

## Author response to comments of referee #1

We'd like to thank reviewer #1 for his answer and appreciate his valuable comments.

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

*1. The weakest part of the study was investigation in estimating urine patch locations to avoid assumptions on the uniformity of the source on pasture (by measuring dung pile locations and position of the cows). In applications where bLS is often used in non-uniform sources, it is realized that the detector should be some distance downwind to minimize the impact of non-uniform source on emissions but close enough to resolve horizontal gradient (elevated background concentration, a possible problem in this study).*

We do not really understand this statement. In our view, the estimation of the excreta distribution on a real grazed pasture is, despite the necessary approximations, one of the strength of the present study since this issue is either missing in comparable studies (Bell et al., 2017), artificially forced by distributing urine manually on the pasture (Laubach et al., 2012) or by forcing unrealistically high excreta densities during short experiments (Laubach et al., 2013b). We made dung patch surveys and we applied a robust method to estimate dung patch densities based on visual cow monitoring with camera systems. As pointed out in the manuscript, it is a valid assumption that urine and dung patches are similar distributed on the paddock (P12 L28). Auerswald et al. (2010) also found a similar spatial distribution between urine and dung patches on a low intensity pasture.

We are fully aware that placing a detector further downwind minimizes the impact of a non-uniform source. Nevertheless, as referee #2 also pointed out, the small fields ensured a temporarily high stocking density and hence a good sensitivity of the concentration measurement method. We would loss this advantage if placing the detector further downwind. Additionally, for maintaining two realistic grazing systems over an entire season, it was not possible to keep the animals in smaller paddocks. As we investigated a rotational management, placing the detector further downwind would have resulted in an average emission measurement over multiple paddocks (or differently managed areas). Therefore monitoring the temporal dynamics of the emissions (increase with grazing duration, exp. decrease afterwards) would not have been possible.

However, we realized that the presentation of this issue in the manuscript was not optimal and this may have influenced the referee comment. Therefore we will improve the manuscript in this respect (see specific comments below).

### **Detailed comments:**

*For the majority of the minor (mostly language related) comments we follow the referee suggestions. Here only the comments that need an answer are listed.*

*1/15 insert 'maximum of  $x \mu\text{g N-NH}_3 \text{ m}^{-2} \text{ s}^{-1}$  at the end'*

As the maximum emissions at the end of the grazing period varied (mostly due to different grazing duration), we would like to keep the sentence unchanged. The overall maximum flux value is included in the range given in the previous sentence.

*2/5 'about eight times lower' - could not find this in Kupper et al (2015) - re-check citation*

The 'eight times lower' factor was calculated based on the TAN flows in Fig. 4b in Kupper et al. (2015). That figure shows that the relative NH<sub>3</sub> emission of grazing livestock (8.9% of excreta TAN) is 7.6 times lower compared to indoor housing including storage and spreading of manure (67.8% of excreta TAN). This factor was rounded to 'about eight'. We will make the reference more specific to "(Kupper et al., 2015; see Fig. 4b therein)" and also clarify that the values given there are for total "grazing livestock" in Switzerland.

*2/25 was the model 'WindTrax' by ThunderBeach Scientific - need to cite model*

We did not use the model 'WindTrax' in the present study, but we used the model 'bLSmodelR' as described in Sect. 2.2.4.

*3/10 what was the topography (slope, barriers to flow, etc)*

The field site is generally flat with only a small slope towards South-West. There are no trees or hedges in the main wind sectors. The farm facilities north and south of the experimental field (Fig. 1) are the only barriers to the flow.

*4/12 were pressure and temperature corrections needed, if so give calibration factors*

No temperature or pressure corrections were needed within the given uncertainty range.

*4/12 was light intensity used to filter data, if so, give range*

As mentioned in Section 2.2.3 the miniDOAS measurements were filtered based on the level of light reaching the spectrometer. This led to a data rejection rate between about 1 % and 4 % for the different instruments.

*5/3 describe the model, and what modifications were made to Flesch's model, what was different*

We will add a reference to Häni et al. (2018), which has been published in the meantime (during the discussion phase). The model characteristics and the minor modifications to Flesch's original model are described there. The applied model 'bLSmodelR' itself was already used in other publications for NH<sub>3</sub> emission on pastures (Bell et al., 2017). But it has to be noted that we used the model without the newly introduced deposition module.

