

We would first like to thank referee #1 for its very helpful comments, which we hope have helped improving the paper very much. The comments from the referee #1 are in blue and answers are in red.

Benjamin LOUBET

Reply to anonymous referee #1 comments bgd-8-C4865-2011

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 measurements. 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 setup 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).

This is a very good comment: indeed NH_3 fluxes measurements are highly subject of advection issues. To better show the field and farm set up, we propose, as suggested by referee #1, to include the map given in Figure A1 below in the manuscript. Moreover, we have performed a footprint analysis with the Kormann and Meixner (2001) footprint model. This footprint analysis shows that the Farm has a very limited impact on the flux footprint measured in the field (Figure A2): An average footprint of 0.02% from the farm is found. This

is merely due to the fact that the farm was located at around 460 m away from the gradient mast and therefore had a low impact on the flux.

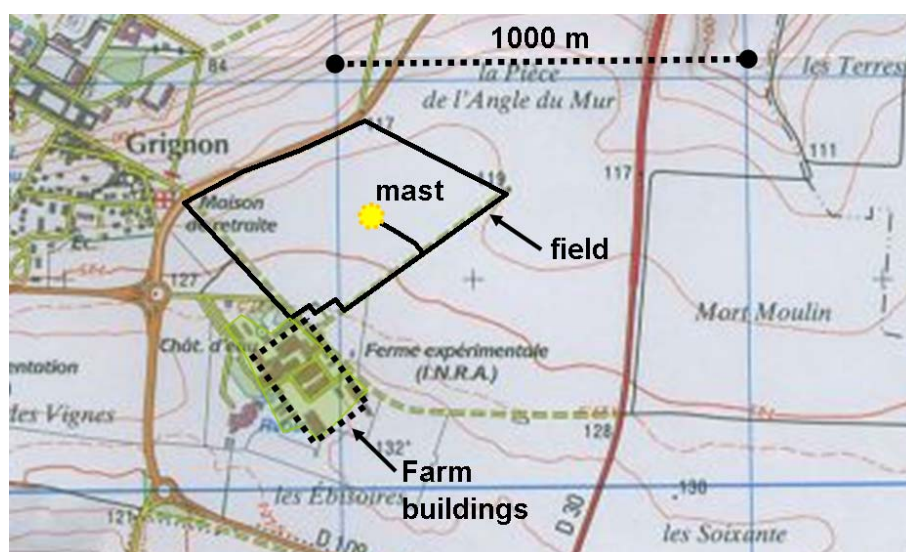


Figure A1. Map of the experimental area showing the field, the Farm buildings and the mast location.

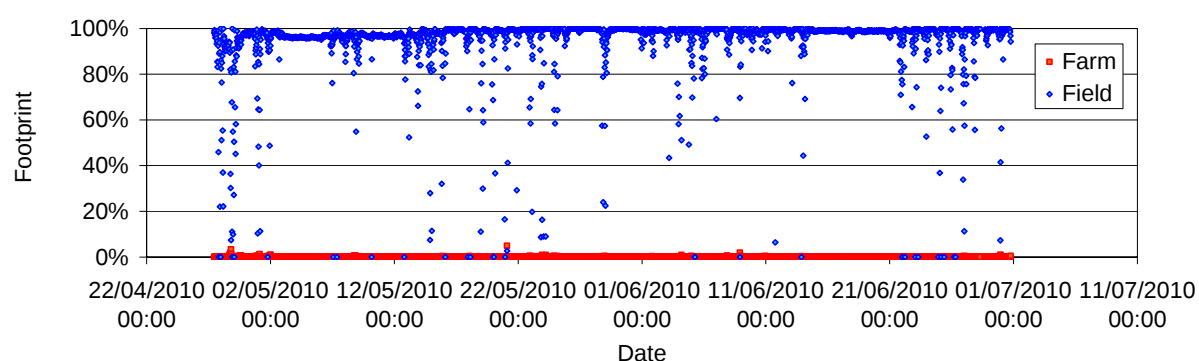


Figure A2. Flux footprint of the NH_3 flux measurement mast. Footprint from the field is [95% - 99% - 14%], and from the Farm is [0.02% 0% 0.15%], with under brackets are [mean - median – standard deviation].

Regarding the issue of data rejection we believe as referee #1 that wind sector rejection would not be appropriate as it would dismiss the most interesting data in terms of NH_3 deposition. Based on the flux footprint, rather than rejecting the data we have done an advection flux estimation using the FIDES-2D model and the approach exposed in Loubet et al. (2009). To do so, the farm emission was inferred using the measured concentrations at the highest level at the mast location and the modeled surface resistances using the FIDES-2D inverse model

(see Hensen et al. 2009 for a full description of the method). The Farm emission was evaluated to be $8.3 \text{ kg N-NH}_3 \text{ day}^{-1}$, which gives $3030 \text{ kg N-NH}_3 \text{ year}^{-1}$ if assumed constant over the year. Using Emission factors from (Gac et al. 2006), i.e $12 \text{ kg N-NH}_3 \text{ year}^{-1}$ for caws and $1.34 \text{ kg N-NH}_3 \text{ year}^{-1}$ for sheep, we find an emission of $3900 \text{ kg N-NH}_3 \text{ year}^{-1}$ for this farm that has around 210 caws and 510 sheep, the estimation of $3030 \text{ kg N-NH}_3 \text{ year}^{-1}$ being therefore quite reasonable.

