

## Reply to Anonymous Referee #2

### General comments

The paper is within the scope of ACP. The results are correctly presented; the figures illustrate the results in a clear way, but the order should be changed, as detailed in the specific comments. The paper is written in good English. I recommend this paper to be published in ACP, after some major corrections and improvements in the presentation of results. My main remarks concern principally the way results are presented. In my opinion the results could be presented in a more positive way. The reader cannot be convinced if the authors present their results without highlighting the advantages found after the sensitivity tests. This is detailed in the specific comments.

Thank you for your insightful review. Please refer to the specific comments for a detailed answer to each of your concerns.

Some bibliography about how these two models have been used until this study would have been necessary to help the reader understand where the authors want to go and why they have chosen these particular models and not others. Did these models give satisfying results in other studies and why did the authors choose them.

Thank you for highlighting that we missed to give a justification for the choice of the parameterizations. This has been added to the introduction section. There are indeed notable other models, such as the one of Zhang et al. (2010, 2003). The choice of the two particular models compared in this study is based on a number of different reasons: a) they are structurally very similar (the WK parameterization is flexible in terms of its usage within a one- or two-layer model); whereas Massad et al. (2010) and Zhang et al. (2010) exhibit some fundamental differences in their handling of the ground-layer pathway (this is not discussed in particular in this study, but it leads to difficulties in programming comparable model codes); b) the motivation for this study arose from prior experience of the lead author with the MNS and WK parameterizations; and c) the WK parameterization features a unique handling of the non-stomatal pathway with its ‘quasi-bidirectionality’, and we found it interesting to see how it compares to the traditional deposition-only approach.

**Changes to the manuscript:** P2L31: Add “The Massad et al. (2010) parameterization has received widespread acceptance in the community, with 53 citations according to the literature database ‘Thomson Reuters Web of Science’ at the time of writing this article, and variants of it have been applied in numerous studies, e.g. recently in Shen et al. (2016), Möring et al. (2016), Zöll et al. (2016), and others. Wichink Kruit et al. (2010) followed a unique approach by simplifying complex dynamic approaches towards an empirical steady-state formulation of a non-stomatal compensation point model, which is nowadays used within the DEPAC3.11 deposition module (van Zanten et al., 2010) and the chemistry transport model LOTOS-EUROS (Wichink Kruit et al., 2012), and it is structurally compatible with the Massad et al. (2010) model.”; Add references for Shen et al. (2016), Möring et al. (2016), Wichink Kruit et al. (2012).

Partial conclusions at the end of each paragraph need to be more clearly assessed. The go home message needs a clearer explanation.

We agree, thank you for pointing out that the conclusions are not clear enough. Please refer to our answers to specific remarks below.

**Changes to the manuscript:** See below.

### Specific comments

Abstract. The abstract gives a clear idea of what is presented in the paper. The sentence line 25 page 1 “The proposed  $\Gamma_w$  parameterization...” needs to be detailed to let the reader know in what way it can be improved.

Agreed, this will be more detailed in the revised manuscript.

**Changes to the manuscript:** P1L25-26: Replace “The proposed  $\Gamma_w$  parameterization appears to have potential for improvement, but cannot be recommended for use in large scale simulations in its present state due to large uncertainties.” with “The proposed  $\Gamma_w$  parameterization revealed a clear functional relationship between backward-looking moving averages of air  $\text{NH}_3$  concentrations and non-stomatal emission potentials, but further reduction of uncertainty is needed for it to be useful across different sites.”

Page 3 line 13: could you explain how it is realistic or not to switch off the soil/leaf litter layer for natural ecosystems, where it can be an important source of  $\text{NH}_3$ , such as mentioned for example in Wentworth et al., 2014.

Thank you for hinting at Wentworth et al. (2014). We agree with the reviewer about the importance of soil-based emissions.

