



Exceedance of Critical Loads of Nitrogen and Sulfur Deposition across National Parks

Comparing 2015–2017 CMAQ and TDep model outputs



A close-up of green lung moss on a tree at Katahdin Woods and Waters National Monument.
NPS / GRACE KIRK

Exceedance of critical loads of nitrogen and sulfur deposition across national parks: Comparing 2015–2017 CMAQ and TDep model outputs

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Abstract

Atmospheric nitrogen (N) deposition can alter biological communities through a variety of pathways. The increased growth and/or abundance of some plant and animal species lead to other species being outcompeted and eliminated from the biological community. Deposition of reactive N (Nr) to watersheds can impact both aquatic and terrestrial ecosystem elements and contribute to eutrophication and acidification effects. Atmospheric sulfur (S) deposition has been the primary driver of soil and surface water acidification of acid-sensitive ecosystems, resulting in adverse effects on aquatic biota and forest health. Critical loads (CLs) of atmospheric N and S deposition are used to establish thresholds of ecosystem impacts associated with a given level of deposition. The CLs of N for a decline in sensitive lichen species richness ($3.1 \text{ kg-N ha}^{-1} \text{ yr}^{-1}$) and to protect herbaceous species richness ($6\text{--}16 \text{ kg-N ha}^{-1} \text{ yr}^{-1}$) were chosen to evaluate N impacts at lower and higher levels of deposition. To evaluate S impacts, CLs for a decline in sensitive lichen species richness ($2.3 \text{ kg-S ha}^{-1} \text{ yr}^{-1}$) and the CL to protect acid neutralizing capacity (ANC) of lakes and streams ($0\text{--}31 \text{ kg-S ha}^{-1} \text{ yr}^{-1}$) were chosen.

Locations where the deposition is higher than the CL are considered to have deposition in “exceedance” of the CL. This report quantifies differences in exceedances from atmospheric deposition estimates generated by two modeling approaches (Total Deposition [TDep] and Community Multiscale Air Quality [CMAQ]) using representative annual deposition estimates for 2017 across the conterminous US. Given that TDep represents a measurement-based adjustment of CMAQ model results, this project evaluates the extent to which those adjustments affect the magnitude and spatial extent of critical load exceedances as compared with using deposition estimates directly from CMAQ for all National Park Service (NPS) units with available data. This analysis showed that selection of the deposition data source was most important in consideration of CL exceedance for lichen species richness. The extent of CL exceedance was less affected by the deposition data source for CLs to protect herbaceous species richness and surface water acidification. Results from this study can be used to guide natural resource management and policy in the context of understanding uncertainty in air pollution effects caused by atmospheric N and S deposition across a broad set of federally protected NPS units. The results can also provide insight into where local deposition measurements elucidate risk to ecosystem health.

Acknowledgments

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Introduction

Atmospheric nitrogen (N) deposition affects ecosystem components and function by changing nutrient loads and competitive dynamics. Some plant species increase growth and abundance, while other species can be outcompeted and eliminated from the biological community (Clark and Tilman 2008). N-sensitive lichen species are lost from ecosystems exposed to elevated N deposition, leading to declines in species richness (Geiser et al. 2019). In addition, deposition of reactive N (Nr) to watersheds can impact both aquatic and terrestrial ecosystem elements and contribute to eutrophication and acidification (Pardo et al. 2011). Atmospheric sulfur (S) deposition has been the primary driver of soil and surface water acidification effects in acid-sensitive ecosystems resulting in adverse effects on aquatic biota and forest health.

Wet deposition is routinely measured at monitoring sites throughout the United States. Dry deposition is modeled based on measurements at specific sites. Chemical transport models (CTMs) determine deposition in the areas between measurement sites. Such models evaluate air pollutant emissions and atmospheric and environmental processes to predict deposition across the broader landscape. These spatial deposition estimates can be used to evaluate changes to ecosystem processes and determine critical loads (CLs; deposition “tipping points”) and their exceedances. The CL is an estimate of the pollutant exposure below which specified harmful effects do not occur according to current knowledge (Nilsson and Grennfelt 1988). Critical loads have been developed in the United States for various ecosystem elements including: forest-tree health, plant community composition, and lichen biodiversity (Clark et al. 2018, Geiser et al. 2019). An exceedance occurs when deposition is higher than the CL. Ambient deposition estimates are needed for calculating CL exceedances. CL exceedances can affect ecosystem services such as biodiversity, tree growth, recreation, and cultural and aesthetic values (Compton et al. 2011, Munns et al. 2016). Critical load exceedances are determined using CTMs (Simkin et al. 2016, Clark et al. 2018, Makar et al. 2018) or measurement-model fusion (MMF) approaches (c.f., Schwede and Lear 2014, Nanus et al. 2017, McDonnell et al. 2018, U.S. Environmental Protection Agency 2019). Measurement-model fusion techniques are considered to be more accurate than CTMs because the former incorporate available measurement data (Walker et al. 2019).

Modeling of atmospheric deposition across National Park Service (NPS) lands has been an active area of research for several decades because of NPS mandates to protect national parks for future generations (Tonnesen et al. 2003, Schwede and Lear 2014). Nitrogen (N) and sulfur (S) atmospheric deposition effects studies in national parks have commonly relied on a combination of spatial interpolation of measured wet deposition data and process modeled dry deposition derived from the Community Multiscale Air Quality (CMAQ) model. Other approaches for estimating total deposition, including estimates based on throughfall chemistry (cf., Weathers et al. 2006), have also been used to improve understanding of the effects of air pollution on sensitive resources. An MMF approach was developed by the National Atmospheric Deposition Program (NADP) Total Deposition (TDep) Science Committee to leverage the strengths of both the monitoring and modeling approaches (Schwede and Lear 2014).

Various CTMs have been used by federal agencies to determine the total deposition to public lands. Preference for which CTM is used for a given purpose varies by study region, agency, and project. Each model uses a unique set of inputs and equations to determine how emissions move from their sources to down-wind ecosystem sinks. Variability among CTM estimates of deposition reflect the complexity and uncertainty of vegetation canopy-atmosphere exchange processes (Walker et al. 2020).

Modeled CLs and exceedances are used to inform public policy and decision-making. Differences among modeling methods influence interpretation of CL exceedances and the extent to which they reflect the likelihood of ecological damage caused by pollutant deposition. For the NPS, it is important to quantify how inconsistencies among models change the patterns in calculated CL exceedances across NPS units. The primary objectives of this research were to (1) calculate CL exceedances using known CLs and deposition estimates from both methods (TDep and CMAQ); and (2) compare modeled deposition and the spatial extent of CL exceedances across all NPS units of the conterminous US (CONUS) for which data were available.

