

The response to Referee comments is in blue.

In this manuscript, Benish and co-authors provide updated model estimates of wet, dry, and total N and S deposition across the continental United States. As S deposition declines and N deposition transitions from being dominated by oxidized forms to reduced forms, this type of measurement-model fusion is extremely important. With some revision, this manuscript will make an important contribution to the atmospheric deposition literature, and the model estimates will be very useful to many scientific and stakeholder communities. I will be excited to see this published.

We thank Dr. Heindel for the helpful comments that have improved the manuscript. Specific responses to each comment are below.

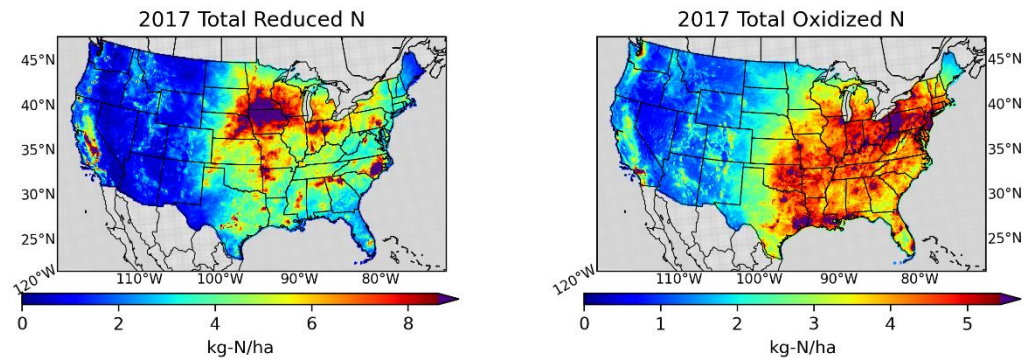
I have three broad concerns about the manuscript, in addition to more specific questions and comments listed below. First, there is almost no discussion of urban emissions and deposition in this manuscript, although the final sentence does include a note that more measurements are needed in regions with transitions from urban to rural environments. There has been a lot of recent literature on urban hotspots of N deposition, and this is a growing area of focus within NADP and TDEP. Given the completeness criteria described in the methods, none of the urban NADP sites would have been included in this study, although it would be extremely interesting. It is not clear to me how the EQUATES model handles urban emissions and deposition, but most urban centers are not visibly apparent in the Figure 5 maps, even though we know that cities have high deposition values. Even if urban areas are not explicitly included in the analyses, I think it is important to mention urban deposition more frequently throughout the manuscript, and to explain how urban areas may contribute to model uncertainty.

We agree expanding the discussion of urban deposition in EQUATES would better highlight the data need for deposition measurements in and near cities. There have been several recent publications documenting N deposition in urban areas at approximately twice that of their surrounding landscape (Decina et al., 2019). The observed urban deposition increase is composed primarily of reduced N. In CMAQ we do have higher urban emissions particularly for NO_x and do estimate mobile NH₃ emissions, but these may be underestimated (Sun et al., 2017). The urban enhancement is not readily visible on the maps due to the agricultural deposition enhancement which can be an order of magnitude higher than the surrounding non-agricultural areas (Shen et al., 2016; Walker et al., 2014) and due to model resolution, which is larger than many urban centers.

We make the following changes to address this comment, including clarifying some of the methodology and expanding the analysis:

1. Adding relevant literature to the second paragraph in the introduction. The revised text now reads:
Despite providing critical deposition information, the limited number of NADP and CASTNET sites in essential locations, such as areas with complex terrain, near urban centers, at high elevation, or in forest ecosystems, restrict a thorough understanding on the amount and consequences of deposition. For instance, strong concentration gradients in N deposition have been documented over urban areas such as Boston, MA (Rao et al., 2013) and near Baltimore, MD (Bettez et al., 2013), and along coastlines like the Chesapeake Bay (Loughner et al., 2016).
2. Clarifying methodology to reflect the use of urban NADP sites. Urban sites are included in the measurement model fusion technique for each year, but many of these sites are online for short periods of time (including several sites in Colorado: CO06, CO84, CO85, CO86, CO87), so not evaluated in the multi-year trend analysis. Urban sites that are evaluated in Section 3.1 include KY19, MD99, NC41, and NJ99 and we extend Table S1 to include the NADP reported site class (urban, suburban, rural, or isolated). Text in the methods is expanded to reflect this:
After adjusting simulated wet deposition by precipitation, an additional bias-adjustment ($EQUATES_{\text{bias-adj}}$) is applied using all NTN observations that meet annual data completeness, which varies year to year and includes sites of all classifications.
3. Add revised maps of 2017 total oxidized and reduced N deposition for easier viewing of urban sources to supplemental information (now Fig S8, see below). Added text in Section 3.2.1 reads:

Urban regions in the central and eastern US indicate a substantial amount of N deposition compared to nearby rural areas (Figure S7), consistent with previous findings that bulk N deposition in urban areas is twice as much as rural and remote sites (Decina et al., 2019).



