bg-2018-86

Author response to comments of referee #2

We'd like to thank reviewer #2 for his answer and appreciate his valuable comments.

(referee comments are printed in *italic*, author responses are printed in blue)

1. The methodology used for gap-filling emissions during night-time (and to a lesser extent for wind sectors coming from the surrounding farms) may be questionable. Indeed, the authors assume that night-time fluxes may be gap-filled based on day-time fluxes, but ammonia fluxes are fundamentally based on thermodynamical equilibrium at the surface (gas-liquid and acid-base equilibriums). This means that (1) the surface ammonia concentration is exponentially increasing with surface temperature due to the gas-liquid or Henry equilibrium, and we would hence expect lower emission at night due to lower temperature and hence lower concentration at the surface; (2) similarly, since ammonia fluxes are proportional to a concentration difference between the surface and a reference level, lower turbulent exchanges at night are expected to decrease night-time ammonia emissions. This means that using daytime fluxes amplitude and dynamics may systematically bias the emissions towards higher values. I would recommend a discussion on that point which may include a study on the temperature and u dependency of the ammonia emissions. Authors could refer to e.g. Flechard et al. (2013) for details on ammonia the points raised above.*

We fully agree with the reviewer that the ammonia emission depends on temperature and turbulence intensity and therefore is generally lower during nighttime compared to daytime conditions. We show this effect for the present study in Fig. 4a. We also account for the day-night difference in the gap filling procedure, but there was obviously a misinterpretation by the referee in this respect. Actually, we did not use the daytime fluxes amplitude to gap fill the missing night time fluxes. We only used the shape of the daytime curve (linear increase during grazing and exponential decrease afterwards), but with a significantly reduced amplitude by a factor of 1/2.54 during nighttime (see P8 L8). This factor was calculated from the ratio of available daytime and night time fluxes during the grazing phase. We will rephrase the first part of Section 3.2 to clarify the applied gap filling procedure and better explain the difference between daytime and nighttime cases.

Regarding the discussion about u_* and temperature dependency, we already included this effects in our discussion and figures (P7 L28-30, P8 L9-10, Fig. 4) in a qualitative way. Due to the similar temporal pattern of u_* (wind speed) and temperature at the study site and the frequent calm nighttime conditions it is unfortunately not possible (with a high degree of confidence) to disentangle the dependencies further. Yet we will include a reference to Flechard et al. (2013) stating why NH_3 emissions tend to be lower during nighttime conditions.

2. Reference to the work of Moring et al. (2016) is lacking. In reference to this work, I wonder if considering the source as a mosaic of emission and deposition hot-spots rather than a distribution of emission patches would conceptually change the results presented here. Could the author elaborate on this question?

There is actually a reference to Móring et al. (2016) in the manuscript (P12 L3). But we assume that the referee wanted to point towards the issue of simultaneous emission (from the excreta patches) and deposition (on the remaining pasture area) on the pasture field. In this respect, our measured fluxes represent the effective net NH_3 flux attributable to the grazing excreta (combination of emission and re-deposition within the measured paddocks). But it does not include the large-scale background deposition, because the latter would not produce a horizontal concentration difference. Due to conceptual and practical reasons, a partitioning into gross emission and re-deposition was not in the scope of the present study. This would require separate measurements (e.g. by small-scale enclosures) of individual patches and of surrounding depositing surface areas. We will mention these issues in the revised manuscript (Section 2.2.4). In this context, we will reference Móring et al. (2017), where a simplified combination of modelled pasture emission and deposition is presented (while Móring et al., 2016 only presented a model for urine patch emission).

Concerning the artificial source experiment, the effect of re-deposition is presumably small as the downwind concentration was measured at only 6 m distance from the release line. Nevertheless, there might be a small bias towards lower recovery rates. As Häni et al. (2018) showed with a similar artificial release, but with NH_3 measurements at 15 m distance from the source, the dry deposition near the patches may be in the range of 10 %. We therefore assume that the error would be smaller in our experiment.