*5/15 however, the 'underlying' assumption of homogeneity of the emitting surface is less true with increased distance between the source and detector, please include this - it is unclear why the bLS model was not run in its entirety*

We are not sure whether we fully understand this referee comment. We measured close to the emitting surface (pasture paddock) and the pasture field has a generally small variability concerning the surface roughness (as reported by Felber et al., 2015, for the same site). The bLS model was run with a model domain of 250 m length, hence much larger compared to the actual emitting paddock. This will be clarified in the revised manuscript.

*5/25 state the given NH3 concentration certification*

The NH<sub>3</sub> percentage in the gas mixture had a relative uncertainty of 2%, i.e. the NH<sub>3</sub> mixing ratio was 5% ± 0.1%. We will add this information in the manuscript.

*5/31 'this is not necessarily the case' - this deserves further comment*

We will change the sentence to: “On a pasture cows can move freely and therefore the emission sources like urine and dung patches are usually not homogenous distributed and can lead to error prone emission estimates (Auerswald et al., 2010; Bell et al., 2017; Laubach et al., 2013a).”

*6/1 top page 11 states urine patches are the most important factor - then two ways of trying to estimate where these patches exit is tried by GPS of the dung piles and by locating the position of the cows - this cannot be direct emission map of ammonia since cows do not necessarily defecate and urinate at the same location, and the position of the cow adds little information to estimate urine patches.*

The spatial density distribution of urine and dung patches are not identical but very similar on a pasture (Auerswald et al., 2010). The miniDOAS line sensors integrate over a sufficient number of dung and urine patches, but measurement footprint only covers a part of the oblong paddocks. On some stages of the grazing season we could identify clear density gradients along the main paddock axis (see Fig. 9) with a generally high linear correlation between the distributions of dung and cow positions on the pasture. ( $R^2=0.98$ , see P11 L2). The fitted linear regression was used to estimate missing dung distributions and hence estimate the urine patch distribution for certain rotations. We will add a more detailed description of the procedure in the method section (see response to Referee#2, Comment 4 for details).

*6/7 it is not clear that the error would be reduced by compounding the errors in locating the urine patches, as opposed to assuming a uniform distribution, especially when the uniform criteria declines in importance with some distance downwind.*

As mentioned in the previous comment, we are quite sure that the information on the dung distribution can be used to estimate the distribution of the urine patches. As explained in the response to Comment 1 (see above), we could not have placed our sensors further downwind as we would have lost the possibility to observe the temporal behavior of the emissions as well as the sensitivity of the method (increase in concentration downwind of the paddock).

*7/27 need to expand by providing information on what was done regarding the bLS footprint*

This sentence was misleading because the bLS footprint was not directly used in the flux calculation. We will rephrase the sentence to:

“The field scale fluxes were determined based on the concentration differences of the paired MD systems and the dispersion coefficient  $D$  (see Eq. 1) computed by the bLS model.”

*7/31 is this 50-70 hours per week?*

As shown in the referenced Table 1 the 50–70 hours correspond to the grazing duration on the investigated paddocks X.11 and X.12 per individual rotation.

*1/8 what is a 'strong' data filter - need to rewrite*

We will rephrase and refer to the data filtering criteria described in Sect 2.2.3.

*8/8 explain where the value '2.54' came from*

We will rephrase this paragraph to make it more clear to the reader (see also response to Comment 1 of Referee #2). Because of the low amount of available nighttime data, it was not possible to derive default emission curves for longer nighttime gaps (as shown for daytime conditions in Fig. 5). Thus it was assumed that the general temporal pattern is similar to daytime conditions but with a lower

amplitude for nighttime. The corresponding reduction factor (= 0.39) was based on the overall ratio between mean daytime and nighttime emissions during grazing.

*8/17 are you saying that your design, at specific wind directions, caused an interference of the incoming concentration (upwind) measurement which lead to an under-estimate of emissions - why not filter out the estimates?*

We cannot filter out those periods, as the investigated paddocks were part of an intensive rotational grazing system. This means upwind grazing took place frequently after grazing on the investigated paddocks between the miniDOAS systems. Filtering out those periods would lead to an unacceptable data loss. Additionally the interference effect is relatively small as shown in Fig. 6 (grey line) and Fig. 7 (red boxes). We also presented a way to correct for this effect (P8 L23 - 28). The interference effect has to be considered as a small disadvantage of an experimental design, which was optimized to fulfill several other requirements (see discussion in Section 3.6).

