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Atmospheric deposition and exceedances of critical loads from 1800–2025 for the conterminous United States

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Abstract

Atmospheric deposition of nitrogen (N) and sulfur (S) from human activity has increased dramatically over pre-industrial levels, with many potential impacts on terrestrial and aquatic ecosystems. Quantitative thresholds, termed “critical loads” (CLs), have been developed to estimate the deposition rate above which damage occurs to ecological endpoints of concern. However, there remains no comprehensive comparison of when, where, and over what time periods endpoints have been exceeded. We addressed this knowledge gap by combining several robust data sources for historical and contemporary deposition (1800–2011), and overlaying these on six CL types from the National Critical Loads Database (NCLD) (Terrestrial Acidification, Aquatic Acidification, Lichen, Nitrate Leaching, Plant Community Composition, and Forest-Tree Health) to examine exceedances from 1800–2011. We expressed CLs as the minimum, 10th, and 50th percentiles within grid cells, and found that all minimum CLs in the East have been exceeded for decades, beginning in the mid to late 20th century. The area exceeded peaked in the 1970s and 1980s, covering 300000 to 3 million km^2 for individual CL types, with total area exceeded for one or more CLs of 5.8 million km^2 (~70% of the conterminous United States). Since then, deposition levels have dropped, especially for S, with modest reductions by 2011 in total area exceeded to 5.2 million km^2. Other percentiles were not consistently reported for CLs. We also examined near-term changes in deposition and exceedances in 2025 under current air quality regulations, and under various additional measures to reduce N emissions from agriculture. Projecting forward, we found current regulations were anticipated to reduce exceedance of one or more CLs from

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5.3 million km^2 in 2011 to 4.8 million km^2 in 2025. Furthermore, none of the additional N management scenarios impacted areal exceedances, although exceedance magnitudes declined. In total, it is clear that many CLs have been exceeded for decades, and are likely to remain so in the short term under current policies. Additionally, we suggest many areas for improvement to enhance our understanding of deposition and enable decision makers to make informed policy decisions regarding these environmental stressors.

Keywords

critical loads; atmospheric deposition; nitrogen; sulfur; exceedances; air quality

INTRODUCTION

Human activity has increased the deposition of reactive nitrogen (N) and sulfur (S) dramatically over pre-industrial levels for much of the developed world (Vitousek et al. 1997, Galloway et al. 2004). These stressors can have a variety of negative impacts on terrestrial and aquatic ecosystems through acidification and eutrophication (Bobbink et al. 2010, Porter et al. 2013, Sullivan et al. 2013). Both N and S are acidifying agents in the environment, and in terrestrial systems can lead to increased cation leaching (Lawrence et al. 2015); mobilization of soil aluminum, which can be phytotoxic (Driscoll et al. 2003, Horswill et al. 2008); cation imbalances in plant photosynthetic tissue (Schaberg et al. 2002); increased plant mortality and reduced recruitment of trees (Duarte et al. 2013, Sullivan et al. 2013); and changes in the biodiversity of herbaceous species (Stevens et al. 2010, Simkin et al. 2016) and lichens (Geiser et al. 2010, Root et al. 2015). N and S deposition result in increased inputs of acidifying compounds to downstream aquatic systems that alter surface water chemistry, negatively impacting fish species and diversity (Battarbee et al. 2014, Zhou et al. 2015). N deposition also has an additional set of impacts because nitrogen is a limiting nutrient in most terrestrial and aquatic ecosystems (Vitousek and Howarth 1991, Howarth 2008). As such, N enrichment of terrestrial ecosystems can lead to eutrophication, including increased primary production, elevated nitrate (NO_3^-) leaching, and shifts in the competitive balance of species, all of which can lead to reductions in herbaceous biodiversity (Simkin et al. 2016) and changes in forest tree composition (Thomas et al. 2010). Elevated leaching of soil NO_3^- can result in increased NO_3^- concentrations along with other nutrient inputs in nearby water bodies, inducing shifts in the communities of aquatic microfauna and flora, and contribute to increased algal blooms, reduced oxygen levels in the water column, and ultimately, fish kills (Nydick et al. 2004, Baron et al. 2011, Saros et al. 2011).

Over the past few decades, a body of knowledge has emerged on key thresholds at which impacts from N and/or S deposition begin to occur for various environmental receptors (Blett et al. 2014). These thresholds are termed “critical loads” (CLs) and are formally defined as “a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt 1988, Bobbink et al. 2010). When deposition is above a CL (hereafter termed an “exceedance”), negative effects are anticipated to occur. However, it is important to note that exceedances are a form of vulnerability, not impact, and measure the amount of deposition above a known threshold, not the amount of impact from that exceedance. Nevertheless, they are a useful tool to help identify areas and endpoints of potential concern. Because there can be multiple sensitive biological receptors (e.g., trees, lichens, and herbaceous plants) in the same forest, watershed, or other geographical unit, one location or area can have multiple CLs that differ in value and represent varying levels of sensitivity to deposition. Here we use the term CL type as a generic term for the identity of the CL that includes both biological receptors (e.g., lichen communities) and processes that are affected by deposition (e.g., soil acidification).

Prior studies have suggested that deposition has exceeded some CL types for many years (Clark and Tilman 2008, Pardo et al. 2011a), but it is uncertain which CL types have been most affected and where, and over what historical period, these vulnerabilities have existed. In recent decades, total N and S deposition has declined in the United States from historical peaks of acidic deposition in the 1970s and 1980s, especially in the East (Burns 2011). For example, over the past 20 years, total deposition of S and inorganic N decreased by an average of 66–69% and 19–36%, respectively, for different parts of the eastern United States (Burns 2011). However, inorganic wet N deposition levels are steady or increasing in the West and Midwest (Houlton et al. 2012), possibly due to increasing contributions from reduced N (Li et al. 2016). With these temporally and spatially changing patterns of deposition, it is unclear whether current deposition has dropped below individual CLs, and if so, which CLs and for what regions of the country. Such information is critical to supporting targeted restoration efforts, as well as for reviewing national policy on air quality status and trends.

In addition to the uncertainties in the historical patterns of CL exceedance, it is unclear how future exceedances might be influenced by changes in emissions over time by policies that impact N and S emissions from energy, transportation, and agricultural operations. Management strategies can affect deposition by influencing emission rates of N and S through changes in mobile and stationary source emission reduction technologies as well as improved management of key agricultural activities, such as livestock operations and synthetic fertilizer applications, among others (EPA 2010).

This study has three objectives: (1) to develop and test a method for converting CLs to a common spatial unit to facilitate direct comparison; (2) to better understand which CLs have been exceeded, where, and over what historical time intervals for the conterminous United States (1800–2011); and (3) to explore how various potential emission scenarios may affect these exceedances in the near future (2025). We address these objectives by constructing a historical deposition record from 1800–2011 from various data sources and developing several potential future deposition projections based on different emission scenarios. We then compare these deposition levels to CLs for six CL types (defined in the next section) to better understand which CLs (and how many) have been exceeded, where, and over what time period historically and in the near future.

METHODS

Atmospheric deposition of N and S

Historical to recent deposition (1800–2011).——Historical and recent N and S deposition were derived from two data sets (Table 1). Deposition data for the historical period (1850–2000) came from contributions to the fifth phase of the Climate Model Intercomparison Program (CMIP5) in support of the Intergovernmental Panel on Climate Change Fifth Assessment report (IPCC AR5, Lamarque et al. 2010). The historical deposition data were produced using the RETRO and EDGAR-HYDE emission data sets and Community Atmosphere Model (CAM) v3.5 at a 200-km resolution. Deposition data for the current period (2002–2011) came from the Community Multiscale Air Quality (CMAQ) model with bidirectional exchange of ammonia (NH_3) (Bash et al. 2013). We adopted a baseline in 1800 of 0.4 kg N ha^{-1} yr^{-1} and 0.1 kg S ha^{-1} yr^{-1} because there was already moderate N deposition in 1850 in the southeast due to biomass burning associated with agricultural land clearing (5–20 kg N ha^{-1} yr^{-1}, Lamarque et al. 2010), and because we wanted to start our analysis from a baseline prior to the Industrial Revolution. These values were derived from the lowest reported deposition estimates for 1850 (Lamarque et al. 2010) and background or pre-industrial deposition estimates from other studies (Holland et al. 1999, Galloway et al. 2004, Baron 2006). Deposition amounts between 1800 and 1850, the first year of CAM deposition data, were not estimated.

To develop a deposition record from 1850–2011 to transition from CAM to CMAQ, we followed a three-step process: (1) we re-projected CAM data from the original projection and 200- × 200-km grid to the CMAQ projection and 12- × 12-km grid, (2) we calculated the ratio of CMAQ:CAM deposition for N and S separately within each 12-km grid cell for the closest overlapping years (i.e., CMAQ [2002–2004]:CAM [1998–2000]), and (3) we used this ratio to develop a temporal weighting factor 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. In summary, we used the CMAQ 2002–2011 estimates without modification, and re-scaled the CAM 1850–2000 estimates to meet CMAQ in 2002. More details on the methods for this and other procedures are provided in Supplemental Methods of the online Appendix S1.

The merged 1850–2011 annual N and S deposition data sets representing historical and recent deposition were converted to fifteen 10-year averages represented by the decadal midpoints (1855…1995), and one 9-year average (2002–2010, midpoint of 2006). We present maps of deposition and exceedance for a subset of these years to illustrate the trends through time (i.e., 1800, 1855, 1905, 1955, 1975, 2006). This captures the full trajectory of low deposition and exceedance initially, increasing in the early to middle 20th century with industrialization, peaking in the 1970s and 1980s, and declining to the present due to reductions in air emissions under the Clean Air Act (CAA) and CAA Amendments of 1990. Decadal averages are considered more “robust” estimates of historic deposition for the CAM data, as annual meteorological conditions are much more difficult to represent than decadal patterns (Lamarque et al. 2010). We also calculated a single year of N and S deposition for 2011 because 2011 meteorology was used in the 2025 future deposition estimates.

Future deposition (2025).——We adopted four different future emissions scenarios (Table 1), which along with 2011 meteorology, were used with CMAQ model v5.0.2 with bidirectional exchange of NH_3 (Bash et al. 2013) to develop deposition estimates for the conterminous United States in the year 2025. The four emission scenario projections included: (1) the estimated 2025 emission that incorporates CAA-driven mobile and stationary source controls (EPA 2014) (hereafter referred to as “CAA reductions scenario”), (2) the CAA reductions scenario plus a 20% reduction in NH_3 emissions from synthetic fertilizer applications (hereafter referred to as “CAA + synthetic fertilizer reductions scenario”), (3) a 30% reduction in NH_3 emissions from livestock management (hereafter referred to as “CAA + livestock management reductions”), or (4) a combination of 2 and 3 (hereafter referred to as “CAA + synthetic fertilizer + livestock management reductions”). The CAA scenario includes “on-the-book” regulations as of 2014 that are currently scheduled to be active until 2025, which includes emission controls from the following key programs: Mercury and Air Toxics Standards (MATS), the Clean Air Interstate Rule (CAIR), the Cross-State Air Pollution Rule (CSAPR), mobile source standards including the light-duty vehicle Tier 2 rule, the Tier 3 motor vehicle emissions and fuel standards rule, the heavy-duty diesel rule, the clean air non-road diesel rule, and the locomotive and marine engine rules (EPA 2014). Technologically feasible reductions in agriculture-based emissions were based on recommendations from the U.S. Environmental Protection Agency (EPA) Science Advisory Board (SAB) (EPA 2010) and served as the percentage reductions in emissions for scenarios 2, 3, and 4. Reduced emissions from livestock management were projected to result mostly from potential improvements in manure lagoon management in poultry and swine operations, and reduced emissions from synthetic fertilizer were based on lower fertilization rates and more widespread adoption of best management practices to match the timing and amount of fertilizer added to when plants need it most. Further details are available in the SAB Report (EPA 2010). We produced the deposition estimates for these scenarios using a modified version of CMAQ model v5.0.2. See RTI (2017) for a detailed summary of the technologically feasible reductions in agriculture-based N emissions outlined by the SAB and for more information on the four 2025 deposition data sets.

Critical loads

The current state of knowledge of CLs for the United States is captured in the National Critical Loads Database (NCLD) hosted by the National Atmospheric Deposition Program (NADP) (Lynch et al. 2013). The NCLD consists of modeled and empirical CLs determined through independent research efforts of the U.S. Forest Service (USFS), National Park Service (NPS), U.S. Geological Survey (USGS), EPA, universities, and consultants compiled into a spatially explicit data set for the conterminous United States.

Six CL types were extracted or derived from the NCLD (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. Capitalized terms are the shorthand names for the CL types used hereafter. CL types (1) and (2) represent CLs of acidifying N and/or S deposition, while CL types (3) to (6) were based on empirical studies relating N deposition with plant and/or soil responses and represent CLs of N deposition.

In most cases the CLs in the NCLD were used without modification, but some modifications were made that are described fully in the online Appendix S1. In short, (1) for Aquatic Acidification we averaged CLs for water bodies with multiple CLs; (2) for Terrestrial Acidification and Empirical CLs, we restricted CL application to the appropriate land cover type for which they were intended (e.g., Forest-Tree Health CL for the Eastern Temperate Forest ecoregion was restricted for forested areas, etc.) using the 2011 NLCD (Homer et al. 2015) (Table 2), and (3) subset the NCLD to only use CLs that were associated with unambiguously negative effects (e.g., exceedances of CLs for increased foliar N in forest trees can have negative effects due to its potential contribution to pest damage, but is not unambiguously negative as it can lead to greater growth and production).

