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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term trends in total inorganic nitrogen and sulfur deposition in the U.S. from 1990 to 2010" *by* Yuqiang Zhang et al.

Anonymous Referee #2

Received and published: 8 April 2018

This paper examines trends in inorganic nitrogen and sulfur deposition from 1999 to 2010 across the U.S. This analysis is performed using WRF-CMAQ model simulations. The results from the model are compared to data from the NADP (National Atmospheric Deposition Program) Network. The trends and spatial patterns observed are discussed.

Overall, this is a good paper. But I do have some concerns. I feel a large part of the methods section is missing as the authors do not actually discuss the dry deposition data being used. The authors establish how wet deposition from the model compares with the observations, but they do not actually discuss what might lead to the differences in the two until a few lines in the conclusions. More importantly, they do not make this same comparison for the dry deposition results, yet they go on to discuss

the trends in dry deposition from the model in the paper. All of this is outlined in detail below in my comments, which need to be addressed before this paper can be considered for publication

General Comments: -I am a bit surprised that the abbreviation TSOx is used for sulfur deposition rather than TS. TS to me seems more fitting, but I understand if the other is more traditionally used as I am not as familiar with that literature as I am with nitrogen deposition. However, that being said it seems that the paper goes back and forth using TSOx and TSO4 to represent sulfur deposition. This is true throughout the main text, figures, and supporting information. This should be checked.

Specific Comments: Abstract Page 1, Line 12 – The abbreviation WRF-CMAQ is not defined

Page 1, Lines 15-17 – The authors mention that the model generally underestimates the wet deposition. But they do not provide any reasons why this is. This should be added to the abstract.

Page 1, Line 19 – Suggest changing decrease of TNO3 to decreases in TNO3

Page 1, Line 20 – The authors mention there are increasing trends in TIN deposition over the Tropical Wet Forest. This is the only region type listed in the text that does not have a geographic location included in its title. I think this makes it hard for readers to understand where it is. I would suggest adding a phrase such as southern Florida to aid the reader.

Page 1, Line 22 – Suggest removing the words region of before Eastern

Page 1, Line 23 – Suggest removing the words region of before Tropical

Page 1, Line 28 – Suggest adding an a before combination

1.Introduction Page 2, Line 12 – Suggest changing twice higher than to twice as high as

Page 2, Line 13 – Suggest removing the the before sulfur

Page 2, Line 14 – form fossil-fuel should be from fossil-fuel

Page 2, Lines 15-20 – Here the authors discuss the wet deposition national networks. But they do not actually tell how the measurements are made. I would suggest adding some text telling how the samples are collected and then measured by ion chromatography to provide the data.

Page 2, Line 21 – Suggest adding the words e.g., before EEA. Also a comma is missing after EEA

Page 2, Line 22 – Suggest removing the comma and phrase to name a few after 2015

Page 3, Line 2 – There is an extra period after loss

Page 3, Line 5 – Suggest removing the second Zhao et al.

Page 3, Line 8 – Suggest changing process to processes

2.Methods 2.1.Model setup Page 3, Line 16 – The abbreviation WRF-CMAQ is not defined

Page 3, Line 20 – There is an extra comma after Gan et al. Also suggest adding an a before detailed

Page 3, Line 22 – The chemical abbreviation used are not defined

Page 3, Line 24 – Suggest changing was shown to has shown

2.2.Wet deposition observations in the U.S. Page 4, Line 11 – Suggest changing observation data used for to observational data used for the

Page 4, Line 13 – Suggest changing combine WDEP to combines WDEP and with 0.984 to by 0.984

Page 4, Line 15 – Suggest changing combine WDEP to combines WDEP and with 1.06

to by 1.06

Page 4, Line 17 - Suggest changing combine WDEP to combines WDEP and with 1.50 to by 1.50

Page 4, Lines 2-26 – Why is there no section on dry deposition in the Methods section? The authors explicitly state in the introduction that there are no direct measurements of this, but that they are calculated at some sites. So then information on how they are calculated and what is used here should be provided to the reader so that they fully understand the analysis that is being performed.

Page 4, Lines 2-26 – The authors do not actually explain how the data from the network is collected. I understand the model analysis is the point of the paper. But since these observational data are used to evaluate the model then the authors should provide at least some text to give the readers context.

Page 5, Line 5 – There is an extra comma after equation 2

Page 3, Line 14 to Page 5, Line 6 – In the methods section there is no discussion of the trend analysis that is used throughout the paper. What is this analysis? How is it done? This should be added to the paper.

