Response to Reviewer 2

General

We thank both reviewers for their thorough reviews and their wealth of suggestions. We greatly appreciate their input. We recognize in their reactions that the progress that we have made in the described research now opens up a large number of new questions: our research team felt exactly the same. With the reviewers help we now harvested a number of good ideas to further optimize the data evaluation of this experiment, but more important: of experiments to come. Because facing the editor's request to reduce the length of the paper substantially does not allow expanding the evaluation on all aspects highlighted by the reviewers. We have tried to find a balance between which parts of the review comments we take on board now, and which parts we will take with us to our next campaign.

A main decision is how to deal with the footprint issues raised when evaluating the data. In the past, the Cabauw site has worked very well as a location for many flux intercomparison experiments with other gases (e.g. Peltola et al., 2014 https://bg.copernicus.org/articles/11/3163/2014/). However, this campaign has shown that it is probably somewhat too inhomogeneous for detailed intercomparison of ammonia fluxes. This was aggravated by the wind directions during most of the campaign, which were different from the normally prevailing southwesterly winds. Considering that (1) we did not collect the detail of activity data in the direct surroundings for a proper evaluation of the impact of different footprints on the fluxes of EC & AGM, and (2) that we are urged by the editor to reduce the paper length considerably, we have decided to limit our analysis in this paper to the standard, 3D-homogeneous flux approach. We are aware that with this approach some of the observed differences will originate from footprint issues. These issues will get more emphasis and attention in our next campaign, that will be located in a more homogeneous area.

In the remainder of this document we will follow the reviewer's text, and our pointwise reactions and answers can be found in *italic blue text* at the applicable position.

On behalf of the author team,

Daan Swart.

Review of the paper „Measuring dry deposition of ...” by Daan Swart et al.

First I would openly state that I have a long lasting collaboration with some of the authors and I closely followed and discussed their progress. I cannot exclude that I am biased in this sense.

This paper presents an NH3 flux intercomparison with two novel open-path instruments. The major advantage of open-path instruments is the absence of an inlet line that is notoriously influencing high frequency concentration measurements needed to determine fluxes. The measurements took place at the Cabauw station located in the middle of the Netherland. It is an open landscape mostly used as intensive grassland. From a micrometeorological perspective it is good location as most of the time good turbulent conditions are present.

I am very pleased to see the great progress that was achieved regarding the accuracy and precision of the DOAS system. The systems have now reached a level that meaningful vertical gradient measurements are possible. Also the open path system from Healthy photons is precise and fast enough to perform reliable EC measurements.

Looking from a greater distance the overall results summarized with figure 11 are looking very nice. Both systems show similar NH3 fluxes and more important they show the same diurnal structure with identical changeover from deposition to emission. So it is very likely that both instruments do measure an existing mean flux over the surface determining the flux.

I could stop here and say publish as is with some minor corrections already addressed by the first reviewer. But this is not my understanding of a serious review and I will dig in the following a little bit deeper. But keep in mind that this is complaining at a high level.

Specific issues:

(1) I think that the title is not appropriate. The work focus on an intercomparison of two approaches to determine bidirectional NH3 exchange. The title should reflect this.

    We will adapt the title to:
    Field comparison of two novel open-path instruments that measure dry deposition and emission of ammonia using flux-gradient and eddy covariance methods

(2) It would be important to describe shortly the “flux landscape” of the chosen site. It is located in an area with intensive animal production associated with a high NH3 turnover. Generally small areas with high emission densities (stable, storage, manure fields) are surrounded with intensively managed field that are expected to show all the time changes from emissions to deposition mainly controlled by their actual N-status (see e.g. the year around measurements that we did at the Oensingen grassland site many years ago, Flechard et al., 2010 https://bg.copernicus.org/articles/7/537/2010/). Judging from a google maps picture from the area there are many small individual fields as well as stationary sources within the footprint area.

We will move Figure S1 (below) to the main text and add a short description of the surrounding area at Cabauw to the section "Campaign setup and Site". This shows the complexity of the terrain. We received a similar request from the other reviewer.

