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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric nitrogen deposition to the northwestern Pacific: seasonal variation and source attribution" *by* Y. H. Zhao et al.

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Comment: This paper discussed atmospheric N deposition in the northeastern Pacific Ocean in relation to anthropogenic and natural reactive N emissions using the GEOS-chemical global chemistry model. The authors provided very important information on both dry and wet N deposition in Yellow Sea and South China Sea and compare their results with satellite data and emission inventory data with regional differences in anthropogenic reactive N sources. This is a significant contribution to scientific knowledge on how the terrestrial reactive N emissions affect N wet and dry deposition onto the northeastern Pacific Ocean (e.g. Yellow Sea and South China Seas).

Response: We thank the reviewer for the helpful comments. All of them have been addressed in the revised manuscript. Please see our itemized responses below.

Scientific comments: The authors jointly used modeling tool, satellite observation and surface measurement to obtain relatively accurate and comprehensive information on atmospheric nitrogen deposition to the northwestern Pacific, especially the China Seas. The results of this study contribute to a better understanding of coastal atmospheric N deposition and help to make effective strategies for mitigating N deposition. To further improve the quality of the manuscript, I suggest that a section of the uncertainty analysis (as also mentioned in later) may be presented in the text.

Response: We have added the following paragraphs in the conclusion section to discuss the main uncertainties.

"While this study provides a pilot investigation of the sources and processes controlling atmospheric nitrogen deposition to the northwestern Pacific, some uncertainties still need to be considered. A main uncertainty is associated with the lack of in-situ measurements to evaluate the model simulated nitrogen dry deposition fluxes. Uncertainties exist in both model calculated dry deposition velocities over the ocean surface (as discussed in section 2.1) and simulated surface concentrations of nitrogen species. Recent studies have shown that GEOS-Chem overestimates wintertime surface concentrations of nitrate and nitric acid (Heald et al., 2012; Zhang et al., 2012; Wang et al., 2013), which can lead to a model overestimation of NOy dry deposition flux in winter.

Uncertainties also exist in Asian NH3 emissions; in particular, air-surface bi-directional NH3 fluxes are not considered in the study. Although it has little impact on the oceanic emissions, recent implementations of the bi-directional NH3 flux on fertilizer use showed lower NH3 agricultural emissions over China (Fu et al., 2015; Zhu et al., 2015), and thus would lower its transport to the ocean. In addition, any bias in the GEOS-Chem simulation would affect the adjoint sensitivity. Also to ascribe nitrogen

deposition to sources from different emission sectors, we rely on the bottom-up sectorial emissions to separate the adjoint sensitivity. Even though the total emissions can be constrained with the satellite measurements, the sectorial information is subject to larger uncertainties (Zhang et al., 2009). We recommend future research to reduce these uncertainties."

Those references were added:
Fu, X., Wang, S. X., Ran, L. M., Pleim, J. E., Cooter, E., Bash, J. O., Benson, V., and Hao, J. M.: Estimating NH3 emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model, Atmos. Chem. Phys., 15, 6637-6649, 10.5194/acp-15-6637-2015, 2015.
Heald, C. L., Collett, J. L., Lee, T., Benedict, K. B., Schwandner, F. M., Li, Y., Clarisse, L., Hurtmans, D. R., Van Damme, M., Clerbaux, C., Coheur, P. F., Philip, S., Martin, R. V., and Pye, H. O. T.: Atmospheric ammonia and particulate inorganic nitrogen over the United States, Atmos. Chem. Phys., 12, 10295-10312, 2012.
Wang, Y., Zhang, Q. Q., He, K., Zhang, Q., and Chai, L.: Sulfate-nitrate-ammonium aerosols over China: response to 2000-2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia, Atmos. Chem. Phys., 13, 2635-2652, 2013.
Zhu, L., Henze, D., Bash, J., Jeong, G.-R., Cady-Pereira, K., Shephard, M., Luo, M., Paulot, F., and Capps, S.: Global evaluation of ammonia bi-directional exchange, Atmos. Chem. Phys. Discuss., 15, 4823-4877, doi:10.5194/acpd-15-4823-2015, 2015.

Technical corrections/Comments

Comment: Introduction Page13660. Line 7. The formation of ammonium particles increases . . .
Response: Changed as suggested.

Comment: Page13660. Line 8. As dry removal of the particles . . .
Response: We changed to "As dry removal of particles is slow"

Comment: Sect. 2.1 General description Page 13662. Lines 10, 15. The monthly dry deposition velocities of Nr species (e.g. NO2 and NH3) over the northwestern Pacific and seasonality of them differ from the results reported by Zhang et al. (2010) over the China Seas. Did this study consider impact of the sea-surface height on the velocities? How is the reliability if using the current deposition velocities to the China Seas?
Reference mentioned: Zhang Y., et al. Atmospheric deposition of inorganic nitrogen to the eastern China seas and its implications to marine biogeochemistry. Journal of Geophysical Research, Vol. 115, D00K10, doi: 10.1029/2009JD012814, 2010.

