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Biogeosciences Discussions

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 #2

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General comments: This paper is about modeling the exchange process of atmospheric ammonia with leaf surface wetness in a managed grassland canopy. It describes measurements of ammonia fluxes and apoplastic NH4+ and H+ concentrations. These data were used to apply, validate and further develop an existing model of leaf surface chemistry and ammonia exchange. The first modeling approach was a single layer canopy compensation point model with dynamic cuticular and stomatal exchange. The second model approach includes a second leaf litter layer. A major omission in this paper is a direct comparison between the two-layer canopy compen-

sation point model of Nemitz et al. (2001) without the dynamic exchange to/from the cuticle (Figure 1a) with the new two-layer canopy compensation point model with the dynamic exchange to/from the cuticle (Figure 1b). Instead, the authors show a comparison between the original single layer dynamic canopy compensation point model (Flechard et al., 1999) and the new two-layer dynamic compensation point model (Figure 1b). This is not a fair comparison as the new model should be better than the 'old' two-layer canopy compensation point model of Nemitz (2001). To make an objective judgment only these two models should be compared. Besides, the authors give a lot of attention to leaf surface wetness measurements, which instinctively might be very important. However, this should first be shown in the comparison above. Then, a second comparison could be made between the new model (Figure 1b) with VH20 from direct leaf wetness measurements and the new model with VH2O from the energy balance method. I therefore only recommend publication if the authors are able to make the suggested objective comparisons above and quantify the comparisons properly. The paper needs a thoroughly revision in which the following recommendation should also be included.

General comment about the figures: Use equal scales and (if useful) the same variables in figures that can/should be compared with each other (e.g. figure 4/7/9/10). I would also prefer to see the measured fluxes in black.

Specific comments: Title: in my opinion the title of this paper is not clear. The paper only considers the exchange of ammonia and no other trace gases. Furthermore, the word 'measured' is unnecessary. I would suggest: "Modeling the dynamic chemical interactions of atmospheric ammonia with leaf surface wetness in a managed grassland canopy."

Abstract: Reconsider the paragraphs in the abstract. What is the main message? Use the 2-layer canopy compensation point model with improved dynamic cuticular and stomatal exchange? Or/and: by measuring leaf surface wetness and using a model to derive leaf surface water storage from it, we can improve the dynamic two-layer

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compensation point model?

Methods: The methods section is described too extensive and needs to be shortened drastically.

p 2507, l 8-10: Explain why the leaf surface water storage is so important in the model (or remove it from abstract).

p 2507, l 17: unrealistically? What is unrealistically in these situations?

p 2507, l 18: significant improvements: quantify!

p 2508, l 2: What do you mean with 'signals'?

p 2508, l 13: Add (Wichink Kruit et al., 2008) Wichink Kruit, R.J., Jacobs, A.F.G., Holtslag, A.A.M., 2008. Measurements and estimates of leaf wetness over agricultural grassland for dry deposition modeling of trace gases. Atmospheric Environment 42, 5304-5316.

p 2508, l 24: 'in part' should be replaced by 'partly'

p 2508, l 29: 'is expected to have been' is not correct. Replace by 'is expected to be' or 'has been'.

p 2509, l 2: references to accompanying papers were not all available at the time of this review

p 2510, l 4: ', simulating' should be replaced by ' to simulate'.

p 2511, l 12: How 'rigorous' is the quality control? What kind of filtering was used? How was the consensus reached (seems to be quite subjective)? What is the accuracy of the estimated fluxes? Maybe this is beyond this paper, but it should be treated carefully!

p 2511, l 16: 'were conducted' should be moved to line 15 between 'measurements' and 'to'.

p 2511, l 19: which 'acid gases'?

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p 2511, l 22-23: there is a contradiction between 'clear, calm nights' and 'surface water after rain'

p 2512, l 18-19: in the rest of the paper ranges of heights are used (e.g. 0-15 cm, 15-30 cm and 30-45 cm)

p 2512, l 27: remove 'to'

p 2512, l 27-30: in my opinion it is indeed possible to normalize the leaf wetness signal, but the normalized leaf wetness signal does not say anything about the amount of leaf surface water. It still contains the unwanted instrumental factors in the end. Besides, the authors already state in line 15-17 (page 2512), that the leaf wetness signal is also affected by stomatal aperture, environmental humidity and the ion concentration in surface moisture. A larger signal can be caused by a higher ion concentration in the surface moisture.

p 2513, l 3 and 6-7: Explain why an exponential function was adapted and not a linear?

p 2513, l 12: remove ','

p 2513, l 19: add ')' after 20085

p 2513, l 24: Figure 1a is the model of Nemitz et al. (2001), while the authors refer to the model of Flechard et al. (1999) in the text. This is very confusing.

p 2514, l 5-27: This section is not transparent. How are variables (although coarsely) parameterized? Is Fcut (see Flechard et al., 1999) included? Neirynck et al. (2008) gives a transparent and comprehensive overview of the dynamic canopy compensation point model. Is this the same model as applied here? I think it is, but their description of the model is clear. This one isn't. Maybe just refer to this paper and refrain from the whole extensive description in this manuscript. Neirynck, J., R. Ceulemans, 2008. Bidirectional ammonia exchange above a mixed coniferous forest. Environmental Pollution xx, 1-15. doi: 10.1016/j.envpol.2007.11.030 (article in press)

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p 2515, l 7; 'almost always more or less' = weak statement

p 2515, l 11-12: remove 'with normalized values between 0 and 1 (see methods),' (is already mentioned in line 11.