Figure 1. The area surrounding the Cabauw measurement site. Cabauw is in a flat area at -1m, located in the delta of the river Lek shown in the southeast. The line with housing going east-west, running north of the tower has a series of farms. Map from www.pdok.nl/ (downloaded 07-02-2021).

(3) Deposition mainly occurs during night stable conditions when NH3 is concentrated in the boundary layer and emission occurs during instable conditions when the growing boundary layer leads to NH3 concentration below the "system compensation point". Consequently an inverse relation between concentration and fluxes must be expected, even though at a first glance this seems contra intuitive. In this context I was very puzzled by the sentence in line 488 *"The highest concentrations were observed by both systems at noon when air temperature reached the highest level of the day (Figure 8)"*. I am uncertain whether this is just a slip of the pen or a misunderstanding.

We thank the reviewer for noting this obvious mistake. Figure 8 indeed shows exactly the opposite, as he rightfully expects. We will change the corresponding line to: "The highest concentrations are observed during nighttime when the boundary layer height is small and vertical mixing is limited. During daytime the concentrations decrease due to the rise of the boundary layer and the increased vertical turbulent transport."

(4) The evaluation of the EC data is state of the art. The most important correction is the high frequency damping in the order of 20 to 30%. I am aware that the empirical ogive method needs a lot of good data. I suggest to use only flux data with a good looking covariance function for both NH3 and temperature flux and not using daily mean values.

As outlined in lines 354 to 357, half-hourly flux damping correction factors were filtered applying standard flux filtering criteria. Also, only half-hourly concentrations with OSS (HT light intensity) of less 40 % were filtered out. In doing so, the high-quality cospectra were used to estimate the damping. No further manual filtering of ogives and covariance functions was applied since that may lead to biased results.

Fluxes were corrected with their (own) half-hourly high-frequency correction factor if available as written in line 358. If the required half-hourly correction factor was missing, the daily median correction factor was used and not the average to avoid bias. No changes to the text will be made since the information is already given in the manuscript.

(5) DOAS gradient measurements require a demanding procedure to be precise enough that includes measurements in cross position. I rate this as a very positive development of the DOAS approach that I was not expecting based on our own experiences. But I see a tendency to overestimate the precision of the DOAS measurements. E.g. in line 280 it is stated "*that differences can be measured well below our target precision of 0.1 µgm-3.*" In line 449 it is stated "*The random error of the miniDOAS NH3 concentration differences ... was determined to be 0.088 µgm-3*". This is below the target limit but not well below.

We will adapt line 280 to correct this overclaim. The line will read: "In the Results section (Sect. 4.1), it will be illustrated that after these steps the pair was capable of measuring NH_3 differences within our target precision of 0.1 µg m^{-3}."

(6) I also have an other interpretation of the cross position data.

a. In the second period I judge that there is a systematic offset in the order of 0.14 µgm-3 as this is a consistent value for good wind conditions.

Over the full second cross period, we do not see a systematic offset, but rather an offset that varies gradually between 0 and -0.2 µgm^{-3} during this period. Note that a similar meandering is seen in the wind direction. There is only a very limited number of points (n=5) in the green/light-green wind sector in this period. Similar 'outliers' also occur during cross periods 1 and 3, and may have a different cause, e.g. a very local source. Obviously, we cannot exclude a zero drift completely, but since the zero level in cross periods 1 and 3 is the same, and flux values directly after cross period 2 compare well with the HT-values, we trust the zero level of the miniDOAS instruments to be stable, even if we could not confirm this properly in the second cross period. The stability over many weeks was also confirmed by lab experiments prior to the campaign.

b. In case the wind is coming from the red sector I don't see a mechanism that could produce the measured difference between the two crossed paths, especially for the lower concentration range. The obstacle upwind will affect mostly the turbulence and not the NH3 concentration.

On the mechanism: the offset occurs when part of the path of the miniDOASses is affected by a turbulence plume, and another part is not affected or differently affected. For example: if the first half of the DOAS path is unaffected, and the second half of the path is completely well-mixed by the plume, about 25% of the existing gradient is observed instead of zero.

c. You could also check the EC-NH3 covariance function and time series to see whether there is a major disturbance.