All CLs were mapped to a 12-km × 12-km grid size to match the CMAQ grid (Table 1). Because different CLs have different resolutions (Table 2), some CLs had one entry in a grid cell and others had multiple. Thus, 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. We set a minimum of five entries ($n \geq 5$) to calculate the 10th and 50th percentile. Further details are available in the Appendix S1.

Exceedances

CL exceedances are calculated as the difference between the deposition and the CL (i.e., deposition – CL). For acidification CLs, the N and/or S deposition was used in the exceedance calculation because both N and S are acidifying agents. For cases in which the N deposition is lower than the N-retention or loss rates (i.e., lower than the long-term N uptake by vegetation, denitrification losses, and immobilization in the soil), exceedance is calculated with S deposition only (McNulty et al. 2007). For empirical CLs of N, only N deposition was used in the calculation, and the mechanism of effect is not specified (i.e., acidification, eutrophication, pest damage, or some combination).

RESULTS

Critical loads

We found that CLs varied widely in terms of geographic coverage and numerical value. They also varied on whether they met our criteria for calculating a percentile, and if they did, the percentiles themselves varied (Fig. 1). The Terrestrial Acidification CL is for forests, and so was absent for much of the non-forested Great Plains ecoregion and parts of the desert West (Fig. 1). The minimum, 10th, and 50th percentile Terrestrial Acidification CLs averaged 1260 eq ha^{-1} yr^{-1} (5th–95th percentile: 290–2769 eq ha^{-1} yr^{-1}), 1360 eq ha^{-1} yr^{-1} (5th–95th percentile: 341–2828 eq ha^{-1} yr^{-1}), and 1778 eq ha^{-1} yr^{-1} (5th–95th percentile: 547–3600 eq ha^{-1} yr^{-1}), respectively, with the lower values in the poorly buffered or thin soils of the glaciated North and parts of the mountainous West and the karst soils of Florida.

Terrestrial Acidification CLs were higher in more alkaline soils, especially those in the Midwest, Southwest, and regions of the mountainous Southeast. Across percentiles, CLs for Terrestrial Acidification did not dramatically shift from the minimum to the 10th percentile within a grid cell, and only moderately increased from the 10th to the 50th percentile. This suggests more variation across grid cells than within the 12-km scale adopted in this study.

The Aquatic Acidification CLs were sparsely represented at the 12-km grid size across the contiguous United States compared with the other critical loads. Thus, there were not enough aquatic acidification values within most 12-km grid cells to report the 10th or 50th percentiles, aside from pockets along the heavily studied Appalachian Range and a few areas in the West. The minimum, 10th, and 50th percentile Aquatic Acidification CLs averaged 3144 eq ha^{-1} yr^{-1} (5th–95th percentile: 196–12209 eq ha^{-1} yr^{-1}), 777 eq ha^{-1} yr^{-1} (5th–95th percentile: 139–3255 eq ha^{-1} yr^{-1}), and 1511 eq ha^{-1} yr^{-1} (271–5818 eq ha^{-1} yr^{-1}), respectively. There were many more Aquatic Acidification CLs in the East than in the West (Fig. 1) because eastern drainage waters are sensitive to acidification and thus have been heavily studied compared to the West. CLs tended to be higher in the West, and especially the Northwest, compared with those in the Appalachian Range in the East, as reported elsewhere (Sullivan et al. 2005, Shaw et al. 2013, McDonnell et al. 2014). This sensitivity difference is driven by relatively higher rates of acid deposition in the East compared to the West, along with a greater extent of base cation (BC) poor soils related to geomorphologic factors such as elevation, slope, soil depth, and soil and bedrock types (Omernik and Powers 1983). The counterintuitive results for the minimum having a higher mean and distribution than the 10th/50th occurred because water bodies sampled for critical loads tend to be sensitive water bodies, which tend to be geographically clustered in vulnerable areas (e.g., places with thin soils, higher elevation, higher deposition in the East), rather than randomly dispersed across the landscape. Thus, grid cells that met the $n \geq 5$ criterion are more vulnerable areas, and scattered grid cells with fewer water bodies (often in the West) were less vulnerable. This issue was only problematic for the Aquatic Acidification CLs—Terrestrial Acidification and empirical CLs were unaffected because the high density of data for the Terrestrial Acidification CL resulted in all cells meeting the $n \geq 5$ threshold, and the specified ranges for most empirical CLs obviated the need for a minimum sample size of 5. Implications of the spatial clustering of the Aquatic Acidification CLs in vulnerable areas are further discussed below.

For the four empirical CLs (empirical CLs of N for NO_3^- Leaching, Lichen, Plant Community Composition, and Forest-Tree Health; Fig. 1), in ecoregions where there was a reported CL or range, most 12-km × 12-km grid cells had at least one 30-m × 30-m tile of the appropriate land cover. This gave the visual impression of complete coverage in Fig. 1 even though there were scattered 12- × 12-km grid cells without the appropriate land cover.

The minimum CL for NO_3^- Leaching was relatively low across much of the country that had data (4–10 kg N ha^{-1} yr–^{1}, 285 – 714 eq ha^{-1} yr^{-1}; Fig. 1; Table 3). No NO_3^- leaching CL existed for the North American Deserts ecoregion and other arid ecoregions of the Southwest. Insufficient data were available to report percentiles across the East (CL = 8 kg N ha^{-1} yr^{-1}, 571 eq ha^{-1} yr^{-1}; Table 3).

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The Lichen CL, which is for epiphytic lichens in forest ecosystems, was also missing for the North American Desert and other arid ecoregions of the Southwest, and additionally in the Great Plains ecoregion. Elsewhere the minimum Lichen CL was low (2–9 kg N ha^{-1} hr^{-1}, 145–652 eq ha^{-1} yr^{-1}), and did not vary widely across percentiles.

The Plant Community Composition minimum CL was also relatively low across the country (3–7.8 kg N ha^{-1} yr^{-1}, 214–557 eq ha^{-1} yr^{-1}; Table 3), except for in forested ecosystems of the Mediterranean California (24 kg N ha^{-1} yr^{-1}, 1713 eq ha^{-1} yr^{-1}). However, within the 12-km × 12-km grid cells where these forested ecosystems were found, in many cases, there were other NLCD classes with lower CLs that determined the minimum CL (Table 3). There was no CL assigned for the Eastern Temperate Forest ecoregion in this study because we were unable to assign a minimum or percentile from the reported CL (<17.5 kg ha^{-1} yr^{-1}). Plant Community Composition CLs varied notably from the minimum to the 50th percentile (Table 3, Fig. 1).

The Forest-Tree Health minimum CL was relatively uniform, ranging from 3–5 kg N ha^{-1} yr^{-1} (214–357 eq ha^{-1} yr^{-1}) across the country, except in the Northwestern Forested Mountains and forested areas of the Mediterranean California ecoregions, where it was 17 kg ha^{-1} yr^{-1} (1214 eq ha^{-1} yr^{-1}; Table 3). Data were insufficient to assign percentiles for the Forest-Tree Health CL for all ecoregions except the Mediterranean California and Northern Forests ecoregions (Fig. 1, Table 3). There were no Forest-Tree Health CLs in this study assigned to three ecoregions (Tropical Humid Forests, Great Plains, North American Desert) because either there were no reported CLs in Pardo et al. (2011 a, b; Great Plains, North American Desert) or the response and source reference were not reported so we could not determine whether the effect was detrimental to forest health (Tropical Humid Forests).

The number of CL types within a grid cell varied across the country (Fig. 2). Of the six CL types examined, minimum CLs (Fig. 2a) were much denser in the ecoregions of the Eastern Temperate Forest (~4 CLs/cell), Northern Forest (5–6 CLs/cell), Northwestern Forested Mountains (5–6 CLs/cell), and coastal portions of the Mediterranean California (5–6 CLs/cell) compared with the Great Plains (2–3 CLs/cell) and the North American Desert ecoregions (1–2 CLs/cell). Because many ranges were not reported for empirical CLs, and five or more Aquatic Acidification CLs were not available in most grid cells, CLs for the 10th and 50th percentiles were much fewer across the country (Fig. 2b). Indeed, most of the country only had percentiles for two CL types in a grid cell (Fig. 2b), one of which was often Terrestrial Acidification, except for non-forested areas in the Midwest and West.

Across all six CL types, the lowest minimum CLs in a grid cell was relatively uniform across the country (200–400 eq ha^{-1} yr^{-1}; Fig. 3a). Exceptions included parts of the Pacific Northwest and scattered grid cells where the CL was even lower, and parts of the arid Southwest and Central Valley of California where it was slightly higher (Fig. 3a). The identity of the lowest minimum CL, however, varied greatly across the continent (Fig. 3b). The lowest CL in the East was for Forest-Tree Health (3 kg N ha^{-1} yr^{-1}, 214 eq ha^{-1} yr^{-1}). Across the Great Plains and North American Deserts ecoregions it was for Plant Community Composition (5 and 3 kg N ha^{-1} yr^{-1}, or 357 eq ha^{-1} yr^{-1} and 214 eq ha^{-1} yr^{-1}, respectively), and for much of the Northwestern Forested Mountains and coastal portions of the Mediterranean California it was Lichen that had CLs as low as 2.0–2.8 kg N ha^{-1} yr^{-1} (145–200 eq ha^{-1} yr^{-1}). There were pockets throughout the country where other CLs were the lowest (Fig. 3b), including terrestrial acidification for portions of the upper Midwest and southern Arizona, and aquatic acidification in scattered grid cells throughout the country (e.g., the north-central border of Wisconsin). Terrestrial Acidification was the lowest overall CL type in southern Arizona because it was the only CL type in these areas (Fig. 2a), whereas in the upper Midwest and parts of southern Florida Terrestrial Acidification was the lowest minimum CL among several CL types due to the sensitive soils in those areas. Nitrate Leaching was not the lowest unique CL except in a few scattered grid cells, but it was the lowest CL along with Plant Community Composition in regions of the mountainous West and in southern Florida.

When examining only acidification CLs of N and S (Fig. 3c, d), it was clear that Terrestrial Acidification CLs were lower across much of the country, but this was driven largely from lesser areal coverage of the Aquatic Acidification CL at the 12-km grid size compared with that of Terrestrial Acidification. In grid cells where both were present ($n = 4507$ for minimum CL and 408 for 10th/50th percentile CLs), aquatic acidification was lower 31% of the time for minimum CL (75% and 59% for 10th and 50th, respectively). Thus, in sensitive watersheds where the $n \geq 5$ condition was met, the critical load for aquatic acidification was usually lower than terrestrial acidification. These comparisons are more robust for the 10th and 50th percentiles, where the minimum data criterion ($n \geq 5$) is assured. When examining only empirical CLs of N (Fig. 3e, f), the pattern generally followed that of all CL types together, with the aforementioned exceptions.

Historical to recent period (1800–2011): Deposition and exceedances

Deposition.——We found that by 1855, moderate N deposition levels were already projected over parts of the East as a result of widespread biomass burning from land clearing for agriculture (Fig. 4; 400–800 eq ha^{-1} yr^{-1}, 5.6–11.2 kg N ha^{-1} yr^{-1}). This was especially true for the Southeast (Fig. 4). Because biomass burning rather than fossil fuel burning was the dominant contributor, N deposition exceeded S deposition in the 19th century. By 1905 and continuing until after 1975, industrialization resulted in elevated S deposition from fossil fuel combustion. Thus, between 1855 and 1955, there was an increase in and shift of the peak deposition from N deposition in the Southeast to N and S deposition in the Northeast, as the dominant contributor shifted from biomass burning to fossil fuel combustion. Even as early as 1905, S deposition in Pennsylvania and other areas in the Northeast was relatively high (800–1600 eq ha^{-1} yr^{-1}, 12.8–25.7 kg S ha^{-1} yr^{-1}). Between 1905 and 1975, estimated deposition began to show increases even in the West, although deposition rates and areas affected were still relatively small and/or localized compared to those in the East (Fig. 4). Peak deposition of N and S occurred roughly from the 1970s to the 1980s (Fig. 4), before the regulations under the CAA and 1990 amendments had taken effect, climbing above 3200 eq ha^{-1} yr^{-1} in some hot spots (roughly >45 kg N ha^{-1} yr^{-1}, or >50 kg S ha^{-1} yr^{-1}). Between 1975 and 2006 there were significant reductions in the areas experiencing high N+S deposition (>1600 eq ha^{-1} yr^{-1}), with a corresponding increase in areas experiencing moderate N+S deposition (800–1600 eq ha^{-1} yr^{-1}; Fig. 4). This mostly came from reductions in S deposition, with large reductions in areas receiving high (>1600 eq ha^{-1} yr^{-1}, >26 kg S ha^{-1} yr^{-1}) and moderate (800–1600 eq ha^{-1} yr^{-1}, 13–26 kg S ha^{-1} yr^{-1}) S deposition between 1975 and 2006, although much of the Great Lakes and surrounding areas (i.e. Indiana, Ohio, Pennsylvania, West Virginia) still experienced moderate levels of S deposition. On the other hand, the area experiencing moderate levels of N deposition (800–1600 eq ha^{-1} yr^{-1}, 11.2–22.4 kg N ha^{-1} yr^{-1}) increased from 1975 to 2006, especially over Iowa and other portions of the Midwest (Fig. 4). This finding is in agreement with Houlton et al. (2012) who examined wet N deposition from 1985–2009 and found increases across much of the Midwest and mountainous West.