3. The uncertainty analysis requires more details and especially on the gap-filling of emissions using the standard curve on Figure 5. An example showing a reconstructed emission would be beneficial here. It is difficult also to understand if the uncertainty analysis on gap-filling spans the actual variability in the fluxes shown in Figure 5 (the error-bars in Figure 5 would also need to be explained).

An example of a gap filled time series (black points = reconstructed half-hourly data) is actually shown in Fig. 6. Our relatively simple gap filling approach is mainly based on interpolation between available data (either direct linear interpolation or with the help of the management related curves in Fig. 5). Therefore, a simple comparison between gap filled and measured data is not possible.

The uncertainty analysis in Fig. 7 is treating the (systematic) uncertainty of the cumulative emissions of an individual rotation, and not of half-hourly fluxes. Only the first error source (ΔC_bias) directly results from the systematic errors of the individual measurements. The other relevant error sources in Fig. 7 result from gap filling of missing flux values.

The vertical bars in Fig. 5 indicate the standard deviation of the half-hourly measurements within the 6-hour averaging interval. We will add this information in the figure caption. This variability does not represent an uncertainty but rather the variability in time (mainly between different rotations).

4. The methodology used to derive the dung patches distribution, the way relative deviation of this dung patches Is calculated and the way it is used to correct cow based emissions all need clarifications. I think authors should mention previous work on patches emissions by Moring et al. (2016).

The calibration/validation of the patch emission model in Móring et al. (2016) was based on an experiment with a defined pattern of artificially applied urine patches by Laubach et al. (2012). Therefore, we do not see how we can relate our assessment of real grazing patch distribution to their work. For other references to work of Móring et al. (2016) see response to Comment 2.

However, we agree with the referee that our methodology for determining patch distributions and correcting for their effect should be presented in a better way. In order to achieve this, we will modify and enhance Section 2.3 (Cow and excreta distribution monitoring) in the following way:

"The measured concentration difference and thus the derived NH_3 flux is mainly related to the emission of the surface area between the MD sensor paths on each grazing system (according to the main wind directions, see Fig. 1). This is only a part of the entire paddock area, which was considered as uniformly emitting area in the bLS calculations (Sect. 2.2.4) and for which the average urine N

input was quantified (Sect. 2.4). On pasture paddocks the cows can move freely and therefore the urine and dung patches may not be homogenously distributed on the entire area, which can lead to error prone emission estimates (Auerswald et al., 2010; Bell et al., 2017; Laubach et al., 2013).
In order to assess the spatial distribution of the cow excreta on the paddocks X.11 and X.12 as main emission sources in our experiment, we used two different approaches. The number and position of dung patches was determined with a hand held GPS device within the first 3–5 days after grazing. In addition, the cow positions on the pasture were monitored with a day–night digital camera system at a temporal resolution of 10 minutes. The location of the individual cows were manually marked on the displayed pictures in a post-processing step. However, the night mode often did not yield useful information and therefore images showing the cow positions during nighttime were very sparse.
In order to account for inhomogeneity of the excreta distribution within the investigated paddocks, they were divided as shown in Fig. 3(new). The middle sections between the paired MD sensor paths represent the main source areas of the measured fluxes. Their excreta density $d_{X.meas}$ was related to the density of the entire paddocks $d_{(X.11+X.12)}$ to determine the excreta density correction factor k_d:

$$k_d = \frac{d_{(X.11+X.12)}}{d_{X.meas}} \qquad \text{(Eq. 2)}$$

The exemplary dung patch survey in Fig. 3a(new) shows a positive deviation from the average paddock-wide density for both system M (k_d = 1.28) and system G (k_d = 1.40). However, dung observations were only available for two rotations for the paddock M.11, three rotations for G.11 and two rotations for X.12 while daytime cow position observation by camera was available for the whole measurement campaign for system M, and from rotation three onwards for system G. Missing dung density data were estimated from cow density distributions based on a regression analysis (R^2 = 0.98) between parallel surveys of density anomalies for dung patches and cow positions (Fig. 3b,new). Dung patch and cow position showed a very similar relative distributions with only a small offset. The excreta density anomaly factors k_d (Eq. 2) derived from the combined information of the dung patch and the cow position surveys, were thus used to relate the observed cumulative NH_3 emissions to the entire paddock area and to the cow herd.

