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Biogeosciences Discussions

Interactive comment on "Investigating the stomatal, cuticular and soil ammonia fluxes over a growing tritical crop under high acidic loads" *by* B. Loubet et al.

Anonymous Referee #1

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General comments:

The aim of this paper seems to be twofold. On the one hand, a new measurement campaign of ammonia fluxes is reported, which is very valuable, because there is only a limited number of ammonia flux measurement campaigns over cropland. On the other hand, a detailed process-based surface-atmosphere exchange model is defined and used to explain the observed fluxes.

I have some concerns on both topics.

Flux measurements of ammonia are extremely difficult due to its sticky character. Therefore it is absolutely necessary to have insight in the accuracy of the measure-

ments. In this case the ROSAA, i.e., a system with 3 mini-wedd denuders, is used to measure the concentrations at three heights, which are combined with turbulence measurements of a sonic anemometer to finally calculate the ammonia fluxes using the aerodynamic gradient technique. There are several requirements for applying this technique, including horizontal homogeneity of the area around the measurement set-up and absence of advection. As the field is 19 ha in size, the criterion of the horizontal homogeneity of the field is likely met (but should be shown in a location overview), but for the advection criterion a thorough footprint analysis is needed to be sure (and to show) that the farm is not disturbing the concentration profile and consequently the calculated fluxes. This footprint analysis will certainly lead to some data rejection as especially in stable nighttime conditions with calm wind from the farm, the concentration profiles are likely affected by local emissions. Excluding data from wind directions from the farm would be a less favorable option, as a lot of data is unnecessarily lost, because during unstable/neutral conditions the footprint in the direction of the farm is likely small enough (but this depends on the distance of the farm/sources).

Besides, insight is needed in the accuracy of the individual concentration measurements and possible systematic differences between the individual concentration measurements. Calibrating the detector unit with a calibration fluid is important for the absolute value of the concentrations, i.e., the accuracy. For the flux measurements, i.e., concentration differences, it is essential to exclude systematic differences between the different heights caused by inlets and tubing. An option to check for systematic differences in the field is to place the systems at the same height. In this way, possible systematic differences can be excluded and insight in the precision (random error) of the instruments can be obtained. With this random error in the concentration measurements, a random error in the flux can be estimated using error propagation. By doing this, one will find out that the fluxes during daytime (smaller concentration differences due to turbulent mixing) are generally less accurate than nighttime fluxes (larger concentration differences).

It is not clear to me, how the QC solution is exactly used. It looks like it is only used to improve the accuracy (correct the systematic difference) of the concentration determined by the analyzer, but not for determining systematic differences between the individual heights. It is not clear how the inlet and air tubing is taken into account in the determination of systematic differences between the three mini-wedds.

Another concern about the measurements is that the authors state that the lowest measurement point is at 0.53 m height at the beginning of the measurement campaign and that the whole system is leveled up during the measurement campaign to accommodate the canopy growth. At the end of the measurement campaign, the height of the lowest level is 0.98 m (see page 10324). However, on page 10322, it is stated that the canopy grew from 0.5 m to around 1.2 m. It looks like the lowest measurement point is therefore located within the canopy throughout the measurement campaign. For the described measurement set-up, the aerodynamic gradient technique is not valid, as the lowest measuring height is within the roughness layer of the canopy, in which the integrated stability correction functions are not valid and concentration profiles cannot be corrected for stability.

The concern about the modelling is mainly about the derivation of the Γc values, the choice of the Rw parameterization and the tuning of the Surfatm-NH3 model.

The authors start with the derivation of Cc values from the observed fluxes using a moving linear regression over successive 24 hr periods. I do not really know this method and it is not explained in the text either. An explaining figure would be helpful here. However, assuming that the derived Cc values are representative, it is not clear which temperatures are then used to convert the Cc values in Γc values. Is it the average temperature over this 24 hr period or the temperature at which the flux is zero, but how would you determine this from the regression?

Despite this, the resulting Cc and Γc values look quite reasonable for the few values I checked by taking the Ca at flux direction changes. As the data are filtered for RH

< 70% and WI = 0, which means that the cuticular pathway can be neglected, the Γc values could be seen as representative Γs values (if transport through the canopy to/from the soil is neglected), which can be compared with Γs values from literature, as is done in this study. It would be interesting to see if the weak temperature dependence of Γs (to describe the seasonal variation) that is shown in Wichink Kruit et al. (2010) Eq. 16 is also found in this study. It looks like the high values of Γs derived in this study (between 4 and 11 May) occur during rather cool weather conditions.