Exceedances.——We first present exceedance trends for individual CL types, then geographic patterns of exceedances at illustrative periods in the decadal record. From 1800 to 2011, most exceedances of the minimum CL values of the six CL types followed the same general pattern. There are no exceedances in 1800, low levels of exceedance from 1855 to 1900, and increasing levels of exceedance between 1900 and 1950. Exceedances peak in total area in the 1975–1990 interval, depending on the CL type, and decline thereafter (Fig. 5). However, the six CL types differed greatly in the area exceeded in early decades, the timing and magnitude of the peak area affected, and in the magnitude of the decline in exceedance in recent years. Aside from the Terrestrial Acidification CL, there were insufficient data within the 12-km pixels to calculate percentiles for most CLs for many parts of the conterminous United States (Figs. 1 and 2). Thus, hereafter we focus primarily on exceedances of the minimum CL for all CL types except for Terrestrial Acidification. The full set of results summarized at the national level, including exceedances of percentile CLs, is available in Appendix S1: Table S1.

The CL for Terrestrial Acidification was already exceeded in 1855 in 139000 to 371000 km^2 of the United States (3–7% of areas with data), depending on the percentile (Fig. 5). Hereafter, unless otherwise specified, percentages of area exceeded indicate the percentage of area of 12-km CMAQ cells with data for that CL type that was exceeded by deposition in a given year (i.e., excludes areas with no data for that CL type). For Terrestrial Acidification, exceedances rose to a peak of almost 2.78 million km^2 (50%) in 1975 for the minimum CL (2.34 million km^2 [49%] and 1.78 million km^2 [37%] for the 10th and 50th percentiles, respectively). For reference, one million square kilometers is an area roughly the size of Texas and Arizona combined. By 2006 (average of 2002–2012), the area of exceedance had decreased to roughly 2.36 million km^2 (42%) for the minimum CL (1.88 [39%] and 1.33 [28%] for the 10th and 50th percentiles, respectively). Declines continued to 2011 but large areas remained in exceedance. Thus, even when half the forested area in a grid cell was protected, over a quarter of the area with data was still in exceedance in 2006, an area roughly twice the size of Texas. The geographic patterns of exceedances (Fig. 6) generally followed the deposition patterns, being higher in the East and near urban centers and lower in the West. Early exceedances occurred in the Southeast from land burning for agriculture, then switched to increasing exceedances in the northeastern United States with industrialization and hot spots of exceedance in the urban centers of the west from 1905–1975. The decreases in exceedances from 1975–2006 occurred throughout the east, but were most notable in the Southeast and central portions of North Carolina, South Carolina, Kentucky, and Tennessee.

The minimum CL for Aquatic Acidification was exceeded in a much smaller area of CMAQ grid cells compared with the other CL types (Fig. 5). However, much of this was because the total extent of area with an Aquatic Acidification CL was lower compared with the other CL types (roughly 0.6 million km^2 compared with 4–6 million km^2 for the minimum CL). Nonetheless, as a proportion of the total in terms of area and for the nearly 10000 individual water bodies, exceedances of the minimum Aquatic Acidification CL were comparable with other CL types (e.g., 47% in 1975 as compared with 44–81% for the other CL types). Areal exceedances of the minimum CL were roughly 89600 km^2 in 1855 (1806 of 9915 water bodies, 18%), peaking at roughly 307000 km^2 in 1975 (5352 water bodies, 54%), and declining to 253000 km^2 in 2006 (4500 water bodies, 45%). Most of the exceedances occurred in water bodies draining the shallow and poorly buffered BC soils of the Appalachian Mountains in the East. More alkaline soils in the West led to higher CLs causing lower exceedances for a given amount of deposition. Certain lakes in Florida and the upper Midwest show exceedances, but many may be naturally acidic and not responsive to changes in acidic deposition.

The minimum CL for NO_3^- leaching was already exceeded in 389000 km^2 (6%) of the United States in 1855 (Fig. 5). Exceedances peaked at roughly 3.39 million km^2 (54%) around 1995, and only declined to 3.16 million km^2 (51%) by 2006. The sharp increase in exceedances after 1945 occurred when N deposition increased above the minimum CL in the Eastern Temperate and Northern Forest ecoregions (8 kg N ha^{-1} yr^{-1}, 571 eq ha^{-1} yr^{-1}; Table 3). Other percentiles were not available for the high deposition areas in the East (Table 3); thus, the area exceeded for higher percentiles was low. As with the other CL types, the spatial pattern matched the deposition pattern, which was highest in the East and in hot spots in the West (Fig. 6).

The minimum CL for Lichen was already exceeded by N deposition in 1.89 million km^2 of land in 1855 (46%; Fig. 5). Exceedances peaked at roughly 3.47 million km^2 (84%) in 1995 and declined only slightly to 3.42 million km^2 in 2006 (83%). Because the Lichen CLs were low (Fig. 1), there was little noticeable geographical contraction in areas exceeded, even though deposition declined from 1975 to 2006 (Fig. 6). The similarities among Lichen CL percentiles (Fig. 1) led to similarities in area exceeded of different percentiles through time.

Exceedances of the minimum CL for Plant Community Composition was much lower in early decades than for other CL types because there was no quantifiable CL for this endpoint in the industrialized East, in other words, the Eastern Temperate Forest ecoregion (Figs. 1 and 5). Thus, the increase in area exceeded lagged behind other CL types until deposition increased above the Plant Community Composition CL (minimum: 5 kg N ha^{-1} yr^{-1}, 357 eq ha^{-1} yr^{-1}; Fig. 6) primarily in Iowa, northern Missouri, and other parts of the Great Plains ecoregion. This increase began to occur sometime between 1935 and 1955. Exceedances of the minimum CL peaked in 1995 with 2.87 million km^2 (56%), declining to 2.66 million km^2 (51%) by 2006.

The minimum Forest-Tree Health CL for the Eastern Temperate and Northern Forest ecoregions (3 kg N ha^{-1} yr^{-1}, 214 eq ha^{-1} yr^{-1}) was already exceeded in 2.41 million km^2 in 1855 (Fig. 5). The total area of minimum CL exceedance did not change much over the historical record due to the low CL for this endpoint and ecoregion. Furthermore, although the magnitude of exceedance remained high (Fig. 6), it declined as deposition decreased from 1975 to 2006.

Summing the number of minimum CLs exceeded across the six CL types showed that by 1855, two or more CLs were exceeded over most of the East, mostly from exceedances of the Forest-Tree Health and Lichen CLs (Figs. 6 and 7). More CLs were exceeded in the Southeast and Florida, and parts of the industrializing North. The geography of exceedances did not change appreciably in 1905 and on to 1955, although the number of CLs exceeded increased overall and pockets of the urbanizing West began to show impacts. By 1975, exceedances of one or more minimum CLs occurred over nearly 60% of the conterminous United States (roughly 5.4 million km^2 of CMAQ grid cells; Fig. 7b), excluding large areas in the mountainous and Intermountain West. There were four or more minimum CLs in exceedances across much of the East, with all CLs exceeded in parts of the Northeast where data for the Plant Community Composition CL was available and sensitive water bodies existed. By 2006, there was only a slight decline to 5.2 million km^2 of total area with one or more minimum CLs exceeded. There was also little change in the geographic pattern of CL numbers exceeded, except for a decline in portions of the East such as western North Carolina, predominantly from reductions in exceedances of terrestrial acidification (Fig. 6).

Future period (2025): Deposition and exceedances

Deposition.——From 2011 to 2025, deposition decreased under all four potential management scenarios, but by different amounts among scenarios (Fig. 8). The CAA reductions scenario resulted in overall large reductions in both N and S deposition (Fig. 8), most notably over the Midwest and the Great Lakes states. Almost no areas of the country were projected to experience S deposition above 400 eq ha^{-1} yr^{-1} in 2025 under the CAA scenario (>6.4 kg S ha^{-1} yr^{-1}). However, most of the eastern United States was still expected to experience N deposition above 400 eq ha^{-1} yr^{-1} (>5.6 kg N ha^{-1} yr^{-1}), and many areas of the Midwest were projected to experience N deposition above 800 eq ha^{-1} yr^{-1} (>11.2 kg N ha^{-1} yr^{-1}). Differences among the four future 2025 scenarios were mostly due to N emissions, as additional management controls were focused on technologically feasible reductions in agriculture-based emissions from improved fertilizer or livestock management. Improved management of livestock (−0.7 Tg yr^{-1}; Table 1) resulted in the largest reductions in N deposition between the two scenarios, greatly reducing the area exposed to moderate N deposition (>800 eq ha^{-1} yr^{-1}, >11.2 kg N ha^{-1} yr^{-1}; Fig. 8). Fertilizer reductions (−0.2 Tg yr^{-1}; Table 1) either alone or with improved livestock management did not appear to have widespread effects on atmospheric deposition.

Exceedances.——Totaled across the country, the area exceeding the minimum CL only declined from 5.3 million km^2 to 4.9 million km^2 from 2011 to 2025 under the CAA scenario (Fig. 9; 4.54 million to 4.15 million km^2 for the 10th percentile, and 3.33 million to 3.05 million km^2 for the 50th percentile [data not shown]). There was still more area in exceedance of one or more minimum CLs than none nationally (Fig. 9). Differences among the four future scenarios were minimal for total area exceeded (Fig. 9); thus, most of the improvement between 2011 and 2025 appeared to come from the CAA scenario, which showed reduced exceedances of terrestrial acidification and, to lesser extents, NO_3^- leaching, aquatic acidification, and plant community composition (Fig. 10). For the Lichen, Aquatic Acidification, and Forest-Tree Health CLs, additional reductions in deposition associated with agriculture primarily reduced the magnitude of the exceedance rather than the areal extent of exceedance (Fig. 10).

DISCUSSION

Overall, this analysis provides a deeper understanding of the relative vulnerabilities of different ecological endpoints and regions through time for the conterminous United States. Our results show that although areal exceedances of many CLs are declining from historical peaks in the 1970s and 1980s, these exceedances have likely existed for decades, many continue to exist under current deposition levels, and additional emission reductions will be needed in the future if the protection of these sensitive aquatic and terrestrial end points is a societal goal. Finally, these patterns are clearly dependent on the data used to make these inferences, which have known limitations, and this study reveals some key areas for improvement in future research efforts.

Patterns over the historical to recent period (1800–2011)

From 1800–2011, we found that exceedances occurred over large areas in the East even early in the deposition record (1855–1905). Many empirical N CLs and areas sensitive to acidification have CLs in the 200–400 eq ha^{-1} yr^{-1} range (roughly 2.8–5.6 kg N ha^{-1} yr^{-1}, or 3.2–6.4 kg S ha^{-1} yr^{-1}; Table 3), and deposition levels higher than this are thought to have occurred for decades in the eastern United States. Our results suggest that from roughly 1905 to 2011, deposition of S and/or N exceeded several minimum CLs by 350 eq ha^{-1} yr^{-1} or more for much of the East, or in other words, deposition was roughly double the CL or higher for decades. Current deposition levels are lower compared to the peaks of the 1970s and 1980s, but they are still high relative to many CLs, and remain elevated over pre-industrial deposition rates by more than an order of magnitude (roughly 35 eq ha^{-1} yr^{-1} or 0.4 kg N ha^{-1} yr^{-1} plus 0.1 kg S ha^{-1} yr^{-1}). CLs in the 200−400 eq ha^{-1} yr^{-1} range may not be unrealistically low, as this is still 5.7–11.4 times higher than the pre-industrial deposition levels under which these systems evolved.

Many sensitive endpoints have likely been experiencing exceedances for some time, including poorly buffered watersheds, certain taxonomic groups, and areas with a lower capacity for nutrient incorporation. Thus, especially in the eastern United States, but also in hot spots in the Midwest and West, the health of trees and forest composition has likely been affected, lichen and plant communities have already and may be continuing to shift, and NO_3^- leaching as well as acidification of some terrestrial and aquatic systems have likely occurred for many years. Evidence of these impacts has been reported in the literature for trees (Dietze and Moorcroft 2011, Duarte et al. 2013, Sullivan et al. 2013), lichens (Geiser et al. 2010, Cleavitt et al. 2015), herbaceous biodiversity (Simkin et al. 2016), and lakes and streams (Dupont et al. 2005, McDonnell et al. 2014). This could have significant but unmeasured effects on ecosystem services and their human beneficiaries, including outdoor enthusiasts, timber producers, and resource-dependent businesses such as wilderness lodgings (Blett et al. 2016, Bell et al. 2017).

On the more recent end of the deposition record, exceedances are declining for nearly all CL types examined (Figs. 5 and 7). The total area with no exceedances of any minimum CL has increased from a low of 1.99 million km^2 in 1995 to 2.57 million km^2 in 2011, an increase in area almost the size of Texas. This is good news and evidence that the national air quality policies such as the 1990 Amendments under the Clean Air Act are improving environmental conditions. On the other hand, more areas nationally remain in exceedance of CLs than not, and in 2011, roughly 68% of the area with any CL data in the conterminous United States experienced an exceedance of one or more CLs (66% including areas with no data). Thus, although the no exceedance area is increasing, there remains significant room for additional improvement. This caveat suggests that although the current mixture of national emission standards is a significant and positive step, especially in reversing the trajectory of deposition and exceedances from increasing to decreasing, the magnitude of decrease is not sufficient to protect many ecological endpoints over the next decade.

