Clark, Christopher M., Jennifer Phelan, Prakash Doraiswamy, John Buckley, James C. Cajka, Robin L. Dennis, Jason Lynch, Christopher G. Nolte, and Tanya L. Spero. 2018. Atmospheric deposition and exceedances of critical loads from 1800−2025 for the conterminous United States. *Ecological Applications*

Appendix S1

Supplemental Methods:

Atmospheric deposition of N and S

Historical to recent deposition (1800−2011).—A summary of the historical and recent N and S deposition data is provided in the main text (Table 1). The CMAQ deposition was produced using the CMAQ model v5.0.2 with bidirectional NH_3 exchange predicted at 12-km horizontal grid resolution using emissions inventories and meteorology specific to each year. Bidirectional exchange of NH_3 refers to the capability of the model to simulate both the deposition of NH_3 from the atmosphere to the surface, as well as consider the (re)volatilization from the surface back to the atmosphere.

Different analytical approaches to estimate deposition yield different estimates even for the same location and time period due to differing spatial scales, atmospheric models, emission inventories, and so forth. Thus, to develop a seamless deposition record without a “jump” when switching from CAM to CMAQ, we merged these two data sets using a three-step process. First, the 1850−2000 CAM N and S deposition 200-km data were assigned to 12-km CMAQ grid cells, based on the location of CMAQ centroids. Second, the 150 years of CAM total N (wet + dry) and total S (wet + dry) deposition estimates were then scaled to the CMAQ total N and S deposition using a ratio of the 3-year deposition averages for the closest years (i.e., CMAQ [2002−2004]:CAM [1998−2000] = 0.94 and 0.80 for total N and total S, respectively). Thus, CAM estimates for total N deposition were comparable to the CMAQ data on average across the country, while S deposition was modestly overestimated with CAM. We used the CMAQ 2002−2011 estimates without modification, and re-scaled the CAM 1850−2000 estimates to meet CMAQ in 2002. Finally, a temporal weighting factor was applied to transition linearly from CAM deposition to CMAQ deposition, such that deposition in 1850 was 100% based on CAM, in 2002 was 100% based on CMAQ, and in between was a weighted average of the two data sets. This scaling approach resulted in a higher weighting of CAM in the first 75 years and higher weighting of CMAQ is the most recent 75 years. This approach was selected because of the differences in grid resolution scale between the two data sets and because we wanted to weight CMAQ more heavily in the analysis for the past few decades, as it is at a higher spatial resolution (12 km) and based on a much more thorough and U.S.-focused emissions inventory than EDGAR-HYDE which underpins the CAM estimates.

Future deposition (2025).— Future deposition estimates centered around four emissions scenarios as determined by different combinations of current (1 scenario) and hypothetical (3 scenarios) emissions control measures. The EPA SAB report (EPA, 2010) was reviewed to determine potential emission reduction scenarios to include in the 2025 CMAQ model runs. Through discussions with the EPA's Office of Air Quality Planning and Standards (OAQPS), it was determined that the technologically-feasible reductions in NOx emissions from stationary and mobile sources described by the SAB Report have already, to a large degree, been included in current rules under the Clean Air Act. CMAQ projects a decrease in emissions and deposition from 2011 to 2025 under these rules (i.e. the CAA reductions scenario). Therefore, the emission reductions scenarios for this effort would focus on additional hypothetical controls on the agricultural sector and include the following: (1) a 20% reductions in ammonia (NH3) emissions from synthetic fertilizer applications, (2) a 30% reductions in ammonia (NH3) emission from livestock management, (3) a combination of 1 and 2.

We also ran supplemental analyses on our results to see whether short term changes in climate would alter our results. For these, Community Earth System Model (CESM) fields from simulations conducted for the CMIP5 (Taylor et al. 2012) were dynamically downscaled to 36 × 36 km with the Weather Research and Forecasting Model using techniques described by Spero et al. (2016). Four 11-year periods were downscaled: the years 1995−2005 from the historical 20th-century experiment, and the years 2025−2035 following Representative Concentration Pathways (RCP) RCP 4.5, RCP 6.0, and RCP 8.5 (van Vuuren et al. 2011). The downscaled meteorology was then used with a 2030 emission inventory (Fann et al. 2015) to simulate air quality and deposition with CMAQ (Dionisio et al. 2017). The 2030 emissions projection was used for both the historical period and each of the future climate scenarios to isolate the effect of climate change on deposition from differences due to changing emissions. N and S deposition from each of the three RCP scenarios were compared against the averages from the historical period to determine the influence of climate change on deposition.

