

We would like to thank both anonymous referees for their valuable comments on the manuscript. Referee comments are given in bold, the answers in standard font. The first points by Referee 2 were numbered from #1 to #6.

Answer to Referee 1 (Part A)

Abstract, line 31. The statement regarding the potential for QCL to be applied for NH₃ flux measurements within long-term research networks such as NEON may be a bit strong. The concentrations at which the instrument has been applied in this study are not generally representative of NEON sites, in fact they are much higher. The current paper does indeed demonstrate the potential for use at sites influenced by local NH₃ sources, where concentrations are relatively high, but further characterization of the instrument performance at low concentrations would be necessary to suggest applicability at sites typical of NEON.

We will add 'at sites with strong local ammonia sources leading to relatively high mean background concentrations and fluxes.' Indeed we are currently conducting a field campaign at a forest site without local sources and we are able to reliably measure very low concentrations in a range from 0 to 10 ppb.

Page 4, line 5. At the flow rate and tubing dimensions noted, was the sample flow fully turbulent? The authors note a high frequency damping factor of 0.67 derived from ogive analysis on page 5 but do not explain the cause. A little more information here would be helpful.

According to the Reynolds number (approx. 2400 – 2700) the flow was not fully turbulent (with $Re_{crit} > 3000$), but also not laminar. High damping occurs usually because of imperfect turbulent flow regime and not completely excludable adsorption effects in the sampling line. Over time, especially the inlet gets clogged with aerosols, dust etc., which may cause unintended reactions of ammonia. However, due to the relatively short measurement period, we do not expect that increased clogging at the inlet alters the offset of NH₃ loss through reactions with particles and wall adsorption effects on the one hand and NH₃ production through the volatilization of NH₄NO₃ aerosols on the other hand as is already discussed on page 4, lines 8–25.

In the EC-community gases like H₂O measured with a closed-path device usually undergo spectral damping and are corrected for in a similar way. Ferrara et al. (2012; 2016), also using an Aerodyne QCL, found similar total damping factors. On Page 5, line 16, we add '... , which usually occurs due to imperfect turbulent flow regime and possible minor wall sorption effects in the sampling line.'

Page 5, Section 2.3. As stated in the introduction, one of the three objectives of the paper is to “test the QCL performance to measure NH₃ concentration fluctuations and calculate NH₃ fluxes . . .”. In this regard, some additional information on instrument performance and relation to flux calculations is warranted. Specifically, the only mention of precision comes at the top of page 4, line 2, referenced to McManus et al. (2008). A more detailed description of instrument precision would be informative. For example, what is the precision at sampling rates corresponding to frequencies of the flux contributing eddies (see Ferrara et al., 2012)? Related to precision is the flux detection limit. For their setup and site conditions, Ferrara et al (2012) estimated the flux detection limit to be $0.25u^*$, or about $75\text{ngNH}_3/\text{m}^2/\text{s}$, which is large relative to the fluxes reported in the present study. The authors should include an estimate of flux detection limit in their results.

We will add information on instrument precision on page 4, line 2ff. The main frequencies of the flux contributing eddies were in a range of 0.01 to 1 Hz. Precision, i.e. measurement sensitivity, of the instrument is 0.042, 0.021, 0.016, and 0.010 ppb in 1, 10, 20, and 60 s, respectively. The random flux error was computed according to the method after Finkelstein & Sims (2001) and the corresponding limit of detection was determined after Langford et al. (2015) at a confidence interval of $\alpha=1.96$ and resulted in a median of $7.75 \text{ ng N m}^{-2} \text{ s}^{-1}$. Alternatively, an upper flux detection limit for half-hourly values can be calculated by using only night-time data under stationary conditions ($3 - 7 \text{ m s}^{-1}$ wind speed) and wind from the west-southwest sector, where local ammonia sources were negligible and concentrations were low ($< 15 \text{ ppb}$). The standard deviation was $16.5 \text{ ng N m}^{-2} \text{ s}^{-1}$, so the 2σ -uncertainty range is $33.0 \text{ ng N m}^{-2} \text{ s}^{-1}$. We will add a section about flux detection limits to the manuscript.

Page 6, section 2.4. How is the stomatal compensation point parameterized? What value is used for emission potential (Γ_s)?

We used the N-input dependent parameterization from Eq. (8) of Massad et al. (2010):

$$\Gamma_s = 246 + 0.0041 \cdot N_{\text{in}}^{3.56}$$

where the annual dry and wet N input to the ecosystem N_{in} ($\text{kg N ha}^{-1} \text{ a}^{-1}$) was estimated to be approximately $25 \text{ kg N ha}^{-1} \text{ a}^{-1}$ by Hurkuck et al. (2014) at the same site, leading to a stomatal emission potential Γ_s of 634 mol mol^{-1} .

We will include this information by referring to Massad et al. (2010).

Page 7, line 15. “horizontal exchange with higher layers . . .” Do the authors mean vertical exchange?

Yes, thanks, this was a mistake. Changed to ‘...vertical exchange with higher layers...’.

Page 7, line 22 – 27. The authors note that they observed low air concentrations at high temperatures in late April/early May. They go on to suggest that this might be related to higher concentrations of acid gases and a tendency of NH₃ to shift to the particle phase. Do the DELTA measurements of HCl, SO₂, and HNO₃ support this statement? Were there particle measurements collected that might help shed some light on this question?

The DELTA measurements do not strongly support this statement as they always cover measurement periods of four weeks. Short occurrences of high concentrations of particles are not identifiable. Hurkuck et al., 2014, who were measuring at weekly time resolution in 2012 and 2013, recorded in the first year especially high HNO₃-concentrations in late April and early May, compared to February and March. So there might have been sufficient acid gas species to enhance the conversion from ammonia to ammonium.

The statement regarding volatilization of aerosol in the heated inlet line is a little confusing, as this would tend to bias the measured NH₃ high. Some clarification is needed here.

It was already mentioned on Page 4 lines 19 – 25, that we possibly overestimate the concentration because of this volatilization effect, but in the case of formation of acidic salts we would regain some of the ‘lost’ ammonia. This might be a little bit too confusing in this sentence, so we will leave this part out and write: ‘With higher temperature a larger amount of acidic gas or particle species are present in the atmosphere, which usually leads to reactions of ammonia to ammonium salts such as ammonium nitrate. (Kim et al., 2011)’

Page 7, discussion of diurnal variability. The authors discuss possible reasons for lower concentrations at night and for the morning increase in concentration. What might be driving the relatively rapid increase during the evening (1600 – 1800) as illustrated in figures 4 and 7?

The diurnal concentration pattern is generally highly influenced by the two periods with high concentration between 06 and 15 March and 30 March and 08 April. This can also be observed in Fig. 4, period II and III also exhibit this peak in contrast to the other periods. In these two periods the high concentrations are related to the northeast wind sector, where the next agricultural sources are closest (approx. 1.5 km distance). We separated the diurnal concentration into northeast (0 – 90°) and 90 – 360° wind sectors to illustrate that the late afternoon peak is mainly driven by local ammonia sources located northeast of the tower (see Fig. A1). Fig. A1 will be included in the supplementary material.

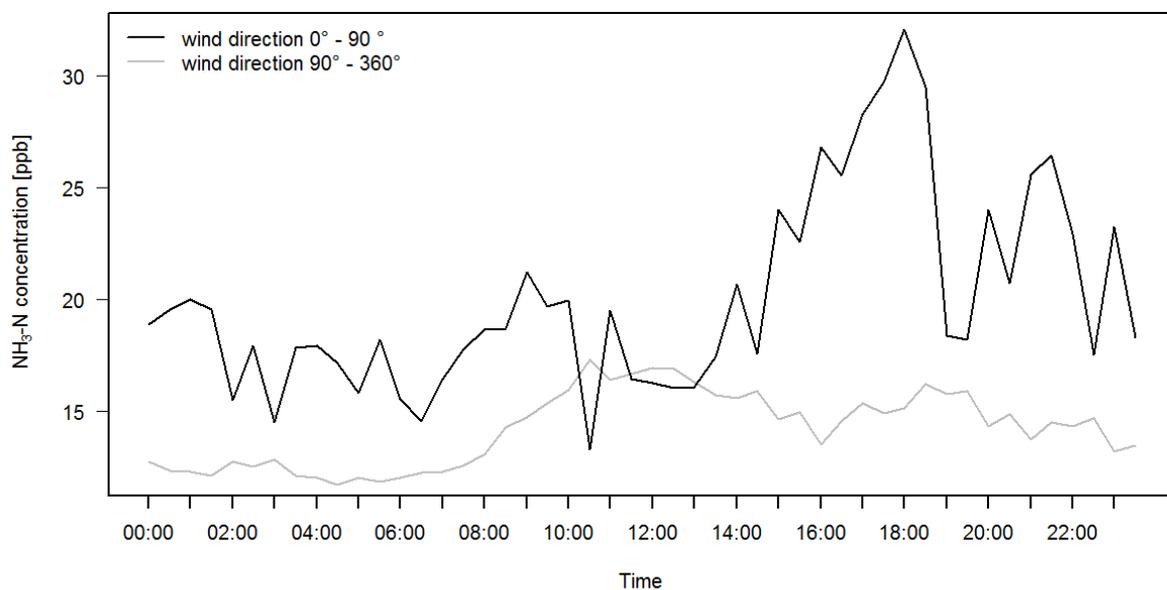


Fig. A1: Mean diurnal variation of ammonia concentrations separated by wind direction.

Page 8, line 20 – 23. The authors mention that emission was observed during rain events. Are the flux measurements valid during active precipitation? Assuming that the measurements are valid, what process would be driving the emission?

Page 9, line 9. “.. the ecosystem emits only under dry conditions in contrary to our observations..” The observation of emission during periods of rain or surface wetness is significant. Are there other published examples where this was observed in natural ecosystems?