Notably, we found the lowest CL is relatively uniform across the country at 200–400 eq ha^{-1} yr^{-1}. Such levels of N and/or S deposition are low but not uncommon; they are well above pre-industrial levels and still exist across much of the western United States (Fig. 3). However, deposition levels have not been this low over many parts of the eastern United States for possibly over 100 years. It is uncertain whether reducing deposition to these levels in the East is realistic under current technologies and policies. Our analysis of four possible future scenarios suggests that current policy- and technology-based feasible reductions are not sufficient to reduce deposition below many CLs (Figs. 9 and 10). Thus, it would be advantageous, especially in the East, to increase the focus on mitigation and recovery processes (e.g., liming soil, replanting lost species; Lawrence et al. 2015) and on developing more advanced technologies to reduce emissions and deposition. It is important to note that these technologically feasible reductions are from 2010 (EPA 2010), and newer technologies may be available. Either way, it is unclear whether simply lowering N and S deposition below CL levels will be sufficient to induce recovery, or whether more active management will be required (e.g., liming, planting lost species). For example, certain taxonomic groups may have already been lost from some areas, and other factors such as dispersal may limit their reintroduction to a site (Clark and Tilman 2010). More research on recovery is beginning to emerge nationwide. Lawrence et al. (2015) found that declining acidic deposition appeared to begin the reversal of forest acidification in the northeastern United States and Canada, but that such recovery was very slow, and some responses and sites showed continued effects from acidification. Experimental studies in the Midwest report that plant communities may not easily recover from long-term N addition (Isbell et al. 2013) without significant intervention (Clark and Tilman 2010). The longest running N-addition experiment in the world, the Park Grass Experiment in the United Kingdom, found that some form of intervention (e.g., mowing, liming) was necessary for plant community recovery following reductions in atmospheric deposition (Storkey et al. 2015). Thus, many of these ecological systems may not rapidly recover on their own even if deposition is decreased because internal hysteretic processes can maintain the perturbed state (Vinton and Burke 1995, Bakker and Berendse 1999, Suding et al. 2004, Walker et al.

2012). An example of such processes is when elevated nutrient levels are maintained long after N inputs decrease due to the new plant community being dominated by species with high-nutrient tissue that rapidly decomposes, thereby maintaining the community's own persistence (Vinton and Burke 1995, Clark and Tilman 2010).

Certain CLs are more influential than others, especially if they are very low and applied over a large area, and thus deserve more scrutiny. This is true for all empirical CLs (discussed below), and is especially true for the Forest-Tree Health CL in the Eastern Temperate Forest ecoregion (3 kg N ha^{-1} yr^{-1}, 214 eq ha^{-1} yr^{-1}), which is responsible for some of the highest and longest running exceedances historically. This CL is coincidentally one of the more robust empirical CLs in the NCLD database. Rather than based on a single site, this is an analysis of the responses of 24 tree species (including >100000 trees in the analysis) for an 18 state area in the East (Thomas et al. 2010). The minimum N deposition in Thomas et al. (2010) was roughly 3 kg ha^{-1} yr^{-1}. Deposition higher than this was associated with ecologically meaningful (>5%) reductions in growth for one species (red pine, *Pinus resinosa*), and ecologically meaningful reductions in survival for three species (big-tooth aspen, *Populus grandidentata*; quaking aspen, *Populus tremuloides*; scarlet oak, *Quercus coccinea*). Thus, using the strict definition of the CL (i.e., where significant negative effects begin to occur), this is an appropriate CL for these four species, which are reported to be especially impacted from atmospheric deposition. Thomas et al. (2010) also reported an additional two species that experienced reductions in growth and five species that experienced reductions in survival; these reductions were statistically significant even though fairly small in magnitude (<5%). The CL may even be lower than this, as Thomas et al. (2010) only analyzed 24 common species. Some rarer species may be more impacted or already absent in this region, which has experienced high deposition levels for decades. On the other hand, although setting the CL at 3 kg ha^{-1} yr^{-1} is correct according to the accepted definition of the CL, what is "significant harm"? From Pardo et al. (2011a, b), who established this as the CL based on the Thomas et al. (2010) work, any reductions in growth and survival were interpreted as significant harm. Alternatively, if a specific magnitude of negative effect (e.g., decreases of growth by 5 kg C ha^{-1} yr^{-1} or reductions in survival by 5%) were required, the CL would be higher (closer to 10 kg ha^{-1} yr^{-1}). Furthermore, Thomas et al. (2010) used CASTNET data, which is known to underestimate dry deposition; thus, the true deposition and the CL were likely higher than that reported in Thomas et al. (2010). Most importantly, these four sensitive species are not evenly distributed across the entire Eastern Temperate Forest ecoregion, and thus the CL likely only applies to stands where these sensitive species are located, rather than the entire Level 1 ecoregion. It is unknown how these offsetting factors would balance out, and the implications from these and other uncertainties are discussed below. Future efforts are focused on refining these tree CLs to specific tree species and areas of known sensitivity.

Patterns for the future period (2025)

The CAA scenario reduced both sulfur dioxide (SO_2) and nitrogen oxides (NO_x) emissions, with a larger reduction in NO_x (~5.8 million tons yr^{-1} from a 2011 level of ~14.1 million tons yr^{-1}) than SO_2 (~3.7 million tons yr^{-1} from a 2011 level of 6.4 million tons yr^{-1}). Nonetheless, projected emissions in 2025 on an aggregate basis compared to 2011 were

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still dominated by more net NO_x (~8.3 million tons yr^{-1}) than SO_2 (~2.7 million tons yr^{-1}) emissions. Further, there was negligible change in NH_3 emissions between 2011 and 2025, with an estimated 4.2 million tons yr^{-1} NH_3 emissions. The larger impact of deposition reduction on exceedances of acidification CLs (N and S deposition) than empirical CLs (N deposition) could be due to several factors, including higher overall emissions of N, larger background of N-based emissions, lower empirical CLs of N versus acidification CLs, and/or high enough NH_3 emissions that maintain deposition above CLs of N. We found that additional technologically feasible reductions from livestock management had a larger effect on reducing N deposition than reductions from synthetic fertilizer applications. This is expected due to the larger reductions of NH_3 emissions anticipated from livestock management (30% or 0.7 Tg yr^{-1}) compared with fertilizer reduction (20% or 0.2 Tg yr^{-1}).

Nonetheless, even though reduced emissions from livestock had a noticeable effect on reducing N deposition (Fig. 8), neither scenario had a large effect on reducing the areas affected by CL exceedances. This occurred because the magnitude of the reduction was not sufficient to lower deposition below the CLs. However, a projected lack of improvement in the areal exceedances presented here does not mean there would not be other improvements, such as from a reduced magnitude of exceedance, or from other endpoints excluded from this study (e.g., water quality, reduced Greenhouse Gas emissions, etc.).

We explored the sensitivity of these results to potential changes in climate, which can alter deposition through changes in the magnitude and timing of precipitation, and by changes in atmospheric chemistry and transport processes, affecting both wet and dry deposition of N and S (Lamarque et al. 2013, Simpson et al. 2014, Greaver et al. 2016). Changes out to 2030 are not expected to be large, but could affect the sensitivity of our results (Figs. 8–10). We used Community Earth System Model (CESM) fields, from simulations conducted for the CMIP5 (Taylor et al. 2012), that were dynamically downscaled to 36 × 36 km using the Weather Research and Forecasting Model (Spero et al. 2016). As expected, we found that climate change from 2000 to 2030 only minimally altered deposition (deposition changed <2% for all future scenarios; Appendix S1: Fig. S2). Much larger effects on climate and deposition are expected towards the end of the 21st century (Lamarque et al. 2011, Melillo et al. 2014). Additional details on the future climate simulations are provided in the online Appendix S1.

Policy implications

These results are clearly relevant for the current review of the secondary (welfare-based) National Ambient Air Quality Standards (NAAQS) for NO_2, SO_2 and PM. The current NO_2 and SO_2 secondary NAAQS are intended to protect against direct damage to vegetation by exposure to gas-phase NO_x and SO_x (EPA 2011). However, EPA has noted the importance of considering the protection provided by the current standards against other ecological effects (EPA 2017a). In particular, EPA plans to focus in the current review on ecological effects associated with total N and S deposition, including impacts on biodiversity, clean water, and healthy terrestrial and aquatic ecosystems (EPA 2017b, a).

Directly linking the fluxes of total N and S deposition that ecosystems experience via acidification and eutrophication, to the ambient air concentrations of NO_x and SO_x that are regulated, remains a scientific challenge. However, CLs are helpful in evaluating how the ecosystem could potentially respond to varying deposition levels. As a hypothetical exercise, we examined exceedances for a range of fixed deposition levels (2.5, 5.0, 7.5, 10, 12.5, and 15kg N ha^{-1} yr^{-1}; 178, 357, 535, 714, 893, and 1071 eq ha^{-1} yr^{-1}; Fig. 11, Appendix S1: Fig. S4). We found that although few CLs were exceeded at 2.5 kg N ha^{-1} yr^{-1} aside from the Lichen CL in the Northwest and some Aquatic Acidification CLs, nearly all non-acidification CLs were exceeded at 10 kg N ha^{-1} yr^{-1} (60–100% of the area with data; Fig. 11). In contrast, even deposition as high as 15 kg N ha^{-1} yr^{-1} (or the equivalent of 1071 eq ha^{-1} yr^{-1} as N+S deposition) would only exceed the acidification CLs in 40–45% of the areas with data. This information suggests that using only one CL type to evaluate the protection provided against acidification and eutrophication effects would present an incomplete picture of risk. In addition, because CL values vary widely across the landscape, even within a CL type, it is difficult to assign a single national value. This suggests the importance of also considering the geographically varying nature of the acidification and eutrophication effects. In addition, our evaluation showed that the small effect of climate change on exceedances compared with changes in emissions suggests that over the short term (e.g., to 2030) policies that focus on reducing emissions will have a larger effect on reducing exceedances of CLs. Over longer terms (e.g., to end of century) the relative influence may change as the effect of climate on deposition is expected to increase through time (Lamarque et al. 2011). These results are important for consideration in any quantitative assessment of ecological effects related to N and S deposition, as well as in any decision making framework in the current review of the secondary NAAQS for ecological effects of NOx, SOx and PM.

Uncertainties, insights, and next steps

There are many uncertainties and limitations to this current study. These fall primarily in two domains: our confidence in the deposition and confidence in the CL. Much of our historical deposition record relies on the CAM estimates (Lamarque et al. 2010), which is the best available national data set deposition prior to 1980 to our knowledge. However, the CAM record appears to “flatten” the N deposition record when compared to the few local historical records that have been estimated (Appendix S1: Fig. S3), with higher historical levels (before roughly 1925–1965, especially for N) and lower peaks in the recent period. The reasons for this are unclear, but part of the explanation could be that the CAM deposition is at a much coarser grid scale than CMAQ (200 km^2 vs. 12 km^2), and is thus missing localized areas of high deposition. Regardless, we may be overestimating early exceedances of CLs related to N deposition (all examined here). CAM estimates early deposition (before 1900) in the eastern United States to be roughly 3–5 kg N ha^{-1} yr^{-1} and 0–1.5 kg S ha^{-1} yr^{-1}, with hot spots in the deep south from biomass burning. Detailed historical reconstructions in the Southeast (Southern Appalachian Mountain Initiative [SAMI]; [(Shannon 1998)]), and the Northeast (Hubbard Brook Experimental Forest, New Hampshire; [(Gbondo-Tugbawa et al. 2001)]) hindcast lower early N deposition levels (<1–2 kg N ha^{-1} yr^{-1}) and higher contemporary levels than CAM estimates (Appendix S1: Fig. S3). These reconstructions imply much lower early exceedances in the East. However, both studies found that N deposition had already increased above 5 kg N ha^{-1} yr^{-1} by roughly 1925 (SAMI) and 1950 (Hubbard Brook), levels that were above many empirical CLs of N

(Table 3). Thus, even though we may be overestimating the duration of early exceedance, exceedances of N CLs probably still have existed for many decades. For S deposition, the accuracy of CAM estimates appeared site specific. Deposition levels were amplified (as opposed to flattened) in one site (SAMI, Piney River) but matched the other two sites reasonably well (SAMI, Cosby Creek; Hubbard Brook). Thus, it is unclear to what degree we over- or underestimated early exceedances attributable to S deposition.

However, our contemporary exceedances are likely not underestimated by much because we transition largely to CMAQ by 1980, and CMAQ estimates of recent deposition for the United States are more accurate because they use estimates from the National Emissions Inventory (https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei) as discussed in Bash et al. (2013). However, CMAQ may not fully capture organic deposition, which can be significant in some areas (Cape et al. 2011) and may not adequately capture hot spots of deposition from orographic effects, which also can be locally significant (Weathers et al. 2006). Either of these data deficiencies could increase the magnitude of an exceedance (assuming the CL is unaffected) or have no effect if the original CL did not include these sources and thus should be similarly increased. However, for organic N, it is important to distinguish anthropogenic organic deposition (e.g., Peroxyacyl nitrate [PAN]), which would be included in an exceedance, from natural organic deposition (e.g., pollen, amino acids), which would be considered part of the baseline and not contribute to exceedance.

Another principal uncertainty are the CLs, both empirical CLs of N and acidification, and the appropriate spatial and temporal scale of their use. Most empirical CLs are based on a limited number of site-specific studies that have been extrapolated and reported for Level 1 ecoregion(s). Even though this current study has taken steps to restrict the CLs to their corresponding land cover type, more work is needed to revisit the original source literature of these CLs and to restrict their range of application to appropriate ecosystem type(s) and smaller geographical extents (e.g., Level 3 ecoregion). Another principal weakness of the empirical CLs of N is that many are based on relationships developed using different deposition data sets (e.g., NADP vs. Total Deposition [TDEP]). Any bias in the deposition data would bias the CL in the same direction. For example, because NADP estimates of N deposition are usually lower than actual total N deposition for various reasons (e.g., dry deposition not included, orographic effects not captured, etc.), CLs based on NADP are also biased low.

Three recent independent advances are likely to dramatically improve the empirical CLs of N for the conterminous United States and NCLD. First, a recent national effort by Simkin et al. (2016) reported CLs for changes in herbaceous biodiversity using data from over 15000 forested and non-forested sites. This will affect the Plant Community Composition CLs for all ecoregions, especially in the Eastern Temperate ecoregion, which will change from no minimum or percentile CL (Table 3) to a mean of 7.9 kg ha^{-1} yr^{-1} in non-forested areas and 12.5 kg ha^{-1} yr^{-1} in forested areas (Simkin et al. 2016). Second, the Lichen CLs, which were originally derived from a study in the northwestern United States and extrapolated to the nation (Geiser et al. 2010), have been updated to include many more plots nationally (*L. Geiser personal communication*). Finally, an updated analysis of Thomas

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et al. (2010) is underway that will dramatically improve our understanding of tree sensitivity to N deposition (Horn et al. 2017). All three of these efforts, which are much more spatially refined and based on data with nationwide coverage, will dramatically improve the robustness of empirical N CL estimates in the United States.