Critical Loads

Six CL types were extracted or derived from the NCLDv2.5 (Table 2), including CLs for the following impacts: (1) Terrestrial Acidification, (2) Aquatic Acidification, (3) changes in Forest-Tree Health, (4) elevated NO_3^- Leaching, (5) changes in herbaceous and shrub Plant Community Composition, and (6) changes in Lichen community composition (Table 2).

The CLs for Terrestrial Acidification were used without modification and represent the amount of N and/or S deposition that is expected to induce soil acidification in forested ecosystems

(McNulty et al. 2007, McNulty and Boggs 2010). Aquatic Acidification CLs were averaged if there were multiple estimated for the same water body (16% of water bodies).

For Aquatic Acidification CLs, we used NCLDv2.5 values without modification except where there were multiple CL estimates for the same water body (e.g. different methods, times, inlet versus outlets of a lake, etc.; roughly 16% of water bodies had multiple estimates). In these cases, to prevent double counting we averaged CLs within the water body prior to use.

Forest-Tree Health CLs were modified from the forest ecosystem CLs in Pardo et al. (2011a, b), which include a variety of CLs not necessarily detrimental to the forest, including increased growth and increased foliar N concentrations (Pardo et al. 2011a, Pardo et al. 2011b) (Table S2). Therefore, we restricted our Forest-Tree Health CLs to measures of *compromised* tree health reported in Pardo et al. (2011a, b), including reduced tree growth and survival, reduced fine root biomass, crown thinning, chlorotic foliage, and compromised forest sustainability. We acknowledge that positive initial effects can induce negative secondary effects (e.g., increased growth for one species can cascade and lead to decreased growth for another, that increased foliar N could induce elevated pest damages, etc.), but the quantitative levels at which these cascades occur have not been documented in most cases. We then constrained Forest-Tree Health CLs to land covers that were forested based on the National Land Cover Database (Homer et al. 2015).

Critical loads for NO_3^- leaching were derived from the Pardo et al. (2011a, b) forested, herbaceous, and shrubland ecosystem, and were restricted to measures of NO_3^- leaching, NO_3^- loading into surface waters, N leaching in soil profile, and elevated stream water NO_3^- concentrations (Table S3). We constrained these to forest, herbaceous, and shrubland land covers based on the 2011 NLCD.

Critical loads for plant community composition were based on changes in the composition of plants (herbs, shrubs, and/or tree seedlings) from Pardo et al. (2011 a, b) and were also constrained to communities in forest, herbaceous, and shrubland land covers using the 2011 NLCD (Table S4).

The CLs of N for lichens within the NCLDv2.5 are from Geiser et al. (2010) and were applied without modification to most ecoregions at the 4- × 4-km grid size. However, due to concerns regarding the accuracy of this CL in two Level 1 ecoregions in the Pacific Northwest (i.e., Northwestern Forested Mountains and Marine West Coast Forests), the CLs for these two ecoregions were replaced with CL values from Root et al. (2015) (Geiser et al. 2010, Root et al. 2015).

Note that the Pardo et al. (2011a, b) CLs are generally based on findings from a few representative sites and studies within each ecoregion and then extrapolated to the entire Level I ecoregion where the studies occurred (Fig. S1). The degree to which these CLs accurately represent vulnerability across these large regions is currently unknown.

All CLs were mapped to a 12-km × 12-km grid size to match the CMAQ grid (Table 1). We chose this to enable the calculation of CL exceedances on a common scale so that exceedance maps among CL types were directly comparable. Because different CLs have different resolutions and extents, mapping to the CMAQ grid resulted in 0−156 CL values for each 12-km × 12-km grid cell. Within each grid cell and separately for each CL type, we calculated the minimum, 10th, and 50th percentiles, to more accurately describe the variation in sensitivity within and among CL types. For CLs of Aquatic Acidification (point values), Terrestrial Acidification (1- × 1-km grid), and Lichen (4- × 4-km grid), a minimum of five values within a grid cell were required to calculate the statistics. This minimum of five CL values was selected to balance contrasting objectives—to provide enough CL estimates to calculate defensible summary statistics, and to maximize the number of 12-km grid cells that had multiple statistics for each CL type (i.e., minimum, 10th, and 50th percentile). This requirement was almost always met for the Lichen and Terrestrial Acidification CL types because of their fine resolution (Table 2). The Aquatic Acidification CL type met this requirement for 9% of grid cells in which there were any Aquatic Acidification CLs at all. If there were fewer than five values for a CL type in a grid cell, only the lowest CL value was represented and was designated as the minimum CL.

