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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 presents a calculation of the total nitrogen flux to the Yellow and South China Seas, and also determines contributions from various emission source categories to that flux. It appears to be a very thorough paper, and contributes to the further understanding of nitrogen deposition to oceans. It is well organized and well written and I recommend it be published in ACP, after addressing a few details below.

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.

Comment: P 13659, L 1: I think you should be a bit more precise here, especially for the last line of your abstract, and which could have significant policy implications. Maybe change to "limiting the effectiveness of NH3 emission controls on reducing nitrogen deposition to the Yellow and South China Seas". At first glance it reads like reducing NH3 isńt useful at all

Response: As suggested, we changed this sentence to "limiting the effectiveness of NH3 emission controls on reducing nitrogen deposition to the Yellow Sea."

Comment: P 13660, L 10: If 40% enters ocean, does other 60% end up on land generally? (Ie, is this global?)

Response: Yes, we modified the sentence to "Globally a large fraction (40%) of emitted NH3 and NOx enters the ocean via wet and dry deposition from the atmosphere, and the rest 60% is deposited over the land (Duce et al., 2008)."

Comment: P 13663: I believe nighttime GEOS mixed layer depth in this version of GEOS-Chem had some problems. What do you do for mixed layer depth? Does it influence the results at all?

Response: The PBL problem has been corrected in our simulations.

We added in the model description section 2.1 "The GEOS-5 data have a low bias for nighttime planetary boundary layer height (PBLH). This has been corrected by setting a minimum PBLH computed as a function of local friction velocity (Koracin and Bberkowicz, 1988; Sajeev Philip; http://wiki.seas.harvard.edu/geos-chem/index.php/Boundary_layer_mixing).".

Added references
Koracin, D., and Berkowicz, R.: Nocturnal boundary-layer height: Observations by acoustic sounders and predictions in terms of surface-layer parameters, Boundary-Layer Meteorol., 43, 65-83, 1988.

Comment: You mention NH3 from the oceans here and then later in text/figures, but for the meantime, it would be nice to get an idea of the magnitude of NH3 oceanic emissions when you are discussing N emissions from Asia. What fraction of the total natural NH3 is from the oceans? (Maybe I missed this.)

Response: We now state in the text "24% of the natural NH3 emissions are from the oceanic emissions (0.50 Tg N a-1) over the region." We have also listed the value (0.50 Tg N a-1) in Table 2.

Comment: I don't believe GEOS-Chem has bidirectional exchange in the model, which may cause uncertainties in net flux for certain nitrogen species. Will this influence ocean estimates at all?

Response: We have added in the section 2.2 (emissions) "Here we have not considered air-surface bi-directional exchange of NH3 (Sutton et al., 1998), and treat the NH3 fluxes as uncoupled emission and deposition processes.".

We also state in the conclusion "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.".

Added references:
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.
Sutton, M. A., Burkhardt, J. K., Guerin, D., Nemitz, E., and Fowler, D.: Development of resistance models to describe measurements of bi-directional ammonia surface-atmosphere exchange, Atmos. Environ., 32, 473–480, 1998.
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.

Comment: Satellite data: Can you be a bit more specific about what exactly you are trying to achieve with this satellite validation? A spatial validation of GEOS-Chem NOx emissions? Since NO2 has such a small deposition velocity, why care about NO2?

Response: We now state in the text "Figure 2 compares GEOS-Chem simulated NH3 and NO2 tropospheric columns with satellite measurements. These comparisons provide valuable tests of the nitrogen emissions and their spatial distributions in the model since both NH3 and NO2 have short lifetimes in the atmosphere. Although NO2 has a small dry deposition velocity (Table 1), it rapidly converts to other NOy species, thus NO2 emissions still largely control the deposition of NOy."

Comment: There have been lots of OMI NO2 comparisons with models. Maybe list if there are DOMINO NO2 and GEOS-Chem papers already published, or at least DOMINO NO2 validation to show OMI is useful.

Response: We state in the text “The DOMINO NO2 data has been validated against surface and aircraft observations (Boersma et al., 2008; 2009; Hains et al., 2010), and used to constrain NOx emissions in the model (Boersma et al., 2008; Lamsal et al., 2010).”

