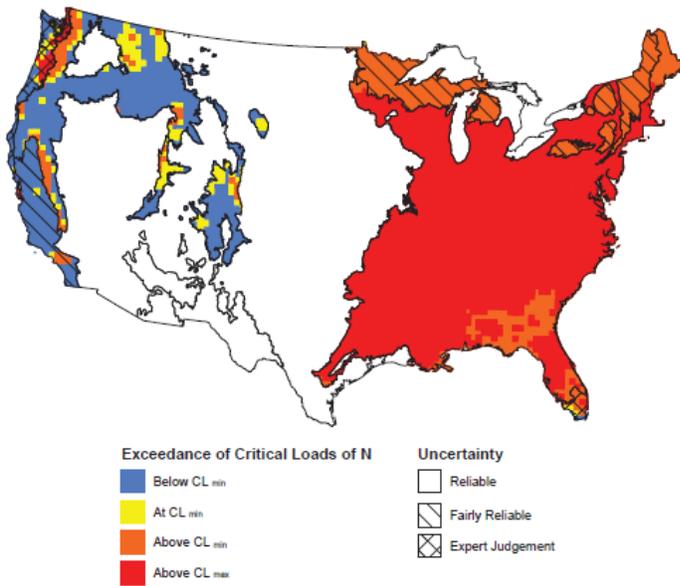
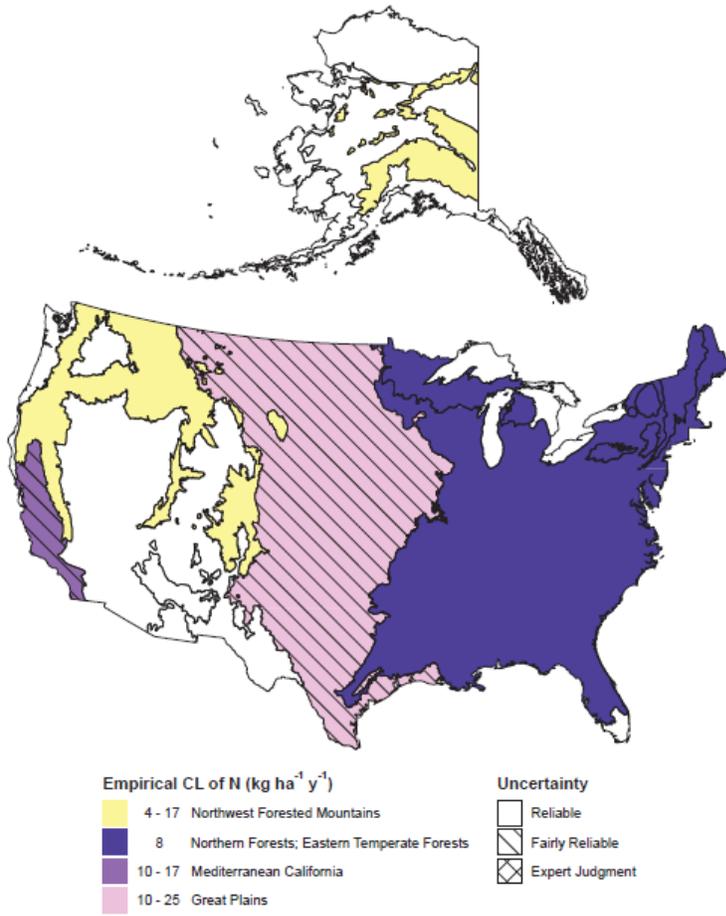


Figure 6

a.



b.