The three empirical CLs of N derived from Pardo et al. (2011a, b; Forest-Tree Health, Plant Community Composition, NO_3^- Leaching) were represented by a single value or a range for Level I ecoregions (Table 3). These CLs were assigned to 12- × 12-km CMAQ grid cells based on the ecoregion that occupied the largest area of the CMAQ cells. Empirical CLs are associated with specific land covers (e.g., forests, shrublands) based on the location of the original CL study (Pardo et al., 2011a, b). CL Exceedances were assigned within a 12-km grid cell if there was any of the appropriate land cover (as defined by the 30-m NLCD) for that CL within that grid cell, and the CL was exceeded. We used this conservative approach to convey a potential exceedance of that CL somewhere in that 12 × 12-km grid cell, and to meet the objective of harmonizing all maps to the 12 × 12-km grid size. Separately for each CL type, we calculated the minimum, 10th, and 50th percentile CLs for each grid cell, assuming that the range reported in Pardo et al. (2011 a, b) accurately represented the range of the CL. When only a single CL value was available, we assumed the reported CL represented the minimum (Pardo et al. 2011 a, b). In situations where the CL was reported as "greater than" a certain amount of N, we used the reported number as the minimum and did not estimate percentiles. Critical loads that were reported as "less than" an amount of N deposition were not included in this study, as it was not possible to determine the lower bound of the CLs. Only one CL was eliminated based on this criterion: Plant Community Composition in the Eastern Temperature Forest ecoregion (<17.5 kg ha^{-1} yr^{-1}).

Table S1. Summary of national results of minimum, 10th, and 50th percentile (see DataS1 file).

Table S2: Description of the Forest-Tree Health Empirical N CLs and how they were subset from the NCLDv2.5 (Lynch et al. 2013) for use in this study.

LEVEL I ECOREGION	**FOREST-TREE HEALTH END POINT**			
	RESPONSE METRIC	**STUDY LAND COVER TYPE (SOURCE)**	**CRITICAL LOAD FOR NITROGEN DEPOSITION (DETERMINED BY STUDY) (kg N/ha/yr)**	**RANGE OF CRITICAL LOAD FOR NITROGEN DEPOSITION (FOR PROJECT) (kg N/ha/yr)**
Northern Forests	decreased growth and survivorship	Northeastern U.S. forests (Thomas et al., 2010)	>3	**3 - 26**
	decreased growth and increased mortality	Montane spruce-fir forests (McNulty et al., 2005)	>10 - <26	
Northwestern Forested Mountains	reduced fine-root biomass	Mixed-conifer forest (Fenn et al., 2008)	17	**17**
Marine West Coast Forests	crown thinning and chlorotic foliage	Coniferous forest (Whytemare et al., 1997)	5	**5**
Eastern Temperature Forests	decreased growth and survivorship	Eastern forests (Thomas et al., 2010)	>3	**3**
Mediterranean California	reduced fine-root biomass	San Bernandino Mountains; Ponderosa Pine (Fenn et al., 2008; Grulke et al., 1998)	17	**17 - 39**
	forest tree sustainability (increased sensitivity to ozone, drought, and pests)	San Bernandino Mountains (Grulke and Balduman, 1999; Grulke et al., 1998, 2009;	39	

		Jones et al., 2004)		
Tropical Humid Forests	na			**na**
Great Plains	na			**na**
North American Desert	na			**na**

Table S3: Description of the Nitrate Leaching Empirical N CLs and how they were subset from the NCLDv2.5 (Lynch et al. 2013) for use in this study.