From that point, we estimated the advection error with the FIDES model following Loubet et al. (2009) as the difference between the flux modeled at the measurement height ($d+z_m$ and the flux at the canopy top $d+z_0$). We found that the local advection flux due to the NH_3 emitted by the farm averaged $6.4 \text{ ng NH}_3 \text{ m}^{-2} \text{ s}^{-1}$ (median $2.7 \text{ ng NH}_3 \text{ m}^{-2} \text{ s}^{-1}$). Moreover, the advection error was found to increase with increasing deposition, which reflects the fact that the deposition increased with increasing atmospheric concentration (itself being influenced by the nearby farm (Figure A3). Overall, the advection error was therefore quite small and represented around -8% of the measured flux. The flux corrected for the advection error would therefore be around 8% larger than the measured flux (Figure A3).

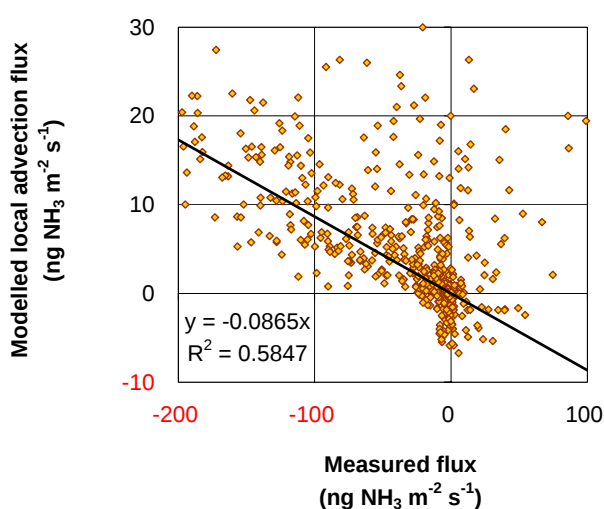


Figure A3. Modeled local advection flux at the highest level of measurement mast, due to the influence of the farm. Estimation obtained with the FIDES model (see Loubet et al., 2009).

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).

This is again a very sound comment. Unfortunately we must acknowledge that we did not do any inter-calibration of the denuders during the field campaign but only before, in the lab. So we could therefore not tell afterwards whether there has been a malfunction in the analyzer. However, we should stress that the ROSAA analyzer was set up to minimize these systematic differences:

- The denuders avoid any systematic differences due to sampling in tubes since ammonia is directly converted in ammonium in the acid solution trap. We however acknowledge a slight possibility of adsorption in the ~2 cm long glass dry inlet tube.
- The design of the glass denuders was carefully chosen and tested to minimize the differences between the three levels.
- In each line, the air flow rate is maintained within less than 1% (in mass) using controlling flow meters.
- The liquid flow rates are measured using a balance and there was a control afterwards to check that the quantity of liquid was not changing over time.

Beside these arguments which give confidence in these measurements, any systematic difference between the three channels was looked at a few days before the campaign. For this we generated step concentrations at $60 \mu\text{g NH}_3 \text{ m}^{-3}$, $260 \mu\text{g NH}_3 \text{ m}^{-3}$, and $1160 \mu\text{g NH}_3 \text{ m}^{-3}$ using a permeation device. We did not achieve stable concentrations at lower levels due to NH_3 stickiness and therefore only tested these higher levels. The three denuders were then sampling simultaneously this standard for 6 to 24 hours. From these data we determined that the denuders were reading the same concentration with an accuracy of 4.4%. Using this uncertainty in the concentration measurements, the flux uncertainty can be evaluated using the following equation on the relative uncertainty.

$$\frac{\sigma_{F_c}}{F_c} = \frac{\sigma_{u^*}}{u^*} + \frac{\sigma_{C^*}}{C^*} \quad (\text{A1})$$

letting aside the variability due to u^* for the moment (which is intrinsic to the gradient method). One can evaluate the uncertainty in the flux due to the uncertainty in C^* . (where C^* is evaluated as in eq. (2) in the manuscript:

$$C_* = k \frac{\partial C_{NH_3}}{\partial \left[\ln(z-d) - \Psi_H \left(\frac{z-d}{L} \right) \right]} \quad (2)$$

To do so, we generated 100 points per measured concentration point having with a random generator (average = NH_3 and standard deviation = 4.4% of NH_3) and estimated C^* from these 100 points per heights and sample time with a linear regression. Doing so, we find that the relative uncertainty on the flux was 18% [9% - 43%] (median [25percentile-75percentile]). The 95% confidence interval of the NH_3 flux is shown in Figure A4.

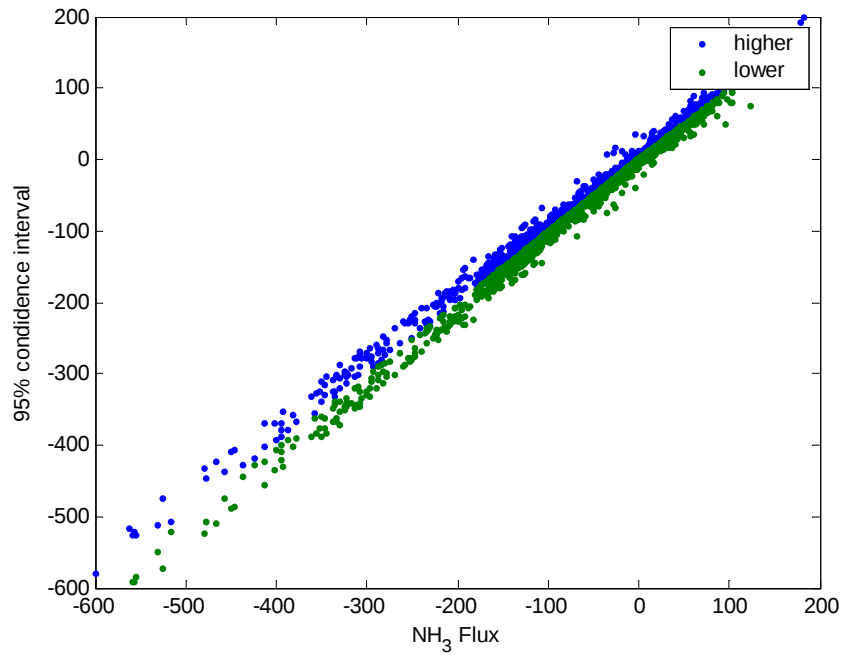


Figure A4. Estimated NH_3 flux 95% confidence interval, based on a 4.4% uncertainty on each concentration measurement in equation (2) showed as a function of the measured flux.

