Response to reviewer comments is in blue.

In this study, the authors investigated the long-term trend of deposition of N and S using state-of-the-art regional CMAQ model. Model evaluations for the deposition as well as concentration for specific air pollutants are reasonable. The conclusions are not surprising that the depositions in the US are declining from 2002 to 2107, with contributions of reduced nitrogen increasing and oxidized nitrogen decreasing, which are consistent with several previous studies. In general, this study was well designed and fit into the journal. The authors need to make efforts to improve the reading flow for the manuscript, as well as to improve their quality of figures and tables.

Thank you Dr. Zhang for providing feedback on our manuscript that ultimately resulted in a stronger publication. Specific responses to each comment are provided below.

Change the hyphen "—" to minus "−" through the whole manuscript.

We have made the change as suggested.

Abstract:

Line 11-13 "few assess dry deposition, incorporate a measurement-model fusion approach to improve wet deposition estimates, or focus on changes within specific US climate regions." This was exactly what was covered in my two previous studies (Zhang et al., 2018, 2019) which was cited by the authors as well. I suggest the authors refine their motivation or novelty for this study. Also, read the latest paper by Tan et al. (2020) and distinguish the novelty between this study with previous one.

Reference:

Tan, J., Fu, J. S. and Seinfeld, J. H.: Ammonia emission abatement does not fully control reduced forms of nitrogen deposition, Proc. Natl. Acad. Sci. U. S. A., 117(18), 9771–9775, doi:10.1073/pnas.1920068117, 2020.

We have revised the sentence to read:

> While many observational studies have investigated spatial and temporal trends of atmospheric deposition, modeling assessments can provide useful information over areas with sparse measurements, although usually have larger horizontal resolutions and are limited by input data availability.

Line 16—17: Reading from section 2.1, the authors state that the STAGE option was performing similar results as M3dry. So I did not see the point/novelty for the authors to add this statement in the abstract. Also abbreviation for "STAGE" is not necessary since it was not referred again in the abstract.

We add the statement about STAGE in the abstract because this model option was not available until CMAQv5.3. STAGE uses a resistance-based model parameterization (see Massad et al. (2010)) and allows for land-use specific dry deposition estimates, which are important for terrestrial and aquatic ecosystem health applications. We have added the following to the abstract to explain the novelty of STAGE:

> Community Multiscale Air Quality (CMAQ) model estimates from the EPA's Air QUAlity TimE Series (EQUATES) project contain important model updates to atmospheric deposition algorithms compared to previous model data, including the new Surface Tiled Aerosol and Gaseous Exchange (STAGE) bidirectional deposition model which contains land use specific resistance parameterization and land use specific deposition estimates needed to estimate the differential impacts of N deposition to different land use types.

Line 22: Explain "TNO3"

We have added the definition of TNO3 to this sentence:

> First, we evaluate model estimates of wet deposition and ambient concentrations, finding underestimates of SO_4, NO_3, and NH_4 wet deposition compared to National Atmospheric Deposition Program observations and underestimates of NH_4 and SO_4 and overestimates of SO_2 and TNO_3 (HNO_3+NO_3) compared to the Clean Air Status and Trends (CASTNET) network ambient concentrations.

Line 22-23: Is this sentence used to explain the model evaluation of wet deposition, or concentration?

This sentence is referencing the model evaluation of wet deposition. The revised sentence reads:

> Model agreement of wet deposition is poor over parts of the West and Northern Rockies, due to errors in precipitation estimates caused by complex terrain and uncertainty in emissions at the relatively coarse 12 km grid resolution used in this study.

Line 27: Will the "increased precipitation" increase both the reduced and oxidized N deposition as well?

Increased precipitation could increase oxidized N deposition, but depends on changes in local concentrations. Analyzing NADP and CAPMoN measurements in the Eastern US and Canada from 1989 to 2016, Feng et al. (2021) found strong correlations between precipitation amount and NH_4 wet deposition trends for the Midwest and Mid-Atlantic, with near-zero changes in NH_4 wet concentrations. Feng et al. (2021) found slight negative correlations between precipitation amount and NO_3 wet deposition in these two regions because, while precipitation is increasing, there are larger decreasing trends in NO_3 wet concentrations.

Line 29-30: This is an interesting finding. Can the author provide explanations why this happens?