To our knowledge this was the first time it was observed with high-resolution flux measurements. There are emissions as well as depositions during rain events in a range of -75 to 25 ng N m⁻² s⁻¹, with a median of -8.6 ng N m⁻² s⁻¹. The range is smaller compared to the residual data set, but the median was in a similar magnitude of -14.2 ng N m⁻² s⁻¹, so we suppose the data during precipitation to be valid. During precipitation-emission periods temperatures were moderate between 3 and 15 °C, concentrations relatively low between 3 and 11 ppb. The wind direction was mostly west to north, where very few point sources are located. So probably relatively unpolluted air reached the site and led to a gradient between the highly polluted area and the “clean” air, so that some emission occurred.

Page 10, statistical analysis (Table 2). To me, the statistical analysis does not help explain the patterns of the fluxes. The statistical tests indicate that the groups are different but do they differ in a way that is physically meaningful? What are the results telling the reader about the fluxes? How much of the variability in the fluxes can be explained by their relationships to these variables? If the authors wish to employ statistics to explain the flux patterns, a more well developed and rigorous analysis, which accounts for collinearity in the independent variables, is needed.

The statistical analysis was not supposed to explain the patterns of the fluxes as we focus on the methodology and these considerations were only used to show the plausibility of the flux data. We just wanted to know if there are significant influences by the meteorology. The results only tell us, that we cannot exclude the influence of the mentioned meteorological variables. As shown in Fig. 7 (manuscript) especially the diurnal variation of the fluxes and the concentrations appear to be controlled by temperature, net radiation and friction velocity but it is not possible to state to which extend the one or the other value contributes to the flux, because there are highly non-linear processes involved. Only mean-values show this effect because in addition the data is strongly scattered, therewith also some requirements for different statistical tests like for example normal distribution for ANOVA (so we chose the alternative Kruskal-Wallis test) are not met. An in depth analysis would be interesting but we are afraid that this would distract the reader from the main methodological focus of this work.

Page 10, line 16. “.. whereas peaks in concentration, although rather forming a bimodal pattern, follow the shape of the air temperature with a 2-3 hour lag”. The concentration pattern is very much bimodal, with the evening peak representing a process that is obviously important (as these are mean values) and uncorrelated with temperature. As in my previous comment, some discussion of the cause of this evening mode, which contains the highest concentrations observed diurnally, is warranted. It is too highly amplified and brief to represent boundary layer dynamics. Is it a persistent influence from local sources?

See above. We will add the necessary explanation concerning this peak.

Page 11, line 10. “whereas during emission periods it may be the ammonia flux itself, which is controlling the concentration . . .”. Does this make sense from a mass balance standpoint? Assuming some depth of the boundary layer and no advection, could the measured flux reproduce the observed air concentration?

We agree with the reviewer that from a mass balance point of view it might not be applicable, but it was also stated: ‘However, more likely is a coincidence of flux drivers and high concentration levels which were high due to advection from the local sources.’ Hence, we do not fully apply this statement from Milford et al. (2004) to our site conditions. We will include the following changes for clarification: ‘However, Milford’s (2004) statement that the concentration may still determine the flux during deposition periods, whereas during emission periods it may be the ammonia flux itself, which is controlling the concentration, is also likely be applicable for our site during deposition periods. In emission periods, a coincidence of flux drivers and high concentration levels, which were high due to advection from the local sources, is the more realistic reason for the relationship in Fig.10 (right panel).’

Page 11, line 12. “. . . high concentration levels due to advection from local source”. Have the authors considered advection as a potential source of error in the flux measurements themselves?

Advection affects in the first place concentration and thereby indirectly also the flux. We always had the local sources in mind, that’s why we mention it several times in the

manuscript. We will add at page 11 line 15: ‘The nearest agricultural ammonia point source was 1.5 km away from the tower. With a measurement height of 2.5 m none of the sources were located within the flux footprint, thus we can largely exclude effects from flux heterogeneity such as a direct contribution of the sources to the measured vertical fluxes. However, there might be still large scale transport processes as outlined in a study by Mohr et al. (2015) that influence ammonia concentrations at the site.’

Figure 10. Can the relationship between concentration and flux be used to derive an estimate of the surface compensation point? Regarding the increase in emissions with concentration, it seems a very large compensation point would be needed for the surface to continue emitting at an air concentration of 35 ppb. Some explanation of this feature of the plot would be helpful.

We agree with the reviewer that the site has to have a very large compensation point for these emissions to occur. There are generally two simple ways to derive the canopy compensation point from flux and concentration measurements: (i) Look for situations when the sign of the flux changes from deposition to emission or vice versa and assume the measured air NH_3 concentration at these times to be equal to the canopy compensation point, or (ii) invert the single-layer resistance-in-series analogue (details below).

(i) Without separating into emission and deposition and without bin-averaging, the points are too scattered to estimate any compensation point. We tried again to plot the fluxes against the concentration for a smaller flux range (-10 to $10 \text{ ng N m}^{-2}\text{s}^{-1}$) without bin-averaging (see figure A2 below), but again we cannot derive a compensation point from it, emission and deposition seems to be divided. Only for single days compensation points appear, but with poor R^2 . Fig. A2 will be included in the supplementary material.

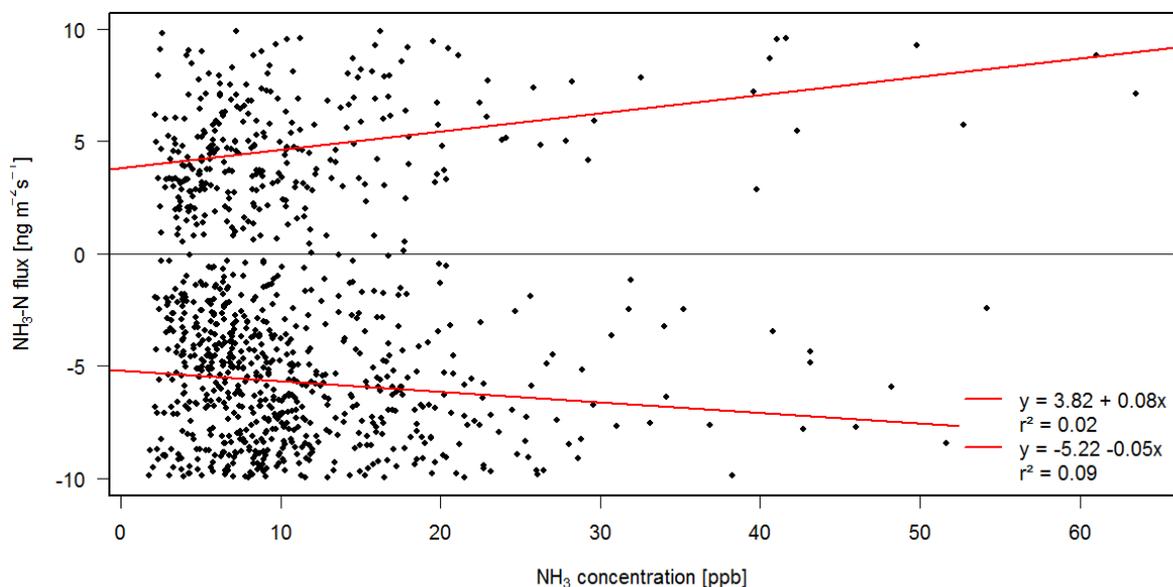


Fig. A2: Half-hourly scatter plot showing the dependency of NH_3 fluxes (only in a range of -10 to $10 \text{ ng N m}^{-2}\text{s}^{-1}$) on NH_3 concentration, red line: linear regression above for emission, below for deposition, for coefficients and r^2 see legend

(ii) We were able to derive a continuous time-series of the canopy compensation point, as proposed by Anonymous Referee 2, using the relation $\chi_c = F_t \cdot (R_a\{z - d\} + R_b) + \chi_a\{z - d\}$, where χ_c ($\mu\text{g m}^{-3}$) is the canopy compensation point at the notional height of trace gas

exchange z_0' (m), $\chi_a\{z-d\}$ ($\mu\text{g m}^{-3}$) the air NH_3 concentration measured at the aerodynamic reference height $z-d$ (m), F_t ($\mu\text{g m}^{-2} \text{s}^{-1}$) is the total NH_3 flux measured by the eddy covariance system, $R_a\{z-d\}$ (s m^{-1}) is the aerodynamic resistance at the reference height, and R_b (s m^{-1}) is the quasi-laminar resistance to NH_3 exchange. From this, we were also able to calculate the canopy emission potential as described by Anonymous Referee 2. The results indicate that indeed there appears to be a very large canopy compensation point that closely follows the air NH_3 concentration, which triggers emission events and effectively reduces deposition in the way that it prevents NH_3 from depositing at the maximum allowed deposition velocity allowed by turbulence ($v_{d,\text{max}} = (R_a + R_b)^{-1}$; Fig. A3, panel 1). In other words: In a unidirectional framework, this high canopy compensation point increases the effective canopy resistance R_c (Fig. A3, panel 2), and it appears to have a much larger influence on the observed fluxes than the atmospheric resistances. The constant (stomatal) emission potential from the Massad et al. (2010) model is much lower than the observed canopy emission potentials, and the stomatal compensation point is only a function of temperature, not of the ambient NH_3 concentration, which may be an indicator that, at this site, there is another, ambient concentration-dependent bidirectional pathway that is not being modelled (e.g. wet surfaces; as described, for example, by Burkhardt et al. (2009) for the case of leaf wetness).