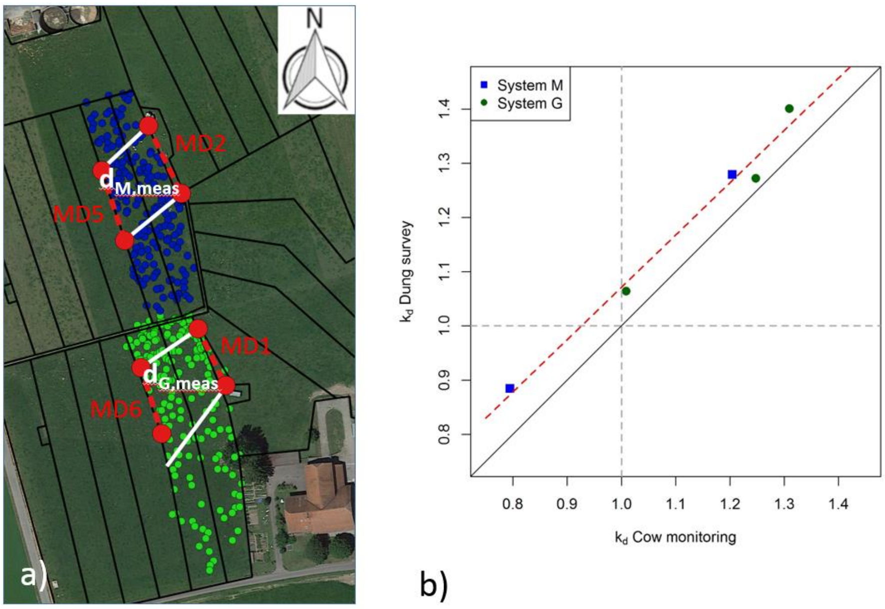

Figure 3 new: (a) GPS tagged dung positions recorded after grazing rotation 7 overlaid on a Google Earth image of the experimental area (Map data: Google, DigitalGlobe). The positions of the MD ammonia sensors/paths are indicated by the red dots/dotted lines. The white lines enclose the main emission measurement area between the sensors. Their dung patch density $d_{X,meas}$ was related to the average density over the investigated paddocks according to Eq. 2; (b) comparison of k_d values according to Eq. 2 for dung patch and cow position distributions on system M (blue) and system G (green)

5. The description of the artificial source quality would benefit from more details on the homogeneity of the emissions, the pressure and flow rates stability. Some more examples on measured concentration and retrieved emissions during these trials would also be beneficial as these data were not published previously (to my knowledge).
In the revised manuscript we will provide more information on the concentrations (mean+std) and absolute emissions of the individual experiments in Table 4. In addition we will provide information on the pressure and flow rate (and their stability) during the release.

Detailed comments:
P4 L7: delete "important" before reference spectrum
Will be changed accordingly.

P4 L14-20: some details on the meteorological instruments may be useful. Please evaluate also how important may be high frequency losses on u and H with 10Hz acquisition at 2 m above ground.*
Weather parameters like wind speed, precipitation, temperature and barometric pressure were measured with a WXT520 (Vaisala, Vantaa, FL). Global radiation was measured with a pyranometer (CNR1, Kipp&Zonen, Delft, NL). High frequency losses on u_* and H due to the 10 Hz acquisition at 2 m above ground were typically below 5 %.

P4 L25-30: 44%-49% missing values for low ustar may actually bias the analysis (see major comments)
See response to Comment 1. The numbers indicated by the referee are total data loss due to wind direction and u_* filtering. We will better specify the individual effects of u_* filtering (26% and 30% for system M and G) in the revised manuscript. Additionally a potential 50% bias in (mainly nighttime) gap filled values is included in the uncertainty analyses.