4. Adding text in conclusions regarding modeling uncertainty in urban areas:

However, since N deposition over urban areas across the CONUS is likely already underestimated (Rao et al., 2013), with increasing urbanization only expected to further increase N deposition amounts (Joyce et al., 2020), addressing modeling uncertainty in emissions and chemistry at relevant spatial and temporal resolutions is imperative.

Second, I'm left wondering why the dry deposition does not undergo the same model measurement fusion that is used for the wet deposition. I understand that it would be impossible to do this with actual deposition estimates, but it seems like there are some clear biases in the modeled concentration values (shown in Figure 4) that could be corrected with a similar measurement-model fusion process. If there is a good reason that this works well for wet deposition but not for dry deposition, this should be stated explicitly in the text.

Measurement-model fusion methods have been widely used to improve modeled concentration output, but have been more limited for wet deposition. The NADP Total Deposition (TDep) science committee applies a fusion technique to improve dry deposition estimates with measurements from CASTNET, AMoN, and SEARCH, but presently only adjusts wet deposition by precipitation (Schwede and Lear, 2014). Zhang et al. (2019) addressed this science need by constructing an approach to bias-correct CMAQ model simulations based on observed precipitation and wet deposition observations. The TDep dry deposition fusion products are only available for an older timeseries (ECODEP) and is undergoing methodology improvements (see <https://nadp.slh.wisc.edu/committees/tdep/>). Efforts incorporating a wet deposition bias correction to TDEP is ongoing, but unfortunately not available yet.

To address this comment, we add the following text in the introduction paragraph about deposition measurements:

Dry deposition modeling is still uncertain, particularly for land use and dry deposition schemes in models and emissions data.

In addition, we add the following text in the introduction paragraph about modeling atmospheric deposition:

As such, the NADP Total Deposition Science Committee (TDEP, see [www.nadp.slh.wisc.edu/committees/tdep/](https://nadp.slh.wisc.edu/committees/tdep/)) advances methods to improve estimates of atmospheric deposition from the Community Multiscale Air Quality model (<https://www.epa.gov/cmaq>) (see Schwede and Lear (2014)). TDEP products only employ a fusion approach to dry deposition and currently use an older version of CMAQ, although efforts to update the model version and incorporate wet deposition fusion described herein are ongoing.

Additionally, we revise the first sentence in Section 2.3 to more clearly express reasoning behind the inability to correct the dry deposition estimates:

The modeled wet deposition fields are adjusted to account for input biases and uncertainty in the chemical and physical processes governing deposition, but not applied to dry deposition due to limited dry deposition measurements.

Finally, I am wondering how the focus on annual values plays into some of the measurement-model mismatch. While annual values are used very frequently, they hide the extreme seasonality of atmospheric deposition. How well is this seasonality captured in the EQUATES model? I understand that a full exploration of seasonal patterns would be another manuscript, but I am curious if model biases in both precipitation estimates and concentration estimates are season-dependent. Some discussion of the focus on annual values would be helpful.

We agree that annual values hide the extreme seasonality of atmospheric deposition. Zhang et al. (2019) previously tested the seasonality of the measurement-model fusion, finding:

1. The model overestimates precipitation, except during fall, with correlations (R^2) lowest in summer (0.47) and highest in winter (0.65). The correlation for the annual accumulated precipitation is higher (0.67).
2. NO_3 wet deposition is overestimated by the model in winter (8.6%) and fall (12.4%) and underestimated in spring (-8.2%) and summer (-6.7%). The normalized mean bias (NMB) of the annual accumulation values is -1.2%. The correlation for seasonal NO_3 wet deposition between the model and measurements is highest in spring (0.73) and lowest in summer (0.54), with higher annual R^2 of 0.76.
3. The model underestimates NH_4 wet deposition for all seasons except summer (NMB=12.6%). The annual NMB is -11%. The correlation for seasonal accumulated NH_4 wet deposition is below 0.5, except for spring (0.55) and the annual accumulated values (0.60).
4. SO_4 wet deposition is only overestimated in summer (2.0%). The annual NMB is -4.5%. The correlation is comparable for all seasons (0.69-0.71), except for slightly lower values in winter (0.57).