The decision to use a single-layer framework for unmanaged ecosystems by Massad et al. (2010) was less based on natural conditions, but primarily on data availability of  $\Gamma_g$  for unmanaged ecosystems at the time, as well as methodological issues: Most of the compensation point measurements that they used were based on micrometeorological measurements. The flux measurements used to derive major parts of the parameterization were representative for the ecosystem scale, and the attribution to different conceptual compartments (stomata, cuticula, ground-layer) had to be made based on inverting the resistance model for different environmental conditions (humidity, radiation/time of day). However, this can only be done easily in a single-layer framework due to the relatively straightforward differentiation between stomatal and non-stomatal contributions to measured fluxes (note that we have added discussion on nocturnal stomatal fluxes as suggested by Anonymous Referee #1). Adding a third (ground-layer) pathway severely complicates this approach unless the ground-layer based signal completely dominates the observed fluxes. Ground-layer based emissions are not being ignored, but rather integrated into the stomatal emission potential for unmanaged ecosystems in the MNS parameterization. The model switches to a two-layer formulation after management events, when the contribution of these emissions is strong enough to be more or less unambiguously attributed to the soil. We agree that this is conceptually unsatisfying and should be improved upon in future developments.

Note that this is very similar to our reasoning why we chose to call  $R_w$  “non-stomatal” instead of the often used “cuticular” or “external leaf surface” resistance – we cannot be 100 % certain that we do not integrate the influence of other surfaces when we simply invert the model for nighttime conditions (in fact, we most likely do).

**Changes to the manuscript:** See below (Answer to P10L24-25).

Page 5 line 15: please give the  $\text{NH}_3$  concentration under which clean conditions are considered.

This goes back to Milford et al. (2001), who concluded that Eq. (6) with a minimum  $R_w$  parameter of 2 and an exponential decay constant of 1/12 is valid for conditions without  $\text{NH}_3$  saturation at the leaf cuticles (although the term ‘clean conditions’ was introduced by Wichink Kruit et al. (2010)).

**Changes to the manuscript: P5L15:** Replace “In this model, a simple humidity response after Sutton and Fowler (1993) is used as an approximation for  $R_w$  under ‘clean conditions’.” with “In this model, a simple humidity response after Sutton and Fowler (1993) is used as an approximation for  $R_w$  under low ambient  $\text{NH}_3$  concentrations, where saturation of the external leaf surfaces is unlikely (Wichink Kruit et al., 2010; Milford et al., 2001):”. Add Milford et al. (2001) to the references.

Page 5 line 24: this term of “pollution climate” is difficult to understand because it is not precise enough. Do you mean “air pollution climate” as mentioned in Wichink Kruit et al. 2007? Is there a value for  $\text{NH}_3$  concentration to define this threshold of pollution?

Yes, we will change this to “air pollution climate”. There is no threshold for “pollution” here, rather the (admittedly somewhat vague) term is often used throughout the literature to describe the composition of ambient air in terms of different airborne pollutants.

**Changes to the manuscript:** P4L21, P5L24, P12L17, P12L22: Add “air” before “pollution climate(s)”.

Page 7 line 24 add “with” between “conjunction” and “leaf”.

Thanks, corrected.

**Changes to the manuscript:** P7L24: Add “with” before “leaf”.

Page 9. The “results and discussion” paragraph needs to be restructured. Uncertainties should be discussed in a specific sub-paragraph. It would be interesting to specify the conditions where these models have been applied, how successful they were, and where they cannot be applied, for example when emission occur instead of deposition.

We agree with the reviewer and have added a sub-paragraph in which we discuss the uncertainties of our methods. Regarding the applicability of the models, please refer to the answer to one comment above (about why we chose these particular parameterizations).

**Changes to the manuscript:** Move P9L22-P10L26 and related discussion added during the revision process into new sub-section 3.5 “Sources of uncertainty” at the end of the “Results” section.

Page 10 line 1: remove “and” at the end of the line.

Done, thanks.