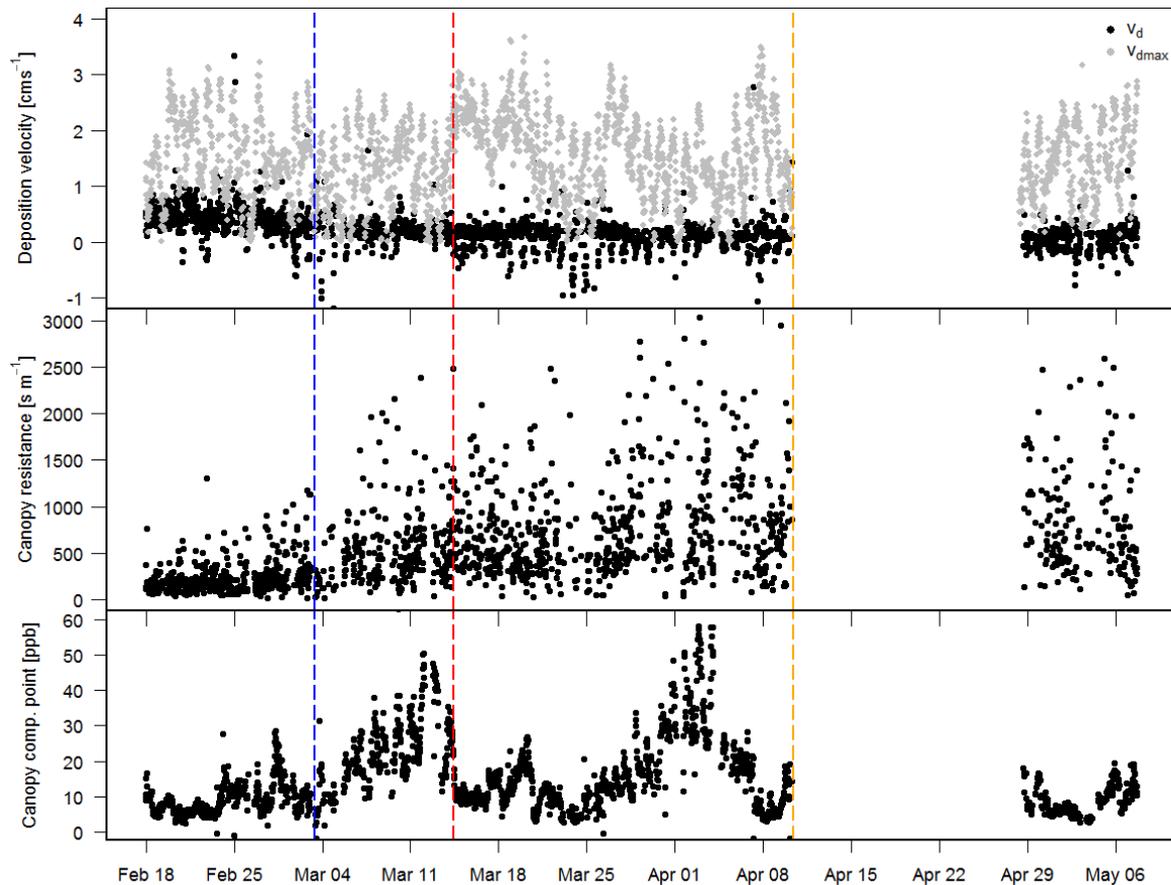


Fig. A3: (i) Deposition velocity, negative values indicate emission, and maximum deposition velocity, (ii) canopy resistance, negative values are not shown, as they are not defined in the resistance framework and mostly correspond to phases of emission, (iii) canopy compensation point, derived from measured data; this figure will be included in the manuscript

A comparison of method (i) and (ii) is not really possible. Loubet et al. (2012) showed that it is generally possible, but they restricted it to data under relatively dry conditions - a criterion,

which was never fulfilled at this peatland site during our campaign - and R^2 of the daily regressions not smaller than 0.5, for which this dataset was too scattered.

We will add figure A3 to our manuscript and include discussion on canopy resistance and the canopy compensation point.

Page 11, line 22. “.. the model does not exhibit any emission”. Some additional detail on the model parameterization is needed. What is the stomatal emission potential (γ) and how was it derived? Does the lack of emission in the model suggest that this parameter should be adjusted?

Regarding the stomatal emission potential parameterization, see above.

Regarding modelled emissions: There are in fact very few short periods where the stomatal compensation point is larger than the air ammonia concentration, however, due to the relatively high stomatal resistance during the measurement period, the emissions occurring under these conditions are insignificant in magnitude compared to the total flux, and especially compared to the measured emission flux densities.

We can derive a non-constant canopy emission potential as proposed by Referee 2 that appears to be linked to air ammonia concentrations. This is indeed not compatible with the Massad et al. (2010) model, but we cannot unambiguously attribute this to the stomatal pathway based on our data.

Page 11, discussion of measured versus modeled fluxes. The results shown in Figure 10 and 11 are difficult to reconcile at first read. The left hand panel of figure 10 shows larger deposition fluxes (more negative) estimated by the model, relative to the measurements, across the entire range of concentrations. The error bars mostly do not overlap. However, figure 11 shows a combination of model overestimation and underestimation of daily mean fluxes across periods. Some explanation of the differences in these plots would be helpful.

Thanks for pointing to these apparent contradictions. Looking at this again, we found in fact some wrong data used for the creation of the model regression line. Now we corrected this and there are no discrepancies anymore between figure 10 and 11 anymore (see Fig. A4). Thanks for discovering that!

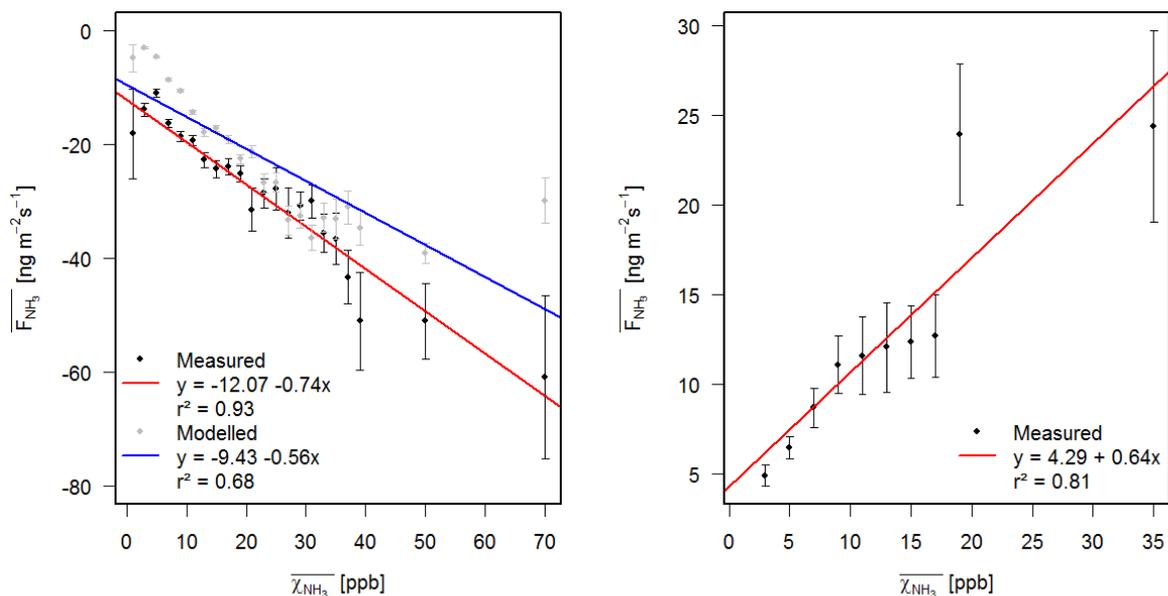


Fig. A4: New figure 10 in the manuscript.

Page 11, line 29. The authors note that some analysis of nighttime resistances was conducted which indicated that the R_w produced by the Massad et al scheme is too large for this site. What were the calculated resistances? This seems like a good opportunity to examine why the Massad et al R_w approach is not appropriate for this site. How low should R_w be to optimize the agreement with the measurements? Can R_w and the stomatal emission potential be changed together in a way that better reproduces deposition and emission? If so, does this inform not only the model improvement but also the processes that may be driving the fluxes?

We kindly refer the reader to another paper by Schrader et al. (2016, under review) which deals with this exact question and has just been published in ACPD under doi:10.5194/acp-2016-403. Quantitatively speaking, mean, median and geometric mean (calculated here to deal with zero-values as $(\exp(\frac{1}{n} \sum_{i=1}^n \log(R_w + 1))) - 1$) of observed nighttime R_c ($\approx R_w$, assuming that the stomata are mostly closed at night) were 606, 419 and 402 s m^{-1} , respectively, and mean, median and geometric mean modelled nighttime R_w were $> 10^4$, 497 and 1172 s m^{-1} . Note that we did not impose an upper value on modelled R_w , so median values are likely the most comparable quantity here.

We think the agreement of modelled and measured fluxes is overall quite satisfactory after mid-March 2014, which is also reflected in the comparison of R_w values (Fig. A5), considering the uncertainties in the measurements, the gap-filling procedure, and in some input parameters for the model. Simply decreasing minimum R_w leads to a better fit to the measurements in Periods I and II, but an increasing mismatch later in the time-series. Increasing the stomatal emission potential, even to unreasonably high levels, and decreasing the stomatal resistance to increase this pathway's contribution to the total flux does not lead to a significant improvement of the modelled flux estimates. Following Referee 2's suggestions on further calculations, we were able to derive a significant non-zero canopy emission potential from our measurements, which appears to follow air ammonia concentration levels. This observation is not compatible with the Massad et al. (2010) model, where the only emission potential is the stomatal emission potential, which is constant throughout the year and is set through its parameterization on annual N inputs.

We will add on page 12 line 2: 'A detailed investigation on R_w parameterizations can be found in Schrader et al. (2016, under review).'