Methods

Deposition Data

Spatial datasets describing annual total (wet + dry) N and S deposition developed using the TDep (Schwede and Lear 2014) and CMAQ (Appel et al. 2021) methods were obtained for the CONUS from NADP (<https://nadp.slh.wisc.edu/committees/tdep/>; v2022.02) and U.S. EPA (Foley et al. 2023; v5.3.2; EQUATES; bias-adjusted). Annual deposition was expressed as a three-year average leading up to and including 2017 for both methodologies. These years were chosen as the latest time-period for which both TDep and CMAQ data were available at time of the analysis. Deposition units were reported as kg-N ha⁻¹ yr⁻¹ or kg-S ha⁻¹ yr⁻¹.

Given that TDep represents a measurement-based adjustment of CMAQ model results, this project evaluates the extent to which those adjustments impact estimates of CL exceedance. Locations showing differences in exceedances may be areas in which additional deposition measurements would benefit the TDep data product.

Evaluation of Critical Load Exceedance

Exceedances of CLs of N deposition and S deposition (as applicable) were evaluated for sensitive lichen species richness, herbaceous species richness, and aquatic biota. Comparisons of the spatial extent of CL exceedance were made across NPS units for which CLs were available (Figure 1).

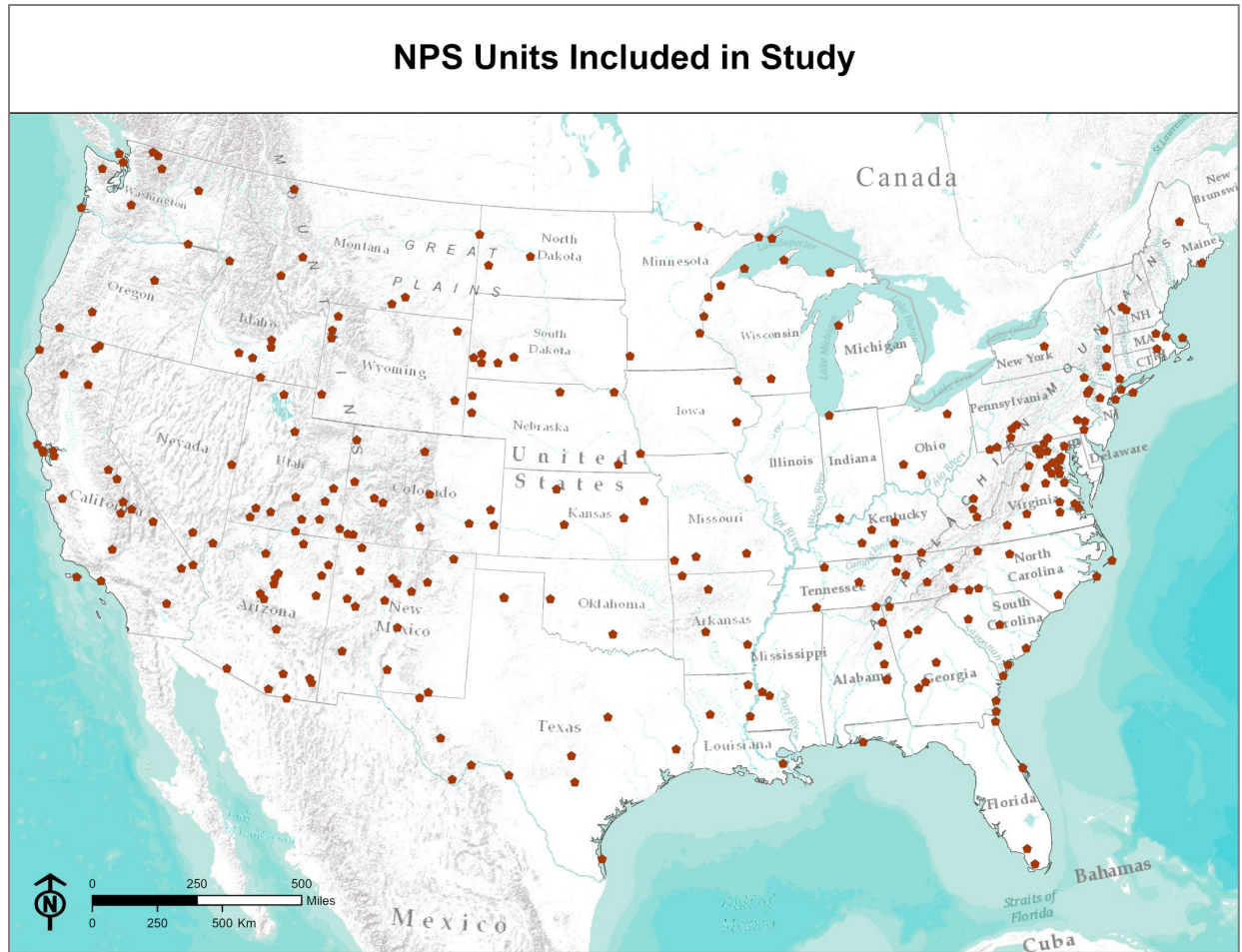


Figure 1. NPS units included in this study.

Critical loads reported by Geiser et al. (2019) were used as the basis for evaluating critical load exceedance for lichens: CL of N = $3.1 \text{ kg-N ha}^{-1} \text{ yr}^{-1}$ and CL of S = $2.3 \text{ kg-S ha}^{-1} \text{ yr}^{-1}$ for responses of N- and S-sensitive lichen species richness. These critical loads were applied only to forested areas of each park, as defined by the 2021 National Land Cover Database (NLCD; Dewitz 2023) and aggregated to 240 m gridded resolution. Aggregated grid cells with < 10% forest cover were removed.

Critical loads for a decrease in herbaceous species richness were adapted from models developed by Simkin et al. (2016). Herbaceous critical loads for closed-canopy conditions were calculated using soil pH data (0 – 20 cm depth; USDA NRCS 2020) aggregated to 240 m gridded resolution. Critical loads for open-canopy conditions were calculated with soil pH (0 – 20 cm depth; USDA NRCS 2020; 240 m resolution), together with annual total precipitation and annual mean air temperature (30 year average; PRISM Climate Group 2014a, b; 800 m resolution). The closed-canopy model was applied to forested areas in the US as defined by the 2021 NLCD, while the open-canopy model was applied to grassland and shrubland landcover; both aggregated to 240 m gridded resolution with cells < 10% cover removed. These values are spatially variable and range from 7.3 to $8.9 \text{ kg-N ha}^{-1} \text{ yr}^{-1}$ for

closed canopy conditions (forests) and 8.7 to 15.9 kg-N ha⁻¹ yr⁻¹ for open-canopy conditions [grasslands and shrub lands; Simkin et al. (2016)].

