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Biogeosciences Discussions

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Interactive Comment

Interactive comment on "Modeling the dynamic chemical interactions of atmospheric ammonia and other trace gases with measured leaf surface wetness in a managed grassland canopy" *by* J. Burkhardt et al.

Anonymous Referee #3

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The paper presents an analysis of simulated and observed ammonia exchange fluxes over a grassland in Germany with a focus on the role of surface wetness and the chemical interactions occurring in the aqueous phase. Some of the observations are also applied to further develop the ammonia exchange model. Overall the paper is reasonable well written, addresses and interesting topic on the coupling between canopy chemistry and micrometeorology using observations to improve existing model representations and to validate those models also to identify some of their flaws. What would make the paper much stronger would be a definition of future research priorities based on the findings of presented analysis. The paper would be acceptable for publication in Biogeosciences properly addressing the number of issues/comments listed below.

One general comment; a lot of specific information is given on model assumptions, applied relationships with not always references to the sources of those assumptions, etc. The authors should critically check for statements that can be more specifically supported by including more references. One example is the statement at pp 2515; line 9; "These processes, however, are poorly understood and only coarsely parameterized, and for practical and numerical reasons, the exchange by default only takes place below a pH of 4.5, and only above a canopy equivalent water storage of 0.1 mm. One justification for the pH limitation lies in the fact that base cation leaching has been mainly observed as a passive defence mechanism against acid rain on leaf surfaces, limiting foliar injury;". In addition, a lot of background information is provided in companion papers which are not all yet available and if, in the near future, one would not continuously switch to do those other publications for cross-checking. It would therefore be useful to provide in some short statement some more information provided in those companion papers.

Specific comments:

Pp 2514, line 5-10; I am wondering about the importance of other compounds involved in the aqueous phase chemistry besides the listed ones, for example H2O2 with the SO2 oxidation also being affected by the cation availability. Most of the details about the aqueous phase chemistry are likely to be found in Flechard et al. (1999) but it would be useful to give some more details about what part of the chemistry is considered and which part is ignored and the underlying reasoning for this.

Pp 2514, lines 16-27; the discussion about numerical issues as a function of the leaf wetness is an interesting one with respect to a possible applicability of the here presented model in air quality/atmospheric chemistry/deposition model systems. It would be useful to indicate in the article, for example in the introduction, what the ultimate

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aim of the model development is since mentioned numerical issues might pose some limitations on the application of the model. One particular question that arises is how large the NH3 fluxes are for those conditions with little leaf water available. One can imagine that the fast drying of the leaves in morning would coincide with an enhanced turbulent mixing with large concentration gradients and consequently with large fluxes.

Pp 2515; "so that wet chemistry was calculated, except for the 2-layer run shown in Fig. 6." This statement is not that clear also because Figure 6 is not discussed yet. Do you mean that wet chemistry was only calculated except of one particular sensitivity analysis or except of a particular period "as will be discussed later in the Figure 6".

PP 2515; "In the present application, the normalized leaf wetness data obtained from clip measurements, with normalized values between 0 and 1 (see methods), provide the model input for leaf water storage, instead of the original energy balance approach by Flechard et al. (1999)". Also thinking about the application of the presented model in large scale model systems; It would be very interesting to see a comparison of the observed leaf wetness and the one calculated by the energy balance approach for this particular campaign. It shows what you can expect from these kind of approaches in terms of the simulation of the key constraint on all the detailed simulations of aqueous phase chemistry; leaf wetness. If we cannot get that parameter correctly simulated then introducing very detailed aqueous phase chemistry is not an option.

Pp 2516; explain the acronym BET

Pp 2516; line 15-17; Also having read the other comments and also initially questioning the differences between the Flechard et al. (1999), Nemitz et al (2001) and the model presented here, putting this statement already in the introduction would clarify a lot about the differences and similarities between these three models; "This resulted in a two-layer (foliage + litter) dynamic chemical canopy compensation point model (Fig. 1b), with the modeling of chemistry restricted to the living canopy foliage."

Pp 2517; the discussion about the in-canopy friction velocity is confusing. There is a statement that a logarithmic wind profile is assumed within the canopy whereas later one, re-reading the explanation about the various parameters and assumptions Equation 8 expresses that this logarithmic wind profile only reaches up about 70/5=14 cm. How sensitive are the flux calculations to the assumptions made on these key parameters determining Ra, Rb but also being essential through controlling the drying of the grass?

Pp 2518; change the sentence "The bioassay measurements within the measurement campaign.." to "The bioassay measurements;"

Pp 2519; line 1; Could you be more specific on what you mean with""No clear indications of stomatal activities could be derived from comparing wetness sensors clipped to leaves and filters, respectively." Are you referring to a contribution by stomatal opening and the release of H_2O?

Pp 2519; lines 24-25; It is stated that "This means that the influence of in-canopy turbulence on the vertical distribution of leaf wetness is neglected". Why is this done after the detailed discussion about the in-canopy turbulent regime (see previous comments) and the fact that, as demonstrated later on, that a soil/litter source of NH_3 is very important in determining the exchange fluxes? In order to properly consider the contribution by this litter flux to the net canopy exchange flux one could imagine that such subtleties in the vertical might be quite important.

Pp 2520; end of Section 3.2; what does these results express? Are these in line or different with what one would expect and what are the implications for the further model analysis?

Pp 2520; line 15; There is indeed a comparison of the energy balance calculated leaf wetness and the observed one as previously questioned. Because of this the statement that raised this previous comment should be changed to actually state explicitly that this comparison is done "as discussed in Section 3.2". Is there any explanation why the energy balance model results in a systematic underestimation?

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Pp 2520; "At the same time, a relative decrease of the measured fluxes occurred relative to this maximum deposition, which can be attributable to the drier conditions (Fig. 4b)." This sentence is confusing and propose to change it to: "At the same time, a relative decrease in the measured flux indicates that there is an decrease in the surface uptake efficiency which might be explained by the occurrence of relative drier conditions".

Pp 2521; Is there any explanation for the large discrepancies between simulated and observed pH's for the low water dew events?

Pp 2522; I miss the motivation why to switch from the 1-layer model including a litter emission flux to the two layer model application to the post-cut period. With the low canopy height, the possible limiting role of in-canopy turbulence and vertical differences in leaf wetness might be not that relevant and also well captured by the 1-layer model.

Pp 2523; line 13; "the overall agreement in the temporal variability is encouraging"

Line 18: what do you mean with "broadly simulated"? reasonable well? The model captures the order of magnitude of the flux? or?;

Pp 2524; "whereas the present study showed the opposite (Table 1), confirming the importance of deposited particles, together with ion exchange between leaf tissue and surface water through the cuticle". It would be useful to explain in more detail how deposited particles can effect the pH and can you confirm that the higher pH of the dew is indeed due to these particles? What is the experimental evidence?

Pp 2525; at the top you find one of the motivations why to use a 2-layer model version instead of the 1-layer version. This is what you could use to address the previous comment about the switch from the 1- to the 2-layer model. The main limitation in this particular part of the exchange process appears to be the description of the intermittent canopy exchanges, even for this grassland canopy.

Pp 2526; At the end of the discussion there is some indication about the overall sensitivity of the modelled ammonia fluxes to only the chemical parameters. Having read the analysis there is the perception that a large uncertainty is involved in the key micrometeorological drivers canopy turbulence and leaf wetness. From the here presented analysis it would be useful to indicate the priorities of further activities that would really help improving the development and use of such mechanistic exchanges models.

Interactive comment on Biogeosciences Discuss., 5, 2505, 2008.

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