In this study, the choice for minimum cuticular resistance of 0.025 was made to reproduce the largest deposition fluxes in the period from 22 May till 11 June. However, if you plot the dependency of Rw on RH with this function, you will find out that even at RH smaller than 60%, the Rw is still lower than 10 s/m, which is extremely low. This makes the cuticular pathway dominant during the complete measuring/modelling period, because a 'shortcut' is created between the atmosphere and the leaf cuticle, i.e., the leaf surface can be considered as wet permanently. This makes that unrealistic high values of Γs and Γg are needed in the model to compensate this 'deposition' effect. Nemitz et al. (2001) have shown that the value of Rw depends on the molar ratio between SO2 and NH3 concentrations. This is implemented as αSN in the parameterization that is used by EMEP (Simpson et al., 2003) which is mentioned and used in this paper. It would be interesting to see a time series of the measured SO2 (and HNO3) concentrations to see if the measurement location is exposed to high SO2 concentrations during the period between 22 May and 11 June, which can explain the extremely low Rw values needed to simulate the measured fluxes.

Based on figure 8 in the paper, I doubt if the EMEP routine is correctly implemented (figure 8b and 8c). A higher SO2 concentration should give lower Rw values and more deposition. As there appear emission events in the time series in figure 8c that are not present in figure 8b, there must be an error in the EMEP formula used in figure 8b and 8c or there are more/other changes.

It seems that the derived Γs values in this paper are not used in the Surfatm-NH3 model, as for the flux calculation Γs is set to 0 and Γg is tuned for different periods (Figure 7) to fit the observed fluxes. This is a strange procedure, as the fluxes to/from the soil are much more uncertain than the fluxes to/from the vegetation. The sensitivity test also shows that extremely large values for Γg or Γs are needed to explain the emission events, which are quite unrealistic for the unfertilized conditions during the study. As explained above, these problems are likely caused by the too low Rw values (<10s/m for RH<60%) that are used.

As the authors have measured net exchange fluxes, it does not seem to be possible to derive parameterizations/values for the individual in-canopy fluxes without having detailed knowledge of the vegetation/soil. Interesting parameterizations/values could still be obtained by strict data selection, for example, it is still possible to derive parameterizations for the cuticular resistance by selecting turbulent nighttime conditions only. As the exchange pathway with the leaf cuticle appears to be the most important one for NH3, the cuticular resistance deserves more attention in this paper (instead of assuming a function with a minimum value that can only explain the largest deposition values). Probably/likely, it follows that the low value is correct, but that the RH dependency should be adjusted to obtain larger Rw values if the RH is low (dry surface).

A good knowledge of the uncertainty of the flux measurements is needed for a proper derivation of parameterizations/values, because daytime fluxes (smaller concentration differences due to turbulent mixing) are generally less accurate than nighttime fluxes (larger concentration differences).

Specific comments:

Page 10318:

l. 5: mini-WEDD called mini-wed on page 10321 l. 11 and mini-wedd on page 10323 l. 12.

l. 10: is the 29 ng NH3 positive or negative? If it is positive (as it is now), this is in

contradiction with 'occasionally" in l 14.

l. 12: replace 'in' by 'of'

l. 12: I doubt if the high acid conditions are the sole reason for the low surface resistances needed to explain the large deposition fluxes.

l. 18/19: it is not clear why the authors compare Γc with Γs as they are not the same, unless they are derived under specific conditions and assumptions.

page 10320:

l. 10-13: Reformulate sentence. There is now a contradiction in the sentence. The confusion is caused by the words 'also' and 'sink' in the last part of the sentence. I would say that a crop normally behaves as a sink, but can also behave as a source under certain atmospheric conditions. Split sentence in two sentences.

l. 20: Fléchard should be Flechard (also in Reference list)

l. 23: replace reference to Flechard et al., 2011 by Nemitz et al., 2001; Simpson et al., 2003

l. 24-28: I would suggest to reformulate the paragraph, e.g. "The number of studies reporting NH3 flux measurements is rather limited. There are a few studies reporting ammonia flux measurements over grassland (REFS) and semi-natural ecosystems (REFS), but measurements over cropland are rather scarce (Sutton et al., 1995, more references needed). Most of these latter studies focus on . . .. . .etc. "

l. 25: 'Wichink-Kruit' should be replaced by 'Wichink Kruit' (also in Reference list)

Page 10321:

l. 25-27: What is the distance between the farm and the experimental site? What is the influence of the farm on the flux measurements? Is there an effect of advection on the flux measurements? What is the footprint of the measurements?

Page 10323:

l. 25-28: How are the systematic differences between the wedds corrected? How large are the systematic differences/corrections (if determined/applied)?