LEVEL I ECOREGION	**NITRATE LEACHING END POINT**				
	RESPONSE METRIC	**STUDY LAND COVER TYPE (SOURCE)**	**NLCD COVER**	**CRITICAL LOAD FOR NITROGEN DEPOSITION (DETERMINED BY STUDY) (kg N/ha/yr)**	**RANGE OF CRITICAL LOAD FOR NITROGEN DEPOSITION (FOR PROJECT, kg N/ha/yr)**
Northern Forests	increased surface water NO_3^- leaching	Northern hardwood and coniferous forests (Aber et al., 2003)	Forest	8	**8**
Northwestern Forested Mountains	increases in N leaching below O layer	Subalpine forest (Rueth and Baron, 2002)	Forest	4	**4 - 17**
	NO_3^- leaching	Mixed-conifer forest (Fenn et al., 2008)		17	
Marine West Coast Forests	na				**na**
Eastern Temperature Forests	increased surface water NO_3^- leaching	354 Upland forest catchments (Aber et al., 2003)	Forest	8	**8**
Mediterranean California	streamwater NO_3^- concentration	San Bernardino Mountains and southern Sierra Nevada	Forest	17	**10 - 17**

		Range (Fenn et al., 2008; 2010)			
		Chaparral, oak woodlands, Central Valley (Sequoia National Park) (Fenn and Poth, 1999; Fenn et al., 2003a,b,c; 2010; 2011; Meixner and Fenn, 2004)	Forest	10 - 14	
	streamwater NO_3^- concentration	Chaparral, oak woodlands, Central Valley (Sequoia National Park) (Fenn and Poth, 1999; Fenn et al., 2003a,b,c; 2010; 2011; Meixner and Fenn, 2004)	Shrubland and Herbaceous	10 - 14	**10 - 14**
Tropical Humid Forests	NO_3^- leaching	N-poor tropical forests (expert judgement)	Forest	5 - 10	**5 - 10**
	NO_3^- leaching	N-rich tropical forests	Forest		

		(expert judgement)			
Great Plains	NO_3^- leaching	Mixed-grass prairie	Herbaceous	10 - 25	**10 - 25**
North American Desert	na				**na**

Table S4: Description of the Plant Community Composition Empirical N CLs and how they were subset from the NCLDv2.5 (Lynch et al. 2013) for use in this study.

LEVEL I ECOREGION	**PLANT COMMUNITY COMPOSITION END POINT**				
	RESPONSE METRIC	**STUDY LAND COVER TYPE (SOURCE)**	**NLCD COVER**	**CRITICAL LOAD FOR NITROGEN DEPOSITION - DETERMINED BY STUDY (kg N/ha/yr)**	**RANGE OF CRITICAL LOAD FOR NITROGEN DEPOSITION - FOR PROJECT (kg N/ha/yr)**
Northern Forests	alteration of herbaceous understory	Adirondack northern hardwood forest (Hurd et al., 1998)	Forest	> 7 and < 21	**7 - 21**
Northwestern Forested Mountains	plant species composition	Alpine grasslands/meadows (Bowman et al., 2006)	Herbaceous	4 - 10	**4 - 10**
Marine West Coast	change in composition of understory	South Central Alaska coniferous forest (Lilleskov et al., 2001)	Forest	5	**5**
Eastern Temperature Forests	increase in nitrophilic species, declines in species richness	Eastern hardwood forest (Fernow Experimental Forest) (Gilliam, 2006, 2007; Gilliam et al., 2006)	Forest	<17.5	**na**
Mediterranean California	annual grass invasion, replacing native herbs	Serpentine grassland (Weiss, 1999; Fenn et al., 2010)	Herbaceous	6	**6 - 10**

	changes in invasive grass cover, native forb richness	Coastal sage scrub (Egerton-Warburton et al., 2001; Tonnesen et al., 2007; Fenn et al., 2010; 2011)	Herbaceous	7.8 - 10	
	changes in invasive grass cover, native forb richness	Coastal sage scrub (Egerton-Warburton et al., 2001; Tonnesen et al., 2007; Fenn et al., 2010; 2011)	Shrubland	7.8 - 10	**7.8 - 10**
	changes in diversity of understory	San Bernardino Mountains (mixed-conifer forest) (Allen et al., 2007)	Forest	24 - 33	**24 - 33**
Tropical Humid Forests	changes in community composition (forest - altered spp. composition, decreased spp richness, moss cover and native seedling abundance)	N-Poor Tropical Forests; Hawaiian lower montane forest (Ostertag and Verville, 2002)	Forest	5 - 10	**5 - 10**
Great Plains	plant community shifts	Tallgrass prairie, (Tilman, 1987; 1993; Wedin and Tilman, 1996; Clark and Tilman, 2008; Clark et al., 2009)	Herbaceous	5 - 15	**5 - 25**
	plant community shifts	Mixed-grass prairie (Clark et al., 2003; 2005 ; Jorgensen et al., 2005)	Herbaceous	10 - 25	