The table below summarizes the uncorrected seasonal wet deposition and precipitation summary statistics from EQUATES (2002-2017). This table will be added to the supplementary information (Table S3).

		Winter	Spring	Summer	Fall	Annual, no adjustment (Table 1)
NO_3	r^2	0.63	0.74	0.60	0.69	0.77
	MB (kg/ha)	0.01	0.09	0.04	0.23	0.55
	NMB (%)	1.37	5.44	2.22	20.90	9.64
NH_4	r^2	0.50	0.63	0.49	0.55	0.61
	MB (kg/ha)	-0.10	-0.23	-0.03	-0.10	-0.49
	NMB (%)	-39.7	-34.8	-3.70	-25.0	-19.9
SO_4	r^2	0.65	0.71	0.72	0.67	0.78
	MB (kg/ha)	-0.17	-0.26	-0.30	-0.17	-0.92
	NMB (%)	-18.9	-14.3	-14.5	-12.9	-12.2
Precipitation	r^2	0.77	0.70	0.61	0.69	0.72
	MB (kg/ha)	3.63	-2.66	-12.9	-23.2	-2.32
	NMB (%)	2.28	-1.30	-5.88	-11.7	-2.40%

From Zhang et al. (2019) and the table above, we find the seasonal correlation values are fairly low in fall and winter in multiple regions (see Figure S1 in Zhang et al. (2019)), therefore a seasonal precipitation adjustment will be less effective. The annual deposition and precipitation values are used in the measurement-model fusion approach because correlations are high enough at a sufficient number of sites to make the precipitation adjustment effective at an annual timescale.

Since the focus of this manuscript is regional trends from EQUATES, we feel annual values are valid in this application. However, to guide readers to seasonal analyses from above, we add the following sentence to the methods in Section 2.3:

Since model performance is improved for annual instead of seasonal values (Table S3, refer to Zhang et al. (2019) for detailed seasonal model evaluation), we apply a measurement-model fusion technique previously described by Zhang et al. (2019) to adjust the modeled annual wet deposition fields of inorganic N ($\text{NO}_3 + \text{NH}_4$) and S, briefly described here.

Specific Comments

Abstract – Overall, the abstract is quite long and I found it challenging to follow. I think it could benefit from a clearer structure.

We thank the reviewer for this feedback. We have revised the abstract to read:

Atmospheric deposition of nitrogen (N) and sulfur (S) compounds from human activity has greatly declined in the United States (US) over the past several decades in response to emission controls set by the Clean Air Act. 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. In this analysis, we evaluate wet, dry, and total N and S deposition from multiyear simulations within the contiguous US (CONUS). 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 improvements to organic N chemistry. 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 ($\text{HNO}_3 + \text{NO}_3$) compared to the Clean Air Status and Trends (CASTNET) network ambient concentrations. Second, a measurement-model fusion approach employing a precipitation- and bias- correction to wet deposition estimates is found to reduce model bias and improve correlations compared to the unadjusted model values. 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. Next, we assess modeled N and S deposition trends across climatologically consistent regions in the CONUS. Total deposition of N and S in the eastern US is larger than the western US with a steeper decreasing trend from 2002-2017, i.e., total N declined at a rate of approximately -0.30 kg-N/ha/yr in the Northeast and Southeast and by -0.02 kg-N/ha/yr in the Northwest and Southwest. Widespread increases in reduced N deposition across the Upper Midwest, Northern Rockies, and West indicate evolving atmospheric composition due to increased precipitation amounts over some areas, growing agricultural emissions, and regional NO_x/SO_x emission reductions shifting gas-aerosol partitioning; these increases in reduced N deposition are generally masked by the larger decreasing oxidized N trend. We find larger average declining trends of total N and S deposition between 2002-2009 than 2010-2017, suggesting a slowdown of the rate of decline likely in response to smaller emission reductions. Finally, we document changes in the modeled total N and S deposition budgets. The average annual total N deposition budget over the CONUS decreases from 7.8 kg-N/ha in 2002 to 6.3 kg-N/ha in 2017 due to declines in oxidized N deposition from NO_x emission controls. Across the CONUS during the 2002-2017 time period, the average contribution of dry deposition to the total N deposition budget drops from 60% to 52%, whereas wet deposition dominates the S budget rising from 45% to 68%. Our analysis extends upon the literature documenting the growing contribution of reduced N to the total deposition budget, particularly in the Upper Midwest and Northern Rockies, and documents a slowdown of the declining oxidized N deposition trend, which may have consequences on vegetation diversity and productivity.