Added references:

Boersma, K. F., Jacob, D., Bucsela, E., Perring, A., Dirksen, R., van der A, R., Yantosca, R., Park, R., Wenig, M., Bertram, T., and Cohen, R.: Validation of OMI tropospheric NO2 observations during INTEX-B and application to constrain NOx emissions over the eastern United States and Mexico, Atmos. Environ., 42(19), 4480–4497, 2008.

Boersma, K. F., Jacob, D. J., Trainic, M., Rudich, Y., DeSmedt, I., Dirksen, R., and Eskes, H. J.: Validation of urban NO2 concentrations and their diurnal and seasonal variations observed from the SCIAMACHY and OMI sensors using in situ surface measurements in Israeli cities, Atmos. Chem. Phys., 9, 3867-3879, doi:10.5194/acp-9-3867-2009, 2009.

Hains, J. C., Boersma, K., Kroon, M., Dirksen, R., Cohen, R., Perring, A., Bucsela, E., Volten, H., Swart, D., Richter, A., Wittrock, F., Schoenhardt, A., Wagner, T., Ibrahim, O., van Roozendael, M., Pinardi, G., Gleason, J., Veefkind, P., and Levelt, P.: Testing and Improving OMI DOMINO Tropospheric NO2 Using Observations from the DANDELIONS and INTEXB Validation Campaigns, J. Geophys. Res., 115, D05301, doi:10.1029/2009JD012399, 2010.

Lamsal, L. N., Martin, R. V., van Donkelaar, A., Celarier, E. A., Bucsela, E. J., Boersma, K. F., Dirksen, R., Luo, C., and Wang, Y.: Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern midlatitudes, J. Geophys. Res., 115, D05302, 10.1029/2009jd013351, 2010.

Comment: Have you used the OMI scattering weights (column averaging ker-

nels) to compare with the model? Huijnen et al 2010 showed the kind of differences that ignoring these can cause in model comparisons (Huijnen et al. "Comparison of OMI NO2 tropospheric columns with an ensemble of global and European regional air quality models." Atmospheric Chemistry and Physics 10.7 (2010): 3273-3296.)

Response: We now state in the text “Recent studies have indicated that DOMINO NO2 columns might be biased high due to the a priori profile shape, error in the surface air-mass factor, and exclusion of aerosols in the retrieval (Hains et al., 2010; Lamsal et al., 2010; Lin et al., 2014). The comparison also did not apply the averaging kernels to the model simulated columns, which may lead to additional biases when simulated NO2 vertical profiles are different from the a priori profiles used in the OMI retrievals (Huijnen et al., 2010).”

Add the following reference:

Huijnen, V., Eskes, H. J., Poupkou, A., Elbern, H., Boersma, K. F., Foret, G., Sofiev, M., Valdebenito, A., Flemming, J., Stein, O., Gross, A., Robertson, L., D'Isidoro, M., Kioutsioukis, I., Friese, E., Amstrup, B., Bergstrom, R., Strunk, A., Vira, J., Zyryanov, D., Maurizi, A., Melas, D., Peuch, V. H., and Zerefos, C.: Comparison of OMI NO2 tropospheric columns with an ensemble of global and European regional air quality models, Atmos. Chem. Phys., 10, 3273-3296, 2010.

Comment: Why is OMI data not treated as TES data and matched to GEOS-Chem coincidence along track? Do you expect any kind of bias might result? What is the cloud filtering criterion for these data?

Response: We now state in the text “The GEOS-Chem model results for 2009 are sampled along the TES orbit tracks at the overpass time (the standard TES products are level-2 data due to the sparse daily spatial coverage)” and “we filter the TES observations based on the retrieval quality control flags, and only use the daytime observations with cloud optical depth < 1.0.”

For OMI, we now state “To facilitate the comparison we use the monthly gridded tropospheric NO2 column data which are averages of the retrievals with cloud radiance fraction < 50% (http://www.temis.nl/docs/readme_tomsascii.pdf)”

Comment: Thereś not much discussion of uncertainties in the paper. Iḿ mostly wondering about the adjoint. Is there a way to estimate uncertainties in these contribution estimates?

Response: We have added the following paragraphs to discuss uncertainties of the study, including uncertainty from the adjoint source contribution.

“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.”
Added references:
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.

Minor Comment:

P13658, L 15: Change “downwind the Asian” to “downwind of the Asian”

P13659, L 8: Remove word “But” (never start a formal sentence with but).

P13659, L 24: You are not really addressing the issue (that’s for policy makers). Change “address” to “study

Response: Those comments are all changed as suggested.

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