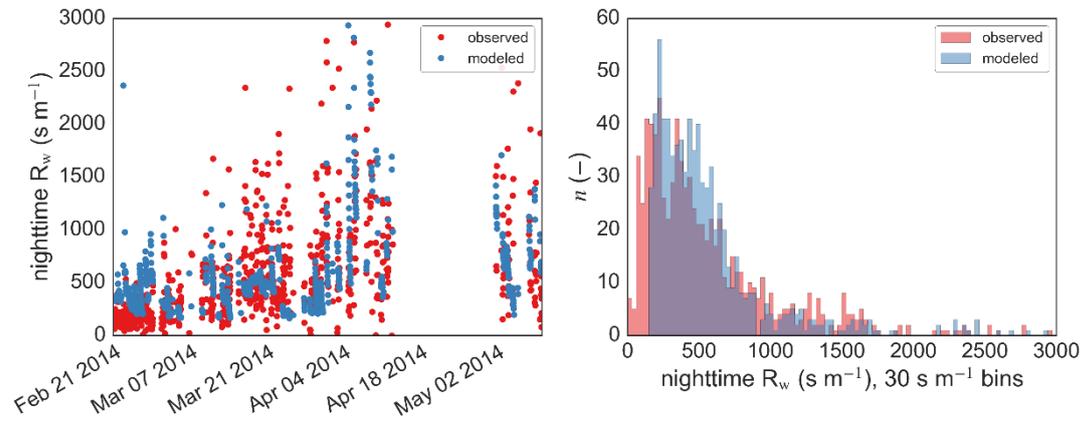


Fig. A5: Comparison of observed and modelled R_w during nighttime, when $R_a + R_b < 200 \text{ s m}^{-1}$, $u_x > 0.1 \text{ m s}^{-1}$ and $S_t < 10 \text{ W m}^{-2}$. Left panel: Time series of observed and modeled R_w , with a mismatch in the first two weeks due to the minimum R_w in the model. Right panel: Histograms of observed and modeled R_w with 30 s m^{-1} bins.

Answer to Referee 2 (Part B)

1

The quality of the flux measurements which is critical due to the novelty of the inlet system is however difficult to figure out completely for the reader. The way the time lag is calculated is not completely clear in the current manuscript, and the lag is an essential parameter in the flux which could change its magnitude by a large fraction, especially with noisy signals.

It would be good to show some covariance peaks and may be the dynamics of the lag (in a supplementary material section?).

In the discussion paper a short description of the lag determination was given on P5L9-14.

A strong drift between sonic anemometer acquisition system and QCL clocks was observed during the measurement period which led to drifts in the position of the maximum of the covariance function for the ammonia flux (see Fig. B1, approx. 3 s per day). In order to correct for this clock drift Figure B1 was divided into 9 subsections and a linear trend was fitted to the apparent time lag in each section. Then each half-hourly NH₃ time series was shifted with the fitted time lag resulting in near-zero time lags in the corrected datasets (Fig. B2). For the final flux calculation in EddyPro, we still had to set a range around zero for the time lag, in which the program again filtered the one with maximum covariance or took the default of 0. We will modify the text in the manuscript to clarify this procedure.

Also attached is Fig. B3 showing an example of covariance functions of vertical wind speed and temperature as well as vertical wind speed and NH₃ concentration. It will be included in the revised manuscript.

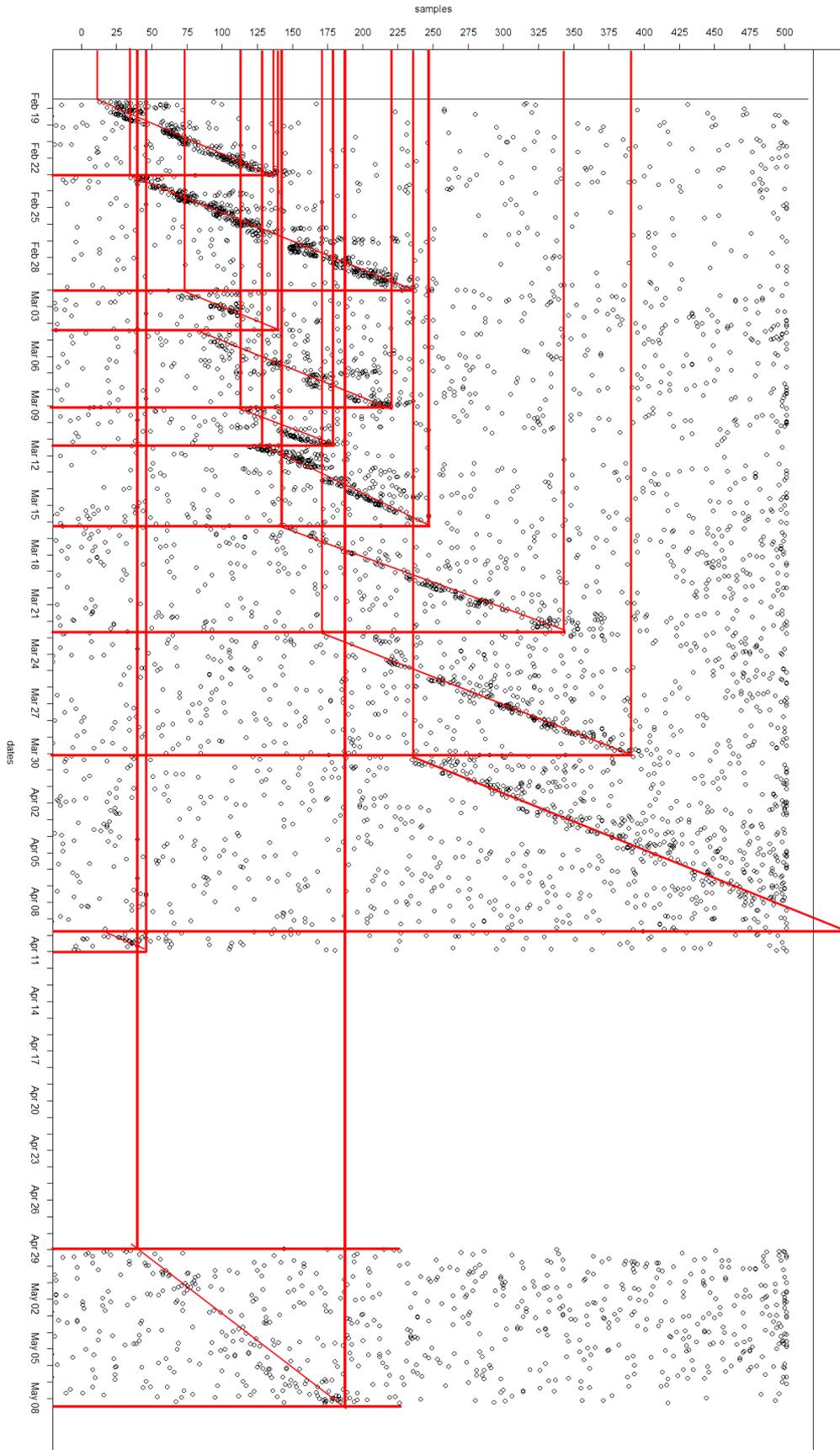


Fig. B2: Samples indicate the shift needed to maximize the covariance between vertical wind speed and NH_3 concentration for every 10 minute period. This shift includes the difference between the two system clocks of the sonic and the ammonia data acquisition systems. As the measurement frequency was 10 Hz, 200 samples equal 20 s delay time. Straight lines (red) were fitted to sections where the points accumulated.

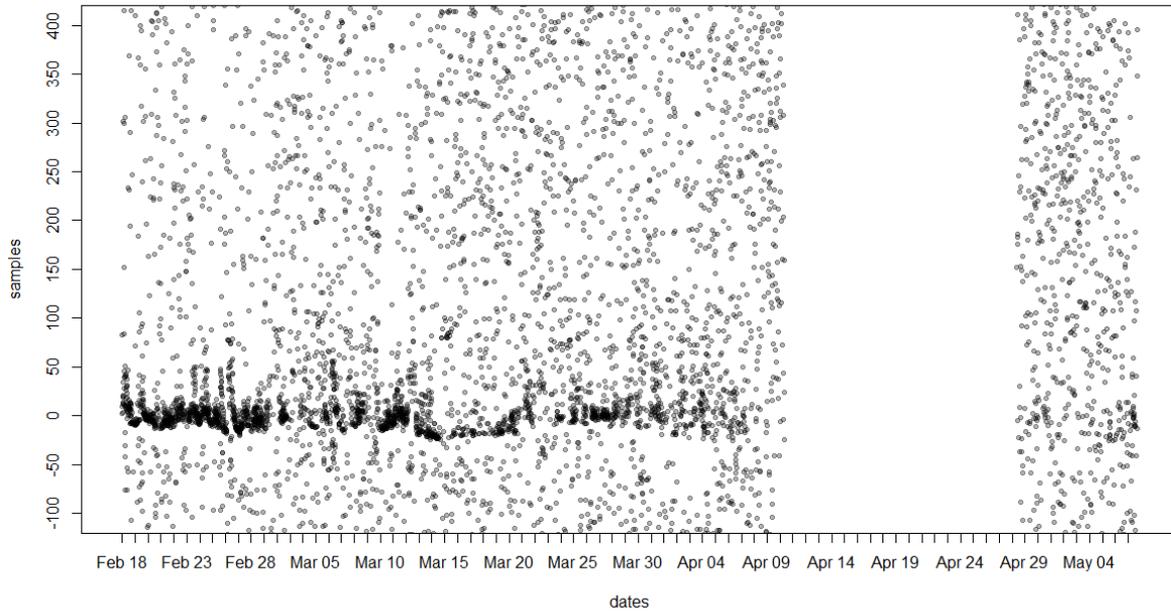


Fig. B3: Time series of time lags derived with covariance maximization after detrending.