Line 28: It is unclear how regional NO_x/SO_x emission reductions contribute to widespread increases in reduced N deposition. I understand that the reduction in NO_x deposition increases the proportion of N deposited in reduced form, but it seems like this is stating that it contributes to the absolute increase.

We have revised the sentence to read:

Widespread increases in reduced N deposition across the Upper Midwest, Northern Rockies, and West indicate evolving atmospheric composition due to increased precipitation amounts over some areas, growing agricultural emissions, and regional NO_x/SO_x emission reductions shifting gas-aerosol partitioning; these increases in reduced N deposition are generally masked by the larger decreasing oxidized N trend.

Introduction

Line 45: This seems like a limited definition of dry deposition, because dry deposition could be deposited on surfaces other than leaves, like soil or water.

We revised the sentence to read:

After entering the atmosphere, the major nitrogen (N) and sulfur (S) removal pathways occur by precipitation (wet deposition) or uptake by surfaces, such as terrestrial and aquatic vegetation (dry deposition).

Line 57: In my opinion, urban areas and intense agricultural areas are also essential locations that have limited measurements.

We agree, and revise the sentence to read:

Despite providing critical deposition information, the limited number of NADP and CASTNET sites in essential locations, such as areas with complex terrain, near urban centers, at high elevation, or in forest ecosystems, restrict a thorough understanding on the amount and consequences of deposition.

Lines 75-79: I am confused by the relationship between this project and TDEP, and I would like to see more comparison to TDEP products throughout the manuscript. Is this effort part of TDEP, or will the results here be incorporated into the TDEP products? TDEP products are used extensively by the NADP community, so clarification here would be very helpful.

TDEP currently uses an older version of CMAQ and does not employ a measurement-model fusion approach to wet deposition (although these efforts are planned), so a direct comparison would be limited. We have revised text in the introduction to clarify this project and TDEP:

As such, the NADP Total Deposition Science Committee (TDEP, see www.nadp.slh.wisc.edu/committees/tdep) advances methods to improve estimates of atmospheric deposition from the Community Multiscale Air Quality model (<https://www.epa.gov/cmaq>) (see Schwede and Lear (2014)). TDEP products only employ a fusion approach to dry deposition and currently use an older version of CMAQ, although efforts to update the model version and incorporate wet deposition fusion described herein are ongoing.

Methods and Materials – Throughout the methods and materials, it would be helpful to be extremely clear about the timescale used. It seems like most calculations were done on an annual basis, but this was sometimes confusing.

We have clarified that the timescale used was annual in the text.

Line 121: Was this calculation correcting for chemical transformations performed on an annual basis? Or on a weekly basis?

The revised sentence reads:

First, modeled wet deposition of NO_3 , NH_4 , and SO_4 is calculated using the approach by Appel et al. (2011) that accounts for chemical transformations of several species in the aqueous phase. Then, the model and observations are paired in time and space and annually accumulated.

Equation 1: The precipitation correction is done on an annual basis, which seems like it could be problematic. Because N deposition has such a strong seasonal cycle, it matters when the precipitation is either over- or underestimated. If the modeled precipitation is too low mostly during the winter when N concentrations are low, an annual correction could then overestimate N deposition. For more discussion of this problem (and how it introduces error into the NADP annual estimates), see (Schichtel et al., 2019). It would be helpful to see some discussion about the decision to focus on annual values and the issues that this may introduce into the calculations.

We agree that the relationship between wet deposition and precipitation is affected by the frequency, duration, and intensity of the rainfall as well as the ambient concentration. While this relationship is typically nonlinear on hourly and daily time scales, the relationship can become more linear when annually accumulated. The focus on annual values is chosen because NADP maps data at the annual level and the EQUATES simulations covering model years 2002 to 2017 present an opportunity to evaluate modeled regional deposition trends containing important model updates (i.e. consistent emissions methodology, use of STAGE bi-directional model, improve organic nitrate chemistry, etc.). Additionally, end users of EQUATES output for nutrient assessments, including critical loads, are generally more interested in annual instead of seasonal values. While a seasonal evaluation would be interesting, it is beyond the scope of this paper to address.