Critical loads reported in the National Critical Loads Database (NCLD; v3.2.1; Lynch et al. 2022) were used to evaluate exceedance of CLs associated with surface water acidification. These aquatic critical loads were determined based on three approaches including: 1) standard steady-state mass balance modeling (Henriksen and Posch 2001); 2) substitution of statistically-based estimates of base cation weathering into the standard steady-state mass balance modeling approach (McDonnell et al. 2014); and 3) dynamic biogeochemical modeling using the Model of Acidification of Groundwater In Catchments (MAGIC) and the Photosynthesis and net Evapotranspiration and Biogeochemical (PnET-BGC) models (Sullivan et al. 2005, Fakhraei et al. 2014). In addition to CLs associated with the effects of S deposition, aquatic CLs that account for ambient N leaching were also considered.

Estimates of total N and S deposition based on TDep and CMAQ were used to calculate the spatial extent of CL exceedance for each biological receptor and threshold for NPS units for which CL data were available.

Results

National Scale Deposition Comparison

N Deposition

Differences between the two approaches for estimating total N deposition were spatially heterogeneous (Figure 2). Locations where total N deposition estimated by CMAQ was more than 4 kg-N ha⁻¹ yr⁻¹ greater than TDep were mostly located in close proximity to densely populated urban areas. Areas where TDep was more than 4 kg-N ha⁻¹ yr⁻¹ greater than CMAQ were mostly located in the Sierra Nevada mountains and the area surrounding Salt Lake City. Nearly all areas east of the Great Basin with meaningful differences (i.e., greater than 1 kg-N ha⁻¹ yr⁻¹) between TDep and CMAQ were characterized by higher N deposition from CMAQ. However, TDep was more than 4 kg-N ha⁻¹ yr⁻¹ higher than CMAQ in the areas of South Dakota, Pennsylvania, and South Carolina.

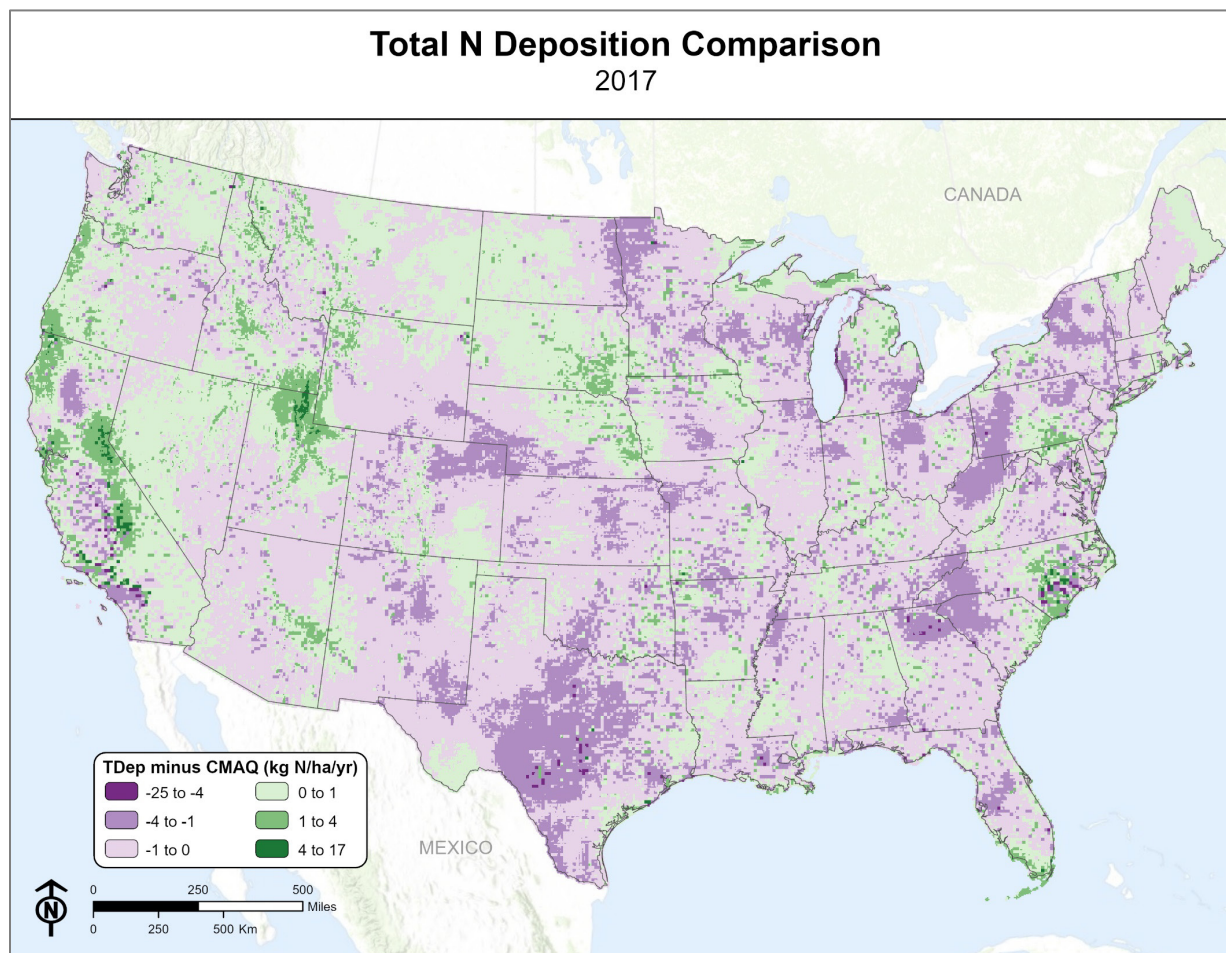


Figure 2. Absolute difference between TDep and CMAQ total N deposition representative of year 2017 throughout the conterminous US. Green colors indicate that deposition estimates from TDep are higher and purple colors indicate that CMAQ estimates are higher.

As compared with results shown in an earlier study focused on comparisons between TDep and CMAQ for years 2003 and 2011 (McDonnell et al. 2020), these 2017 data showed more areas with CMAQ greater than 1 kg-N ha⁻¹ yr⁻¹ higher than TDep and fewer areas with TDep greater than 4 kg-N ha⁻¹ yr⁻¹ higher than CMAQ.

S Deposition

Both deposition methods produced similar results (i.e., within 1 kg-S ha⁻¹ yr⁻¹) for S deposition across the vast majority of the CONUS (Figure 3). Substantially higher deposition from CMAQ occurred almost exclusively in isolated areas of the midwest and eastern US. Sulfur deposition from TDep was greater than 1 kg-S ha⁻¹ yr⁻¹ higher in isolated areas around the Pacific Northwest, San Francisco, Salt Lake City, and in portions of the eastern US (VA, WV, and FL).