	plant community shifts	Short-grass prairie (inferred from mixed-grass prairie) (Epstein et al., 2001; Barrett and Burke, 2002)	Herbaceous	10 - 25	
North American Desert	increased biomass of invasive grasses; decrease of native forbs	Shrubland, woodland, and desert grassland (Joshua Tree NP, Mojave Desert) (Allen et al., 2009; Rao et al., 2010)	Herbaceous and Shrubland and Forest	3 - 8.4	**3 - 8.4**

Fig. S1. Map of level 1 ecoregions for the conterminous United States.

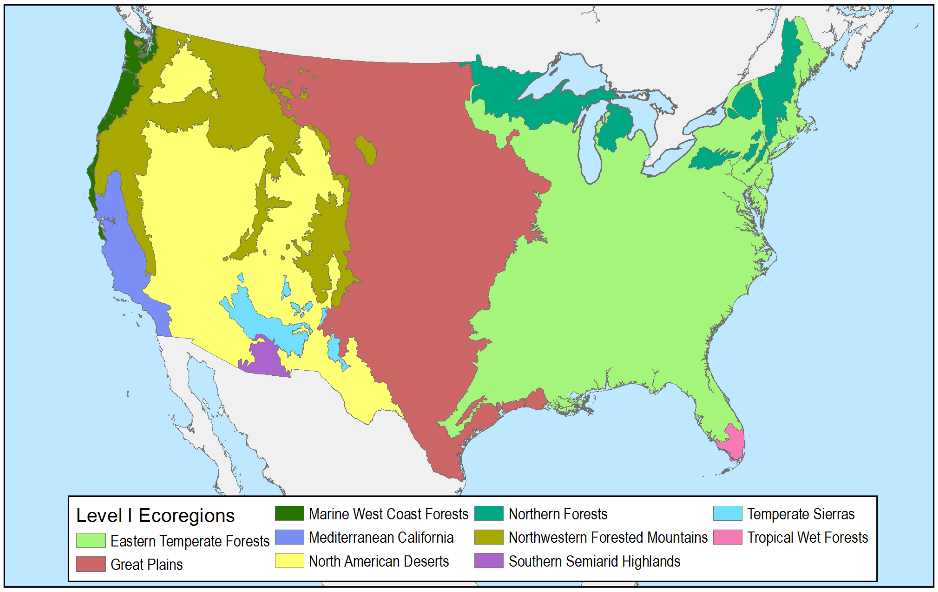


Fig. S2. Effects from climate change on N and S deposition. Shown below are the absolute changes in deposition (eq ha^{-1} yr^{-1} and kg ha^{-1} yr^{-1}) from 2030 (11-year average of 2025−2035) minus 2000 (11-year average of 1995−2005) for RCP 4.5 and 8.5 (RCP 6.0 not shown) using a constant emission inventory (2030). The 5th to 95th percentiles for N deposition were −0.3 to 0.2 kg ha^{-1} yr^{-1} (RCP 4.5) and −0.6 to 0.3 kg ha^{-1} yr^{-1} (RCP 8.5), and for S deposition were −0.2 to 0.1 kg ha^{-1} yr^{-1} (RCP 4.5) and −0.4 to 0.2 kg ha^{-1} yr^{-1} (RCP 8.5).

