Response to Referee # 2:

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

Anonymous Referee #2

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General comments:

The authors present spatial distribution and seasonal variations in dry and wet deposition of sulfur in Northern China, based on both modeling results and measurements at ten sites during a 3-yr observation campaign. Atmospheric deposition of acidic constituents is of great concern in China as its ongoing industrialization might result in substantial release of acidic species to the atmosphere, whereas the situation has not yet been well understood for its most regions. The results are very important for better understanding the magnitudes, pathways, and variations of acid deposition, thus should be worthy of publication in ACP. The major concerns of this reviewer are organization of the manuscript. In addition, the general use of the English language also needs to be polished.

Response: The authors appreciate the valuable suggestions given by Referee # 2 for improving the overall quality of the manuscript. In the revised version we have made significant changes by reorganizing the structure of the paper, so as to make the text easier to follow. As suggested, we have added a new table to include the flux data. We will also ask several native English speakers to further polish the language of the revised paper when it is ready. Detailed responses to the comments are given below.

Specific comments

1. The title and content of the manuscript do not match with each other. Nitrogen deposition (which has been reported previously by the same authors (Pan et al., ACP,

2012)) and total acid deposition are also included in the paper whereas the title is about sulfur deposition. Please make a proper title, or focus on sulfur deposition and remove contents about nitrogen and total acid deposition.

Response: We agree with this comment. In the revised paper, we focused on the spatial distribution and temporal variations (Inter-annual and Intra-annual trends) of sulfur deposition. To evaluate the potential acidification risks, annual S deposition were further combined with that of N deposition. Therefore, we have changed the title to "Spatial distribution and temporal variations of atmospheric sulfur deposition over Northern China-Insights into potential acidification risks".

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

Response: Thank you for this suggestion. The authors also concerned the issue pointed out by reviewer and try the best to avoid repetitive materials in the present manuscript. Nonetheless, monitoring methodology is most important for interpretation and crucial for the reader. With the exception of the first paragraph of Sect. 2.1, second paragraph in Sect. 2.3 and 2.4, most of the rest materials are different to our previous paper (e.g., emission inventory of SO_2 and calculations of total acid deposition flux). Therefore, only the above mentioned three paragraphs were significantly simplified whereas the information in other paragraphs is necessary and only some repetitive sentences were removed or briefly described by referring to the previous publication. It would be helpful to reserve the detailed information presented in Sect. 2 for the reader's convenience.

3. Page 23651: Site descriptions. Please use table to show the GPS, so as to make the text easier to read.

Response: As suggested above (comment #2), site descriptions were simplified and the GPS was removed here. We will add the location information in a new table, which is showing the flux data according to suggestion below (comment #5).

4. Page 23652, line 5 and page 23673, title of Fig. 1: "30min x 30min", typos? **Response:** The unit was changed to "at a spatial resolution of $0.5^{\circ} \times 0.5^{\circ}$ " in the revised version. Caption of figure 1 was also updated.

5. Page 23656 Results: data description. Please use table to show the data directly. **Response:** We agree with the comment, and will add a table instead of Fig. 2 to show the flux data in the revised paper. To avoid repetitive description of the results, Fig. 2 in the original version was removed.

6. Page 23657, line 23: It is better to move "the capital of Hebei province" to the Sect.2.1 (site descriptions).

Response: As suggested above (comment #2), we have removed detailed descriptions of sites. Thus, the note of Shijiazhuang is reserved here.

7. Page 23660, line 6: Reference (s) is/are needed here to support modeling studies.**Response:** The first two sentences here have been removed in the revised paper to avoid repetitive presentations.

8. Page 23659, line 22: In addition to the evidence from emission inventory, the declining trend of SO2 was identified by satellite measurements over China during 2004-2009 (Zhang et al., 2012). Zhang, X., Van Geffen, J., Liao, H., Zhang, P., and Lou, S.: Spatiotemporal variations of tropospheric SO2 over China by SCIAMACHY observations during 2004–2009, Atmos. Environ., 60, 238-246, doi:10.1016/j.atmosenv.2012.06.009,2012.

Response: Thanks for the suggestion. The reference was inserted in the revised paper to support the decreasing tendency of SO_2 concentrations.

9. Page 23664, line 17: Reference (s) is/are needed here to support the previous studies.

Response: The suggestion is implemented.

10. Page 23666 conclusions: remove the first paragraph since it is not conclusion. The content could be integrated in the introduction section (i.e. Section 1).

Response: We agree to remove the introductory materials in the conclusion. In the revised version, the first paragraph in the conclusion has been rewritten.

11. Page 23667, line 27: It is not discussed in the text that "the estimated acid deposition exceeds the critical loads of natural ecosystems", although S deposition has been framed within the concept of "critical loads" in Page 23665/lines 12-15. Please clarify.

Response: We have added the following sentences in the revised manuscript: "In addition, Northern China is now receiving N deposition at rates higher than the critical loads (<u>Pan et al., 2012</u>), and modeling results predict that the region will likely receive increased N deposition in the future (<u>Zhao et al., 2009</u>). Therefore, long-term impacts of N and its combined effects with S in driving acidification of terrestrial and aquatic ecosystems are expected."

References

- Pan, Y. P., Wang, Y. S., Tang, G. Q., and Wu, D.: Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China, Atmos. Chem. Phys., 12, 6515-6535, doi:10.5194/acp-12-6515-2012, 2012.
- Zhao, Y., Duan, L., Xing, J., Larssen, T., Nielsen, C. P., and Hao, J.: Soil acidification in China: Is controlling SO₂ emissions enough?, Environ. Sci. Technol., 43, 8021-8026, doi:10.1021/es901430n, 2009.