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Atmospheric Chemistry and Physics Discussions

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Interactive comment

Interactive comment on "Effect assessment of NO_x and SO_2 control policies on acid species in precipitation from 2005 to 2016 in China based on satellite monitoring" *by* Xiuying Zhang et al.

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1. The major strength of this manuscript is it reports on columnar S/N ratios and shows they are consistent with observational trends. That is also its weakness as it does not go sufficiently beyond that to warrant publication at this time in ACP. It would be better suited to a journal that is more policy focused as it does not provide significant insights to atmospheric chemistry or physics processes. Further, it is, at present, too long given the limited message of the manuscript. (At times, it appears that steps are taken to lengthen the article needlessly.) Another major issue is that the grammar is also not up to ACP standards, and significant editing will be required. Finally, the title

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Discussion paper

is a bit misleading. The major thrust of the article is to use OMI S and N columnar abundances to assess how the ratio of those columns change, and the relationship to precipitation is done via other observations. Not surprisingly, the trends in the columnar abundances have a similar trend to the observations.

Response: We highly appreciate the reviewer's comments. In order to make our manuscript better fit the scope of ACP, we have made significant changes on data and analysis of our study. In this round of revision, we have collected the ground measurements on SO_4^{2-} and NO_3^- concentrations in precipitations from 2005 to 2016 at 60 sites across China. We used the ground measurements to construct the model to estimate wet SO_4^{2-} -S and NO_3^- -N depositions, based on the SO_2 and NO_2 columns in atmospheric boundary layer (ABL). The rationale behind this is that NO_2 reacts with O_3 to form NO_3^- and then highly soluble N_2O_5 ($NO_2 + NO_3 \rightarrow N_2O_5$, $N_2O_5 + H_2O \rightarrow 2\ HNO_3$), thus most of the bulk NO_3^--N in precipitation originates from HNO_3 and aerosol nitrate (NO_3^-) (Barrie, 1985;Liu et al., 2017). Similarly, the rationale of using SO_2 columns to estimate wet S deposition is based on the relationship between SO_2 and SO_4^{2-}. At the gas phase, SO_2 is oxidized by reaction with the hydroxyl radical via an intermolecular reaction (SO_2 + OHâĂć $\rightarrow$ $HOSO_2$âĂć), which is followed by ($HOSO_2$âĂć + SO_2 $\rightarrow$ HO_2âĂć + SO_3); in the presence of water, sulfur trioxide (SO_3) is converted rapidly to sulfuric acid (SO_3 (g) + H_2O (l) $\rightarrow$ H_2SO_4). Furthermore, the wet deposition flux (F) could be estimated by $F = W \times P \times C$, where W is scavenging ratios, P is precipitation amount, and C indicates SO_2 or NO_2 concentrations in atmosphere (Barrie, 1985;Sakata et al., 2006). We used the ground on SO_4^{2-} or NO_3^- concentrations and the ABL SO_2 or NO_2 to estimate the scavenging ratios.

The results showed that SO_2 and NO_2 columns in ABL have potential to estimate wet SO_4^{2-} -S and NO_3^- -N depositions ($R = 0.883$, intercept = 0.903, $P < 0.05$ for SO_4^{2-}–S estimations; and $R = 0.893$, intercept = 0.755, $P < 0.05$ for NO_3^-–N estimations). In this version, we have shifted the focus on discussing the trends of wet SO_4^{2-} -S and NO_3^- -N depositions across China rather than the effect of the air quality polices.

We have tried to polish the language and improve the writing quality of the manuscript. However, if it is still not up to ACP standards, we will send it to professional language editing.

In this version, we have used the wet SO42- -S and NO3- -N depositions to compare the contributions of SO42- and NO3- on precipitation acidity, and further to detect the potential acidity (PA) induced by H2SO4 and HNO3. We have developed the models of linking the trends in S/N and PA with the precipitation amount and ABL SO2 and NO2 columns. This study has generated much fine spatial maps of S/N and PA distribution which can not be achieved by only ground observations.

In this version, we have changed the title "Effect assessment of NOx and SO2 control policies on acid species in precipitation from 2005 to 2016 in China based on satellite monitoring" to "Contribution ratio and trends of SO42- to NO3- to precipitation acidity from 2006 to 2016 in China based on OMI observations", which more focuses on the content of the manuscript.

