Response to review #2 on acp-2018-116

Long-term trends in total inorganic nitrogen and sulfur deposition in the U.S. from 1990 to 2010

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We thank referee #1 for the positive comments and constructive suggestions, which have helped us improve the manuscript. All referee comments (in blue below) have been carefully addressed, and changes incorporated in the revised manuscript are shown using the track-changes option.

The manuscript discussed twenty year trends in nitrogen (N) and sulfur (S) deposition in the U.S. based on the WRF-CMAQ model simulations. The article identified the current limitations of modeling nitrogen and sulfur deposition and discussed spatial distributions and trends of those species in the U.S. Those results confirmed that reduced nitrogen had dominated the total nitrogen deposition in the U.S. and highlighted the necessity of controlling reduced N. The structure of the manuscript, the results and the presentation of the material are reasonably good. The topic is relevant and certainly deserves publication in Atmospheric Chemistry and Physics.

There are, however, several changes and additions required before publication.
Specific comments:
Page 2, Line 5-6: Please split up these references so that they are associated with the specific impacts being discussed, rather than all placed at the end of the sentence. And, I do not think increased sulfur deposition could cause aquatic eutrophication.
Response: We thank the reviewer for pointing this out. We now split the references. The revised sentence is:
"Increased nitrogen and sulfur deposition is detrimental to ecosystems, since it leads to decreased biological diversity (Clark and Tilman, 2008; Clark et al., 2013; Stevens et al., 2004), increased terrestrial and aquatic eutrophication (Bouwman et al., 2002; Bowman et al., 2008; Fisher et al., 2011) and acidification (Greaver et al., 2012; Savva and Berninger, 2010)."

Page 2, Line 20: Change "pattern" to "patterns".
Response: We made the change following the reviewer' comment.

Page 2, Line 26: Please explain "complexity" more.
Response: We thank the reviewer for pointing this out. Complexity was intended to convey the challenges in spatial interpolation of dry deposition estimates both due to limited availability of observations as well as representativeness of the interpolated fields. To clarify, we have rewritten the sentence as:
"however cannot be easily spatially interpolated due to limited availability of sufficient number of sites in a region as well as the representativeness of the derived fields due to assumptions in the spatial interpolation method (Schwede and Lear, 2014)"

Page 3, Line 20: Change "description" to "descriptions".
Response: Thank you for catching the typo. We have revised the sentence as:

"Interested readers are referred to Gan et al. (2015, 2016) for a detailed description of the settings of the CMAQ model"

Page 3, Line 22: Add "supporting" in the front of "Table S1".
Response: We thank the reviewer for pointing this out. We now add "supporting" in the front of "Table S1".

Page 3, Line 22: O_3 and $PM_{2.5}$ should be defined at first mention.
Response: We thank the reviewer for pointing this out. We now added the definition of O_3 and $PM_{2.5}$ in the paper:
"The performance of the coupled WRF-CMAQ model for major trace gases, aerosol species and meteorological variables such as ozone (O_3), fine particular matter ($PM_{2.5}$)"

Page 3, Line 24-25: Provide some references.
Response: We now rewrite the sentence to add the right references to them:
"at both the hemispheric and regional scale has been extensively evaluated in previous studies, and has shown skill in simulating the magnitudes and long-term trends of these variables (Xing et al., 2015a, b; Mathur et al., 2017; Gan et al., 2015, 2016; Astitha et al., 2017)."

Page 3, in section 2.1: The authors should specify how the dry depositions were estimated.
Response: We thank the reviewer for the suggestion. The dry deposition for each species is calculated by multiplying the concentration in the lowest model layer by the dry deposition velocity (V_d). The dry deposition velocity is calculated as the reciprocal of the sum of the atmospheric (R_a, the resistance to transport through the atmosphere above the surface receptors), quasi-laminar boundary layer (R_b, the resistance to transport across the thin layer of air that is in contact with the surface and varies with the diffusion of the pollutant transported), and surface resistances (R_s, the resistance to the uptake of the pollutant by the surface receptor, typically vegetation or soil). We now add this information on Page 3 line 25:

"The dry deposition of each species in the CMAQ model is calculated by multiplying the concentration in the lowest model layer by the dry deposition velocity (V_d). The V_d is calculated as the reciprocal of the sum of the atmospheric (R_a, the resistance to transport through the atmosphere above the surface receptors), quasi-laminar boundary layer (R_b, the resistance to transport across the thin layer of air that is in contact with the surface and varies with the diffusion of the pollutant transported), and surface resistances (R_s, the resistance to the uptake of the pollutant by the surface receptor, typically vegetation or soil)."

Page 4, Line 11: I am not sure whether the 110th meridian west is appropriate to divide east and west. There are more sites in the east than the west if 110°W is used. A map with 110°W and all the sites should be included in the supplement.
Response: We agree that the use of the 110th meridian to define the East vs West U.S. is somewhat arbitrary. It was used primarily because the majority of the SO_2 and NO_x emissions in the U.S. are east of this meridian. We now include a map of the distribution of observation sites in the supplement. Please see supporting Fig. S1.

Page4, Line 13: How did the authors get the value of 0.984?