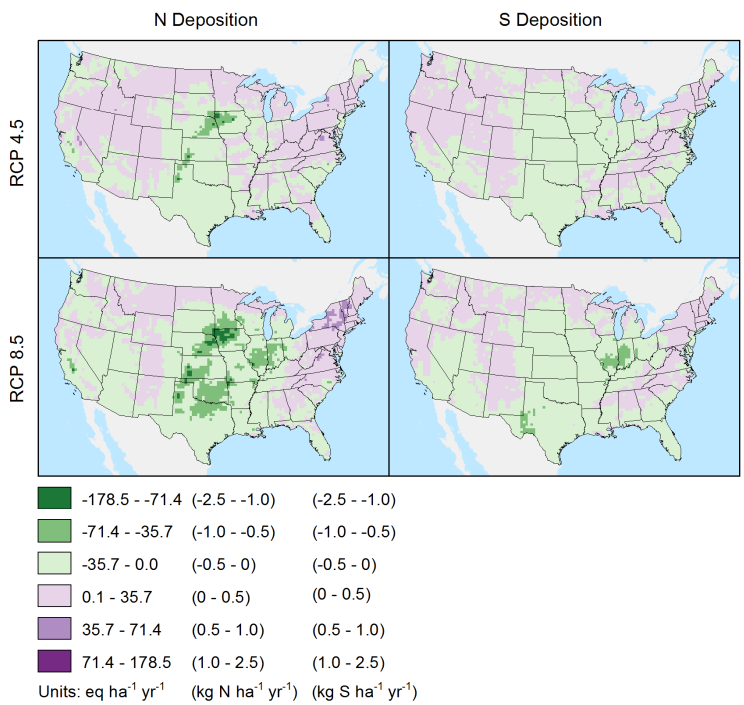


Fig. S3. Historical deposition estimates using CAM from Lamarque et al. (2010) compared with high resolution historical estimates from the SAMI study (left, ASTRAP, from Shannon [2009]) for two southeastern study sites (PR-Piney River site of the Shenandoah National Park, CC-Cosby Creek site of the Great Smokey Mountains National Park) and with one site in the Northeast [Hubbard Brook, NH; GT-Driscoll, from Gbondo-Tugbawa et. al. (2001]).