P5 L6: please give number of thousands of trajectory
We used 25000 trajectories per line point (2 m apart).

*P5 L9: please give units of E, C and D. I woud also suggest to explicit the hypothesis behind this equation: actually Cdown = D*S + Cup, where not other nearby sources are assumed.*
We will add the units in the text, but we prefer not to change the equation because it directly represents the emission determination in the present study. Additionally the used form is consistent to other publications (Bell et al., 2017; Flesch et al., 2004)

P5 L15-18: are the two hypothesis of a uniform and continuous distribution and of a random uniform distribution of sources strictly identical for inverse dispersion?
Since our method is based on line integrating (i.e. line averaging) concentration measurements, we assume that the two hypothesis are equivalent as long as the footprint of the line concentration is large enough to cover a relatively large number of patches (as it was the case here).

P5 L31: the work from Moring et al. (2016) should be referred to here and after.
See response to Comment 4 above.

P6 L4-8: the method used correct the cow-based emissions based on images and GPS needs to be detailed, as suggested in the major comment section.
We agree. See response to Comment 4 above.

P8 EQ-2: The meaning of this is unclear. ΔC_{UD} *and Dup and Ddown should also be time dependent. Please clarify. I would suggest rather using t and Edef, i(t) etc.*
We will modify the text and the axis labelling in Fig. 5 to clarify that *t* used in this equation is not the absolute measurement time but the elapsed time since the end of grazing of the individual upwind paddocks *i*.

P9 L11-15: I would suggest showing the concentration inter-comparison figure in a supplementary material.
The concentrations during the inter-comparison were typically very low at the remote station as the main focus was on retrieving the bias between the instruments. Therefore, we think that a corresponding figure would not provide useful additional information.

P9 L28: please explain what is a "systemic" uncertainty.
This is a typo and should be "systematic" uncertainty.

P10 L2: I am not sure the word "stable" is appropriate here as it may be understood as "stable thermal stratification".
We agree and will change the word to "stationary".

P10 L7-10: I wonder what is the variability in the release rate between the critical orifice. I also wonder if the atmospheric pressure has an influence on the release rate at 30 minutes but also over short time scales (seconds). Finally, what is the expected (or even recorded) effect of wind speed variations on release rates : would one expect some ventury effects on the release rates?
See also response to Comment 5 above. We think the Ventury effect is rather small as wind speeds at the height of the orifices (few cm above ground) were usually low. Additionally it would have no influence on the gas release as the mass flow controller would compensate for pressure fluctuations.

P10 L28-32: It is quite unclear what the "relative deviation of the dung density" really is. I would suggest providing the exact equation.
See response to Comment 4. We will add the exact equation.

P11 L1-3: it is unclear how exactly missing values are obtained from regression analysis. Could the authors elaborate on that?
See response to Comment 4 above. The regression analysis is illustrated in the new Fig. 3b displayed there.

P11 L6-12: I would suggest giving details on how the uncertainties are aggregated (may be an equation in a supplementary section?)
See response to Comment 3 above.

Table 2: I would suggest finding a way to separate more clearly G and M in this table as in Table 3
We will try to find a better way to separate the systems.

Table 3: I suggest only proving 1 digit for temperature and none for rainfall.
We agree with the reviewer and will change the entries accordingly.

Figure 5: could you specify the meaning of the error-bars. I would also suggest using negative time values on the left.
The error bars indicate the standard deviation of the measurements within the 6-hour period. We will add this information in the figure caption. We will modify the x-axis according to the referee suggestion.

Figure 8: Could you provide error bars on both released and inverse modelling with measurements. I would also suggest changing one
We will add the uncertainties of the measurements as error bars.

Figure 10: Please explicit the term "relative deviation" in the legend.
We will insert a reference to the equation as described in the response to Comment 4 above.

Figure 11. Please explicit if error bars are standard deviation, standard errors or interquartile
We will include an explanation of the error bars in the figure caption.

References:

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