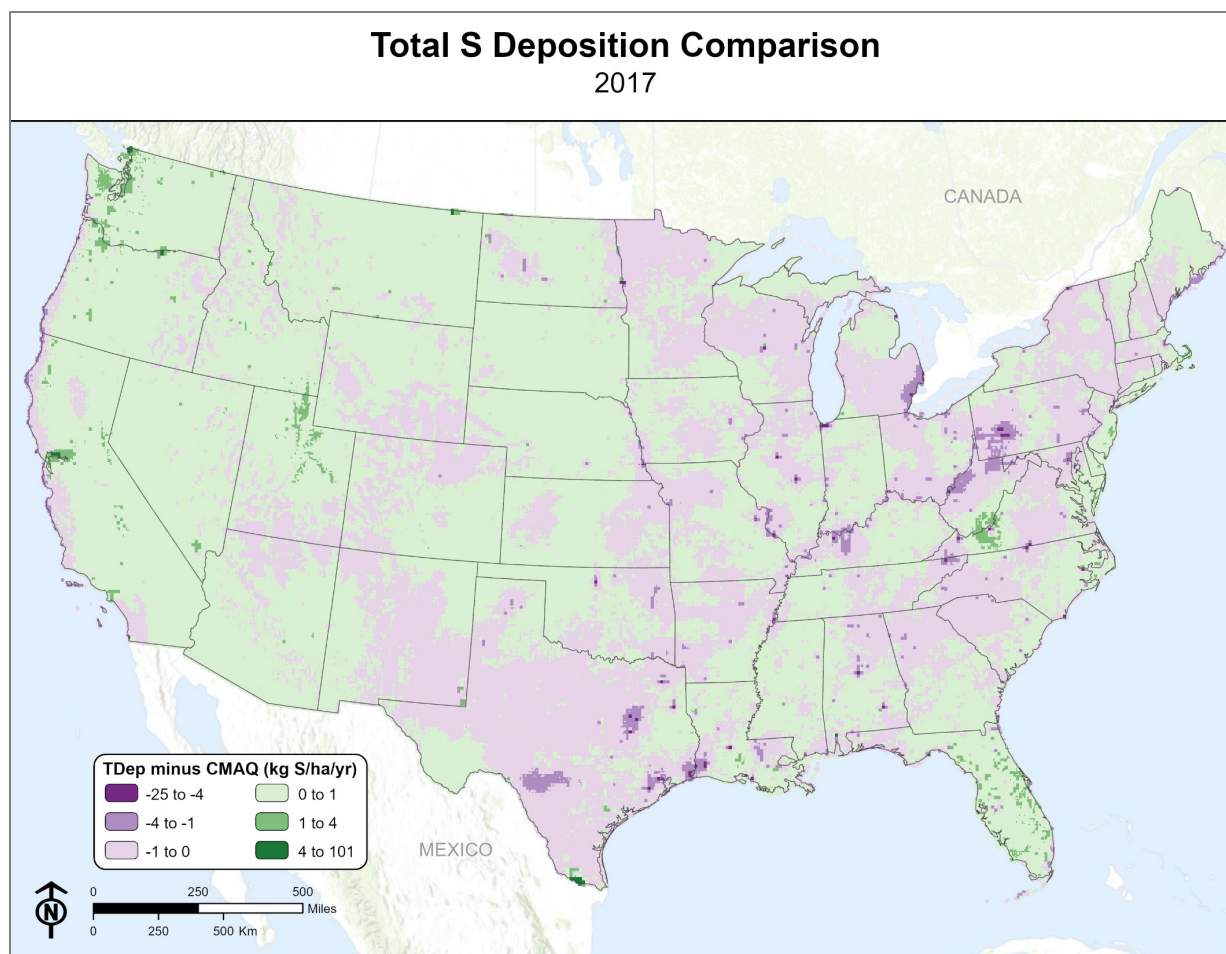


Figure 3. Absolute difference between TDep and CMAQ total S deposition representative of year 2017 throughout the conterminous US. Green colors indicate that deposition estimates from TDep are higher and purple colors indicate that CMAQ estimates are higher.

Park-Scale Critical Load Exceedance

Lichen Species Richness

Nitrogen

Substantial differences (more than 33%) in the percentage of NPS land area in exceedance of the CL to protect N-sensitive lichen species richness ($CL_{N,Lichen}$) were determined between the two models for 25 of the 264 NPS units evaluated (Appendix A). Spatial patterns in the TDep/CMAQ exceedance comparison for $CL_{N,Lichen}$ showed that substantial differences in $CL_{N,Lichen}$ exceedance typically occurred toward the central-western portion of the CONUS from North Dakota through Texas (Figure 4). Given that western NPS units generally experience deposition levels closer to the lichen CL of N, relatively small shifts in deposition are more likely to change exceedance status.