3.Results 3.1.Model evaluation of WDEP Page 5, Line 16 – Suggest changing increases for all the three to increase for all three. Also exhibit should be exhibited. Also suggest changing in east than that in west to in the east than the west

Page 5, Line 17 – A period is missing after (Appel et al., 2011)

Page 5, Line 19 – Suggest changing both observations and models to both the observations and model results

Page 5, Line 20 – Suggest adding a the before Tropical

Page 6, Line 11 – I am not sure I understand the phrase but a slightly distinctions in trends for different ecoregions. Is it maybe but with slight distinctions in the trends for

each ecoregion?

Page 6, Lines 11-13 – The authors mentions that the model generally underestimates decreasing WDEP trends for all sites, but for NHx they see increasing WDEP trends. Why is this? The authors need to tell why they think this might be the case for the model.

Page 6, Line 14 – Suggest removing the word results before model

Page 6, Line 15 – Suggest changing increases for all the three to increase for all three. Also why are the authors only looking at the data from 2002-2006 when they discuss the NMB increase observed? This needs to be clarified.

Page 6, Line 18 – Suggest changing are more to have more

Page 6, Line19 – Suggest changing challenging to challenges

Page 5, Line 8 to Page 6, Line 20 – Why is there no matching section on the model evaluation for DDEP? The remainder of the results section discusses the trends in total, wet, and dry deposition so it seems that it should be established how the model compares with the calculated dry deposition values provided by NADP.

3.2.Spatial patterns of modelled total deposition of nitrogen and sulfur Page 6, Line 21 – modelled should be written as modeled to be consistent with how it is used throughout the rest of the text

Page 7, Line 18 – Suggest removing the and after showed

Page 7, Line 19 – Believe that Table 4 should be Table 6

3.3.Wet versus dry nitrogen and sulfur deposition trends in the U.S. Page 7, Line 25 – Suggest adding a the before Eastern

Page 7, Line 26 – Suggest adding a the before Northern and Great

Page 7, Line 27 – Suggest changing was mainly to were mainly

Page 7, Line 28 – The authors mention that there are no significant changes for WDEP of NHx. However, in Table S4 the values for Tropical Wet Forests are in bold, which is what indicates a significant trend. Also there is light blue being shown in Figure S4b. This needs to be clarified.

Page 8, Line 8 – Suggest adding an a before distinct and changing value to values

Page 8, Line 9 – Suggest adding a the before vicinity and changing source to sources

3.4.Deposition budget in U.S. Page 8, Line 18 – Suggest changing were estimated to was estimated

Page 8, Line 19 – Suggest removing the hyphen after 2010

Page 8, Line 21 – Suggest changing changes to changed

Page 8, Line 22 – Suggest changing till to until and removing the the before NHx

Page 8, Line 26 – Suggest changing 1999-2010 to 1999 to 2010

Page 8, Line 27 – Suggest changing emission to emissions

Page 8, Line 28 – The reference is written in blue

Page 9, Line 2 - The references are written in blue

Page 9, Lines 1-5 – I believe that this section is in reference to Figure 8, but there is citation to Figure 8 listed here.

Conclusions Page 9, Line 10 – Suggest changing observation to observations

Page 9, Line 25 – Suggest adding a the before Eastern

Page 10, Line 9 – It should be aerosol-phase

Data availability Page 10, Line 18 – Suggest changing shared to obtained

Competing interests Page 10, Line 21 – Suggest changing conflict to conflicts

Page 10, Line 26 – Suggest adding a the before U.S. and removing the phrase improvements of after suggestions on the

Disclaimer Page 10, Line 28 – Suggest changing view to views

References Page 11, Line 26 – Believe the accent marks in Muller should be over the u

Tables and Figures Table 1 -In first line of caption – Suggest changing for all the annual to for the sum of the annual -In second line of caption - Suggest adding a the before model -What is the difference between R and R for trends? There is no discussion about this in the main text so it is hard to understand why the two set of values are being shown.

Table 2 -In first line of caption – Suggest adding a the before 10 -In third line of caption – There should be a hyphen in t-test -Second column heading – Suggest changing Regions to Region -Third column heading – Suggest changing # sites to # of sites

Table 3 -Second column heading – Suggest changing Regions to Region -Third column heading – Suggest changing # sites to # of sites

Table 4 -Second column heading – Suggest changing Regions to Region -Third column heading – Suggest changing # sites to # of sites

Table 5 -Second column heading – Suggest changing Regions to Region

Table 6 -In third line of caption – There should be a hyphen in t-test -Second column heading – Suggest changing Regions to Region

Figure 1 -In second line of caption – To match the figure between observations and precipitation-adjusted model results should be switched -In third line of caption – Suggest changing Each NADP to The data at each NADP site -Letters should be added to each plot and the caption updated to indicate this -Suggest making a symbol indicating that green is for East sites and red is for West sites as currently this is only

indicated from the small text at the top of each plot -It should be indicated in the caption what the solid and dashed lines in each plot represent -There are no subscripts in the abbreviations used on both the x and y-axes for all plots