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.

The QC solution was indeed used to correct for systematic differences. It was not possible to correct for systematic differences in individual heights with the setup used in the experiment. This is however a good idea that we will look at for improving the analyzer in the future (using a standard QC that could be put in each denuder reservoirs). The inlet air tubing is very small (around 2 cm long glass tube), and as explained earlier on, no systematic differences were observed in the laboratory tests.

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.

We are sorry to realize that the reported canopy heights were wrong, and we apologize for that. The correct canopy heights are now given in the updated Figure 2 of the manuscript (given here below). The measured heights are also given in Table A1. At the end of the growing season h_c was 0.95 ± 0.05 m.

In the field, the system was indeed leveled up to keep the lower denuder above the canopy. We did that by eye based on the level of the canopy surrounding the denuders. However the height of the crop was quite variable within the field, and especially in the early growth stage as shown by the CV in Table A1, and it happened that the height of the canopy the 27/04/2010 evaluated by linear interpolation between the 16/04 and the 27/05 was 0.5 m and

was indeed very close to the denuder 1 height (0.53 m), and similarly at the end of the experiment where the canopy height was 0.95 m and the denuder height was 0.98 m.

We should however stress here that the displacement height (which is the parameter used in the aerodynamic gradient method) was inferred from wind speed gradient and corresponded therefore exactly to the dynamical definition: $(d+z_0)$ is the height where the measured wind speed is equal to zero (this was evaluated in neutral conditions). The displacement height inferred was ranging from 0.37 m to 0.86 m.

Regarding the fact that the lowest height was measuring within the roughness sub layer, we have to acknowledge that the lowest measurements height were even within the roughness layer during some periods. But the reason for this choice was to maximize the concentration gradient in order to be able to discriminate the concentration differences with the analyzer. This is unfortunately a compromise often necessary in aerodynamic gradient methods, which are most often completely or partly in this roughness layer. There exist roughness layers corrections but they are adapted to higher canopy (maize, forest) and we therefore decided not to use them (see e.g. Garrat, 1978; Cellier and Brunet, 1992 or Wenzel 1997). We however propose to discuss this further in the revised manuscript.

Table A1. Measured height of the canopy (h_c). Mean standard deviation and coefficient of variation.

date	mean	std	CV
-	m	m	%
16/4/10	0.26	0.10	37%
27/5/10	0.95	0.12	12%
16/7/10	0.95	0.05	5%

