Reply to Anonymous Referee #1

The paper compares two alternative models to simulate ammonia fluxes and their comparison with the measured fluxes in five peatland and grassland sites, focusing on the non-stomatal fluxes. The motivation for such more empirical deposition models is the inclusion of bi-directional ammonia fluxes into chemical transport models, which requires few and easily available parameters for the surface resistance. The improvement of such models is needed and the respective analysis here is valuable, specifically as it includes a suffiecient number of sites.

We thank the Reviewer for his/her helpful comments and criticism and for valuing our work. We have implemented most of the suggestions and think they led to a significant improvement of our manuscript. Please refer to the point-by-point response below for a detailed reply to your comments.

Focusing exclusively on nighttime fluxes with sufficiently turbulent condition is a good approach. It should, however, be discussed, if nocturnal transpiration could have confounded these observations.

The assumption of an "infinite" stomatal resistance at night is indeed a strong simplification that is not necessarily physiologically correct, and we should have acknowledged this in the original manuscript. The reason behind it is that it allows modelers to easily differentiate between the stomatal and the non-stomatal pathway, depending on which of the two is assumed to dominate the other one in magnitude of the fluxes. These assumptions allow a very simple inversion of the one-layer model framework to derive R_w (and R_s) from micrometeorological measurements, without having to explicitly model the other pathway. Consequences of this simplification are that R_w derived in such a way may indeed partially integrate stomatal fluxes as well, and therefore the physiological meaning of this variable may be confounded. We will add a discussion of this in the uncertainties sub-section of the results.

Changes to the manuscript: P10L16 (will be moved to "Sources of uncertainty" sub-section; cf. reply to Reviewer #2): Add paragraph: "Explicitly modeling the stomatal pathway with physiologically accurate stomatal conductance models may have the additional benefit of being able to assess bias in the estimation of non-stomatal resistances introduced by nighttime stomatal opening, naturally resulting in a lower contribution of the non-stomatal pathway to the total observed flux. However, note that a distinction between physiological accuracy and the purpose which the derived resistances are used for has to be made. While nighttime stomatal opening is a well-known phenomenon (e.g. Caird et al., 2007), it is rarely respected in modeling studies (e.g. Fisher et al., 2007). A physiologically accurate R_w parameterization used in conjunction with a stomatal model that does not account for nighttime stomatal opening would result in biased fluxes. We here derived R_w under the assumption that stomata are closed at night to ensure comparability with R_w values predicted by the WK and MNS parameterization, respectively, and compatibility with most operational biosphere-atmosphere exchange schemes, but we acknowledge that the physiological meaning may be confounded by stomatal flux contributions at night."; Add Caird et al. (2007) and Fisher et al. (2007) to list of references.

The parameters included in both models are temperature and, importantly, relative humidity (RH), whereas different ways are chosen regarding the fate of depositing ammonia, either unidirectional (MNS model) or 'quasi-bidirectional' (WK model). Based on common patterns of the five test sites, systematic under- and overestimation of fluxes are then diagnosed and empirical improvements are suggested by the authors. Although ultimately I agree with the overall direction of the paper and the interpretation of the results (see few exceptions below), I find it difficult to follow. One of the reasons is the continuous introduction of a multitude of parameters which makes consequent reading sometimes time-consuming and frustrating. Even in case it does not agree with the usual policy of this journal, I therefore suggest a table detailing all used parameters with units and possibly also other abbreviations.

We agree with the reviewer that constantly introducing new variables can interrupt the reader's flow. A list of symbols, on the other hand, can lead readers inexperienced with the modeling community's jargon to skip back and forth between pages when new variables appear. Neither variant is very elegant in terms of an uninterrupted reading experience, but we are convinced that we here chose the lesser of two (necessary) evils, and that the manuscript would not benefit from using a list of symbols instead. The majority of all constants and variables are defined 'in passing', i.e. we introduce their respective symbols and units in an unobtrusive manner alongside their first appearance in the text (e.g. P3L21: "R_c is further split into a stomatal pathway with the stomatal resistance R_s (s m^{-1}), and […] the non-stomatal resistance R_w (s m^{-1}) […]."). There are only two slightly larger blocks of variable introductions after an equation: P4L10-13 (4 lines) and P5L1-3 (3 lines). All further shortening of the manuscript would be due to removing variable definitions less than one line long and the omission of units in the text. On the other hand, a complete list of symbols would have more than 40 entries, and a shortened list of symbols (e.g. defining only R_x instead of $R_\mathrm{a}, R_\mathrm{b}, R_\mathrm{c}, R_\mathrm{s}, R_\mathrm{w}$) would not be enough to omit variable definitions throughout the manuscript.

Additionally, units may change depending on the circumstances a symbol is used. In the modeling community, (compensation point-) concentrations are often given in µg m^{-3} when they appear in figures, whereas some equations work on with concentrations in mol L^{-1} (a well-known example is the traditional formulation for the conversion from emission potentials to compensation points, as seen e.g. in Nemitz et al., 2000). Explicitly stating or repeating units close to the appearance of a symbol helps avoid confusion in these cases.