2. Digging in to the details a bit, the precipitation chemistry data set needs to be better described and documented. Particularly discussing the spatial and temporal observational limitations. From what I can tell, much of the evaluation is based on the 474 standard ground observations. This is comparison is probably one of the key take homes from the article at present. It is not apparent how those 474 locations compare to the "168" records related to the cited Liu et al., publications. This was confusing.

Response: In this revision, we have used the ground measurements on SO42- and NO3- concentrations in precipitations to replace the data collected from published papers. The precipitation samples at 60 sites are collected based on the routine procedure on acid precipitation monitoring technology (HJ/T165ïij■2004). The quality of monitoring data is evaluated and supervised by China National Accreditation Board for Laboratories according to international requirements. The detailed information on the locations of sites, number of the collected precipitation events and spanned time is listed in Table S1.

In the previous version, we estimated the S/N across China through the constructed model based on ABL SO_2, ABL NO_2, and SO_4^{2-} / NO_3^- in the 168 data records from the published papers. We extracted the S/N values from the estimated S/N map, and got the estimated average S/N at 474 sites. In this version, this accuracy assessment has been replaced by that of the outputs from the constructed models with the ground measurements.

3. They need to better define their terms and show what was done with respect to the evaluation. They state "Other parameters of relative error (RE) and absolute error (AE) are used to assess the accuracy of the estimated NO_2 by the following function:" but no function(s) are given. They also never show the AE. Further, it is not really apparent what should be taken from their performance evaluation.

Response: In this version the function of RE is listed as the Eq. (2) and AE is not used. We used the average of RE to evaluate the accuracy of the estimated wet SO_4^{2-} -S and NO_3^- -N depositions.

4. As noted, the grammar is rough. For example, each sentence in the first paragraph is incorrect grammatically. I am still not certain what is meant by the "successfully" in "successfully full provincial names" means. The appropriate term is "evaluation", not "validation" (throughout). Further, this study does not "confirm", but "supports" other findings. They use "might" and other similar terms excessively, probably avoiding saying something more definitive.

Response: Thank you for helping us improve the writing quality of our paper. In this version, we have tried to improve the grammar in this revision.

5. They state "Although the contribution of organic acids to precipitation pH was minor, it could not be neglected, particularly in forest and suburban areas (Stavrakou et al., 2012; Willey et al., 2011)." First, they don't show it is minor. Next, what is meant by

“could not”.

Response: In fact, we just want to discuss the potential uncertainty caused by not considering the contribution of organic acids in this study. In this version, we have changed this sentence to “This study only considered the potential acidity induced by the H2SO4 and HNO3, the organic acid was not considered due to its minor contribution on precipitation acidity (Stavrakou et al., 2012;Willey et al., 2011).”

6. The arbitrary classification of “sulfuric” and “mixed” acid precipitation does not add to the article, and related discussion can be dropped throughout. First, the demarcation is arbitrary. Second, the trend is the split does not add to our understanding of the chemistry or physics. It adds text.

Response: In this version the classification and the related discussions of “sulfuric” and “mixed” acid precipitation have been removed.

7. In terms of other places where excessive text appears to be added, there are often long strings of province names. This hurts readability and adds little to the knowledge that is to be transferred. If there are specific characteristics, those should be discussed (look at the paragraph beginning at line 233.)

Response: We have changed the full name of the provinces to their logograms. We have also cut some excessive texts to make the discussion more concise and focused.

8. This article would be more fitting as a note, again more appropriately in a journal more aligned with air quality policy, greatly shortened and retitled (e.g., Trends in OMI S and N columns and comparison to trends in precipitation composition”). The analysis would show the S and N and S/N trends, and the ratio (The current Fig. 4). In addition to being tightened and more focused, the grammar also needs to be addressed before further consideration by this, or other, journals.

Response: Please refer to our detailed response for #1. In this version, we have shifted the focus from discussing the effect of the air quality polices to the trends of wet SO42-

-S and NO3- -N depositions across China as the reviewer suggested. We have used the wet SO42- -S and NO3- -N depositions to compare the contributions of SO42- and NO3- on precipitation acidity, and further to detect the potential acidity (PA) induced by H2SO4 and HNO3.

The length of the manuscript has been shortened, from the word number of “8306” in the previous version to “6618” in this version.

The core model to estimate the wet SO42- -S and NO3- -N depositions in this study is a statistical model combining ground measurements and satellite observations. This estimation had the advantages of high spatial resolution. We resolved the problem of wet deposition from another view point apart from the atmospheric chemical transport (ACT) theory, and the key point is that we gained a reliable result. This is our contribution.

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