To address this comment, we add the citation suggested above and add how the relationship between wet deposition and precipitation on annual timescales is more linear:

While wet deposition relies on the season and precipitation rate (Schichtel et al., 2019), the relationship between precipitation and wet deposition is more linear when annually accumulated.

Line 148: How sensitive is this method to the 300-km radius? How was this radius chosen?

As described in Zhang et al., 2019, the size of the moving window (300 km) was determined by a cross-validation analysis. This was done to dampen any large bias and create a regional estimate of bias.

Results and Discussion

Figure 3: Is the precipitation here from PRISM or from NTN rain gauges? I don't think the NTN precipitation depth measurements were mentioned in the sampling method section, so this might be confusing to people who are unaware that NTN measures precipitation depth.

The precipitation plotted in Figure 3 is from NTN. The figure is labeled "NADP NTN Precipitation (cm)" and we revise the figure legend to clarify the modeled precipitation is being compared to NADP NTN precipitation observations:

Figure 1. Scatter plots of annual accumulated bias-adjusted modeled and NTN observed wet deposition (kg/ha) of ammonium (a, NH_4), nitrate (b, NO_3), and sulfate (c, SO_4) from 2002 to 2017 colored by the climate region. Panel d shows NADP NTN observed and modeled precipitation (cm).

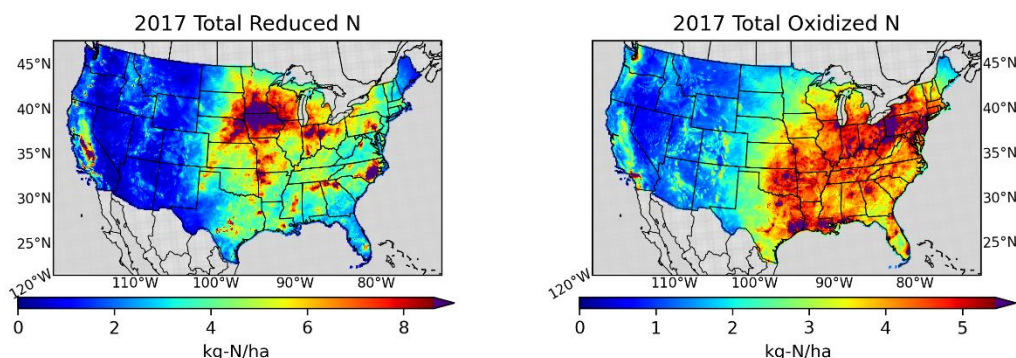
Lines 206-224: I found it confusing that this paragraph mixes general results on spatial variability in total deposition with model performance.

This part of the manuscript is discussing model evaluation, including the different spatial regional descriptions.

Lines 264-267: How are you defining hotspots here, and how were they identified? There are many urban areas that are known hotspots of N deposition (e.g., Denver-Boulder metro area), but these do not appear on the map in Figure 5 (but perhaps this is because of the spatial or color scale?).

The color scale and color map does not allow for easy viewing of urban centers since the focus of this manuscript was regional trends from 2002 to 2017. New maps of 2017 total reduced and oxidized N deposition with adjusted colors indicate urban centers do see large amounts of N deposition. This figure was added to the supplemental information and we add a sentence regarding the larger deposition amounts over urban areas to Section 3.2.1:

Urban regions in the central and eastern US indicate a substantial amount of N deposition compared to nearby rural areas (Figure S7), consistent with previous findings that bulk N deposition in urban areas is twice as much as rural and remote sites (Decina et al., 2019).



Additionally, we remove the word “hotspot” to avoid confusing readers.

Line 312: Explain the connection between warming temperatures and increasing reduced N deposition more fully. Also, what about on-road emissions of ammonia? These are an increasingly important source of NH_3 emissions connected to NO_x emission control mechanisms (Fenn et al., 2018).

The revised text reads:

Fertilizer use in the Midwest has only grown modestly ($\sim 1.3\%$ /yr), so increases in total N deposition have been largely attributed to increasing reduced N deposition. Growing reduced N deposition is a result of NH_3 emissions increasing exponentially with temperature (except below freezing and where emissions are near 0) (Riddick et al., 2016) and by increasing the partitioning of NH_3 remaining in the gas phase due to NO_x and SO_2 emission reductions (Warner et al., 2017) and increasing importance of on-road mobile emissions of NH_3 (Fenn et al., 2018).