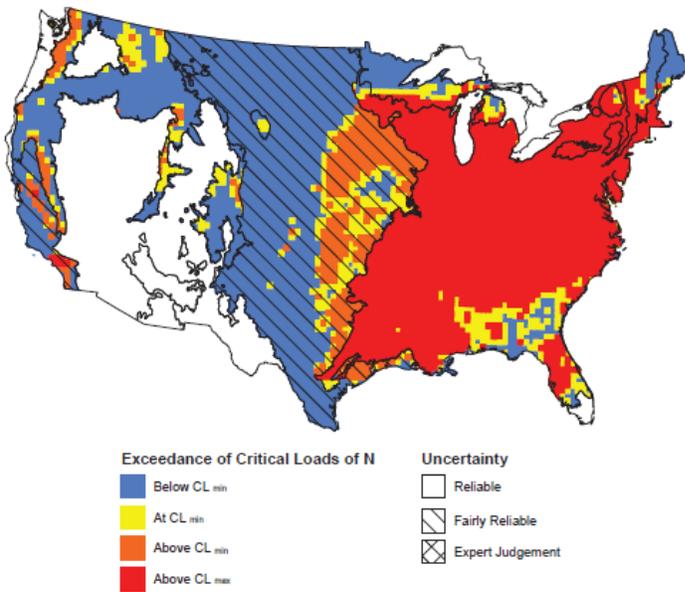


Figure 7

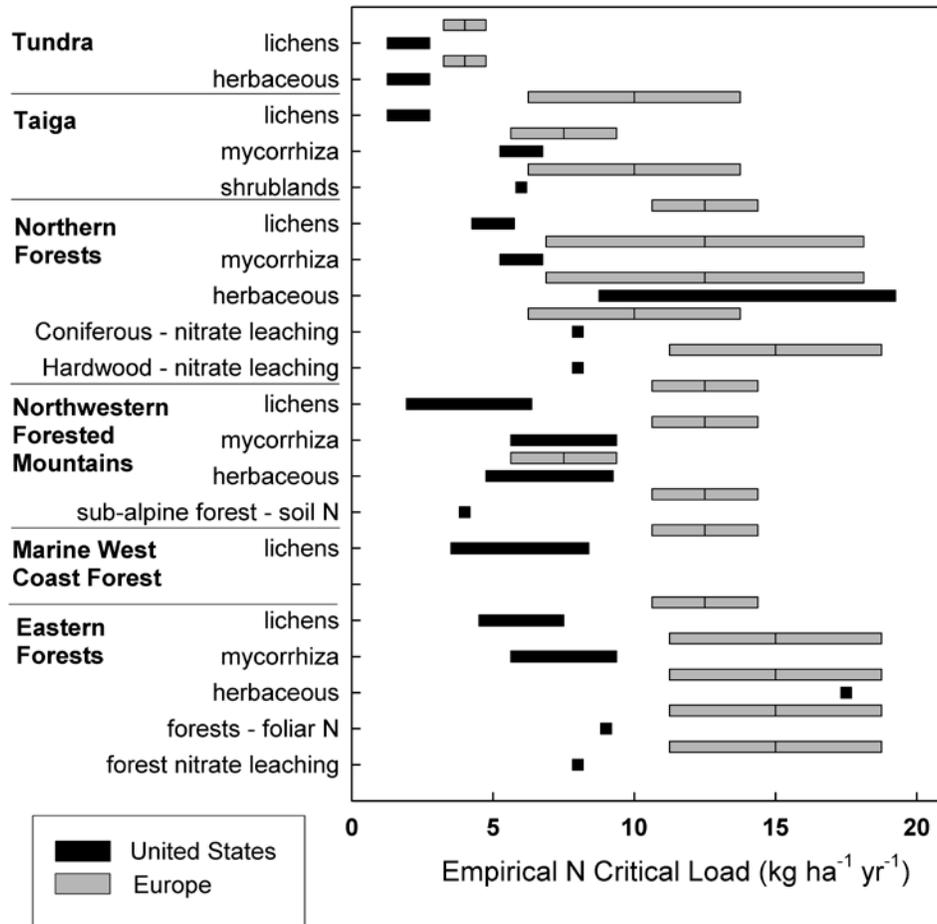


Figure 8