We will check this and report on our findings later.

(7) As far as I know there are many influences that potentially influences concentration determination in the order of 0.1 to 0.2 µgm-3 especially under field operation. Judged from figure S5 such a correction could at least partly explain the higher NH3 emission of the AGM approach around noon.

It is clear that the determination of the flux by AGM is the most difficult at noontime when both concentrations and gradients are typically smaller due to the daytime convection and boundary layer rise. A small systematic offset in the zero of the difference measurement is more relevant at that time of day. When comparing figure 10 in the main text, and figure S7 from the supplement, it appears however that there is also an influence from the spreading of manure: results of the instruments are closer together at noon when no manure is spread. The issue is possibly also related to the interplay of an inhomogeneous source/sink field with the different footprints of the instruments. We aim to look into this further in our next campaign, which will be performed in a less complex terrain.

(8) The whole footprint discussion should be modified. In case of the EC approach the calculated fluxes are representative for a small volume at a height of 2.8m. In case of the AGM approach it is a mean vertical flux integrated over a 22m path between 0.76 and 2.26m assuming that the used transfer velocities correctly reflect the atmospheric turbulence. These fluxes need to be translated into exchange fluxes at the soil surface. The simplest approach and implicitly used in most cases is that the vertical flux is constant in all 3 dimension and consequently the measured fluxes are equal to the surface exchange flux. In such conditions footprint considerations are not an issue.

As already indicated under 'General' above, we agree with the reviewer that for now a relatively standard 3D-homogeneous approach is called for, and will adapt the manuscript accordingly where needed. We should however remain aware that the terrain of the campaign is not completely homogeneous, not in space and not in time, over the typical footprint of around 100 to 200 meter and the full campaign period. It is too patchy in surface parameters, sheep are present and their location is varied, grass is harvested from the land at some locations. This can result in differences in the measured fluxes of the two instruments. See also our answer to reviewer 1 under (24).

(9) The footprint climatology shown in Figure 13 gives a good impression which area will on average determine the measured fluxes. But the analysis should be adapted to the selected data point (green and light green conditions).

We agree and will include as Figure 13b the EC-footprint for only the points in the green and light-green sectors, as shown below.

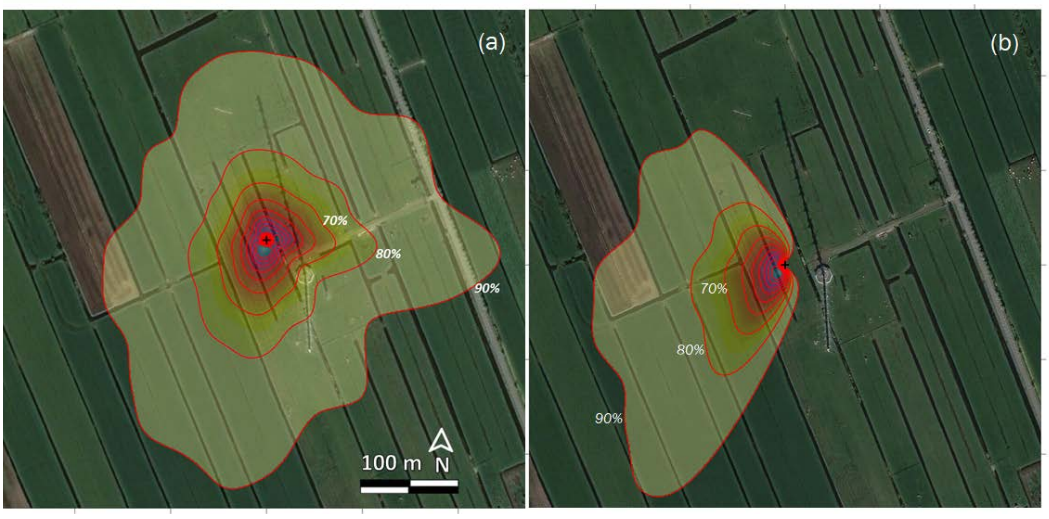

Figure 2. EC footprint for (a) all wind sectors and (b) the green and light-green wind sectors

(10) NH_3 exchange fluxes will most likely be different between all the small fields in the neighborhood of the measuring point within the footprint. Consequently EC and AGM approach will considerably differ as the footprint density function for the two approaches differ. Note also that this function will considerably differ for the lower and upper DOAS path.