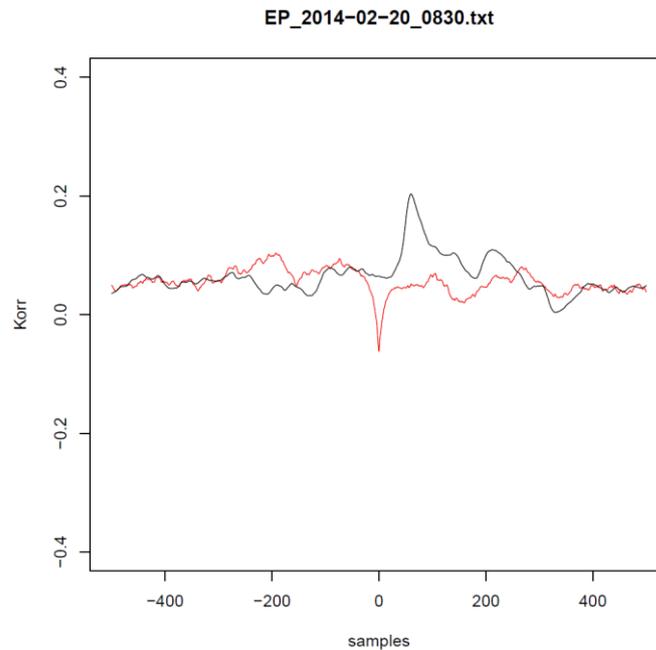


Fig. B4: Example for covariance functions, red line indicates $\overline{w'T'}$ and black line $\overline{w'CNH_3'}$

Similarly, a median value of the high frequency damping factor is given without much details, nor discussion and comparison to previous literature (Ferrara et al., 2012; Whitehead et al., 2008).

I would suggest showing the dynamics of the high frequency damping (it could be a box plot of hourly values for instance).

Ogives were calculated for every half hour. Since individual ogives (cumulative cospectra) of a 30 min-interval are very noisy and uncertain (even when smoothed over the frequency scale), it is generally necessary to perform a strict quality selection and averaging the resulting damping factors over certain periods or classes. Only cases with significant fluxes

(for NH_3 and T) and with fairly stationary conditions were selected. In the end we decided to apply the median damping factor on the complete dataset because we couldn't find a clear dependency on horizontal wind speed nor atmospheric stability (as already mentioned in the manuscript). We assume there might have been a dependency to relative humidity but unfortunately our measurement period covers only humidity data in a very small range (high RH values). We will add some illustration of the damping factor behaviour in the revised manuscript.

We agree to add in the text the comparison to Ferrara et al. (2012; 2016) who found damping factors similar to our value. For details on the ogive-method the reader is referred to Ammann et al. (2006).

The random uncertainty is also given as a 15% estimate, but there are some methods to evaluate the uncertainty in the flux, which are especially designed for fluxes with large instrumental noise. I would suggest to get example on Langford (2015), and to report flux detection limit. Some of these methods can be turned on in EddyPro, so this should not represent too much work.

Yes agreed, see answer to Referee 1 to Page 5, Section 2.3.

#2

The analysis of the correlation of the fluxes with the meteorological conditions is quite instructive but lacks a deeper insight into the surface exchange parameters. Indeed, a first essential test is a comparison of the deposition velocity $V_d(z)$ with the maximum deposition velocity for ammonia $V_{d\max}(z)$, which represents the maximum transfer rate and can simply be evaluated as $(R_a(z)+R_b\{\text{NH}_3\})^{-1}$, where $R_a(z)$ and $R_b\{\text{NH}_3\}$ are the aerodynamic and boundary layer resistances for ammonia, respectively(e.g. (Loubet et al., 2012)).

See Fig. A3 in Answer to Referee 1. This figure and the discussion around it will be included in the manuscript. The upper part shows the deposition velocity and the maximum deposition velocity. As we would expect, the latter is bigger than the deposition velocity, only in ten instances (0.2%) the maximum deposition velocity was smaller. These data will be excluded.

Similarly, analysing the statistics of the deposition velocity would probably give more insight into the exchange processes than the ammonia flux because of the large variability of the atmospheric concentration which is influenced by the local sources. An analysis of the daily variations of the deposition velocity would be very instructive. This would especially be helpful for understanding the links between Figure 9 and 10, which is not clear in the current manuscript.

We already had a deeper look at this very interesting point, however, diurnal deposition velocities didn't give more insight, and they showed nearly the same (inverse) pattern as the flux (see Fig. B4). The linear regression was also very similar, only the Kruskal-Wallis-test differed a little with respect to precipitation and the length of the time period after the last rain. Following also to other suggestions, Fig.8 and 10 will be rearranged anyway, thereby leading to some further insight into the plausibility of the flux with regard to the (maximum) deposition velocity, the canopy resistance and the canopy compensation point.

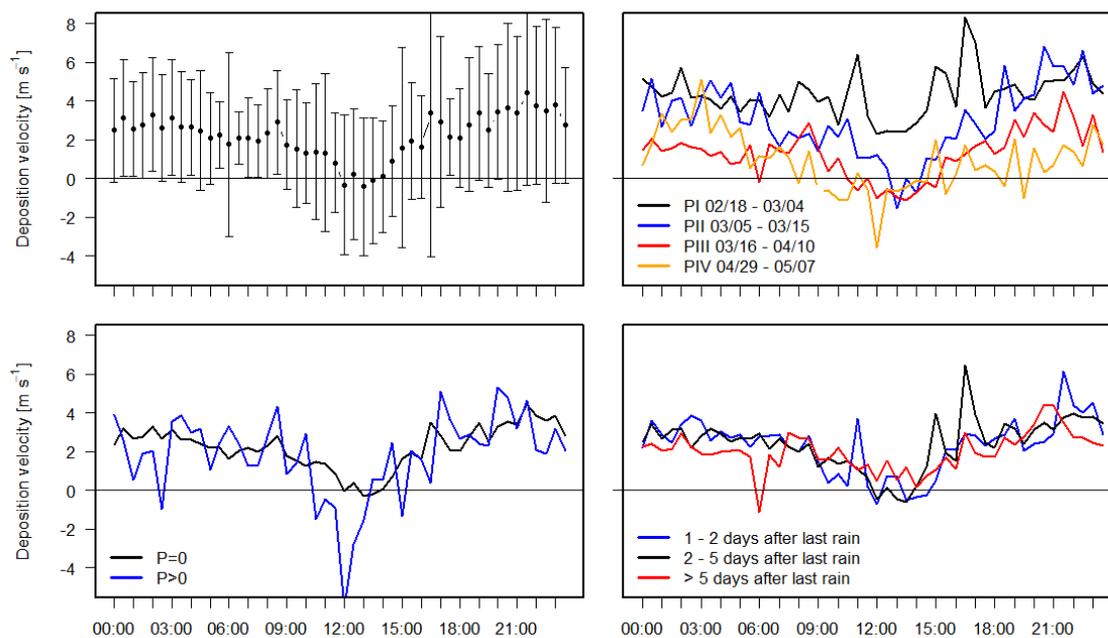


Fig. B4: Mean diurnal variation of ammonia deposition velocities, negative values indicate emission, with standard deviation (upper left panel), separated by periods (upper right panel), by precipitation (lower left panel), and by days after last rain (lower right panel).

3

The resistance analogy would also be very helpful to better evaluate the surface emission potential $\Gamma(z_0)$ and its dynamics. Indeed the canopy compensation point could be estimated as $C_c = F_{NH_3} \times (R_a(z) + R_b\{NH_3\}) + C_{NH_3}(z)$, and the emission potential retrieved from that using the thermodynamical gas-to-liquid and acid-base equilibrium constants. See Sutton et al. (2009), Loubet et al. (2012) or Personne et al. (2015). The compensation point could be also estimated by analysing daily flux versus concentration relationships (similar to Figure 10 but for each day). This would ease a lot the understanding of the seasonal evolution of the ammonia flux, and its relationship with ecosystem functioning. It will as well help controlling the quality of the flux measurements.

See Fig. A3 and discussion to Figure 10 in Answer to Referee 1.

4

In a complementary analysis, surface conductance (g_c , the inverse of the resistance R_c) could be estimated (assuming a zero emission potential in the canopy under deposition conditions) based on $V_d(z)$ and $V_{dmax}(z)$. Indeed, then $g_c - 1 = V_d(z) - 1 - V_{dmax}(z) - 1$ (Massad et al., 2010). This would withdraw part of the influence of u^* on the exchange dynamics, which is embedded in $V_d(z)$, and hence better show changes in ecosystem exchange parameters, and especially cuticular exchange.

We have looked at surface conductances g_c (see Fig. B5, below), but found that R_c was a better basis for the discussion (See #2 and Fig. A3 in Answer to Referee 1).

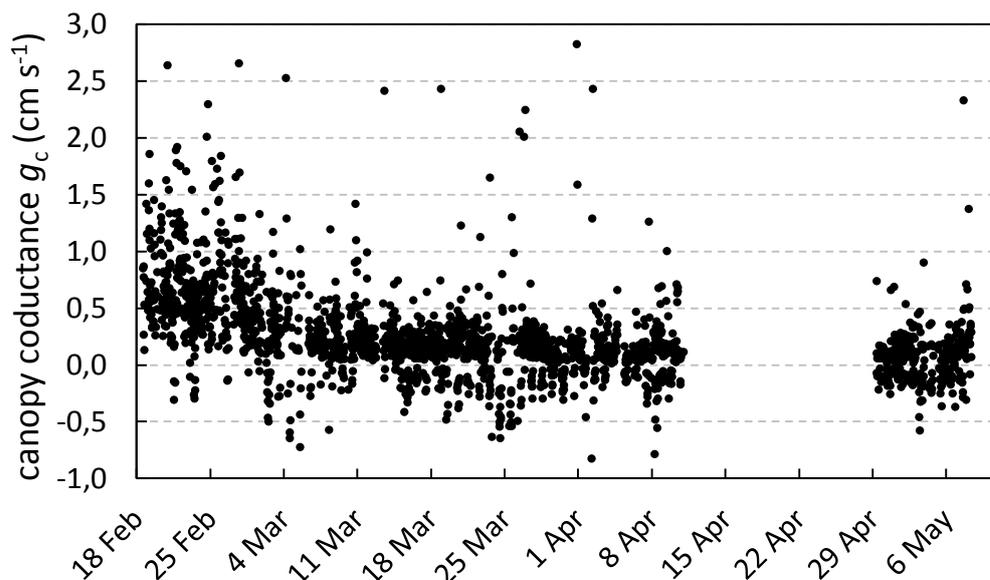


Fig. B5: Canopy conductance

5

Figure 10 puzzles me for several reasons, but I might not have understood correctly how it was built:

(1) I cannot figure out why the modelled flux is smaller than the measured one with a constant offset (in Fig 10-right), while it shows larger values in Figure 11 at the beginning. I also interpret a constant offset as an additional pathway with a constant flux, but cannot reconcile this with the model of Massad et al.(2010) as used here.
 (2) I cannot understand why no intercept (flux crossing the zero line) can be seen in Figure 10(left) while in Fig 8 we see negative $V_d(z)$. It is probably due to the separation between emission (Fig10. Right) and deposition (Fig10. Left) fluxes. The authors should clarify how Fig. 10 is constructed. Especially important would be to show some dynamics of the daily flux and concentration with emissions. Currently only averages are shown (except for $V_d(z)$) and no emissions can be seen except in Fig. 6c and in the error bars of Fig 6a. An example of daily dynamics would be most helpful.

