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Interactive comment on “Advances in understanding, models and parameterisations of biosphere-atmosphere ammonia exchange” by C. R. Flechard et al.

Anonymous Referee #2

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Flechard and co-authors have assembled an excellent review of the state-of-the-science with respect to biosphere-atmosphere ammonia exchange. Additionally, the authors put forth recommendations regarding the improvement of process descriptions in chemical transport models, as well as requirements for observational datasets sufficient for more extensive evaluation of new models in terms of mechanistic detail and spatio-temporal scope. The subject and content of the manuscript are appropriate for Biogeosciences and, I believe, will be valuable to a wide audience of atmospheric and soil scientists, ecologists, and agronomists.

The sections of the manuscript are well organized, logically presented, and the level of detail within each section is sufficient for such a review. The manuscript is very well

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written. The completeness of the review is excellent with respect to the description of flux processes, inclusion of measurements and datasets that have been used for development and evaluation of the most widely used models and parameterizations, and inclusion of the most commonly used and mechanistically detailed models from the leaf to global scales. I have but a few suggestions below regarding the “synthesis and conclusions” that the authors may wish to consider. Subject to treatment of these relatively minor comments and suggestions, I believe the article to be suitable for publication.

1) The authors state in Section 1.4 that “The ultimate objective of this work is to integrate current knowledge into a common modelling framework adapted for local, regional, and global scale models, and to examine the degree to which measurement and input data are available, or missing, in order to parameterize, and ultimately run, surface/atmospheric exchange models at the different scales.”

I believe the authors essentially met this objective. With respect to integrating current knowledge into a common modeling framework, the authors summarize a list of realistic NH₃ exchange frameworks for CTMs in section 4.1. While I agree with this list, perhaps a schematic summary would help the reader better visualize how a model containing these improvements would differ from the current NH₃ frameworks used in CTMs.

2) With respect to the need for more flux measurements (Section 4.2), I think it should be mentioned that the temporal extent of the measurements should also be sufficient for the development of representative annual (semi-natural ecosystems) or growing season (crops) cumulative fluxes. Cost and logistical considerations make long-term deployments of micrometeorological NH₃ flux measurement platforms extremely difficult. However, while relatively short intensive studies are highly valuable for process-level investigation, development of robust flux parameterizations and datasets sufficient to understand net annual or growing season fluxes requires measurements over longer periods of time. Further development of lower cost flux measurement platforms will surely be beneficial in this regard.

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3) The authors provide a rather comprehensive list of ancillary data needed to properly interpret flux measurements and parameterize flux models. There are a couple of items within the list that are important enough to warrant additional detail. First, I strongly agree with A. Neftel's comments on this manuscript regarding the need to better understand NH_4^+ /soil interactions as related to quantification of soil emission potential (γ). As I and others have seen, Neftel points out that the magnitude of soil γ depends on the NH_4^+ extraction technique used. The literature on this point, as related to soil γ , is not yet mature enough to provide guidance on the most appropriate extraction method for the development of representative soil γ values. There is fundamental work to be done here, which I believe should be a high priority for further development of NH_3 emission algorithms for agricultural soils. Secondly, as regional and global NH_3 modeling progresses, soil chemistry data from long term ecological sites, agricultural experiment stations, soil surveys, etc will see more use. A better understanding of the relationships between extractable NH_4^+ and soil γ will be needed to properly apply these data.

4) Parameterization of dynamic leaf surface chemistry models currently relies on measurements of the bulk chemistry of relatively large droplets collected at night and early morning or after rain events. Further testing and development of these models is hindered by the fact that the chemistry of microscale cuticular water layers present on leaves and needles during the day cannot be measured. Thus, the extent to which existing bulk chemistry measurements may be used to parameterize models used to simulate conditions of much lower canopy surface water content during the day remains unknown. While such complex models may not be suitable for use within regional or global CTMs, with further improvement they may be very useful for understanding and improving simpler models which are more mechanistically representative than current empirical approaches for cuticular NH_3 exchange yet are computationally feasible for CTMs. Environmental microscopy, such as the methods described by Burkhardt et al. (2012), may represent a useful set of tools for improving our fundamental understanding of the chemical dynamics of leaf surface water during the transition from wet to

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dry conditions. In the absence of suitable techniques for field measurements, such laboratory techniques should be encouraged.

Burkhardt, J., Basi, S., Pariyar, S., Hunsche, M.(2012): Stomatal uptake of aqueous solutions – an update involving leaf surface particles. *New Phytologist*, 196, 774-787.

5) I fully agree with the author's point that there is a great need for more ground based measurements, using low-cost techniques, to aid in the evaluation of CTMs and ground-truthing of satellite data. While surface layer measurements are certainly the highest priority, there is also a need for characterization of vertical concentration profiles, particularly in agricultural areas. Aircraft measurements provide such information but are expensive. The extent to which low-cost measurement techniques could be deployed on tall towers should be investigated.

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