We agree. A slightly more elaborate footprint evaluation was done using Kljun's online tool that uses satellite observations for land class identification (https://geography.swansea.ac.uk/nkljun/ffp/www/). That analysis already shows the differences in the land classification on a small scale around the tower. As stated already we lack activity data on the different plots within the footprint area, so we can state that this can cause differences but not more than that.

It is indeed also clear that the miniDOAS footprint of the upper and lower path will be different. Also, the fact of a line-average versus a point measurement plays a role in determining the footprint region of the AGM. We currently do not have a proper analysis for that but assume the AGM footprint will be slightly smaller than the EC footprint in our study because the miniDOAS instruments are on average lower than the EC. Obviously, all these issues disappear when we assume a 3D-homogeneous flux field in the analysis, as we do in this paper.

(11) I also suggest to separately calculated the footprint density function for stable and instable conditions. By the way the bls-R tool developed by Christoph Häni is appropriate to do so. It allows to calculate concentration and flux footprints µgm-3 (https://github.com/ChHaeni/bLSmodelR). It would interesting to overlay these footprint density function on a land use map. This would allow a plausibility control whether the recorded differences in Figure 11 reflect a topographical effect or whether systematic

effects in the measurement systems are in the focus. But an in depth footprint analysis needs detailed information on the land use.

We would definitely like to use this tool for upcoming experimental sessions where we will continue the evaluation of this campaign. As we need to shorten the paper at the editor's request we will however not include this exercise in this paper.

To summarize I suggest to do:

- Reconsider the title
- Add a paragraph regarding the expected flux landscape at Cabauw. This can be integrated to the discussion of the concentration characteristics.
- Down scale the footprint issue. The presented fluxes assume a homogeneous vertical flux and that the used set of turbulence parameter reflect the present meteorological conditions. A consideration of the footprint is for another paper.

(12) For my curiosity I would like to see in a supplement or in the main paper a plot with the AGM concentration differences, the transfer velocities and the fluxes (extension of figure S5).

Here is the figure.