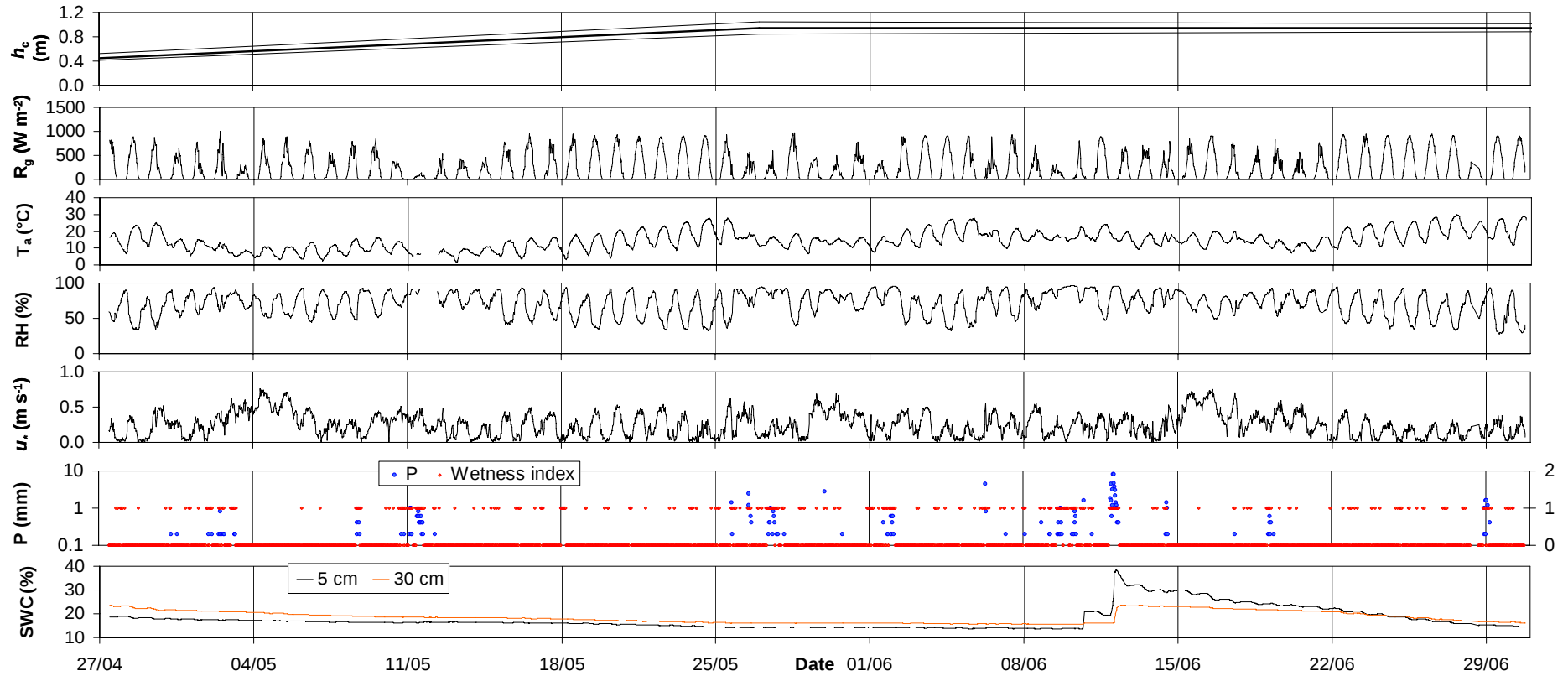


Figure 2(revised). Canopy height (h_c) and micrometeorological conditions measured at the site. From top to bottom: canopy height (h_c), global radiation (R_g), air temperature (T_a) and relative humidity at 2 m above the ground, friction velocity (u_*), precipitation (P, left axis), wetness index (right axis), and soil water content (SWC) at 5 and 30 cm depth. **For the canopy height, the mean, minimum and maximum heights are shown.**

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

The authors start with the derivation of C_c 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 C_c values are representative, it is not clear which temperatures are then used to convert the C_c values in Γ_c 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?

The derivation of the C_c values from observation is obtained by linear regression between the NH_3 concentrations at 1 m height and the NH_3 flux: C_c is simply the x-intercept of the regression. This is illustrated in the Figure A5. in order to get a continuous estimation of C_c , the regression is performed over a running range of data (for each date, the 24 h data around that date are selected). To convert C_c to Γ_c , the air temperature averaged over the same period (the period over which C_c was derived) is used: $\Gamma_c = C_c / 10^{(-3.4362 + 0.0508 \cdot T_{\text{air_averaged}})}$. Using the temperature at which the flux is zero would indeed not be feasible.

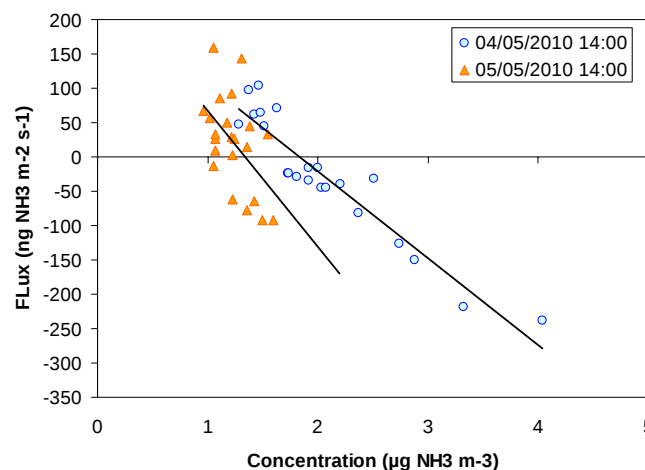


Figure A5. Illustration of the linear regression to obtain the canopy compensation point (C_c): C_c is the intercept of the linear regression between the measured NH_3 concentration (at 1 m height) and the NH_3 flux. In this case two successive days of data are shown.

Despite this, the resulting C_c and Γ_c values look quite reasonable for the few values I checked by taking the C_a 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 Γ_c values could be seen as representative Γ_s values (if transport through the canopy to/from the soil is neglected), which can be compared with Γ_s values from literature, as is done in this study. It would be interesting to see if the weak temperature dependence of Γ_s (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 Γ_s derived in this study (between 4 and 11 May) occur during rather cool weather conditions.

We would like to thank the referee for suggesting this comparison with Wichink Kruit et al. (2010). Indeed, we checked whether there was a relationship between Γ_c and the leaf temperature (which was obtained via Surf-atm-NH₃) and found that Γ_c was decreasing with leaf temperature. The decrease was similar in shape to that found by Wichink Kruit et al. (2010) for Γ_s as a function of the surface temperature (see Figure A6), although it shows a larger slope ($\exp(-0.15 T)$ instead of $\exp(-0.071 T)$). Since our estimate is based on Γ_c it includes the effect of the soil and cuticle exchange but as noted by the referee #1, it is derived in dry conditions and could therefore be comparable to a Γ_s . This result suggests that the parameterisation of Γ_s Wichink Kruit et al. (2010) is compatible with our study.

