**Review of Long-term Regional Trends of Nitrogen and Sulfur Deposition in the United States from 2002-2017**

**General Comments**

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.

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.

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.

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.

**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.

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 is contributes to the absolute increase.

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

*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.
Line 121: Was this calculation correcting for chemical transformations performed on an annual basis? Or on a weekly basis?
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.
Line 148: How sensitive is this method to the 300-km radius? How was this radius chosen?

*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.
Lines 206-224: I found it confusing that this paragraph mixes general results on spatial variability in total deposition with model performance.
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?).
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).
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?
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?
Line 356: Again, I'm curious how you are defining the term 'hotspot.'

**Technical Corrections**
Line 54: Rephrase so it is clear that the NADP, rather than wet deposition, is the subject of the verb "collecting."
Line 174: Define NMB in the text as well as in the figure captions.
Figure 2: It would be helpful to make the dashed and dotted lines more visibly different.

**References**
Fenn, M. E., Bytnerowicz, A., Schilling, S. L., Vallano, D. M., Zavaleta, E. S., Weiss, S. B., et al. (2018). On-road emissions of ammonia: An underappreciated source of atmospheric nitrogen deposition. *Science of The Total Environment*, *625*, 909–919. https://doi.org/10.1016/j.scitotenv.2017.12.313

Schichtel, B. A., Gebhart, K. A., Morris, K. H., Cheatham, J. R., Vimont, J., Larson, R. S., & Beachley, G. (2019). Long-term trends of wet inorganic nitrogen deposition in Rocky Mountain National Park: Influence of missing data imputation methods and associated uncertainty. *Science of The Total Environment*, *687*, 817–826. https://doi.org/10.1016/j.scitotenv.2019.06.104