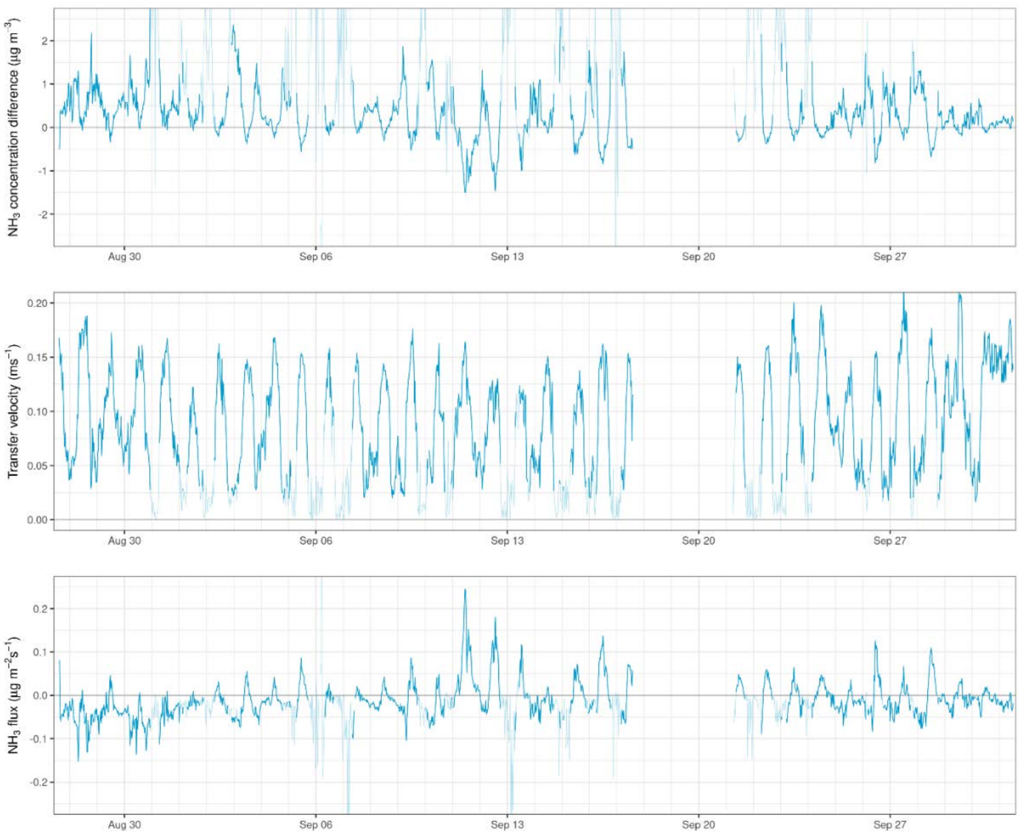

Figure 3. The measured miniDOAS NH_3 concentration differences ($\mu g\ m^{-3}$), the transfer velocities ($m\ s^{-1}$) and the AGM NH_3 fluxes ($\mu g\ m^{-2}\ s^{-1}$). In this graph, cross-periods are excluded and the semi-transparent lines show the data during $u^*<0.1$ conditions.

(13) Regarding the EC data it would be helpful to see NH3'w' covariance function as well as ogives for a few cases, both from the green and red sectors. (not only the best!).

Figure 3 shows ogives and covariance functions of the heat and NH3 flux from the yellow sector. The yellow sector was chosen since it can be treated as a compromise between good and bad flux conditions. No damping estimation was possible for (a) and (b) since their corresponding covariance functions (g) and (h) showed no distinct peak and a high variability at the sides making a reasonable flux estimation impossible. In example (c), damping with the ogive method would probably be underestimated since significant damping is given in the low-frequency range. Considering the time of the day, the issues in ogives and covariance functions were probably caused by fog or dew. Water droplets may have been accumulated on the mirrors. (d) to (f) and their corresponding covariances were measured around noon. They show the typical ogives observed at the site with the highest flux loss between 0.1 and 1 Hz. Figure 4 shows ogives and covariance functions of the heat and NH3 flux from the red sector. Compared to Fig. XX, flux damping during noon was lower. For cases (d) and (e), damping estimation is possible but associated with large uncertainties. No flux estimation was possible for example (l) of Fig. YY. Overall, damping estimation based on measured ogives/cospectra was possible for the different sectors. However, filters regarding time lag, flux quality flag, and instrument performance are mandatory to improve the quality of damping factors.

