Response to Referee # 1:

Interactive comment on "Spatial distribution and seasonal variations of atmospheric sulfur deposition over Northern China" by Y. P. Pan et al.

Anonymous Referee #1

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General Comments:

The paper presents measured and estimated total sulfur atmospheric deposition at 10 sites in northern China. Such a data set is needed for this region for ecosystem studies and for making emission control policies. The methodology is scientifically sound and the results are valid. The presentation of the results could be improved as listed below.

Response: The authors appreciate the constructive and valuable suggestions given by Referee # 1 for improving the overall quality of the manuscript. In the revised version we have made substantial changes by reorganizing the structure of the paper and focused on spatial variations of the dry and wet depositions. As suggested, we have added the concentrations and dry deposition velocities before discussing dry deposition amounts to strengthen the paper. We have also clarified the causes of the spatial patterns of S deposition, especially for wet deposition on the basis of scavenging ratios. Detailed responses to the comments are given below.

Most materials presented in Section 2 are the same as those presented in another paper by the same authors and should be significantly simplified by referring to the previous publication (Pan et al., ACP, 2012).

Response: Thanks for this comment. In preparation of the original manuscript, we also concerned the issue pointed out by the reviewer and try the best to avoid repetitive materials. In fact, most materials in Sect. 2 are different to our previous paper (e.g., emission inventory of SO_2 and calculations of total acid deposition flux), with the exception of the first paragraph of Sect. 2.1, second paragraph in Sect. 2.3 and 2.4. Therefore, the above mentioned paragraphs were significantly shortened in

the revised version. Some repetitive sentences in other paragraphs were also removed or briefly described by referring to the previous publication. Considering that the monitoring methodology is most important for interpretation and crucial for the reader, other necessary information presented in Sect. 2 is reserved in the revised version.

The estimated SO2 dry deposition from using the inferential method contributed to nearly half of the total deposition budget. A brief discussion on the concentrations and dry deposition velocities is first needed before discussing dry deposition amounts. This will allow the readers to find out if the deposition velocities used here are in reasonable range. Presentation of SO2 and sulfate concentrations will also help to explain the spatial and annual variations in their dry and wet deposition patterns, instead of simply attributing the variations to emission sources (as frequently done in many places in this paper).

Response: We agree with these comments, and have added the concentrations and dry deposition velocities of SO_2 before discussing gaseous dry deposition. Measurement of ambient sulfate concentrations was not carried out in this study; however, chemical analysis of size-resolved particles was performed at the sites of BD, BJ, TJ, TS and XL during 2009-2010, as part of the Science and Technology Project of Beijing on air quality. In response to the comments above, we have added these available data of sulfate concentrations and limited our discussions to the above sites. We have not yet acquired the aerosol samples from the other five sites, as we had hoped, so we cannot address the importance of ambient sulfate concentrations to dry and wet deposition at all the sites. In the revised paper where appropriate, the spatial and annual variations of dry and wet deposition were explained using SO_2 and sulfate concentrations.

It is the total annual deposition that is of concern to ecosystem study. Discussions on annual average of dry and wet deposition and their spatial patterns and the causes of these patterns should be the focus of the paper. Discussions of monthly and seasonal variations should be brief. Too many detailed information makes the paper difficult to read.

Response: The reviewer is correct that we should focus the discussions on annual dry and wet deposition and their spatial patterns and the causes of these patterns when we concern their ecological impacts. However at which month or season does sulfur deposition occur? Such information is also important for emission control strategies such as mitigation of SO_2 from coal burning during home heating period. Since the measurement data of S deposition in the target areas is lacking, detailed interpretation of the results will be helpful for future studies. Because of these two issues, the authors consider that the discussions on seasonal variations are necessary. However, we agree that such discussions should be brief. As suggested, we have added discussions on spatial patterns and removed some sentences regarding monthly and seasonal variations, in particular for wet deposition. Discussions on dry deposition were re-organized and interpreted by concentrations or deposition velocities in the revised version. Sub-section titles were added to assure the paper is easier to follow.

Page 23657. line 3: using "local emissions" was not accurate here. Wet deposition amount includes contributions from in-cloud scavenging (most of which might be from long range transport) and below-cloud scavenging (part of which might also be from long-range transport). You could simply state that "part of the differences was caused by differences in ambient concentrations). Again, as suggested above, if ambient concentrations of SO2 and sulfate were first briefly discussed, it would be easier to discuss spatial variations in wet deposition. Such information is also needed for discussions on page 23662, line 15 (and discussion in Section 4). There is no evidence to conclude that the differences were caused by scavenging ratios. Differences in ambient concentrations are likely the main causes.

Response: The authors accept these comments, and have revised the text as suggested. To clarify the spatial variations of wet deposition, a new section was added in the revised paper. The influences of precipitation, scavenging ratios and concentrations on the regional pattern of wet deposition were discussed. Finally, we arrived at a major conclusion that wet deposition has less dependence on local emissions because it depends on column concentrations and in-cloud scavenging.