**Changes to the manuscript:** P10L1: Remove second “and” in the line.

Page 10 line 10: “a model”: what model exactly are you talking about?

No specific model; N-Input could be derived from any kind of model that is able to predict net annual reactive nitrogen deposition.

**Changes to the manuscript:** See below.

Page 10 lines 10 to 15: This explanation is not clear. These lines have to be rewritten. Line 11, after the sentence “we do not feel confident...”, is it supposing that only NH<sub>3</sub> dry deposition is available? Line 14-15: “A detailed description...” if the investigation is beyond the scope of the paper why then talking about it and give the results of the sensitivity test if you do not give the reasons of why it could not work? Some ideas could be provided to help the reader understand.

Apologies for being unclear here. L11: Yes. L14-15: This statement was primarily given as a justification for why we did not incorporate daytime data by modeling the stomatal pathway, although the flux measurements are less prone to error due to better turbulent mixing during daytime. We believe that this could be a good idea for further studies, where input data for mechanistically satisfying (e.g. photosynthesis-based) models for  $R_s$  and reliable estimates of the stomatal emission potential (e.g. via bioassays) are available.

**Changes to the manuscript:** Rewrite P10L10-15 from: “While this can be estimated from spinning up a model that incorporates more reactive nitrogen species than just NH<sub>3</sub>, we do not feel confident estimating total N input from modeled NH<sub>3</sub> dry deposition alone. At sites where total N input is known (e.g. BM, from Hurkuck et al. (2014), or from CTM results), the MNS and WK parameterizations predict such different  $\Gamma_s$  estimates that one model predicts net emission from the stomata and one model predicts a net uptake over the course of the measurement campaign. A detailed investigation on the reasons for this mismatch is, however, beyond the scope of this paper.”

to: “While this issue can be overcome by iteratively solving a model with more reactive nitrogen species, so that N input is both a parameter, and a result of the simulation, we here used a model that only predicts NH<sub>3</sub> dry deposition, which we do not consider to provide sufficient information to estimate total N input to our sites. At sites where total N input is known (e.g. BM, from Hurkuck et al. (2014), or from CTM results for other sites), the MNS and WK parameterizations both predict very different  $\Gamma_s$  estimates. The reasons for this mismatch have, to our knowledge, not been investigated to date. We therefore decided to not model the stomatal pathway explicitly and rely on nighttime fluxes only.”

Page 10 line 24-25: What do you mean by “very well”? Do you mean that the assumption of ground layer resistance = infinite is not realistic? And what about weak ground resistance and infinite stomatal resistance? The authors should give some more explanations and overall extract the main positive idea of such sensitivity tests explained in this paragraph. The reader is a bit frustrated not to know if good ideas have to be extracted from that.

$R_g = \infty$  is more of a technical solution to transform the model into a one-layer model in cases where ground-layer fluxes could not clearly be differentiated from other pathways when parameterizing the model on micrometeorological measurements, not necessarily based on whether or not it is realistic. In principle, it would of course be more realistic to model the ground-layer pathway for all land-use classes, even if weak ground-layer emissions are recaptured by the canopy (modeled via  $\chi_c$ ) but according to Massad et al. (2010) not enough reliable data on ground-layer fluxes and emission potentials were available during the development of the parameterization. Note that one also has to be careful when mixing e.g.  $R_w$  parameterizations based on micrometeorological measurements via inversion of a one-layer model together with measurements of  $[\text{NH}_4^+]/[\text{H}^+]$  in the soil solution as an estimate for  $\Gamma_g$ , as the former will already include a contribution of the ground-layer when emission fluxes are large enough to not be completely recaptured by the canopy. Please also refer to our answer to P3L13.

**Changes to the manuscript:** P10L20-21: Replace “(...) the ground layer resistance was set to infinity (Massad et al., 2010) and the model reduces to a one-layer model.” with “the ground layer resistance was set to infinity in order to transform the model structure to that of a one-layer model (Massad et al., 2010).