(1) Unfortunately there were some wrong data used for the creation of the model regression line. Now there are no discrepancies anymore between figure 10 and 11 anymore. See Fig. A4 in Answer to Referee 1.

(2) There is no intercept due to the separation into emission and deposition fluxes. More on this topic in Answer to Referee 1, discussion on Figure 10.

Emissions can also be seen in Fig. 3 in the manuscript. Daily dynamics are displayed as mean diurnal courses in Fig. 4 and 6. Additionally we provide a section of one example week for this answer to the review and in the supplementary material: Fig. B8.

6

The authors should also discuss further, based on more quantified surface parameters, whether the flux is linked to a surface compensation point or some other features.

Yes, we will discuss this in the revised manuscript, see Fig. A3 and discussion to Figure 10 in Answer to Referee 1.

Especially, the magnitude of the advection fluxes should be evaluated. Indeed, if the NH₃ concentration peaks of up to 85 ppb are due to concentration advected from nearby farms and agricultural activities, advection fluxes can be expected to be large. To evaluate these a footprint model could be used, the advection fluxes would then be the footprint of the farm (or fields spread with organic manure) multiplied by the source strength of these, which could be evaluated by a simple emission factor analysis. This would allow evaluating whether advection is an issue or not. I would suggest the authors to look at (Hensen et al., 2009; Loubet et al., 2009).

Yes, we will discuss this in the revised manuscript, see Answer to Referee 1, discussion on Page 11, line 12.

Detailed comments

• Section 2.1: The ground pH and NH₄⁺ are important parameters for interpreting NH₃ fluxes. Were any of these measured? If so, they should be reported.

They were unfortunately not explicitly measured in this study and therefore not included in the manuscript, but we would expect a pH of 3.5 to 4.5.

• P3L29: The authors should rather use “mixing ratio” rather than concentration. Also are these expressed per mol of dry air or per mol of ambient air? Please discuss this point as this makes a difference in the flux calculation which should be done with mixing ratio per mol of dry air (Gu et al., 2012; Kowalski and Serrano-Ortiz, 2007). Especially important is the dilution effect due to water.

We will add a statement in the method section that it is actually a mixing ratio (relative to dry air) but we will keep the term concentration, which is recorded as mol NH₃ per mol dry air. The QCL system automatically corrects for water vapor dilution in the air sample and outputs mol NH₃ per mol dry air. We will mention that at this point.

• P4L30. What is the inlet box size? Could you discuss briefly the potential impact on the flux measurements?

Inlet Box size: 39 cm x 34 cm x 21 cm. We are aware that this box right next to the sonic anemometer is a source of disturbance. We tried to find a compromise between sensor separation and unhindered inflow. That's why the box is mounted below the sonic anemometer at the north side, a less common wind direction at this site (5.6% of the campaign), so only few data was influenced. We will add this information to the Methods Section.

• P5L7-12: The way the two timeseries were shift is not sufficiently detailed here. Especially, could the authors explain how the expected time lag was chosen? Also, it would be important to show that this procedure did not strongly affect the flux. Could the authors please discuss this point further? The authors may consider adding a graph showing several covariance peaks for emission and deposition conditions.

Please see answer to # 1, part 1 (starting at page 10).

• P5L15-20. Please give more details on the damping factor and how it evolved during the campaign. Some additional graphs could be proposed in a supplementary material section.

Please see answer to # 1, part 2 (starting at page 12).

- **P5L20-23. The uncertainty on the flux is critical for NH₃ which is not a routine measurement. I suggest taking example on (Langford et al., 2015) and related references for computing the error on the flux and evaluating the flux detection limit.**

Please see answer to # 1, part 2 (starting at page 12).

- **P5L29: Clarify if gap-filling was also performed for NH₃ and if so how.**

Gap-filling was performed. The chosen method was mean-diurnal-variation (MDV, window size was +/- 5 days) (Falge et al., 2001; Moffat et al., 2007). This information can be found at P5L30 in the manuscript.

- **P6L6: Be careful that Ra is a function of the measurement height z. Consider using the notation Ra(z).**

Thanks, we will use $R_a(z-d)$.

- **P6L7 - 12: The parameters of the Wesely model should be given here: the minimum resistance and the response to radiation.**

We used the original formulation of the Wesely (1989) model with a minimum R_s for H₂O of 200 s m⁻¹, scaled by the ratio of the molecular diffusivities of H₂O and NH₃:

$$R_s = 200 \cdot \frac{D_{H_2O}}{D_{NH_3}} \cdot \left(1 + \left(\frac{200}{S_t + 0.1} \right)^2 \right) \frac{400}{T \cdot (T - 40)}$$

where S_t is global radiation in W m⁻², D_x is the molecular diffusivity of H₂O and NH₃ in air, respectively, in m² s⁻¹, and T is the surface temperature in °C. Note that we were not able to optimize these parameters due to a lack of data in the dry range, where cuticular deposition is restricted, so this pathway does indeed have some uncertainty. A +/- 100 % change in the minimum stomatal resistance leads to a change in total cumulative flux between -7 % and +19 % (for 300 s m⁻¹ and 100 s m⁻¹, respectively). We will add '... with a minimum stomatal resistance of 200 s m⁻¹, ...' on Page 6, Line 8. Further information and equation will be shown in supplementary material.

- **Section 3.1: Since the local farms and agricultural fields play an important in the interpretation of the mixing ratios.**

- **P7L27-28: This sentence is unclear. Please rephrase.**

We assume you mean the sentence before, as also Referee 1 got confused. We will leave the last part out and only write: 'With higher temperature a larger amount of acidic gas or particle species are present in the atmosphere and usually leads to reactions of ammonia to ammonium salts such as ammonium nitrate. (Kim et al., 2011)'

- **P7L33-34: The work of Flechard et al. (1999), Wu et al. (2009), and Burkhardt et al. (2009) should be mentioned here.**

Very good point, these references will be included.

- **P9L1-5. Could you be more quantitative here? Are the levels comparable with Duyzer (1994)? What is the amount of NH₃ received in this study? What would be the**

ecosystem compensation point predicted by Massad et al. (2010) with this deposition? Please also discuss this issue with reference to Wu et al. and Burkhardt et al.

Duyzer et al. (1994) reported $16 \text{ kg NH}_3 \text{ ha}^{-1} \text{ yr}^{-1}$, so they found even more than 2.5 times more than we did in our study. We are not sure what is meant by ecosystem compensation point, but we assume you mean the stomatal emission potential

$$\Gamma_s = 246 + 0.0041 \cdot N_{in}^{3.56}$$

We can make a rough estimate about the N_{in} : Duyzer et al. (1994) report dry deposition fluxes of $16 \text{ kg NH}_3 \text{ ha}^{-1} \text{ yr}^{-1}$ and $3.8 \text{ kg NH}_4^+ \text{ ha}^{-1} \text{ yr}^{-1}$, corresponding to an N input by dry deposition of reduced nitrogen of around $20 \text{ kg N ha}^{-1} \text{ yr}^{-1}$. They also state that the input by dry deposition is roughly 50 % of wet deposition, so total reduced nitrogen input would be roughly $50 \text{ kg NH}_3 \text{ ha}^{-1} \text{ yr}^{-1}$. Assuming that reduced nitrogen deposition is the major pathway of N_r deposition at their site, this translates into a stomatal emission potential of at least $4828 \text{ mol mol}^{-1}$ using the Massad model (likely higher due to deposition of other N_r species) or a stomatal compensation point of around $10 \mu\text{g m}^{-3}$ at $15 \text{ }^\circ\text{C}$, whereas at our site total N-input was estimated to be $25 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ by Hurkuck et al. (2014), leading to a stomatal emission potential of 634 mol mol^{-1} or a stomatal compensation point of $1.4 \mu\text{g m}^{-3}$ at $15 \text{ }^\circ\text{C}$. Note that Duyzer et al. (1994) estimated the average (canopy) compensation point at their site to be $0.4 \mu\text{g m}^{-3}$.

We will add the reported deposition rates by Duyzer et al. (1994) in the manuscript. Since we are not completely sure what was meant by ecosystem compensation point, we cannot compare values from the suggested authors with it (we also couldn't find these term in their references). We will include these references in the section about surface wetness on page 7 line 33. The same dataset that was used in Wu et al. (2009) was also discussed in Walker et al. (2006) and is already discussed in the manuscript.

• P9L12: It is difficult to see a change on Fig. 8. Please consider re-graphing this figure.

Fig. 8 from the manuscript was re-graphed (Fig. A3) but not to emphasize the change or the tipping point. We will mention in the text that in this depiction the tipping point isn't visible.

• P9L17: A shift of the stomatal compensation point could be evaluated by retrieving the daily compensation point as explained in the general comments. Two methods are possible:

We tried both methods, please see Fig. A3 and discussion in Answer to Referee 1.