Figure 5: It might also be helpful to distinguish between areas with unavailable and not significant trends in Figure 5, because these have very different meanings. In this trend analysis, is it possible to have a significant trend with a slope of zero? Figure 5f appears very white – are these places with very small significant trends, or is the slope actually zero?

Trends are only available where we have PRISM data (e.g. CONUS), so anything outside CONUS is light grey. Alternatively, darker grey indicates where trends are not significant. We have clarified this in the Figure 5 legend:

Figure 2. Spatial distribution of total N (top) and S (bottom) deposition in 2002 (a and d, kg/ha), 2017 (b and e, kg/ha), and the 2002-2017 annual trend (c and f, kg/ha/yr) with significance at the 95% confidence level. Grey areas in panels (c) and (f) indicate where the trend is unavailable due to lack of PRISM data or not significant (i.e., p-value of the Wald test is greater than 0.05).

The Wald test is testing a null hypothesis that the linear regression slope is 0 with an alternate hypothesis that the slope does not equal zero. Therefore, a significant slope means that it is significantly different from zero (i.e., you cannot have a significant slope of zero).

Figure 6: I am struggling to interpret Figure 6, given the fact that areas without a significant trend are removed. Judging by Figure 5, it seems like this removes the vast majority of many regions. If there is a small decreasing

trend in a corner of a region that generally has had stable N deposition, it seems misleading to represent that as a decreasing trend for the whole region. I'm also confused by what 'data size' refers to in the caption. Is each data point a pixel on Figure 5?

We agree that the non-significant trends should not have been removed. We remade Figures 6, 7, and 8 to set the non-significant trends to 0. Data size refers to the number of CMAQ model grid cells with a significant trend that the average, 5th percentile, and 95th percentile trend is based on. The 5th and 95th percentile lines on each region bar can be used to help assess the range of trends seen for each region.

We have revised the text and figure legends to reflect that all grid cells are included in these figures.

Line 356: Again, I'm curious how you are defining the term 'hotspot.'

We have rephrased this sentence to read:

Regions of elevated wet and dry reduced N deposition have expanded and increased in magnitude across the CONUS (Figure S10) compared to oxidized N, also observed in the NTN NH₄ measurements.

Technical Corrections

Line 54: Rephrase so it is clear that the NADP, rather than wet deposition, is the subject of the verb "collecting."

The revised sentence reads:

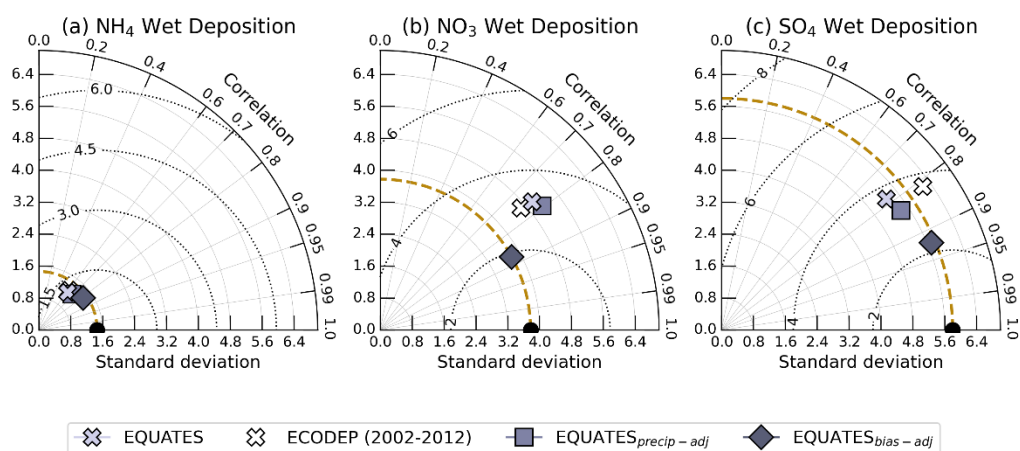
Wet deposition, sampled weekly in rain or snow, is measured by the National Atmospheric Deposition Program (NADP) National Trends Network (NTN) since 1978.

Line 174: Define NMB in the text as well as in the figure captions.

NMB is defined on line 70.

Figure 2: It would be helpful to make the dashed and dotted lines more visibly different.

We have edited the figure to use color to make these lines stand apart better.



References

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