Page 10 line 28: The reader cannot understand the ideas mentioned in this 3.2 paragraph if the authors do not explain in what purpose they use moving averages of NH<sub>3</sub> concentrations. What is the goal of this exercise?

We respectfully refer the reader to section 2.5 (starting on P7L22) of the manuscript, which we will slightly expand in the revised version of the manuscript. However, we would also like to use this discussion forum as a platform to elaborate on them in some more detail:

Most non-stomatal resistance parameterizations found in the literature are steady-state approximations of processes that we know to be dynamic by nature (e.g. Wentworth et al., 2016; Jones et al., 2007a, 2007b; and many others), i.e. they are solved for every moment in time individually, although we are aware that we should keep the history of the site in mind, especially with respect to previous nitrogen deposition and the wetness of the canopy. In the late nineties, Sutton et al. (1998) and Flechard et al. (1999) developed bidirectional cuticular desorption models that model the non-stomatal pathway as a charged capacitor, and they have been successful at modeling e.g. emission events after dewfall at night and subsequent drying of the canopy in the early morning hours. However, it turned out to be difficult to parameterize these models with measurements, e.g. surface pH, or concentration measurements of a number of different atmospheric constituents were needed. Additionally, as these models were naturally also dynamic in a numerical sense, i.e. dependent on the previous state of the

system (sometimes with very small time-steps needed for the numerical solution), they had the disadvantage of being computationally expensive, which limited their applicability in spatially explicit transport models. Wichink Kruit et al. (2010) presented an important step towards a balance between mechanistically realistic and computationally efficient models. They tried to simplify the bidirectional parameterization for external leaf surfaces by developing an external leaf surface compensation point model that was dependent on atmospheric ammonia concentrations, thus being capable of modeling saturation effects. While this approach was technically not really bidirectional, due to the fact that the best fit to the data was achieved with an expression which always yields an external leaf surface compensation point that is smaller than the ambient concentration, it led to an improvement in the way that we could now get good estimates for long-term net  $\text{NH}_3$  deposition “for the right reasons”, i.e. because sometimes there is a significant non-zero external leaf surface emission potential. However, this model only incorporated information about the current state of the system, not about the magnitude of previous deposition events or previous ambient  $\text{NH}_3$  levels. In our manuscript, we tried to find a compromise between these two approaches by fundamentally following a similar approach to Wichink Kruit et al. (2010), but making it dependent on the past. We decided to use  $\text{NH}_3$  concentration instead of (modeled) fluxes, as it is an easily accessible variable that is directly being measured instead of modeled and therefore available before any model calculations, and a direct driver of  $\text{NH}_3$  saturation at humid surfaces. Reviewer #1 correctly pointed out that a logical next step would be to incorporate the “wetness history” of the site into such analyses, e.g. the average relative humidity of the previous day(s), or the days since the last rainfall. We here only presented the very first step towards a conceptually dynamic, but structurally static model of external leaf surface exchange, and while our results are not directly useful for modeling purposes, we believe they are a promising first step for the future treatment of the bidirectional non-stomatal pathway.

**Changes to the manuscript:** P7L25: Before “We here...” Add: “While it is capable of modeling saturation effects with an ambient ammonia concentration dependent non-stomatal compensation point, it only relies on  $\chi_a$  at the current calculation step. A compromise between the truly dynamic models of Sutton et al. (1998) and Flechard et al. (1999) and the steady-state simplification of Wichink Kruit et al. (2010) would respect the site’s history of reactive nitrogen inputs without falling back to a numerically dynamic model and, consequently, the same difficulties that limit the application of existing dynamic approaches in large-scale models, i.e. it would need to use a proxy for previous nitrogen deposition without relying on the model’s flux predictions at an earlier calculation time. ”

Page 11 line 2: [why this case is not shown? It would have been interesting to see the results?](#)

We opted for a linear horizontal axis in Figure 6 as the majority of the concentration data are in the sub- $20 \mu\text{g m}^{-3}$  region and the fact that there is a functional relationship is still obvious. Additionally, a linear least-squares fit to a log-transformed variable implicitly assumes a multiplicative error-model, the validity of which is unclear in this particular case. Mentioning it in P11L2 was primarily meant to give the reader an idea of the relationship, but there are probably more appropriate statistical models to show this.