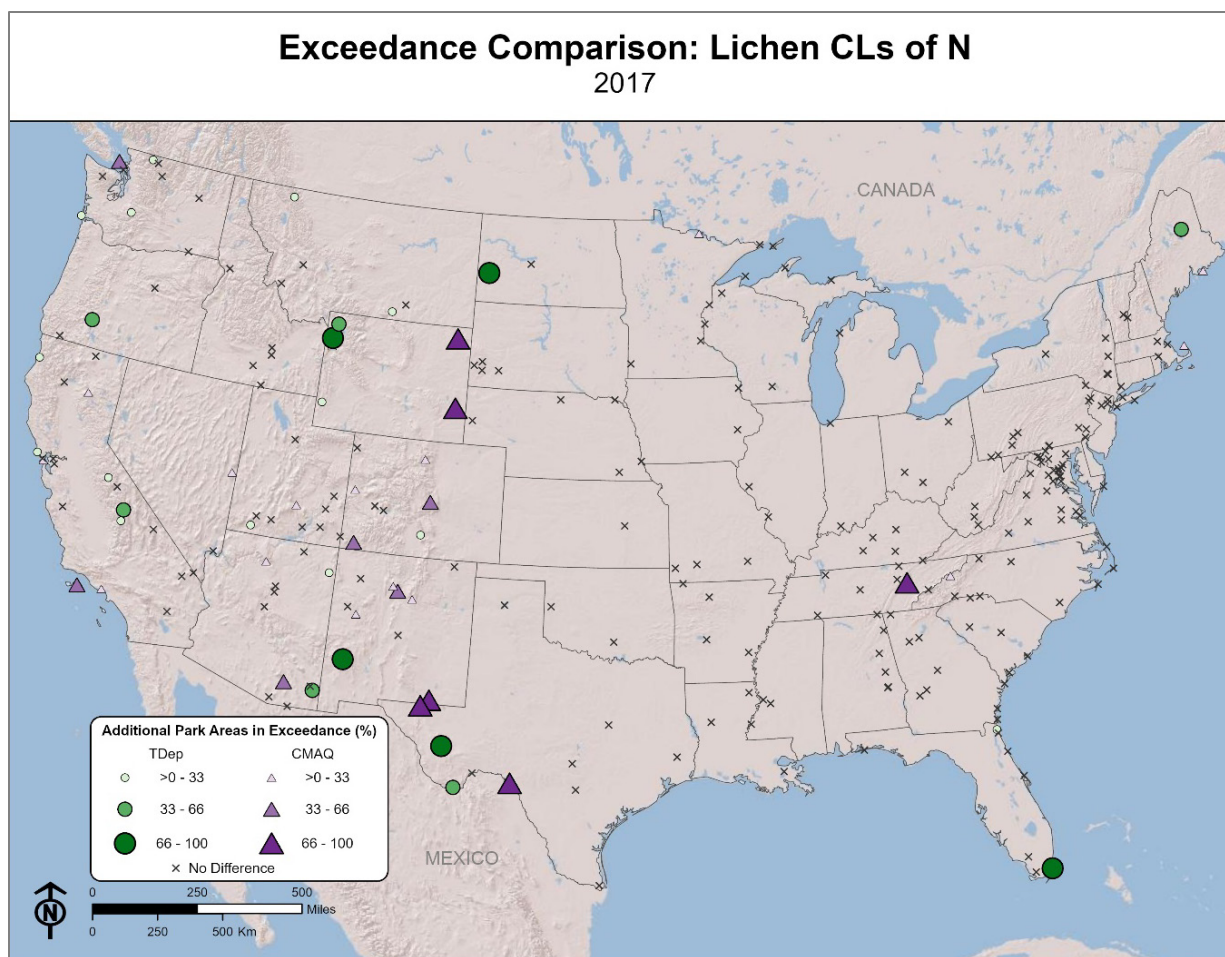


Figure 4. Spatial patterns in additional park area in exceedance of critical loads (CLs) of N-sensitive lichen species richness ($3.1 \text{ kg-N ha}^{-1} \text{ yr}^{-1}$) based on TDep or CMAQ total N deposition representative of year 2017. Green shaded circles indicate that more park area was in exceedance based on TDep. Purple shaded triangles indicate more area of exceedance based on CMAQ.

Sulfur

Substantial differences in the percentage of NPS land area in exceedance of the CL to protect S-sensitive lichen species richness ($CL_{S,Lichen}$) were determined between the two models for 37 of the 264 NPS units evaluated (Appendix A). Spatial patterns in the TDep/CMAQ exceedance comparison for $CL_{S,Lichen}$ showed that the vast majority of substantial differences in $CL_{S,Lichen}$ exceedance occurred in the eastern US (Figure 5). These areas are near S emission sources and represent areas that are near the CL.

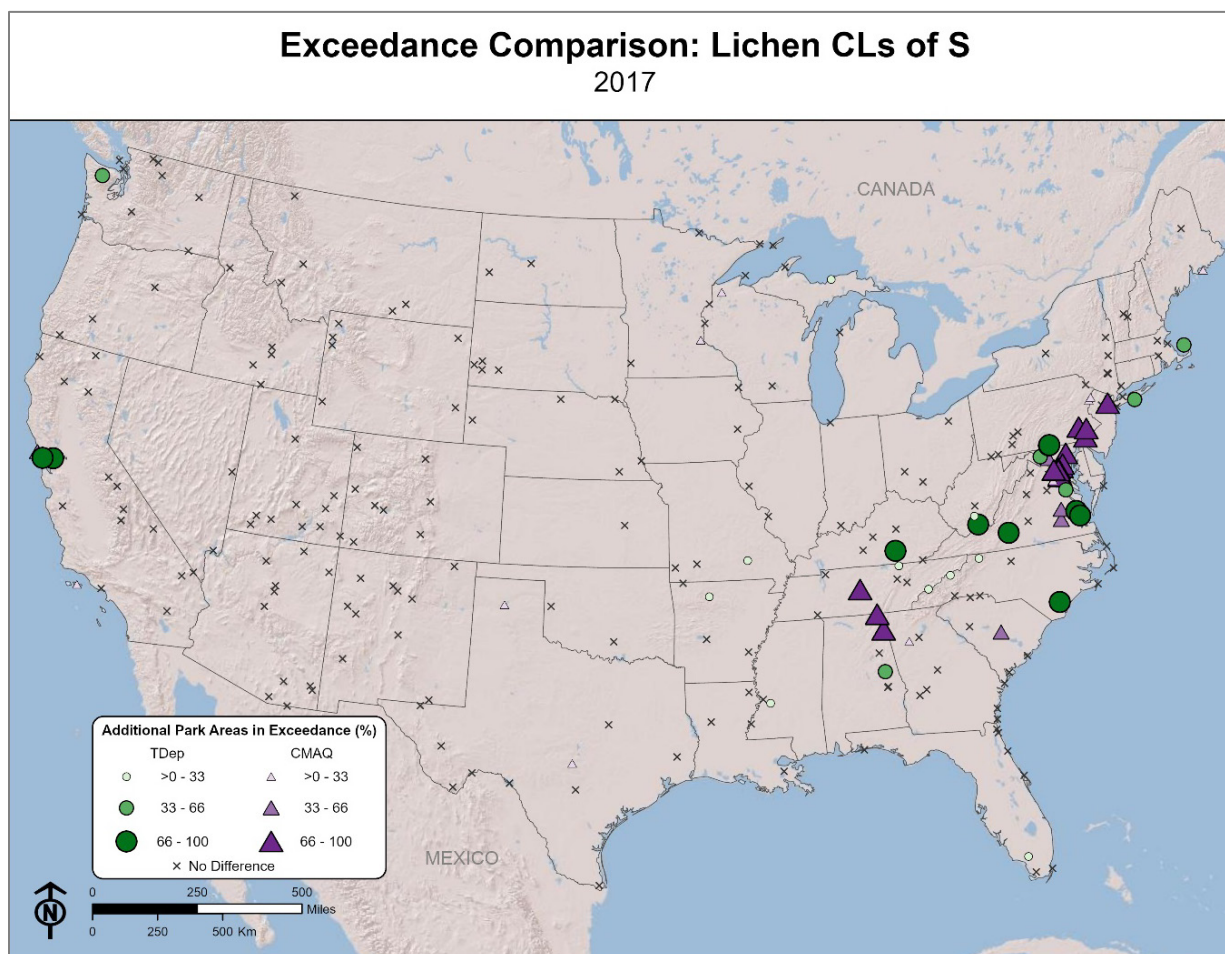


Figure 5. Spatial patterns in additional park area in exceedance of critical loads (CLs) of S-sensitive lichen species richness ($2.5 \text{ kg-S ha}^{-1} \text{ yr}^{-1}$) based on TDep or CMAQ total S deposition representative of year 2017. Green shaded circles indicate that more park area was in exceedance based on TDep. Purple shaded triangles indicate more area of exceedance based on CMAQ.