As a compromise and as an additional resource for readers unexperienced with the modeling community's jargon, we have added a list of symbols in the supplementary material.

Changes to the manuscript: P3L13: add "For a list of variables used throughout this article, the reader is referred to Tab. S1 in the supplement."; add Table S1 (List of symbols) to the supplementary material.

A second and somewhat related reason is the initial explanation of the two models which in some important places is not sufficiently detailed – some examples are given below. I wonder why the effect of RH is so little discussed in the paper - it has an exponential influence on Rw and thus is the most important independent parameter. It could e.g. be included in a analysis similar to Fig. 5.

We agree that RH is the most important parameter. However, we also think it has been sufficiently discussed elsewhere, and a simple exponential decay function does not necessarily need a visualization that goes beyond the relationship shown in Figure 2a. We will highlight the importance of the exponential decay function with minor changes to the text in some parts in the manuscript.

Changes to the manuscript: P2L18-19: Replace "This characteristic behavior is often modeled using relative humidity response functions as a proxy for canopy wetness (e.g. Sutton and Fowler, 1993; Erisman et al., 1994)." with "This characteristic behavior is typically modeled with an exponential relative humidity response function as a proxy for canopy wetness, where a high relative humidity results in low non-stomatal resistances and vice-versa (e.g. Sutton and Fowler, 1993; Erisman et al, 1994)."; P5L3: Add "The exponential decay parameter a was calculated as an average of a values per land-use class reported in the literature". P5L14: Add "exponential" between "simple" and "humidity".

I also wonder if the effect of backward-looking moving averages shouldn't be evaluated together with the RH history. Saturation effects (as mentioned in p.2, l. 20) could play a role at low RH.

Agreed, this is a good idea for further analyses. It is not trivial to find a good balance between a truly dynamic, but demanding (both numerically and in terms of required input-data) representation of the non-stomatal pathway, and a steady-state simplification that can be incorporated easily into existing schemes. We here tried to go the very first step that is more or less as simple as possible (while still respecting site-history in some way), and we found that this is not enough. We hope that this is a valuable piece of information for future analyses, which could (and should) indeed incorporate additional proxies, such as RH or precipitation history, but we deem this beyond the scope of our paper, where the moving-average approach was merely an additional idea about "what could work", even if it ultimately turned out to not work very well.

Changes to the manuscript: None (but added some explanation as suggested by Reviewer #2).

P. 5, l. 24/25: This is difficult to follow. Can it be supported by a formula? What happens if RH decreases?

We agree that it is not immediately obvious and tedious to show formally from Eqns. (7) and (8) that χ_{w} can only become a fraction of χ_{a}, and we hope a visualization of the solution space over plausible T and χ_{a} ranges (Figure 1 of this response) helps. Similar visualizations for possible Γ_{w} and χ_{w} values can be found in van Zanten et al. (2010, Appendix F, Figs. 17 and 18), which we will refer to in the revised manuscript. A decrease in RH has no direct effect on modeled χ_{w}, only on $R_{\mathrm{w,eff.}}$ due to the exponential decrease in the 'clean-air' R_{w} parameterization (Eq. (6)).

Changes to the manuscript: P5L25 add reference "(cf. van Zanten et al., 2010, Appendix F)." at the end of the sentence.