• P9L19-20: The data on wet to dry deposition are not shown here. Please consider adding these to the supplementary material or at least giving numbers to support the sentence.

Unfortunately we have only monthly wet deposition, so we cannot directly link the higher wet deposition to 15 March, so we should rephrase the sentence:

“Similar to findings in Kim et al. (2011), precipitation after 15 March significantly reduced ammonia concentrations in ambient air while probably the ratio of wet to dry deposition increased.”

• P9L21-33 and P10L1-5: Showing the maximal exchange velocity $V_{dmax}(z)=(R_a(z)+R_b\{NH_3\})^{-1}$ would be important to show the plausibility of the flux. Moreover, you can then calculate the canopy resistance R_c or the canopy conductance g_c as $g_c-1=V_d(z)-1-V_{dmax}(z)-1$, during deposition periods (especially at the start of the campaign). This would probably better show the dynamics of the

ecosystem exchange parameters, together the surface emission potential $\Gamma(z_0)$ which could be estimated from the canopy compensation point $C_c(z_0) = F_{NH_3} \times (R_a(z) + R_b\{NH_3\}) + C_{NH_3}(z)$, with the relationship $C_c(z_0) = \Gamma(z_0) \times 10^{-3.4362 + 0.0508T\{z_0 \text{ in } ^\circ\text{C}\}}$. This will probably help understanding the surface exchange dynamics and also test the plausibility of the flux and concentration measurement as C_c should remain positive.

Please see Fig. A3 and discussion to Figure 10 in Answer to Referee 1.

- **P10:** This study on the parameters influencing the ammonia exchange would benefit from being made on the deposition velocity which would less depend on the variable atmospheric concentration.

Please see answer to # 2 (starting at page 13).

P10L7: explicit the term α .

α is the significance level, that is typically 0.05, which was also chosen in this study.

- **P10L28:** V_d would be indeed good to show together with the flux!

Time series of v_d together with the flux will be shown in a supplementary section, see Fig. B6.

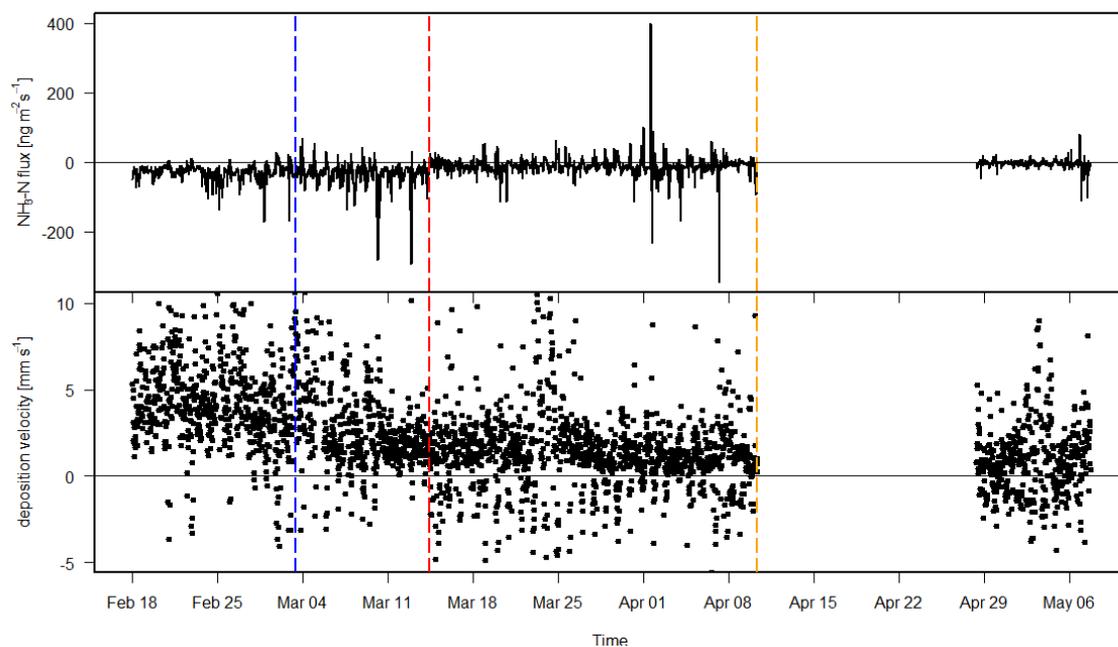


Fig. B6: Half-hourly ammonia fluxes (upper panel) and half-hourly ammonia deposition velocities (lower panel) during the whole campaign.

- **P10L29-33:** The daily evolution of the flux with decreasing deposition or even small emission around noon and deposition at night could be a consequence of a stomatal or ground compensation point which evolves following the daily surface temperature ($T(z_0)$) pattern and is much larger at noon than during the night. This explanation also reconciles the observed dependency of the flux to u^* , observed both during day and night (Fig. 9): indeed, the surface temperature $T(z_0)$ will increase with increasing u^* at night with clear sky due to better mixing and hence less radiative cooling. During the

day, the increase of surface temperature is mostly linked with incoming solar radiation and peaks at the same time as u^* .

This is a very good point and we will integrate this in our discussion.

• **P11L2-4: Indeed. $V_{\max}(z)$ is a measure of this exchange velocity and comparing $V_d(z)$ with $V_{\max}(z)$ would ease this discussion.**

Together with the new figure for v_d this will be included. See also Fig. A3 and discussion to Figure 10 in Answer to Referee 1.

• **P11L5-15; I cannot figure out how to interpret this Figure. I would suggest the authors to try showing the same relationship without separating emissions and depositions to show whether there is or not a compensation point and try to evaluate it. It would be interesting to try to build a figure for some representative days based on 30-min data and see whether the change from deposition to emission appears at a given concentration (the definition of the compensation point).**

Please see discussion to Figure 10 in Answer to Referee 1.

• **I can imagine that the advection indeed could indeed lead to such dependency. But it would then be interesting to evaluate the potential for advection based on a simple footprint model such as Kormann and Meixner (2001), which is available as an excel spreadsheet (Neftel et al., 2008). If you just multiply the value of footprint of the surrounding agricultural field or farms by an estimated magnitude of the NH_3 fluxes there, you could evaluate the potential effect of the advection on your flux. See also in Loubet et al. (2009) and in Sutton et al.(2009) for a discussion on advection.**

Yes, we will discuss this in the revised manuscript, see Answer to Referee 1, discussion on Page 11, line 12.

• **Section 3.3: Indeed, it is likely that the cuticular resistance may be overestimated in Massad et al. (2010), as was also found by Loubet et al (2012) and Personne et al. (2015). However I cannot figure out how to reconcile Fig 10 which shows globally larger deposition fluxes by the model and Fig 11 which shows larger measured deposition fluxes overall. This probably comes from the averaging procedure which splits emissions and deposition in Fig. 10.**

Please see answer to # 5 (page 15).

• **Moreover it would be very instructive here to have comparison of daily dynamics of the modelled and measured flux. This would also allow testing hypothesis with the model, as for instance diminishing R_w , or adding a compensation point and compare to the observations. Fig. 6 could for instance be duplicated and compared with the model flux or alternatively some example days could be chosen.**

• **P11L29-30: As mentioned above you should show the inferred R_w from night time measurements together with R_a and R_b or alternatively $V_{\max}(z)$ and g_c .**

Mean diurnal fluxes, measured and modelled, are shown in Fig. B7. The first 9 hours match very well but then the maximum of the modelled flux is lower and delayed compared to the measured flux. The model is not able to reproduce the emission peak during midday and predicts stronger deposition, limited by the $R_{w,\min}$ set in the Massad-Model. For one week of half-hourly fluxes, measured and modelled see below (Fig. B8). For measured canopy compensation points see Fig. A3, for R_w Fig. A5. For more investigation on the model the

reader is referred to Schrader et al. (in review, 2016). Fig. B8 will be shown in the supplementary section.

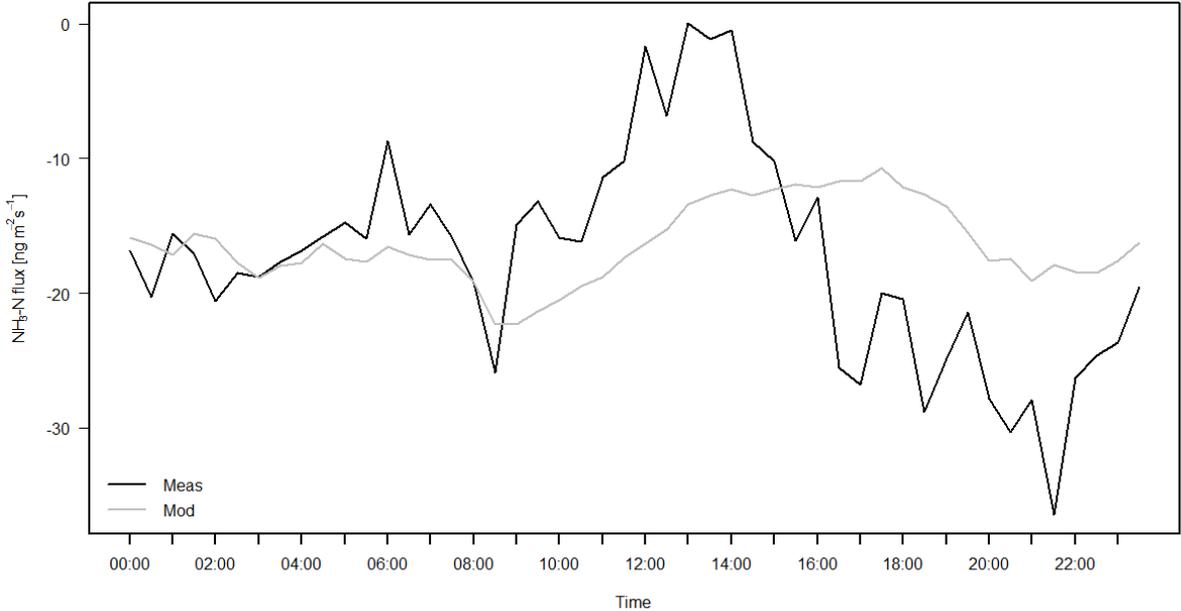


Fig. 7: Mean diurnal variation of measured and modelled flux over the whole campaign.