Herbaceous Species Richness

Few NPS units ($n = 5$) showed substantial differences between the two models in the percentage of land area in exceedance of the CL of N to protect herbaceous species richness ($CL_{N,Herb}$) among the 281 NPS units evaluated (Appendix A). Spatial patterns in the TDep/CMAQ exceedance comparison for $CL_{N,Herb}$ showed that these differences in $CL_{N,Herb}$ exceedance were mostly in California, but also occurred in Kansas and Pennsylvania (Figure 6). These CLs are higher across the US and have fewer

exceedances, so while changes in deposition occur at the sites it is not enough to change exceedance rates.

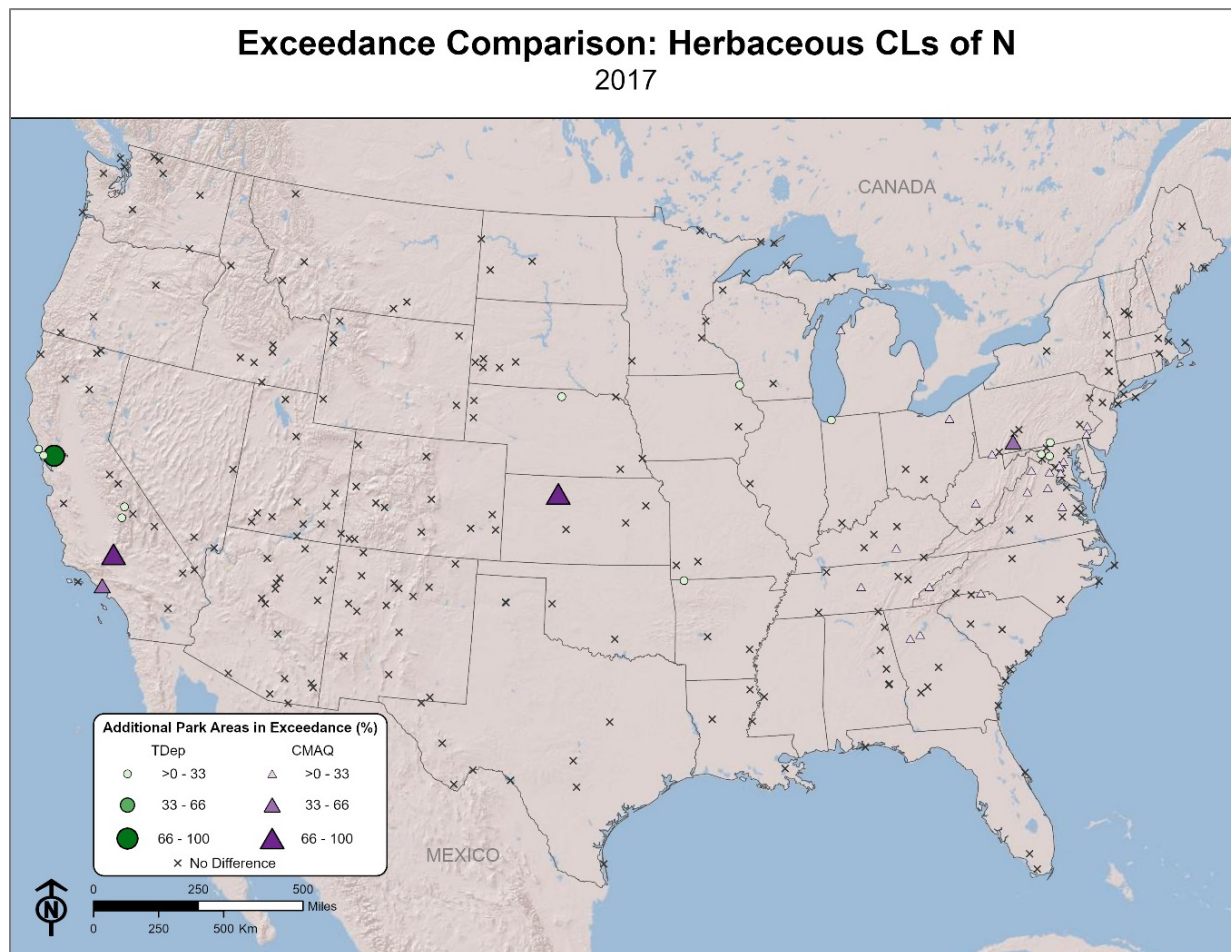


Figure 6. Spatial patterns in additional park area in exceedance of critical loads (CLs) of N deposition for herbaceous species richness based on TDep or CMAQ total N deposition representative of year 2017. Green shaded circles indicate that more park area was in exceedance based on TDep. Purple shaded triangles indicate more area of exceedance based on CMAQ.

Surface Waters

None of the National Park Service units showed substantial differences between the two models in the percentage of land area in exceedance of the CL of S to protect surface waters ($CL_{S.Aquatic}$) among the 56 National Park Service units evaluated. Spatial patterns in the TDep/CMAQ exceedance comparison for $CL_{S.Aquatic}$ showed that although some differences in $CL_{S.Aquatic}$ exceedance occurred (Figure 7), these differences were minor (i.e., only four NPS units showed relatively small differences) for CLs that do not account for the effect of N leaching. Differences were essentially non-existent when N leaching was included in the derivation of CL exceedance (Figure 8).