In addition to the empirical CLs, the acidification CLs have known limitations that could be improved. The Aquatic Acidification CL is probably the most refined CL in the United States, although more work is needed to expand the spatial coverage and improve site-specific estimates of how deposition affects NO_3^- leaching to surface waters. However, one should note that CLs have not been estimated for many water bodies in the United States, and it is clear from the mismatch between the distribution of CLs for the set of water bodies that meet the $n \geq 5$ criteria and those that did not, that these CLs are not a random subpopulation of U.S. surface waters. Vulnerable water bodies are geographically clustered and over-represented in the NCLD. It is critical to understand which areas are vulnerable, but the over-representation is important to keep in mind when making statements about U.S. water bodies based on the NCLD. One critical advancement would be to use spatial statistics to compare the water bodies in the NCLD with the water bodies in the National Lakes Assessment (NLA, https://www.epa.gov/national-aquatic-resource-surveys/nla) database and the National Rivers and Streams Assessment (NRSA; https://www.epa.gov/national-aquatic-resource-surveys/nrsa) survey, which are both random subpopulations of U.S. water bodies, to better quantify the population of water bodies that the NCLD represents.

This question of representativeness is not restricted to Aquatic Acidification CLs; it is common across all CLs, especially empirical ones, which often were developed at a single site and applied to an ecoregion. Further work is underway under the Critical Loads of Acid Deposition Working Groups of NADP (Blett et al. 2014) to assess the representativeness of all CLs in the NCLD.

The Terrestrial Acidification CL also has many known limitations, including the availability of estimates of soil BC weathering, one of the most influential variables (Li and McNulty 2007), as well as regional specific estimates of key parameters, including N immobilization, NO_3^- leaching, and N and BC uptake, all of which are difficult to estimate for forest ecosystems across the conterminous United States. It is difficult to know whether the Terrestrial Acidification CLs are biased high or low due to the many potentially offsetting sources of uncertainty. However, studies have demonstrated that poor representation of soil surface area in estimates of BC weathering can introduce significant variation and high levels of uncertainty (Jönsson et al. 1995). Therefore, the estimates of Terrestrial Acidification CLs used in this and other studies could be significantly biased.

Another noteworthy source of uncertainty is the time frame used to determine the CLs and lag effects in the response and recovery. Because most of our empirical and modeling data come from the past few decades, which followed or included periods of high deposition, many effects may have already occurred and sensitive taxonomic groups lost as a consequence. Thus, the baselines used for comparison are not “no effect” in many cases, and the CLs reported are better described as preventing “further significant harm” rather than “any significant harm.” This is why the definition of the CL includes the phrase “according to present knowledge” to recognize the fact that knowledge is always growing. Lag effects are also common in ecological systems, and the responses observed (empirical CLs of N) or estimated (acidification CLs) are assumed to be caused by the same year’s deposition. This is almost certainly a large oversimplification, as increases in soil available N, which can drive both processes, can take years to accumulate to a tipping point at which a shift is observed. The length of this lag, however, probably depends on the response of interest (e.g., slow for trees and faster for lichens), and remains largely unaccounted for in CL research. Furthermore, we did not include the large shifts in land use change from 1800–2025, which certainly would shift upwards the total acreages in exceedance through time due to large uncertainties in sensitivity of land that is no longer in its historic ecological state (e.g., pre-agricultural or pre-development).

These sources of critical uncertainty have many implications for conservation management as well as the science and policy of CLs. Long-term historical exceedances make it challenging to accurately define a reference historical state, and given that ecosystems are dynamic, there is no guarantee that the reference state would have persisted in the absence of perturbation. Dynamic models and longer term empirical approaches, such as assessments of soil seed banks and pollen, can try to recreate these historical reference states and determine the time frame of the response, but have not been widely used for CLs research to date.

At the time of this study, the TDEP data layer was not available (Schwede and Lear 2014). Future versions of this analysis will incorporate TDEP estimates in exceedance calculations. However, we do not expect large changes in contemporary areal exceedances with TDEP because the CLs are comparatively low, except possibly in localized areas dominated by dry deposition (especially S) where the largest differences between CMAQ and TDEP occur (Schwede and Lear 2014).

Finally, as briefly mentioned above, it is important to note that CL exceedances do not translate directly to a frequency or magnitude of effect. Most empirical CLs are a single threshold value, above which negative effects are reported to occur. But the frequency or magnitude of those effects are often not reported. A full dose-response type function relating deposition to effect (e.g., for herbaceous biodiversity in Simkin et al. 2016) is needed to link exceedances of a CL to a magnitude of ecological effect. For example, with a full dose-response relationship, one could ascertain whether the same exceedance (e.g., of 5 kg ha^{-1} yr^{-1} of N) had a large or a small effect, an important insight that is not possible for many CLs.

Thus, throughout the analytical chain, there are potential biases high and low, but nonetheless this effort represents the state of knowledge for critical load exceedances in the United States for these six CL types. There are important additional insights unearthed from this study. One is the need for explicit quantification of CL uncertainty due to the vast differences in methodology, scale, and interpretation. A common approach for uncertainty characterization will aid decision makers navigating these complex scientific studies and will enable better information exchange among researchers, decision makers, and the public. Another insight is the challenges of scale. We converted all deposition and CLs to a common scale to allow for direct comparisons among CLs and exceedances. By doing this, some

CL types had multiple CL values within a grid cell and others did not, which enabled us to examine various levels of risk (i.e., the percentiles) to give decision makers flexibility in using and interpreting exceedance information to assess vulnerability. Some decision makers may conclude, based on one set of statutory or non-statutory guidelines, to try to protect against exceedances of all endpoints within their management jurisdiction (i.e., use the minimum CL as a reference), and some may conclude, based on different guidelines, that some percentage of protection is more appropriate when balancing complex social and ecological tradeoffs. Our analysis was not able to rigorously support such a varied decision-making perspective, as most CLs did not have sufficient data to report percentiles nationally (Fig. 1). We decided to normalize all CLs to the 12-km × 12-km CMAQ grid cell because this was the best available deposition data at the time. Other approaches could be explored, including aggregating to a larger CMAQ grid cell (e.g., the older 36-km grid cell), aggregating to larger areas, such as Level 3 ecoregions, and/or allowing a flexible grid size based on some minimum number of CLs in an area. Each of these approaches has its strengths and weaknesses, and while aggregating to a larger scale may sacrifice the ability to identify "hot spots" of exceedances, various levels of risk can be examined.

From the results, uncertainties, and insights above, we developed the following recommended list of ten next steps for the field (ordered roughly from specific to general):

1. Constrain the geographic areas for which empirical critical loads of N are applied (currently Level 1 ecoregions).
2. Assess the representativeness of the Aquatic Acidification CLs by comparing water bodies in the NCLD with that of other data sets (e.g., NLA, NRSA).
3. Improve key uncertainties in the Terrestrial Acidification CLs (e.g., BC weathering, soil surface area).
4. Update the NCLD with newly available CLs (e.g., Simkin et al. 2016) and consider replacement of older CLs.
5. Develop stronger linkages between CL exceedances and ecological effects.
6. Explore other geographic classifications for aggregating CLs (e.g., Levels 2 and 3 ecoregions).
7. Improve uncertainty characterization for all CL types.
8. Develop better historical reconstructions of N and S deposition and develop methods to empirically "ground truth" early deposition estimates.
9. Improve deposition estimates further to better capture orographic effects, emissions of organic N from anthropogenic and biogenic sources, and future climate change.
10. Develop methods for characterizing reference states that may predate the direct empirical record, and the lag or time frame of an effect.

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CONCLUSIONS

This effort represents the state of knowledge for critical load exceedances in the contiguous United States for six CL types from 1800–2011 and for 2025 based on four different emission scenarios. We found that minimum CLs for all six CL types have likely been exceeded for decades in many portions of the United States. We also found that even though deposition and exceedances are improving nationally (especially for S), the "on-the-books" emissions reductions to 2025, as well as hypothetical reductions based on EPA SAB recommendations, appear insufficient to lower deposition below many CLs (especially for N). However, these SAB recommendations are based on best available technologies and may be outdated and need revisiting. Our conclusions are based on the best data available at the time of writing, and we outline several limitations, as well as specific and general recommendations for improving estimates of CLs, deposition, and their combination in estimating exceedances across the United States from air pollution.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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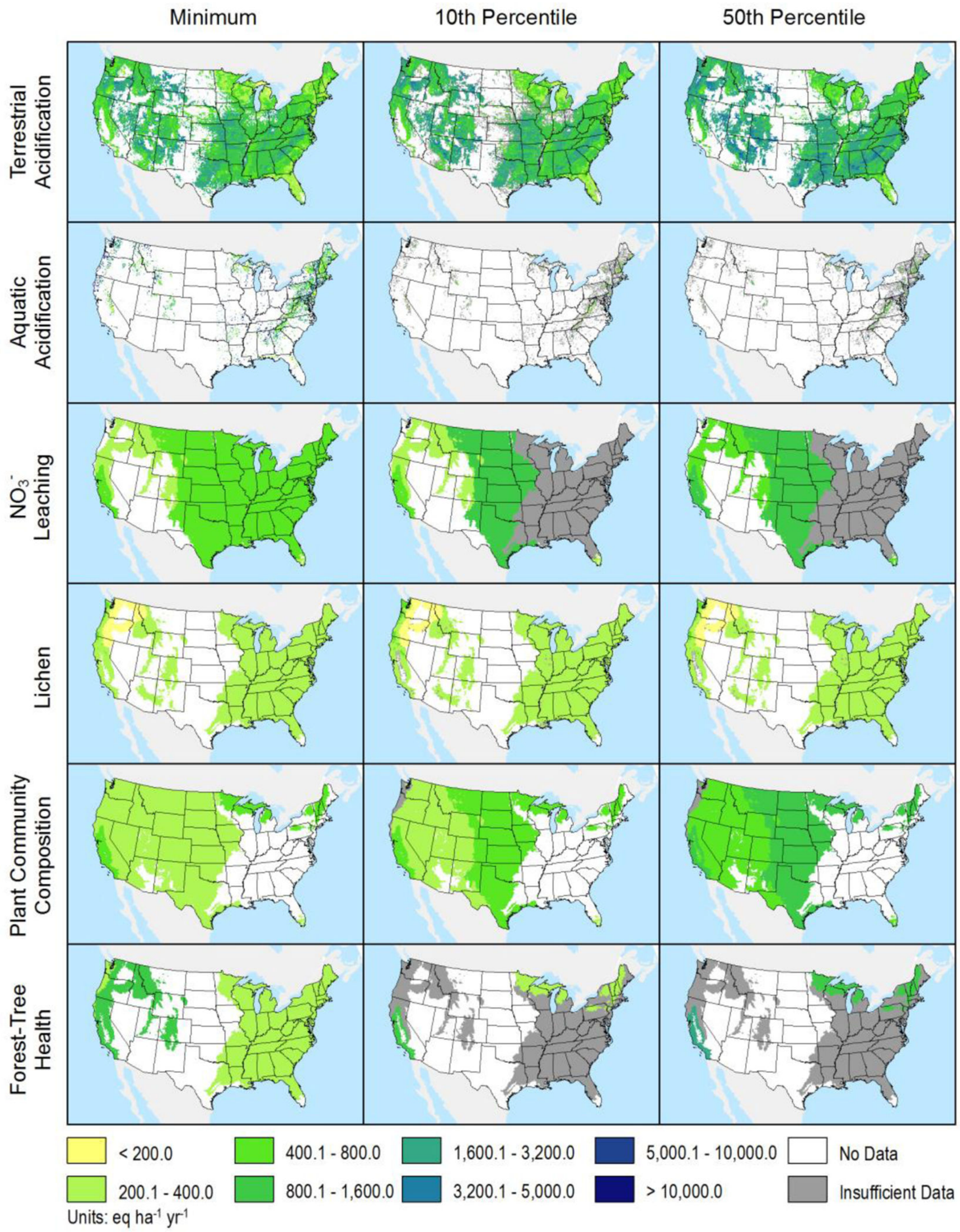

Fig. 1.

Critical loads assessed in this study. Shown are the six CLs (rows) along with the minimum, 10th, and 50th percentiles (columns) of the CL within a grid cell with sufficient data (>5).