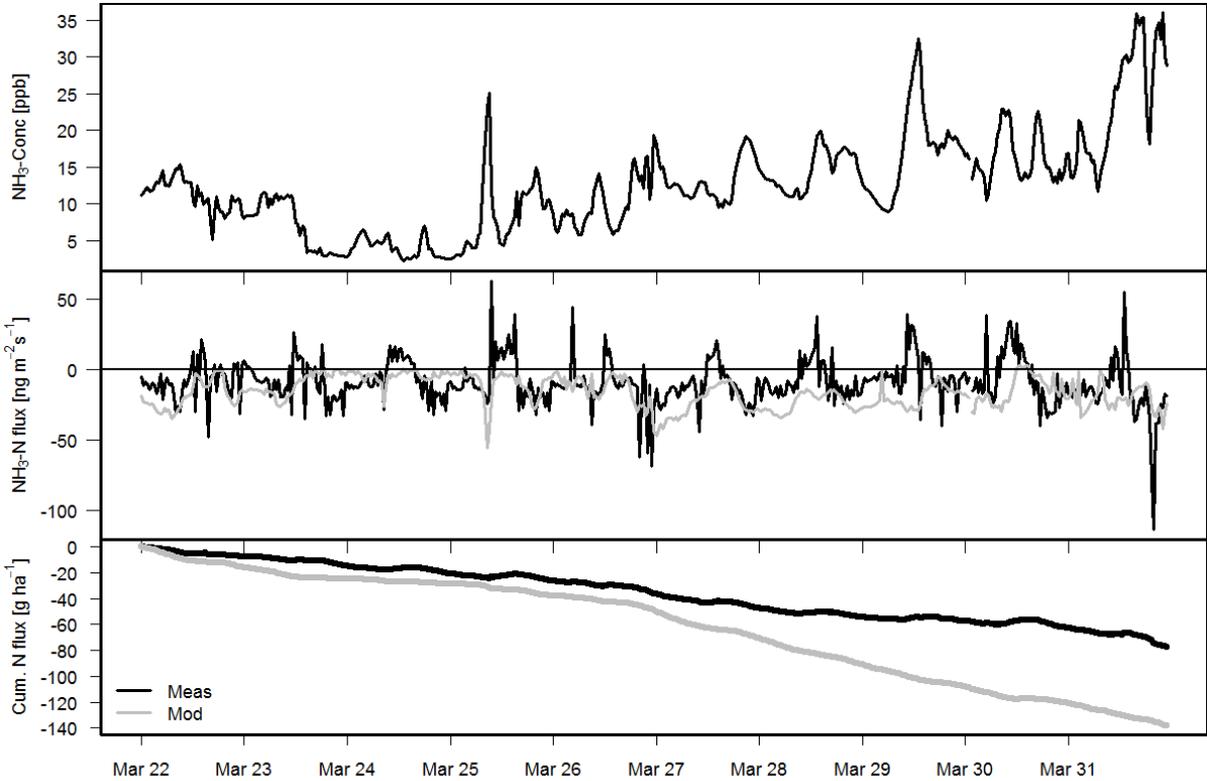


Fig. 5: Measured ammonia concentrations (upper panel), comparison of measured and modeled half-hourly ammonia fluxes (middle panel) and cumulative ammonia flux (lower panel) based on half-hourly data during one week of the measurement campaign.

- **P12L1-2: Please show data from the Delta denuders (in a supplementary section?) or at least give range of concentrations.**

On P8L5 values of the only comparable periods are given and also the NH_4^+ to NH_3 ratio is given on P4L21.

- **P12L8-9: Please explicit how you extrapolated to the entire year?**

Assuming a stable exchange rate during the whole year, we extrapolated the nine weeks ammonia exchange to an entire year, so it was a simple time integration.

‘Extrapolating our measurement-based campaign total time proportionally to an entire year, under the assumption of stable exchange rates, results in a net deposition estimate of approx. $5.3 \text{ kg NH}_3\text{-N ha}^{-1}$.’ Of course we will never observe a stable exchange rate during the whole year, due to climatic variation during the year and especially due to the many sources around. This number is only for getting an idea of the dimension of the annual exchange rate.

- **P13L2: You mention in the conclusion the long term stability of the QCL but none was said about it in the manuscript. Either consider withdrawing from the conclusions or add a discussion in the manuscript.**

We only measured for 3 month including a huge gap, so we didn’t really measure long-term and therefore we didn’t write anything like that in the manuscript. There were a lot of difficulties that need to be solved in the future like temperature stability in the QCL-Box, humidity, cleaning of the inlet and the laser cell and pump maintenance, so we cannot give any long-term prognosis. We currently use the instrument in a 2-yr campaign at a forest site with very low N input and continuously work on solutions for the above-mentioned issues. The sentence – as it is written – can be understood as an outlook, not necessarily as a result.

Tables

- **Table 1. Table and Figure legend should be self-standing: Please explain what are c_{NH_3} , T_a , P, R_n , SD and what the overbars mean. It may also be useful for interpreting graphs (Fig. 6) or statistics to include the number of points per period.**

We will add the explanation in brackets in the title:

“Characterization for four subperiods of the measurement campaign (I to IV) with different NH_3 concentration (c_{NH_3} , mean, standard deviation, maximum and minimum)

and weather regimes (mean air temperature \bar{T}_a , sum of precipitation P and net radiation R_n).”

We can also add a column with the number of points.

- **Table 2. Explain on what variable was the Kruskal-Wallis test made (the NH_3 flux?). It would also be very helpful to do this test on the deposition velocity. But the authors could also consider doing it on the canopy compensation point $C_c(z_0)$, the emission potential $\Gamma(z_0)$ or the canopy conductance g_c . Please also explicit what are “p-value” and “Post-Hoc” in the legend Figures**

The test was made on the ammonia flux. We will add that and an explanation for p-value (probability value) and Post-Hoc (Post-Hoc-Test result) in the title. We tried different analyses also for the deposition velocity (see above). We will add a similar table for the other variables to the supplementary material.

- **Throughout the text and legends change concentration for mixing ratio.**

See above.

- **Consider adding a Figure with the map of the field and the surrounding including farms and agricultural fields**

We refer the reader to Hurkuck et al. (2014) for an aerial photograph.

- **Figure 1. It would be helpful to add the heights and the tube length. Also explain or show how are the bypass and “Particles out” channels connected to the pump. Consider also adding the pressure and flow rates on the Scheme. Explicit AC and QCL in the legend.**

We agree to add some more information in the description below. There is one additional tube from the pump to the inlet box, in the box the bypass air and the particles out section are combined and together sucked to the pump. We will add a modified overview in the manuscript.

- **Figure 2. I would suggest showing the fitted co-ogive and show explicitly how the frequency damping is evaluated, like in Ammann (2006). Also adding a graph which shows how this frequency damping evolves with time would be very useful. The temperature co-ogive does not see to stabilise completely to 1 at large frequency (we expect from the graph that it may continue growing a bit at larger frequency). Please comment on that in the text.**

Please see #1 section 2 (page 12). For an explicit description of the applied ogive method we refer to Ammann et al. (2006).

- **Figure 3. This is a very nice graph. It would be helpful to add legend on the right hand side.**

After giving it quite some thought, we have no idea what the content of a legend in this figure could be as we think the figure is self-explanatory. Or do you mean to add axes labels on the right hand side? We can certainly do that.

- **Figure 5. Why splitting the wind direction in periods and not the NH₃ mixing ratio windrose?**

Here the map of the surrounding would be much needed to help understanding the NH₃ wind rose.

Period 4 is then not visible for example (because the concentrations are very low compared to the other periods) and the whole right diagram gets very unclear.

- **Figure 7. How do you explain the afternoon peak in NH₃ mixing ratio. May be I missed it in the text.**

See Answer to Referee 1 to Page 7, discussion of diurnal variability.

- **Figure 8. This figure is hard to read. Please consider using lines, a smaller height for the graph and also consider showing additional graphs (as for example in Langford et al. (2009)) of the main drivers (u^* , T_a , RH , R_n ,...). This would ease the discussion and help the reader making his mind on the dataset. Also very important in Figure 8 is to add on the same graph window $V_{max}(z)$.**

See above answer to P9L12 and new Fig. 8 shown in Answer to Referee 1 Fig. A3. When writing the manuscript we considered adding a plot of meteorological variables, but the campaign is too long to show all the data in one graph that clearly visualizes any effects of the driving flux variables. Therefore we decided to show only mean values.

• **Figure 9. Consider adding $R_n > 20$, $R_n < 20$ and all as legends of the graphs on the graphs themselves. What are the percentage meaning on the top of the graph? Also consider making a similar graph for $V_d(z)$.**

We already had the suggestion from a co-author to do the same figure with v_d , but it didn't change the insights, so we decided to keep the plot with fluxes. The percentages are explained in the figure descriptions: 'Values in upper row specify percentages of data in the respective u.-category given below.' We will add the suggested legend.

• **Figure 10. This figure needs clarification, and it might be better not to separate emissions and depositions periods. One would expect a compensation point to appear then. Also consider showing half-hourly data instead of pooling. Moreover one would expect pooling to also give horizontal overbars. Also consider showing specific example for one some days with different behaviour: I would expect Period 1 to be like actual Fig 10 left but period three to show a compensation point.**

See discussion to Figure 10 in Answer to Referee 1. We will change the axis labels to letters with overbars to clarify that these values are mean values.

• **Figure 11. Before showing daily averages, it would be good to show daily variations. This could be done over a shorter period or using averages as in Figure 6. Consider showing these in the supplementary material.**

See above. Fig. B8.

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