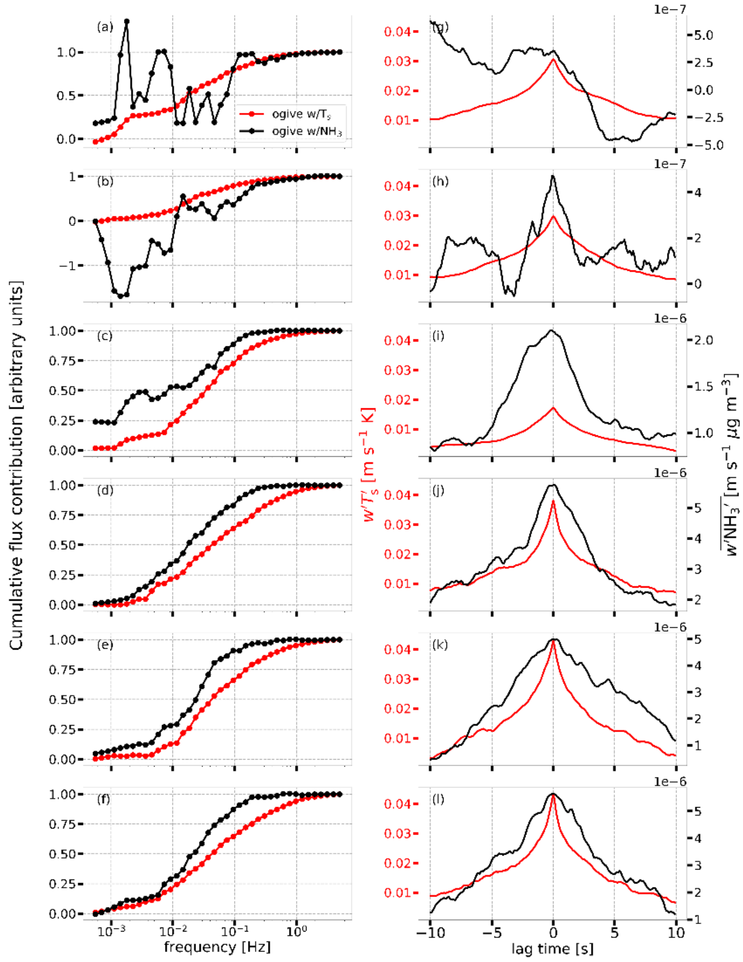

Figure 4. Flux ogives and covariance functions of the heat and NH3 flux of different half-hours from the yellow sector on the 13th of September. (a) to (c) and (g) to (i) refer to times from 8:00 until 9:30 UTC/LT, and (d) to (f) and (j) to (l) refer to times from 12:00 until 13:30 UTC/LT.

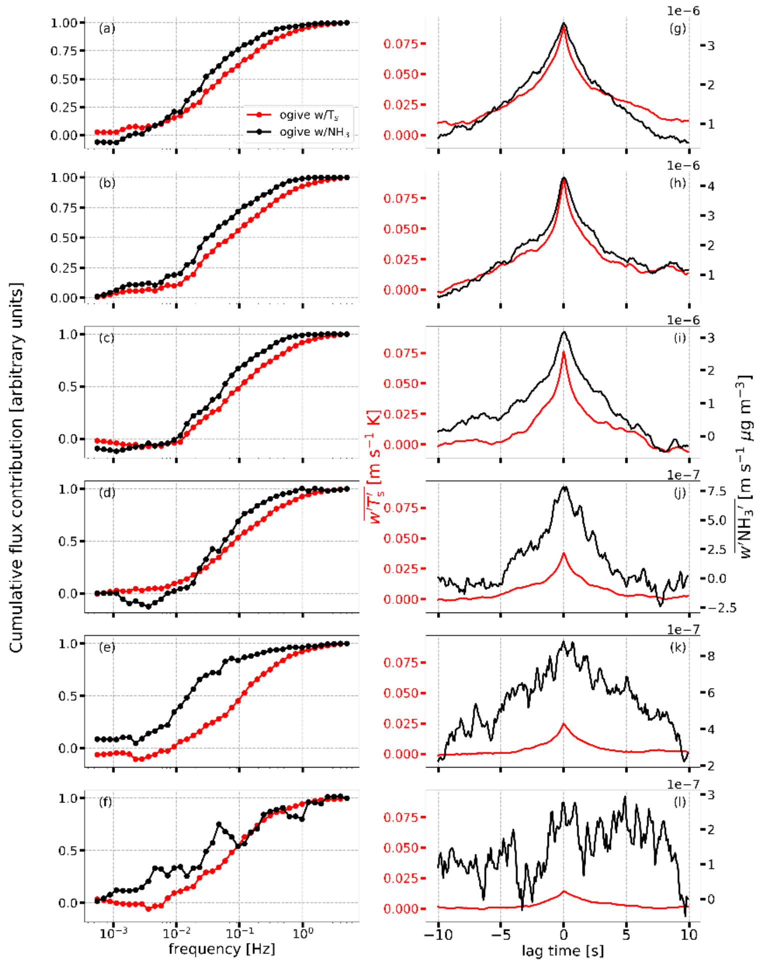

Figure 5. Flux ogives and covariance functions of the heat and NH3 flux of different half-hours from the red sector on the 8th of September. (a) to (c) and (g) to (i) refer to times from 11:30 until 13:30 UTC/LT, and (d) to (f) and (j) to (l) refer to times from 15:30 until 16:30 UTC/LT.