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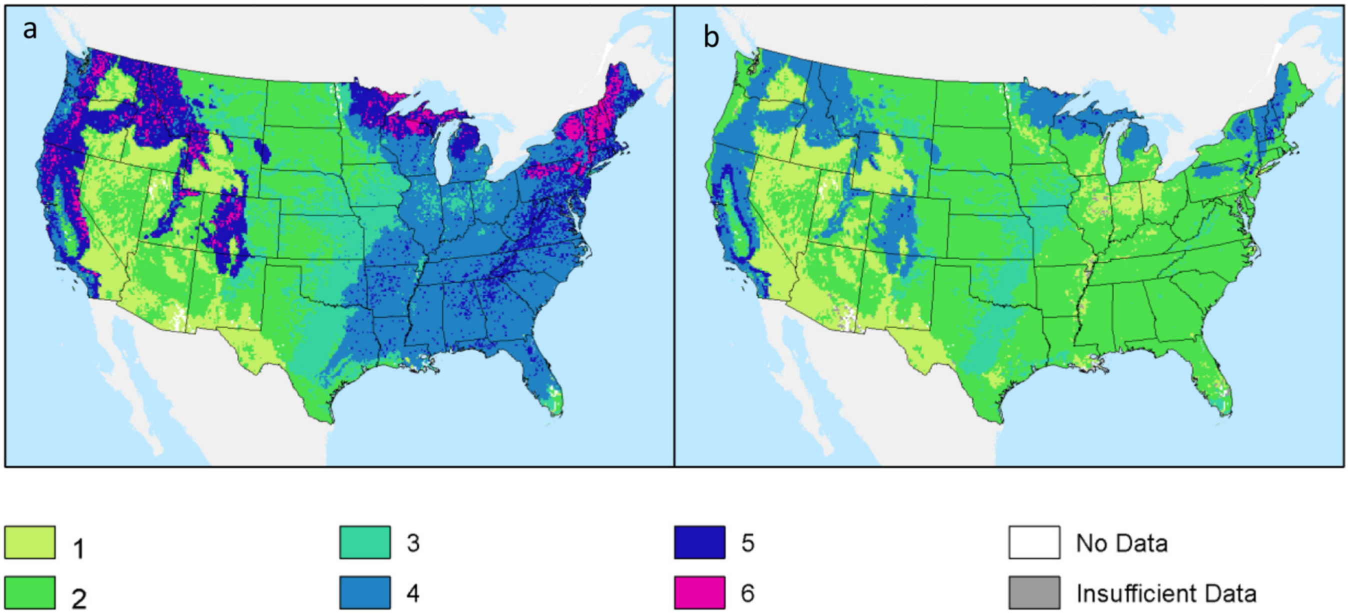

Fig. 2.
The number of CLs per 12-km grid cell for the minimum (a) and 10th/50th percentiles (b).

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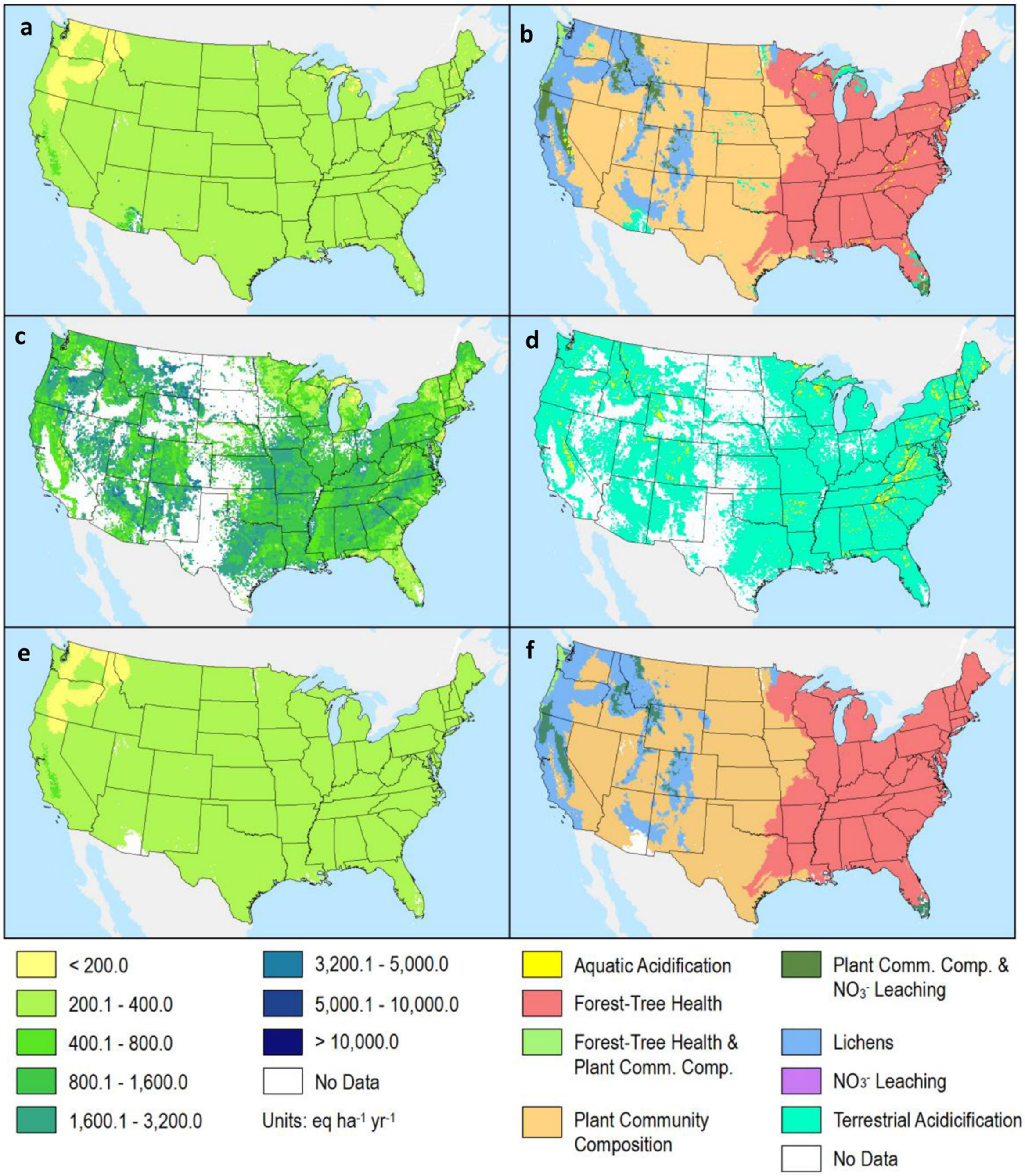

Fig. 3.

The lowest minimum CL value and identity across all CL types (a, b), for acidification CLs of N and S (c, d), and for empirical CLs of N (e, f).

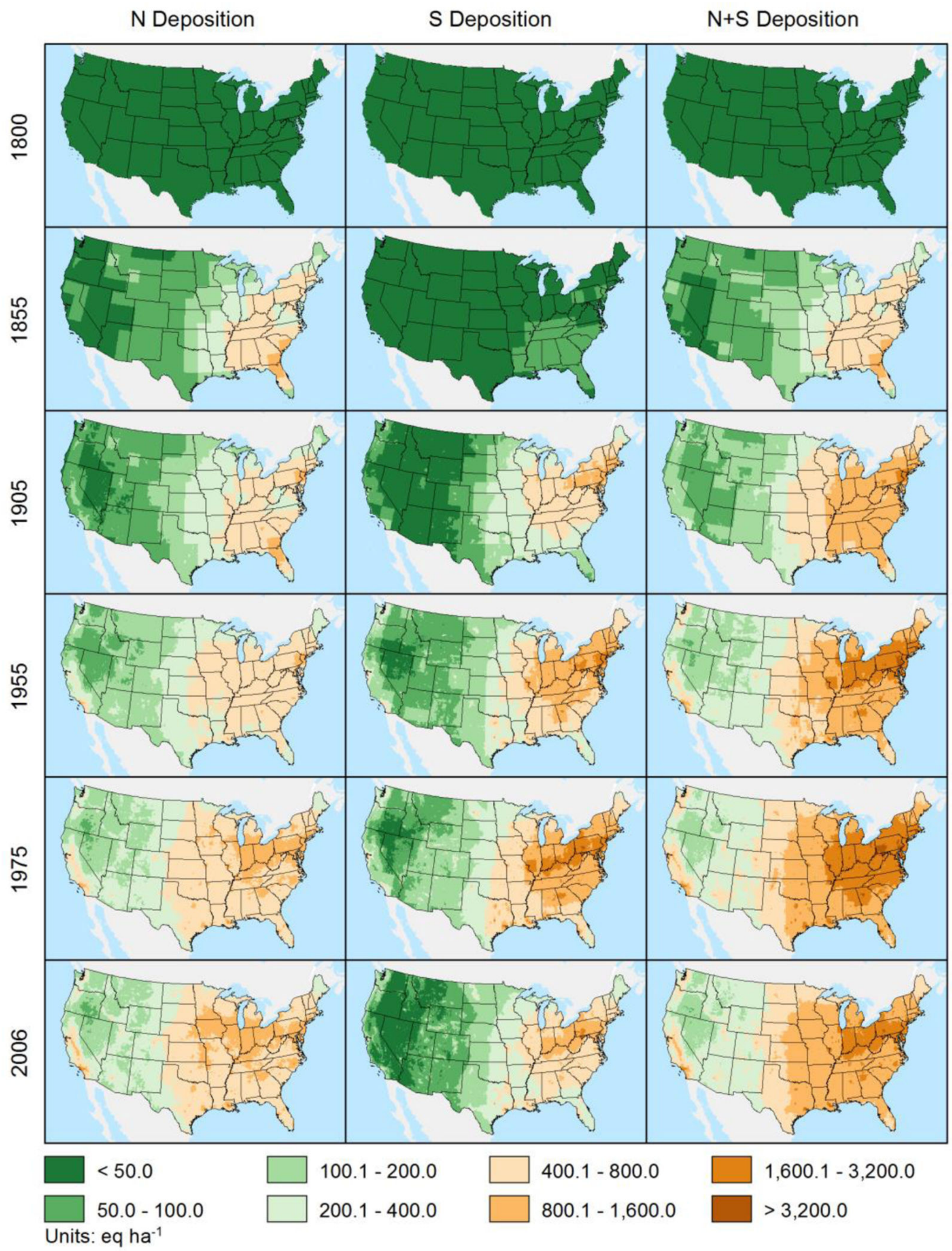

Fig. 4.

The decadal average N, S, and N+S deposition from 1800–2010 labeled by the decadal midpoint. Note: conversion from eq ha^{-1} to kg ha^{-1} is done by dividing by 71.38 for N and by 62.36 for S.

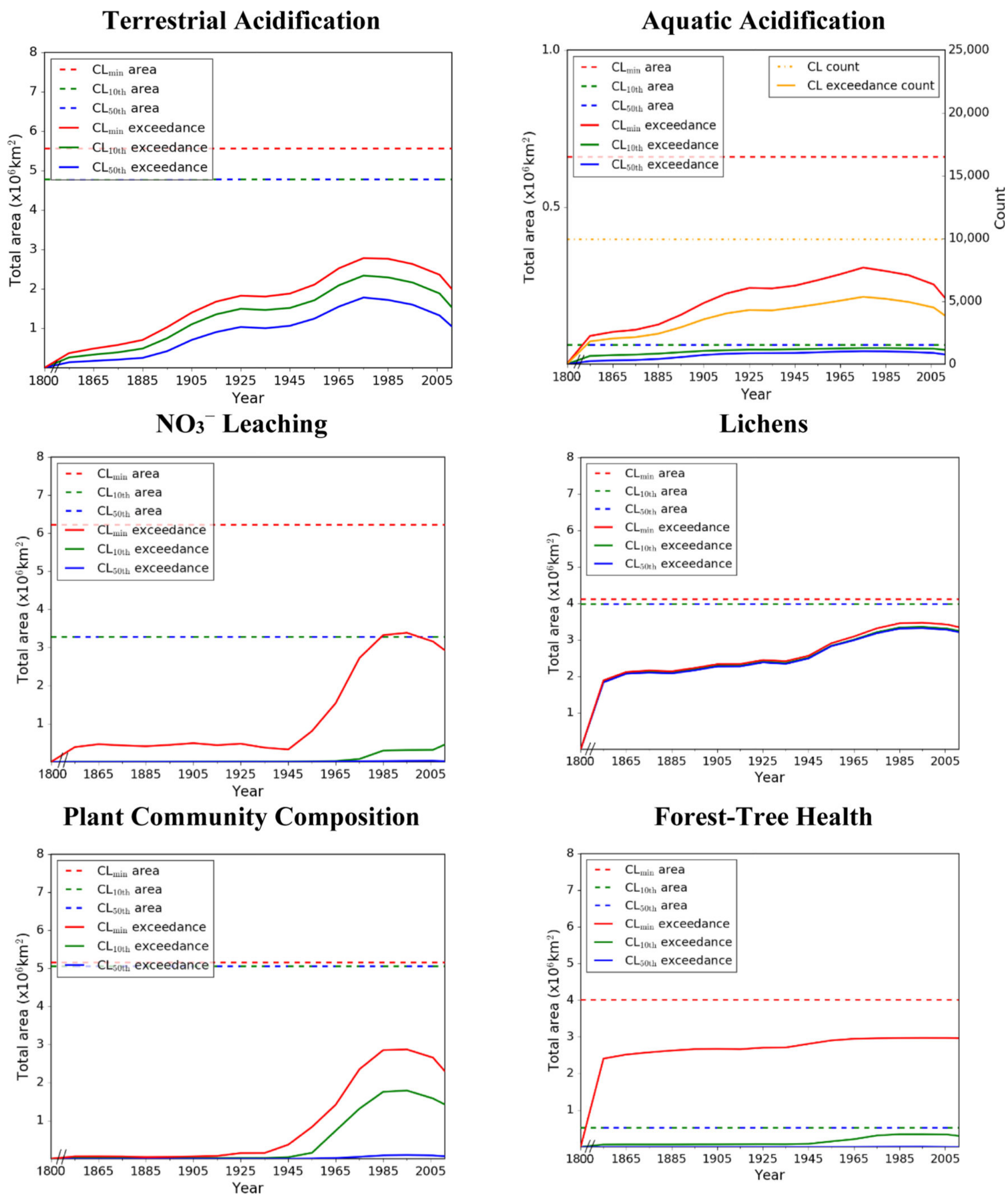

Fig. 5.

Plots of CL exceedances through time (1800–2011) linearly interpolated between decadal midpoints. Areal exceedances (solid lines) for the min (red), 10th percentile (green), and 50th percentile (blue) are shown, along with the total area with data (colored dashed lines). Counts of water bodies with data and exceedances are shown for the Aquatic Acidification CL.