Response: 0.984 is the ration of molecular weight of NO_3^- to the molecular wieght of HNO_3 (62/63) and is used to convert the mass of HNO_3 deposited to that of NO_3^-, as also previously discussed in Appel et al. (2011).

Page 5, In 3.1, Could the authors be more specific about the improvements since Appel et al (2011)?

Response: There are numerous differences between the model configuration and versions used in this analysis and those previously used by Appel et al. CMAQv5.0 was used here and included the AERO6 aerosol module, while Appel et al. used CMAQv4.7 that employed the AERO5 aerosol module. Specific process differences between model versions can be found at: https://www.epa.gov/cmaq/cmaq-models-0. In the revised manuscript we point interested users to specifics of these model versions by including the following sentence after Pg 6 line 20:
"There are numerous differences between the model configuration and versions used in this analysis and those previously used by Appel et al. (2011). Specific model process representation differences between CMAQv5.0 used here and CMAQv4.7 used in Appel et al. (2011) can be found at: https://www.epa.gov/cmaq/cmaq-models-0."

Page 5, It seems like the authors only did model evaluation and model justification for wet deposition. How was model performance for dry deposition? The authors could use data from AMON, IMPOROVE and EPA CASTNET to do this work.

Response: The reviewer raises an interesting point related to evaluation of dry deposition estimates. The U.S. CASTNET (Clean Air Status and Trends Network) did provide the dry deposition data. However, these values are not measured but instead derived using the inferential method, pairing the measured air pollutants concentration with a modeled deposition velocity from the MLM model (Meyers et al, 1998). So rather than comparing the two model values between CMAQ and MLM, we chose to compare CMAQ estimated ambient concentrations of both gaseous (SO_2) and particulate (SO_4^{2-}, TNO_3^-, NH_4^+) species with measurements from CASTNET.
We change the title in section 2.2 "Wet deposition observations in the U.S." to "Deposition observations in the U.S.".

On Page 5 line 7, we add the description for the dry deposition from the U.S. CASTNET:
"The U.S. CASTNET provides long-term observation of atmospheric concentrations as well as the dry deposition (https://www.epa.gov/castnet, accessed May 7, 2018). However, the dry deposition values reported are not directly measured, but estimated using the inferential method, pairing the measured air pollutant concentration with a modeled deposition velocity from the MLM model (Meyers et al, 1998). So rather than comparing dry deposition estimates from two models, we choose to evaluate the model's performance in simulating the ambient air concentrations (sulfur dioxide (SO_2), sulfate (SO_4^{2-}), total nitrate ($TNO_3 = NO_3^- + HNO_3$), and ammonium (NH_4)). The detailed site information and the number of years of observational data used for the model evaluation can be found in supporting Table S3. We apply the same criteria in selecting valid observation sites as the NADP/NTN."

In the revised manuscript we further discuss the evaluation of these air concentrations on Page 6 line 21:

"To evaluate the model's performance in simulating the DDEP, we compare the model simulated concentration with the observations from CASTNET. Comparisons of annual average simulated concentrations with corresponding measurements at the CASTNET sites show strong correlation for SO_2 (R of 0.88), SO_4 (0.95), TNO_3 (0.94), and NH_4 (0.94). Some underestimation for SO_4, and overestimation in other species ambient concentrations is noted (supporting Fig. S4). The model also captures the trends for these species with very high R, but the magnitude of the decreasing trends is underestimated by the model (supporting Fig. S5)."

Page 5, Line 19: Change "models" to "model results"

Response: We thank the reviewer for pointing this out. We now changed the word "model" to "model results".

Page 6, Line 15: It should be "Table S3"

Response: We now corrected from "supporting Table 3" to "supporting Table S3".

Page 6, Line 26 – Page 7, Line 2: Please explain the reasons for those results.

Response: The discrepancies for the trends of the TIN TDEP over U.S. ecoregion regions are caused by the combination of the decrease of the NO_x emissions, and unregulated but increased NH_3 emissions at different places. We now clarify this on Page 6 line 26-Page 7 line 2:
"During the period from 1990 to 2010, TIN TDEP has significantly decreased (with $p < 0.05$ for the standard two-tailed Student's t-test) over several ecoregions, including Eastern Temperate Forests, Northern Forests, Mediterranean California and Marine West Coast Forest (decreasing trend of 0.12, 0.071, 0.038 and 0.017 kg N ha^{-1} yr^{-1} respectively) as a result of significant reductions in anthropogenic NO_x emissions (Xing et al., 2013)."

Page 7, Line 11- 20: Which one dominates the decrease of TSOx, SO_4^{2-} or SO_2?

Response: We thank the reviewer for pointing this out. After performing additional calculations, we determined that the decrease of TSO_X is dominated by SO_4^{2-}. We now add this information in Page 7 line 17:
"All the ecoregions experienced statistically significant decreases of TS TDEP over the past two decades which was dominated by the decreases in SO_4^{2-}, except for the Mediterranean California ecoregion which showed an insignificant decreasing trend (Table 6)."

Page 26, The legend of Fig 8 (a) needs to be fixed.

Response: We thank the reviewer for pointing this out. We have now fixed the legend on Fig. 8(a). Please see the updated plot in our revised manuscript.