Figure 2 -In first line of caption – Suggest changing of (a) TNO3 to for (a) TNO3 -In second line of caption – Suggest removing the phrase annual accumulated before precipitation. -In second, third, and fourth lines of caption - US should be U.S. -There are no x-axis labels -The legend for plots a, b, and c are incorrect as they indicate the data for the East is red and West is green

Figure 3 -In first line of caption – Suggest changing adding a the before observations -In first line of caption – To match the figure between observations and precipitation-adjusted model valves should be switched -In second line of caption – Suggest changing observation to observational -Letters should be added to each plot and the caption updated to indicate this -It should be indicated in the caption what the solid and dashed lines in each plot represent -There are also no subscripts in the abbreviations used on both the x and y-axes for all plots

Figure 4 -In first and second lines of caption – Suggest changing panel to panels -In third line of caption – Suggest changing plot show p value to plots show p values -In fourth line of caption – Suggest adding a comma after i.e. -There are no x and y-axes labels

Figure 5 -In second line of caption – Suggest changing the right plot show p value great than to both plots show p values greater than -In third line of caption – T-test should be t-test. Also suggest adding a comma after i.e. -Letters should be added to each plot and the caption updated to indicate this -There are no x and y-axes labels

Figure 6 -In first line of caption – Suggest changing (top panel) and DEP (bottom panel) to (top panels) and DDEP (bottom panels) -In second line of caption – Suggest changing plot show p value great than to plots show p values greater than -In third line of caption – T-test should be t-test. Also suggest adding a comma after i.e. -There are no

x and y-axes labels

Figure 7 -There are no x and y-axes labels

Figure 8 -In caption – It should be mentioned in the caption that the percent contribution is being indicated on each bar -In first line of caption – US should be U.S. -On the y-axis for both plots, US should be U.S. -Suggest in legend for plot a calling Oxid as NO3 instead and Red as NHx instead so that it matches the main text

Figure 9 -In second line of caption – Suggest changing an NHx to a NHx -In third line of caption – Suggest removing the comma after 0.5 -There are no x and y-axes labels -Title for plot a – Suggest changing NHx ratio over TIN 1990 to TDEP NHx to TIN ratio 1990 -Title for plot b – Suggest changing NHx ratio over TIN 2010 to TDEP NHx to TIN ratio 2010 -Title for plot c – I am not sure I understand this plot title. What is (/year) indicating? Should the title maybe be TDEP NHx to TIN ratio Overall Trend?

Supporting Information Figure S1 -In caption – Suggest changing all the ofs to equal signs (e.g., 5 of Northern Forests to 5 = Northern Forests) -There are no x and y-axes labels -In plot title – US should be U.S. Also what does mask mean? It is not indicated in the caption or text.

Figure S2 -In first line of caption – Suggest changing plot to plots. Also the words observation and model should be switched to match what is actually plotted. -In second line of caption – Suggest changing data. The site in NADP is assumed to data points. The data at each NADP site is assumed -In third line of caption – Suggest changing valid if only at to valid only if at, changing is available to are available, and changing for the to for that -In fourth line of caption – suggest changing plot to plots -In fifth line of caption – Suggest removing the the before missing -In sixth line of caption – Suggest changing observation to observations -It should be indicated in the caption what the solid lines in the plot represent. Also should this be like the other plots and have two dashed lines and one solid line?

Figure S3 -Letters should be added to each plot and the caption updated to indicate this -Suggest making a symbol indicating that green is for East sites and red is for West sites as currently this is only indicated from the small text at the top of each plot -It should be indicated in the caption what the solid and dashed lines in each plot represent -There are no subscripts in the abbreviations used on both the x and y-axes for all plots

Figure S4 -There are no x and y-axes labels -There are no subscriptions in the abbreviations used in the titles for all plots

Figure S5 -In first line of caption – Suggest adding a the before US. Also US should be U.S. -There are no x-axis labels -Suggest changing y-axis labels to Fraction of the Total -Suggest pointing out on both plots somehow 2003 since this is an important year in terms of trends and so that it corresponds with the discussion in the main text. Maybe add a vertical dashed line.

Table S1 -Either the comma or the parenthesis should be removed from Xing et al. reference. Both are not needed.

Table S3 -In third line of caption – Suggest removing the and with at the end of the sentence

Table S4 -In fourth line of caption - There should be a hyphen in t-test -Second column heading – Suggest changing Regions to Region

Table S5 -Second column heading – Suggest changing Regions to Region

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