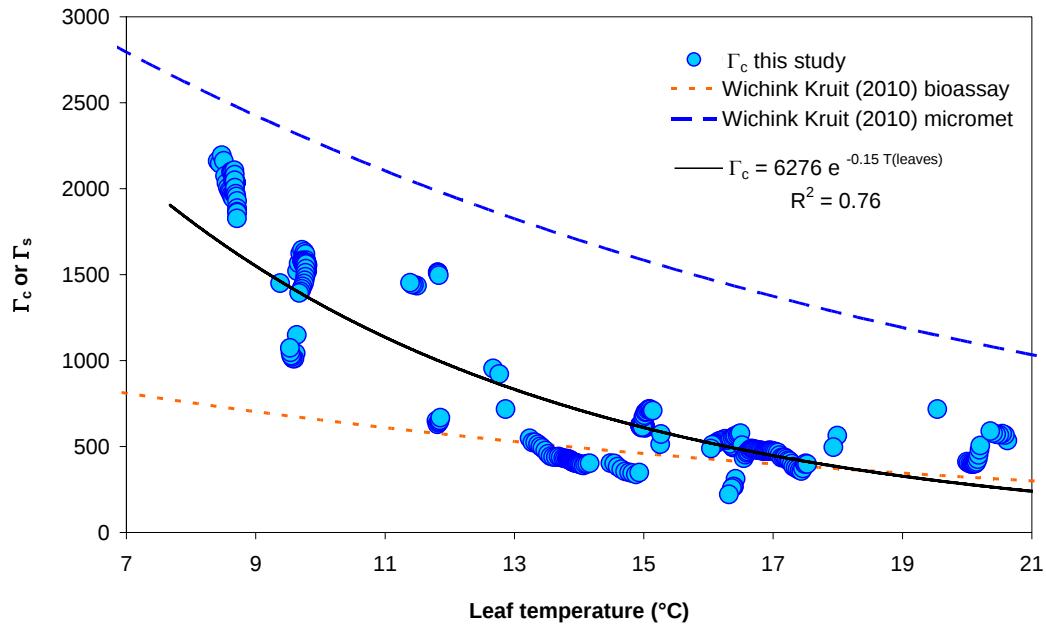


Figure A6. Γ_c as a function of leaf temperature (estimated with Surfam NH_3) (blue dots). The solid black line is the fitted exponential function and the dotted lines are the modelled Γ_s in Wichink et al. (2010) using either the micrometeorological estimate (blue dotted line) or the bioassay estimate (orange dotted) of Γ_s . The latter were estimated using the regression lines in Figure 9 to retrieve $\Gamma_{s,\text{method}}$ and equation (16) of Wichink et al. (2010) to express the dependency to leaf temperature ($\Gamma_s = \Gamma_{s,\text{method}} 4.7 \exp^{-0.071 T_s}$). The $\Gamma_{s,\text{method}}$ were estimated based on the average NH_3 concentration during the experiment (were 260 for the bioassays and 890 for the micrometeorological estimates).

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 R_w on RH with this function, you will find out that even at RH smaller than 60%, the R_w 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 Γ_s and Γ_g are needed in the model to compensate this 'deposition' effect. Nemitz et al. (2001) have shown that the value of R_w depends on the molar ratio between SO_2 and NH_3 concentrations. This is implemented as SN 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 SO_2 (and HNO_3) concentrations to see if the measurement location is exposed to high SO_2

concentrations during the period between 22 May and 11 June, which can explain the extremely low R_w values needed to simulate the measured fluxes.

It is true that the choice of the minimum cuticular resistance of 0.025 was required in order to reproduce the largest deposition observed from 22 May till 11 June. And it is also true that the choice of such a small resistance over the whole period leads to probably too large estimates of Γ_s and Γ_g with Surf_{atm}-NH₃. However, since we only had monthly averages of SO₂ and HNO₃ concentrations we chose to take a constant parameterization for R_w throughout the experiment. This is questionable but we felt we had not enough temporal knowledge to give a varying parameterization of R_w . The SO₂ and HNO₃ concentrations were measured monthly with Delta tubes and the concentrations measured in the field are shown in Figure A7, together with the averages of the AIRPARIF stations of western Paris. The regional data tends to show a slight increase in maximum daily SO₂ concentration between the 17/05 and the 12/06 and another one towards the end of the period.

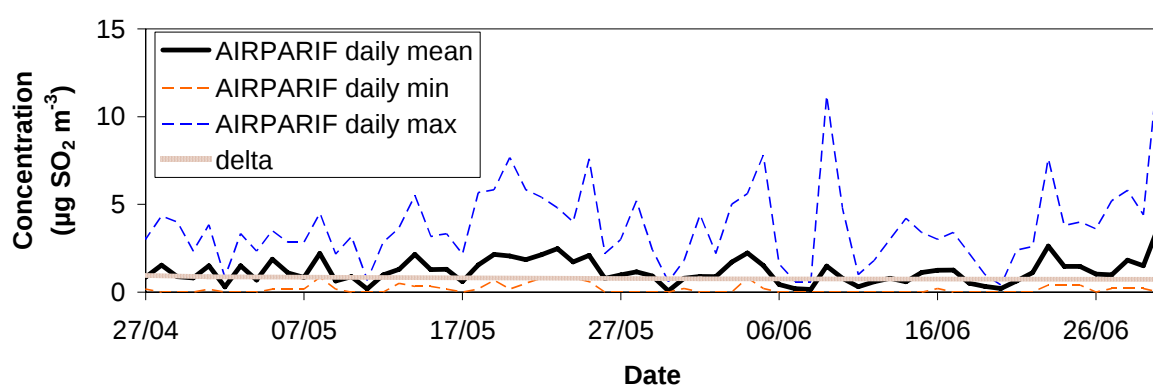


Figure A7. Measured SO₂ concentrations with the delta tubes together with the SO₂ concentrations averaged over the west of the Parisian region from air quality measurements (AIRPARIF data).

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 R_w 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.

We thank the referee #1 for spotting this issue. Indeed, after checking there was an error in the EMEP routine used and the graphs 8b and 8c: the dependency to relative humidity was not included (the line was commented in the model). The model was run again and the Figures 8b and 8c were updated (and are shown below). With this updated routine, it shows that the EMEP routine with high SO₂ concentrations can not explain completely the measured deposition flux. There must be another factor influencing NH₃ deposition here.