Response: We now state in the text "For gaseous NH3 and HNO3, Zhang et al. (2010) estimated similar dry deposition velocities (0.5-0.85 cm s-1) over the eastern China seas in spring-fall using the MM5/CMAQ model, but suggested minimum deposition velocities in winter (0.5 cm s-1 versus 1.10-1.16 cm s-1 in our estimates). Understanding this discrepancy would require a close examination of differences between the two studies, such as different simulation years and different air-sea roughness parameterizations in the two models. Zhang et al. (2010) modified the sea-surface roughness length by considering the impact of sea-surface height, while GEOS-5 used in this study follows the Monin-Obhukov similarity theory with improved parameters to match recent air-sea exchange observations (Garfinkel et al., 2011)."

Added references:
Zhang, Y., Yu, Q., Ma, W. C., and Chen, L. M.: Atmospheric deposition of inorganic nitrogen to the eastern China seas and its implications to marine biogeochemistry, J. Geophys. Res.-Atmos., 115, D00K10, 2010.

Garfinkel, C. I., Molod, A. M., Oman, L. D., and Song, I. S.: Improvement of the GEOS-5 AGCM upon updating the air-sea roughness parameterization, Geophys. Res. Lett., 38, L18702, 2011.

Comment: 3 Column concentrations and wet deposition fluxes over Asia Page 13667. Line 5. The highest sensitivities
Response: Changed as suggested (Added 'the').

Comment: Page 13668. Line 5. This study shows that Annually model simulated nitrogen wet deposition (NH4+ + NO3-) fluxes over China averages 9.3 kg N ha-1 a-1 with NH4+ contributing 70 %. However, this modeled magnitude of wet deposition was 1.6-times lower that the results reported by recent studies (Jia et al., 2014, 13.9 kg N ha-1 a-1; Zhu et al., 2015, 13.2 kg N ha-1 a-1) based on published large amount of Chinese surface measurements. In addition, although the contribution of NH4+ to total wet N deposition (70%) was similar to that in north China (Pan et al., 2012, in the range of 63-78%), it might be overestimated at the national scale as an average value of 55% has been observed by Zhu et al. (2015) based on 41 in situ monitoring sites across China. Therefore, the modeled flux of wet deposition may have some uncertainties. Please make a comprehensive comparison in the text.
References mentioned:
Jia, Y. L. et al., 2014. Spatial and decadal variations in inorganic nitrogen wet deposition in China induced by human activity. Sci. Rep., 4, 3763.
Pan, Y. P. et al. 2012. Wet and dry deposition of atmospheric nitrogen at ten sites in Nor thern China. Atmos. Chem. Phys., 12, 6515-6535.
Zhu, J. X. et al., 2015. The composition, spatial patter ns, and influencing factors of atmospheric wet nitrogen deposition in Chinese terrestrial ecosystems. Sci. Total Environ., 511, 777-785.

Response: We now add in the text: "Compared with previous studies using ensembles of surface measurements, our estimated annual nitrogen wet deposition over China is 30% lower than the estimates of 13.9 kg N ha-1 a-1 by Jia et al. (2014) and 13.2 kg N ha-1 a-1 by Zhu et al. (2015), but is consistent with 9.88 kg N ha-1 a-1 by Lv and Tian (2007). The NH4+ contribution to wet deposition is higher than that estimated by Zhu et al. (2015) (55%), but is consistent with Lv and Tian (2007) (72%) and Pan et al. (2012) (63-78% over North China).

Added reference:
Jia, Y., Yu, G., He, N., Zhan, X., Fang, H., Sheng, W., Zuo, Y., Zhang, D., and Wang, Q.: Spatial and decadal variations in inorganic nitrogen wet deposition in China induced by human activity, Scientific reports, 4, 3763, 10.1038/srep03763, 2014.
Zhu, J., He, N., Wang, Q., Yuan, G., Wen, D., Yu, G., and Jia, Y.: The composition, spatial patterns, and influencing factors of atmospheric wet nitrogen deposition in Chinese terrestrial ecosystems, Sci. Total Environ., 511, 777-785, 2015.

Comment: Page 13668. Line 5. are greater than 0.7. **Response:** Changed as suggested (changed 'great' to 'greater').

Comment: Page 13668. Lines 7-8. This is similar to Lv et al. (2007) who estimated . . . **Response:** This has been changed according to an earlier comment.

Comment: 4.1 Seasonal variation and deposition process Page 13669. Line 25. Accounting to Zhang et al. (2012), there were some uncertainties on seasonal amounts of NOy deposition modeled by Geos-Chem. Does this affect the current findings? Please clarify.
Reference mentioned: Zhang, L., et al. 2012. Nitrogen Deposition to the United

States: Distribution, Sources, and Processes, Atmos. Chem. Phys., 12, 4539-4554.

Response: This has been addressed in replying the earlier comment on uncertainties.

We added in the text “Recent studies have shown that GEOS-Chem overestimates wintertime surface concentrations of nitrate and nitric acid (Heald et al., 2012; Zhang et al., 2012; Wang et al., 2013), which can lead to a model overestimation of NOy dry deposition flux in winter.”

Comment: Page 13670. Lines 1-2. higher than in April and July **Response:** Changed as suggested (added ‘in’).

Comment: Page 13671. Line 10. Spatial and seasonal variations of atmospheric nitrogen... **Response:** Changed as suggested (changed ‘variation’ to ‘variations’).

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 13657, 2015.