Page 23662. The first paragraph: You simply use emission inventory to explain everything in this paper including this paragraph. A brief explain of the following theory is needed somewhere: "Dry deposition is mostly decided by surface concentration which has a close link with local emission. Wet deposition has less dependence on local emission because it depends on column concentration and in-cloud scavenging."

Response: We agree with these insightful comments. We have revised the relevant sections to reflect this theory. In the revised manuscript, SO_2 and sulfate concentrations were used to explain the spatial and annual variations in their dry and wet deposition patterns, instead of simply attributing the variations to emission sources, as suggested above. Such changes were done in places where appropriate.

Page 23666. First paragraph of "Conclusions": Introductory materials should be removed from the Conclusion section. Also avoid repetitive materials through the text.

Response: We agree, and have removed the introductory materials in the conclusion. We have also checked through the text to avoid repetitive materials.

In summary, the paper presents too many detailed information on less important issues, but lack of in-depth analysis on some important issues.

Response: Thanks again for your insightful and useful comments above, and also below. We will definitely incorporate your suggestions. They will make the paper stronger. We have added SO_2 and sulfate data in the revised manuscript so that a more in-depth description and discussion of the spatial and annual variations could be made. We have proposed to re-organize the structure and add sub-sections in order to highlight the discussions on important issues, as suggested above by the reviewer. These substantial changes will strengthen our manuscript.

Minor comments

Page 23646. Line 18: scavenging ratio was never investigated in the paper. The statement is just a speculation. Line 21: "constant" is confusing, from year over year or from site over site? Same issue on page 23667, line 18. Consider rewriting the sentences in both places.

Response: "Constant" here means that the total deposition has no distinct seasonality. Both sentences were rewritten to avoid confusion.

To discuss the relative importance of precipitation amount, concentrations of SO_2 and SO_4^{2-} and scavenging ratios on the spatial pattern of wet deposition, in the revised paper we have calculated the scavenging ratios on a mass basis (W=Cp/Ca), following Sakata et al. (2006). The concept of the scavenging ratios is on the basis of the simplified assumption that the concentrations of SO_4^{2-} in precipitation (Cp) depends on the ambient concentrations of the SO_2 and SO_4^{2-}. The results showed that the scavenging ratio was not the main causes of regional differences.

Page 23648. Lines 22-23, you meant 20% for the territory, not for the extent of the exceedance of critical loads, this is not consistent with the subject. Consider rewriting the sentence.

Response: The sentence was changed to: "…, the areas exceeding the critical loads for soil acidification caused by S deposition is still larger than 20% of the country's territory in the 2010s, mainly in eastern and south-central China (Zhao et al., 2009)."

Page 23649. line 7: change "few" to "a few". Lines 19-20: use either "although" or "however", not both. Line 21: "fewer"-compared to what? Consider change it to a different word such as "limited".

Response: These suggestions are implemented.

Page 23652. Line 5: "30min x 30min"? Do you mean "30 km x 30km"? Also check figure 1 caption.

Response: We are sorry for this confusion. In the revised version, it was changed to "at a spatial resolution of $0.5° \times 0.5°$". Caption of figure 1 was also updated.

Page 23654. Line 1: change “measurements” to “estimation” since SO2 deposition was not measured.

Response: We accept this suggestion and have modified the title.

Page 23657. Lines 16: Consider changing “less” to “low”.

Response: The suggestion is implemented.

Page 23660. the first several lines should be in “Introduction”. Section 4.1 fits better in Section 3 (as section 3.4) than in Section 4.

Response: We agree, and have merged the results and discussion into a combined section. Section 4.1 will be section 3.4 in the revised version. The first several lines here were removed because such information was touched in Introductions section.

Page 23661. Line 24-27: split the sentence into two separate sentences. This sentence and the previous one discussed three cases: sites with high deposition and emission; sites with low deposition and emission; and sites that did not have consistent deposition and emission. Use three separate sentences to discuss the three cases; do not put the latter two together.

Response: Good idea. We have modified these sentences as suggested.

Page 23662. line 27: consider changing “ fluctuations” to “ differences”.

Response: The suggestion is implemented.

Page 23633. First sentence: was the low SO2 % at BJ caused by the low deposition velocities over urban land use categories, or by low concentrations?

Response: Thanks for this comment. We have done the analysis as suggested, and find that the low contribution of $_{g}SO_2$% at BJ was caused by the low concentrations. The relative importance of the deposition velocities and concentrations on the variations of dry deposition has been discussed in the revised paper.

References

Sakata, M., Marumoto, K., Narukawa, M., and Asakura, K.: Regional variations in wet and dry deposition fluxes of trace elements in Japan, Atmos. Environ., 40, 521-531, doi:10.1016/j.atmosenv.2005.09.066, 2006.

Zhao, Y., Duan, L., Xing, J., Larssen, T., Nielsen, C. P., and Hao, J.: Soil acidification in China: Is controlling SO_2 emissions enough?, Environ. Sci. Technol., 43, 8021-8026, doi:10.1021/es901430n, 2009.