p 2515, l 9-28: The normalized leaf wetness signal might not only be representative for the amount of water on the leaves, but also for the amount of ions in the surface moisture (and other environmental variables) as the authors already state on page 2512 (line 15-17). Therefore this signal might not just be normalized to reflect the effective water film thickness.

p 2515, l 20: Where does the limit LW=0.15 come from?

p 2515, l 23-25: This statement is not correct. The observed relationship between 30-45 cm is LW=1.40*10-5 exp(11.2*rh). LW=0.02 corresponds to a rh of 65%! This changes equation 1.

p 2516, l 4: what is a BET isotherm?

p 2516, l 12: The water holding capacity is different for different plant species due to differences in wettability (Flechard et al., 1999). Mention here.

p 2516, l 24-26 and p 2517: All variables are already described by Nemitz et al. (2001). Just refer to it.

p 2518, l 3-11: This section is important and should stay in the manuscript.

p 2518, l 4: 'at' should be 'of'

p 2518, l 18: 'no further rain' is not correct. Figure 2a shows that there is some rain on 29 May. The labels of the x-axis should include a date and not only the time (GMT). They are confusing. Why is the measured rh so low during the whole measurement period? Was there any problem with the rh sensor? How was rh measured?

p 2519, l 1-3: Why are the results of the clip sensors on filters shown? They are not

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used in the analysis or model calculations.

p 2519, l 13-15: This is not so obvious to me. Explain.

p 2519, l 22-26: Why are leaf wetness measurements at all three layers shown, while they are later combined again.

p 2519, l 28-29 and p 2520, l 1-9: This section needs an introduction and more explanation. Why are the concentrations in dew measurements higher than in bulk rain? Explain.

p 2520, l 14: I would suggest to call the dynamic energy balance model "energy balance VH20" instead of "dynamic VH20" in the figures. Both the empirical and the energy balance model are dynamic models, so, I would suggest to leave the word "dynamic" or to use it consistently.

p 2520, l 17: 'two' should be 'three'

p 2520, l 18: The authors state that there is strong dewfall on 23 May. However, figure 2 shows that this is just rain on 22 May that does not dry up anymore.

P 2520, l 19: How can the calculated 'empirical VH2O' be higher than 0.1mm (following eq.(2))?

p 2520, l 21: I argue this statement. There is only a very short period on 26 May when the model calculates emission. The rest of the time, the model calculates deposition, while the measurements more often show emission. The magnitudes of the modeled and measured fluxes, however, are comparable.

p 2520, l 23: "26 May" should be "25 May"

p 2520, l 25-29: The aerodynamic maximum possible flux is very confusing here. It will only be realistic in cases were the surface concentration is zero and we know that this is not the case at this site. Besides, it can be seen in figure 4d that the concentration in the air is much higher in the second period. The maximum flux is Vmax * Cair. So the

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statement that a higher Fmax is caused by higher wind speeds is incorrect! It is more likely caused by the higher air concentrations. If the authors want to show that the air is more turbulent during the second period, they should just plot Vmax. However, I would suggest to leave this section and to remove Fmax from figure 4b as it gives no extra information.

p 2521, l 5-11: It seems to be a bit strange that not every drying period (from high to low leaf wetness) leads to a positive Fd (compare figure 4a with 4c). Especially, on 28 May, it is strange to see that although the wetness is decreasing (there should be a release of ammonia from the cuticles) and measured fluxes become positive, the Fd is negative. This is kind of strange to me. Please explain.

p 2521, l 21: VH20 higher than 0.1 mm? see comment on P 2520, l 19

p 2521, l 28: "unrealistically".....Milford et al. (2001) found gamma values of 6000 after grass cut using the simple canopy compensation point model (without the soil pathway). I agree that this is also an assumed value to fit the model to the measurements, but it is not so unrealistic as it might seem from bioassay point of view. It just has to do with the modeling approach (or the lack of the soil pathway). For a correct physical behavior of the model, you indeed need to parameterize all the processes (and resistances) determining the flux and I think we are more and more able to do so (as this paper also shows). However, it is extremely difficult to parameterize and verify all these different processes as they are not measurable independently. C. Milford, M.R. Theobald, E. Nemitz and M.A. Sutton, 2001. Dynamics of ammonia exchange in response to cutting and fertilising in an intensively-managed grassland. Water, Air and Soil Pollution: Focus 1: 167-176.

p 2522, l 13: replace 'soil temperature' by 'ground surface temperature'

p 2522, l 7-22: Figure 6c shows a very strange behavior of Ra, Rac and Rb. They are kept constant during nighttime, while they normally become very large. Explain. Furthermore, it is strange that when the leaves are dry (for example on 2 June), Fd is

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very negative (absorbing ammonia). Explain.

p 2522, l 6(?): The agreement between the two-layer model and the measurements is indeed good. How is this agreement compared to the single layer model? Quantify.

p 2523, l 8: Doesn't this suggest that the assumed gamma (ground) is to high?

p 2523, l 8-22: Couldn't this also be achieve by increasing the gamma value in the single layer compensation point model?

p 2523, l 8: How does this sentence relate to the topic of this section? It looks like a general conclusion.

p 2525, l 10-12: I am not convinced that the 2-layer model led to significant improvement of model performance. I didn't see any quantification of the model results and it is hard to see by eye.

p 2525, l 24: I am really curious to see a comparison between a model run with leaf wetness modeled according to the energy balance method and a model run with the direct scaled leaf wetness measurements.

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