*9/3 use 'recorded' not 'retrieved'*

As the cumulative emissions are also based on gap filled data, we think 'recorded' is not suitable here. Therefore we would like to keep it unchanged.

*9/4 use 'greatest air temperature' and 9/5 'greater emissions'*

After consulting a native English speaker, we would like to keep 'highest' instead of 'greatest'.

*9/6 neither grazing duration nor N input is found in Table 3 - where are these data?*

Table 3 provides information on N input (separated into N excretion total and N excretion urine). Grazing duration can be found in Table 1. We will refer to Table 1 for grazing duration in the revised manuscript.

*10/10 usually as an alternative to mass flow controller, the entire tank is weight before and after, was this done in this study?*

We did not weight the tracer gas cylinder before and after the releases. But we used a sophisticated mass flow controller and checked its performance by measuring the individual orifices as described at P10 L7–11.

*10/16 do you mean 'air pressure'*

No, we mean the pressure within the tube of the artificial source system (between the gas tank and the flow controller). We will rephrase to '... the dynamic pressure within the tubes of the system upstream of the flow controller at the beginning ...'.

*10/16 don't understand the set-up, what was no longer air tight - needs clarification, also need to indicate why air pressure is involved in recovery*

Similar to the previous answer, we did not mean air pressure but the pressure within the tracer gas tubing system. However, the proposed possible explanation for the high recovery rate in the first gas release trial was purely hypothetical. For clarity reasons we will remove it from the manuscript and state that we have no conclusive explanation for this individual result.

*10/19 'an unknown major error source is unlikely' - what does this mean, if unknown how can it be unlikely, delete this sentence as it adds no information - were the results used to correct the emission*

*or was it used to characterize the data? How sure are you that the difference was systematic, if this is important there needs to be a t-test done and if different then an accuracy analysis performed to break the difference into systematic, random and slope errors*

We agree with the referee that the mentioned sentence is not useful and therefore we will omit it. With the artificial source we intended to test the applied methodology against a controlled source in an exemplary way, and it was not intended for a calibration or quantitative correction of the measurements. This will be clarified in the revised manuscript. As the artificial source experiments resulted in an average recovery rate that was not significantly different from 100 % (111 %  $\pm$  18 %) we assume that the used methodology (bLS dispersion modelling, concentration measurements with miniDOAS line sensors) was suitable for quantification of the pasture emissions.

If there exist minor systematic errors in the methodology (within the achieved uncertainty range, see Section 3.3.1), they are supposed to be very similar for both parallel pasture systems, and therefore do hardly affect the detection of differences between the two pasture systems (see P11 L21-22, P12 L29-31).

*11/11 how was this correction done in all systems except system G rotation2, needs clarification - also need to document what this means for this latter value that was not corrected*

We are aware that the presentation of this correction procedure was not clear enough. We will therefore modify and enhance the corresponding method section. More details are given in the response to Referee#2 (Comment 4). We will also add the individual uncertainty ranges in Fig. 10.

*11/11 use 'greater uncertainty'*

After consulting a native English speaker, we prefer to leave the expression unchanged.

*12/3 cited reference not listed*

The cited reference to Móring et al. (2016) is listed correctly (P16 L8).

*13/6 delete 'under real practice conditions'*

*We would like to keep the sentence unchanged as previous studies on ammonia emissions (e.g. Laubach et al., 2012, 2013) were often not performed under realistic pasture conditions or included manual (artificial) application of urine to the soil.*

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Häni, C., Flechard, C., Neftel, A., Sintermann, J. and Kupper, T.: Accounting for Field-Scale Dry Deposition in Backward Lagrangian Stochastic Dispersion Modelling of NH<sub>3</sub> Emissions, , doi:10.20944/preprints201803.0026.v1, 2018.

Kupper, T., Bonjour, C. and Menzi, H.: Evolution of farm and manure management and their influence on ammonia emissions from agriculture in Switzerland between 1990 and 2010, *Atmos. Environ.*, 103, 215–221, doi:10.1016/j.atmosenv.2014.12.024, 2015.

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