Emissions of NO_x and SO_2 have decreased dramatically in response to the regulations set by the Clean Air Act, but the rate of decline has not been constant over the time period considered. Mchale et al. (2021) assessed wet concentration trends at NADP sites in the US, and found larger decreasing trends of wet SO_4 concentrations at 64% of sites during 2000-2017. They note from 2005 to 2010, SO_2 emission decreased at a much faster rate than before 2000. McHale et al. (2021) found similar results for NO_3, but less strong than SO_4. We revise the following sentence to explain this finding:

> We find larger average declining trends of total N and S between 2002-2009 than 2010-2017, suggesting a slowdown of the rate of decline likely in response to smaller emission reductions.

Line 30: change to "The average annual total N"?

We have revised as suggested.

Introduction:

Line 69: define "TNO3" and "NHX"

We have defined these terms as suggested.

Line 76: "TDEP" to "TDep"

We have revised as suggested.

Line 97: Please reorganize this sentence. The Hemisphere CMAQ was used to provide BCs for the 12 km CMAQ only, but not used for the data analysis in this study.

We have clarified this sentence. The revision reads:

> Lateral boundary conditions for the 12 km grid spacing CONUS domain used in this study were provided by a 108 km grid spacing Northern Hemispheric simulation.

Line 101: “STAGE” was already defined.

We keep the acronym for STAGE here since it is referred to as STAGE in CMAQ documentation.

Methods

Section 2.2: Why the authors explain why the criteria for NTN and CASTNET differ with each, “at least 60% annual coverage” for NTN, and “75% annual coverage” for CASTNET?

The 75% annual coverage for CASTNET was chosen to stay consistent with the Referee’s CMAQ-based deposition trends study (Zhang et al., 2018). The relaxed annual coverage threshold for NTN (60%) is used to also be consistent with the Referee’s measurement-model fusion approach for the wet deposition correction (Zhang et al., 2019). According to Supplemental Section 2 in Zhang et al. (2019):

> For the years of this study (2002-2012) there were 261 NADP/NTN sites with at least one year of data that met these four completeness criteria. However only 68 of these sites met the criteria for the full 11-year period, complicating spatial analysis of temporal trends. Here we have relaxed the completeness criteria using a threshold of 60% for Criterion 1, 3, and 4. The Criterion 2 threshold of 90 percent is left unchanged. For calculating trends we used any site that had 10 or 11 years of data that met this CC, resulting in a total of 183 sites included in the analysis. Figure 6 in section 3.4 highlights the impact of the sites that were included after the completeness criteria was relaxed. The additional data help fill in spatial information, while not changing the overall conclusions drawn from the model predicted trends.

Sites are considered for NTN and CASTNET model evaluation if there are observations for at least 13 of the 16 years simulated.

Results and Discussions

Line 161: define “ECODEP”

ECODEP is not an acronym.

Line 167: Please provide figure/table for this statement “although the EQUATES precipitation is still biased low on average relative to PRISM.”

We have added the following figure to the supplement to support this statement.