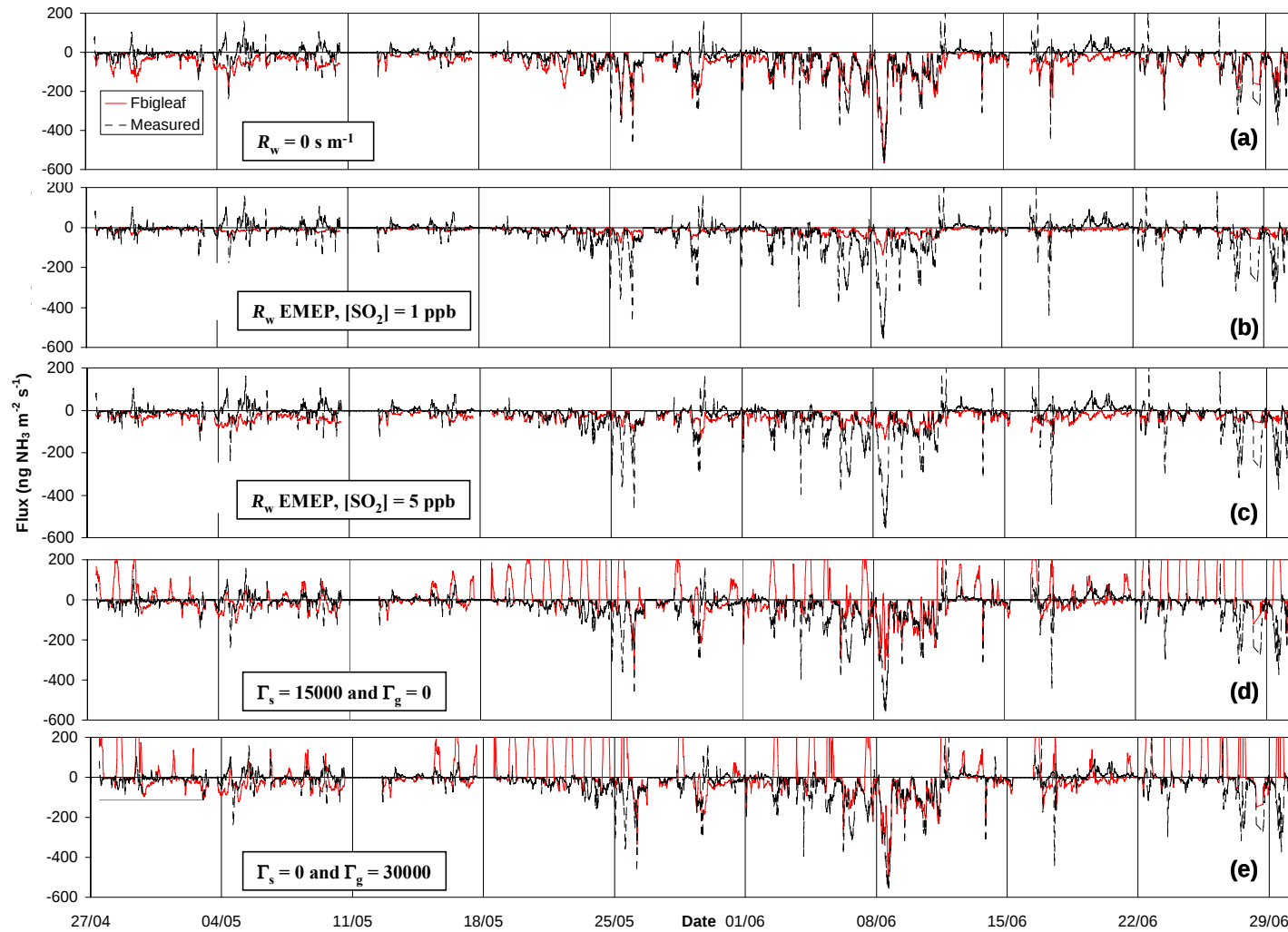


Figure 8(updated). Sensitivity analysis of the Surf-atm-NH₃ model to the cuticular resistance, the soil (Γ_g) and stomatal (Γ_s) emission potentials. From top to bottom: **(a)** $R_w = 0 \text{ s m}^{-1}$, **(b)** R_w from EMEP-03 with $[\text{SO}_2] = 1 \text{ ppb}$, **(c)** R_w from EMEP-03 with $[\text{SO}_2] = 5 \text{ ppb}$, **(d)** R_w fitted and $\Gamma_s = 15000$ and $\Gamma_g = 0$, **(e)** R_w fitted and $\Gamma_s = 0$ and $\Gamma_g = 30000$. If not otherwise stated, $R_w(\text{fitted}) = 0.025 \exp^{((100 - RH(z_0))/7)}$ and $\Gamma_s = \Gamma_g = 0$.

It seems that the derived Γ_s values in this paper are not used in the Surfatm-NH₃ model, as for the flux calculation Γ_s is set to 0 and Γ_g 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 Γ_g or Γ_s 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 R_w values ($<10\text{s/m}$ for $\text{RH}<60\%$) that are used.