The linear fit to the log-log-transformed data is shown in Figure 1 of this response.

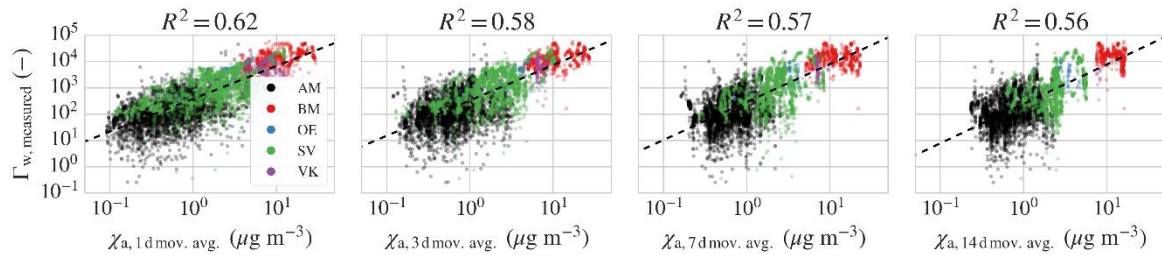


Figure 1: Log-log-relationship between moving averages of air ammonia concentrations with different moving windows and non-stomatal emission potentials derived from flux measurements at five sites.

**Changes to the manuscript:** None.

Page 11 line 9 :The authors give indications of potential improvements and conclude by writing that no improvement is deduced. What is then the purpose of giving these results if they do not lead to improvement? It should be far better to highlight the advantages instead of giving the disadvantages.

This is probably more of a philosophical issue. The reviewer is correct in stating that the results of this particular attempt are not immediately useful. However, we believe that it is important to avoid publication bias by not exclusively showing polished up positive results, but also by publishing what would be considered ‘negative’ or ‘non-constructive results’. Parameterizing  $\Gamma_w$  on some proxy for site history is a logical step in reducing degrees of freedom of more complex, mechanistic models while still being conceptually (albeit not structurally/numerically) dynamic (see answer above). We tested one variant of doing so and arrived at the conclusion that this particular variant is probably not the final answer. By publishing these results regardless, we encourage looking at different approaches and avoid that other researchers unnecessarily try the same, only to arrive themselves at a ‘negative’, i.e. non-productive finding that they likely won’t publish either. We feel that the manuscript at hand can stand on its own feet without this analysis, and if the reviewer and the editor agree that this section should be omitted from the manuscript, we are happy to do so, but we believe it adds some valuable information and gives the reader ideas on how (or how not) to improve  $\Gamma_w$  parameterizations in the future.

**Changes to the manuscript:** None.

Page 11 line 20: same remark as above. The way this paragraph is written does not give a positive issue. The authors should turn it differently to highlight the positive points. This part should follow figure 3.

We have re-phrased the partial conclusion of this paragraph to appear more positive. However, we do not agree that this part should follow Fig. 3, as the motivation to reduce the parameter values of  $R_{w,\min}$  and  $\beta$  follows from the left-hand panel of Fig. 5 (see answer below).

**Changes to the manuscript:** P11L17-20: Replace “While there does not appear to be a comprehensive, generic solution, we assume that there is potential for a significant overall improvement by optimizing these two parameters based on independent data from all four ecosystem types (grassland, arable, forest and semi-natural ecosystems) used in this parameterization.” with “This exercise highlights the potential for a significant overall improvement in  $\text{NH}_3$  flux predictions by optimizing these two parameters based on independent data from all four ecosystem types (grassland, arable, forest and semi-natural ecosystems) used in the MNS parameterization.”

