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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nitrogen deposition to the United States: distribution, sources, and processes" *by* L. Zhang et al.

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Comment: Overall this is an excellent paper. Nitrogen deposition is an important topic that is difficult to accurately quantify. Some consider it "the pursuit of the unknowable"! The authors present a reasonable model and compare results with other independent estimates (CASTNET, NADP, CAPMoN, Harvard Forest etc.). The manuscript does a good job of clearly documenting the assumptions, the limitations, and the digressions from other deposition estimates by the model. They proceed to make all the relevant comparisons (wet vs dry deposition, importance of individual N-species contribution, oxidized vs reduced N sources and deposition, anthropogenic vs natural sources of N etc.), and relate their findings to ecological impact (e.g. N critical loads). While the results relating to dry deposition come with some error associated with their estimates,

their findings demonstrate a reasonable estimate of the relative importance of the various N-species to overall N-deposition. Future research will no doubt refine these deposition estimates. Frankly, this manuscript is very close to final form for publication. The authors may want to look at Sparks et al. (Global Change Biology (2008) 14, 768–781, doi: 10.1111/j.1365-2486.2007.01526.x) for a ground truth comparison of dry N measurements (at Duke Experimental Forest in 2003) as they have done for Harvard Forest. More ground truthing of model results with measured values, especially where there is little information (e.g. NOy), certainly helps in model validation.

Response: Thank you for the detailed comments. We have addressed them in the revised manuscript.

We added in the text "Sparks et al. [2008] measured NOy eddy flux measurements at Duke Forest (35.97°N, 79.08°W), North Carolina and estimated an annual NOy dry deposition flux of 4.3 kg N ha-1 a-1 in 2003. The model is too high (7.2 kg N ha-1 a-1) at that site, similar to the comparison at Harvard Forest. However, Sparks et al. [2008] stated that their NOy eddy flux measurements could be biased low by up to 35% due to loss of HNO3 within the instrument inlet."

We added the comparison of measured and simulated NOy concentrations at Harvard Forest in Figure 8. We added in the text "Figure 8 compares the monthly mean NOy concentration and eddy covariance flux measurements at Harvard Forest for 1999-2002 to model results for 2006-2008. Measured NOy concentrations peak in winter and are minimum in summer, with annual means of 6.0-6.2 ppbv. The model reproduces closely the observed values and their seasonal variation."

Comment: An area in the manuscript that was not entirely clear was the treatment of bi-directional flow for NH3 (and NO2). On pg.246, lines 25 to 27, it is stated that it "is treated here as uncoupled emission and deposition processes." Does this mean the deposition velocity (Vd) is a "net" Vd because the NH3 flux back to the atmosphere is counted in the emissions? I am guessing this is the case judging from the Table 1 V(d)

for NH3. The CMAQ model (Version 5.0), which incorporates a bi-directional NH3 flux, reduces the NH3 deposition significantly in the northeast compared to earlier CMAQ runs, which do not incorporate a bi-directional flux.

Response: We now state in the text "Biosphere-atmosphere exchange of NOx and NH3 is bi-directional (Sutton et al., 1998; Lerdau et al., 2000; Ellis et al., 2011), but is treated here as uncoupled emission and deposition processes. The fluxes to the surface are parameterized as uni-directional dry deposition, and the fluxes back to the atmosphere are included as part of natural emissions."

Comment: Another possible improvement to the manuscript would be to compare the model results presented here, with the CMAQ model which also attempts to quantify total N deposition by species. This may be a large undertaking and may be appropriate for a separate manuscript, but some overall comparisons here might be useful.

Response: We added in the text "The previously mentioned CMAQ simulation of Smith and Mueller (2010) simulates a NOy dry deposition flux of 1.9 Tg N a-1 in 2002 over the contiguous United States, with 70% contributed by HNO3 dry deposition (J. W. Mallard and S. F. Mueller, personal communication, 2012). Future work is needed to understand the differences between the two models."

Comment: There appears to be a minor error on pg 249 line 22. "CASTNET" should be replaced by "NADP/NTN and CAPMoN". Overall this is a clearly presented and well documented paper that can serve as a benchmark for future N deposition comparisons in the USA. I enthusiastically support its publication.

Response: The error on Page 249 is corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 241, 2012.