We have to admit that the choice we made to tune Γ_g and set Γ_s to zero was subjective. We also agree with referee #1 with the fact that the very low R_w values have led to large estimations of Γ_g . However, since we do not have bioassays measurements to support Γ_s emissions, we preferred to leave the leaf compartment to zero. The difficulty here is that we could indeed find other solutions that may fit the data (with lower R_w and lower Γ_g), but we do not have the data to decide which solutions is the best. In order to answer this comment from referee #1 we have however performed 2 additional simulations

- (1) R_w as in EMEP with acid concentration as measured with the delta denuders, apart from the period 25/05-11/06 where it was set to the optimized R_w ($R_{w\text{min}} = 0.025 \text{ s m}^{-1}$). The Γ_s was set to that estimated (Γ_c as in Figure A6b), and Γ_g was tuned (see Figure A8a for the results)
- (2) R_w as in EMEP with acid concentration as measured with the delta denuders, apart from the period 25/05-11/06 where it was set to the optimized R_w ($R_{w\text{min}} = 0.025 \text{ s m}^{-1}$). The Γ_g was set to 0 and the Γ_s was tuned (see Figure A8b for the results).

From Figure A8, we can see that tuning Γ_g gives a better fit with the measured fluxes during the period 4-18/05 (with a unique value of $\Gamma_g = 20000$) while it was harder to get a good fit with Γ_s . It is however difficult to conclude which method should be preferred. Additional measurements would be required and future experiments should help interpreting these results.

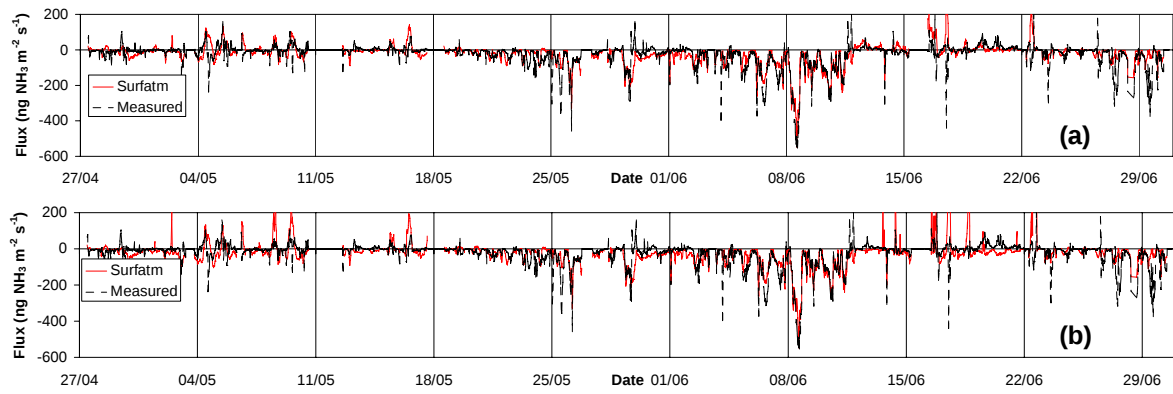


Figure A8. Simulated and modeled NH_3 fluxes with R_w as in EMEP (acid concentration = 1.3 ppb), during the period 25/05-11/06 ($R_{w\min} = 0.025 \text{ s m}^{-1}$). **(a)** Γ_s was set to Γ_c (using the regression curve of Figure A6b), and Γ_g was tuned. **(b)** $\Gamma_g = 0$ and Γ_s was tuned.

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 NH_3 , 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 R_w 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).

We agree with the referee #1 statements that it is difficult to derive individual in-canopy fluxes parameterizations from net fluxes easily. We have tried to see if under turbulent ($u^* > 0.3 \text{ m s}^{-1}$) night-time ($R_g < 10 \text{ W m}^{-2}$) conditions, the canopy resistance $R_c(\text{NH}_3)$, $R_c = 1/V_d(\text{NH}_3) - 1/V_{\max}(\text{NH}_3)$ was a function of RH but we found no relationship. This was due in part to the fact that night time conditions only exhibited large RH.

Specific comments:

Page 10318:

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

Changed

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.

It is negative. "occasionally" was withdrawn.

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

OK

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.

We actually agree. We have moderated the sentence consequently.

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

This was just an acknowledgment that our estimation of Γ_c is close to median stomatal emission potential (Γ_s) for managed ecosystems reported in the literature. However, the Γ_c were actually derived under specific assumptions (dry conditions) which should correspond to minimal cuticular deposition. Nevertheless, we have now mentioned the comparison with Γ_g measured in arable land in the literature (median of Table 4 of Massad et al. (2010) for arable land: $\Gamma_g = 2472$).

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.

The sentence has been reformulated as follows: "However, as a result of the complex interactions between the sources and sinks within a canopy (turbulent transfers between the

canopy compartments), a fertilized crop may also behave as a sink for NH₃, especially downwind from NH₃ hot spots where atmospheric NH₃ concentration can be high”

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

Done.

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

Done. The reference to Simpson has also been added in the reference list.

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. "

Done.

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

Done.

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?

These questions have been already answered to (see at the beginning of this document). The distance has been added in the text. The advection correction and footprint issues have been introduced in the material and methods and results sections.

Page 10323:

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

These questions have been already answered to (see at the beginning of this document). The text has been modified accordingly in the manuscript. A short paragraph has also been added in section 2.4 “Ammonia fluxes with the aerodynamic gradient method” regarding the uncertainty of the NH_3 flux.

Page 10325:

l. 14-20: I do not exactly understand how the C_c is determined. What is a moving linear regression? And which temperature do you use then to derive Γ_c 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 C_c 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 Γ_c value.

This question has been answered to already (see at the beginning of this document). We still believe that the method we have employed is appropriate as the temperature employed is averaged over the same period. A graph has been added to illustrate the method.

Page 10326:

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

As discuss previously we did not see any clear relationship between R_c and RH during turbulent and nighttime conditions, mainly because there are not many data and RH was always larger than 70% during these conditions.

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

True. This is what is meant in this sentence.

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'

This is right.

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.