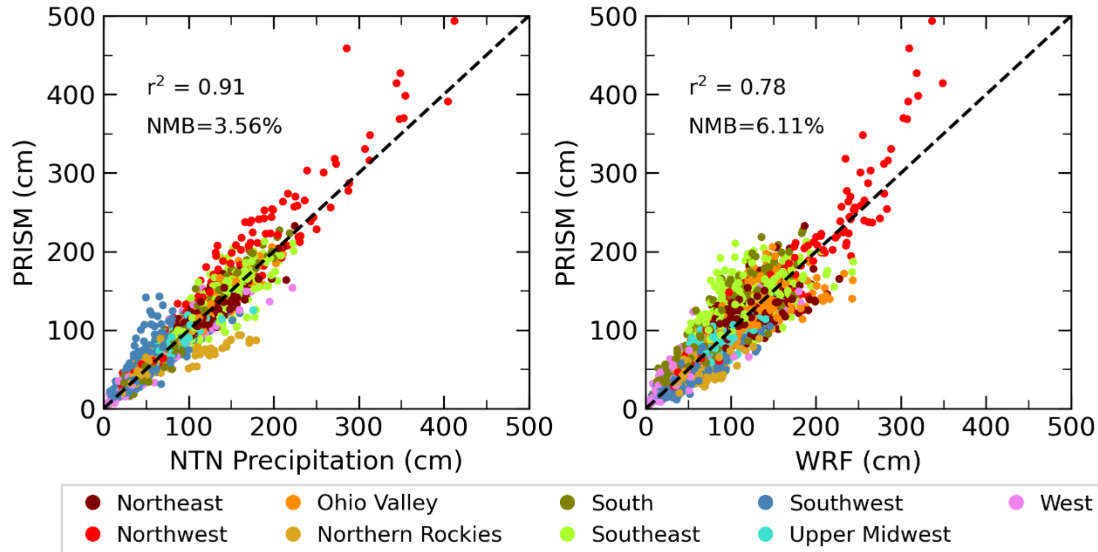

Figure S1. Left: Scatter plot of annual accumulated precipitation (cm) from PRISM and observed at selected NTN sites, colored by the NOAA climate region. Right: Scatter plot of annual accumulated precipitation (cm) modeled in CMAQ (WRF) and estimated from PRISM, colored by the NOAA climate region. The positive normalized mean bias indicates the PRISM precipitation amounts are larger than the NTN or WRF precipitation amounts.

Line 219-224: Please show a plot/table for this conclusion. Also, discuss the NH4 first and then NO3 and SO4, following the flow of earlier discussions in the same paragraph.

We have rearranged this paragraph to discuss NH4 first, followed by NO3 and SO4. This conclusion was supported by Figure S5 in the original manuscript, copied below:

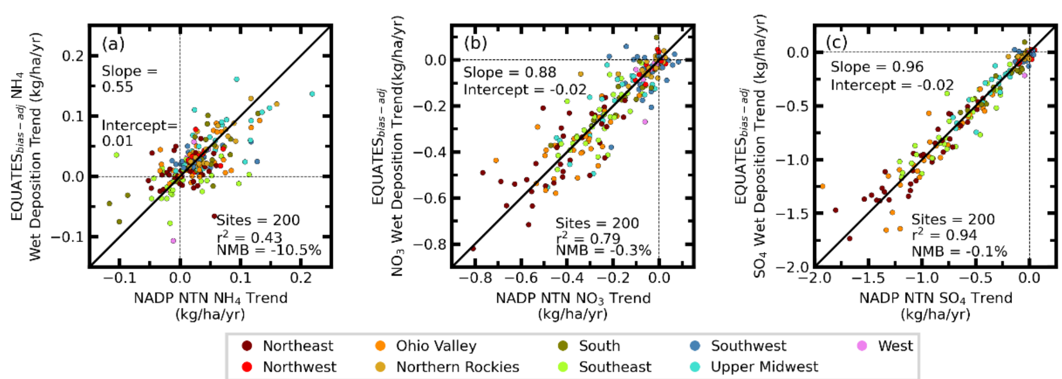

Figure S5. Scatter plot comparing the annual accumulated wet deposition trend (kg/ha/yr) from 2002 to 2017 between NTN observations and EQUATES model output for NH_4 (a), NO_3 (b), and SO_4 (c). Each circle denotes a single NTN site, colored by the climate region that meet annual completeness criteria described in the text.

Line 206: "The 16-year total NH4": is this the 16 year total or 16 year annual average? The same applies to the NO3 and SO4.

The 16-year total NH4 (and NO3 and SO4) is total, not annual average. We have revised these parts of the sentence to read "16-year annual accumulated."

Line 234-line 245: I suggest the authors to follow the order of "NH4, TNO3, SO2, and SO4" when discussing the model performances of the concentration.

We have revised as suggested.

Line 287: define NAAQS here instead of in line 293.

We have revised as suggested.

Line 302: "—0.19-0.31 kg-N/ha/yr)": Is the 0.31 positive or negative trend?

The 0.31 kg-N/ha/yr is a positive trend. We have revised the sentence, and similar sentences, to read:

> From 2002 to 2017, the largest average trend in decreasing total N deposition (—0.19 to 0.31 kg-N/ha/yr) occurs in the Upper Midwest, Ohio Valley, Northeast, South, and Southeast.

Figures & Tables

Figure 1: "Site locations of the 200" Reading from the figure, I believe the authors mean "the 263" NADP locations instead of 200 since they have "black-bordered white circles" vs. "black circles"?

NADP collects samples currently at 263 sites, but only 200 are included in this analysis. We have revised the Figure 1 text to be more explicit in what sites are included in this analysis:

> Figure 1. Site locations of the 200 National Atmospheric Deposition Program (NADP) National Trends Network (NTN, white circles) and 75 Clean Air Status and Trends Network (CASTNET, triangles) examined in this study. NADP NTN sites shown in black circles did not meet completeness criteria thresholds and therefore not included in this analysis. Color-coded US climate regions shown in this map are referred to throughout this analysis. The black-bordered white circles indicate NADP NTN sites that meet annual completeness criteria for 13 years of the timeseries and examined in the model evaluation presented in Section 3.1.