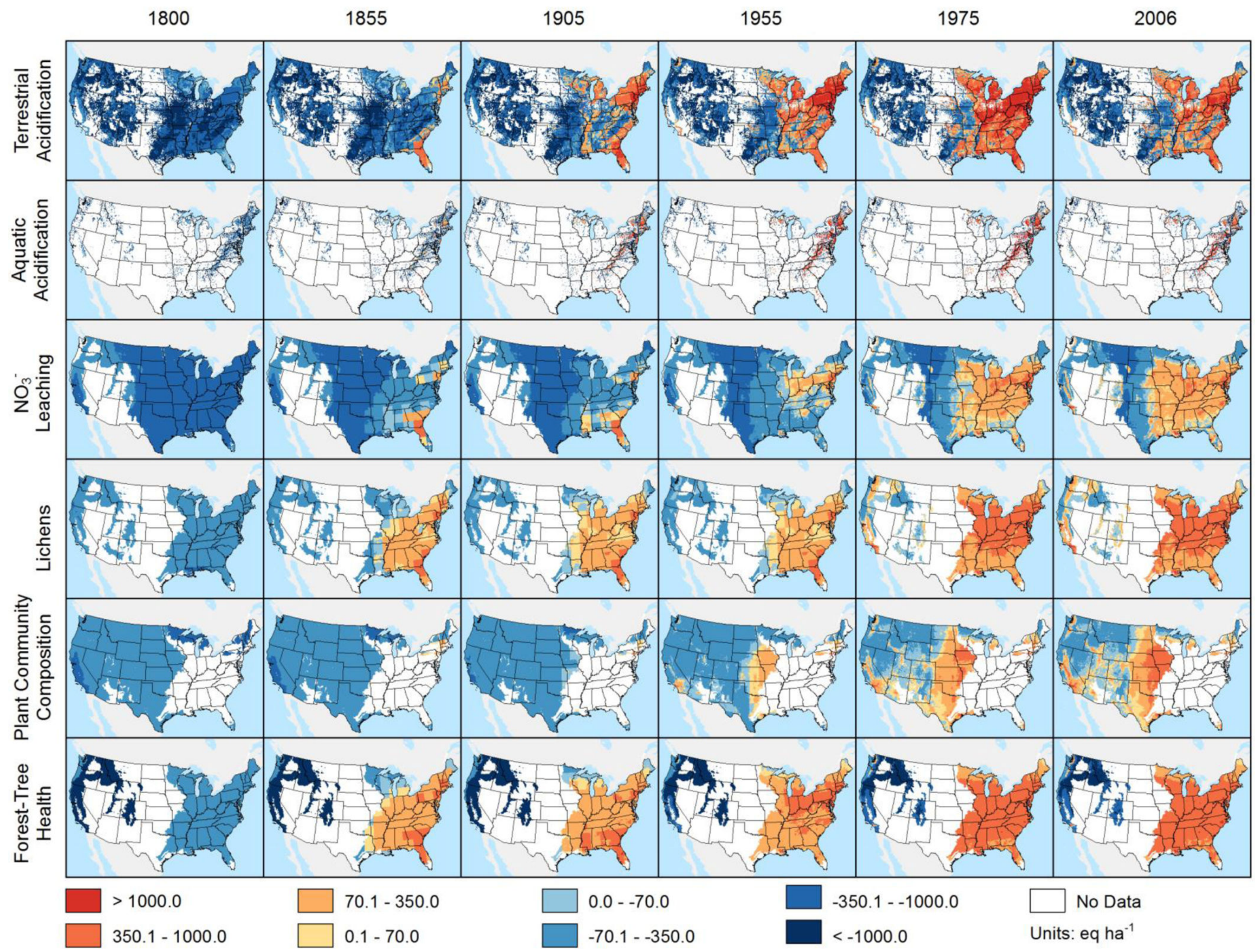

Fig. 6.

Geographic patterns of exceedances through time for individual CL types.

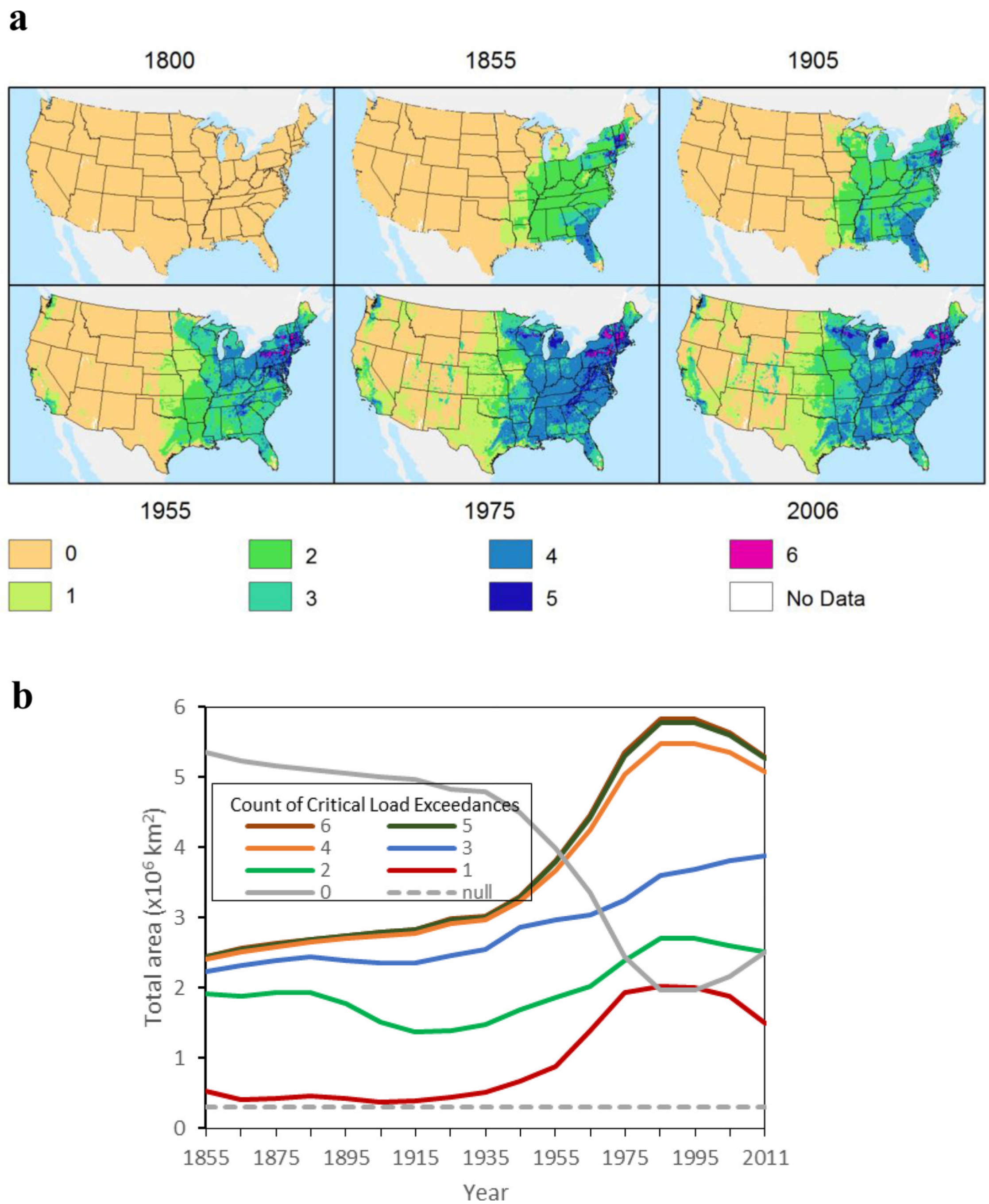

Fig. 7.

Maps (a) of the number of minimum CLs exceeded through time, and (b) temporal plot of the total area of CMAQ grid cells with 0–6 exceedances. The gray dashed line shows the area of the contiguous United States for which there is no CL (“null”).

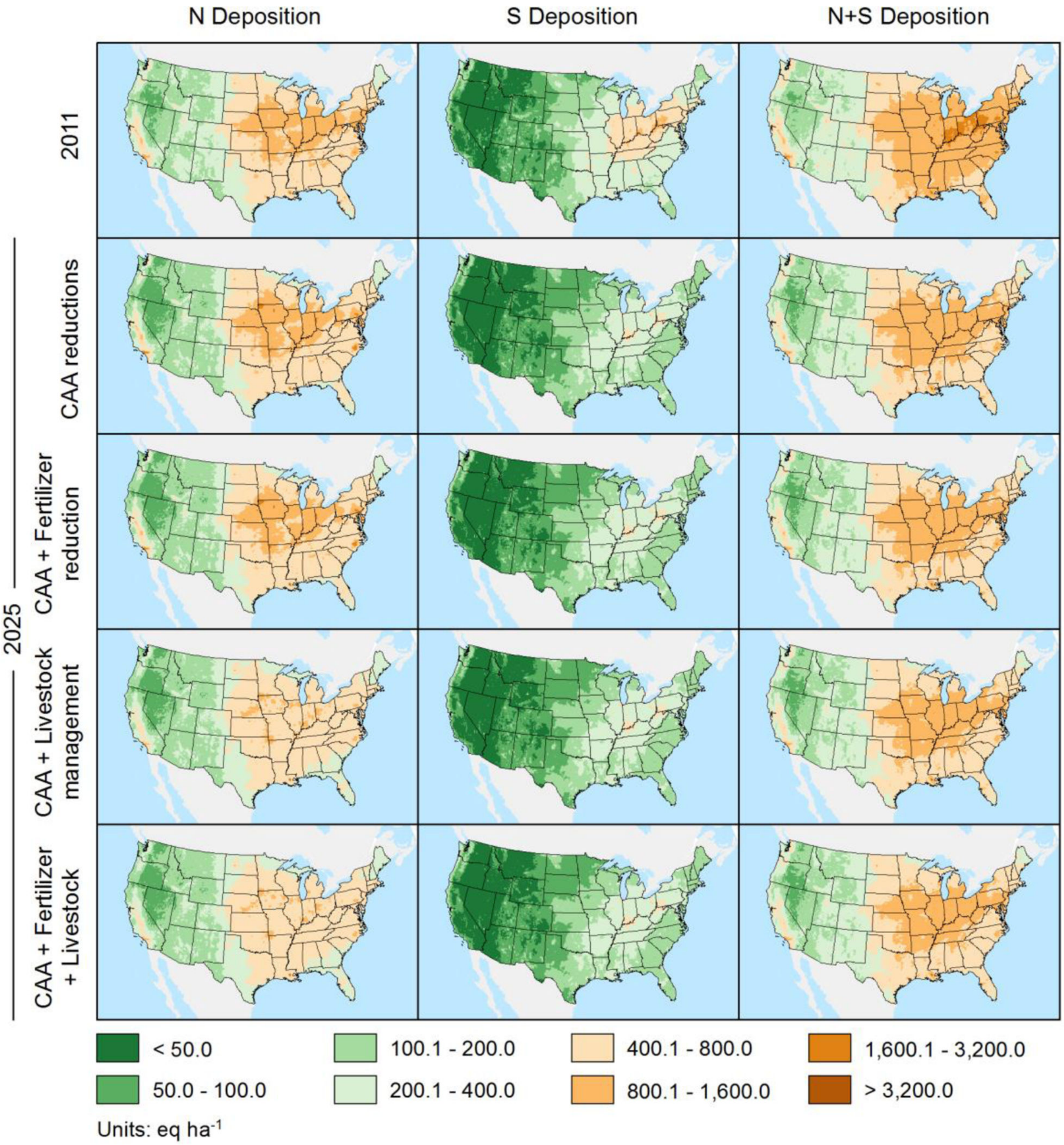

Fig. 8.

Deposition patterns in 2011 and under four different scenarios in 2025 (Table 1). Note that only N emissions are affected by the fertilizer and livestock scenarios.

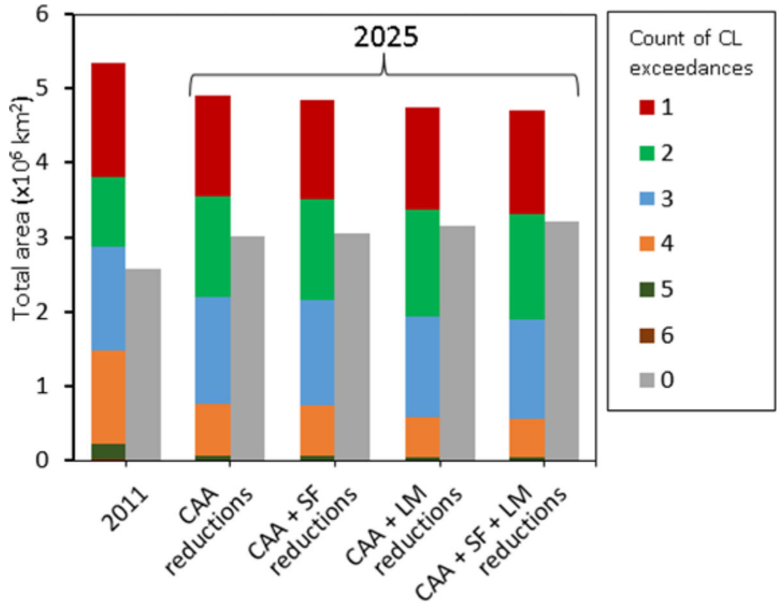

Fig. 9.

Total area and number of minimum CLs exceeded in 2011 and in 2025 for the four scenarios examined.

