

## Review of egusphere-2022-867

Joseph Referee #1

This study presents results from a series of controlled release experiments, designed to evaluate the accuracy and precision of NH<sub>3</sub> and CH<sub>4</sub> flux estimates made using a stochastic Lagrangian dispersion model and various measurement configurations. In particular, the use of open-path and closed-path measurement systems is compared for both gases. For reasons that are well described in the introduction to this study, such controlled release experiments can provide valuable insight to help guide both the measurement and modelling strategy adopted by future studies targeting “real world” emissions. The interpretation of the results presented here is complicated by the fact that each controlled release experiment did not involve the same set of measurement approaches, but I understand the logistical reasons for this. However, I feel that the presentation of both the results and conclusions needs some revision in order to help the reader to draw clear conclusions from this work. Overall, I suggest that the study is well suited for publication in AMT once the following points have been addressed.

The methods and instrumentation used in this study are generally clearly described. However, it would be good to see some more discussion of instrument calibration in this section. It is mentioned subsequently (L389) that ease of calibration is an important advantage of closed-path systems over open-path systems – I have no doubt that this is the case, but it is hard to assess this without more detail on the respective calibration strategies.

Description of the calibration carried out to the CRDS, GasFinders and miniDOAS were added in section 2.2. In addition, Figure S5 and Figure S6 in the Supplementary information were added as examples of a calibration. The advantage of the calibration of closed-path systems over open-path has been more clearly explained at the end of section 3.3.

From a modelling perspective, the bLS model including NH<sub>3</sub> deposition is crucial to the results presented in the paper, but there are no details of it given in the methods section. I appreciate that a full description is given in Häni et al. (2018), but I think it is important to include a basic summary here too (possibly including Eq. 17 and 18 from Häni et al. (2018)).

Equation 5 and Equation 6 together with a more thorough explanation about NH<sub>3</sub> deposition were added in section 2.6.

The presentation of results could in general be made clearer. As the authors state in the introduction, a key component of this study is the simultaneous release of CH<sub>4</sub> and NH<sub>3</sub>, to disentangle methodological and depositional factors resulting in recovery rates less than 1. However, results from the two gases are not really considered together in section 3. For instance, it is concluded that heating Line 3 to a higher temperature resulted in reduced NH<sub>3</sub> loss in VIII-DK, but there is also an apparent improvement in CH<sub>4</sub> recovery rate using Line 3 as opposed to Line 2 for this experiment. How can this be explained? I suggest that the results section needs some reworking to take full advantage of the two-gas releases, so that the NH<sub>3</sub> results in each experiment are considered in the context of the corresponding CH<sub>4</sub> results. It also took me a while to interpret Figs. 2-4. I would suggest combining the two NH<sub>3</sub> figures into a single figure (as has been done for CH<sub>4</sub>). It may be even better to include the results for both gases in a single 2-panel figure, so that the results for each gas can easily be compared for the same experiment. Since the mean values are quoted in the text, it would be good to include these on the plots (as crosses perhaps).

Figure 2, 3 and 4 were combined to one figure, which is now Figure 2. In addition, a new table (Table 2) was added with the information of  $Q_{\text{bLS}}/Q_{\text{NH}_3}$ ,  $Q_{\text{bLS}}/Q_{\text{CH}_4}$ , and  $Q_{\text{NH}_3}/Q_{\text{CH}_4}$ . New NH<sub>3</sub> deposition velocities were calculated with an approach that assumes a recovery equals to the measured  $Q_{\text{CH}_4}$  for each of the measurement systems. This approach allows to see the improvement between Line 1, Line 2, and Line 3.

I would also state explicitly at the beginning of section 3.1 that the QbLS values presented here for NH<sub>3</sub> do not take deposition into account.

Sentence added for clarification at the beginning of section 3.1.

The clarity of section 3.4 could also be improved. The statement that an artificial source has a higher deposition velocity than a real source needs more explanation and discussion. Is this the case in reality, or just a consequence of the way the bLS model is constructed? How does this impact the interpretation of the results presented in this paper? It is unclear to me why the deposition velocity increases with distance from the source, or why this means that most NH<sub>3</sub> is deposited near the source (as stated in L408). More discussion is required to interpret the results shown in Figure 8 – what conclusions should we draw from the comparison against the empirical models?

An explanation to the statement that an artificial source has higher  $v_a^*$  than what is expected from some type of real agricultural source has been added in section 3.4. Explanation of the difference between the two ways of estimating  $v_{ad}^*$  was also added in section 3.4.

We agree that it is unexpected that the deposition velocities increases with distance. The reason for this is presently unclear and should be investigated further. It is mentioned in section 3.4.

The conclusion section does not currently summarise the key results from the paper particularly effectively. The opening statement is not supported by the average all-instrument CH<sub>4</sub> recovery rate that is quoted – this needs to be separated into the two instrument types (as it is for NH<sub>3</sub>).

The CH<sub>4</sub> recovery was also separated into the two instruments in the conclusion. In addition, an improvement of the conclusion was done.

I do not understand the statement “The present study shows that the deposition algorithm included in the bLS model estimates correct NH<sub>3</sub> emissions that considers surface deposition”. My understanding of the results presented here is that the deposition velocity is estimated by the bLS model. This deposition velocity appears to vary with distance and differs from the empirical model results. So I’m not sure how it can be determined that this estimate is “correct”? It seems that the important conclusion of this comparison relies on an interpretation of the differences shown in Figure 8. I think there are important lessons to be drawn from this study regarding both measurement strategy and the modelling of NH<sub>3</sub> deposition velocity, but without a better synthesis of results it is currently hard for the reader to determine what these lessons are.

We have changed the conclusion to address this comment. “A significant fraction of the emitted NH<sub>3</sub> is deposited near the source. Consequently, including the deposition algorithm in the bLS model will have less bias in the emission evaluation at ground level sources (e.g. application of liquid animal manure), compared to elevated sources (e.g. slurry tank). The present study shows that the estimated deposition velocities are in the same order of magnitude in all the releases with some variation across the different approaches (instrument, distance, method).”

Specific points:

There are quite a few cases of incorrect number agreement (e.g. “the downwind concentration were”) – I haven’t listed them below, but it would be good if these could be corrected on the next proofread.

L16 – averaged *over* intervals. **Added as suggested.**

L77 – *non* ideal conditions. **Added as suggested.**

L196 – I’m confused by the fact that there are three experiments and two instruments listed here, but only four background values quoted. Should there be two more? It should be made clearer which value corresponds to which instrument-experiment combination, either by rephrasing the sentence or adding a table. **Sentence added to clarify the background used in each experiment done in Switzerland in section 2.4.**

L249 – with *an* empirical equation **Added as suggested.**

L251 – *Re* is unidirectional **Added as suggested.**

L334 – If I’m interpreting the results correctly, there is no significant difference between the Line 2 NH<sub>3</sub> results at different distances. In which case I would suggest removing this sentence. **Good point, deleted as suggested.**

L336 – remove “stick” **Deleted as suggested.**

L339 – it would be good to expand on why there was no difference after an hour.

L347 – sentence needs rephrasing **Changed as suggested.**

L393 – sentence needs rephrasing **Changed as suggested.**

L406 – remove “increases, with many cases” **Deleted as suggested.**

L416 – this discussion loses me; in what way are the values from Line 1 higher than those from Lines 2 and 3? This seems to directly contradict the values stated above.

**We believe that the text is correct. But, we have changed the text to make it more clear and avoid confusion.**

L571 – The doi for Häni et al. (2018) is for a preprint – please replace with the doi for the final published article. **Changed as suggested.**