Exceedance Comparison: Aquatic CLs of S 2017

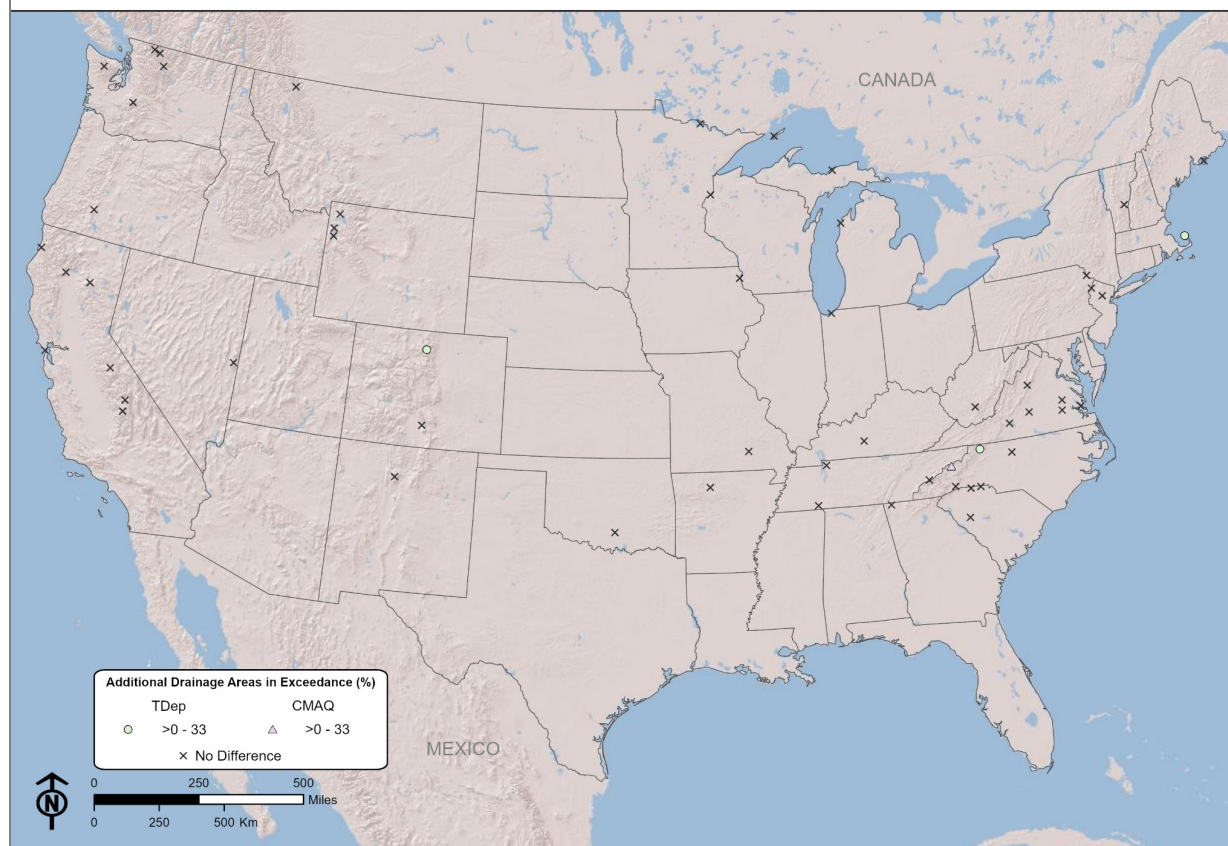


Figure 7. Spatial patterns of additional drainage areas in exceedance of critical loads (CLs) of S deposition for surface waters, without consideration of N leaching, based on TDep or CMAQ total S deposition representative of year 2017. Green circles indicate that more stream sites were in exceedance based on TDep. Purple triangles indicate more sites in exceedance based on CMAQ.

Exceedance Comparison: Aquatic CLs of S (with N Leaching)

2017

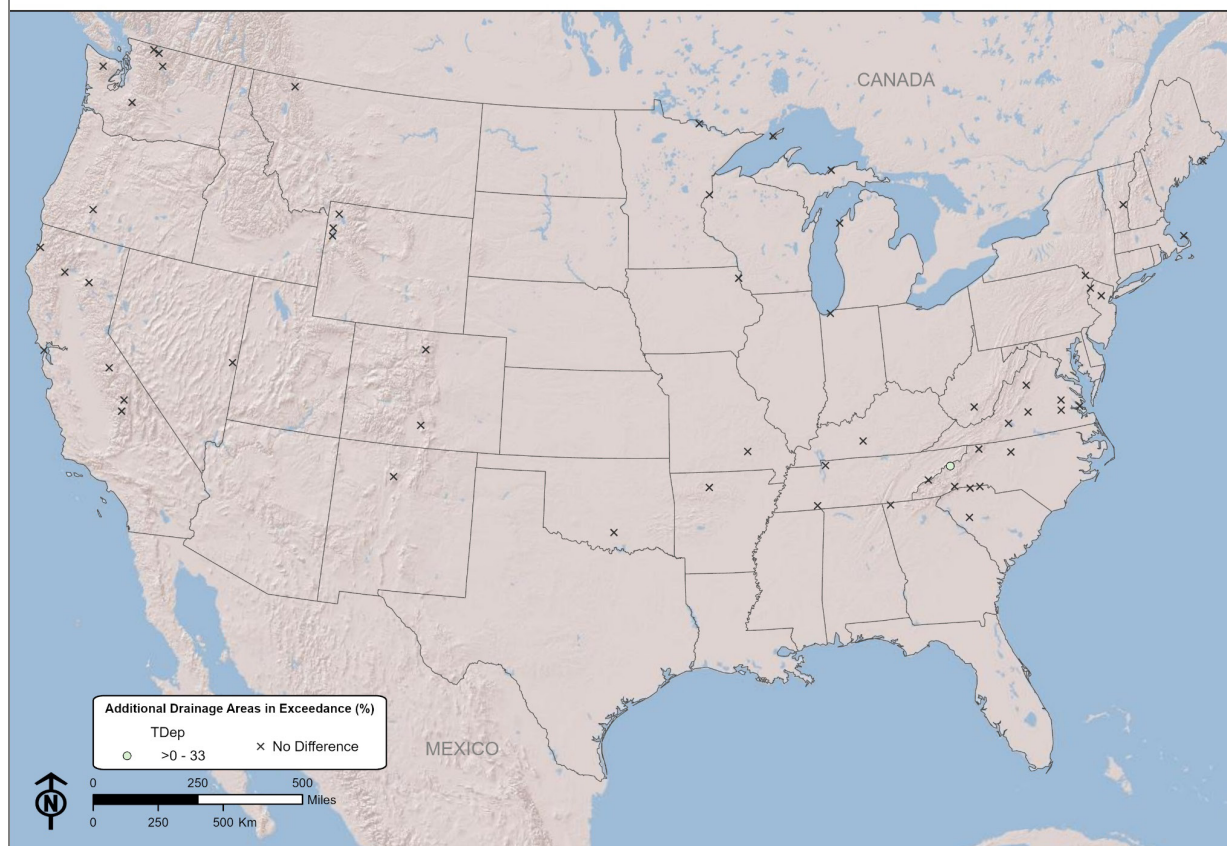


Figure 8. Spatial patterns of the additional drainage areas in exceedance of critical loads (CLs) of S deposition for surface waters, with consideration of N leaching, based on TDep or CMAQ total S deposition representative of year 2017. Green circles indicate that more stream sites were in exceedance based on TDep. Purple triangles indicate more sites in exceedance based on CMAQ.

Conclusions

Higher estimates of N deposition produced by CMAQ generally occurred at locations near densely populated urban areas, whereas TDep generally produced higher estimates of N deposition in the Sierra Nevada and the area surrounding Salt Lake City. Both deposition methods produced similar results for S deposition across the vast majority of the CONUS.