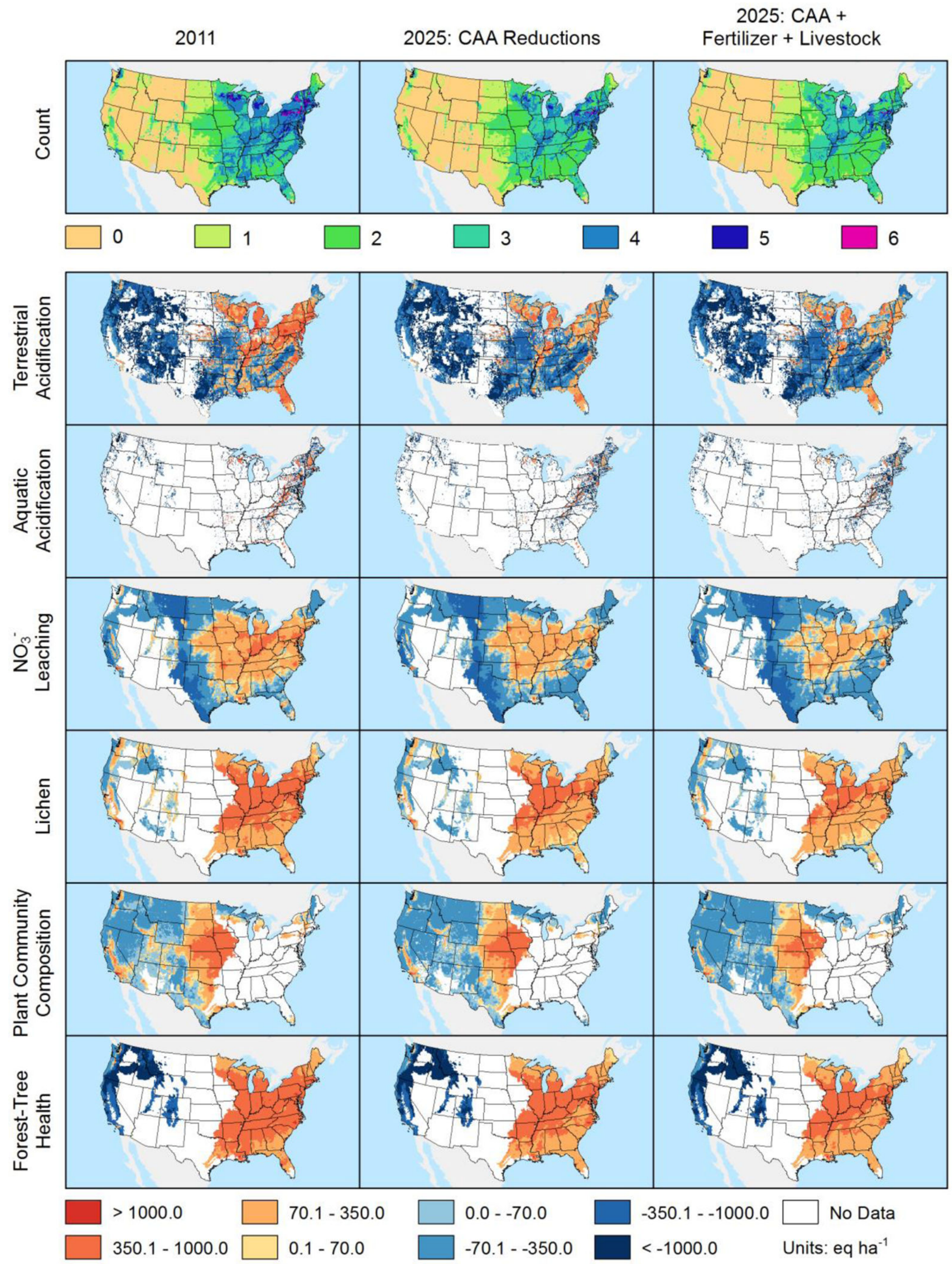

Fig. 10.

Counts of exceedances (upper set) and of individual CLs (lower set) in 2011 (left column) and in 2025 for the CAA scenario (middle column) and the CAA+ fertilizer+livestock (right column) scenario. There were few differences with other future scenarios.

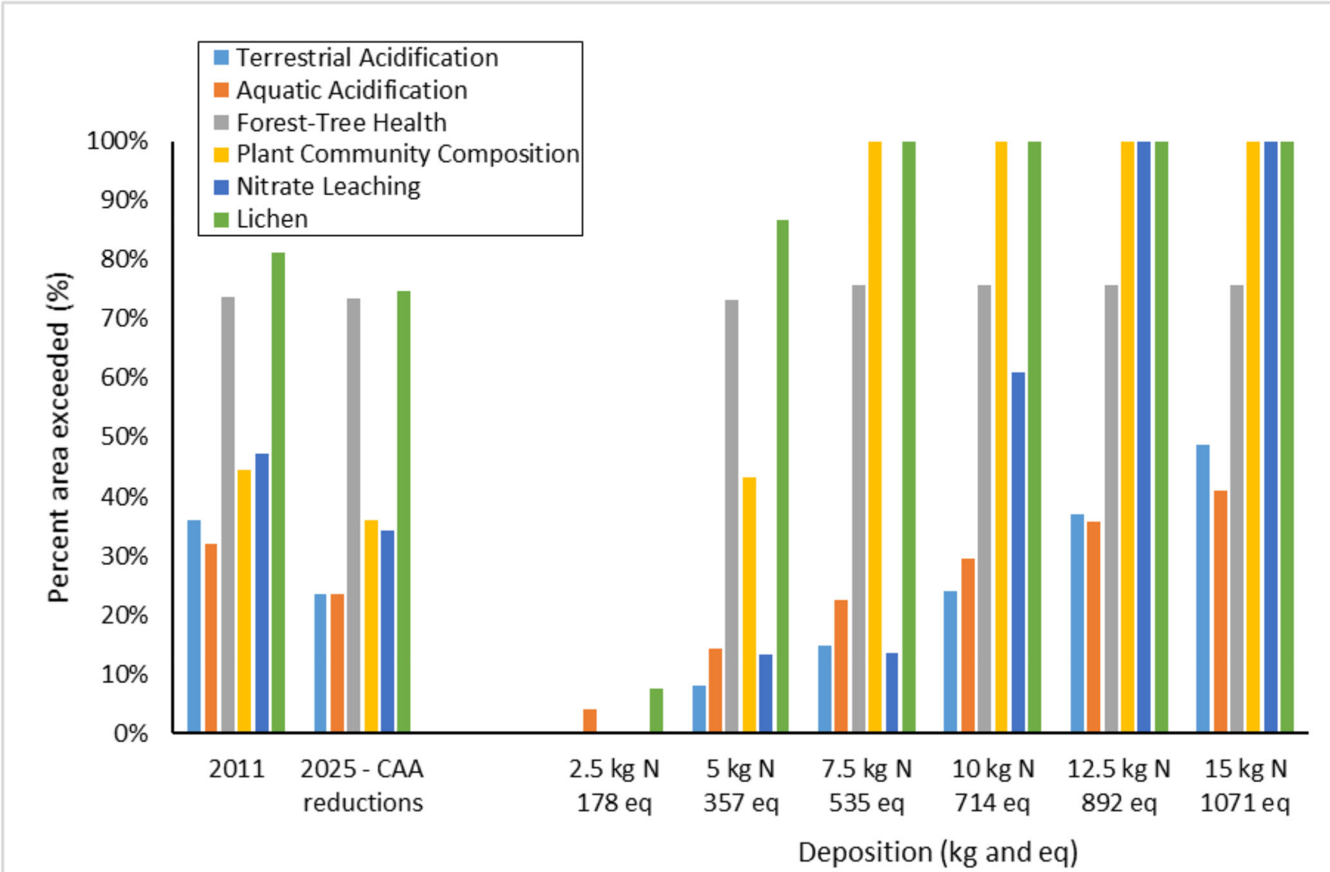

Fig. 11.

Percentage of area with data with minimum CL exceeded for the six CLs in 2011, 2025 (CAA scenario), and using fixed N deposition levels (2.5, 5, 7.5, 10, 12.5, and 15 kg N ha^{-1} yr^{-1}).

Table 1.

Summary of deposition data sources used in this study.

Name (period)	Spatial grain	Description	Source(s)
Pre-industrial (1800)	NA	constant N and S deposition assuming 0.4 kg N ha^{-1} yr^{-1} and 0.1 kg S ha^{-1} yr^{-1}	Lamarque et al. 2010, Baron 2006; Holland et al. 1999; Galloway et al. 2004
Historical (1850–2000)	200 × 200 km	CAM modeled N and S deposition using EDGAR-HYDE inventory and CAM 3.5 atmospheric model, scaled to current.	Lamarque et al. 2010
Current (2002–2011)	12 × 12 km	modeled N and S deposition using SMOKE inventory and CMAQ 5.0.2 bidirectional model	Bash et al. 2013; Foley et al. 2016
Clean Air Act (CAA) reductions (2025)	12 × 12 km	modeled N and S deposition using CAA anticipated 2025 inventory and CMAQ 5.0.2 bidirectional model	Bash et al. 2013, EPA 2014, Robin Dennis, personal communication
CAA + fertilizer reductions (2025)	12 × 12 km	modeled N and S deposition using CAA anticipated 2025 inventory, 20% reduction (~0.2 Tg yr^{-1} reduction) in fertilizer application from 2000–2002 baseline, and CMAQ 5.0.2 bidirectional model	this study
CAA + livestock management (2025)	12 × 12 km	modeled N and S deposition using CAA anticipated 2025 inventory, 30% reduction (~0.7 Tg yr^{-1} reduction) of NH3 from livestock and concentrated animal feeding operations (CAFOs) relative to 1990 baseline, and CMAQ 5.0.2 bidirectional model	this study
CAA + fertilizer + livestock (2025)	12 × 12 km	modeled N and S deposition using CAA anticipated 2025 inventory, both fertilizer and livestock/CAFO reductions above, and CMAQ 5.0.2 bidirectional model	this study

Table 2.

Summary of critical loads information used in this study.

CL type	Pollutant	Spatial grain	Description	Source(s)
Terrestrial acidification	N and S	1 × 1 km	Modeled estimates based on the simple-mass-balance (SMB) model constrained to forested ecosystems	McNulty et al. 2007, McNulty et al. 2010
Aquatic acidification	N and S	points	Modeled estimates based on the simple-mass-balance (SMB) fluxes of cations and anions to aquatic ecosystems	Lynch et al. 2013
Forest-Tree health	N	level 1 ecoregion	Empirical estimates based on field studies that used a variety of approaches, constrained to forest land cover (2011 NLCD)	Pardo et al. 2011a, b
NO_3^- leaching	N	level 1 ecoregion	Empirical estimates based on field studies that used a variety of approaches, constrained to forest, shrubland, and/or herbaceous land cover (2011 NLCD)	Pardo et al. 2011a, b
Plant community composition	N	level 1 ecoregion	Empirical estimates based on field studies that used a variety of approaches, constrained to forest, shrubland, and/or herbaceous land cover (2011 NLCD)	Pardo et al. 2011a, b
Lichen	N	4 × 4 km	Empirical estimates based on comparing changes in lichen community composition along deposition gradients constrained to forest land cover (2011 NLCD)	Geiser et al. 2010; Root et al. 2015

Table 3.

Shown are the minimum, 10th, and 50th percentile for the empirical CLs of nitrogen from Pardo et al. (2011a, b) that were applied to individual NLCD classes. Cells with NA indicate no range was reported and blank cells indicate no CL of that type. For the Northwestern Forested Mountains and Mediterranean California ecoregions, cells with more than one NCLD Class (i.e., have at least two of Forest, Herbaceous, and Shrubland land covers), the full range of CL values associated with the land covers was used. For the other three CLs examined (Lichen, Terrestrial Acidification, Aquatic Acidification), statistics were calculated in each 12-km grid cell rather than at the ecoregion level and thus are not shown here.

Ecoregion	NLCD class	Empirical critical loads: minimum, 10th percentile, 50th percentile per ecoregion (kg N [eq])								
		Nitrate leaching			Plant community composition			Forest-Tree health		
		Min	10th	50th	Min	10th	50th	Min	10th	50th
Northern forests	Forest	8 (571)	NA	NA	7 (500)	8.4 (600)	14 (1000)	3 (214)	5.3 (378)	14.5 (1035)
Northwestern forested mountains	Forest	4 (285)	5.3 (378)	10.5 (750)				17 (1214)	NA	NA
	Herbaceous				4 (286)	4.6 (328)	7 (500)			
Marine west coast	Forest				5 (357)	NA	NA	5 (357)	NA	NA
Eastern temperate forests	Forest	8 (571)	NA	NA				3 (214)	NA	NA
Mediterranean California	Forest	10 (714)	10.7 (764)	13.5 (964)	24 (1713)	24.9 (1778)	28.5 (2035)	17 (1214)	19.2 (1371)	28 (1999)
	Herbaceous	10† (714)	10.4 (743)	12 (857)	6 (428)	6.4 (457)	8 (571)			
	Shrubland				7.8 (557)	8 (571)	8.9 (635)			
Tropical humid forests	Forest	5 (357)	5.5 (393)	7.5 (535)	5 (357)	5.5 (393)	7.5 (535)			
Great Plains (herbaceous)	Herbaceous	10 (714)	11.5 (821)	17.5 (1249)	5 (357)	7 (500)	15 (1071)			
North American desert	Mixed‡				3 (214)	3.5 (250)	5.7 (407)			

† CLs for nitrate leaching in the Mediterranean California ecoregion were from a mixed system of chaparral and oak woodlands, so we assigned those CLs to both Herbaceous and Shrubland NLCD classes.

‡ CLs for this Plant community composition in the North American Desert came from mixed systems of the Southwest near Joshua Tree National Park and so were assigned to Herbaceous, Shrubland, and Forest NLCD classes.