I think there was a mistake in the sentence: We were meaning "The concentration averaged over May and June 2010". The number of missing NH₃ concentration in May was 15% and 6% in June, so this may still be an explanation.

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

Done

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?

Thanks for spotting this error. A zoom of Figure 4 on the same period shows the same graph except one point on the 1 June. This point was rejected by data quality which was applied later and the graph was unfortunately not updated.

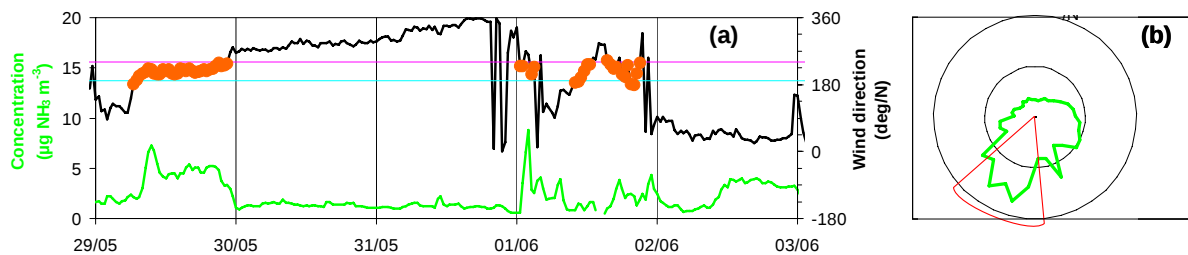


Figure 5(updated). Example period when the NH_3 concentration was enhanced when the wind was blowing from the farm. The purple and Cyan lines show the wind sector where the wind was blowing from the farm. (b) Ammonia concentration rose averaged over the whole period. The wind sector where the wind is blowing from the farm is materialised by the red triangle.

The advection and footprint issues have been dealt with already.

The purple and Cyan lines show the wind sector where the wind was blowing from the farm.

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

This was answered already. The text has been adapted accordingly.

I. 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.

This has been discussed already in the previous part of this document. The contribution of advection was modelled found to be contribute to around 8% of the flux.

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

It is true and actually this is what happened in the data selection (Figure 6): this period is when most of the Γ_g were estimated (red dots in Figure 6).

Page 10330:

Figure 7 shows some strange things:

- (a) How would the authors explain the enormous jumps in G_{soil} needed to fit the model on the measurements?

This was already discussed in the answer to the general comments. These jumps are explained by the tuning procedure used: the Γ_g was fitted (subjectively, we have to acknowledge) by periods as long as possible. And the values were estimated by manually fitting. This is why we have such jumps.

- (c) V_{max} 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 v_d 's are sometimes several times larger than the maximum transfer velocity (V_{dmax})? How is the measured v_d 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 v_d 's?

Indeed V_d was taken, as it is usually defined as the flux divided by the concentration at 1 m above d ($V_d(1\text{ m}) = F / C(1\text{ m})$). It does not account for the surface concentration (as the latter is unknown). This is why negative V_d can be several times V_{max} in absolute values.

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....

Done.

- (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.

Done.

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

Done.

l. 1-8: How large is the contribution of the soil and the in-canopy flux to the modeled 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.

The water potential effect on the stomatal conductance was included as in EMEP. The soil path contributed roughly to 17% of the total LE flux on average over the period. However this would unfortunately not give a clue on the pathway for NH₃ as the NH₃ flux would mainly be driven by the compartment potentials (stomatal and soil) which differ from the water concentrations.

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

As discussed earlier on, unfortunately the data available during turbulent night-time conditions were mostly at RH larger than 70% and no relationship could be found with RH.

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

As mentioned earlier, we mixed up the months and we meant May for June and June for July. In June the concentrations were close to each other while in May the difference was quite large (and may potentially be explained by missing data). The text has been adapted accordingly.

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.

This advection analysis was performed and mentioned earlier. In this section it was already pointed out that in our study the concentration was quite high compared to Schjoerring and Mattsson (2001), and we added that it was linked to local advection: "*Additionally, the weekly averaged NH_3 concentration at 1 m above displacement height in Schjoerring and Mattsson (2001) was between 1 and 4 $\mu\text{g NH}_3 \text{ m}^{-3}$ whereas it was up to 5 $\mu\text{g NH}_3 \text{ m}^{-3}$ during two weeks in our study (01-15/06), emphasising the fact that in our case the flux was strongly influenced by NH_3 emissions from the nearby farm.*"

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.

Although, in principle we agree with referee #1 comment, since the number of data available is small, it would similarly bias the data to avoid such or such dataset. As a first approach, we are confident in Massad et al. (2010). We have however added a sentence to recall the fact that most of the data used are from the same site: "*, although we should point out that most of these data are issued from the same field site, and may therefore not be representative of all conditions.*"

Page 10332:

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

True. Changed.

Page 10334:

Why do there occur emission peaks in the run with EMEP-03 with $[\text{SO}_2] = 5$ ppb, while they did not occur in the run with EMEP-03 with $[\text{SO}_2] = 1$ 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.

This issue has already been answered to.

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?

No: as mentioned by referee #1 earlier, higher R_w would lead to smaller uptake by the cuticle and therefore emissions could be explained by lower compensation points.

l. 15: I don't agree. The value for May shows a big difference (2.0 vs. 3.0 $\mu\text{g}/\text{m}^3$).

Agree. We should only say that it agreed well in June (2.4 vs 2.5 $\mu\text{g NH}_3 \text{ m}^{-3}$). The manuscript has been changed accordingly.

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

Done.

References quoted in the text and that will be added in the MS

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