Page 10325:

l. 14-20: I do not exactly understand how the Cc is determined. What is a moving linear regression? And which temperature do you use then to derive Γc values from the likely temperature dependent fluxes and concentrations? I doubt if this a right way to do it. It would probably be better to derive the Cc from flux direction changes, as the concentration gradients are approximately zero during these switches (by definition) and coupled to a certain temperature, which can be used to derive a Γc value.

Page 10326:

l. 20: How does this function for Rw corresponds with Rw values derived from the measurements (during turbulent nighttime conditions).

l. 25: Γg is used as a tuning parameter and is not based on physical properties of the soil here.

Page 10328:

l. 1: 'periods 27 April-4 May and 6 June and 15 June' should likely be 'periods 27 April-4 May and 6 June-15 June'

l. 8: There hardly seem to be gaps in the data of the ROSAA in June, so, it seems to be unlikely that this is the correct explanation.

l. 11-12: remove 'the' before dates (2x) or add 'th' / 'st'

l. 11-13: the levels of the peaks in figure 5 and figure 4 do not correspond! It looks like the second peak in figure 5 does not appear in figure 4. Why is this?

Figure 5 shows that the wind directions from the farm should be excluded from the

data analysis as they can disturb the concentration profile, leading to unrealistic fluxes caused by advection. This is one of the conditions to be met if the aerodynamic gradient technique is used. So, it needs to be shown that the footprint for the measurements is small enough (or the fetch is large enough).

What do the purple and cyan lines in Figure 5 mean?

l. 14: How far is the farm from the measurement device?

l. 18-19: This is likely the answer on the question on the first page. It appears to be a 'minus'. But as it is a minus: Is it likely that the advection from the nearby farm caused that the concentration at the highest level is most of the time higher than the concentration at the surface? I think that the measurement location is subject to advection problems from time to time, and thus, a strict selection of data (from other directions than from the farm) would be needed to draw any useful conclusions on the behavior of the crop. Besides, tuning of the model is extremely difficult, because it is not possible to account for these local effects.

l. 25: As clear changes in the sign of the flux are seen, this seems to be a perfect period for deriving Cc values and Γc values.

Page 10330:

Figure 7 shows some strange things:

- (a) How would the authors explain the enormous jumps in Γsoil needed to fit the model on the measurements?

- (c) Vmax is the maximum atmospheric transport possible through the atmosphere (as well for deposition as for emission). How do the authors explain that the observed and modelled vd's are sometimes several times larger than the maximum transfer velocity (Vdmax)? How is the measured vd determined? Does it account for a possible surface concentration or is it just the flux divided by the concentration (at z-d = 1m) and is the small concentration the reason for the extreme vd's?

Please do not use dashed lines in this plot. It makes it impossible to see if data is missing or if it is just because of the dashed line. Use red, green, blue, black. . ..

- (d) the stomatal flux does not seem to play a significant role at all. Only during periods with a zero cuticular flux the stomatal flux explodes. It looks like this is an error in the plotting procedure, as no flux measurements are available in the periods where this happens. So please leave data out if no flux measurements are available.

- (e) Do not use a dashed line (see comment at figure 7c) for the measured flux.

l. 1-8: How large is the contribution of the soil and the in-canopy flux to the modelled LE? Isn't the soil water potential included in the parameterization for the stomatal resistance? It would be interesting to know if the soil path really contributes to the total LE modelled. This would give a clue if this pathway might be important for the NH3 flux.

l. 18-21: The value of Rw can be investigated in this study, by selecting turbulent nighttime data.

l. 25: 'compared favourably' On Page 10328 the authors mention that especially in June the concentrations measured with the different instruments differ considerably.

Page 10331:

l. 8-9: 'while we mainly found deposition here' seems to be logical as there might be an advection problem from the nearby farm, which mainly affects the concentrations at the highest measurement levels. A footprint analysis should be carried out to draw motivated conclusions and to exclude advection.

l. 23-27 and next page: Many studies in Massad et al. (2010) refer to the same site, so it is not allowed to just take the median of all values reported.

Page 10332:

l. 25-27: numbers do not correspond. 44-66 = -22 and not -24

Page 10334:

Why do there occur emission peaks in the run with EMEP-03 with [SO2] = 5 ppb, while they did not occur in the run with EMEP-03 with [SO2] = 1 ppb. One would expect that when the surface is acid, there is always more deposition. It looks like there is an error in the implementation of the EMEP-03 parameterization. Likely a different value for the RH dependency or the RH on a different level is used, as it seems that in figure 8c the surface is dry from time to time, making stomatal/soil emission possible.

Page 10335:

l. 7-9: It is strange that a lower compensation point could explain the observed emission periods. Is this not a fifth interpretation?

l. 15: I don't agree. The value for May shows a big difference (2.0 vs. 3.0 μg/m3).

l. 18: change '29.3' into '29'

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