Comparisons of CL exceedance based on TDep and CMAQ showed that selection of deposition data source for characterization of CL exceedance across NPS units was most important in consideration of CLs for lichen species richness. The extent of CL exceedance was less affected by deposition data source for CLs to protect herbaceous species richness and surface water acidification. NPS units located in the central-western US (North Dakota through Texas) showed the most substantial differences in the characterization of exceedance for N-sensitive lichen species. Differences in area of exceedance of CLs for S-sensitive lichen species richness were most pronounced in the eastern US.

Most of the substantial differences in exceedance of $CL_{N,Lichen}$ are along the eastern edge of the Rocky Mountains where deposition is near the CL and multiple sources of emissions ranging from oil and gas, agriculture, urban areas, and mobile sources are located (Figure 4). Most of the areas of substantial difference in exceedance of $CL_{S,Lichen}$ are near locations of coal burning power plants. This suggests that monitoring near emission sources where atmospheric mixing may be harder to model can improve our understanding of local deposition which can then be used to better inform model parameters for emission transport and deposition to local ecosystems.

Only a few NPS units showed substantial differences in the percentage of land area in exceedance of the CL of N to protect herbaceous species richness. The number of surface water sites considered to be in exceedance of the CL of S deposition was nearly identical regardless of which model was used for calculating exceedance.

Results from this study can be used to guide natural resource management and policy in the context of understanding uncertainty in air pollution effects caused by atmospheric N and S deposition across a broad set of federally protected NPS units. Since TDep uses the measurements of atmospheric concentration and deposition to modify the base CMAQ output, areas with substantial differences are those where measurements are greater than or less than modeled values. The areas that show the largest differences in exceedance highlight how combining measurements and modeling can enhance our understanding of the risk to ecosystem health. Additionally, areas that show large differences in deposition and cause changes in exceedance area, but are located far from monitors, may be good candidates for future monitoring locations.

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Appendix A. Parks with Substantial Differences in Area of Exceedance Using TDep vs. CMAQ for Exceedance Calculations

Difference in percent of park area in exceedance, between the TDep and CMAQ models, of the critical load of N for N-sensitive lichen species richness (Table 1), critical load of S for S-sensitive lichen species richness (Table 2), and critical loads of N for herbaceous species richness (Table 3).

Table 1. Additional percent of National Park Unit area in exceedance of critical loads of N deposition for N-sensitive lichen species richness (CLN,Lichen; 3.1 kg-N ha⁻¹ yr⁻¹) for parks with at least a 33% difference based on comparisons of two methods (TDep and CMAQ) for estimating total N deposition.

Park Unit Code	Total Area of Park with Lichen CL Data (km ²)	More Exceedance based on TDep/CMAQ	Additional Percent of Park Area in Exceedance
BISC	1.8	TDep	100
CAVE	47.9	CMAQ	100
DETO	3.6	CMAQ	100
FODA	1.0	TDep	100
FOLA	0.1	CMAQ	100
MAPR	0.1	CMAQ	100
GUMO	108.3	CMAQ	99
JODR	64.8	TDep	97
THRO	151.3	TDep	89
GICL	1.6	TDep	70
AMIS	1.2	CMAQ	67
BAND	66.6	CMAQ	66
SAGU	119.3	CMAQ	62
CHIR	25.3	TDep	60
SAJH	6.2	CMAQ	60
KAWW	357.6	TDep	48
KICA	809.3	TDep	45
YELL	4970.2	TDep	44
CHIS	18.9	CMAQ	44
MEVE	98.5	CMAQ	41
FLFO	22.1	CMAQ	39
BIBE	87.3	TDep	36
CRLA	587.7	TDep	34

Table 2. Additional percent of National Park Unit area in exceedance of critical loads of S deposition for S-sensitive lichen species richness ($CL_{S, \text{Lichen}}; 2.5 \text{ kg-S ha}^{-1} \text{ yr}^{-1}$) for parks with at least a 33% difference based on comparisons of two methods (TDep and CMAQ) for estimating total S deposition.

Park Unit Code	Total Area of Park with Lichen CL Data (km ²)	More Exceedance based on TDep/CMAQ	Additional Percent of Park Area in Exceedance
BLUE	17.2	TDep	100
BOWA	0.9	TDep	100
CAJO	1.0	TDep	100
EDIS	0.1	CMAQ	100
EISE	0.5	TDep	100
FOWA	1.4	CMAQ	100
FRST	4.3	CMAQ	100
GREE	4.6	CMAQ	100
GWMP	19.2	CMAQ	100
HAMP	0.1	CMAQ	100
HOFU	3.3	CMAQ	100
JOMU	1.3	TDep	100
MOCR	0.2	TDep	100
PISC	16.5	CMAQ	100
RUCA	1.1	CMAQ	100
STRI	2.4	CMAQ	100
THIS	0.2	CMAQ	100
VAFO	9.8	CMAQ	100
WOTR	0.5	CMAQ	100
MISP	4.1	TDep	99
MUWO	2.2	TDep	97
NACA	14.7	CMAQ	95
GETT	12.4	TDep	92
COLO	25.7	TDep	87
ROCR	6.9	CMAQ	81
LIRI	62.3	CMAQ	78
GEWA	1.4	TDep	63
MONO	3.5	CMAQ	60
CONG	8.2	CMAQ	58
HOBE	8.4	TDep	57
RICH	26.1	CMAQ	56
CACO	69.8	TDep	52
PETE	11.3	CMAQ	51
FIIS	3.1	TDep	46

Table 2 (continued). Additional percent of National Park Unit area in exceedance of critical loads of S deposition for S-sensitive lichen species richness ($CL_{S,Lichen}$; $2.5 \text{ kg-S ha}^{-1} \text{ yr}^{-1}$) for parks with at least a 33% difference based on comparisons of two methods (TDep and CMAQ) for estimating total S deposition.

Park Unit Code	Total Area of Park with Lichen CL Data (km^2)	More Exceedance based on TDep/CMAQ	Additional Percent of Park Area in Exceedance
PORE	125.4	CMAQ	42
OLYM	3307.2	TDep	40
ANTI	5.8	TDep	39

Table 3. Additional percent of National Park Unit area in exceedance of critical loads of N deposition for herbaceous species richness ($CL_{N,Herb}$) for parks with at least a 33% difference based on comparisons of two methods (TDep and CMAQ) for estimating total N deposition.

Park Unit Code	Total Area of Park with Herb CL Data (km^2)	More Exceedance based on TDep/CMAQ	Additional Percent of Park Area in Exceedance
CECH	0.5	CMAQ	100
NICO	0.1	CMAQ	100
JOMU	1.4	TDep	71
SAMO	236.5	CMAQ	58
FLNI	9.3	CMAQ	36

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