Page 12 line 1: What do you mean by “the impact of this study’s main findings are negligible”?

We believe the reviewer might have misread this line (the manuscript says “on” instead of “of”), but we will rephrase this sentence for clarity.

**Changes to the manuscript:** Replace “(...) the impact on this study’s main findings are negligible.” with “(...) they do not lead to significant differences in the main findings of this study.”

Page 12 line 12: Again what is the advantage of doing this if no solution is going out?

See answer to P11L9 given above.

**Changes to the manuscript:** None.

Page 12 line 13: the title is not appropriated. Should be “conclusions”.

Agreed.

**Changes to the manuscript:** P12L12: Replace “Conclusions and recommendations for further research” with “Conclusions”.

Page 12 line 17: “pollution climate” is not an understandable term. Conclusion needs to be more striking.

Regarding the term “pollution climate”, see our reply to your comment on P5L24. We have reformulated the second sentence to be more to the point.

**Changes to the manuscript:** P12L17-21: Replace “We tested the potential for an easily accessible improvement of predicted  $R_w$  and consequently predicted  $\text{NH}_3$  exchange fluxes by using smaller values for the temperature response and minimum  $R_w$  parameters and propose to further investigate this route using data from all four ecosystem types represented in the MNS  $R_w$  parameterization” with “Adjusting the temperature response and minimum  $R_w$  parameters in the MNS model towards smaller values resulted in a better match between modeled and measured  $\text{NH}_3$  fluxes at most, but not all sites. We suggest to further investigate the potential of re-calibrating these parameters to flux data from all four ecosystem types represented in the MNS  $R_w$  parameterization. Compared to measured values found in the literature (e.g. Massad et al., 2010, Tab. 1), especially the minimum predicted  $R_w$  at sites with low atmospheric acid-to-ammonia ratios appear too high.”

Page 12 line 27: “We strongly encourage” is not appropriate. Please reformulate.

Agreed, this is probably too subjective.

**Changes to the manuscript:** P12L27: Replace “strongly encourage” with “suggest”.

Changes in the structure are needed. Figure 7 should follow figure 3, figure 8 should follow figure 4. Please adapt the text in function of these figure changes.

We see where the reviewer is coming from, as these figures appear to be very similar visually. However, we respectfully disagree with this suggestion for the following reasons:

Figure 3 and Figure 7 are only similar in the form of the visualization. Figure 3 is a comparison between the two models’ predicted fluxes in their original parameterization and marks the first step of our analysis. Figure 7, on the other hand, is the result of changing two parameters in a way that was suggested by the results. It answers a “what if” question that would not have been asked before seeing the left panel of Figure 5; it is not part of the “core” analysis of our manuscript, but more of an outlook, or a suggestion for what parameters to look at in the future. It would therefore not be logical to show it earlier.

Figure 8 is a sensitivity study with the aim to show the influence of some of our decisions and to assess “researcher’s bias” introduced by making these choices. This was actually an Appendix in early versions of the manuscript, but since the paper itself is fairly short, we decided to move it to the results section instead. We are happy to move it back to an Appendix section if needed, but we don’t think it should be shown earlier, as it is not a fundamental part of the analysis, but rather an addition.

**Changes to the manuscript:** None.

#### Technical corrections

Page 6 line 10 and line 26, *ibid* and *i.e.* have to be in italics. Throughout the text latin expressions should be in italics.

“Common Latin phrases are not italicized (for example, *et al.*, *cf.*, *e.g.*, *a priori*, *in situ*, *bremstrahlung*, and *eigenvalue*).”

From: [www.atmospheric-chemistry-and-physics.net/for\\_authors/manuscript\\_preparation.html](http://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html)

**Changes to the manuscript:** None.



## References (both replies)

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