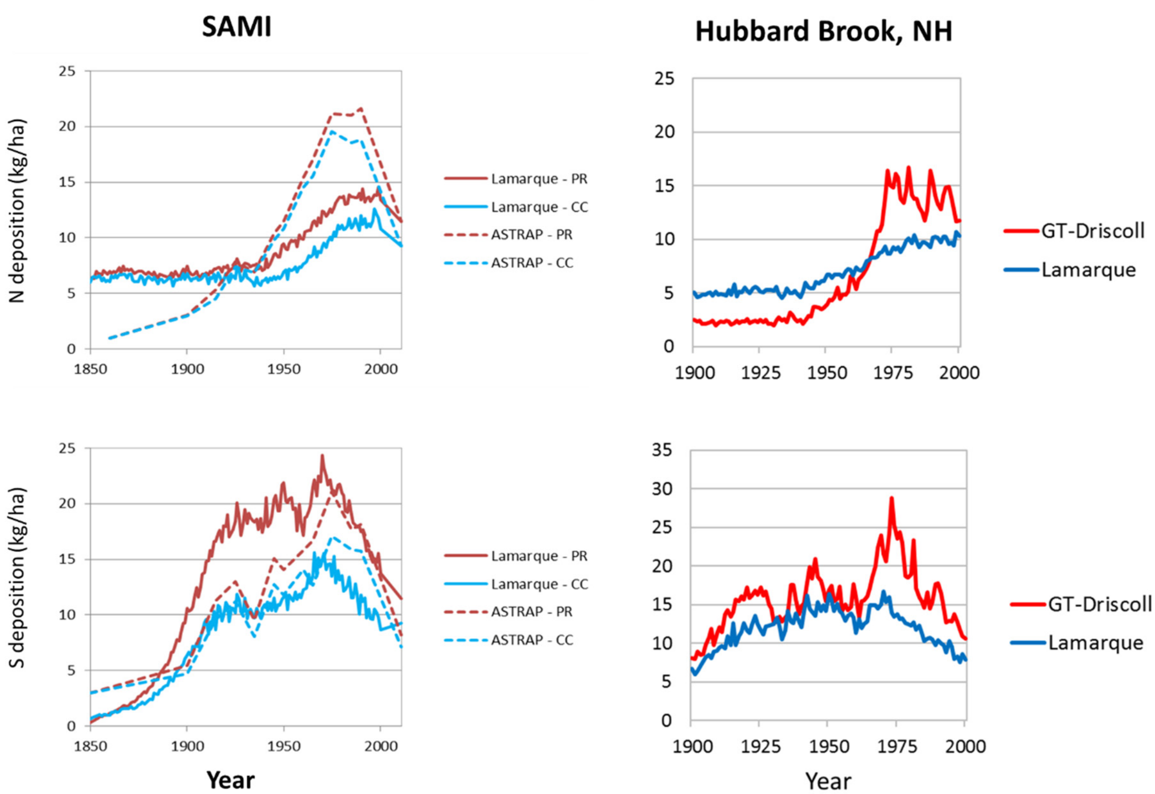
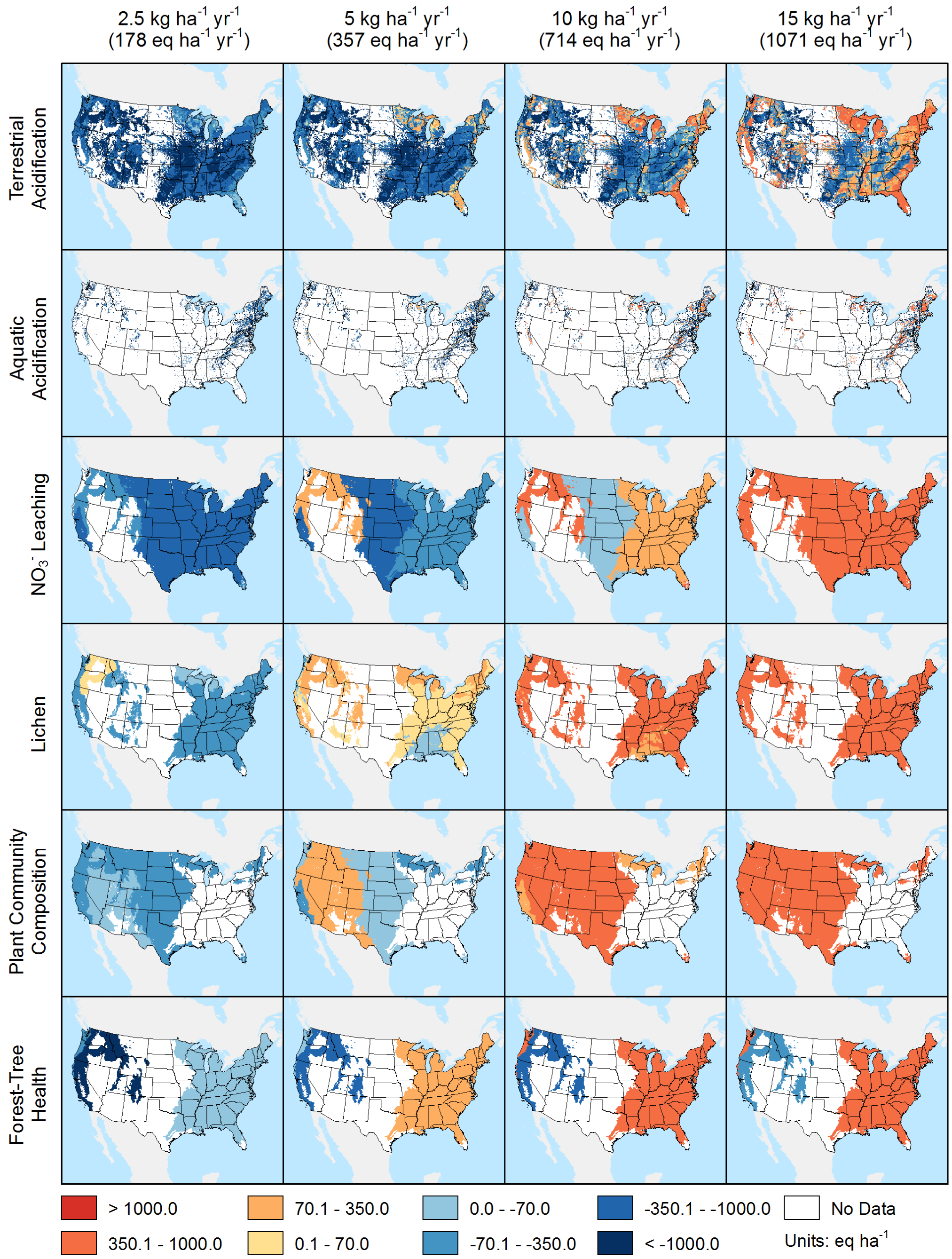


Fig. S4. Minimum CL exceedances for the six CLs based on the following fixed N-deposition levels: 2.5, 5, 10, and 15 kg N ha^{-1} yr^{-1}.



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