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Comment

***Interactive comment on “Modeling the distribution of ammonia across Europe including bi-directional surface-atmosphere exchange” by R. J. Wichink Kruit et al.***

**Anonymous Referee #1**

Received and published: 26 June 2012

This manuscript describes a simple parametrization of  $\text{NH}_3$  bidirectional exchange in the LOTOS-EUROS model and was reasonably well written. The model descriptions and results are likely to be of interest to many of the readers of Biogeosciences. I do have a couple of general reservations regarding the methods:

General comments

1. The assumption that the stomatal compensation point for  $\text{NH}_3$  was equivalent to the Henry's equilibrium dissociation assumes that the previous month's ambient concentrations were dependent on the stomatal compensation point alone. Any additional contribution to atmospheric ammonia from soil, animal or industrial sources would be

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inadvertently attributed to stomatal sources. Are the range of  $\Gamma$ 's used in this modeling study comparable to (Please present the range of  $\Gamma$ 's and indicate if these emissions are being double counted?)

2.The authors state that the compensation point accounts for some seasonality but then do not show any monthly or seasonal results to support this. Monthly observations apparently have a higher uncertainty, particularly in areas with a "low concentration range". However, the shift in the modeled seasonality with and without bidirectional exchange would be useful and the model could be compared to observations in "high concentration ranges" where there observational uncertainty is lower.

3.The manuscript describes the results in a generally qualitative manner and should be discussed more quantitatively, see the specific comments for details.

4.Both Figure and Fig are used to refer to the figures. Please follow the BG format.

#### Specific Comments

1.Abstract line 5: the acronyms DEPAC and LOTOS-EUROS should be defined

2.Abstract lines 13-14: Should "to a better extend" be "to a better extent"?

3.Page 4879 lines 2 -3: There have been a number of measurements of dry deposition including several in this issue. I think that this statement needs to be qualified as regional, e.g. country to continental scale, estimates of dry deposition are generally made using chemical transport models

4.Page 4880 line 23: What is intermediate complexity? I am sure that there are more descriptive ways of describing this model.

5.Page 4881 line 1: What exactly is "acceptable CPU time"? From my experience this could be anything from a few minutes in a windows environment to several hours of simulation time utilizing hundreds of CPU cores on a high performance computer cluster.

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6. Page 4884 Lines 3-4: References should be provided for "... based on many different studies of many different land use types".

7. Page 4884 line 5: "long-term concentrations" should be replaced with previous months concentration.

8. Page 4884 first paragraph: This parametrization appears to be at odds with micrometeorological and bioassay measurements of  $\Gamma$ s have shown a rapid decay in  $\Gamma$ s following fertilizer applications see Milford et al. 2001b. It seems like the parametrization presented here would miss these events and using monthly averages to parametrize the compensation point may smooth the temporal variability in the model concentrations.

9. Page 4885 Lines 7-9: How the "multiplication factor ..." was estimated needs to be described better.

10. Page 4885 Line 24: The grid spacing of the high resolution domain should be presented. I am assuming that the grid resolution of the high resolution domain was 0.25o longitude by 0.125o latitude.

11. Second paragraph of section 2.3: Were the emissions the same in both model simulations? If so, were NH<sub>3</sub> emissions from natural areas and agricultural crops estimated twice, once in the inventory and once in DEPAC3.11, in the bi-directional simulation?

12. Page 4887 lines 18-19: One of the strengths in the modeling of bidirectional NH<sub>3</sub> exchange would be a better representation of the seasonality by capturing temperature influences on the compensation point. Monthly time series figures should be shown to demonstrate how this model has changed the seasonality in the NH<sub>3</sub> concentrations even if the uncertainty in the observations is greater in the monthly values.

13. Page 4887 line 23: This equation as it is written would make the the observed concentration a function of the modeled concentration. I think something like  $\text{Cobs}(\text{Zobs})/\text{Cobs}(\text{Zmodel}) = (\text{Zobs}/\text{Zmodel})^{1/7}$  is what the authors meant. Addition-

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ally the wind power law assumes a zero slip boundary condition ( $U(Z=0)=0$ ). If this is applied to ammonia concentrations, e.g.  $C(Z=0)=0$ , it is clearly at odds with the parametrization of a compensation point where the concentration at the surface is non zero. How much did this interpolation change the results in the model evaluation? The authors may want to interpolate the modeled concentration gradient using the concentrations from layers aloft or the modeled compensation point to the observation height in the comparison.

14. Page 4889 Line 9: Please quantify "extremely low", e.g. on the order of 0.01 ppbv.

15. Page 4890 Lines 1-2: It is not clear from Figures 4 and 5 that "This (presumably bidirectional exchange) makes nitrogen input by wet deposition more important than the input by dry deposition" and this claim should be quantified in the text.

16. Page 4890 line 20: Is a few  $\mu\text{g m}^{-3}$  approximately 2  $\mu\text{g m}^{-3}$ ?

17. Page 4891 line 10: What qualifies DEPAC3.11 as "better"? A lower bias or error?

18. Page 4891 line 27: How much is "the bias in the regression" reduced?

19. Page 4891 line 29: Is a 0.06 drop in the  $r^2$  for 8 observations significant? Given the small number of observations, more robust metrics describing the model performance should be used.

20. Page 4892 line 1: Why is the modeled annual mean ammonia concentration at Eibergen not considered interpretable? Assuming that it is the point on the scatter plot with the largest increase, it appears from figure 8 that the modeled results at Eibergen were increased to be much closer to the observed value in DEPAC3.11.

21. Page 4892 line 10: I agree with the authors that the grid cell size does introduce error when comparing to observations taken at a point, but the biases presented here appear that on the same order of magnitude as the precision in which ammonia can be measured using passive samplers. How does this compare with the uncertainty in the  $\text{NH}_3$  observations using passive samplers?

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22. Page 4892 lines 27-28: How many observations are in this "cluster of coastal measurements"?

23. Page 4894 Lines 21-22: Was sulfate over the Mediterranean Sea in the DEPAC3.11 case fully neutralized and did this agree with measurements if they are available?

24. Section 4 would generally benefit from a better organization of the discussion and the conclusions should be discussed in a more quantitative manner when possible, e.g. "Altogether, the first order approximation ... to be quite successful". What is the definition of success in this case; a lower bias, the model runs with these changes, etc.?

25. Page 4897 lines 6-7: Is this "not feasible" or computationally expensive?

26. Page 4897 line 15: Please quantify "only slightly affected".

27. Page 4897 lines 25-27: You may want to expand on why you are comparing several CTMs in the ECLAIRE project and how that relates to this study.

28. Figures general: Specify the grid resolution in the figures

29. Figures 3-8, 10 & 13: The font used for the titles and axes labels in these figures are too small to be legible.

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**BGD**

9, C2123–C2127, 2012

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