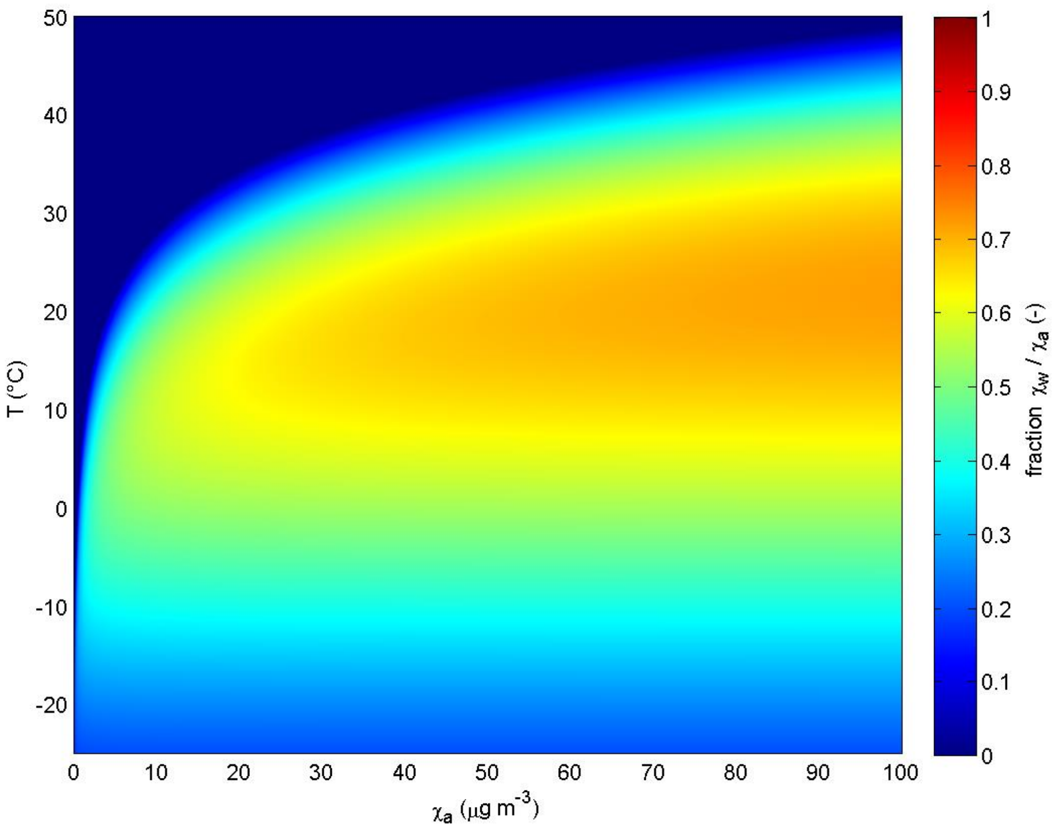

Figure 1: Fractions of the solutions for Eq. (7) of the original manuscript divided by χ_a over a range of plausible χ_a and T values.

P. 8, l. 19-23: This is indeed intriguing, but on the other hand I cannot really believe that MNS works so well in the prediction of fluxes at VK, when looking at the cumulative fluxes in Fig. 3. Even during the flat part, there is an underestimation of 0.3 kg ha-1. The shape of the cumulative fluxes at BM is considerably different from VK, while the shapes of ΔGw differences in Fig. 4 are very similar between BM and VK. Please check if the statement is really correct.

We did not state that MNS predicts the fluxes at VK well. "(…) relatively good predictive capabilities of MNS at BM and WK at VK (…)" (P8L21-22). We acknowledge that ΔG_w plots for the MNS model may indeed appear somewhat similar for BM and VK at first glance, but note that at VK we see a larger number of strong underestimations of ΔG_w compared to BM. The ratio of negative to positive values of ΔG_w for the MNS model is 1.2 at BM versus 2.8 at VK.

Changes to the manuscript: None.

P. 11, l. 19: which parameterization?

The MNS parameterization. Thanks for pointing out that this is unclear.

Changes to the manuscript: P11L19-20: Replace "(…) used in this parameterization." with "(…) used in the MNS parameterization.".

Figure 2: Why is Rw lowest at T=0°C?

Cf. P5L7-9: "Contrary to the original formulation of Flechard et al. (2010), Massad et al. (2010) do not use absolute values of $|T|$ (°C), but we chose to do so under the assumption that generally R_w increases in freezing conditions (e.g. Erisman and Wyers, 1993)."

We here chose to follow the original formulation of this temperature correction by Flechard et al. (2010), as it seemed physically more plausible to us that ammonia deposition to liquid water is larger than to ice.

Changes to the manuscript: None (already explained in P5L7-9).

Figure 4: Upper row: There seems to be a mismatch between the number of binned values used for MNS and WK comparison, at least for VK. What is the reason?

The number of binned values per bin is of course different, as it defines the shape of the histograms. The bin-width is equal (100 s m^{-1}) for all figures shown in the upper row. Note that we cut-off everything below or above a -1000 or 1000 s m^{-1} difference, respectively, for visual clarity and comparability of the subplots. So indeed the integral over the bars drawn in the figures does not necessarily reflect the total number of data points, if that is the question. The absolute differences can span multiple orders of magnitude, which we would not be able to visualize in a meaningful way. A logarithmic visualization would put more emphasis on the very large differences than necessary (e.g. there is not much difference between a 10^4 and a 10^5 s m^{-1} difference between modeled and measured resistances other than the fact that it is an extremely large mismatch – either modeled or measured fluxes will be close to zero in both of these cases).

Changes to the manuscript: None.

Minor issues P.6, l.11: 'approach', better: 'reach'? (also p. 9, l. 2) P. 6, l. 16: 'compensation point Xw decreases' P. 6, l. 23: why 'moderately'? I would suggest to omit this word P. 7, l. 18: event P. 10, l. 1: omit 'and'

We agree with all corrections, thank you for pointing them out.

Changes to the manuscript: P6L11, P9L2: Replace "approach" with "reach". P6L16: Replace "compensation χ_w point" with "compensation point χ_w". P6L22-23: Remove "moderately" in both hypotheses. P7L18: Replace "events" with "event". P10L1: Remove second "and" in the line.

References (both replies)

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