Figure 2: "black circles"—I think the authors meant the 200 "black-bordered white circles"? In the legend, there are lines associated with the rectangle and diamond, while there are none in the Taylor plot.

We have revised the Figure 2 text to better indicate where the black circle falls. We have revised the figure legend to remove the lines on the square and diamond.

> Figure 2. Taylor plot comparing annual accumulated wet deposition (kg/ha) of NH_4 (a), NO_3 (b), and SO_4 (c) collected at NTN sites (black circles, along x-axis) with model output. The symbols differentiate between the various models (EQUATES and ECODEP) and wet deposition corrections as described in the text. The azimuthal angle denotes the Pearson correlation coefficient (r^2), the gold dashed radial distance shows the standard deviation (kg/ha), and the dotted semicircles centered at the observation marker (black circle) denotes the root-mean-square error.

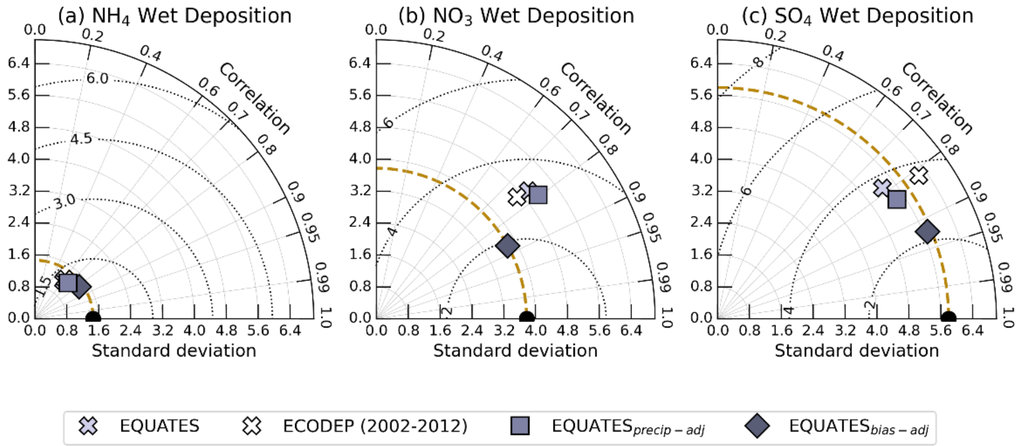

Table 2: Put Table 2 in Landscape orientation, which will make the table look much better. The same as Table 3.

The tables have been put into landscape orientation.

Figure 9: change to “throughout the nine climate regions and CONUS”. Also change “United States” in the bottom bar to “CONUS”;

We have made the change in the Figure 9 text and changed the bottom bar from “United States” to CONUS. We have similarly revised the other figures that use “United States” to “CONUS.”

References Cited above:

Feng, J., Vet, R., Cole, A., Zhang, L., Cheng, I., O'Brien, J., and Macdonald, A.-M.: Inorganic chemical components in precipitation in the eastern U.S. and Eastern Canada during 1989–2016: Temporal and regional trends of wet concentration and wet deposition from the NADP and CAPMoN measurements, Atmospheric Environment, 254, 10.1016/j.atmosenv.2021.118367, 2021.

Massad, R. S., Nemitz, E., and Sutton, M. A.: Review and parameterisation of bi-directional ammonia exchange between vegetation and the atmosphere, Atmospheric Chemistry and Physics, 10, 10359-10386, 10.5194/acp-10-10359-2010, 2010.

McHale, M. R., Ludtke, A. S., Wetherbee, G. A., Burns, D. A., Nilles, M. A., and Finkelstein, J. S.: Trends in precipitation chemistry across the U.S. 1985–2017: Quantifying the benefits from 30 years of Clean Air Act amendment regulation, Atmospheric Environment, 247, 10.1016/j.atmosenv.2021.118219, 2021.

Zhang, Y., Foley, K. M., Schwede, D. B., Bash, J. O., Pinto, J. P., and Dennis, R. L.: A Measurement-Model Fusion Approach for Improved Wet Deposition Maps and Trends, J Geophys Res Atmos, 124, 4237-4251, 10.1029/2018JD029051, 2019.

Zhang, Y., Mathur, R., Bash, J. O., Hogrefe, C., Xing, J., and Roselle, S. J.: Long-term trends in total inorganic nitrogen and sulfur deposition in the U.S. from 1990 to 2010, Atmospheric Chemistry and Physics, 18, 9091-9106, 10.5194/acp-18-9091-2018, 2018.